Pump-probe cross-correlation-induced resonances in four-wave mixing

G. S. Agarwal
School of Physics, University of Hyderabad, Hyderabad 500 134, India

C. V. Kunasz and J. Cooper
Joint Institute for Laboratory Astrophysics, University of Colorado and National Bureau of Standards, Boulder, Colorado 80309-0440

(Received 8 December 1986)

A rigorous theory of the stochastic field-induced resonances in four-wave mixing with cross-correlated pump and probe fields is developed. The behavior of the extra resonance for a range of fluctuation and collisional parameters is discussed. Our results differ significantly from those obtained from a decorrelated theory.

Agarwal and Kunasz[1] analyzed in great detail the effects of the fluctuations of the pump field on four-wave mixing[2] and predicted that the effects of pump fluctuations would be very different from the effects of collisions due to the atoms of the buffer gas, although both collisions and fluctuations produce relaxation of the system. They also predicted a redistribution of the coherent radiation, i.e., that the generated radiation has frequencies other than those obtained by four-wave mixing. The redistributed radiation exhibits additional resonances due to pumping fluctuations.

Prior *et al.*[3] discovered that a cross correlation[4] of the pump and probe fields can lead to additional resonances[5,6] in four-wave mixing. The situation considered by Prior *et al.* is distinct from that considered by Agarwal and Kunasz since the latter work assumed a nonfluctuating probe. Prior *et al.* base their work on the ensemble average of the density matrix elements ρ_{kl} of the atomic system, which are then used to calculate the third-order susceptibility $\chi^{(3)}$. This ensemble average, denoted by $\langle\ \rangle, \langle \rho_{kl} \rangle$, is taken over the fluctuations of the correlated pump and probe fields. Let $\langle P \rangle$ be the induced polarization at the frequency $2\omega_1-\omega_2$. Prior *et al.* argue that since $\langle P \rangle$ exhibits *additional resonances,* e.g., at $\omega_1-\omega_2$, equal to the separation ω_{12} of two excited states connected by dipole transition with the ground state, the four-wave-mixing signal (via $\chi^{(3)}$) would exhibit such a resonance if the pump and probe were cross correlated. The argument implicitly assumes that the four-wave-mixing signal I_D is related to $\langle P \rangle$ by

$$I_D \propto |\langle P \rangle|^2 . \tag{1}$$

Let us look more closely at the argument of Prior *et al.* leading to the fluctuation-induced resonances in four-wave mixing. Consider the equation for the off-diagonal element of ρ,

$$\dot{\rho}_{kl} = -\Gamma_{kl}\,\rho_{kl} - i\omega_{kl}\,\rho_{kl} + (\text{field terms}) . \tag{2}$$

The phase fluctuations from the field-dependent terms can be removed[7] in most cases by defining $\tilde{\rho}_{kl} = \rho_{kl} e^{-i\Phi_{kl}(t)}$ so that

$$\dot{\tilde{\rho}}_{kl} = -i[\omega_{kl} + \Gamma_{kl} + \dot{\Phi}_{kl}(t)]\tilde{\rho}_{kl} + (\text{field terms}) , \tag{3}$$

where the field terms are now phase independent. If the phases associated with the fields are given by the phase diffusion model, i.e., $\dot{\Phi}_{kl}$ is a δ-correlated Gaussian random process,

$$\langle \dot{\Phi}_{kl}(t)\dot{\Phi}_{kl}(t') \rangle = 2\gamma_{kl}\delta(t-t') , \tag{4}$$

then the ensemble average of $\tilde{\rho}$ is given by[7]

$$\langle \dot{\tilde{\rho}}_{kl} \rangle = -(i\omega_{kl} + \tilde{\Gamma}_{kl})\langle \tilde{\rho}_{kl} \rangle + (\text{field terms}) , \tag{5}$$

where

$$\tilde{\Gamma}_{kl} = \Gamma_{kl} + \gamma_{kl} . \tag{6}$$

Let us now consider the case of a three-level system interacting with two fields. Let the field with phase Φ_1 (Φ_2) interact between the levels $|1\rangle$ ($|2\rangle$) and $|3\rangle$. Then from the above analysis it is clear that

$$\begin{aligned} &\tilde{\Gamma}_{13} = \Gamma_{13} + \gamma_{c1}, \quad \tilde{\Gamma}_{23} = \Gamma_{23} + \gamma_{c2} , \\ &\tilde{\Gamma}_{12} = \Gamma_{12} + \gamma_{c1} + \gamma_{c2} - 2\gamma_{\mathrm{CC}} . \end{aligned} \tag{7}$$

Here γ_{ci} gives the bandwidth of ith field and γ_{CC} is the cross correlation between two fields. *Assuming* that the four-wave-mixing signal can be obtained from Eq. (1), then the argument of Bloembergen *et al.* will show the existence of additional resonances if

$$\tilde{\Gamma}_{13} + \tilde{\Gamma}_{23} - \tilde{\Gamma}_{12} \neq 0 , \tag{8}$$

i.e.,

$$(\Gamma_{13} + \Gamma_{23} - \Gamma_{12}) + 2\gamma_{\mathrm{CC}} \neq 0 . \tag{9}$$

Thus the extra resonance can arise either from collisions ($\Gamma_{13}+\Gamma_{23}\neq\Gamma_{12}$) or from the cross correlation of the pump and probe ($\gamma_{\mathrm{CC}}\neq 0$). In general, cross correlations and collisions will together determine the characteristics of the extra resonance.

The prediction based on Eq. (9) for $\gamma_{\mathrm{CC}}\neq 0$ cannot lead to the correct behavior of the extra resonance since the validity of Eq. (1) is doubtful. In the presence of fluctuations, one should first calculate $|P|^2$ and then take the ensemble average, i.e., a correct procedure[1,8] will be to use[9]

$$I \propto \langle \, |P|^2 \rangle \neq |\langle P \rangle|^2 \; . \tag{10}$$

This result is true in the limit of small samples (specifically to a length less than c/γ_t where γ_t is a typical decay time[2]) since the propagation effects (via Maxwell-Bloch-type equations) are not considered. Yet, incoherently generated radiation, as we shall see later, also contributes to the small-sample intensity. For large samples the problem gets extremely complicated. In the following we implicitly restrict ourselves to the small-sample limit.

In this paper we present the main results of our investigation[10] of the effects of pump-probe cross correlation with regard to fluctuation-induced resonances in four-wave mixing. We present a theory derived from first principles and based on the relationship (10). We verify that a consistent theory based on Eq. (10) does indeed lead to the existence of fluctuation-induced resonances in four-wave mixing if both pump and probe are correlated. We also discuss the differences between the theories based on Eqs. (1) and (10). Note that the theory based on Eq. (1) is in a sense a decorrelated theory as it presumes a factorization like $\langle \rho_{kl}\rho_{ij} \rangle = \langle \rho_{kl} \rangle \langle \rho_{ij} \rangle$.

We follow the procedure of Agarwal and Kunasz (AK) but modify their equations to account for the cross correlation between the pump and probe. First we summarize the AK procedure.

(i) Since there is considerable redistribution of the generated radiation, a spectral analysis of the redistributed radiation is carried out and contributions to the spectral peak at (or within a few linewidths of) $2\omega_1-\omega_2$ are selected.

(ii) Each laser is assumed to have stabilized amplitude but a phase that undergoes diffusion. It is further assumed for simplicity that pump and probe are *completely* cross correlated, i.e.,

$$\begin{aligned} &\Phi_i(t)=\Phi(t), \quad \langle \dot\Phi \rangle = 0 \; , \\ &\langle \dot\Phi_i(t)\dot\Phi_j(t') \rangle = \langle \dot\Phi(t)\dot\Phi(t') \rangle = 2\gamma_c \delta(t-t') \; . \end{aligned} \tag{11}$$

Moreover, $\dot\Phi$ is a Gaussian random process.

(iii) Using the theory of multiplicative stochastic processes and following the method of Ref. 7, the ensemble averages $\langle \rho_{ij}(t) \rangle$ and $\langle \rho_{ij}(t)\rho_{kl}(t) \rangle$ are calculated. (These quadratic forms are needed in the evaluation of the mean of the four-wave-mixing signal.)

Now we recall the equations of AK (in their notation) which are to be modified. (A reader not interested in the detail may ignore this and go to the results.) In the work of AK, the basic density matrix equation is written in the form

$$\begin{aligned} &\frac{\partial \sigma}{\partial t} = M\sigma + I + e^{-i\delta t}(M_+\sigma + I_+) + e^{i\delta t}(M_-\sigma + I_-) \; , \\ &\delta = \omega_1 - \omega_2 \; . \end{aligned} \tag{12}$$

The matrices $M_\pm$ (M) describe the interaction with the probe (pump) field. All relaxation parameters are contained in the matrix M. The phase fluctuations of the fields are in the matrices M and $M_\pm$. By using transformations similar to that used in connection with Eq. (3), the explicit time dependence of these matrices can be removed. The resulting ensemble averages of the redefined density matrix elements involve additional matrices $\gamma_c F^2$ which depend on phase fluctuations. The complete cross correlation between the pump and probe changes some of those F matrices. For example $\psi^\pm$, which gives the linear response with respect to the probe and all order response with respect to the pump, is now given by [cf. AK, Eq. (3.7)]

$$\begin{aligned} &\psi^\pm = (\gamma_c F^2 - M \mp i\delta)^{-1}(M_\pm \langle \psi^{(0)} \rangle + I_\pm) \; , \\ &\langle \psi^{(0)} \rangle = (\gamma_c F^2 - M)^{-1} I \; . \end{aligned} \tag{13}$$

The ensemble average of the quadratic forms is still given by AK, Eqs. (3.16) and (3.17) but the matrix U is now given by [cf. AK, Eq. (3.13)]

$$[\mp i\delta + \gamma_c(F_\alpha+F_\beta)^2]U^{\pm\mu}_{\alpha\beta} - \sum_i (M_{\alpha i}U^{\pm\mu}_{i\beta} + M_{\beta i}U^{\pm\mu}_{i\alpha}) = g^{\pm\mu}_{\alpha\beta} \; . \tag{14}$$

Physically, U's are the first-order (with respect to the probe) contributions to the quadratic forms $\langle \psi_\alpha \psi_\beta \rangle$. The spectrum of the redistributed radiation is [cf. AK, Eq. (4.16)]

$$\begin{aligned} S^-_{pg} = &\sum_j [z - M + \gamma_c(F+1)^2]^{-1}_{pj} W^-_{gj} \\ &+ \sum_{j,r,l} [z-M+\gamma_c(F+1)^2]^{-1}_{pj} M^-_{jr} [z-M+\gamma_c(F+1)^2 - i\delta]^{-1}_{rl} [U^{\mu+}_{gl} + I_l \psi^+_g (z+\gamma_c - i\delta)^{-1}] \\ &+ \sum_j [z-M+\gamma_c(F+1)^2]^{-1}_{pj} (I_-)_j (z+\gamma_c - i\delta)^{-1} \psi^+_g \; . \end{aligned} \tag{15}$$

Here, W's give the second-order contributions to the quadratic forms $\langle \psi_\alpha \psi_\beta \rangle$. Since the spectrum has many peaks resulting from the eigenvalues of M which result in the redistribution of radiation, we isolate the contributions to the spectral peak in the vicinity of $2\omega_1-\omega_2$. Such a peak arises from poles in the expression Eq. (15) such that $\mathrm{Im} z \sim \delta$ and $-\omega_{12}$ (which is of the same order as δ).

We show our results in Figs. 1–3. Here we have chosen the same parameters as used originally in the experiments[5] on Na D lines. Figure 1 clearly shows that the correlated fluctuations of the pump and probe lead to the extra resonance in four-wave mixing *even* if there are no collisions. The next two figures show the combined effects of fluctuations and collisions. Figures 2 and 3 show that the peak-to-background ratio improves considerably if $\Gamma_p \neq 0$, $\gamma_c \neq 0$. In Fig. 4 we show the decorrelat-

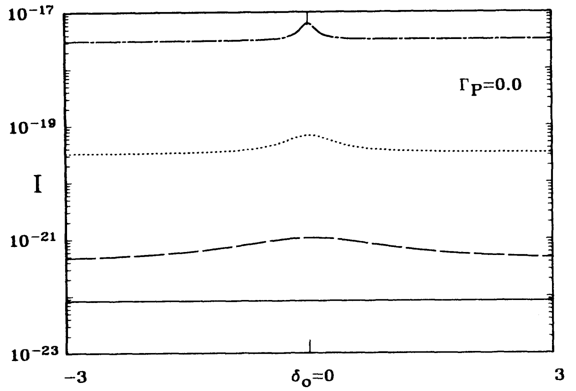

FIG. 1. Intensity I of the fluctuation-induced extra resonance in arbitrary units as a function of the detuning parameter $\delta_0=(\omega_1-\omega_2+\omega_{12})/\gamma$ for various values of the laser bandwidth parameter $\gamma_c/\gamma=0$, 1, 10, and 100, with γ_c values increasing from bottom to top. The other parameters have been chosen as $\gamma_1=\gamma_2=\gamma=2\pi$ (10) MHz, $\Delta_2/\gamma=-1.5\times10^3$, $\omega_{12}/\gamma=5.1\times10^4$, $\Gamma_1=\Gamma_2=\gamma(\Gamma_p+\frac{1}{2})$, $\Gamma_0=\gamma(\Gamma_p+1)$, $\Gamma_p=0$. For convenience, all the dipole matrix elements have been set at unity. The computed signal is proportional to the intensity of the probe and to the square of the pump's intensity. The actual δ_0 values for $\gamma_c=10$ and 100 are, respectively, three and ten times those shown.

ed result I_D obtained from the theory of Prior *et al.* and its comparison with our theory. In the context of the three-level model, the results of Prior *et al.* can be obtained from ψ^+, as given by Eq. (13). It is clear that the differences between the two theories are quite significant. The peak-to-background ratio in the decorrelated theory is rather high. The differences in the two theories[11] continue to be important until the collisional contribution to the Γ's becomes large in comparison to γ_c. These differences can be understood if we recall that the laser fluctuations lead to considerable redistribution of the generated radiation via nonlinear mixing and that the redistributed radiation exhibits rather dominant fluctuation-induced resonances. The decorrelated theory essentially ignores the redistribution effects and adds all the coherent contributions to the extra resonances. Only a spectral analysis of the generated radiation can lead to a proper understanding of the radiation at $2\omega_1-\omega_2$.

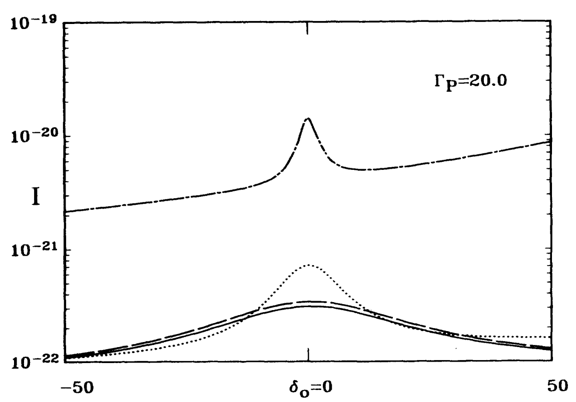

FIG. 2. Same as in Fig. 1, but with $\Gamma_p=20$, so that the signal now shows the presence of both fluctuations and collisions. The actual δ_0 values for $\gamma_c=10$ and 100 are, respectively, three and ten times those shown.

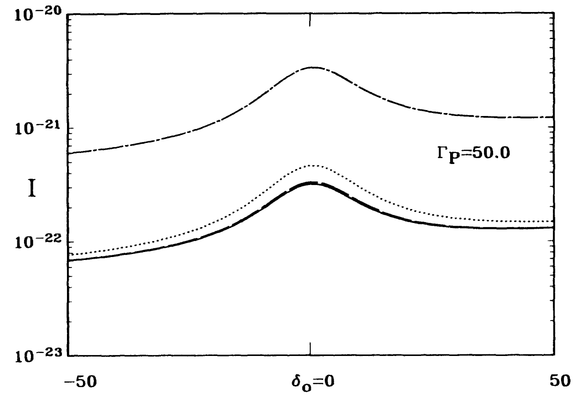

FIG. 3. Same as in Fig. 1, but with $\Gamma_p=50$. The actual δ_0 values are as shown for all γ_c.

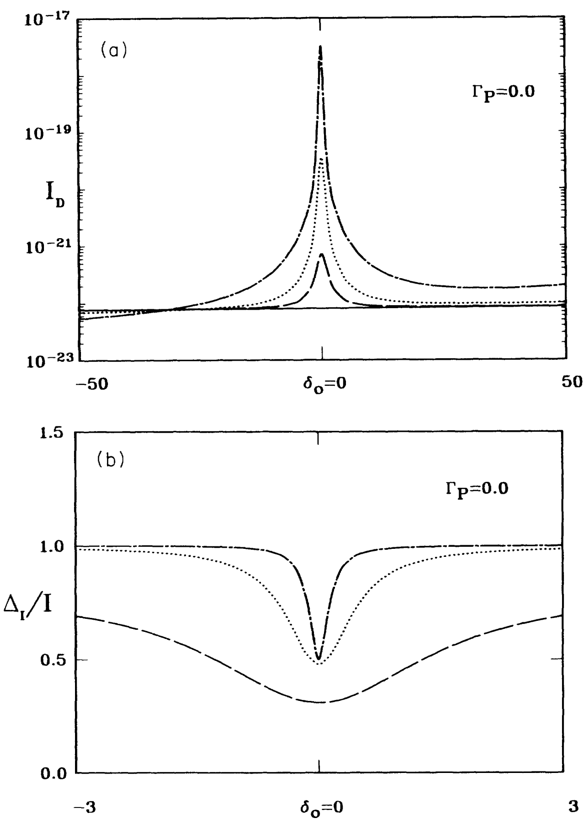

FIG. 4. (a) The signal I_D as obtained from the decorrelated theory. All the parameters are the same as in Fig. 1. The actual δ_0 values for $\gamma_c=10$ and 100 are three and ten times those shown. (b) Differences between our theory and the decorrelated theory $\Delta I/I\equiv(I-I_D)/I$. δ_0 values displayed are in a narrower range than in (a), but δ_0 values for $\gamma_c=10$ and 100 are still multiplied by three and ten, respectively.

In conclusion, we would like to emphasize that the stochastic field-induced resonances, as discovered by Prior *et al.*, should also be seen in higher-order nonlinear mixing experiments with correlated pump and probe. It would also be interesting to examine the effect of cross correlation on near-resonant nonlinear mixing experiments.

This work was supported in part by National Science Foundation Grant No. PHY-86-04504 through the University of Colorado, and the computations were done on the Joint Institute for Laboratory Astrophysics (JILA) VAX 8600. One of us (C.V.K.) thanks Dr. Ragini Saxena for extensive discussions.

---

[1]G. S. Agarwal and C. V. Kunasz, Phys. Rev. A **27**, 996 (1983).

[2]Effects of fluctuations and saturation on resonant degenerate four-wave mixing have been considered by G. Alber, J. Cooper, and P. Ewart, Phys. Rev. A **31**, 2344 (1985).

[3]Y. Prior, I. Schek, and J. Jortner, Phys. Rev. A **31**, 3775 (1985).

[4]Complete cross correlation between pump and probe is very important in high-resolution spectroscopic work [D. G. Steel and S. C. Rand, Phys. Rev. Lett. **55**, 2285 (1985); J. E. Thomas, P. R. Hemmer, S. Ezekiel, C. C. Leiby, R. H. Picard, and C. R. Willis, *ibid.* **48**, 867 (1982)] and other problems such as population trapping [B. J. Dalton and P. L. Knight, J. Phys. B **15**, 3997 (1982); S. Swain, J. Opt. Soc. Am. B **2**, 1666 (1985); G. S. Agarwal and N. Nayak, J. Phys. B **19**, 3375 (1986)].

[5]Y. Prior, A. R. Bogdan, M. Dagenais, and N. Bloembergen, Phys. Rev. Lett. **46**, 111 (1981); A. Bogdan, M. Downer, and N. Bloembergen, Phys. Rev. A **24**, 623 (1981).

[6]Recently, Y. H. Zou and N. Bloembergen, Phys. Rev. A **34**, 2968 (1986), have presented experimental evidence for the stochastic field-induced resonances.

[7]G. S. Agarwal, Phys. Rev. A **18**, 1490 (1978).

[8]Other authors, notably among them S. Mukamel and E. Hanamura [Phys. Rev. A **33**, 1099 (1986)]; and E. Hanamura and S. Mukamel [J. Opt. Soc. Am. B **3**, 1124 (1986)], have also emphasized the usage of $\langle |P|^2 \rangle$ rather than $|\langle P \rangle|^2$ in the perturbative calculation of the four-wave-mixing signal produced by a medium with fluctuating pump fields. Strong spatial correlations in the medium also necessitate the usage of $\langle |P|^2 \rangle$. It may be noted that the model of AK can also be used for studying pump-induced saturation effects.

[9]Note that for fluorescence from a single atom, the total intensity which consists of incoherent and coherent components is proportional to the excited-state population and not to the absolute square of the dipole moment, see B. R. Mollow, Phys. Rev. **188**, 1969 (1969).

[10]We treat the case of forward four-wave mixing only. Similar results are expected to hold for phase conjugation geometry if the absorption length is small and if saturation effects are ignored.

[11]Y. Prior and A. Levine (unpublished) have informed us that for a certain model of the correlations, the result of Ref. 3 is recovered even when one uses $I \propto \langle |P|^2 \rangle$.