Nonlinear spectroscopy with cross-correlated chaotic fields

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A general theory of four-wave mixing in correlated pump and probe fields is developed. The form of the signals, when the probe is delayed, is also given. The theory is applicable to both degenerate and nondegenerate four-wave mixing. The cross correlation between pump and probe leads to an extra contribution to the signal. This extra contribution is easily isolated in an experiment where the probe is delayed with respect to the pump and carries useful information about the physical parameters of the system. Various applications of the theory are discussed. The theory is shown to lead to the existence of fluctuation-induced resonances in a natural way.

I. INTRODUCTION

It has been recently recognized that the incoherence and, in general, the partial coherence of the pump and probe fields can lead to important spectroscopic applications. For example, Yajima and co-workers[1] have emphasized that extremely fast dephasing phenomena can be studied by doing four-wave mixing with incoherent light. Steel and Rand[2] have demonstrated the existence of ultranarrow resonances in four-wave mixing in ruby. These resonances are possible only if the pump and probe are completely correlated in phase; otherwise, the independent fluctuations of the pump and probe would worsen the resolution.[3] Other important applications are possible only if the input fields are highly stabilized or various pump and probe fields are correlated. It has also been demonstrated that the fluctuations of the pump and probe can lead to new resonances[4–7] in nonlinear mixing. The cross correlation of the pump and probe is quite significant in such studies. It is obvious that one needs a theory of nonlinear-spectroscopic experiments involving fluctuating pump and probe fields. Moreover, one should formulate the theory in such a way that any cross correlation between the pump and probe is fully accounted for. The theory will depend on the statistical nature of the input fields. It may be noted that in most nonlinear-spectroscopic experiments one uses a Nd-YAG (yttrium aluminum garnet) laser for the pump field. Such a laser's output has many modes and it can be modeled by a chaotic field. Thus it necessitates a theory of spectroscopic experiments with chaotic pump and probe fields. Considerable work[8–13] has already been done on the coherent anti-Stokes Raman scattering signals in *uncorrelated* (i.e., *statistically independent*) pump and probe fields modeled by a Gaussian stochastic process. It has also been possible to account for the delay between two pump beams.[12] In view of the importance of the cross correlation here we address ourselves to the more general question of how to describe nonlinear mixing experiments if the pump and probe fields are modeled by correlated Gaussian random processes. In this work we ignore the saturation effects and consider only the steady-state nonlinear spectroscopic phenomena.

The organization of the paper is as follows. In Sec. II we present the general result for four-wave mixing signals in terms of convolution integrals. The additional terms due to the cross correlation arise in the signal. In Sec. III we present the form of the signal for the case when the pump and probe are delayed with respect to each other. Spectroscopic consequences for the case of broadband pump and probe are discussed. In Sec. IV we discuss the modifications in the formulas of Sec. II for the case of nondegenerate four-wave mixing. We present in Sec. V applications of formulas to fluctuation-induced extra resonances in four-wave mixing which have been previously studied[4,7] for the phase-diffusion model of the input fields.

II. THEORY OF THIRD-ORDER NONLINEAR-OPTICAL PHENOMENA IN CORRELATED PUMP AND PROBE FIELDS

In this section we derive the expressions for the four-wave-mixing signals produced by a system interacting with cross-correlated pump and probe fields. Consider the input fields represented by their Fourier transforms $\varepsilon(\omega)$,

$$E(t)=\int \varepsilon(\omega)e^{-i\omega t}\frac{d\omega}{2\pi} . \tag{2.1}$$

Then the induced polarization can be expressed as

$$P(t)=\int P(\omega)e^{-i\omega t}\frac{d\omega}{2\pi} ,$$

$$P(\omega)=\int\int\int \chi^{(3)}(\omega_1,\omega_2,\omega_3)\varepsilon(\omega_1)\varepsilon(\omega_2)\varepsilon(\omega_3)\times\delta(\omega-\omega_1-\omega_2-\omega_3)d\omega_1 d\omega_2 d\omega_3 . \tag{2.2}$$

In writing (2.2) we ignore for simplicity all vectorial indices. Note that if the fields are written as the sum of pump $\varepsilon^{(l)}$ and probe $\varepsilon^{(s)}$,

$$\varepsilon=\varepsilon^{(l)}+\varepsilon^{(s)} , \tag{2.3}$$

then the four-wave-mixing signal is obtained from

$$P(\omega)=3\int d\{\omega\}\chi^{(3)}(\omega_1,\omega_2,-\omega_3)\varepsilon^{(l)}(\omega_1)\varepsilon^{(l)}(\omega_2)\times[\varepsilon^{(s)}(\omega_3)]^*\delta(\omega_1+\omega_2-\omega_3-\omega)\ . \quad (2.4)$$

We should bear in mind that $\chi^{(3)}$ in (2.4) has the usual permutation symmetry. For fluctuating fields the ε's are stochastic functions of the frequency and therefore $P(\omega)$ is a stochastic quantity. The spectrum of the generated signal can be obtained from the spectrum of the polarization fluctuations. The spectrum $S(\omega)$ of the polarization fluctuations, defined by

$$\langle P(\omega)P^*(\omega')\rangle=\delta(\omega-\omega')S(\omega)\ , \quad (2.5)$$

can be related to the sixth-order correlation function of the electric fields,

$$S(\omega)=9\int d(\omega)d(\omega')\delta(\omega-\omega_1-\omega_2+\omega_3)\delta(\omega-\omega_1'-\omega_2'+\omega_3')\chi^{(3)}(\omega_1,\omega_2,-\omega_3)\times[\chi^{(3)}(\omega_1',\omega_2',-\omega_3')]^*\Gamma(\omega_1',\omega_2',\omega_3,\omega_1,\omega_2)\ . \quad (2.6)$$

Here the sixth-order correlation function Γ of the fields is defined by

$$\langle\varepsilon^{(l)}(\omega_1)\varepsilon^{(l)}(\omega_2)[\varepsilon^{(s)}(\omega_3)]^*[\varepsilon^{(l)}(\omega_1')]^*[\varepsilon^{(l)}(\omega_2')]^*\varepsilon^{(s)}(\omega_3')\rangle=\delta(\omega_1+\omega_2+\omega_3'-\omega_3-\omega_1'-\omega_2')\Gamma(\omega_1',\omega_2',\omega_3,\omega_1,\omega_2)\ . \quad (2.7)$$

The relation (2.7) is the analog of the Wiener-Khintchin theorem for higher-order correlations and depends on the stationarity of the field. If the input fields are chaotic then the sixth-order correlation function Γ can be expressed in terms of the second-order correlation function of the field defined by

$$\langle\varepsilon^{(\alpha)}(\omega)[\varepsilon^{(\alpha)}(\omega')]^*\rangle=I^{(\alpha)}(\omega)\delta(\omega-\omega'),\quad \alpha=l,s \quad (2.8)$$

and the cross correlation $C(\omega)$,

$$\langle\varepsilon^{(l)}(\omega)[\varepsilon^{(s)}(\omega')]^*\rangle=\delta(\omega-\omega')C(\omega)\ . \quad (2.9)$$

In writing (2.9) we have assumed that the fields $\varepsilon^{(l)}$ and $\varepsilon^{(s)}$ have the same optical frequency. A careful application of the moment theorem for Gaussian random processes enables us to obtain Γ in terms of $I^{(\alpha)}(\omega)$ and $C(\omega)$,

$$\begin{aligned}\Gamma(\omega_1',\omega_2',\omega_3',\omega_1,\omega_2)=&\ \delta(\omega_2-\omega_2')\delta(\omega_3-\omega_3')I^{(l)}(\omega_1)I^{(l)}(\omega_2)I^{(s)}(\omega_3')+\delta(\omega_2-\omega_1')\delta(\omega_3'-\omega_3)I^{(l)}(\omega_1)I^{(l)}(\omega_2)I^{(s)}(\omega_3')\\&+\delta(\omega_2-\omega_3)\delta(\omega_3'-\omega_2')I^{(l)}(\omega_1)C(\omega_2)C^*(\omega_2')+\delta(\omega_2-\omega_3)\delta(\omega_1'-\omega_3')I^{(l)}(\omega_1)C(\omega_2)C^*(\omega_1')\\&+\delta(\omega_2-\omega_1')\delta(\omega_2'-\omega_3')C(\omega_1)I^{(l)}(\omega_2)C^*(\omega_2')+\delta(\omega_2-\omega_2')\delta(\omega_1'-\omega_3')I^{(l)}(\omega_2)C(\omega_1)C^*(\omega_1')\ . \quad (2.10)\end{aligned}$$

Note that the last four terms in (2.10) arise due to the cross correlation between the pump and probe. Upon substituting (2.10) in (2.6) and making use of the permutation symmetry of $\chi^{(3)}$, we get our key result for $S(\omega)$,

$$\begin{aligned}S(\omega)=&\ 18\int d\{\omega\}I^{(l)}(\omega_1)I^{(l)}(\omega_2)I^{(s)}(\omega_3)\delta(\omega-\omega_1-\omega_2+\omega_3)\,|\chi^{(3)}(\omega_1,\omega_2,-\omega_3)|^2\\&+36\int d\omega_1\delta(\omega-\omega_1)\left|\int d\omega_2C(\omega_2)\chi^{(3)}(\omega_1,\omega_2,-\omega_2)\right|^2I^{(l)}(\omega_1)\ . \quad (2.11)\end{aligned}$$

If there is no cross correlation between the pump and probe, then (2.11) reduces to

$$S(\omega)=18\int d(\omega)I^{(l)}(\omega_1)I^{(l)}(\omega_2)I^{(s)}(\omega_3)\times\delta(\omega-\omega_1-\omega_2+\omega_3)\times|\chi^{(3)}(\omega_1,\omega_2,-\omega_3)|^2\ . \quad (2.12)$$

The result (2.12) has been used extensively in the past[8-13] in connection with coherent anti-Stokes Raman spectroscopy (CARS) in fluctuating pump fields. It has also been used to examine the existence of various types of fluctuation-induced resonances[6] in nonlinear mixing. In what follows we concentrate on the second term (2.11) which arises from the cross correlation between the pump and probe. Let us denote this contribution by $S^{(c)}(\omega)$,

$$S^{(c)}(\omega)=36\int d\omega_1\delta(\omega-\omega_1)I^{(l)}(\omega_1)\times\left|\int d\omega_2C(\omega_2)\chi^{(3)}(\omega_1,\omega_2,-\omega_2)\right|^2\ . \quad (2.13)$$

It is important to note that the terms dependent on the cross correlation involve the susceptibility $\chi^{(3)}(\omega_1,\omega_2,-\omega_2)$, which is actually responsible for two-photon processes like Raman processes. Thus the four-wave mixing acquires a contribution from the integrated sum of the Raman-like susceptibilities. We will see that the fluctuation-induced resonances in four-wave mixing, i.e., in radiation at frequency $2\omega_l-\omega_s$, actually originate from such Raman-like susceptibilities. The fluctuation-induced resonances of Ref. 6 occur in a different frequency range and do not require the presence of the cross correlation. Finally, the vectorial properties, which, for instance are, important in problems involving magnetic degeneracies, Zeeman coherence, etc., can be restored by inspection; for example, (2.13) is to be replaced by

$$S_{\mu\nu}^{(c)}(\omega)=36\int d\omega_1\delta(\omega-\omega_1)I_{\alpha\alpha'}^{(l)}(\omega_1)\times\int d\omega_2\chi_{\mu\alpha\beta\gamma}^{(3)}(\omega_1,\omega_2,-\omega_2)C_{\beta\gamma}(\omega_2)\times\int d\omega_3\chi_{\nu\alpha'\beta'\gamma'}^{(3)}(\omega_1,\omega_3,-\omega_3)C_{\beta'\gamma'}^*(\omega_3)\ , \quad (2.14)$$

where quantities like $I^{(l)}_{\alpha\alpha'}$ are defined by

$$\langle \varepsilon^{(l)}_{\alpha}(\omega)[\varepsilon^{(l)}_{\alpha'}(\omega')]^* \rangle = \delta(\omega-\omega')I^{(l)}_{\alpha\alpha'}(\omega) \; . \quad (2.15)$$

We conclude this section by examining the form of (2.13) for the case of an isolated Raman resonance whence $\chi^{(3)}$ has the form[14]

$$\chi^{(3)}(\omega,\omega_2,-\omega_2) = A + \frac{B}{(\omega_2-\omega-\omega_R+i\Gamma)} \; . \quad (2.16)$$

The integral in (2.13) can be evaluated explicitly for the case of a Lorentzian spectral profile

$$I(\omega)=C(\omega)=\frac{\gamma_c/\pi}{\gamma_c^2+(\omega-\omega_l)^2}I \; , \quad (2.17)$$

with the result

$$S^{(c)}(\omega)=\frac{\gamma_c/\pi}{\gamma_c^2+(\omega-\omega_l)^2}I^3 \times \left| A + \frac{B}{\omega_l-\omega-\omega_R+i(\Gamma+\gamma_c)} \right|^2 . \quad (2.18)$$

The contribution of the cross correlation to the total signal $S^{(c)}=\int d\omega\, S^{(c)}(\omega)$ is equal to

$$S^{(c)}I^3 = |A|^2 + \frac{|B|^2}{\omega_R^2+\Gamma'^2}\left[1+\frac{\gamma_c}{\Gamma+\gamma_c}\right] + 2\,\mathrm{Re}\left[\frac{A^*B}{-\omega_R+i\Gamma'}\right], \quad \Gamma'=\Gamma+2\gamma_c \; . \quad (2.19)$$

Thus the fluctuations not only change the linewidth from Γ to $\Gamma'=\Gamma+2\gamma_c$ but there is a genuine fluctuation-induced contribution in the resonant term coming from the factor $\gamma_c/(\Gamma+\gamma_c)$ in the parentheses.

III. NONLINEAR MIXING WITH PUMP AND PROBE DELAY

In this section we show how the basic formulation of Sec. II can be generalized to include the delay between the pump and probe fields. It has been demonstrated by several authors that the four-wave-mixing signal as a function of delay can yield various dephasing times, particularly if the pump and probe fields are derived from the same fluctuating source with negligibly small correlation time. Note that if the fields were purely monochromatic, then the four-wave-mixing signal in the long-time limit would be independent of the delay between the pump and probe. This is because the steady state of a system is generally independent of the initial state or the initial preparation. The system can "remember" its initial preparation due to a finite correlation between pump and probe. The treatment given here also forms the basis of the incoherent spectroscopy.[1]

For simplicity we only treat the case of degenerate four-wave mixing. The field ε in (2.3) is to be replaced by

$$\varepsilon(t)=\varepsilon^{(l)}(t)+\varepsilon^{(s)}(t-\delta) \; , \quad (3.1)$$

where δ is the delay between the pump and probe. The Fourier transform of $\varepsilon(t)$ will be

$$\varepsilon(\omega)=\varepsilon^{(l)}(\omega)+\varepsilon^{(s)}(\omega)e^{i\omega\delta}, \quad \delta>0 \; , \quad (3.2)$$

which implies that Eq. (2.4) is to be modified by replacing $\varepsilon^{(s)}_{(\omega)}$ by $\varepsilon^{(s)}(\omega)e^{i\omega\delta}$. Thus, in place of (2.6), the spectrum of the polarization fluctuations will now be

$$S(\omega)=9\int d(\omega)d(\omega')\delta(\omega-\omega_1-\omega_2+\omega_3)\delta(\omega-\omega_1'-\omega_2'+\omega_3')\chi^{(3)}(\omega_1,\omega_2,-\omega_3) \times[\chi^{(3)}(\omega_1',\omega_2',-\omega_3')]^* e^{-i\omega_3\delta+i\omega_3'\delta}\Gamma(\omega_1',\omega_2',\omega_3,\omega_1,\omega_2) \; . \quad (3.3)$$

This can be further simplified for chaotic fields by using Eq. (2.10) for the sixth-order correlation function of the input field. The result of such a calculation is

$$S(\omega)=18\int d\{\omega\}I^{(l)}(\omega_1)I^{(l)}(\omega_2)I^{(s)}(\omega_3)\delta(\omega-\omega_1-\omega_2+\omega_3)\,|\chi^{(3)}(\omega_1,\omega_2,-\omega_3)|^2 + 36\int d\omega_1\delta(\omega-\omega_1)I^{(l)}(\omega_1)\left|\int d\omega_2 C(\omega_2)\chi^{(3)}(\omega_1,\omega_2,-\omega_2)e^{-i\omega_2\delta}\right|^2 . \quad (3.4)$$

Note that the first term in (3.4) is independent of the delay between the pump and probe. It is the term arising from the cross correlation between the pump and probe that depends on the delay δ. In the scan of the signal as a function of δ, the first term in (3.4) only contributes to the background and thus the interesting contribution will come only from the term depending on $C(\omega)$, i.e., from

$$S^{(c)}(\omega)=36I^{(l)}(\omega)\left|\int d\omega_2 C(\omega_2)\chi^{(3)}(\omega,\omega_2,-\omega_2) \times e^{-i\omega_2\delta}\right|^2 . \quad (3.5)$$

The general form of $S^{(c)}(\omega)$ can be evaluated depending on the form of the spectral profile of the input field. Analytical results can be obtained for Lorentzian spectral profiles. For other profiles, such as Gaussian, the integrals are to be evaluated numerically. The total signal is the integrated sum of (3.5).

If the field has a very short correlation time, i.e., $I^{(l)}(\omega)\approx C(\omega)\approx I_0$, then the delay-dependent contribution to the signal becomes

$$S^{(c)}=\int S^{(c)}(\omega)d\omega \approx 36I_0^3\int d\omega\left|\int d\omega_2 e^{-i\omega_2\delta}\chi^{(3)}(\omega,\omega_2,-\omega_2)\right|^2 . \quad (3.6)$$

Thus the delay dependence of the signal is governed by the complex poles of the Raman-like susceptibility $\chi^{(3)}(\omega,\omega_2,-\omega_2)$. For example, a pole at $\omega_2=\omega_0-i\Gamma$ in the lower half of the complex ω_2 plane will yield a term of the form $e^{-2\Gamma\delta}$. The scan of the signal as a function of δ will therefore yield the width Γ. In general, $S^{(c)}$ will be a sum of several decaying exponentials, since $\chi^{(3)}$ can have several complex poles. For large delay times, the term with the smallest relaxation width will survive.

We next evaluate (3.6) explicitly for the case of four-wave mixing in a two-level system. The nonlinear susceptibility $\chi^{(3)}(\omega,\omega_2,-\omega_2)$ is well known for a two-level system:

$$\chi^{(3)}(\omega_2,-\omega_2,\omega)=\frac{\chi}{(\omega_0-\omega-i\Gamma)}\left[\frac{2}{i\gamma}\left[\frac{1}{\omega_0-i\Gamma-\omega_2}+\frac{1}{-\omega_0-i\Gamma+\omega_2}\right]+\frac{2}{(\omega-\omega_2+i\gamma)}\left[\frac{1}{\omega_0-i\Gamma-\omega}+\frac{1}{-\omega_0-i\Gamma+\omega_2}\right]\right] . \tag{3.7}$$

We assume that $\delta\neq0$. The only poles in the lower half of the complex plane contribute. Thus the nonvanishing contribution comes from $\omega_2=\omega_0-i\Gamma$ with the result

$$S^{(c)}=36I_0^3\,|\chi|^2\int\frac{d\omega}{(\omega_0-\omega)^2+\Gamma^2}\left[\frac{4\pi}{\gamma}\right]^2|e^{-i\omega_0\delta-\Gamma\delta}|^2$$
$$=\frac{(36)(16)\pi^3I_0^3}{\gamma^2\Gamma}e^{-2\Gamma\delta},\quad \delta>0 . \tag{3.8}$$

This result is in agreement with the work of Morita and Yajima.[1] The result is more complicated for $\delta<0$ because two poles at $\omega_2=\omega_0+i\Gamma$, $\omega_0+i\gamma$ contribute, which leads to three exponentials in $S^{(c)}$. Next consider a typical CARS experiment involving a single isolated resonance. In this case $\chi^{(3)}$ is given by

$$\chi^{(3)}(\omega,\omega_2,-\omega_2)\cong A+\frac{B}{\omega_2-\omega-\omega_R+i\Gamma} . \tag{3.9}$$

The integral in (3.6) is easily evaluated with the result

$$S^{(c)}(\omega)=36I_0^3\,|B|^2(2\pi)^2e^{-2\Gamma\delta},\quad \delta>0 . \tag{3.10}$$

It is important to note that the nonresonant background term A does not contribute to $S_c(\omega)$. Thus the spectroscopy with incoherent fields enables one to suppress the effects of the unwanted nonresonant terms.

IV. NONDEGENERATE FOUR-WAVE MIXING IN CROSS-CORRELATED FIELDS

We next treat the nondegenerate four-wave mixing in chaotic pump and probe fields with cross correlation. We will obtain a result analogous to (2.11). We now write the pump and probe fields as

$$E^{(l)}(t)=\varepsilon^{(l)}(t)e^{-i\omega_l t}+\text{c.c.} ,$$
$$E^{(s)}(t)=\varepsilon^{(s)}(t)e^{-i\omega_s t}+\text{c.c.} \tag{4.1}$$

The envelopes $\varepsilon^{(l)}(t)$ and $\varepsilon^{(s)}(t)$ are stationary random processes. In subsequent equations $\varepsilon^{(l)}(\omega)$ stands for the Fourier transform of $\varepsilon^{(l)}(t)$ and the spectrum of $\varepsilon^{(l)}(t)$ is centered around zero frequency. In view of (4.1), the induced polarization is now given by [cf. Eq. (2.4)]

$$P(\omega)=3\int d\{\omega\}\chi^{(3)}(\omega_1+\omega_l,\omega_2+\omega_l,-\omega_3-\omega_s)\times\varepsilon^{(l)}(\omega_1)\varepsilon^{(l)}(\omega_2)[\varepsilon^{(s)}(\omega_3)]^*\times\delta(\omega_1+\omega_2-\omega_3+2\omega_l-\omega_s-\omega) . \tag{4.2}$$

The spectrum of the coherently generated radiation can be calculated in the usual way leading to

$$S(\omega)=9\int d\{\omega\}d\{\omega'\}\chi^{(3)}(\omega_1+\omega_l,\omega_2+\omega_l,-\omega_3-\omega_s)[\chi^{(3)}(\omega_1'+\omega_l,\omega_2'+\omega_l,-\omega_3'-\omega_s)]^*\times\Gamma(\omega_1',\omega_2',\omega_3',\omega_1,\omega_2)\delta(\omega_1+\omega_2-\omega_3+2\omega_l-\omega_s-\omega)\delta(\omega_1'-\omega_2'-\omega_3'+2\omega_l-\omega_s-\omega) , \tag{4.3}$$

where Γ is the sixth-order correlation function of the envelopes of the pump and probe fields. For chaotic fields this correlation function is still given by Eq. (2.10) with $I(\omega)$ and $C(\omega)$ standing for the second-order correlation functions of the field envelopes. Upon substituting (2.10) in (4.3) and making use of the permutation symmetry of $\chi^{(3)}$, we get our key result for the nondegenerate four-wave-mixing signals in correlated chaotic fields,

$$S(\omega)=18\int d\{\omega\}I^{(l)}(\omega_1)I^{(l)}(\omega_2)I^{(s)}(\omega_3)\delta(\omega-\omega_1-\omega_2+\omega_3-2\omega_l+\omega_s)\,|\chi^{(3)}(\omega_1+\omega_l,\omega_2+\omega_l,-\omega_3-\omega_s)|^2+36I^{(l)}[\omega-(2\omega_l-\omega_s)]\left|\int C(\omega_2)d\omega_2\chi^{(3)}(\omega+\omega_s-\omega_l,\omega_2+\omega_l,-\omega_2-\omega_s)\right|^2 . \tag{4.4}$$

Note that for monochromatic but correlated chaotic fields (4.4) reduces to

$$S(\omega)=6\delta(\omega-2\omega_l+\omega_s)\,|\,3\chi^{(3)}(\omega_l,\omega_l,-\omega_s)\,|^2 \times (I^{(l)})^2 I^{(s)} . \quad (4.5)$$

The numerical factor 6 is the kinematical enhancement factor that arises due to (a) chaotic nature and (b) complete correlation of the pump and probe fields. The term dependent on the correlation between the pump and probe shows that the four-wave mixing at $\omega\approx 2\omega_l-\omega_s+x$ can result in (a) absorption of a photon in the vicinity of ω_l and (b) emission of a photon in the vicinity of ω_s, such that the net difference between the absorption and emission process remains $\omega_l-\omega_s$. This is due to the cross correlation between the pump and probe. These two processes are to be accompanied by the absorption of another photon of frequency ω_l+x.

V. APPLICATION OF THE CONVOLUTION RESULT TO FLUCTUATION-INDUCED RESONANCES IN FOUR-WAVE MIXING

We now show how the formulation of Sec. II can be used to explain the existence of the fluctuation-induced resonances that arise in four-wave mixing with cross-correlated pump and probe fields. Such resonances have been recently[4,7] discussed by assuming the phase-diffusion model[15] for the pump and probe fluctuations. Our present discussion will show their existence for chaotic fields, thus establishing the general existence of the fluctuation induced-resonances for the different statistical natures of the pump and probe fields.

For the sake of brevity we consider a V system with two excited Zeeman states labeled $|1\rangle$ and $|2\rangle$. The ground state will be denoted by $|3\rangle$. We also restrict our analysis to the case of degenerate four-wave mixing. The susceptibility $\chi^{(3)}(\omega_1,-\omega_1,\omega_2)$ is known for the three-level model. We quote its simplified form for the case of radiative relaxation of the excited states at the rates γ_i,

$$\chi^{(3)}(\omega_1,-\omega_1,\omega_2)\,|\,\varepsilon_1^2\,|\,\varepsilon_2=(d_{31}\sigma_1+d_{32}\sigma_2) , \quad (5.1)$$

where the optical coherences σ_1 $(=\rho_{13})$ and σ_2 $(=\rho_{23})$ are given by

$$\sigma_1=(i\delta+\Gamma_1+i\Delta_1)^{-1}(-ig_2\sigma_5-2ig_1\sigma_7-ig_1\sigma_8 -i\lambda_2\Psi_5-2i\lambda_1\Psi_7-i\lambda_1\Psi_8) ,$$

$$\sigma_2=(i\delta+\Gamma_2+i\Delta_2)^{-1}(-ig_1\sigma_6-ig_2\sigma_7-2ig_2\sigma_8 -i\lambda_1\Psi_6-i\lambda_2\Psi_7-2i\lambda_2\Psi_8) , \quad (5.2)$$

$$\Psi_5=g_1g_2^*(\Gamma_1+i\Delta_1)^{-1}(\Gamma_2-i\Delta_2)^{-1}=\Psi_6^* ,$$

$$\Psi_7=|\,g_1\,|^2/(\Gamma_1^2+\Delta_1^2),\quad \Psi_8=|\,g_2\,|^2/(\gamma_2^2+\Delta_2^2) , \quad (5.3)$$

$$\sigma_5=\lambda_1g_2^*(\Gamma_2-i\Delta_2)^{-1}(\Gamma_1+i\Delta_1+i\delta)^{-1} ,$$

$$\sigma_6=\lambda_2g_1^*(\Gamma_1-i\Delta_1)^{-1}(\Gamma_2+i\Delta_2+i\delta)^{-1} ,$$

$$\sigma_7=\lambda_1g_1^*(\Gamma_1-i\Delta_1)^{-1}(\Gamma_2+i\Delta_1+i\delta)^{-1} ,$$

$$\sigma_8=\lambda_2g_2^*(\Gamma_2-i\Delta_2)^{-1}(\Gamma_2+i\Delta_2+i\delta)^{-1},\quad \Gamma_i=\gamma_i/2 . \quad (5.4)$$

Here various detunings and the coupling constants are given by

$$\Delta_1=\omega_{13}-\omega_1,\quad \Delta_2=\omega_{23}-\omega_1,\quad \delta=\omega_1-\omega_2 ,$$

$$g_1=\mathbf{d}_{13}\cdot\boldsymbol{\varepsilon}_1,\quad g_2=\mathbf{d}_{23}\cdot\boldsymbol{\varepsilon}_1 ,$$

$$\lambda_1=\mathbf{d}_{13}\cdot\boldsymbol{\varepsilon}_2,\quad \lambda_2=\mathbf{d}_{23}\cdot\boldsymbol{\varepsilon}_2 . \quad (5.5)$$

The expression (5.1) is to be used in (2.13) to obtain the contribution to the signal from the cross correlation of the pump and probe. The Hanle resonance corresponds to $\Delta_1=\Delta_2$. Such a resonance can arise from terms like $(\Gamma_1+i\Delta_1)^{-1}(\Gamma_2-i\Delta_2)^{-1}$. We thus focus our attention on the following term in expression (5.1):

$$H\equiv(i\delta+\Gamma_1+i\Delta_1)^{-1}(-i\lambda_2g_1g_2^*)(\Gamma_1+i\Delta_1)^{-1}\times(\Gamma_2-i\Delta_2)^{-1}d_{31} +(i\delta+\Gamma_2+i\Delta_2)^{-1}(-i\lambda_1g_1g_2)(\Gamma_1-i\Delta_1)^{-1}\times(\Gamma_2+i\Delta_2)^{-1}d_{32} . \quad (5.6)$$

One needs to evaluate integrals like

$$\int d\omega_1\frac{\gamma_c/\pi}{(\omega_1-\omega_0)^2+\gamma_c^2}\,\frac{1}{(\omega_{13}-\omega_1-i\Gamma_1)(\omega_{23}-\omega_1+i\Gamma_2)} =\left[1-\frac{2i\gamma_c}{\omega_{12}-i(\Gamma_1+\Gamma_2)}\right]\frac{1}{[\omega_{13}-\omega_0-i(\Gamma_1+\gamma_c)][\omega_{23}-\omega_0+i(\gamma_c+\Gamma_2)]} . \quad (5.7)$$

Thus for a large value of the detuning $\Delta_1\approx\Delta_2\approx\Delta$ and upon ignoring the vectorial character of the dipole matrix elements, we find that the term (5.6) contributes the following to the four-wave-mixing signal:

$$S_c\propto\frac{I^3}{\Delta^6}\left|1+\frac{2\gamma_c(\Gamma_1+\Gamma_2)}{\omega_{12}^2+(\Gamma_1+\Gamma_2)^2}\right|^2+\cdots . \quad (5.8)$$

The other terms in (5.8) contribute to the background which also depends on γ_c. It is clear from (5.8) that the Hanle resonance in a four-wave-mixing signal can result from the correlated fluctuations of the pump and probe, even in the absence of the dephasing collisions.[16] Note that the width of the Hanle resonance is independent of γ_c. This is in contrast to the pressure-induced resonance[16] in systems, which generally has a width proportional to the pressure.

In conclusion we have developed a general theory[17,18] of nonlinear spectroscopy with correlated pump and probe fields. Many applications of this theory are possible. We have demonstrated its applicability to delayed pump-probe experiments, and to the fluctuation-induced resonances.

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[18]We have also shown that the contribution $S^{(c)}(\omega)$ can be measured directly in degenerate four-wave mixing by homodyning the coherently generated signal with the pump [G. S. Agarwal, in *Proceedings of the International Laser Science Conference III,* edited by W. C. Stwalley and M. Lapp (AIP, New York, 1988)].