# Suppression of Amplified Spontaneous Emission by the Four-Wave Mixing Process

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Two-photon-resonant excitation of the sodium $3d$ level can lead to the generation of new frequencies either by amplified spontaneous emission at the $3d \rightarrow 3p$ transition frequency or by a resonantly enhanced four-wave mixing process. Competition between these two processes has been observed. The four-wave mixing process can suppress amplified spontaneous emission by preventing the excitation of the $3d$ level due to an interference between two different pathways connecting the ground ($3s$) and $3d$ states.

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In this Letter, we present the results of an experimental study that demonstrates competition between amplified spontaneous emission (ASE) and the four-wave mixing (FWM) process. An intense laser beam was tuned to the $3s \rightarrow 3d$ two-photon-allowed transition of the sodium atom and the radiation emitted by the vapor cell was examined in both the forward and backward directions. We observed that when the input laser was tuned off resonance, a FWM process led to the generation of radiation near the $3d \rightarrow 3p$ and $3p \rightarrow 3s$ transition frequencies (see Fig. 1). When the laser is tuned precisely to resonance, population can be transferred to the $3d$ level, inverting it with respect to the $3p$ level and leading to ASE at the $3d \rightarrow 3p$ transition frequency. Under conditions of intense resonant excitation, the gain of the ASE process calculated in the absence of competition is many orders of magnitude larger than that of FWM (see below). Although ASE is thus expected to dominate, we have observed experimentally that for appropriate focusing of the laser beam only FWM is present. When the FWM process is inhibited through the use of counterpropagating pump beams, the normal result of ASE is observed. We interpret these observations to be a consequence of a previously unrecognized competition between the ASE and FWM processes. The fields generated by the FWM process create a new pathway connecting the $3s$ and $3d$ levels. This pathway can interfere destructively with that involving only the frequency of the incident field, prohibiting the transfer of population to the $3d$ level. The nature of the suppression is described by the solutions of the density matrix equations for a three-level atomic system.

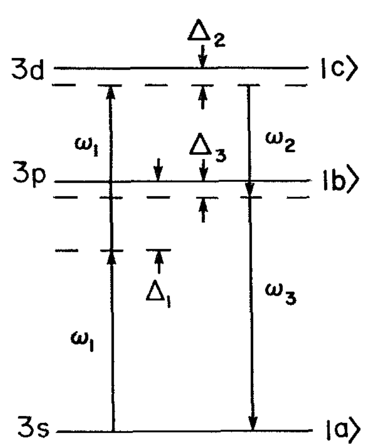


FIG. 1. Energy-level diagram showing the four-wave mixing process and the notation used in the calculation.

Competition between coherent and incoherent optical processes has recently been the subject of intense interest. The only previously reported example of such competition has been the suppression of multiphoton ionization by third-harmonic generation. This effect was observed by Miller *et al.*[1] and by Glownia and Sander,[2] and has been successfully explained by Jackson and Wynne,[3] and others.[4–7] However, these theoretical explanations presuppose that one of the optical fields is strongly absorbed by the nonlinear medium, and hence do not apply to our system. Our work demonstrates that competition effects can occur under conditions more general than those previously believed to be necessary.

Our experimental setup consists of a pulsed dye laser of energy $\leq 500$ $\mu$J, pulse length of 1.5 ns, and spectral width of 0.7 cm$^{-1}$. The 6855-Å output of this laser was focused to a $\sim 100$-$\mu$m spot size and into a heat-pipe oven containing $\sim 10^6$ sodium atoms per cubic centimeter. Radiation generated within the cell could be directed to a spectrometer having a resolution of 0.3 Å.

The ASE[8,9] and FWM[10] processes have different spatial and spectral signatures, which allow the processes to be unambiguously distinguished. As a result of phase-matching considerations, FWM can occur only in the forward direction. For the number densities and detunings used in our experiment, this radiation has the shape of a cone surrounding the transmitted laser beam [see Fig. 2(a)].[11,12] When ASE occurs it has equal gain in the forward and backward directions, and has the shape of a filled-in disk. We observe cone-shaped emission in the forward direction even when tuned precisely to resonance, which shows that FWM, and not ASE, is occurring. FWM and ASE

 

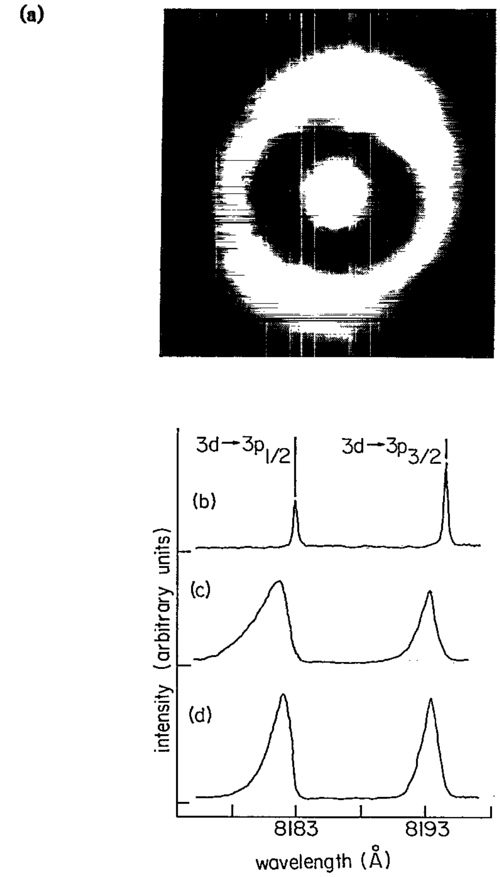


FIG. 2. (a) Spatial distribution of the radiation emitted near the $3p \rightarrow 3s$ transition frequency due to FWM. (b) Spectrum of the radiation (ASE) emitted in the backward direction. (c) Spectrum of the radiation generated by FWM in the forward direction for a detuned laser. (d) Spectrum of the radiation emitted in the forward direction with the laser tuned precisely to the two-photon resonance.

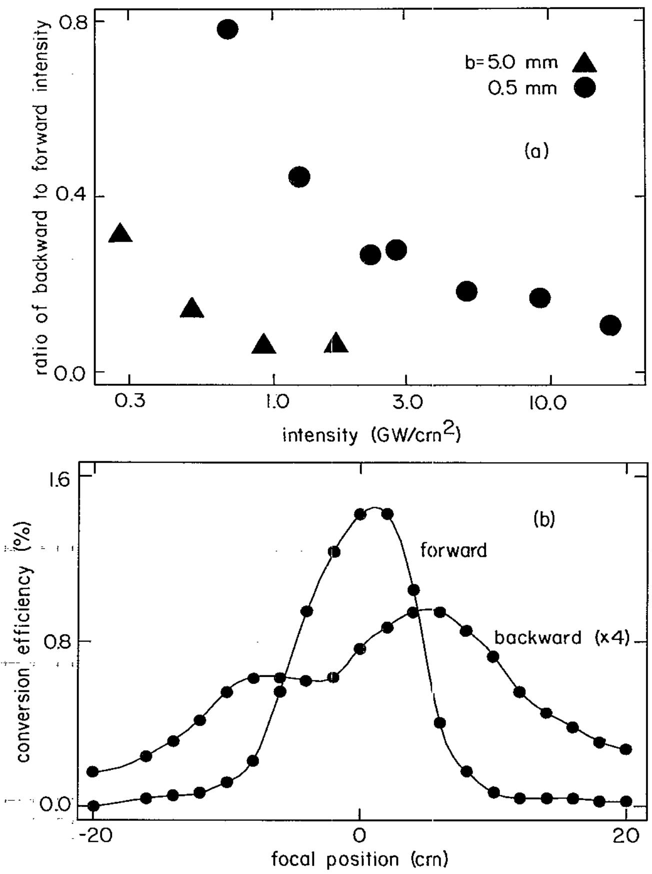


FIG. 3. (a) Ratio of the intensity of the emission in the backward direction to that in the forward direction vs incident laser intensity for two values of the beam confocal parameter. The limiting values of unity and zero correspond to pure ASE and pure FWM, respectively. (b) Intensities of the emission in the forward and backward directions vs the position of the beam waist in the cell. Note that ASE is suppressed when FWM is enhanced.

can also be distinguished spectrally. The spectrum of ASE has features only at the $3d \rightarrow 3p_{3/2}$ and $3d \rightarrow 3p_{1/2}$ transitions frequencies [see Fig. 2(b)], whereas the FWM process produces a broader output spectrum that peaks $\sim 1.5$ Å to the short wavelength side of resonance and tails off to shorter wavelengths [see Fig. 2(c)]. These spectral characteristics occur as the result of phase-matching considerations.[13] Figure 2(d) shows the spectrum of the light emitted in the forward direction when the input laser was tuned precisely to the two-photon resonance. The spectrum is seen to be that of FWM and not ASE.

We interpret the absence of ASE when tuned precisely to the two-photon resonance to be a consequence of competition between the ASE and FWM processes. To verify this interpretation, we instead excited the $3d$ level using counterpropagating pump waves of slightly different frequency, adjusted so that the sum of their frequencies was equal to the $3s \rightarrow 3d$ transition frequency and that their combined intensity was the same as that used in the single-beam experiments in which suppression of ASE was observed. FWM involving one photon from each pump wave could not occur as a result of phase-matching constraints, and furthermore FWM involving two photons from the same pump wave was very inefficient because there was no two-photon resonance enhancement. Under these conditions, ASE was observed, demonstrating that it is the existence of efficient FWM that leads to the suppression of ASE.

Since FWM is a phase-matched process, whereas ASE is not, the competition between ASE and FWM depends on the focusing characteristics of the incident laser beam. Figure 3(a) shows the ratio of the emission in the backward direction (ASE) to the total emission in the forward direction (ASE plus FWM) plotted as a function of the maximum intensity of the incident laser beam and for focusing into the center of the cell.

The ratio is seen to decrease with laser intensity because ASE is suppressed more fully as the gain of the FWM process increases. The ratio also decreases as the confocal parameter of the incident beam increases because the fields generated by the FWM process can grow to a larger value in a longer interaction length. Figure 3(b) shows how the intensity of the forward and backward emission changes as the position of the beam waist is varied within the cell. The confocal parameter (5 mm) and intensity (1 GW $cm^{-2}$) have been chosen so that strong competition occurs when the beam waist is in the center of the cell. Note the decrease in the backward emission (ASE) for focal positions for which FWM occurs efficiently.

The nature of the competition between ASE and FWM can be understood by calculating the steady-state population of level $c$ and the magnitudes of the nonlinear polarizations giving rise to FWM. We calculate these quantities by solving the density-matrix equations of motion for a three-level atomic system to fourth order in perturbation theory. We assume that the atomic system is subjected to fields at frequencies $\omega_1$, $\omega_2$, and $\omega_3$, with the convention that the field at frequency at $\omega_j$ is given by $E_j \exp(ik_jz - i\omega_jt) + \text{c.c.}$ We find, using the notation defined in Fig. 1, that for the case of exact two-photon resonance ($\Delta_2 = 0$) and in the limit $\Delta_1 >> \Delta_3 >> \Gamma_{ba}$, where $\Gamma_{ba}$ is the dipole dephasing rate of the $b \rightarrow a$ transition, the dominant (i.e., most resonant) contributions to the nonlinear polarization are given by

$$P^{\rm NL}(\omega_1) = 2\chi^{\rm FWM} E_2 E_3 E_1^* e^{-i\Delta kz} + 2\chi_s^{\rm TPA} |E_1|^2 E_1, \quad (1a)$$

$$P^{\rm NL}(\omega_2) = \chi^{\rm FWM} E_1^2 E_3^* e^{i\Delta kz} + \chi_d^{\rm TPA} |E_3|^2 E_2, \quad (1b)$$

$$P^{\rm NL}(\omega_3) = \chi^{\rm FWM} E_1^2 E_2^* e^{i\Delta kz} + \chi_d^{\rm TPA} |E_2|^2 E_3, \quad (1c)$$

where the nonlinear susceptibilities for FWM and for two-photon absorption (TPA) involving the same and different frequencies are given by

$$\chi^{\rm FWM} = \frac{\Delta_1}{\Delta_3}\chi_s^{\rm TPA} = \frac{\Delta_3}{\Delta_1}\chi_d^{\rm TPA} = \frac{iN|\mu_{ba}|^2|\mu_{bc}|^2}{\hbar^3 \Delta_1 \Delta_3 \Gamma_{ca}}, \quad (2)$$

where $\mu_{ij}$ denotes the $ij$ matrix element of the dipole operator and $N$ is the atomic number density. In the same limits we find that the probability that a given atom is in state $c$ is given by

$$\rho_{cc}^{(4)} = \frac{1}{\hbar N \gamma_c}\{\text{Im}\chi_s^{\rm TPA}|E_1|^4 + \text{Im}\chi_d^{\rm TPA}|E_2|^2|E_3|^2 + \text{Im}[\chi^{\rm FWM}(E_2E_3E_1^{*2}e^{-i\Delta kz} + \text{c.c.})]\}, \quad (3)$$

where $\gamma_c$ is the inverse of the lifetime of level $c$. This expression contains an interference term which for appropriate values of the complex field amplitudes can cause the population in level $c$ to vanish. The condition for this to occur is

$$A_2A_3/A_1^2 = \Delta_3/\Delta_1, \quad (4a)$$

and

$$2\phi_1 - \phi_2 - \phi_3 + \Delta kz = \pi, \quad (4b)$$

where the real amplitude and phase is defined by $E_i = A_i \exp(i\phi_i)$. At the entrance face of the medium only the field $E_1$ is present, but as a result of the FWM process the fields $E_2$ and $E_3$ can be created by growing from noise. The spatial evolution of the fields is governed by coupled amplitude equations of the form

$$\frac{\partial E_i}{\partial z} = \frac{2\pi i \omega_i^2}{k_i c^2} P^{\rm NL}(\omega_i), \quad i = 1, 2, 3. \quad (5)$$

Since the intensity of the wave at $\omega_i$ is given by $S_i = (cn_i/2\pi)|E_i|^2$, the spatial evolution of the intensity is given through the use of Eq. (5) by

$$\frac{\partial S_i}{\partial z} = -\frac{\omega_i}{2}\text{Im}[E_i^* P^{\rm NL}(\omega_i)], \quad i = 1,2,3, \quad (6)$$

where $P^{\rm NL}(\omega_i)$ is given by Eq. (1). An important solution of these equations is that for which $\partial S_i/\partial z = 0$ for $i = 1$, 2, and 3. Under these conditions, the waves *neither* exchange energy among themselves nor lose energy to the medium. The condition for this to occur is seen by inspection to be precisely that for $\rho_{cc}^{(4)}$ to vanish, as given by Eq. (4).

In order to determine the spatial evolution of the complex field amplitudes, we have performed a numerical integration of the coupled amplitude equations (5). We find that under very general conditions the fields evolve in such a way as to satisfy the conditions (4) for the suppressions of excitation of level $c$. A typical solution is shown in Fig. 4. Our laser operated in approximately ten longitudinal modes, only one of which was resonant with the two-photon transition. The calculation assumes an input field of magnitude 80 esu, which corresponds to the field strength of a single mode. The calculated gain[14] of the FWM process is 200 $cm^{-1}$, allowing the $\omega_2$ and $\omega_3$ fields to grow rapidly from noise. However, the maximum conversion efficiency is only $\sim 10^{-3}$. The anomalously low conversion efficiency of parametric mixing processes enhanced by an exact intermediate resonance has been discussed previously (for third harmonic generation) by Kildal and Brueck.[15] The measured efficiency of the FWM process is $\sim 0.02$. We believe that this disagreement results from the following effects: (1) the incident laser beam is multimode and thus not all of the incident radiation is exactly at the two-photon resonance, (2) the generated radiation is ejected in a

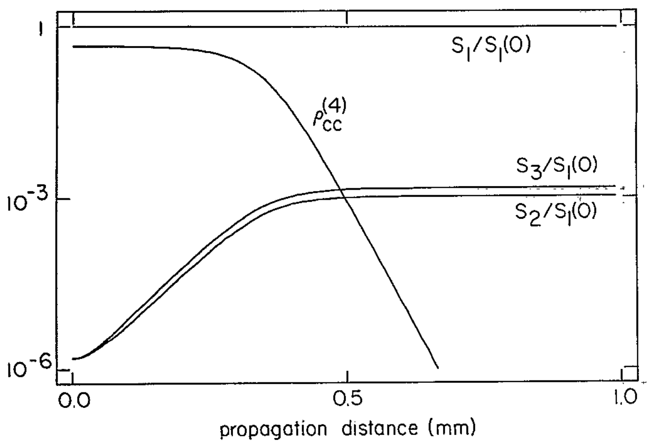


FIG. 4. Results of a numerical integration of the coupled equations showing the spatial variation of the field intensities and the population of level $c$, for $\Delta k=0$, $\Delta_1/2\pi c=2392$ $\text{cm}^{-1}$, $\Delta_2/2\pi c=0$, $\Delta_3/2\pi c=4.5$ $\text{cm}^{-1}$, $N=5\times10^{16}$ $\text{cm}^{-3}$, $\Gamma_{ca}=1.3\times10^{10}$ $\text{s}^{-1}$, $\gamma_c=6\times10^{7}$ $\text{s}^{-1}$, and $A_1=80$ esu.

cone from the interaction region as a result of phase-matching requirements, and (3) our experiment was conducted with field intensities for which saturation effects not included in our theory may be of some importance. Figure 4 also shows that $\rho_{cc}^{(4)}$ drops to a small value in a fraction of a millimeter. ASE is thus suppressed because the inversion between levels $c$ and $b$ is decreased as a result of the presence of the fields frequencies $\omega_2$ and $\omega_3$. The prevention of excitation of level $c$ is a form of population trapping[16] in which the population is trapped in the ground state.

We have also studied ASE and FWM when tuned to the $4d$ level and have found that strong competition does not exist under our experimental conditions. The calculated gain for the FWM process involving the $4d$ and $3p$ levels is comparable to that for the $3d$ and $3p$ levels. However, the gain cross section for ASE involving the $4d\rightarrow4p$ transition is $4\times10^{-11}$ $\text{cm}^2$, whereas that for the $3d\rightarrow3p$ transition is $5\times10^{-12}$ $\text{cm}^2$. We believe that competition is not observed when tuned to the $4d$ level because the $E_2$ and $E_3$ fields cannot grow rapidly enough to suppress ASE when its gain is sufficiently large. However, detailed calculations would be necessary to determine the nature of competition under these conditions.

In conclusion, we have observed competition between ASE and FWM when tuned to the sodium $3d$ state and have developed a theoretical model that explains these observations.

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