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Theory of resonant degenerate four-wave mixing with broad-bandwidth lasers

G. Alber, J. Cooper, and P. Ewart*
Joint Institute for Laboratory Astrophysics, University of Colorado and National Bureau of Standards, Boulder, Colorado 80309
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The effects of finite laser bandwidth on resonant degenerate four-wave mixing (DFWM) are calculated with use of a model in which the intense, counterpropagating pump beams are characterized by a chaotic field, the probe beam is weak and monochromatic, and the medium consists of a gas of two-level atoms. We present a steady-state solution in the limit where the pump-laser bandwidth exceeds all other atomic relaxation rates. Although the mean intensity due to the fluctuating fields is spatially independent (no steady-state standing-wave pattern is established), the analytic results indicate that, for intensities above the saturation intensity I_{sat}, spatially periodic saturation effects are important. Increasing bandwidth is shown to lead to an increase in the effective saturation intensity resulting in lower phase-conjugate reflectivity for $I < I_{\text{sat}}$ than for coherent pump fields, in contrast to the results for narrow-bandwidth chaotic fields. The resonant DFWM line shape is also calculated and compared to the coherent result. We comment on the application of the model to other four-wave-mixing processes employing broad-bandwidth lasers.

I. INTRODUCTION

Degenerate four-wave mixing (DFWM) has been studied experimentally and theoretically both as a means of producing phase-conjugate reflection and for high-resolution spectroscopy. The field has been reviewed recently in the book edited by Fisher.[1] Resonantly enhanced DFWM induced by saturable absorption is of particular spectroscopic interest. Abrams and Lind have developed a theory of this process in absorbing media which treats the case of monochromatic pump and probe waves, the so-called coherent case.[2,3] In many situations, especially those involving pulsed lasers, the radiation field has a finite bandwidth and this can lead to significant modification of the interactions. The effects of finite laser bandwidth have been studied extensively in the cases of laser-induced resonance fluorescence and multiphoton absorption and ionization.[4–7]

Much less work on bandwidth effects has been done for parametric processes such as four-wave mixing. Saxena and Agarwal have calculated the effects of photon statistics on DFWM in the somewhat idealized case of a chaotic field with a bandwidth much narrower than the natural width of the atomic transition.[8] Field fluctuations have also been studied theoretically for PIER (pressure-induced extra resonances) and CARS (coherent anti-Stokes Raman scattering) and experimentally for CARS.[9] Laser bandwidth effects were observed qualitatively in excited-state DFWM primarily as an increase in the effective saturation intensity over that expected for a monochromatic field.[10] Saturation effects in DFWM have also been studied in the coherent case.[11,12]

Calculation of the saturation behavior of a medium consisting of two-level atoms is conveniently done using a density matrix approach. The nonlinear response of the medium to two strong pump waves interacting with a peak probe wave in four-wave mixing is described by the saturated susceptibility. Such a calculation has been presented by Boyd *et al.*[13] Their results are consistent with the theory of Abrams and Lind[2] in which the susceptibility was derived in a nonperturbative calculation which included the strong field amplitudes to all orders but the weak probe and reflected fields to only first order.

The calculations of Ref. 13 did not treat the case of intense counterpropagating beams. However, it is exactly this situation which often prevails in DFWM and leads to the complications of standing waves and associated spatial variations in the field amplitudes. The polarization responsible for the reflected wave must be correctly spatially averaged and this modifies the response significantly at intensities which exceed the saturation intensity I_{sat}.[2,3] When the driving laser fields have stochastic fluctuations then the atomic density matrix elements also acquire stochastic variations. The resulting atom-field variables responsible for the radiated fourth wave must then be averaged over both the spatial variations and the field fluctuations. The general solution of this problem for arbitrary intensities and arbitrary bandwidths is a formidable mathematical problem since, due to the inevitable scattering between modes of the fields, an infinite hierarchy of coupled equations is involved. However, in some special cases it is possible to obtain an analytic expression for the mean intensity of the reflected wave.

In this paper we consider DFWM for the case of intense, broadband pump waves interacting with a weak monochromatic probe. We begin in Sec. II by defining the basic equations of motion for our model for coherent pump fields and secondly for incoherent or fluctuating pump fields. We will assume that the pump wave fluctuations can be described by a chaotic field.[14] In Sec. III we derive an expression for the DFWM reflectivity in the limit of very-broad-bandwidth pumps where, in spite of the short timescale of the associated fluctuations, we allow for saturation effects in the medium. We conclude in Sec. IV with a discussion of the broad-bandwidth results by comparing them with those obtained in the monochromatic or coherent case.

II. BASIC EQUATIONS

A. Coherent case

We consider a typical four-wave-mixing geometry as illustrated in Fig. 1. Two counterpropagating pump beams of the same frequency ω_1 and field strengths $\mathscr{E}_1(\mathbf{x},t)$ and $\mathscr{E}_2(\mathbf{x},t)$ traverse a medium composed of two-level atoms. The pump frequency is resonant with the transition from the ground state $|g\rangle$ of energy E_g to the excited state $|e\rangle$ of energy $E_e \approx E_g + \hbar\omega_1$. Neglecting collisional mixing of rotationally degenerate atomic substrates, the atoms may be modeled by such a two-level system with a dipole moment $\langle e|\mu|g\rangle$. A weak probe beam of frequency ω_3 and wave vector $\mathbf{k}_3$ crosses the pumped region at a small angle and the nonlinear coupling generates a fourth wave at frequency $\omega_4 = 2\omega_1 - \omega_3$ and with a wave vector $\mathbf{k}_4 = -\mathbf{k}_3$. This generated wave is the phase conjugate of the probe beam of amplitude $\mathscr{E}_3(\mathbf{x},t)$.

The interaction of the gas atoms with the classical electromagnetic field produced by the three laser fields,

$$\mathbf{E}(\mathbf{x},t) = \sum_{j=1}^{3} \mathscr{E}_j(\mathbf{x},t) e^{-i(\omega_j t - \mathbf{k}_j \cdot \mathbf{x})} + \text{c.c.} , \tag{1}$$

may be described within the dipole approximation. $\omega_2 = \omega_1, \omega_3$ and $\mathbf{k}_2 = -\mathbf{k}_1, \mathbf{k}_3$ are the frequencies and wave vectors, respectively, of the coherent laser fields and $\mathscr{E}_j(\mathbf{x},t)$, $j = 1$, 2, and 3, are their slowly varying electric field amplitudes. In the case considered here we have $\mathscr{E}_1(\mathbf{x},t) = \mathscr{E}_2(x,t)$ and a standing wave is set up by the counterpropagating pump fields.

We consider the interaction of the atoms with the arbitrarily strong laser fields $(\omega_1, \mathbf{k}_1)$ and $(\omega_1, -\mathbf{k}_1)$ up to all orders in the fields (within the rotating-wave approximation) and treat the influence of the weak probe-laser field $(\omega_3, \mathbf{k}_3)$ on the atomic dynamics in lowest-order perturbation theory. We find with the method outlined by Georges *et al.*[15] for the macroscopic density operator $\rho(\mathbf{x},t)$ the following system of equations:

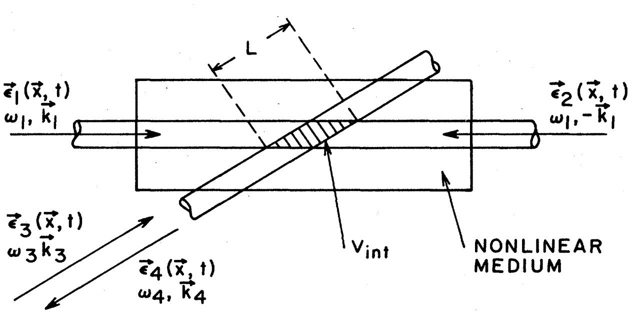


FIG. 1. Geometry for DFWM. L is the length of the interaction volume V_{int} in the direction of the generated wave $\mathscr{E}_4(\mathbf{x},t)$. $\mathscr{E}_1(\mathbf{x},t)$ and $\mathscr{E}_2(\mathbf{x},t)$ are the counterpropagating pump beams and $\mathscr{E}_3(\mathbf{x},t)$ is the input probe beam.

$$\begin{aligned}
&\left[\frac{d}{dt} + \kappa\right](\rho_{gg}^{(0,0)} - \rho_{ee}^{(0,0)})(\mathbf{x},t) = \kappa - 2\,\mathrm{Im}[\Omega^*(\mathbf{x})\rho_{eg}^{(1,0)}(\mathbf{x},t)] ,\\
&\left[\frac{d}{dt} - i\Delta + \frac{\kappa}{2} + \gamma\right]\rho_{eg}^{(1,0)}(\mathbf{x},t) = i\tfrac{1}{2}\Omega(\mathbf{x})(\rho_{gg}^{(0,0)} - \rho_{ee}^{(0,0)})(\mathbf{x},t) ,\\
&1 = \rho_{gg}^{(0,0)}(\mathbf{x},t) + \rho_{ee}^{(0,0)}(\mathbf{x},t) ,\\
&\left[\frac{d}{dt} + i(\Delta - \delta) + \frac{\kappa}{2} + \gamma\right]\rho_{ge}^{(0,-1)}(\mathbf{x},t)\\
&\qquad\qquad = -\frac{i}{2}\Omega_3^*(\rho_{gg}^{(0,0)} - \rho_{ee}^{(0,0)})(\mathbf{x},t) ,\\
&\left[\frac{d}{dt} - i\delta + \kappa\right](\rho_{gg}^{(1,-1)} - \rho_{ee}^{(1,-1)})(\mathbf{x},t)\\
&\qquad\qquad = i\Omega^*(\mathbf{x})\rho_{eg}^{(2,-1)}(\mathbf{x},t) - i\Omega(\mathbf{x})\rho_{ge}^{(0,-1)}(\mathbf{x},t)\\
&\qquad\qquad\quad + i\Omega_3^*\rho_{eg}^{(1,0)}(\mathbf{x},t) ,\\
&\left[\frac{d}{dt} - i(\Delta + \delta) + \frac{\kappa}{2} + \gamma\right]\rho_{eg}^{(2,-1)}(\mathbf{x},t)\\
&\qquad\qquad = i\tfrac{1}{2}\Omega(\mathbf{x})(\rho_{gg}^{(1,-1)} - \rho_{ee}^{(1,-1)})(\mathbf{x},t) ,
\end{aligned} \tag{2}$$

where we have defined the density matrix elements $\rho_{ij}^{(n,m)}(\mathbf{x},t)$, $i,j = g,e$, which are slowly varying in time, by

$$\rho(\mathbf{x},t) = \sum_{n,m=-\infty}^{\infty} \rho^{(n,m)}(\mathbf{x},t) e^{-in\omega_1 t} e^{-im(\omega_3 t - \mathbf{k}_3 \cdot \mathbf{x})} . \tag{2$'$}$$

$\Delta = (E_g + \hbar\omega_1 - E_e)/\hbar$ is the detuning of the counterpropagating pump beams from the atomic transition frequency. $\delta = \omega_1 - \omega_3$ is the relative detuning of the weak probe-laser frequency from ω_1. κ and γ are decay rates due to spontaneous emission and collisional dephasing processes. $\Omega_3 = (2/\hbar)\langle e|\mu|g\rangle \cdot \mathscr{E}_3$ is the Rabi frequency due to the third probing laser where we assume a space- and time-independent electric field amplitude. The Rabi frequency due to the coherent pump beams taking account of the standing-wave nature of the fields is

$$\Omega(\mathbf{x}) = \frac{2}{\hbar}\langle e|\mu|g\rangle \cdot 2\mathscr{E}_1 \cos(\mathbf{k}_1 \cdot \mathbf{x}) . \tag{2$''$}$$

The pump amplitude $\mathscr{E}_1$ is also assumed to be time and space independent. The spatial dependence of this Rabi frequency on the length scale $1/|\mathbf{k}_1|$ in the direction of the pump beams leads to a similar spatial dependence of the density matrix equations. In Eqs. (2) loss mechanisms such as ionization as well as effects due to the motions of the atoms in the medium are neglected.

The propagation of the electromagnetic field of Eq. (1) through the gaseous medium would in general be described by Maxwell's equations with macroscopic polarizations determined by the density matrix Eqs. (2).[2,3] In the following we shall restrict ourselves to the case of optically thin conditions so that all kinds of pulse propagation effects due to amplitude or phase changes may be neglected. In this situation Eqs. (2) must be solved with the initial conditions

$$\rho_{i,j}(\mathbf{x}, t=0)=\delta_{ig}\delta_{jg}, \quad i,j=g,e \ . \tag{2'''}$$

In the case of coherent laser fields, which we have been considering so far, the intensity of the generated fourth wave is given in the stationary limit by[2,3]

$$I_4 \propto \int_{V_{\rm int}} d^3x\, d^3x' \rho_{eg}^{(2,-1)}(\mathbf{x}, t\rightarrow\infty) \times[\rho_{eg}^{(2,-1)}(\mathbf{x}', t\rightarrow\infty)]^* \ . \tag{3}$$

In the integration over the region $V_{\rm int}$, $\mathbf{x}$ and $\mathbf{x}'$ represent the positions of only those atoms such that $\mathbf{x}-\mathbf{x}'$ is parallel to $\mathbf{k}_4$. Thus, this integral reduces to two one-dimensional integrals over a length L, as shown in Fig. 1. The spatial integrations involved in formula (3) can result in important effects. In particular, if the atoms interact with an intense standing wave, which induces a spatial variation of the density matrix equations on a length scale of order $1/|\mathbf{k}_1|$, a significant modification is made to the phase-conjugate reflectivity. Using Eqs. (2) and (3) we recover the usual results.[2,3]

B. Incoherent case

In this subsection we investigate the influence of laser fluctuations on the mean generated intensity $\langle I_4\rangle$. In particular, we shall concentrate on the case in which the counterpropagating laser beams $(\omega_1,\mathbf{k}_1)$ and $(\omega_1,-\mathbf{k}_1)$ exhibit amplitude fluctuations, which may be described by a chaotic field. The weak probe beam $(\omega_3,\mathbf{k}_3)$ is still assumed to be coherent. Our observable of interest is then given by

$$\langle I_4\rangle \propto \int_{V_{\rm int}} d^3x\, d^3x' \langle \rho_{eg}^{(2,-1)}(\mathbf{x}, t\rightarrow\infty) \times[\rho_{eg}^{(2,-1)}(\mathbf{x}', t\rightarrow\infty)]^* \rangle \ , \tag{4}$$

where $\langle\ \rangle$ represents the averaging over the fluctuations of the pump beams. It is the simultaneous averaging over laser fluctuations and the interaction region which complicates the theoretical treatment of laser fluctuations and gives rise to the kind of effects discussed in this paper.

We consider two counterpropagating multimode laser beams with mean frequency ω_1, constant directions of propagation $\mathbf{k}_1/|\mathbf{k}_1|$ and $-\mathbf{k}_1/|\mathbf{k}_1|$, the same polarization and randomly varying phases. Assuming that the statistics of both fluctuating laser fields are not affected by the propagation through the medium and that the interaction length L' in the direction $\mathbf{k}_1/|\mathbf{k}_1|$ is small in the sense $L'<c/nb$, where b is the bandwidth of the laser fields, the corresponding electromagnetic field is given by

$$\mathbf{E}(\mathbf{x},t)=\sum_{\mu}^{\infty}(\mathcal{E}_\mu^r e^{-i\phi_\mu^r}e^{i\mathbf{k}_1\cdot\mathbf{x}}+\mathcal{E}_\mu^l e^{-i\phi_\mu^l}e^{-i\mathbf{k}_1\cdot\mathbf{x}}) \times\mathbf{e}e^{-i(\varpi_\mu-\omega_1)t}e^{-i\omega_1 t}+\text{c.c.} \tag{5}$$

with $|\mathbf{k}_1|=n\omega_1/c$ and the refractive index n. We want to emphasize that the restriction to a small interaction length L', as just mentioned, allows us to neglect all retardation effects, i.e., we may say

$$\exp[\pm i(\varpi_\mu-\omega_1)n\mathbf{k}_1\cdot\mathbf{x}/c\,|\mathbf{k}_1|]\approx 1 \ .$$

$\mathcal{E}_\mu^{r,l}$ and $\phi_\mu^{r,l}$ are the real electric field amplitudes and phases of mode μ with frequency ϖ_μ, polarized in the direction $\mathbf{e}$, which are propagating to the right (r) or left (l). According to Eq. (5) the fluctuating electromagnetic field is considered to consist of an infinite number of modes propagating in opposite directions. Phases associated with different modes μ as well as different propagation directions are assumed to be uncorrelated, i.e.,

$$\langle\phi_\mu^i\phi_\nu^j\rangle=0, \quad i,j=r,l \ , \tag{5'}$$

and equally distributed within the interval $[0,2\pi]$ in each mode. Equation (5) then describes two counterpropagating uncorrelated chaotic fields.[14]

Solving the density matrix Eqs. (2) with the Rabi frequency of Eq. (2″) replaced by

$$\Omega(\mathbf{x},t)=\frac{2}{\hbar}\langle e|\boldsymbol{\mu}\cdot\mathbf{e}|g\rangle \times\sum_\mu(\mathcal{E}_\mu^r e^{-i\phi_\mu^r}e^{i\mathbf{k}_1\cdot\mathbf{x}}+\mathcal{E}_\mu^l e^{-i\phi_\mu^l}e^{-i\mathbf{k}_1\cdot\mathbf{x}}) \times e^{-i(\varpi_\mu-\omega_1)t} \tag{6}$$

and taking the averages in Eq. (4) yields the mean intensity of the generated fourth wave in the presence of fluctuating counterpropagating laser beams. For arbitrary bandwidths and intensities of the electromagnetic field of Eq. (5) this task is complicated. However, for some limiting cases $\langle I_4\rangle$ may be calculated analytically. In the following we shall concentrate on such a case, where although the correlation time of the fluctuations is very short we allow for saturation of the atomic transition from $|g\rangle$ to $|e\rangle$.

III. LARGE-BANDWIDTH SOLUTION

In this section we outline the calculation of the mean generated intensity $\langle I_4\rangle$ in the case of two counterpropagating uncorrelated multimode laser fields [see Eqs. (5) and (5′)] with large bandwidths interacting with a coherent weak probe beam. In particular, we make the following assumptions.

(1) The left and right propagating fluctuating fields of mean frequency ω_1 have the same mean intensity, i.e., $\sum_\mu|\mathcal{E}_\mu^r|^2=\sum_\mu|\mathcal{E}_\mu^l|^2$ and a Lorentzian spectrum with bandwidth b.

(2) The bandwidth is larger than all other rates determining the time evolution of the atoms within the medium, i.e.,

$$b \gg [\langle|\Omega(\mathbf{x},t)|^2\rangle]^{1/2},\kappa,\gamma$$

with $\Omega(\mathbf{x},t)$ given by Eq. (6).

In the long-time limit $(t\gg 1/\kappa)$ the quantity $\rho_{eg}^{(2,-1)}(\mathbf{x},t)[\rho_{eg}^{(2,-1)}(\mathbf{x}',t)]^*$ can be expressed in terms of the product of population differences

$$(\rho_{gg}^{(0,0)}-\rho_{ee}^{(0,0)})(\mathbf{x},t)(\rho_{gg}^{(0,0)}-\rho_{ee}^{(0,0)})(\mathbf{x}',t)$$

[see Eq. (A3) of Appendix A]. Due to the large bandwidth b we are considering here in the calculation of $\langle \rho_{eg}^{(2,-1)}(\mathbf{x},t)[\rho_{eg}^{(2,-1)}(\mathbf{x}',t)]^* \rangle$, all field averages which vary on the rapid timescale $1/b$ may be decorrelated from the averages of the product of the population differences which vary much more slowly, but not necessarily from products of coherences. This corresponds to the usual decorrelation approximation,[16] which is appropriate for treating interactions of large-bandwidth laser fields with atoms. With the procedure outlined in Appendix A, we finally find, in the long-time limit, the following expression:

$$\langle \rho_{eg}^{(2,-1)}(\mathbf{x},t)[\rho_{eg}^{(2,-1)}(\mathbf{x}',t)]^* \rangle = C\frac{\cos^2[\mathbf{k}_1\cdot(\mathbf{x}-\mathbf{x}')]}{\alpha+\cos^2[\mathbf{k}_1\cdot(\mathbf{x}-\mathbf{x}')]}\frac{1}{\beta+\cos^2[\mathbf{k}_1\cdot(\mathbf{x}-\mathbf{x}')]}\ , \qquad (7)$$

with

$$C=|\Omega_3|^2\tfrac{1}{4}\langle|\Omega|^2\rangle^2[J_1+J_2+J_3+J_4+2\,\mathrm{Re}(J_5)+2\,\mathrm{Re}(J_6)] \times\frac{\kappa^2}{\langle|\Omega|^2\rangle^2(b/\kappa)/(\delta^2+b^2)} \times\frac{1}{2\langle|\Omega|^2\rangle^2\{b/[(\kappa/2)+\gamma]\}/(\Delta^2+b^2)}\ ,$$

$$\langle|\Omega|^2\rangle=\left|\frac{2}{\hbar}\langle e|\boldsymbol{\mu}\cdot\mathbf{e}|g\rangle\right|^2\sum_\mu|\mathscr{E}_\mu^r|^2 =\left|\frac{2}{\hbar}\langle 2|\boldsymbol{\mu}\cdot\mathbf{e}|g\rangle\right|^2\sum_\mu|\mathscr{E}_\mu^l|^2$$

and the saturation parameters

$$\alpha=\frac{(\Delta+\delta)^2+[\kappa/2+\gamma+\langle|\Omega|^2\rangle b/(\delta^2+b^2)]^2}{\langle|\Omega|^2\rangle^2(b/\kappa)/(\delta^2+b^2)}\ ,$$

$$\beta=\frac{[\kappa+2\langle|\Omega|^2\rangle b/(\Delta^2+b^2)]^2}{2\langle|\Omega|^2\rangle^2\{b/[(\kappa/2)+\gamma]\}/(\Delta^2+b^2)}\ .$$

The functions J_k, $k=1,2,\ldots,6$, are dependent on κ, γ, and b and the detunings Δ and δ and are given in Eq. (B1) of Appendix B. An interesting feature of Eq. (7) is the fact that, although contrary to the coherent case of Eq. (2″), the mean intensity of the electromagnetic field of Eq. (5) is spatially independent, the mean product of the nonlinear polarizations does show a spatial dependence. This is due to the fact that a fourth-order-in-the-field correlation function, which is no longer spatially independent, determines the mean product of the nonlinear polarizations [see Eq. (A3)]. This spatial dependence becomes extremely important when the saturation parameters α and β become small. In DFWM ($\Delta=\delta=0$) this is the case as soon as $\langle|\Omega^2|\rangle \gtrsim b\kappa$ and the atomic transition begins to saturate.

Carrying out the spatial integrations as in formula (4) we finally find the mean reflectivity R to be given by

$$R=\langle I_4\rangle/I_3 \propto\frac{L^2}{|\Omega_3|^2}\frac{C}{\beta-\alpha}\left[\left[\frac{\beta}{1+\beta}\right]^{1/2}-\left[\frac{\alpha}{1+\alpha}\right]^{1/2}\right]\ , \qquad (8)$$

where the spatial integration over the interaction length $L \gg 1/|\mathbf{k}_1|$ has been done with the help of the approximate relation

$$\int_L dx\,f(x)\approx L\frac{2|\mathbf{k}_1|}{\pi}\int_0^{\pi/2|\mathbf{k}_1|}dx\,f(x)\ , \qquad (8')$$

where $f(x)$ is an arbitrary function. R describes the reflectivity as long as the angle between $\mathbf{k}_1$ and $\mathbf{k}_3$ is small. It is worth noting that in the extreme case where this angle is $\pi/2$, the condition $\mathbf{k}_1\cdot\mathbf{k}_3=0$ implies that $\mathbf{k}_1\cdot(\mathbf{x}-\mathbf{x}')=0$, since only those atoms at $\mathbf{x}$ and $\mathbf{x}'$ such that $\mathbf{x}-\mathbf{x}'$ is parallel to $\mathbf{k}_4=-\mathbf{k}_3$ contribute to the integral in Eq. (4). Thus, in this case the term $\cos^2[\mathbf{k}_1\cdot(\mathbf{x}-\mathbf{x}')]$ in Eq. (7) is equal to unity over the whole integration path.

IV. RESULTS AND DISCUSSION

To distinguish more clearly the effects of large bandwidths of the pump waves, we may compare our results with those of the coherent case. In the absence of field fluctuations the spatially integrated polarization, derived from the steady-state solution for $\rho_{eg}^{(2,-1)}(\mathbf{x},t\to\infty)$ in Eqs. (2), leads to a reflected intensity of the form

$$I_4^c\propto L^2\left|B\frac{1}{\beta_c-\alpha_c}\left[\left[\frac{\beta_c}{1+\beta_c}\right]^{1/2}-\left[\frac{\alpha_c}{1+\alpha_c}\right]^{1/2}\right]\right|^2\ , \qquad (9)$$

where

$$B=\frac{-\frac{1}{2}\bar{\Omega}^2\Omega_3^*[\delta+i(\kappa+2\gamma)]\kappa[\Delta-i(\kappa/2+\gamma)]}{[\Delta-\delta-i(\kappa/2+\gamma)]|\bar{\Omega}|^4(\kappa/2+\gamma)}$$

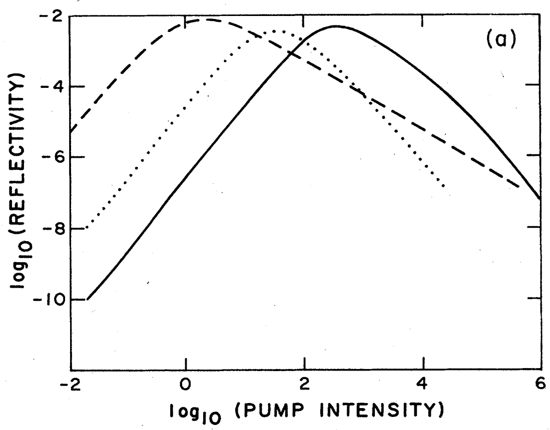
and

$$\alpha_c=-\frac{[\Delta+\delta+i(\kappa/2+\gamma)](\delta+i\kappa)}{\frac{1}{2}|\bar{\Omega}|^2}\ ,$$

$$\beta_c=\frac{\kappa[\Delta^2+(\kappa/2+\gamma)^2]}{|\bar{\Omega}|^2(\kappa/2+\gamma)}\ ,$$

$$\bar{\Omega}=\frac{2}{\hbar}\langle e|\boldsymbol{\mu}|g\rangle\cdot 2\mathscr{E}_1\ .$$

This is equivalent to the result presented by Abrams *et al.*[3] for monochromatic pump waves. The right-hand side (rhs) of Eq. (9) is proportional to the reflectivity and is plotted in Fig. 2(a) as a function of pump intensity. We also plot our results for the broadband pump case based on Eq. (8). We note that for pump intensities below the coherent saturation intensity the reflectivity generated by broadband pump waves is reduced compared to that for monochromatic pumps. This is in contrast to the results of Saxena and Agarwal,[8] who considered the case of pump waves characterized by a narrow-bandwidth chaotic field, where the statistical fluctuations enhanced the re-



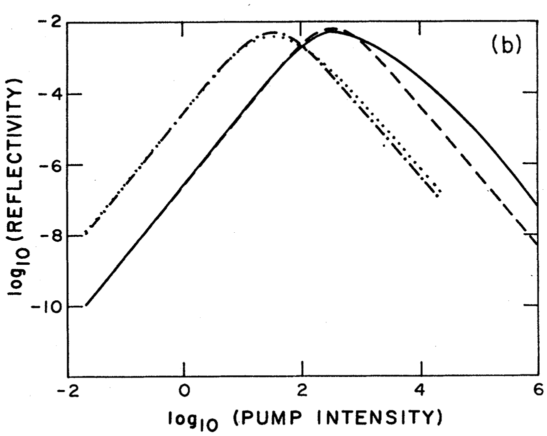


FIG. 2. (a) DFWM reflectivity as a function of pump wave intensity, neglecting atomic motion. (— — —) coherent case; reflectivity given by rhs of Eq. (9) divided by $L^2|\Omega_3|^2$ and the pump intensity by $\frac{1}{2}|\overline{\Omega}|^2$. (· · · · and ——) incoherent case for $b/\kappa=10^2$ and 10^4, respectively; reflectivity given by rhs of Eq. (8) divided by L^2 and the pump intensity by $2\langle|\Omega|^2\rangle$. All quantities are in units of κ and plotted for the case $\kappa=\gamma$. (b) Effects of atomic motion on DFWM for broadband pump fields modeled by replacing $\cos^2[\mathbf{k}_1\cdot(\mathbf{x}-\mathbf{x}')]$ by $\frac{1}{2}$ in Eq. (7). (· · · ·) no motion and (—·—·—·) motion effects included for $b/\kappa=10^2$. (——) no motion and (— — —) motion effects included for $b/\kappa=10^4$. All parameters and units as in (a).

flectivity over the pure coherent case. Our results show clearly that the effective saturation intensity, defined by the maximum in the reflectivity curve in Fig. 2, increases with increasing bandwidth. This is in agreement with the qualitative results of Ewart and O'Leary.[10]

We note that for intensities exceeding the saturation value in the coherent case $R\propto 1/I_1$ whereas for the incoherent, broadband case $R\propto 1/\langle I_1\rangle^2$. It is interesting also that for a forward scattering four-wave-mixing process, where there is no standing-wave field, even for coherent input waves the generated intensity should scale as $1/I_1^2$ when $I_1 \gg I_{\rm sat}$.

Thus far, we have not included the effects of atomic motion. In general for arbitrarily strong, counterpropagating pump beams, and even for the coherent case, this is a complex problem. Bloch and Ducloy have considered the saturation effects on the DFWM line shapes in the case of a Doppler-broadened medium but where only one pump beam is strong.[12] Their treatment and the discussion of atomic-motion effects by Wandzura[17] deal with monochromatic pump and probe beams. Atomic-motion effects in certain limits are also discussed for the coherent case by Abrams *et al.*[3] The essential physical problem is how to describe the interaction of moving atoms with the standing wave set up by the highly correlated counterpropagating pump beams. In the case considered here, although the pump beams are considered to be totally uncorrelated, spatial variation of the saturation behavior cannot be ignored. These spatial saturation effects are described by the terms $\cos^2\mathbf{k}_1\cdot(\mathbf{x}-\mathbf{x}')$ in our solution as given in Eq. (7). In general, the effects of atomic motion are complicated to investigate since we need to average over both space and velocity. However, we may get some insight into the effects of atomic motion by taking the following, heuristic approach. If we consider a time scale which is long compared to an inverse Doppler width (i.e., the mean time to move a wavelength) the motion of the atoms ($\mathbf{x}\rightarrow\mathbf{x}_0+\mathbf{v}t$) effectively washes out the spatial grating structures in the medium. Then the effects may be represented by replacing the spatial saturation terms $\cos^2[\mathbf{k}_1\cdot(\mathbf{x}-\mathbf{x}')]$ in Eq. (7) by a factor of $\frac{1}{2}$. This should be valid when the Doppler width is greater than the spontaneous decay rate κ, since our solution [Eq. (7)] requires times greater than $\sim 1/\kappa$. The results of this process are illustrated in Fig. 2(b) where we see that for $I_1 < I_{\rm sat}$ motional effects are unimportant. However, when $I_1 \gtrsim I_{\rm sat}$, and saturation effects become apparent, the effect of atomic motion is to reduce the reflectivity, as might be expected from a "washing out" process. From Eq. (8) in particular we see that in the limit of large b and for very strong fields, $I_1 \gg I_{\rm sat}$ ($|\alpha|,\beta \ll 1$), the reflectivity for all beams on resonance ($\Delta=\delta=0$) has the form

$$R\propto L^2C\frac{1}{\sqrt{\kappa/b}+\sqrt{(\kappa+2\gamma)/b}}\ .$$

If we neglect the spatial saturation terms, equivalent to including atomic motion, then we have

$$R\propto 2L^2C\ ;$$

we therefore find that including the Doppler effect would reduce the reflectivity by a factor of $\sim\sqrt{\kappa/b}+\sqrt{(\kappa+2\gamma)/b}$.

The frequency dependence of DFWM in the case considered here will be determined by tuning the coherent probe frequency ω_3 through the resonance whilst keeping the strong pump beams on resonance, i.e., $\Delta=0$. In Fig. 3(a) we plot the line shape in the coherent case for various pump beam intensities. We note the three-peaked structure characteristic of ac Stark splitting. Figure 3(b) shows the results obtained from the present calculation. The main feature of our result is simply a power broadening of

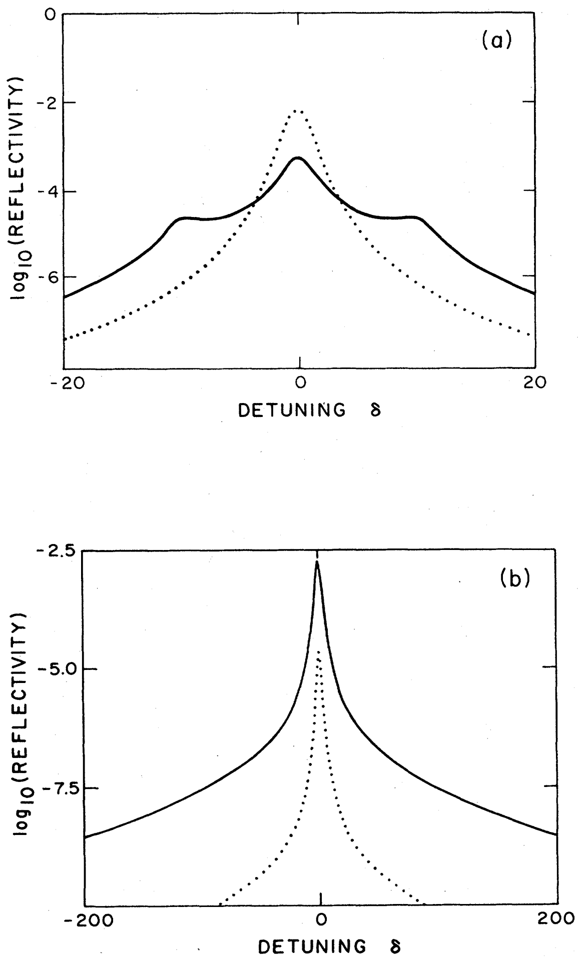


FIG. 3. Line shapes of DFWM for different pump beam intensities for $\Delta=0$ and $\kappa=\gamma$. (a) Coherent case: pump intensity represented by $\frac{1}{2}|\tilde{\Omega}|^2/\kappa^2=1$ (dotted curve) and 10^2 (solid curve). (b) Incoherent case with $b/\kappa=10^2$: pump intensity represented by $2\langle|\Omega|^2\rangle/\kappa^2=1$ (dotted curve) and 10^2 (solid curve). The reflectivities plotted for coherent and incoherent cases are the same as in Fig. 2. The detuning δ is in units of κ.

the line shape and a smearing out of the ac Stark structure since $b \gg \Omega$.

In conclusion, we have calculated the effects of finite laser bandwidth on resonant DFWM in the limit of the bandwidth exceeding all other atomic relaxation rates. We have treated explicitly the case of uncorrelated, counterpropagating pump beams and have shown that even in this situation, spatial saturations effects play an important role for intensities which exceed the effective saturation intensity. Our results show how this effective saturation intensity increases with increasing bandwidth. We have shown also that the spectral behavior of the resonant DFWM process is modified for finite bandwidth pump beams so that ac Stark splitting is masked by the power broadening of the medium's response. It is a straightforward matter to apply our method of calculation to other four-wave-mixing processes, such as CARS where the complications of counterpropagating beams, encountered in this work, are absent.

A quantitative test of our results would require a variable bandwidth laser source which could be characterized by a chaotic field and a monochromatic probe field, say, from a single-mode laser. In principle, a broadband probe field should provide qualitatively similar behavior to that predicted by the present analysis. In this case one would expect the response to a broadband probe to be described in lowest order by a convolution of the coherent probe response and the probe-laser line shape. Ideally, the interaction should be studied in a steady-state situation, i.e., with cw lasers (or with a laser with pulse duration much larger than $1/\kappa$) resonant with a nondegenerate two-level atom.

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APPENDIX A

In this appendix we outline the derivation of Eq. (7). Laplace transforming the density matrix Eqs. (2) we find with $\omega_\mu \equiv \bar{\omega}_\mu - \omega_1$

$$\left[z+\Delta+\delta+i\left(\frac{\kappa}{2}+\gamma\right)\right]\rho_{eg}^{(2,-1)}(\mathbf{x},z)-\frac{1}{2}\sum_{\mu,\nu}\frac{\tilde{\Omega}_\mu(\mathbf{x})\tilde{\Omega}_\nu^*(\mathbf{x})}{z-\omega_\mu+\delta+i\kappa}\rho_{eg}^{(2,-1)}(\mathbf{x},z-\omega_\mu+\omega_\nu)$$

$$=-\frac{1}{4}\Omega_3^*\sum_{\mu,\nu}\left[\frac{\tilde{\Omega}_\mu(\mathbf{x})}{z-\omega_\mu+\delta+i\kappa}\,\frac{\tilde{\Omega}_\nu(\mathbf{x})}{z-\omega_\mu-\omega_\nu-\Delta+\delta+i\left(\frac{\kappa}{2}+\gamma\right)}+\frac{\tilde{\Omega}_\mu(\mathbf{x})}{z-\omega_\mu+\delta+i\kappa}\,\frac{\tilde{\Omega}_\nu(\mathbf{x})}{z-\omega_\mu+\Delta+i\left(\frac{\kappa}{2}+\gamma\right)}\right]$$

$$\times(\rho_{gg}^{(0,0)}-\rho_{ee}^{(0,0)})(\mathbf{x},z-\omega_\mu-\omega_\nu)\,,$$

$$(z+i\kappa)(\rho_{gg}^{(0,0)}-\rho_{ee}^{(0,0)})(\mathbf{x},z)=-\frac{\kappa}{z}+i+i\,\mathrm{Im}\left[\sum_{\mu,\nu}\frac{\tilde{\Omega}_\mu^*(\mathbf{x})\tilde{\Omega}_\nu(\mathbf{x})}{z+\omega_\mu+\Delta+i\left(\frac{\kappa}{2}+\gamma\right)}(\rho_{gg}^{(0,0)}-\rho_{ee}^{(0,0)})(\mathbf{x},z-\omega_\nu+\omega_\mu)\right]\,, \tag{A1}$$

where

$$\rho(\mathbf{x},z)=\int_0^\infty dt\, e^{izt}\rho(\mathbf{x},t)$$

and

$$\tilde{\Omega}_\mu(\mathbf{x})=\frac{2}{\hbar}\langle e\,|\,\mu\,|\,g\rangle\cdot\mathbf{e}(\mathscr{E}^r_\mu e^{-i\phi^r_\mu}e^{+i\mathbf{k}_1\cdot\mathbf{x}}+\mathscr{E}^l_\mu e^{-i\phi^l_\mu}e^{-i\mathbf{k}_1\cdot\mathbf{x}})\;,$$

in accordance with our assumption $L'<c/nb$. As we are interested in the long-time limit we neglect z in comparison with κ everywhere in the denominators of Eq. (A1). Transforming back to the time domain we obtain the expressions ($t\gg 1/\kappa$)

$$\begin{aligned}
&\left[\Delta+\delta+i\left[\frac{\kappa}{2}+\gamma\right]-\tfrac{1}{2}\sum_{\mu,\nu}\frac{\tilde{\Omega}_\mu(\mathbf{x})\tilde{\Omega}^*_\nu(\mathbf{x})}{\delta-\omega_\mu+i\kappa}e^{i(\omega_\nu-\omega_\mu)t}\right]\rho^{(2,-1)}_{eg}(\mathbf{x},t)\\
&\quad=-\tfrac{1}{4}\Omega_3^*\sum_{\mu,\nu}\left[\frac{\tilde{\Omega}_\mu(\mathbf{x})}{\delta-\omega_\mu+i\kappa}\frac{\tilde{\Omega}_\nu(\mathbf{x})}{-\omega_\mu-\omega_\nu-\Delta+\delta+i\left[\frac{\kappa}{2}+\gamma\right]}+\frac{\tilde{\Omega}_\mu(\mathbf{x})}{\delta-\omega_\mu+i\kappa}\frac{\tilde{\Omega}_\nu(\mathbf{x})}{\Delta-\omega_\mu+i\left[\frac{\kappa}{2}+\gamma\right]}\right]\\
&\qquad\times e^{-i(\omega_\mu+\omega_\nu)t}(\rho^{(0,0)}_{gg}-\rho^{(0,0)}_{ee})(\mathbf{x},t)\;,\\
&(\rho^{(0,0)}_{gg}-\rho^{(0,0)}_{ee})(\mathbf{x},t)=\frac{\kappa}{\kappa-\operatorname{Im}\left[\sum_{\mu,\nu}\frac{\tilde{\Omega}^*_\mu(\mathbf{x})\tilde{\Omega}_\nu(\mathbf{x})}{\omega_\mu+\Delta+i\left[\frac{\kappa}{2}+\gamma\right]}e^{i(\omega_\mu-\omega_\nu)t}\right]}\;.
\end{aligned}\tag{A2}$$

From these equations, we derive the following expression for the product of polarizations:

$$\begin{aligned}
&\left\{(\Delta+\delta)^2+\left[\frac{\kappa}{2}+\gamma\right]^2-\tfrac{1}{2}\sum_{\mu,\nu}\frac{\tilde{\Omega}_\mu(\mathbf{x})\tilde{\Omega}^*_\nu(\mathbf{x})}{\delta-\omega_\mu+i\kappa}e^{+i(\omega_\nu-\omega_\mu)t}\left[\Delta+\delta-i\left[\frac{\kappa}{2}+\gamma\right]\right]\right.\\
&\quad-\tfrac{1}{2}\sum_{\mu,\nu}\frac{\tilde{\Omega}^*_\mu(\mathbf{x}')\tilde{\Omega}_\nu(\mathbf{x}')}{\delta-\omega_\mu-i\kappa}e^{+i(\omega_\mu-\omega_\nu)t}\left[\Delta+\delta+i\left[\frac{\kappa}{2}+\gamma\right]\right]\\
&\quad\left.+\tfrac{1}{4}\sum_{\substack{\mu,\nu,\\ \rho,\sigma}}\frac{\tilde{\Omega}_\mu(\mathbf{x})\tilde{\Omega}^*_\nu(\mathbf{x})}{\delta-\omega_\mu+i\kappa}\frac{\tilde{\Omega}^*_\rho(\mathbf{x}')\tilde{\Omega}_\sigma(\mathbf{x}')}{\delta-\omega_\rho-i\kappa}e^{+i(\omega_\nu-\omega_\mu-\omega_\sigma+\omega_\rho)t}\right\}\rho^{(2,-1)}_{eg}(\mathbf{x},t)[\rho^{(2,-1)}_{eg}(\mathbf{x}',t)]^*\\
&=\tfrac{1}{16}\,|\,\Omega_3\,|^2\sum_{\mu,\nu}\tilde{\Omega}_\mu(\mathbf{x})\tilde{\Omega}_\nu(\mathbf{x})\left[\frac{1}{\delta-\omega_\mu+i\kappa}\frac{1}{-\omega_\mu-\omega_\nu-\Delta+\delta+i\left[\frac{\kappa}{2}+\gamma\right]}+\frac{1}{\delta-\omega_\mu+i\kappa}\frac{1}{\Delta-\omega_\mu+i\left[\frac{\kappa}{2}+\gamma\right]}\right]\\
&\qquad\times e^{-i(\omega_\mu+\omega_\nu)t}\sum_{\rho,\sigma}\tilde{\Omega}^*_\rho(\mathbf{x}')\Omega^*_\sigma(\mathbf{x}')\left[\frac{1}{\delta-\omega_\rho-i\kappa}\frac{1}{-\omega_\rho-\omega_\sigma-\Delta+\delta-i\left[\frac{\kappa}{2}+\gamma\right]}\right.\\
&\qquad\qquad\left.+\frac{1}{\delta-\omega_\rho-i\kappa}\frac{1}{\Delta-\omega_\rho-i\left[\frac{\kappa}{2}+\gamma\right]}\right]\\
&\qquad\times e^{+i(\omega_\rho+\omega_\sigma)t}(\rho^{(0,0)}_{gg}-\rho^{(0,0)}_{ee})(\mathbf{x},t)(\rho^{(0,0)}_{gg}-\rho^{(0,0)}_{ee})(\mathbf{x}',t)\;.
\end{aligned}\tag{A3}$$

Taking on both sides of this equation the average over the fluctuations of the pump-laser fields and decorrelating all field averages from $\rho^{(2,-1)}_{eg}(\mathbf{x},t)[\rho^{(2,-1)}_{eg}(\mathbf{x}',t)]^*$ and $(\rho^{(0,0)}_{gg}-\rho^{(0,0)}_{ee})(\mathbf{x},t)(\rho^{(0,0)}_{gg}-\rho^{(0,0)}_{ee})(\mathbf{x}',t)$ according to the large-bandwidth limit yields an expression for the mean value $\langle\rho^{(2,-1)}_{eg}(\mathbf{x},t)[\rho^{(2,-1)}_{eg}(\mathbf{x}',t)]^*\rangle$. In the evaluation of the remaining field averages we take into account the fact that left- and right-propagating waves as well as different modes are uncorrelated.

The fifth term on the left-hand side of Eq. (A3) is, for example, evaluated as follows. Taking the average over the expression of fourth order in the field, both terms with $\mu=\rho$ and $\nu=\sigma$ and $\mu=\nu$ and $\rho=\sigma$ are nonvanishing, as different modes are uncorrelated. We therefore have to consider, for example, terms of the form

$$\langle \tilde{\Omega}_\mu(\mathbf{x})\tilde{\Omega}^*_\mu(\mathbf{x}')\rangle = \left|\frac{2}{\hbar}\langle e\,|\boldsymbol{\mu}|\,g\rangle\cdot\mathbf{e}\right|^2 (\mathcal{E}^r_\mu\mathcal{E}^r_\mu e^{i\mathbf{k}_1\cdot(\mathbf{x}-\mathbf{x}')}+\mathcal{E}^l_\mu\mathcal{E}^l_\mu e^{-i\mathbf{k}_1\cdot(\mathbf{x}-\mathbf{x}')}$$
$$+\mathcal{E}^r_\mu\mathcal{E}^l_\mu\langle e^{-i(\phi^r_\mu-\phi^l_\mu)}\rangle e^{i\mathbf{k}_1\cdot(\mathbf{x}+\mathbf{x}')}+\mathcal{E}^l_\mu\mathcal{E}^r_\mu\langle e^{-i(\phi^l_\mu-\phi^r_\mu)}\rangle e^{-i\mathbf{k}_1\cdot(\mathbf{x}+\mathbf{x}')})\,. \tag{A4}$$

The third and fourth terms involved in the averaging over the uncorrelated phases of left- and right-propagating waves average to zero. As we are assuming a Lorentzian spectrum of bandwidth b and the same intensities for the left- and right-propagating pump waves [see assumption (1) of Sec. III], we further replace expressions such as

$$\sum_\mu [(\mathcal{E}^r_\mu)^2 e^{i\mathbf{k}_1\cdot(\mathbf{x}-\mathbf{x}')}+(\mathcal{E}^l_\mu)^2 e^{-i\mathbf{k}_1\cdot(\mathbf{x}-\mathbf{x}')}]f(\omega_\mu) \tag{A5}$$

by

$$2\cos[\mathbf{k}_1\cdot(\mathbf{x}-\mathbf{x}')]\sum_\mu (\mathcal{E}^r_\mu)^2\frac{1}{\pi}\int_{-\infty}^{\infty}dz\frac{b}{z^2+b^2}f(z)$$

for arbitrary functions f. In this way we finally arrive at Eq. (7).

APPENDIX B

The functions J_k, $k=1,2,\ldots,6$, of Eq. (7) are given by

$$J_1=\left[\frac{1}{\pi}\right]^2\int_{-\infty}^{\infty}dz\,dw\frac{b}{z^2+b^2}\frac{b}{w^2+b^2}\frac{1}{(z-\delta)^2+\kappa^2}\times\frac{1}{(z+w+\Delta-\delta)^2+\left[\frac{\kappa}{2}+\gamma\right]^2}\,,$$

$$J_2=\frac{1}{\pi}\int_{-\infty}^{\infty}dz\frac{b}{z^2+b^2}\frac{1}{(z-\delta)^2+\kappa^2}\times\frac{1}{(z-\Delta)^2+\left[\frac{\kappa}{2}+\gamma\right]^2}\,,$$

$$J_3=\left|\frac{1}{\pi}\int_{-\infty}^{\infty}dz\frac{b}{z^2+b^2}\frac{1}{z-\delta-i\kappa}\frac{1}{z-\Delta-i\left[\frac{\kappa}{2}+\gamma\right]}\right|^2\,, \tag{B1}$$

$$J_4=\left[\frac{1}{\pi}\right]^2\int_{-\infty}^{\infty}dz\,dw\frac{b}{z^2+b^2}\frac{1}{w^2+b^2}\times\frac{1}{z-\delta-i\kappa}\frac{1}{w-\delta+i\kappa}\times\frac{1}{(z+w+\Delta-\delta)^2+\left[\frac{\kappa}{2}+\gamma\right]^2}\,,$$

$$J_5=\left[\frac{1}{\pi}\right]^2\int_{-\infty}^{\infty}dz\,dw\frac{b}{z^2+b^2}\frac{b}{w^2+b^2}\frac{1}{z-\delta-i\kappa}\times\frac{1}{z+w+\Delta-\delta-i\left[\frac{\kappa}{2}+\gamma\right]}\frac{1}{z-\delta+i\kappa}\times\frac{1}{z-\Delta+i\left[\frac{\kappa}{2}+\gamma\right]}\,,$$

$$J_6=\left[\frac{1}{\pi}\right]^2\int_{-\infty}^{\infty}dz\,dw\frac{b}{z^2+b^2}\frac{b}{w^2+b^2}\frac{1}{z-\delta-i\kappa}\times\frac{1}{z+w+\Delta-\delta-i\left[\frac{\kappa}{2}+\gamma\right]}\frac{1}{w-\delta+i\kappa}\times\frac{1}{w-\Delta+i\left[\frac{\kappa}{2}+\gamma\right]}\,.$$

The evaluation of these integrals is straightforward but tedious. We shall not present the analytical results here as they are rather involved.

*On leave from University of Oxford, Oxford OX1 3NP, United Kingdom.

[1]*Optical Phase Conjugation*, edited by R. A. Fisher (Academic, New York, 1983).

[2]R. L. Abrams and R. C. Lind, Opt. Lett. 2, 94 (1978); 3, 205 (1978).

[3]R. L. Abrams, J. F. Lam, R. C. Lind, D. G. Steel, and P. F. Liao, in Ref. 1, p. 211.

[4]B. R. Mollow, Phys. Rev. **175**, 1555 (1968).
[5]G. S. Agarwal, Phys. Rev. A **1**, 1445 (1970).
[6]H. J. Kimble and L. Mandel, Phys. Rev. A **15**, 689 (1977).
[7]P. Zoller and P. Lambropoulos, J. Phys. B **13**, 69 (1980).
[8]R. Saxena and G. S. Agarwal, Opt. Lett. **8**, 566 (1983).
[9]L. A. Rahn, R. L. Farrow, and R. P. Lucht, Opt. Lett. **9**, 223 (1984); N. K. Dutta, J. Phys. B **13**, 411 (1980); G. S. Agarwal and J. Cooper, Phys. Rev. A **26**, 2761 (1982).
[10]P. Ewart and S. V. O'Leary, J. Phys. B (to be published).
[11]J. P. Woerdman and M. F. H. Schuurmans, Opt. Lett. **6**, 239 (1981).
[12]D. Bloch and M. Ducloy, J. Opt. Soc. Am. **73**, 635 (1983); **73**, 1844 (1983).
[13]R. W. Boyd, M. G. Raymer, P. Narum, and D. J. Harter, Phys. Rev. A **24**, 411 (1981).
[14]P. Zoller, G. Alber, and R. Salvador, Phys. Rev. A **24**, 398 (1981), in particular, Chap. II.
[15]A. T. Georges, P. Lambropoulos, and J. H. Marburger, Phys. Rev. A **15**, 300 (1977).
[16]See, for example, P. Agostini, A. T. Georges, S. E. Wheatley, P. Lambropoulos, and M. D. Levenson, J. Phys. B **11**, 1733 (1978).
[17]S. M. Wandzura, Opt. Lett. **4**, 208 (1979).