Chapter 4

Two-photon lasers

by

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§ 1. Introduction

An amazing property of laser light that makes it so useful for a variety of applications is its high directionality, spectral purity, and high power. This high degree of spatial and temporal coherence arises from a complex interplay between several physical processes taking place within a laser, such as the fundamental light–matter interactions of absorption, spontaneous emission and stimulated emission, and the effects of feedback due to the reflecting surfaces of the optical resonator (Mandel and Wolf [1995]). The degree of coherence of light generated by lasers can be altered significantly and often in a surprising manner by modifying the properties of the resonator or the light–matter interactions. Of great current interest is the generation of specific quantum states of light that possess desired correlations among the emitted photons (related to the $n$th-order spatial and temporal coherence functions characterizing the light) for applications in quantum information (Bouwmeester, Ekert and Zeilinger [2000]), for example.

One fascinating technique for modifying the coherence properties of laser light is to operate the device in the ‘cavity quantum electrodynamics regime’ where only a small number of atoms and photons interact with and are strongly coupled to the optical resonator. For example, the threshold, stability, and quantum-statistical properties of a single-atom maser (Messchede, Walther and Müller [1985]) or laser (An, Childs, Dasari and Feld [1994]) operating in this regime are very different from their multi-atom counterparts. The coherence properties of laser light can also be modified by exploiting different types of light–matter interactions. For example, it has been known since the early days of quantum mechanics that there exist two-photon analogies to the standard one-photon processes of absorption, emission, and stimulated emission.

To understand how two-photon effects can change the properties of laser light, I first review the characteristics of the one-photon processes shown in figs. 1a–c depicting the interaction of light with an atom possessing an upper state $|b'\rangle$ and lower state $|a'\rangle$. Spontaneous emission (fig. 1a) occurs when an atom in the upper state decays at a rate $A^{1γ}$ to the lower state and emits a photon whose frequency $\omega$ is essentially equal to the transition frequency $\omega_{b'a'}$. For a typical allowed electric dipole transition, $A^{1γ} \approx 10^8$ s$^{-1}$. Absorption (fig. 1b) occurs when an atom in the lower state is promoted to the upper state and the photon is annihilated, where
one-photon processes

\[ \begin{align*}
(a) & \quad \omega' \quad \omega' \\
(b) & \quad \omega' \\
(c) & \quad \omega' \\
\end{align*} \]

two-photon processes

\[ \begin{align*}
(d) & \quad \omega'' \quad \omega'' \\
(e) & \quad \omega'' \\
(f) & \quad \omega'' \\
(g) & \quad \omega'' \\
\end{align*} \]

Fig. 1. Light–matter interactions. One-photon (a) spontaneous emission, (b) absorption, and (c) stimulated emission. Two-photon (d) spontaneous emission, (e) absorption, (f) singly stimulated emission, and (g) stimulated emission.

the absorption rate $W_{a'b'}^{1\gamma}$ is proportional to the incident photon flux. Stimulated emission (fig. 1c) occurs when an incident photon forces an atom to jump from the upper state to the lower state and two photons are scattered by the atom. The scattered photons have the same frequency, phase and direction as the incident photon, which gives the laser its unique coherence properties. The stimulated emission rate $W_{b'a'}^{1\gamma}$ is also proportional to the incident photon flux, and proceeds most efficiently when $\omega = \omega_{b'a'}$ and when the states have opposite parity (electric-dipole-allowed transition). Based on thermodynamic considerations, it was shown by Einstein [1917] that $W_{a'b'}^{1\gamma} = W_{b'a'}^{1\gamma}$.

By analogy, consider the two-photon processes that occur when light interacts with an atom possessing an upper state $|b\rangle$ and lower state $|a\rangle$, which typically have the same parity so that one-photon transitions between the states are electric-dipole forbidden. Spontaneous emission (fig. 1d) occurs when an atom in the upper state decays to the lower state at a rate $A_{a'b'}^{2\gamma}$ and emits two photons of frequency $\omega'$ and $\omega''$. For this process to be allowed, the atom must possess additional auxiliary states $|i\rangle$ with opposite parity (real or continuum states). Note that the frequencies of the scattered photons can take on any value so long as $\omega' + \omega'' \approx \omega_{ha}$, where $\omega_{ha}$ is the two-photon transition frequency. The dashed line in the figure represents a ‘virtual state’ that has opposite parity (associated with the real states $|i\rangle$) whose lifetime is determined by the Heisenberg uncertainty principle, crudely given by $\hbar/|\Delta E|$, where $\Delta E$ is the energy difference between the virtual state and the closest opposite-parity
auxiliary state. For typical metastable states where two-photon spontaneous emission is the dominant decay mechanism, $A^{2\gamma} \approx 7 \text{s}^{-1}$. Two-photon absorption (fig. 1e) occurs when an atom in the lower state is promoted to the upper state and two incident photons are annihilated, where the absorption rate $W^{2\gamma}_{ab}$ is proportional to the flux of photons at frequency $\omega'$ multiplied by the flux of photons at frequency $\omega''$. A new decay mechanism for the upper state that has no correspondence to any of the one-photon process is singly-stimulated spontaneous emission (fig. 1f). In this process, an incident photon of frequency $\omega' \neq \frac{1}{2} \omega_{ba}$ induces the emission of two photons at $\omega'$ and a single photon at the complementary frequency $\omega'' = \omega_{ba} - \omega'$. In the stimulated emission process (fig. 1g), two incident photons force the atom to the lower state and four photons are scattered by the atom. The stimulated emission rate $W^{2\gamma}_{ba} = W^{2\gamma}_{ab}$ is also proportional to the flux of photons at frequency $\omega'$ multiplied by the flux of photons at frequency $\omega''$, and proceeds most efficiently when $\omega' + \omega'' \approx \omega_{ba}$. The scattered photons have the same frequency, phase and direction as the incident photons, which give the laser coherence properties different from those of normal one-photon lasers. For the case when there is only a single monochromatic beam of light incident on the atom so that $\omega' = \omega'' \approx \frac{1}{2} \omega_{ba}$, the degenerate two-photon stimulated emission rate is proportional to the square of the incident photon flux. Working independently, both Sorokin and Braslau [1964] and Prokhorov [1965] suggested that it would be of great interest to develop a 'two-photon laser' based on the two-photon stimulated emission process. While replacing the standard one-photon stimulated emission process in a laser by a high-order one might be expected to give rise to subtle differences observable only at the quantum level, it has been predicted that there will be dramatic changes in both the microscopic and macroscopic laser behaviors even when many atoms participate in the laser process. One reason for these differences is that the stimulated emission rate depends quadratically on the incident photon flux (for the degenerate two-photon laser where $\omega' = \omega''$), resulting in an inherently nonlinear light–matter interaction.

As an example, consider the effects of such a nonlinearity on the threshold behavior of the degenerate two-photon laser. Briefly, the threshold condition for all lasers is that the round-trip gain must equal the round-trip loss. For one-photon lasers this yields the well-known result that lasing will commence when a uniquely defined minimum inversion density (proportional to the gain) is attained via sufficient pumping. The situation is more complicated for the two-photon laser because the unsaturated gain increases with increasing inversion density and with increasing cavity photon number. This results in a threshold condition
specified by a uniquely defined minimum inversion density and cavity photon number so that it cannot turn on unless quantum fluctuations or an injected field bring the intracavity light above the critical value. In addition, once the minimum photon number exists in the cavity, the photon number undergoes a run-away process, growing rapidly until the two-photon transition is saturated (Sorokin and Braslau [1964]). Therefore, the two-photon laser operates in the saturated regime (a source of optical nonlinearity) even at the laser threshold, giving rise to the possibility that the laser will display dynamical instabilities.

The primary purpose of this chapter is to review the research on two-photon quantum processes (§§2, 4), leading up to the development and characterization of continuous-wave two-photon masers (§ 5) and lasers (§§6, 7). An experimental perspective will be emphasized, with a brief description of simple theoretical models of two-photon lasers to develop a conceptual foundation for their behavior (§ 3). The vast body of theoretical research on two-photon lasers will not be covered in detail, highlighting only the early work and some of the more recent results (§ 8).

§ 2. Two-photon processes

The concept of stimulated emission of photons was first postulated by Einstein [1917] in the development of an alternate derivation of Planck's black-body radiation law, but only encompassed the idea of the one-photon process shown in figs. 1a–c. The first discussion of two-quantum processes in the interaction between electromagnetic radiation and atoms appeared in an initial report by Göppert-Mayer [1929] and in a later article (Göppert-Mayer [1931]) that summarized the results of her doctoral dissertation. In these studies, the interactions of the atom and field were treated fully quantum-mechanically using second-order perturbation theory (one of the earlier uses of this approach).

The concept of two-photon quantum processes was used soon thereafter to explain the decay rate of metastable atomic states. Breit and Teller [1940] predicted that the dominant mechanism for the decay of the hydrogen 2S_{1/2} state is via two-photon spontaneous emission (fig. 1d) to the 1S_{1/2} ground state, giving rise to a predicted lifetime of the order of 0.1 s. The probability of such spontaneous two-photon transitions is low, and it is smaller than those of allowed one-photon transitions by a factor of the order of α(αZ)^2 (≈ 10^{-6} for hydrogen), where α is the fine-structure constant and Z is the atomic number (Bethe and Salpeter [1977]). The lifetime predictions were later refined by Shapiro and Breit [1959] and Tung, Ye, Salamo and Chan [1984]; confirmation
of the predictions took many years and required the use of sophisticated experimental techniques because perturbations from external static electric fields and collisions significantly shorten the lifetime, thereby obscuring the two-photon decay process. Observation of two-photon absorption and stimulated emission processes (figs. 1e and 1g, respectively) required the advent of the laser, which could produce intensities high enough to increase the transition probability to detectable levels. In this section, I briefly review the early research investigating two-quantum processes.

2.1. Spontaneous emission

The study of two-photon spontaneous emission has been of interest for a number of years because of its possible role in dictating the decay of metastable states (Breit and Teller [1940]), for a possible explanation for the continuous spectrum observed in planetary nebulae (Spitzer and Greenstein [1951]), and for the search of a possible electric dipole moment of the electron (Salpeter [1958]). The first direct detection of two-photon spontaneous emission was by Lipeles, Novick and Tolk [1965] using the $2S_{1/2}$ state of singly ionized helium. They used ionized helium rather than atomic hydrogen because the two-photon lifetime and Stark quenching rate are both more than an order of magnitude smaller. They verified the prediction by Spitzer and Greenstein [1951] that the photons are emitted in a continuous broad spectrum peaked at $\frac{1}{2}\omega_{\text{ho}}$ and followed a $(1 + \cos^2 \theta)$ angular distribution for polarization-insensitive detectors, where $\theta$ is the angle between the wavevectors of the scattered photons. A review of the status of the experiments as of the late 1960s was given by Novick [1969]. In contrast to the decay of the hydrogenic $2S_{1/2}$ state, the emission spectrum and angular distribution are expected to be quite different for the two-photon decay of the helium-like $2^3S$ triplet state due to quantum interferences arising in the two-photon matrix element (Dalgarno [1969]), although the predicted lifetime of 10 years for atomic helium imposes severe experimental challenges.

The decay rate of metastable states due to two-photon processes can be enhanced significantly by singly stimulated spontaneous emission using an intense laser beam (see fig. 1f) as discussed by Lipeles, Gampel and Novick [1962] and later by Abella, Lipeles and Tolk [1963]. The first experiment to observe such emission was by Yatsiv, Rokni and Barak [1968]¹, who studied

¹ Note the typographical error in the title of the paper by Yatsiv, Rokni and Barak [1968]: proton should be replaced by photon.
the interaction of intense laser beams with a dense potassium vapor. To say
the least, their experiment is rather complicated because multiple intense fields
were used to populate the upper state as well as to singly stimulate the
spontaneous emission. Specifically, a beam generated by a ruby laser and a
second beam generated by Raman-shifting some of the ruby light in nitrobenzene
were overlapped and passed through a 1-m-long heated potassium vapor cell.
By coincidence, the sum of the ruby laser frequency and the Raman-shifted
frequency is essentially equal to the \( 4S_{1/2} \) to \( 6S_{1/2} \) two-photon transition
frequency; the \( 6S_{1/2} \) and \( 4S_{1/2} \) states served as the upper and lower states shown
in fig. 1f for studying the singly stimulated spontaneous emission process. The
field at frequency \( \omega' \) needed to induce this process was simultaneously self-
generated in the potassium vapor by stimulated atomic Raman scattering of
the ruby laser light from the potassium \( 4P_{3/2} \) state (Rokni and Yatsiv [1967]),
which was populated by exciting the \( 4S_{1/2} \rightarrow 4P_{3/2} \) transition whose resonance
frequency overlaps, by coincidence, the frequency of the nitrobenzene-shifted
ruby laser light. Evidence for singly stimulated two-photon emission was the
observation of the complementary frequency \( \omega'' = \omega_{na} - \omega' \), which was
10 cm\(^{-1}\) above the \( 5P_{3/2} \rightarrow 4S_{1/2} \) resonance line. In hindsight, there is a chance
that this emission was due to a different process known as phase-matched
four-wave mixing (Shen [1984]) where the incident laser beams and the self-
generated atomic-Raman-shifted light interacted via a third-order nonlinear
susceptibility to generate the complementary frequency. It is known that two-
photon resonant four-wave mixing occurs readily in strongly driven atomic
vapors (see, for example, Malcuit, Gauthier and Boyd [1985]), including the
specific transitions studied by Yatsiv, Rokni and Barak [1968] (Efthimiopoulos,
Movsessian, Katharakis, Merlemis and Chrissopoulou [1996]).

To circumvent the complexity in the possible interpretation of the experiment
by Yatsiv, Rokni and Barak [1968], Braunlich and Lambropoulos [1970]
investigated singly stimulated two-photon emission using a beam of metastable
deuterium atoms and a single intense, 55-joule laser beam generated by a
neodymium-doped-glass laser at a wavelength of 1.054 \( \mu \)m. The atomic beam
of metastable deuterium atoms was produced by a charge-exchange reaction
between a beam of deuterons and a Cs vapor. Evidence for the enhanced emission
was provided by the observation of light scattered by the atoms at a wavelength
of 1,373 Å, which is equal to the wavelength of the complementary photon. They
observed on average \( \sim 30 \) photons per 100 laser shots at this wavelength, which
agrees well with the theoretically predicted rate of 15 photons per 100 laser shots
considering the uncertainty in the efficiency of the collection optics. Related
experiments aimed at generating intense vacuum-ultraviolet light were conducted
by Zych, Lukasik, Young and Harris [1978] using a high-density helium glow-discharge. In this experiment, the $2s^1S \leftrightarrow 1s^2^1S$ transition of metastable helium underwent singly stimulated two-photon emission by irradiating the atoms with pulses from a picosecond Nd:YAG laser at a wavelength of 1.064 μm, leading to the spontaneous generation of photons at a wavelength of 637 Å.

Research on spontaneous two-photon emission continues to date, mainly for understanding the spectral distribution of light emitted by astrophysical objects and for fundamental tests of physics. For example, Stancil and Copeland [1993] have investigated theoretically the dependence of the lifetime of the hydrogenic $2S_{1/2}$ state in an ultra-strong magnetic field that might be produced in the vicinity of white dwarfs or pulsars. They found that the rate can be enhanced enormously, to a value approaching $10^8 \text{s}^{-1}$ at fields of the order of $10^6 \text{T}$, because it is resonantly enhanced by the sublevels of the $2P$ states that are Zeeman-shifted to energies between the $1S$ and $2S$ states by the magnetic field. Also, knowledge of the lifetimes of metastable helium-like ions is important for fundamental experiments using highly charged heavy-ion accelerators, including parity violation in helium-like uranium (Munger and Gould [1986]) and measurement of the nuclear magnetic moments of Coulomb-excited nuclear states (Labzowsky, Nefiodov, Plunien, Soff and Liesen [2000]).

A general relativistic theory for the two-photon spontaneous emission rates in helium-like ions in support of these experiments was recently presented by Sanots, Patte, Parente and Indelicato [2001].

2.2. Absorption

Of all the two-photon processes, absorption has found the widest range of applications, from precision measurement of physical constants (Niering, Holzwarth, Reichert, Pokasov, Udem, Weitz, Hänsch, Lemonde, Santarelli, Abgrall, Laurent, Salomon and Clairon [2000]) to a new type of high-resolution microscopy that has revolutionized the study of living organisms in three spatial dimensions (Denk, Strickler and Webb [1990]). It was first observed experimentally by Kaiser and Garrett [1961] soon after the development of the laser. They passed the red light of a ruby laser beam (wavelength 694 nm) through a CaF$_2$:Er$^{2+}$ crystal and observed blue fluorescent light (wavelength 425 nm) emanating from the crystal. Their explanation of the fluorescence generation is as follows: the Er$^{2+}$ ion is promoted from the ground 4f state to the broad 5d excited state via two-photon absorption, then relaxes to the bottom of the 5d band, then decays to the ground state via one-photon spontaneous
emission of a blue fluorescent photon. Support for their conjecture was the observation that the intensity of the fluorescent light scales quadratically with the intensity of the ruby laser beam. In addition, an estimate for the two-photon absorption cross-section based on the experimental parameters was in surprisingly good agreement with the theoretical estimate of Kleinman [1962], who showed how to make simplifying assumptions in evaluating the complete theory of Göppert-Mayer [1931] so that it could be compared to experiments. They also pointed out that the blue light could not be due to the phase-matched process of second-harmonic generation, which was reported only four weeks before their experiment (Franken, Hill, Peters and Weinreich [1961]), because the CaF$_2$:Er$^{2+}$ crystal possesses a center of inversion and hence second-harmonic generation should be forbidden.

Numerous observations of two-photon absorption were reported by several groups with access to a ruby laser soon after the experiments of Kaiser and Garrett [1961]. One notable experiment by Abella [1962] used a ruby laser beam to excite the $6S_{1/2} \rightarrow 9D_{3/2}$ two-photon transition in a dilute rubidium vapor, which subsequently decayed back to the ground state via two step-wise one-photon spontaneous emission events. Because the atomic structure was well known for rubidium, a quantitative comparison to theoretical predictions could be made. It was observed that the fluorescent intensity was a factor of 100 smaller than expected, even correcting for the multi-mode nature of the ruby laser beam, suggesting that non-radiative quenching of the $9D_{3/2}$ was taking place. Similarly, Hall, Robinson and Branscomb [1965] studied photodetachment of I$^-$ via two-photon excitation. Since I$^-$ only has a single bound level, the two-photon-excited detachment rate is sensitive to the bound-to-continuum transition matrix elements. They found that the experimentally measured rate was a factor of 36 larger than the predicted value (Geltman [1962]), suggesting that treating the free-electron states as plane waves is not appropriate.

There have been many subsequent observations of two-photon absorption in every imaginable material and for a wide range of applications. Several discussions are available in textbooks such as those by Levenson [1982] and Stenholm [1984].

2.3. Stimulated emission and lasing

Soon after the realization of the first laser in 1960, there was an explosion of research on every new and imaginable effect that might occur when light from a ruby laser was focused to high intensity and passed through matter in
various forms and states. In addition, researchers were scrambling to develop new laser sources to extend the accessible wavelength range and to tailor the properties of laser light for various possible applications. It is therefore not too surprising that the concept for the two-photon laser was developed independently and essentially at the same time by scientists in the USA at the IBM Research Laboratories and in the former USSR at the Lebedev Institute in Moscow.

The first public discussion of the work on two-photon lasers coming out of the Lebedev Institute appears to have been in December 1964 during the Nobel prize acceptance speech of Prokhorov [1965], who was being recognized for his contribution to the development of the one-photon laser. In his speech, he summarized the research leading up to the discovery of the laser, then went on to describe enthusiastically the possibility of developing a two-photon laser. He was especially interested in two properties of such quantum oscillators: They should display a faster growth of the field density in comparison to usual lasers, and they should produce simultaneously two different frequencies $\omega'$ and $\omega''$ (where $\omega' + \omega'' = \omega_{pa}$). Since the specific lasing frequencies are set by the boundary conditions imposed on the electromagnetic field by the surfaces comprising the optical resonator, such a laser could be a source of broadly and continuously tunable radiation, which would have been of great use in the area of molecular spectroscopy and controlling chemical reactions. It was also mentioned that an auxiliary laser would be needed to provide a sufficient number of photons to initiate lasing, as described briefly in §1. He closed with a statement that the development of a tunable two-photon laser would be difficult, but that it was extremely interesting and might revolutionize the chemical industry. In hindsight, his comment on the difficulty of achieving two-photon lasing was quite appropriate; it took over fifteen years for researchers to overcome the several technical challenges that delayed the development of the first two-photon quantum oscillators.

It appears that work on the two-photon laser at the Lebedev Institute pre-dates the Nobel lecture considerably. Specifically, the published lecture (Prokhorov [1965]) cites a patent disclosure with a filing date of December 1963. The patent was granted by the Soviet government sometime between the time of a brief paper describing the possibility of a two-photon laser by Selivanenko [1966] (which was submitted for publication in May 1964, one month after the work in the United States was first published), and a longer paper by Kirsanov and Selivanenko [1967] in which the patent was referenced. The longer paper gives more details of the operating characteristics of the two-photon laser and contrasts its behavior to a parametric oscillator. Note that the term two-photon oscillator was often used in the Soviet literature to describe both a two-photon laser and
a parametric oscillator even though their behaviors are very different (see, for example, Gurevich and Kheifets [1967]). I have not yet been able to obtain a copy of the two-photon laser patent, with my search hindered by the recent passing of Prof. Prokhorov, but it seems to make claims about the ability of the two-photon laser to generate intense continuously tunability pulses of light, based on the later published reports. Additional information summarizing the work on two-photon lasers from the Lebedev Institute around this time period can be found in the publication by Butylkin, Kaplan, Khronopulo and Yakubovich [1977].

The IBM group (Sorokin and Braslau [1964]) also appreciated the potential usefulness of the two-photon laser for generating intense pulses of light. The first paper focused on the operating characteristics of a degenerate two-photon laser, including the generation of a giant pulse. They also devised a different technique for achieving a critical photon density (called ‘priming photons’) for initiating lasing. Rather than providing priming photons from an auxiliary laser beam that was injected into the resonator, as suggested by Prokhorov, they envisioned a laser medium containing two different species of ions doped in a solid host, one possessing a one-photon transition (transition frequency $\omega_A$) and the other possessing a two-photon transition (transition frequency $\omega_B$). A population inversion is obtained in both species by exciting the ions to broad absorption bands via flash-lamp pumping, which subsequently decay to the upper laser transitions, as shown in fig. 2a for the case of a non-degenerate two-photon laser. One-photon lasing on the A-species ions gives rise to emission at
frequency $\omega_A$, which triggers two-photon emission at both $\omega_A$ and $\omega_C$ such that ($\omega_A + \omega_C \approx \omega_B$). They proposed that lasing could be obtained with technology available at the time using CaF$_2$:Yb$^{3+}$ as the two-photon medium and one of many rare-earth ions doped in one of several crystals as the one-photon medium for supplying the priming photons. Note that Sorokin and Braslau [1964] only treated the case of a degenerate two-photon laser, which was later generalized by Garwin [1964] to account for non-degenerate operation of the laser. Both reports are summarized in greater detail by Smith and Sorokin [1966].

They used a simple rate-equation model to describe the operating characteristics of the combined effects of both atomic species and the coupling of the atoms to the optical resonator, as described in greater detail in § 3. For degenerate operation of the two-photon laser, they found that two-photon amplification gives rise to an extremely fast increase in the number of laser photons once a critical number of photons $q_{\text{min}}$ in the resonator is attained due to the one-photon lasing species. They described this as an avalanche effect, leading to a giant pulse that continues until most of the energy is extracted from the B-species atoms. Figure 2b shows the predictions of Sorokin and Braslau [1964] for the temporal evolution of the photon number $q$ as a function of the initial number of priming photons $q(0)$ provided by the A-species atoms. Once $q(0) > q_{\text{min}}$, it is seen that the photon number increases by three orders of magnitude on sub-nanosecond time scales, then decreases exponentially at the cavity decay rate. Gordon and Moskvin [1976] also predicted giant-pulse operation in a two-photon laser for essentially the same configuration, although they appear to be unaware of the previous work of Sorokin and Braslau [1964].

In a related theoretical study on the short-pulse generation capabilities of a two-photon laser, Letokhov [1968] suggested that passing a short laser pulse through a two-photon amplifier will give rise to a dramatic pulse shortening whose ultimate duration would only be limited by saturation of the two-photon transition (see also Selivanenko [1966]). Letokhov also points out that significant pulse shortening can be expected for intensities much lower than that needed to operate a two-photon laser, and hence an amplifier based on two-quantum processes might be more useful than a laser. More recently, Heatley, Ironside and Firth [1993] investigated theoretically ultrashort-pulse generation in a two-photon laser in which the bandwidth of the gain overlapped many longitudinal modes of the optical resonator. Taking into account linear dispersion and two-photon gain saturation, they found that the laser is capable of producing sub-picosecond pulses via phase locking of the longitudinal modes.

The two-photon laser appeared to be an experimentalist's dream because it should be continuously tunable over a broad range (recall that there were only
fixed-frequency lasers in the early to mid 1960s), operate at high power and store large energies. The high energy extraction capabilities were so promising that two-photon lasers and amplifiers were thoroughly scrutinized for use in laser-induced thermonuclear fusion experiments (Carman [1975]). On a more fundamental side, the two-photon laser challenges our understanding of the interaction of light with matter because it is a highly nonlinear, far from equilibrium system that cannot be analyzed easily using standard perturbation techniques. Unfortunately, achieving two-photon lasing was stymied by a lack of suitable gain media. In § 4, the early experimental work on measuring two-photon amplification and lasing will be summarized after a brief introduction to simple models of the two-photon processes in the next section.

§ 3. Simple models of amplification and lasing

Before reviewing the experimental work on two-photon amplification and lasing, it is useful to consider simple models of these systems to gain an understanding of how they are different from one-photon systems and why it has been difficult to realize them in experiments. I first consider a rate-equation model of the interaction, then go on to consider how coherent effects enter in two-photon processes.

3.1. Rate-equation model

In a situation where there is large dephasing of the coherences of the atomic energy levels, a simple rate-equation model of lasers is known to give results that are not too different from that obtained by more complete approaches (Allen and Eberly [1987]). Sorokin and Braslau [1964] used this approach in the first description of the operating characteristics of the two-photon laser.

For simplicity, consider degenerate operation of a two-photon laser (mode volume \( V \)) filled homogeneously with atoms possessing a two-photon transition as shown in figs. 1d–g. The number of photons in the cavity is denoted by \( q \) and the total number of atoms in the upper (lower) energy level is denoted by \( N_b \) (\( N_a \)) so that the inversion is given by \( \Delta N = N_b - N_a \). The primary difference between one- and two-photon lasers is that the stimulated emission rate

\[
W^{(2)} = B^{(2)} q^2.
\]
is proportional to the square of the photon number, where $B^{(2)}$ is the two-photon rate coefficient. Following Concannon and Gauthier [1994], the photon number and inversion evolve according to

$$\frac{dq}{dt} = B^{(2)} q^2 \Delta N - \kappa [q - q_{\text{inj}}(t)], \quad (3.2)$$

and

$$\frac{d\Delta N}{dt} = -2B^{(2)} q^2 \Delta N - \gamma_\parallel (\Delta N - \Delta N_o), \quad (3.3)$$

where $\Delta N_o$ is the inversion density in the absence of the field due to the pump process, $\gamma_\parallel$ is the atomic inversion decay rate, $\gamma_\parallel \Delta N_o$ is the pump rate, $\kappa$ is the cavity decay rate, and $q_{\text{inj}}(t)$ is the photon number injected into the cavity by an external source. It is seen from eq. (3.2) that the photon number increases due to the two-photon stimulated emission process and by injection from the external source, and decreases due to linear loss through the cavity mirrors. The possibility of two-photon spontaneous emission processes at the laser frequency is ignored because the emission rates are extremely small in the optical regime (Holm and Sargent [1986]). This approximation is not valid for two-photon masers where the stimulated and spontaneous rates are comparable (Davidovich, Raimond, Brune and Haroche [1987]). From eq. (3.3), it is seen that the inversion decreases in response to the stimulated emission process and due to other radiative (at frequencies distinct from the laser frequency) and non-radiative decay mechanisms, and increases due to the pump process.

An crude understanding of the laser turn-on behavior can be obtained by investigating the initial transient behavior of the laser using eq. (3.2) under the assumption that the inversion is not depleted during the turn-on, as discussed by Sorokin and Braslau [1964] and Schubert and Wiederhold [1979], and summarized in Schubert and Wilhelmi [1986]. With an initial inversion $\Delta N(0)$ and $q_{\text{inj}} = 0$, the photon number increases when the initial number of photons in the cavity is greater than

$$q_{\text{min}} = \frac{\kappa}{B^{(2)} \Delta N(0)}. \quad (3.4)$$

For an initial photon number $q(0)$ exceeding the minimum value, the temporal evolution of the laser is given by

$$q(t) = \frac{q_{\text{min}}}{1 - [1 - q(0)/q_{\text{min}}]e^{\gamma t}}, \quad (3.5)$$
which diverges in a time

$$t_{\text{div}} = \frac{1}{\kappa} \ln \left[ \frac{q(0)/q_{\text{min}}}{q(0)/q_{\text{min}} - 1} \right].$$

(3.6)

Of course, saturation of the inversion prevents any divergence in the photon number. Nevertheless, eq. (3.6) serves as a reasonable estimate of the time at which the photon number undergoes the explosive growth shown in fig. 2b. For the laser materials considered by Sorokin and Braslau [1964], they estimated that $\kappa = 2.5 \times 10^8 \text{ s}^{-1}$, $B^{(2)} = 3.6 \times 10^{-25} \text{ s}^{-1}$, and $\Delta N(0) = 2 \times 10^{18}$ atoms, so that $q_{\text{min}} = 3.5 \times 10^{14}$ photons, which they believed could have been obtained with existing technology. A later report (Smith and Sorokin [1966]) mentions that $B^{(2)}$ was probably overestimated by at least an order of magnitude.

Even though the photon number can experience growth when $q(0) > q_{\text{min}}$, sustained two-photon laser oscillation can occur only when there also exists a sufficient number of inverted atoms in the resonator. The conditions for sustained laser operation must take into account the mutual interaction of the photons and atoms, which can be determined by investigating the steady-state solutions to eqs. (3.2) and (3.3) and their stability, following the work of Ning [1991] and Concannon and Gauthier [1994]. It is found that the steady-state inversion is given by

$$\Delta N_{ss} = \frac{\Delta N_0}{1 + q_{ss}^2/q_{\text{sat}}^2},$$

(3.7)

where

$$q_{\text{sat}} = \sqrt{\frac{\gamma_{||}}{2B^{(2)}}}$$

(3.8)

is the two-photon saturation photon number. Equation (3.7) is dramatically different from the steady-state inversion for a one-photon laser (Svelto [1989]) where the inversion is clamped above the laser threshold. The saturation photon number typical for the rare-earth elements can be found using $\gamma_{||} = 1.6 \times 10^5 \text{ s}^{-1}$ and the rate coefficient given above, yielding $q_{\text{sat}} = 4.7 \times 10^{14}$ photons. The saturation intensity is given in terms of the saturation photon number through the relation $I_{\text{sat}} = c h \omega q_{\text{sat}}/\lambda$. For $\omega = 2.7 \times 10^{15} \text{ s}^{-1}$, $\lambda = 10^{-6} \text{ m}$, and $q_{\text{sat}} = 4.7 \times 10^{14}$ photons, one has $I_{\text{sat}} = 3.8 \times 10^{10} \text{ W/m}^2$, an intensity that was easily attainable from lasers in the mid 1960s.
The steady-state solution for the photon number is very different from that of one-photon lasers. Concannon and Gauthier [1994] found three solutions given by

\[ q_{ss}^0 = 0, \]  

and

\[ q_{ss}^\pm = \frac{\gamma_0}{4k} \left[ \Delta N_0 \pm \sqrt{\Delta N_0^2 - \frac{16q_{sat}^2k^2}{\gamma_0^2}} \right], \]  

for the case when \( q_{inj}(t) = 0 \). The physically meaningful (real) steady-state solutions represented by these equations are shown in fig. 3 where they are plotted as a function of the pump rate.

The threshold conditions for sustained laser action can be determined from eq. (3.10) by finding the minimum value of \( \Delta N_0 \) that admits a positive photon number. Upon inspection, the minimum inversion is given by

\[ \Delta N_0^{th} = \frac{4q_{sat}k}{\gamma_0}. \]  

This yields \( q_{ss}^\pm = q_{sat} \) and \( \Delta N_{ss} = \frac{1}{2} \Delta N_0^{th} \) at threshold. For the estimated laser parameters given above, \( \Delta N_0^{th} = 2.9 \times 10^{18} \) inverted atoms. To turn the laser on at the minimum threshold inversion (\( \Delta N_0 = \Delta N_0^{th} \)), an initial photon number approximately equal to

\[ q_{min} \approx \frac{k}{B^{(2)}\Delta N_0^{th}} = \frac{1}{2} q_{sat} \]  

must be present in the cavity. The photon number and inversion will evolve toward their steady-state values given above, as governed by eqs. (3.3) and (3.2).
Equations (3.11) and (3.12) represent the dual threshold conditions for achieving two-photon lasing, in agreement with the heuristic discussion of the threshold behavior presented in § 1. A similar condition has been found for the case of a non-degenerate two-photon laser by Hoshimiya, Yamagishi, Tanno and Inaba [1978]. For the case when photons are injected into the resonator over many cavity lifetimes, the minimum number of injected photons can be somewhat less than that given by eq. (3.12), as discussed below (see eq. 3.13).

The discontinuous threshold behavior shown in fig. 3 is indicative of a first-order phase transition (sometimes referred to as a hard mode of excitation; see Butylkin, Kaplan, Khronopulo and Yakubovich [1977]), which is very different from the smooth turn-on behavior of normal one-photon lasers. Note from fig. 3a that the two-photon gain is saturated at threshold, again in sharp contrast to the typical one-photon laser which operates very far below saturation. Figure 3b also shows that the inversion is never constant, unlike the behavior of one-photon lasers where the inversion clamps above threshold (Svelto [1989]).

Based on past experience with one-photon lasers (see, for instance, Weiss and Vilaseca [1991]), it is expect that the steady-state solutions may be unstable because the laser operates in the saturated regime. Concannon and Gauthier [1994] performed a linear stability analysis of the steady-state solutions and found that: (1) the zero-photon solution \( (q_{ss}^0, \Delta N_{ss}^0) \) is always stable (two-photon spontaneous emission, neglected here, can destabilize this solution); (2) the \( (q_{ss}^-, \Delta N_{ss}^-) \) solution, where the photon number decreases with increasing pump rate, is always unstable; (3) the \( (q_{ss}^+, \Delta N_{ss}^+) \) solution is always stable for a 'good' cavity (\( \gamma_\parallel/\kappa > 1 \)); and (4) the \( (q_{ss}^+, \Delta N_{ss}^+) \) solution is unstable for a 'bad' cavity (\( \gamma_\parallel/\kappa < 1 \)) for pumping just above threshold but stabilizes for higher pump rates. Ovadia and Sargent [1984], Ning and Haken [1989a] and Ning [1991] have found that the degenerate laser without ac Stark shifts is stable for a good cavity, may display instabilities for a moderately good cavity, and is always unstable for a bad cavity when coherent effects are taken into account, as will be described in greater detail in § 8.

To address how an injected field that remains on for several cavity lifetimes can turn on the laser, Concannon [1996] determined the steady-state solutions of eqs. (3.2) and (3.3) and their stability properties when a continuous-wave beam of photons is injected into the cavity \( (q_{inj} \neq 0) \). The stability of the solutions can be quite complex, especially in the bad-cavity limit. However, the analysis is straightforward if the laser is initially 'off', with \( \Delta N_o > \Delta N_o^{th} \) and \( q_{inj} \) is increased slowly. This behavior is illustrated in fig. 4a where the three solutions for the steady-state photon number are shown when \( q_{inj} \neq 0 \) and \( \Delta N_o = 1.2\Delta N_o^{th} \). The low-power solution (dashed curve) increases as \( q_{inj} \) increases and is stable
Fig. 4. Predicted behavior of the two-photon laser with an injected signal. (a) Photon number as a function of the injected number of photons. (b) Minimum number of photons that have to be injected into the two-photon laser to switch it from the off to the on state. From Concannon and Gauthier [1994].

until it reaches a critical value $q_{\text{inj}}^{\text{th}} \approx 0.11q_{\text{sat}}$. This point defines the minimum injected photon number necessary to initiate lasing. Increasing $q_{\text{inj}}$ beyond $q_{\text{inj}}^{\text{th}}$ forces the laser to switch from the low-power solution (now unstable) to the high-power solution (stable); the laser turns on. The behavior of the laser as $q_{\text{inj}}$ decreases depends on the quality of the cavity. For a good cavity, the laser continues to operate at high power as $q_{\text{inj}}$ decreases. For a bad cavity, the laser only continues to operate at high power if the solution is stable when $q_{\text{inj}} = 0$. Concannon and Gauthier [1994] studied the stability behavior for other values of the pump rate and found that the injection threshold $q_{\text{inj}}^{\text{th}}$ decreases for higher values of the pump rate as shown in fig. 4b and is given approximately by

$$
\frac{q_{\text{inj}}^{\text{th}}}{q_{\text{sat}}} \approx \frac{\Delta N_o}{\Delta N_o^{\text{th}}} - \frac{1}{2} \sqrt{4 \left( \frac{\Delta N_o}{\Delta N_o^{\text{th}}} \right)^2 - 1} 
$$

(3.13)

when $\Delta N_o \geq \Delta N_o^{\text{th}}$. At the minimum pump rate ($\Delta N_o = \Delta N_o^{\text{th}}$) one has $q_{\text{inj}}^{\text{th}} \approx 0.134q_{\text{sat}}$. In practice, a pulse (peak photon number $q_{\text{inj}}^{\text{p}}$) is injected into the laser rather than a continuous-wave beam. When the laser can (cannot) adiabatically follow the temporal variation of the pulse, $q_{\text{inj}}^{\text{p}} = q_{\text{inj}}^{\text{th}}$ ($q_{\text{inj}}^{\text{p}} \geq q_{\text{inj}}^{\text{th}}$).

The transient behavior of the laser described above was explored by Concannon and Gauthier [1994] through numerical integration of eqs. (3.2) and (3.3). Figure 5 shows how the laser responds to injected trigger pulses for a good cavity ($\gamma/\kappa = 2$) when the pump rate is greater than the threshold pump rate ($\Delta N_o = 1.2\Delta N_o^{\text{th}}$) and when there are no photons in the cavity initially. For a weak trigger pulse (peak amplitude $q_{\text{inj}}^{\text{p}} = 0.1q_{\text{sat}}$, fig. 5a) the laser is not driven above threshold, while for a slightly stronger pulse (peak amplitude $q_{\text{inj}}^{\text{p}} = 0.12q_{\text{sat}}$,}
Fig. 5. Predicted temporal evolution of the photon number for (a) weak and (b) slightly stronger injected signal. The two-photon laser turns on (b) only when a sufficient number of photons are injected into the cavity. From Concannon and Gauthier [1994].

fig. 5b) the laser is driven above threshold and attains a constant amplitude after the injected pulse switches off. This is in good agreement with the injection threshold $q_{\text{inj}}^{\text{th}} \approx 0.11 q_{\text{sat}}$ for this pump rate using eq. (3.13).

The most important conclusion that can be drawn from the rate-equation model is that the intensity circulating in the laser resonator is always greater than or equal to the two-photon saturation intensity. Hence, the gain medium must have a low two-photon saturation intensity to avoid the high intensities that tend to magnify competing nonlinear optical processes. Fortunately, low saturation intensities go hand-in-hand with large two-photon gain, as discussed below.

3.2. Coherent effects in two-photon amplification

The rate-equation model described above does a reasonable job of capturing some of the essential characteristics of the two-photon laser. Additional important features arise from the coherent driving of the two-photon dipole moment (Takatsuji [1975]), which becomes relevant when the dipole dephasing rate is comparable to the population decay rate. To determine the influence of these coherent effects, a semi-classical theory of the interaction of the laser field with the atoms is quite revealing as described by Allen and Stroud [1982] and Meystre and Sargent [1991], for example. Following Gauthier and Concannon [1994], consider the interaction of a monochromatic field $E(r,t) = [\epsilon E \exp(-i\omega t) + \text{c.c.}]$ with a three-level atomic system possessing an inverted two-photon transition $|b\rangle \leftrightarrow |a\rangle$ as shown in fig. 6a. Since these states have the same parity, they are not connected by a normal one-photon transition and hence the electric-dipole matrix element connecting them is zero. A real intermediate level $|i\rangle$ with opposite parity is located near the virtual
intermediate level of the two-photon transition (detuning $\Delta_{ia} = \omega - \omega_{ia}$, Bohr frequency $\omega_{ia}$ for the $|i\rangle \leftrightarrow |a\rangle$ transition) to enhance the two-photon transition rate. Since $\Delta_{ia}$ is assumed to be small, other intermediate levels can be ignored. The electric-dipole matrix element of the $|b\rangle \leftrightarrow |i\rangle$ ($|i\rangle \leftrightarrow |a\rangle$) transition is denoted by $\mu_{bi}$ ($\mu_{ia}$) and the corresponding Rabi frequency is given by $\Omega_{bi} = 2\mu_{bi} \cdot eE/h$ ($\Omega_{ia} = 2\mu_{ia} \cdot eE/h$). Population in the upper level can decay spontaneously to the intermediate level (rate $\gamma_{bi}$) which subsequently decays to the lower level (rate $\gamma_{ia}$). For simplicity, assume that $\gamma_{ia} \gg \gamma_{bi}$ so that essentially no population builds up in the intermediate level; Roldán, de Valcárcel and Vilaseca [1993] has explored the consequences of this approximation. Note that this model can be generalized to account for many intermediate states (Meystre and Sargent [1991]), but the physics of the problem is primarily embodied in this simple analysis.

The only nonzero elements of the density matrix are: the populations $\rho_{aa}$ and $\rho_{bb}$; the one-photon coherences $\rho_{bi}$ and $\rho_{ia}$; and the two-photon coherence $\rho_{ba}$. The equations of motion for the density matrix are simplified using the one- and two-photon rotating-wave approximations (Narducci, Eidson, Furcinitti and Eteson [1977], Allen and Stroud [1982]) and by adiabatic elimination of the one-photon coherences (performed formally by setting $\dot{\rho}_{bi} = \dot{\rho}_{ia} = 0$), which is valid as long as $\Delta_{ia} \gg \gamma_{bi}, \Omega_{bi}, \Omega_{ia}$. It is found that the equations of motion for the two-photon population inversion $w = (\rho_{bb} - \rho_{aa})$ and the slowly varying two-photon coherence $\sigma_{ba} = \rho_{ba} \exp(i2\omega t)$ are given by

$$\frac{dw}{dt} = -\gamma_{p}(w - w_{eq}) - i[\Omega^{*} \sigma_{ba} - \sigma_{ab} \Omega],$$

(3.14)
and
\[ \frac{d\sigma_{ba}}{dt} = [i(\Delta_2 - \delta_s) - \gamma_\perp] \sigma_{ba} - i \frac{\hbar}{2} \Omega, \]  \hspace{1cm} (3.15)\]

In these equations, the inversion decay rate is denoted by \( \gamma_\parallel \simeq (\gamma_{bi} + R) \), \( w_{eq} \) is the equilibrium inversion in the absence of the field, \( \Delta_2 = 2\omega - \omega_{ba} \) is the two-photon detuning, \( \gamma_\perp \) is the two-photon coherence dephasing rate,
\[ \Omega = \frac{\Omega_{bi} \Omega_{ia}}{2\Delta_{ia}} = \frac{(\mu_{bi} \cdot \epsilon)(\mu_{ia} \cdot \epsilon)}{2\hbar^2 \Delta_{ia}} |E|^2 \]  \hspace{1cm} (3.16)\]
is the two-photon Rabi frequency, and
\[ \delta_s = \frac{|\Omega_{bi}|^2 - |\Omega_{ia}|^2}{4\Delta_{ia}} = \frac{(|\mu_{bi} \cdot \epsilon|^2 - |\mu_{ia} \cdot \epsilon|^2)}{\hbar^2 \Delta_{ia}} |E|^2 \]  \hspace{1cm} (3.17)\]
is the ac Stark shift of the two-photon transition (Liao and Bjorkholm [1975]), where \( \epsilon \) is the polarization unit vector. Note that eqs. (3.14) and (3.15) are nearly identical to the corresponding equations for a driven two-level atom and are often collectively referred to as the two-photon two-level model (Meystre and Sargent [1991]).

Important new features of this model are the proportionality of the two-photon Rabi frequency to the intensity rather than the field amplitude, and the presence of the ac Stark shift \( \delta_s \) of the transition frequency, which is also proportional to the intensity of the field. As can be seen from eq. (3.15), maximal atom–field coupling occurs when the detuning of the field is adjusted to the ac-Stark-shifted resonance (\( \Delta_2 = \delta_s \)) rather than to the bare-atom resonance (\( \Delta_2 = 0 \)). This shift plays an important role in the nonlinear and quantum-dynamical properties of two-photon lasers, as will be discussed in § 8, and gives rise to optical bistability when a two-photon absorber is placed in an optical resonator (Giacobino, Devaud, Biraben and Grynberg [1980]), and to cavity-less optical bistability for beams counterpropagating through a two-photon amplifier (Gingras and Denariez-Roberge [1985]).

The optical field induces a polarization in the atoms as determined by eqs. (3.15) and (3.14), which acts back on the field in a self-consistent manner through Maxwell’s equations. The macroscopic polarization of the medium is given by \( \mathbf{P}(r, t) = \eta \langle \hat{\mathbf{p}} \rangle = \eta \text{Tr}(\hat{\rho} \hat{\mathbf{p}}) \), where \( \eta \) is the atomic number density and the angle brackets denote a quantum-mechanical expectation value. Concannon [1996] has investigated the case of a continuous-wave beam of light passing through a low-
gain two-photon amplifier and finds that the output \(I_{\text{out}}\) and input \(I_{\text{in}}\) intensities are related through

\[
\frac{I_{\text{out}}}{I_{\text{in}}} - 1 = \frac{I_{\text{in}}G^{(2)}L}{1 + (I_{\text{in}}/I_{\text{sat}})^2},
\]

where the two-photon gain coefficient is given by

\[
G^{(2)} = \frac{\hbar \omega \eta(R - \gamma_{bi})}{2I_{\text{sat}}^2},
\]

and the saturation intensity is defined through the relation

\[
\frac{1}{I_{\text{sat}}^2} = \frac{(|\vec{m}_{bi} \cdot \vec{e}|^2 |\vec{m}_{ia} \cdot \vec{e}|^2 \gamma_{\perp}}{\varepsilon^2 c^2 \hbar^4 \Delta_{ia}^2 \gamma_{\parallel}[(\Delta_2 - \delta_5)^2 + \gamma_{\perp}^2].
\]

Figure 7 shows the predicted laser beam amplification as a function of the input intensity. It is seen that there is no gain for zero input intensity, in contrast to a one-photon amplifier where the gain is independent of the input intensity until saturation sets in. From the figure, it is also seen that the gain takes on its maximum value \(I_{\text{sat}}G^{(2)}L/2\) at \(I_{\text{in}} = I_{\text{sat}}\). In light of the scaling of \(G^{(2)}\) (see eq. 3.19), the maximum gain is inversely proportional to the saturation intensity, which can be minimized by taking \(\Delta_2 = \delta_5\), using transitions with large dipole matrix elements, and using an atom with nearly degenerate transitions frequencies so that \(\Delta_{ia}\) is small (or by operating the laser in the non-degenerate mode with the frequencies adjusted so that \(\Delta_{ia}\) is small).

While the situation of a continuous-wave beam leads to a simplified analysis, it is also of interest to consider how pulses of light propagate through a two-photon amplifier since it is easier to attain the high peak intensities needed to
reach the saturation intensity with pulsed lasers. Narducci, Eidson, Furcinitti and Eteson [1977] considered the case of an ultrashort pulse propagating through a two-photon amplifier, where the pulse width is much smaller than the transition dephasing rate. They found that there are no asymptotically stable solutions; the pulse grows in amplitude, compresses in duration, and eventually breaks up into multiple pulses.

The results obtained from the density-matrix formalism are connected to the rate equations used in the previous section under conditions when there is large dephasing of the two-photon coherence \((\Gamma \gg \Delta_2, \delta_s, \gamma)\). In this case, the coherence can be adiabatically eliminated from eq. (3.14) using eq. (3.15) with \(\dot{\delta}_{\text{pa}} = 0\). By comparison with eq. (3.3), it is found that the two-photon rate coefficient is given by

\[
B^{(2)} = \frac{\hbar^2 \omega^2 c^2 \gamma_i}{32 V^2 I_{\text{sat}}^2}.
\]  

(3.21)

From eq. (3.21), it is straightforward to determine the saturation photon number using eq. (3.8) and the threshold inversion using eq. (3.11).

It was an understanding of the scaling of \(G^{(2)}\) on the dipole matrix elements and the intermediate-state detuning that led to the development of the first two-photon quantum oscillators.

3.3. Competing processes

Based on the preliminary estimates by Sorokin and Braslau [1964] as described above, it would appear to be straightforward to build a two-photon laser. Unfortunately, experimental realization of this new quantum oscillator was hindered by competing nonlinear optical effects that prevented the attainment of an inversion on the two-photon transition or obscured the lasing process. To lowest order in perturbation theory, the two-photon stimulated emission process is a third-order nonlinear optical effect and hence any other third-order effect can compete with it. Competing processes include self-focusing of the transverse profile of a laser beam (Carman [1975]), which degrades the spatial coherence of the beam and limits its intensity, and photo-ionization of the atoms in the gain medium by excitation of the atom from the state \(|b\rangle\) to the continuum by absorbing one or more photons (Bay, Elk and Lambropoulos [1995]). It was found in studies of candidate two-photon amplifying media that the competing effects usually dominate.

Another primary competing process is normal one-photon lasing that can occur on the \(|b\rangle \leftrightarrow |i\rangle\) transition for the case when the state \(|i\rangle\) is lower
in energy than the state $|b\rangle$. For large $\Delta_{in}$, the two-photon lasing frequency is spectrally isolated from the possible one-photon lasing frequency and the latter can be avoided by using an optical resonator constructed with mirrors that are highly transparent at the one-photon resonance and highly reflective at the two-photon lasing frequency when the one-photon gain is sufficiently low. For small $\Delta_{in}$, which enhances the two-photon stimulated emission rate, the possible one- and two-photon lasing frequencies will be close. The one-photon lasing can be avoided by using a short optical resonator constructed with ultra-low-loss, high-reflectivity mirrors, such as that used in cavity quantum electrodynamics experiments (see, for example, An, Sones, Fang-Yen, Dasari and Feld [1997]), that selectively enhances the two-photon laser frequency and not the one-photon laser frequency. For this approach to succeed, no mode of the cavity (longitudinal or transverse) can overlap with the one-photon amplification resonance, which is difficult to achieve in practice because the mode frequencies tend to be dense except in mode-degenerate cavities.

Even without an optical resonator, the one-photon amplification process can deplete the two-photon inversion when the gain is high, giving rise to the quantum-initiated processes of superfluorescence or amplified spontaneous emission (superfluorescence occurs when the inversion is created on a time scale that is fast in comparison to the dipole dephasing time of the transition whereas amplified spontaneous emission dominates when the excitation is slow, as discussed by Malcuit, Maki, Simkin and Boyd [1987]). These competing processes were first pointed out by Nakatsuka, Okada and Masahiro [1974] and discussed in detail by Grischkowsky, Luy and Liao [1975] in their proposal for creating a two-photon inversion by the adiabatic rapid-passage technique. The threshold for superfluorescence or amplified spontaneous emission occurs when the one-photon intensity gain coefficient $G^{(1)} \approx 30/L$ so that the single-pass increase in the intensity of a beam propagating through the medium of length $L$ is $\sim \exp(30)$. The only way to avoid this competing process is to reduce the atomic number density (which also reduces the two-photon gain$^\dagger$). To estimate the limitation imposed by this competing process, consider a two-photon transition where $|\mu_{bi} \cdot \epsilon| \approx |\mu_{in} \cdot \epsilon|$ and the two-photon coherence dephasing rate is similar to the population decay rate ($\gamma_\perp \approx \gamma_\parallel$). The ratio of the one-photon gain coefficient to the maximum two-photon gain is then given approximately by

$$\frac{G^{(1)} L}{\frac{1}{2} (G^{(2)} I_{sat} L)} \approx \frac{4|\Delta_{in}|}{\gamma_{bi}^\perp},$$

(3.22)

where $\gamma_{bi}^\perp$ is the coherence dephasing rate for the upper transition. For one
proposed scheme involving the $6P_{3/2} \leftrightarrow 4P_{3/2}$ two-photon transition of atomic potassium (Bay and Lambropoulos [1994]), this ratio is of the order of $10^2$ for degenerate two-photon lasing so that the maximum two-photon gain is $\sim 3 \times 10^{-4}$ when the one-photon gain is set to the superfluorescent limit ($G^{(1)}L \approx 30$). To achieve lasing using such a small gain requires the use of an ultra-low-loss optical resonator. Petrosyans and Lambropoulos [1999] have shown that two-photon lasing can coexist with one-photon lasing that occurs on an adjacent mode of the optical resonator, suggesting that this competing effect need not be devastating.

Another important competing process, identified by Carman [1975], is stimulated anti-Stokes Raman scattering of the two-photon laser field from the inverted two-photon transition, as shown schematically in fig. 6b. In this process, a photon is annihilated and an anti-Stokes photon is generated as the atom undergoes a transition from state $|b\rangle$ to state $|a\rangle$, thereby depleting the atomic inversion and removing photons from the two-photon laser field. For most materials, it was found that the rate of anti-Stokes Raman scattering far exceeds that for two-photon stimulated emission because the intermediate virtual state of the former process is closer to a real atomic transitions $|i'\rangle$ in comparison to the latter so that it enjoys a larger resonance enhancement. For example, Głownia, Arjavalingam and Sorokin [1985] found that stimulated anti-Stokes Raman scattering dominates two-photon stimulated emission in laser-pumped vapor-phase triethylenediamine. Carman [1975] suggested that two-photon amplification might dominate when an intense pulse produced by a CO$_2$ laser (wavelength $10.6 \mu$m, used in laser-fusion experiments at Los Alamos in the 1970s and 1980s) passes through a gas of excited iodine, acting as an "afterburner" by increasing the energy by a factor of ten.

Yet another competing process that may have played a role in the first experiments on singly stimulated two-photon emission and two-photon lasing is parametric wave mixing. It can arise in experiments when the laser beams that are used to create the two-photon inversion interact simultaneously with the photons involved in the stimulated-emission process, and is especially of concern when the pumping and stimulating fields travel along the same axis. In wave-mixing processes, the atom begins and ends in the same quantum state, there is a well-defined phase relation among the interacting fields, and the momentum of the fields must be conserved for efficient generation (the phase-matching condition). Ascertain the presence of parametric wave mixing can be complicated by the presence of superfluorescence or amplified spontaneous emission that produce intense beams of light self-generated in the medium whose phase fluctuates because the process is initiated or sustained by quantum noise.
The self-generated fields can then interact with the incident pump laser beams via parametric wave mixing, generating new fields that might obscure or prevent two-photon lasing. In most situations, parametric wave mixing can be suppressed using an experimental geometry where the pump laser beam(s) propagate in a direction orthogonal to the beam(s) undergoing two-photon amplification or lasing because the interaction cannot be phase-matched. This technique was used by Gauthier, Wu, Morin and Mossberg [1992] in the realization of the dressed-state two-photon laser and by Pfister, Brown, Stenner and Gauthier [2001] in the realization of the Raman two-photon laser.

Competing nonlinear optical process limited the usefulness of most candidate two-photon gain media, hampering research on two-photon lasers, although not for lack of effort (see, for example, Bethune, Lankard and Sorokin [1978] and Glownia, Gnass and Sorokin [1996]). In the intervening years, researchers developed one-photon lasers that had high energy, high power, short pulse durations, and tunability. Thus, many of the original applications envisioned for two-photon lasers were filled by one-photon lasers. Only with the understanding that two-photon lasers should produce beams of light with novel and potentially useful quantum-statistical properties was interest in them rekindled, as will be described in § 8.

§ 4. Two-photon amplification and lasing

Overcoming the competing effects described in the previous section required a thorough understanding of two-photon processes and novel experimental techniques. The first reported observation of laser beam amplification due to the stimulated emission process was by Loy [1978] at IBM who created a transient inversion on the ammonia ν₂ = 0→ ν₂ = 2→ (J = 5, K = 4, M = ±5) two-photon transition and succeeded in amplifying a CO₂ laser beam (10 μm wavelength) by ~0.2% over a brief 25 ns time interval. The inversion was created using the method of two-photon adiabatic inversion in a manner similar to that put forth by Grischkowski and Loy [1975] and Grischkowski, Loy and Liao [1975], also of IBM.

In the experiment, two laser pulses counterpropagated through a 40-cm-long low-pressure ammonia cell to both create and probe the inversion. As the pulses entered the cell, the two-photon transition frequency was linearly swept through resonance with the sum frequency of the two beams via the dc Stark effect over an interval of 200–300 ns, and then was quickly swept back to the its original frequency over an interval of ~ 50 ns. Complete inversion of the two-photon transition occurred when the sweep interval was short in comparison to
the lifetime of the upper state (~4\,\mu s at an ammonia pressure of 9\,\text{mTorr}) and the rate of the frequency change was adiabatic, that is, less than the square of the two-photon Rabi frequency (see eq. 3.16). The first condition was satisfied by keeping the ammonia pressure low (9\,\text{mTorr}) and the second condition was satisfied by adjusting the time over which the transition frequency was swept and the intensities of the laser pulses. During the 200–300\,\text{ns} linear sweep of the transition frequency, both conditions were satisfied; most of the population of the lower energy state was transferred to the excited state and large two-photon absorption of the laser pulses was observed. During the resetting of the transition frequency, most of the population was returned to the lower state, resulting in a brief interval over which the laser pulse was amplified. At higher ammonia vapor pressures, the magnitude of the two-photon gain decreased because the inversion decay rate increased from collisions so that the inversion decayed before the resetting of the atomic frequency. Because the ammonia system displayed low gain and low energy-storage capabilities, Loy concluded that it would not be a useful candidate for building a two-photon laser.

Taking a different approach to the problem, Schlemmer, Frolich and Welling [1980] investigated continuous-wave non-degenerate two-photon amplification that was resonantly enhanced by an intermediate atomic level. In their experiment, they measured simultaneous lasing on the neon 3s_2 \leftrightarrow 3p_4 (3.39\,\mu m transition wavelength) and 3p_4 \leftrightarrow 2s_2 (2.40\,\mu m transition wavelength) cascade transition using two optical resonators, each of which supported normal one-photon lasing on one of the transitions. They observed that the power tuning curve of one of the lasers contained an off-resonance feature when the frequency of the other laser was tuned to the edge of its tuning range so that the sum of the two laser frequencies was equal to the 3s_2 \leftrightarrow 2s_2 two-photon transition frequency. For this detuned operation and a ring-laser geometry, they also found that the emission direction for the two lasers was unidirectional and in opposite directions, which they attributed to the reduction of the Doppler effect for counterpropagating beams. While it is clear that two-photon amplification was playing a role in their experiment, they did not attempt to ascertain the relative contributions of two-photon and off-resonant one-photon amplification. I am not aware of any follow-up experimental research to determine whether two-photon lasing could be obtained in this system for larger intermediate-state detunings, which is unfortunate because the neon system appears to be very promising for making a non-degenerate two-photon laser in the infrared part of the spectrum.

Soon after these experiments in ammonia, Nikolaus, Zhang and Toschek [1981] reported the observation of two-photon lasing in a laser-pumped,
high-density lithium vapor. In the experiment, 4 ns pulses generated by two tunable dye lasers counterpropagated through a lithium heat pipe operated at a temperature of 720°C, corresponding to an atomic number density of \( \sim 10^{16} \) cm\(^{-3} \). The frequency \( \omega_1 \) of dye laser 1 was tuned to the high-frequency side of the \( 2s^2S \leftrightarrow 2p^2P^0 \) one-photon transition, and the frequency \( \omega_2 \) of dye laser 2 was adjusted so that \( \omega_1 + \omega_2 \) was equal to the \( 2s^2S \leftrightarrow 4f^2F^0 \) transition. The laser pulses transferred most of the population to the \( 4f^2F^0 \), giving rise to prompt superfluorescent emission that quickly transferred population to the \( 3d^2D \) state, resulting in a transient two-photon inversion between the \( 3d^2D \) upper state and the \( 2s^2S \) ground state. Note that two-photon excitation of the \( 4f^2F^0 \) should be forbidden because the \( 2p^2P^0 \leftrightarrow 4f^2F^0 \) transition is electric-dipole forbidden; they presumed that it occurred due to the small electric quadrupole moment connecting the states. Subsequent detailed analysis by Sparbie, Boller and Toschek [1996] indicated that the lasers actually excited a nearby Li\(_2\) molecular state, which rapidly dissociated, leaving atomic lithium in the excited \( 4f^2F^0 \) as well as other nearby states.

Once a two-photon inversion was established, photons from dye laser 1 at frequency \( \omega_1 \) singly stimulated emission at frequency \( \omega_x \) that was observed to propagate along the same axis, but opposite to the propagation direction of the stimulating beam. In a single pass through the vapor, the light at frequency \( \omega_x \) attained a maximum peak power of 30 W. Nikolaus, Zhang and Toschek [1981] referred to this light as ‘laser emission’, although no optical resonator surrounded the medium\(^2\); it is more reminiscent of the super-radiant light generated by nitrogen or excimer lasers (Svelto [1989]). Nikolaus, Zhang and Toschek [1981] stated that the interaction can not be due to a parametric wave-mixing process because such an interaction could not be phase-matched with a geometry of counterpropagating pump beams, and the emission showed no phase correlation with the beam produced by dye laser 2, although they did not describe how this measurement was performed. In a second experiment, they injected light from a third dye laser whose frequency \( \omega_3 \) was set to precisely one-half the \( 3d^2D \leftrightarrow 2s^2S \) two-photon transition frequency. They observed that the beam was amplified by up to 20%, which they attribute to two-photon amplification.

These experiments using laser-pumped lithium vapor were met by some skepticism from the IBM group. Jackson and Wynne [1982] conducted experiments in

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\(^2\) This experiment is similar to the earlier observation of singly stimulated two-photon emission by Yatsiv, Rokni and Barak [1968], with the exception that there is greater flexibility in choosing the frequency \( \omega_1 \) because it is generated by a tunable laser.
laser-pumped sodium vapor and observed light generated at $\omega_k$ whose frequency characteristics were analogous to that observed by Nikolaus, Zhang and Toschek [1981]. Rather than attributing the generated light to two-photon emission, they showed that it could be due to parametric wave mixing. The interaction can be phase-matched even with the counterpropagating pump-beam geometry, but only for the case when the generated light is emitted counterpropagating with the pump beam at $\omega_1$, consistent with their observation. Jackson and Wynne [1982] noted that two-photon emission should be generated in both directions, inconsistent with their observation. In a separate experiment, they were not able to observe two-photon amplification when a third laser was tuned to the analogous two-photon transition. Since Nikolaus, Zhang and Toschek [1981] only reported on measurements of generated light that counterpropagated with $\omega_1$, the claim of Jackson and Wynne [1982] could not be tested.

Following up on the experiments of Jackson and Wynne [1982], Gao, Eidson, Squicciarini and Narducci [1984] observed singly stimulated two-photon emission on the sodium $8P \leftrightarrow 3P$ two-photon transition, which was inverted by single-photon excitation of the $3S \leftrightarrow 8P$ transition. Their experiment was unique in that they only attempted to obtain two-photon emission after the pump laser pulse had exited the vapor but before the inversion decayed via cascade one-photon spontaneous emission, thereby suppressing all possible phase-matched interactions involving the pump beam. They observed singly stimulated emission at 1.14 $\mu$m that was correlated and counterpropagated with the stimulating beam at 0.74 $\mu$m. They did not observe any singly stimulated emission that copropagated along the direction of the stimulating beam. They attributed this asymmetry of the emission to the near cancelation of the Doppler broadening of the two-photon transition, thereby increasing the gain, which occurs only for the counterpropagating geometry. Thus, they demonstrated that an asymmetry in the emission direction does not necessarily indicate a phase-matched interaction, as put forth by Jackson and Wynne [1982].

Many years later, Sparbier, Boller and Toschek [1996] performed a series of detailed experiments in laser-pumped lithium to shed additional light on the emission observed by Nikolaus, Zhang and Toschek [1981]. One of the main findings is that the pump lasers actually excited a lithium dimer state that dissociated, leaving behind atomic lithium in the $4f^2F^0$ state. They suggest that this excitation mechanisms suppressed parametric wave-mixing processes involving this state since dissociation is believed to be an incoherent process that does not depend on the relative phase of the exciting fields under their experimental conditions. In addition, analysis of the spectrum and state of polarization of the generated light and its dependence on the tuning of the
pump laser beam frequencies revealed that two-photon emission and parametric wave mixing coexisted with nearly equal strength. They observed that the emission direction was asymmetric, consistent with the experiments of both Jackson and Wynne [1982] and Gao, Eidson, Squicciarini and Narducci [1984]. They did not attempt to observe the two-photon emission process after the pump laser beams had exited the lithium vapor cell to test whether the presence of the pump beams was required for the observed emission, but it seems clear that several scattering processes, including two-photon stimulated emission, occurred simultaneously.

These early experiments bring to light the great difficulties in constructing two-photon lasers in the presence of competing effects and point to the need for a drastically different experimental approach.

§ 5. The two-photon maser

The crucial advance that revolutionized research on two-photon quantum oscillators was the use of experimental techniques developed for research in cavity quantum electrodynamics (CQED); see, for example, Berman [1994]. In such experiments, only a few atoms at a time, often produced by an atomic beam source or a cloud of cooled and trapped atoms, pass through a small-volume ultrahigh-$Q$ resonator. No windows can be placed in the resonator to contain the atoms because the small reflection or scattering losses from the window surfaces would seriously degrade the cavity $Q$, and hence the resonator must be placed in the same vacuum chamber as the atom source. In the mid 1980s, experiments were underway at l’Ecole Normale Supérieure in Paris and at the Max-Planck-Institut für Quantenoptik in Garching that combined the ultralarge electric-dipole moments characteristic of alkali-metal Rydberg atomic transitions in the microwave part of the spectrum with ultrahigh-$Q$ superconducting cavities for studying the strong-coupling regime of CQED.

In the CQED research most related to the present discussion, Brune, Raimond and Haroche [1987] investigated the possibility of building a two-photon maser based on $nS_{1/2} \leftrightarrow (n - 1)S_{1/2}$ two-photon transitions that are enhanced

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$^3$ I note that some authors (see, e.g., Ovadia and Sargent [1984], Swain [1988] and Ning and Haken [1989a]) classify an experiment by Gryenberg, Giacobino and Biraben [1981] as an observation of two-photon lasing. However, this experiment used two-photon excitation of an atomic state that served as the upper level of a normal one-photon laser.
by the \((n - 1)P_{3/2}\) intermediate state, where \(n\) is the principal quantum number. They discovered that the Rydberg states of the alkali-metal atoms are especially favorable for enhancing the degenerate-frequency two-photon stimulated emission rate because the intermediate state detuning \(\Delta_{ia}\) is nearly zero for one value of the principal quantum number, denoted by \(n_o\), due to the slow variation of the quantum defects of the energy levels as a function of \(n\). Thus, by taking \(n = n_o \pm 1\), it is possible to spectrally separate the degenerate two-photon resonance from the competing one-photon resonance while keeping the rate large. For rubidium, \((n_o - 1) = 38\) and \(\Delta_{ia}/2\pi = 39\) MHz for \((n - 1) = 39\), which should be compared to the two-photon maser frequency \(\omega/2\pi = 68.42\) GHz, and cavity linewidths of the order of 1 kHz for a cavity \(Q\) of \(10^8\) that were available at the time.

Using typical parameters for the rubidium system and available superconducting microwave resonators, Brune, Raimond and Haroche [1987] estimated an amazingly low threshold for two-photon masing of only one atom and a few tens of photons on average in the resonator! Thus, the two-photon maser would operate in what is known as the ‘micromaser’ regime. They envisioned that a dilute stream of atoms would pass through the resonator in a time \(t_{int}\) on average with a mean time \(t_{at}\) between atoms, where \(t_{at} \geq t_{int}\) so that there is at most one atom in the cavity on average. The cavity lifetime \(t_{cav} = Q/\omega\) was assumed to be much longer than both \(t_{at}\) and \(t_{int}\) so that the atoms, one at a time, would interact with the cavity photons that had been left behind from previous atoms. The threshold conditions for masing, expressed in terms of these quantities, are given by

\[
\Omega t_{int} = -\frac{\omega (\mu_{bi} \cdot \epsilon)(\mu_{ia} \cdot \epsilon)}{4V\epsilon_0 c \hbar \Delta_{ia}} \bar{q} t_{int} \simeq \pi, \tag{5.1}
\]

\[
t_{at} \simeq \frac{2t_{cav}}{\bar{q}}, \tag{5.2}
\]

where \(\bar{q}\) is the mean cavity photon number and \(\Omega\) is the two-photon Rabi frequency, given approximately by eq. (3.16). Threshold conditions (5.1) and (5.2) are equivalent to the conditions (3.4) (with \(\Delta N(0) = \Delta N_{0}^2\)) and (3.11), respectively, discussed in § 3. For the rubidium parameters given above, \(t_{int} = 25\) \(\mu s\), \((\mu_{bi} \cdot \epsilon) = 1,443ea_o\), \((\mu_{ia} \cdot \epsilon) = 1,479ea_o\), \(Q = 10^8\) \((t_{cav} = 0.23\) ms\), and \(V = 70\) mm\(^2\), the threshold conditions predict \(t_{at} = 14.3\) \(\mu s\) and \(\bar{q} = 32\).

Brune, Raimond and Haroche [1987] also pointed out that the two-photon spontaneous emission rate for this atomic transition is relatively large so that the threshold condition for the minimum number of photons can be satisfied by spontaneously generated photons emitted into the cavity and does not require
injection of an external field. They developed a full quantum theory for the two-photon maser that takes into account the ac Stark shifts, and predicted that the time it takes for the mean photon number to go above threshold will show a significant lethargy because the probability for a large enough quantum fluctuation was somewhat low. In follow-up studies, Davidovich, Raimond, Brune and Haroche [1987] and Davidovich, Raimond, Brune and Haroche [1988] further developed and analyzed a quantum theory of the two-photon micromaser. Using a master equation approach for describing the complete state of the field in the cavity, they found that the photon-number statistics are sub-Poissonian for a wide range of atomic injection times and hence the device could be used to generate squeezed states of the electromagnetic field (see § 8).

5.1. Realization of the two-photon maser

The first demonstration of a two-photon maser based on this proposal was reported by Brune, Raimond, Goy, Davidovich and Haroche [1987] who used the rubidium atomic transitions discussed in the example above. The energy-level structure for the two-photon transition and the experimental setup is shown in fig. 8. A collimated thermal atomic beam of rubidium atoms passed through three laser beams that propagated in a direction orthogonal to the motion of the atoms to cancel Doppler broadening. The lasers sequentially excited the

![Fig. 8. Two-photon maser. Top panel: energy-level diagram of Rb showing the relevant states and excitation pathway. Bottom panel: experimental setup showing the atomic and optical pumping beams, superconducting microwave cavity, and field-ionization region. From Brune, Raimond, Goy, Davidovich and Haroche [1987].](image-url)
atoms from the ground $5S_{1/2}$ state to the $40P_{3/2}$ Rydberg state just before they entered a liquid-He-cooled superconducting niobium microwave resonator with a $Q$ of $10^8$ at a temperature of 1.7 K. Just as the atoms entered the resonator structure, but before they entered the resonator itself, a coherent microwave field transferred the atoms from the $40P_{3/2}$ state to the $40S_{1/2}$ state, which served as the upper state of the two-photon transition. The excited atoms entered the resonator and interacted with the cavity field. The frequency of the resonator was adjusted precisely to the two-photon transition frequency (measured to be $68.41587 \pm 0.00001$ GHz in a separate experiment) by mechanical deformation of the resonator. Note that the lifetime of the upper state is long in comparison to the transit time of an atom through the resonator so that the number of atoms in this state should remain constant unless the interaction between the atom and cavity de-excites the atom due to lasing processes, for example.

For such a high-$Q$ resonator, very few photons are emitted by the cavity so the researchers instead measured the number of atoms in each atomic state. After the atoms exited the resonator, they passed through field-ionization plates upon which a linearly increased voltage was applied. In this manner, the ionization current is directly proportional to the applied voltage, which in turn is directly proportional to time during the voltage sweep. Figure 9a shows the ionization current as a function of time for a low flux of atoms $[1/t_{at} = (8 \pm 4) \times 10^4$ atoms s$^{-1}]$, where it is seen that rubidium atoms were only found in the 40P and 40S states, indicating that the maser was below threshold. For larger atomic fluxes $[1/t_{at} = (2 \pm 1) \times 10^5$ atoms s$^{-1}$, fig. 9b], more than half the atoms that entered in the 40S state exited in the 39S state, indicating that the laser was above threshold. Furthermore, the measured threshold atomic flux

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*Fig. 9. Two-photon maser operating characteristics. Population of the atomic states when the maser operates (a) below threshold and (b) above threshold. (c) Tuning the two-photon maser through the gain resonance. From Brune, Raimond, Goy, Davidovich and Haroche [1987].*
agreed well with the predicted value of \(1/t_{\text{at}} = 1/14.3 \mu s = 7 \times 10^4 \text{ atoms s}^{-1}\). The maser also displayed a very strong dependence on the frequency of the microwave resonator as shown in fig. 9c when the atomic flux was high. The relative transfer of population from the upper to the lower state was monitored and it was found that high transfer (indicating masing) occurred over a tuning range of only \(\sim 40 \text{ kHz}\). This tuning range is much smaller than the detuning \(\Delta_{\text{iu}}\) from the competing detuned one-photon \(|b\rangle \leftrightarrow |i\rangle\) resonance, providing further evidence that they were observing pure two-photon masing. Finally, they observed very long turn-on times of the maser (up to a few seconds) when the atomic flux was set close to the threshold value and the cavity initially started in the vacuum state, consistent with the predictions of Brune, Raimond and Haroche [1987].

This one experimental result has led to numerous theoretical investigations of the two-photon micromaser because the physical parameters of the experimental system are well known and only a small number of atoms and photons are present in the resonator. It has become a universal system for investigating the nonlinear interaction between light and matter from first principles. Recent theoretical research, for example, includes a study of cooperativity when two atoms are present in the resonator simultaneously (Ashraf and Toor [2000]), the influence of atomic decay on the linewidth of a two-photon micromaser (Hamza and Qamar [2000]), the effects of two-photon atomic-motion-induced amplification of radiation (Zhang and He [1999]), and the existence of trapping states and the generation of a photon-number state (Alexanian, Bose and Chow [1998]). A complete review of the body of research on theoretical aspects of two-photon micromasers is beyond the scope of this chapter.

§ 6. The dressed-state two-photon laser

The major breakthrough in the development of optical two-photon quantum oscillators came with the realization that it is possible to 'engineer' a near-ideal two-photon gain medium that possesses a small and adjustable detuning, as discussed by Lewenstein, Zhu and Mølmer [1990]. It consists of a two-level atom with an electric-dipole-allowed transition driven by an intense, continuous-wave, near-resonant laser field. The coupling is so strong between the atom and the intense field that it make more sense to think of it as a composite atom-field system (a 'dressed' atom) rather than separate entities. By combining dressed atoms with optical CQED experimental techniques (Kimble [1994]), they showed that it is possible to realize a two-photon laser that operates with only
a few thousand atoms and photons in the cavity, and that it is possible to avoid competing nonlinear effects by using an appropriate experimental geometry.

It has been known for quite some time that the absorption spectrum of a weak probe beam interacting with a collection of dressed atoms has a spectrally narrow gain feature for the case when the dressing laser frequency is detuned from the bare atomic resonance frequency (Boyd [1992]). Consider a two-level atom with a ground (excited) level denoted by $|g\rangle$ ($|e\rangle$) driven by an intense ‘dressing’ laser field $E_d(t) = [E_d \exp(-i\omega_d t) + \text{c.c.}]$. The atomic states have opposite parity with transition matrix element $\mu_{eg}$. The population decay rate (coherence dephasing rate) is denoted by $\gamma_{\parallel}^{eg}$ ($\gamma_{\perp}^{eg}$) and it is assumed for simplicity that the atom experiences only natural broadening so that $\gamma_{\perp}^{eg} = \frac{1}{2} \gamma_{\parallel}^{eg}$. The interaction strength of the driven-atom system is characterized by the generalized Rabi frequency $\Omega'_d = \sqrt{\Omega_d^2 + \Delta_d^2}$, where $\Omega_d = 2\mu_{eg} \cdot E_d/\hbar$ is the resonant Rabi frequency and $\Delta_d = \omega_d - \omega_{eg}$ is the detuning of the driving field from the atomic resonance frequency $\omega_{eg}$. The Rabi frequency can be of the order of $6 \times 10^9$ s$^{-1}$ for a strong optical transition and a 1 W laser beam collimated to a diameter of $\sim 1$ mm.

Figure 10 shows an example of a weak-probe gain spectrum calculated with the density-matrix formalism (Boyd [1992]). The gain feature occurs at a probe-beam frequency $\omega = \omega_d - \Omega'_d$ for $\Delta_d < 0$, its width is equal to that of the undriven transition $\gamma_{\perp}^{eg}$, and the maximum gain is equal to $\sim 0.05 \alpha_{eg}$, where $\alpha_{eg}$ is the line-center absorption coefficient of the undriven system. Agarwal [1990] showed that a laser based on this one-photon gain feature is identical in every respect to a normal laser, even including the quantum-statistical properties of the generated light, although later studies show that Bloch–Siegert-like shifts can significantly
modify the squeezing spectrum of the laser (Zakrzewski, Lewenstein and Mossberg [1991]). Several research groups have successfully constructed one-photon lasers using dressed atoms, as summarized in the more recent work by Lezama, Zhu, Kanskar and Mossberg [1990]. Dressed atoms display phase-insensitive optical amplification (i.e., not involving a phase-matching condition) when the angle between the propagation direction of the dressing and the propagation direction of the probe field or cavity axis is large and as close as possible to 90° (orthogonal geometry). For the case where the pump and probe beams are nearly colinear, it is known that dressed atoms can give rise to large probe-beam amplification due to phase-matched four-wave mixing, as discussed by Boyd, Raymer, Narum and Harter [1981].

The absorption spectrum of a dressed atom becomes complex when the probe-beam intensity increases as shown in fig. 10b, calculated using the procedures described by Agarwal and Nayak [1986]. Several new, intensity-dependent gain features appear that occur at \( \omega = \omega_0 - \frac{\Omega'_d}{m} (m = 1, 2, 3, \ldots) \). The existence of these features has been known for some time; they were originally referred to as “subharmonic resonances” (see, for example, the more recent work of Papademetriou, Chakmakjian and Stroud [1992]). The interpretation that these features correspond to multi-photon stimulated emission resonances was first pointed out by Lewenstein, Zhu and Mossberg [1990]. In particular, they showed that the feature occurring at \( \omega = \omega_0 - \frac{1}{2} \Omega'_d \) arises from two-photon stimulated emission and could be used to realize a two-photon laser. As in the one-photon gain feature, it is possible to enhance multi-wave mixing processes near these multi-photon gain features for nearly colinear laser beam geometries. Thus, an orthogonal geometry is needed to suppress wave-mixing processes.

A basic understanding of the origin of the gain features can be obtained from a perturbation-theory analysis of the interaction of the probe beam and the driven atom. It is found that the gain features arise from hyper-Raman scattering processes, as shown in fig. 11. In both processes, probe-beam photons (solid arrows) are created at the expense of pump-beam photons (dashed arrows) when the fields induce a transition from the ground level \(|g\rangle\) to the excited level \(|e\rangle\). Hence, the ‘upper’ (‘lower’) laser level is actually the ground (excited) level of the atom for the driven-atom gain medium. The ‘inversion’ between the upper and lower laser levels is maintained by spontaneous emission from \(|e\rangle\) to \(|g\rangle\). Note that there is never an inversion between the excited and ground levels of the atom. In the one-photon amplification process (fig. 11a), one probe-beam photon is created and two dressing-field photons are annihilated, while in the two-photon amplification process (fig. 11b), two probe-beam photons are created and three dressing-field photons are annihilated. This analysis does not accurately predict
the location of the gain resonances shown in fig. 10b because the perturbative analysis does not converge for such a strong atom–field interaction.

A quantitative (and intuitive) understanding of the origin of the gain features can be obtained using the dressed-state interpretation of the driving-field–atom interaction. The analysis properly accounts for the interaction of the intense pump field with the atom and treats the probe field and spontaneous emission as perturbations. As described by Cohen-Tannoudji, Dupont-Roc and Grynberg [1992] and Compagno, Passante and Persico [1995], the dressed states are the energy eigenstates of the Hamiltonian describing the two-level atom, the single quantized mode of the dressing field, and their interaction. They are linear superpositions of the atom–field product states and are denoted by $|\pm, n\rangle$, where $n$ is the photon occupation number (volume) of the dressing mode. The dressed states consist of an infinite ladder of levels separated by the energy of a dressing photon $\hbar \omega_d$, and each rung of the ladder is a doublet. For a driving field produced by an intense continuous-wave laser beam, described as a coherent state, only a narrow distribution of dressed levels high up the ladder are occupied. They are ‘locally periodic’ since their relative populations and the splitting of the doublets are independent of $n$. In this case, the doublet splitting is given by the generalized Rabi frequency $\Omega_d'$.

The interaction of the dressed states with the other field modes is treated using perturbation theory. It is found that all matrix elements are zero except for those that connect one rung of the ladder with its nearest neighbor. Considering the dressed-state selection rules, it is found that each level on a rung of the ladder decays spontaneously to both levels one step down the ladder. After several spontaneous emission events, the relative populations of a dressed-state doublet reach equilibrium in response to spontaneous decays into and out of the doublet, and the relative population distribution is independent of $n$ due to the local periodicity. When the dressing field is on resonance ($\Delta_d = 0$), both levels are
equally populated; for off-resonance driving the populations become unbalanced. For example, the states \(|-, n\rangle\) are more heavily populated than the states \(|+, n\rangle\) when \(\Delta_d < 0\), as shown in fig. 12.

A probe beam (frequency \(\omega\)) interacting with the dressed atom experiences gain or absorption when the populations of the dressed levels are unbalanced just as a bare atom will absorb or amplify a beam of light. It can be seen from fig. 12a that a probe beam interacting with the \(|-, n + 1\rangle \leftrightarrow |+, n\rangle\) transition will be amplified due to one-photon stimulated emission when \(\omega = \omega_d - \Omega_d^\prime\) for \(\Delta_d < 0\). This is precisely the spectral location of the gain feature shown in fig. 10a. Analogously, one-photon absorption will occur on the \(|-, n\rangle \leftrightarrow |+, n + 1\rangle\) transition.

An intense probe beam can induce multi-photon transitions among the dressed levels. In particular, a probe beam interacting with the \(|-, n + 1\rangle \leftrightarrow |+, n - 1\rangle\) will be amplified due to two-photon stimulated emission when \(\omega = \omega_d - \frac{1}{2} \Omega_d^\prime\) as shown in fig. 12b. Most importantly, the \(|+, n\rangle\) and \(|-, n\rangle\) intermediate levels are nearly degenerate with the virtual level of the two-photon transition, thereby enhancing significantly the two-photon rate coefficient. In addition, the two-photon gain is spectrally distinct from the regions of large one-photon gain; hence, a high-finesse optical resonator can selectively enhance the two-photon gain. It is precisely for these reasons that dressed atoms make a near ideal two-photon gain medium. Note that the driven-atom system can support \(m\)-photon gain due to \(m\)-photon stimulated emission when it undergoes a transition from one rung on the ladder to \(m\) rungs below.
Extending the proposal of Lewenstein, Zhu and Mossberg [1990], a thorough theoretical investigation of the dressed-state two-photon laser was undertaken as described in a series of theoretical papers by Zakrzewski, Lewenstein and Mossberg [1991]. Their effective Hamiltonian approach to derive semiclassical two-photon-laser equations includes the effect of the ac Stark shifts arising from the two-photon cavity field, but assumes that the cavity linewidth is narrow so that the two-photon process can be distinguished from the competing one-photon process. Interestingly, they find that the one- and two-photon processes are strongly anti-correlated so that once the two-photon laser turns on, it will suppress to some degree the coherence needed for one-photon lasing. In addition, they find that the ac Stark shift narrows considerably the domain of parameters over which stable laser operation exists, and causes significant intensity-dependent ‘pulling’ of the laser frequency.

Based on an analysis of the steady-state solutions of the laser equations, they determined the threshold for sustained continuous-wave two-photon lasing. The threshold depends on the detuning of the dressing field from the atomic resonance and takes on its minimum value when $\Delta_d = -0.4\Omega_d$. At this optimum value, the threshold number of dressed atoms in the cavity is

$$N_{th} \simeq 25.4 \frac{\Omega_d \kappa}{g^2}, \quad (6.1)$$

and the number of photons in the cavity at the laser threshold is

$$q_{th} \simeq 2.36 \frac{\Omega_d \gamma_{eg}}{g^2}, \quad (6.2)$$

where $\kappa$ is the cavity decay rate, $g = \sqrt{6\pi c^3 \gamma_{eg}^2 / \omega_{eg}^2 V_c}$ is the atom–cavity coupling constant corresponding to the Rabi frequency for a single photon in the cavity, $V_c = \pi w_0^2 L_c$ is the effective mode volume, $w_o$ is the 1/e radius of the field in the cavity, and $L_c$ is the cavity length. To estimate the threshold requirements, consider creating dressed atoms using the barium $^1S_0 \leftrightarrow ^1P_0$ transition ($\omega_{eg} = 3.4 \times 10^{15}$ rad s$^{-1}$, $\gamma_{eg} = 1.19 \times 10^8$ rad s$^{-1}$) driven with an intense laser beam ($\Omega_d = 2.6 \times 10^9$ rad s$^{-1}$) and the use of a high-finesse optical cavity with $w_o = 66 \mu m$, $L_c = 5 cm$, and $\kappa = 5.3 \times 10^6$ rad s$^{-1}$. For this experimental arrangement, $N_{th} \simeq 4.5 \times 10^4$ atoms.

\[4\] Note that Lewenstein, Zhu and Mossberg [1990] define $g$ as the Rabi frequency for a single photon in the cavity. Many other researchers define it to be one-half the single-photon Rabi frequency.
and $g_{\text{th}} \approx 9.3 \times 10^4$ photons, which was attainable with available technology. Note that this theory of dressed-state lasers assumes that the two-level atom only experiences spontaneous-emission broadening and that the resonator is in a ring configuration.

A stability analysis of the steady-state solutions revealed that the laser should be stable, at least in the vicinity of the threshold. Further above threshold, multiple effects destabilize the dressed-state two-photon laser operation. Zakrzewski, Lewenstein and Mossberg [1991] predicted that the laser will display instabilities far above threshold due to the presence of ac Stark shifts induced by the cavity field. In addition, off-resonant one-photon amplification and lasing is expected to disrupt two-photon laser operation at an atom number approximately equal to 15 times the threshold value, which still gives a considerable range for investigating its behavior. This contrasts the behavior of two-photon lasers based on inverted three-level atoms; those are unstable just above threshold and become stable far above threshold, as will be described in §8.

A later study by Zakrzewski and Lewenstein [1992] investigated the operating characteristics of the two-photon laser in the so-called ‘bad-cavity’ limit, where the cavity decay rate exceeds all other decay rates of the system. Under these conditions, the effective-Hamiltonian approach used by the researchers is no longer valid, which required formulating the problem in terms of generalized effective theories that are based on a hierarchy of macroscopic variables. They find that competition between two- and one-photon lasing can give rise to self-pulsing and even chaos in the intensity of the beam generated by the laser. For the case of dressing the atom with a short pulse so that the energies of the dressed states are now a function of time, Zakrzewski, Segal and Lewenstein [1992] found that the light emitted by the cavity has different frequencies arising from the various dressed energy levels, which interfere and give rise to a very complex field generated by the laser.

Connecting the results for the dressed-state laser to the earlier discussion in §3, it is found that the two-photon rate coefficient is given by

$$B^{(2)} \approx 0.03 \frac{|\mathbf{\mu}_{eg} \cdot \mathbf{e}|^4}{V^2 \hbar^2 e_0^2 (\Omega_d/2)^2 \gamma_{\text{un}}}$$

(6.3)

at the optimum dressing field detuning. It is seen that the dressed-state and three-level-atom rate coefficients are similar with $\Delta_{aa} \to \frac{1}{2} \Omega_d'$ by comparing eqs. (6.3) and (3.21). Thus, the rate coefficient for a dressed atom can be considerably larger than that for a typical three-level atom with fixed energy levels because $\Omega_d'$ can be small. Note that there is an optimum value of $\Omega_d'$ for a particular
experimental situation since the one- and two-photon gain features will start to overlap as $Q'_d$ becomes small.

6.1. Measurement of continuous-wave two-photon amplification

To test the proposal of Lewenstein, Zhu and Mossberg [1990], Zhu, Wu, Morin and Mossberg [1990] used dressed $^{138}$Ba as the two-photon amplification medium. Under appropriate conditions, $^{138}$Ba is a near ideal two-level atom because the $^1S_0 \leftrightarrow ^1P_0$ transition has zero nuclear spin and hence is not complicated by hyperfine interactions, and it possesses a large electric-dipole moment with a transition wavelength accessible by high-power tunable dye lasers. One minor complication is the presence of other barium isotopes (22% abundance in natural barium), but their resonances are all to the high-frequency side of the $^{138}$Ba $^1S_0 \leftrightarrow ^1P_0$ transition frequency.

In the experiments, an atomic beam source of barium was used to reduce the Doppler broadening of the transition. The Doppler effect broadens and reduces the two-photon gain feature, and requires the use of a higher-intensity dressing field because the Rabi frequency needs to exceed the Doppler width to see appreciable laser beam amplification. On the other hand, the use of an atomic beam limits the available atomic number density and the length of the gain medium. To compensate for loss in gain, they surrounded the dressed atoms with a 1-cm-long optical enhancement cavity (a confocal Fabry–Pérot optical resonator) to increase the effective path length of the medium by approximately the cavity finesse (200 in their experiment). The dressed-state gain was determined by measuring the transmission of a probe beam that passed through the cavity. The axis of the atomic beam and enhancement cavity, and the propagation direction of the dressing laser beam, were mutually orthogonal to suppress wave-mixing-type gain processes, as shown in the top panel of fig. 12.

The atomic beam was rapidly turned on and off using a mechanical chopper so that lock-in signal-averaging techniques could be employed. During the interval when the atomic beam was blocked, a standard modulation technique was used to actively lock the frequency of the probe laser to the empty-cavity resonance to compensate for laser frequency drift. The gain measurements were complicated by their choice of a locking scheme because the dispersion of the dressed atoms shifts the frequency of cavity. Therefore, the probe-beam transmission was always below what it would have been in the absence of dispersive effects, mimicking absorption. Locking the probe-beam frequency to the cavity resonance in the presence of the dressed atoms was not easily
implemented due to the intensity-dependent gain and dispersive effects arising from the multi-photon stimulated emission and saturation processes, causing the locking loop to be unstable.

Figure 13a shows the probe-beam gain spectrum for the case of a weak probe beam, $\Omega_d/2\pi \simeq 340\,\text{MHz}$, and $\Delta_d \simeq -100\,\text{MHz}$ so that $\Omega_d'/2\pi \simeq 354\,\text{MHz}$. It is seen that there is a large gain feature when the frequency difference between the probe and dressing fields is approximately equal to $-\Omega_d'$, indicating the one-photon stimulated emission process between the states $|+, n\rangle \leftrightarrow |-, n + 1\rangle$ shown in fig. 12a. For a higher probe beam power that is sufficiently strong to induce the two-photon stimulated emission process $|+, n + 1\rangle \leftrightarrow |-, n - 1\rangle$ shown in fig. 12b, a new feature appears in the gain spectrum at approximately $-\frac{1}{2} \Omega_d'$, as seen in fig. 13b. Careful inspection of the data reveals that the feature represents reduced absorption rather than gain, which they attributed to the pseudo-absorption effect arising from the dispersion of the dressed atoms, as discussed above. The dashed line in the figure shows the theoretical predictions for the experiment taking into account the dispersion, dressing-field inhomogeneities, the standing-wave probe-field pattern within the cavity, and the barium isotopes. It is seen that the agreement between the measurements and predictions is very
good. From their analysis, they infer that they would have observed an increase of the probe-beam transmission of approximately 8% in the absence of dispersive effects (pseudo-absorption).

These observations represent the first measurement of continuous-wave two-photon stimulated emission in the optical part of the spectrum and support the conjecture of Lewenstein, Zhu and Mossberg [1990] that dressed atoms can be used to achieve two-photon lasing. More recently, Lange, Agarwal and Walther [1996] observed enhanced two-photon decay from dressed Rydberg atoms in the microwave part of the spectrum, suggesting that the general dressed-atom approach can also be of use for developing a new class of two-photon masers. I also note that the so-called 'Doppleron' resonances observed in the velocity distribution of atoms moving in an intense optical standing wave (Bigelow and Prentiss [1990], Tollett, Chen, Story, Ritchie, Bradley and Hulet [1990]) can be interpreted in terms of multi-photon stimulated emission, suggesting that a two-photon laser could be realized using cold-atom and CQED methods. In addition, Manson, Changjiang Wei and Martin [1996] have observed multi-photon resonances in the driven nuclear magnetic resonance response within the ground-state hyperfine levels of the nitrogen-vacancy center in diamond, which may be useful in realizing dressed-state masers in a solid material.

6.2. Realization of the continuous-wave two-photon laser

The first observation of continuous-wave two-photon lasing was reported by Gauthier, Wu, Morin and Mossberg [1992] using an experimental arrangement very similar to that used by Zhu, Wu, Morin and Mossberg [1990] to measure two-photon amplification. To achieve lasing, the cavity losses were reduced and the number of dressed atoms in the cavity was increased. The cavity losses were reduced by replacing the enhancement cavity by a confocal cavity with higher-reflectivity mirrors \( w_0 = 66 \mu \text{m}, L_c = 5 \text{ cm}, \text{ and } \kappa = 5.3 \times 10^6 \text{ rad s}^{-1} \). The optical finesse for this resonator was 1800, corresponding to a \( Q \) of \( 6.4 \times 10^8 \). In addition, the barium atomic beam was replaced by one that could produce higher atomic number densities and had a larger diameter so that \( N \approx 2 \times 10^5 \) atoms were present in the cavity mode, although at the cost of increasing the residual Doppler width to \(~40\text{ MHz}\) (to be compared to the 19 MHz natural width of the transition and the 1.7 MHz cavity linewidth). The possibility of obtaining two-photon lasing is indicated by the fact that \( N \gtrsim N_{\text{th}} \) given above.

Gauthier, Wu, Morin and Mossberg [1992] conducted two preliminary experiments to characterize the dressed-atom laser system. In one experiment,
the optical power emitted from one end of the cavity was measured as the cavity resonance frequency $\omega_c$ was tuned through the dressed-atom resonances. As seen in fig. 14a, it was found that normal one-photon dressed-atom lasing occurred when $\omega_c \approx \omega_d - \Omega_d'$ and that no lasing occurred when $\omega_c \approx \omega_d - \frac{1}{2} \Omega_d'$ (the predicted frequency of maximum two-photon gain). This measurement confirmed that the one-photon gain feature was spectrally removed from the region where high two-photon gain was expected and hence it would not obscure two-photon lasing.

The other experiment measured the gain of the system directly by observing the transmission of an intense laser beam as it passed through the cavity in the presence of the dressed atoms, as shown in fig. 14b. The pseudo-absorption present in the experiment of Zhu, Wu, Morin and Mossberg [1990] was not a factor in this experiment because the probe-laser frequency was scanned through the dressed-atom–cavity resonance and the maximum transmission was recorded. A pronounced, intensity-dependent gain feature was observed when $\omega_c \approx \omega_d - \frac{1}{2} \Omega_d'$ that coincides with the predicted location of the two-photon gain feature. Note that the tail of the one-photon gain feature overlaps somewhat with the two-photon gain feature; they estimated that $\sim 35\%$ of the laser beam amplification was due to one-photon stimulated emission. It appears that this non-ideal behavior did not seriously disrupt the lasing behavior, as described below, which is consistent with the prediction by Zakrzewski, Lewenstein and Mossberg [1991] that two-photon lasing should suppress the coherence needed for one-photon lasing.

Based on the discussion in the previous sections, two-photon lasing should not occur in this system until both a sufficient number of inverted atoms and a sufficient number of photons are in the cavity. To observe two-photon lasing, they adjusted the cavity frequency to the peak of the observed two-photon gain feature.
shown in fig. 14b and injected trigger pulses (duration $\sim 1.4 \mu s$) into the laser resonator. The time-resolved cavity output power just prior to and following the injected pulse was repeatedly recorded for various trigger-pulse powers. As shown in fig. 15a, the number of injected trigger photons ($q_{\text{inj}} \approx 3.4 \times 10^3$) was insufficient to initiate two-photon lasing, and the cavity output power decayed to zero after the injected pulse was turned off. For higher injected powers, the cavity output power remained high after the trigger pulse was turned off as shown in fig. 15b. They estimated that there were $\sim 1.4 \times 10^5$ intercavity photons once the laser was turned on and the trigger pulse had left the cavity, which is in reasonable agreement with the predicted value of $q_{\text{th}} \approx 9.3 \times 10^4$ photons. Spiking behavior was observed while the injected field was present, a behavior that is apparently not accounted for in the theory of two-photon lasers. Figure 15c shows the observed two-photon laser behavior over a longer time scale for the case when a stronger trigger field was injected, which appears to suppress the initial spiking in the photon number. Finally, they observed complex temporal evolution of the power generated by the laser when the dressing field was tuned closer to the atomic resonance, as shown in fig. 15d. They proposed that this behavior might be due to the beating of distinct transverse cavity modes, an instability driven by competition between the two-photon and off-resonant one-photon gain processes, or due to instabilities driven by ac Stark shifts induced by the lasing field.
The trigger-induced transition to a state of nonzero cavity output power is entirely consistent with the expected threshold behavior of a two-photon laser as discussed in § 3 (see fig. 4, above), demonstrating conclusively that two-photon lasing was indeed achieved. To date, no additional experiments have been performed to fully characterize the stability properties or the quantum-statistical properties of the dressed-state two-photon laser.

§ 7. The Raman two-photon laser

Continuous-wave two-photon amplification and lasing are also possible in driven multi-level atoms, which opens up the possibility of identifying the generic properties of two-photon lasers and operation on different states of polarization. For example, Concannon, Brown, Gardner and Gauthier [1997] considered laser beam amplification in a laser-driven thermal vapor of potassium atoms when the dressing-laser frequency was tuned in the vicinity of the $4S_{1/2} \leftrightarrow 4P_{1/2}$ transition. They demonstrated that two-photon gain arises in the system by a process they called two-photon Raman scattering, shown schematically in fig. 16a. In this process, intense dressing (dashed arrows) and probe (solid arrows) fields stimulate the atom to make a transition from the initial state $|g\rangle$ to the final state $|g'\rangle$ by absorbing two photons from the dressing field (frequency $\omega_d$) and adding two new photons to the probe field (frequency $\omega$) via virtual intermediate states. Energy conservation requires that $\omega = \omega_d - \frac{1}{2} \Delta_{gg'}$, where $\hbar \Delta_{gg'}$ is the energy difference between $|g\rangle$ and $|g'\rangle$. In the experiments, the states $|g\rangle$ and $|g'\rangle$ corresponded to the $4S_{1/2}$ ($F = 1$) and $4S_{1/2}$ ($F = 2$) hyperfine states, respectively, where $\Delta_{gg'}/2\pi = 462$ MHz for $^{39}$K. To obtain continuous-wave two-photon amplification based on this stimulated emission process, a steady-state

Fig. 16. Raman two-photon scattering. (a) Scattering diagram showing the Raman two-photon process. (b) Gain spectrum for an intense probe beam propagating through a vapor of laser-driven potassium atoms. From Concannon, Brown, Gardner and Gauthier [1997].
imbalance must exist between the states $|g\rangle$ and $|g'\rangle$ so that $N_g > N_{g'}$, which is accomplished by optical pumping of the atom by the intense dressing field. Note that this process is similar to the multi-photon scattering diagram shown in fig. 11 for the perturbative explanation of two-photon amplification in driven two-level atoms. It is possible to understand the origin of the two-photon amplification process using the dressed-state basis for the driven three-level atom, but such an analysis is not all that much simpler than using the bare-atom basis.


In the experiment, potassium vapor was contained in a 7-cm-long evacuated pyrex cell with uncoated, near-normal-incidence optical windows heated to a temperature of 150°C which produced a number density of approximately $10^{13}$ atoms/cm$^3$. Because they used natural-abundance potassium, gain and absorption features were also observed due to scattering from $^{41}$K where $\Delta_{gg'} = 254$ MHz. The dressing laser beam was linearly polarized, collimated to a diameter of 150 $\mu$m (intensity FWHM) as it passed through the cell, had a power of 850 mW at the entrance to the cell, and was tuned approximately 2.4 GHz to the low-frequency side of the D1 transition \([4S_{1/2} (F = 2) \rightarrow 4P_{1/2} (F = 1)]\) occurring near $\lambda = 769.9 \text{ nm}$. The probe beam was collimated to a diameter of 65 $\mu$m and had a polarization orthogonal to that of the pump field, which resulted in the maximum two-photon gain. The probe beam nearly copropagated with the pump beam (crossing angle 12 mrad) so that the two-photon Raman scattering process was nearly Doppler-free while minimizing the effects of parametric wave mixing that could potentially compete with the two-photon gain process.

Figure 16b shows the probe-beam gain spectrum for a probe beam intensity that was high enough to induce the multi-photon Raman processes. They observed a narrow, intensity-dependent gain feature at $\omega \simeq \omega_d - \frac{1}{2} \Delta_{gg'}$ \([(\omega - \omega_d)/2\pi \simeq -231 \text{ MHz}]]$, which they attributed to the two-photon Raman scattering process shown in fig. 16a. The continuous-wave two-photon gain was as large as 30%! Also apparent in fig. 16b is a feature at $\omega \simeq \omega_d - \Delta_{gg'}/3$ \([(\omega - \omega_d)/2\pi \simeq -154 \text{ MHz}]]$, corresponding to 5% three-photon gain.

Subsequent research by Brown [1999] using laser-driven potassium atoms in
a vapor cell revealed that the probe beam experienced significant beam steering due to a nonlinear waveguide structure created by the interaction of the atoms and the dressing laser field. It was suspected that the probe beam was guided precisely along the propagation direction of the dressing laser field even when the crossing angle outside the vapor cell was set fairly large, opening the possibility that parametric multi-wave mixing effects (Trebino and Rahn [1987]) might be responsible for some of the observed laser-beam amplification. Therefore, the vapor-cell experiments were abandoned and an atomic beam apparatus was constructed (Brown [1999]) so that a mutually orthogonal experimental geometry could be used to suppress wave-mixing effects, similar to the geometry employed by Zhu, Wu, Morin and Mossberg [1990] and Gauthier, Wu, Morin and Mossberg [1992] in the laser-driven barium experiments.

Pfister, Brown, Stenner and Gauthier [1999] demonstrated two-photon amplification in laser-driven potassium atoms using the mutually orthogonal geometry shown in fig. 17a. The interactions are somewhat more complex because the magnetic sublevels of the potassium hyperfine states have to be taken into account for this geometry, as shown schematically in fig. 17b to lowest order in perturbation theory. Laser-beam amplification occurred when two circularly polarized dressing-field photons were annihilated and two linearly polarized probe photons were created as the atom underwent a transition from the |g22⟩ to the |g20⟩ Zeeman sublevels. The atomic states are denoted by |αF,αM⟩, where α = g for the potassium 42S1/2 and α = e for the 42P1/2 levels, and F and M are the quantum numbers for the total angular momentum and its projection along

\[ M_\alpha = -2 \quad -1 \quad 0 \quad 1 \quad 2 \]
the \( z \) quantization axis (taken as the propagation direction of the dressing field). The necessary inversion between the states \( |g22\rangle \) and \( |g20\rangle \) was maintained using auxiliary optical pumping beams that continuously transferred population from all hyperfine states into \( |g22\rangle \).

In their experiment, the atoms are produced by an atomic beam with a half-angle divergence of 30 mrad, giving rise to a residual Doppler width of 30 MHz, a diameter of 2.5 mm and an atomic number density of \( 2 \times 10^{11} \) atoms \( \text{cm}^{-3} \) in the interaction region. The atoms were dressed with a circularly polarized laser beam propagating along the quantization axis with an intensity of 25 W cm\(^{-2} \) and its frequency tuned to the blue side of the \( |g11\rangle \rightarrow |e2M\rangle \) transition by 522 MHz. A weak uniform magnetic field \( (B_z \ll 10^{-4} \tau) \) was applied in the interaction region to overwhelm stray magnetic fields. A \( z \)-polarized probe beam collimated to a radius \( w_o = 90 \mu m \) interacted with the dressed atoms and experienced intensity-dependent gain for sufficiently high probe beam intensities.

They measured the two-photon gain directly (without the use of an enhancement cavity), a difficult experiment because the two-photon signal was as small as \( 10^{-5} \), while, on the other hand, the probe power was high enough to saturate typical silicon detectors or the associated electronic preamplifiers. They solved these problems by using high-power photodiodes in a difference-detection setup. The probe beam was split into two beams of equal power, one of which (the signal beam) was sent through the vacuum chamber and the driven atoms, while the other (reference) beam was sent directly to its detector. The power of each beam was converted to a current by a photodiode that had a linear response for powers up to several tens of milliwatts (Hamamatsu S3994), the two photocurrents were subtracted at their mutual junction, and the resulting difference current was converted to a voltage.

Figure 18a shows the probe-beam gain spectrum, where several gain and absorption features are evident. They identified the origin of all resonances by measuring their spectral location and their dependence the probe beam power. The small feature located at a probe-pump detuning of \( \omega \simeq \omega_d - \frac{1}{2} \Delta g_r \) = 232 MHz was attributed to the two-photon Raman process shown in fig. 18b. The continuous-wave two-photon amplification was only about \( \sim 1.5 \times 10^{-4} \) ! Note that the gain features are very narrow and comparable to the residual Doppler width of the resonance, so that the various resonances were highly spectrally isolated, in contrast to the behavior of the dressed-state two-photon laser (see fig. 14).

To further demonstrate that the feature was due to a genuine two-photon process, they measured its dependence on the probe-beam intensity, as shown in fig. 18b. A clear linear increase of the gain was observed for powers up to 3 mW (corresponding to an intensity of 24 W cm\(^{-2} \)), for which a saturation plateau
occurred at a maximum peak height of $4 \times 10^{-4}$. The observed linear dependence below saturation is a central result of their experiment: it demonstrates directly that photons are emitted two at a time. They noted that the height of the feature is not the net two-photon gain because it overlaps with the wing of a large absorption feature due to transitions from $|g22\rangle$ to $|e22\rangle$. They estimated that the maximum gain was about 50% of the peak height. Using the simple model for two-photon amplification (see eq. 3.18), they estimated that $G^{(2)} = 9 \times 10^{-5} \text{ cm W}^{-1}$ and $I_{\text{sat}} = 36 \text{ W cm}^{-2}$. The solid line is the fit of this simple model to their observations.

In a subsequent study, Fernández-Soler, Font, Vilaseca, Gauthier, Kul’minskii and Pfister [2002] investigated theoretically laser-beam amplification in driven potassium atoms using a semi-classical description of the interaction that accounted for most of the hyperfine level structure (transitions involving states with $M < 0$ were neglected). They determined a complex ‘gain’ coefficient

$$
\widetilde{G} = \frac{U}{\beta} \left[ \rho_{\epsilon g22} + \sum_{i=0,1} \sum_{j,k=1,2} \rho_{\epsilon \bar{g} \bar{g} \bar{g} \bar{g} \bar{g}} \right], \quad (7.1)
$$

where $U$ is the unsaturated gain parameter and $\beta$ is proportional to the probe-beam Rabi frequency. Equation (7.1) was obtained by summation of all the one-photon atomic coherences $\rho_{ul}$ induced on the transitions $u \equiv |eFM\rangle \rightarrow l \equiv |gF'M\rangle$. The imaginary part of eq. (7.1), or the gain factor $G$, represents the relative increase in the probe-field amplitude per unit of time, and its real part is proportional to the induced change in refractive index experienced by the probe beam.
The solid curve in fig. 19 shows the gain factor as a function of the probe-pump detuning $\Delta_p$ for the conditions reported in the experiments described above with no adjustable parameters except for an overall vertical scale factor. The experimental trace is shown as crosses in the figure (note that the vertical scale is offset for clarity). It is seen that there is very good agreement between the predictions of the model and the experiments, indicating that the model will be useful for making new predictions for two-photon amplification and lasing in laser-driven potassium atoms. Interestingly, they found that interferences between different quantum pathways were constructive for the two-photon process and that the interferences helped to reduce the ac Stark shifts of the two-photon transition.

Continuous-wave two-photon lasing in laser-driven potassium atoms was first observed by Pfister, Brown, Stenner and Gauthier [2001], who combined the experimental apparatus from the measurements of amplification with a low-loss optical resonator. They used a linear (standing-wave) cavity consisting of two high-reflectivity ultra-low-loss mirrors with radius of curvature 5 cm set close together at a distance of $L_c = 1.5$ cm so that it is operated in a transverse-mode-degenerate sub-confocal configuration ($w_o = 66 \mu$m). Pinholes were placed in front of each mirror to suppress higher-order transverse modes that were not degenerate with the lower-order ones due to spherical aberrations and, with these pinholes, they measured an optical finesse of $\approx 1.5 \times 10^4$, resulting in $\kappa = 2.1 \times 10^6$ s$^{-1}$. Within the cavity mode volume, they estimated that there were $7 \times 10^6$ atoms. The dressing laser beam had an intensity of $\sim 25$ W cm$^{-2}$ and
its frequency was tuned to the blue side of the $|g1M\rangle \leftrightarrow |e2M\rangle$ transition by 512 MHz.

They determined the location of the two-photon resonance by injecting a probe beam into the cavity and measuring its transmission. To initiate lasing, they set the cavity frequency to the peak of the observed two-photon transition frequency, momentarily blocked the dressing beam to quell any pre-existing two-photon lasing, and measured the power of the light emitted from the two-photon laser resonator using an avalanche photodiode. In the absence of the probe-laser beam, they observed essentially no light emitted from the resonator, indicating that quantum fluctuations did not provide a sufficient number of photons in the cavity to satisfy the lasing criteria. To trigger lasing, they injected 1.2-$\mu$s-long pulses of light into the resonator; fig. 20a shows the case when a below-threshold pulse was injected into the resonator. The dashed vertical lines indicate the duration of the injected pulse; they estimated that $q_{\text{inj}} = 1.8 \times 10^4$ photons were injected into the cavity. Note that there was significant spiking in the power transmitted through the cavity, even though the input pulse was smooth, similar to that shown in fig. 20 for the dressed-state two-photon laser. For slightly higher trigger-pulse powers (fig. 20b, $q_{\text{inj}} = 3.4 \times 10^4$), the cavity photon number grows substantially, displaying considerable spiking, and remains high for a few microseconds, but
was still insufficient to initiate lasing, indicating that they were very close to satisfying both two-photon laser threshold criteria.

The two-photon laser turned on for $q_{\text{inj}} = 5.2 \times 10^4$ (fig. 20c), where the power emitted by the cavity rose to approximately $0.2 \, \mu \text{W}$ and remained at this value, corresponding to an average cavity photon number of $3.5 \times 10^5$ and an intracavity intensity of $41.3 \, \text{W cm}^{-2}$ at the cavity waist. This intercavity intensity is slightly higher than the two-photon saturation intensity measured for this process in the amplification experiments described above, indicating that the laser runs fully saturated near the threshold condition as discussed in §3. A measurement by Brown [1999] of the transverse profile of the beam emitted by the laser showed that it appeared to be a lowest-order Gaussian mode. Figure 20d shows the power emitted from the resonator on a longer time scale for a larger injected number of photons and during a different experimental run but under essentially identical conditions. The experimental data of fig. 20 give convincing evidence that the device is indeed a two-photon laser. These observations are consistent with theoretical analysis of the Raman two-photon laser by Fernández-Soler, Font, Vilaseca, Gauthier, Kulminskii and Pfister [2002], who find that the laser emission line shape is frequency-pushed due to the ac Stark effect and has the form of a closed curve composed of a stable and an unstable branch.

Since the optical resonator was highly isotropic and the magnetic sublevels of the potassium atom support multi-photon transitions for more than just a linearly polarized cavity field, Pfister, Brown, Stenner and Gauthier [2001] investigated the polarization characteristics of the generated light. They placed a linear polarizer oriented in the $\hat{z}$-direction in the beam and found that the two-photon laser displayed polarization instabilities even though the total emitted power was nearly constant on the time scale of the instability. It is seen in fig. 21a that the state of polarization underwent very regular oscillations of period $0.11 \, \mu$s, with a 50% depth of modulation. Note that the oscillations commenced even while the injected trigger pulse was present. Similar dynamical behavior was observed for all polarizer orientations, suggesting that the state of polarization is elliptical with an ellipticity of 0.5 and a precessing major axis. They also found that the laser dynamics were quite sensitive to the applied magnetic field: an increase

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5 The photon numbers and intercavity intensities given here are different from the values given in Pfister, Brown, Stenner and Gauthier [2001]. Errors were found in the determination of these quantities from the measured power of the beam emitted from the cavity. Here, $q = \frac{P_{\text{out}}}{\hbar \omega_0 c}$ and $I = \frac{8P_{\text{out}}}{\sqrt{2} \pi w_0^2}$, which takes into account the fact that the cavity is symmetric and an equal power is emitted from the opposite side of the cavity.
Fig. 21. Polarization instabilities in the Raman two-photon laser. Temporal evolution of the light emitted from the cavity for (a) small and (b) larger applied static magnetic field strengths. From Pfister, Brown, Stenner and Gauthier [2001].

by as little as 0.5 Gauss was sufficient to generate a much more complicated pattern. Figure 21b shows the complex, possibly chaotic behavior they observed for a magnetic field strength of 2.0 G.

They suggested two possible mechanisms that might be responsible for the observed instability. One arises from the multiple frequency-degenerate final lasing states and interference between multiple quantum pathways for different states of polarization for the emitted cavity photons, as shown schematically in fig. 22. As the laser turns on and begins to saturate the pathway shown in fig. 22a (since they injected a z-polarized trigger field), an x-polarized field can begin to grow on the unsaturated pathways shown in fig. 22b and fig. 22c, which is enhanced by the existing z-polarized cavity field due to singly stimulated emission. Another possible mechanism is the presence of standing waves; it is well known that counterpropagating laser beams in conjunction with a tensor nonlinear optical interaction can give rise to polarization instabilities with a reduced instability threshold (Gauthier, Malcuit and Boyd [1988]). They suggested that application of a strong magnetic field to lift the degeneracy of the different final states might suppress the instability. Also mentioned is the possibility that the laser might produce polarization-entangled twin beams of light for precision measurement and quantum communication applications.

These experiments demonstrate that multi-level structure in a two-photon gain medium gives rise to novel behavior in the state of polarization of the generated beam. Further experiments are needed to fully explore the quantum-statistical and nonlinear dynamics behavior of this new type of quantum oscillator.
Fig. 22. Raman two-photon scattering in optically pumped and laser-driven potassium atoms showing the possible quantum pathways connecting initial and final lasing state for different states of polarization supported by the cavity. From Pfister, Brown, Stenner and Gauthier [2001].

§ 8. Quantum-statistical and nonlinear dynamical properties

As demonstrated in the previous sections, the two-photon laser is a highly nonlinear system that displays unusual properties in comparison to other quantum oscillators. For this reason, it has attracted considerable theoretical attention since the original proposals of Prokhorov [1965] and Sorokin and Braslau [1964]. Describing the complete body of theoretical research on two-photon lasers is beyond the scope of this chapter and deserves a review in itself. To give a flavor of the excitement for this work, I highlight the areas concerning the quantum-statistical properties of the light generated by the laser and its nonlinear dynamical properties, describing some of the early research and a few recent results.

8.1. Quantum-statistical properties and squeezing

Since the two-photon stimulated emission and absorption processes shown in
fig. 1 require two incident photons to induce the atom to make a transition to the lower state, it is expected that the quantum-statistical properties of the incident light will affect the transition rate. Also, it is expected that light passing through two-photon amplifiers or absorbers, or generated by a two-photon laser, will have interesting properties. Early work considered the case when thermal radiation or light generated by a normal one-photon laser interacts with a two-photon transition. A one-photon laser operating well above threshold is fairly well characterized by a coherent state, where the photon-counting probability distribution is Poissonian, and the dispersion of the quadrature amplitudes is equal and at the minimum value allowed by the uncertainty principle (see Ch. 18 of Mandel and Wolf [1995]). Thermal radiation, on the other hand, is characterized by Bose–Einstein statistics and hence displays photon bunching. It was predicted by Lambropoulos, Kikuchi and Osborn [1966] and later demonstrated by Shiga and Imamura [1967] that the two-photon absorption rate is highest for a thermal field because the probability for two photons to be incident simultaneously at an atom is higher for bunched light. Similarly, he predicted that a beam of light traveling through an unsaturated two-photon amplifier will experience greater gain when its statistics are thermal-like (Lambropoulos [1967]). Surprisingly, he found that fluctuations of a thermal beam of light tended to decrease whereas the fluctuations tended to increase for a coherent state, which was attributed to the presence of both spontaneous emission as well as singly stimulated emission. He thus concluded that a two-photon amplifier is not coherent in the sense that it tends to increase the fluctuations of the field amplitudes for a coherent input. These predictions were later confirmed by McNeil and Walls [1974] using a different approach.

Fully quantum-mechanical modelling of the degenerate single-mode two-photon laser was first undertaken by McNeil and Walls [1975], who determined the stationary density operator for the light field in the cavity and found that the photon-distribution functions are narrower than that for normal one-photon lasers. To simplify the analysis, they assumed that photons are coupled out of the cavity two-at-a-time so that detailed balance could be invoked when solving the equations describing the probability flow. [For the simplified model described in § 3, the photon decay term in eq. (3.2) would be \(-\kappa q^2\) rather than \(-\kappa q\) for such output coupling of photons.] Unfortunately, this assumption totally changes the threshold characteristics of the two-photon laser so that the discontinuous behavior shown in fig. 3 is replaced by a smooth turn-on of the laser, analogous to a second-order phase transition. Bulsara and Schieve [1979] obtained similar results using a semiclassical stochastic approach, and Nayak and Mohanty [1979] generalized the theory to take into account Doppler broadening, but both studies
also considered the unphysical situation when detailed balance is maintained. When McNeil and Walls [1975] relaxed the assumption of detailed balance, they predicted that the laser should produce coherent light obeying Poisson statistics using a perturbative solution to the problem, which was later confirmed using a $Q$-function approach (Görtz and Walls [1975]). In a later study, Zubairy [1980] found quite the opposite using non-perturbative techniques: the relative fluctuations of the number of photons in a two-photon laser are larger than that for a coherent state.

In view of the earlier work, it was quite surprising when Yuen [1975] predicted that an ideal degenerate single-mode two-photon laser operating far above threshold should produce a squeezed state of the electromagnetic field (see also Yuen [1976]). In particular, he found that the field was best described by a so-called ‘two-photon coherent state’, which is a minimum-uncertainty state but with the unusual properties that the variance of one quadrature of the field can be made arbitrarily small (squeezed) at the expense of increasing the fluctuations in the other quadrature. Such behavior has no analogy in classical physics and hence it is thought of as a purely quantum-mechanical effect. Squeezing is predicted to occur when the Hamiltonian has a quadratic form so that it has contributions depending on the square of the photon creation and annihilation operators. When the laser operates far above threshold, he reasoned that the operators describing the raising and lowering of the atomic excitation could be treated as c-numbers and the resulting Hamiltonian would give rise to squeezing. Rowe [1978] arrived at a similar conclusion independently. Soon thereafter, it was shown that two-photon quantum states had the potential for improving the noise characteristics of optical communication channels (Hirota [1977] and Yuen and Shapiro [1978]). Note that a squeezed field can display sub- or super-Poissonian statistics depending on the phase of the squeezing operator (see Ch. 21 of Mandel and Wolf [1995], for example), so that its existence is not necessarily contradictory with the earlier findings described above. Yuen’s predictions resulted in renewed interest in two-photon lasers for testing the foundations of quantum optics in a situation where the light–matter interaction is highly nonlinear, even though the originally envisioned applications for tunable high-power lasers were no longer as relevant.

While there was great general interest in squeezed states, there arose considerable controversy as to whether a squeezed state is generated by a two-photon laser. Lugliato and Strini [1982] and Reid and Walls [1983] developed a more accurate model of the atomic-pumping and cavity-loss mechanisms and found that there was no squeezing in the resonantly tuned laser with an injected signal due to the effects of spontaneously emitted photons. Their model was
based on earlier work by Sczaniecki [1980]. Reid, McNeil and Walls [1981] extended this model and found that the two-photon laser displays extreme photon bunching just above threshold, consistent with the findings of Bandilla and Voigt [1982], Herzog [1983] and Cheng and Haken [1988]. On the other hand, Hu and Sha [1991] predicted that squeezing should occur when the laser is tuned away from the two-photon resonance. In addition, Ashraf and Zubairy [1989] found that the spontaneous-emission contribution to the laser linewidth is twice as large as that for a normal one-photon laser, but that the gain contribution to the diffusion coefficient is independent of the mean photon number. While the degenerate laser appears not to generate quantum beams of light, Swain [1988] and Zubairy [1982] found that there exist quantum correlations between the photons in the two modes of a non-degenerate two-photon laser, which, they suggest, might be of use in precision measurement and laser gyroscopes.

All of these earlier models are based on an ‘effective Hamiltonian’, where the atom possesses only two energy levels connected by a two-photon transition. In a series of papers, Wang and Haken [1984] described how to obtain the effective Hamiltonian from a microscopic one that considers the interaction of the fields with the upper and lower energy levels as well as the intermediate states, such as the simplified energy-level diagram shown in fig. 6a for a three-level atom. Unfortunately, they dropped the terms representing the ac Stark shift (see eq. 3.17) in their analysis. Boone and Swain [1989, 1990] demonstrated that the ac Stark shifts make an important contribution to the off-diagonal density matrix elements describing the non-degenerate two-photon laser, implying that predictions for the linewidth, frequency shifts, squeezing spectrum, and stability boundaries are incorrect when using effective Hamiltonians. They found that the effective-Hamiltonian approach is valid near the laser threshold for determining the diagonal density matrix elements, but fails far above threshold because power broadening and shifts of the transitions increase the contribution of the off-resonance one-photon processes. On the other hand, the effective-Hamiltonian approach can never correctly describe the off-diagonal elements. One prediction of their work is that the laser linewidth is a factor of two larger than that expected using an effective Hamiltonian. Similar conclusions were made by Lu [1990] for degenerate operation of the laser. Other research incorporating the effects of the intermediate levels includes studies of the photon-number probability distribution (Zhu and Li [1987]), the correlation between the two modes of non-degenerated lasers when the atoms are injected with an initial atomic coherence (Lu, Zhao and Bergou [1989], Majeed and Zubairy [1991, 1995]), and the emission spectrum in the absence of atomic and cavity damping (Nasreen and Razmi [1993]). Bay, Elk and Lambropoulos [1995] has found that ac Stark shifts
away from the transition frequency, even for a good cavity. In addition, the threshold for instabilities is lowered significantly by including the ac Stark effect (Ovadia, Sargent and Hendow [1985]), which gives rise to intensity-dependent dispersion and phase-conjugate scattering of the laser field off the two-photon coherence.

In a later series of studies from the Universität Stuttgart group, they also found that the laser can display self-pulsing instabilities near the laser threshold that stabilize only far above threshold (Ning and Haken [1989a]). Near the second laser threshold where it makes a bifurcation from self-pulsing to stable lasing, they used the slaving principle and normal-form techniques to arrive at a nonlinear theory governing the laser dynamics (Ning and Haken [1989b]). Their analysis reveals that the bifurcation can be of the normal Hopf type (super-critical), displaying a period-doubling route to chaos, or it can be a co-dimension-two Hopf type (sub-critical), displaying two limit cycles. Related phenomena have been found by Yang, Hu and Xu [1994] for the case when an external field is injected into the laser. In addition, they showed that the self-pulsing behavior can be characterized through use of a geometrical phase (Ning and Haken [1992a,b]).

Recent research has more fully explored, for example, the dependence of the instability boundaries on the effects of off-resonant one-photon amplification (Roldán, de Valcárcel and Vilaseca [1993]), tristability in good-cavity lasers with an injected signal (Urchueguía, Espinosa, Roldán and de Valcárcel [1996]), standing waves occurring in Fabry–Pérot lasers (Espinosa, Vilaseca, Roldán and de Valcárcel [2000]), departures from the mean-field approximation (Abdel-Aty and Obada [2001]), and specific models of the two-photon maser (Davidovich, Raimond, Brune and Haroche [1988]), the dressed-state two-photon laser (Zakrzewski, Lewenstein and Mossberg [1991], Zakrzewski and Lewenstein [1992]), and the Raman two-photon laser (Fernández-Soler, Font, Vilaseca, Gauthier, Kul'minskii and Pfister [2002]). Especially interesting is the novel behavior displayed by wide-aperture two-photon lasers where transverse effects are important. Vilaseca, Torrent, García-Ojalvo, Brambilla and San Miguel [2001] predicted that the laser will spontaneously form two-dimensional bright localized structures in a dark background. The bright spots, called cavity solitons, arise from the two-photon emission and are embedded in a background of emission due to off-resonant one-photon gain. Figure 23 shows the predicted intensity and phase for such a laser in a regime where there is turbulent motion of solitons that appear and disappear spontaneously. They suggest that these coherent structures could be useful for all-optical parallel signal transmission and processing.
Fig. 23. Predicted two-photon cavity solitons in an active optical medium. (a) Intensity and (b) phase of the transverse profile of the beam emitted from the two-photon laser in the turbulent regime. From Vilaseca, Torrent, García-Ojalvo, Brambilla and San Miguel [2001].

§ 9. Future prospects

The two-photon laser is a unique class of quantum oscillator that has an unusual threshold behavior, displays nonlinear dynamical instabilities, and is predicted to produce beams of light with interesting quantum statistical properties. Unfortunately, achieving two-photon lasing has proven to be rather difficult and there is not yet a detailed comparison of its operating characteristics with the numerous theoretical predictions. I hope that my research group will remedy this situation in the near future using the Raman two-photon setup described in § 7. I also anticipate that researchers will begin to explore the strong atom–cavity coupling regime where a single atom passing through a cavity can be induced to emit two or more photons at a time on demand (see Bertet, Osnaghi, Milman, Auffeves, Maioli, Brune, Raimond and Haroche [2002] and Kuhn, Henrrich and Rempe [2002]), thereby giving us the ability to ‘quantum engineer’ states of the electromagnetic field.

Acknowledgments

I gratefully acknowledge very enjoyable collaborations on two-photon laser research with William Brown, Hope Concannon, Juanjo Fernández-Soler, Josep Lluís Font, Jeff Gardner, Alexandar Kul’minskii, Maciej Lewenstein, Steve Morin, Thomas Mossberg, Olivier Pfister, Michael Stenner, Ramon Vilaseca, Qilin Wu, and Yifu Zhu, and fruitful discussions with Robert Boyd, Michael
Raymer, Peter Sorokin, and John Thomas. I thank Lorenzo Narducci for encouraging me to write this review, and the long-term financial support of the US Army Research Office and the National Science Foundation, especially the current grant No. PHY-0139991.

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