Instabilities in Four-Wave Mixing

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Abstract

Recent research on instabilities in the four-wave mixing process is summarized. The four-wave mixing process can cause a laser beam to become unstable to the growth of new frequency components as the beam propagates through a nonlinear medium. Similar instabilities can occur in more complicated optical systems such as lasers and bistable devices. The nature of the instability is very different for nonlinearities resulting from one- and two-photon resonances in the susceptibility describing four-wave mixing. The four wave mixing process can lead to chaotic behavior in a self-pumped phase conjugate mirror.

Introduction

There has recently been great interest in instabilities that can occur in optical systems. This interest has occurred both because the presence of instabilities can limit the performance of optical systems and, more fundamentally, because optical instabilities present a particularly clean example of unstable behavior in a system governed by straightforward physical principles. The present paper is concerned with instabilities associated with the four-wave mixing process. Such instabilities are of particular interest because they are due solely to the nonlinear response of the material system and are not associated with the interaction of the nonlinear system with an external cavity. These instabilities also provide some insight into the nature of the instabilities associated with more complicated systems such as lasers and optical bistable devices in which optical instabilities have been studied extensively.

The simplest manifestation of the four-wave mixing instability is the growth from noise of new frequency components in an initially monochromatic laser beam as it passes through a nonlinear optical medium. This instability had been predicted on the basis of theoretical arguments in the early 1970s by Mollow and by McCall. As illustrated in Fig. 1, the nature of this instability is the simultaneous annihilation of two photons of frequency \( \omega_1 \) accompanied by the creation of photons of frequencies \( \omega_2 \) and \( \omega_3 \), where

\[
2\omega_1 = \omega_2 + \omega_3
\]

(1)

Since \( \omega_1 - \omega_2 = \omega_3 - \omega_1 \), the new frequency components constitute frequency sidebands symmetrically displaced from the frequency \( \omega_1 \) of the incident field. These sidebands grow due to parametric amplification processes pumped by the incident laser field. Figure 1 shows the four-wave mixing process for both of one- and two-photon resonance because, as will be discussed below, the nature of the instability is quite different in these two cases.

![Energy level diagrams showing the resonances of the four-wave mixing process for (a) one-photon resonance and (b) two-photon resonance.](image)

**Fig. 1.** Energy level diagrams showing the resonances of the four-wave mixing process for (a) one-photon resonance and (b) two-photon resonance.

Instabilities in Propagation through One-Photon-Resonant Media

As mentioned in the Introduction, instabilities in the propagation of laser radiation through one-photon resonant media were originally predicted by Mollow and McCall and have subsequently been discussed by many workers. This instability often manifests itself
as the generation of sidebands displaced from the incident laser frequency by the generalized Rabi frequency associated with the atomic response. The generalized Rabi frequency is expressed as

$$\Omega' = (a^2 + a^2)^{1/2},$$

where $\Omega = E/h$ is the Rabi frequency given in terms of the atomic dipole transition moment $\mu$ and the amplitude $E$, of the applied optical field, and where $\Delta = \omega_2 - \omega_0$ is the detuning of the incident laser from the atomic transition frequency $\omega_0$. The reason why the parametric gain is greatest at the Rabi sidebands can be understood intuitively in terms of the energy-level diagram shown in Fig. 2a. In the presence of an intense laser field, the atomic energy levels become modified as a consequence of the ac Stark effect. The ground and excited states are each split into a doublet of separation equal to $\Omega'$ and are shifted to an energy separation of $\omega_1$. Shown in Fig. 2b is the theoretically predicted gain of the four-wave mixing process as a function of the detuning of the generated wave from the incident laser. This theoretical prediction is obtained by solving the density matrix equations of motion exactly for a two-level atom driven by the incident laser field, obtaining the lowest-order correction to the atomic response in the simultaneous presence of the fields at $\omega_2$ and $\omega_3$, and using the atomic response as a nonlinear source term in the driven wave equation for the optical field. This curve was plotted for the case where $(\omega_1 - \omega_0)T_2 = 6$, $\Omega' T_2 = 25$, and $T_2/T_1 = 2$, where $T_2$ and $T_1$ are respectively the dipole dephasing time and population relaxation time, and where $\omega_0 L = 100$, where $\omega_0$ is the line-center unsaturated absorption coefficient and $L$ is the length of the interaction region.

The Rabi sideband instability was observed experimentally by Harter et al in an experiment performed in atomic sodium vapor. These authors verified that the transmitted laser beam contained the sideband frequencies $\omega_1 + \Omega'$ and $\omega - \Omega'$, and that the sideband detuning scaled with laser intensity and detuning as predicted by Eq. (2). They also showed that for large laser intensities the lower frequency sideband was emitted in the form of a cone surrounding the transmitted laser beam, and interpreted this result as a consequence of non-collinear phase matching. The nature of this conical emission has been discussed by numerous other workers.

Since the two sidebands are emitted by a phase-matched four-wave mixing process, they are emitted with a well-defined phase relationship, and consequently the superposition of the central component with the two sidebands constitutes a temporally modulated field of a well-defined modulation form. This modulated optical field can be represented as

$$E(t) = 2 \text{Re} \left[ (E_0 + \delta E(t)) \exp[i(\omega_1 t + \phi)] \right]$$

where we assume $E_0$ to be real. Then, depending on whether $\delta E$ is real, imaginary, or complex, the modulation form is respectively amplitude modulation (AM), frequency modulation (FM), or a combination thereof. Physically, the phase of $\delta E$ depends on the relative phases of the two sidebands. Hillman et al.12 and Kruemer et al.13 have shown that in the limit where the sidebands are weak compared to the central component, there always exist two forms of modulation that remain unchanged as the beam propagates through the nonlinear medium. These forms are known as natural modes of propagation through a nonlinear medium. These natural modes have the property that the amplitudes of the two sidebands can change due to the nonlinear interaction but the relative amplitudes and phases remain unchanged. For the case

Fig. 2. (a) Atomic energy levels as modified by the ac Stark effect. (b) Gain experienced by a weak probe beam as predicted by the model described in the text. Note the strong resonant response at the Rabi sidebands.
where the central component is exactly resonant with the atomic transition, the natural modes are pure amplitude modulation and pure frequency modulation, although more generally they are of a combination of these two forms of modulation. If the sidebands are allowed to grow from noise, the mode with the largest gain will always dominate. For the case of the Rabi-sideband instability with the incident field exactly on resonance, the natural mode with the largest gain has the form of amplitude modulation.

The presence or absence of dephasing collisions has a profound effect on the nature of the nonlinear response that leads to instabilities. This dependence is seen most dramatically in the absorption lineshape experienced by a probe wave in the presence of a strong, fixed-frequency pump wave. As described originally by Schwartz and Tan" and subsequently by others, the probe absorption profile contains a hole of width $\frac{1}{T_2}$ centered on the laser frequency. This feature is reminiscent of the spectral hole burning that occurs in inhomogeneously broadened transitions, but has a different physical origin, population oscillations. The theoretically predicted probe absorption profile obtained through the solution of the density matrix equations of motion for a two-level atom are shown for two cases in Fig. 3.

![Fig. 3. Probe absorption lineshape for a homogeneously broadened two-level atom for (a) $T_2/T_1=2$ and (b) $T_2/T_1=0.1$. In each case the upper curve shows the profile in the absence of saturation and the lower curve shows the response when the atom is partially saturated so that the saturation parameter $\beta^2 T_1 T_2$ is equal to 0.25. The horizontal axis is normalized to $\Gamma_{ba}=1/T_2$.](image)

The spectral hole due to population oscillations occurs only for $T_2 < T_1$, that is, in the presence of dephasing collisions. The occurrence of this spectral feature was observed by Hillman et al. in an experiment performed in ruby. Due to the long ground state recovery time of ruby, the spectral hole had a very narrow width of 37 Hz. Additional experiments involving other materials have demonstrated the existence of spectral holes as narrow as 750 millihertz.

The existence of spectral holes such as that shown in Fig. 3b has important implications to the stability characteristics of homogeneously broadened lasers. As the pumping rate of the laser is increased, the laser would be expected to reach threshold first at line center, at which point gain saturation occurs so that at equilibrium the gain is equal to loss. However, Fig. 3b shows that although the gain may be well saturated at line center, regions of large gain can occur in the wings of the line. (For the case of an inverted medium, the vertical axis represents probe amplification, not absorption.) This gain can allow sidemodes to grow and cause the laser to run unstably. An experiment to verify that even homogeneously lasers can become unstable due to such hole-burning effects was performed by Hillman et al. using a rhodamine 6G dye laser. They also found that at still higher laser intensities the laser bifurcates into a new regime of stable operation characterized by the simultaneous occurrence of two modes separated by the Rabi frequency associated with the material response. These observations were found to be in good agreement with a theory that includes the effects of both gain saturation and four-wave mixing processes that couple together the symmetrically displaced sidemodes.

**Instabilities in Propagation through Two-Photon-Resonant Media**

The nature of the instabilities in the four-wave mixing process is somewhat different from that discussed above for the case in which the applied radiation makes a two-photon resonance with the material system, as illustrated in Fig. 1b. The reason for the difference is two-fold: (1) The two-photon Rabi frequency is not in general equal to the ac...
Stark shift of the interacting levels, in contrast to the case of one photon resonance where the Rabi frequency and level shifts are equal.\(^4\) (2) Additional pathways of de-excitation are usually available in the two-photon case.

An experiment conducted by Krasinski et al\(^6\) has demonstrated instabilities for the case of a laser beam tuned close to the 3s–3d two-photon-allowed transition of atomic sodium vapor. Strong emission was observed in the forward direction near the 3d–3p and 3p–3s transition frequencies. The emission near each transition was approximately 10 A broad and was observed to be emitted in the form of a cone surrounding the transmitted laser beam. The measured cone emission angle is shown in Fig. 4 as a function of the emission wavelength for several different sodium number densities. In order to demonstrate that the origin of this conical emissions was a phase-matched four-wave mixing process, Krasinski et al have compared the measured emission angle with that predicted theoretically on the basis of the assumption that all components of the photon momenta are conserved in the four-wave mixing process, as shown in the phase-matching diagram of the inset to Fig. 4. The agreement between the solid theoretical curves and the data are quite good except very close to resonance where absorption effects not included in the theory are important.

Fig. 4. Cone emission angle as a function of the wavelength of the emitted radiation. The laser was tuned to the 3s–3d two-photon-allowed resonance of the sodium atom. The data are in good agreement with the theoretical curves obtained under the assumption that the radiation is emitted by a phase-matched four-wave mixing process in which the wave vectors are arranged as shown in the inset. The atomic number densities are (i) \(3.8 \times 10^{16}\) cm\(^{-3}\), (ii) \(1.8 \times 10^{16}\) cm\(^{-3}\), and (iii) \(7.4 \times 10^{15}\) cm\(^{-3}\).

In additional experiments, Malcuit et al\(^2\) have shown that under two-photon-resonant excitation the four-wave mixing process can be sufficiently strong to lead to the suppression of other nonlinear optical processes, even those that nominally are expected to have larger gain and hence to dominate. Their experiment entailed tuning a laser to the 3s–3d transition of atomic sodium and measuring the relative intensities of the four-wave mixing signal and of that due to amplified spontaneous emission (ASE) between the 3d upper level and the 3p intermediate level. The ASE signal is a probe of the population in the 3d excited state. Some of the data of Malcuit et al are reproduced in Fig. 5. As the laser intensity is increased, thereby increasing the gain of the four-wave mixing process, the relative intensity of the ASE is observed to decrease. These and other data demonstrate that the four-wave mixing process once fully developed can inhibit the excitation of the 3d level. Malcuit et al present a calculation that predicts the suppression of excitation of the 3d level under their experimental conditions. This calculation is based on the solution to the coupled amplitude equations of nonlinear optics with nonlinear susceptibilities calculated from the density matrix equations of motion for the three-level system consisting of the 3s, 3p, and 3d levels. Intuitively, the reason for suppression of the 3d population is that the four-wave mixing process leads to the creation of new fields at frequencies \(\omega_2\) and \(\omega_1\). These new fields create a new pathway for two-photon excitation of the 3d level. Since these new fields are generated by a phase-matched process, they possess a definite phase relationship with respect to the incident field at \(\omega_i\), and the phase relationship is such that the pathway associated with the absorption of one photon of frequency \(\omega_2\) and one photon of frequency \(\omega_1\) interferes destructively with that associated with the absorption of two \(\omega_i\) photons. Competition effects of the sort described by Malcuit et al could have important implications for the design of extreme ultraviolet lasers that rely on multiphoton excitation for their pumping. It has been suggested\(^2\) that the absence of population inversion reported recently in an experiment\(^2\) involving barium vapor may have been due to related competition effects.
Instabilities with Counterpropagating Waves

The instabilities discussed above that occur when a single laser beam passes through a nonlinear medium do not lead to chaotic evolution of the system. In contrast, there is good theoretical evidence that chaotic instabilities can result from the mutual interaction of two counterpropagating waves.

Winful and Marburger\textsuperscript{24} showed in 1980 that hysteresis and optical bistability could occur in the polarization of the transmitted beams in degenerate four-wave mixing involving counterpropagating vector fields in an isotropic Kerr medium. Later, Lytel\textsuperscript{25} showed that multistability could occur, and Kaplan and Law\textsuperscript{26} showed that the solution possessed multiple isolated branches known as isolas. These results suggest but do not prove that the solutions can show chaos in certain branches of the parameter space. More recently, Yumoto and Otsuka\textsuperscript{27} and Gregori and Wabnitz\textsuperscript{28} showed that at least for the case of anisotropic media the solutions can become chaotic in the sense of being very sensitive to the length of the interaction region. In order to determine whether counterpropagating waves in an isotropic medium show instabilities in its time evolution, Gaeta et al.\textsuperscript{29} have performed a numerical integration of the propagation equations given in references 25 and 26 including time dependence. An example of the solutions they obtain is shown in Fig. 6. This solution shows periodic oscillations of the intensity associated with one polarization component of the output.

Fig. 5. Ratio of the emitted intensity in the forward direction to that in the backward direction as a function of the laser intensity, for a laser tuned to the 3s-3d transition of atomic sodium vapor for two different laser confocal parameters $b$. Ratios of unity and zero correspond respectively to pure ASE and pure four-wave mixing. ASE is seen to be suppressed by the four-wave mixing process under conditions of large laser intensity.

Fig. 6. Temporal evolution of the intensity associated with one polarization component of the field transmitted by a Kerr nonlinear medium for the case of counterpropagating waves of equal intensity. The intensity is such that the nonlinear refractive index due to each wave separately leads to a phase shift of 4 radians in one passage through the interaction region.
The instabilities discussed in the previous paragraph occur only in the polarization characteristics of the transmitted fields. Silberberg and Bar-Joseph have shown that chaotic fluctuations in the total intensity of each transmitted beam can occur if the nonlinear response of the medium is characterized by a Debye relaxation equation with a nonzero response time. They show that the instability occurs because the counterpropagating waves interfere to create a grating that acts as a distributed feedback structure, and that four-wave mixing (in the limit often referred to as two-beam coupling) provides gain for the growth of new frequency components.

Instabilities in Phase Conjugation

One of the primary methods of producing the phase conjugate of an optical wavefront is by means of degenerate four-wave mixing. Instabilities might therefore be expected to be present in the phase conjugation process. Light-induced critical behavior in phase conjugation has been predicted theoretically by Flytzanis and Tang, and instabilities in a phase conjugate resonator have been described by Valley and Dunning.

Chaotic instabilities have recently been observed by Narum et al in a self-pumped barium titanate phase-conjugate mirror. Self-pumped phase conjugation in barium titanate was originally discovered by Feinberg. According to the model developed to explain this effect, the incident laser beam breaks up into two beams due to a self-focusing process. Due to total internal reflection at the surfaces of the crystal, these beams are redirected so that they each intercept the incident laser beam a second time and thereby form two interaction regions in which each phase conjugation can occur due to the usual four-wave mixing process. Since the two pump beams grow from noise in a highly nonlinear process and interact strongly with the incident and conjugate beams, the output beam might be expected to show chaotic fluctuations.

Some of the results of the experiment of Narum et al are shown in Fig. 7. The output intensity is seen to display wild fluctuations, even though the input laser is relatively noise free and even though great care was taken to ensure that the phase conjugate signal was not allowed to feed back into the laser. This data has been analyzed by the method of Grassberger and Procaccia, and the results are shown in Fig. 6b. The slope and spacing of the straight-line portions of these curves suggest that the nonlinear system is evolving on a strange attractor of dimension 2.5 and that the Kolmogorov entropy of the system is approximately 0.7 bits per second.

Instabilities in Stimulated Scattering

Milonni et al have shown that chaos can occur in stimulated Raman scattering due to the coupling of the Stokes and anti-Stokes waves by four-wave mixing processes, if they make the assumption that the anti-Stokes but not the Stokes wave shows Raman gain. Randall and Albritton have shown that chaos can occur in stimulated Brillouin scattering if a boundary reflective to light is placed near the interaction region, and present experimental results that are in qualitative agreement with their theory. Bar-Joseph et al have studied similar instabilities for the case of stimulated Brillouin scattering in an optical fiber.
Conclusions

Instabilities can occur due to four-wave mixing processes in the propagation of laser beams through nonlinear optical media which can lead to unstable operation of lasers and phase conjugate devices.

Acknowledgements

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References