Multimode Atomic Pattern Formation via Enhanced Light-atom Interactions

by

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Dissertation submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in the Department of Physics in the Graduate School of Duke University 2016
Abstract

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Abstract

The nonlinear interaction between light and atoms is an extensive field of study with a broad range of applications in quantum information science and condensed matter physics. Nonlinear optical phenomena occurring in cold atoms are particularly interesting because slowly-moving atoms can spatially organize into density gratings, which is useful for studying light-matter interactions in structured materials. In this thesis, I describe a novel nonlinear optical effect that arises when cold atoms spatially bunch in an optical lattice. I show that employing this spatial atomic bunching provides access to a unique physical regime with reduced thresholds for nonlinear optical processes and enhanced material properties. Using this method, I experimentally observe the nonlinear optical phenomenon of transverse optical pattern formation at record-low powers. These transverse optical patterns are generated by a wave-mixing process that is mediated by the cold atomic vapor. The optical patterns are highly multimode and induce rich non-equilibrium atomic dynamics. In particular, I find that there exists a synergistic interplay between the generated optical patterns and the atoms, wherein the scattered fields help the atoms to self-organize into new, multimode structures that are not externally imposed on the atomic sample. These self-organized structures in turn enhance the power in the optical patterns. My work represents the first direct observation of multimode self-organization in atoms. I characterize the atomic motional dynamics using parametric resonance and Bragg scattering techniques, and I show that the atoms self-organize
into gratings characterized by both sub- and super-wavelength lattice constants. I show that the multimode generated fields induce Sisyphus cooling in all three spatial dimensions, which is the first observation of spontaneous three-dimensional cooling. I also provide a self-consistent theoretical model for studying light-atom interactions in an optical lattice, and I show that by using tightly-bunched atoms, one can enhance the nonlinear refractive index by more than two orders of magnitude c.f. a homogeneous gas. I also develop the first stability analysis for pattern formation that allows for tight atomic bunching. My work represents a unique means by which to study nonlinear optics and non-equilibrium dynamics at ultra-low required powers.
This thesis is dedicated to my family

with many thanks for their inexhaustible support and encouragement.
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1.1 **Single-mode self-organization.** Panel 1: Atoms inside a single-mode cavity organize in a 1D optical lattice applied orthogonal to the cavity axis. Panel 2: A small field starts to build inside the cavity. Panel 3: When the cavity field is sufficiently strong, the dipole force bunches the atoms into self-organized structures. This figure is adapted from Ref. [Baumann *et al.* (2010)].

1.2 **Overview of experimental setup.** (a) I apply two counterpropagating optical fields along the long (\(\hat{z}\)) axis of the atoms in a linear polarization configuration. Optical field polarizations are defined with green arrows/circles. The dimensions of the cloud are typically length \(L = 3\) cm and width \(w \approx 400\) \(\mu\)m. After Sisyphus cooling, the atoms are tightly bunched in the applied 1D optical lattice so that they form pancake-shaped structures, as depicted in the rectangle. (b) Above a threshold \(\chi_{\text{eff}}\), new optical fields are generated along a cone of half-angle \(\theta \approx 3 – 10\) mrad. (c) Examples of various optical patterns. The central spot is bleed-through pump light. The small ring closely surrounding the pump spot is a beam reshaping effect that arises when the pump size is comparable to \(w\). All other spots around the allowed ring of emission are the optical patterns. (d) Example of an interference pattern between a pump field and a nearly counterpropagating pattern-forming optical field. (e) Example of an interference pattern between a pump field and a nearly copropagating pattern-forming optical field. (f) For a two- (six-) spot optical pattern, the self-organized structures within each pancake are stripes (hexagonal spots).

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2.2 **Light-shifted energy states.** When an optical field of frequency \( \omega \) and detuning \( \Delta = \omega - (\omega_e - \omega_g) \) is incident on an atom, the ground and excited state energies shift by \( \Delta E_{e,g} \).

2.3 **Radiation pressure force.** When an atom absorbs a photon of wavevector \( \vec{k} \), it gets a momentum kick in the direction of the incoming photon, resulting in an average momentum transfer of \( \Delta \vec{p}_{\text{in}} = \hbar \vec{k} \). Subsequent spontaneous emission can occur in any direction, with an average momentum transfer over many emission events of \( \Delta \vec{p}_{\text{out}} \rightarrow 0 \). Thus, the total average momentum transfer to an atom per scattering event is \( \Delta \vec{p} = \hbar \vec{k} \), which gives rise to the radiation pressure force.

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3.1 **Enhancing transverse perturbations.** 1. Counterpropagating optical fields are applied to a \( \chi^{(3)} \) nonlinear optical material. 2. Spontaneous emission events give rise to transverse perturbations in the light-matter interaction. 3. Certain transverse perturbations are enhanced via wave-mixing processes inside the material. 4. For a sufficiently high \( \chi^{(3)} \), these wave-mixing processes generate macroscopic optical fields, which we can transverse optical patterns.
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7.4 **Results of stability analysis.** Single-beam intensity normalized by the resonant saturation intensity as a function of (a) the detuning normalized by the natural linewidth for the specific case of an optical depth of 62, $L = 3$ cm, $\tilde{T} = 3/146$, and red detunings with a free parameter value $p = 6$, and (b) the optical depth for the specific case of an optical depth of $\tilde{\Delta} = -5$, $L = 3$ cm, $\tilde{T} = 3/146$, and a free parameter value $p = 54$. The blue curve represents the predictions from Eq. 7.66 with free parameter $\tilde{I} \rightarrow p\tilde{I}$, and the red points represent experimental data. The red rectangle represents the statistical error due to the initial intensity measurement, the detuning measurement, and the confidence interval of the fit to the OD measurement. (See App. A.)

A.1 **Magneto-optical trap.** (a) Picture of the vacuum chamber with the rectangular coils underneath the cell. (b) Images (from both above and side perspectives) of the cloud of atoms fluorescing during the application of the cooling and trapping beams. These photos were taken by Joel Greenberg and included in Ref. [Greenberg (2012)].

A.2 **Schematic of MOT beams.** Ti-Sapph light is sent through an AOM. The first-order diffracted light is sent to the MOT. The repump light from the DFB laser is combined with the radial cooling and trapping beams from the Ti-Sapph. The radial cooling and trapping beams are sent through telescopes that use cylindrical lenses (CTS), and the third cooling and trapping beam is sent through a regular telescope (TS). The radial beams at the cell are elliptical with dimensions $\sim 5$ cm by $\sim 2$ cm, and the third beam is circular with diameter $\sim 2$ cm. The radial beams are sent through quarter-wave plates to create circularly polarized fields and are then retroreflected by a gold mirror at the bottom of the vacuum chamber. The third beam, which is applied at an angle $10^\circ$ relative to the magnetic field zero, is retroreflected off a mirror, where a quarter-wave plate (“$\lambda/4$”) is used to preserve the polarization. The magnetic field coils (not shown) are under the vacuum chamber and create a quadrupolar field with a zero-field region inside the chamber.
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List of Abbreviations and Symbols

Symbols

- $I_p$  Pump beam intensity.
- $I_{\text{sat}}$  Resonant saturation intensity.
- $I_{s\Delta}$  Off-resonant saturation intensity.
- $\tilde{I}$  Intensity normalized by the off-resonant saturation intensity.
- $\Delta$  Frequency detuning from the resonant atomic frequency.
- $\Gamma$  Natural linewidth.
- $\tilde{\Delta}$  $\Delta/\Gamma$.
- $T$  Atomic temperature.
- $\tilde{T}$  Atomic temperature normalized by the Doppler temperature.
- $k$  Wavenumber in vacuum.
- $k'$  Wavenumber inside a material of index of refraction $n$: $k' = kn$.
- $L$  Length of the cloud of atoms.
- $w$  Beam waist.

Abbreviations

- MOT  Magneto-optical trap.
- MPO  Mirrorless parametric oscillation.
- 4WM  Four-wave-mixing.
Acknowledgements

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Introduction

The spontaneous emergence of patterns in non-equilibrium systems is a phenomenon that occurs on nearly all scales of nature, including the formation of galaxies, the flocking behaviors of birds and fish, and cellular growth. Since the pioneering work of Turing [Turing (1952)], it became evident that pattern formation can be triggered on an infinitesimal scale, where small perturbations give rise to an instability that cascades into macroscopic ordering—also known as self-organization.

Self-organization has also been studied extensively in coupled light-atom systems, where the initial perturbations are enhanced by a nonlinear interaction between light and atoms. In recent years, self-organization in cold-atom systems has attracted substantial interest because such slowly moving atoms can form into real-space atomic patterns [Black et al. (2003)], which has led to numerous experiments studying phase transitions [Baumann et al. (2010)] and spontaneous symmetry-breaking [Baumann et al. (2011)].

Self-organization of cold atoms has been observed in single-mode cavities with both thermal [Black et al. (2003)] and Bose-condensed [Baumann et al. (2010)] atoms. In these experiments, atoms bunch in an optical lattice applied orthogonal to the
**Figure 1.1: Single-mode self-organization.** Panel 1: Atoms inside a single-mode cavity organize in a 1D optical lattice applied orthogonal to the cavity axis. Panel 2: A small field starts to build inside the cavity. Panel 3: When the cavity field is sufficiently strong, the dipole force bunches the atoms into self-organized structures. This figure is adapted from Ref. [Baumann et al. (2010)].

cavity axis, as shown in the first panel of Fig. 1.1. Then, via spontaneous emission, a small field starts to build up inside the cavity. Eventually, the field becomes strong enough to bunch the atoms into new structures that are distinct from the applied lattice, as shown by the vertical structures in panel 3 of Fig. 1.1. These new structures that form along the single-mode cavity axis are termed “self-organized” because they are not imposed by externally-applied optical fields.

Self-organization of ultracold atoms in multimode geometries allows access to different non-equilibrium physics, such as continuous symmetry-breaking [Gopalakrishnan et al. (2009)]. To work in a multimode geometry, one must either use a multimode optical cavity or work in a naturally-multimode free-space system. However, in order to observe self-organizing instabilities, one requires strong nonlinear light-atom interactions [Tesio et al. (2014)]. While multimode optical cavities allow multiple passes of optical fields through the atoms and, subsequently, enhanced light-atom interaction strengths, they are experimentally difficult to use because they require sub-wavelength-precision alignment [Kollár et al. (2015)]. Free-space systems have the advantage of experimental simplicity, but in order to achieve strong light-atom interactions in free space, one must enhance the nonlinear optical response of
atoms to incident optical fields.

My contribution to the fields of nonlinear optics and atomic physics is the experimental realization of coupled multimode optical-atomic pattern formation by utilizing enhanced light-atom interaction strengths in free space. By using ultracold atoms that are bunched in an optical lattice and allowing them to interact strongly with the lattice-forming optical fields, it is has been shown experimentally that one can enhance the nonlinear light-atom interaction strength \cite{Greenberg2011, Greenberg2012b}.

Above a threshold nonlinear refractive index, an instability gives rise to the generation of new optical fields, which I refer to as optical patterns. By working in the strong-coupling regime, there is a synergistic interaction between the optical patterns and the atoms such that the atoms self-organize into structures that depend on the geometry of the optical patterns. I observe experimentally that the atoms self-organize into real-space patterns under the influence of the dipole forces imposed by the spontaneously-generated fields, which represents the first direct observation of multimode atomic self-organization. I also show that the self-organization of these atomic patterns is enhanced by spontaneous three-dimensional (3D) Sisyphus cooling that arises due to the interaction among the pattern-forming optical fields and the applied lattice fields. This represents the first observation of spontaneous 3D cooling, which is useful for achieving longer coherence times in cold-atom experiments \cite{Raithel1997b}. While other cold-atom systems have observed the spontaneous emission of multimode optical fields \cite{Labeyrie2014, Greenberg2012b}, they have not measured real-space multimode atomic self-organization nor 3D cooling.

In this thesis, I also present a self-consistent theoretical model describing ultracold atoms in an optical lattice, and I show that one can enhance the nonlinear refractive index beyond that which is achievable in a homogeneous gas by allowing the atoms
to tightly bunch at the lattice sites [Schmittberger and Gauthier (2014)]. By using this method to enhance the light-atom interaction strength, I am able to study pattern formation at ultra-low light levels. I also extend this model to develop the first stability analysis for predicting the threshold for pattern formation in a gas of tightly bunched atoms.

In this chapter, I review previous work involving ultracold atoms in optical lattices, and I describe the unique regime in which I work c.f. these systems. I then review other optical and atomic systems that observe pattern formation and briefly describe the physical phenomena that give rise to the pattern-forming instability. Finally, I provide an overview of the work presented in this thesis and describe its contributions in the context of previous work in nonlinear optics and atomic physics.

1.1 Optical lattices

Soon after the techniques of magneto-optical trapping were established, there was a surge of experiments that took advantage of the benefits posed by sub-Doppler-cooled atoms; e.g., such slowly-moving atoms do not suffer from extra absorption due to Doppler-broadened linewidths [Brown et al. (1997); Metcalf and van der Straten (1999)]. In addition, by 1995, sub-Doppler cooling techniques had been combined with evaporative cooling to achieve Bose-Einstein condensation [Bradley et al. (1995)]—a milestone that fused the worlds of atomic physics and condensed matter physics. However, even in the early years between the first magneto-optical trap and a Bose-Einstein condensate (BEC), researchers used sub-Doppler-cooled atoms to spatially trap atoms in optical lattices [Jessen et al. (1992)].

Atoms become spatially trapped in an optical lattice when their thermal energy is less than the dipole potential energy of the lattice, which is a function of the intensity of the applied optical fields. Thus, sub-Doppler cooling allows researchers to trap atoms in optical lattices without the need for ultra-high laser intensities.
Spatially bunched atoms give rise to a spatially dependent refractive index, which allows for experiments involving Bragg scattering [Birkl et al. (1995); Weidemüller et al. (1995)] and studies of photonic band gaps [Deutsch et al. (1995); Petrosyan (2007); Schilke et al. (2012b)], for example.

In the regime where the lattice-forming optical fields interact strongly with the bunched atoms, the atoms can modify the properties of the optical fields. This light-atom interaction is a synergistic process, wherein the optical fields act to bunch the atoms and modify their effective refractive index $n$, which in turn modifies the effective wavenumber of the optical fields $k' = kn$, where $k$ is the wavenumber in vacuum. Synergistically-enhanced light-atom interactions can give rise to collective effects, such as superradiance [Dicke (1954)] and collective atomic recoil lasing [Bonifacio and Salvo (1994)], as well as reduced thresholds for nonlinear optical processes, such as four-wave-mixing [Muradyan et al. (2005); Saffman and Wang (2008)].

It is a common belief that in order to reach the regime of enhanced light-atom interactions, one must use an optical cavity [Ritsch et al. (2013)]. While optical cavities enhance the light-atom coupling strength, there are alternative methods for achieving strong light-atom interactions that do not require the use of a cavity.

In 1995, it was shown theoretically [Deutsch et al. (1995)] that atoms that are tightly bunched in a free-space optical lattice give rise to enhanced synergistic coupling strengths. In Ref. [Deutsch et al. (1995)], the authors approximate the tightly-bunched atoms to be infinitesimally-thin sheets of dielectric material (i.e., temperature $T = 0$), and they show that under certain experimental conditions, the atoms bunch at the intensity maxima of the optical lattice, which results in enhanced light-atom interaction strengths. However, one does not require zero-temperature atoms in order to achieve enhanced coupling strengths using this method.

I show theoretically that one can also achieve enhanced light-atom interaction strengths in finite-temperature systems using cold, thermal atoms that are bunched
in an optical lattice [Schmittberger and Gauthier (2014)], which allows me to experimentally study nonlinear effects, such as self-organization, at low required intensities. One way to quantify the strength of the light-atom interaction is via the effective susceptibility $\chi_{\text{eff}}$, where $n \approx 1 + \chi_{\text{eff}}/2$. I show that by allowing finite-temperature atoms to bunch in the intensity maxima of an optical lattice, one can enhance $\chi_{\text{eff}}$ by more than two orders of magnitude c.f. a homogeneous gas.

In order to achieve this regime of enhanced light-atom interaction strengths, I show it is necessary to use ultracold atoms, so that the atoms tightly bunch at the lattice sites, as well as small detunings, where the optical fields are tuned close to the resonant atomic transition frequency, so that the lattice-forming optical fields interact strongly with the bunched atoms. While it is common to use small detunings to study low-light-level nonlinear optics [Boyd (2008)], most researchers in the lattice community use large detunings, i.e., many orders of magnitude larger than the natural linewidth of the atomic transition. The lattice community typically only uses the lattice-forming optical fields to bunch the atoms, and they then apply other optical fields to perform experiments, e.g., for Bragg scattering off the gratings of bunched atoms [Schilke et al. (2012b)]. For these applications, the use of large detunings can be beneficial because it minimizes the synergistic light-atom coupling so that $k' \approx k$, which simplifies the Bragg condition for efficient scattering. However, to achieve enhanced light-atom interaction strengths in free space, I use the lattice-forming optical fields for dual purposes: to bunch the atoms and to drive nonlinear optical processes. As a result, it is necessary for me to use small detunings so that the optical fields interact strongly with the atoms.

In this regime of ultracold atoms and small detunings, one can make $\chi_{\text{eff}}$ large and achieve enhanced light-atom coupling strengths in free space, which allows me to study multimode, nonlinear optical phenomena without the need for a cavity. For sufficiently large $\chi_{\text{eff}}$, there is a transverse instability that generates new, multimode...
optical fields and lattices of bunched atoms, which I refer to as optical and atomic pattern formation, respectively.

1.2 Pattern formation

The transverse instability that gives rise to pattern formation is triggered by a perturbation either in the atomic density distribution or the transverse profile of the applied optical fields, e.g., an atomic fluctuation or spontaneous emission into the transverse plane. This instability generates new, weak optical fields that propagate in the transverse plane as well as weak atomic bunching in structures that are distinct from those in the applied optical lattice. The result is a synergistic, runaway enhancement of the instability, wherein the new optical fields generate additional atomic bunching and cooling, and those bunched structures in turn give rise to increased scattering and power in the generated optical fields.

In my experiment, I apply counterpropagating optical fields down the long (\( \hat{z} \)) axis of a cloud of sub-Doppler-cooled atoms that are linearly polarized perpendicular to one another, as depicted in Fig. 1.2(a), which I refer to as a “lin\_lin” polarization configuration. This cloud of atoms is about 3 cm long and \(~400 \mu\text{m}\) in diameter, and the atoms are initially cooled to a temperature \(T \sim 30 \mu\text{K}\). The applied optical fields force the atoms to undergo an additional cooling process, known as Sisyphus cooling, which cools the temperature component of the atoms along the \( \hat{z} \)-axis to \(T_z \sim 2–3 \mu\text{K}\). The lin\_lin polarization configuration gives rise to two superimposed dipole potentials that together have dipole potential minima occurring every \( \lambda'/4 \approx 195 \text{ nm} \) (instead of \( \lambda'/2 \), as would be the case for parallel polarizations), where \( \lambda' = 2\pi/k' \). This is discussed further in Ch. 2.

After Sisyphus cooling, the atoms are tightly bunched in the optical lattice so that they form pancake-like structures, as depicted in the inset of Fig. 1.2(a). Above a threshold value of \( \chi_{\text{eff}} \) \((i.e., \text{for sufficiently high-intensity fields, cold atoms, and/or}) \)
Figure 1.2: Overview of experimental setup. (a) I apply two counterpropagating optical fields along the long (\(\hat{z}\)-) axis of the atoms in a lin\(\perp\)lin polarization configuration. Optical field polarizations are defined with green arrows/circles. The dimensions of the cloud are typically length \(L = 3\) cm and width \(w \sim 400\) \(\mu\)m. After Sisyphus cooling, the atoms are tightly bunched in the applied 1D optical lattice so that they form pancake-shaped structures, as depicted in the rectangle. (b) Above a threshold \(\chi_{\text{eff}}\), new optical fields are generated along a cone of half-angle \(\theta \sim 3 - 10\) mrad. (c) Examples of various optical patterns. The central spot is bleed-through pump light. The small ring closely surrounding the pump spot is a beam reshaping effect that arises when the pump size is comparable to \(w\). All other spots around the allowed ring of emission are the optical patterns. (d) Example of an interference pattern between a pump field and a nearly counterpropagating pattern-forming optical field. (e) Example of an interference pattern between a pump field and a nearly copropagating pattern-forming optical field. (f) For a two- (six-) spot optical pattern, the self-organized structures within each pancake are stripes (hexagonal spots).
small detunings), a transverse instability generates new optical fields that emerge at a small angle $\theta \sim 3$ to 10 mrad relative to the applied fields, as depicted in Fig. 1.2(b). The cylindrical symmetry of the generated fields conserves momentum in the wave-mixing process. Figure 1.2(c) shows example images of the generated optical fields, which form multi-spot optical patterns when imaged in the far-field. I obtain different types of patterns by changing $\chi_{\text{eff}}$ or by changing the alignment of the pump fields. However, I also observe that the atoms can generate different patterns under the same experimental conditions. This is discussed further in Ch. 4.

While the pancakes of Fig. 1.2(a) are imposed structures of bunched atoms, I show in Ch. 5 that within each pancake, there exist additional structures that are self-organized. The self-organized structures emerge according to the dipole potentials generated by the interference between the applied fields and the pattern-forming optical fields. Two examples of these interference patterns are shown in Figs. 1.2(d) and (e), and there exist complimentary patterns for all pairs of applied and generated fields. The interference pattern generated by an applied field and a nearly counter-propagating generated field overlaps spatially to within 1% of the imposed pancakes depicted in Fig. 1.2(a), with a period close to $\lambda'/4 \approx 195$ nm, as shown in Fig. 1.2(d). The interference pattern between an applied field and a nearly copropagating generated field are nearly orthogonal to the others and have a spacing of $\sim 50$ to $100$ $\mu$m, depending on $\theta$, as shown in Fig. 1.2(e). These interference patterns generate new dipole potential wells throughout the atoms. Where these dipole potentials overlap with the imposed pancake structures, there exist self-organized atomic bunching, as simulated in Fig. 1.2(f) for a two-spot and six-spot optical pattern. Because the atomic self-organization is synergistically enhanced by the optical patterns, the geometry of the self-organized atomic structures depend on the geometry of the optical patterns, which gives rise to multimode self-organization via coupled optical-atomic pattern formation.
In the remainder of this section, I describe the history of optical and atomic pattern formation in other systems. I describe the general nonlinear optical principles that give rise to optical pattern formation, and I provide an overview of past studies of transverse optical patterns. I then describe the unique consequences of studying pattern formation in cold atoms, and I review other work that explores the synergistic interaction between optical fields and spatially bunched atoms.

1.2.1 Mirrorless parametric oscillation

Optical pattern formation is more generally described by the nonlinear optics community as a form of “parametric oscillation.” Parametric oscillation describes a light-matter interaction that generates new, coherent optical fields, similar to a laser. While the gain mechanism in a typical laser requires inversion of atomic population in multi-level energy schemes, gain in parametric oscillators is instead acquired via nonlinear optical wave-mixing processes [Fleischhauer et al. (2000)].

In order for parametric oscillators to “lase,” they must operate in the strong-coupling regime, using either a cavity or enhanced light-atom interactions in free space. When working in free-space, parametric oscillators are termed “mirrorless.” Mirrorless parametric oscillation (MPO) is well known to generate optical pattern formation in a warm atomic vapor [Yariv and Pepper (1977); Firth and Paré (1988); Firth et al. (1990)]. In warm atoms, the instability that triggers MPO is enhanced by the formation of spin (“polarization”) gratings rather than density gratings. After applying counterpropagating optical fields, warm atoms are optically pumped into certain spin states according to the local electric field, which gives rise to a spatially varying refractive index, similar to the formation of density gratings in cold atoms. Despite the difference in the nature of the nonlinearity for warm and cold atoms, the result is the same: a transverse instability is enhanced by MPO and gives rise to pattern formation.
1.2.2 Optical pattern formation

Transverse optical pattern formation has been studied theoretically and experimentally for over three decades. After the first theoretical prediction that optical fields counterpropagating through a nonlinear material could exhibit a transverse instability [Silberberg and Bar-Joseph (1982)], multiple experiments followed that study the spatial [Khitrova et al. (1988)] and temporal [Gauthier et al. (1988)] nature of the transverse instability in a warm atomic vapor. More recently, it was shown that these transverse optical patterns could be controlled by a weak probe beam and used as a low-light-level all-optical switch [Dawes et al. (2005)]. All of these experiments used warm atoms, but different physics is accessible when using cold atoms as the nonlinear optical material.

In cold atoms, the generation of pattern-forming optical fields also gives rise to new dipole potentials into which the atoms can self-organize and form new structures. The first theoretical description of pattern formation in cold atoms was described in Ref. [Muradyan et al. (2005)], and while subsequent experiments by the same research group did not observe pattern formation, they did observe other transverse effects such as self-focusing [Saffman and Wang (2008)].

We reported the first observation of transverse optical pattern formation in cold atoms [Greenberg et al. (2011)]. As I show in this thesis, in addition to self-organization, the use of cold atoms also gives rise to reduced power thresholds for pattern formation c.f. warm atoms because of the different nature of the nonlinearity.

Our observation of cold-atom pattern formation was quickly followed by the observation of MPO in a different setup [Schilke et al. (2012a)] that also generated transverse optical fields. In Ref. [Schilke et al. (2012a)], they instead apply two sets of counterpropagating optical fields: one pair generates a dipole trap to cool and bunch the atoms in a 1D optical lattice, and the other pair acts as the pump
beams to seed the MPO process. In contrast, I use only one set of counterpropagating optical fields, which simultaneously cools and traps the atoms and acts as the pump beams for the MPO process. The difference between our systems lies in the atomic temperature and the need for ultracold atoms: the atoms in Ref. [Schilke et al. (2012a)] are approximately $\sim 10^2$ warmer than in my system and necessitate the use of a dipole trap to bunch the atoms. With much colder temperatures, the pump beams in my experiment are sufficient to create atomic bunching. The importance of using ultracold temperatures to reach the threshold for the pattern-forming instability is discussed further in Chs. 4 and 7.

1.2.3 Atomic pattern formation

With the use of ultracold temperatures, the formation of optical patterns is synergistically coupled to the formation of new gratings of bunched atoms (atomic patterns), which synergistically enhance one another during the MPO enhancement. This type of light-atom synergy was first described in the context of cold atoms in Ref. [Inouye et al. (1999)], which explored the relationship between optical fields and the corresponding matter waves in a BEC. In this experiment, the researchers studied a phenomenon called superradiant Rayleigh scattering, which is in many ways analogous to MPO or degenerate four-wave-mixing [Greenberg (2012)]. Superradiance is considered a “collective” phenomenon because the generated optical power scales with $N^2$, where $N$ is the atom number. In contrast, the generated power depends linearly on $N$ in four-wave-mixing. However, the initial gain mechanism in both systems is that of a wave-mixing instability. Superradiant Rayleigh scattering is a form of superradiance involving only frequency-degenerate optical fields; other superradiance experiments explore non-degenerate processes, i.e., Raman scattering [Bohnet et al. (2012)].

The work of Wolfgang Ketterle’s group [Inouye et al. (1999)] provides a direct
Superradiant Rayleigh scattering in a BEC. Based on simulations and data in Ref. [Inouye et al. (1999)]. (a) A single optical field is applied to an elongated BEC. Atoms undergo collective recoil due to the absorption and superradiant scattering of optical fields. (b) Recoiled atoms can undergo subsequent recoil-induced scattering.

measurement of the effects of scattered optical fields on the density distribution of atoms. In this experiment, they apply a single optical field to the side of an elongated BEC, as depicted in Fig. 1.3(a). Above a threshold intensity, they observe a collective recoil of the atomic sample, which arises from momentum conservation in collective scattering of light. As the atoms continue to scatter light from the incident optical field, they undergo subsequent collective atomic motion, which gives rise to additional collective scattering, as depicted in Fig. 1.3(b). If one images the scattered optical fields, one can obtain information about the locations of the atoms as well as the density of atoms at each location, where higher atom numbers correspond to higher-intensity scattered fields.

My experiment explores a different geometry and temperature regime, but it nevertheless employs the same initial gain mechanism and the idea that optical fields scattered by the atoms provide information about the atomic structure. As simulated in Fig. 1.2(f), the geometry of the optical patterns corresponds directly to the density distribution of the atoms. This light-atom synergy was also explored at length by Joel Greenberg—one of Daniel Gauthier’s previous graduate students who built and
used the same magneto-optical trap (MOT) that I use. In Greenberg’s experiment, he used a different beam geometry, as depicted in Fig. 1.4(a), where he pumped at a large (\(\sim10^\circ\)) angle relative to the long axis of the cloud. He observed collective emission of optical fields along the long axis of the atomic sample. Because of his applied beam geometry, the generated fields were not phase-matched to take the form of transverse optical patterns. However, he did observe multimode emission, as shown in Fig. 1.4(b). There are many similarities between my experiment and Greenberg’s, including the observation of a wave-mixing instability and the use of an elongated cloud of atoms to achieve enhanced light-atom interactions. In addition, Greenberg’s experiment suffers from less pump-beam distortion and absorption because his pump beams are much larger than the length of the cloud of atoms and thus only effectively propagate through the width (\(w \sim 400 \mu m\)) of the sample. In contrast, my applied optical fields must propagate down the entire length (\(L = 3 \text{ cm}\)) of the cloud of atoms.

My work is distinguished from that of Joel Greenberg’s in the following ways: (1) I observe the spontaneous initiation of three competing wave-mixing processes to generate multi-spot optical patterns c.f. one wave-mixing process in [Greenberg (2012)]; (2) I perform \textit{in situ} measurements of the motional states of the atoms, which verifies that the atoms self-organize into real-space atomic patterns on multiple spatial scales; and (3) I observe spontaneous 3D Sisyphus cooling, which allows the patterns to persist for more time, helps facilitate self-organization, and enhances the nonlinear refractive index of the atomic sample.

1.3 Overview

This thesis describes my experimental and theoretical investigation of optical and atomic pattern formation in a sample of cold atoms driven by counterpropagating optical fields. A summary of my unique findings, which I outline throughout this


Figure 1.4: **Off-axis pumping.** Experimental setup and results from Ref. [Greenberg (2012)]. (a) By pumping at an angle \( \sim 10^\circ \) relative to the long axis of the atoms, fields generated by MPO emerge along the long axis. (b) The generated fields can be multimode, where \( \theta_d \approx 3 \text{ mrad} \).

The general progression of this thesis is as follows.

I first provide an overview of light-atom interactions and the nonlinear optical effects that give rise to pattern formation. In Ch. 2, I introduce the concepts in nonlinear optics that are relevant to my system, and I derive the general material susceptibility for a sample of two-level atoms. I then introduce the two main forces that optical fields can impose on atoms: the radiation pressure force and the dipole force. I show that these forces give rise to a sub-Doppler cooling scheme, known as Sisyphus cooling, which is a crucial component of my experiment because it enhances the nonlinear refractive index of my sample. Finally, I show that the dipole force imposed by counterpropagating optical fields gives rise to atomic bunching for sufficiently cold atoms, and I derive the steady-state density distribution for atoms in an optical lattice. In Ch. 3, I provide an overview of transverse optical pattern formation. I discuss the gain mechanism for pattern formation as well as the requirements of phase-matching for efficient generation of the optical fields.

In Ch. 4, I provide my experimental characterization of transverse optical pat-
Table 1.1: **Summary of findings.** This lists the unique contributions I have made, the impact of each one for future research, a summary of previous related work, and the chapter in which I describe each finding.

<table>
<thead>
<tr>
<th>My Contribution</th>
<th>Impact</th>
<th>Previous Work</th>
<th>Chapter</th>
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<tbody>
<tr>
<td>Observe pattern formation in cold atoms (with Joel Greenberg, [Greenberg et al. (2011)]).</td>
<td>First observation of pattern formation in cold atoms, which gives rise to record-low power thresholds and the opportunity to study synergistic effects between the optical fields and real-space atomic bunching.</td>
<td>Previously only studied in homogeneous warm atoms [Dawes et al. (2010)].</td>
<td>Ch. 4</td>
</tr>
<tr>
<td>Observe self-organization of atoms into real-space gratings with both sub- and super-wavelength lattice constants [Schmitberger and Gauthier (2016b)].</td>
<td>First observation of self-organization in a multimode geometry, which allows access to new non-equilibrium physics.</td>
<td>Self-organization has previously only been observed in single-mode geometries [Black et al. (2003); Baumann et al. (2011)].</td>
<td>Ch. 5</td>
</tr>
<tr>
<td>Observe spontaneous 3D Sisyphus cooling [Schmitberger and Gauthier (2016b)].</td>
<td>First observation of spontaneous 3D cooling, which allows for longer coherence times using a simplified, 1D applied beam geometry.</td>
<td>Other researchers have applied optical lattices in 3D to achieve 3D cooling [Raithel et al. (1997b)].</td>
<td>Ch. 5</td>
</tr>
<tr>
<td>Develop self-consistent model for light-atom interactions in 1D optical lattice [Schmitberger and Gauthier (2014)].</td>
<td>Predicts enhanced light-atom interaction strengths achievable by using atomic bunching and small detunings. Unifies finite-temperature nonlinear optics models with zero-temperature optomechanical models.</td>
<td>Previous models are restricted to certain atomic bunching regimes [Muradyan et al. (2005); Deutsch et al. (1995)].</td>
<td>Ch. 6</td>
</tr>
<tr>
<td>Perform theoretical stability analysis for pattern-forming optical fields with multi-level atoms [Schmitberger and Gauthier (2016a)].</td>
<td>First stability analysis for pattern formation in cold, tightly-bunched atoms. Shows how bunching-induced nonlinearity gives rise to reduced intensity thresholds.</td>
<td>Previous analyses are for warm atoms [Firth and Paré (1988)] or weakly bunched atoms [Muradyan et al. (2005)].</td>
<td>Ch. 7</td>
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tern formation in cold atoms and an analysis of the threshold for observing pattern formation. I show that the atoms are tightly bunched in the applied optical lattice during pattern formation. By reaching this tight-bunching regime and allowing the atoms to interact strongly with the lattice-forming optical fields, I observe pattern formation at record-low powers. I describe my measurements of the threshold conditions for pattern formation and my observation of continuous symmetry-breaking, which is a hallmark of multimode non-equilibrium phenomena.

In Ch. 5, I describe my experimental investigation of the atomic patterns. I use parametric resonance and Bragg scattering techniques to measure the spatial and motional properties of the self-organized atomic gratings. I show that the synergistic interaction between the optical patterns and the atoms generates self-organization on both sub- and super-wavelength scales, which represents the first direct observation of multimode atomic self-organization in real space. I extract both an effective dipole potential well depth of the self-generated gratings as well as an atomic temperature of atoms in those gratings. I conclude that, despite only applying optical fields in 1D, the 3D geometry of the optical patterns facilitates spontaneous 3D Sisyphus cooling, which represents the first observation of spontaneous cooling. I show that 3D Sisyphus cooling aids in atomic self-organization and longer coherence times for pattern formation.

To describe my observations theoretically, I begin with a simplified model to describe the refractive index of two-level atoms at thermal equilibrium in a 1D optical lattice. Previous models for atoms in optical lattices either assume the interaction between the atoms and the lattice-forming optical fields is weak [Asbóth et al. (2005); Petrosyan (2007); Wu et al. (2008); Nunn et al. (2010); Schilke et al. (2012b)] or are restricted to a certain regime of atomic bunching, i.e., only weakly bunched atoms [Muradyan et al. (2005)] or infinitely thin sheets of tightly bunched atoms [Deutsch et al. (1995); Asbóth et al. (2007, 2008)]. In Ch. 6, I derive a model
that allows for strong light-atom coupling strengths and is also valid for atoms that are homogeneous, weakly bunched, and tightly bunched. This model represents a unification between the zero-temperature models commonly used in the optomechanical physics community and the finite-temperature models used in the nonlinear optics community. I show that by using optical fields that are frequency-detuned below the atomic resonant frequency and by using sub-Doppler cooled atoms, the resulting tight atomic bunching gives rise to a more than two orders of magnitude enhancement in the nonlinear refractive index \(c.f.\) a homogeneous gas. This enhancement in the nonlinear refractive index allows for studies of low-light-level nonlinear optics, and it allows me to observe optical pattern formation at ultra-low powers.

In Ch. 7, I extend this model to include multi-level atoms and present a theoretical description of pattern formation. Pattern formation has previously been described theoretically in warm-atom systems [Firth and Paré (1988)] and weakly-bunched cold-atom systems [Muradyan et al. (2005)], but my use of cold atoms requires a new model that accounts for atomic bunching. I describe the modification to the nonlinear refractive index in the presence of the transverse optical patterns as well as the formation of self-organized atomic patterns. Despite the fact that Sisyphus cooling aids in self-organization, I do not explicitly account for cooling in this model. Instead, I use the experimentally-measured temperature of the atoms after they have been cooled. I then perform a stability analysis and derive the threshold condition for the instability that gives rise to pattern formation.

In Ch. 8, I conclude my findings and describe possible future directions for my work.
Since the invention of the ruby laser granted access to the nonlinear response of materials to light [Maiman (1960); Franken et al. (1961)], the field of nonlinear optics has been a driving force in studying fundamental physics and developing optical technologies. Nonlinear optics describes any light-matter interaction in which a material’s response to an incident optical field depends nonlinearly on the optical field strength. The properties of these nonlinear materials are modified by incident optical fields, and they, in turn, can modify the properties of the optical fields passing through them in a synergistic way. Nonlinear light-matter interactions give rise to phenomena that are inaccessible in the linear optical regime, such as optical frequency conversion, wave-mixing, and the generation of optical solitons [Boyd (2008)]. Nonlinear optics enables optical fields to effectively interact with one another by means of interacting with a nonlinear optical material, which is the fundamental principle that facilitates the concepts and experiments presented in this thesis.

In this chapter, I provide an introduction to nonlinear optics and describe theoretically the interaction of optical fields with atoms. This chapter provides the foundation for the theoretical descriptions of light-atom interactions and pattern
formation that I use in the remainder of this thesis. In the first section, I discuss the regime of nonlinear optics in which I work. In the second section, I derive the general material susceptibility of a cloud of two-level atoms upon interaction with light—a parameter that I use in subsequent chapters to describe the strength of light-atom interactions. I then go on to discuss mechanical effects of light on atoms, which are necessary for understanding nonlinear optical effects in cold atoms, which I study. I describe the two main optical forces on atoms: the radiation pressure force and the dipole force. I discuss how they give rise to sub-Doppler cooling schemes, such as Sisyphus cooling, which I use experimentally to enhance the light-atom interaction strength and study pattern formation at low light levels. Finally, I show that atoms under the influence of the dipole force can spatially bunch into real-space density gratings. I show in Ch. 6 that this bunching-induced nonlinearity gives rise to reduced thresholds for pattern formation and highly nonlinear light-atom interactions.

2.1 Nonlinear optical effects

A material is considered nonlinear when an optical field with electric field \( \vec{E} \) induces a polarization \( \vec{P} \) that depends nonlinearly on the electric field strength. The material polarization defines the macroscopic dipole moment per unit volume and is defined generally as

\[
\vec{P} = \epsilon_0 \chi \vec{E},
\]

where \( \epsilon_0 \) is the permittivity of free space and \( \chi \) is the tensor material susceptibility. If \( \chi \) depends on \( |\vec{E}| \), then the polarization is a nonlinear function of the optical field strength, and the material is nonlinear.

The nonlinear optical material on which I focus in this thesis is a sample of rubidium atoms. Atoms possess inversion symmetry, and the polarization of such
where $\chi^{(n)}$ is termed the $n$th-order susceptibility. The second, fourth, and higher-order even nonlinear susceptibilities vanish in materials that possess inversion symmetry because such terms are associated with field terms that depend on the direction of the applied electric field. Materials that lack inversion symmetry, such as optical crystals that have a well-defined optical axis, have nonzero even-order nonlinear susceptibilities.

In order to study nonlinear optical effects, the nonlinear (field-dependent) terms in $\chi^{(n)}$ must be sufficiently large to affect the light-matter interaction non-negligibly. These nonlinear terms can be made large either by enhancing $\chi^{(j)}$ ($j > 1$) or by increasing the electric field strength by using high optical intensities. In many materials, the nonlinear susceptibilities are small, and one requires large optical field strengths in order to study nonlinear optical effects. However, there are ways to enhance $\chi^{(j)}$ ($j > 1$) in certain materials so that one can study nonlinear optics at lower light levels. I provide examples of these methods in Chs. 4 and 6.

The lowest-order nonlinear susceptibility in Eq. 2.2 is $\chi^{(3)}$, which is typically the strongest nonlinearity in a sample of atoms. In many materials, higher-order nonlinear terms are orders of magnitude weaker than $\chi^{(3)}$, and many researchers neglect them. This is an especially good approximation in samples of warm atoms, where nonlinear processes such as four-wave-mixing [Dawes et al. (2005)] or electromagnetically induced transparency (EIT) [Schmidt and A. Imamoğlu (1996)] are inherently defined by the $\chi^{(3)}$ nonlinearity. In some materials, like a gas of tightly-bunched atoms, $\chi^{(5)}$ or higher-order terms are also large and cannot be neglected [Boyd et al. (2009); Greenberg and Gauthier (2012a)]. As I show in Ch. 6, when cold atoms spa-
tially bunch in an optical lattice, the fifth-order nonlinear term in the polarization is comparable to the third-order term and cannot be neglected for an accurate description of the light-atom interaction [Schmittberger and Gauthier (2014)]. In general, any finite nonlinear susceptibility can drive nonlinear optical processes, which modifies the optical response of the material and allow optical fields to interact with one another.

2.1.1 Driving nonlinear optical processes

Optical fields interact with one another via a nonlinear material of polarization $\vec{P}$. This is described mathematically via the wave equation,

$$\nabla^2 \vec{E} - \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{P}}{\partial t^2}, (2.3)$$

where $c$ is the speed of light in vacuum. This equation states that an electric field component $\vec{E}$ can be generated or enhanced by a polarization component $\vec{P}$ that has the same (or nearly the same) spatial and temporal variation. In other words, if the material polarization $\vec{P}$ contains terms that propagate with a wavevector $\vec{k}$ and oscillate with a frequency $\omega$, then an optical field with wavevector $\vec{k}$ and frequency $\omega$ can be generated or enhanced inside the material.

In linear optics and neglecting absorption, the wave equation indicates that a material can only impose a linear phase shift on an incident optical field $\vec{E}$. This phase shift depends on the linear susceptibility defined in the material polarization, and a field will have the same frequency before and after propagating through a linear material.

In nonlinear optics, Eq. 2.3 gives rise to more complicated dynamics due to the nonlinear nature of $\vec{P}$. The light-matter interaction described by $\vec{P}$ can drive a new electric field component $\vec{E}$ that was not necessarily applied to the nonlinear
optical material. Some examples of this include harmonic generation and four-wave-mixing, in which the nonlinear light-atom interaction generates new optical fields that propagate with a different direction or frequency than the optical fields that were sent into the nonlinear optical material [Boyd (2008)]. Mathematical examples of how the wave equation describes such nonlinear optical processes are given in Chs. 3 and 7.

In general, Eq. 2.3 provides an intuitive understanding of the synergistic nature of nonlinear optical processes. An incident optical field generates a polarization \( \vec{P} \), which modifies the susceptibility \( \chi \). This new polarization can, in turn, create or enhance the optical field’s properties defined by \( \vec{E} \). In this thesis, I use the material susceptibility \( \chi \) as a measure of the strength of the light-atom interaction that I study. The nonlinear terms in \( \chi \) mediate the synergistic interaction between optical fields and the nonlinear optical material.

2.2 Deriving the material susceptibility for a two-level atom

In order to understand the nonlinear nature of \( \chi \) for a sample of atoms, we must understand its definition and physical origin. I begin with a simple Hamiltonian that describes the interaction of a two-level atom with an optical field. I then calculate the density matrix equations and, subsequently, the material polarization and susceptibility. This section is based largely on the description of a two-level atom in Ch. 6 of Ref. [Boyd (2008)]. This section provides a basis for the theoretical work that I present later in this thesis as well as an intuitive framework for understanding the light-atom interactions that I study. In Ch. 7, I extend this treatment to describe the polarization for a multi-level atom.
2.2.1 Optical field interacting with a two-level atom

I denote the atomic ground state by $g$ and the excited state by $e$, as shown in Fig. 2.1. The Hamiltonian for this system is

$$\hat{H} = \hat{H}_0 + \hat{V}(t),$$

where

$$\hat{H}_0 = \begin{pmatrix} E_g & 0 \\ 0 & E_e \end{pmatrix}$$

is the atomic Hamiltonian, with $E_g$ and $E_e$ the energies of the ground and excited states, respectively. The “interaction Hamiltonian”

$$\hat{V}(t) = -\vec{\mu} \cdot \vec{E}(t)$$

defines the energy of the interaction between an atom of dipole moment $\vec{\mu}$ and an applied electric field $\vec{E}(t)$. The dipole moment couples the ground state $|g\rangle$ and the excited state $|e\rangle$ according to $\vec{\mu} = -q \langle e|\vec{r}|g\rangle$, where the elementary charge is $q$. I assume that the atomic state wavefunctions have definite parity so that $\mu_{gg} = \mu_{ee} = 0$. Therefore, the interaction term reduces to

$$\hat{V}(t) = \begin{pmatrix} 0 & -\vec{\mu}_{ge} \cdot \vec{E}(t) \\ -\vec{\mu}_{eg} \cdot \vec{E}(t) & 0 \end{pmatrix}.$$
The density matrix for the state of this system is defined by

\[ \hat{\rho} = \begin{pmatrix} \rho_{gg} & \rho_{ge} \\ \rho_{eg} & \rho_{ee} \end{pmatrix}, \quad (2.8) \]

where \( \rho_{ge} = \rho_{eg}^* \). The time evolution of the density matrix is described by

\[ \dot{\rho}_{eg} = -\frac{i}{\hbar} \sum_\nu (H_{\nu e} \rho_{\nu g} - \rho_{\nu e} H_{\nu g}). \quad (2.9) \]

I define \( \omega_{eg} = (E_e - E_g)/\hbar \), so that Eq. 2.9 becomes

\[ \dot{\rho}_{eg} = -i \omega_{eg} \rho_{eg} - \frac{i}{\hbar} \sum_\nu (V_{\nu e} \rho_{\nu g} - \rho_{\nu e} V_{\nu g}). \quad (2.10) \]

One can verify from this equation that \( \dot{\rho}_{gg} + \dot{\rho}_{ee} = 0 \), which shows that the total population is conserved. It also follows that \( \rho_{gg} + \rho_{ee} = 1 \), so that the probabilities of the total state occupations sum to unity.

2.2.2 Including relaxation processes

In order to account for relaxation processes, Eq. 2.10 must be modified. Consider that level \( e \) decays to level \( g \) at a rate \( 1/T_1 \). Also assume that the characteristic linewidth of the transition is \( \Gamma_{eg} = 1/T_2 \). Equation 2.10 can then be expanded into the following set of equations:

\[ \dot{\rho}_{eg} = - \left( i \omega_{eg} + \frac{1}{T_2} \right) \rho_{eg} + \frac{i}{\hbar} V_{eg} (\rho_{ee} - \rho_{gg}), \quad (2.11) \]

\[ \dot{\rho}_{ee} = -\rho_{ee} \frac{T_1}{T_2} - \frac{i}{\hbar} (V_{eg} \rho_{ge} - \rho_{eg} V_{ge}), \quad (2.12) \]

and

\[ \dot{\rho}_{gg} = \rho_{ee} \frac{T_1}{T_2} + \frac{i}{\hbar} (V_{eg} \rho_{ge} - \rho_{eg} V_{ge}). \quad (2.13) \]
It follows that

\[
\frac{d}{dt} (\rho_{ee} - \rho_{gg}) = -(\rho_{ee} - \rho_{gg}) - (\rho_{ee} - \rho_{gg})^{(eq)} \frac{2i}{\hbar} \frac{1}{T_1} (V_{eg} \rho_{ge} - \rho_{eg} V_{ge}),
\]

where \((\rho_{ee} - \rho_{gg})^{(eq)}\) is the population difference in thermal equilibrium.

If we assume \(\mu_\parallel E(t)\), Eq. 2.6 becomes

\[
V_{eg} = -\mu_{eg} (E e^{-i\omega t} + E^* e^{i\omega t}).
\]

It is very difficult to solve Eqs. 2.11 and 2.14 with this form of \(V_{eg}\), but the math becomes much easier under the rotating wave approximation. This approximation says that if \(\rho_{eg}\) oscillates at \(e^{-i\omega t}\), then the \(e^{i\omega t}\) term does not drive \(\rho_{eg}\) efficiently. Under the rotating wave approximation,

\[
V_{eg} \approx -\mu_{eg} E e^{-i\omega t}.
\]

Then,

\[
\dot{\rho}_{eg} = -\left( i\omega_{eg} + \frac{1}{T_2} \right) \rho_{eg} - \frac{i}{\hbar} \mu_{eg} E e^{-i\omega t} (\rho_{ee} - \rho_{gg})
\]

and

\[
\frac{d}{dt} (\rho_{ee} - \rho_{gg}) = -(\rho_{ee} - \rho_{gg}) - (\rho_{ee} - \rho_{gg})^{(eq)} \frac{2i}{\hbar} \frac{1}{T_1} \left( -\mu_{eg} E e^{-i\omega t} \rho_{ge} + \rho_{eg} \mu_{ge} E^* e^{i\omega t} \right).
\]

The steady-state solutions of Eqs. 2.17 and 2.18 are obtained by setting the left-hand-sides equal to zero, so that

\[
\rho_{ee} - \rho_{gg} = \frac{(\rho_{ee} - \rho_{gg})^{(eq)} [1 + (\omega - \omega_{eg})^2 T_2^2]}{1 + (\omega - \omega_{eg})^2 T_2^2 + (4/\hbar^2)|\mu_{eg}|^2 |E|^2}
\]

and

\[
\rho_{eg} = \frac{\mu_{eg} E e^{-i\omega t} (\rho_{ee} - \rho_{gg})}{\hbar (\omega - \omega_{eg} + i/T_2)}.
\]
2.2.3 Defining the material polarization

The polarization is defined as the density times the off-diagonal elements of the density matrix:

\[ \vec{P}(t) = \eta \langle \vec{\mu} \rangle = \eta (\vec{\mu}_{ge}\rho_{eg} + \vec{\mu}_{eg}\rho_{eg}), \] (2.21)

where \( \eta \) is the density. This can be written as

\[ \vec{P}(t) = \vec{P} e^{-i\omega t} + \text{c.c.} \] (2.22)

From Eqs. 2.1 and 2.20, it can be shown that

\[ \chi = \frac{\eta |\mu_{eg}|^2(\rho_{ee} - \rho_{gg})}{\epsilon_0 \hbar (\Delta + i/T_2)}, \] (2.23)

where the detuning \( \Delta = \omega - \omega_{eg} \). Note that this is now a scalar quantity where the tensor product in Eq. 2.1 is simplified in Eq. 2.21 for a two-level atom interacting with a monochromatic field. Using Eq. 2.19, this becomes

\[ \chi = \left[ \frac{\eta |\mu_{eg}|^2(\rho_{ee} - \rho_{gg})^{(eq)} T_2}{\epsilon \hbar} \right] \frac{\Delta T_2 - i}{1 + \Delta^2 T_2^2 + |\Omega|^2 T_1 T_2}, \] (2.24)

where \( |\Omega|^2 = 4|\mu_{eg}|^2|E|^2/\hbar^2 \) is the square of the Rabi frequency \( \Omega \). In the low-intensity, steady-state regime, it is a good approximation to take all the population to be in the ground state, so that \( (\rho_{ee} - \rho_{gg})^{(eq)} = -1 \), and

\[ \chi = -\frac{2|\mu_{eg}|^2}{\epsilon \hbar \Gamma} \eta \frac{2\Delta/\Gamma - i}{1 + 4\Delta^2/\Gamma^2} \frac{1}{1 + \tilde{I}}, \] (2.25)

where I have neglected collisional dephasing and assumed \( T_1 = T_2/2 \). Here, \( \tilde{I} = \Omega^2/[2(\Delta^2 + \Gamma^2/4)] \) is the intensity normalized by the off-resonant saturation intensity \( I_{s\Delta} = \epsilon_0 c \hbar^2/[2|\mu_{eg}|^2 T_1 T_2 (1 + \Delta^2/T_1 T_2)] \), and I’ve simplified the natural linewidth \( \Gamma = \Gamma_{eg} \), where

\[ \Gamma = \frac{\omega_{eg}^3 |\mu_{eg}|^2}{3\pi \epsilon_0 c \hbar^3}. \] (2.26)
I define $k_{eg} = \omega_{eg}/c$, and the susceptibility becomes

$$
\chi = -\frac{6\pi}{k_{eg}^3} \eta \frac{2\Delta/\Gamma - i}{1 + 4\Delta^2/\Gamma^2} \frac{1}{1 + \Gamma}.
$$

Equation 2.27 provides the basis of much of the theoretical work presented in the remainder of this thesis. In this simplified case, the susceptibility is related to the index of refraction via $n = \sqrt{1 + \chi}$. The susceptibility describes how optical fields couple to both the internal (spin) states of the atom (via $\mu_{eg}$) as well as the external (motional) states of the atoms (via $\eta$). In a sample of warm atoms, the density distribution is homogeneous, and $\eta$ is simply equal to the average atomic density. For a sample of cold atoms in a spatially varying intensity distribution (i.e., an optical lattice), the density distribution acquires a spatial dependence when the atoms become trapped in the lattice. This density distribution is defined explicitly in Ch. 6. However, in order to understand how the optical fields couple to the external degrees of freedom of an atom, it is first necessary to understand the types of forces that optical fields can impose on atoms.

### 2.3 Optical forces on atoms

Optical fields impose a force on the center-of-mass motion of atoms, which can in turn give rise to cooling or heating. The manipulation of the center-of-mass motion combined with cooling techniques play a crucial role in the physics that I present in this thesis. Namely, in Ch. 6, I show that the ability to move, trap, and cool atoms using optical fields results in an enhanced nonlinear susceptibility, which allows us to realize nonlinear optical effects at low light levels. In this section, I derive the basic light forces on a sample of two-level atoms due to the presence of either a single optical field or the presence of counterpropagating optical fields (i.e., an optical lattice). I show that when this formalism is extended to a multi-level atom descrip-
tion, the application of an optical lattice gives rise to atomic cooling under certain experimental conditions. The work in this section is based largely on Ref. [Metcalf and van der Straten (1999)].

2.3.1 Setting up the problem: Light shifts

If we first consider a plane wave of wavevector $\vec{k} = k\hat{z}$ and electric field amplitude $E_0$, the interaction Hamiltonian of Eq. 2.6 becomes

$$\hat{V} = \hbar \Omega \cos(\vec{k}z - \omega t),$$

where

$$\Omega = \frac{\vec{\mu} \cdot \vec{E}_0}{\hbar}$$

is the Rabi frequency. The probability of finding an atom in the states $|g\rangle$ or $|e\rangle$ oscillates in time at a frequency $\sqrt{\Omega^2 + \Delta^2}$, which depends on the light-atom coupling strength [Metcalf and van der Straten (1999)]. If the optical field is exactly tuned to the atomic resonance ($\Delta = 0$), the atomic population oscillates exactly at the Rabi frequency. Finite detunings result in a weaker light-atom coupling and give rise to faster population oscillations but also a reduced probability of excitation.

The presence of the optical field distorts the electron cloud surrounding the atom and results in a shifting of the atomic energy levels, known as a “light shift.” The magnitude and direction of this light shift depend on the detuning and the Rabi frequency, where the ground and excited state energy levels are shifted by an amount

$$\Delta E_{e,g} = \frac{\hbar}{2} \left( -\Delta \mp \sqrt{\Omega^2 + \Delta^2} \right).$$

For red detunings ($\Delta < 0$), the energy levels are shifted apart, as depicted in Fig. 2.2. For blue detunings, the ground and excited states move closer together. This detuning dependence has implications for sub-Doppler cooling schemes, such as Sisyphus cooling, which is discussed in subsection 2.3.3.
Figure 2.2: **Light-shifted energy states.** When an optical field of frequency $\omega$ and detuning $\Delta = \omega - (\omega_e - \omega_g)$ is incident on an atom, the ground and excited state energies shift by $\Delta E_{e,g}$.

### 2.3.2 Deriving the force

In addition to giving rise to energy shifts in the internal states of an atom, optical fields also exert forces on the external degrees of freedom of atoms. These external forces depend on the interaction Hamiltonian from Eq. 2.6 according to

$$F_z = -\left\langle \frac{\partial \hat{V}}{\partial z} \right\rangle$$

for applied field(s) along $\hat{z}$. Using Eq. 2.29 and the property $\left\langle \hat{A} \right\rangle = \text{Tr}(\hat{\rho} \hat{A})$ with $\hat{\rho}$ defined in Eq. 2.8, this becomes

$$F_z = \hbar \left( \frac{\partial \Omega}{\partial z} \rho_{eg}^* + \frac{\partial \Omega^*}{\partial z} \rho_{eg} \right).$$

This expression is simplified using Eq. 2.20, where I again take the low-intensity approximation and assume all the population is in the ground state so that $(\rho_{ee} - \rho_{gg})^{eq} = -1$.

It is useful to analyze the result of this simplification under two cases: a single optical field and counterpropagating optical fields. Each of these cases gives rise
to one of the two primary forces that optical fields impose on atoms: the radiation pressure force and the dipole force.

The radiation pressure force

In the case of a single optical field incident on a two-level atom at rest, where the electric field goes as $\vec{E}(z, t) = E_0(z)e^{-i\omega t} + \text{c.c.}$ with $E_0(z) = E_0e^{ikz}$, Eq. 2.32 simplifies to

$$\vec{F}_{\text{rad}} = \frac{\hbar k(\Gamma/2)(I/I_{\text{sat}})}{1 + I/I_{\text{sat}} + 4\Delta^2/\Gamma^2} \hat{z},$$

(2.33)

where $I_{\text{sat}} = \frac{\hbar^2\Gamma^2\epsilon_0}{2|\mu|^2}$ is the resonant saturation intensity. This force is known as the radiation pressure force, and it is the total force on an atom due to a scattering event (i.e., absorption from the optical field followed by spontaneous emission). The direction of this force is parallel to the wavevector of the optical field because of momentum conservation in the absorption event, as shown in Fig. 2.3. Spontaneous emission can occur in all directions, and thus the average force due to spontaneous emission after many scattering events is approximately zero. At high intensities (large $s_0$), this force saturates to a maximum value of $\hbar k\Gamma/2$.

The radiation pressure force is dissipative because energy is removed from the system via spontaneous emission. This force is a key component to cooling atoms, and it is discussed further in Sec. 2.3.3.

The dipole force

In contrast to the dissipative radiation pressure force, another type of force that optical fields can impose on atoms is the dipole force, which is conservative. This force arises due to the spatially varying light shifts in the atomic energy states, shown in Fig. 2.2. With an example electric field amplitude $E_0(z) = E_0\cos(kz)$, Eq. 2.32
Figure 2.3: Radiation pressure force. When an atom absorbs a photon of wavevector $\vec{k}$, it gets a momentum kick in the direction of the incoming photon, resulting in an average momentum transfer of $\Delta \vec{p}_{\text{in}} = \hbar \vec{k}$. Subsequent spontaneous emission can occur in any direction, with an average momentum transfer over many emission events of $\Delta \vec{p}_{\text{out}} \to 0$. Thus, the total average momentum transfer to an atom per scattering event is $\Delta \vec{p} = \hbar \vec{k}$, which gives rise to the radiation pressure force.

reduces to

$$\vec{F}_{\text{dip}} = \frac{\hbar k \Delta (I/I_{\text{sat}}) \sin(2kz)}{1 + 2(I/I_{\text{sat}}) \cos^2(kz) + 4\Delta^2/\Gamma^2} \hat{z},$$

(2.34)

where $I = 2\varepsilon_0 c |E_0(z)|^2$ is the total intensity and $I_{\text{sat}} = \varepsilon_0 c \hbar^2 / (2|\mu_{eg}|^2 T_1 T_2)$ is the resonant saturation intensity. This force is conservative and related to the dipole potential via $\vec{F}_{\text{dip}} = -\nabla U_{\text{dip}}$, where

$$U_{\text{dip}} = \frac{\hbar \Delta}{2} \log \left[ 1 + \frac{2(I/I_{\text{sat}}) \cos^2(kz)}{1 + 4\Delta^2/\Gamma^2} \right].$$

(2.35)

Atoms are forced to minimize their energy in the presence of the dipole potential, and thus atoms are attracted to the intensity maxima of a red-detuned ($\Delta < 0$) optical lattice and the intensity minima of a blue-detuned ($\Delta > 0$) optical lattice. For atoms with a thermal energy $k_B T \ll U_{\text{dip}}$, with $k_B$ Boltzmann’s constant and $T$ the atomic temperature, the atoms spatially bunch into the dipole potential minima, which gives rise to a spatially varying density distribution. This spatial bunching is
the primary mechanism that gives rise to the enhanced light-atom interactions that I present in this thesis, and I provide more explicit definitions for the dipole potential in Chs. 4, 6 and 7. In the regime where the off-resonant saturation parameter $\tilde{I} = I/I_{sat}(1 + 4\Delta^2/\Gamma^2) \ll 1$, the dipole potential reduces to the light shift $\Delta E_g$ in Eq. 2.30. In the special case of equal-intensity counterpropagating optical fields, the radiation pressure force is absent because the probability of absorbing light from each optical field is equal, and the dipole force is the only optical force on the atoms.

In addition, while the dipole force is conservative and cannot directly give rise to atomic cooling, there are sub-Doppler cooling schemes that involve the dipole force. One of these cooling schemes, known as Sisyphus cooling, combines the effects of the dipole force and the dissipative radiation pressure force. This cooling mechanism gives rise to enhanced bunching and, subsequently, enhanced light-atom interaction strengths.

### 2.3.3 Sisyphus cooling

Sisyphus cooling is one example of a scheme that is used to cool atoms below the Doppler limit. Doppler cooling cools atoms to the so-called “Doppler temperature,” which is 146 $\mu$K for rubidium [Metcalf and van der Straten (1999)]. While this is already many orders of magnitude colder than atoms in a room-temperature vapor, I show in Ch. 6 that sub-Doppler temperatures give rise to enhanced light-atom interaction strengths. Therefore, many researchers use sub-Doppler cooling schemes like Sisyphus cooling in order to achieve sub-Doppler or “ultracold” temperatures.

The effects that I have described thus far assume a two-level atomic excitation scheme, but to understand Sisyphus cooling, it is necessary to describe a multi-level atom. The purpose of this subsection is simply to provide an intuitive understanding of the Sisyphus mechanism and how it gives rise to cooling. Thus, here I simply provide the equations that result from this multi-level atom formalism; I provide
**Figure 2.4: Sisyphus cooling.** (a) The excitation scheme for a $J = 1/2 \rightarrow J' = 3/2$ atomic transition, shown with the circular polarizations that excite the stretched state transitions. The numbers denote the $m_J$ sublevels. (b) Pictoral representation of Sisyphus cooling. Above the graph, there is a depiction of counterpropagating optical fields in a $\text{lin} \perp \text{lin}$ polarization configuration, which gives rise to a spatially varying electric field polarization, represented by the red arrows and circles. The graph depicts the spatially varying AC Stark shifts undergone by the $m_J = \pm 1/2$ ground states with the applied optical fields. The gray circles represent an atom moving through space. The red arrows represent absorption events that are most probable when an atom reaches a region with pure $\hat{\sigma}^\pm$-polarization. The black arrows indicate spontaneous emission where the atom decays into a different ground state.

A more explicit derivation of how the multi-level description modifies the effective susceptibility in Ch. 7.

I consider the energy scheme depicted in Fig. 2.4(a), which shows the fine structure for a $J = 1/2 \rightarrow J' = 3/2$ atomic transition. While we cannot realistically ignore the hyperfine structure, the fine structure excitation scheme provides a simplified picture for understanding Sisyphus cooling. In addition, using a fine-structure model has been shown to be a good approximation for describing light-atom interactions in schemes where stretched-state transitions are the dominant excitation [Greenberg et al. (2011)], as they are in my experiment. This is discussed further in Chs. 5 and 7.

In Fig. 2.4(a), the red arrows represent possible excitations due to light that is
polarized with a combination of $\hat{\sigma}^+$ and $\hat{\sigma}^-$ polarizations. The black arrows represent spontaneous emission events where the atom decays into a different ground state.

For counterpropagating optical fields that are linearly polarized orthogonal to one another—the “lin⊥lin” polarization configuration—the total electric field goes as

$$\overrightarrow{E} = F(z)e^{i(kz-\omega t)}\hat{x} + e^{i\phi} B(z)e^{i(-kz-\omega t)}\hat{y},$$

(2.36)

where $\phi$ is some relative phase between the fields. We define circular polarization unit vectors as

$$\hat{\sigma}^+ = -\frac{\hat{x} + i\hat{y}}{\sqrt{2}} \quad \text{and} \quad \hat{\sigma}^- = \frac{\hat{x} - i\hat{y}}{\sqrt{2}},$$

(2.37)

so that

$$\hat{x} = \frac{-\hat{\sigma}^+ + \hat{\sigma}^-}{\sqrt{2}} \quad \text{and} \quad \hat{y} = \frac{i\hat{\sigma}^+ + \hat{\sigma}^-}{\sqrt{2}}.$$

(2.38)

Then the electric field can be rewritten as

$$\overrightarrow{E} = \left\{ \left[ -F(z)e^{ikz} + B(z)e^{-ikz} \right] \frac{\hat{\sigma}^+}{\sqrt{2}} + \left[ F(z)e^{ikz} + B(z)e^{-ikz} \right] \frac{\hat{\sigma}^-}{\sqrt{2}} \right\} e^{-i\omega t},$$

(2.39)

where I have taken $\phi = -\pi/2$ so that the electric field is $\hat{\sigma}^-$-polarized at $z = 0$. For equal intensity fields ($|F(z)|^2 = |B(z)|^2$), this becomes

$$\overrightarrow{E} = \left\{ -i\sqrt{2} E_0 \sin(k'z)\hat{\sigma}_+ + \sqrt{2} E_0 \cos(k'z)\hat{\sigma}_- \right\} e^{-i\omega t},$$

(2.40)

where I’ve taken $F(z) = E_0 e^{i\delta z}$, $B(z) = E_0 e^{-i\delta z}$, and $k' = k + \delta$ is the wavenumber of the optical fields inside the atoms, where $\delta$ is a function of the effective index of refraction. One can see that the electric field polarization is periodic in $k'z$ for this polarization configuration. At $z = 0$, the light is $\hat{\sigma}^-$-polarized. At $z = \lambda'/4$, with $k' = 2\pi/\lambda'$, the light is $\hat{\sigma}^+$-polarized. Halfway between these regions of pure circular polarizations, the light is purely linearly polarized. This periodic modulation in the polarization is depicted at the top of Fig. 2.4(b).
Because the polarization of the electric field is spatially varying, the Rabi frequency for a given $g \rightarrow e$ transition, $\Omega_{eg} = \vec{\mu}_{eg} \cdot \vec{E}/\hbar$, is also spatially varying due to the dot product between the atomic dipole and the electric field. As a result, the light shifts $\Delta E_g$ of Eq. 2.30 are also spatially varying due to the dependence on $\Omega$. In the low-intensity limit ($\tilde{I} \ll 1$), these light shifts that give rise to the dipole force are approximately

$$\Delta E_g \approx \hbar \Delta \tilde{I} C_{ge}^2,$$

(2.41)

where $C_{ge}$ is the Clebsch-Gordon coefficient for a given $g \rightarrow e$ transition. For the $J = 1/2 \rightarrow J' = 3/2$ transition, the stretched-state transitions ($m_J = \pm 1/2 \rightarrow m_{J'} = \pm 3/2$) have $C_{\pm 1/2, \pm 3/2} = 1$, and the $m_J = \pm 1/2 \rightarrow m_{J'} = \mp 1/2$ transitions have $C_{\pm 1/2, \pm 1/2} = \sqrt{1/3}$. Recalling $\vec{E}$ defined in Eq. 2.40, it is apparent that the light shifts of both $m_J = \pm 1/2$ ground states are spatially varying. The phase of the spatial variation for each ground state is also separated by $\pi$, as depicted in Fig. 2.4(b).

The dipole force induced by these counterpropagating optical fields thus gives rise to two superimposed dipole potentials: $U(z) = U_0^+ \sin^2(k'z) + U_0^- \cos^2(k'z)$, where $U_0^\pm$ correspond to the dipole potentials for the $m_J = \pm 1/2$ ground states. For an atom moving through this dipole potential well landscape with polarization-dependent light shifts, the atom can undergo optical pumping processes, described by the radiation pressure force in Eq. 2.33, which ultimately cause the atom to lose (or gain) kinetic energy. When the optical fields are red-detuned (blue-detuned), the atom undergoes cooling (heating) as a result of optical pumping into ground states of different energies [Castin et al. (1991)]. Since I utilize these methods for cooling, I restrict my discussion to that of red-detuned optical fields.

In order to understand the dissipative nature of these optical pumping processes, it is useful to visually follow an atom (depicted by the gray circle) in Fig. 2.4(b). At $z = 0$, I take the atom to be in the $m_J = -1/2$ ground state. As it moves
through space (along $\hat{z}$ in this example), the local electric field polarization changes. Eventually, it reaches a location (at $\lambda'/4$) where the total electric field is $\hat{\sigma}^+$-polarized. Here, the atom can be excited into the $m_J = +1/2$ excited state, following the polarizations defined in the energy scheme of Fig. 2.4(a). From this excited state, the atom can decay back to either ground state. If the atom decays to the $m_J = +1/2$ ground state via spontaneous emission, then the atom “falls” into a different energy landscape, as depicted by the black arrow in Fig. 2.4(b). At $z = \lambda'/4$, the $m_J = +1/2$ ground state has a slightly lower energy than the $m_J = -1/2$ ground state from which the atom was optically pumped. As a result, the atom loses energy by undergoing this absorption-emission event.

This cooling scheme is named for the Greek mythological figure Sisyphus, who was forced to endlessly push a boulder up a hill. In this cooling scheme, a moving atom is constantly climbing up the sides of potential wells, only to be optically pumped into a different ground state and forced to climb a new potential. After many ($\sim 10^3$) optical pumping events, an atom can be cooled quite effectively.

In my experiment, the atoms are initially cooled to $30 - 40 \, \mu\text{K}$ in a MOT [Greenberg et al. (2007)]. I then apply red-detuned, lin,lin counterpropagating optical fields to the atoms along $\pm \hat{z}$, so that they undergo Sisyphus cooling to a final temperature along $\hat{z}$ of $T_z = 2 - 3 \, \mu\text{K}$. This final temperature is about an order of magnitude larger than the recoil temperature for rubidium (391 nK)—the energy associated with a recoil kick imparted to the atom by scattering a photon—which is the expected minimum temperature achievable via Sisyphus cooling [Dalibard and Cohen-Tannoudji (1989)].

It is also important to note that Sisyphus cooling is maximally effective when the atom traverses a distance of $\lambda'/4$ during one optical pumping time ($\tau_p = 2(\Gamma/2\pi)s_0/9$), i.e., when the initial atomic velocity is $v_c = \lambda'/4\tau_p$. For $\tilde{I} = 0.5$, this critical velocity corresponds to a temperature of $\sim 60 \, \mu\text{K}$, which is only a factor of $1/2 - 2/3$ from our
initial temperature. For much higher initial temperatures, the atoms are moving too quickly to undergo efficient optical pumping, and the dominant cooling mechanism is Doppler cooling [Dalibard and Cohen-Tannoudji (1989)]. Similarly, for much higher optical field intensities, the optical pumping time is too fast, and the critical velocity for Sisyphus cooling is much lower than one can achieve in a MOT. Thus, we are able to use Sisyphus cooling effectively by using low optical field intensities so that the initial temperatures in our MOT correspond closely to the critical velocity where Sisyphus cooling is efficient.

2.4 Density distribution in a uniform optical lattice

Sisyphus cooling helps the atoms become trapped in an optical lattice when the dipole potential well depth overcomes the thermal energy of the atoms. When the atoms become spatially trapped, the density $\eta$ in Eq. 2.27 becomes a spatially-dependent function. In order to derive the density distribution for bunched atoms in the presence of a dipole potential, I assume that the atoms are initially Brownian particles, and I solve the Fokker-Planck equation for such particles in an external potential. In this section, I make the assumption that the atoms are in thermal equilibrium. While this is not the case in my experiment due to Sisyphus cooling, it is a good approximation after most of the atoms have undergone cooling [Greenberg and Gauthier (2012a)]. I discuss this approximation further in Ch. 7. The following section is based largely on Refs. [Risken (1984); Cross (2013); Orlandini (2013)].

2.4.1 The Fokker-Planck Equation

One treatment of Brownian motion is given by the Fokker-Planck equation. The complete solution of a system of Brownian atoms would involve solving the equations of motion for each atom. Because it is (generally) very difficult to do this, it is common practice to instead describe the system using stochastic macroscopic variables [Risken
The Fokker-Planck equation is an equation of motion for the distribution function of the stochastic fluctuations of the macroscopic variables.

The general Fokker-Planck equation in one spatial dimension (along \( \hat{x} \)) has the form

\[
\frac{\partial p}{\partial t} = \left( -\frac{\partial}{\partial x} D^{(1)}(x) + \frac{\partial^2}{\partial x^2} D^{(2)}(x) \right) p,
\]

where \( D^{(1)} \) is the drift coefficient and \( D^{(2)} > 0 \) is the diffusion coefficient. It is useful to rewrite the Fokker-Planck equation as a continuity equation for the probability distribution \( p(x, t) \), which takes the form

\[
\frac{\partial p(x, t)}{\partial t} = \hat{L}_{FPP}(x, t),
\]

where

\[
\hat{L}_{FP} = -\frac{\partial}{\partial x} D^{(1)}(x, t) + \frac{\partial^2}{\partial x^2} D^{(2)}(x, t).
\]

I introduce the probability current

\[
j(x, t) = \left[ D^{(1)}(x, t) - \frac{\partial}{\partial x} D^{(2)}(x, t) \right] p(x, t),
\]

so that Eq. 2.43 becomes

\[
\frac{\partial p(x, t)}{\partial t} = -\frac{\partial}{\partial x} j(x, t).
\]

**Stationary Solutions of the Fokker-Planck equation**

The long-time-limit solution (i.e. the “stationary” solution) of Eq. 2.46 is defined as

\[
\frac{\partial}{\partial t} p_s(x, t) = 0.
\]

For homogeneous diffusive processes, \( D^{(1)}(x, t) \) and \( D^{(2)}(x, t) \) do not explicitly depend on time. In this case, the stationary solution of the Fokker-Planck equation also
implies (with $d j_s(x)/dx = 0$)
\[
\frac{d}{dx} \left[ D^{(1)}(x)p_s(x) \right] - \frac{\sigma^2}{\partial x^2} \left[ D^{(2)}(x)p_s(x) \right] = 0. \tag{2.48}
\]

If we assume that flux is conserved in a region of interest, which is a good approximation in a standard MOT with many atoms, then $j_s(x) = 0$ in that region, and
\[
D^{(1)}(x)p_s(x) = \frac{\partial}{\partial x} \left[ D^{(2)}(x)p_s(x) \right]. \tag{2.49}
\]

Letting $g(x) = D^{(2)}(x)p_s(x)$ and $A(x) = D^{(1)}(x)/D^{(2)}(x)$, this equation becomes
\[
\frac{\partial}{\partial x} g(x) = A(x)g(x), \tag{2.50}
\]
which has the solution $g(x) = \exp \left[ \int A(x)dx \right]$. Therefore, enforcing normalization with $N_0$,
\[
p_s(x) = \frac{N_0}{D^{(2)}(x)} \exp \left[ \int_a^x \frac{D^{(1)}(x')}{D^{(2)}(x')} dx' \right]. \tag{2.51}
\]

In steady-state, the drift and diffusion coefficients go as [Orlandini (2013)]
\[
D^{(1)}(x) = \frac{F(x)}{m\gamma} \tag{2.52}
\]
and
\[
D^{(2)}(x) = \frac{\sigma^2}{2\gamma^2}, \tag{2.53}
\]
where $\gamma$ is the damping coefficient and $\sigma$ is defined via $\langle F(t_1)F(t_2) \rangle = \sigma^2 \delta(t_1 - t_2)$. Therefore, with a force that is related to a scalar potential as $\vec{F}(\vec{r}) = -\nabla U(\vec{r})$, this becomes
\[
p_s(x) = \frac{2\gamma^2 N_0}{\sigma^2} \exp \left[ -\frac{2\gamma}{m\sigma^2} (U(x) - U(a)) \right]. \tag{2.54}
\]
In the steady-state, equilibrium solution, the fluctuation-dissipation relation $m\sigma^2 = 2k_BT\gamma$ holds [Orlandini (2013)]. Using this relation, the stationary Fokker-Planck solution for the probability density in the presence of a potential is

$$p_s(x) = N\exp\left[-\frac{U(x)}{k_BT}\right],$$  \hspace{1cm} (2.55)

where I have absorbed all constants from Eq. 2.54 into a new normalization constant $N$. In Ch. 6, I show that when $|U(x)| \gg k_BT$ for sub-Doppler-cooled atoms in an optical lattice of dipole potential $U(x)$, the atoms become spatially trapped at the dipole potential minima of the optical lattice.

### 2.5 Summary

In this chapter, I provide an introduction to the terms and concepts in nonlinear optics that I use throughout this thesis. I introduce the material polarization, which is the fundamental parameter that describes light-atom interactions. I then derive the material susceptibility for a two-level atom, which is a measure of the strength of the light-atom interaction and determines the nonlinear optical nature of a material. I also introduce the two basic forces that optical fields impose on atoms: the radiation pressure force and the dipole force. The dipole force gives rise to spatial trapping of atoms in an optical lattice, which is central to the experiments and theory I present in this thesis. I also describe Sisyphus cooling, which arises when one accounts for dissipative radiation in an optical lattice. Finally, I derive the steady-state solution of the Fokker-Planck equation, which describes the probability distribution for atoms at thermal equilibrium in an optical lattice. In Ch. 6, I use this definition of the density distribution to describe how optical fields couple to the external degrees of freedom of a gas of ultracold atoms, and I describe how this gives rise to enhanced light-atom interaction strengths and reduced power thresholds for studying transverse pattern formation.
Introduction to Transverse Optical Pattern Formation

Transverse effects in nonlinear optical systems have been a subject of interest for many years. Early studies of transverse optical pattern formation provided insights into the nonlinear dynamics of warm atoms, four-wave-mixing processes, and transverse instabilities [Silberberg and Bar-Joseph (1982)]. More recently, transverse optical pattern formation in warm atoms was used to create a 600-photon optical switch, which has applications in building low-light-level optical communication devices [Dawes et al. (2005); Dawes (2008)].

Transverse optical pattern formation relies upon a transverse instability that is enhanced via wave-mixing processes. The purpose of this chapter is to introduce wave-mixing and the nonlinear optical effects that give rise to pattern formation. The concepts presented in this chapter can be found in other sources [Boyd (2008); Chiao et al. (1966); Yariv and Pepper (1977); Firth et al. (1990)]. I summarize the concepts here to provide an intuitive understanding of the physics that gives rise to transverse pattern formation. This framework is useful for understanding the experimental and theoretical work on transverse pattern formation presented in the
remainder of this thesis.

In this chapter, I describe four-wave-mixing processes and how they generate new optical fields. I then discuss the concept of phase-matching in four-wave-mixing, which enhances the efficiency of the wave-mixing process. I also describe the phenomenon of weak-wave retardation, which explains why the fields generated in transverse optical pattern formation propagate at an angle relative to the applied fields. Finally, I describe a theoretical example of four-wave-mixing based on Ref. [Yariv and Pepper (1977)], which provides a framework for understanding the conditions under which new optical fields can be generated in a wave-mixing process.

3.1 Introduction to transverse wave-mixing instabilities

Transverse optical patterns are optical fields that emerge spontaneously from a gas of atoms driven with counterpropagating optical fields. In Sec. 1.2, I introduce my experimental setup, wherein I apply counterpropagating optical fields to an elongated cloud of ultracold atoms. Above a threshold nonlinear refractive index, a wave-mixing instability generates new optical fields that have a transverse wavevector component.

To understand the physics of this wave-mixing instability, I simplify the picture and focus on four-wave-mixing, which is a third-order ($\chi^{(3)}$) nonlinear optical process. However, I note that in my experiment, higher-order nonlinear effects can also contribute to transverse optical pattern formation, where $\chi^{(5)}$ nonlinearities give rise to six-wave-mixing, $\chi^{(7)}$ to eight-wave-mixing, etc.

The system I consider in this chapter is depicted in panel 1 of Fig. 3.1, where counterpropagating optical fields are applied to a $\chi^{(3)}$ nonlinear optical material. In this system, there exist transverse perturbations, either from the applied pump fields (e.g., light spontaneously emitted into the transverse plane) or via atomic fluctuations (e.g., atoms moving with a transverse velocity component). In panel 2 of Fig. 3.1, I depict these transverse perturbations as spontaneously emitted fields. Above a
Figure 3.1: **Enhancing transverse perturbations.** 1. Counterpropagating optical fields are applied to a $\chi^{(3)}$ nonlinear optical material. 2. Spontaneous emission events give rise to transverse perturbations in the light-matter interaction. 3. Certain transverse perturbations are enhanced via wave-mixing processes inside the material. 4. For a sufficiently high $\chi^{(3)}$, these wave-mixing processes generate macroscopic optical fields, which we can transverse optical patterns.

threshold nonlinear refractive index, some of these small transverse perturbations can be enhanced by nonlinear optical wave-mixing processes, as shown in panel 3. The particular weak-field perturbations that are enhanced are those that meet a phase-matching condition with the applied pump fields, which conserves momentum in the wave-mixing process, and which I discuss in Sec. 3.2. As the wave-mixing process is seeded by the pump fields, the power scattered into the transverse fields grows exponentially, which generates macroscopic transverse optical patterns, as shown in panel 4 of Fig. 3.1.

### 3.1.1 Wave-mixing processes

Four-wave-mixing describes a phenomenon where three optical fields interact in a nonlinear optical material and generate a fourth optical field. Recall that optical fields effectively “interact” with one another by modifying the nonlinear refractive index of a material, which affects subsequent light-matter interactions.
Figure 3.2: **Frequency-degenerate four-wave-mixing.** (a) The energy scheme for $^{87}\text{Rb}$, where I typically apply an optical field of frequency $\omega$ detuned from the $|g\rangle = 5^2S_{1/2}(F = 2) \rightarrow |e\rangle = 5^2P_{3/2}(F = 3)$ transition by $\Delta = -4$ to $-10\Gamma$. (b) A pictorial representation of the four-wave-mixing scheme that I study, where three optical fields of frequency $\omega$ interact with a nonlinear optical material to generate a fourth, frequency-degenerate optical field.

The four-wave-mixing process that I study is depicted in Fig. 3.2. In Fig. 3.2(a), I show the optical pumping scheme that I apply experimentally, where I pump the atoms with optical fields that are red-detuned from the $|g\rangle = 5^2S_{1/2}(F = 2) \rightarrow |e\rangle = 5^2P_{3/2}(F = 3)$ transition in $^{87}\text{Rb}$. I typically use optical field detunings in the range $\Delta = -4$ to $-10\Gamma$, which corresponds to 24 to 60 MHz below the resonant transition frequency. As discussed further in Ch. 6, the use of such small detunings is useful for enhancing the light-atom interaction strength. I do not typically work closer to the resonant frequency than $\Delta = -4\Gamma$ because the increased absorption gives rise to increased spontaneous emission events that reduce the efficiency of the wave-mixing process. In addition, I always use red-detuned optical fields for two reasons: Sisyphus cooling, which gives rise to enhanced bunching, only works for red detunings, and the wave-mixing process is only phase-matched in our experimental regime for red detunings. The importance of phase-matching is discussed further below and in Ch. 7.
Upon pumping the atoms with counterpropagating optical fields at this red-detuned frequency $\omega$, there is a threshold above which wave-mixing processes occur efficiently and generate the transverse optical patterns. The four-wave-mixing process that occurs in my system is frequency-degenerate four-wave-mixing, which is depicted in Fig. 3.2(b). There are other types of four-wave-mixing schemes that are non-degenerate, and which involve, for example, two-photon transitions to higher energy states or Raman transitions to different ground states. In my system, I have always observed degenerate four-wave-mixing, which I expect simply dominates because it gives rise to balanced radiation pressure in all spatial dimensions and retains more bunched atoms.

To understand how three optical fields “interact” to generate a fourth in this wave-mixing process, it is useful to first consider the interaction of two distinct optical fields with the atoms. Figure 3.3(a) shows the interference pattern (gray) that arises due to an applied pump beam (field 1) and one of the weak fields generated by the transverse perturbation (field 2). This interference pattern gives rise to a spatial modulation in the index of refraction of the atoms. In this beam geometry, the spatially modulated index of refraction is then perfectly phase-matched to scatter the other pump field (field 3) into the direction of another transverse field (field 4). This particular geometry is known as backward four-wave-mixing or phase-conjugation, the latter name derived from the fact that the output field is the phase-conjugate of one of the input fields.

There are two other types of four-wave-mixing that occur in my experiment, the geometries for which are depicted in Figs. 3.3(b) and (c). Figure 3.3(b) shows forward four-wave-mixing, which arises due to the interference of a pump field with a nearly copropagating perturbative field. The induced modulation in the refractive index is nearly phase-matched to scatter subsequent light from the same pump beam into the direction of the fourth field. “Nearly” phase-matched processes are defined
Figure 3.3: **Four-wave-mixing geometries.** (a) Backward four-wave-mixing: A pump field and a nearly counterpropagating perturbative field induce gratings in the material. The counterpropagating pump field is then phase-matched to scatter off these gratings into the direction of the fourth field. (b) Forward four-wave-mixing: A pump field and a nearly copropagating perturbative field induce gratings in the material. The same pump field is nearly phase-matched to scatter off these gratings into the direction of the fourth field. (c) Parametric oscillation: The pump fields induce gratings in the material. A perturbative field is nearly phase-matched to scatter off these gratings into the direction of the fourth field.

below, and they are simply wave-mixing processes that have a slight phase mismatch but still contribute strongly to the enhancement of an optical field. Another nearly phase-matched four-wave-mixing process is depicted in Fig. 3.3(c), where a perturbative field can scatter off the modulated index of refraction formed by the applied counterpropagating pump fields.

These modulations in the refractive index can correspond to one of two types of gratings: polarization gratings or density gratings. Polarization gratings, also known as “spin” gratings, arise due to a spatial variation in either the intensity or the optical field polarization, where the atoms interact more strongly with regions of high intensity or where the field is well-aligned with the dipole moment of the
atom. Polarization gratings make up the spatially dependent part of the saturable nonlinearity, which is the only type of nonlinearity that gives rise to four-wave-mixing in warm atoms. In cold atoms, density gratings can also form when the dipole potential formed by the optical fields exceeds the thermal energy of the atoms. These density gratings correspond to the bunching-induced nonlinearity, which is discussed further in Chs. 4 and 6. In my experiment, the gratings depicted in Fig. 3.3 are primarily density gratings, and the saturable nonlinearity has a negligible effect. (See Ch. 6.)

It is also important to note that Fig. 3.3 depicts only those four-wave-mixing geometries that enhance one of the pattern-forming optical fields. There are analogous processes that happen simultaneously to enhance all of the fields that go on to form the transverse optical patterns.

3.2 Phase-matching

In order for the pattern-forming four-wave-mixing processes to occur efficiently, they must meet a condition known as phase-matching. Phase-matching conserves momentum in the wave-mixing process, and it requires that the wavevectors of the four fields involved possess the appropriate magnitudes and directions according to

\[ \vec{k}_1 + \vec{k}_2 = \vec{k}_3 + \vec{k}_4. \] (3.1)

In certain situations, there exists a small phase mismatch \( \Delta \vec{k} \) among the fields, so that the phase-matching condition becomes \( \vec{k}_1 + \vec{k}_2 = \vec{k}_3 + \vec{k}_4 - \Delta \vec{k} \). As long as \( \Delta \vec{k} \) is small, these nearly phase-matched processes, which include those depicted in Figs. 3.3(b) and (c), still contribute to the four-wave-mixing process. The exact value of \( \Delta \vec{k} \) in my experiment is defined in Ch. 7.

In order to satisfy the condition in Eq. 3.1, one must consider both the wavevector direction as well as the magnitude \( |\vec{k}_j| = n_j \omega/c \), where \( n_j \) is the effective index
of refraction experienced by the optical field. Because the index of refraction is intensity-dependent, it is different for the pump fields and the weaker pattern-forming optical fields. Phase-matching among strong and weak optical fields is described by a phenomenon known as weak-wave retardation.

3.2.1 Weak-wave retardation

Weak-wave retardation was first introduced in 1966 in Ref. [Chiao et al. (1966)], and it describes the interaction of a strong field and a weak field propagating through a $\chi^{(3)}$ material, as depicted in Fig. 3.4(a). This interaction can generate four-wave-mixing processes analogous to those in my experiment, where there are exactly two strong pump fields and two weak pattern-forming optical fields involved in the four-wave-mixing process. In order to define the phase-matching condition for these four optical fields, it is necessary to define the effective index of refraction experienced by the strong and weak fields. I define the index of refraction experienced by a pump field to be

$$n_p = n_0 + n_2 I,$$  \hspace{1cm} (3.2)

where $n_0$ is the linear refractive index, $n_2 = 3\chi^{(3)}/(4\pi n_0^2 \varepsilon_0 c)$ is the coefficient of the nonlinear refractive index, and $I$ is the total optical field intensity [Boyd (2008)]. The concept of weak-wave retardation states that for pump fields experiencing an index of refraction defined in Eq. 3.2, then the index of refraction experienced by the weak fields is

$$n_w = n_0 + 2n_2 I,$$  \hspace{1cm} (3.3)

\textit{i.e.}, the nonlinear refractive index for the weak fields is twice that of the strong fields [Chiao et al. (1966)]. This is a special case of the nonlinear optical phenomenon known as cross-phase modulation, which describes how optical fields interact with one another in a nonlinear optical material. By making the assumption that one field is much weaker than the other, one can show that the nonlinear optical phase shift
Figure 3.4: Weak-wave retardation. (a) The system under consideration in Ref. [Chiao et al. (1966)]: a strong pump field of wavenumber $k_p$ and a weak optical field of wavenumber $k_w$ are applied to a third-order nonlinear material. (b) Phase-matching a four-wave-mixing process involving these two fields: For a nonlinear refractive index $n_2 > 0$, $k_w > k_p$, and this process can be phase-matched by allowing $\vec{k}_w$ to propagate at a slight angle relative to $\vec{k}_p$. For a nonlinear refractive index $n_2 < 0$, $k_w < k_p$, and this process can never be phase-matched for the fixed pump beam directions.

Based on Eqs. 3.2 and 3.3, it is clear that $|\vec{k}_w| < |\vec{k}_p|$ if $n_2 > 0$, and $|\vec{k}_p| > |\vec{k}_w|$ if $n_2 < 0$. The phase-matching conditions for these two scenarios are depicted in Fig. 3.4(b). Note that these are pictorial representations of the phase-matching condition defined in Eq. 3.1. If $n_2 > 0$, then the wavevectors of the weaker fields are longer than those of the pumps, and the phase-matching condition can be met if the weaker fields propagate at an angle relative to the applied pump fields. However, if $n_2 < 0$, then the wavevectors of the weaker fields are shorter, and the weak fields can never meet a phase-matching condition with the applied pump fields, whose directions are fixed. Weak-wave retardation therefore provides an explanation for why the pattern-forming optical fields emerge at a small angle relative to the pump fields: the intensity-dependent wavevector requires that angle in order to satisfy the phase-matching condition.
The phase-matching condition required by weak-wave retardation defines a specific angle $\theta$ at which the pattern-forming fields propagate relative to the applied pump fields. This angle is chosen by the initial transverse perturbations that initiate the wave-mixing process, where emission of optical fields at an angle $\theta$ has the highest scattering rate because it satisfies momentum conservation. This angle $\theta$ also depends on the effective index of refraction in order to meet the phase-matching condition. In addition, the nonlinear part of the refractive index must be sufficiently high in order for four-wave-mixing to occur efficiently and enhance the transverse perturbations.

Weak-wave retardation also imposes a restriction on the refractive index, i.e., it requires $n_2 > 0$. The condition that $n_2 > 0$ requires that $\chi^{(3)} > 0$, since [Boyd (2008)]

$$n_2 = \frac{3\chi^{(3)}}{4\eta_0^2 c}. \tag{3.4}$$

This condition is only satisfied for warm atomic vapors using blue detunings, which supports previous work that only observes pattern formation in warm atoms using fields tuned about the resonant frequency [Dawes et al. (2005)]. As I show in Chs. 4 and 6, however, this condition can be satisfied for both blue and red detunings using spatially bunched, cold atoms, which gives rise to a different dependence of $\chi^{(3)}$ on the detuning. However, weak-wave retardation enforces that the nonlinear refractive index must be positive to generate transverse patterns.

### 3.3 Threshold for wave-mixing

The minimum nonlinear refractive index at which pattern formation occurs is called the “threshold” refractive index. I derive this threshold condition for transverse optical pattern formation in cold atoms in Ch. 7. However, in order to understand this threshold condition and how it depends on the nonlinear refractive index, it is
Figure 3.5: **Backward four-wave-mixing configuration.** This is the configuration considered in Ref. [Yariv and Pepper (1977)], where two pump fields ($A_1$ and $A_2$) are applied at an angle relative to an elongated $\chi^{(3)}$ material of length $L$. This work studies the backward four-wave-mixing processes that generate and enhance the weak fields $A_3$ and $A_4$.

useful to first review a simplified system that also possesses a threshold for generating new optical fields.

The system that I review in this section was first considered in 1977 in Ref. [Yariv and Pepper (1977)]. Yariv and Pepper consider the light-atom interaction depicted in Fig. 3.5, where pump fields $A_1$ and $A_2$ are applied at an angle to an elongated $\chi^{(3)}$ material of length $L$. In this geometry, they consider a relatively large angle, so that backward four-wave-mixing is the only type of wave-mixing process that can be phase-matched. (Note: In transverse optical pattern formation, the other two types of four-wave-mixing processes depicted in Fig. 3.3 are not perfectly phase-matched and cannot occur spontaneously for large angles.) The weak fields that are generated/enhanced in the four-wave-mixing process are $A_3$ and $A_4$.

In this system, the total electric field inside the $\chi^{(3)}$ material is

$$\vec{E}(r, t) = \vec{A}_1(r)e^{i(k_1r - \omega t)} + \vec{A}_2(r)e^{i(k_2r - \omega t)} + \vec{A}_3(z)e^{i(-k_3z - \omega t)} + \vec{A}_4(z)e^{i(k_4z - \omega t)} + \text{c.c.}$$

(3.5)

The terms in the nonlinear polarization that are phase-matched to $\vec{A}_3(z)$ and $\vec{A}_4(z)$ are

$$\vec{P}^{\text{NL}}_3 = \epsilon_0 \chi^{(3)} \vec{A}_1(r)\vec{A}_2(r)\vec{A}_4^*(z)$$

(3.6)
and

\[ \vec{P}_{NL}^4 = \epsilon_0 \chi^{(3)}(\vec{A}_1(\vec{r})) \vec{A}_2(\vec{r}) \vec{A}_3^*(z), \]  

(3.7)

respectively. One can then solve the wave equation in Eq. 2.3 for each of these field components.

The solution of the wave equation yields the “gain” for \( \vec{A}_3(z) \) and \( \vec{A}_4(z) \). Gain is a measure of the power that a field acquires upon interaction with a nonlinear optical material. The gain for \( \vec{A}_3(z) \) and \( \vec{A}_4(z) \) are defined as

\[ G_{A_3} = \left| \frac{A_3(0)}{A_3(L)} \right|, \]  

(3.8)

and

\[ G_{A_4} = \left| \frac{A_4(L)}{A_4(0)} \right|, \]  

(3.9)

i.e., the output field amplitude divided by the input field amplitude. The gain for these fields is greater than 1 when an external source (like the pump fields) scatter into the direction of \( \vec{A}_3(z) \) or \( \vec{A}_4(z) \). In this case, this scattering process occurs via four-wave-mixing.

The authors in Ref. [Yariv and Pepper (1977)] show that in the special case of a finite input at \( A_4(0) \) but with \( A_3(L) = 0 \), the solutions to the wave equation for these field components are

\[ A_3(0) = -i \frac{\kappa^*}{|\kappa|} \tan(|\kappa|L) A_4^*(0) \]  

(3.10)

and

\[ A_4(L) = \frac{A_4(0)}{\cos(|\kappa|L)}, \]  

(3.11)

where \( \kappa = 2\pi k \chi^{(3)} A_1 A_2 / n \). In the limit where

\[ |\kappa|L = \pi/2, \]  

(3.12)
these equations become

\[
\left| \frac{A_3(0)}{A_4^*(0)} \right| \rightarrow \infty \tag{3.13}
\]

and

\[
\left| \frac{A_4(L)}{A_4(0)} \right| = G_{A_4} \rightarrow \infty. \tag{3.14}
\]

This condition of “infinite gain” is also known as “parametric oscillation” (i.e., “MPO” introduced in Ch. 2) or “lasing,” where even with zero input \([A_4(0) = 0]\), one can produce a finite field \([A_4(L) > 0]\) as a result of the nonlinear optical scattering process. This regime of infinite gain is what I observe in my pattern formation experiment, where after only applying counterpropagating optical fields in one dimension, I observe the formation of multiple, new optical fields with wavevectors that are distinct from the input wavevectors.

3.4 Summary

In this chapter, I describe the nonlinear optical effects that give rise to transverse optical pattern formation. I provide an overview of previous theoretical and experimental work on transverse optical pattern formation, and I present a theoretical example of how four-wave-mixing generates new optical fields. This theoretical framework becomes more complicated when using cold atoms, rather than warm atoms, as the nonlinear optical material. In the following chapters, I describe the additional complexities and advantages of using cold atoms to study pattern formation.
Transverse Optical Pattern Formation in Cold Atoms

Early studies of transverse optical pattern formation were performed in warm atomic vapors. In 2011, we reported the first observation of transverse optical pattern formation in cold atoms [Greenberg et al. (2011)]. Pattern formation in cold atoms gives rise to fundamentally different material properties than in warm atoms. In warm atoms, the nonlinear refractive index that gives rise to pattern formation arises due to polarization (spin) gratings induced in the atoms by optical pumping. In my system of cold atoms, the dominant contribution to the nonlinear refractive index is the bunching of atoms into real-space density gratings. Atomic bunching gives rise to enhanced light-atom interaction strengths, which I discuss further in Ch. 6.

In this chapter, I describe my experimental observation of transverse optical pattern formation in cold atoms, which was first reported in Ref. [Greenberg et al. (2011)]. I first describe the nonlinear refractive index that I induce in the atoms by applying counterpropagating optical fields, and I characterize the motional states of atoms in the imposed 1D optical lattice. I provide evidence of the importance of atomic bunching in this applied lattice to generating transverse patterns. I then char-
acterize the transverse optical patterns in terms of the threshold nonlinear refractive index as well as the multimode nature of the emergent patterns. I show that by using spatially bunched cold atoms instead of a homogeneous gas of atoms, one can achieve an orders of magnitude reduction in the power required for observing transverse optical pattern formation. I also discuss characteristics of our system that distinguish it from similar cold-atom experiments [Labeyrie et al. (2014)], including ultra-low thresholds and the observation of a continuous rotation of the optical patterns.

I note that I devote Chs. 5 and 7 to discussing the interplay between the optical patterns and the atomic density gratings. This chapter is devoted solely to introducing transverse optical pattern formation, its characterization in terms of the threshold nonlinear refractive index and the induced bunching in the 1D lattice, and the properties exhibited by the optical patterns. Chapter 5 goes on to characterize the properties of the atoms during optical pattern formation and describe the synergistic interaction between the transverse optical patterns and the atoms.

4.1 Overview of experiment

I first cool and trap the atoms in an anisotropic MOT (see App. A) to a temperature $T \approx 30 \, \mu\text{K}$. I then turn off the cooling and trapping beams in the MOT and apply counterpropagating, linear optical fields along the long ($\hat{z}$-) axis of the cloud, as depicted in Fig. 4.1(a). The atoms undergo Sisyphus cooling to a final temperature along $\hat{z}$ of $T_z = 2 - 3 \, \mu\text{K}$ [Greenberg et al. (2011)]. Below threshold, the temperature along $\hat{r}$ remains $T_{\text{rad}} \approx 30 \, \mu\text{K}$. After cooling, the atoms are tightly bunched in the applied lattice, as depicted by the pancake-shaped structures in Fig. 4.1(a). For red-detuned optical fields, the atoms tightly bunch at the intensity maxima of the optical lattice, which enhances the refractive index. (See Ch. 6.)

Above threshold nonlinear refractive index, a transverse instability generates new optical fields that propagate at a small angle $\theta \approx 3 - 10 \, \text{mrad}$ relative to the $\hat{z}$-axis,
Figure 4.1: Overview of pattern formation experiment. (a) Counterpropagating optical fields are applied in a linear polarization configuration (shown with green circles and arrows) to an anisotropic cloud of sub-Doppler-cooled $^{87}$Rb atoms. The atoms undergo Sisyphus cooling and bunch into pancake-shaped structures, as shown in the rectangle. (b) Above a threshold nonlinear refractive index, new optical fields are generated that propagate at a small angle $\theta \approx 3 - 10$ mrad relative to the applied fields. These fields can propagate anywhere along a cone of half-angle $\theta$. (c) When imaged in the transverse plane, I observe multi-spot optical patterns or ring-like patterns, where the central spot is bleedthrough pump light.

defined by the phase-matching condition discussed in Sec. 3.2. This phase-matching condition allows the optical fields to propagate anywhere along a cone of half-angle $\theta$, as depicted in Fig. 4.1(b). These generated fields appear in the transverse plane (imaging along $\hat{z}$) as multi-spot optical patterns or as ring-like patterns, as shown in Fig. 4.1(c). The central spot in these images is bleedthrough pump light. I can obtain these different types of optical patterns by adjusting the alignment of the applied optical fields, the shape of the atomic cloud, or the refractive index (e.g., by adjusting the applied field intensity or detuning). However, as I show in Sec. 4.4, I also observe different types of optical patterns under essentially the same experimental conditions, which is expected behavior for non-equilibrium processes like pattern formation.
4.1.1 Threshold for pattern formation

The threshold nonlinear refractive index for pattern formation can be approximated based on the condition for “infinite gain” in Eq. 3.12. To reach this condition, one requires a sufficiently strong nonlinear light-atom interaction. In the case of the backward four-wave-mixing geometry of Ref. [Yariv and Pepper (1977)], one requires that $\chi^{(3)}$, $|A_1|$, $|A_2|$, and $L$ are sufficiently large to reach the condition in Eq. 3.12, which I refer to as the threshold for parametric oscillation. In my experiment, the threshold condition for pattern formation is more complicated because there are multiple wave-mixing processes as well as higher-order nonlinear susceptibilities that give rise to parametric oscillation in multiple directions (i.e., many pattern-forming optical fields). In Ch. 7, I formally derive the equations for threshold in my experiment. However, the condition $|\kappa|L = \pi/2$ is still a good approximation, and it is useful to examine it for a qualitative understanding of the threshold for pattern formation.

One can rewrite Eq. 3.12 more generally as approximately $k \langle \chi_{\text{eff}}^{\text{NL}} \rangle L \approx \pi/2$, where $\chi_{\text{eff}}^{\text{NL}}$ is the nonlinear part of the effective susceptibility and $\langle \rangle$ denotes a spatial average. In the weak-bunching regime, where the dipole potential energy of the lattice is comparable to the thermal energy of the atoms, the nonlinear susceptibility for two-level atoms is

$$\chi_{\text{eff}}^{\text{NL}}(z) \approx -\chi_{\text{lin}} \left[ \frac{\Delta I(z) - \langle I(z) \rangle}{I_{s\Delta}} + \frac{I(z)}{I_{s\Delta}} \right], \quad (4.1)$$

where $\chi_{\text{lin}} = -6\pi(2\Delta)n_a/[k_{eg}^3(1 + 4\Delta^2)]$ is the linear susceptibility, $\Delta = \Delta/\Gamma$, $I(z)$ is the applied intensity distribution, $I_{s\Delta}$ is the off-resonant saturation intensity, and $\tilde{T} = T/T_D$. Equation 4.1 is derived in Ch. 6.

The temperature-dependent term in Eq. 6.4 corresponds to the bunching-induced nonlinearity, which generates density gratings and is the dominant nonlinearity in my experiments. The third term corresponds to the saturable (Kerr) nonlinearity,
which generates polarization gratings. Based on Eq. 4.1, one can enhance $\langle \chi_{NL}^{a}(z) \rangle$ and reach the threshold for pattern formation at low intensities by using high atomic densities (maximizing $n_a$), small detunings (minimizing $|\Delta|$), and cold atoms (minimizing $\tilde{T}$).

Realizing the threshold for parametric oscillation at low intensities is of interest for studying low-light-level nonlinear optical effects, as discussed in Ch. 1. To minimize the intensity threshold for pattern formation (i.e., parametric oscillation in my experiment), I use large optical depths $\text{OD} \sim 50 - 100$ (where $\text{OD} = 6\pi n_a L / k_{e\gamma}^2$), small detunings ($|\Delta| = 3$ to 10), and sub-Doppler-cooled atoms ($\tilde{T} \sim 3/146$).

The typical relationship that I measure between the threshold power for pattern formation and the detuning is shown in Fig. 4.2(a). The threshold intensity increases for larger detunings because $|\chi_{eff}|$ is smaller for larger $|\Delta|$, and thus one requires higher powers to meet the threshold for pattern formation. For detunings closer to the resonant frequency than $\Delta = -4\Gamma$, the threshold intensity is also higher. I attribute this to the detrimental effects of absorption on the light-atom interaction, which also reduces $\chi_{eff}$, according to the imaginary part of Eq. 2.27. In contrast, Greenberg observes the minimum threshold at $\Delta = -3\Gamma$ [Greenberg (2012)]. I attribute this difference to the fact that absorption plays a larger role in my beam geometry, where the pump beams propagate down the entire 3 cm length of the atomic cloud. In Ref. [Greenberg (2012)], the pump beams are much larger than the size of the cloud and propagate at a large angle relative to the long axis. As a result, the number of atoms with which each pump beam interacts per optical cross section is smaller than in my experiment and is thus less affected by absorption.

The minimum power at which I have observed pattern formation is 420 nW per pump beam at $\Delta = -4\Gamma$, as shown in Fig. 4.2(b), for an optical depth $\sim 100$. This is a factor of $\sim 10^3$ smaller than the minimum threshold power observed for pattern
Figure 4.2: **Threshold vs. detuning.** The threshold power per pump beam as a function of detuning, where $\Delta < 0$. This data was taken with a $1/e^2$ beam diameter of 156 $\mu$m and (a) an optical depth of OD $\sim 60$ in gray circles. (b) The data from (a) is shown alongside OD $\sim 20$ (red squares) and OD $\sim 100$ (blue diamonds). The error bars represent the typical statistical uncertainty for all data points.

formation in warm atoms [Dawes (2008)], which is a manifestation of the enhanced nonlinearity achievable by using cold atoms and is discussed further in Ch. 6. The threshold power is smaller for larger optical depths because $|\chi_{\text{NL}}^{\text{eff}}|$ $\propto$ OD, which is supported by the trend in Fig. 4.2(b) for increasing OD.

The corresponding minimum intensity at which I have observed pattern formation is $\sim 0.8$ mW/cm$^2$, which is more than an order of magnitude smaller than the intensity threshold for other cold-atom systems exhibiting pattern formation [Labeyrie *et al.* (2014)]. The lower threshold in my system is due to the lower atomic temperature ($2 - 3$ $\mu$K c.f. $\sim 60$ $\mu$K in Ref. [Labeyrie *et al.* (2014)]). Lower atomic temperatures enhance the bunching induced nonlinearity in Eq. 4.1, which allows me to meet the threshold condition of Eq. 3.12 at lower intensities.

According to the weak-wave retardation phenomenon discussed in Sec. 3.2, one can only reach the threshold for pattern formation in the weak-bunching regime when $\langle \chi_{\text{eff}}^{\text{NL}}(z) \rangle > 0$ in Eq. 4.1 [Chiao *et al.* (1966)]. This condition is always satisfied for blue detunings ($\chi_{\text{lin}} < 0$), and it is satisfied for red detunings ($\chi_{\text{lin}} > 0$) when the
bunching-induced nonlinearity is greater in magnitude than the saturable nonlinearity. In addition, as I show in Ch. 6, the use of red detunings instead of blue detunings gives rise to a greater $\chi_{\text{eff}}$ in the tight-bunching regime, where the dipole potential energy greatly exceeds the thermal energy of the atoms, and which is the regime in which I work experimentally. For this reason, along with the fact that Sisyphus cooling only cools for red detunings [Castin et al. (1991)], I only use red-detuned optical fields in my experiments.

4.2 Atomic bunching in the imposed 1D lattice

The nonlinear refractive index is greatly enhanced in the tight-bunching regime when many sub-Doppler-cooled atoms organize at the intensity maxima of the applied 1D lattice. Because the bunching-induced nonlinearity in Eq. 4.1 can be made much larger than the saturable nonlinearity in the sub-Doppler regime, I expect that atomic bunching is the dominant nonlinear contribution to reaching the threshold condition for generating patterns. To verify that the atoms are bunched in the applied lattice, I perform a step-change in the intensity of the applied pump beams. This non-adiabatic change in the dipole potential well depth results in “breathing-mode oscillations” [Raithel et al. (1997a)], wherein the power scattered by the atoms oscillates in time according to the motional vibrational frequency of the new dipole potential well.

To perform this series of experiments, I detect the power in the pattern-forming optical fields and use them as probe beams scattered off the applied lattice. I only work above threshold for pattern formation in this experiment when the atoms are tightly bunched in the applied lattice. Working below threshold, the atoms are only weakly bunched, and I find that the reflectivity of an externally applied probe beam is too small to detect the signature of the bunched atoms.

By working above threshold, where some of the applied pump power is lost to the
generated fields, I incur errors in the expected dipole potential well depth. However, the pattern-forming optical fields are nearly phase-matched to scatter off the applied lattice gratings, as shown in Fig. 3.3(c), which gives rise to a good signal strength. I do not detect a signature from the other gratings depicted in Figs. 3.3(a) and (b) because they are much shallower than the applied pump-pump dipole potentials, i.e., they form due to the interference of a strong pump field and a weak generated field. As I show in Ch. 5, these self-generated gratings are more than an order of magnitude weaker, and thus the signature of the applied dipole potentials dominates the breathing mode oscillations.

The breathing-mode oscillations are visible in Fig. 4.3(a). Here, the atoms are cooled and trapped in the lattice during $-200 \mu s < t < 0 \mu s$ with applied intensities of 2 mW/cm$^2$ per pump and $\Delta = 2\pi \times (40) \text{ MHz}$. At $t = 0$, the intensity in each pump beam is increased to 13 mW/cm$^2$. The resulting oscillations are fit to an exponentially increasing function with decaying periodic oscillations $\propto [A \exp(-t/\tau_1) \cos(\omega t + \phi) + B \exp(t/\tau_2)]$, where $\omega = 2\pi \times 255 \text{ kHz}$ in this experiment. The exponential increase corresponds to the increase in the power generated by the wave-mixing process due to the increase in pump intensity.

I took similar sets of data for different final pump intensities. I relate these to a dipole potential magnitude $U_0 = \hbar |\Delta| I_{\text{tot}}/I_{\Delta}$ from the spatially dependent part of Eq. 2.35 in the low-intensity limit, where $I_{\text{tot}}$ is the total applied intensity of the pump beams. The results are shown in Fig. 4.3(b) with a fit to the expected motional frequency of a harmonic potential $\omega/2\pi = \sqrt{2k^2U_0/m}$, normalizing $U_0$ by the recoil energy $E_r = \hbar^2k^2/2m$. The deviation of the experimental data from this fit at lower intensities likely arises because of the error in estimating $I_{\text{tot}}$ due to a beam reshaping effect, which is discussed in Sec. 4.4, as well as the loss of power to generating the optical patterns. This error is discussed in more detail in Sec. 5.1.2. The results of Fig. 4.3(b) indicate that the atoms are bunched in the pump-pump lattice above
Figure 4.3: **Breathing mode oscillations.** (a) The power in the generated fields as a function of time, where a step-change in the pump intensity occurs at \( t = 0 \). For \(-200 \mu s < t < 0\), the total pump intensity is 2 mW/cm\(^2\). For \( t > 0 \), the total pump intensity is 13 mW/cm\(^2\). (b) The frequency of the breathing-mode oscillations extracted from data similar to plot (a) for various final dipole potential well depths \( U_0 \), normalized by the recoil energy \( E_r \).

threshold for pattern formation, which enhances \( \langle \chi_{\text{NL}}^\text{eff}(z) \rangle > 0 \) in Eq. 4.1 and gives rise to pattern formation at low light levels.

4.2.1 Pushing the atoms

To further investigate the importance of bunched atoms on pattern formation, I apply a 50-\( \mu \)s-wide pulse of radiation pressure to the atoms along \( \hat{z} \) by increasing the power in one of the pump beams from 4 to 7 mW/cm\(^2\). The results of this are shown in Fig. 4.4, which shows a typical temporal signal for power in the optical patterns, but where I turn on the radiation-pressure pulse at time \( t_{\text{on}} = 200 \mu s \). I attribute the initial spike to an increase in scattering efficiency due to the increased pump power before the atoms are pushed out of the lattice. The power in the optical patterns then decreases substantially while the atoms are being pushed during the 50-\( \mu \)s pulse. Once the pulse is turned off at \( t_{\text{off}} = 250 \mu s \), the power in the patterns begins to increase as the atoms re-bunch in the lattice.

This experiment shows the importance of applying equal-intensity, counterprop-
Figure 4.4: **Pushing atoms in the pump-pump lattice.** At time $t_{\text{on}} = 200 \, \mu s$, a 50 $\mu s$-wide pulse was applied along the direction of one of the pump beams. This imposes a radiation pressure on the atoms, pushing them out of their original dipole potential wells. The signal recovers after the pulse is removed and the atoms can re-bunch.

agating optical fields to the atoms. When the atoms experience no net radiation pressure, more atoms can bunch in the lattice, which gives rise to more efficient pattern formation. As I show in Chs. 6 and 7, high atomic densities and tight atomic bunching give rise to enhanced light-atom interaction strengths, which allows me to observe effects like self-organization in free space without the need for a cavity.

4.2.2 *Bistability and the effect of Sisyphus cooling*

Atomic bunching in the applied lattice is aided by Sisyphus cooling, which cools the atoms from an initial temperature of 30 $\mu K$ to a final temperature along $\hat{z}$ of $T_z = 2 - 3 \, \mu K$. Without Sisyphus cooling, the bunching-induced nonlinearity in Eq. 4.1 would be an order of magnitude smaller, resulting in a greatly reduced light-atom interaction strength. Because Sisyphus cooling is dissipative, as discussed in Sec. 2.3.3, it gives rise to a hysteresis in the nonlinear refractive index of the atoms; *i.e.*, once the atoms are bunched, they have different properties than atoms that
have not yet undergone maximum cooling and bunching. This hysteresis manifests itself as a “transient bistability” in the scattering rate for pattern formation, which is a well-known effect in atoms [Bonifacio and Lugiato (1978)] and arises due to the saturable nature of the nonlinearity.

Bistability is a feature observed in many non-equilibrium systems, including those that exhibit superradiance [Bowden and Sung (1979)] and self-organization [Asbóth et al. (2005)]. In my experiment, the bistable behavior arises in a dual nature in the scattering efficiency of the wave-mixing process depending on whether the atoms require cooling and bunching from the pump fields, or whether they are already cooled and bunched.

To verify the bistable behavior in my system and thus quantify one effect of Sisyphus cooling, I perform an experiment to extract the power in the pattern-forming optical fields for the case where atoms are “pre-bunched.” In this case, I use the pump scheme depicted in Fig. 4.5(a), where I apply high-intensity pump beams (14 mW/cm² per beam) for 200 µs, which cool and bunch the atoms. I then make a step reduction in the pump intensity and measure the power in the pattern-forming optical fields near threshold. The power in the pattern-forming optical fields as a function of pump intensity for this pre-bunched case is shown as the red circles in Fig. 4.5(b). I compare this to the “normal” case in blue, where the atoms are not pre-bunched, and the pump beams must act to both cool and bunch the atoms as well as generate the pattern-forming optical fields.

By employing this “pre-bunching” method, the scattering efficiency is higher, i.e., the power generated by the wave-mixing process is higher for a given applied intensity. These two situations give rise to different scattering efficiencies because they are characterized by different nonlinear refractive indices. Atoms that are “pre-bunched” using high-intensity fields have a different density distribution than atoms that still require cooling and bunching to reach the threshold for pattern formation,
which alters $\chi^{NL}_{\text{eff}}$. I note that it is difficult to directly compare the minimum threshold in this case because the “pre-bunched” case also generates Bragg structures, which are discussed in Sec. 5.2. The bistability in the scattering rate demonstrates that cooling and bunching give rise to a qualitatively different nonlinear refractive index, which verifies the importance of the bunching-induced nonlinearity to reaching the threshold for pattern formation at low intensities.

4.3 Transient optical pattern formation

After cooling and for sufficiently tight atomic bunching, the nonlinear refractive index reaches the threshold for pattern formation, approximated by $k \langle \chi^{NL}_{\text{eff}} \rangle L \approx \pi/2$ via Eq. 3.12. Once the patterns begin to form, there is an exponential buildup of the power in the patterns, as shown in Fig. 4.6. Figure 4.6(a) shows the results of a typical pattern formation experiment, where I detect the power in the generated fields as a function of time, and the pump beams are turned on at time $t_{\text{on}} = 0$. 
Figure 4.6: **Exponential power increase.** (a) Power in the generated fields as a function of time for $I_p = 12$ mW/cm$^2$ and $\Delta = 2\pi \times (-24)$ MHz. (b) Zooming in on the formation of the patterns, the exponential fit (red, solid curve) to a function of the form $a\exp[(t - t_\delta)/\tau] + b$ has an exponential constant $\tau = 5.8 \mu s$.

In this transient regime, where I usually work, I load the MOT for 97 ms and then shut the cooling and trapping beams (the “MOT” beams) off. I then turn on the pump beams for 3 ms to perform experiments. (See Appendix A for additional experimental details and detection procedures.)

Figure 4.6(b) provides a closer look at the behavior of the generated fields close to time $t_{on}$, when the MOT beams are turned off and the pump beams are turned on. Once a finite field begins to form, there is an exponential increase in the power generated in the wave-mixing process, which is the expected behavior for such wave-mixing instabilities [Yariv and Pepper (1977); Agrawal (2008)].

The maximum power is typically reached $50 \mu s - 200 \mu s$ after $t_{on}$, which corresponds to the time it takes for the atoms to undergo Sisyphus cooling from their interaction with the pump beams. (See Sec. 5.3.2 for a discussion of this timescale.) I thus interpret the peak of the signal to correspond to the time at which the nonlinear refractive index effectively “saturates,” wherein once most of the atoms have undergone Sisyphus cooling, the bunching-induced nonlinearity reaches its peak value. The decay of the power in the optical patterns then arises due to loss of atoms from the
Figure 4.7: Pattern power vs. time. Power in pattern-forming optical fields in the transient regime as a function of time for 4 $\mu$W pump powers and $\Delta = 2\pi \times (-42)$ MHz. Here, optical pattern formation persists for $\sim 2.4$ ms.

The patterns typically persist for 1 – 2.4 ms (see Fig. 4.6(a) and Fig. 4.7). The oscillatory behavior in the power output arises from pattern rotation out of the detector’s collection region (see Sec. 4.4.1) as well as Larmor precession of the atoms (see Sec. 5.2.2). The observation of transient pattern formation persisting for $\sim 2.4$ ms is much longer than in other transient wave-mixing experiments in cold atoms [Greenberg and Gauthier (2012b)], and it is evidence that the atoms undergo three-dimensional cooling, as discussed further in Sec. 5.3.2.

4.4 Continuous symmetry-breaking

The observation of pattern formation in cold atoms is not only of interest for its reduced intensity thresholds, but also for its access to studying multimode, non-equilibrium phenomena at low temperatures. Multimode systems that exhibit pattern formation are expected to exhibit unique non-equilibrium phenomena, such as continuous symmetry-breaking [Gopalakrishnan et al. (2009)], that are inaccessible in the single-mode regime. While continuous symmetry-breaking has been observed in warm-atom systems exhibiting pattern formation [Dawes (2008)], cold-atom sys-
tems provide a synergistic coupling between the optical field geometry and the real-space density distribution of atoms, which is of great interest in condensed matter physics [Gopalakrishnan et al. (2011)]. As I show in Ch. 5, the atoms self-organize into real-space structures according to the geometry of the optical patterns, which I refer to as coupled optical-atomic pattern formation. Because my system is the first to directly observe multimode atomic pattern formation, it is the first to access multimode non-equilibrium phenomena, such as continuous symmetry-breaking, at low temperatures.

When I detect the generated fields on a camera, they appear as multi-spot optical patterns, as shown in Fig. 4.1(c), which represents a breaking of a continuous symmetry. If all possible modes were excited, one would expect to see a continuous ring of generated light. The observation this continuous symmetry-breaking has been shown to be energetically favorable over the symmetric ring pattern [Geddes et al. (1994)].

I obtain the different multi-spot patterns in Fig. 1.2(c) by changing the alignment of the input pump fields or by adjusting $n$, e.g. by changing the optical depth or the pump intensity, which changes the phase-matching condition for the wave-mixing process.

I also find that the atoms can spontaneously choose different broken symmetries under the same experimental conditions. For example, each pair of optical patterns shown in Fig. 4.8 were taken in contiguous experimental runs (100 ms apart). In between each image, the MOT was re-loaded, but all other experimental parameters are the same for each pair. These shot-to-shot fluctuations in consecutive experimental runs are a hallmark of non-equilibrium phenomena [Black et al. (2003); Baumann et al. (2010)], where symmetry-breaking results in the spontaneous excitation of different modes.

The maximum number of modes supported by a system is determined by the
Fig. 4.8: **Shot-to-shot pattern fluctuations.** Images of optical patterns taken in contiguous experimental runs (100 ms apart) under the same experimental conditions. The first example shows a 12-spot pattern and a 6-spot pattern. The second example shows a 10-spot pattern and a nearly continuous pattern with multiple radial modes. The third example shows a 4-spot pattern and a broken ring pattern. The central spot is bleed-through pump light and the small ring immediately surrounding it is a beam reshaping effect.

Fresnel number $\mathcal{F}$, defined as

$$\mathcal{F} = \frac{w^2}{L\lambda}. \quad (4.2)$$

Because $\mathcal{F} \approx 6.83$ in my system, it can support a maximum of approximately 6 modes, which is exhibited in the 12-spot patterns of Fig. 4.8. “One mode” corresponds to a two-spot optical pattern, where two fields are always generated on opposite sides of the cone of emission in order to conserve momentum in the wave-mixing process.

The small ring closely surrounding the central pump spot in the optical patterns shown in Fig. 4.8 is a beam reshaping effect that arises because the pump beam size is comparable to the width of the atomic cloud. This gives rise to a characteristic diffraction pattern of concentric circular fringes in the far field [Saleh and Teich (2007)]. For the case of $\Delta/\Gamma = -5$ and a single-pump intensity of 0.5 mW/cm², this diffraction pattern in the far-field is shown in Fig. 4.9. The first image shows the case where the applied counterpropagating pumps are left on all the time—even during the loading of the MOT. The second image shows the case where the applied
counterpropagating pumps are only turned on during a 1 ms period when the MOT beams are off. The third image shows the case of a single probe beam applied to the cloud of atoms. The fourth image is for comparison with the size of the pump in the far field when there are no atoms. The ring generated by this reshaping effect has consequences for measuring the dipole potential wells in the atoms, which is discussed further in Sec. 5.1.2.

4.4.1 Pattern rotation

The generated transverse optical patterns are not necessarily stationary, and they can in fact rotate around the allowed cone of emission during a single experiment. This pattern rotation is observed visually on a CCD (charge-coupled device) camera (see App. A), and the timescales are studied on an oscilloscope. To measure this pattern rotation on an oscilloscope, I split the path of the generated fields using a 50-50 beam splitter, and I place apertures in each path to select two distinct spatial modes of emission, as depicted in the inset of Fig. 4.10. The output of these two modes are sent to two different detectors. One example of the rotation of the optical patterns between these spatial modes is shown in Fig. 4.10. Here, the vertical axis is the relative amplitude of the generated field power that reaches the detectors, and the horizontal axis is time, where the pump beams are turned on at time 0. The
Figure 4.10: Pattern rotation. The blue and gray circles show the power in the generated light that propagates through two distinct spatial locations on the cone of emission, as depicted in the inset. The observed anticorrelation (correlation factor -0.2) as a function of time indicates a rotation of the optical pattern between these two modes.

The gray curve shows the power emitted into one spatial mode, and the blue curve shows the power emitted into the other. The correlation factor between these modes is -0.2, which indicates temporal anti-correlation. The fastest timescale for fluctuations between modes is \(\sim 50 \mu s\), which is the order of the time it takes for an atom to move to the next-nearest pancake and thus contribute to exciting a different optical pattern.

This smooth rotation of patterns among different spatial locations is a manifestation of the degeneracy among the multiple modes of our system and is also observed in other pattern-forming systems [Dawes (2008)]. In my cold-atom pattern-forming experiment, fluctuations in the atomic density distribution give rise to a continuous re-organization of atoms into different spatial modes. The result is a continuous movement of the optical patterns, where the amplitude of the generated light in a particular mode is maximized once most of the atoms have re-organized into a particular spatial mode. The initial symmetry-breaking of the optical pattern is therefore not fixed, and other solutions of the broken symmetry are easily excitable.
4.4.2 Correlations among optical patterns

There also exist strong phase and temporal correlations among the generated modes of the optical patterns, which is expected for systems exhibiting transverse effects [Gatti and Mancini (2001)] and for conserving momentum in the wave-mixing process [Greenberg et al. (2011)]. While characterizing these correlations is not essential to understanding the main focus of this thesis, it is useful for motivating potential future directions of this work, which I discuss briefly in this subsection as well as in Ch. 8.

Phase correlations

There exist strong phase correlations between the generated fields in a single spatial mode. In order to measure the relative phase of the generated fields, I interfere the optical patterns with a large Gaussian beam on a CCD camera (see App. A), and I see the results shown in Fig. 4.11 for two- and four-spot optical patterns. The orientation of the optical patterns is simulated in the left panels, and the data is shown in the right panels.

Figure 4.11: Phase of generated fields. Left panels: Orientation of optical patterns shown to the right. Right panels: The interference of two- and four-spot optical patterns with a large Gaussian field. The central spot is bleed-through pump light.
Following the circular fringes, there is a continuous interference pattern for the two-spot pattern, which indicates that the fields generated in a two-spot optical pattern have the same relative phase. In a four-spot pattern, however, there are discontinuities in the path of the circular interference fringes, which indicate that there are $\pi$-phase shifts between neighboring spots. This is a common phase relationship for systems exhibiting transverse optical structure [Hall (1990)], \textit{e.g.} in a TEM$_{11}$ mode, where neighboring lobes are $\pi$ out-of-phase.

Temporal correlations

The dynamics of the optical patterns are also strongly correlated in time on both sides of the atomic cloud, \textit{i.e.}, between those patterns propagating nearly along $+\hat{z}$ and $-\hat{z}$. These correlations are most apparent when measuring the power of the optical patterns as a function of time while operating in the steady-state regime, when the optical patterns typically persist for $\gtrsim 2$ sec \textit{c.f.} up to 2.4 ms in the transient regime.

To operate in the steady-state regime, I load the MOT for 97 ms, and then I reduce the intensity of the cooling and trapping beams by 85%. By leaving on the cooling and trapping beams at a reduced intensity, I retain more atoms, which allows the patterns to persist longer.

To study the correlations among the optical patterns in the steady-state regime, I set up two detectors on opposite sides of the cloud of atoms, and I observe the results shown in Fig. 4.12. This experimental data was taken in collaboration with Joel A. Greenberg. Figure 4.12(a) shows a 1-sec. window of the detected optical pattern power, where the blue and gold curves show the power reaching the two detectors. In Fig. 4.12(b), I zoom in to show a smaller timescale, where the power correlations are more apparent. The correlation factor between these data sets is 0.72, which indicates a strong correlation in the optical pattern power emitted on opposite sides of the atomic cloud. Due to these long coherence times, my system may be useful as
**Figure 4.12: Steady-state correlations.** (a) Power in pattern-forming optical fields on each side of the cloud (blue and gold indicate different sides) of atoms as a function of time. In this experiment, the patterns can persist for more than 1 sec by leaving the cooling and trapping beams for the MOT on at 15% of the normal loading intensity. (b) A close-up of figure (a) to show the strong correlation (correlation factor 0.72) between the fields generated on each side of the cloud.

mirrorless laser for optical metrology [Meiser and Holland (2010)]. In addition, with strong intensity and phase correlations in the macroscopic optical patterns, it is likely that close to threshold, the atoms generate twin photons on each side of the cloud, which is a known characteristic of four-wave-mixing schemes [Boyer et al. (2008)]. Thus, one future direction of my work is to use my system to generate entangled or hyperentangled photon pairs [Yan and Zhu (2013)], where hyperentanglement is feasible due to the strong polarization correlations among the patterns.

4.5 Summary

This chapter provides an experimental characterization of the transverse optical patterns that I study. I analyze the threshold condition for pattern formation and show...
that atomic bunching can give rise to enhanced light-atom interactions and reduced power requirements for generating patterns. I verify that the atoms are bunched in the applied optical lattice during pattern formation, and I characterize the effects of perturbing and modifying the amount of bunching on the power in the optical patterns. I also characterize the properties of the transverse optical patterns and relate them to previous studies of pattern formation. The work in this chapter represents a detailed description of the first experimental observation of pattern formation in cold atoms [Greenberg et al. (2011)], as described in the first row of Table 1.1.

In this chapter, I restrict my discussion of atomic bunching to the applied optical lattice, which is crucial for reaching the threshold for pattern formation. However, the interaction of the pattern-forming optical fields with the applied fields gives rise to dipole forces that can bunch atoms into new, self-organized structures. In the next chapter, I measure the properties of these self-organized atoms, thereby demonstrating the first direct observation of multimode, real-space atomic self-organization.
Atomic Pattern Formation and Spontaneous Three-Dimensional Cooling

Self-organization of atoms into real-space structures has been observed in single-mode geometries [Black et al. (2003)], which has been of great interest for studying phase transitions [Baumann et al. (2010)] and non-equilibrium phenomena such as spontaneous symmetry-breaking [Baumann et al. (2011)]. As described in Ch. 1, the observation of self-organization in multimode geometries allows access to different physics, such as continuous symmetry-breaking, compliant lattices, and phase transitions that are inaccessible in single-mode systems [Gopalakrishnan et al. (2009)].

In this chapter, I present the first direct observation of multimode self-organization of cold atoms into real-space structures. This self-organization arises due to the synergistic coupling between the transverse optical fields and the center-of-mass degrees of freedom of the atoms. As the transverse fields begin to form due to the wave-mixing instability, they impose a dipole force on the atoms, which forces the atoms to both cool and bunch into new structures with transverse components. These structures in turn scatter more power into the generated optical fields, which results in a
runaway enhancement of both the power in the optical patterns and the amount of bunching of the atoms into corresponding atomic patterns. These atomic patterns are purely self-organized because they are not imposed externally.

In this chapter, I show that the mutual interaction between the transverse optical fields and the atoms generates both multimode self-organization of atoms on multiple spatial scales as well as spontaneous 3D Sisyphus cooling. While other cold-atom systems have observed the spontaneous formation of multimode optical fields [Schilke et al. (2012a); Greenberg and Gauthier (2012b); Labeyrie et al. (2014)], these experiments do not provide information about the motional states of the atoms. Here, I provide direct in situ measurements of the motional atomic energy states for both the imposed and self-organized atomic structures. I also show that, despite only applying optical fields in 1D, pattern formation spontaneously generates Sisyphus cooling in all three spatial dimensions. Three-dimensional cooling schemes have previously been studied using applied 3D lattices in order to achieve longer coherence times [Raithel et al. (1997b)], but it has not yet been observed to occur spontaneously. This work represents the first observations of both real-space multimode self-organization and spontaneous 3D Sisyphus cooling. My work is also distinguished in that I observe this self-organization at ultra-low intensities (c.f. Refs. [Schilke et al. (2012a); Labeyrie et al. (2014)]), which allows for studies of multidimensional, low-light-level nonlinear optics.

This chapter is organized as follows. I characterize the self-organized atomic structures using parametric resonance and Bragg scattering techniques. I then develop a heuristic model to relate the decay of a Bragg-scattered signal to an atomic temperature. Finally, I describe my measurements of the atomic temperature in the self-organized gratings and show that the atoms have undergone 3D cooling. This chapter is based largely on my work in Ref. [Schmittberger and Gauthier (2016b)].
5.1 Self-organization: Emergent bunching

There are two types of structures into which atoms bunch in this experiment: imposed and self-organized. In Sec. 4.2, I describe the imposed structures, which arise due to bunching of atoms into the applied 1D lattice. The self-organized structures form above threshold for pattern formation due the synergistic coupling between the generated optical fields and the atoms.

There are two types of self-organized atomic patterns: short- and long-wavelength gratings. The short-wavelength gratings, depicted in Fig. 5.1(a), arise due to the interference of a pattern-forming optical field with a nearly counterpropagating pump field and have a spacing of \( d_s \approx \pi/[2k'\cos(\theta/2)] \approx 195 \text{ nm} \). These gratings overlap strongly with the imposed pump-pump grating of spacing \( d_p = \pi/2k' \), and sit at only a small angle relative to the pancakes depicted in Fig. 1.2(a). The long-wavelength gratings, depicted in Fig. 5.1(b), arise due to the interference of a pattern-forming optical field with a nearly co-propagating pump field and have a spacing of \( d_\ell \approx \pi/[2k'\sin(\theta/2)] \approx 80 \mu\text{m} \). I only depict these two examples for simplicity, but there exist complimentary gratings for all combinations of generated fields and pump fields. Where these gratings overlap with the pancakes depicted in Fig. 4.1(b), the atoms self-organize into new structures, as simulated by Fig. 5.1(c).

I define the grating wavevectors for the pump-pump, short-wavelength, and long-wavelength gratings as \( \vec{g}_p = \pm 2k'\hat{z}, \vec{g}_s = k'\{\pm[1 + \cos(\theta)]\hat{z} \pm \sin(\theta)\hat{r}\}, \) and \( \vec{g}_\ell = k'\{\pm[1 - \cos(\theta)]\hat{z} \pm \sin(\theta)\hat{r}\}, \) respectively. I define \( \zeta_{s,\ell} = U_{\text{dip},(s,\ell)}/k_B T_{s,\ell} \), where \( U_{\text{dip},(s,\ell)} \) defines the effective dipole potential of the short-wavelength (s) and long-wavelength (\( \ell \)) gratings, and \( T_{s,\ell} \) refers to the atomic temperature along \( \vec{g}_{s,\ell} \). When \( \zeta_{s,\ell} > 1 \), the atoms bunch into the self-generated dipole potentials, resulting in self-organization. I measure \( U_{\text{dip},(s,\ell)} \) and \( T_{s,\ell} \) in the following sections, which provides the first direct observation of multimode self-organization of cold atoms.
Figure 5.1: **Self-generated patterns.** (a) The interference of a pump field and a nearly counterpropagating generated field form short-wavelength interference patterns of spacing $d_s \approx 195 \text{ nm}$, whose dipole potential wells have grating wavevectors along the direction $\hat{g}_s$. (b) The interference of a pump field and a nearly copropating generated field form long-wavelength interference patterns of spacing $d_\ell \approx 80 \text{ \mu m}$, whose dipole potential wells have grating wavevectors along the direction $\hat{g}_\ell$. (c) A simulation of the self-generated atomic patterns in a given pancake [c.f. Fig. 1.2(a)]: A two-spot optical pattern generates striped atomic patterns. Higher-order optical patterns generate more complicated atomic patterns.

5.1.1 Parametric resonances

To measure that the atoms undergo self-organization, I measure the motional vibrational frequencies of atoms during pattern formation. I perform parametric driving experiments to modulate the optical lattices (both imposed and self-generated) at variable frequencies. While shaking the lattices, I measure the power in the pattern-forming optical fields. When a parametric resonance is excited, atoms heat out of the lattice, thus reducing the efficiency of the wave-mixing process and the power in the generated fields. I expect to observe three distinct resonances due to the different material gratings [$U_{\text{dip},(p,s,\ell)}$, where $U_{\text{dip},p}$ is the imposed pump-pump potential].

I assume the dipole potential wells to be harmonic, so that the motional frequencies of atoms in the lowest energy bound states oscillate according to

$$\omega_{\text{vib},(p,s,\ell)} \approx \sqrt{\frac{\pi^2 U_{\text{dip},(p,s,\ell)}}{2m^2 d_{(p,s,\ell)}^2}}. \quad (5.1)$$
Parametric resonance occurs at a frequency $2\omega_{\text{vib}}$ when modulating along the direction of the grating wavevectors [Raithel et al. (1997a)]. Because $\hat{g}_f$ is nearly orthogonal to $\hat{g}_p$ and $\hat{g}_s$, I use different experimental methods to parametrically drive atoms in these gratings.

**Short-wavelength gratings**

To modulate the potential along $\pm \hat{z}$ and drive atoms in both the imposed and short-wavelength gratings, I use an electro-optic phase modulator (New Focus 4002 Broadband Phase Modulator, DC-100 MHz, 500-900 nm) placed in the path of one of the pump fields. This drives a periodic phase shift at an adjustable frequency $\omega_{\text{mod}}$, which shakes the optical lattice. During these parametric resonance experiments, I slightly misalign the pump beams so that I observe a stationary, two-spot pattern, in order to avoid detection errors due to the pattern rotation effect discussed in Sec. 4.4. I operate these experiments at a detuning $\Delta = -2\pi \times (28 \pm 2) \text{ MHz}$.

I observe two distinct resonances, as depicted in Fig. 5.2(a). The high-frequency resonance at $\omega_{\text{mod}}/2\pi \approx 632 \pm 30 \text{ kHz}$ (width $258 \pm 26 \text{ kHz}$) corresponds to the pump-pump (applied) lattice, where the error is due to the confidence interval of the Lorentzian fit. This agrees with the expected value of $2\omega_{\text{vib}}/2\pi \simeq 686(+123/-109) \text{ kHz}$ from Eq. 5.1 for $U_{\text{dip},p} = U_0$ with experimental parameters $\Delta \simeq -5\Gamma$ and $I/I_{s\Delta} = 0.28$. The low-frequency resonance at $\omega_{\text{mod}}/2\pi = 92 \pm 1 \text{ kHz}$ (width $33 \pm 2 \text{ kHz}$) corresponds to the short-period self-organized gratings, which agrees with the expected value of $\omega_{\text{mod}}/2\pi = 95 (+12/-9) \text{ kHz}$ for a measured generated field intensity of $(17 \pm 6) \mu\text{W/cm}^2$. These predicted values and the associated errors are discussed in more detail in Sec. 5.1.2. The observation of this low-frequency motional resonance provides unambiguous proof that the atoms self-organize into structures that are not imposed on the atoms by the applied fields, which is a major result of my research.
Driving parametric resonances. The amplitude of the generated light when modulating the phase of one of the pump beams at $\omega_{\text{mod}}/2\pi$ by $\pm \pi/37$, normalized by the amplitude in the unperturbed ($\omega_{\text{mod}}/2\pi = 0$) case. Experimental data is shown in gray circles, and the best fit to a sum of Lorentzian functions is the blue curve. (a) Driving along $\hat{z}$ using an electro-optic phase modulator. (b) Driving along $\hat{r}$ by intensity-modulating an elliptical probe beam of intensity $\tilde{I} = 2 \times 10^{-3}$ applied to the side of the atomic cloud. The error bars indicate the confidence interval of the fit function at the locations of parametric resonances, the values of which are quoted in the text.

From Eq. 5.1 and normalizing the dipole potential energy by $k_B/2$, $U_{\text{dip,p}} \sim 635 \, \mu\text{K}$ and $U_{\text{dip,s}} \sim 13 \, \mu\text{K}$. The self-generated dipole potential wells are therefore approximately 50 times shallower than the imposed pump-pump potential wells. Since atoms in the short-wavelength gratings oscillate nearly along $\hat{z}$, I expect the temperature of atoms in these gratings to be close to the temperature $T_z = 2 - 3 \, \mu\text{K}$ for atoms in the pump-pump grating [Greenberg et al. (2011)], which will be verified in Sec. 5.3.2. Thus, $\zeta_s = U_{\text{dip,s}}/T_s > 1$, and it makes intuitive sense that atoms can spatially bunch into these self-generated dipole potentials.

The best fit for Fig. 5.2(a) also indicates that there is another low-frequency resonance at $\omega_{\text{mod}}/2\pi = 177 \pm 15 \, \text{kHz}$, which I attribute to the dipole potential generated by the pump beam reshaping effect discussed in Sec. 4.4. This resonance is discussed further in Sec. 5.1.2.
**Long-wavelength gratings**

To parametrically drive atoms in the long-period gratings, I apply a weak elliptically-shaped optical field to the side of the atomic cloud (along $-\hat{r}$) of intensity $\bar{I} = 2 \times 10^{-3}$, length $\sim 4 \text{ cm}$, and width $\sim 1.2 \text{ mm}$. I then periodically modulate its amplitude by modulating the output of an acousto-optic modulator (see App. A), though which this beam is sent before being shaped using an elliptical telescope. This periodic modulation of the beam’s power invokes a periodic perturbation to the dipole potential. To detect the long-period, low-frequency parametric resonances, I operate in the steady-state regime, as described in Sec. 4.4.2.

As shown in Fig. 5.2(b), I observe a parametric resonance when modulating the amplitude at a frequency $\omega_{\text{mod}}/2\pi = 134 \pm 2 \text{ Hz}$ (width $50 \pm 7 \text{ Hz}$), which agrees with the expected value of $\omega_{\text{mod}}/2\pi = 191 (+78/-62) \text{ Hz}$ (to be discussed in in Sec. 5.1.2). The observation of this resonance indicates that I observe self-organization on a second, vastly different spatial scale, where the long-wavelength gratings are separated by orders of magnitude more than the optical wavelength.

The self-generated dipole potential for the long-wavelength gratings is $U_{\text{dip,} \ell} = 8 \pm 4 \mu\text{K}$. Because the long-wavelength grating wavevector $\hat{g}_\ell \approx \hat{r}$, one might expect $T_\ell \approx T_{\text{rad}} = 30 \mu\text{K}$, which would imply $\zeta_\ell < 1$ and negligible bunching. However, as I show in Sec. 5.3.2, the interaction between the pattern-forming optical fields and the atoms gives rise to cooling along $\hat{g}_\ell$ as well, which gives rise to $\zeta_\ell > 1$ and allows self-organization of atoms into the long-wavelength gratings.

For atoms in both the short- and long-wavelength gratings, the location of the parametric resonance is a function of the index of refraction, i.e., it increases with increasing pump intensity and vice-versa. Overall, I only work well above threshold where I have good signal-to-noise in measuring the power in the pattern-forming optical fields. For $\bar{I} = 8 \times 10^{-2}$ to 0.2, I observe parametric resonance for the short-
wavelength grating between 90 and 133 kHz and for the long-wavelength grating between 11 and 134 Hz.

5.1.2 Predicting the resonances

To assign the parametric resonances in Fig. 5.2 to self-organized structures, I calculate the expected value of the resonance using the intensity that generates the dipole potentials. The dipole potential energy

$$U_0 = \frac{\hbar \Delta I}{I_{sat}(1 + (2\Delta/\Gamma)^2)}$$

is derived for multi-level atoms in Sec. 7.2.2. The intensity of a single pump field is $I_p \sim 32 \pm 7$ mW/cm$^2$, where the error is due to the measured beam size $(100 \pm 15 \text{ \textmu m})$ after the beam reshaping effect discussed in Sec. 4.4. This measurement error is due to the error in the Gaussian fit on the camera, where the confidence interval is only to within one pixel size $[(16 \text{ \textmu m})^2]$, and to the confidence in the characterization of the imaging system in relating the far-field image size to the size of the beam at the cloud of atoms. This value of $I_p$ accounts for the reduction in pump power from 11.2 $\mu$W to 10 $\mu$W that results from the pump-beam reshaping, i.e., I take the pump-pump resonance to arise only due to the strong central pump spot, and I calculate the potentials that arise due to the ring-beam from the pump beam reshaping separately.

I calculate the intensity of a single generated field by measuring the beam size in the far field and predicting its near-field size based on a calibration of our imaging system. (See App. A.) I find the output intensity of a single generated field is $\sim 0.017 \pm 0.006$ mW/cm$^2$, where the error is due to a slight asymmetry in size between the two spots in the optical pattern. However, because the wave-mixing process gives rise to an exponential increase in the generated field intensity across the length of the atomic cloud, the output intensity is not a good representation of the generated field intensity inside the atomic cloud, where self-organization occurs. Thus, I use the
approximate intensity at the center of the cloud of atoms to predict the parametric resonance. Assuming an exponential increase and a cloud length of $L = 3$ cm, the intensity at $L = 1.5$ cm is 18% of the output intensity, or $I_g = (3 \pm 1) \times 10^{-3}$ mW/cm$^2$.

To predict the parametric resonances, I use these intensities in $U_{dip,(p,s,\ell)} = U_0$ from Eq. 5.2 with the resonant saturation intensity $I_{\text{sat}} = 1.3$ mW/cm$^2$ and the intensities $I$ defined below. For this experiment, Eq. 5.2 is also modified from that which is derived in Sec. 7.2.2 to assume that the atoms only pump the stretched-state transitions. This assumption is valid well above threshold, where I operate these experiments, when the atoms are tightly bunched in regions of pure $\hat{\sigma}^\pm$ polarizations. With this assumption, the Clebsch-Gordon coefficient parameter contained in the saturation intensity of Eq. 7.21 becomes $C^2 \rightarrow 1$, and the intensities are as follows. For the pump-pump (imposed) gratings, $I = I_p$. For the self-generated gratings, $I = 2\sqrt{T_p\sqrt{T_g}}$, where the factor of 2 accounts for the fact that there exist two sets of self-generated gratings everywhere; for example, the dipole potentials formed by the interference between $F(z)e^{ikz}$ and $b_+(z,r)e^{ik(-z\cos\theta-r\sin\theta)}$ and those between $B(z)e^{-ikz}$ and $f_-(z,r)e^{ik(z\cos\theta+r\sin\theta)}$ overlap spatially.

From this analysis, I predict the parametric resonances $2\omega_{\text{vib},s} = 686 \ (+123/ -109)$ kHz, $2\omega_{\text{vib},s} = 95 \ (+12/ -9)$ kHz, and $2\omega_{\text{vib},\ell} = 191 \ (+78/ -62)$ Hz, which agree with the measured values defined above. The larger error range for the long-period resonance is due to the error in $\theta = 4 \pm 1$ mrad, which again arises due to the confidence interval in the characterization of the imaging system.

I attribute the additional resonance observed at $\omega_{\text{mod}}/2\pi = 177 \pm 15$ kHz (width $213 \pm 34$ kHz) in Fig. 5.2(a) to the dipole potential that arises due to the interference of a pump beam with a nearly counterpropagating ring field, which arises from the beam reshaping effect. (See Sec. 4.4.) I model this ring using an LG$_{10}$ mode, which has a peak intensity of $\sim 36\%$ of the total intensity. Thus, because the ring contains
\( \sim 10\% \) of the power contained in the central pump spot, I approximate the peak intensity of the ring to be \( 0.1 \times 0.36 = 3.6\% \) of the pump intensity. Thus, using the same methods as above with \( I = \sqrt{0.036I_p} \sqrt{I_p} \), I predict a parametric resonance at \( 172 (+38/−27) \) kHz due to this beam reshaping effect, which agrees with the measured value of \( 177 ± 15 \) kHz. I neglect the dipole potentials due to a ring and a nearly copropagating pump field because the lattice spacing of these gratings is \( \sim 250 \mu \text{m} \) with a grating wavevector direction nearly along \( \hat{r} \), which implies that there only exist 0 or 1 such gratings inside the width of the cloud of atoms.

### 5.2 Bragg scattering

While parametric resonances provide information about the dipole potential well depth of the self-generated gratings, Bragg scattering has been shown to provide information about both the spatial coherences of lattice structure [Miyake et al. (2011)] as well as temperature information about the atoms trapped in the lattice [Mitsunaga et al. (1998)]. I use Bragg scattering to extract an atomic temperature along \( \hat{g}_s \) and \( \hat{g}_\ell \), where the sensitivity of the Bragg angle allows me to distinguish between the short- and long-wavelength gratings. Measuring the atomic temperature in the self-organized gratings is useful for ensuring that \( \zeta_{s,\ell} > 1 \), which gives rise to bunching in the self-generated gratings, and for use in theoretical predictions, which I describe in Ch. 7.

In the Bragg scattering experiment, I apply the pump beams for 200 \( \mu \text{s} \) in order to allow the optical/atomic patterns to form and reach the peak power in the generated fields/maximum bunching. I place an aperture so that I only collect light from a fraction of one of the emission cones, as depicted in Fig. 4.1(b). I then shut off the pump fields and inject a weak probe beam along either the \( ±\hat{z}\)-direction. To distinguish between the short- and long-wavelength gratings, I use the probe geometries shown in Fig. 5.3. A probe traveling along \( +\hat{z} (-\hat{z}) \) will only reach the
Figure 5.3: **Bragg scattering probe geometries.** The position of the detector
(black semi-circle) is shown with (a) short-wavelength gratings, which will scatter a
probe traveling along $-\hat{z}$ into the direction of the detector, and (b) long-wavelength
gratings, which will scatter a probe traveling along $+\hat{z}$ into the direction of the
detector.

During this experiment, I also apply a weak (0.3 G) external magnetic field to
oppose the Earth’s magnetic field and minimize Larmor precession, which otherwise
gives rise to oscillations in the decay signal. This will be discussed further in
Sec. 5.2.2. External magnetic fields only play a role for this experiment where the
optical lattice is absent; I find that magnetic fields do not inhibit nor enhance pattern
formation, during which the dipole potential is the dominant energy scale.

Typical results of the Bragg-scattering experiment are shown in Fig. 5.4. In this
experiment, the pump beams are shut off at time $t = -65$ ns, and the probe beam
is turned on at $t = 0$. Figure 5.4(a)/(b) shows the result from a probe beam applied
in the $-\hat{z}/+\hat{z}$-direction, thus scattering off the short-/long-wavelength gratings.

The initial scattering amplitude at time 0, which is proportional to the strength
of the Bragg grating [Agrawal (2008)], is approximately two times higher for the
short-wavelength gratings *c.f.* the long-wavelength gratings. This implies that the
short-wavelength gratings contain approximately twice as many atoms as the long-
wavelength gratings. With the observation (above) that the short-wavelength dipole
potentials are approximately two times deeper than the long-wavelength potentials,
this is consistent if atoms in both gratings are the same temperature, which I show
below is indeed the case.

Once the pump beams are shut off, the atoms expand ballistically out of their
Figure 5.4: **Bragg-scattered signals.** The power in the scattered probe beam as a function of time. Here, the pump beams ($\bar{I} = 0.17$) are on from $-200 \mu s \leq t \leq -65$ ns so that patterns form. A probe beam is turned on at $t = 0$ in the (a) $-\hat{z}$-direction and (b) $+\hat{z}$-direction. The red (dashed) curve provides a best fit to a Gaussian decay, where (a) $\tau_s = 0.91 \mu s$ and (b) $\tau_l = 120 \mu s$. The rectangular error bars at (a) $t = 0.5 \mu s$ and (b) $t = 100 \mu s$ indicate the typical confidence interval of the fit function at any given time.

gratings, which gives rise to a decay in the power of the Bragg-scattered signal. The short-wavelength gratings typically decay over $\tau_s = 0.8 - 1.4 \mu s$, and the long-wavelength gratings decay over $\tau_l = 100 - 170 \mu s$. These decay timescales correspond to the time it takes for the nonlinear refractive index to reduce to $1/e$ of its initial value as a result of ballistic expansion. Because $d_l \gg d_s$, the peak density in the long-wavelength gratings decays more slowly, which gives rise to the vastly different timescales $\tau_l \gg \tau_s$.

5.2.1 **Characterization of Bragg-scattered probe**

To optimize the power in the Bragg-scattered probe, it is useful to enhance the reflectivity of the Bragg gratings and to use a probe frequency that is degenerate with the pump beam frequencies. I enhance the reflectivity of the Bragg scattered gratings by increasing the pump intensities, which increases the imposed and self-generated dipole potential well depths. I show in Fig. 5.5 the dependence of the

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1 I have also verified that slow light effects in this regime delay a pulse of light by less than 200 ns and are therefore not responsible for decays on these timescales.
Figure 5.5: Efficiency of Bragg scattering. The maximum power in the Bragg scattered beam as a function of the pump intensity. The error bars represent the statistical deviation for three experiments. I quote power here rather than intensity because the decay of the probe is faster than the inverse frame rate of my camera, and it is difficult to capture the spatial distribution of the scattered probe.

scattered probe power as a function of pump intensity, which verifies that the grating reflectivity is enhanced with higher pump powers.

I also repeat these Bragg scattering experiments using probe beams of different frequencies relative to the pump frequency at $\Delta/2\pi = -25$ MHz. Figure 5.6 shows that the scattered power decreases for larger deviations from the pump frequency, which detunes the probe from the optimal Bragg condition [Agrawal (2008)]. The dip around $\Delta/2\pi = 0$ is due to absorption.

Non-diffuse scattering

To verify that the scattered signal in Fig. 5.4 arises due to Bragg scattering and not simply diffuse scattering of the probe in all directions, I set up an experiment to measure the spatial dependence of the power in the scattered probe. Because the probe scatters off the gratings formed by the interference between the generated fields and the pump fields, the probe should only scatter into the direction in which a generated fields propagates. If it were to scatter in other directions, that would
Figure 5.6: **Bandwidth of Bragg scattering.** The power in the Bragg scattered beam normalized by the input probe power as a function of the detuning of the probe beam. The pump beam detuning is $\Delta/2\pi = -25$ MHz.

indicate that the probe is undergoing diffuse scattering rather than Bragg scattering.

I perform this Bragg scattering experiment as described above with a stationary two-spot optical pattern. I place an aperture between the atoms and the detector, as shown in the inset of Fig. 5.7, so that I collect only a large portion of the generated field (red), a partial portion of a generated field (green), or part of the cone of emission into which a generated field does not propagate (blue). As shown in Fig. 5.7, the decreasing amplitude in scattered power for these respective aperture placements indicates that the probe is indeed undergoing Bragg scattering rather than diffuse scattering.

### 5.2.2 Larmor precession

The Bragg scattering experiments also suffer from higher sensitivity to external magnetic fields because once the lattice is removed, the dipole potential is no longer the dominant energy scale, and internal-state Zeeman effects play a larger role. An example of the effects of external magnetic fields on the Bragg-scattered signal is shown
in the blue curve of Fig. 5.8(a), where the oscillations arise due to Larmor precession of the atoms. The frequency of the oscillations in this experiment are \(~156\) kHz, which corresponds to an average field of \(0.22\) G for \(^{87}\text{Rb}\), where the \(F = 2\) ground state has a Zeeman splitting \(0.70\) MHz/G, which is close to the Earth field.

I use a gaussmeter (LakeShore Cryotronics, Model 460 with 3-axis probe) to measure the strength of the background field near the vacuum chamber. The external field (due to the Earth and any stray magnetization of the optical table) is \(\vec{B}_{\text{ext}} \approx (0.3 \text{ G})(\hat{z}) + (0.1 \text{ G})(-\hat{y})\), where \((-\hat{y})\) is into the direction of the floor of the laboratory. I place Helmholtz coils around the vacuum chamber to oppose the direction of the external magnetic fields. When I apply a 0.3 G field along \(-\hat{z}\) to oppose the external field, I observe the gold curve in Fig. 5.8(a), where the oscillations due to Larmor precession are largely suppressed. I typically did not apply a field along \(+\hat{y}\) to oppose that component because it plays a smaller role, and applying a field in
that direction moved the location of the trap.

In addition, the power of the Bragg-scattered signal is enhanced at time 0 in the case where the external magnetic fields are suppressed. This indicates that Larmor precession is detrimental to the Bragg-scattered signal. I thus apply an external magnetic field along $\hat{z}$ to cancel the background field component when I use the Bragg scattering data to extract a temperature, as described in Sec. 5.3.

I also find that, when I apply this external field to oppose the background, the oscillations that I typically see in the temporal signal of the pattern-forming optical fields are suppressed, as shown in Fig. 5.8(b). However, this external field does not affect the temporal persistence of the wave-mixing process nor the threshold for pattern formation.

5.3 Three-dimensional Sisyphus cooling

To measure the temperature of atoms along $\hat{g}_s$ and $\hat{g}_t$, I use a heuristic model to relate the decay timescale of the Bragg scattered signal to an atomic temperature. In this section, I describe this heuristic model and apply it to the Bragg scattering data.
discussed in Sec. 5.2. Based on these results, I then show that the atoms undergo spontaneous Sisyphus cooling in the radial direction, resulting in far-sub-Doppler-cooled temperatures in 3D.

5.3.1 Heuristic model of the atomic temperature

The temporal decay of the Bragg-scattered signal results from ballistic expansion of atoms out of their gratings. The atoms move ballistically, rather than diffusively, because the mean free path is much longer than the distance scales of relevance for atoms in a given experiment.

The mean free path is \( (8\pi a^2 n'_a)^{-1} \). Here, \( n'_a \) is the peak density of bunched atoms in a given dipole potential well, and \( a \) is the characteristic scattering length, where \( 90a_0 \leq a \leq 106a_0 \) for \(^{87}\text{Rb} \) with \( a_0 \) the Bohr radius [Pethick and Smith (2008)]. I take \( n'_a = 5 \times 10^{10} \) atoms/cm\(^3\) for atoms in the pump-pump gratings, where the typical average density is \( n_a \approx 10^{10} \) atoms/cm\(^3\). The mean free path is therefore \( \approx 23 \) mm, which is much longer than \( d_p \approx d_s \approx 195 \) nm or \( d_\ell \approx 80 \) \( \mu \)m. Therefore, once released from the lattice, the atoms expand ballistically.

Ballistic expansion is described by a Gaussian-type decay \( \propto \exp(-t^2/\tau_j^2) \) [Mitsunaga et al. (1998)]. The characteristic decay constant \( \tau_j \) depends on the atomic temperature \( T_j \), where \( j \in \{ p, s, \ell \} \) denotes atoms in the pump-pump, short-wavelength, or long-wavelength gratings, respectively. The decay constant \( \tau_j \) defines the time it takes for the peak density in a particular grating to shrink by a factor of \( 1/e \).

The density distribution of the short-period gratings before releasing the lattice is approximately

\[
\eta_s(z, r) \approx \eta_s(z) \approx \frac{n_a}{2I_0(-\zeta_s)} \left[ e^{-\zeta_s \cos(2k'_w z)} + e^{-\zeta_s \cos(2k'_w z - \pi)} \right]. \tag{5.3}
\]

To model this density distribution before ballistic expansion, I use the measured value of \( U_{\text{dip},s} \) from the parametric resonance experiments in Sec. 5.1.1 to calculate \( \zeta_s \). I
Figure 5.9: Density distribution before and after ballistic expansion. The blue (solid) curve shows the density distribution $\eta_s(z)$ before ballistic expansion for example parameters $I/I_s\Delta = 0.01$, $\Delta/\Gamma = -5$, and $T/T_D = 3/146$. The green (dashed) curve shows the density distribution $\eta'_s(z)$ at a time $\tau_s$ after ballistic expansion begins, where the peak density is reduced by $1/e$.

Note that $k'_w \gtrsim k'$ because the pattern-forming optical fields experience a different index of refraction according to weak-wave retardation. (See Sec. 3.2.) However, it is a good approximation to take $k'_w \approx k'$ for the relevant distance scales in this problem.

To model the density distribution after ballistic expansion, when the lattice is no longer present, I consider a periodic distribution weakly modulated about the average density, i.e., $\eta'_s(z) = n_a \left[ 1 + f(t)\cos(4k'z) \right]$, where $f < 1$. The constraint $f < 1$ is valid in this experiment after ballistic expansion where the peak density has reduced by a factor of $1/e$. This constraint also maintains normalization in $\eta'_s(z)$. I calculate the magnitude of $|f(t = \tau_s)|$ such that the peak density of $\eta'_s(z)$ is $1/e$ that of $\eta_s(z)$. Figure 5.9 shows example density distributions at $t = 0$ (blue, solid) and $t = \tau_s$ (green, dashed).

To apply these density distributions to a temperature model, I fit the “before” ($t = 0$) and “after” ($t = \tau_s$) density distributions between $-\lambda'/8 \leqslant z \leqslant \lambda'/8$ to Gaussian envelopes. These fits provide the effective widths of the grating, which I take to be the characteristic distance of travel $d$ for an atom during ballistic expansion over this timescale. I use this characteristic distance in a simple kinematics model.
Heuristic model of the atomic temperature. The predicted decay constant $\tau_{(s,\ell)}$ as a function of $T_{(s,\ell)}$ for example parameters $\tilde{\Delta} = -5$ and $I = 0.7 \text{ mW/cm}^2$.

of $v_0 = d/\tau_s$, where $v_0 = \sqrt{3k_B T/m}$. I keep $T_s$ as an unknown parameter in $\zeta_s$ and find its numerical solution as a function of $\tau_s$. For example parameters $\tilde{\Delta} = -5$ and $I/I_{s\Delta} = 0.01$, I show this predicted relationship in Fig. 5.10(a). I also use the same procedure to relate $\tau_l$ and $T_l$ for atoms in the long-period gratings. These results are shown in Fig. 5.10(b).

5.3.2 Extracting the temperature

From these relationships, I extract $T_{(s,\ell)}$ from the experimentally-measured temporal decay constants of the Bragg-scattered signals in Fig. 5.4. I performed multiple Bragg scattering experiments to extract $T_{(s,\ell)}$ from the decay constants for various initial pump intensities. I show these results in Fig. 5.11 for atoms in both the short- and long-wavelength gratings. The atomic temperature along $\hat{g}_s$ for atoms in the short-wavelength gratings is $T_s \approx 1.8 - 2.8 \mu$K for all pump intensities. These low temperatures are unsurprising because the short-wavelength gratings overlap spatially to within 1% of the pump-pump gratings, and atoms in these gratings have the advantage of undergoing early Sisyphus cooling due to the applied pump fields. The slight upward trend is likely the result of the fact that Sisyphus cooling operates optimally at a critical intensity defined by the relation $kv_c = 1/\tau_p$, where
The atomic temperature along (a) $\hat{g}_s$ and (b) $\hat{g}_\ell$ extracted from the heuristic model for ballistic expansion as a function of different (pre-expansion) pump intensities.

$\tau_p = 9 I_{\text{sat}} / (2 \Gamma I)$ and $v_c$ is the “critical” speed at which an atom undergoes the most efficient Sisyphus cooling, i.e., when it moves a distance $\lambda / 4$ in a pumping time $\tau_p$ [Castin et al. (1991); Dalibard and Cohen-Tannoudji (1989)]. Higher intensities give rise to higher scattering rates that limit the efficiency of the cooling process.

Figure 5.11(b) shows that atoms in the long-wavelength gratings have also cooled to $T_l \approx 1.9 \mu K$ despite having a momentum state nearly along $\hat{r}$. This result indicates that the atomic temperature along $\hat{g}_\ell$ has cooled by an order of magnitude from the initial temperature in the MOT of $T_{\text{rad}} = 30 \mu K$.

Such transverse cooling is known to arise due to a weak transverse dipole force in a 1D optical lattice [Blatt et al. (2009)]. Under the influence of this weak dipole force, it would take approximately 600 $\mu s$ for the atoms to move a distance $d_l$. However, this force alone cannot be responsible for our observed temperatures, because I observe the signature of these long-wavelength gratings as soon as 20 $\mu s$ after turning on the pump beams; i.e., I observe $\zeta_\ell > 1$ just 20 $\mu s$ after turning on the pump beams, as shown in Fig. 5.12.

In contrast, Sisyphus cooling is the only mechanism that can cool and trap atoms so quickly. The expected damping time for Sisyphus cooling due to the interaction
Figure 5.12: **Scattering off the long-wavelength gratings.** The Bragg-scattered signal off the long-wavelength gratings in the cases where the pump beams were on for 200 µs (blue), 50 µs (purple), and 20 µs (green). One can see that the long-wavelength gratings have already begun to form even in the case where the pump beams are only applied for 20 µs. The oscillations near $t = 0$ are residual effects due to Larmor precession, discussed in Sec. 5.2.2, where in this experiment, there is no externally applied field along $\hat{z}$.

The oscillations near $t = 0$ are residual effects due to Larmor precession, discussed in Sec. 5.2.2, where in this experiment, there is no externally applied field along $\hat{z}$.

of a pump beam and a generated field is [Dalibard and Cohen-Tannoudji (1989)]

$$t_d = \left(\frac{2\Gamma I}{9I_{\text{sat}}}\right)^{-1} \approx 10 \, \mu\text{s}. \quad (5.4)$$

Therefore, despite only applying optical fields only in a 1D lattice configuration, I observe spontaneous 3D Sisyphus cooling, which cools atoms to far-sub-Doppler temperatures within $\sim 20 \, \mu\text{s}$.

This represents the first observation of spontaneous 3D Sisyphus cooling and is one of the main results of my research. This cooling effect allows me to observe pattern formation in the transient regime for up to 2.4 ms, as shown in Fig. 4.7. This is comparable to the work in Refs. [Greenberg and Gauthier (2012b); Greenberg (2012)], where the wave-mixing process typically only persists for $200 - 400 \, \mu\text{s}$ due to the absence of cooling in the direction orthogonal to the applied pumps, causing atoms to expand and the wave-mixing process to decay more quickly. Thus, spontaneous 3D cooling gives rise to longer coherence times in this experiment.
5.4 Conclusions

In conclusion, I have performed the first observation of the self-organization of atoms in a real-space, multimode geometry and of spontaneous 3D Sisyphus cooling, which are two major results of my research described in rows 2 and 3 of Table 1.1. In this chapter, I characterize the properties of atoms in the self-organized gratings by using parametric resonance and Bragg scattering techniques. I describe my heuristic model for relating the temporal decay of the Bragg scattered signal to an atomic temperature. I show that for all gratings, the dipole potential energy exceeds the thermal energy of the atoms, which allows for real-space bunching into the self-generated gratings.

In the following chapters, I develop a self-consistent model for pattern formation, which shows how I achieve sufficiently strong light-atom interactions in free space to observe the phenomenon of self-organization. I begin by describing bunching in a 1D applied optical lattice, which provides the framework for understanding how atomic bunching gives rise to enhanced light-atom interaction strengths even in free space. In Ch. 7, I apply this model to a full theoretical characterization of pattern formation in cold atoms, which incorporates atomic bunching into the self-organized structures.
Transverse optical pattern formation has been studied theoretically in both warm atoms [Silberberg and Bar-Joseph (1982); Firth et al. (1990)] and cold atoms [Muradyan et al. (2005); Tesio et al. (2014)]. In general, these techniques consider counterpropagating optical fields incident on a sample of atoms. They show that for a threshold nonlinear refractive index, transverse perturbations can generate macroscopic optical fields that form multi-spot optical patterns.

The first theoretical description of transverse pattern formation in cold atoms [Muradyan et al. (2005)] considers two-level atoms and is restricted to the regime of weak atomic bunching. A more recent description does not account for the formation of real-space gratings of atoms and also does not account for the synergistic interaction between the atoms and the optical fields [Tesio et al. (2014)]. To describe pattern formation in my system, I require a theoretical description that allows for tight atomic bunching and accounts for strong, synergistic light-atom interactions.

To develop a model for pattern formation, I begin by introducing a theoretical description of atoms in a 1D lattice. This theoretical description is unique in that it is
both valid for both weakly- and tightly-bunched atoms and accounts for the synergistic interaction of the atoms with the lattice-forming optical fields. Existing models for similar schemes either assume the light-atom interaction to be weak [Asbóth et al. (2005); Petrosyan (2007); Wu et al. (2008); Nunn et al. (2010); Schilke et al. (2012b)], which is valid in the far-detuned regime, or they account for the light-atom synergy but approximate the atoms to be infinitely thin sheets of dielectric material [Deutsch et al. (1995); Asbóth et al. (2007, 2008)]. In contrast, the model that I describe in this chapter both accounts for finite-temperature effects and describes the regime of strong light-atom interactions. As a result, this model provides a connection between the zero-temperature models of the optomechanical physics community [Deutsch et al. (1995); Asbóth et al. (2008)] and the finite-temperature models of the nonlinear optics community [Muradyan et al. (2005); Saffman and Wang (2008); Greenberg and Gauthier (2012b)].

While the model I describe in this chapter does not describe pattern formation, it provides a self-consistent description of the refractive index of atoms driven by counterpropagating optical fields. This model is highly simplified from my experimental conditions; i.e., it describes two-level atoms at thermal equilibrium in a linear 1D optical lattice. In my experiment, I use a linear 1D optical lattice, which requires a multi-level-atom description, and the atoms undergo cooling and are thus not at thermal equilibrium.

Despite these simplifications, I devote an entire chapter to this model because it provides rich insights into the effects of atomic bunching on the light-atom interaction strength. These insights are necessary for understanding how I study nonlinear optical effects at low light levels and achieve strong light-atom interactions in free space, which ultimately give rise to my observation of multimode self-organization. The methods that I present in this chapter for enhancing the light-atom interaction strength have been proven to work experimentally [Greenberg and Gauthier
(2012b)], but this model provides a formalism for predicting the magnitude of the enhancement of the nonlinear refractive index achievable by using bunched, sub-Doppler-cooled atoms. This model is also an important stepping stone in developing a full theoretical description for pattern formation. In Ch. 7, I extend the model from this chapter to account for multi-level atoms in a linear optical lattice, and I show how atomic bunching in multiple spatial dimensions also enhances the light-atom interaction strength. This chapter is based largely on my previously published work [Schmittberger and Gauthier (2014)].

In Sec. 6.1, I motivate the development of this simplified model and summarize the insights it provides into light-atom interactions. Sec. 6.2, I provide an overview of the theoretical model and calculate the coupled amplitude equations for the lattice-forming optical fields. In Sec. 6.3, I analyze the results of the coupled amplitude equations and derive expressions for the light-atom interaction strength in different regimes of atomic bunching. In Sec. 6.4, I analyze how different types of optical nonlinearities interfere with one another and enhance or inhibit the light-atom interaction strength.

6.1 Introduction: Enhancing light-atom interaction strengths

The ability to enhance the response of a nonlinear optical material to incident optical fields is a broad goal in both classical and quantum optics. On the classical side, enhanced light-atom interaction strengths allow one to explore nonlinear optical effects at low light levels, which has applications in slow light [Harris and Hau (1999)] and reducing the threshold for all-optical switching using transverse optical patterns [Dawes et al. (2008)], for example. On the quantum side, one ultimate goal is to enhance light-atom interaction strengths to the point where one can realize nonlinear optical effects at the single-photon level, which has important applications in developing quantum logic gates and building quantum memories. Such single-photon nonlinear-
ities have recently been observed [Birnbaum et al. (2005); Tanji-Suzuki et al. (2011); Peyronel et al. (2012); Baur et al. (2014)], which represents substantial progress in the field of quantum information science.

In order to enhance light-atom interaction strengths, common techniques include placing atoms inside optical cavities or hollow fibers [Black et al. (2003); Bajcsy et al. (2009)], employing electromagnetically induced transparency (EIT) [Eisaman et al. (2005)], and using Rydberg blockade [Dudin and Kuzmich (2012); Parigi et al. (2012)]. In this chapter, I describe a different method for enhancing light-atom interaction strengths, which is applicable even for two-level atoms in free space. I show theoretically that by cooling atoms to sub-Doppler temperatures and allowing them to spatially bunch in an optical lattice, one can enhance the nonlinear susceptibility by more than two orders of magnitude relative to that which is achievable in a homogeneous gas.

I study three different parameter regimes according to the ratio of the dipole potential energy to the thermal energy of the atoms, as depicted in Fig. 6.1. In Regime I, there is weak or no atomic bunching because the thermal energy of the atoms greatly exceeds the dipole potential energy of the lattice. In Regime II, most of the atoms are trapped in the lattice because the dipole potential energy is on the order of the thermal energy of the atoms. In Regime III, the atoms tightly bunch in the lattice because the dipole potential energy greatly exceeds the thermal energy.

In addition, the model I present in this chapter provides insights into how atomic bunching can give rise to high-order nonlinear optical effects. I show that when there exists substantial atomic bunching, it is necessary to include fifth- or higher-order nonlinearities in order to accurately describe the light-atom interaction. The presence of these strong high-order optical nonlinearities enhances the light-atom interaction strength and allows for studies of nonlinear optics at low light levels.

I note that this model has certain restrictions. I neglect Doppler broadening,
so that this model is only valid for sub-Doppler temperatures. To describe a homogeneous gas (Regime I), this model is only valid for very low intensities, where the thermal energy of the atoms exceeds the dipole potential energy. I also do not account for Bose condensation in Regime III, where atom-atom correlations and quantum scattering are important [Pethick and Smith (2008)].

6.2 Theoretical model

I consider a gas of two-level atoms in a 1D optical lattice, which is described by the effective susceptibility from Eq. 2.27, reproduced here:

$$\chi_{\text{eff}}(z) = \frac{-6\pi}{k_{eg}^3} \eta(z) \frac{2\tilde{\Delta} - i}{1 + 4\Delta^2} \left( \frac{1}{1 + I(z)/I_{s\Delta}} \right), \quad (6.1)$$

where $\eta(z)$ is the density distribution of atoms and $\tilde{\Delta} = \Delta / \Gamma$. The total intensity is $I(z) = 2\varepsilon_0 c \langle \tilde{E}(z, t) \cdot \tilde{E}^*(z, t) \rangle_t$, where $\varepsilon_0$ is the permittivity of free space, $c$ is the speed of light in vacuum, and $\langle \rangle_t$ denotes a time average. I recall $I_{s\Delta} = I_{\text{sat}}(1 + 4\tilde{\Delta}^2)$ is the off-resonant saturation intensity. I limit this model to the case where $|\tilde{\Delta}| \gtrsim 3$, so that I consider only the real part of $\chi_{\text{eff}}(z)$ and hence neglect absorption. For the purposes of this model, I also consider the atoms to be in steady-state and thermal
equilibrium, and I assume they do not experience a net radiation pressure force. In the experiments described in this thesis, the atoms are not in thermal equilibrium because they undergo Sisyphus cooling. However, above the “décrochage” intensity (typically $I \approx I_{\text{sat}}$), where the majority of atoms have undergone cooling, it is a good approximation to describe the gas using a Maxwell-Boltzmann distribution characterized by the post-cooling temperature [Greenberg and Gauthier (2012a); Greenberg et al. (2011)]. I use this approximation here and in Ch. 7.

The factor $(1 + I(z)/I_{s\Delta})^{-1}$ in Eq. 6.1 corresponds to the saturable nonlinearity, which is the only nonlinearity that gives rise to optical pattern formation in warm atoms. In cold atoms, $\eta(z)$ depends on the intensity and gives rise to another type of nonlinearity: the bunching-induced nonlinearity.

I take the density distribution $\eta(z)$ to be the steady-state solution of the Fokker-Planck equation given in Eq. 2.55, which I rewrite here as

$$\eta(z) = n_a \tilde{\eta} \exp \left[ -U(z)/k_B T \right],$$

where $n_a$ is the average atomic density, $\tilde{\eta}$ is a normalization constant, $U(z)$ is the dipole potential, $k_B$ is Boltzmann’s constant, and $T$ is the atomic temperature. The dipole potential for a two-level atom in the low-intensity regime ($\langle I(z) \rangle /I_{\text{sat}} < 1$, where $\langle \rangle$ denotes the spatial average) reduces to the AC Stark Shift defined in Eq. 2.30, where for a two-level atom, I take $C_{ge}^2 = 1$. Thus, the ratio of the dipole potential energy to the thermal energy goes as

$$\frac{U(z)}{k_B T} \approx \frac{\Delta I(z)}{I_{s\Delta} T},$$

where $\tilde{T} = T/T_D$ is the temperature normalized by the Doppler temperature $T_D = \hbar \Gamma / 2k_B$.
6.2.1 The weak bunching limit

It is useful to first analyze a limiting case of Eq. 6.1, where in the weak-bunching, low-intensity limit, \( \max[U(z)]/k_B T \ll 1 \) and \( \max[I(z)]/I_{s\Delta} \ll 1 \), and Eq. 6.1 is approximately

\[
\chi_{\text{eff}}(z) \approx \chi_{\text{lin}} \left[ 1 - \left( \frac{\bar{\Delta} I(z) - \langle I(z) \rangle}{I_{s\Delta}} + \frac{I(z)}{I_{s\Delta}} \right) \right],
\]

(6.4)

which was first introduced in Eq. 4.1. The first term in Eq. 6.4 is the linear susceptibility. The second term in Eq. 6.4 corresponds to the bunching-induced nonlinearity, which is the dominant nonlinearity in the experiments described in this thesis, where \( \bar{T} \approx 3/146 \) and \( |\bar{\Delta}| = 3 - 10 \). In this term, the spatially independent part of the intensity is subtracted because only the spatially dependent part of the dipole potential energy contributes to the bunching-induced nonlinearity, as I show in Sec. 6.3.

The third term in Eq. 6.4 corresponds to the saturable (Kerr) nonlinearity.

While both the bunching-induced nonlinearity and the saturable nonlinearity scale with \( I(z)/I_{s\Delta} \), further examination of Eq. 6.4 shows that the bunching-induced nonlinearity may be made much larger than the saturable nonlinearity by using sub-Doppler temperatures. In Secs. 6.3 and 6.4, I move past the weak-bunching approximation (no longer assuming that \( \max[U(z)]/k_B T \ll 1 \)), and I calculate \( \chi_{\text{eff}} \) for a general degree of atomic bunching. I show that the bunching-induced nonlinearity gives rise to greatly enhanced light-atom interaction strengths for sub-Doppler-cooled atoms.

6.2.2 The coupled amplitude equations

I consider a one-dimensional optical lattice, as depicted in Fig. 6.1, whose total applied electric field is

\[
\vec{E}(z, t) = \vec{F}(z, t)e^{i(kz - \omega t)} + \vec{B}(z, t)e^{i(-kz - \omega t)} + \text{c.c.},
\]

(6.5)
where \( k \) is the wavenumber of the optical fields in vacuum. The periodicity of the density distribution of atoms in this optical lattice depends directly on the periodicity of the intensity distribution inside the atomic medium [Deutsch et al. (1995); Asbóth et al. (2008)]. I therefore define the density distribution \( \eta(z) \) via the Floquet expansion

\[
\eta(z) = n_a \sum_{j=-\infty}^{\infty} \tilde{\eta}_j(z)e^{j2\pi k z}.
\]

The coefficients \( \tilde{\eta}_j(z) \) have a slowly-varying position dependence that accounts for the synergistic interaction between the atoms and the optical fields; these coefficients are derived in Sec. 6.3. Many theoretical models assume the light-atom interaction is small and take the periodicity of the density distribution to be equal to that of the vacuum intensity distribution [Asbóth et al. (2005); Petrosyan (2007); Schilke et al. (2012b)]. This is a good approximation when the optical fields are far-detuned and the wavenumber of the optical fields while propagating through the atoms \( k' \approx k \).

By accounting for the spatial dependence of \( \tilde{\eta}_j(z) \), I account for the dependence of the wavevectors on the index of refraction \( (k' = nk) \). As a result, my model is self-consistent, and it accounts for strong light-atom interactions, where one can observe nonlinear optical effects at low light levels.

The light-atom interaction is described via the polarization \( \vec{P} = \epsilon_0 \chi_{\text{eff}}(z) \vec{E} \) using Eqs. 6.1 and 6.6 [Boyd (2008)]. I consider the low-intensity regime where \( \max[I(z)]/\bar{I}_{s\Delta} \ll 1 \), so that the Taylor expansion \( (1 + I(z)/\bar{I}_{s\Delta})^{-1} \approx (1 - I(z)/\bar{I}_{s\Delta}) \) is valid. For \( |\Delta| \gtrsim 3 \), this approximation requires only \( \max[I(z)]/I_{\text{sat}} \lesssim 1 \).

I solve the wave equation from Eq. 2.3 for the electric field in Eq. 6.5. I consider parallel optical field polarizations \( \langle \vec{F}(z) || \vec{B}(z) \rangle \) for simplicity in this chapter. In the experiments I describe in this thesis, I use a lin\perp lin polarization configuration, which requires a multi-level atom formalism, described in Ch. 7.

I make the rotating wave approximation, in which I average over a long time
c.f. $2\pi/\omega$ and thus select terms oscillating only near $\pm\omega$. I also assume steady-state (time-independent) electric field amplitudes. The wave equation then gives rise to the coupled amplitude equations

$$\frac{\partial F}{\partial z} = \frac{ik}{2} \chi_{\text{lin}} \left\{ \tilde{\eta}_0(z) - \frac{4\epsilon_0 c}{I_{s\Delta}} \left( |F|^2 + |B|^2 \right) \tilde{\eta}_0(z) + FB^* \tilde{\eta}_{-1}(z) + F^* B \tilde{\eta}_1(z) \right\} F +$$

$$\left[ \tilde{\eta}_1(z) e^{2ikz} - \frac{4\epsilon_0 c}{I_{s\Delta}} \left( |F|^2 + |B|^2 \right) \tilde{\eta}_1(z) e^{2ikz} + FB^* e^{2ikz} \tilde{\eta}_0(z) + F^* B e^{-2ikz} \tilde{\eta}_2(z) e^{4ikz} \right] B e^{-2ikz} \right\} (6.7)$$

and

$$\frac{\partial B}{\partial z} = -\frac{ik}{2} \chi_{\text{lin}} \left\{ \tilde{\eta}_0(z) - \frac{4\epsilon_0 c}{I_{s\Delta}} \left( |F|^2 + |B|^2 \right) \tilde{\eta}_0(z) + FB^* \tilde{\eta}_{-1}(z) + F^* B \tilde{\eta}_1(z) \right\} B +$$

$$\left[ \tilde{\eta}_{-1}(z) e^{-2ikz} - \frac{4\epsilon_0 c}{I_{s\Delta}} \left( |F|^2 + |B|^2 \right) \tilde{\eta}_{-1}(z) e^{-2ikz} +$$

$$FB^* e^{2ikz} \tilde{\eta}_{-2}(z) e^{-4ikz} + F^* B e^{-2ikz} \tilde{\eta}_0(z) \right] F e^{2ikz} \right\} (6.8)$$

where $F \equiv F(z)$, $B \equiv B(z)$. Here, I have only retained terms that are phase-matched or nearly phase-matched, i.e., that have a spatial variation close to $e^{ikz}$ in Eq. 6.7 or $e^{-ikz}$ in Eq. 6.8. In Eqs. 6.7 and 6.8, the first term in square brackets on the right-hand-side describes the dispersion of the optical fields as they propagate through the atomic medium, i.e., the self-phase shift. The other term represents the nonlinear optical coupling between the forward and backward fields.
6.3 Uniform optical lattice

I simplify Eqs. 6.7 and 6.8 by considering the case of equal-intensity counterpropagating fields. This special case suppresses the radiation pressure force so that the atoms only experience the dipole force that gives rise to atomic bunching.

Under these equal-intensity conditions, each optical field experiences the same effective susceptibility $\chi_{\text{eff}}$. Thus, each optical field has a wavevector magnitude $k' = nk$ inside the atomic medium, where $n \approx 1 + \chi_{\text{eff}}/2$ [Boyd (2008)]. Here, $\chi_{\text{eff}}$ is spatially independent and contains only those terms from Eq. 6.1 that are phase-matched to efficiently drive a self-phase shift and/or nonlinear coupling between the fields. The spatial dependence of the optical field amplitudes is then described by

$$F(z) = \tilde{F} e^{i k (\chi_{\text{eff}}/2) z} \quad \text{and} \quad B(z) = \tilde{B} e^{-i k (\chi_{\text{eff}}/2) z},$$

(6.9)

where $\tilde{F}$ and $\tilde{B}$ are independent of $z$. Because the dipole potential depends on the intensity distribution, I define

$$\eta_j(z) = n_a \tilde{\eta}_j e^{i 2ik (\chi_{\text{eff}}/2) z},$$

(6.10)

so that

$$\eta(z) = n_a \sum_{j=-\infty}^{\infty} \tilde{\eta}_j e^{i 2ik' z}.$$  

(6.11)

The coefficients $\tilde{\eta}_j$ are now independent of $z$ because the periodicity of the density distribution exactly equals the periodicity of the intensity distribution inside the atomic medium. I note $\eta_j = \eta_{-j}$, which is proven in App. B using the shift theorem.

For equal-intensity applied fields, $F(-L/2) = B(L/2)$ for a medium of length $L$. Solving Eqs. 6.7 and 6.8 under these conditions gives rise to the solution for the effective susceptibility

$$\chi_{\text{eff}} = \chi_{\text{lin}} \left[ \tilde{\eta}_0 + \tilde{\eta}_{\pm 1} - \frac{1}{2} (3\tilde{\eta}_0 + \tilde{\eta}_{\pm 1} + 3\tilde{\eta}_{\pm 1} + \tilde{\eta}_{\pm 2}) \right],$$

(6.12)
where \( \tilde{I} = \langle I(z) \rangle / I_{\Delta} \), and

\[
\tilde{\eta}_j = \frac{1}{\lambda'/2} \int_{-\lambda'/4}^{\lambda'/4} \eta(z) \frac{e^{-j2k'z}}{n_a} dz,
\]

(6.13)

where \( \lambda' = 2\pi/k' \).

In order to calculate the Fourier coefficients, it is important to note that in Eq. 6.2, the position-independent part of \( U(z) \) can be absorbed into a new normalization constant \( \tilde{\eta}' \). The position-independent part of the dipole potential does not contribute to the dipole force and thus does not give rise to atomic bunching. The Fourier coefficients are therefore described by

\[
\tilde{\eta}_j = \frac{\tilde{\eta}' k'}{\pi} \int_{-\pi/2k'}^{\pi/2k'} \exp \left[ -\zeta \cos(2k'z) \right] e^{-j2k'z} dz,
\]

(6.14)

where

\[
\zeta = \frac{\Delta \bar{I}}{I}.
\]

(6.15)

From Eq. 6.14,

\[
\tilde{\eta}_j = \tilde{\eta}' I_j(-\zeta),
\]

(6.16)

where \( I_j \) are modified Bessel functions of the first kind of order \( j \). The normalization constant \( \tilde{\eta}' \) can be calculated explicitly by integrating the density distribution over one period, \( i.e., \)

\[
\frac{\lambda'}{2} = \tilde{\eta}' \int_{-\pi/2k'}^{\pi/2k'} \exp \left[ -\zeta \cos(2k'z) \right] dz,
\]

(6.17)

which gives \( \lambda'/2 = \pi \tilde{\eta}' I_0(-\zeta)/k' \). Thus,

\[
\tilde{\eta}' = \frac{1}{I_0(-\zeta)}.
\]

(6.18)

Previous finite-temperature models do not explicitly determine this normalization constant; they either take it to be fixed by experimental parameters [Muradyan et al. 109]
Combining Eqs. 6.14 and 6.18, the Fourier coefficients are

\[ \tilde{\eta}_j = \frac{I_j(-\zeta)}{I_0(-\zeta)}. \] (6.19)

The first-order Fourier coefficient is a measure of the amount of atomic bunching. I therefore define the bunching parameter

\[ b = |\tilde{\eta}_1|, \] (6.20)

which can be used to distinguish the three bunching regimes depicted in Fig. 6.1. This is analogous to the bunching parameter used to describe other parametric oscillation processes in cold atoms [Bonifacio et al. (1994)]. In this case, however, I only consider the first-order Fourier coefficient because it is the only component that directly couples the forward and backward waves. The bunching parameter \( b \in [0, 1] \), where \( b = 0 \) describes a homogeneous gas and \( b = 1 \) corresponds to “perfect bunching,” or the case where the density distribution consists of infinitesimally thin sheets of atoms, which is the case considered in Ref. [Deutsch et al. (1995)].

Figure 6.2 shows the dependence of the bunching parameter \( b \) on \( |\zeta| \). Here, I define the regimes of atomic bunching from Fig. 6.1 using reasonable but arbitrary ranges of \( b \). Regime I, where the gas is effectively homogeneous, is defined by \( b < 0.2 \) \((|\zeta| < 0.4)\). Regime II describes the region where \( 0.2 \leq b \leq 0.8 \) \((0.4 \leq |\zeta| < 2.9)\), where the thermal energy of the atoms is the same order of magnitude as the dipole potential energy of the optical lattice. Regime III, where \( b > 0.8 \) \((|\zeta| > 2.9)\), corresponds to tight atomic spatial localization. This is attainable in a typical MOT [Birkl et al. (1995)]. For reference, based on the measurements presented in Sec. 4.1.1, I observe
the minimum threshold for pattern formation at $\eta^\pm_1 \simeq 0.7 (|\zeta| \simeq 2)$, where the atoms are nearing the tight-bunching regime.

Two examples of the normalized density distribution

$$
\eta(z) = \frac{n_a}{I_0 (-\zeta)} \exp \left[ \frac{U(z) - \langle U(z) \rangle}{k_B T} \right]
$$

are shown in Fig. 6.3 for two different bunching regimes. When using red- ($\tilde{\Delta} < 0$) versus blue- ($\tilde{\Delta} > 0$) detuned optical fields, the locations of density maxima are phase-shifted by $z = \lambda/4$ relative to one another. This is because atoms are attracted to the dipole potential minima, which correspond to the intensity maxima (minima) for red (blue) optical lattices. An example of Regime I is shown in Fig. 6.3(a), where the density distribution is only weakly modulated. Figure 6.3(b) shows the density distribution for Regime III, where the atoms are tightly bunched in the potential wells and the local density in each well exceeds the average density. In fact, it is close to and in Regime III where the first Bragg scattering experiments for atoms in optical lattices were performed: In Ref. [Weidemüller et al. (1995)], the authors worked with $b \simeq 0.7 (|\zeta| \simeq 2)$, and in Ref. [Birkl et al. (1995)], the authors used $b \simeq 0.93 (|\zeta| \simeq 7)$.
Figure 6.3: Example density distributions. Density distributions for (a) $b = 0.01$ ($\zeta = 0.2$) and (b) $b = 0.86$ ($\zeta = 4$). The red, solid curves show red detunings ($\Delta < 0$), and the blue, dashed curves show blue detunings ($\Delta > 0$).

From Eqs. 6.12 and 6.19, I determine the effective susceptibility experienced by the optical fields, which is the basis of the analysis presented in the remainder of this chapter. The effective susceptibility, which is one measure of the light-atom interaction strength, is

$$
\chi_{\text{eff}} = \chi_{\text{lin}} \left[ 1 + \frac{I_1(-\zeta)}{I_0(-\zeta)} - \frac{\tilde{I}}{2} \left( 3 + 4 \frac{I_1(-\zeta)}{I_0(-\zeta)} + \frac{I_2(-\zeta)}{I_0(-\zeta)} \right) \right], \quad (6.22)
$$

where the temperature-dependent terms correspond to the bunching-induced nonlinearity, and the other intensity-dependent terms correspond to the saturable nonlinearity. In the remainder of this chapter, I show that in certain parameter regimes, the bunching-induced nonlinearity gives rise to greatly enhanced light-atom interaction strengths. I also study how $\chi_{\text{eff}}$ varies with $\zeta$ in the different parameter regimes, so that one can use this model to determine how to maximize the light-atom interaction strength for a certain atomic temperature.

In Regime I, a Taylor expansion to first order in $\tilde{I}$ is valid, which simplifies the expression for the effective susceptibility to

$$
\frac{\chi_{\text{eff}}}{\chi_{\text{lin}}} \approx -\frac{\Delta}{|\Delta|} \left[ 1 - \frac{\Delta \tilde{I}}{2T} - \frac{3}{2} \tilde{I} \right] \quad \text{for } b < 0.2. \quad (6.23)
$$
The factor of 1/2 appearing in the nonlinear terms (c.f. Eq. 6.4) arises because only one term in the exponential form of the intensity distribution is phase-matched in the wave equation.

In the tight-bunching regime, a Taylor expansion is no longer valid. However, a good approximation of $\chi_{\text{eff}}$ for $b > 0.8$ is an asymptotic expansion, which gives rise to

$$\frac{\chi_{\text{eff}}}{|\chi_{\text{lin}}|} \approx \begin{cases} 
2 - \frac{T}{2|\Delta|I} - 4\tilde{I} + \frac{2\tilde{T}}{|\Delta|} & \text{if } \tilde{\Delta} < 0 \\
-\frac{\tilde{T}}{2|\Delta|I} - \frac{3\tilde{T}}{4|\Delta|} & \text{if } \tilde{\Delta} > 0.
\end{cases}$$

(6.24)

The behavior of $\chi_{\text{eff}}$ in these two example bunching regimes is plotted in Fig. 6.4 as a function of both $\tilde{I}$ and $b$ along with the Taylor and asymptotic expansions used in Eqs. 6.23 and 6.24. Figures 6.4(a) and (b) depict example curves that describe the effective susceptibility both with and without bunching. In order to describe the case of a homogeneous gas, I take $\tilde{T} = 1$ in Eq. 6.23. While this regime can still generate weak bunching, I only consider very low intensities in Fig. 6.4, where an atomic sample characterized by the Doppler temperature is essentially homogeneous.

Because the susceptibility is shown as a function of intensity, the slope of each curve in Fig. 6.4 depends directly on the third-order nonlinear optical susceptibility $\chi^{(3)}$. From Eq. 6.23, $\chi^{(3)}$ in Regime I is given by

$$\chi^{(3)} \approx \frac{\tilde{\Delta}}{|\Delta|} \frac{2\epsilon_0 c |\chi_{\text{lin}}|}{I_s\Delta} \left[ \frac{\tilde{\Delta}}{2\tilde{T}} + \frac{3}{2} \right] \text{ for } b < 0.2,$$

(6.25)

where the first term corresponds to the bunching-induced nonlinearity and the second corresponds to the saturable (Kerr) nonlinearity.

In the case of a homogeneous gas ($b \to 0$), $\chi^{(3)} = 3\epsilon_0 c |\chi_{\text{lin}}|\tilde{\Delta}/I_s\Delta|\tilde{\Delta}|$. Therefore, $\chi^{(3)} < 0$ ($\chi^{(3)} > 0$) for red (blue) detunings, which corresponds to a self-defocusing.
Figure 6.4: Effective susceptibility in different bunching regimes. The effective susceptibility normalized by the linear susceptibility as functions of $\tilde{I}$ and $b$ for $|\Delta| = 3$ in the (a) red-detuned case and (b) blue-detuned case. The solid curve is an example of sub-Doppler-cooled atoms, where I’ve taken $\tilde{T} = 3/146$, which corresponds to $T = 3 \mu K$ for rubidium where $T_D = 146 \mu K$. The long, dashed line is the case $\tilde{T} = 1$. The triangles represent the Taylor series expansion used to obtain Eq. 6.23, and the circles represent the asymptotic expansion used in Eq. 6.24. The vertical, dashed lines delineate the boundaries between Regimes I, II, and III.

(self-focusing) nonlinearity. These different types of nonlinearities have important implications for nonlinear optical processes. For example, transverse optical pattern formation can only occur when $\chi^{(3)} > 0$ [Chiao et al. (1966)]. Thus, our model predicts that, for a homogeneous gas, transverse optical pattern formation can only occur for blue detunings, which is consistent with experiments [Dawes et al. (2008)].

Despite the fact that homogeneous atomic samples give rise to strong light-atom interactions and, subsequently, low-light-level nonlinear optical effects [Schmidt and A. Imamoglu (1996); Dawes et al. (2008)], it is clear from Eq. 6.25 that the nonlinear susceptibility can be further enhanced by using sub-Doppler-cooled atoms. Using such low temperatures allows for atomic bunching in the optical lattice. The effect of bunching is apparent from the steep slopes in Fig. 6.4 for $b < 0.2$ for the sub-Doppler-cooled case. In fact, at an example detuning $|\Delta| = 5$, $\chi^{(3)}$ is more than two orders of magnitude larger for a gas of rubidium atoms at $\tilde{T} = 3 \mu K$ than in the
homogeneous case.

In Regime III, $\chi^{(3)}$ is only enhanced c.f. a homogeneous gas for red detunings. From Eq. 6.24, $\chi^{(3)}$ in the tight-bunching regime is

$$\chi^{(3)} \simeq \begin{cases} \frac{2\epsilon_0 c|\chi_{\text{lin}}|}{I_s \Delta} \left( \frac{T}{2|\Delta|I^2} - 4 \right) & \text{if } \tilde{\Delta} < 0 \\ \frac{2\epsilon_0 c|\chi_{\text{lin}}|}{I_s \Delta} \frac{T}{2|\Delta|I^2} & \text{if } \tilde{\Delta} > 0. \end{cases} \quad (6.26)$$

Here, $\chi^{(3)}$ is intensity-dependent because $\chi_{\text{eff}}$ contains high-order nonlinearities that cannot be neglected. This is discussed further in Sec. 6.4. For high intensities in this tight-bunching regime, Eq. 6.26 simplifies to $\chi^{(3)} \rightarrow -8\epsilon_0 c|\chi_{\text{lin}}|/I_s \Delta$ for red detunings and $\chi^{(3)} \rightarrow 0$ for blue detunings. In the high-intensity limit, $\chi^{(3)}$ is independent of the atomic temperature because once the atoms are tightly confined, the bunching-induced nonlinearity plays a less substantial role. In addition, for red detunings, $\chi^{(3)}$ is a factor of $8/3$ larger than in the homogeneous case, which arises because the atoms are tightly bunched at the intensity maxima. In a homogeneous gas, some atoms overlap spatially with the intensity zeroes and do not contribute to the light-atom interaction.

However, for blue detunings in the tight-bunching regime, the atoms spatially organize at the intensity zeroes. In the limit of perfect bunching ($b \rightarrow 1$), both $\chi_{\text{eff}}$ and $\chi^{(3)}$ approach zero, which simply describes the fact that when sitting at the intensity zeroes, the atoms cannot interact with the optical fields. This is supported by the high-intensity limit of Fig. 6.4(b), where bunching increases as the dipole potential well becomes deeper.

As we increase the intensity for red detunings, it is apparent in Fig. 6.4(a) that there is a local maximum in $\chi_{\text{eff}}$ in Regime III. From Eq. 6.26, this local maximum
This critical point represents the transition from a self-focusing to a self-defocusing nonlinearity for increasing intensities. This corresponds physically to the condition at which the saturable nonlinearity begins to dominate over the bunching-induced nonlinearity. In other words, once the atoms are tightly bunched, the effect of the bunching-induced nonlinearity effectively “saturates” because the external degrees of freedom are exhausted. At this point, the internal-state, saturable nonlinearity dominates the light-atom interaction.

However, for atoms in a blue optical lattice, there is no such critical point. In this case, increasing the amount of bunching only reduces the number of atoms that can interact with the optical fields. This agrees with many experiments that operate in the tight-bunching regime, which find that red-detuned optical lattices are more favorable for observing nonlinear optical processes [Greenberg and Gauthier (2012b); Greenberg et al. (2011); Gattobigio et al. (2006); Arnold et al. (2012); Deng et al. (2010)], including my own experiment, where I only observe pattern formation for red detunings.

In order to better understand the competing nature of the bunching-induced nonlinearity and the saturable nonlinearity that gives rise to this critical point, it is useful to investigate the constructive or destructive interference of these nonlinearities and how they enhance or inhibit the overall light-atom interaction strength, described by $\chi_{\text{eff}}$. 
6.4 Interference between competing nonlinearities

I decompose $\chi_{\text{eff}}$ into

$$\chi_{\text{eff}} = \chi_{\text{lin}} + \chi_{\text{bunching}} + \chi_{\text{SN}} + \chi_{\text{bunching+SN}},$$  \hspace{1cm} (6.28)

which define the contributions due to linear effects, the bunching-induced nonlinearity, the saturable nonlinearity, and the combined effects of these two nonlinearities, respectively. From Eq. 6.22, these are defined as

$$\chi_{\text{bunching}} = \chi_{\text{lin}} \frac{I_1(-\zeta)}{I_0(-\zeta)},$$  \hspace{1cm} (6.29)

$$\chi_{\text{SN}} = -\frac{3\chi_{\text{lin}} \bar{I}}{2},$$  \hspace{1cm} (6.30)

and

$$\chi_{\text{bunching+SN}} = -\chi_{\text{lin}} \frac{I}{2} \left[ 4 \frac{I_1(-\zeta)}{I_0(-\zeta)} + \frac{I_2(-\zeta)}{I_0(-\zeta)} \right].$$  \hspace{1cm} (6.31)

It is important to note that the numerical factor of 3 in $\chi_{\text{SN}}$ is absent in $\chi_{\text{bunching}}$. This factor corresponds to the spatially independent part of the intensity, which polarizes the atoms and contributes to the saturable nonlinearity, but does not contribute to the dipole force that gives rise to bunching.

Another important difference between $\chi_{\text{SN}}$ and $\chi_{\text{bunching}}$ is the characteristic timescale of each nonlinearity. The response time of $\chi_{\text{SN}}$ is approximately $1/\Gamma$ (e.g., 26 ns for rubidium). However, the response time of $\chi_{\text{bunching}}$ is much slower — approximately the time it takes for an atom to move a distance $\lambda'/2$. This timescale depends on the atomic temperature; e.g., it is $\sim 20 \mu$s for rubidium atoms at 3 $\mu$K. These vastly different timescales allow researchers to determine the dominant nonlinearity in experiments [Labeyrie et al. (2014)].

The linear susceptibility and the various nonlinear susceptibilities defined in Eqs. 6.29-6.31 are illustrated in Fig. 6.5 as a function of $\zeta$. In Figs. 6.5(a) and
Figure 6.5: Interference of linear and nonlinear susceptibilities. (a) $\chi_{\text{lin}}/\chi_{\text{lin}}$ (dashed line) and $\chi_{\text{bunching}}/\chi_{\text{lin}}$ (solid curve). (b) $\chi_{\text{SN}}/\chi_{\text{lin}}$ (dashed line) and $\chi_{\text{bunching+SN}}/\chi_{\text{lin}}$ (solid curve).

(b) for blue detunings ($\zeta > 0$), the contributions $\chi_{\text{lin}}$ and $\chi_{\text{bunching}}$ destructively interfere with one another, as do $\chi_{\text{bunching+SN}}$ and $\chi_{\text{SN}}$. This is consistent with the high-intensity limiting case described above, which results in a very small $\chi_{\text{eff}}$ for large $\zeta$.

For red detunings ($\zeta < 0$), $\chi_{\text{lin}}$ and $\chi_{\text{bunching}}$ interfere constructively, as do $\chi_{\text{SN}}$ and $\chi_{\text{bunching+SN}}$. However, each of these sets of terms have opposite signs and together reduce the overall $\chi_{\text{eff}}$. However, these destructively interfering processes have different dependences on the intensity, and thus the dominant contribution to $\chi_{\text{eff}}$ depends on whether one is below or above the critical point of Eq. 6.27.

In the limit $\tilde{T} \to 0$, the density distribution corresponds to that of infinitely thin sheets of atoms, which is the same distribution described in Refs. [Deutsch et al. (1995); Asbóth et al. (2008)]. These works show that the lattice constant is smaller than (identical to) the periodicity of the vacuum intensity distribution for red (blue) detunings, as shown in Fig. 6.6. Taking $\tilde{T} \to 0$ in Eq. 6.22, the wavevector in the
Figure 6.6: **Light-atom synergy in the zero-temperature limit.** Based on Ref. [Deutsch et al. (1995)]. Intensity distribution as a function of $d = \lambda'/2$ for a) in vacuum, b) with atoms (black circles) in a blue optical lattice, and c) with atoms (black circles) in a red optical lattice.

Medium $k' = k(1 + \chi_{\text{eff}}/2)$ becomes

$$k' = \begin{cases} k \left[ 1 + \chi_{\text{lin}} \left( 1 - 2\tilde{T} \right) \right] & \text{if } \tilde{\Delta} \leq 0 \\ k & \text{if } \tilde{\Delta} > 0. \end{cases} \quad (6.32)$$

Since the lattice constant goes as $d = \lambda'/2$, my model agrees with the results of the zero-temperature models [Deutsch et al. (1995); Asbóth et al. (2008)]. I note that I do not account for Bose condensation in this model, and the limit $\tilde{T} \to 0$ simply reproduces the density distributions considered in Refs. [Deutsch et al. (1995); Asbóth et al. (2008)].

It is important to note that $\chi_{\text{bunching}}$ and $\chi_{\text{bunching+SN}}$ contain high-order nonlinear contributions, *i.e.*, $\chi_{\text{bunching+SN}}$ is fifth-order at lowest-order. Such high-order nonlinearities are neglected in most nonlinear optical models that work in the low-intensity regime [Boyd (2008); Muradyan et al. (2005)]. However, these high-order nonlinearities are crucial for describing the tight-bunching regime. If I had not retained these terms, I would not have reproduced the results in the zero-temperature models of Refs. [Deutsch et al. (1995); Asbóth et al. (2008)]. These terms are necessary for con-
necting the zero-temperature models commonly used by the optomechanical physics community to the finite-temperature models used in the nonlinear optics community.

6.5 Summary

In this chapter, I show that atomic bunching at the intensity maxima of an optical lattice gives rise to an enhanced nonlinear optical response even at very low optical intensities. I find that the third-order nonlinear susceptibility can be made more than two orders of magnitude larger for sub-Doppler-cooled two-level atoms than for a homogeneous gas, which is consistent with experiments that work in this regime [Greenberg et al. (2011); Greenberg and Gauthier (2012b)].

This model also provides new insights into methods for enhancing the light-atom interaction strength. I find that atomic bunching gives rise to high-order nonlinearities that enhance the nonlinear refractive index, and allows studies of nonlinear optical effects at low light levels. By using tightly bunched atoms and small detunings to optimize the light-atom interaction strength, I observe transverse optical pattern formation at low required powers. It is also by using the methods presented in this chapter that I achieve strong light-atom coupling without the need for a cavity, which allows me to work in a naturally multimode free-space system and study multimode self-organization.

This model is useful for developing a description of pattern formation because it accounts for the synergistic interaction between the atoms and the lattice-forming optical fields, and it provides the first self-consistent theoretical description of finite-temperature atoms that are tightly bunched in an optical lattice, as described in the fourth row of Table 1.1. To extend this model for describing my pattern formation experiment, I must incorporate the effects of a multi-level atomic excitation scheme as well as the addition of pattern-forming optical fields.
The model in Ch. 6 provides a self-consistent theoretical description of sub-Doppler-cooled atoms that are spatially bunched in a 1D optical lattice. To apply this model to the experiment described in this thesis, I must incorporate a multi-level atom description and account for the lin_\perp lin polarization configuration that I use. As I show in this chapter, the multi-level structure gives rise to a more complicated dipole potential energy distribution, which modifies the refractive index of atoms in the applied lattice.

For sufficiently tight bunching in the applied 1D lattice, there is a transverse instability that gives rise to spontaneous pattern formation, as described in Ch. 4. To model transverse pattern formation, I must also account for the presence of the generated optical fields and corresponding atomic patterns that synergistically enhance each another during pattern formation. By incorporating these transverse effects, I define the refractive index of the atoms in the presence of patterns, from which I can derive the threshold conditions for pattern formation.

The standard technique for predicting the threshold refractive index for pattern formation is performing a stability analysis. A stability analysis for transverse optical...
patterns is similar to the techniques presented in Sec. 3.3 for deriving the threshold condition for parametric oscillation (Eq. 3.12), but with the additional complication of generating multiple optical fields. Previous works that perform stability analyses for transverse pattern formation are either restricted to warm atoms [Firth et al. (1990)] or to the weak-bunching regime in cold atoms [Muradyan et al. (2005)]. Here, I present the first stability analysis for pattern formation in cold atoms that is valid for both weakly- and tightly-bunched atoms.

In the model I present in this chapter, I note that I do not account for Sisyphus cooling, which gives rise to a non-equilibrium gas [Jersblad et al. (2004)]. Instead, I approximate the momentum distribution of the atoms using a simplified Maxwell-Boltzmann distribution characterized by the temperature of the atoms after they have undergone Sisyphus cooling.

In this chapter, I first describe theoretically the interaction of counterpropagating optical fields with multi-level atoms. I show that a multi-level atom description is necessary to describe a linear polarization configuration. I then go on to extend the model in Ch. 6 to study the nonlinear refractive index for atoms in a 1D optical lattice in the regime of strong light-atom interactions with multi-level atoms. Finally, I extend this model to two spatial dimensions to derive the refractive index above threshold for pattern formation. I show that there exist transverse perturbations in both the intensity and atomic density distributions. Finally, I perform a stability analysis to derive the threshold condition under which pattern formation occurs and compare these predictions to my experimental data. This chapter is based largely on my work in Ref. [Schmittberger and Gauthier (2016a)].
7.1 Applied lin⊥lin optical lattice

In my experiment, I apply counterpropagating optical fields in a lin⊥lin polarization configuration with electric field \( \vec{E} = \vec{E}_0(z)e^{-i\omega t} + \text{c.c.} \), where

\[
\vec{E}_0(z) = F(z)e^{ikz}\hat{x} + e^{i\phi}B(z)e^{-ikz}\hat{y},
\]  

(7.1)

where \( k \) is the wavevector in vacuum and \( \phi \) is an arbitrary relative phase between the fields. I take \( \phi = -\pi/2 \) in order to set the field polarization to \( \hat{\sigma}^- \) at \( z = 0 \), which I use to define a self-consistent density distribution, as shown in Fig. 7.1. Here, I show the electric field configuration described by Eq. 7.1 with \( \phi = -\pi/2 \) and the corresponding locations of density maxima for an example bunching parameter \( b = |I_1(\zeta)|/I_0(\zeta) = 0.8 \).

The right and left circular polarization vectors are

\[
\hat{\sigma}^+ = -\frac{\hat{x} + i\hat{y}}{\sqrt{2}} \quad \text{and} \quad \hat{\sigma}^- = \frac{\hat{x} - i\hat{y}}{\sqrt{2}}.
\]  

(7.2)

The electric field can therefore be rewritten as

\[
\vec{E}_0(z) = \left\{ \left[ -F(z)e^{ikz} + B(z)e^{-ikz} \right] \frac{\hat{\sigma}^+}{\sqrt{2}} + \left[ F(z)e^{ikz} + B(z)e^{-ikz} \right] \frac{\hat{\sigma}^-}{\sqrt{2}} \right\} e^{-i\omega t}.
\]  

(7.3)

I define \( F(z) = \tilde{F}e^{i\delta z} \) and \( B(z) = \tilde{B}e^{-i\delta z} \), where \( e^{i\delta z} \) explicitly defines the slow spatial variation of the optical field amplitudes inside the atomic medium. I also then define \( k' = k + \delta \) as the wavevector of the optical fields inside the medium, where from the methods presented in Ch. 6, \( \delta = k\chi_{\text{eff}}/2 \). Then,

\[
\vec{E} = \left\{ \left[ -\tilde{F}e^{ik'z} + \tilde{B}e^{-ik'z} \right] \frac{\hat{\sigma}^+}{\sqrt{2}} + \left[ \tilde{F}e^{ik'z} + \tilde{B}e^{-ik'z} \right] \frac{\hat{\sigma}^-}{\sqrt{2}} \right\} e^{-i\omega t}.
\]  

(7.4)

For equal intensity optical fields, \( \tilde{F} = \tilde{B} = \tilde{E}_p \), and

\[
\vec{E}_0(z) = \left\{ -i\sqrt{2}\tilde{E}_p\sin(k'z)\hat{\sigma}^+ + \sqrt{2}\tilde{E}_p\cos(k'z)\hat{\sigma}^- \right\} e^{-i\omega t}.
\]  

(7.5)
The applied electric field from Eq. 7.1 has a periodically-varying polarization. The choice of phase $\phi = -\pi/2$ selects a $\hat{\sigma}^-$-polarization at $z = 0$. The corresponding density distribution for example bunching parameter $b = 0.8$ shows that atoms bunch at locations of circular polarizations.

I only consider the time-independent intensity $I(z)$, because $e^{\pm i\omega t}$ terms correspond to two-photon transitions, which the atoms cannot see. The intensity therefore goes as

$$I(z) = 2\epsilon_0 c |\tilde{E}|^2 \rightarrow I_{\text{tot}} = 4\epsilon_0 c |\tilde{E}_p|^2.$$

This is independent of position, which is expected for a lin-lin-counterpropagating configuration [Castin et al. (1991)]. However, the dipole potential has a position dependence due to the spatial dependence of the optical pumping configuration from the stretched states. The saturable nonlinearity also has a position dependence due to the fact that optical transitions can saturate. I must therefore treat a multi-level atom picture in order to describe the light-atom interaction in the lin-lin polarization configuration.
7.2 Defining the material polarization

The material polarization is derived in Ch. 2 for a two-level atom. To extend this description to a multi-level atomic scheme, one needs to account for all possible optical transitions. In the multi-level-atom case, the material polarization from Eq. 2.19 is modified according to [Boyd (2008)]

$$\vec{P}(z) = \sum_{g,e} \eta(z, r) \frac{\vec{\mu}_{ge} (\vec{\mu}_{eg} \cdot \vec{E})}{\hbar (\Delta + i/T_2)} \frac{(\rho_{ee} - \rho_{gg})^{(eq)} [1 + (\omega - \omega_{eg})^2 T_2]}{1 + (\omega - \omega_{eg})^2 T_2^2 + (4/\hbar^2) |\vec{\mu}_{eg} \cdot \vec{E}|^2 T_1 T_2}. \quad (7.7)$$

I consider the $J = 1/2 \rightarrow 3/2$ spin model for $^{87}\text{Rb}$. While I cannot realistically ignore the hyperfine structure, a fine-structure model is known to provide a good qualitative understanding of the experiment when most (~90%) of the atoms are tightly bunched at regions of pure $\hat{\sigma}^+ / \hat{\sigma}^-$ polarization and pumped into the stretched states [Greenberg et al. (2011)]. I take all the population to be evenly distributed between the two ground states so that the polarization becomes

$$\vec{P}(z) = \sum_{g,e} -\vec{\mu}_{ge} (\vec{\mu}_{eg} \cdot \vec{E}) (\Delta - i\Gamma/2) \frac{1}{\hbar (\Delta^2 + \Gamma^2/4)} \eta(z) \left( 1 + \frac{8}{\hbar^2 \Gamma^2} \frac{|\vec{\mu}_{eg} \cdot \vec{E}|^2}{1 + 4\Delta^2/\Gamma^2} \right)^{-1}. \quad (7.8)$$

This model ignores Raman transitions between the $m_J = \pm 1/2$ ground states, which is a good approximation when the atoms are tightly bunched at regions of $\hat{\sigma}^\pm$ polarization. However, this is not a good approximation for other experiments that use higher-temperature atoms [Labeyrie et al. (2014)], and any extension of this model to describe those systems should account for these transitions.

In the limit where the intensity is much less than the saturation intensity, the Taylor expansion $1/(1 + x) \approx 1 - x$ is valid, and Eq. 7.8 becomes

$$\vec{P}(z) = \sum_{g,e} -\vec{\mu}_{ge} (\vec{\mu}_{eg} \cdot \vec{E}) (\Delta - i\Gamma/2) \frac{1}{\hbar (\Delta^2 + \Gamma^2/4)} \eta(z) \left[ 1 - \frac{8}{\hbar^2 \Gamma^2} \frac{|\vec{\mu}_{eg} \cdot \vec{E}|^2}{1 + 4\Delta^2/\Gamma^2} \right]. \quad (7.9)$$
The last term in square brackets represents the saturable nonlinearity. The density distribution $\eta(z)$ contains intensity-dependent terms that give rise to the bunching-induced nonlinearity. In the following subsections, I investigate these nonlinearities in the multi-level-atom picture to show how this differs from the two-level-atom, lin||lin configuration discussed in Ch. 6.

### 7.2.1 The saturable nonlinearity

Despite having a spatially-uniform intensity in the lin||lin polarization configuration, the saturable nonlinearity is spatially dependent due to the spatially-varying electric field polarization. From Eq. 7.9 and with $I_{\text{sat}} = \hbar^2 \Gamma^2 \epsilon_0 c/2 |\mu|^2$, the saturable nonlinearity is

$$\frac{8}{\hbar^2 \Gamma^2} \frac{\langle \vec{\mu}_{eg} \cdot \vec{E} \rangle^2}{[1 + 4\Delta^2/\Gamma^2]} = \frac{4\epsilon_0 c |\vec{\mu}_{eg} \cdot \vec{E}|^2}{I_{s\Delta} |\mu|^2}, \quad (7.10)$$

which has a spatial dependence that depends on the $g \to e$ transition and the local electric field. I define $I^+(z)$ and $I^-(z)$, where, with $F(z) = \tilde{F}e^{i\delta z}$ and $B(z) = \tilde{B}e^{-i\delta z}$,

$$\frac{I^+(z)}{I_{s\Delta}} = \frac{2\epsilon_0 c}{I_{s\Delta}} \left[ \tilde{F}^2 + \tilde{B}^2 - 2\tilde{F} \tilde{B} \cos(2k'z) \right] + \frac{12\epsilon_0 c}{3 I_{s\Delta}} \left[ \tilde{F}^2 + \tilde{B}^2 + 2\tilde{F} \tilde{B} \cos(2k'z) \right], \quad (7.11)$$

and

$$\frac{I^-(z)}{I_{s\Delta}} = \frac{2\epsilon_0 c}{I_{s\Delta}} \left[ \tilde{F}^2 + \tilde{B}^2 + 2\tilde{F} \tilde{B} \cos(2k'z) \right] + \frac{12\epsilon_0 c}{3 I_{s\Delta}} \left[ \tilde{F}^2 + \tilde{B}^2 - 2\tilde{F} \tilde{B} \cos(2k'z) \right]. \quad (7.12)$$

Equations 7.11 and 7.12 define the saturable nonlinearity for atoms in the $m_J = \pm 1/2$ ground states, respectively. The first term in each represents a stretched state transition, and the second term represents a transition to the other allowed excited state. When the atoms are tightly bunched at regions of pure $\hat{\sigma}^{\pm}$-polarization, the atoms only undergo stretched state transitions, and the second term in Eqs. 7.11
and 7.12 is negligible. This has implications when operating well above threshold, as discussed in Sec. 5.1.2. For the purposes of this chapter, however, I assume that below and close to threshold, all transitions are possible. Thus I use Eqs. 7.11 and 7.12 to simplify Eq. 7.9 and define the light-atom interaction in the lin\perp\perp lin polarization configuration.

7.2.2 The lin\perp\perp lin dipole potential

In addition to the saturable nonlinearity, Eq. 7.9 also contains the bunching-induced nonlinearity in the definition of \( \eta(z) \). The density distribution \( \eta(z) \) depends on the ratio of the dipole potential to the thermal energy of the atoms. The dipole potential \( U(z) \) in the lin\perp\perp lin polarization configuration is more complicated than in the lin||lin polarization configuration because of the spatially varying electric field polarization discussed in Sec. 2.3.3. This periodically varying polarization can be treated as the sum of the two light shifts experienced by atoms in the \( m_J = \pm 1/2 \) ground states [Castin et al. (1991)], such that

\[
U(z) = U_p^+(z) + U_p^-(z),
\]

where

\[
U_p^\pm(z) = \frac{4\Delta(\vec{\mu}^\pm \cdot \vec{E})^* (\vec{\mu}^\pm \cdot \vec{E})}{\hbar \Gamma^2 (1 + (2\Delta/\Gamma)^2)}
\]

define the dipole potentials for atoms in the \( m_J = \pm 1/2 \) ground states, respectively. For atoms in the \( m_J = +1/2 \) ground state, there are two transitions that contribute to the dipole potential: the \( m_J = +1/2 \rightarrow m_{J'} = +3/2 \) transition (Clebsch-Gordon coefficient=1), and the \( m_J = +1/2 \rightarrow m_{J'} = -1/2 \) transition (Clebsch-Gordon coefficient= \( \sqrt{1/3} \)). I calculate these separately according to \( U_p^+(z) = \ldots \)
\[
U_{p,1/2\rightarrow+3/2}(z) + U_{p,1/2\rightarrow-1/2}(z). \text{ Here,}
\]

\[
U_{p,1/2\rightarrow+3/2}(z) = \frac{4\Delta(-\mu^+ F(z)e^{ikz})*(-\mu^+ F(z)e^{ikz})}{\hbar \Gamma^2(1 + (2\Delta/\Gamma)^2)^{\sqrt{2}\sqrt{2}}} + \frac{4\Delta(\mu^+ B(z)e^{-ikz})*(\mu^+ B(z)e^{-ikz})}{\hbar \Gamma^2(1 + (2\Delta/\Gamma)^2)^{\sqrt{2}\sqrt{2}}} + \frac{4\Delta(-\mu^+ F(z)e^{ikz})*(\mu^+ B(z)e^{-ikz})}{\hbar \Gamma^2(1 + (2\Delta/\Gamma)^2)^{\sqrt{2}\sqrt{2}}} + \frac{4\Delta(\mu^+ B(z)e^{-ikz})*(-\mu^+ F(z)e^{ikz})}{\hbar \Gamma^2(1 + (2\Delta/\Gamma)^2)^{\sqrt{2}\sqrt{2}}} \quad (7.15)
\]

and

\[
U_{p,1/2\rightarrow-1/2}(z) = \frac{4\Delta(\mu^- F(z)e^{ikz})*(\mu^- F(z)e^{ikz})}{\sqrt{3}\sqrt{3}\hbar \Gamma^2(1 + (2\Delta/\Gamma)^2)^{\sqrt{2}\sqrt{2}}} + \frac{4\Delta(\mu^- B(z)e^{-ikz})*(\mu^- B(z)e^{-ikz})}{\sqrt{3}\sqrt{3}\hbar \Gamma^2(1 + (2\Delta/\Gamma)^2)^{\sqrt{2}\sqrt{2}}} + \frac{4\Delta(-\mu^- F(z)e^{ikz})*(\mu^- B(z)e^{-ikz})}{\sqrt{3}\sqrt{3}\hbar \Gamma^2(1 + (2\Delta/\Gamma)^2)^{\sqrt{2}\sqrt{2}}} + \frac{4\Delta(\mu^- B(z)e^{-ikz})*(-\mu^- F(z)e^{ikz})}{\sqrt{3}\sqrt{3}\hbar \Gamma^2(1 + (2\Delta/\Gamma)^2)^{\sqrt{2}\sqrt{2}}} \quad (7.16)
\]

I take \(|\mu^+|^2 = |\mu^-|^2 = |\mu|^2\) and keep the Clebsch-Gordon coefficients as numerical coefficients. From the methods in Ch. 6, I let \(F(z) = \tilde{F}e^{i\delta z}\) and \(B(z) = \tilde{B}e^{-i\delta z}\), so that

\[
U_{p,1/2\rightarrow+3/2}(z) = \frac{2\Delta|\mu|^2(\tilde{F}^2 + \tilde{B}^2)}{\hbar \Gamma^2(1 + (2\Delta/\Gamma)^2)} - \frac{2\Delta|\mu|^2\tilde{F}\tilde{B}e^{-2ik'z}}{\hbar \Gamma^2(1 + (2\Delta/\Gamma)^2)} - \frac{2\Delta|\mu|^2\tilde{F}\tilde{B}e^{2ik'z}}{\hbar \Gamma^2(1 + (2\Delta/\Gamma)^2)}. \quad (7.17)
\]

I define \(I_F = 2\epsilon_0c\tilde{F}^2\) and \(I_B = 2\epsilon_0c\tilde{B}^2\), and I assume equal-intensity pump fields \(I_F = I_B = I_p\), so that

\[
U_{p,1/2\rightarrow+3/2}(z) = \frac{\hbar I_p}{I_{\text{sat}}(1 + (2\Delta/\Gamma)^2)} - \frac{\hbar I_p\cos(2k'z)}{I_{\text{sat}}(1 + (2\Delta/\Gamma)^2)}. \quad (7.18)
\]

Analogously,

\[
U_{p,1/2\rightarrow-1/2}(z) = \frac{\hbar I_p}{3I_{\text{sat}}(1 + (2\Delta/\Gamma)^2)} + \frac{\hbar I_p\cos(2k'z)}{3I_{\text{sat}}(1 + (2\Delta/\Gamma)^2)}. \quad (7.19)
\]

Thus, the dipole potential for atoms in the \(m_J = +1/2\) ground state is

\[
U_p^+(z) = \frac{4\hbar I_p}{3I_{\text{sat}}(1 + (2\Delta/\Gamma)^2)} - \frac{2\hbar I_p\cos(2k'z)}{3I_{\text{sat}}(1 + (2\Delta/\Gamma)^2)}. \quad (7.20)
\]

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I redefine this in terms of the total intensity $I_{\text{tot}} = 2I_p$ and define

$$U_0 = \frac{\hbar \Delta I_{\text{tot}}}{I_{\text{sat}}(1 + (2\Delta/\Gamma)^2)},$$

(7.21)

so that the dipole potential can be rewritten as

$$U_p^+(z) = \frac{2}{3}U_0 - \frac{1}{3}U_0\cos(2k'z),$$

(7.22)

which agrees with the results of Refs. [Dalibard and Cohen-Tannoudji (1989); Castin et al. (1991)]. An analogous procedure for atoms in the $m_J = -1/2$ ground state leads to

$$U_p^-(z) = \frac{2}{3}U_0 + \frac{1}{3}U_0\cos(2k'z).$$

(7.23)

I introduce the variable $C^2 = |C_{1/2,3/2}|^2 - |C_{-1/2,1/2}|^2 = |C_{-1/2,-3/2}|^2 - |C_{1/2,-1/2}|^2 = 2/3$, which is the difference of the square of the Clebsch-Gordon coefficients for the two possible transitions, so that these can be rewritten as

$$U_p^+(z) = U_0 - C^2U_0\cos^2(k'z)$$

(7.24)

and

$$U_p^-(z) = U_0 - C^2U_0\sin^2(k'z).$$

(7.25)

From these equations, it is clear that there exist two superimposed dipole potentials that are phase-shifted by $\pi/2$. These correspond to the phase-shifted potentials depicted in Fig. 2.4.

7.2.3 The bunching-induced nonlinearity

The density distribution associated with the dipole potential in Eq. 7.13 goes as

$$\eta(z) = \bar{\eta}\exp\left[\frac{-U(z)}{k_BT}\right].$$

(7.26)
Based on the spatial periodicity of the superimposed dipole potentials defined in Eqs. 7.24 and 7.25, I define the Floquet expansion

$$\eta(z) = \eta^+(z) + \eta^-(z) = \sum_{j=-\infty}^{\infty} \eta_j^+ e^{2i(k'z-\pi/2)*j} + \sum_{j=-\infty}^{\infty} \eta_j^- e^{2ik'z*j},$$  

(7.27)

where

$$\eta_j^- = \frac{1}{\lambda'/2} \int_{-\lambda'/4}^{\lambda'/4} \tilde{\eta} \exp \left[ -\frac{U\mp(z)}{k_BT} \right] e^{-2ik'z*j} dz. \quad (7.28)$$

I define $\eta_j^-$ this way because there is a dipole potential minimum for red detunings that is symmetric about $z = 0$ and thus corresponds to a density maximum at $z = 0$. Also note that according to the shift theorem (see Sec. B.2), the Fourier coefficients $\eta_j^- = \eta_j^+$. Thus, $\eta_j^-$ becomes

$$\eta_j^- = \frac{1}{\lambda'/2} \int_{-\lambda'/4}^{\lambda'/4} \tilde{\eta} \exp \left[ -\frac{2U_0/3 - U_0 \cos(2k'z)/3}{k_BT} \right] e^{-2ik'z*j} dz. \quad (7.29)$$

I absorb the spatially independent part of the dipole potential into a new normalization constant

$$\eta' = \tilde{\eta} \exp \left[ -\frac{2U_0}{3k_BT} \right]. \quad (7.30)$$

I define $\tilde{I} = I_p/I_s\Delta$ and use the expansion $e^{-2ik'z*j} = \cos(2k'z * j) - i\sin(2k'z * j)$, so that Eq. 7.29 becomes

$$\eta_j^- = \frac{\eta'}{\lambda'/2} \int_{-\lambda'/4}^{\lambda'/4} \exp \left[ -\frac{C^2 \Delta \tilde{I}}{\tilde{T}} \cos(2k'z) \right] \cos(2k'z * j) dz, \quad (7.31)$$

where the integral containing the $\sin(2k'z * j)$ term yields zero due to the multiplication of the even and odd functions. I define

$$\zeta = C^2 \Delta \tilde{I}/\tilde{T}, \quad (7.32)$$

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\[ \lambda' = 2\pi/k', \quad \tilde{z} = 2k'z, \quad \text{and} \quad d\tilde{z} = 2k'dz, \] so that

\[ \eta_j^- = \frac{\eta'}{2\pi} \int_{-\pi}^{\pi} \exp \left[ -\zeta \cos(\tilde{z}) \right] \cos(\tilde{z} \cdot j) d\tilde{z}. \] (7.33)

This yields

\[ \eta_j^- = \eta' I_j(-\zeta), \] (7.34)

where \( I_j(x) \) are modified Bessel functions of the first kind of order \( j \).

The normalization constant \( \eta' \) is calculated independently by equating the areal density to the integral of the density distribution over one period. In this case, unlike the linear case in Ch. 6, the areal density goes as \( N_0 = n_a \cdot (\lambda'/4) \), where \( n_a \) is the average atomic density, because each pancake of atoms occurs every \( \lambda'/4 \). Therefore,

\[ \frac{n_a\lambda'}{4} = \eta' \int_{-\pi/2k'}^{\pi/2k'} \exp \left[ -\zeta \cos(2k'z) \right] dz. \] (7.35)

This yields \( \eta' = n_a/[2I_0(-\zeta)] \), so the Fourier coefficients are

\[ \eta_j^- = \frac{n_a I_j(-\zeta)}{2 I_0(-\zeta)}. \] (7.36)

The factor of 1/2 here, which is different than the linear case, corrects for the apparent double-counting of \( \eta_0 \) that occurs in our original definition of \( \eta(z) \). Applying this normalization constant to Eq. 7.27, the density distribution \( \eta(z) \) is properly normalized for the linear polarization configuration.

The density distribution

\[ \eta(z) = \eta^+(z) + \eta^-(z) = \frac{n_a I_j(-\zeta)}{2 I_0(-\zeta)} \left[ \sum_{j=-\infty}^{\infty} e^{2i(k'z-\pi/2)\ast j} + \sum_{j=-\infty}^{\infty} e^{2ik'z\ast j} \right] \] (7.37)

contains highly nonlinear terms in the polarization of Eq. 7.9 for the tight-bunching regime; i.e., when \( |\zeta| \gtrsim 0.8 \), the Bessel functions of Eq. 7.37 give rise to non-negligible
χ(5) and higher-order terms. In order to solve for the explicit dependence of the index of refraction on ζ, it is necessary to solve the wave equation for the coupled forward and backward fields \( F(z)e^{ikz} \) and \( B(z)e^{-ikz} \).

### 7.3 Calculating the refractive index for a linear lattice

When solving the wave equation, transitions from the two ground states decouple from one another, so that

\[
\vec{P} = \vec{P}^+_{g=+1/2} + \vec{P}^-_{g=-1/2} = \vec{P}^+ + \vec{P}^-, \tag{7.38}
\]

where

\[
\vec{P}^+ = P_{g=+1/2;\varepsilon=+3/2}\hat{\sigma}^+ + P_{g=+1/2;\varepsilon=-1/2}\hat{\sigma}^-
\]

and

\[
\vec{P}^- = P_{g=-1/2;\varepsilon=-3/2}\hat{\sigma}^- + P_{g=-1/2;\varepsilon=+1/2}\hat{\sigma}^+. \tag{7.39}
\]

I define

\[
P^+ = \sum_e -\frac{|\vec{\mu}^+_{e,g=+1/2}|(|\vec{\bar{\mu}}^+_{e,g=+1/2} \cdot \vec{E}^1|)\Delta - i\Gamma/2}{\hbar(\Delta^2 + \Gamma^2/4)} \left[ 1 - \frac{8}{\hbar^2 \Gamma^2} \frac{|\vec{\bar{\mu}}^+_{e,g=+1/2} \cdot \vec{E}^1|^2}{1 + 4\Delta^2/\Gamma^2} \right] \eta^+(z) \tag{7.40}
\]

and an analogous expression for \( P^- \). I then solve the wave equation for the amplitudes \( F(z) \) and \( B(z) \). (See App. B.) From the solution to the wave equation, the phase shift \( \delta = k' - k \), where \( F(z) = \tilde{F}e^{i\delta z} \) and \( B(z) = \tilde{B}e^{-i\delta z} \), is

\[
\delta = \frac{k'}{4} \chi_{\text{lin}} C^2 \left[ \left( 1 - 3\tilde{I} \right) + \left( 1 - 2\tilde{I} \right) \frac{I_1(-\zeta)}{I_0(-\zeta)} - 2\tilde{I} \frac{I_2(-\zeta)}{I_0(-\zeta)} \right]. \tag{7.41}
\]

Because Eq. 7.42 calculates the phase shift experienced by one circular component of the total applied field, which is half the total applied field, the total phase shift experienced by the pure linearly polarized light is twice this. (See Sec. B.1.1 for the
Therefore, the total phase shift is $2\delta$, so that the index of refraction in a \textit{lin} optical lattice goes as $n \approx 1 + \chi_{\text{eff}}/2$ with

$$
\chi_{\text{eff}} = \chi_{\text{lin}} C^2 \left[ \left( 1 - 3\bar{I} \right) + \left( 1 - 2\bar{I} \right) \frac{I_1(-\zeta)}{I_0(-\zeta)} - 2\bar{I} \frac{I_2(-\zeta)}{I_0(-\zeta)} \right]. \quad (7.43)
$$

There are multiple qualitative differences between this case and the \textit{lin} case described in Ch. 6. For blue detunings ($\zeta > 0$) in the zero-temperature limit ($\zeta \to \infty$),

$$
\lim_{\zeta \to \infty} \chi_{\text{eff}} = -3\chi_{\text{lin}} C^2 \bar{I}. \quad (7.44)
$$

Therefore, since $\bar{I} \ll 1$, there is a finite but small light-atom interaction for blue detunings in the zero temperature limit. In contrast, in the \textit{lin}, two-level-atom case from Ch. 6, this limit went to zero because the atoms tightly bunch at the intensity zeroes. In contrast, the intensity is nonzero everywhere in the \textit{lin} polarization configuration. Thus, Eq. 7.44 implies that for blue detunings in the zero temperature limit, the atoms still always have a finite probability of interacting with the fields. For example, an atom in the $m_J = -1/2$ state minimizes its energy in the presence of a blue-detuned optical lattice by spatially bunching at a location of \hat{\sigma}^- polarization. In this case, the atoms are not pumped into the stretched state. However, the atom can still be pumped via the weaker transition into the $m_J = +1/2$ excited state. Therefore, the conditions are never optimal for the atom to interact strongly with the optical fields. However, the atoms may still always interact with them.

For red detunings ($\zeta < 0$) in the zero temperature limit ($\zeta \to -\infty$),

$$
\lim_{\zeta \to -\infty} \chi_{\text{eff}} = \chi_{\text{lin}} C^2 \left\{ 2 - 7\bar{I} \right\}. \quad (7.45)
$$

So the atoms still experience a much larger phase shift in the zero-temperature limit for red detunings \textit{c.f.} blue detunings. This makes sense because for red detunings the
atoms spatially organize into the locations that allow them to continuously pump into the stretched states. This is the situation that is optimized for atoms to interact strongly with the optical fields, as opposed to the blue-detuned case.

The general index of refraction in the $\text{lin} \perp \text{lin}$ polarization configuration is $n = n_{\text{lin}} + n_{\text{NL}}$, where $n_{\text{lin}} = 1 + \chi_{\text{lin}} C^2/2$ is the linear refractive index, and

$$n_{\text{NL}} = \frac{\chi_{\text{lin}} C^2}{2} \left[ -3\bar{I} + \left( 1 - 2\bar{I} \right) \frac{I_1(-\zeta)}{I_0(-\zeta)} - 2\bar{I} \frac{I_2(-\zeta)}{I_0(-\zeta)} \right]$$

(7.46)

is the nonlinear refractive index. Above a threshold value of $n_{\text{NL}}$, there exist transverse perturbations that give rise to transverse pattern formation. In order to derive this threshold condition, I perform a stability analysis to derive the conditions under which transverse perturbations experience sufficient gain to form transverse patterns.

7.4 Stability analysis

A stability analysis is a standard technique for deriving the threshold condition for transverse optical pattern formation in warm atoms [Firth and Paré (1988); Petrossian et al. (1992); Gaeta and Boyd (1993)]. However, in my cold-atom case, the expressions for the material polarization are more complicated than in the warm-atom case because I must also account for the bunching-induced nonlinearity.

To simplify the stability analysis, I consider a two-spot optical pattern, whose beam geometry is depicted in Fig. 7.2, and whose electric field goes as

$$\bar{E}(z,r,t) = \left[ F(z)e^{ikz}\hat{x} + e^{i\phi} B(z)e^{-ikz}\hat{y} + e^{i\phi} \alpha f_+(z,r)e^{ik(\cos \theta_2 - \sin \theta r)}\hat{y} + \right. \left. + e^{i\phi} \alpha f_-(z,r)e^{ik(\cos \theta_2 + \sin \theta r)}\hat{y} + \alpha b_+(z,r)e^{ik(-\cos \theta_2 - \sin \theta r)}\hat{x} + \right. \left. + \alpha b_-(z,r)e^{ik(-\cos \theta_2 + \sin \theta r)}\hat{x} \right] e^{-i\omega t} + \text{c.c.,} \quad (7.47)$$

where I use $\alpha$ to keep track of the order of the small-amplitude generated fields. I use these polarizations because these are the relative polarizations I observe experimen-
**Figure 7.2:** Optical field geometry for stability analysis. This depicted the beam geometry I consider for the stability analysis: A two-spot optical pattern on either side of the atomic cloud, where fields with a forward/nearly forward wavevector are denoted $F(z)$, $f_+(z,r)$ and $f_-(z,r)$, and fields with a backward/nearly backward wavevector are denoted $B(z)$, $b_+(z,r)$ and $b_-(z,r)$, as defined in Eq. 7.47.

Finally, however, it is straightforward to extend the analysis I present in this chapter and in App. B to study alternative polarization configurations. I assume that the phase $\phi = -\pi/2$ so that the polarization of all fields at $z = 0$ is $\hat{\sigma}^-$-polarized. We can rewrite this in terms of circular polarizations according to

$$
\vec{E}(z,r,t) = \left\{ \begin{array}{l}
-F(z)e^{ikz} + B(z)e^{-ikz} + \alpha f_+(z,r)e^{i(k\cos\theta z - k\sin\theta r)} + \\
\alpha f_-(z,r)e^{i(k\cos\theta z + k\sin\theta r)} - \alpha b_+(z,r)e^{i(-k\cos\theta z - k\sin\theta r)} - \alpha b_-(z,r)e^{i(-k\cos\theta z + k\sin\theta r)} \end{array} \right\} \frac{\hat{\sigma}^+}{\sqrt{2}} + \\
\left[ F(z)e^{ikz} + B(z)e^{-ikz} + \alpha f_+(z,r)e^{i(k\cos\theta z - k\sin\theta r)} + \alpha f_-(z,r)e^{i(k\cos\theta z + k\sin\theta r)} + \\
\alpha b_+(z,r)e^{i(-k\cos\theta z - k\sin\theta r)} + \alpha b_-(z,r)e^{i(-k\cos\theta z + k\sin\theta r)} \right] \frac{\hat{\sigma}^-}{\sqrt{2}} e^{-i\omega t} + \text{c.c.} \quad (7.48)
$$

Recall the polarization defined in Eq. 7.38. With the modified electric field from Eq. 7.48, the density distribution

$$
\eta(z,r) = n_0 \tilde{\eta} \exp \left[ - \frac{U^+(z,r) + U^-(z,r)}{k_B T} \right] \quad (7.49)
$$

can be separated into those components of the dipole potential arising solely due to the pump-pump gratings and those arising due to the generated pump-pattern.
gratings according to
\[
\eta(z, r) = n_a \tilde{n} \exp \left[ -\frac{U^+_\text{pumps}(z) + U^-_{\text{pumps}}(z)}{k_B T} \right] \exp \left[ -\frac{U^+_p(z, r) + U^-_{pp}(z, r)}{k_B T} \right].
\] (7.50)

The first exponential that defines the pump-pump gratings is defined by Eq. 7.37. I take the normalization constant to be that defined only due to the pump-pump gratings, which is a good approximation close to threshold. To derive the threshold condition, I am interested in the regime where the generated fields are very weak. In this case, the pump-probe dipole potentials are very shallow, so that the Taylor expansion
\[
\exp \left[ -\frac{U^+_p(z, r) + U^-_{pp}(z, r)}{k_B T} \right] \approx 1 - \frac{U^+_p(z, r) + U^-_{pp}(z, r)}{k_B T}
\] (7.51)
is valid. Equation 7.51 defines the transverse perturbations to the density distribution. Atoms that bunch into these perturbative gratings are termed “self-organized” because they are not imposed by externally applied optical fields, as discussed in Ch. 5. These terms further enhance the material polarization, which implies that the self-organized atomic structures enhance the light-atom interaction during pattern formation. In Eq. 7.51,
\[
U^\pm_{pp}(z, r) = \frac{4\Delta(\vec{\mu}^\pm \cdot \vec{E})^* (\vec{\mu}^\pm \cdot \vec{E})}{\hbar \Gamma^2 (1 + (2\Delta/\Gamma)^2)} \bigg|_{O(\alpha)}
\] (7.52)
but where again I only retain those terms of order \(\alpha\) that give rise to self-organized gratings. I neglect terms of order \(\alpha^2\) or higher because those correspond to the interference of two generated fields, which are orders of magnitude weaker and negligible close to threshold.

With \(k_B = \hbar \Gamma/2T_D\), \(\bar{T} = T/T_D\), and \(\bar{\Delta} = \Delta/\Gamma\), the polarization components
become

\[
\tilde{P}^+(z, r) = -\frac{2\tilde{\mu}^+_e(\tilde{\mu}^+_e \cdot \tilde{E})(2\tilde{\Delta} - i)}{h\Gamma(1 + 4\tilde{\Delta}^2)} \left[ 1 - \frac{8}{h^2\Gamma^2 (1 + 4\tilde{\Delta}^2)} |\tilde{\mu}^+_e \cdot \tilde{E}|^2 \right] \left( \sum_{j=-\infty}^{\infty} \eta_j e^{2i(k'z - \pi/2)\eta_j} \right) \left( 1 - \frac{8\tilde{\Delta}(\tilde{\mu}^+_e \cdot \tilde{E}_{pp})(\tilde{\mu}^+_e \cdot \tilde{E}_{pp})}{(h\Gamma)^2 T(1 + 4\Delta^2)} \right),
\]

(7.53)

and

\[
\tilde{P}^-(z, r) = -\frac{2\tilde{\mu}^-_e(\tilde{\mu}^-_e \cdot \tilde{E})(2\tilde{\Delta} - i)}{h\Gamma(1 + 4\tilde{\Delta}^2)} \left[ 1 - \frac{8}{h^2\Gamma^2 (1 + 4\tilde{\Delta}^2)} |\tilde{\mu}^-_e \cdot \tilde{E}|^2 \right] \left( \sum_{j=-\infty}^{\infty} \eta_j e^{2ik'z\eta_j} \right) \left( 1 - \frac{8\tilde{\Delta}(\tilde{\mu}^-_e \cdot \tilde{E}_{pp})(\tilde{\mu}^-_e \cdot \tilde{E}_{pp})}{(h\Gamma)^2 T(1 + 4\Delta^2)} \right),
\]

(7.54)

where the component \((\tilde{\mu}^\pm \cdot \tilde{E}_{pp})^*(\tilde{\mu}^\pm \cdot \tilde{E}_{pp})\) must consist of exactly one pump field term and one weak field term. I expand Eqs. 7.53 and 7.54 in App. B. I then extract terms that are phase-matched to the generated fields in order to solve the wave equation. Recall the wave equation is

\[
\nabla^2 \tilde{E} - \frac{1}{c^2} \frac{\partial^2 \tilde{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \tilde{P}}{\partial t^2}.
\]

(7.55)

Under the rotating wave approximation, the left-hand-side of the wave equation for the \(f_+(z, r)\) weak field component that is \(\hat{\sigma}^-\)-polarized is

\[
2ik\cos\theta \frac{\partial f_+}{\partial z} e^{i(k\cos\theta z - k\sin\theta r - \omega t)} \frac{\hat{\sigma}^-}{\sqrt{2}}
\]

(7.56)

where I take the optical field amplitude variation in \(r\) to be very small, so that \(\partial f_+ / \partial r \to 0\). I define new variables

\[
f_+(z, r) = f'_+(z, r) e^{i(k' - k\cos\theta)z},
\]

(7.57)
\begin{align}
  f_-(z, r) &= f'_-(z, r)e^{i(k' - k\cos\theta)z}, \\
  b_+(z, r) &= b'_+(z, r)e^{-i(k' - k\cos\theta)z},
\end{align}

and

\begin{align}
  b_-(z, r) &= b'_-(z, r)e^{-i(k' - k\cos\theta)z}.
\end{align}

I define the following quantities:

\begin{align}
  \xi_0 &= \frac{k\chi_{\text{lin}}}{2\cos\theta} C^2, \\
  A &= 2\tilde{\eta}_0 - \frac{\tilde{\Delta}I_p}{2TI_{s\Delta}} \frac{2}{3} (5\tilde{\eta}_0 + 4\tilde{\eta}_1) - \frac{I_p}{2I_{s\Delta}} \frac{4}{3} (5\tilde{\eta}_0 + 4\tilde{\eta}_1) + \frac{\tilde{\Delta}}{T} \left( \frac{I_p}{2I_{s\Delta}} \right)^2 \frac{2}{9} (42\tilde{\eta}_0 + 52\tilde{\eta}_1 + 14\tilde{\eta}_2), \\
  B &= -\frac{\tilde{\Delta}I_p}{2TI_{s\Delta}} \frac{2}{3} (5\tilde{\eta}_0 + 4\tilde{\eta}_1) - \frac{I_p}{2I_{s\Delta}} \frac{2}{3} (5\tilde{\eta}_0 + 4\tilde{\eta}_1) + \frac{\tilde{\Delta}}{T} \left( \frac{I_p}{2I_{s\Delta}} \right)^2 \frac{2}{9} (42\tilde{\eta}_0 + 52\tilde{\eta}_1 + 14\tilde{\eta}_2), \\
  C &= \tilde{\eta}_1 - \frac{\tilde{\Delta}I_p}{2TI_{s\Delta}} \frac{1}{3} (5\tilde{\eta}_0 + 8\tilde{\eta}_1 + 5\tilde{\eta}_2) - \frac{I_p}{2I_{s\Delta}} \frac{2}{3} (5\tilde{\eta}_0 + 8\tilde{\eta}_1 + 5\tilde{\eta}_2) + \frac{\tilde{\Delta}}{T} \left( \frac{I_p}{2I_{s\Delta}} \right)^2 \frac{1}{9} (56\tilde{\eta}_0 + 91\tilde{\eta}_1 + 56\tilde{\eta}_2 + 13\tilde{\eta}_3), \\
  D &= -\frac{\tilde{\Delta}I_p}{2TI_{s\Delta}} \frac{1}{3} (5\tilde{\eta}_0 + 8\tilde{\eta}_1 + 5\tilde{\eta}_2) - \frac{I_p}{2I_{s\Delta}} \frac{1}{3} (5\tilde{\eta}_0 + 8\tilde{\eta}_1 + 5\tilde{\eta}_2) + \frac{\tilde{\Delta}}{T} \left( \frac{I_p}{2I_{s\Delta}} \right)^2 \frac{1}{9} (56\tilde{\eta}_0 + 91\tilde{\eta}_1 + 56\tilde{\eta}_2 + 13\tilde{\eta}_3).
\end{align}
Then by symmetry from Eq. B.66 in App. B, the coupled amplitude equations are as follows:

\[
\frac{\partial}{\partial z} \begin{pmatrix} f'_+ \\ f'_- \\ b'_+ \\ b'_- \end{pmatrix} = \begin{pmatrix} i\xi_0 A - i\delta_k & i\xi_0 D & i\xi_0 C & i\xi_0 B \\ -i\xi_0 D & -i\xi_0 A + i\delta_k & -i\xi_0 B & -i\xi_0 C \\ -i\xi_0 C & -i\xi_0 B & -i\xi_0 A + i\delta_k & -i\xi_0 D \\ i\xi_0 B & i\xi_0 C & i\xi_0 D & i\xi_0 A - i\delta_k \end{pmatrix} \begin{pmatrix} f'_+ \\ f'_- \\ b'_+ \\ b'_- \end{pmatrix}.
\] (7.66)

I solve this equation for the boundary conditions \( f'_+(-L/2) = f''_-(L/2) = b'_+(+L/2) = b''_-(+L/2) = 0 \) using the methods in Refs. [Silberberg and Bar-Joseph (1982); Firth et al. (1990)]. For the example case of an optical depth of 20 and a detuning \( \Delta = -5 \), the solution is shown in Fig. 7.3 for the predicted pump intensity as a function of the phase mismatch \( \Delta k = k[1 - \cos(\theta)] \). The blue (lower) curve represents the minimum solution. The orange (higher) curve represents another higher numerical solution. This solution shows that at \( k[1 - \cos(\theta)] = 0 \) (\( \theta = 0 \)), the intensity required to generate new optical fields approaches infinity. However, at a finite angle, one can generate new optical fields. This is consistent with the concept of weak wave retardation discussed in Sec. 3.2, which tells us that the wave-mixing process that generates new fields requires a finite angle for phase-matching.

The minimum point on the blue curve in Fig. 7.3 at \( k[1 - \cos(\theta)] \simeq 120 \) provides a theoretical prediction for the angle of emission of the generated fields: \( \theta \simeq 5 \) mrad, which is consistent with typical experimental observations of \( \theta = (4 \pm 1) \) mrad. The minimum point also predicts the minimum intensity threshold for these parameters: \( I_p/I_{\text{sat}} \simeq 0.1 \).

I solve Eq. 7.66 for other detunings and optical depths to build theoretical curves from the extracted solutions for the minimum predicted intensities. I show these theoretical results (blue) with experimental data points (red) in Fig. 7.4 as functions of detuning and optical depth.

In these theoretical curves, I incorporate a free parameter \( p \) that I use to adjust
Figure 7.3: Predicted intensity vs. phase mismatch. Single-beam intensity normalized by the resonant saturation intensity as a function of the phase mismatch $\Delta k = k(1 - \cos \theta)$, where again $k$ is the vacuum wavevector and $\theta$ is the angle between the generated fields and the applied (pump) fields. This is for the specific case of an optical depth of 20, $\bar{\Delta} = -4$, $L = 3$ cm, and $\bar{T} = 3/146$.

Figure 7.4: Results of stability analysis. Single-beam intensity normalized by the resonant saturation intensity as a function of (a) the detuning normalized by the natural linewidth for the specific case of an optical depth of 62, $L = 3$ cm, $\bar{T} = 3/146$, and red detunings with a free parameter value $p = 6$, and (b) the optical depth for the specific case of an optical depth of $\bar{\Delta} = -5$, $L = 3$ cm, $\bar{T} = 3/146$, and a free parameter value $p = 54$. The blue curve represents the predictions from Eq. 7.66 with free parameter $I_0 \rightarrow pI_0$, and the red points represent experimental data. The red rectangle represents the statistical error due to the initial intensity measurement, the detuning measurement, and the confidence interval of the fit to the OD measurement. (See App. A.)
the effective intensity $\bar{I} \rightarrow p\bar{I}$. The use of a free parameter allows me to adjust the scale of the predicted curves in order to test whether the predicted curvature matches my experimental data. The deviation of the experiment from the scale of the theoretical predictions may arise due to multiple factors: 1. There are statistical errors in the initial measurements of $\bar{I}$, $\Delta$, $L$, and the OD. 2. There are additional statistical errors in the measurement of $\bar{I}$ due to the beam reshaping effect discussed in Sec. 4.4. 3. There are additional statistical errors in $L$ because the generated fields may emerge from the cloud before propagating its full length, thus reducing the effective length of the cloud. 4. This model assumes perfectly counterpropagating pump beams, and thus any slight misalignment of the pump beams reduces the efficiency of the wave-mixing process and increases the intensity threshold. 5. This model assumes a uniform atomic density across the pump beams, which is not the case when the pump beam size is comparable to the width of the atomic cloud. 6. I do not explicitly account for Sisyphus cooling, and 7. I neglect absorption in this model.

With the use of a free parameter, the predicted theoretical curves fit well to the experimental data in Fig. 7.4.

7.5 Conclusions

In this chapter, I provide a theoretical description for multi-level atoms in an optical lattice. I derive the index of refraction for sub-Doppler-cooled atoms in a lin⊥lin optical lattice, and I discuss how this differs from the lin∥lin case. I then extend this model to a two-dimensional geometry with multiple optical fields in order to describe two-spot optical pattern formation. I then perform a stability analysis in order to derive the threshold condition for generating patterns.

This work represents the first stability analysis that is valid for all regimes of atomic bunching, as described in the last row of Table 1.1. As I show in Chs. 4, 5,
and 6, this description is necessary to describe pattern formation in my experiment, where the atoms are tightly bunched in the applied optical lattice. This model also shows the importance of accounting for transverse perturbations in the density distribution, \textit{i.e.}, the self-organized atomic structures, which enhances the refractive index of the atoms during and above threshold for pattern formation and give rise to multimode atomic self-organization at low light levels.
In this thesis, I describe a novel technique for studying non-equilibrium phenomena in a multimode geometry. I show that by enhancing the light-atom interaction strength using atomic bunching, I observe coupled optical-atomic pattern formation in a free-space, laser-driven gas of cold atoms at low intensities. This work represents the first observation of multimode self-organization and spontaneous 3D cooling, which has implications for nonlinear optics, fundamental atomic physics, condensed matter physics, and quantum information science, as I discuss below.

8.1 Summary

In Ch. 1, I provide an introduction and motivation for the work presented in this thesis. I show that previous studies of self-organization in cold atoms work in a single-mode geometry, and I describe the new physics that is accessible in a multimode geometry [Gopalakrishnan et al. (2009)]. To observe self-organization in a free-space, multimode geometry, I establish that it is necessary to achieve enhanced light-atom interaction strengths so that a single pass of an optical field is sufficient to generate atomic bunching into new structures. I also describe the previous work involving
strong light-atom interactions in optical lattices [Deutsch et al. (1995)], as well as work in both optical [Silberberg and Bar-Joseph (1982); Dawes et al. (2005)] and atomic [Inouye et al. (1999)] pattern formation. I then provide an outline and a summary of the novel work presented in this thesis.

Chapters 2 and 3 provide an introduction to light-atom interactions and pattern formation, which are useful for understanding the physics, equations, and terminology used in the subsequent chapters. In Ch. 2, I derive the material susceptibility for a two-level atom, which is the quantity that I use throughout this thesis as a measure of the light-atom interaction strength. I also discuss the mechanical light forces on atoms, and I show how these forces give rise to Sisyphus cooling. In addition, I derive the steady-state solution to the Fokker-Planck equation, which I use to describe the density distribution of atoms in my experiment. In Ch. 3, I introduce the relevant nonlinear optics of pattern formation, including the importance of phase-matching and weak-wave retardation.

Chapter 4 provides a description of my experiment, a qualitative analysis of the threshold conditions for pattern formation, and a characterization of the transverse optical patterns. I show that atomic bunching in the applied 1D lattice is essential for reaching the threshold for pattern formation in cold atoms at low intensities. I describe my observations of pattern formation at record-low powers, continuous symmetry-breaking of the patterns, and the phase and temporal correlations of the patterns. This work represents the first observation of pattern formation in cold atoms.

In Ch. 5, I show that the formation of optical patterns in my system necessarily corresponds to the formation of atomic patterns. I describe my novel observation of self-organization in a multimode geometry as well as spontaneous 3D cooling. I measure the motional states of atoms and verify that the atoms bunch into new structures that depend on the geometry of the multimode optical patterns. I also
measure the temperature of atoms in these gratings and show that atoms also undergo Sisyphus cooling in the radial direction, which gives rise to longer coherence times.

Chapters 6 and 7 provide a theoretical characterization of my experiment. In Ch. 6, I present a new, self-consistent model for atoms in optical lattices that allows for tight bunching and strong interactions between the atoms and the lattice-forming optical fields. This work represents a unification of the finite-temperature models used in nonlinear optics [Muradyan et al. (2005)] and the zero-temperature models used in the optomechanical physics community [Deutsch et al. (1995)]. In Ch. 7, I extend this model to account for multi-level atoms and pattern formation. I show that the bunching-induced nonlinearity gives rise to enhanced light-atom interaction strengths and low-intensity thresholds for realizing optical and atomic pattern formation.

8.2 Future directions

The main results of this thesis are the experimental observations of pattern formation in cold atoms, multimode self-organization, and spontaneous 3D cooling, as well as the formal theoretical treatment of a bunching-induced enhancement of light-atom interactions. This work is at the intersection of the fields of nonlinear optics, fundamental atomic physics, and condensed matter physics. In this section, I describe potential applications of my work in these fields as well as in quantum information science.

On the theoretical side, the self-consistent model presented in Ch. 6 for atoms in optical lattices represents a novel way to predict the nonlinear refractive index in tightly bunched cold atoms. This work represents a unification of models used in nonlinear optics and optomechanical physics, and it provides new insights regarding how to achieve enhanced light-atom interaction strengths. For the lattice community, this model also provides a basis for predict the nonlinear optical phase shift imposed
on atoms by the lattice-forming optical fields, which may open new avenues for lattice physics, including a different regime of studying photonic band gaps, for example.

In the experiment, after applying optical fields in only 1D, I observe the spontaneous formation of multiple optical fields at low-light-levels. In addition to pattern formation, this provides a simplified system in which one can potentially observe other low-light-level, multidimensional nonlinear optical effects, such as high-dimensional optical soliton formation [Fibich et al. (2007)] or filamentation [Fibich and Klein (2011)].

Because I observe pattern formation at low intensities and with long coherence times due to spontaneous 3D cooling, it is also feasible to use my system as a source of correlated light for continuous-variable quantum information protocols [Boyer et al. (2008)]. Due to the strong phase and temporal correlations among the generated optical fields as well as the observed polarization correlations, my system may also be used to generate hyperentangled photon pairs [Yan and Zhu (2013)]. To verify this, a future experiment may involve measuring quantum correlations among the generated fields close to the threshold for pattern formation.

Even though I do not use Bose-condensed atoms, cold but finite-temperature systems are also known to exhibit interesting phases, such as spin glasses [Gopalakrishnan et al. (2011)]. My work therefore represents an important step towards studying non-equilibrium phenomena in multimode geometries. In addition, there currently exist efforts to study multimode self-organization of Bose-condensed atoms in a multimode cavity [Kollár et al. (2015)], which is an exciting project that is expected to produce novel phases of matter, and which is the ideal system in which to study phenomena such as glassiness and frustration [Gopalakrishnan et al. (2009)]. My work represents a thermal precursor to this work, just as single-mode self-organization in Bose-condensed atoms [Baumann et al. (2010)] was preceded by its observation in thermal atoms [Black et al. (2003)].
Appendix A

Experimental Setup and Procedures

In this appendix, I provide an overview of my experimental setup and procedures. I describe the magneto-optical trap (MOT), lasers, and detectors that I use. I also describe how I measure the frequency of the optical fields I use in my experiment and the imaging system I use for the optical patterns.

A.1 Magneto-optical trap

To obtain a long nonlinear optical material, I use a 2D MOT that generates a cloud of atoms of length $L = 3$ cm. The details of the MOT that I use can be found in Refs. [Greenberg et al. (2007); Greenberg (2012)], but I provide a brief overview here. A picture of the vacuum chamber is shown in Fig. A.1(a). The glass walls of the chamber allow high flexibility for experimentation because optical fields can be applied through 4 sides of the cube. At the bottom of the chamber is a gold mirror, which is used for retroreflecting the cooling and trapping beams, as discussed below. Underneath the vacuum chamber are four rectangular coils, wrapped with $N = 200$ turns [Greenberg et al. (2007)]. The outer coils use current 1.94 A, and the inner
coils use a current 1.79 A. Together, they form a quadrupolar magnetic field with a zero-field region \( \sim 2 \text{ cm} \) above the center of the coils. This zero-field region is inside the vacuum chamber and traps the atoms. With the application of cooling and trapping optical fields, discussed below, this MOT forms a highly anisotropic cloud of atoms, as shown in Fig. A.1(b), of length \( \sim 3 \text{ cm} \) and width \( \sim 400 \mu \text{m} \).

The optical field geometry for the MOT is shown in Fig. A.2, from a perspective along \(-\hat{y}\), according to the axes shown in Fig. A.1(b). A titanium-sapphire 899 ring laser by Coherent (“Ti-Sapph”) is tuned to \( \Delta \approx -\Gamma \) and used for cooling and trapping. A distributed feedback laser (DFB) is tuned to the \( F = 1 \rightarrow F' = 2 \) transition in \(^{87}\text{Rb}\) and used for “repumping” atoms to the \( F = 2 \) ground state that fall into the \( F = 1 \) ground state. The collimated Ti-Sapph power is initially sent through an acousto-optic modulator (AOM, IntraAction, model 403). The first-
The first-order diffracted light is sent to the MOT, so that the optical fields at the MOT can be turned on and off quickly by triggering the AOM with +1 or 0 V using the IntraAction signal processor (model DE). I generate the desired +1/0 V waveform using a LabView program with a National Instruments Digital I/O Acquisition Board housing a NI CB-2162 Digital Waveform Generator.

After the AOM, the Ti-Sapph light (∼150 mW power) is split among three fields: two “radial” and one that is applied nearly along the long axis of the atoms (at an
angle $10^\circ$). The intensity of these fields is $\sim 8$ mW/cm$^2$ at the vacuum chamber. The repump light ($\sim 35$ mW power) is combined with the radial beams. The beam sizes are enlarged by telescopes, where the radial-beam telescopes use an elliptical lens for generating fields with a more uniform intensity across the length of the atoms. The radial beams are retro-reflected off a gold mirror at the bottom of the vacuum chamber. The third cooling beam is retroreflected off a mirror. By applying this third beam at a small angle relative to the magnetic field zero, the cloud of atoms can be made long c.f. spherical magneto-optical traps [Greenberg et al. (2007)].

A.1.1 Optical depth

With a long cloud of atoms, this MOT can achieve optical depths up to $\sim 100$. The optical depth (OD) is related to the transmission of a probe beam via

$$ T = \frac{I_{\text{out}}}{I_{\text{in}}} = e^{-\text{OD}}, \quad (A.1) $$

where $I_{\text{in}}$ is the intensity sent into the cloud of atoms and $I_{\text{out}}$ is the output intensity. When the probe beam is tuned to the atomic resonant frequency, the probe beam will undergo absorption, and $I_{\text{out}} < I_{\text{in}}$. When the probe beam is far-detuned from an atomic resonance, the probe beam will be relatively unaffected, and $I_{\text{out}} \rightarrow I_{\text{in}}$. Thus, measuring the full optical depth requires measuring the absorption at different frequencies.

I detect the output power of a probe beam sent down the long axis of the cloud of atoms. I modulate the amplitude of the MOT’s cooling and trapping beams according to the timing scheme shown in Fig. A.3(a), where the MOT beams are on most of the time to retain the majority of atoms. I scan the frequency of the probe beam across the $F = 2 \rightarrow F' = 3$ resonance of $^{87}$Rb. I extract data points only when the MOT beams are off, which otherwise shift the resonance. The resulting transmission of the scan is a Lorentzian lineshape, which relates to the optical depth.
Figure A.3: Measuring the optical depth. (a) The timing scheme during measurement of the optical depth generated by Labview and sent to the IntraAction signal processor using the National Instruments Digital I/O Acquisition Board. The MOT’s cooling and trapping beams are left on most of the time, with a ~50 µs shut-off every 10 ms. Meanwhile, the probe beam scans across resonance. (b) The transmission of a probe beam as a function of the detuning $\Delta$ relative to the $F = 2 \rightarrow F’ = 3$ resonant frequency. The data points are collected only during times when the MOT beams are off. The blue curve is the best fit to Eq. A.2, which gives $OD = 49.3$.

According to

$$T(\nu) = \exp\left\{-OD/(1 + [2\pi(\nu - \nu_0)/\Gamma]^2)\right\}, \quad (A.2)$$

where $\nu_0$ defines the location of the resonant frequency shown by $\Delta = 0$ in Fig. A.3(b). The frequency is calibrated to the time of the scan by relating the center of the $F = 2 \rightarrow F’ = 3$ resonance to the center of the $F = 2 \rightarrow F’ = 2$ resonance. The best fit in Fig. A.3(b) is $OD = 49.3$.

A.2 Pump beams

For the pump and probe beams that I describe in this thesis, I use a Toptica DL Pro diode laser. I send this light through an IntraAction AOM (model ATM-2301A2, driver model DE), and I fiber-couple the first-order diffracted light. I use fiber-splitters (Thorlabs model FC780-50B-APC) to extract some light for the frequency measurement (see Sec. A.2.1) and to split the remaining power evenly for the two applied pump fields.
Figure A.4: Schematic of pump beams. Each pump field encounters a half-waveplate ("$\lambda/2$"), which is used to adjust the power through the polarizers, shown as black ellipses with the set polarizations in the green circles/arrow. The pump beams are focused into the cloud of atoms using lenses of $f = 30$ cm. The 50/50 beam splitters are used to extract the generated fields for detection.

The beams are collimated exiting the fiber ports, depicted by the black rectangles in Fig. A.4. The lenses of focal length $f = 30$ cm are used to focus the beams into the vacuum chamber, where the center of the vacuum chamber is located at the focal point of each lens. The half-waveplates are used to balance the power that passes through the polarizers. The polarizers are the last optics that the pump beam encounters before the vacuum chamber. For most experiments, where I use a lin.lin polarization configuration, the polarizers are cross-polarized, as shown in the figure.

A.2.1 Frequency measurement

The frequency of the pump beams is detected using a New Focus photodiode (model 1801, bandwidth 250 MHz). Part of the pump light is split off using fiber splitters (Thorlabs model FC780-90B-APC) before it reaches the setup shown in Fig. A.4. This is overlapped on a 50/50 beam splitter with light from the Ti-Sapph, and the combined light is fiber-coupled for good mode-matching. This fiber-coupled light is sent to the photodiode whose output is sent to a spectrum analyzer (Agilent, model 4440A). The spectrum analyzer displays the frequency difference (beat note)
A lens located $d_0 \approx 58$ cm from the vacuum chamber is used to Fourier transform the fields into the plane of the DALSA camera. Typically $d_1 \approx f \approx 6.5$ cm is used to capture the optical pattern.

between the two laser fields incident on the detector. The Ti-Sapph is tuned to the $F = 2 \rightarrow F' = 3$ resonant frequency. Then, the measured beat note with the DL Pro is used to measure the frequency detuning of the pump beams relative to this resonant frequency.

### A.3 Imaging system

To image the optical patterns, I use the system shown in Fig. A.5, where $d_0 \approx 58$ cm and $d_1 \approx f \approx 6.5$ cm. I calibrate the magnification using a Thorlabs beam profiler (model BP209-VIS) to measure the size of the beam at the vacuum chamber. The DALSA camera (CCD, model CA-D1-0128T-STDL) has a pixel size of $16 \mu m \times 16 \mu m$ and a frame rate of up to 500 frames/sec, with which 1-3 pictures of the optical patterns are taken per 3 ms experiment. The output of the camera is processed using Labview with a National Instruments PCI-1424 CAD1T interface.

To verify that this system images the far-field plane, I place a clear film with an array of thin ($\sim 50 \mu m$-wide) lines of spacing 1 mm at the input to the vacuum chamber. I then send a probe beam down the optical axis of the imaging system. The result is shown in Fig. A.6, where (a) shows the case without the film, and (b) shows the case with the film. A simulation of the transparency used is shown in Fig. A.6(c). Figure Fig. A.6(b) is the Fourier transform of a Gaussian beam passing through the array of lines shown in Fig. A.6(c), which verifies that the imaging plane
Figure A.6: Far field calibration. (a) A probe beam sent through the vacuum chamber and down the axis of the imaging system shown in Fig. A.5. (b) The image of the probe beam at the camera after placing a clear film with an array of lines just outside the vacuum chamber. (c) A simulation of the film transparency, with a line spacing of 1 mm and a line thickness of $\sim 50 \mu m$.

is in the far field. The thickness of the lines in this case corresponds to the distance between the lines of the film.

A.4 Detectors

To measure the power in the generated fields, I use a Hamamatsu Avalanche Photodiode (APD, model C5460), of bandwidth 10 MHz and gain $1.5 \times 10^6 \text{ V/W}$. For experiments described in this thesis that required two detectors, I also use a variable-gain Hamamatsu Photomultiplier Tube (PMT, model H6578-20).
Appendix B

Multi-level Atom Theory

In this appendix, I provide additional algebra for the work shown in Ch. 5. This includes both calculations for the index of refraction of the pump-pump lattice in the multi-level-atom, lin⊥lin polarization configuration, as well as for the stability analysis.

B.1 Calculations for the applied optical lattice

From the work shown in Secs. 7.2.1 and 7.2.3, Eq. 7.38 becomes, with \( \vec{P} = \vec{P}^+ + \vec{P}^- \), \( \vec{P}^+ = P_{g=+1/2;\epsilon=+3/2}^{+} \hat{\sigma}^+ + P_{g=+1/2;\epsilon=-1/2}^{+} \hat{\sigma}^- \), and \( \vec{P}^- = P_{g=-1/2;\epsilon=-3/2}^{-} + P_{g=-1/2;\epsilon=+1/2}^{-} \hat{\sigma}^+ \),

\[
P_{g=+1/2;\epsilon=+3/2} = -\frac{|\mu|^2}{\sqrt{2}} \frac{\left[ -F(z)e^{ikz} + B(z)e^{-ikz} \right]}{\sqrt{h^2(\Delta^2 + \Gamma^2/4)}} (\Delta - i\Gamma/2).
\]

\[
\left[ 1 - \frac{8|\mu|^2}{2h^2\Gamma^2} \frac{\left[ |F(z)|^2 + |B(z)|^2 - F(z)B^*(z)e^{2ikz} - F^*(z)B(z)e^{-2ikz} \right]}{1 + 4\Delta^2/\Gamma^2} \right] \cdot \sum_{j=-\infty}^{\infty} \eta_j^+ e^{2i(k'z-\pi/2)j} \quad (B.1)
\]
and

\[ P_{g=e=+1/2, \epsilon=-1/2} = -\frac{1}{3} \frac{\mu^2}{\sqrt{2}} \frac{[F(z)e^{ikz} + B(z)e^{-ikz}] (\Delta - i\Gamma/2)}{\sqrt{2}\hbar(\Delta^2 + \Gamma^2/4)} \cdot \sum_{j=-\infty}^{\infty} \eta_j^+ e^{2i(k'x-j\pi/2)} \eta_j \]  \hspace{1cm} \text{(B.2)}

with analogous equations for the \( m_J = -1/2 \) ground state.

The wave equation is

\[ \nabla^2 \vec{E} - \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{P}}{\partial t^2} \]  \hspace{1cm} \text{(B.3)}

Recall that the electric field is

\[ \vec{E} = \left\{ \begin{array}{l} \left[-F(z)e^{ikz} + B(z)e^{-ikz}\right] \frac{\vec{\sigma}^+}{\sqrt{2}} + \left[F(z)e^{ikz} + B(z)e^{-ikz}\right] \frac{\vec{\sigma}}{\sqrt{2}} \right\} e^{-i\omega t} + \text{c.c.} \]  \hspace{1cm} \text{(B.4)}

I consider only \( e^{-i\omega t} \) terms and make the slowly-varying amplitude approximation. This gives rise to the following coupled-amplitude equations:

\[ \nabla^2 \left[-F(z)e^{i(kz-\omega t)} + B(z)e^{i(kz-\omega t)} \right] = -\frac{\omega^2}{\epsilon_0 c^2} \frac{\tau^2}{\sqrt{2}} \left( \vec{P}^+ + \vec{P}^- \right) \cdot \vec{\sigma}^+ e^{-i\omega t} \]  \hspace{1cm} \text{(B.5)}

and

\[ \nabla^2 \left[F(z)e^{i(kz-\omega t)} + B(z)e^{i(kz-\omega t)} \right] = -\frac{\omega^2}{\epsilon_0 c^2} \frac{\tau^2}{\sqrt{2}} \left( \vec{P}^+ + \vec{P}^- \right) \cdot \vec{\sigma}^- e^{-i\omega t}. \]  \hspace{1cm} \text{(B.6)}
These become

\[
\begin{align*}
\left[k^2 F(z)e^{i(kz-\omega t)} - 2ik \frac{\partial F(z)}{\partial z} e^{i(kz-\omega t)} - \frac{\partial^2 F(z)}{\partial z^2} e^{i(kz-\omega t)} - k^2 B(z) e^{i(-kz-\omega t)} + \\
- 2ik \frac{\partial B(z)}{\partial z} e^{i(-kz-\omega t)} + \frac{\partial^2 B(z)}{\partial z^2} e^{i(-kz-\omega t)} \right] + \\
\frac{\omega^2}{c^2} (-F(z)e^{i(kz-\omega t)} + B(z)e^{i(-kz-\omega t)}) = \frac{-\omega^2}{\epsilon_0 c^2} \sqrt{2} (\vec{P}^+ + \vec{P}^-) \cdot \hat{\sigma}^+ e^{-i\omega t}
\end{align*}
\]

\[\text{(B.7)}\]

and

\[
\begin{align*}
\left[-k^2 F(z)e^{i(kz-\omega t)} + 2ik \frac{\partial F(z)}{\partial z} e^{i(kz-\omega t)} + \frac{\partial^2 F(z)}{\partial z^2} e^{i(kz-\omega t)} - k^2 B(z) e^{i(-kz-\omega t)} + \\
- 2ik \frac{\partial B(z)}{\partial z} e^{i(-kz-\omega t)} + \frac{\partial^2 B(z)}{\partial z^2} e^{i(-kz-\omega t)} \right] + \\
\frac{\omega^2}{c^2} (F(z)e^{i(kz-\omega t)} + B(z)e^{i(-kz-\omega t)}) = \frac{-\omega^2}{\epsilon_0 c^2} \sqrt{2} (\vec{P}^+ + \vec{P}^-) \cdot \hat{\sigma}^- e^{-i\omega t}.
\end{align*}
\]

\[\text{(B.8)}\]

Making the rotating wave approximation and noting that \(k = \omega/c\), these become

\[
\begin{align*}
\frac{\partial F(z)}{\partial z} e^{ikz} + \frac{\partial B(z)}{\partial z} e^{-ikz} = \frac{-ik}{2\epsilon_0} \sqrt{2} (\vec{P}^+ + \vec{P}^-) \cdot \hat{\sigma}^+
\end{align*}
\]

\[\text{(B.9)}\]

and

\[
\begin{align*}
\frac{\partial F(z)}{\partial z} e^{ikz} - \frac{\partial B(z)}{\partial z} e^{-ikz} = \frac{ik}{2\epsilon_0} \sqrt{2} (\vec{P}^+ + \vec{P}^-) \cdot \hat{\sigma}^-.
\end{align*}
\]

\[\text{(B.10)}\]
Keeping only the phase-matched or nearly phase-matched terms, Eq. B.7 becomes

\[
\frac{\partial F(z)}{\partial z} e^{ikz} + \frac{\partial B(z)}{\partial z} e^{-ikz} = \frac{ik |\mu|^2 (\Delta - i\Gamma/2)}{2\epsilon_0 \hbar (\Delta^2 + \Gamma^2/4)}.
\]

\[
\left\{ \begin{array}{l}
- (\eta_0^+ + \eta_1^+ e^{-2ik'z + i\pi}) F(z) e^{ikz} + (\eta_0^- + \eta_1^- e^{2ik'z - i\pi}) B(z) e^{-ikz} \\
1 - \frac{4|\mu|^2}{\hbar^2 \Gamma^2} \left[ \frac{|F(z)|^2}{1 + 4\Delta^2/\Gamma^2} \right] + \\
- \frac{4|\mu|^2}{\hbar^2 \Gamma^2} \left[ \frac{- (\eta_0^- + \eta_1^+ e^{2ik'z - i\pi}) F(z) B(z) e^{-ikz} - (\eta_0^- + \eta_1^- e^{2ik'z - i\pi}) B(z) F(z) e^{-3ikz}}{1 + 4\Delta^2/\Gamma^2} \right] + \\
- \frac{4|\mu|^2}{\hbar^2 \Gamma^2} \left[ \frac{- (\eta_0^- + \eta_1^- e^{2ik'z}) F(z) B(z) e^{-ikz} + (\eta_0^- + \eta_1^- e^{2ik'z}) B(z) F(z) e^{-ikz}}{1 + 4\Delta^2/\Gamma^2} \right] \\
\end{array} \right\} + \frac{ik |\mu|^2 (\Delta - i\Gamma/2)}{2\epsilon_0 \hbar (\Delta^2 + \Gamma^2/4)}.
\]

\[
\left\{ \begin{array}{l}
- (\eta_0^- + \eta_1^- e^{-2ik'z}) F(z) e^{ikz} + (\eta_0^- + \eta_1^- e^{2ik'z}) B(z) e^{-ikz} \\
1 - \frac{4|\mu|^2}{3\hbar^2 \Gamma^2} \left[ \frac{|F(z)|^2}{1 + 4\Delta^2/\Gamma^2} \right] + \\
- \frac{4|\mu|^2}{3\hbar^2 \Gamma^2} \left[ \frac{- (\eta_0^- + \eta_1^- e^{2ik'z}) F(z) B(z) e^{-ikz} + (\eta_0^- + \eta_1^- e^{2ik'z}) B(z) F(z) e^{-3ikz}}{1 + 4\Delta^2/\Gamma^2} \right] + \\
- \frac{4|\mu|^2}{3\hbar^2 \Gamma^2} \left[ \frac{- (\eta_0^- + \eta_1^- e^{-4ik'z}) F(z) B(z) e^{-2ikz} - (\eta_0^- + \eta_1^- e^{4ik'z}) F(z) B(z) e^{-2ikz}}{1 + 4\Delta^2/\Gamma^2} \right] \\
\end{array} \right\}, \quad (B.11)
\]
with a similar equation for Eq. B.8. Decoupling this into phase-matched terms, Eq. B.11 becomes

\[
\frac{\partial F(z)}{\partial z} e^{ikz} = \frac{ik}{2\epsilon_0} \frac{\mu^2(\Delta - i\Gamma/2)}{\hbar(\Delta^2 + \Gamma^2/4)} \left\{ -\eta_0^+ F(z) e^{ikz} - \eta_1^+ e^{2ik'z} B(z) e^{-ikz} \right\} \left[ 1 - \frac{4|\mu|^2 [ |F(z)|^2 + |B(z)|^2 ]}{\hbar^2 \Gamma^2 (1 + 4\Delta^2/\Gamma^2)} \right] +
\]

\[
-\frac{4|\mu|^2}{\hbar^2 \Gamma^2} \left[ -\eta_0 F(z) |B(z)|^2 e^{ikz} - \eta_1^+ e^{2ik'z} B(z) |F(z)|^2 e^{-ikz} \right] \left( 1 + 4\Delta^2/\Gamma^2 \right) +
\]

\[
\frac{1}{3} \left[ -\eta_0^+ F(z) e^{ikz} + \eta_1^- e^{2ik'z} B(z) e^{-ikz} \right] \left[ 1 - \frac{4|\mu|^2 [ |F(z)|^2 + |B(z)|^2 ]}{3\hbar^2 \Gamma^2 (1 + 4\Delta^2/\Gamma^2)} \right] +
\]

\[
-\frac{4|\mu|^2}{3\hbar^2 \Gamma^2} \left[ -\eta_0^+ F(z) |B(z)|^2 e^{ikz} + \eta_1^- e^{2ik'z} B(z) |F(z)|^2 e^{-ikz} \right] \left( 1 + 4\Delta^2/\Gamma^2 \right) +
\]

\[
\frac{4|\mu|^2}{3\hbar^2 \Gamma^2} \left[ \eta_1^- e^{-2ik'z} F(z) F(z) B^*(z) e^{3ikz} \right] \left( 1 + 4\Delta^2/\Gamma^2 \right) +
\]

\[
-\frac{4|\mu|^2}{3\hbar^2 \Gamma^2} \left[ -\eta_2^- e^{4ik'z} F^*(z) B(z) e^{-2ikz} B(z) e^{-ikz} \right] \left( 1 + 4\Delta^2/\Gamma^2 \right) \right\} \right. \)

(B.12)
Assuming equal-intensity pump beams \(|F(z)|^2 = |B(z)|^2\), this becomes

\[
\frac{\partial F(z)}{\partial z} = \frac{ik}{2\epsilon_0 \hbar \Gamma} \left( \frac{2|\mu|^2(2\Delta/\Gamma - i)}{1 + 4\Delta^2/\Gamma^2} \right) \cdot \frac{2}{3} \left\{ -2\eta_0^+ F(z) - \eta_1^+ B(z)e^{2i(k'-k)z} + \right.
\]

\[
\frac{12|\mu|^2|F(z)|^2}{\hbar^2 \Gamma^2 (1 + 4\Delta^2/\Gamma^2)} \left[ 2\eta_0^+ F(z) + \eta_1^+ B(z)e^{2i(k'-k)z} \right] +
\]

\[
\left( \frac{4|\mu|^2}{\hbar^2 \Gamma^2 (1 + 4\Delta^2/\Gamma^2)} \left[ \eta_{-1}^+ e^{-2i(k'-k)z} F(z)^2 B^*(z) + 2\eta_2^+ e^{-4i(k'-k)z} F^*(z)B(z)B(z) \right] \right) \right\}, \quad (B.13)
\]

and from Eq. B.8,

\[
\frac{\partial B(z)}{\partial z} = \frac{ik}{2\epsilon_0 \hbar \Gamma} \left( \frac{2|\mu|^2(2\Delta/\Gamma - i)}{1 + 4\Delta^2/\Gamma^2} \right) \cdot \frac{2}{3} \left\{ 2\eta_0^+ B(z) + \eta_{-1}^+ e^{-2i(k'-k)z} F(z) +
\]

\[
- \frac{12|\mu|^2|F(z)|^2}{\hbar^2 \Gamma^2 (1 + 4\Delta^2/\Gamma^2)} \left[ 2\eta_0^+ B(z) + \eta_{-1}^+ e^{-2i(k'-k)z} F(z) \right] +
\]

\[
- \frac{4|\mu|^2}{\hbar^2 \Gamma^2 (1 + 4\Delta^2/\Gamma^2)} \left[ \eta_1^+ e^{2i(k'-k)z} B(z)B(z)F^*(z) + \right.
\]

\[
\left. \left. 2\eta_2^+ e^{-4i(k'-k)z} F(z)B^*(z)B(z) \right] \right\}. \quad (B.14)
\]

I define \(\delta = k' - k\) and let

\[F(z) = \tilde{F}e^{i\delta z}, \quad (B.15)\]

and

\[B(z) = \tilde{B}e^{-i\delta z}. \quad (B.16)\]

Also then

\[
\frac{\partial F(z)}{\partial z} = i\delta \tilde{F}e^{i\delta z} \quad (B.17)
\]
and

$$\frac{\partial B(z)}{\partial z} = -i\delta \tilde{B} e^{-i\delta z}. \quad (B.18)$$

Then Eqs. B.13 and B.14 simplify to the solution for the phase shift:

$$\delta = \frac{k}{2\varepsilon_0} \frac{2|\mu|^2(2\Delta/\Gamma - i)}{\hbar \Gamma (1 + 4\Delta^2/\Gamma^2)} \cdot \frac{2}{3} \left\{ -2\eta_0 - \eta_1 + \frac{12|\mu|^2|\tilde{F}|^2}{\hbar^2 \Gamma^2 (1 + 4\Delta^2/\Gamma^2)} [2\eta_0 + \eta_1] + \frac{4|\mu|^2\tilde{F}\tilde{B}}{\hbar^2 \Gamma^2 (1 + 4\Delta^2/\Gamma^2)} [\eta_{-1} + 2\eta_2] \right\}. \quad (B.19)$$

Rewriting this in terms of the total intensity $I_{\text{tot}} = 4\varepsilon_0 c |\tilde{E}_p|^2$, where $\tilde{E}_p = \tilde{F} = \tilde{B}$, this becomes

$$\delta = \frac{k}{2\varepsilon_0} \frac{2|\mu|^2(2\Delta/\Gamma - i)}{\hbar \Gamma (1 + 4\Delta^2/\Gamma^2)} \cdot \frac{2}{3} \left\{ -2\eta_0^+ - \eta_1^+ + \frac{3|\mu|^2 I_{\text{tot}}}{\hbar^2 \Gamma^2 \varepsilon_0 c (1 + 4\Delta^2/\Gamma^2)} [2\eta_0^+ + \eta_1^+] + \frac{|\mu|^2 I_{\text{tot}}}{\hbar^2 \Gamma^2 \varepsilon_0 c (1 + 4\Delta^2/\Gamma^2)} [\eta_{-1}^+ + 2\eta_2^+] \right\}. \quad (B.20)$$

The saturation intensity goes as

$$I_{\text{sat}} = \frac{\hbar^2 \Gamma^2 \varepsilon_0 c}{2|\mu|^2} \rightarrow 2I_{\text{sat}} = \frac{\hbar^2 \Gamma^2 \varepsilon_0 c}{|\mu|^2}. \quad (B.21)$$

The off-resonant saturation intensity is

$$I_{s\Delta} = I_{\text{sat}} (1 + 4\Delta^2/\Gamma^2), \quad (B.22)$$

and I define $\tilde{I} = I_{\text{tot}}/I_{s\Delta}$. Therefore,

$$\delta = \frac{k}{2\varepsilon_0} \frac{2|\mu|^2(2\Delta/\Gamma - i)}{\hbar \Gamma (1 + 4\Delta^2/\Gamma^2)} \cdot \frac{2}{3} \left\{ -2\eta_0^+ - \eta_1^+ + \frac{3}{2} \tilde{I} [2\eta_0^+ + \eta_1^+] + \frac{\tilde{I}}{2} [\eta_{-1}^+ + 2\eta_2^+] \right\}. \quad (B.23)$$

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Note that the absorption coefficient goes as $\alpha_0 = 2k n_o |\mu|^2 / (\varepsilon_0 h \Gamma)$ and the linear susceptibility $\chi_{\text{lin}} = - (\alpha_0/k)((2\Delta/\Gamma - i)/(1 + 4\Delta^2/\Gamma^2))$. Therefore, the phase shift is

$$\delta = - \frac{k}{4} \chi_{\text{lin}} \frac{2}{3} \left\{ (-1 + 3\tilde{I}) + (-1 + 2\tilde{I}) \frac{I_1(-\zeta)}{I_0(-\zeta)} + 2\tilde{I} \frac{I_2(-\zeta)}{I_0(-\zeta)} \right\}, \tag{B.24}$$

which simplifies to Eq. 7.42.

### B.1.1 Total phase shift versus phase shift of circular component

Equation 7.42 defines the phase shift experienced by one of the circularly-polarized components of one of the input fields. Because the input field is actually linearly polarized, I must calculate the total phase shift experienced by the input field. The electric field of one of the input fields inside the material goes as

$$B_p(z) e^{i(-kz - \omega t)} \hat{y} = B(z) e^{i(-kz - \omega t)} \left( e^{i\phi} \frac{\hat{\sigma}^+ + \hat{\sigma}^-}{\sqrt{2}} \right), \tag{B.25}$$

or

$$B(z) e^{i(-kz - \omega t)} \hat{y} = \left( \frac{\tilde{B} e^{-i\delta z} \hat{\sigma}^+ + \tilde{B} e^{-i\delta z} \hat{\sigma}^-}{\sqrt{2}} \right) e^{i(-kz - \omega t)}. \tag{B.26}$$

It follows that

$$B(z) e^{i(-kz - \omega t)} \hat{y} = \tilde{B} e^{i(-k + 2\delta)z - \omega t} \hat{y}. \tag{B.27}$$

Therefore, the total phase shift experienced by one of the input fields is $\delta_{\text{tot}} = 2\delta$, or

$$\delta_{\text{tot}} = \frac{k}{2} \chi_{\text{lin}} \frac{2}{3} \left\{ (-1 + 3\tilde{I}) + (-1 + 2\tilde{I}) \frac{I_1(-\zeta)}{I_0(-\zeta)} + 2\tilde{I} \frac{I_2(-\zeta)}{I_0(-\zeta)} \right\}. \tag{B.28}$$

### B.1.2 Proof for field amplitudes

To obtain Eq. B.28, I also make the assumption $F(z) = \tilde{F} e^{i\delta z}$ and $B(z) = \tilde{B} e^{-i\delta z}$. This assumption is only valid if $|F(z)|^2$ and $|B(z)|^2$ are constants of the motion
(independent of $z$). To prove this, I consider

$$\frac{\partial |F(z)|^2}{\partial z} = \frac{\partial F(z)}{\partial z} \cdot F^*(z) + F(z) \frac{\partial F^*(z)}{\partial z}, \quad (B.29)$$

or

$$\frac{\partial |F(z)|^2}{\partial z} = (i\delta F(z)) \cdot F^*(z) + F(z) (-i\delta F^*(z)). \quad (B.30)$$

When $\delta$ is real, this implies

$$\frac{\partial |F(z)|^2}{\partial z} = 0. \quad (B.31)$$

The same procedure also proves that $\partial |B(z)|^2 / \partial z = 0$. This implies that both $|F(z)|^2$ and $|B(z)|^2$ are constants of the motion when $\delta$ is real.

When $\delta$ has an imaginary component (i.e. when I account for absorption and keep the $i$-dependence in $\chi_{\text{lin}}$), $|F(z)|^2$ and $|B(z)|^2$ are not necessarily constants of the motion. I can, however, show that $|F(z)|^2 + |B(z)|^2$ is a constant of the motion if $|F(z)|^2 = |B(z)|^2$. I define $\delta = \delta' + i\delta''$, so that

$$F(z) = \bar{F} e^{(i\delta' - \delta'')z} \quad (B.32)$$

and

$$B(z) = \bar{B} e^{(-i\delta' + \delta'')z}. \quad (B.33)$$

Then

$$\frac{\partial (|F(z)|^2 + |B(z)|^2)}{\partial z} = (i\delta' - \delta'')|F(z)|^2 +$$

$$( -i\delta' - \delta'')|F(z)|^2 + ( -i\delta' + \delta'')|B(z)|^2 + (i\delta' + \delta'')|B(z)|^2, \quad (B.34)$$

and

$$\frac{\partial (|F(z)|^2 + |B(z)|^2)}{\partial z} = -2\delta''|F(z)|^2 + 2\delta''|B(z)|^2. \quad (B.35)$$

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So in the case of equal intensity fields, \( \partial(|F(z)|^2 + |B(z)|^2)/\partial z = 0 \), and the above equations are still valid even when we account for absorption. However, for unbalanced optical field intensities, this is not valid, and the above integrals become much more difficult to solve with the additional \( z \)-dependence.

### B.2 Applying the shift theorem

The solution in Eq. B.28 also assumes that the Fourier coefficients \( \eta_j^+ = \eta_j^- \). To prove this, I define the generic atomic density

\[
\eta(z) = \eta^+(z) + \eta^-(z) = \sum_{j=-\infty}^{\infty} \bar{\eta}_j^+ e^{2ik'z*} + \sum_{j=-\infty}^{\infty} \bar{\eta}_j^- e^{2ik'z*}, \quad (B.36)
\]

where

\[
\bar{\eta}_j^- = \frac{1}{\lambda'/2} \int_{-\lambda'/4}^{\lambda'/4} \bar{\eta}\exp\left[\frac{-U^\pm(z)}{k_B T}\right] e^{-2ik'z*} dz. \quad (B.37)
\]

Here,

\[
U^+(z) = U_0 - C^2 U_0 \cos^2(k'z) \quad (B.38)
\]

and

\[
U^-(z) = U_0 - C^2 U_0 \sin^2(k'z). \quad (B.39)
\]

Therefore, \( U^-(k'z) = U^+(k'z - \pi/2) \). The shift theorem says that if

\[
F(k'z) = \int f(k'z)e^{-2ik'z*} dz, \quad (B.40)
\]

then

\[
\int f(k'z - k'z_0)e^{-2ik'z*} dz = e^{-2ik'z_0*} F(k'z). \quad (B.41)
\]

Therefore,

\[
\bar{\eta}_j^+ = \eta_j^- e^{-2ik'z_0*} = \bar{\eta}_j^+ e^{-2i(\pi/2)*}. \quad (B.42)
\]
I then define $\eta^+_j$ via

$$\tilde{\eta}^+_j = \eta^-_j e^{-2i(\pi/2)\phi_j} = \eta^+_j e^{2i(-\pi/2)\phi_j},$$  \hspace{1cm} (B.43)

such that

$$\tilde{\eta}^+_j e^{2ik'z\phi_j} = \eta^+_j e^{2i(k'z-\pi/2)\phi_j},$$  \hspace{1cm} (B.44)

which returns the density distribution defined in the above sections, with $\eta^+_j = \eta^-_j$.

### B.3 Calculations for the stability analysis

Accounting for more optical fields to solve for the polarization in the presence of optical patterns, I obtain Eqs. 7.53 and 7.54. To solve for the threshold condition at which these fields are generated, I solve the wave equation for each generated field. For simplicity in this appendix, I only present the calculations for terms that are phase-matched or nearly phase-matched to $f_+ (z, r) e^{i(k \cos \theta z - k \sin \theta r - \omega t)}$.

The expansion of Eq. 7.54 goes as

$$\vec{P}^-(z, r) = -\frac{2\tilde{\mu}_g^- (\tilde{\mu}_g^- \cdot \vec{E}) (2\Delta - i)}{h\Gamma (1 + 4\Delta^2)} \left[ \sum_{j=-\infty}^{\infty} \eta^-_j e^{2ik'z\phi_j} \right] +$$

$$-\frac{2\tilde{\mu}_g^- (\tilde{\mu}_g^- \cdot \vec{E}) (2\Delta - i)}{h\Gamma (1 + 4\Delta^2)} \left[ \sum_{j=-\infty}^{\infty} \eta^-_j e^{2ik'z\phi_j} \right] \left( -\frac{8\Delta (\tilde{\mu}_g^- \cdot \vec{E}_{pp})^* (\tilde{\mu}_g^- \cdot \vec{E}_{pp})}{(h\Gamma)^2 T (1 + 4\Delta^2)} \right) +$$

$$-\frac{2\tilde{\mu}_g^- (\tilde{\mu}_g^- \cdot \vec{E}) (2\Delta - i)}{h\Gamma (1 + 4\Delta^2)} \left[ \frac{8|\tilde{\mu}_g^- \cdot \vec{E}|^2}{h^2 \Gamma^2 (1 + 4\Delta^2)} \right] \left[ \sum_{j=-\infty}^{\infty} \eta^-_j e^{2ik'z\phi_j} \right] +$$

$$-\frac{2\tilde{\mu}_g^- (\tilde{\mu}_g^- \cdot \vec{E}) (2\Delta - i)}{h\Gamma (1 + 4\Delta^2)} \left[ \frac{8|\tilde{\mu}_g^- \cdot \vec{E}|^2}{h^2 \Gamma^2 (1 + 4\Delta^2)} \right] \left[ \sum_{j=-\infty}^{\infty} \eta^-_j e^{2ik'z\phi_j} \right] \left( \frac{8\Delta (\tilde{\mu}_g^- \cdot \vec{E}_{pp})^* (\tilde{\mu}_g^- \cdot \vec{E}_{pp})}{(h\Gamma)^2 T (1 + 4\Delta^2)} \right). \hspace{1cm} (B.45)$$

I simplify this equation and keep only the real part (ignoring absorption). The component of the polarization corresponding to the $g = -1/2 \rightarrow e = -3/2$ transition...
\[
\bar{P}_{g=-1/2,e=-3/2}^{-}(z, r) = -\frac{4|\mu|^2 \tilde{\Delta}}{\sqrt{2} \hbar \Gamma (1 + 4\Delta^2)} \left[ \sum_{j=-\infty}^{\infty} \eta_j^{-} e^{2ik'z* j} \right] E^{-} + \\
- \frac{4|\mu|^2 \tilde{\Delta}}{\sqrt{2} \hbar \Gamma (1 + 4\Delta^2)} \left[ - \frac{4\Delta|\mu|^2}{(\hbar \Gamma)^2 T (1 + 4\Delta^2)} \right] \left[ \sum_{j=-\infty}^{\infty} \eta_j^{-} e^{2ik'z* j} \right] E^{-}(E^{-})^* E_{pp} + \\
- \frac{4|\mu|^2 \tilde{\Delta}}{\sqrt{2} \hbar \Gamma (1 + 4\Delta^2)} \left[ - \frac{4|\mu|^2}{\hbar^2 \Gamma^2 (1 + 4\Delta^2)} \right] \left[ \sum_{j=-\infty}^{\infty} \eta_j^{-} e^{2ik'z* j} \right] E^{-}(E^{-})^* E^{-} + \\
- \frac{4|\mu|^2 \tilde{\Delta}}{\sqrt{2} \hbar \Gamma (1 + 4\Delta^2)} \left[ - \frac{4|\mu|^2}{\hbar^2 \Gamma^2 (1 + 4\Delta^2)} \right] \left[ \sum_{j=-\infty}^{\infty} \eta_j^{-} e^{2ik'z* j} \right] E^{-}(E^{-})^* E_{pp}^* E_{pp}^{-}.
\]

where \( E^{-} = \left[ F(z)e^{i(kz-\omega t)} + B(z)e^{i(-kz-\omega t)} + \sigma f_{+}(z, r)e^{i(k\cos\theta z - k\sin\theta r - \omega t)} + \right. \\
\left. \sigma f_{-}(z, r)e^{i(k\cos\theta z + k\sin\theta r - \omega t)} + \sigma b_{+}(z, r)e^{i(-k\cos\theta z - k\sin\theta r - \omega t)} + \sigma b_{-}(z, r)e^{i(-k\cos\theta z + k\sin\theta r - \omega t)} \right], \\
and where I have extracted the \( 1/\sqrt{2} \) from the dipole moment-field dot products into the coefficients of each term. Also recall that the combination \((E_{pp}^{-})^* E_{pp}^{-}\) must consist of exactly one pump field term and one weak field term.

Those terms that are at order \( \alpha \) and phase-matched or nearly phase-matched to
For the last term, there will be many possible combinations of each set of phase-matched or nearly phase-matched terms because this is a very high-order nonlinear
term containing $\chi^{(5)}$ and $\chi^{(7)}$ nonlinearities. This term goes as

$$\frac{\text{last}}{e^{i(k\cos \theta - k\sin \theta - \omega t)}} = \frac{4|\mu|^2 \tilde{\Delta}}{\sqrt{2}\hbar \Gamma (1 + 4\Delta^2)} \left[ \frac{4|\mu|^2}{\hbar^2 \Gamma^2 (1 + 4\Delta^2)} \right] \left( \frac{4\tilde{\Delta}|\mu|^2}{(\hbar \Gamma)^2 T(1 + 4\Delta^2)} \right) \cdot \left\{ \begin{array}{l}
\eta_0 |F(z)|^4 f_+ + 2\eta_0 |F(z)|^2 |B(z)|^2 f_+ + \\
+ 3\eta_0 |F(z)|^2 F(z)B(z) b^*_b + 2\eta_0 |F(z)|^2 F(z)^* B(z) b_+ e^{2i(k - k\cos \theta)z} + \\
\eta_0 |F(z)|^2 F(z)^2 f^*_* e^{2i(k - k\cos \theta)z} + 2\eta_1 |F(z)|^2 B(z) F(z) f_+ + \\
2\eta_{-1} |F(z)|^2 F(z) B(z) f_+ + 3\eta_1 |F(z)|^2 B(z)^2 b^*_b + \eta_{-1} |F(z)|^2 F(z)^2 b^*_b + \\
+ \eta_1 |F(z)|^4 b_+ e^{2i(k' - k\cos \theta)z} + 2\eta_1 |F(z)|^2 |B(z)|^2 b_+ e^{2i(k' - k\cos \theta)z} + \\
3\eta_1 |F(z)|^2 F(z) B(z) f^*_* e^{2i(k' - k\cos \theta)z} + 2\eta_2 |F(z)|^2 B(z) F^*(z) b_+ e^{2i(k' - k\cos \theta)z} + \\
3\eta_2 |F(z)|^2 B(z)^2 f_+ e^{4ik'z - 2ik\cos \theta}z \end{array} \right\} + \\
+ \eta_0 |B(z)|^2 F(z) B(z) b^*_b + 2\eta_0 |B(z)|^2 F(z)^* B(z) b_+ e^{2i(k - k\cos \theta)z} + \\
3\eta_0 |B(z)|^2 F(z)^2 f^*_* e^{2i(k - k\cos \theta)z} + 2\eta_1 |B(z)|^2 B(z) F(z) f_+ + \\
2\eta_{-1} |B(z)|^2 F(z) B(z) f_+ + \eta_1 |B(z)|^2 B(z)^2 b^*_b + 3\eta_{-1} |B(z)|^2 F(z)^2 b^*_b + \\
+ 2\eta_1 |B(z)|^2 |F(z)|^2 b_+ e^{2i(k' - k\cos \theta)z} + \eta_1 |B(z)|^4 b_+ e^{2i(k' - k\cos \theta)z} + \\
3\eta_1 |B(z)|^2 F(z) B(z) f^*_* e^{2i(k' - k\cos \theta)z} + \\
+ 2\eta_2 |B(z)|^2 B(z) F^*(z) b_+ e^{4ik'z - 2ik\cos \theta}z + \eta_2 |B(z)|^2 B(z)^2 f^*_* e^{4ik'z - 2ik\cos \theta}z \right\} + \\
+ \eta_2 B(z)^2 F^*(z)^2 f^*_* e^{4ik'z - 4ikz} + \eta_{-2} F(z)^2 (B^*(z))^2 f_+ e^{4ik'z - 4ikz} + \\
\eta_2 B(z)^3 F^*(z)^2 b^*_b e^{4ik'z - 4ikz} + \eta_{-2} F(z)^3 B^* b^*_b e^{4ik'z - 4ikz} + \\
\eta_{-1} F(z)^2 (B^*(z))^2 b_+ e^{-2ik'z + 4ikz - 2ik\cos \theta}z + \eta_{-1} F(z)^3 B^*(z) f^* e^{-2ik'z + 4ikz - 2ik\cos \theta}z + \\
\eta_3 B(z)^2 (F^*(z))^2 b_+ e^{6ik'z - 4ikz - 2ik\cos \theta}z + \eta_3 B(z)^3 F^*(z) f^* e^{6ik'z - 4ikz - 2ik\cos \theta}z \right\}.$$

(4.8)
The component of the polarization for the other transition from the $m_j = -1/2$ ground state is

\[
\tilde{p}^{-}_{g=-1/2,e=+1/2}(z, r) = -\frac{4|\mu|^2 \tilde{\Delta}}{\sqrt{2} \hbar \Gamma(1 + 4\Delta^2)} \frac{1}{3} \left[ \eta_0 f_+ - \eta_1 b_+ e^{2i(k' - k \cos \theta)z} \right] + \\
- \frac{4|\mu|^2 \tilde{\Delta}}{\sqrt{2} \hbar \Gamma(1 + 4\Delta^2)} \frac{1}{9} \left( -\frac{4\tilde{\Delta}|\mu|^2}{(\hbar \Gamma)^2 T(1 + 4\Delta^2)} \right) \left\{ \eta_0(|F(z)|^2 + |B(z)|^2)f_+ + \\
+ 2\eta_0 F(z) B(z) b^*_+ + \eta_0 F(z) B(z)^* b_+ e^{2i(k - k \cos \theta)z} + \eta_0 F(z)^2 f^*_e e^{2i(k - k \cos \theta)z} + \\
- \eta_1 B(z) F(z)^* f_+ - \eta_1 F(z) B(z)^* f_+ - \eta_1 B(z)^2 b^*_+ - \eta_1 F(z)^2 b^*_+ + \\
- \eta_1(|F(z)|^2 + |B(z)|^2)b_+ e^{2i(k' - k \cos \theta)z} - 2\eta_1 F(z) B(z) f^*_e e^{2i(k' - k \cos \theta)z} + \\
+ \eta_2 B(z) F^*(z) b_+ e^{(4ik' - 2ik - 2ik \cos \theta)z} + \eta_2 B(z)^2 f^*_e e^{4ik'z - 2ikz - 2ik \cos \theta} \right\} \\
- \frac{4|\mu|^2 \tilde{\Delta}}{\sqrt{2} \hbar \Gamma(1 + 4\Delta^2)} \left[ -\frac{4|\mu|^2}{\hbar^2 \Gamma^2(1 + 4\Delta^2)} \right] \frac{1}{9} \left[ 2\eta_0(|F(z)|^2 + |B(z)|^2)f_+ + \\
+ 2\eta_0 F(z) B(z) b^*_+ + 2\eta_0 F(z) B(z)^* b_+ e^{2i(k - k \cos \theta)z} + \eta_0 F(z)^2 f^*_e e^{2i(k - k \cos \theta)z} + \\
- 2\eta_1 B(z) F(z)^* f_+ - 2\eta_1 F(z) B(z)^* f_+ - \eta_1 B(z)^2 b^*_+ - \eta_1 F(z)^2 b^*_+ + \\
- 2\eta_1(|F(z)|^2 + |B(z)|^2)b_+ e^{2i(k' - k \cos \theta)z} - 2\eta_1 F(z) B(z) f^*_e e^{2i(k' - k \cos \theta)z} + \\
+ 2\eta_2 B(z) F^*(z) b_+ e^{2i(k' - k \cos \theta)z} + \eta_2 B(z)^2 f^*_e e^{4ik'z - 2ikz - 2ik \cos \theta} \right) + \\
- \frac{4|\mu|^2 \tilde{\Delta}}{\sqrt{2} \hbar \Gamma(1 + 4\Delta^2)} \left[ -\frac{4|\mu|^2}{\hbar^2 \Gamma^2(1 + 4\Delta^2)} \right] \left( -\frac{4\tilde{\Delta}|\mu|^2}{(\hbar \Gamma)^2 T(1 + 4\Delta^2)} \right) \frac{1}{27} \cdot P_{\text{high-order}}, \quad (B.49)
\]
where

\[
P_{\text{high-order}} = \left\{ \begin{array}{l}
\eta_0 |F(z)|^4 f_+ + 2\eta_0 |F(z)|^2 |B(z)|^2 f_+ + \\
+ 3\eta_0 |F(z)|^2 F(z)B(z)b^* - 2\eta_1 |F(z)|^2 B(z)F(z) f_+ \\
\eta_0 |F(z)|^2 F(z)^2 f e^{2i(k' - kcos\theta)z} - 2\eta_1 |F(z)|^2 B(z)F(z) f_+ \\
- 2\eta_1 |F(z)|^2 F(z)B(z)f_+ - 3\eta_1 |F(z)|^2 B(z)^2 b^* - \eta_1 |F(z)|^2 B(z)^2 b^* + \\
- \eta_1 |F(z)|^4 b^* e^{2i(k' - kcos\theta)z} - 2\eta_1 |F(z)|^2 B(z)^2 b^* + \\
- 3\eta_1 |F(z)|^2 F(z)B(z) f^* e^{2i(k' - kcos\theta)z} + \\
+ 2\eta_2 |F(z)|^2 B(z)F^*(z)b^* e^{2i(k' - kcos\theta)z} + 3\eta_2 |F(z)|^2 B(z)^2 f^* e^{4ik'z - 2ikcos\theta} \\
\end{array} \right. 
\]
I make the non-depleted pump approximation and take $F(z) = \tilde{F} e^{i\delta z}$ and $B(z) = \tilde{B} e^{-i\delta z}$. I also then take $k' = k + \delta$. The above equations are combined via $\tilde{P} = \tilde{P}_{g=-1/2; e=-3/2} + \tilde{P}_{g=-1/2; e=+1/2}$ with $\tilde{F} = \tilde{B}$ to be

\[
\frac{\tilde{P}_{g=-1/2; e=-3/2} + \tilde{P}_{g=-1/2; e=+1/2}}{e^{(k\cos\theta_2 - k\sin\theta_2 - \omega_t)}} = - \frac{4|\mu|^2 \tilde{\Delta}}{\sqrt{2} \hbar \Gamma(1 + 4 \Delta^2)} \left( \frac{4 \tilde{\Delta} |\mu|^2}{(\hbar \Gamma)^2 \tilde{T}(1 + 4 \Delta^2)} \right) \tilde{E}_p^2 \left[ \left( \frac{4}{9} \eta_0 + \frac{8}{9} \eta_1 + \frac{8}{9} \eta_{-1} \right) f_+ + \left( \frac{10}{9} \eta_0 + \frac{8}{9} \eta_1 + \frac{8}{9} \eta_{-1} \right) f^*_+ e^{2i(k' - k\cos\theta)z} + \left( \frac{10}{9} \eta_0 + \frac{8}{9} \eta_1 + \frac{10}{9} \eta_2 \right) f^- e^{2i(k' - k\cos\theta)z} \right] + \\
- \frac{4|\mu|^2 \tilde{\Delta}}{\sqrt{2} \hbar \Gamma(1 + 4 \Delta^2)} \left( \frac{4 \tilde{\Delta} |\mu|^2}{(\hbar \Gamma)^2 \tilde{T}(1 + 4 \Delta^2)} \right) \tilde{E}_p^2 \left[ \left( \frac{4}{9} \eta_0 + \frac{8}{9} \eta_1 + \frac{8}{9} \eta_{-1} \right) f_+ + \left( \frac{10}{9} \eta_0 + \frac{8}{9} \eta_1 + \frac{8}{9} \eta_{-1} \right) f^*_+ e^{2i(k' - k\cos\theta)z} + \left( \frac{10}{9} \eta_0 + \frac{8}{9} \eta_1 + \frac{8}{9} \eta_{-1} \right) f^- e^{2i(k' - k\cos\theta)z} \right] + \\
- \frac{4|\mu|^2 \tilde{\Delta}}{\sqrt{2} \hbar \Gamma(1 + 4 \Delta^2)} \left( \frac{4 \tilde{\Delta} |\mu|^2}{(\hbar \Gamma)^2 \tilde{T}(1 + 4 \Delta^2)} \right) \tilde{E}_p^4 \left\{ \left( \frac{6}{27} \eta_0 + \frac{26}{27} \eta_1 + \frac{26}{27} \eta_{-1} + \frac{28}{27} \eta_2 + \frac{28}{27} \eta_{-2} \right) f_+ + \left( \frac{6}{27} \eta_0 + \frac{26}{27} \eta_1 + \frac{26}{27} \eta_{-1} + \frac{28}{27} \eta_2 + \frac{28}{27} \eta_{-2} \right) f^*_+ e^{2i(k' - k\cos\theta)z} + \left( \frac{6}{27} \eta_0 + \frac{26}{27} \eta_1 + \eta_{-1} + \frac{28}{27} \eta_2 + \frac{26}{27} \eta_{-2} \right) f^- e^{2i(k' - k\cos\theta)z} \right\}. \quad (B.51)
\]
The wave equation is
\[ \nabla^2 \vec{E} - \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \vec{P}}{\partial t^2}. \]  \tag{B.52}

I also investigate the left-hand-side of the wave equation for that weak field component:
\[ \left[ \frac{\partial}{\partial z} \left[ \frac{\partial f_+(z, r)}{\partial z} e^{i(k \cos \theta z - k \sin \theta r - \omega t)} + ik \cos \theta f_+(z, r) e^{i(k \cos \theta z - k \sin \theta r - \omega t)} \right] \right. \]
\[ \left. + \frac{\partial}{\partial x} \left[ \frac{\partial f_+(z, r)}{\partial x} e^{i(k \cos \theta z - k \sin \theta r - \omega t)} - ik \sin \theta f_+(z, r) e^{i(k \cos \theta z - k \sin \theta r - \omega t)} \right] \right] \frac{\hat{\sigma}^-}{\sqrt{2}}. \tag{B.53} \]

Under the rotating wave approximation, this becomes
\[ \left[ 2ik \cos \theta \frac{\partial f_+(z, r)}{\partial z} e^{i(k \cos \theta z - k \sin \theta r - \omega t)} - k^2 \cos^2 \theta f_+(z, r) e^{i(k \cos \theta z - k \sin \theta r - \omega t)} + \\
- 2ik \sin \theta \frac{\partial f_+(z, r)}{\partial x} e^{i(k \cos \theta z - k \sin \theta r - \omega t)} - k^2 \sin^2 \theta f_+(z, r) e^{i(k \cos \theta z - k \sin \theta r - \omega t)} + \\
k^2 f_+(z, r) e^{i(k \cos \theta z - k \sin \theta r - \omega t)} \right] \frac{\hat{\sigma}^-}{\sqrt{2}}. \tag{B.54} \]

or
\[ \left[ 2ik \cos \theta \frac{\partial f_+(z, r)}{\partial z} e^{i(k \cos \theta z - k \sin \theta r - \omega t)} - 2ik \sin \theta \frac{\partial f_+(z, r)}{\partial x} e^{i(k \cos \theta z - k \sin \theta r - \omega t)} \right] \frac{\hat{\sigma}^-}{\sqrt{2}}. \tag{B.55} \]

I take the optical field amplitude variation in $x$ to be very small, such that $\partial f_+ / \partial x \rightarrow 0$. Then the left-hand-side of the wave equation for this field term is
\[ 2ik \cos \theta \frac{\partial f_+}{\partial z} e^{i(k \cos \theta z - k \sin \theta r - \omega t)} \frac{\hat{\sigma}^-}{\sqrt{2}}. \tag{B.56} \]
The full wave equation for those terms that are phase-matched or nearly phase-matched to \( f_+(z, r) e^{i(kz \cos \theta - k_r \sin \theta - \omega t)} \) is then

\[
2ik \cos \theta \frac{\partial f_+}{\partial z} = -\frac{\omega^2}{\epsilon_0 c^2} \frac{4|\mu|^2 \tilde{\Delta}}{\hbar \Gamma (1 + 4\Delta^2)} \left\{ \frac{4}{3} \eta_0 f_+ + \frac{2}{3} \eta_1 b_+ e^{2i(k' - k \cos \theta) z} + \right.
\]
\[
\left. + \left( -\frac{4 \Delta |\mu|^2}{(\hbar \Gamma)^2 (1 + 4\Delta^2)} \right) \tilde{E}_p^2 \left[ \left( \frac{2}{9} \eta_0 + \eta_1 + \eta_{-1} \right) f_+ + \left( \frac{2}{9} \eta_0 + \frac{8}{9} \eta_1 + \frac{8}{9} \eta_{-1} \right) b_+^* \right] + \right.
\]
\[
\left. + \left( \frac{10}{9} \eta_0 + \frac{8}{9} \eta_1 + \frac{10}{9} \eta_2 \right) b_+ e^{2i(k' - k \cos \theta) z} + \left( \frac{10}{9} \eta_0 + \frac{8}{9} \eta_1 + \frac{10}{9} \eta_2 \right) f_+^* e^{2i(k' - k \cos \theta) z} \right] + \right.
\]
\[
\left. + \left( \frac{2}{9} \eta_0 + \frac{8}{9} \eta_1 + \frac{10}{9} \eta_2 \right) b_+ e^{2i(k' - k \cos \theta) z} + \left( \frac{10}{9} \eta_0 + \frac{8}{9} \eta_1 + \frac{10}{9} \eta_2 \right) f_+^* e^{2i(k' - k \cos \theta) z} \right] + \right.
\]
\[
\left. + \left[ \frac{4|\mu|^2}{\hbar^2 \Gamma^2 (1 + 4\Delta^2)} \right] \left( \frac{4 \Delta |\mu|^2}{(\hbar \Gamma)^2 (1 + 4\Delta^2)} \right) \tilde{E}_p^4 \right. \\
\left. \left[ \left( \frac{6}{27} \eta_0 + \frac{4}{27} \eta_1 + \frac{26}{27} \eta_{-1} + \frac{28}{27} \eta_2 + \eta_{-2} \right) f_+^* + \right. \right.
\]
\[
\left. + \left( \frac{6}{27} \eta_0 + \frac{4}{27} \eta_1 + \frac{26}{27} \eta_{-1} + \frac{28}{27} \eta_2 + \eta_{-2} \right) b_+^* \right. \\
\left. + \left( \frac{4}{27} \eta_0 + \frac{26}{27} \eta_1 + \frac{26}{27} \eta_{-1} + \frac{26}{27} \eta_2 + \frac{26}{27} \eta_3 \right) b_+ e^{2i(k' - k \cos \theta) z} + \right.
\]
\[
\left. + \left( \frac{4}{27} \eta_0 + \frac{26}{27} \eta_1 + \frac{26}{27} \eta_{-1} + \frac{26}{27} \eta_2 + \frac{26}{27} \eta_3 \right) f_+^* e^{2i(k' - k \cos \theta) z} \right]\}. \quad (B.57)
\]

I define \( \tilde{\eta}_j = \eta_j / (n_a / 2) \). I recall \( \tilde{E}_p^2 = I_{tot} / (4\epsilon_0 c) \). I recall \( 2I_{sat} = \hbar^2 \Gamma^2 \epsilon_0 c / |\mu|^2 \).
Then,

\[
\begin{align*}
\frac{\partial f_+}{\partial z} &= - \frac{ik}{2\epsilon_0 \cos \theta} \frac{4 |\mu|^2 \tilde{\Delta}}{\hbar \Gamma (1 + 4 \Delta^2)^2} \left( \frac{4}{3} \eta_0 f_+ + \frac{2}{3} \tilde{\eta}_1 b_+ e^{2i(k' - k \cos \theta)z} + \right. \\
&\quad + \left( - \frac{\tilde{\Delta} I_{\text{tot}}}{2 I_{\text{sat}} \Gamma (1 + 4 \Delta^2)} \right) \left[ \left( \frac{2}{9} \eta_0 + \frac{8}{9} \tilde{\eta}_1 + \frac{8}{9} \tilde{\eta}_2 \right) f_+ + \left( \frac{2}{9} \eta_0 + \frac{8}{9} \tilde{\eta}_1 + \frac{8}{9} \tilde{\eta}_2 \right) b_- \right] \\
&\quad + \left( - \frac{I_{\text{tot}}}{2 I_{\text{sat}} (1 + 4 \Delta^2)} \right) \left[ \left( \frac{2}{9} \eta_0 + \frac{8}{9} \tilde{\eta}_1 + \frac{8}{9} \tilde{\eta}_2 \right) f_+ + \left( \frac{2}{9} \eta_0 + \frac{8}{9} \tilde{\eta}_1 + \frac{8}{9} \tilde{\eta}_2 \right) b_- \right] \\
&\quad + \left( \frac{I_{\text{tot}}}{2 I_{\text{sat}} (1 + 4 \Delta^2)} \right) \left( \frac{\tilde{\Delta} I_{\text{tot}}}{2 I_{\text{sat}} \Gamma (1 + 4 \Delta^2)} \right) \left\{ \left( \frac{2}{9} \eta_0 + \frac{8}{9} \tilde{\eta}_1 + \frac{8}{9} \tilde{\eta}_2 \right) f_+ + \left( \frac{2}{9} \eta_0 + \frac{8}{9} \tilde{\eta}_1 + \frac{8}{9} \tilde{\eta}_2 \right) b_- \right\} \\
&\quad + \left( \frac{I_{\text{tot}}}{2 I_{\text{sat}} (1 + 4 \Delta^2)} \right) \left( \frac{\tilde{\Delta} I_{\text{tot}}}{2 I_{\text{sat}} \Gamma (1 + 4 \Delta^2)} \right) \left\{ \left( \frac{2}{9} \eta_0 + \frac{8}{9} \tilde{\eta}_1 + \frac{8}{9} \tilde{\eta}_2 \right) f_+ + \left( \frac{2}{9} \eta_0 + \frac{8}{9} \tilde{\eta}_1 + \frac{8}{9} \tilde{\eta}_2 \right) b_- \right\}
\end{align*}
\]
Since $\tilde{\eta}_j = \tilde{\eta}_{-j}$, this becomes

$$
\frac{\partial f_+}{\partial z} = \frac{ik \chi_{\text{lin}}}{4 \cos \theta} \left( \frac{4}{3} \tilde{\eta}_0 f_+ + \frac{2}{3} \tilde{\eta}_1 b_+ e^{2i(k' - k \cos \theta)z} + \left( -\frac{\Delta I_{\text{tot}}}{2TI_{s\Delta}} \right) \left[ \left( 2 \frac{10}{9} \tilde{\eta}_0 + 2 \frac{8}{9} \tilde{\eta}_1 \right) f_+ + \left( \frac{10}{9} \tilde{\eta}_0 + 2 \frac{8}{9} \tilde{\eta}_1 + \frac{10}{9} \tilde{\eta}_2 \right) b_+ e^{2i\delta_k z} + \left( \frac{10}{9} \tilde{\eta}_0 + 2 \frac{8}{9} \tilde{\eta}_1 + \frac{10}{9} \tilde{\eta}_2 \right) f^*_+ e^{2i\delta_k z} \right] \right) + \left[ -\frac{I_{\text{tot}}}{2LI_{s\Delta}} \right] \left( 4 \frac{10}{9} \tilde{\eta}_0 + 4 \frac{8}{9} \tilde{\eta}_1 \right) f_+ + \left( 2 \frac{10}{9} \tilde{\eta}_0 + 2 \frac{8}{9} \tilde{\eta}_1 \right) b_+ e^{2i\delta_k z} + \left( \frac{10}{9} \tilde{\eta}_0 + 2 \frac{8}{9} \tilde{\eta}_1 + \frac{10}{9} \tilde{\eta}_2 \right) f^*_+ e^{2i\delta_k z} \right] + \left[ \frac{I_{\text{tot}}}{2LI_{s\Delta}} \right] \left( \frac{\Delta I_{\text{tot}}}{2TI_{s\Delta}} \right) \cdot \left[ \left( 6 \frac{28}{27} \tilde{\eta}_0 + 8 \frac{26}{27} \tilde{\eta}_1 + 2 \frac{28}{27} \tilde{\eta}_2 \right) f_+ + \left( 6 \frac{28}{27} \tilde{\eta}_0 + 8 \frac{26}{27} \tilde{\eta}_1 + 2 \frac{28}{27} \tilde{\eta}_2 \right) b_+ e^{2i\delta_k z} + \left( 6 \frac{28}{27} \tilde{\eta}_0 + 8 \frac{26}{27} \tilde{\eta}_1 + 2 \frac{28}{27} \tilde{\eta}_2 \right) f^*_+ e^{2i\delta_k z} \right] \right) \right), \quad (B.59)
$$

where $\delta_k = k' - k \cos \theta$.

I define the new variables

$$
f_+(z, r) = f'_+(z, r)e^{i(k' - k \cos \theta)z}, \quad (B.60)
$$

$$
f_-(z, r) = f'_-(z, r)e^{i(k' - k \cos \theta)z}, \quad (B.61)
$$

$$
b_+(z, r) = b'_+(z, r)e^{-i(k' - k \cos \theta)z}, \quad (B.62)
$$

and

$$
b_-(z, r) = b'_-(z, r)e^{-i(k' - k \cos \theta)z}. \quad (B.63)
$$
Then
\[
\frac{\partial f_+}{\partial z} = \frac{\partial f_+(z, r)}{\partial z} e^{i(k' - k \cos \theta)z} + i \delta_k f'_+(z, r) e^{i(k' - k \cos \theta)z}, \tag{B.64}
\]
and
\[
\begin{align*}
\frac{\partial f'_+}{\partial z} + i \delta_k f'_+ &= \frac{i k \chi}{4 \cos \theta} \left( \frac{4}{3} \tilde{\eta}_0 f'_+ + \frac{2}{3} \tilde{\eta}_1 b'_+ + \left( - \frac{\Delta I_{\text{tot}}}{2TI_{s\Delta}} \right) \left[ \left( \frac{10}{9} \tilde{\eta}_0 + \frac{8}{9} \tilde{\eta}_1 \right) f'_+ + \left( \frac{2}{9} \tilde{\eta}_0 + \frac{8}{9} \tilde{\eta}_1 + \frac{10}{9} \tilde{\eta}_2 \right) b'_+ + \left( \frac{10}{9} \tilde{\eta}_0 + \frac{8}{9} \tilde{\eta}_1 + \frac{10}{9} \tilde{\eta}_2 \right) f'_* \right] \right) \\
&\quad + \left( \frac{2}{9} \tilde{\eta}_0 + \frac{8}{9} \tilde{\eta}_1 \right) b'_* + \left( \frac{10}{9} \tilde{\eta}_0 + \frac{8}{9} \tilde{\eta}_1 + \frac{10}{9} \tilde{\eta}_2 \right) b'_+ + \left( \frac{10}{9} \tilde{\eta}_0 + \frac{8}{9} \tilde{\eta}_1 + \frac{10}{9} \tilde{\eta}_2 \right) f'_* \right) \\
&\quad + \left[ \frac{I_{\text{tot}}}{2I_{s\Delta}} \right] \left( \frac{\Delta I_{\text{tot}}}{2TI_{s\Delta}} \right) \left[ \left( \frac{6}{27} \tilde{\eta}_0 + \frac{26}{27} \tilde{\eta}_1 + \frac{28}{27} \tilde{\eta}_2 \right) f'_+ + \left( \frac{28}{27} \tilde{\eta}_0 + \frac{26}{27} \tilde{\eta}_1 + \frac{28}{27} \tilde{\eta}_2 \right) b'_* + \left( \frac{4}{27} \tilde{\eta}_0 + \frac{26}{27} \tilde{\eta}_1 + \frac{28}{27} \tilde{\eta}_2 + \frac{26}{27} \tilde{\eta}_3 \right) b'_+ + \left( \frac{4}{27} \tilde{\eta}_0 + \frac{26}{27} \tilde{\eta}_1 + \frac{28}{27} \tilde{\eta}_2 + \frac{26}{27} \tilde{\eta}_3 \right) f'_* \right] \right) \Bigg). \tag{B.65}
\end{align*}
\]
Simplifying further, one obtains

\[ \frac{\partial f'_i}{\partial z} + i\delta_k f'_i = \frac{i k \chi_{\text{lin}} 2}{4 \cos \theta} \left\{ \right. \\
\left. 2\bar{\eta}_0 f'_+ + \bar{\eta}_1 b'_+ + \left( -\frac{\bar{\Delta} I_{\text{tot}}}{2T I_{s\Delta}} \right) \left[ \left( \frac{5}{3} \bar{\eta}_0 + \frac{4}{3} \bar{\eta}_1 + \frac{5}{3} \bar{\eta}_2 \right) f'_+ + \left( \frac{2}{3} \bar{\eta}_0 + \frac{2}{3} \bar{\eta}_1 \right) f'_+ \right] + \right. \\
\left. + \left( \frac{5}{3} \bar{\eta}_0 + \frac{4}{3} \bar{\eta}_1 \right) b'^* + \left( \frac{5}{3} \bar{\eta}_0 + \frac{4}{3} \bar{\eta}_1 + \frac{5}{3} \bar{\eta}_2 \right) b'^* + \left( \frac{2}{3} \bar{\eta}_0 + \frac{2}{3} \bar{\eta}_1 + \frac{5}{3} \bar{\eta}_2 \right) f'^* \right] + \right. \\
\left. + \left[ -\frac{I_{\text{tot}}}{2I_{s\Delta}} \right] \left[ \left( \frac{4}{3} \bar{\eta}_0 + \frac{4}{3} \bar{\eta}_1 \right) f'_+ + \left( \frac{2}{3} \bar{\eta}_0 + \frac{2}{3} \bar{\eta}_1 \right) b'^* + \left( \frac{5}{3} \bar{\eta}_0 + \frac{4}{3} \bar{\eta}_1 + \frac{5}{3} \bar{\eta}_2 \right) f'^* \right] + \right. \\
\left. + \left[ \frac{I_{\text{tot}}}{2I_{s\Delta}} \right] \left( \frac{\bar{\Delta} I_{\text{tot}}}{2T I_{s\Delta}} \right) \left[ \left( \frac{14}{9} \bar{\eta}_0 + \frac{13}{9} \bar{\eta}_1 + \frac{14}{9} \bar{\eta}_2 \right) f'_+ + \left( \frac{6}{9} \bar{\eta}_0 + \frac{8}{9} \bar{\eta}_1 + \frac{2}{9} \bar{\eta}_2 \right) b'^* + \right. \\
\left. \left( \frac{4}{9} \bar{\eta}_0 + \frac{14}{9} \bar{\eta}_1 + \frac{13}{9} \bar{\eta}_3 \right) b'^* + \left( \frac{4}{9} \bar{\eta}_0 + \frac{14}{9} \bar{\eta}_1 + \frac{13}{9} \bar{\eta}_3 \right) f'^* \right] \right\}. \\
\text{(B.66)} \]

I then multiply this and the analogous equations for \( f_- (z, r) \), \( b_+ (z, r) \), and \( b_- (z, r) \) by 2 in order to account for the other circular component of the optical field polarization to define the total phase shift. This gives rise to Eq. 7.66.
Bibliography


Schmittberger, B. L. and Gauthier, D. J. (2016b), “Spontaneous emergence of free-space optical and atomic patterns,” (manuscript under review).


Biography

Bonnie Lee Schmittberger was born in Princeton, NJ on July 4, 1988. She lived in East Windsor, NJ as a baby, but the majority of her childhood was spent in Holland, PA. There, she attended Holland Elementary, Holland Middle School, and Council Rock High School South, where she was a member of the National Honor Society and proud member of both the orchestra and the marching band. She matriculated at Bryn Mawr College in 2006, where she spent four wonderful years studying physics, math, language, and art history. She graduated magna cum laude with an A.B. in Physics in 2010.

Bonnie matriculated at Duke University in 2010, where she immediately joined the Quantum Electronics Lab under Prof. Daniel J. Gauthier. During her time at Duke, she was named a Walter Gordy Fellow in both 2010 and 2015. She acted as both president (2012-2013) and vice president (2013-2014) of the Graduate Student Organization in the Physics Department. She also worked as a liaison with the Emerging Leaders in Science & Society (ELISS) program of the American Association for the Advancement of Science, which she hopes will be a valuable program to all graduate students for many years to come.

Bonnie received her A.M. degree in Physics from Duke University in 2013. She received her Ph.D. in Physics from Duke in 2016 for her work on optical and atomic pattern formation in cold atoms. In 2016, she moved to Maryland to join the group of Dr. Paul D. Lett as a postdoctoral researcher at the Joint Quantum Institute.
List of Publications

- B.L. Schmittberger and D.J. Gauthier, ‘Modeling pattern formation in ultra-cold atoms,’ (manuscript in preparation) (2016)

- B.L. Schmittberger and D.J. Gauthier, ‘Spontaneous emergence of free-space optical and atomic patterns,’ (manuscript under review) (2016)


List of Presentations

Oral Presentations:

- B.L. Schmittberger and D.J. Gauthier, ‘Probing Atomic Dynamics and Structures Using Optical Patterns,’ June 10, 2015, DAMOP, Columbus, OH.

- (Invited) B.L. Schmittberger and D.J. Gauthier, ‘Enhancing the nonlinearity at ultra-low light levels using spatial bunching of cold atoms,’ February 10, 2015, SPIE Photonics West, San Francisco, CA.

- B.L. Schmittberger and D.J. Gauthier, ‘Optical Pattern Formation in Spatially Bunched Atoms: A Self-Consistent Model and Experiment,’ June 5, 2014, DAMOP, Madison, WI.


• B.L. Schmittberger, J.A. Greenberg, and D.J. Gauthier, ‘Pattern Formations for Optical Switching Using Cold Atoms as a Nonlinear Medium,’ June 17, 2011, DAMOP, Atlanta, GA.

Poster Presentations:

• B.L. Schmittberger and D.J. Gauthier, ‘Realizing optical pattern formation at ultra-low light levels via atomic bunching,’ March 9-10, 2015, Fitzpatrick Institute for Photonics Annual Symposium, Duke University, Durham, NC.


• B.L. Schmittberger and D.J. Gauthier, ‘A Self-Consistent Model for Optical Pattern Formation in Cold Atoms,’ March 11-12, 2014, Fitzpatrick Institute for Photonics Annual Symposium, Duke University, Durham, NC.

• B.L. Schmittberger, J.A. Greenberg, and D.J. Gauthier, ‘Optical Pattern Formation in Cold Atoms: Explaining the Red-Blue Asymmetry,’ June 05, 2013, DAMOP, Quebec City, Quebec, Canada.

• B.L. Schmittberger, J.A. Greenberg, and D.J. Gauthier, ‘Optical Pattern Formation in Cold Atoms: The Role of Sisyphus Cooling,’ March 11, 2013, Fitz-
• B.L. Schmittberger, J.A. Greenberg, and D.J. Gauthier, ‘Free-space, multi-mode self-organization of cold, thermal atoms via transverse optical patterns,’ August 15, 2012, Quantum Science Conference, Gordon Research Conferences, Easton MA.

• B.L. Schmittberger, J.A. Greenberg, and D.J. Gauthier, ‘Optical Pattern Formation and Self-Organization of Cold Atoms,’ October 10, 2011, Fitzpatrick Institute for Photonics Annual Symposium, Duke University, Durham, NC.