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Frequency spectrum of the signal wave in resonant four-wave mixing induced by broad-bandwidth lasers

D. R. Meacher, P. G. R. Smith, and P. Ewart
Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX1 3PU, United Kingdom

J. Cooper
Joint Institute for Laboratory Astrophysics, University of Colorado and National Institute of Standards and Technology, Boulder, Colorado 80309
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We develop a theory of near-resonant four-wave mixing induced by broad-bandwidth chaotic fields in a medium composed of two-level atoms. By solving the equations of motion for the elements of the atomic density matrix using an appropriate decorrelation approximation, we derive an analytic expression for the frequency spectrum of the signal wave for the case of the bandwidth of the fluctuations of the pump field exceeding the other relaxation rates in the problem. The probe wave is considered weak and of arbitrary bandwidth. The theory is valid for pump intensities up to and exceeding the bandwidth-dependent saturation value. Finally, we show how the frequency spectrum is modified if account is taken of atomic motion. The theoretical results are of direct relevance to practical experiments involving broad-bandwidth pulsed lasers and employing an atomic vapor as the nonlinear medium.

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I. INTRODUCTION

Recent work has demonstrated the important effects of the stochastic phase and amplitude fluctuations of the driving fields in laser-induced atom-field interactions. Studies have been carried out on the effects of field fluctuations on the saturation and Stark splitting of an atomic resonance, on the spectrum of the resulting resonance fluorescence, and their effects in the related phenomenon of the reversal of the asymmetry of the Autler-Townes doublet in double-optical-resonance-type experiments [1,2]. The effect of a finite laser bandwidth on the shape of Hanlé resonances [3] and on the efficiency of multiphoton absorption and ionization have also been calculated and observed in experiments [4].

Nonlinear parametric processes are also strongly affected by the fluctuations of the driving fields. These include practically important schemes such as coherent anti-Stokes Raman scattering (CARS) and degenerate four-wave mixing (DFWM). The effects of field fluctuations in this area have been reviewed by Reintjes [5]. The effects of pump laser fluctuations on CARS line shapes have been calculated [6], and it has been demonstrated that the fluctuations of the lasers employed can be the limiting factor in the precision of temperature measurements based on CARS spectra [7]. The DFWM reflectivity induced by chaotic pump fields has been calculated for the somewhat idealized case of the laser bandwidth being much less than the atomic linewidth [8]. More recently we developed a theory of FWM induced by broad-bandwidth chaotic fields [9]. We showed that, in contrast to the narrow-bandwidth case, increasing the bandwidth of the pump fluctuations leads, at low intensity, to a reduced reflectivity compared to the coherent case and furthermore to an increased effective saturation intensity for the process. The theory was extended to treat the time-dependent case of FWM induced by broad-bandwidth lasers of arbitrary temporal pulse shape and the theoretical predictions tested in experiments [10] employing sodium vapor as the nonlinear medium and a variable bandwidth dye laser to form the pump waves. In addition, the pulse-shortening effects in DFWM induced by intense broad-bandwidth lasers were investigated.

DFWM has been extensively studied as a method of achieving phase conjugation and as a tool in high-resolution spectroscopy [11]. The Doppler-free nature of the response of a resonant medium in coherent DFWM has been pointed out by a number of authors. This has stimulated investigations of DFWM line shapes in spectroscopic studies in which the laser frequency is tuned through the atomic resonance [12]. DFWM has recently emerged as a potentially useful optical diagnostic technique for combustion studies [13]. Furthermore, it has been demonstrated that the spectrum of the FWM signal in experiments with broad-bandwidth lasers may yield useful spectroscopic data over a wide frequency range [14] in analogy with the technique of multiplex CARS. This opens up the possibility of deriving concentration and temperature measurements from spectra produced by a single laser shot and thereby significantly enhancing the temporal resolution of the technique. Of importance to these broad-bandwidth experiments is the spectral content of the generated radiation rather than its intensity. It is to the calculation of this aspect of the interaction that we address ourselves in this paper. The theoretical model is based on that used in our previous work [9] on the saturation behavior of DFWM induced by broad-bandwidth lasers. In the following section we show how this formalism can be extended to calculate the amplitude autocorrelation of the signal field. The spectrum of the generated radiation is then calculated from a Laplace transform of the autocorrelation function. Finally, we consider the effects of atomic motion on the power spectrum of the reflected radiation.

II. THEORY

We consider the laser fields to interact in the usual near-collinear geometry [10] with an ensemble of two-level atoms with ground- and excited-state energies ε_g and ε_e, respectively, and $\varepsilon_e-\varepsilon_g=\hbar\omega_{eg}$. The pump and probe fields are assumed chaotic, with a Lorentzian spectral distribution and center frequencies ω_1 and ω_3. The bandwidth b of the pump field is larger than any other relaxation rate in the problem: $b \gg |\Omega|, |\Omega_3|, \kappa, \Gamma$, where κ and Γ are the longitudinal and transverse relaxation rates of the atom and $\Omega(x,t)$ and $\Omega_3(x,t)$ are the Rabi frequencies associated with the pump and probe fields, respectively. We treat the cases of probe bandwidth p as zero or large in the sense defined for b above. The probe field is assumed weak, $|\Omega_3| \ll \Gamma$, and we neglect, for the present, the motion of the atoms.

A. Basic equations

The evolution of the atomic density operator is given by the Liouville equation

$$i\hbar\frac{\partial\rho}{\partial t}=[H,\rho]+i\hbar[\mathcal{D},\rho] \;, \tag{1}$$

where

$$H=H_0+\hbar[\Omega(x,t)e^{-i\omega_1 t}+\Omega_3(t)e^{-i\omega_3 t}+\text{c.c.}] \;.$$

To derive the basic equations we assume, for the moment, coherent fields and write

$$\hbar\Omega(x,t)=2\mu_{ge}E_1\cos k_1x, \quad \hbar\Omega_3(t)=2\mu_{ge}E_3 \;. \tag{2}$$

H_0 is the unperturbed atomic Hamiltonian and $\mathcal{D}$ the damping matrix. We expand the density operator as a sum of components at harmonics of the pump and probe field frequencies

$$\rho_{ij}=\sum_{m,n}\rho_{ij}^{mn}e^{-im\omega_1 t}e^{-in(\omega_3 t-k_3x)} \tag{3}$$

and obtain, on substituting into (1), the following set of equations

$$\left[\frac{d}{dt}+\kappa\right]\rho_0(x,t)=\kappa-2\,\mathrm{Im}[\Omega^*(x,t)\rho_1(x,t)] \;, \tag{4a}$$

$$\left[\frac{d}{dt}+\beta\right]\rho_1(x,t)=\frac{i}{2}[\Omega(x,t)\rho_0(x,t)+\Omega_3(x,t)\rho_4(x,t)] \;, \tag{4b}$$

$$\left[\frac{d}{dt}+\beta^*\right]\rho_2(x,t)=\frac{-i}{2}[\Omega^*(x,t)\rho_0(x,t)+\Omega_3^*(x,t)\rho_4(x,t)] \;, \tag{4c}$$

$$\left[\frac{d}{dt}+c\right]\rho_3(x,t)=\frac{-i}{2}[\Omega_3^*(x,t)\rho_0(x,t)+\Omega^*(x,t)\rho_4(x,t)] \;, \tag{4d}$$

$$\left[\frac{d}{dt}+d\right]\rho_4(x,t)=i[\Omega^*(x,t)\rho_5(x,t)-\Omega(x,t)\rho_3(x,t)+\Omega_3^*(x,t)\rho_1(x,t)] \;, \tag{4e}$$

$$\left[\frac{d}{dt}+a\right]\rho_5(x,t)=\frac{i}{2}\Omega(x,t)\rho_4(x,t) \;, \tag{4f}$$

where we have written

$$\begin{aligned}
&\rho_0(x,t)\equiv\rho_{gg}^{0,0}(x,t)-\rho_{ee}^{0,0}(x,t) \;,\\
&\rho_1(x,t)\equiv\rho_{eg}^{1,0}(x,t) \;,\\
&\rho_2(x,t)\equiv[\rho_{eg}^{1,0}(x,t]^* \;,\\
&\rho_3(x,t)\equiv\rho_{ge}^{0,-1}(x,t) \;,\\
&\rho_4(x,t)\equiv\rho_{gg}^{1,-1}(x,t)-\rho_{ee}^{1,-1}(x,t) \;,\\
&\rho_5(x,t)\equiv\rho_{eg}^{2,-1}(x,t)
\end{aligned}$$

and

$$\begin{aligned}
&a=\Gamma+i\Delta_4 \;,\\
&\beta=\Gamma-i\Delta \;,\\
&c=\Gamma+i\Delta_3 \;,\\
&d=\kappa-i\delta \;,\\
&\Delta_3=\Delta-\delta \;,\\
&\Delta_4=\Delta+\delta
\end{aligned}$$

with $\Gamma=\kappa/2+\gamma$ and γ is the collisional dephasing rate. Δ is the detuning of the pump waves, $\Delta=-(\varepsilon_e-\varepsilon_g-\hbar\omega_1)/\hbar$, and δ the pump-probe detuning given by $\delta=\omega_1-\omega_3$.

To derive an expression for the amplitude of the generated wave, we express the induced polarization P as

$$P=\sum_{m,n}P^{mn}e^{-im\omega_1 t}e^{-in(\omega_3 t-k_3x)}$$

and substitute this in the nonlinear wave equation. Picking out terms oscillating at frequency $2\omega_1-\omega_3$ we obtain, in the slowly varying envelope approximation, for the amplitude of the signal wave

$$A_4(t)=\frac{2\omega_1-\omega_3}{\sqrt{2}\epsilon_0 c}\int_0^l dx\, P^{2,-1}(x,t) \;, \tag{5}$$

where l is the length of the interaction zone. The positive frequency part of the induced polarization is given in terms of the atomic density operator by

$$P=N\mu_{ge}\rho_{eg} \;,$$

where N is the number density of interacting atoms and μ_{ge} the electric dipole matrix element connecting the atomic ground and excited states. Thus

$$A_4(t)=\frac{(2\omega_1-\omega_3)N}{\sqrt{2}\epsilon_0 c}\int_0^l dx\, \mu_{ge}\rho_5(x,t)e^{-i(2\omega_1-\omega_3)t}\ . \tag{6}$$

The intensity of the generated wave is

$$I_4(t)=\frac{(2\omega_1-\omega_3)^2|\mu_{eg}|^2N^2}{2\epsilon_0 c}\int_0^l\int_0^l dx\, dx'\langle\rho_5(x,t)\rho_5(x',t)\rangle\ , \tag{7}$$

where now we assume fluctuating fields and the angle brackets represent an average over the spatial and temporal variations of the induced polarization caused by the stochastic driving fields.

The power spectrum of the signal wave is found from a Laplace transform, $\mathcal{L}$, with respect to τ of the amplitude autocorrelation function:

$$\begin{aligned}P(\omega)&=\mathrm{Re}\left[\mathcal{L}\frac{\langle A_4(t+\tau)A_4^*(t)\rangle}{\langle|A_4(t)|^2\rangle}\right]\\&=\mathrm{Re}\frac{\int_0^l\int_0^l dx\,dx'\mathcal{L}\{\langle\rho_5(x,t+\tau)[\rho_5(x',t)]^*\rangle e^{-i(2\omega_1-\omega_3)\tau}\}}{\int_0^l\int_0^l dx\,dx'\langle\rho_5(x,t)[\rho_5(x',t)]^*\rangle}\ .\end{aligned} \tag{8}$$

The first step in evaluating (8) involves calculating an expression for $\rho_5(x,t)$ from (2) and forming and averaging the product $[\rho_5(x',t)]^*\rho_5(x,t)$. In Ref. [9] we showed that this product is given by integral expressions of the form

$$\langle[\rho_5(x',t)]^*\rho_5(x,t)\rangle=e^{-2\Gamma t}\int_{-\infty}^{t}\int_{-\infty}^{t}dt_1dt_2\langle\Omega^*(x,'t_1)\Omega(t_2)\rho_4(x',t_1)\rho_4(x,t_2)\rangle e^{-\beta^*t_1-\beta t_2}\ ,$$

where

$$\begin{aligned}\rho_4(t)+\tfrac{1}{2}&\left[\int_{-\infty}^{t}\int_{-\infty}^{t_1}dt_1dt_2e^{-\alpha(t-t_1)}[e^{-\beta_3(t_1-t_2)}\Omega^*(t_1)\Omega(t_2)+e^{-\beta_2^*(t_2-t_1)}\Omega(t_1)\Omega^*(t_2)]\rho_4(t_2)\right]\\&=-\frac{i}{2}\left[\int_{-\infty}^{t}\int_{-\infty}^{t_1}dt_1dt_2e^{-\alpha(t-t_1)}[e^{-\beta(t-t_1)}\Omega_3^*\Omega(t_2)+e^{-\beta_2^*(t-t_1)}\Omega_3^*\Omega(t_1)]\rho_0(t_2)\right]\ ,\\\rho_0(t)=1-\mathrm{Re}&\left[\int_{-\infty}^{t}\int_{-\infty}^{t}dt_1dt_2e^{-\beta(t_1-t_2)}e^{-\kappa(t_1-t)}\Omega^*(t_1)\Omega(t_2)\rho_0(t_2)\right]\ .\end{aligned} \tag{9}$$

In principle these equations could be averaged over the fluctuations of Markovian driving fields of arbitrary bandwidth using the procedures outlined by Georges and Georges, Lambropoulos and Zoller [2]. Georges calculated the intensity and spectrum of resonance fluorescence for the case of chaotic, phase-diffusing, and Gaussian amplitude driving fields. The intensity was obtained effectively by averaging the last of Eqs. (9) for ρ_0. The calculation of the spectrum was more complicated and yielded an analytic result only in the limit of small average Rabi frequencies. In the present case of four-wave mixing driven by chaotic fields the analysis is even more cumbersome, and therefore we limit the discussion to the case of the pump bandwidth exceeding all other relaxation rates in the problem, a model which admits of simple closed-form expressions for the intensity and frequency spectrum of the scattered radiation, which allows us to treat intensities of the driving fields that saturate the atomic medium, and which is experimentally realistic. A further advantage is that it permits us in a simple way to include the effects of atomic motion.

B. Broad-bandwidth solution

To evaluate (8) in the limit of broad-bandwidth pump fields, we first define

$$B_{ij}(x,x';t,\tau)=[\rho_i(x',t)]^*\rho_j(x,t+\tau)\ .$$

We start off by deriving an expression for $\langle B_{55}(x,x';t,0)\rangle$, which appears in the denominator of (8) as follows: A set of coupled differential equations for the temporal evolution of the $B_{ij}(x,x';t,0)$ can easily be derived from the set of equations (4) for the density matrix elements. The first few, which we give to illustrate their structure, are

$$\left[\frac{d}{dt}+2\Gamma\right]B_{55}=-\frac{i}{2}[\Omega^*(x',t)B_{54}-\Omega(x,t)B_{45}]\ , \tag{10a}$$

$$\left[\frac{d}{dt}+a+d^*\right]B_{54}=-\frac{i}{2}[2\Omega(x',t)B_{55}-\Omega(x,t)B_{44}]\ , \tag{10b}$$

$$\left[\frac{d}{dt}+a^*+d\right]B_{45}=\frac{i}{2}[2\Omega^*(x,t)B_{55}-\Omega^*(x',t)B_{44}]\ . \tag{10c}$$

These equations may be formally integrated to give

$$\langle B_{55}(x,x';t,0)\rangle=\frac{i}{2}\int_{-\infty}^{t}dt_1 e^{-2\Gamma(t-t_1)}\langle \Omega(x,t_1)B_{45}(x,x';t_1,0)\rangle-\langle \Omega^*(x',t_1)B_{54}(x,x';t_1,0)\rangle\ , \tag{11a}$$

$$\langle \Omega^*(x',t)B_{54}(x,x';t,0)\rangle=\int_{-\infty}^{t}dt_1\frac{i}{2}e^{-(a+d^*)(t-t_1)}$$
$$-2\langle \Omega^*(x',t)\Omega(x',t_1)B_{55}(x,x';t_1,0)\rangle+\langle \Omega(x',t)\Omega^*(x,t_1)B_{44}(x,x';t_10)\rangle\ , \tag{11b}$$

$$\langle \Omega(x,t)B_{45}(x,x';t,0)\rangle=\int_{-\infty}^{t}dt_1\frac{i}{2}e^{-(a^*+d)(t-t_1)}$$
$$-\langle \Omega(x,t)\Omega^*(x',t_1)B_{44}(x,x';t_1,0)\rangle+2\langle \Omega(x,t)\Omega^*(x,t_1)B_{55}(x,x';t_1,0)\rangle\ . \tag{11c}$$

To evaluate the averages over field fluctuations of the products in the integrands, we decorrelate the atomic terms and the more rapidly fluctuating field terms writing, for example,

$$\langle \Omega(x,t)\Omega^*(x',t_1)B_{44}(x,x';t_1,0)\rangle$$
$$=\langle \Omega(x,t)\Omega^*(x',t_1)\rangle\langle B_{44}(x,x';t,0)\rangle\ . \tag{12}$$

This decorrelation procedure, while exact for driving fields described by the phase-diffusion model [2], is applicable to chaotic fields only in the large-bandwidth limit. As this limit is approached, the atomic system becomes increasingly insensitive to field correlations of order higher than one, and the models then give identical results. A similar argument applies in the limit of low pump intensity in which the atom responds mainly to first-order correlations of the pump field. To give an indication of the magnitude of the error incurred in using a decorrelation approximation appropriate to a phase-diffusing field for a chaotic field we may examine the difference in the population inversion induced in the two models for fields of the same bandwidth and mean intensity. Georges [2] showed that the largest discrepancy between the result for a phase-diffusing field and that for a chaotic field occurs for intermediate intensities, $\Omega_{\rm rms}\simeq\Gamma$. This discrepancy is approximately 19% of the phase-diffusion-model result for $b/\Gamma=0.1$, 16% for $b/\Gamma=1$, and 5% for $b/\Gamma=10$. We therefore expect the error to be unimportant in the regime that concerns us in this work of $b/\Gamma\gg1$.

An important aspect of our procedure is that the complete set of equations (11) is decorrelated simultaneously. Thus we take into account correlations in products of atomic and field terms that would be lost if we solved for $\rho_5(x,t)$ using the set of equations (4) and then formed and averaged the product $B_{55}(x,x';t,0)$.

First-order field autocorrelations such as that appearing in (12) are given by

$$\langle \Omega(x_1,t_1)\Omega^*(x_2,t_2)\rangle=2|\Omega|^2\cos[k_4(x_1-x_2)]e^{-b|t_1-t_2|}\ , \tag{13}$$

where $|\Omega|^2$ is the mean-square Rabi frequency associated with each pump beam.

The integral equations (11), which involve convolutions of exponential functions of time with averaged products of atomic and field variables, can now be solved for the steady state by taking Laplace transforms and using $F(t\to\infty)=\lim_{s\to0}s\mathcal{L}[F(t)]$. The result is a set of linear equations, 31 in all, of which the first few are

$$\langle B_{55}(x,x';t,0)\rangle=\frac{i}{4\Gamma}[\langle \Omega(x,t)B_{45}\rangle-\langle \Omega^*(x',t)B_{54}\rangle]\ , \tag{14a}$$

$$\langle \Omega^*(x',t)B_{54}(x,x';t,0)\rangle=\frac{i|\Omega|^2}{b+d^*+a}[-2\langle B_{55}\rangle+\langle B_{44}\rangle\cos k_4(x-x')]\ , \tag{14b}$$

$$\langle \Omega(x,t)B_{45}(x,x';t,0)\rangle=\frac{i|\Omega|^2}{b+d+a^*}[2\langle B_{55}\rangle-\langle B_{44}\rangle\cos k_4(x-x')]\ . \tag{14c}$$

The bandwidth of the probe appears in the integral equations (11) by way of averaged products of the form $\langle B_{kk}(x,x';t_1,0)\Omega_3(x,t_1)[\Omega_3(x,t)]^*\rangle$. For the case of p large, or, provided the probe field is weak, for arbitrary p, the probe field and the atomic terms may be decorrelated in a similar manner to that used for products of pump field and atomic terms in (12). Obviously, this is also true for $p=0$. The autocorrelation of the probe field is then evaluated according to

$$\langle \Omega_3(x,t+\tau)[\Omega_3(x,t)]^*\rangle=|\Omega_3|^2\exp-p|\tau|\ .$$

The set of coupled equations (14) may readily be solved to give an analytic expression for the quantity $\langle B_{55}(x,x';t,0)\rangle$, which is related to the intensity of the reflected wave and for other averaged products of field and atomic variables such as $\langle \Omega(x,t)B_{54}(x,x';t,0)\rangle$, $\langle \Omega_3^*(t)\langle \Omega(x,t)B_{51}(x,x';t,0)\rangle\rangle$, etc.

From the expression for $\langle B_{55}(x,x';t,0)\rangle$ we find an expression for $\langle B_{55}(x,x';t,\tau)\rangle$. First of all we use the fact that $\langle B_{55}(x,x';t,0)\rangle$ is stationary to drop the t argument and set

$$B_{55}(x,x';\tau)=[\rho_5(x',0)]^*\rho_5(x,\tau)\ .$$

Then, using Eq. (4f), integrating and taking the average over the field fluctuations we obtain the exact result

$$\langle B_{55}(x,x';\tau)\rangle-\langle B_{55}(x,x';0)\rangle e^{-a\tau} = \int_0^\tau dt \frac{i}{2}\langle \Omega(x,t)B_{54}(x,x';t)\rangle e^{-a(\tau-t)} . \quad (15)$$

Taking a Laplace transform with respect to τ and setting

$$b_5(s)=\mathcal{L}\langle B_{55}(x,x';\tau)\rangle ,$$
$$b_4(s)=\mathcal{L}\langle \Omega(x,\tau)B_{54}(x,x';\tau)\rangle ,$$
$$\mathcal{B}_5(x,x')=\langle B_{55}(x,x';0)\rangle ,$$
$$\mathcal{B}_4(x,x')=\langle \Omega(x,0)B_{54}(x,x';0)\rangle$$

yields

$$(s+a)b_5(s)=\mathcal{B}_5+\frac{i}{2}b_4(s) . \quad (16a)$$

Similarly we obtain

$$(s+b+d)b_4(s)=\mathcal{B}_4+2i|\Omega|^2 b_5(s)+ib_3(s) \quad (16b)$$

and

$$(s+b+\beta)b_3(s)=\beta_3 , \quad (16c)$$

where we have used the exact result [2]

$$\langle \Omega(x,\tau)B_{54}(x,x';0)\rangle = e^{-b\tau}\langle \Omega(x,0)B_{54}(x,x';0)\rangle$$

and

$$b_3(s)=\mathcal{L}\langle \Omega_3^*(\tau)\langle \Omega(x,\tau)B_{51}(x,x';\tau)\rangle\rangle ,$$
$$\mathcal{B}_3(x,x')=\langle \Omega_3^*(0)\langle \Omega(x,0)B_{51}(x,x';0)\rangle\rangle .$$

III. RESULTS

Equations (16) may be solved for $b_5(s)$. To lowest order in Γ/b the solution is

$$b_5(s)=\frac{1}{s+a}\frac{\mathcal{B}_5(x,x')}{1+[|\Omega|^2/(s+a)][1/(s+b+d)]} . \quad (17)$$

Substituting (17) in (8) and canceling the spatial integrals that appear in the denominator and numerator finally yields for the power spectrum,

$$P(\omega)=\mathrm{Re}\left[\frac{1}{s+a}\frac{1}{1+[|\Omega|^2/(s+a)][1/(s+b+d)]}\right]_{s\to i(\omega-\omega_{eg}-\Delta_4)} . \quad (18)$$

The spectrum for low pump intensity $|\Omega|^2/b\kappa \ll 1$ is thus a simple Lorentzian of half-width at half maximum Γ centered at the frequency of the atomic resonance,

$$P(\omega_4)=\frac{\Gamma}{\Gamma^2+\omega_4^2} , \quad \omega_4=\omega-\omega_{eg} . \quad (19)$$

An interesting feature is that this is independent of the probe bandwidth and detuning for $p,\Delta_3,\Delta<b$, in contrast to the monochromatic case $b=0$, when $\omega_4=\omega-(2\omega_1-\omega_3)$. This is because if the frequency bandwidth of the pump field exceeds the detuning of the monochromatic probe field, a perturbation theory calculation of the third-order susceptibility is dominated by diagrams of the form of Fig. 1, in which the intermediate step is two-photon resonant for $\omega_1'=\omega_3$. For pump intensities approaching or exceeding the bandwidth-dependent saturation value given by $|\Omega|^2=b\kappa$ the frequency spectrum of the reflected wave becomes power broadened.

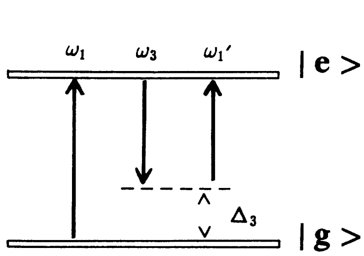

FIG. 1. Schematic diagram showing one process contributing to the induced third-order polarization.

The frequency spectrum of the reflected wave is shown for a few values of the saturation parameter $|\Omega|^2/b\kappa$ in Fig. 2.

IV. ATOMIC MOTION

Degenerate four-wave mixing is commonly employed as a spectroscopic technique in gaseous media. It is therefore useful to determine the effect of the atomic motion on the spectral properties of the reflected radiation. The foregoing analysis may be readily modified to take this into account. The density matrix must now be averaged over the distribution of atomic velocities along

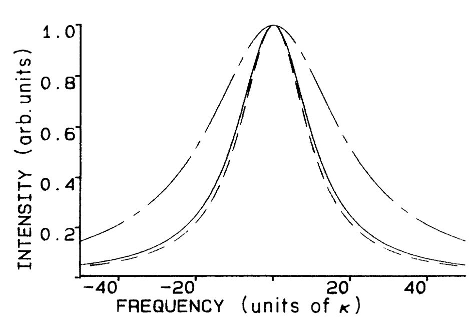

FIG. 2. Graph showing the spectral power density of the reflected wave as a function of the frequency offset from the atomic resonance for $p=b=1000\kappa$, $\Gamma=10\kappa$, $\Delta=\delta=0$, and for $|\Omega|^2/b\kappa=0.1$, dashed line; 1, continuous line; 10, dot-dashed line.

the axis defined by the intersecting beams as well as over the fluctuations of the driving laser fields. We therefore replace the expansion (3) with

$$\rho_{ij}=\int_{-\infty}^{\infty}p(v)\tilde{\rho}_{ij}(v,t)dv\ ,$$
$$\tilde{\rho}_{ij}(v,t)=\sum_{m,n}\rho_{ij}^{mn}(x,v,t)$$
$$\times\exp[-im_1(\omega_1 t-k_1x-k_1vt)$$
$$-im_2(\omega_1 t+k_1x+k_1vt)$$
$$-in(\omega_3 t-k_3x-k_3vt)]\ ,$$
$$m=m_1+m_2\ , \tag{20}$$

where $p(v)$ is the probability density for the longitudinal velocity distribution,

$$p(v)=\frac{2\omega_{eg}}{c\Delta_d}\left[\frac{\ln 2}{\pi}\right]^{1/2}\exp-\frac{4\omega_{eg}^2}{c^2}\frac{(\ln 2)v^2}{\Delta_d^2}$$

and Δ_d is the Doppler width. We define

$$B_{ij}(x,x';v,v';t,\tau)=[\rho_i(x',v',t)]^*\rho_j(x,v,t+\tau)\ ,$$

and, as before, derive coupled equations for $\langle B_{ij}(x,x';v,v';t,0)\rangle$. Using these we derive Laplace transformed equations, counterparts of (16), for $b_5(s,v,v')$. We obtain, again to lowest order in Γ/b,

$$b_5(s,v,v')=\tilde{b}_5(s,v)\mathcal{B}_5(x,x';v,v')\ , \tag{21}$$

where

$$\tilde{b}_5(s,v)=\frac{1}{s+a+ik_3v}\,\frac{1}{1+[|\Omega|^2/(s+a+ik_3v)][1/(s+b+d+ik_3v)]}\ .$$

The frequency spectrum is given from (8) by

$$P(\omega)=\mathrm{Re}\frac{\int_0^1\int_0^1\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}p(v)p(v')b_5[i(\omega-\omega_{eg}-\Delta_4),v,v']dv\,dv'dx\,dx'}{\int_0^1\int_0^1\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}p(v)p(v')\mathcal{B}_5(x,x',v,v')dv\,dv'dx\,dx'}\ . \tag{22}$$

For arbitrary Doppler width Δ_d, $\mathcal{B}_5$ is a complicated function of v and v'. The solution of the set of equations for $\mathcal{B}_5$, the counterparts of Eqs. (14) including the Doppler effect, may be carried through in detail, but the analysis is extremely tedious and we do not include it here. Instead we present an approximate result that is valid for the case of the Doppler width Δ_d, large compared to κ but small compared to b. We first reformulate the velocity integrals appearing in (22) as a product of integrals over v and $\Delta v\equiv v-v'$ and carry out the integration over Δv using a suitable contour in the complex plane. We notice the atomic motion modifies the denominators appearing in (14) which are now of the form $2\Gamma+ik_3\Delta v$, $b+\Gamma+\kappa+i(\Delta+k_3\Delta v+\epsilon k_1 v)$, where $\epsilon=\pm 1$, etc. As described in Ref. [9] we consider only the contribution to the integral of the most significant poles and, in this way, derive an approximate expression for the integral of $\mathcal{B}_5(x,x';v,v')$ over Δv. This is given in Eq. (22) of Ref. [9]:

$$\int_{-\infty}^{\infty}p(\Delta v)d\Delta v\mathcal{B}_5(x,x';v,v')=\frac{\kappa\Theta^2}{16\Delta_d(1+2\Theta)^3}\,\frac{\gamma_0}{(\Delta_3-k_3v)^2+\gamma_0^2}\,\frac{1}{f^2(g+f)}\left[f+\frac{f-g}{1+2f^2/[\kappa(1+2\Theta)]}\right]\ , \tag{23}$$

where

$$f=(\Gamma+\kappa\Theta)^{1/2}\ ,$$
$$g=\left[f^2-\frac{\kappa\Theta^2}{1+2\Theta}\right]^{1/2}\ ,$$
$$\gamma_0=2\Gamma+\kappa(1+\Theta)+p$$

and Θ is the saturation parameter given by

$$\kappa\Theta=\frac{b|\Omega|^2}{b^2+\Delta^2}\ .$$

Here we are only interested in the dependence of the right-hand side of (23) on the velocity v, since the other factors cancel with the denominator of (22). Thus the frequency spectrum is

$$P(\omega)=\mathrm{Re}\int_{-\infty}^{\infty}p(v)dv\,\tilde{b}_5[i(\omega-\omega_{eg}-\Delta_4),v]$$
$$\times\frac{\gamma_0}{(\Delta_3-k_3v)^2+\gamma_0^2}\ . \tag{24}$$

The velocity dependence of $\mathcal{B}_5(x,x';v,v')$ is insignificant for small Doppler widths $\Delta_d\ll\Delta_3$ or when the total width γ_0 is large. The spectrum then approximates to a simple convolution of the power broadened spectrum (17) with the Doppler line shape. This is shown in Fig. 3, in which the spectrum is plotted for $p=b$ for various values of the pump laser intensity and Doppler width. The spectra are here normalized to the same peak intensity; the absolute reflectivity in each case is found by integrating the expression (23) over the distribution of velocities v. This just gives the result (22) of Ref. [9]. Figure 4 shows the spectrum for zero-probe bandwidth $p=0$ and constant probe detuning from resonance and for a few values of the Doppler width. The variation in the spectrum with Doppler width shown in Fig. 4 is as follows: For small Doppler width $\Delta_d<\Delta_3$, the spectral response is Doppler broadened as mentioned above. However, for $\Delta_d>\Delta_3$ and $p=0$ the spectrum is a Lorentzian of width Γ at frequency $\omega_4=\omega_{eg}-\Delta_3$. Thus the effect of the Doppler motion on the spectral response in the $p=0$ case

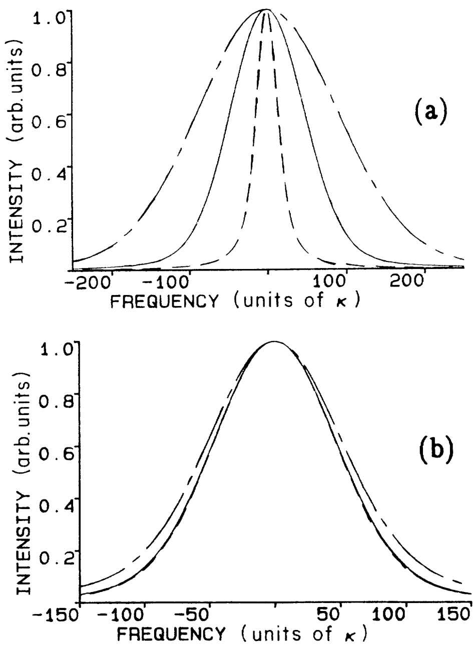

FIG. 3. Graphs showing the spectral power density of the reflected wave for the case $p=b=1000\kappa, \Gamma=10\kappa, \Delta=\delta=0$, (a) for the low-intensity case $|\Omega|^2/b\kappa=0.1$ and Doppler widths $\Delta_d=10\kappa$, dashed line; 50κ continuous line; 100κ, dot-dashed line; (b) for Doppler width $\Delta_d=50\kappa$ and $|\Omega|^2/b\kappa=0.1$, dashed line; 1, continuous line; 10, dot-dashed line.

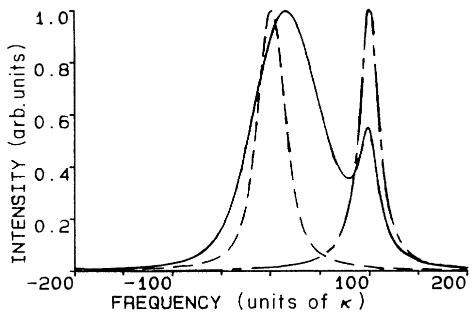

FIG. 4. Graphs showing the spectral power density of the reflected wave for the case of monochromatic probe $p=0$, broad-bandwidth pumps $b=1000\kappa$ and $\Gamma=10\kappa$, pump detuning $\Delta=0$, probe detuning $\Delta_3=-100\kappa$, and Doppler widths $\Delta_d=10\kappa$, dashed line; 30κ continuous line; 100κ dot-dashed line.

is to shift the center frequency of the signal with respect to that of the Doppler-free case given by (18) rather than broaden the distribution of radiated frequencies. This is because the diagram of Fig. 1 now applies in the rest frame of a particular velocity class provided Δ_3 is replaced by Δ_3+k_3v. The contributions to the polarization of this diagram have then to be averaged over all velocity classes. The generated signal is dominated by the contribution of the velocity class for which the probe is resonant, $\Delta_3+k_3v=0$, and the frequency of the detected signal is therefore shifted from the atomic resonance by an amount Δ_3 and has a spectral width determined by the homogeneous width of the transition. As shown in Fig. 4, for intermediate Doppler widths the spectral profile has a double peak.

V. CONCLUSION

We have found analytic expressions for the frequency spectrum of the reflected wave in near-resonant four-wave mixing for the case of broad-bandwidth pump waves interacting in a near-collinear geometry with a weak probe wave in a medium composed of two-level atoms. The solution is valid for an arbitrary bandwidth of the weak probe wave. Results have been obtained for the Doppler-free case and for the situation in which atomic motion is important.

This work represents a study of an experimentally realizable model of a nonlinear-optical parametric process and is of direct relevance to spectroscopic four-wave experiments employing broad-bandwidth lasers and gaseous nonlinear media. Experimental work to test the predictions of this theory is underway.

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