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Four-wave mixing with time-delayed correlated fields of arbitrary bandwidths and pump intensities

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We consider the generation of four-wave-mixing signals by an ensemble of two-level atoms interacting with broadband, correlated, pump and probe fields in the forward geometry. We show how the calculation of the four-wave-mixing signal reduces to the solution of six coupled Langevin equations that are numerically integrated using Monte Carlo methods. We account for the pump-induced saturation effects although the probe is weak. The finite correlation time of the pump and probe fields is also included. For weak and δ-correlated pump fields, we reproduce the results of Morita and Yajima [Phys. Rev. A **30**, 2525 (1984)]. We find that even for short correlation times of the fields, the four-wave-mixing signal, as a function of the delay between the pump and the probe fields, starts reviving with increase in the pump intensity. This revival as a function of the pump-probe delay time becomes more pronounced with further increase in the pump intensity.

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I. INTRODUCTION

Conventional techniques for measuring dephasing times of atoms are based on the method of photon echoes or variations thereof. All collisional broadening studies give the rate for destruction of coherence, i.e., T_2 times. These techniques rely on the use of femtosecond laser pulses to measure subpicosecond lifetimes and the smallest dephasing times that can be measured are limited by the width of the laser pulses available. In the past few years, incoherent broadband, correlated, pump and probe pulses have been used to study dephasing times of atoms [1–5] in the picosecond or subpicosecond regime. This method was originally proposed by Morita and Yajima [1], who studied the four-wave-mixing signal as a function of the time delay between the pump and the probe fields. They discovered that in the limit of weak pump and probe fields and in the limit of zero correlation time of the field fluctuations, i.e., δ-correlated fluctuations, the decay of the four-wave-mixing signal as a function of the delay between the pump and the probe directly yields the dephasing time of atomic media when the probe follows the pump in time. These authors established that when using incoherent broadband light, the smallest dephasing times that can be measured are limited not by the temporal duration of the light (as with pulsed lasers) but by the correlation time associated with the fluctuating light. This is indeed a powerful technique, since it is easier to produce incoherent light with a short correlation time than to produce ultrashort light pulses, especially over a broad spectral range.

Some theoretical generalizations of the work of Morita and Yajima have since appeared [6–9]. One of us [6] calculated the four-wave-mixing signal for fields with arbitrary bandwidths but within the framework of third-order perturbation theory. Thus both the pump and probe fields were treated as weak fields in that work. The delay dependence of the four-wave-mixing signal was expressed in terms of the third-order susceptibility of the medium and the results derived in that work were also applicable to multilevel systems. The four-wave-mixing signals for multilevel systems irradiated by pulses with short correlation time have also been calculated by Mi *et al.* [7] and by Hartmann and co-workers [10]. Related work on the effect of time-delayed beams on fluctuation-induced resonances in four-wave-mixing has been reported by Kofman, Levine, and Prior [11].

The calculation of the signal when the pump and the probe are both intense is a very complicated problem and considerable attention has been focused of late on the strong pump situation. The usual perturbative approaches are no longer valid and one has to resort to other methods for studying the strong-field cases. Beach, DeBeer, and Hartmann [2] carried out experiments in the strong-field regimes while Tchenio *et al.* developed diagrammatic methods to handle the case of strong pump and weak probe [9]. Finkelstein and Berman [12] have addressed this problem theoretically and reported analytical results while Tchenio *et al.* [13] have reported experimental and theoretical results for this situation. In related context, Gheri, Marte, and Zoller [14] have reported analytical results for the absorption of a time-delayed, weak probe by two-level atoms in the presence of a strong pump field, where the pump and probe are assumed to have correlated phase fluctuations. One of the outstanding problems with strong fields is that even if the correlation time of the field fluctuations is short, the system reacts differently from the weak-field case, because for strong fields the Rabi flopping time may become short compared to the correlation time of the field and thus bring in new features. This aspect has been discussed at length in the context of resonance fluorescence [15,16]. If the pump fluctuations are approximated by a telegraphic signal, i.e., say, the amplitude is treated as a discontinuous Markov process, the problem can be handled analytically [15]. However, the real difficulty arises when the pump is treated as a chaotic field. This is indeed the case in most experiments related to spectroscopic applications where a multimode laser is used and it is well known that the field from such a laser can be accurately modeled as a chaotic field. (In view of the current tremendous interest in the field of chaos as related to the dynamics of nonlinear systems, we point out that the use of the word "chaotic" here implies a thermal field.)

In the work of Finkelstein and Berman [12] dealing with strong fields, the authors consider the depletion of the ground state by the strong pump, but require the bandwidth of the pump to be larger than the Rabi frequency. However, in our work, we can incorporate not only the depletion of the ground state but also account for the Rabi frequency being larger than the bandwidth of the field. This situation is also considered in the work of Gheri, Marte, and Zoller [14] in the context of phase fluctuations of the field.

In the present paper, we analyze the four-wave-mixing signals in the forward geometry produced by an ensemble of two-level atoms. The pump and the probe are treated as chaotic fields and are derived from the same source. Our calculations take into account arbitrary pump intensities while the probe is taken to be weak, as is the case in most experiments related to four-wave mixing. The probe is usually weak in such works since it is used only to "read" the response of the atomic system excited by the strong pump. We also investigate the effects of the finite correlation time of the field fluctuations. Our calculations rely on Monte Carlo methods to compute the four-wave-mixing signals as a function of the time delay between the pump and the probe [16]. We present detailed numerical results demonstrating the dependence of the signal on the correlation time of the field fluctuations and on the intensity of the pump.

The rest of the paper is as follows: in Sec. II we present the theoretical model used to calculate the dependence of the four-wave-mixing signal on the time delay between the pump and probe fields, when fully correlated fields interact with an ensemble of two-level atoms. We show how the problem reduces to the solution of six coupled Langevin equations. In Sec. III we discuss the numerical method for the solution of these equations and details of the Monte Carlo method are reviewed. Section IV contains the results of our study and the conclusions drawn from this work.

II. FOUR-WAVE MIXING IN TWO-LEVEL SYSTEMS IN CORRELATED FIELDS

In this section we discuss the theoretical model on which this work is based. We consider the interaction of an ensemble of two-level atoms, with resonant frequency ω_0, with broadband fields of the form

$$E(t)=\exp(-i\omega_l t+i\mathbf{k}_l\cdot\mathbf{r})\hat{\epsilon}\epsilon(t) +\exp(-i\omega_l t+i\mathbf{k}_s\cdot\mathbf{r})g\hat{\epsilon}\epsilon(t-\tau) \ . \tag{2.1}$$

Clearly the composite electric field consists of two separate fields. The first part of this expression represents the pump (with subscripts l) and the second part refers to the probe (with subscripts s). ω_l is the laser frequency, and since the pump and probe are derived from a single source, is the same for both fields. $\mathbf{k}_l$ and $\mathbf{k}_s$ represent the wave vectors for the pump and probe, respectively, while $\hat{\epsilon}$ is the unit polarization vector. The time delay between the pump and the probe is represented by τ, which can be varied as desired. The factor g in Eq. (2.1) accounts for the weak probe (i.e., $g \ll 1$ in general).

The system thus described interacts with a pump field in the direction $\mathbf{k}_l$ and a probe field in the direction $\mathbf{k}_s$, both fields obtained from the same source but time delayed with respect to one another. Since all the fields are at the same frequency, this is a case of degenerate four-wave-mixing, and we consider the signal at frequency ω_l and in the direction $2\mathbf{k}_l-\mathbf{k}_s$. The envelope of the electric field $\epsilon(t)$ is considered to be a stochastic function of time, t. The pump and probe are fully correlated since they are derived from the same source. The probe is assumed to be weak while the pump is of arbitrary magnitude.

At this point we need to choose a model to represent the pump field fluctuations. Since chaotic fields are a universal feature of multimode lasers, we consider the electric field to be chaotic in nature. Thus $\epsilon(t)$ is taken to be a Gaussian process with a correlation time $1/\Gamma$ and has the properties

$$\begin{aligned}&\langle\epsilon(t)\rangle=0,\quad \langle\epsilon^*(t)\epsilon(t')\rangle=|\epsilon_0^2|\exp(-\Gamma|t-t'|)\ ,\\ &\langle\epsilon(t)\epsilon(t')\rangle=0\\ &\qquad=\langle\epsilon^*(t)\epsilon^*(t')\rangle\ .\end{aligned}\tag{2.2}$$

These expressions indicate that the electric field $\epsilon(t)$ is a Gaussian process with zero mean, variance of $|\epsilon_0^2|$, and a correlation time of the fluctuations given by $1/\Gamma$.

A similar model was adopted by Morita and Yajima [1] and they reported analytical results for the behavior of the four-wave-mixing signal versus pump-probe delay. Their results were obtained for the case of weak pump fields and zero correlation time of the broadband field, i.e., for δ-correlated fluctuations ($\Gamma\rightarrow\alpha$). Agarwal [6] presented results for weak pump but for finite correlation times.

The dynamical behavior of the atoms interacting with a field of the form described by Eq. (2.1) is given by the usual optical Bloch equations. Let Ψ_1, Ψ_2, and Ψ_3 denote the components of the atomic dipole moments, and the population inversion, respectively. In our notation, if we represent the density-matrix operator by ρ then the off-diagonal elements of the density matrix, ρ_{21} and ρ_{12}, are given by Ψ_1, and Ψ_2, respectively, and Ψ_3 equals $\frac{1}{2}(\rho_{11}-\rho_{22})$, i.e., the difference in population between the excited state and the ground state. We now make the rotating-wave approximation and transform to a frame rotating with the frequency of the pump field such that the Bloch equations can now be written as

$$d\tilde{\Psi}/dt = M\tilde{\Psi}+I \ , \tag{2.3}$$

where

$$M=\begin{bmatrix} -1/T_2+i\Delta & 0 & -2ix^*(t) \\ 0 & -1/T_2-i\Delta & 2ix(t) \\ -ix(t) & ix^*(t) & -1/T_1 \end{bmatrix} . \tag{2.4}$$

Here, Δ is the detuning between the atomic frequency and the laser frequency, i.e., $\Delta=\omega_0-\omega_l$,

$$\begin{aligned} &x(t)=\mathbf{d}\cdot\hat{\epsilon}/\hbar\{\epsilon(t)+\exp[i(\mathbf{k}_s-\mathbf{k}_l)\cdot\mathbf{r}]g\epsilon(t-\tau)\} \ , \\ &\tilde{\Psi}_1=\exp(-i\omega_l t+i\mathbf{k}_l\cdot\mathbf{r})\Psi_1 \ , \\ &\tilde{\Psi}_3=\Psi_3 \ , \\ &I_1=I_2=0 \ , \\ &I_3=-1/2T_1 \ , \end{aligned} \tag{2.5}$$

and we define

$$u=T_2/T_1 \ . \tag{2.6}$$

In (2.4), T_1 and T_2 denote, respectively, the longitudinal and transverse relaxation times of the atomic system. (All time units in our work have been normalized to T_2.) We calculate the four-wave-mixing signal to first order in the probe field. We thus write

$$\tilde{\Psi}=\tilde{\Psi}^{(0)}+\tilde{\Psi}^{(1)}+\cdots \ , \tag{2.7}$$

where $\tilde{\Psi}^{(0)}$ and $\tilde{\Psi}^{(1)}$ are given by

$$d\tilde{\Psi}^{(0)}/dt=M^{(0)}\tilde{\Psi}^{(0)}+I \tag{2.8}$$

and

$$d\tilde{\Psi}^{(1)}/dt=M^{(0)}\tilde{\Psi}^{(1)}+M^{(1)}\tilde{\Psi}^{(0)} \ . \tag{2.9}$$

Here $M^{(0)}$ is obtained from (2.4) by setting $g=0$, and $M^{(1)}$ is given by (2.5) with $1/T_2=\Delta=1/T_1=0$ and

$$x(t)=\mathbf{d}\cdot\epsilon/\hbar g\epsilon(t-\tau)\exp[i(\mathbf{k}_s-\mathbf{k}_l)\cdot r] \ . \tag{2.10}$$

Note that (2.9) holds for times t greater than or equal to τ, since the probe starts acting at time τ. We rewrite the solution of Eq. (2.9) as

$$\tilde{\Psi}^{(1)}=\exp[i(\mathbf{k}_s-\mathbf{k}_l)\cdot\mathbf{r}]A+\exp[-i(\mathbf{k}_s-\mathbf{k}_l)\cdot r]F \ . \tag{2.11}$$

Clearly the column matrix is given by the solution of

$$dF/dt=M^{(0)}F+ig(\mathbf{d}\cdot\epsilon/\hbar)\epsilon^*(t-\tau)\begin{bmatrix} -2\tilde{\Psi}_3^{(0)}(t) \\ 0 \\ \tilde{\Psi}_2^{(0)}(t) \end{bmatrix} , \tag{2.12}$$

where $\tilde{\Psi}^{(0)}$'s are to be obtained from the solution of Eq. (2.8). Equations (2.8) and (2.12) are Langevin equations with the stochastic modulation appearing in multiplicative form. The four-wave-mixing signal for homogeneously broadened media can be shown to be proportional to S, which is defined by

$$S=\lim_{t\to\infty}\langle F_2^*(t)F_2(t)\rangle \ , \tag{2.13}$$

where the brackets refer to stochastic averaging with respect to the fluctuations of the field $\epsilon(t)$. In the present work, we calculated the signal S for arbitrary intensity and bandwidth of the pump. Note that Eq. (2.8) leads to $\tilde{\Psi}^{(0)}$ which is an infinite-order functional of the stochastic field and this indeed is the source of complication when one attempts to calculate S analytically. In this paper, we use Monte Carlo methods to numerically integrate the Langevin equations (2.8) and (2.12) and the numerical technique is described in Sec. III.

III. NUMERICAL PROCEDURE

As stated earlier, the inclusion of arbitrary pump intensities and bandwidths makes the problem very difficult to solve analytically and we resort to numerical methods to obtain the dynamical behavior of the atom. To carry out the Monte Carlo simulations, we first need to produce the complex, stochastic electric field, a method for which is outlined here. The electric field $x(t)$ is assumed to follow an Ornstein-Uhlenbeck process, i.e., it can be represented by exponentially correlated (colored) Gaussian noise, with the properties

$$\langle x(t)\rangle=0, \quad \langle x(t)x^*(t')\rangle=D\Gamma\exp(-\Gamma|t-t'|) \ , \tag{3.1}$$

where Γ is the inverse of the correlation time and $D\Gamma$ is the variance of $x(t)$. This colored noise has a Lorentzian spectral profile with a full width at half maximum (FWHM) of 2Γ.

The algorithm used to generate $x(t)$ is detailed in Ref. [17] and is briefly described here. The first step is to produce the complex, Gaussian δ-correlated (white) noise g_w, which is the source term for the colored noise. g_w has the well-known properties

$$\langle g_w(t)\rangle=0, \quad \langle g_w(t)g_w^*(t')\rangle=2D\delta(t-t') \ , \tag{3.2}$$

which completely determine all of its statistical properties. It is easily produced by the Box-Mueller algorithm:

$$g_w=[-2D\Delta t\ln(a)]^{1/2}\exp(2\pi ib) \ , \tag{3.3}$$

where a and b are computer generated, uniformly distributed random numbers between 0 and 1 and Δt is the integration step size [used in integrating (2.8) and (2.12)].

Exponentially correlated colored noise as described in Eq. (3.1) is obtained from the equation

$$dx/dt = -\Gamma x + \Gamma g_w \tag{3.4}$$

in which g_w is the Gaussian white noise as defined before. It has been shown previously [17] that by integrating (3.4) we get

$$x(t+\Delta t) = x(t)\exp(-\Gamma\Delta t) + h(t) , \tag{3.5}$$

where h depends on g_w and is Gaussian with zero mean and a second moment given by

$$\langle |h(t,\Delta t)|^2 \rangle = D\Gamma[1-\exp(-2\Gamma\Delta t)] . \tag{3.6}$$

Thus to generate the colored noise $x(t)$, we first produce h by the formula

$$h = \{-D\Gamma[1-\exp(-2\Gamma\Delta t)]\ln(a)\}^{1/2}\exp(2\pi i b) , \tag{3.7}$$

where, as before, a and b are computer generated, uniformly distributed random numbers between 0 and 1. The exponentially correlated noise is then obtained from expression (3.5).

The two Langevin equations (2.8) and (2.12) were solved numerically using the colored noise generated above. An Euler method was used for the numerical stochastic integration, which is very accurate provided the time step of the numerical integration is much smaller than all other relevant time scales appearing in the problem. For most of this work we chose a time step of $\Delta t = 0.0001 T_2$. To investigate the effects of δ-correlated fluctuations, the above noise algorithm was used in the limit of $\Gamma \gg D$, instead of explicitly using the Box-Mueller algorithm to produce the white noise.

In our work the noise modulation appears in multiplicative form and requires caution when dealing with the white-noise limit. During the Euler method integration, we are restricting ourselves to terms in the first order with respect to Δt. Suppose for illustration purposes that the Langevin equation we are integrating is of the form

$$dy(t)/dt = m[y(t)] + n[y(t)]\beta(t) , \tag{3.8}$$

where $m[y(t)]$ and $n[y(t)]$ are functions of y and $\beta(t)$ is the white-noise term with zero mean and a variance of $2D$, which appears in the multiplicative form. It is shown in Ref. [18] that in the correct form for numerically integrating this equation to first order in Δt, the integration step is

$$y(t+\Delta t) = y(t) + m[y(t)]\Delta t + n[y(t)]w(t) + \{\tfrac{1}{2}n[y(t)]dn[y(t)]/dy(t)\}w(t)^2 , \tag{3.9}$$

where $w(t) = \beta(t')dt'$ is a Gaussian random number with zero mean and variance of $2D\Delta t$, which can be obtained from the Box-Mueller algorithm. It is emphasized in Ref. [18] that the last term in Eq. (3.9) vanishes for additive white noise and also plays a diminishing role for colored noise (we refer the interested reader to Ref. [18] for further details of this aspect). However, we have used this general form (3.9) throughout our work since we have used the same algorithm to produce both the white and colored noise.

The set of Langevin equations given by (2.8) was first solved for time $0 < t < \tau$, with the initial conditions $\tilde{\Psi}_1(t=0)=0$, $\tilde{\Psi}_2(t=0)=0$, and $\tilde{\Psi}_3(t=0)=-0.5$ and the values of the electric field $x(t)$ stored. After a time τ, Eqs. (2.12) were simultaneously solved with (2.8) and the values of the stored electric field were used in (2.12). This accounted for the time delay between the pump and the probe fields. The initial conditions for (2.12) were $F_1(0)=F_2(0)=F_3(0)=0$. The delay in our work was changed from 0 to 4 time units. For a given delay, the computed values of F_2 were allowed to reach a steady state and from these steady-state values the four-wave-mixing signal was obtained as per Eq. (2.13). The signal in our results represents an averaging over several thousand trajectories, each with a different set of random numbers to ensure that the results are not influenced by small statistics. The signal thus computed was plotted as a function of the delay between the pump and the probe.

IV. RESULTS

In this section we report the results on the dependence of the four-wave-mixing signal on the pump-probe time delay, when correlated fluctuating fields interact with an ensemble of two-level atoms. As stated earlier, our Monte Carlo methods allow us to examine the regime of arbitrary pump intensities and arbitrary bandwidths of the fluctuating fields. Figures 1–9 contain the results of our numerical computations for a wide range of parameter values, corresponding to the broadband field. In our numerical work, we write

$$|\mathbf{d}\cdot\boldsymbol{\epsilon}/\hbar|^2 \langle \epsilon^*(t)\epsilon(\tau)\rangle = D\Gamma\exp(-\Gamma|t-\tau|) , \tag{4.1}$$

where our units are normalized to T_2 such that to convert to real time scales (say, for comparison to experiments) one would use τ/T_2, ΓT_2, and $D\Gamma T_2^2$. Thus two independent parameters D and Γ characterize the pump and the probe fields. The parameter $D\Gamma$ is a measure of the strength of the pump field. Note that in the limit $\Gamma\to\infty$ (4.1) goes over to $2D\delta(t-\tau)$. Throughout our work the probe is of course assumed to be weak and so the four-wave-mixing signal is proportional to g^2. All results displayed in this paper depict the signal normalized to unity at zero delay.

In Fig. 1 we show the four-wave-mixing signal as a function of the delay time τ between the pump and the probe. This figure is for the case when Γ is large (≈ 100) and when both the pump and the probe are weak ($D=0.0001$). This corresponds to the case studied by Morita and Yajima [1], where all fields are weak and the field fluctuations are δ correlated. We compare our results with the analytic expressions given by Morita and Yajima [1] for two cases, when the ratio u ($\approx T_2/T_1$) is zero and when $u=2$. Clearly, the results from our Monte Carlo simulations are in full agreement with the analytical results of Morita and Yajima.

Figure 2(a) illustrates the effect of increasing the strength of the pump ($D=0.1$) while keeping the correlation time fixed ($\Gamma=100$). We display the results for three cases, when $u=0$, 1, and 2. There is an obvious deviation from Morita and Yajima's results because the weak-field approximation is now violated. The exponential fit of these curves shows that for $u=0$, the decay is essentially determined by power broadening [see Fig. 2(b)], which is plotted on a semilogarithmic scale). With an increase in u, the decay of the signal is faster and also the background signal increases with an increase in u.

In Fig. 3 we compare the signals for a fixed value of $\Gamma=100$, $T_1=T_2$, and for different intensities of the pump. The values of D we depict are 0.01, 0.1, and 0.5 in increasing order of the pump strengths. Clearly, with an increase in the pump intensity, the signal decays faster and has a higher background level. Also, we notice a revival of the four-wave-mixing signal for large values of the pump intensity. This revival behavior is more clearly seen in Fig. 4. This figure corresponds to the case of fixed pump intensity but different coherence times of the pump. We have chosen the pump intensity, i.e., the product ΓD to be 10 in our dimensionless units and varied the coherence time of the pump $(1/\Gamma)$ over values 0.01, 0.1, and 1. The revival of the signal is very prominent for large values of the coherence time of the pump. The signal is in fact quite large for delay times much larger than the coherence time of the pump. The pump here is strong enough to saturate the atomic transition. It should be pointed out that this revival behavior is qualitatively similar to the behavior predicted by Tchenio *et al.* [13] and by Finkelstein and Berman [12], and experimentally observed by Tchenio *et al.* [13]. Figure 5 is similar to Fig. 4 but now the parameter $D\Gamma$ is chosen to be 1, i.e., the pump field is weaker than in the case shown in Fig. 4. For small values of the coherence time, i.e., fast pump field fluctuations, we see no revival of the signal and the signal decays in accordance with the dephasing time. But even for this weaker pump field situation, for large values of the pump coherence time (small Γ) we see a revival of the signal. In this figure the revival is seen even when the coherence time of the pump is comparable to the dephasing time.

In Fig. 6 we show the behavior of the signal for a fixed value of D of 0.1, but for different coherence times of the pump. For short coherence times $(\Gamma=100)$, the signal shows exponential decay. With the increase in the coherence time of the pump the signal decays slower as a func-

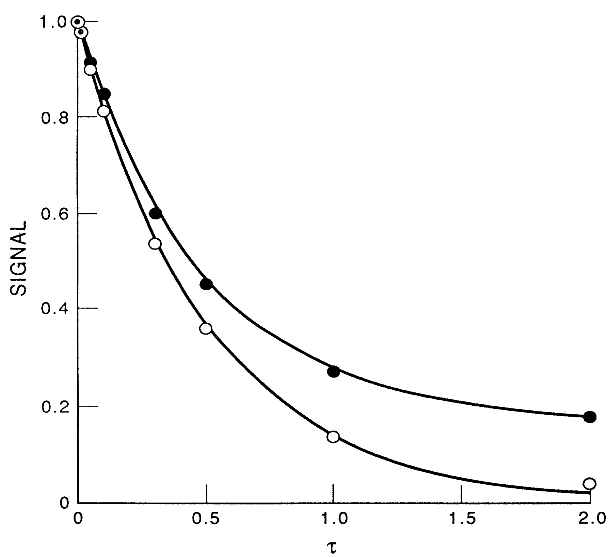

FIG. 1. Four-wave-mixing signal (dimensionless) as a function of the delay (units of T_2) between pump and probe for weak and δ-correlated fields. The smooth lines are the analytic results of Ref. [1] while the open and solid circles are from the methods of this paper for $u=0$ and 2, respectively, and $\Gamma=100$ and $D=0.0001$.

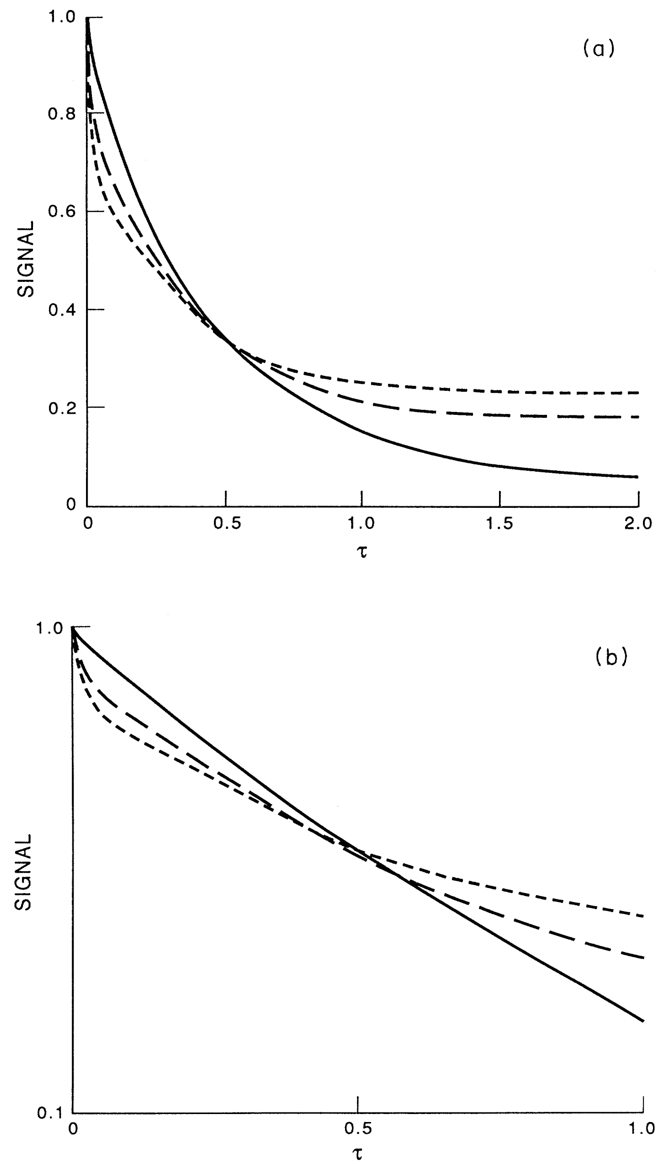

FIG. 2. (a) Four-wave-mixing (FWM) signal (dimensionless) vs delay (units of T_2) for strong fields with $\Gamma=100$ and $D=0.1$ for $u=0$ (solid), $u=1$ (long dash) and $u=2$ (short dash). (b) (a) on a semilogarithmic plot.

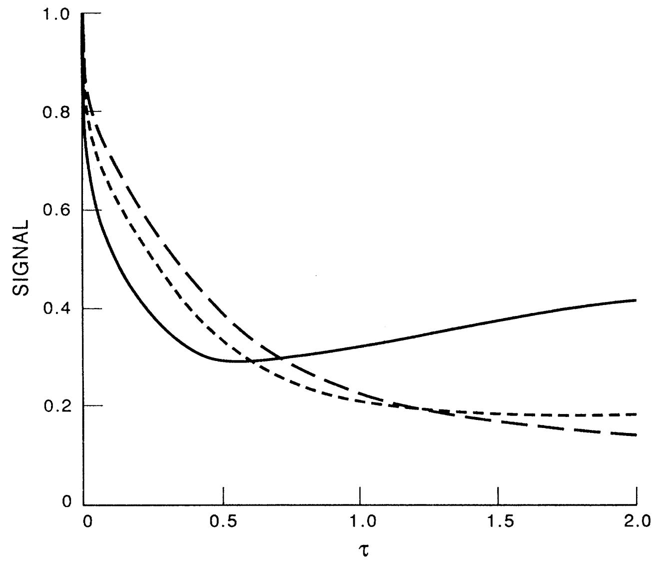

FIG. 3. FWM signal (dimensionless) vs τ (units of T_2) for $u=1$, $\Gamma=100$, and three different pump intensities. $D=0.01$ (long dash), $D=0.1$ (short dash) and $D=0.5$ (solid).

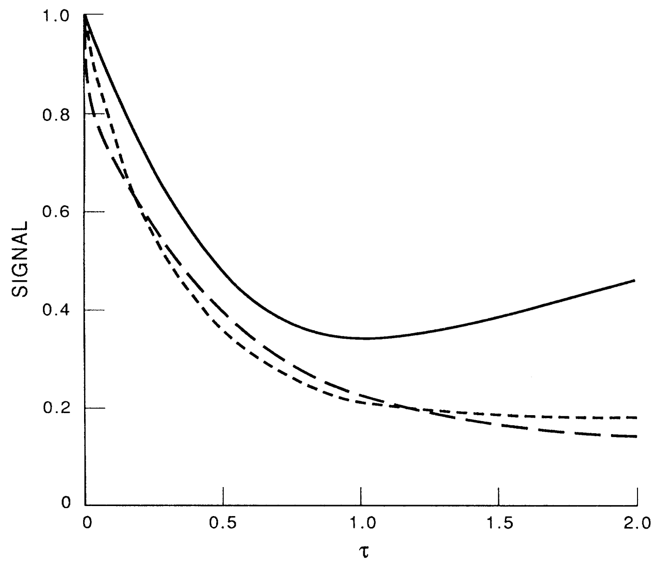

FIG. 5. FWM signal (dimensionless) vs τ (units of T_2) for $\Gamma D=1$ and $u=1$. $\Gamma=100$, $D=0.01$ (long dash), $\Gamma=10$, $D=0.1$ (short dash), and $\Gamma=1$, $D=1$ (solid).

tion of the pump-probe delay. This behavior is consistent with that displayed in Fig. 3. In both cases, we find that with an increase in the parameter $D\Gamma$, the signal decays faster. However, the background signal level, i.e., the signal for large delays, is the same in Fig. 6 for all values of Γ while in Fig. 3 it increases with increase in D. From these results, it seems the background signal level may be a function of D alone. Figure 7 is similar to Fig. 3, except now the coherence time of the pump $(1/\Gamma)$ is fixed at 1 instead of 0.01 and D is varied over 10, 1, and 0.1. Consistent with our previously noted observations, the signal decays faster with an increase in the product $D\Gamma$, the background signal level increases with an increase in D, and in fact for $\Gamma=1$ and $D=10$ we see the revival of the signal for large delays.

In all of the results discussed above, we have con-

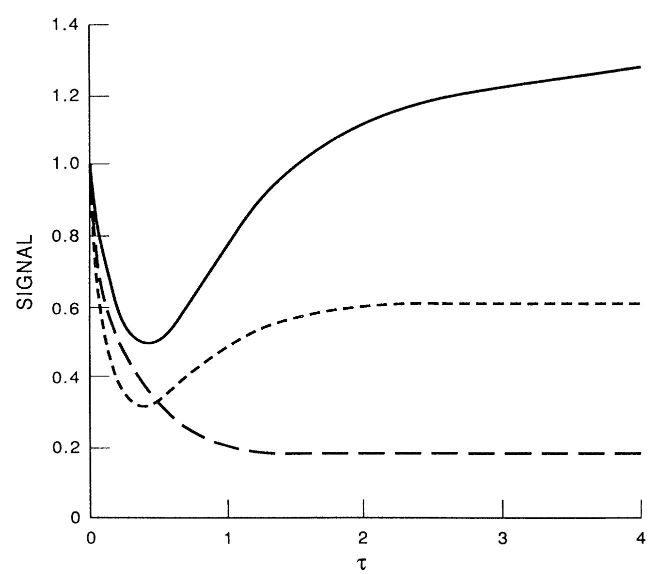

FIG. 4. FWM signal (dimensionless) vs τ (units of T_2) for fixed pump intensity (ΓD) of 10 and $u=1$. $\Gamma=100$, $D=0.1$ (long dash), $\Gamma=10$, $D=1$ (short dash), and $\Gamma=1$, $D=10$ (solid).

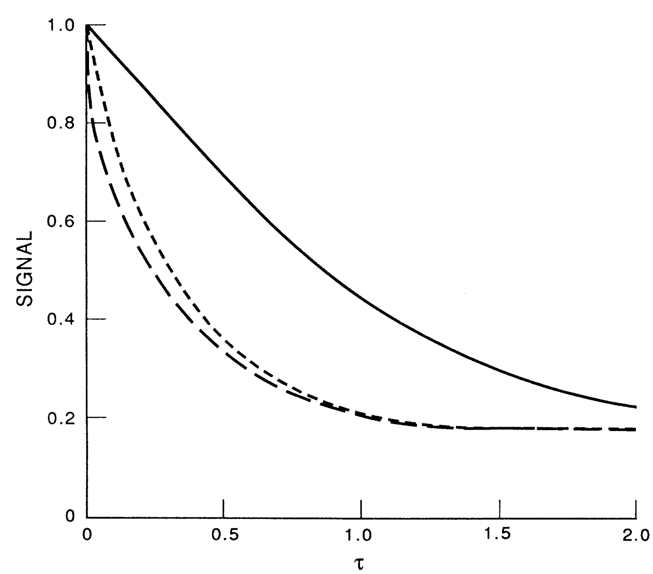

FIG. 6. FWM signal (dimensionless) vs τ (units of T_2) for different coherence times of the pump. $u=1$, $D=0.1$, and $\Gamma=100$ (long dash), $\Gamma=10$ (short dash), $\Gamma=1$ (solid).

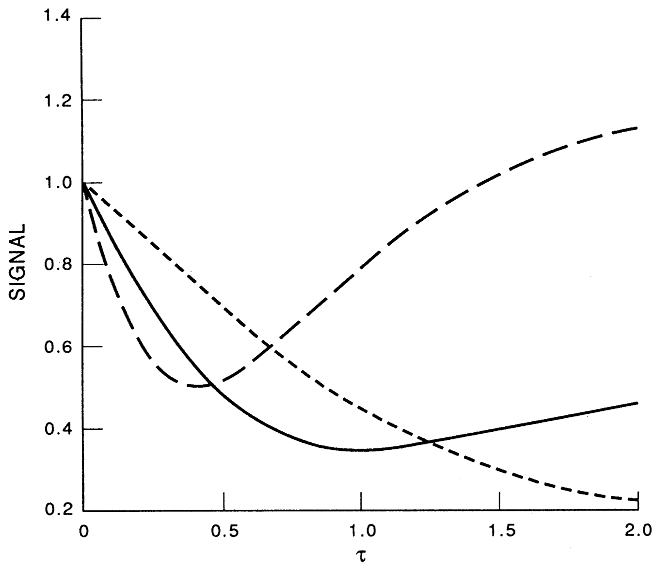

FIG. 7. FWM signal (dimensionless) vs τ (units of T_2) for $u=1$, $\Gamma=1$ and $D=0.1$ (short dash) $D=1$ (solid) and $D=10$ (long dash).

sidered the case of only positive τ values, i.e., the probe field is assumed to follow the pump field. It is, however, fairly straightforward to modify our Eqs. (2.8) and (2.12) to compute the signal for negative τ, i.e., when the pump follows the weak probe. To do this, one needs to replace $x(t)$ by $x(t-\tau)$ for all $\tau \le t \le \infty$ in Eq. (2.8) and replace $x^*(t-\tau)$ by $x^*(t)$ for all $0 \le t \le \infty$ in Eq. (2.12). After this modification the resulting six coupled Langevin

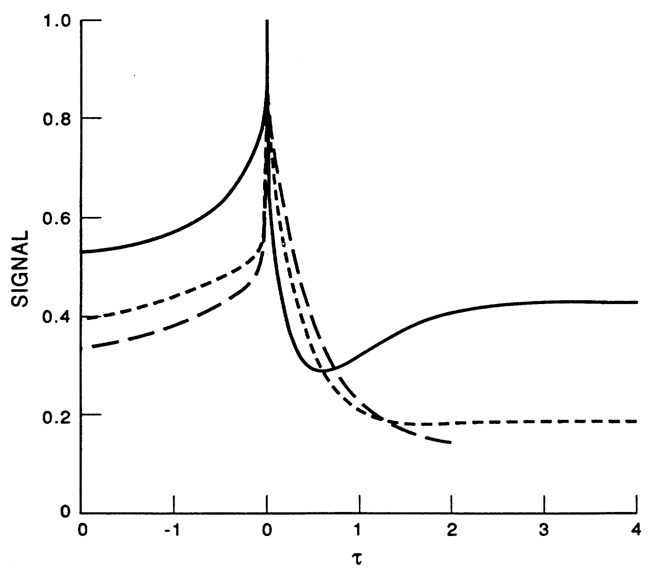

FIG. 8. FWM signal (dimensionless) vs τ (units of T_2) for both positive and negative values of τ and $u=1$, $\Gamma=100$. $D=0.01$ (long dash), $D=0.1$ (short dash), and $D=0.5$ (solid).

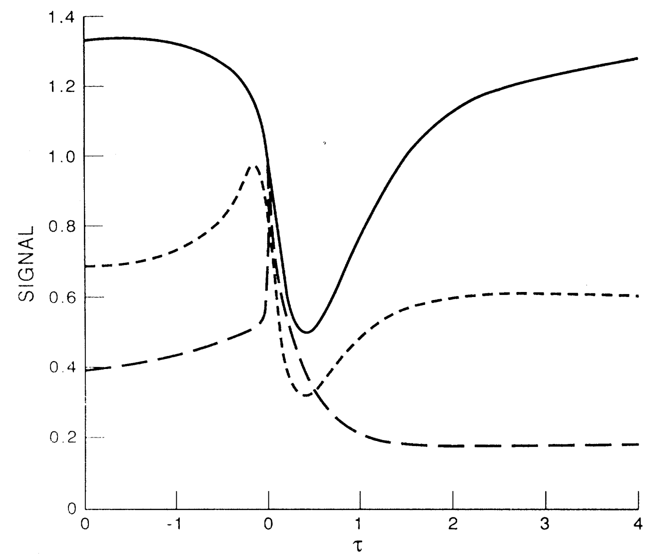

FIG. 9. FWM signal (dimensionless) vs τ (units of T_2) for $u=1$, $\Gamma D=10$ and $\Gamma=100$, $D=0.1$ (long dash), $\Gamma=10$, $D=1$ (short dash), and $\Gamma=1$, $D=10$ (solid).

equations can be solved precisely as before to obtain the behavior of the four-wave-mixing signal as a function of negative delay time τ. In Fig. 8 we show the case when the pump fluctuations are on a very fast time scale ($\Gamma=100$) and for D values of 0.01, 0.1, and 0.5. The qualitative behavior is identical to that seen for positive τ. In Fig. 9 is shown the effect of the pump intensity on the signal. The product $D\Gamma$ is kept a constant of 10 and the revival of the signal for large values of the coherence time is evident even for negative values of τ.

In conclusion, we have studied the dependence of the four-wave-mixing signal in forward geometry on the time delay between correlated pump and probe fields. For weak fields and δ-correlated fluctuations, we obtain excellent agreement between our numerical calculations and the analytic results of Morita and Yajima. Our Monte Carlo methods also investigate the regime of arbitrary pump intensities and arbitrary bandwidths of the broadband fields. For weak pump fields, we see a decay of the four-wave-mixing signal as a function of the pump-probe delay on a time scale representative of the dephasing time of the atoms. For large pump intensities, we see a revival of the signal at large delays, which can be larger than the signal at zero delay.

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APPENDIX

In this appendix we present a method for approximate estimation of the four-wave-mixing signal when the pump and probe are either fully correlated (i.e., $\tau=0$) or when the pump and probe are completely uncorrelated (i.e., $\tau\rightarrow\infty$). The approach is strictly valid when the bandwidth of the field is much smaller than the natural width of the atomic transition. In this limit, averaging over the fluctuations reduces to averaging with respect to the intensity distribution function of the chaotic field. This distribution function for a chaotic field is given by [19]

$$p(I)=1/\langle I\rangle\exp(-I\langle I\rangle)\ ,$$

where $\langle I\rangle$ is the average intensity of the fluctuating field. The solution of the optical Bloch equations (2.8) and (2.12) for a monochromatic field (i.e., $\Gamma=0$) yields the following form of the four-wave-mixing signal.

$$S=I_{\rm pump}^2 I_{\rm probe}/(1+I/I_{\rm sat})^4\ ,$$

where $I_{\rm pump}$ is the intensity of the pump, $I_{\rm probe}$ is the intensity of the probe, and $I_{\rm sat}$ is the saturation intensity. The signal for $\tau=0$ is given by

$$S=\int_0^\infty dI\, I^3 p(I)/(1+I/I_{\rm sat})^4$$

and the signal for $\tau\rightarrow\infty$ is given by

$$S=\langle I\rangle\int_0^\infty dI\, I^2 p(I)/(1+I/I_{\rm sat}^4)\ ,$$

where the pump and probe are decorrelated. As an illustration, we find that for $\Gamma=1$ and a pump intensity of 0.1 (i.e., $D=0.1$), the signal at $\tau=0$ obtained from the simulations is 1.834×10^{-3} and at large τ is 9.383×10^{-4}. The corresponding signals obtained from the above expressions are 1.858×10^{-3} and 8.110×10^{-4}. For $\Gamma=1$ and pump intensity of 1 (i.e., $D=1$), the simulations give 2.385×10^{-2} at $\tau=0$ and 1.246×10^{-2} at large τ while the Gaussian integrals above give values of 4.597×10^{-2} and 4.125×10^{-2}. The quantitative agreement between simulations and the Gaussian integrals is not expected to be good since the Gaussian integrals are strictly valid for Γ equal to zero. However, the qualitative behavior of the signal for zero delay and large delay can be estimated from these integrals and is in reasonable agreement with our simulations.

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