Nondegenerate four-wave mixing in a Doppler-broadened resonant medium

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Received August 9, 1980

Third-order susceptibilities are calculated for a Doppler-broadened two-level system. The dependence of four-wave mixing on the angle θ between the pump and signal fields is studied, and the reflection coefficient is shown to have a narrow field of view that is inversely proportional to the ratio of the Doppler width to the homogeneous linewidth. For the collinear geometry, $\theta = 0°$, the frequency dependence of the nondegenerate case is analyzed and shown capable of yielding a real-time narrow-bandwidth optical filter whose bandwidth is limited by the homogeneous linewidth.

1. INTRODUCTION

Recently there has been a considerable amount of interest in the use of resonantly enhanced systems[1–5] to generate phase-conjugate signals through four-wave mixing. Several authors have analyzed four-wave mixing in a homogeneously broadened two-level system,[6,7] and a recent paper[8] has included the effects of atomic motion in several special cases. This paper examines nondegenerate four-wave mixing in a Doppler-broadened two-level system and presents general results that can be used to analyze the efficiency of four-wave mixing in spectroscopy, optical filters,[7] and other applications.

2. CALCULATIONS

Using the geometry of Yariv and Pepper[9] shown in Fig. 1, the fields are taken as plane waves:

$$E_i(r_i,t) = \tfrac{1}{2}A_i(r_i)\exp[i(\omega_i t - \mathbf{k}_i \cdot \mathbf{r}_i)] + \text{c.c.}, \tag{1}$$

where r_i is the distance along $\mathbf{k}_i$. The mixing involves two intense counterpropagating pump waves E_1 and E_2 of the same frequency ω and two weak counterpropagating waves E_3 and E_4 with frequencies ω_3 and ω_4, respectively. We have

$$\mathbf{k}_1 + \mathbf{k}_2 = 0, \qquad \omega_3 + \omega_4 = 2\omega. \tag{2}$$

The two-level system is characterized by a dipole moment μ, an energy splitting $\hbar\omega_0$, and longitudinal and transverse relaxation times T_1 and T_2, respectively.[10] The density matrix equations are solved to third order by perturbation theory[10] for an atom of velocity $\mathbf{v}$ to determine the induced polarizations at ω_3 and ω_4.

$$P(\omega_3 = 2\omega - \omega_4, \mathbf{v}_N) = \frac{c}{4\pi\omega_3}\{-i\alpha_3(\mathbf{v}_N)A_3 + \kappa_3{}^*(\mathbf{v}_N)A_4{}^* \exp[i(\Delta k)z]\}\exp[i(\omega_3 t - \mathbf{k}_3\cdot\mathbf{r})],$$

$$P(\omega_4 = 2\omega - \omega_3, \mathbf{v}_N) = \frac{c}{4\pi\omega_4}\{-i\alpha_4(\mathbf{v}_N)A_4 + \kappa_4{}^*(\mathbf{v}_N)A_3{}^* \exp[i(\Delta k)z]\}\exp[i(\omega_4 t - \mathbf{k}_4\cdot\mathbf{r})] \tag{3}$$

The constants appearing in Eqs. (3) are given by

$$\alpha_3(\mathbf{v}_N) = \frac{-i\alpha_0}{(\delta - \nu - \hat{n}_3\cdot\mathbf{v}_N - i)}, \qquad \alpha_4{}^*(\mathbf{v}_N) = \frac{i\alpha_0}{(\delta + \nu - \hat{n}_4\cdot\mathbf{v}_N + i)},$$

$$\begin{aligned}\kappa_3{}^*(\mathbf{v}_N) = \frac{\alpha_0}{2}\frac{A_1A_2}{E_s{}^2}\Bigg\{&\frac{-i}{(\delta - \hat{n}_1\cdot\mathbf{v}_N - i)[1 - ia\nu + ia\mathbf{v}_N\cdot(\hat{n}_4 - \hat{n}_1)](\delta - \nu - \hat{n}_3\cdot\mathbf{v}_N - i)}\\ &+ \frac{-i}{(\delta - \hat{n}_2\cdot\mathbf{v}_N - i)[1 - ia\nu + ia\mathbf{v}_N\cdot(\hat{n}_4 - \hat{n}_2)](\delta - \nu - \hat{n}_3\cdot\mathbf{v}_N - i)}\\ &+ \frac{i}{(\delta + \nu - \hat{n}_4\cdot\mathbf{v}_N + i)[1 - ia\nu + ia\mathbf{v}_N\cdot(\hat{n}_4 - \hat{n}_1)](\delta - \nu - \hat{n}_3\cdot\mathbf{v}_N - i)}\\ &+ \frac{i}{(\delta + \nu - \hat{n}_4\cdot\mathbf{v}_N + i)[1 - ia\nu + ia\mathbf{v}_N\cdot(\hat{n}_4 - \hat{n}_2)](\delta - \nu - \hat{n}_3\cdot\mathbf{v}_N - i)}\Bigg\},\end{aligned}$$

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$$\kappa_4(\mathbf{v}_N) = \frac{\alpha_0}{2}\frac{A_1{}^*A_2{}^*}{E_s{}^2}\left\{\frac{i}{(\delta - \hat{n}_1\cdot\mathbf{v}_N + i)[1 - ia\nu + ia\mathbf{v}_N\cdot(\hat{n}_1 - \hat{n}_3)](\delta + \nu - \hat{n}_4\cdot\mathbf{v}_N + i)} + \frac{i}{(\delta - \hat{n}_2\cdot\mathbf{v}_N + i)[1 - ia\nu + ia\mathbf{v}_N\cdot(\hat{n}_2 - \hat{n}_3)](\delta + \nu - \hat{n}_4\cdot\mathbf{v}_N + i)} + \frac{-i}{(\delta - \nu - \hat{n}_3\cdot\mathbf{v}_N - i)[1 - ia\nu + ia\mathbf{v}_N\cdot(\hat{n}_1 - \hat{n}_3)](\delta + \nu - \hat{n}_4\cdot\mathbf{v}_N + i)} + \frac{-i}{(\delta - \nu - \hat{n}_3\cdot\mathbf{v}_N - i)[1 - ia\nu + ia\mathbf{v}_N\cdot(\hat{n}_2 - \hat{n}_3)](\delta + \nu - \hat{n}_4\cdot\mathbf{v}_N + i)}\right\}, \quad (4)$$

where $\delta = (\omega - \omega_0)T_2$ is the normalized detuning of the pump fields (E_1 and E_2) from line center, $\nu = (\omega_4 - \omega)T_2$ is the normalized detuning of the signal field from the pump fields, $a = T_1/T_2$, $E_s{}^2 = \hbar^2/T_1T_2\mu^2$ is the line-center saturation intensity, $\alpha_0 = 4\pi\mu^2\Delta N_0T_2k_0/2\hbar$ is the line-center homogeneous-broadening absorption coefficient of a gas with the same density as the subject gas, k_0 is the magnitude of the wave number at frequency ω_0, $\Delta k = 2(\omega_4 - \omega)/c$, $\hat{n}_i = \mathbf{k}_i c/\omega_i$ is the normalized wave vector, and $\mathbf{v}_N = \omega_0 T_2\mathbf{v}/c$ is the normalized velocity.

The probability function for the velocity distribution is given by

$$\rho(\mathbf{v}_N) = \frac{1}{(\pi u_N{}^2)^{3/2}}\exp[-(v_N/u_N)^2], \quad (5)$$

where u_N is the normalized Doppler velocity spread. The macroscopic polarizations that are used in Maxwell's equations are derived by summing the contributions from all velocities $\mathbf{v}$. The expressions obtained are identical with Eqs. (3) but with the coupling constants given by

$$\begin{aligned}\alpha_3 &= \int\rho(\mathbf{v}_N)\alpha_3(\mathbf{v}_N)\mathrm{d}^3v_N,\\ \alpha_4{}^* &= \int\rho(\mathbf{v}_N)\alpha_4{}^*(\mathbf{v}_N)\mathrm{d}^3v_N,\\ \kappa_4 &= \int\rho(\mathbf{v}_N)\kappa_4(\mathbf{v}_N)\mathrm{d}^3v_N,\\ \kappa_3{}^* &= \int\rho(\mathbf{v}_N)\kappa_3{}^*(\mathbf{v}_N)\mathrm{d}^3v_N. \end{aligned} \quad (6)$$

These coefficients are a function of δ and ν as well as of the wave directions $\hat{n}_1$, $\hat{n}_2$, $\hat{n}_3$, $\hat{n}_4$, but no longer of $\mathbf{v}_N$.

Neglecting pump depletion and using the adiabatic approximation,[7] the mode equations describing the evolution of waves A_3 and A_4 are

$$\begin{aligned}\frac{\mathrm{d}A_3}{\mathrm{d}z} &= \alpha_3A_3 + i\kappa_3{}^*A_4{}^*\exp[i(\Delta k)z],\\ \frac{\mathrm{d}A_4{}^*}{\mathrm{d}z} &= -\alpha_4{}^*A_4{}^* + i\kappa_4A_3\exp[-i(\Delta k)z]. \end{aligned} \quad (7)$$

3. NUMERICAL RESULTS

The frequency and angular dependencies of the reflected signal are studied for the case of a single input wave $A_4{}^*(0)$ in the Doppler-broadened region ($u_N \gg 1$). The reflection coefficient[7] R is defined as

$$R \equiv |A_3(0)|^2/|A_4(0)|^2 \quad (8)$$

and can be approximated by

$$R \simeq |k_3L|^2(1 - e^{-2\alpha_R L})^2/4\alpha_R{}^2L^2 \quad (9)$$

with $2\alpha_R = \mathrm{Re}(\alpha_3 + \alpha_4{}^*)$. The values of the coupling constants are determined from Eqs. (6) by numerical integration on an IBM 370/3032 computer. The parameters are chosen to be $a = 1/2$, $u_N = 100$, $2L/cT_2 = 0.01$, $A_1A_2/E_s^2 = 0.1$, $\alpha_0 L\sqrt{\pi}/u_N = 0.1$.

The angular dependence of the reflected signal is shown for the degenerate case, $\nu = 0$. Figure 2 shows the reflection coefficient plotted versus $\sin\theta$ (θ is the angle between $\mathbf{k}_1$ and $\mathbf{k}_4$) for the case in which $\delta = 0$. For large θ, R is inversely proportional to $(u_N \sin\theta)^4$, in excellent agreement with the work of Wandzura.[8] For small angles, Fig. 3 shows the normalized reflectivity versus $\sin\theta$ for several values of the pump detuning δ. This figure disagrees with the results presented by Wandzura,[8] who used a simple formula to interpolate between his results at $\theta = 0°$ and at large angles. The narrow field of view shown in this figure is inherent to any Doppler-broadened system as a result of the angular bandwidth's being inversely proportional to u_N. As δ increases, the field of view does increase, but this is realized only with a concomitant decrease in the absolute value of R, as shown in Fig. 4, where the reflection coefficient is plotted versus the detuning δ for the case in which $\theta = 0°$. From Fig. 4, one notes that the re-

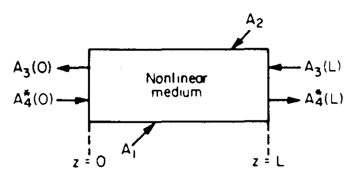

Fig. 1. Geometry for nearly degenerate four-wave mixing (assuming nondepleting pump waves).

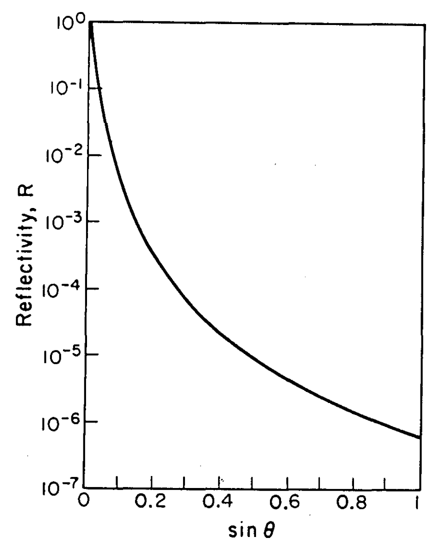

Fig. 2. Reflectivity versus $\sin\theta$ for $\delta = 0$ and $\nu = 0$. The curve is normalized to unity at $\theta = 0°$.

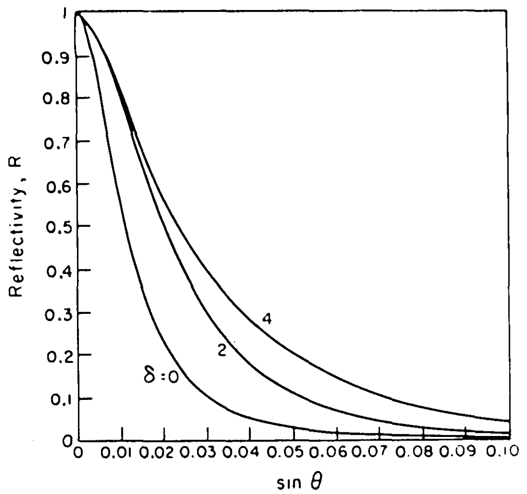

Fig. 3. Reflectivity for degenerate four-wave mixing versus sin θ for several values of the pump detuning δ. All curves are normalized to unity at $\theta = 0°$.

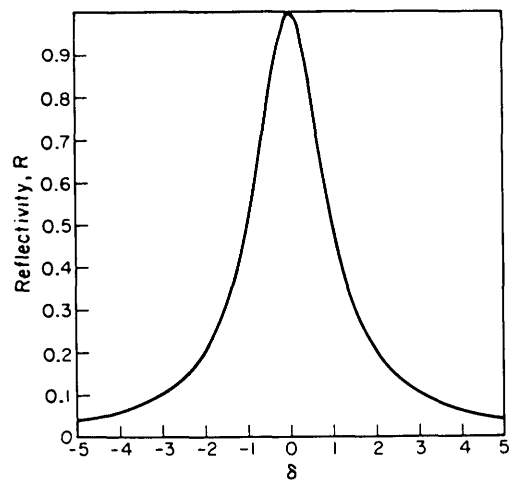

Fig. 4. Reflectivity for degenerate four-wave mixing versus pump detuning δ for the collinear case $\theta = 0°$. The curve is normalized to unity at $\delta = 0$.

flectivity peaks on resonance, $\delta = 0$, and has a line shape determined by the homogeneous linewidth. A recent paper by Elci and Rogovin[11] suggests that there should be a dip in the intensity of the phase-conjugate wave at line center ($\delta = 0$). This prediction results from the incorrect method they used in order to include the effects of Doppler broadening on the third-order susceptibility.

To obtain an approximate analytic expression for R, the Gaussian distribution in Eq. (5) is modeled by a polynomial[12] and used in Eqs. (6) to obtain the coupling coefficients. For small positive angles $[R(\theta)/R(\theta = 0°) > 0.1]$, the coupling coefficient is given by

$$\kappa_3{}^*L = \frac{\alpha_0 L}{2}\frac{A_1A_2}{E_s{}^2}\frac{ic_1}{u_N\cos(\theta/2)} \times \left\{\frac{1}{[1+2ac_2u_N\sin(\theta/2)][1+c_2u_N\sin(\theta/2)+i\delta]} + \frac{ic_3e^{i\theta_1}}{[1+2ac_4u_N\sin(\theta/2)e^{i\theta_2}][1+c_4u_N\sin(\theta/2)e^{i\theta_2}+i\delta]} + \frac{-ic_3e^{-i\theta_1}}{[1+2ac_4u_N\sin(\theta/2)e^{-i\theta_2}][1+c_4u_N\sin(\theta/2)e^{-i\theta_2}+i\delta]}\right\} \tag{10}$$

with

$$c_1 = 1.878693 \qquad \theta_1 = 4.1182°,$$
$$c_2 = 0.819668 \qquad \theta_2 = 51.7614°,$$
$$c_3 = 0.3937158,$$
$$c_4 = 1.257383.$$

The agreement of Eq. (10) with the computer-integrated results for $\kappa_3{}^*$ is better than 1% for $u_N > 10$. For $\theta = 0°$, Eq. (10) reduces to

$$\kappa_3{}^*L \simeq \frac{\alpha_0 L}{2}\frac{A_1A_2}{E_s{}^2}\frac{\sqrt{\pi}}{u_N}\frac{1}{\delta - i}. \tag{11}$$

Since the reflectivity is the largest for the collinear geometry $\theta = 0°$, the frequency dependence (filter function) of R in the nondegenerate case will be studied for that geometry. Figure

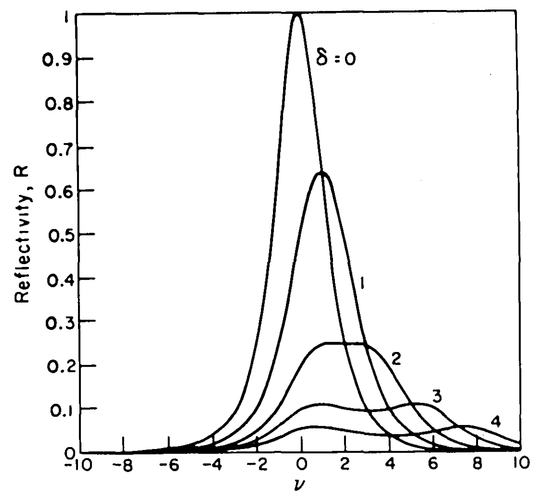

Fig. 5. Reflectivity versus signal detuning ν for several values of the pump detuning δ. The curves are normalized to $\delta = \nu = 0$.

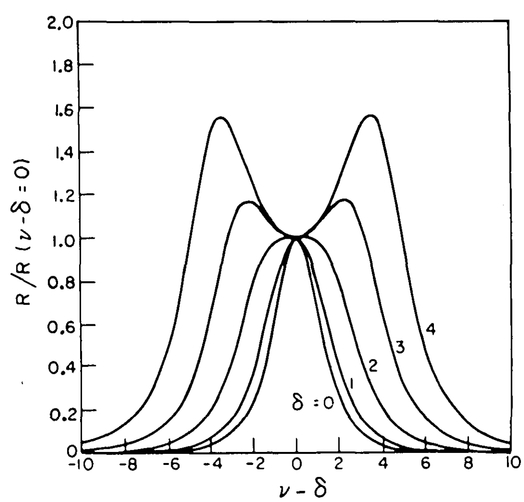

Fig. 6. Reflectivity versus the detuning $\nu - \delta$ for several values of the pump detuning δ. All curves are normalized to unity at $\nu - \delta = 0$.

5 shows the reflection coefficient plotted versus signal detuning ν for several values of the pump detuning δ. As δ becomes larger, the reflectivity has a double-peaked structure with the two peaks near $\nu = 0$ and $\nu = 2\delta$. This is shown better in Fig. 6, where R, normalized to unity, is plotted versus $\nu - \delta$. This behavior is unique to a Doppler-broadened system and does not appear in a system of stationary atoms.[7] The frequency response suggests that an active narrow-bandwidth optical filter can be constructed using a Doppler-broadened system. The bandwidth of the filter depends on the homogeneous linewidth and monotonically increases with pump detuning δ.

Using Eqs. (6) with $\theta = 0°$, the coupling constants can be approximated by

$$\kappa_4 = \alpha_0 \frac{A_1{}^* A_2{}^*}{E_s{}^2} \frac{\sqrt{\pi}}{u_N} \frac{1}{(1 - ia\nu)(2\delta + \nu + 2i)},$$

$$\kappa_3{}^* = \alpha_0 \frac{A_1 A_2}{E_s{}^2} \frac{\sqrt{\pi}}{u_N} \frac{1}{(1 - ia\nu)(2\delta - \nu - 2i)}. \qquad (12)$$

This is valid for δ and $\nu < 0.1 u_N$.

4. CONCLUSION

The primary effects of Doppler broadening are to reduce the effective absorption cross section and to impart an angular dependence to the reflection coefficient. The angular dependence would severely limit the field of view of devices based on this system. However, it is important to note that the linewidth of the reflection coefficient approaches the homogeneous linewidth at small angles.

The frequency response of the Doppler-broadened system in the nondegenerate case has demonstrated how four-wave mixing can yield an active narrow-bandwidth optical filter whose frequency characteristics depend on the homogeneous linewidth. The field of view of such a device would be limited.

J. Nilsen thanks the Fannie and John Hertz Foundation for support of graduate studies. J. Nilsen is a Fannie and John Hertz Foundation Fellow. This research also was supported by the U.S. Army Office, Durham, N.C.

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