Four-wave mixing in stochastic fields: Fluctuation-induced resonances

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The effect of pump fluctuations on various coherent processes that arise in three-level systems interacting with two external fields is examined. Such coherent processes include the forward Hanle effect and various four-wave mixing effects such as the generation of phase-conjugate signals. A general formulation that enables one to calculate the influence of laser linewidth on the coherent signals produced in various directions is presented. Ensemble averages, over laser temporal fluctuations, of various physical quantities, such as atomic polarization, are calculated. The spectrum of polarization fluctuations is shown to consist of several new features which lead to coherent radiation in different directions depending on the resonant frequencies in the polarization fluctuations. The influence of pump linewidth on pressure-induced extra resonance (PIER) is treated in detail. The possibility of producing a laser-fluctuation-induced coherent signal at one of the atomic frequencies is examined. This new signal, which is produced in a direction different from that of the PIER signal, but has the same type of resonant character as PIER, is found to have significantly different pressure dependence than the PIER signal. The results of our numerical computations are qualitatively explained in terms of the convolutions of products of third-order susceptibilities and pump field-correlation functions.

I. INTRODUCTION

The interaction of an atomic system with two different laser fields leads to a variety of important phenomena, many of which fall under the general class of four-wave mixing[1] effects. Other interesting results of such interaction are seen in saturated absorption and Hanle studies,[2,3] etc. Some of these effects, such as the forward Hanle effect and four-wave mixing, are coherent effects in that various atoms cooperate to produce a macroscopic polarization, whereas others, such as optical double resonance and Raman gain spectroscopy, are of the incoherent type since they are basically single-atom effects. Many of these are now used as important spectroscopic tools in the study of the characteristics of atomic/molecular systems and hence it is desirable to examine how the statistical fluctuations[4,5] of the laser light can affect the results of the interaction of the atom with fields. Previous calculations[4,5] have shown the important effect of laser bandwidth on the outcome of experiments involving single-atom phenomena such as resonance fluorescence, double resonance, and Raman scattering. The effect of laser field fluctuations on coherent effects such as those arising in the context of four-wave mixing and the forward Hanle effect seems to have received very little attention. The only problem that appears to have been examined is that of resonant coherent anti—Stokes-Raman scattering.[6]

In the present study, we examine the effect of laser bandwidth and statistics on the coherent effects in three-level systems with special emphasis on the recently discovered collisionally induced coherence[7–10] effects in four-wave mixing. In Sec. II, we present the basic equations for a three-level system in the field of two lasers with frequencies ω_1 and ω_2. Each laser is assumed to be fluctuating and we characterize each by its statistical properties. The atom has both radiative and nonradiative sources of relaxation. The model is very general and is capable of describing a diversity of phenomena such as four-wave mixing, phase conjugacy,[11] Hanle effects, and saturated absorption. A simple expression for the third-order nonlinear susceptibility for the three-level model is also presented. In Sec. III we derive equations that yield the ensemble average of the density matrix elements and the quadratic forms involving density matrix elements. Such ensemble averages can be used to obtain the average value of the atomic polarization and the fluctuations in polarization. In Sec. IV we compute the spectrum of the polarization fluctuations which we then use to derive the spectrum of the emitted coherent radiation. We will, throughout this paper, use the terminology *coherent radiation* for the radiation produced by the cooperative interaction among various atoms, even though the emitted radiation is not strictly coherent in that it would have statistical fluctuations due to the laser temporal fluctuations. The generation of coherent radiation depends on the various phase-matching conditions. We first separate out the PIER signal and treat the influence of pump line width on PIER in detail. We then show that a coherent signal can arise due to laser temporal fluctuations. The new coherent signal predicted here has an extraordinary pressure dependence. In Sec. V we compare the results of our numerical computations with those following from the general form of third-order nonlinear susceptibility[7] and certain types of convolution integrals[6] involving the product of $\chi^{(3)}$ and the higher-order pump correlation functions. In particular, we present the analytic form of the pressure dependence of the fluctuation-induced resonance (FIER). It is important to note that it is possible to discriminate experimentally between the PIER signal and the fluctuation-induced contribution by the phase-matching condition.

II. BASIC DYNAMICAL EQUATIONS FOR FOUR-WAVE MIXING IN A THREE-LEVEL MODEL

Keeping in view the recent experimental and the theoretical activity related to the pressure-induced resonances in four-wave mixing and the generation of phase-conjugated signals, we discuss the response equations for the behavior of a system of three-level atoms in the presence of two fluctuating fields. In Fig. 1, we schematically show various interactions along with various incoherent processes. We write the total electric field incident on the system in the form

$$\vec{\epsilon}=\vec{\epsilon}_1\exp\left[-i\Phi_1\left[t-\frac{\hat{k}_1\cdot\vec{r}}{v}\right]-i\omega_1 t+i\vec{k}_1\cdot\vec{r}\right]+\vec{\epsilon}_2\exp(-i\omega_2 t+i\vec{k}_2\cdot\vec{r})+\text{c.c.}\ , \qquad (2.1)$$

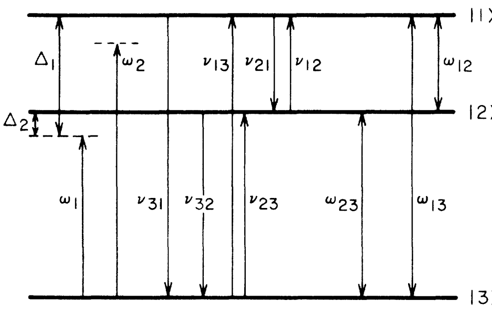

FIG. 1. Schematic representation of the energy levels and the various coherent and incoherent transitions.

where the field of the laser at ω_1 is assumed to have a fluctuating phase[12] $\Phi_1(t)$ and v is the velocity of the light in the medium. We will ignore the dispersion of the refractive index. The justification for writing the field in the form (2.1) is given in the Appendix. The fluctuating phase is assumed to be a Gaussian random variable with zero mean and with the correlation function

$$\dot{\Phi}_1(t)=\mu_1(t)\ ,$$
$$\langle\mu_1(t)\mu_1(t')\rangle=2\gamma_{c1}\delta(t-t')\ . \qquad (2.2)$$

Thus each laser is assumed to have a random frequency modulation, i.e., a phase-diffusion model is used for the laser at ω_1. In what follows, we assume that the field $\vec{\epsilon}_1$ is a strong pump, whereas $\vec{\epsilon}_2$ is only a weak one. In the context of four-wave mixing two photons are taken from the field $\vec{\epsilon}_1$ and one from $\vec{\epsilon}_2$. Now taking the usual form of the radiation matter interaction $-\vec{d}\cdot\vec{E}$, making the rotating wave approximation and the transformation to rotating frame so as to eliminate the fast periodic time dependence from the equations of motion, we find that the density matrix equations, for the μth atom located at the position $\vec{R}_\mu$, can be written in the following matrix form:

$$\frac{\partial\sigma^{(\mu)}}{\partial t_\mu}=M(t_\mu)\sigma^{(\mu)}+I(t_\mu)+e^{-i\delta t_\mu+i\theta_\mu}[M_+(t_\mu)\sigma^{(\mu)}+I_+(t_\mu)]+e^{i\delta t_\mu-i\theta_\mu}[M_-(t_\mu)\sigma^{(\mu)}+I_-(t_\mu)]\ ,$$
$$\delta=\omega_1-\omega_2 \qquad (2.3)$$

where t_μ *now refers to the reduced time* $[t-(\hat{k}_1\cdot\vec{R}_\mu/v)]$, which depends on the position of the μth atom. In Eq. (2.3) the elements of the eight-component vector σ are related to the density matrix elements by

$$\sigma_1=\rho_{13}e^{i\omega_1 t_\mu},\ \ \sigma_2=\rho_{23}e^{i\omega_2 t_\mu},\ \ \sigma_3=\sigma_1^*,\ \ \sigma_4=\sigma_2^*,\ \ \sigma_5=\rho_{12},\ \ \sigma_6=\sigma_5^*\ ,$$
$$\sigma_7=\rho_{11},\ \ \sigma_8=\rho_{22},\ \ \rho_{33}=1-\rho_{11}-\rho_{22}\ , \qquad (2.4)$$

and θ_μ denotes the phase factor $[(\omega_2/v)\hat{k}_1-\hat{k}_2]\cdot\vec{R}_\mu$. Other matrices in (2.3) are

$$M=\begin{bmatrix} -\Gamma_1-i\Delta_1 & 0 & 0 & 0 & -ig_2 & 0 & -2ig_1 & -ig_1 \\ 0 & -\Gamma_2-i\Delta_2 & 0 & 0 & 0 & -ig_1 & -ig_2 & -2ig_2 \\ 0 & 0 & -\Gamma_1+i\Delta_1 & 0 & 0 & ig_2^* & 2ig_1^* & ig_1^* \\ 0 & 0 & 0 & -\Gamma_2+i\Delta_2 & ig_1^* & 0 & ig_2^* & 2ig_2^* \\ -ig_2^* & 0 & 0 & ig_1 & -\Gamma_0-i(\Delta_1-\Delta_2) & 0 & 0 & 0 \\ 0 & -ig_1^* & ig_2 & 0 & 0 & -\Gamma_0+i(\Delta_1-\Delta_2) & 0 & 0 \\ -ig_1^* & 0 & ig_1 & 0 & 0 & 0 & \begin{pmatrix} -\nu_1 \\ -\nu_{13} \end{pmatrix} & \begin{pmatrix} \nu_{12} \\ -\nu_{13} \end{pmatrix} \\ 0 & -ig_2^* & 0 & ig_2 & 0 & 0 & \begin{pmatrix} +\nu_{21} \\ -\nu_{23} \end{pmatrix} & \begin{pmatrix} \nu_2 \\ -\nu_{23} \end{pmatrix} \end{bmatrix}$$

$$\Delta_1=\omega_{13}-\omega_1,\quad \Delta_2=\omega_{23}-\omega_1 \tag{2.5}$$

and the nonvanishing elements of M_+ and M_- are

$$\begin{aligned} &M_{+36}=i\lambda_2^*,\ M_{+37}=2i\lambda_i^*,\ M_{+38}=i\lambda_1^*,\ M_{+45}=i\lambda_1^*,\ M_{+47}=i\lambda_2^*\ ,\\ &M_{+48}=2i\lambda_2^*,\ M_{+51}=-i\lambda_2^*,\ M_{+62}=-i\lambda_i^*,\ M_{+71}=-i\lambda_i^*,\ M_{+82}=-i\lambda_2^*\ ; \end{aligned} \tag{2.6}$$

$$\begin{aligned} &M_{-15}=-i\lambda_2,\ M_{-17}=-2i\lambda_1,\ M_{-18}=-i\lambda_1,\ M_{-26}=-i\lambda_1,\ M_{-27}=-i\lambda_2\ ,\\ &M_{-28}=-2i\lambda_2,\ M_{-54}=i\lambda_1,\ M_{-63}=i\lambda_2,\ M_{-73}=i\lambda_1,\ M_{-84}=i\lambda_2\ . \end{aligned} \tag{2.7}$$

The nonvanishing elements of the column matrices $I, I_\pm$ are

$$\begin{aligned} &I_1=ig_1,\ I_2=ig_2,\ I_3=-ig_1^*,\ I_4=-ig_2^*,\ I_7=+\nu_{13},\ I_8=\nu_{23}\ ,\\ &I_{+3}=-i\lambda_1^*,\ I_{+4}=-i\lambda_2^*,\ I_{-1}=i\lambda_1,\ I_{-2}=i\lambda_2\ . \end{aligned} \tag{2.8}$$

In the above equations various field-matter couplings are

$$\begin{aligned} &g_1(t_\mu)=g_1e^{-i\Phi_1(t_\mu)},\ g_2(t_\mu)=g_2e^{-i\Phi_1(t_\mu)},\ g_1=-(\vec{d}_{13}\cdot\vec{\epsilon}_1)\ ,\\ &\lambda_1=-(\vec{d}_{13}\cdot\vec{\epsilon}_2),\ g_2=-(\vec{d}_{23}\cdot\vec{\epsilon}_1),\ \lambda_2=-(\vec{d}_{23}\cdot\vec{\epsilon}_2)\ . \end{aligned} \tag{2.9}$$

The parameter ν_{ij} gives the transition rate per unit time from the level $|j\rangle$ to $|i\rangle$ due to all sources of relaxation such as spontaneous emission, collisions, etc.; for example, ν_1 will be equal to $\nu_{31}+\nu_{21}$. The decay constants Γ's of the off-diagonal elements are related to ν's by

$$\Gamma_1=\Gamma_1^{\mathrm{ph}}+\tfrac{1}{2}(\nu_1+\nu_{23}+\nu_{13}),\ \Gamma_2=\Gamma_2^{\mathrm{ph}}+\tfrac{1}{2}(\nu_2+\nu_{23}+\nu_{13}),\ \Gamma_0=\Gamma_0^{\mathrm{ph}}+\tfrac{1}{2}(\nu_1+\nu_2)\ , \tag{2.10}$$

where Γ^{ph} represents the contribution to Γ due to phase-interrupting collisions. The impact approximation for collision rates has been assumed.

If the frequency modulation of the field at ω_1 is ignored, then the steady-state solution of (2.3) to first order in λ's is

$$\sigma^{(\mu)}=\sigma^{(0)}+e^{i\delta t_\mu-i\theta_\mu}\sigma_-^{(1)}+e^{-i\delta t_\mu+i\theta_\mu}\sigma_+^{(1)}+\cdots\ , \tag{2.11}$$

$$\sigma^{(0)}=-M^{-1}I\ , \tag{2.12}$$

$$\sigma_\pm^{(1)}=(\mp i\delta-M)^{-1}(M_\pm\sigma^{(0)}+I_\pm)\ . \tag{2.13}$$

It may be noted that $\sigma^{(0)}$ and $\sigma_\pm^{(1)}$ are independent of the position of the μth atom. If $\vec{p}^{\,(\mu)}$ is the positive-frequency part of the polarization produced by the μth atom, then the coherent contribution to the intensity with a given polarization $\hat{\epsilon}$ and in the *direction* $\vec{k}$ can be shown to be proportional to

$$I_{\vec{k}}=\left|\sum_\mu \vec{p}^{(\mu)}\cdot\hat{\epsilon}\,e^{-i\vec{k}\cdot\vec{R}_\mu}\right|^2=\sum_{\mu\nu}(\vec{p}^{\,(\mu)}\cdot\hat{\epsilon})(\vec{p}^{\nu}\cdot\hat{\epsilon})^*e^{-i\vec{k}\cdot(\vec{R}_\mu-\vec{R}_\nu)}$$

which in terms of density matrix elements becomes

$$I_{\vec{k}} = \sum_{\mu\nu} (\vec{d}_{31}\cdot\hat{\epsilon}\rho_{13}^{(\mu)} + \vec{d}_{32}\cdot\hat{\epsilon}\rho_{23}^{(\mu)})(\vec{d}^{\,*}_{31}\cdot\hat{\epsilon}^{\,*}\rho_{13}^{(\nu)*} + \vec{d}^{\,*}_{32}\cdot\hat{\epsilon}^{*}\rho_{23}^{(\nu)*})e^{-i\vec{k}\cdot(\vec{R}_\mu - \vec{R}_\nu)}$$
$$= \sum_{\mu\nu} (\vec{d}_{31}\cdot\hat{\epsilon}\sigma_1^{(\mu)} + \vec{d}_{32}\cdot\hat{\epsilon}\sigma_2^{(\mu)})(\vec{d}^{\,*}_{31}\cdot\hat{\epsilon}^{*}\sigma_3^{(\nu)} + \vec{d}^{\,*}_{32}\cdot\hat{\epsilon}^{*}\sigma_4^{(\nu)})e^{-i(\vec{k}-\vec{k}_1)\cdot(\vec{R}_\mu - \vec{R}_\nu)} . \quad (2.14)$$

The factor

$$\exp[i\vec{k}_1\cdot(\vec{R}_\mu - \vec{R}_\nu)]$$

arises since the reduced times for the μth and the νth atom differ by

$$(t-\hat{k}_1\cdot\vec{R}_{\mu/\nu}) - (t-\hat{k}_1\cdot\vec{R}_{\nu/v}) .$$

Note that the magnitude of the vector $\vec{k}$ is determined by the frequency around which the oscillations of the dipole moment occur. The elements $\sigma_i^{(\mu)}$ depend on the wave vectors and coordinates of atoms. The four-wave mixing contributions arise from terms in the density matrix that are of first order in λ, i.e., by the contribution (2.13). On substituting (2.13) in (2.14), we find a complicated dependence on atomic positions. This dependence cancels for certain directions of observation and coherent signals result. The four-wave signal in the direction $2\vec{k}_1 - \vec{k}_2$ can be obtained from (2.14) if we use the replacements

$$\sigma_{1,2}^{(\mu)} \to (\sigma_+^{(1)})_{1,2}, \quad \sigma_{3,4}^{(\nu)} \to (\sigma_-^{(1)})_{3,4} . \quad (2.15)$$

However, if we were to use the replacements

$$\sigma_{1,2}^{(\mu)} \to (\sigma_-^{(1)})_{1,2}, \quad \sigma_{3,4}^{(\nu)} \to (\sigma_+^{(1)})_{3,4} , \quad (2.16)$$

then (2.14) would yield a contribution in the direction $\vec{k}_2$ at frequency ω_2. Such a contribution may be used, for example, in studies of the forward Hanle effect. Note that so far we have made no assumption regarding the pump beam's (ω_1) intensity. The high-intensity results are important in the generation of phase-conjugate signals in which case *ac* Stark splitting has received considerable attention.[11] The lowest-order contribution in the direction $(2\vec{k}_1 - \vec{k}_2)$ is easily evaluated. One finds that

$$\sigma_{-1}^{(1)} = -ig_2g_1\lambda_2^*(\Gamma_1+i\Delta_1-i\delta)^{-1}(\Gamma_1+i\Delta_1)^{-1}(\Gamma_2-i\Delta_2-i\delta)^{-1} \times \left\{1+\frac{\Gamma_1+\Gamma_2-\Gamma_0}{\Gamma_0+i(\Delta_1-\Delta_2-\delta)}\right\} - ig_1(-i\delta+i\Delta_1+\Gamma_1)^{-1}(2A+B) , \quad (2.17)$$

$$\sigma_{-2}^{(1)} = -ig_1g_2\lambda_1^*(\Gamma_2+i\Delta_2-i\delta)^{-1}(\Gamma_2+i\Delta_2)^{-1}(\Gamma_1-i\Delta_1-i\delta)^{-1} \times \left\{1+\frac{\Gamma_1+\Gamma_2-\Gamma_0}{\Gamma_0+i(\Delta_2-\Delta_1-\delta)}\right\} - ig_2(-i\delta+i\Delta_2+\Gamma_2)^{-1}(A+2B) , \quad (2.18)$$

with

$$A = g_1\lambda_1^*(-i\delta+\nu_1)^{-1}[(\Gamma_1+i\Delta_1)^{-1}(\Gamma_1-i\Delta_1-i\delta)^{-1}](2\Gamma_1-i\delta) ,$$
$$B = g_2\lambda_2^*(-i\delta+\nu_2)^{-1}[(\Gamma_2+i\Delta_2)^{-1}(-i\delta-i\Delta_2+\Gamma_2)^{-1}](2\Gamma_2-i\delta) . \quad (2.19)$$

The coherent radiation in the direction $2\vec{k}_1 - \vec{k}_2$ and at frequency $(2\omega_1-\omega_2)$ is now

$$|\vec{d}_{31}\cdot\hat{\epsilon}\sigma_{+1}^{(1)} + \vec{d}_{32}\cdot\hat{\epsilon}\sigma_{+2}^{(1)}|^2 .$$

As is well known from the work of Bloembergen and coworkers,[7] pressure-induced resonances arise due to the nonvanishing of $\Gamma_1+\Gamma_2-\Gamma_0$. The term in the curly bracket in (2.18) is the same as that of Bloembergen *et al.*[8] Let us now compare the magnitude of the peak at $\Delta_1-\Delta_2=\delta$ with the contribution from terms like A and B:

$$B \sim g_2\lambda_2^*(i\Delta_2)^{-1}(-i\delta)^{-1}(-i\delta-i\Delta_2)^{-1}(-i\delta)$$
$$= g_2\lambda_2^*/\Delta_2(\delta+\Delta_2) \sim g_2\lambda_2^*/\Delta_2\Delta_1 \quad \text{for } \Delta_1,\Delta_2,\delta \gg \Gamma$$

whereas the peak amplitude (at $\delta=\Delta_1-\Delta_2$) is approximately

$$(g_1g_2\lambda_2^*/\Delta_1^2\Delta_2)\{1+[(\Gamma_1+\Gamma_2-\Gamma_0)/\Gamma_0]\} .$$

Hence for good resolution of the peak at $\Delta_1-\Delta_2=\delta$, one must have $\Delta_1 \ll \Delta_2$. The contribution from A

terms can be eliminated by choosing the polarizations of the fields properly. Similarly for a good resolution of the peak at $\Delta_1-\Delta_2=-\delta$, one must choose detunings such that $\Delta_1 \gg \Delta_2$. Bloembergen *et al.* have performed a detailed study of the pressure-induced resonance $\Delta_1-\Delta_2=-\delta$ under the condition $\Delta_1 \gg \Delta_2$.

III. ENSEMBLE AVERAGES FOR DENSITY MATRIX ELEMENTS AND THEIR QUADRATIC FORMS OVER LASER FLUCTUATIONS

In this section we treat in detail the effect of laser fluctuations on four-wave mixing. The fluctuations of the laser are described by the model (2.2). Because of the fluctuations of Φ_i, the density matrix elements become random functions and such random functions have to be averaged, over laser fluctuations, in order to obtain the mean dipole moments and other physical quantities. Since the intensity (coherent part) is a quadratic function of the density matrix elements, it is obvious that the average intensity will be obtained by averaging the quadratic forms rather than by constructing quadratic forms from the averaged density matrix elements. The fluctuations can be handled using the technique developed in Ref. 4 which essentially finds transformations that make the structure of the dynamical equations for density matrix elements similar to the equations for multiplicative stochastic processes. We now introduce the set ψ of elements ψ_i defined by

$$\begin{aligned}&\psi_1^{(\mu)}=\sigma_1^{(\mu)}e^{+i\Phi_1},\ \ \psi_2^{(\mu)}=\sigma_2^{(\mu)}e^{+i\Phi_1},\ \ \psi_3^{(\mu)}=\sigma_3^{(\mu)}e^{-i\Phi_1},\ \ \psi_4^{(\mu)}=\sigma_4^{(\mu)}e^{-i\Phi_1},\\&\psi_5^{(\mu)}=\sigma_5^{(\mu)},\ \ \psi_6=\sigma_6^{(\mu)},\ \ \psi_7^{(\mu)}=\sigma_7^{(\mu)},\ \ \psi_8^{(\mu)}=\sigma_8^{(\mu)} .\end{aligned} \tag{3.1}$$

The equation of motion (2.3) now leads to equations for the ψ_i, which in matrix form read as

$$\begin{aligned}\frac{\partial\psi^{(\mu)}}{\partial t_\mu}=&M\psi^{(\mu)}+I+e^{i\delta t_\mu-i\theta_\mu+i\Phi_1(t_\mu)}(M_-\psi^{(\mu)}+I_-)\\&+e^{-i\delta t_\mu+i\theta_\mu-i\Phi_1(t_\mu)}(M_+\psi^{(\mu)}+I_+)+i\mu_1(t_\mu)F\psi^{(\mu)} ,\end{aligned} \tag{3.2}$$

where all the matrices M, I, M_+, I_+, M_-, and I_- are obtained from the corresponding matrices $M(t)$, $I(t)$, $M_+(t)$, $I_+(t)$, $M_-(t)$, and $I_-(t)$ by letting $e^{\pm i\Phi_i(t)}\rightarrow 1$. All the matrices are now time independent. The matrix F has nonzero elements given by

$$F_{11}=F_{22}=-F_{33}=-F_{44}=1 . \tag{3.3}$$

If we now define

$$\psi^{(\mu,n)}=e^{in\Phi_\mu}\psi^{(\mu)},\ \ \Phi_\mu=\Phi_1(t_\mu) , \tag{3.4}$$

then, on using the results from the theory of multiplicative processes (Ref. 4, Appendix B), one can show that the ensemble average of ψ over laser fluctuations obeys equations of the form

$$\begin{aligned}\langle\dot\psi^{(\mu,n)}\rangle=&M\langle\psi^{(\mu,n)}\rangle+I\langle e^{in\Phi_\mu}\rangle+e^{i\delta t_\mu-i\theta_\mu}(M_-\langle\psi^{(\mu,n+1)}\rangle+I_-\langle e^{i(n+1)\Phi_\mu}\rangle)\\&+e^{-i\delta t_\mu+i\theta_\mu}(M_+\langle\psi^{(\mu,n-1)}\rangle+I_+\langle e^{i(n-1)\Phi_\mu}\rangle)-\gamma_{c1}(F+n)^2\langle\psi^{(\mu,n)}\rangle .\end{aligned} \tag{3.5}$$

The above set of equations for $\langle\psi^{(n)}\rangle$ can now be solved using perturbation techniques. For example, up to first order in λ, one has

$$\begin{aligned}&\langle\psi^{(\mu,n)}\rangle=(\gamma_{c1}F^2-M)^{-1}I\delta_{n,0},\ \ O(1)\\&\langle\psi^{(\mu,\pm1)}\rangle=e^{\mp i\delta t_\mu\pm i\theta_\mu}\psi^{\pm},\ \ O(\lambda)\end{aligned} \tag{3.6}$$

$$\begin{aligned}\psi^{\pm}=&[+\gamma_{c1}(F\pm1)^2-M\mp i\delta]^{-1}\\&\times(M_\pm\langle\psi^{(0)}\rangle+I_\pm) ,\end{aligned} \tag{3.7}$$

which may be compared with the results (2.12) and (2.13) in the absence of any laser temporal fluctuations. It may be noted that results like (3.7) are quite useful, for example, in studying the rate of energy absorption[3] from a probe beam in the presence of a pump beam, when both pump and probe are fluctuating. This is because the energy absorption depends linearly on the induced polarization, which can be obtained from certain elements of ψ.

As noted earlier, the coherent radiation is determined by the quadratic forms involving density matrix elements; therefore we have to construct equations for quantities like $\langle \psi^{(\mu)}\psi^{(\nu)}\rangle$ where the indices μ and ν refer to any pair of two atoms. The phase factor θ depends on the location of a given atom. Since we are only interested in the four-wave mixing signal, we present our calculations only to second order in λ. On writing down equations for $\psi_\alpha^{(\mu)}$ and $\psi_\beta^{(\nu)}$ and on using the results from the theory of multiplicative stochastic processes, we find up to second order in λ, with superscripts on $\langle\ \rangle$ denoting the order of perturbation

$$\langle \psi_\alpha^{(\mu)}\dot{\psi}_\beta^{(\nu)}\rangle^{(2)} = -\gamma_{c1}(F_\alpha+F_\beta)^2\langle \psi_\alpha^{(\mu)}\psi_\beta^{(\nu)}\rangle^{(2)} + \Bigg[I_\alpha\langle \psi_\beta^{(\nu)}\rangle^{(2)} + \sum_i M_{\alpha i}\langle \psi_i^{(\mu)}\psi_\beta^{(\nu)}\rangle^{(2)} + e^{i\delta t_\mu}(I_-)_\alpha e^{-i\theta_\mu}\langle \psi_\beta^{(\nu)} e^{i\Phi_\mu}\rangle^{(1)}$$
$$+ e^{-i\delta t_\mu}(I_+)_\alpha e^{i\theta_\mu}\langle \psi_\beta^{(\nu)} e^{-i\Phi_\mu}\rangle^{(1)} + \sum_i e^{i\delta t_\mu - i\theta_\mu} M_{\alpha i}^{(-)}\langle \psi_i^{(\mu)}\psi_\beta^{(\nu)} e^{i\Phi_\mu}\rangle^{(1)}$$
$$+ e^{-i\delta t_\mu + i\theta_\mu}\sum M_{\alpha i}^{(+)}\langle \psi_i^{(\mu)}\psi_\beta^{(\nu)} e^{-i\Phi_\mu}\rangle^{(1)} + \begin{bmatrix}\alpha\leftrightarrow\beta\\ \mu\leftrightarrow\nu\\ \theta_\mu\to\theta_\nu - q_{\nu\mu}\end{bmatrix}\Bigg],\quad q_{\nu\mu} = \frac{\delta\hat{k}_1}{v}\cdot(\vec{R}_\mu - \vec{R}_\nu)\ . \tag{3.8}$$

The phase factor $q_{\nu\mu}$ arises since the reduced time for the νth atom is different from that for the μth atom. First-order terms like $\langle \psi_\beta^{(\nu)} e^{\pm i\Phi}\rangle^{(1)}$ are already given by (3.7). First-order terms like $\langle \psi_\alpha^{(\mu)}\psi_\beta^{(\nu)} e^{\pm i\Phi}\rangle^{(1)}$ are obtained by equations similar to (3.8):

$$\langle \psi_\alpha^{(\mu)}\dot{\psi}_\beta^{(\nu)} e^{i\Phi_\mu}\rangle^{(1)} = -\gamma_{c1}(F_\alpha+F_\beta+1)^2\langle \psi_\alpha^{(\mu)}\psi_\beta^{(\nu)} e^{i\Phi_\mu}\rangle^{(1)} + \Bigg[I_\alpha\langle \psi_\beta^{(\nu)} e^{i\Phi_\mu}\rangle^{(1)} + M_{\alpha i}\langle \psi_i^{(\mu)}\psi_\beta^{(\nu)} e^{i\Phi_\mu}\rangle^{(1)} + e^{i\delta t_\mu}(I_-)_\alpha e^{-i\theta_\mu}\langle \psi_\beta^{(\nu)} e^{2i\Phi_\mu}\rangle^{(0)}$$
$$+ e^{-i\delta t_\mu}(I_+)_\alpha e^{i\theta_\mu}\langle \psi_\beta^{(\nu)}\rangle^{(0)} + \sum_i e^{i\delta t_\mu - i\theta_\mu} M_{\alpha i}^{(-)}\langle \psi_i^{(\mu)}\psi_\beta^{(\nu)} e^{2i\Phi_\mu}\rangle^{(0)}$$
$$+ \sum_i e^{-i\delta t_\mu + i\theta_\mu} M_{\alpha i}^{(+)}\langle \psi_i^{(\mu)}\psi_\beta^{(\nu)}\rangle^{(0)} + \begin{bmatrix}\alpha\leftrightarrow\beta\\ \mu\leftrightarrow\nu\\ \theta_\mu\to\theta_\nu - q_{\nu\mu}\end{bmatrix}\Bigg]\ . \tag{3.9}$$

Zeroth-order terms like $\langle \psi_i^{(\mu)}\psi_\beta^{(\nu)} e^{\pm i\Phi_\mu}\rangle^{(0)}$, $\langle \psi_\beta^{(\nu)} e^{\pm 2i\Phi_\mu}\rangle^{(0)}$ are all zero in the steady state, whereas expectation values like $\langle \psi_\alpha^{(\mu)}\psi_\beta^{(\nu)}\rangle^{(0)}$ are given by

$$\langle \psi_\alpha^{(\mu)}\dot{\psi}_\beta^{(\nu)}\rangle^{(0)} = -\gamma_{c1}(F_\alpha+F_\beta)^2\langle \psi_\alpha^{(\mu)}\psi_\beta^{(\nu)}\rangle^{(0)} + \left[I_\alpha\langle \psi_\beta^{(\nu)}\rangle^{(0)} + \sum_i M_{\alpha i}\langle \psi_i^{(\mu)}\psi_\beta^{(\nu)}\rangle^{(0)} + \begin{bmatrix}\alpha\leftrightarrow\beta\\ \mu\leftrightarrow\nu\end{bmatrix}\right]\ . \tag{3.10}$$

In deriving Eqs. (3.8) and (3.9) we have assumed that the transit time $|(R_\mu - R_\nu)/v|$ is much smaller than the time scales associated with the system. Hence the differences between t_μ and t_ν in slowly varying quantities are ignored. The typical time scales associated with the system will be of the order Γ_i^{-1}, γ_{c1}^{-1}. In vapors where R_μ and R_ν are random, the transit time is generally much smaller than the observation time. This condition is equivalent to $L \ll l_c$, where L is the cell length and l_c is the coherence length, i.e., the length over which the atoms cooperate. From the structure of equations like (3.8) and (3.9), one can show that in the steady state $\langle \psi_\alpha^{(\mu)}\psi_\beta^{(\nu)}\rangle^{(2)}$ has the structure, in the limit $t\to\infty$,

$$\langle \psi_\alpha^{(\mu)}\psi_\beta^{(\nu)}\rangle^{(2)} = e^{-i(\theta_\mu - \theta_\nu + q_{\nu\mu})} W_{\alpha\beta}^{+} + e^{i(\theta_\mu - \theta_\nu + q_{\nu\mu})} W_{\alpha\beta}^{-} + \cdots\ , \tag{3.11}$$

where ellipses denote either the terms oscillating at δ or the terms that do not depend on the relative phase factor $(\theta_\mu - \theta_\nu)$. We will see in Sec. IV that the spatial structure of (3.11) in terms of the atomic positions is crucial in the determination of the coherent signals that may be generated in various directions. We now simplify

Eqs. (3.8)–(3.11) so that these can be used for numerical evaluation. The steady-state solution of (3.9) can be shown to have the structure

$$\langle \psi_\alpha^{(\mu)} \psi_\beta^{(\nu)} e^{\pm i\Phi_\mu} \rangle^{(1)} \cong e^{\mp i\delta t_\mu} (e^{\pm i\theta_\mu} U_{\alpha\beta}^{\pm\mu} + e^{\pm i(\theta_\nu - q_{\nu\mu})} U_{\alpha\beta}^{\pm\nu}) , \tag{3.12}$$

where

$$[\mp i\delta + \gamma_{c1}(F_\alpha + F_\beta \pm 1)^2 + \gamma_{c2}] U_{\alpha\beta}^{\pm\mu} - \sum_i (M_{\alpha i} U_{i\beta}^{\pm\mu} + M_{\beta i} U_{\alpha i}^{\pm\mu}) = g_{\alpha\beta}^{\pm,\mu} , \tag{3.13}$$

and

$$g_{\alpha\beta}^{\pm\mu} = I_\beta \psi_\alpha^{\pm} + \sum_i (M^{\pm})_{\alpha i} \langle \psi_i^\mu \psi_\beta^\nu \rangle^{(0)} + (I_\pm)_\alpha \langle \psi_\beta^\nu \rangle^{(0)} . \tag{3.14}$$

The quantities $U_{\alpha\beta}^{\pm\nu}$ are also determined by (3.13) but with an inhomogeneous term $g_{\alpha\beta}^{\pm\nu}$ which is related to $g_{\alpha\beta}^{\pm\mu}$ by

$$g_{\alpha\beta}^{\pm\nu} = g_{\alpha\beta}^{\pm\mu} \Big|_{\substack{\alpha \leftrightarrow \beta \\ \mu \leftrightarrow \nu}} . \tag{3.15}$$

Using now the structure of (3.12) and (3.11), we obtain, from (3.8), equations for $W^{\pm}$,

$$\left[\gamma_{c1}(F_\alpha + F_\beta)^2 W_{\alpha\beta}^{\pm} - \sum_i M_{\alpha i} W_{i\beta}^{\pm} - \sum_i M_{\beta i} W_{i\alpha}^{\pm} \right] = C_{\alpha\beta}^{\pm} , \tag{3.16}$$

$$C_{\alpha\beta}^{\pm} = (I_\mp)_\alpha \psi_\beta^{\pm} + (I_\pm)_\beta \psi_\alpha^{\mp} + \sum (M^{\mp})_{\alpha i} U_{i\beta}^{\pm,\nu} + \sum (M^{\pm})_{\beta i} U_{\alpha i}^{\mp\mu} . \tag{3.17}$$

We have now derived a set of equations for the ensemble average of the density matrix elements and their quadratic forms which describes the interaction of two fluctuating pump beams with the medium. These equations can be used to study a variety of phenomena. For example, the forward-scattering Hanle effect (where the probe beam has been suppressed by a crossed analyzer) can be described using the W^+ elements whereas, as seen in Sec. IV, the four-wave mixing is determined using the W^- contributions in (3.11). In the analysis given above the pump beam could be of arbitrary intensity.

IV. THE EFFECT OF LASER TEMPORAL FLUCTUATIONS ON PRESSURE-INDUCED RESONANCES AND THE GENERATION OF FLUCTUATION-INDUCED RESONANCES

In Sec. III we calculated the mean values of the quadratic forms involving density matrix elements. These forms can be used to obtain the intensity in the direction $(2\vec{k}_1 - \vec{k}_2)\, v/\omega$, where ω represents a typical frequency at which the system radiates. Since the phase-matching conditions are crucial for the production of a coherent signal, it is important to know the frequencies at which the system radiates. If lasers were strictly monochromatic, then the system would radiate (coherently) only at $2\omega_1 - \omega_2$ in the direction $(2\vec{k}_1 - \vec{k}_2)\, v/(2\omega_1 - \omega_2)$. The fluctuations of the field induce the system to radiate at other frequencies as well. To see this, let us consider the simplest possible situation involving linear susceptibilities, wherein the relationship between the induced polarization and the impressed electric field, at ω_l, is

$$P(\omega) = \chi^{(1)}(\omega) E(\omega) . \tag{4.1}$$

On using the Wiener-Khintchine theorem and (4.1), we find that the spectrum $S_p(\omega)$ of polarization fluctuations is related to the spectrum $\Gamma(\omega)$ of the applied field by

$$S_p(\omega) = |\chi^{(1)}(\omega)|^2 \Gamma(\omega) \tag{4.2}$$

which goes over to

$$\to |\epsilon|^2 |\chi^{(1)}(\omega_l)|^2 \delta(\omega - \omega_l)$$

in the limit of monochromatic incident fields. The equation (4.2) shows at which additional frequencies the system can radiate due to the fluctuations in the incident field. In fact, the system radiates at all the natural frequencies [determined by the poles of $\chi^{(1)}(\omega)$] as well as at ω_l. In the absence of laser fluctuations only the coherent component at ω_l is produced. Thus some fluctuations are needed to see atomic frequency excitations. A similar situation occurs in the context of four-wave mixing, details of which will be discussed in the next section using $\chi^{(3)}$. It may be noted that the recent four-wave mixing experiments pick up the contributions at frequencies in the immediate neighborhood of $(2\omega_1 - \omega_2)$. In order to relate our theory to such experiments, we obviously have to calculate the details of the spectrum in the direction $(2\vec{k}_1-\vec{k}_2)v/(2\omega_1-\omega_2)$ and in the neighborhood of $(2\omega_1-\omega_2)$. It turns out, however, that our calculations of the complete spectrum show also the possibility of a fluctuation-induced coherent signal which is produced in a direction different from $(2\vec{k}_1-\vec{k}_2)v/(2\omega_1-\omega_2)$.

In order to calculate the spectrum of polarization fluctuations and thereby the spectrum of the emitted radiation, it is convenient to rewrite (3.2) as a 9×9 matrix equation with the ninth element of ψ defined as unity:

$$\dot{\psi}^{(\nu)}=L\psi^{(\nu)}+\exp[i\delta t_\mu-i(\theta_\nu-q_{\nu\mu})+i\Phi(t_\nu)]K_-\psi^{(\nu)} +\exp[-i\delta t_\mu+i(\theta_\nu-q_{\nu\mu})-i\Phi(t_\nu)]K_+\psi^{(\nu)}+i\mu_1(t_\nu)f\psi^{(\nu)}\,, \tag{4.3}$$

where

$$L=\begin{bmatrix} M & I\\ 0 & 0\end{bmatrix},\quad K_\pm=\begin{bmatrix} M^\pm & I_\pm\\ 0 & 0\end{bmatrix},\quad f=\begin{bmatrix} f & 0\\ 0 & 0\end{bmatrix}. \tag{4.4}$$

Note that for the purpose of the evaluation of the spectrum we have to know the time correlation function $\langle P_\alpha^{(\nu)}(t+\tau)P_\beta^\mu(t)\rangle$. Equations for such correlation functions can be constructed using (4.3) and the results from the theory of multiplicative stochastic processes:

$$\frac{d}{dt_\mu}\langle\psi^{(\nu)}g\rangle=L\langle\psi^{(\nu)}g\rangle-\gamma_{c1}f^2\langle\psi^{(\nu)}g\rangle+e^{i\delta t_\mu-i(\theta_\nu-q_{\nu\mu})}K_-\langle\psi^{(\nu)}e^{i\Phi_\nu}g\rangle +e^{i\delta t_\mu+i(\theta_\nu-q_{\nu\mu})}K_+\langle\psi^{(\nu)}e^{-i\Phi_\nu}g\rangle\,, \tag{4.5}$$

where

$$\langle\psi^{(\nu)}g\rangle=\lim_{\tau\to\infty}\langle\psi^{(\nu)}(t+\tau)g(\tau)\rangle\,. \tag{4.6}$$

Here we focus our attention on the steady-state results, though transient response can also be calculated by similar methods. The equations for correlation functions can be solved to various orders in λ. The results to second order in λ will be presented below. Using (4.5), we obtain the set of equations, for various correlation functions to different orders in λ,

$$\frac{d}{dt_\mu}\langle\psi^{(\nu)}e^{i\Phi_\nu}g\rangle^{(2)}=[L-\gamma_{c1}(f+1)^2]\langle\psi^{(\nu)}e^{i\Phi_\nu}g\rangle^{(2)}+K_-e^{-i(\theta_\nu-q_{\nu\mu})}\langle\psi^{(\nu)}e^{2i\Phi_\nu+i\delta t_\mu}g\rangle^{(1)} +K_+e^{i(\theta_\nu-q_{\nu\mu})}\langle\psi^{(\nu)}e^{-i\delta t_\mu}g\rangle^{(1)}\,, \tag{4.7}$$

$$\frac{d}{dt_\mu}\langle\psi^{(\nu)}e^{2i\Phi_\nu+i\delta t_\mu}g\rangle^{(1)}=[L-\gamma_{c1}(f+2)^2+i\delta]\langle\psi^{(\nu)}e^{2i\Phi_\nu+i\delta t_\mu}g\rangle^{(1)}+K_-e^{-i(\theta_\nu-q_{\nu\mu})}\langle\psi^{(\nu)}e^{3i\Phi_\nu+2i\delta t_\mu}g\rangle^{(0)} +K_+e^{i(\theta_\nu-q_{\nu\mu})}\langle\psi^{(\nu)}e^{i\Phi_\nu-i\delta t_\mu}g\rangle^{(0)}\,, \tag{4.8}$$

$$\frac{d}{dt}\langle\psi^{(\nu)}e^{-i\delta t_\mu}g\rangle^{(1)}=[L-\gamma_{c1}(f^2)-i\delta]\langle\psi^{(\nu)}e^{-i\delta t_\mu}g\rangle^{(1)}+K_-e^{-i(\theta_\nu-q_{\nu\mu})}\langle\psi^{(\nu)}e^{i\Phi_\nu}g\rangle^{(0)} +K_+e^{i(\theta_\nu-q_{\nu\mu})}\langle\phi^{(\nu)}e^{-i\Phi_\nu-2i\delta t_\mu}g\rangle^{(0)}\,, \tag{4.9}$$

$$\frac{d}{dt}\langle\psi^{(\nu)}e^{in\Phi_\nu}g\rangle^{(0)}=[L-\gamma_{c1}(f+n)^2]\langle\psi^{(\nu)}e^{in\Phi_\nu}g\rangle^{(0)}\,. \tag{4.10}$$

All the relevant steady-state values have been computed in Sec. III. On denoting the Laplace transform of $\psi(t)$ by $\hat{\psi}(z)$, we find that (4.7) leads to

$$\mathscr{L}\langle\psi^{(\nu)}e^{i\Phi}g\rangle^{(2)}=[Z-L+\gamma_{c1}(f+1)^2]^{-1} \times\{\langle\psi^{(\nu)}e^{i\Phi_\nu}g\rangle^{(2)}+K_-e^{-i(\theta_\nu-q_{\nu\mu})}[Z-L+\gamma_{c1}(f+2)^2-i\delta]^{-1}\langle\psi^{(\nu)}e^{2i\Phi_\nu+i\delta t_\mu}g\rangle^{(1)} +K_+e^{i(\theta_\nu-q_{\nu\mu})}(Z-L+\gamma_{c1}f^2+i\delta)^{-1}\langle\psi^{(\nu)}e^{-i\delta t_\mu}g\rangle^{(1)}\}\,, \tag{4.11}$$

where we have made use of the property [cf. Eq. (3.6)]

$$\langle \psi^{(\nu)} e^{in\Phi_\nu} g \rangle^{(0)} = 0 \quad \text{for } n \neq 0 \; . \tag{4.12}$$

If we now define the Laplace transform of the correlation functions by

$$S_{pg}(Z) \equiv \int_0^\infty dt\, e^{-Zt} \lim_{\tau\to\infty} \langle \psi_p^{(\nu)}(t+\tau)\psi_g^{(\mu)}(\tau)\rangle \; , \tag{4.13}$$

then we see, following the arguments similar to those in Sec. III [cf. Eq. (3.11)], that these transforms have the structure

$$S_{pg}(Z) = e^{-i(\theta_\mu - \theta_\nu + q_{\nu\mu})} S_{pg}^{(+)} + e^{i(\theta_\mu - \theta_\nu + q_{\nu\mu})} S_{pg}^{(-)} + \cdots \; . \tag{4.14}$$

The term relevant for four-wave mixing is $S_{pg}^{(-)}$. In order to simplify (4.10), we express 9×9 matrices like $(Z-L)^{-1}$ in terms of 8×8 matrices $(Z-M)^{-1}$ using the relation

$$\left[Z - \begin{bmatrix} A & B \\ 0 & C \end{bmatrix} \right]^{-1} = \begin{bmatrix} (Z-A)^{-1} & (Z-A)^{-1}B(Z-C)^{-1} \\ 0 & (Z-C)^{-1} \end{bmatrix} , \tag{4.15}$$

where C is a number. A lengthy calculation using (4.11)–(4.15) then leads to

$$\begin{aligned} S_{pg}^{(-)} = {} & \sum_j [Z - M + \gamma_{c1}(F+1)^2]_{pj}^{-1} W_{gj}^{-} \\ & + \sum_{jrl} [Z - M + \gamma_{c1}(F+1)^2]_{pj}^{-1} M_{jr}^{-1} [Z - M + \gamma_{c1}(F+2)^2 - i\delta]_{rl}^{-1} [U_{gl}^{\mu +} + I_l \psi_g^{+}(Z + 4\gamma_{c1} - i\delta)^{-1}] \\ & + \sum_j [Z - M + \gamma_{c1}(F+1)^2]_{pj}^{-1} (I_-)_j (Z + 4\gamma_{c1} - i\delta)^{-1} \psi_g^{+} \; , \end{aligned} \tag{4.16}$$

where ψ_g^+ is given by Eq. (3.7). Let us now relate the Laplace transform $S_{pg}^{(-)}$ to the physical spectrum. As indicated earlier, the time scale associated with the μth atom is $t - (\hat{k}_1 \cdot \vec{R}_\mu / v)$, whereas the time scale for the νth atom is $t - (\hat{k}_1 \cdot \vec{R}_\nu / v)$, so that the time scale associated with the νth atom relative to the μth atom will be $[\hat{k}_1 \cdot (\vec{R}_\mu - \vec{R}_\nu)/v]$. Thus in the actual calculation of the time correlation function we should change τ to

$$\{\tau + [\hat{k}_1 \cdot (\vec{R}_\mu - \vec{R}_\nu)/v]\} \; .$$

Note also that all the calculations have been done in a rotating frame, which would lead to factors like

$$\exp\{-i\omega_1[t - (\hat{k}_1 \cdot \vec{R}_\mu / v)]\} \exp\{i\omega_1[t - (\hat{k}_1 \cdot \vec{R}_\nu / v)]\} = \exp[i\omega_1 \hat{k}_1 \cdot (\vec{R}_\mu - \vec{R}_\nu)/v] \; .$$

However, since one should not double count the phase factors associated with $\hat{k}_1 \cdot (\vec{R}_\mu - \vec{R}_\nu)/v$ as these also occur in the equal-time expectation value (3.11), one can show that the physical two-time correlation can be obtained by just changing

$$\tau \to \tau + [\hat{k}_1 \cdot (\vec{R}_\mu - \vec{R}_\nu)/v]$$

in the correlation function computed from (4.14) and by dropping the phase factors like $e^{\pm q_{\nu\mu}}$.

Equation (4.16) is our final expression for the steady-state correlation function. The appearance of a large number of poles in (4.16) is significant. The pole structure is determined by the eigenvalues of the matrix M, the laser bandwidth and the parameter $\delta = \omega_1 - \omega_2$. The spectrum of the polarization fluctuations will be determined from a linear combination of terms like (4.16), and thus such a spectrum will have peaks at

$$\omega = \omega_1 + \text{Im}\beta_i$$

and (4.17)

$$\omega = \omega_1 + \text{Im}\beta_i + \delta \; ,$$

where the β_i's are the eigenvalues of the 8×8 matrix M. The additional ω_1 in (4.17) arises because the calculations leading to (4.16) were done in the rotating frame [cf. Eq. (2.4)]. Thus the emitted radiation, in principle, can have spectral peaks at $\omega_1 + \text{Im}\beta_i$, and $2\omega_1 - \omega_2 + \text{Im}\beta_i$ with the peak at $2\omega_1 - \omega_2$ corresponding to the usual four-wave mixing signal in the absence of laser fluctuations.

Let us now present an explicit relation between the spectrum of the emitted radiation and the polarization fluctuations. The spectrum is related to the two-time correlation function of the function

$$\sum \vec{p}^{(\mu)} \cdot \hat{\epsilon} e^{-i\vec{k}\cdot\vec{R}_\mu}$$

[cf. (2.14)], where $\vec{k}$ is the direction of observation and $\vec{p}^{(\mu)}$ is the positive-frequency part of the polarization produced by the μth atom. Using the standard relations between Fourier transforms and Laplace transforms and keeping in view the discussion following (4.17), one can show that the spectrum of the radiation in the direction

$$\{2\vec{k}_1-\vec{k}_2+[(\omega-2\omega_1+\omega_2)\hat{k}_1/v]\}v/\omega\equiv\hat{n}_\omega$$

will be

$$S_{\hat{n}_\omega}(\omega)=\frac{1}{\pi}\operatorname{Re}[\,|\vec{d}_{31}\cdot\hat{\epsilon}\,|^2S_{31}^{(-)}(z)+|\,\vec{d}_{32}\cdot\hat{\epsilon}\,|^2S_{42}^{(-)}(z)+(\vec{d}_{31}\cdot\hat{\epsilon})(\vec{d}_{32}^{\,*}\cdot\hat{\epsilon}^{\,*})S_{41}^{(-)}(z)$$
$$+(\vec{d}_{32}\cdot\hat{\epsilon})(\vec{d}_{31}^{\,*}\cdot\hat{\epsilon}^{\,*})S_{32}^{(-)}(z)]\,|_{z=i(\omega-\omega_1)}\ . \qquad (4.18)$$

The direction in which the coherent signal is produced depends, of course, on the resonance frequencies in $S_{\alpha\beta}^{(-)}$. Such resonant frequencies are given by (4.17). The area under any of these peaks can be obtained by evaluating the corresponding residue. For example, the total intensity in the direction $\hat{n}_\Omega$ due to the spectral peak at Ω, produced by a pole z_Ω of $S_{\alpha\beta}^{(-)}(z)$ such that $\operatorname{Im}z_\Omega=(\Omega-\omega_1)$, will be

$$I_{\hat{n}_\Omega}=\operatorname{Re}\lim_{z\to z_\Omega}(z-z_\Omega)[\,|\vec{d}_{31}\cdot\hat{\epsilon}\,|^2S_{31}^{(-)}(z)+|\,\vec{d}_{32}\cdot\hat{\epsilon}\,|^2S_{42}^{(-)}(z)+(\vec{d}_{31}\cdot\hat{\epsilon})(\vec{d}_{32}^{\,*}\cdot\hat{\epsilon}^{\,*})S_{41}^{(-)}(z)$$
$$+(\vec{d}_{32}\cdot\hat{\epsilon})(\vec{d}_{31}^{\,*}\cdot\hat{\epsilon}^{\,*})S_{32}^{(-)}(z)]\ . \qquad (4.19)$$

If we choose $\Omega=(2\omega_1-\omega_2)$, then (4.19) will be the usual four-wave mixing contribution. Note also that if the Ω's are widely separated, then the coherent signals will be produced only for certain Ω's. Thus if in a given situation the incident wave vectors are adjusted so that the phase-matching condition is satisfied for some Ω, then the coherent signal will be produced only for that particular value of Ω.

Having presented the general formulation for four-wave mixing effects in fluctuating fields, we now present the results of our numerical computations. We will simplify the analysis by assuming that the pump fields and the detunings are such that the saturation effects are not important. In order to relate to the experiments[8] on PIER, we use parameters appropriate to those used in experiments on sodium *D* lines:

$$\gamma_1=\gamma_2=\gamma,$$
$$\gamma/2\pi=10\text{ MHz},$$
$$\frac{\Delta_2}{\gamma}=\frac{\omega_{23}-\omega_1}{\gamma}=-1.5\times10^3\ ,$$
$$\omega_{12}/\gamma=5.1\times10^4,$$
$$\Delta_1-\Delta_2=\omega_{12},$$
$$\Gamma_1=\Gamma_1^{\text{ph}}+\tfrac{1}{2}\gamma\ ,$$
$$\Gamma_2=\Gamma_2^{\text{ph}}+\frac{\gamma}{2},$$
$$\Gamma_0=\Gamma_0^{\text{ph}}+\gamma,$$
$$\Gamma_1^{\text{ph}}\sim\Gamma_2^{\text{ph}}\sim\Gamma_0^{\text{ph}}\sim\Gamma_p\gamma\ ,$$

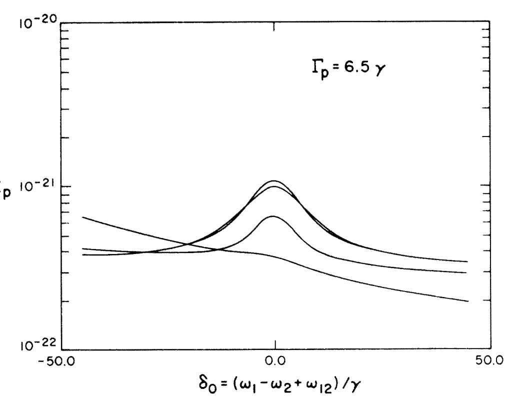

FIG. 2. The dependence of the PIER signal I_p, when $\omega_1-\omega_2=-\omega_{12}$, on collisional linewidth Γ_p and the laser (at ω_1) linewidth γ_{c1}, as a function of $\delta_0\equiv(\omega_1-\omega_2+\omega_{12})/\gamma\gamma_{c1}$ values for the four curves are, in increasing order (in units of the radiative linewidth γ), 0, 1, 10, and 100 with the uppermost curve corresponding to $\gamma_{c1}=0$. The collisional linewidth has been taken to be $\Gamma_p=6.5\gamma$. The actual δ_0 values for $\gamma_{c1}=10\gamma$ and 100γ are, respectively, 3 and 10 times those shown here.

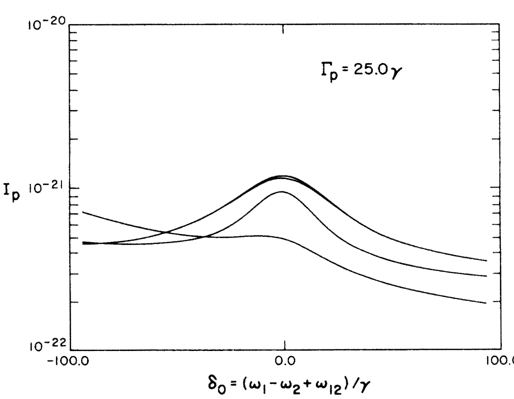

FIG. 3. Same as in Fig. 2 but $\Gamma_p=25\ \gamma$ and δ_0 values for $\gamma_{c1}=10\gamma$ and 100γ are, respectively, 2 and 5 times those shown here.

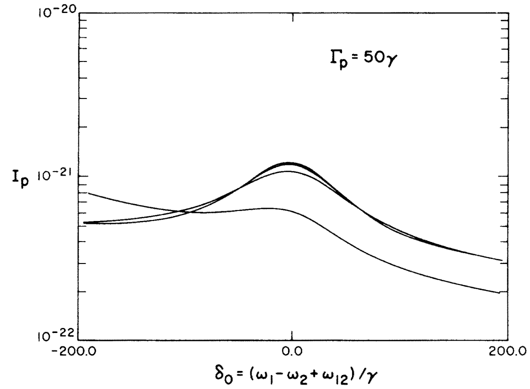

FIG. 4. Same as in Fig. 2 but $\Gamma_p = 50\gamma$ and δ_0 values for $\gamma_{c1} = 100$ are 2.5 times those shown here. The curves corresponding to $\gamma_{c1} = 0$ and γ are almost indistinguishable for such large values of Γ_p.

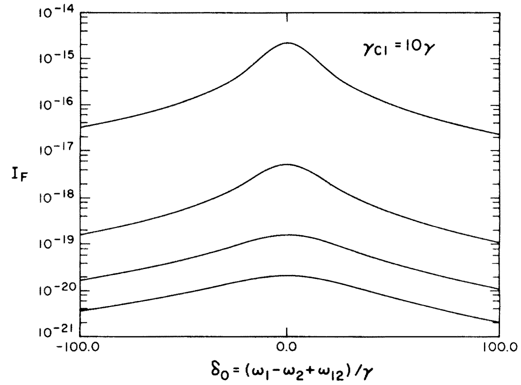

FIG. 6. Same as in Fig. 5, but for a moderate value of laser linewidth $\gamma_{c1} = 10\gamma$. Here δ_0 values for $\Gamma_p = 6.5\gamma$, 25γ, and 50γ are, respectively, 1.5, 2, and 3 times those shown here.

and we assume that there are no state-changing collisions.

We first examine the signal at $2\omega_1 - \omega_2$ and study the behavior of the pressure-induced extra resonance at $\omega_1 - \omega_2 = -\omega_{12}$ as a function of pressure and the pump linewidth. We denote this signal by I_p and show the results of numerical computations in Figs. 2–4. Since we are doing these computations by ignoring saturation effects, the only poles that contribute to the signal at $2\omega_1 - \omega_2$ with $\omega_{12} \cong -(\omega_1 - \omega_2)$ correspond to $\text{Im}z = \delta = (\omega_1 - \omega_2)$ and $\text{Im}z \sim -\omega_{12} \sim (\omega_1 - \omega_2)$. We have checked that in the absence of any collisions ($\Gamma_p = 0$), I_p does not show any resonant character at $\omega_1 - \omega_2 = -\omega_{12}$ even though the pump (laser at ω_1) has been assumed to have finite linewidth. As is seen from Figs. 2–4, the signal I_p broadens with peak values diminishing as the pump linewidth increases. For

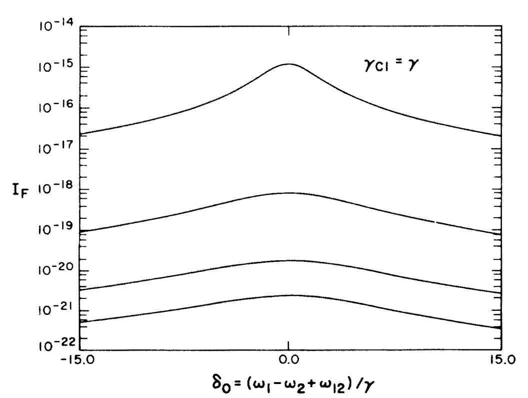

FIG. 5. The dependence of the FIER signal I_F when $\omega_1 - \omega_2 = -\omega_{12}$ on Γ_p and γ_{c1}. Here $\gamma_{c1} = \gamma$ and Γ_p values are in increasing order $\Gamma_p = 0$, 6.5γ, 25γ, and 50γ with the topmost curve corresponding to $\Gamma_p = 0$. The δ_0 values for $\Gamma_p = 6.5\gamma$, 25γ, and 50γ are, respectively, 3, 8, and 15 times those shown here.

large values of γ_{c1}, the resonant character becomes much less pronounced.

As mentioned earlier and as one can see from (4.19), laser temporal fluctuations can induce coherent signals at the atomic excitation frequencies. The question which now arises is can such a signal also exhibit the resonance at $\omega_1 - \omega_2 = -\omega_{12}$? A careful analysis of the denominators that appear in (4.16) shows that such a resonance is indeed possible if we choose $\Omega = \omega_{23}$. We denote such a resonant signal by I_F. It is easily seen that I_F arises from the poles in (4.16) such that $\text{Im}z \sim \Delta_2$, i.e., from eigenvalues β_i of the M matrix such that $\text{Im}\beta_i \sim \Delta_2$, or

$$\text{Im}\beta_i \sim \Delta_2 - \delta \sim \Delta_2 + \omega_{12} \sim \Delta_1 .$$

We will henceforth refer to such a resonance as "fluctuation-induced extra resonance" (FIER). In Figs. 5–7 the behavior of the FIER signal is displayed for various values of laser bandwidth and collisional parameters γ_{c1} and Γ_p. In the limit $\gamma_{c1} \rightarrow 0$, I_F vanishes. Signals I_F broaden with an in-

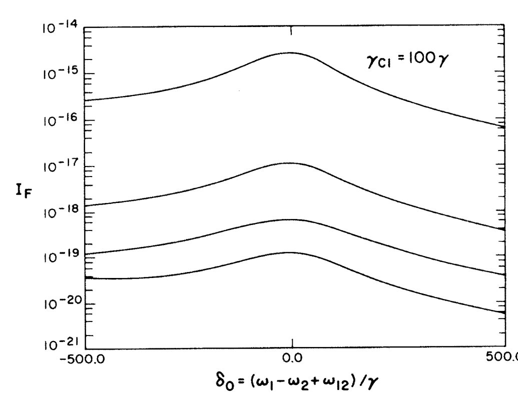

FIG. 7. Same as in Fig. 5, but for a large value of the laser linewidth $\gamma_{c1} = 100\gamma$. Here δ_0 values corresponding to $\Gamma_p = 50\gamma$ are 1.5 times those shown here.

crease in Γ_p. For moderate values of γ_{c1}, the peak height of I_F increases with γ_{c1}. The FIER peak height always decreases sharply with pressure, whereas the PIER peak height is roughly independent of pressure for $\Gamma_p \gg \gamma$. It is interesting to note, for example, from a comparison of Figs. 2 and 5, that the FIER contribution is much bigger than the PIER contribution for moderate values of pressure. It should be borne in mind that the PIER and FIER signals are produced in different directions since the phase-matching conditions for the two are different:

$$\hat{n}=(2\vec{k}_1-\vec{k}_2)v/|2\omega_1-\omega_2| \quad \text{(PIER)}$$

$$=[2\vec{k}_1-\vec{k}_2+\hat{k}_1(\omega_{23}-2\omega_1+\omega_2)/v]/|\omega_{23}| \quad \text{(FIER)} \quad (4.20)$$

and hence either the PIER or the FIER signal can be seen.

V. APPLICATION OF THE THIRD-ORDER SUSCEPTIBILITY $\chi^{(3)}(\omega_a,\omega_b,-\omega_c)$ TO THE STUDY OF THE EFFECT OF LASER BANDWIDTH ON FOUR-WAVE MIXING SIGNALS

In Sec. IV, we have seen how fluctuations can induce extra resonances which were also shown to have a different type of behavior than the PIER signal when the buffer gas pressure is increased. In order to understand some of these features, it is instructive to examine the general form of the third-order nonlinear susceptibility. The general form[7] of $\chi^{(3)}$ contains a large number of terms; however, a number of these drop out since we work with the Hamiltonian in the rotating wave approximation. In this case the $\chi^{(3)}$ of Bloembergen *et al.* simplifies to

$$\chi^{(3)}_{\mu\alpha\beta\gamma}(-\omega_p,\omega_a,\omega_b,-\omega_c)=\frac{\hbar^3}{6}\sum_{kj}(d_{3k}d_{k3}d_{3j}d_{j3})\frac{\alpha\gamma\beta\mu}{(\omega_{j3}-\omega_p)(\omega_{k3}-\omega_a)(-\omega_a+\omega_c)}$$
$$+\frac{\beta\gamma\alpha\mu}{(\omega_{j3}-\omega_p)(\omega_{k3}-\omega_b)(-\omega_b+\omega_c)}+\frac{\alpha\mu\beta\gamma[1+K_2(\omega_b-\omega_c,\omega_a-\omega_c)]}{(\omega_{k3}-\omega_a)(\omega^*_{j3}-\omega_c)(\omega_b-\omega_c)}$$
$$+\frac{\beta\mu\alpha\gamma[1+K_2(\omega_a-\omega_c,\omega_b-\omega_c)]}{(\omega_{k3}-\omega_b)(\omega^*_{j3}-\omega_c)(\omega_a-\omega_c)}, \quad (5.1)$$

where $\alpha\mu\beta\gamma$ stands for $(d_{3k})_\alpha(d_{k3})_\mu(d_{3j})_\beta(d_{j3})_\gamma$ and the frequencies ω_{kj} are complex, i.e., ω_{kj} appearing in (5.1) are equal to $\omega_{kj}+i\Gamma_{kj}$, with Γ_{kj} representing the decay of the off-diagonal element ρ_{kj}. In Eq. (5.1), K_2 stands for

$$K_2(\Omega_1,\Omega_2)=\frac{i(\Gamma_{kj}-\Gamma_{kg}-\Gamma_{gj})\Omega_1}{(\omega_{kj}-\Omega_2)(\omega^*_{3k}+\omega_p)}, \quad \omega_p=\omega_a+\omega_b-\omega_c . \quad (5.2)$$

As emphasized by Bloembergen *et al.*, the terms involving K_2 vanish if the system has only radiative relaxation. We will now show how the above $\chi^{(3)}$ can be used to get results similar to those obtained in Sec. IV for the PIER and FIER signals.

It has been shown elsewhere[6] that the spectrum of the polarization fluctuations is related to the fourth-order correlation function of the field at ω_1 and second-order correlation function of the field at ω_2. If one further assumes that the field at ω_1 is Gaussian, then its fourth-order correlation function can be expressed in terms of the product of second-order correlations. Assuming further, for simplicity, that the field at ω_2 is a monochromatic field, then the spectrum of the emitted radiation in the direction $\hat{k}$ can be shown to be proportional to

$$S(\omega)=18\sum\int d\omega_a\int d\omega_b\chi^{(3)}_{i\alpha\beta\gamma}(\omega_a,\omega_b,-\omega_2)\chi^{(3)*}_{j\alpha'\beta'\gamma'}(\omega_a,\omega_b,-\omega_2)\hat{\epsilon}_i\hat{\epsilon}^*_j$$
$$\times\Gamma_{\alpha\alpha'}(\omega_a)\Gamma_{\beta\beta'}(\omega_b)\delta(\omega-\omega_a-\omega_b+\omega_2)\epsilon_{2\gamma}\epsilon^*_{2\gamma'}$$
$$\times\exp\left[-i(\vec{R}_\mu-\vec{R}_\nu)\cdot\left[\vec{k}+\vec{k}_2-\hat{k}_1\frac{n(\omega_a)\omega_a+n(\omega_b)\omega_b}{c}\right]\right]. \quad (5.3)$$

The factors involving atomic positions appear for reasons given in the Appendix. The multiplying factor 18 on the right-hand side of (5.3) is due to a numerical factor of 2 which arises from the Gaussian nature of the field at ω_1 and a factor of 9 which depends on the way susceptibilities are defined. The integrated spectrum I will be

$$I=\sum 18 \int d\omega_a \int d\omega_b \chi^{(3)}_{i\alpha\beta\gamma}(\omega_a,\omega_b,-\omega_2)\chi^{(3)*}_{j\alpha'\beta'\gamma'}(\omega_a,\omega_b,-\omega_2)\hat{\epsilon}_i\hat{\epsilon}_j^{*}\Gamma_{\alpha\alpha'}(\omega_a)\Gamma_{\beta\beta'}(\omega_b)\epsilon_{2\gamma}\epsilon^{*}_{2\gamma'} \times \exp\left[-i(\vec{R}_\mu-\vec{R}_\nu)\cdot\left[\vec{k}+\vec{k}_2-\hat{k}_1\frac{n(\omega_a)\omega_a+n(\omega_b)\omega_b}{c}\right]\right] . \tag{5.4}$$

For the Lorentzian spectrum of the field at ω_1 we have

$$\Gamma_{\alpha\alpha'}(\omega_a)=\frac{\gamma_{c1}\epsilon_{1\alpha}\epsilon^{*}_{1\alpha'}}{\pi[(\omega_a-\omega_1)^2+\gamma_{c1}^2]} . \tag{5.5}$$

Note that in the above we have assumed the field at ω_1 to be Gaussian, an assumption which is different from that of the model used in Secs. II–IV. One would, however, expect that the lowest-order results for four-wave mixing would be qualitatively similar for the two models, as far as the effect of laser bandwidth is concerned. Even in the rotating wave approximation $\chi^{(3)}$ contains a large number of terms which lead to a large number of spectral peaks in $S(\omega)$, as also discussed in Sec. IV, unless the laser at ω_1 is also monochromatic. We concentrate our attention on those terms from $\chi^{(3)}$ that are important in describing a given class of phenomena.

A. Pressure-induced extra resonance

Let us first examine how the fluctuations change the PIER signal. Picking the term corresponding to PIER from $\chi^{(3)}$, and for the moment concentrating our attention only on this term, we can write the PIER contribution $S^{(P)}(\omega)$ to the spectrum as

$$S_P(\omega)=T_P(\omega)(\Pi \text{ dipole matrix elements})N^2 \tag{5.6}$$

with

$$T_P(\omega)=\sum_{\mu,\nu}\int\int d\omega_a d\omega_b \delta(\omega-\omega_a-\omega_b+\omega_2)\frac{(\Gamma_0-\Gamma_1-\Gamma_2)^2}{N^2[(\omega_{13}-\omega_2)^2+\Gamma_1^2]}\frac{1}{[(\omega-\omega_{23})^2+\Gamma_2^2]}$$
$$\times\left[\frac{1}{(\omega_{21}-\omega_a+\omega_2)^2+\Gamma_0^2}\frac{1}{(\omega_{23}-\omega_a)^2+\Gamma_2^2} + \frac{1}{(\omega_{21}-\omega_a+\omega_2-i\Gamma_0)(\omega_{21}-\omega_b+\omega_2+i\Gamma_0)[(\omega_{23}-\omega_a)-i\Gamma_2][\omega_{23}-\omega_b+i\Gamma_2]}\right]$$
$$\times\frac{\gamma_{c1}}{\pi[\gamma_{c1}^2+(\omega_a-\omega_1)^2]}\frac{\gamma_{c1}}{\pi[\gamma_{c1}^2+(\omega_b-\omega_1)^2]}$$
$$\times\exp\left[-i(\vec{R}_\mu-\vec{R}_\nu)\cdot\left[\vec{k}+\vec{k}_2-\hat{k}_1\frac{n(\omega_a)\omega_a+n(\omega_b)\omega_b}{c}\right]\right] . \tag{5.7}$$

Here, for simplicity, we have dropped the tensorial indices. The four-wave mixing contribution can only arise from values of ω_a and ω_b in the integrand in the neighborhood of ω_1. So we look for poles in the vicinity $\omega_a \sim \omega_1$, $\omega_b \sim \omega_1$, and then the phase-matching condition for the PIER signal becomes $\vec{k}=2\vec{k}_1-\vec{k}_2$,

$$|k|=n(2\omega_1-\omega_2)|2\omega_1-\omega_2|/c .$$

It should be remembered that since we are looking for the PIER signal near $\omega_{12}\sim-(\omega_1-\omega_2)$, resonances like $[(\omega_{21}-\omega_a+\omega_2)^2+\Gamma_0^2]^{-1}$ in the integrand should be properly treated. Taking out all the slowly varying terms from the integrand, we find that the area T_P of the peak at $\omega=2\omega_1-\omega_2$ is

$$T_P\approx\frac{2(\Gamma_1+\Gamma_2-\Gamma_0)^2}{(\omega_{13}-\omega_2)^2(\omega_{23}-\omega_1)^2(2\omega_1-\omega_2-\omega_{23})^2}\left[\frac{[1+(\gamma_{c1}/2\Gamma_0)]}{(\gamma_{c1}+\Gamma_0)^2+(\omega_{21}-\omega_1+\omega_2)^2}\right] . \tag{5.8}$$

Equation (5.8) shows the effect of the laser fluctuations on one of the contributions to PIER. The overall kinematical factor 2 arises due to the assumed Gaussian nature of the field at ω_1, i.e., due to $\langle I^2\rangle = 2\langle I\rangle^2$. Note that the laser linewidth makes the character of the numerator in (5.8) a little more complicated; for example, the weight factor, instead of being $[1+(\gamma_{c1}/\Gamma_0)]$, is $[1+(\gamma_{c1}/2\Gamma_0)]$. There is another contribution to PIER that arises due to the interference between the background term and the PIER term. In fact, $\chi^{(3)}$ as far as the behavior of the PIER signal (resonance at $\omega_1-\omega_2=-\omega_{12}$) is concerned, can be approximated by

$$\frac{1}{(\omega_{13}-\omega_2)(\omega_{23}-\omega_1)(2\omega_1-\omega_2-\omega_{23})}\left[1+\frac{i}{2}(\Gamma_0-\Gamma_1-\Gamma_2)\left[\frac{1}{(\omega_{21}-\omega_a+\omega_2-i\Gamma_0)}+\frac{1}{(\omega_{21}-\omega_b+\omega_2)-i\Gamma_0}\right]\right].$$

Then the full contribution to PIER can be shown to be

$$T_P \approx \frac{2}{(\omega_{13}-\omega_2)^2(\omega_{23}-\omega_1)^2(2\omega_1-\omega_2-\omega_{23})^2} \times \left[1+\frac{(\Gamma_1+\Gamma_2-\Gamma_0)}{(\omega_{21}-\omega_1+\omega_2)^2+\Gamma_0^2}\left[(\Gamma_1+\Gamma_2+\Gamma_0)+\frac{\gamma_{c1}}{2\Gamma_0}(\Gamma_1+\Gamma_2+3\Gamma_0)\right]\right]. \tag{5.9}$$

If $\gamma_c=0$, then (5.9) leads to the standard expression. Note that there is no simple substitution rule like $\Gamma_0\to\Gamma_0+\gamma_{c1}$ that would enable one to obtain a result like (5.9) from the result in the absence of laser bandwidth effects. Equation (5.9) also agrees with the result[13] obtained recently from an effective two-level description of PIER. The peak height behaves as

$$\left[\frac{\Gamma_1+\Gamma_2}{\Gamma_0}-1\right]\left[\frac{\Gamma_1+\Gamma_2}{\Gamma_0}+1+\frac{\gamma_{c1}}{2\Gamma_0}\left[\frac{\Gamma_1+\Gamma_2}{\Gamma_0}+3\right]\right].$$

For the Na experiments of Bloembergen and coworkers,

$$\Gamma_1=\tfrac{1}{2}\gamma+\kappa p,\quad \Gamma_2=\tfrac{1}{2}\gamma+\kappa p\ , \qquad \Gamma_0=\gamma+\kappa p\ , \tag{5.10}$$

where p is the pressure and γ^{-1} gives the lifetime of states $|1\rangle$ and $|2\rangle$. Since the peak height of PIER is

$$\left[1+\frac{\gamma}{\kappa p}\right]^{-2}\left[3+\frac{2\gamma}{\kappa p}+\frac{(\gamma_{c1}/\kappa p)}{2[1+(\gamma/\kappa p)]}\left[\frac{4\gamma}{\kappa p}+5\right]\right],$$

and for large values of pressure $\kappa p \gg \gamma$, the peak height goes as

$$\left[3+\frac{5}{2}\left[\frac{\gamma_{c1}}{\kappa p}\right]\right]$$

suggesting that the effect of linewidth could be significant if $\gamma_{c1}\sim\kappa p$. The area under the peak is then

$$\frac{(\Gamma_1+\Gamma_2-\Gamma_0)\pi}{(\Gamma_0+\gamma_{c1})} \times \left[(\Gamma_1+\Gamma_2+\Gamma_0)+\frac{\gamma_{c1}}{2\Gamma_0}(\Gamma_1+\Gamma_2+3\Gamma_0)\right],$$

which in the limit of large pressure $\kappa p \gg \gamma$, reduces to

$$\pi\kappa p\left[1+\frac{\gamma_{c1}}{\kappa p}\right]^{-1}\left[3+\frac{5}{2}\frac{\gamma_{c1}}{\kappa p}\right].$$

It has to be remembered that this result holds only for the Gaussian model of the laser fluctuations and is in contrast to the behavior exhibited by the phase-diffusion model of the laser.

B. Fluctuation-induced extra resonance (FIER)

We next show that due to laser fluctuations one can see the resonance at $\omega_{12}=\omega_2-\omega_1$ in four-wave mixing experiments. The four-wave signal is not at a combination $2\omega_1-\omega_2$ of the impressed external fields at ω_1 and ω_2 but at one of the natural frequencies of the system. Specifically, such a signal is at the frequency $\omega=\omega_{23}$. Since the new resonance at $\omega_{12}=\omega_2-\omega_1$ arises due to laser fluctuations, we will term this the fluctuation-induced extra resonance. Of course, such a resonance will *not* contribute to the usual four-wave mixing experiments since in the usual experiments, one looks at the signal at $2\omega_1-\omega_2$, and the frequencies ω_{23} and $2\omega_1-\omega_2$ could differ considerably and in addition the phase-matching conditions for the two signals are different. For example, in Na experiments where ω_1, say, is near ω_{23} and ω_2 near ω_{13}, then $(2\omega_2-\omega_1)$ and ω_{23} would differ by $\sim 17\ \text{cm}^{-1}$. We will show that the new resonance will have very different pressure dependence than the PIER. This new term arises from $(\omega_{23}-\omega)^{-1}(\omega_{13}-\omega_b)^{-1}(-\omega_b+\omega_2)^{-1}$ in $\chi^{(3)}$. Denoting the contribution of such a term to $S(\omega)$ by $T_F(\omega)$, we have

$$T_F(\omega)=\sum_{\mu,\nu}\frac{2}{(\omega_{23}-\omega)^2+\Gamma_2^2}\times\int d\omega_a\frac{\gamma_{c1}}{\pi[\gamma_{c1}^2+(\omega_a-\omega_1)^2]}\frac{\gamma_{c1}}{N^2\pi[\gamma_{c1}^2+(\omega+\omega_2-\omega_a-\omega_1)^2]}\times\frac{1}{(\omega-\omega_a)^2}\frac{1}{[(\omega_{13}-\omega-\omega_2+\omega_a)^2+\Gamma_1^2]}\times\exp\left[-i(\vec{R}_\mu-\vec{R}_\nu)\cdot\left[\vec{k}+\vec{k}_2-\hat{k}_1\frac{n(\omega_a)\omega_a+(\omega+\omega_2-\omega_a)n(\omega+\omega_2-\omega_a)}{c}\right]\right], \tag{5.11}$$

which shows a spectral peak at $\omega=\omega_{23}$. The phase-matching condition for this spectral peak is

$$n(\omega_{23})(\omega_{23}/c)\hat{k}+\vec{k}_2=\hat{k}_1[n(\omega_1)\omega_1+(\omega_{23}+\omega_2-\omega_1)n(\omega_{23}+\omega_2-\omega_1)/c]\ .$$

Such a spectral peak exists only in the presence of laser fluctuations, since in the limit $\gamma_{c1}\to 0$, the integral in (5.11) leads to a single spectral peak at $\omega=2\omega_1-\omega_2$. The area T_F under the spectral peak at $\omega=\omega_{23}$ will be

$$T_F\approx\frac{(2)(\pi)\gamma_{c1}}{\pi\Gamma_2}\int d\omega_a\frac{\gamma_{c1}}{\pi[\gamma_{c1}^2+(\omega_a-\omega_1)^2]}\frac{1}{(\omega_{23}-\omega_a)^2}\times\frac{1}{[\gamma_{c1}^2+(\omega_{23}+\omega_2-\omega_a-\omega_1)^2]}\frac{1}{(\omega_{13}-\omega_{23}-\omega_2+\omega_a)^2+\Gamma_1^2}\ . \tag{5.12}$$

Expression (5.12) obviously leads to a resonant contribution at $\omega_{12}=\omega_2-\omega_1$, due to the pole of the integrand at $\omega_a\sim\omega_1$:

$$T_F\approx 2\left[\frac{\gamma_{c1}}{\Gamma_2}\right]\frac{[1+(\gamma_{c1}/\Gamma_1)]}{(\omega_{23}-\omega_1)^2(\omega_{23}-2\omega_1+\omega_2)^2[(\omega_1-\omega_2-\omega_{21})^2+(\Gamma_1+\gamma_{c1})^2]}+\cdots, \tag{5.13}$$

where the ellipses represents other nonresonant terms. The peak height of the FIER goes as

$$\left[\frac{\gamma_{c1}}{\Gamma_2}\right]\frac{1}{\Gamma_1(\Gamma_1+\gamma_{c1})},$$

which for large pressure ($\kappa p\gg\gamma$) goes as $(\gamma_{c1})/(\kappa p)^3[1+(\gamma_{c1}/\kappa p)]$, while the width

$$\sim[(\gamma_{c1})+(\gamma/2)+\kappa p]\to\kappa p[1+(\gamma_{c1}/\kappa p)]\ .$$

The area under the FIER peak is $\gamma_{c1}/\Gamma_1\Gamma_2\sim\gamma_{c1}/(\kappa p)^2$. The pressure dependence of the FIER signal is in distinct contrast to that of the PIER signal. Our numerical results, presented in Sec. IV, are in agreement with such a pressure dependence. On comparing (5.9) and (5.13), we see that

$$T_P^{\text{peak}}/T_F^{\text{peak}}\sim\left[\frac{\gamma_{c1}}{\Gamma_2}\right]^{-1}\frac{\Gamma_1(\Gamma_1+\gamma_{c1})}{(\omega_{13}-\omega_2)^2}\left[\frac{\Gamma_1+\Gamma_2}{\Gamma_0}-1\right]\left[\frac{\Gamma_1+\Gamma_2}{\Gamma_0}+1+\frac{\gamma_{c1}}{2\Gamma_0}\left[\frac{\Gamma_1+\Gamma_2}{\Gamma_0}+3\right]\right]. \tag{5.14}$$

Since generally $\Gamma_1/(\omega_{13}-\omega_2)\ll 1$, it is clear from (5.14) that T_F would dominate over T_P. As noted earlier [following Eqs. (5.7) and (5.11); see also Eq. (4.20) which is a special case of these phase-matching conditions when $n(\omega)\approx n$, $v=c/n$] the phase-matching conditions for the two signals are different and hence it is possible to discriminate between the two signals experimentally.

We have thus shown how many of the results of numerical work in Sec. IV can be qualitatively understood in terms of the general form of the nonlinear susceptibility $\chi^{(3)}(\omega_p,\omega_a,\omega_b,-\omega_c)$. The quantitative results can only be obtained by using the full expression for $\chi^{(3)}$ and by evaluating the integral in (5.3) exactly.

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APPENDIX: STRUCTURE OF A PLANE WAVE WITH FLUCTUATING ENVELOPE

In this appendix we discuss the structure of a plane wave whose envelope is a stochastic function. The structure should be consistent with Maxwell's equations. Consider a plane wave

$$\vec{\mathrm{E}}=\vec{\epsilon}e^{-i\omega_1 t+i\vec{\mathrm{k}}_1\cdot\vec{\mathrm{r}}}+\mathrm{c.c.}\ , \tag{A1}$$

whose field envelope $\vec{\epsilon}$ is a slowly varying function of space and time and is fluctuating. Then, on making the Fourier decomposition of $\vec{\mathrm{E}}$ and using the Maxwell equations, we find that $\vec{\mathrm{E}}$ must have the form

$$\vec{\mathrm{E}}=\int d\omega\,\vec{\tilde{\epsilon}}(\omega)\exp(-i\omega\{t-[n(\omega)\hat{k}\cdot\vec{\mathrm{r}}/c]\}),\ \ \tilde{\epsilon}^*(\omega)=\tilde{\epsilon}(-\omega)\ , \tag{A2}$$

where $n(\omega)$ is the refractive index of the medium at frequency ω. We have thus expressed the field as a superposition of plane waves all propagating in the direction $\hat{k}$ but having different frequencies. Clearly the wave equation $[\nabla^2+n^2(\omega)\omega^2/c^2]\vec{\mathrm{E}}=0$ for $\vec{\mathrm{E}}$ is satisfied. $\vec{\tilde{\epsilon}}(\omega)$ is now a stochastic variable and hence the correlation function of $\vec{\mathrm{E}}$ can be written as

$$\begin{aligned}\langle\vec{\mathrm{E}}(\vec{\mathrm{R}}_\mu,t)\vec{\mathrm{E}}(\vec{\mathrm{R}}_\nu,t+\tau)\rangle=\int\int d\omega_1 d\omega_2\langle\vec{\tilde{\epsilon}}(\omega_1)\vec{\tilde{\epsilon}}(\omega_2)\rangle\\ \times\exp(-i\omega_1\{t-[n(\omega_1)\hat{k}\cdot\vec{\mathrm{R}}_\mu/c]\})\exp(-i\omega_2\{t+\tau-[n(\omega_2)\hat{k}\cdot\vec{\mathrm{R}}_\nu/c]\})\ ,\end{aligned} \tag{A3}$$

which, on using the Wiener-Khintchine theorem

$$\langle\tilde{\epsilon}(\omega_1)\tilde{\epsilon}(\omega_2)\rangle=\delta(\omega_1+\omega_2)\tilde{\Gamma}(\omega_1)$$

and $n(-\omega)=n^*(\omega)$, reduces to

$$\langle E_\alpha(\vec{\mathrm{R}}_\mu,t)E_\beta(\vec{\mathrm{R}}_\nu,t+\tau)\rangle=\int d\omega\,\tilde{\Gamma}_{\alpha\beta}(\omega)\exp\{i\omega\tau+[i\omega n(\omega)\hat{k}/c]\cdot(\vec{\mathrm{R}}_\mu-\vec{\mathrm{R}}_\nu)\}\ , \tag{A4}$$

where we have also assumed that $n(\omega)$ is real. In particular for equal time ($\tau=0$), we get

$$\langle E_\alpha(\vec{\mathrm{R}}_\mu,t)E_\beta(\vec{\mathrm{R}}_\nu,t)\rangle=\int d\omega\,\tilde{\Gamma}_{\alpha\beta}(\omega)\exp[i\omega n(\omega)(\hat{k}/c)\cdot(\vec{\mathrm{R}}_\mu-\vec{\mathrm{R}}_\nu)]\ . \tag{A5}$$

The phase factors that appear in (5.3) are due to the structure (A5) of the equal-time correlation function of the field. The results can be further simplified if the dispersion of $n(\omega)$ can be ignored. In such a case it is clear from the foregoing that for the phase-diffusion model of the laser, we should write the field in the form

$$\vec{\mathrm{E}}=\vec{\epsilon}\exp\{-i\omega_1[t-(\hat{k}_1\cdot\vec{\mathrm{r}}/v)]-i\Phi_1[t-(\hat{k}_1\cdot\vec{\mathrm{r}}/v)]\}+\mathrm{c.c.},\ \ v=\frac{c}{n} \tag{A6}$$

where $\Phi_1(x)$ represents a function of variable x. This is the form we have used in our analysis in Sec. II.

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