

Comparative study of four-wave mixing in chaotic and phase-diffusing fields

G. S. Agarwal

School of Physics, University of Hyderabad, Hyderabad 500 134, India

Gautam Vemuri, C. V. Kunasz, and J. Cooper

*Joint Institute for Laboratory Astrophysics, University of Colorado,
and National Institute of Standards and Technology, Boulder, Colorado 80309-0440*

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We consider the dependence of the four-wave-mixing signal from an ensemble of homogeneously broadened, two-level atoms on the statistics of the pump field. Two models for the pump field are considered: the chaotic field and the phase-diffusing field. A Monte Carlo simulation procedure is used to integrate the equations describing the four-wave-mixing process numerically. This technique allows consideration of arbitrary bandwidths for the pump and incorporates pump-induced saturation effects. Results are presented for the strength of the four-wave-mixing signal as a function of the pump intensity for both resonant and off-resonant cases. It is shown that the four-wave-mixing signal is sensitive to the statistics of the pump, particularly for strong fields.

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I. INTRODUCTION

Four-wave mixing in fluctuating fields continues to be the subject of extensive investigations in spite of the fact that a large body of literature on this subject already exists [1–10]. This is because of the complex nature of the underlying equations which render analytic solutions only in some simple cases. Saxena and Agarwal [1] considered the phase-conjugate geometry and studied the effects of a chaotic pump (probe being monochromatic) on the phase-conjugate reflectivities. They, however, assumed that the bandwidth of the pump, Γ , was much smaller than the atomic width, γ . Cooper and co-workers [4,5,8] studied the case of degenerate four-wave mixing (DFWM) in the regime when $\Gamma \gg \gamma$. In this limit they were able to use the decorrelation approximation and predicted several features of the four-wave-mixing signal, e.g., the increase in the pump intensity that was required to saturate the DFWM signals with increasing bandwidths. Kaczmarek and co-workers further generalized the theory to study the DFWM signals produced by pulsed fields [8,9]. Meacher *et al.* reported measurements of signals as a function of pump intensity [8]. Kaczmarek, Meacher, and Ewart also presented experimental results [9] on the temporal evolution of DFWM signals generated by broadband pulsed lasers. The theory was not able to treat the case when $\Gamma < \gamma$ and when the probe could be correlated with the pump (this could happen when the pump and probe fields are derived from a single source). However, Agarwal [6] within the framework of the third-order perturbation theory showed how one can account for pump-probe correlations as well as for arbitrary bandwidths. This theory was, however, valid for weak fields and could not incorporate pump-induced saturation effects. Correlated pump-probe fields are important in many applications, for example, in relaxation studies with time-delayed [10–18] pump and

probe fields. Recently, calculations involving time-delayed pump and probe fields have been generalized to account for saturation effects [13,14,17] under various conditions on the pump bandwidth. In particular, Tchenio and co-workers [13], using diagrammatic techniques, calculated the signals produced by time-delayed pulses, when one of the pulses was strong and the other weak. These workers included the effects of inhomogeneous broadening but neglected effects of homogeneous broadening. They assumed that the correlation time of the field was short so that the decorrelation approximation could be used. With use of the decorrelation approximation Finkelstein and Berman [14] were able to obtain analytical results for four-wave-mixing signals for the case of three broadband pulses of which two were overlapping and the third one was well separated. They also examined the signals for the case when the sample is irradiated by two broadband pulses and both pulses are strong enough to cause saturation. They could also include Doppler effect in their analysis. The works mentioned above, while dealing with strong fields, could not incorporate the condition of the Rabi frequency of the pump being larger than the bandwidth of the field. Vemuri *et al.* [17] used Monte Carlo methods to obtain coherent signals and thus could avoid the use of the decorrelation approximation, at the same time being able to study arbitrary bandwidths and pump intensities.

The effects of phase fluctuations on degenerate as well as nondegenerate four-wave mixing have been studied using third-order perturbation theory [2,3]. Such studies for correlated pump and probe fields led to the existence of fluctuation-induced resonances. It is desirable to find the signals when the pump-induced saturation is important. Moreover, it would be interesting to examine additional features of the four-wave-mixing (FWM) signals due to the non-Lorentzian nature of the pump line shapes.

From the foregoing it is clear what the open questions that need addressing are. We need to develop a theory of DFWM which accounts for saturation effects, pump-probe correlations, and arbitrary bandwidth of the pump, without resorting to a decorrelation approximation. The results are expected to be sensitive to whether we consider phase fluctuations or amplitude fluctuations.

In a series of recent papers, Zoller and co-workers [19] have demonstrated the sensitivity of atomic response to field statistics. In particular, for two statistically different field models with identical second-order correlation functions, atomic observables which are dependent on higher than second-order field correlation functions showed dramatic differences from model to model. Four-wave mixing is dependent on the sixth-order correlations of the electric field and hence should display similar sensitivity to field statistics. This serves as an additional impetus for us to undertake a comparative study of four-wave mixing in chaotic and phase-diffusing fields.

In this paper we will use Monte Carlo simulations to study the above-mentioned effects. Both chaotic and phase-diffusing fields with Lorentzian spectra are considered and for the phase-diffusion case the spectrum can also be non-Lorentzian. We consider the forward geometry for FWM and restrict our study to stationary two-level atoms and hence neglect Doppler effects.

II. BASIC EQUATIONS AND MONTE CARLO SIMULATIONS

We consider the interaction of an ensemble of two-level atoms of frequency ω_0 with an electromagnetic field of frequency ω_1 . The electromagnetic field consists of two parts: (i) pump propagating in the direction \mathbf{k}_1 and (ii) probe in the direction \mathbf{k}_p . The total field can be written in the form

$$\begin{aligned} \mathbf{E}(r, t) = & \boldsymbol{\varepsilon}_1(t) e^{-i\phi_1(t)} e^{i\mathbf{k}_1 \cdot \mathbf{r} - i\omega_1 t} \\ & + \boldsymbol{\varepsilon}_p(t) e^{i\phi_p(t)} e^{i\mathbf{k}_p \cdot \mathbf{r} - i\omega_1 t} + \text{c.c.} \end{aligned} \quad (2.1)$$

The amplitudes and phases are in general fluctuating quantities. We will consider the following models for fluctuating fields.

A. Chaotic fields

Here the complex amplitude $\boldsymbol{\varepsilon}_1 e^{-i\phi_1} = \tilde{\boldsymbol{\varepsilon}}_1$ is a Gaussian random process with the properties

$$\begin{aligned} \langle \tilde{\boldsymbol{\varepsilon}}_1(t) \rangle &= 0, \\ \langle \tilde{\boldsymbol{\varepsilon}}_1(t) \tilde{\boldsymbol{\varepsilon}}_1^*(t') \rangle &= D\Gamma e^{-\Gamma|t-t'|}, \\ \langle \tilde{\boldsymbol{\varepsilon}}_1(t) \tilde{\boldsymbol{\varepsilon}}_1(t') \rangle &= 0 = \langle \tilde{\boldsymbol{\varepsilon}}_1^*(t) \tilde{\boldsymbol{\varepsilon}}_1^*(t') \rangle, \end{aligned} \quad (2.2)$$

where D is the strength of the fluctuations and $1/\Gamma$ is the time scale of the fluctuations. The product $D\Gamma$ is the variance of the Gaussian random process and is identified with the intensity of the field. The chaotic field model as described by (2.2) has a Lorentzian spectral profile with a full width at half maximum (FWHM) of 2Γ and is an accurate representation of the field from a thermal source

or a multimode laser. The probe field $\boldsymbol{\varepsilon}_p(t) e^{-i\phi_p(t)} = \tilde{\boldsymbol{\varepsilon}}_p(t)$ can either be monochromatic or it can be derived from the pump laser itself, as in Ref. [17]. In the latter case it is fully correlated with the pump and can be written as

$$\tilde{\boldsymbol{\varepsilon}}_p(t) = g\tilde{\boldsymbol{\varepsilon}}_1(t) \quad (2.3)$$

where $g \ll 1$. In what follows, the probe will be treated perturbatively while the pump will be treated to all orders and thus pump-induced saturation effects are accounted for.

B. Phase-diffusing fields

We now assume that there are no amplitude fluctuations but the phase ϕ_1 fluctuates such that $d\phi_1/dt = \mu$ is a Gaussian random process with the properties

$$\langle \mu(t) \rangle = 0 \quad \text{and} \quad \langle \mu(t)\mu(t') \rangle = b\beta e^{-\beta|t-t'|}, \quad (2.4)$$

where b is the strength of the frequency fluctuations and $1/\beta$ is the time scale of the frequency fluctuations. For values of $\beta \gg b$, the field line shape is a Lorentzian with a FWHM of $2b$, while for $\beta \ll b$ it is a Gaussian with a FWHM related to $(b\beta)^{1/2}$. The probe is taken to be either monochromatic or fully correlated with the pump phase. In the correlated case we set $\phi_p(t) = \phi_1(t)$.

In the frame rotating with the frequency of the pump, ω_1 , the Bloch equations for the atomic polarization $\psi_1 = \rho_{21}$, $\psi_2 = \rho_{12}$, and the inversion $\psi_3 = \frac{1}{2}(\rho_{11} - \rho_{22})$ can be written as

$$\frac{d\tilde{\boldsymbol{\psi}}}{dt} = \mathbf{M}\tilde{\boldsymbol{\psi}} + \mathbf{I}, \quad (2.5)$$

where

$$\mathbf{M} = \begin{pmatrix} -1/T_2 + i\Delta & 0 & -2ix^*(t) \\ 0 & -1/T_2 - i\Delta & 2ix(t) \\ -ix(t) & ix^*(t) & -1/T_1 \end{pmatrix} \quad (2.6)$$

and

$$\begin{aligned} \Delta &= \omega_0 - \omega_1, \quad \tilde{\boldsymbol{\psi}}_1 = e^{-i\omega_1 t + i\mathbf{k}_1 \cdot \mathbf{r}} \boldsymbol{\psi}_1, \quad \tilde{\boldsymbol{\psi}}_3 = \boldsymbol{\psi}_3, \\ I_1 &= I_2 = 0, \quad I_3 = -1/2T_1, \\ x(t) &= (\mathbf{d} \cdot \boldsymbol{\varepsilon} / \hbar) [\boldsymbol{\varepsilon}_1(t) + e^{i(\mathbf{k}_p - \mathbf{k}_1) \cdot \mathbf{r}} \boldsymbol{\varepsilon}_p(t)]. \end{aligned} \quad (2.7)$$

As mentioned earlier, the probe is weak and so we expand the Bloch vector $\tilde{\boldsymbol{\psi}}$ in powers of the probe field

$$\tilde{\boldsymbol{\psi}} = \tilde{\boldsymbol{\psi}}^{(0)} + \tilde{\boldsymbol{\psi}}^{(1)} + \dots, \quad (2.8)$$

where

$$\frac{d\tilde{\boldsymbol{\psi}}^{(0)}}{dt} = \mathbf{M}^{(0)}\tilde{\boldsymbol{\psi}}^{(0)} + \mathbf{I}, \quad (2.9)$$

$$\frac{d\tilde{\boldsymbol{\psi}}^{(1)}}{dt} = \mathbf{M}^{(0)}\tilde{\boldsymbol{\psi}}^{(0)} + \mathbf{M}^{(1)}\tilde{\boldsymbol{\psi}}^{(0)}, \quad (2.10)$$

and where $\mathbf{M}^{(0)}$ is obtained from (2.6) by setting $\tilde{\boldsymbol{\varepsilon}}_p = 0$ in $x(t)$ [Eq. (2.7)]. The matrix $\mathbf{M}^{(1)}$ is obtained from (2.6) by letting $1/T_2$, Δ , and $\tilde{\boldsymbol{\varepsilon}}_1 \rightarrow 0$. The solution for $\tilde{\boldsymbol{\psi}}^{(1)}$ can be written in the form

$$\tilde{\psi}^{(1)} = \exp[i(\mathbf{k}_p - \mathbf{k}_1) \cdot \mathbf{r}] A + \exp[-i(\mathbf{k}_p - \mathbf{k}_1) \cdot \mathbf{r}] F, \quad (2.11)$$

where F is the solution of

$$dF/dt = M^{(0)}F + ig(\mathbf{d} \cdot \boldsymbol{\varepsilon} / \hbar) \boldsymbol{\varepsilon}_p^*(t) \begin{pmatrix} -2\varphi_3^{(0)}(t) \\ 0 \\ \varphi_2^{(0)}(t) \end{pmatrix}. \quad (2.12)$$

Here $\tilde{\psi}^{(0)}(t)$ is given by (2.9). Note that both (2.12) and (2.9) are Langevin equations as the fields $\tilde{\varepsilon}_1$ and $\tilde{\varepsilon}_p$ are fluctuating. The steady-state four-wave-mixing signal is obtained from

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

where $\langle \rangle$ denotes stochastic averaging over the fluctuations of the pump and the probe. The stochastic averaging is done for different models of pump and probe fluctuations by Monte Carlo techniques. We adopt the Monte Carlo technique in the form developed by Fox *et al.* [20] and as used by Vemuri *et al.* [17] for calculating signals involving time-delayed pump-probe fields. Using this technique we can calculate the FWM signals for a very wide range of pump intensities and bandwidths.

III. NUMERICAL RESULTS

In this section we present the results obtained by Monte Carlo simulation methods.

A. Chaotic fields

We consider the case when the probe is monochromatic. We assume that both the pump and probe are on resonance with atomic transition. All frequencies are measured in units of the atomic linewidths $1/T_2$. We also assume radiative relaxation, i.e., $T_2/T_1=2$. Figure 1 shows the variation of the signal with the pump intensity $D\Gamma$ [Eq. (2.2)] for different values of the pump bandwidth. The case of $\Gamma=100$ is the extreme broadband case and can be compared with the work of Cooper *et al.* [5]. [It should be noted that Ref. [5] deals with FWM signals with counterpropagating pump fields in a medium of finite length (like a vapor cell) and hence effects due to a standing wave in the medium are important, while our work assumes a forward geometry and a thin medium

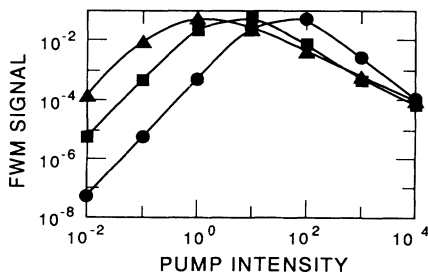


FIG. 1. FWM signal vs pump intensity for chaotic fields. Circles are for field bandwidth, $\Gamma=100$; squares for $\Gamma=10$; and triangles for $\Gamma=1$.

(like an atomic beam) and hence we neglect any effects due to a standing wave.) For this case, we find that below saturation the FWM signal increases as I_p^2 (I_p is pump intensity). This is precisely the behavior predicted in Ref. [5], which further predicted for very high intensities a decrease as I_p^{-2} (which is modified to $I_p^{-3/2}$ when standing-wave effects are included). For pump intensities higher than the saturation intensity, it is difficult to estimate a power law from our data. To get an estimate of the power law here one has to calculate the signal for higher values of I_p , but the CPU time for these was prohibitive. Figure 1 also clearly demonstrates the increase in the pump intensity needed to reach saturation with increasing bandwidths. Once again, this result is consistent with that of Ref. [5], which predicts below the saturation intensity an increase proportional to the square of the bandwidth. On comparing the signal for pump intensities of 10^{-2} and 10^{-1} for Γ of 100 and 10, we find similar qualitative behavior.

Figure 1 also shows that the saturation curve is much broader for pump bandwidths of the order of the atomic transition width. This is the regime where the decorrelation approximation is no longer valid and hence we expect different behavior. While the signal does increase as I_p^2 for I_p less than the saturation intensity, the behavior of the signal for pump intensities greater than the saturation intensity is drastically different from that predicted by the decorrelation theory. This is evident from the different slopes for the curves and bandwidths of 100 and 1 for intensities larger than the saturation intensity.

Figure 2 gives the saturation curves for the case of $\Gamma T_2=1$ but for different values of the detuning parameter Δ . Larger Δ requires higher values of the pump intensity to yield maximum signal. This is to be expected, since it is much harder to saturate the transition with off-resonant excitation. Figure 3 shows a scan of the signal as a function of Δ , for small pump bandwidth and large pump intensity. We find that signal first decreases with Δ , but then shows a revival before decreasing again. It should be borne in mind that the line shape of the pump remains Lorentzian for all values of Γ , a property of the chaotic field.

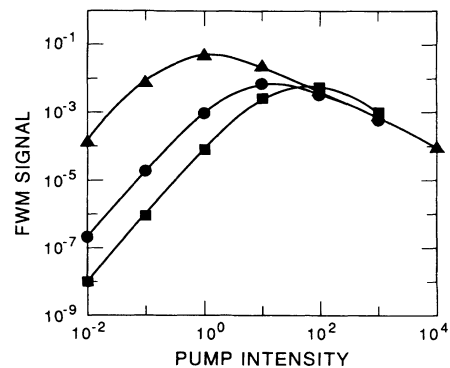


FIG. 2. FWM signal vs pump intensity for chaotic fields and fixed detuning, Δ of pump from atomic transition frequency. $\Gamma=1$ and the triangles are for $\Delta=0$, circles for $\Delta=5$, and squares for $\Delta=10$.

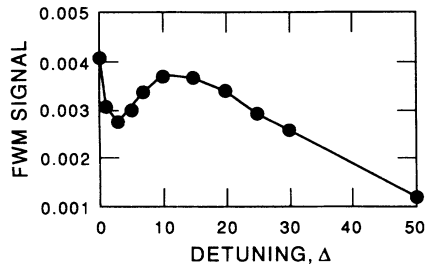


FIG. 3. FWM signal vs pump detuning, Δ for chaotic fields with $\Gamma=1$ and $D=100$.

B. Phase-diffusing fields

We first consider the case when the probe is monochromatic and hence is not correlated with the pump. Figure 4 shows the behavior of the FWM signal as a function of the pump intensity. Clearly the saturation behavior depends on the strength of the frequency fluctuations. For $b=1$ and $\beta=10$, the pump line shape is a Lorentzian to a good approximation. As mentioned earlier, it is a characteristic of the phase-diffusion model that for $\beta \gg b$ the line shape is a Lorentzian while for $\beta \ll b$ it is Gaussian, with intermediate values of b and β giving Voigt profiles. In Fig. 4, the laser bandwidth is moderate compared to the atomic linewidth. Once again we find that with an increase in the pump bandwidth there is an increase in the saturation intensity. We find, however, that the behavior of the FWM signal is not very sensitive to the β parameter for fixed value of b (not shown).

While the decorrelation approximation as used in Ref. [5] is not valid for moderate to small bandwidths in the case of the chaotic fields, it is valid for arbitrary bandwidths in the case of phase-diffusing fields, provided β is much larger than any other rate in the problem. Hence we expect the results of Ref. [5] to apply to our results here if this condition is satisfied. In fact, we find that for $\beta=10$ and b of 1 and 3, the FWM signal increases as the square of the pump intensity for intensities below the saturation intensity. In Fig. 4, the decorrelation approximation can be used to obtain the curves with $\beta=10$, $b=1$ and $\beta=10$, $b=3$.

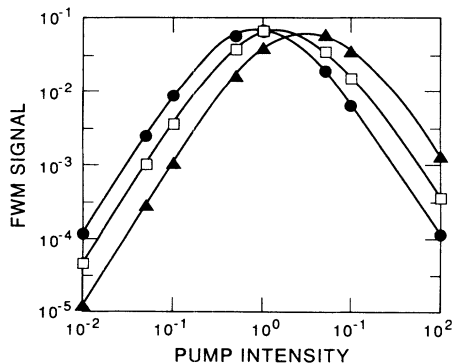


FIG. 4. FWM signal vs pump intensity for phase diffusing fields. For all three curves $\beta=10$ and circles are for $b=1$, squares for $b=3$, and triangles for $b=10$.

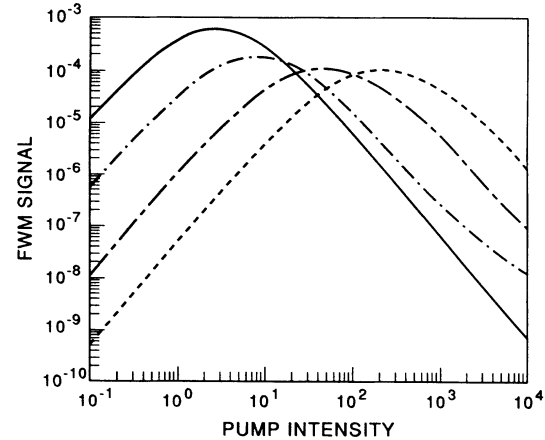


FIG. 5. FWM signal vs pump intensity as obtained from the analytic work of Ref. 2. The solid curve is for $b=0$, the dash-dotted curve for $b=10$, the dashed curve for $b=100$, and the dotted curve for $b=500$. Pump and probe are on resonance with atomic transition.

For the pure phase-diffusion model, i.e., when $\beta \rightarrow \infty$ and the line shapes become Lorentzian, it is more convenient to use the theory of multiplicative processes to obtain the DFWM signals. This theory is discussed at length in Ref. [2]. The theory enables one to obtain deterministic equations for single- and two-atom averages. The two-level results can be obtained as a special case of the three-level system. The off-resonant signal is calculated in Ref. [2], i.e., the case when both the pump and probe are far off resonance, whereas in what follows we look at resonant signals. The saturation behavior of the signals obtained in this manner is shown in Figs. 5 and 6 for different bandwidths of the pump. Figure 5 shows the signals when the pump is on resonance with the atomic transition, while Fig. 6 is the case when $\Delta=20$. Note that the parameter b now gives the linewidth of the pump.

It is usually difficult to quantitatively compare the sig-

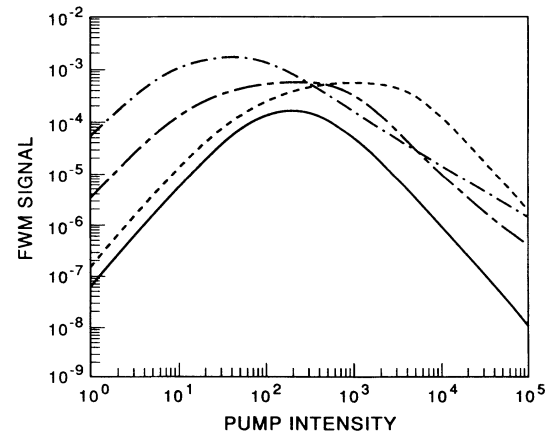


FIG. 6. FWM signal vs pump intensity as obtained from Ref. [5] for pump detuning, $\Delta=20$. The solid curve is for $b=0$, the dash-dotted curve for $b=10$, the dashed curve for $b=100$, and the dotted curve for $b=500$.

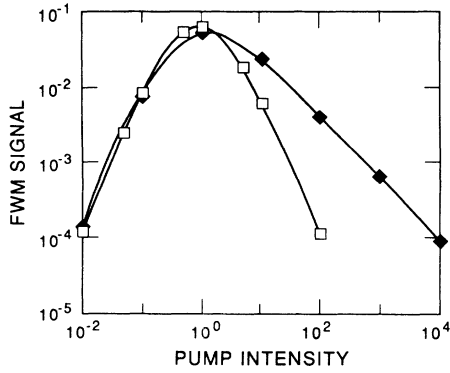


FIG. 7. Comparison of FWM signal vs pump intensity for chaotic and phase diffusing fields with identical bandwidths and band shapes. For the phase-diffusing field, $\beta=10$ and $b=1$ and for the chaotic field $\Gamma=1$.

nals obtained from chaotic fields and phase-diffusing fields except for some special parameter values. The chaotic field has two independent parameters, the bandwidth Γ and the strength of the noise D with the product ΓD specifying the pump intensity. The line shape is always Lorentzian for all values of Γ and D . The phase-diffusing field on the other hand has three independent parameters, the spectral density of the frequency fluctuations b , the time scale of fluctuations, $1/\beta$, and the Rabi frequency of the field, Ω (square root of the intensity). Here the line shapes can be Lorentzian, Gaussian, or intermediate between the two, depending on the relative values of b and β . The only situation in which a fair comparison of the saturation behavior for the two models can be made is when $\beta \gg b$ for the frequency fluctuations. In Fig. 7, we show a comparison of the FWM signals as a function of the pump intensity for the two models, when the laser line shape is Lorentzian for both models and is of the order of the atomic linewidth. Thus for chaotic field we choose $\Gamma=1$ and for the phase-diffusing field we choose $b=1$ and $\beta=10$. We find that the differences in

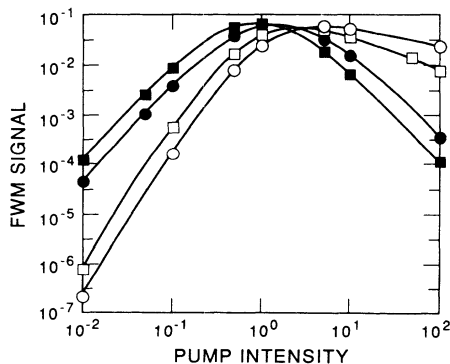


FIG. 8. Comparison of FWM signal vs pump intensity for monochromatic and nonmonochromatic probe. The squares are for $\beta=10$ and $b=1$ while the circles are for $\beta=10$ and $b=3$. The solid markers are for monochromatic probe while the open markers are for the probe correlated with the pump.

the signals are more pronounced at higher pump intensities.

Finally, we consider the effect of cross correlations between the phase fluctuations of the pump and probe fields. Figure 8 shows a comparison of the saturation behavior for correlated and uncorrelated pump and probe fields for two different field bandwidths. The noisy probe is less effective at low pump intensities but becomes more effective at higher pump intensities. Further, the increase in the FWM signal with pump intensity for intensities below the saturation intensity scales faster than a square law while the decrease in the signal above the saturation intensity scales slower than the corresponding monochromatic probe signal.

IV. DISCUSSION

We have studied the effect of the pump field statistics on the saturation behavior of four-wave-mixing signals produced by an ensemble of homogeneously broadened two-level atoms. A comparison of the signal generated by chaotic fields with the signals generated by phase-diffusing fields is presented. We also present results for the chaotic fields with bandwidths comparable to the atomic linewidths, a regime where the decorrelation approximation is not valid. These results, as might be expected, are significantly different from those obtained for broadband fields (as in Ref. [5]).

For the phase-diffusing fields, our results display the need for increasing pump intensities to achieve saturation of the FWM signal as the pump bandwidth increases. The results are in agreement with the decorrelation theory of Ref. [5] since this approximation is valid for phase-diffusing fields. A direct comparison of the signals produced by an ensemble of two-level atoms interacting with chaotic or phase-diffusing fields is also presented. The saturation behavior is dramatically different for the two field models, even though they have identical bandwidths and band shapes and hence identical second-order field correlation functions. This result is not surprising in view of the recent works of Zoller and co-workers [19] demonstrating that atomic observables which depend on higher-order field correlations can be very sensitive to differences in the field statistics. Since FWM depends on sixth-order field correlations, it falls into that category.

The Monte Carlo methods used in this work also allow us to incorporate the effects of correlated pump and probe fields with arbitrary bandwidths. As an illustration we have presented the saturation behavior of the FWM signal when the pump and probe have correlated phase fluctuations. We find the saturation behavior for correlated pump and probe is quite different from the behavior with a monochromatic probe. In the context of phase fluctuations, our numerical methods are complementary to the analytic methods developed by Gheri, Marte, and Zoller [18] where they study the case of $\beta \rightarrow \infty$. Further, our numerical methods can be easily applied to study correlated pump and probe fields for the chaotic field model, as shown in Ref. [17].

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