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# Effect of pump fluctuations on line shapes in coherent anti-Stokes Raman scattering

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The theory of coherent anti-Stokes Raman scattering (CARS) is extended to include the effect of pump fluctuations. The intensities and spectra of lines in resonant CARS are calculated to all orders in fields assuming a phase-diffusion model for waves at the two pump frequencies. The bandwidth of the two lasers enters in a much more complicated way than following a simple scaling of $T_1$ or $T_2$. Various resonances in CARS spectra due to dynamic splitting of the energy levels are discussed for a range of detunings, field intensities, and bandwidths. In contrast to the usual spectra in strong fields, the Rabi sidebands appear as dispersion-shaped structures. The laser linewidth is shown to change dramatically the CARS line shape. The case of no saturation is also treated, thus allowing for the inclusion of more general line shapes and fluctuations of the pump waves and the nonlinear susceptibility tensor $\chi^{(3)}$. Gaussian statistics of the pump field are shown to lead to enhancement factors in CARS intensity, similar to those appearing in the context of multiphoton absorption processes.

## I. INTRODUCTION

Four-wave mixing is being increasingly used to study the properties of atomic-molecular systems.[1,2] Important applications in producing phase conjugated signals have also been found.[3] Bloembergen, Lotem, and Lynch[1] recently studied the most general form of the third-order, nonlinear susceptibility $\chi^{(3)}$ valid for arbitrary relaxation parameters of the system. They also discussed the additional resonances that $\chi^{(3)}$ can have, due to the nonradiative nature of relaxations, and such resonances have recently been seen by Prior, Bogdan, Dagenais, and Bloembergen.[4] It is quite clear from the discussion of Bloembergen *et al.*[1] and the recent observation of Prior *et al.*[4] that a proper discussion of line shapes could be given provided all sources of relaxations have been accounted for. Recently the calculations of line shapes in coherent anti-Stokes Raman scattering (CARS) have been extended by Henesian and Byer[5] to include Doppler broadening. However, very little has been done as far as the effect of laser temporal fluctuations on the CARS line shape is concerned, as most treatments assume the incident fields to be strictly monochromatic (see, however, the work of Dutta[6]). In this paper we shall show that such fluctuations of the pump waves could significantly affect the nature of the CARS line shape, particularly in resonant situations. The CARS line shape is shown to be very sensitive to the fluctuations in the pump laser at $\omega_1$. We also examine some intensity dependent effects as they also form a source of linewidth in certain circumstances. In particular, we discuss the structure of CARS spectra when the fields are strong enough to saturate the Raman transition. In this case, Rabi sidebands are shown to appear as dispersion-shaped structures. The changes in the Raman spectra due to the increase in pump intensities and laser temporal fluctuations have already been discussed in detail by several workers.[7,8] Raymer *et al.*[8] has also considered the situation where propagation effects are important. In the main body of this paper we deal with a specific model of the laser and the resonant CARS, as in this case exact results for CARS signals and spectra can be obtained. The nonresonant part of $\chi^{(3)}$ leads to important interference effects in CARS. In the Appendix we discuss how some general results on line shape can be obtained using the general form of $\chi^{(3)}$, provided we ignore the saturation effects. We also comment on the generalizations to higher-order CARS[9] in fluctuating fields.

The basic equations describing resonant CARS have been obtained by several workers and good reviews exist.[2] Figure 1 gives schematically the energy levels of the system and various transitions involved in the generation of the CARS signal. The field amplitude of the CARS signal at frequency $\omega_a = 2\omega_1 - \omega_2$ can be written as

$$\vec{E}_a(t) = \mathscr{E}_a(t) e^{i\vec{k}_a \cdot \vec{r} - i(2\omega_1 - \omega_2)t} + \text{c.c.} , \tag{1.1}$$

and the pump fields are given by similar expressions,

$$\vec{E}_j(t) = \vec{\mathscr{E}}_j(t) e^{i\vec{k}_j \cdot \vec{r} - i\omega_j t} + \text{c.c.}, \quad j = 1,2 . \tag{1.2}$$

The difference in the frequencies of the pump fields $\omega_1$, $\omega_2$ is assumed to be close to the Raman resonance frequency $\omega_R$, i.e., $\omega_1 - \omega_2 \cong \omega_R$. The growth of $\mathscr{E}_a$ in the quasistatic approximation is described by[2,6]

$$\frac{\partial \mathscr{E}_a}{\partial z} = i\alpha Q \mathscr{E}_1(\eta) e^{i\Delta k z} \sigma_{12}(\eta) + \frac{i\alpha}{N} \mathscr{E}_1^2(\eta) \mathscr{E}_2^*(\eta) e^{i\Delta k z} \chi_{\text{NR}} , \tag{1.3}$$

$$\frac{\partial \sigma_{12}}{\partial \eta} = -\left[\frac{1}{T_2} - i\delta\right] \sigma_{12} + iq \mathscr{E}_1(\eta) \mathscr{E}_2^*(\eta) n , \tag{1.4}$$

$$\frac{\partial n}{\partial \eta} = -\frac{n - n_0}{T_1} + 2iq [\mathscr{E}_1^*(\eta) \mathscr{E}_2(\eta) \sigma_{12} - \text{c.c.}] , \tag{1.5}$$

where

$$\begin{aligned} \eta &= t - z/v , \\ \alpha &= \frac{2\pi N}{c}(2\omega_1 - \omega_2) , \\ \Delta \vec{k} &= \vec{k}_a + \vec{k}_2 - 2\vec{k}_1 , \\ \delta &= (\omega_1 - \omega_2 - \omega_R) , \\ Q &= \frac{1}{\hbar} \sum_n d_{1n} d_{n2} \left[\frac{1}{\omega_{n2} + \omega_2} + \frac{1}{\omega_{n2} - \omega_a}\right] , \\ q &= \frac{1}{\hbar^2} \sum_n d_{2n} d_{n1} \left[\frac{1}{\omega_{n2} - \omega_2} + \frac{1}{\omega_{n2} + \omega_1}\right] . \end{aligned} \tag{1.6}$$

Here $\chi_{\text{NR}}$ is the nonresonant part of the susceptibility $\chi^{(3)}$, $N$ is the number of atoms/molecules, and $T_1$ and $T_2$ are the longitudinal and transverse relaxation times. $\sigma_{12}$ is the off-diagonal element of the density matrix between levels 1 and 2 which

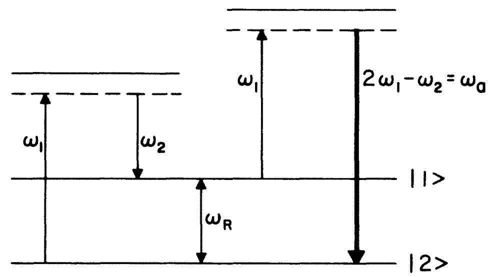


FIG. 1. Schematic of level configuration and the interaction of the pump fields at frequencies $\omega_1$ and $\omega_2$ in the generation of the CARS signal at frequency $\omega_a$.

determines the resonant contribution to the susceptibility $\chi^{(3)}$, $n$ is the difference between the populations of levels 1 and 2, $n_0$ is the equilibrium value of this difference, and $d_{ij}$ in the definitions of $Q$ and $q$ denotes the dipole moment matrix element between states $i$ and $j$. Equations (1.4) and (1.5) are the effective Bloch equations for the present problem with the effective field $\mathscr{E}_1 \mathscr{E}_2^*$. It may be noted that Bloch equations in the context of two-photon processes have been worked out and studied at length by several authors.[10] The coupling between the levels 1 and 2 is provided through intermediate transitions. It is assumed that all the waves have the same group velocity $v$ and that the amplitude $\mathscr{E}_a$ is much smaller than the amplitudes of the pump fields. From Eq. (1.3) we can immediately write

$$\mathscr{E}_a(z,\eta) = \left[\frac{e^{i\Delta k z} - 1}{i\Delta k}\right] i\alpha Q \mathscr{A}(\eta) \tag{1.7}$$

and

$$\mathscr{A}(\eta) = \left[\mathscr{E}_1(\eta)\sigma_{12}(\eta) + \mathscr{E}_1^2(\eta)\mathscr{E}_2^*(\eta)\frac{\chi_{\text{NR}}}{NQ}\right] . \tag{1.8}$$

Equations (1.4) and (1.5), in the limit of monochromatic and deterministic fields, lead to the following expression for $\mathscr{A}(\eta)$:

$$\mathscr{A}(\eta) = \mathscr{E}_1^2(\eta)\mathscr{E}_2^*(\eta)\left[\frac{\chi_{\text{NR}}}{NQ} + \frac{iqn_0}{(1/T_2) - i\delta}\right] . \tag{1.9}$$

On combining Eq. (1.9) with Eq. (1.7) we obtain the standard CARS signal.[2] It is clear from Eq. (1.7) that CARS intensity and line shape at a given point $z$ would be given by

$$\mathscr{I} \equiv \lim_{\eta\to\infty} \langle \mathscr{A}^*(\eta)\mathscr{A}(\eta)\rangle \alpha^2 Q^2 \left[\frac{\sin(\Delta k z/2)}{(\Delta k/2)}\right]^2 , \tag{1.10}$$

$$C_a(\tau) \equiv \lim_{\eta\to\infty} \langle \mathscr{A}^*(\eta)\mathscr{A}(\eta+\tau)\rangle \alpha^2 Q^2 \times \left[\frac{\sin(\Delta k z/2)}{(\Delta k/2)}\right]^2 . \tag{1.11}$$

The averaging in the above expressions is with respect to the fluctuations of the pump fields. In the following sections we show how $\mathscr{I}$ and $C_a(\tau)$ could be computed for arbitrary bandwidths and the intensities of the pump fields.

## II. DEPENDENCE OF CARS SIGNAL ON LASER LINEWIDTHS AND INTENSITIES

In this section we obtain CARS signals (intensities) as a function of the pump intensities, line widths, and the detuning $\delta$. Equations (1.3)–(1.5) are nonlinear stochastic equations, since both $\mathscr{E}_1(\eta)$ and $\mathscr{E}_2(\eta)$ are stochastic in nature. The stochastic nature of these fields can be taken to be prescribed by Langevin equations. In general, one does not expect the exact solutions of the set (1.3)–(1.5) for an arbitrary model of the pump unless one ignores saturation effects (see the Appendix). However, if we adopt the phase-diffusion model for each pump wave, which is characteristic of a laser operating far above threshold, then it is possible to obtain exact solutions. For this model the pump amplitudes can be written as

$$\mathscr{E}_j(\eta) = \mathscr{E}_j e^{-i\Phi_j(\eta)}, \quad j=1,2 \tag{2.1}$$

where $\Phi_j(\eta)$ represents a diffusion process, i.e.,

$$\frac{d\Phi_i}{d\eta} = \mu_i(\eta),$$
$$\langle \mu_i(\eta)\rangle = 0, \tag{2.2}$$
$$\langle \mu_i(\eta)\mu_j(\eta')\rangle = 2\gamma_i \delta_{ij}\delta(\eta-\eta') ,$$

and $\mu_j(\eta)$ represents a Gaussian random process. We have assumed that the pump waves at $\omega_1$, $\omega_2$ are uncorrelated. Such a model has been extensively used earlier in the calculations involving resonance fluorescence spectra,[11,12] and in optical double resonance.[13] Eberly[12] has emphasized the type of "substitution rules" that in some cases might be used to account for the pump fluctuations. However, there are many situations where no simple substitution rules can be used and each physical situation has to be analyzed separately. The calculation of the CARS signal turns out to be such a case.

From Eqs. (1.8) and (2.1), we find that

$$\mathscr{A}(\eta) = \mathscr{E}_1 \left[\sigma_{12}(\eta) e^{-i\Phi_1(\eta)} + \frac{\mathscr{E}_1\mathscr{E}_2\chi_{\mathrm{NR}}}{NQ} e^{-2i\Phi_1(\eta)+i\Phi_2(\eta)}\right] , \tag{2.3a}$$

and hence the intensity (1.10) can be rewritten as

$$\mathscr{I} = |\mathscr{E}_1|^2 \alpha^2 Q^2 \left[\frac{\sin(\Delta k z/2)}{(\Delta k/2)}\right]^2 I ,$$
$$I = \langle X_1 X_2\rangle + |B|^2 + B\langle X_2\rangle + B^*\langle X_1\rangle . \tag{2.3b}$$

Here we have introduced the following definitions:

$$X_1 = \sigma_{12} e^{i(\Phi_1-\Phi_2)} , \quad X_2 = X_1^* , \quad X_3 = n ,$$
$$B = \frac{-q_0}{b}, \quad q_0 = \mathscr{E}_1\mathscr{E}_2 q, \quad b = -\frac{NQq}{\chi_{\mathrm{NR}}} . \tag{2.4}$$

Using Bloch equations (1.4) and (1.5) and the model (2.1) and (2.2) for the pump waves, we obtain the following equations for the variables $X_i$'s:

$$\frac{\partial X_1}{\partial\eta} = -\left[\frac{1}{T_2} - i\delta - i\mu_1 + i\mu_2\right] X_1 + i q_0 X_3 , \tag{2.5a}$$

$$\frac{\partial X_3}{\partial\eta} = -\frac{1}{T_1}(X_3 - n_0) + 2iq_0(X_1 - X_2) , \tag{2.5b}$$

$$\frac{\partial}{\partial\eta}(X_1X_1) = -\left[\frac{2}{T_1} - 2i\delta - 2i\mu_1 + 2i\mu_2\right](X_1X_1) + 2iq_0(X_1X_3) , \tag{2.5c}$$

$$\frac{\partial}{\partial\eta}(X_1X_2) = -\frac{2}{T_2}(X_1X_2) + iq_0(X_3X_2 - X_3X_1) , \tag{2.5d}$$

$$\frac{\partial}{\partial\eta}(X_3X_3) = -\frac{2}{T_1}(X_3X_3) + 4iq_0(X_1X_3 - X_2X_3) + \frac{2n_0}{T_1}X_3 , \tag{2.5e}$$

$$\frac{\partial}{\partial\eta}(X_1X_3)=-\left[\frac{1}{T_1}+\frac{1}{T_2}-i\mu_1-i\mu_2-i\delta\right](X_1X_3)+2iq_0(X_1X_1-X_1X_2)+iq_0(X_3X_3)+\frac{n_0}{T_1}X_1\ , \qquad (2.5f)$$

together with the equations obtained from complex conjugation.

The set of Eqs. (2.5) is stochastic in nature as the $\mu$'s are stochastic variables. These stochastic equations can be transformed into mean value equations by using the theory of multiplicative stochastic processes.[11] The resulting equations are found to be

$$\frac{\partial}{\partial\eta}\langle X_1\rangle=-\left[\frac{1}{T_2}+\gamma_L-i\delta\right]\langle X_1\rangle+iq_0\langle X_3\rangle\ , \qquad (2.6a)$$

$$\frac{\partial}{\partial\eta}\langle X_3\rangle=-\frac{1}{T_1}(\langle X_3\rangle-n_0)+2iq_0(\langle X_1\rangle-\langle X_2\rangle)\ , \qquad (2.6b)$$

$$\frac{\partial}{\partial\eta}\langle X_1X_1\rangle=-2\left[\frac{1}{T_2}+2\gamma_L-i\delta\right]\langle X_1X_1\rangle+2iq_0\langle X_1X_3\rangle\ , \qquad (2.6c)$$

$$\frac{\partial}{\partial\eta}\langle X_1X_2\rangle=-\frac{2}{T_2}\langle X_1X_2\rangle+iq_0(\langle X_2X_3\rangle-\langle X_3X_1\rangle)\ , \qquad (2.6d)$$

$$\frac{\partial}{\partial\eta}\langle X_3X_3\rangle=-\frac{2}{T_1}\langle X_3X_3\rangle+4iq_0(\langle X_1X_3\rangle-\langle X_2X_3\rangle)+\frac{2n_0}{T_1}\langle X_3\rangle\ , \qquad (2.6e)$$

$$\frac{\partial}{\partial\eta}\langle X_1X_3\rangle=-\left[\frac{1}{T_1}+\frac{1}{T_2}+\gamma_L-i\delta\right]\langle X_1X_3\rangle+\frac{n_0}{T_1}\langle X_1\rangle+iq_0(\langle X_3X_3\rangle+2\langle X_1X_1\rangle-2\langle X_2X_1\rangle)\ , \qquad (2.6f)$$

plus the equations obtained from complex conjugation.

In Eqs. (2.6) $\gamma_L$ denotes the sum of the linewidths associated with each pump,

$$\gamma_L=\gamma_1+\gamma_2\ . \qquad (2.7)$$

It is interesting to note that the linewidths of both the pump waves enter symmetrically in the above equations. Equations (2.6) can be solved in the steady state with the result

$$\langle X_1\rangle=\langle X_2\rangle^*=iq_0\left[\frac{1}{T_2}+\gamma_L-i\delta\right]^{-1}\langle X_3\rangle\xrightarrow{q_0\to 0}iq_0n_0\left[\frac{1}{T_2}+\gamma_L-i\delta\right]^{-1}\ , \qquad (2.8a)$$

$$\langle X_3\rangle=n_0\left[1+\frac{4q_0^2T_1(1/T_2+\gamma_L)}{\delta^2+(1/T_2+\gamma_L)^2}\right]^{-1}\xrightarrow{q_0\to 0}n_0+O(q_0^2)\ , \qquad (2.8b)$$

$$\langle X_1X_2\rangle=q_0T_2\mathrm{Im}\langle X_1X_3\rangle\xrightarrow{q_0\to 0}\frac{q_0^2n_0^2T_2(1/T_2+\gamma_L)}{\delta^2+(1/T_2+\gamma_L)^2}\ , \qquad (2.8c)$$

$$\langle X_2X_2\rangle=-iq_0\langle X_2X_3\rangle\left[\frac{1}{T_2}+2\gamma_L+i\delta\right]^{-1}\xrightarrow{q_0\to 0}-q_0^2n_0^2\left[\frac{1}{T_2}+2\gamma_L+i\delta\right]^{-1}\left[\frac{1}{T_2}+\gamma_L+i\delta\right]^{-1}\ , \qquad (2.8d)$$

$$\langle X_1X_3\rangle=\frac{iq_0n_0\langle X_3\rangle}{|A|^2+q_0(A+A^*)(2T_1+T_2)}[A^*C+q_0^2(2T_1+T_2)(C-C^*)]\xrightarrow{q_0\to 0}iq_0n_0^2\left[\frac{1}{T_2}+\gamma_L-i\delta\right]^{-1}\ , \qquad (2.8e)$$

where

$$A=\left[\frac{1}{T_1}+\frac{1}{T_2}+\gamma_L\right.$$
$$\left.-i\delta+2q_0^2\left[\frac{1}{T_2}+2\gamma_L-i\delta\right]^{-1}\right],$$
$$C=1+\left[\frac{T_1}{T_2}+T_1(\gamma_L-i\delta)\right]^{-1}. \quad (2.8f)$$

The final result for the intensity is obtained by substituting (2.8) in (2.3). This result has been obtained without any restrictions on the pump intensities or their linewidths. It should also be noted that the laser linewidth does not appear in any simple fashion in the expressions obtained here, i.e., the CARS signal in the presence of pump fluctuations cannot be obtained by a simple scaling of $T_1$ and $T_2$. However, the behavior of the expectation values such as $\langle X_1\rangle$ and $\langle X_3\rangle$ is obtained by simple scaling $1/T_2\rightarrow 1/T_2+\gamma_L$. The CARS intensity in the limit of no saturation ($I\propto q_0^2$) does not quite follow the scaling relation $1/T_2\rightarrow 1/T_2+\gamma_L$ because of the appearance of the additional factor $T_2$ in (2.8c). It might be thought that laser bandwidth effect is similar to a relaxation effect. However, that is not the case since the usual relaxation theory is based on two major assumptions: (i) the weakness of the interaction with the reservoir, i.e., the interaction that is responsible for relaxation; and (ii) the smallness of the correlation time of the reservoir relative to other time scales in the problem. This latter assumption enables one to obtain equations that are local in time without enhancing the dimensionality of our space. However, in our present problem the phase of the field, which determines the phase of the dipole moment of the system, remains correlated over a long time, since $\langle\phi(t+\tau)\phi(t)\rangle=2\gamma_L t$.

The behavior of the CARS signal $I$ [Eq. (2.3b)] as a function of $\delta$ for various values of the pump strength $q_0$ and for various values of $\gamma_L$ is presented in Figs. 2 and 3. The parameter $b$ in these figures is defined by Eq. (2.4). For weak fields (Fig. 2) we find that the intensity distribution has the usual dispersionlike profile. With increasing pump linewidth $\gamma_L$ the peak at $\delta=0$ broadens. The effect of pump fluctuations is much more pronounced on the minimum in the intensity profile which starts disappearing rapidly as $\gamma_L$ is increased. Similar behavior continues even with moderate field strengths. However, significant differences start appearing when $q_0$ becomes of the

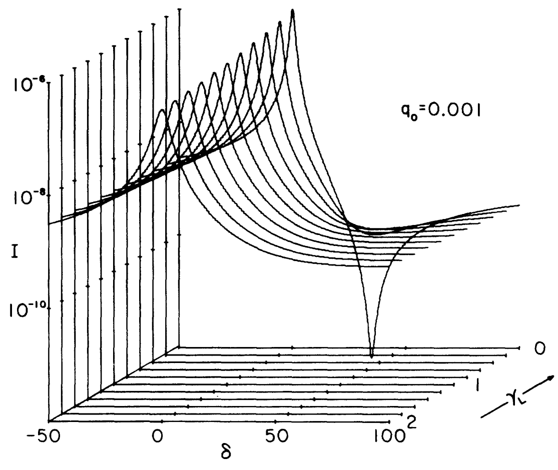


FIG. 2. The variation of the CARS signal intensity [Eq. (1.10)] with detuning $\delta$ for $q_0=0.001$ and several different values of $\gamma_L=\gamma_1+\gamma_2$. We have taken $T_1=T_2$, $n_0=-1$, $b=35$, and both $\delta$ and $\gamma_L$ have been expressed in units of $T_1$. The parameter $b$ is defined by Eq. (2.4). Note that the curve on the extreme right corresponds to $\gamma_L=0$.

order of $1/T_2$ (Fig. 3). The peak at $\delta=0$ starts splitting. The minimum in the intensity profile also has a different structure. As $\gamma_L$ is increased the peaks at $\delta=0$ show broadening due to both the pump intensity and the pump linewidths. This behavior is illustrated in Fig. 3. When the pump fields become quite large ($q_0\gtrsim 10$) the intensity distribution does not show any interesting features. The splitting observed when $q_0\sim 1$ is because of the behavior of $\langle X_1X_2\rangle$ as a function of $\delta$, as the expression for $\langle X_1X_2\rangle$ given by Eq. (2.8c) involves a denominator containing higher powers of $\delta$ than 2. The other terms in the expression for the intensity $I$ show only a power-broadening effect. The

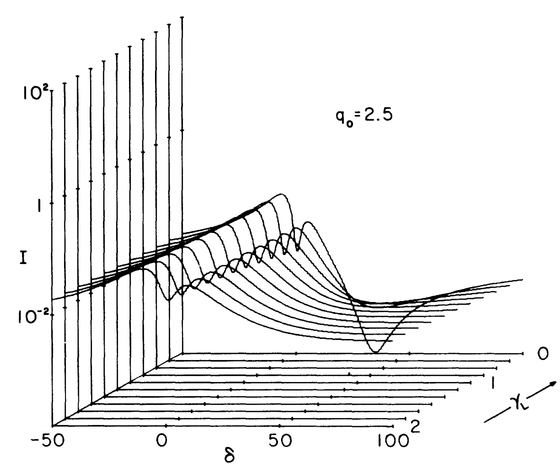


FIG. 3. Same as Fig. 2 for $q_0=2.5$.

splitting of the peak at $\delta=0$ is rapidly wiped out due to power broadening as pump strength $q_0$ is increased. The final behavior of the CARS signal is, of course, determined from $\langle X_1X_2\rangle$ and the interference effects arising from the nonresonant part of the susceptibility.

## III. CONTRIBUTION OF PUMP LINEWIDTHS TO CARS LINEWIDTH AND SPECTRA

Having studied in detail the behavior of CARS intensities as a function of the pump intensities and linewidths, we now calculate the CARS spectra which are obtained by taking the Fourier transform of the two-time amplitude correlation function defined by Eq. (1.11). For this purpose we find it convenient to define the following new variables:

$$\psi_1(\eta)=\sigma_{12}(\eta)e^{-i\Phi_1(\eta)}\ , \tag{3.1a}$$

$$\psi_2(\eta)=e^{-3i\Phi_1(\eta)+2i\Phi_2(\eta)}\sigma_{21}(\eta)\ , \tag{3.1b}$$

$$\psi_3(\eta)=ne^{-2i\Phi_1(\eta)+i\Phi_2(\eta)}\ , \tag{3.1c}$$

and

$$\psi_4(\eta)=e^{-2i\Phi_1(\eta)+i\Phi_2(\eta)}\ . \tag{3.1d}$$

In terms of these new variables the correlation function defined by (1.11) can be rewritten as

$$C_a(\tau)=\Gamma_a(\tau)\alpha^2Q^2|\mathscr{E}_1|^2\left[\frac{\sin(\Delta kz/2)}{(\Delta k/2)}\right]^2 ,$$

with

$$\Gamma_a(\tau)=[\langle\psi_1(\eta+\tau)\psi_1^*(\eta)\rangle + |B|^2\langle\psi_4(\eta+\tau)\psi_4^*(\eta)\rangle + B\langle\psi_4(\eta+\tau)\psi_1^*(\eta)\rangle + B^*\langle\psi_1(\eta+\tau)\psi_4^*(\eta)\rangle]\ . \tag{3.2}$$

Starting from Bloch equations (1.4) and (1.5) we find the following stochastic equations of motion for the variables $\psi_i$'s:

$$\frac{\partial\psi_1}{\partial\eta}=-\left[\frac{1}{T_2}-i\delta+i\mu_1\right]\psi_1+iq_0\psi_3\ , \tag{3.3}$$

$$\frac{\partial\psi_2}{\partial\eta}=-\left[\frac{1}{T_2}+i\delta+3i\mu_1-2i\mu_2\right]\psi_2-iq_0\psi_3\ , \tag{3.4}$$

$$\frac{\partial\psi_3}{\partial\eta}=\left[-2i\mu_1+i\mu_2-\frac{1}{T_1}\right]\psi_3+\frac{n_0\psi_4}{T_1} + 2iq_0(\psi_1-\psi_2)\ , \tag{3.5}$$

$$\frac{\partial\psi_4}{\partial\eta}=(-2i\mu_1+i\mu_2)\psi_4\ . \tag{3.6}$$

The variables $\psi$ satisfy a closed set of Langevin equations. Similar equations are satisfied by their complex conjugates. The correlation functions of the $\psi$ variables can be obtained from their Markovian property and from the general results on multiplicative stochastic processes, which, for example, lead to

$$\frac{\partial}{\partial\eta}\langle\psi\rangle=\begin{bmatrix} -\frac{1}{T_2}-\gamma_1+i\delta & 0 & iq_0 & 0\\ 0 & -\frac{1}{T_2}-i\delta-9\gamma_1-4\gamma_2 & -iq_0 & 0\\ 2iq_0 & -2iq_0 & -4\gamma_1-\gamma_2-\frac{1}{T_1} & \frac{n_0}{T}\\ 0 & 0 & 0 & -4\gamma_1-\gamma_2 \end{bmatrix}\langle\psi\rangle \tag{3.7}$$

$$\equiv M\langle\psi\rangle\ , \tag{3.8}$$

where $\langle\psi\rangle\equiv(\psi_1,\psi_2,\psi_3,\psi_4)^T$ is a column matrix and $M$ is a $4\times4$ matrix defined by Eq. (3.7). It follows from Eq. (3.8) that

$$\langle\psi_1(\eta+\tau)\rangle=[e^{M\tau}]_{11}\langle\psi_1(\eta)\rangle+[e^{M\tau}]_{12}\langle\psi_2(\eta)\rangle+[e^{M\tau}]_{13}\langle\psi_3(\eta)\rangle+[e^{M\tau}]_{14}\langle\psi_4(\eta)\rangle\ , \tag{3.9}$$

and on using the Markov property of $\psi$'s we find the following expression for the correlation function of $\psi_1$,

$$\begin{aligned}\langle \psi_1(\eta+\tau)\psi_1^*(\eta)\rangle &= [e^{M\tau}]_{11}\langle \psi_1(\eta)\psi_1^*(\eta)\rangle + [e^{M\tau}]_{12}\langle \psi_2(\eta)\psi_1^*(\eta)\rangle \\ &\quad + [e^{M\tau}]_{13}\langle \psi_3(\eta)\psi_1^*(\eta)\rangle + [e^{M\tau}]_{14}\langle \psi_4(\eta)\psi_1^*(\eta)\rangle \\ &= [e^{M\tau}]_{11}\langle X_1X_2\rangle + [e^{M\tau}]_{12}\langle X_2X_2\rangle + [e^{M\tau}]_{13}\langle X_2X_3\rangle + [e^{M\tau}]_{14}\langle X_2\rangle \ . \end{aligned} \tag{3.10}$$

In deriving (3.10) we have used the definitions (3.1) and (2.4). Using Eq. (3.10) in Eq. (3.2) we can obtain the amplitude correlation function for the field at $2\omega_1-\omega_2$. The spectrum of the field can then be shown to be proportional to

$$S(\omega)\equiv 2\,\mathrm{Re}\Gamma[i(2\omega_1-\omega_2-\omega)] \ , \tag{3.11}$$

where

$$\begin{aligned}\Gamma(z) &= m_{11}\langle X_1X_2\rangle + m_{12}\langle X_2X_2\rangle + m_{13}\langle X_3X_2\rangle + m_{14}\langle X_2\rangle + |B|^2 m_{44} + Bm_{44}\langle X_2\rangle + B^*m_{11}\langle X_1\rangle \\ &\quad + B^*m_{12}\langle X_2\rangle + B^*m_{13}\langle X_3\rangle + B^*m_{14} \ . \end{aligned} \tag{3.12}$$

The matrix elements $m_{ij}$ are given by

$$m_{11}=D^{-1}(z)\left[2q_0^2+\left[z+\frac{1}{T_2}+i\delta+9\gamma_1+4\gamma_2\right]\left[z+\frac{1}{T_1}+4\gamma_1+\gamma_2\right]\right] , \tag{3.13a}$$

$$m_{12}=2q_0^2D^{-1}(z) \ , \tag{3.13b}$$

$$m_{13}=iq_0D^{-1}(z)\left[z+\frac{1}{T_2}+i\delta+9\gamma_1+4\gamma_2\right] , \tag{3.13c}$$

$$m_{14}=\frac{iq_0n_0}{T_1}D^{-1}(z)\left[z+\frac{1}{T_2}+i\delta+9\gamma_1+4\gamma_2\right](z+4\gamma_1+\gamma_2)^{-1} , \tag{3.13d}$$

$$m_{44}=(z+4\gamma_1+\gamma_2)^{-1} \tag{3.13e}$$

with $D(z)$ given by

$$D(z)=4q_0^2\left[z+\frac{1}{T_2}+5\gamma_1+2\gamma_2\right]+\left[z+\frac{1}{T_2}-i\delta+\gamma_1\right]\left[z+\frac{1}{T_2}+i\delta+9\gamma_1+4\gamma_2\right]\left[z+\frac{1}{T_1}+4\gamma_1+\gamma_2\right] . \tag{3.14}$$

Expression (3.12) gives the resonant CARS spectra for arbitrary values of $|\mathscr{E}_1|$, $|\mathscr{E}_2|$, $\delta$, and $\gamma_1$, $\gamma_2$. Again note that the pump linewidths do not enter in any simple manner. In the limit of $\gamma_i \rightarrow 0$ one of course has

$$s(\omega)=\mathrm{const}\,\delta(\omega-2\omega_1+\omega_2) \ . \tag{3.15}$$

This is because in the derivation of the spectra and the definition (1.11) we have ignored single atom/molecule contributions in favor of the coherent contributions from a collection of atoms/molecules. The single atom contributions were studied earlier in the context of resonance fluorescence.[14] We have also not included the Doppler broadening in our treatment. The weak field limit of the result (3.13) can be shown to be equivalent to the known result of Dutta,[6] obtained by a different method. Dutta, however, has not presented detailed plots for the line shapes, which as we show below, depend critically on the relative magnitudes of $\gamma_1$, $\gamma_2$.

The behavior of the CARS spectrum defined by Eqs. (3.11)–(3.14) is shown in Figs. 4 and 5 for several different values of $\gamma_1$, $\gamma_2$, and $q_0$. It should be noted that all the additional resonances in the spectra represent the Rabi splitting of the energy levels. These could be seen in the resonant CARS situation only because of the laser temporal fluctuations, in the absence of which (and any other relaxations) they do not show up. This is because the strength of these resonances depends on the fluctuations. The situation is somewhat similar to that encountered in the experiments of Prior *et al.*[4] where the additional relaxation effects due to pressure broadening enabled one to see extra resonances. In our case the Rabi splitting of the spectra could be seen only because of the "relaxation effects" implied by laser fluctuations. It should also be

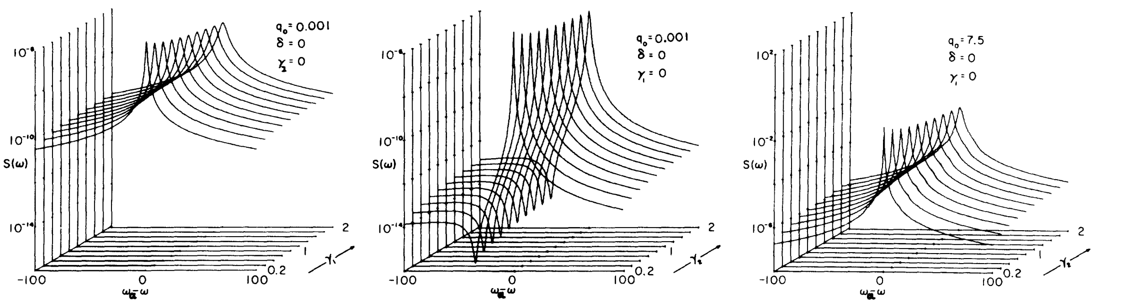


FIG. 4. The variation of the CARS spectral density $S(\omega)$ [Eq. (3.11)] with $\omega$ for $\delta=0$ and several different values of $q_0$, $\gamma_1$, and $\gamma_2$. Other parameters for this figure are the same as for Fig. 2.

emphasized that these resonances are rather broad and could be resolved only if $q_0$ were very large compared to $\gamma_1$ and $\gamma_2$. For example, a simple analysis shows that the approximate roots of the polynomial $D(z)$ [Eq. (3.14)] are

$$z_0 \simeq -\frac{1}{T_1} - \left[(4\gamma_1+\gamma_2) + \frac{4q_0^2(\gamma_1+\gamma_2)}{\delta^2+4q_0^2}\right] \tag{3.16a}$$

and

$$z_\pm \simeq -\frac{1}{T_1} \pm i(\delta^2+4q_0^2)^{1/2} - \frac{1}{2}\left[9\gamma_1+3\gamma_2+\frac{\delta^2(\gamma_1+\gamma_2)}{\delta^2+4q_0^2} \mp \frac{2\delta(4\gamma_1+\gamma_2)}{(\delta^2+4q_0^2)^{1/2}}\right] . \tag{3.16b}$$

It is evident from the structure of (3.13) and (3.14) that the fluctuations of the pump wave at $\omega_1$ are more important than the fluctuations of the wave at $\omega_2$. This is because at least two photons should be absorbed from the wave at $\omega_1$ and only one photon from the wave at $\omega_2$ to generate the CARS signal at $2\omega_1-\omega_2$.

The behavior of the spectral density $S(\omega)$ for $\delta=0$, i.e., when the Raman transition frequency $\omega_R$ coincides with the two-photon transition frequency $\omega_1-\omega_2$ is displayed in Fig. 4 for several different values of $q_0$, $\gamma_1$, and $\gamma_2$. It is found that

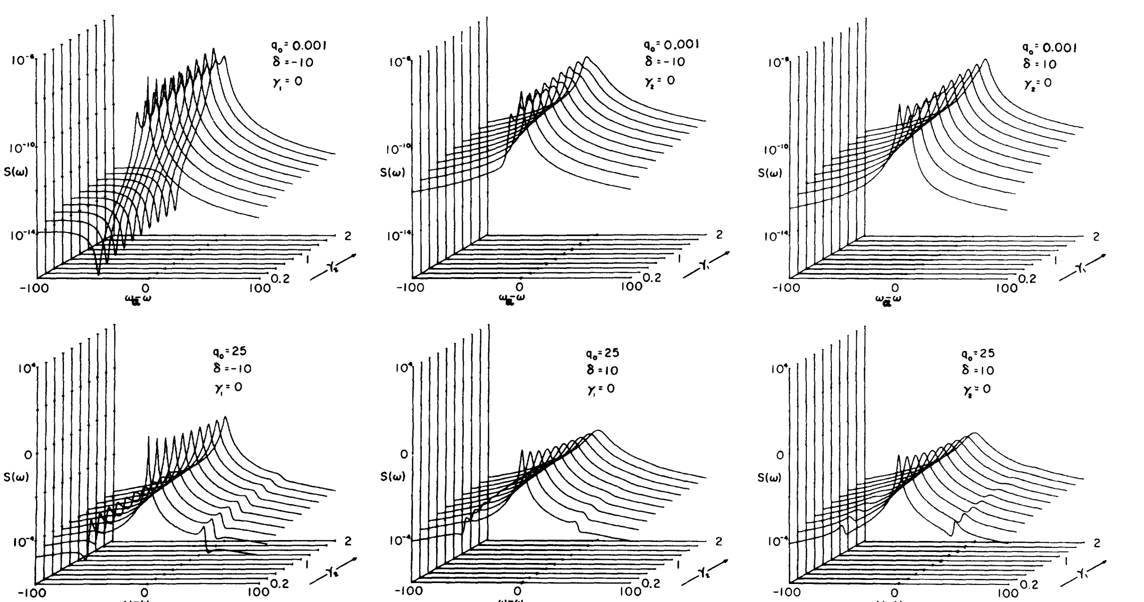


FIG. 5. The variation of the CARS spectral density $S(\omega)$ [Eq. (3.11)] with $\omega$ for $\delta=-10$ and $\delta=10$ for several different values of $q_0$, $\gamma_1$, and $\gamma_2$. Other parameters for this figure are the same as for Fig. 2.

for a monochromatic field at frequency $\omega_2$ there is only a single pronounced peak at $\omega-\omega_a=0$ for all values of the field strength $q_0$ and the effect of increasing bandwidth $\gamma_1$ is to broaden this peak. However, the behavior of $S(\omega)$ is remarkably different when the field at frequency $\omega_1$ is monochromatic. In this case, for small values of the effective Rabi frequency ($q_0=0.001$) $S(\omega)$ has a dispersionlike structure at $\omega=\omega_a$. For sufficiently large field strengths, $S(\omega)$ acquires a well-defined peak rather than a dispersionlike structure. There remains a central peak at $\omega-\omega_a=0$ and two additional peaks corresponding to Rabi splitting of the energy levels start to make their pressure felt (see the figure corresponding to $q_0=7.5$). With increasing $\gamma_2$ these two peaks are rapidly washed out. It is clear from Fig. 4 and the preceding discussion that the fluctuations of the beam at frequency $\omega_1$ are significantly more important than the fluctuations of the beam at frequency $\omega_2$ in modifying the behavior of $S(\omega)$.

Figure 5 illustrates the behavior of $S(\omega)$ when the Raman transition frequency $\omega_R$ is different from the two-photon transition frequency ($\omega_1-\omega_2=\omega_R+\delta$) for several different values of $q_0$, $\gamma_1$, $\gamma_2$, and $\delta$. We find that in weak fields ($q_0=0.001$) $S(\omega)$ has two peaks at $\omega_a-\omega\cong 0$ and $\omega_a-\omega\cong\delta$. The peak $\omega_a-\omega\sim\delta$ corresponds to anti-Stokes frequency, i.e., $\omega=\omega_1+\omega_R$. This is because, in the second order in $q_0$, the system has a finite probability to be found in the state $|1\rangle$. Then the absorption of $\omega_1$ photon produces the anti-Stokes frequency $\omega_1+\omega_R$. The effect of increasing bandwidths is to broaden and wash out these peaks. The peak at $\omega_a-\omega\cong 0$ disappears faster than the peak at $\omega_a-\omega\cong\delta$. However, in the case $\gamma_1=0$ the spectral density $S(\omega)$ has a dispersionlike structure in addition to the type of structure one sees for $\gamma_2=0$. The different structure of $S(\omega)$ in the two cases is due to different roles played by the two linewidths. This additional structure is wiped out as field strength is increased. In strong fields $S(\omega)$ has the expected triplet structure due to Rabi splitting of the energy levels. However, we find that the spectrum is asymmetric with respect to the sign of $\delta$. It is interesting to note that now we have well-defined dispersionlike structures for Rabi sidebands. The dispersionlike structures are less pronounced for $\gamma_2=0$ and start disappearing with increase in $\gamma_1$. It is also seen by a comparison of the $\gamma_1=0$ and $\gamma_2=0$ cases, that $\gamma_1$ is much more effective than $\gamma_2$ in modifying the structure of $S(\omega)$. It is amply clear now that the observed differences in the behavior of spectra for $\gamma_1=0$ and $\gamma_2=0$ cases point to the breakdown of any simple scaling law and justify a detailed discussion of the role of laser fluctuations in CARS spectra calculation. It is easy to infer from Figs. 4 and 5 and the subsequent discussion the effects of both fields being nonmonochromatic simultaneously. For example, when both pump fields are fluctuating most of the extra structure that $S(\omega)$ has for the $\gamma_1=0$ case will be washed out very rapidly, and the behavior of $S(\omega)$ will resemble the behavior for the $\gamma_2=0$ case. Another noteworthy feature of the behavior of $S(\omega)$ is that it is not a symmetric function of $\delta$ and $\omega$.

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## APPENDIX: CARS LINE SHAPES AS CONVOLUTION OF THE SUSCEPTIBILITY TENSOR $\chi^{(3)}$ AND THE CROSS-SPECTRAL TENSORS OF THE FIELD

In this appendix we comment on the structure of the line shapes in cases where saturation effects are not important. It is then possible to consider more general fluctuations and the line shapes of the pump fields. We will show that, when saturation effects can be ignored, the CARS line shape is related to the fourth- and the second-order cross-spectral density tensors of the fields at $\omega_1$ and $\omega_2$, respectively. Similar results were obtained earlier[15] in the context of multiphoton processes. Such results would be particularly useful if the line shape of the pump field were described more accurately by, say, a Gaussian rather than by a Lorentzian.

It can be shown that in general the Fourier component of the induced polarization $P^{(3)}$ at frequency $\omega$ can be written as

$$P_\mu^{(3)}(\omega)=\int\int\int d\omega_1 d\omega_2 d\omega_3 \delta(\omega-\sum_i \omega_i)\chi^{(3)}_{\mu\alpha\beta\gamma}(\omega_1,\omega_2,\omega_3)E_\alpha(\omega_1)E_\beta(\omega_2)E_\gamma(\omega_3) . \quad (A1)$$

Here the Greek subscripts denote the Cartesian components of the fields and the susceptibility tensor and all the Fourier transforms have been defined by

$$X(t)=\int X(\omega)e^{-i\omega t}d\omega,\ \ X(\omega)=\int \frac{dt}{2\pi}X(t)e^{i\omega t} . \quad (A2)$$

The susceptibility tensor $\chi^{(3)}_{\mu\alpha\beta\gamma}(\omega_1,\omega_2,\omega_3)$ has the intrinsic permutation symmetry with respect to the interchange of its indices $(\alpha\omega_1,\beta\omega_2,\gamma\omega_3)$. For CARS in monochromatic fields the total pump field $\vec{E}(t)$ can be written as

$$\vec{E}(t)=\vec{\mathscr{E}}^{(1)}e^{-i\omega_1 t}+\vec{\mathscr{E}}^{(2)}e^{-i\omega_2 t}+\text{c.c.} , \quad (A3)$$

where the superscript $i$ denotes the field at frequency $\omega_i$. It can then be shown that the component of the induced polarization at the CARS frequency $2\omega_1-\omega_2$ will be given by

$$P_\mu^{(3)}(\omega)=3\delta(\omega-2\omega_1+\omega_2)\chi^{(3)}_{\mu\alpha\beta\gamma}(\omega_1,\omega_1,-\omega_2)\mathscr{E}_\alpha^{(1)}\mathscr{E}_\beta^{(1)}\mathscr{E}_\gamma^{(2)*} . \quad (A4)$$

In deriving this result we have used the permutation symmetry of $\chi^{(3)}$ and the Fourier transform of Eq. (A3) in Eq. (A1). Relation (A4) helps in identifying the susceptibility tensor $\chi^{(3)}$ when it occurs in relations like Eq. (A4) involving monochromatic fields, and in relations like (A1). For the case of fluctuating pump waves, the amplitudes $\mathscr{E}^{(1)}$ and $\mathscr{E}^{(2)}$ will not be deterministic but would be stochastic processes. However $\mathscr{E}^{(i)}$ would only be slowly varying functions of time $t$. It is therefore convenient to write

$$\vec{E}(t)=\vec{E}^{(+)}(t)+\vec{E}^{(-)}(t),\ \ \vec{E}^{(\pm)}(\omega)=\int \frac{dt}{2\pi}e^{i\omega t}\vec{E}^{(\pm)}(t) , \quad (A5)$$

where $\vec{E}^{(+)}(\omega)$ $[\vec{E}^{(-)}(\omega)]$ contains only positive (negative) frequency components. For a stationary field we find on using the Wiener-Khintchine theorem that

$$\langle E_\alpha^{(+)}(\omega_1)E_\beta^{(+)*}(\omega_2)\rangle=\delta(\omega_1-\omega_2)\Gamma_{\alpha\beta}(\omega_1)=\langle E_\alpha^{(+)}(\omega_1)E_\beta^{(-)}(-\omega_2)\rangle , \quad (A6)$$

where

$$\Gamma_{\alpha\beta}(\omega)=\int \frac{dt}{2\pi}e^{i\omega t}\langle E^{(+)}(t)E^{(-)}(0)\rangle . \quad (A7)$$

In view of (A1), (A4), and (A5) it is now clear that if we allow the fluctuations of the field, then in place of relation (A4) we will have

$$P_\mu^{(3)}(\omega)=3\int\int\int d\{\omega_i\}\delta\left[\omega-\sum_i\omega_i\right]\chi^{(3)}_{\mu\alpha\beta\gamma}(\omega_1,\omega_2,\omega_3)E_\alpha^{(1)(+)}(\omega_1)E_\beta^{(1)(+)}(\omega_2)E_\gamma^{(2)(-)}(\omega_3) , \quad (A8)$$

where $P_\mu^{(3)}(\omega)$ will now be fluctuating. Its spectrum, however, will be peaked at $2\omega_1-\omega_2$. From Eq. (A8) we easily obtain for the fluctuations of $P_\mu^{(3)}(\omega)$

$$\langle P_\mu^{(3)}(\omega)P_\nu^{(3)*}(\omega')\rangle=9\int d\{\omega_i\}d\{\omega_i'\}\delta\left[\omega-\sum_i\omega_i\right]\delta\left[\omega'-\sum_i\omega_i'\right]\chi^{(3)}_{\mu\alpha\beta\gamma}(\omega_1,\omega_2,\omega_3)\chi^{(3)*}_{\nu\alpha'\beta'\gamma'}(\omega_1',\omega_2',\omega_3')$$
$$\times\langle E_\alpha^{(1)(+)}(\omega_1)E_\beta^{(1)(+)}(\omega_2)[E_{\alpha'}^{(1)(+)}(\omega_1')E_{\beta'}^{(1)(+)}(\omega_2')]^*\rangle\langle E_\gamma^{(2)(-)}(\omega_3)[E_{\gamma'}^{(2)(-)}(\omega_3')]^*\rangle , \quad (A9)$$

where we have assumed that the pump waves at $\omega_1$ and $\omega_2$ are uncorrelated with each other. Since the field $\vec{E}(t)$ [Eq. (A3)] has been assumed to be stationary, it can be shown following Mandel and Mehta[16] that the fourth-order correlation of the field $\vec{E}^{(1)}$ in the frequency domain has the following structure:

$$\langle E_\alpha^{(1)(+)}(\omega_1)E_\beta^{(1)(+)}(\omega_2)(E_{\alpha'}^{(1)(+)}(\omega_1'))^*(E_{\beta'}^{(1)(+)}(\omega_2'))^*\rangle=\delta(\omega_1+\omega_2-\omega_1'-\omega_2')\Gamma^{(1)}_{\alpha\beta\alpha'\beta'}(\omega_1,\omega_2,\omega_1') , \quad (A10)$$

where

$$\Gamma^{(1)}_{\alpha\beta\alpha'\beta'}(\omega_1,\omega_2,\omega'_1)=\int \frac{d\{t_i\}}{(2\pi)^3}\langle E^{(1)(+)}_{\alpha}(t_1)E^{(1)(+)}_{\beta}(t_2)E^{(1)(-)}_{\alpha'}(t_3)E^{(1)(-)}_{\beta'}(0)\rangle \exp(i\omega_1 t_1+i\omega_2 t_2-i\omega'_1 t_3) . \tag{A11}$$

On substituting (A10) and (A6) in Eq. (A9) and on simplification we obtain

$$\langle P^{(3)}_{\mu}(\omega)P^{(3)*}_{\nu}(\omega')\rangle=\delta(\omega-\omega')\Gamma^{(P)}_{\mu\nu}(\omega),\quad \omega>0 \tag{A12}$$

where the spectrum $\Gamma^{(P)}_{\mu\nu}(\omega)$ of the polarization fluctuations is given by

$$\Gamma^{(P)}_{\mu\nu}(\omega)=9\int d\{\omega_i\}d\omega'_1 d\omega'_2\delta(\omega-\omega_1-\omega_2+\omega_3)\delta(\omega_1+\omega_2-\omega'_1-\omega'_2)\chi^{(3)}_{\mu\alpha\beta\gamma}(\omega_1,\omega_2,-\omega_3)$$
$$\times\chi^{(3)*}_{\nu\alpha'\beta'\gamma'}(\omega'_1,\omega'_2,-\omega_3)\Gamma^{(2)}_{\gamma'\gamma}(\omega_3)\Gamma^{(1)}_{\alpha\beta\alpha'\beta'}(\omega_1,\omega_2,\omega'_1) . \tag{A13}$$

Equation (A13) is our final expression which gives the spectrum of CARS polarization fluctuations in terms of $\chi^{(3)}$ and the fourth-order cross-spectral density tensor of the field at $\omega_1$. Further simplifications in formula (A13) can be made if the field statistics are known and various special cases follow from Eq. (A13) depending on the bandwidth of the fields. For example, if the field at frequency $\omega_1$ obeys Gaussian statistics, the fourth-order correlation function can be expressed in terms of second-order ones, and then, depending on the line shapes of the pump beams at $\omega_1$ and $\omega_2$, $\Gamma^{(P)}_{\mu\nu}(\omega)$ can be computed.[6,17] For the Gaussian case one discovers kinematical enhancement factors like $n!$, which are reminiscent of those occurring in the context of multiphoton processes.[15] The derivation of Eq. (A13) is quite general and is not necessarily restricted to resonant CARS, and it follows that out of the 48 terms contributing to $\chi^{(3)}$ as many terms may be retained as may be required from physical considerations.

Finally, we mention that the approach presented here could also be used to discuss higher-order CARS which, for example, has been recently studied by Compaan *et al.*[9] The anti-Stokes radiation in this case is generated at frequency $3\omega_1-2\omega_2$, and it can be shown that the spectrum of polarization fluctuations will now be related to the sixth-order correlation function of the field at $\omega_1$ and the fourth-order correlation function of the field at $\omega_2$. The kinematical enhancement factors, when the fields are Gaussian, would now be $3!2!=12$.


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