

High-pump-power effects on the four-wave mixing signal for a two-level system coupled with a thermal bath

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Abstract

We present a formalism to incorporate high order effects of the pump field on the four-wave mixing signal for a two-level system immersed in a thermal bath. We derive analytical expressions for the nonlinear polarization in terms of the field amplitude and thermal noise parameters.

Key words: Four-wave mixing; stochastic; two-level.

Efectos de alta potencia de bombeo sobre la señal de mezcla de cuatro ondas para un sistema de dos niveles acoplados con un baño térmico

Resumen

Presentamos un formalismo para incorporar efectos de alto orden del campo de bombeo sobre la señal de mezcla de cuatro ondas para un sistema de dos niveles inmerso en un baño térmico. Derivamos expresiones analíticas para la polarización no lineal en términos de la amplitud del campo y de los parámetros del ruido térmico.

Palabras clave: Dos niveles; estocástico; mezcla de cuatro ondas.

Introduction

Four-wave mixing (FWM) is a fruitful area for the study of ultrafast phenomena, where two pump fields and one probe field interact via the third-order susceptibility $\chi^{(3)}$ to generate the signal field (1) Different radiative and nonradiative processes take place during the field-matter nonlinear interaction. For optical transitions in completely isolated atoms or molecules the

dominant contribution to the longitudinal and transversal relaxation times, T_1 and T_2 , respectively, comes from radiative effects associated with reaction or zero-point fluctuations of the free electromagnetic field. The radiative effects are certainly not the only source and, usually, they are not the dominant source of relaxation fluctuations and incoherences. The effects of such fluctuations (associated to the electromagnetic fields) have been studied in several nonlin-

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ear optical processes including resonance fluorescence (2), multiphoton absorption (3), multiphoton ionization (4) and stimulated Raman scattering (5). In other cases where the relaxation mechanism involves changes in the transition frequency of the molecular system, the equations governing the time evolution of the Bloch vector are integro-differential in nature, and defined as Optical Stochastic Bloch equations (OSBE). Most of the work done in nonlinear optics with the Optical Conventional Bloch equations (OCBE) considers the transition frequency of the two-level system ω_0 as a parameter. This assumption is valid in regimes of weak solute-solvent interaction. However, if one is interested in the generation of radiation from a two-level system driven by a monochromatic field and immersed in a condensed medium the assumption is no longer valid. Under those circumstances, molecular collisions induce frequency shifts, and due to the nature of the collisions, they lead to a stochastic modulation of the frequency. This shift in the transition frequency becomes a function of time and can be represented as $\xi(t) = \omega_0 + \sigma(t)$, where $\sigma(t)$ comprises all the stochasticity of ξ . These fluctuations have been treated by different theories, which correspond to different mathematical models for the frequency fluctuations: Gaussian-Markovian model (6), non-Markovian models (7), and a "random-telegraph model" (8).

It is the purpose of this work to generalize our previous treatment on the stochasticity of the heat reservoir and its effect on the two-level molecular system, in FWM technique, in the case where the pump-field is considered at all orders. We obtain analytical expressions for the induced macroscopic polarization in a spatial local regime.

Theory

Let us consider a molecule represented by a two-level system defined by the states $|a\rangle$ and $|b\rangle$. As a result of the effect of the

heat reservoir surrounding the molecule, the transition frequency of the system, ξ is well characterized as a stochastic variable. The time evolution of the population of the system and the coherence between them is given by the well known OSBE. This is:

$$\dot{\rho}(t) = M_{\xi}(t)\rho(t) \tag{1}$$

$$\text{where } \rho(t) = \begin{pmatrix} \rho_{ba}(t) & 0 \\ \rho_{ab}(t) & 0 \\ \rho_D(t) & \rho_D^{(0)}/T_1 \end{pmatrix} \text{ and}$$

$$M_{\xi}(t) = \begin{pmatrix} A & 0 & \frac{i}{\hbar} \bar{\mu}_{ba} \cdot \vec{E} \\ 0 & B & \frac{i}{\hbar} \bar{\mu}_{ab} \cdot \vec{E} \\ \frac{2i}{\hbar} \bar{\mu}_{ab} \cdot \vec{E} & \frac{2i}{\hbar} \bar{\mu}_{ba} \cdot \vec{E} & 1/T_1 \end{pmatrix}$$

$$A = (i\xi - 1/T_2) \frac{i}{\hbar} (\bar{\mu}_{aa} - \bar{\mu}_{bb}) \cdot \vec{E}$$

$$B = (i\xi - 1/T_2) \frac{i}{\hbar} (\bar{\mu}_{aa} + \bar{\mu}_{bb}) \cdot \vec{E}$$

where we have defined $\rho_D(t) = \rho_{aa}(t) - \rho_{bb}(t)$; $\rho_D^{(0)}$ is the equilibrium population difference; $\bar{\mu}_{ba} = \bar{\mu}_{ab}^*$ and $\bar{\mu}_{ii}$ ($i = a, b$) are the transition and permanent dipole moments, respectively; finally $\vec{E}(t)$ corresponds to the amplitude of the total classical external field, given by $\vec{E}(t) = \vec{E}_1(t) + \vec{E}_2(t)$, where is defined $\vec{E}_m(t) = \vec{E}_m(\omega_m) \exp(i\omega_m t)$ c.c., with $\vec{E}_m(\omega_m) = (\vec{E}_{om}/2) \exp(i(\vec{k}_m \cdot \vec{r}))$.

Considering Eq. [1] not taking into account the $\bar{\mu}_{ii}$ in the molecule, and making use of a perturbation expansion to first order in the probe beam (index 2), but at all orders in the pump-field (index 1), we obtain the following relationships for the coherence that oscillate at frequency of interest $\omega_3 = 2\omega_1 - \omega_2$ associated to the FWM generate signal:

$$\rho_{ba}(\omega_3) = \frac{2i|\bar{\mu}_{ba}|^2|\bar{\mu}_{ab}|}{\hbar^3 D_3} \frac{1}{D_1} \frac{1}{D_2^*} \rho_D^{dc} E_1(\omega_1) E_1(\omega_1) E_2^*(\omega_2) \quad [2]$$

$$\text{where } \Gamma_1(\Delta) = 2|\mu_{11}|^2 \frac{1}{D_3} \frac{1}{D_2^*};$$

$$D_j = \frac{1}{T_2} i(\xi - \omega_j) \text{ for } j = 1, 2, 3, \text{ given by:}$$

$$\rho_D^{dc} = \rho_D^{(0)} \frac{|D_1|^2 T_2^2}{|D_1|^2 T_2^2 + 4S}. \quad [3]$$

Here, S is the saturation parameter that represents a measure of the radiation-matter intensity coupling, defined by $S = |\mu_{11}|^2 T_1 T_2$ where $\mu_{11} = \bar{\mu}_{ba} \cdot \bar{E}_1(\omega_1) / \hbar$ is the Rabi frequency. In comparison with Eq. [2], but considering one pump field at all orders, we have studied the effects of the solvent on the optical properties (9). In Eq. [2] the crucial assumption is that the transition frequency ξ is the slow variation during the time needed for the system to reach the steady-state regime. For simplicity, we consider in the present work the resonant case for the optical frequencies, this is $\omega_1 \sim \omega_2$, and is related with a maximum of the population effect (10). Under these circumstances, we obtain:

$$\rho_{ba}(\omega_3) = \frac{i\lambda}{D_\xi |D_\xi|^2} E_1(\omega_1) E_1(\omega_1) E_2^*(\omega_1) \frac{1}{D_1} Y_\xi^{-1}, \quad [4]$$

$$\text{where } D_\xi = D_m \quad (m=1, 2, 3); \lambda = 4|\mu_{ba}|^2 \mu_{ab} T_1 \rho_D^{(0)} \hbar^3 T_2^{-1}; Y_\xi = 4S / T_2^2 |D_\xi|^2.$$

Considering typical magnitudes of the relaxation times, and transition dipole moments, associated to organic molecules (11), it is possible consider that $|Y_\xi| \gg 1$. After some algebraic manipulations, we can write:

$$\rho_{ba}(\omega_3) = i\lambda E_1(\omega_1) E_1(\omega_1) E_2^*(\omega_1) \sum_{n=0}^M \Lambda_n(S) \langle \xi^{(n)} \rangle_\xi \quad [5]$$

$$\text{where } \Lambda_n(S) = (1)^n (n-1) 4S / T_2^{2-n} \text{ and } \langle \xi^{(n)} \rangle_\xi = \frac{1}{D_\xi |D_\xi|^{2-2n}} \text{ for } n = 0, 1, 2, 3, \dots$$

In the above summation M is the order required to achieve numerical convergence of the induce coherence. Considering the shift in the transition frequency it becomes a quasi-function of time, the average of the coherence, over the distribution of ξ will be given by:

$$\langle \rho_{ba}(\omega_3) \rangle_\xi = i\lambda E_1(\omega_1) E_1(\omega_1) E_2^*(\omega_1) \sum_{n=0}^M \Lambda_n(S) \langle \xi^{(n)} \rangle_\xi \quad [6]$$

In the above equation for the coherence, its ensemble has to be performed over all realizations of the events ξ . In order to do so, van Kampen (12-14) has proposed a method, where he formally solves the stochastic differential equation as if it were a deterministic one and then takes an average over the realizations of the stochastic variable. A different approach is based on taking the same average before solving the Optical Bloch equations. In the latter case, the set of differential equations obtained will contain a multiplicative noise term (which may be described as an Ornstein-Uhlenbeck process (OUP)), the set has to be solved in order to obtain an equation for the average of $\rho_{ba}(\omega)$. In order to solve the OSBE, we have explored two different approaches. On the one hand, we have first taken an average of the OSBE and later on, using a theorem due to Novikov, we have performed the average involving the multiplicative noise (15,16). We have solved the OSBE perturbatively, as if it they were a set of OCBE, and then we have taken an average of the formal solution of the coherence over the realizations of ξ (9,17). For those cases when high-power is considered we have decided to use a method similar to the second one mentioned above.

That is, we solved the equations as if they were deterministic, and only then, acknowledging the fact that $\rho_{ba}(\omega)$ depends on the realizations of ξ , we took its ensemble-average over the distribution of states with molecular frequencies within ξ and $\xi \pm d\xi$. Thus, solving the stochastic equations and by taking ensemble-average, instead of taking a time-average, we have by-passed the problem of solving the averages involving the multiplicative noise term.

The ensemble average values, Eq. [6] can be calculated using a method described extensively in the previous work (9), and where the solvent-solute interaction is modeled according to an OUP with intensity γ and exponential correlation function with decaying rate τ . The first term of the summation in Eq. [6] is given by:

$$\left\langle \left\langle \frac{1}{D_\xi |D_\xi|^2} \right\rangle_\xi \right\rangle_\xi \quad [7]$$

In order to take the average in Eq. [7] we first define the series $\tilde{\sim}^{(n)}_\xi = 1 / D_\xi |D_\xi|^{2n}$, for $n = 0, 1, 2, 3, \dots$, whose average can be calculated as follows. Consider the first term in the series $\tilde{\sim}^{(0)}_\xi$, its average is (9):

$$\left\langle \tilde{\sim}^{(0)}_\xi \right\rangle_\xi = \frac{1}{\sqrt{\gamma\tau}} \text{Re} \langle \omega_0 \rangle + i \text{Im} \langle \omega_0 \rangle \quad [8]$$

where $\omega_0 = 1 - \text{erf}(u) \exp u^2$ with

$$u = T_2^{-1} i \Delta_1 (2\gamma\tau)^{1/2}; \Delta_1 = \omega_1 - \omega_0, \quad [9]$$

and $\text{erf}(u)$ meaning error function evaluated at u . Full details of this before average, are given in ref. (9). With this result in hand, it is possible to evaluate the average given in Eq. [7], that is:

$$\left\langle \tilde{\sim}^{(1)}_\xi \right\rangle_\xi = \left\langle \left\langle \frac{1}{D_\xi |D_\xi|^2} \right\rangle_\xi \right\rangle_\xi, \quad [10]$$

which can be expressed as:

$$\left\langle \left\langle \frac{1}{D_\xi |D_\xi|^2} \right\rangle_\xi \right\rangle_\xi = \left\langle \text{Re} \langle \omega_0 \rangle \right\rangle_\xi + i \left\langle \text{Im} \langle \omega_0 \rangle \right\rangle_\xi. \quad [11]$$

After some algebraic manipulations, and using the tools described in ref. (9), we can write:

$$\left\langle \text{Re} \langle \omega_0 \rangle \right\rangle_\xi = \frac{T_2}{2\sqrt{\gamma\tau}} T_2 \frac{1}{T_2\gamma\tau} \text{Re} \langle \omega_0 \rangle + \frac{\Delta_1}{\gamma\tau} \text{Im} \langle \omega_0 \rangle = \frac{2}{\sqrt{2\pi\gamma\tau}} \quad [12.a]$$

$$\text{Im} \langle \omega_0 \rangle = \frac{1}{\gamma\tau} \frac{1}{T_2} \text{Im} \langle \omega_0 \rangle - \Delta_1 \text{Re} \langle \omega_0 \rangle \quad [12.b]$$

The other terms of the expansion, Eq. [6] are of the general form:

$$\left\langle \left\langle \frac{1}{D_\xi |D_\xi|^{2n}} \right\rangle_\xi \right\rangle_\xi = \left\langle \text{Re} \langle \omega_n \rangle \right\rangle_\xi + i \left\langle \text{Im} \langle \omega_n \rangle \right\rangle_\xi \quad [13]$$

$n = 1, 2, 3, \dots$

where:

$$\left\langle \text{Re} \langle \omega_n \rangle \right\rangle_\xi = \frac{T_2^2}{2n} \left\langle \text{Re} \langle \omega_{n-1} \rangle \right\rangle_\xi - \frac{T_2}{2n} \frac{1}{(1/T_2)} \left\langle \text{Re} \langle \omega_{n-1} \rangle \right\rangle_\xi$$

$$- \sum_{m=0}^{n-1} \binom{n-1}{m} A_{m,n} \frac{T_2^{(2n-m)}}{2^n n!} \frac{1}{(1/T_2)^m} \left\langle \text{Re} \langle \omega_m \rangle \right\rangle_\xi$$

and

$$\left\langle \text{Im} \langle \omega_n \rangle \right\rangle_\xi = \frac{T_2}{2n} \frac{1}{\Delta_1} \left\langle \text{Re} \langle \omega_{n-1} \rangle \right\rangle_\xi - \frac{T_2^n}{2^n n!} \frac{1}{\Delta_1^n} \left\langle \text{Re} \langle \omega_0 \rangle \right\rangle_\xi,$$

and where the coefficients $A_{m,n}$ can be selected from Table 1. The Eq. [6] is rewrite as:

$$\left\langle \rho_{ba}(\omega_3) \right\rangle_\xi = \text{Re} \langle \rho_{ba}(\omega_3) \rangle_\xi + i \text{Im} \langle \rho_{ba}(\omega_3) \rangle_\xi, \quad [14]$$

where

Table 1
Values of the coefficient $A_{m,n}$

n	$m=0$	$m=1$	$m=2$	$m=3$	$m=4$	$m=5$	$m=6$
0	1						
1	1	1					
2	3	3	1				
3	15	15	6	1			
4	105	105	45	6	1		
5	945	945	420	141	21	1	
6	—	—	—	—	—	—	1

$$\operatorname{Re}\langle\rho_{ba}(\omega_3)\rangle_{\xi} = \lambda E_1(\omega_1)E_1(\omega_1)E_2^*(\omega_1) \Lambda_n(\mathcal{S}) \langle \operatorname{Im} \left(\frac{\rho_{ba}(\omega_3, \omega_0)}{\xi} \right) \rangle_{\xi} \quad \bar{P}(\omega_3) = N \left\langle \left\langle \rho_{ba}(\omega_3, \omega_0) \right\rangle_{\xi} \bar{\mu}_{ab} \right\rangle \quad [16]$$

and

$$\operatorname{Im}\langle\rho_{ba}(\omega_3)\rangle_{\xi} = \lambda E_1(\omega_1)E_1(\omega_1)E_2^*(\omega_1) \Lambda_n(\mathcal{S}) \langle \operatorname{Re} \left(\frac{\rho_{ba}(\omega_3, \omega_0)}{\xi} \right) \rangle_{\xi}.$$

The Fourier component of the total induced macroscopic complex polarization for inhomogeneous linewidth in general terms, is calculated from:

$$\bar{P}(\omega_3) = N \int_{-\infty}^{\infty} d\omega_0 \rho_{ba}(\omega_3, \omega_0) g(\omega_0) \bar{\mu}_{ab} \quad [15]$$

$$N \left\langle \left\langle \rho_{ba}(\omega_3, \omega_0) \right\rangle_{g(\omega_0)} \bar{\mu}_{ab} \right\rangle$$

where the external bracket $\langle \dots \rangle$ denotes an average over molecular orientations of the system and the internal one, represents the average over the distribution of molecular frequencies $g(\omega_0)$. In the case studied in the present article, we have considered a frequency ω_0 , whose value changes due to interactions with the surrounding media. Here, an average has to be taken respect to the possible realizations of the molecular frequency ξ due to the interactions with the bath. Hence, an observable variable, for instance, the complex polarization will be of the form:

Notice that the formal similarities between these two expressions for the polarization are striking. However, as it should be clear by now, the physical situations behind each case are quite different. The similarities relying only on the fact that an average over different probability density functions has to be taken in each case. Using Eq. [14], we write:

$$\bar{P}(\omega_3) = \lambda \bar{\mu}_{ab} E_1(\omega_1)E_1(\omega_1)E_2^*(\omega_1) \Lambda_n(\mathcal{S}) \langle \operatorname{Im} \left(\frac{\rho_{ba}(\omega_3, \omega_0)}{\xi} \right) \rangle_{\xi}$$

$$i\lambda \bar{\mu}_{ab} E_1(\omega_1)E_1(\omega_1)E_2^*(\omega_1) \Lambda_n(\mathcal{S}) \langle \operatorname{Re} \left(\frac{\rho_{ba}(\omega_3, \omega_0)}{\xi} \right) \rangle_{\xi} \quad [17]$$

Within the scalar approximation and by considering both, the rotating-wave approximation and the steady-state approximation, we obtain:

$$P(\omega_3) = P^{(SV)}(\omega_3) + P^{coup}(\omega_3), \quad [18]$$

where $P^{(SV)}(\omega_3)$ is the solvent polarization at frequency ω_3 ; $P^{coup}(\omega_3)$ represents the value in the line center of the induced macroscopic polarization associated to the FWM process that oscillate at frequency ω_3 , which is

referred to the coupling process between the probe and pump fields. Without considering the tensorial property of the susceptibility, we write:

$$P^{coup}(\omega_3) = \chi^{coup}(\omega_3) E_1(\omega_1) E_1(\omega_1) E_2^*(\omega_2) \quad [19]$$

where $\chi^{coup}(\omega_3)$ can be expressed as:

$$\chi^{coup}(\omega_3) = \text{Re} \chi^{coup}(\omega_3) + i \text{Im} \chi^{coup}(\omega_3) \quad [20]$$

$$\text{where } \text{Re} \chi^{coup}(\omega_3) = \lambda \mu_{ab} \sum_{n=0}^M \Lambda_n \langle \text{Im} \chi_{\xi}^{(n)} \rangle_{\xi}$$

$$\text{and } \text{Im} \chi^{coup}(\omega_3) = \lambda \mu_{ab} \sum_{n=0}^M \Lambda_n \langle \text{Re} \chi_{\xi}^{(n)} \rangle_{\xi}.$$

In Eq. (20) we can write the two types of contributions in FWM process. Firstly, the third-order process (indicated by Λ_0), and secondly the high-order, associated to the saturative effects (indicated by Λ_n with $n \neq 0$). The real and imaginary parts of $\chi^{coup}(\omega_3)$ will be expressed as:

$$\text{Re} \chi^{coup}(\omega_3) = \chi_{\text{Re}[\chi]}^{(3)}(\omega_3) + \chi_{\text{Re}[\chi]}^{eff}(\omega_3) \quad [21]$$

$$\text{and } \text{Im} \chi^{coup}(\omega_3) = \chi_{\text{Im}[\chi]}^{(3)}(\omega_3) + \chi_{\text{Im}[\chi]}^{eff}(\omega_3) \quad [22]$$

where

$$\chi_{\text{Re}[\chi]}^{(3)}(\omega_3) = \lambda \mu_{ab} \langle \text{Im} \chi_{\xi}^{(0)} \rangle_{\xi};$$

$$\chi_{\text{Re}[\chi]}^{eff}(\omega_3) = \lambda \mu_{ab} \sum_{n=1}^M \Lambda_n \langle \text{Im} \chi_{\xi}^{(n)} \rangle_{\xi}$$

$$\chi_{\text{Im}[\chi]}^{(3)}(\omega_3) = \lambda \mu_{ab} \langle \text{Re} \chi_{\xi}^{(0)} \rangle_{\xi};$$

$$\chi_{\text{Im}[\chi]}^{eff}(\omega_3) = \lambda \mu_{ab} \sum_{n=1}^M \Lambda_n \langle \text{Re} \chi_{\xi}^{(n)} \rangle_{\xi}.$$

The subscripts $\text{Re}[\chi]$ and $\text{Im}[\chi]$ state that these magnitudes were derived from the real and imaginary parts of the susceptibility, respectively. The superscript (3) denotes the third order contribution in the susceptibility, for which the FWM process

occur; the superscript “*eff*” corresponds to the effective scalar susceptibility at frequency ω_3 , responsible for the saturative effects. With all these results at hand, the complex macroscopic polarization, is finally rewritten as:

$$P(\omega_3) = P^{(SV)}(\omega_3) + \chi^{(3)}(\omega_3) E_1(\omega_1) E(\omega_1) E_2^*(\omega_2) + \chi^{eff}(\omega_3) E_1(\omega_1) E(\omega_1) E_2^*(\omega_2) \quad [23]$$

where $\chi^{(3)}(\omega_3) = \chi_{\text{Re}[\chi]}^{(3)}(\omega_3) + i \chi_{\text{Im}[\chi]}^{(3)}(\omega_3)$ and $\chi^{eff}(\omega_3) = \chi_{\text{Re}[\chi]}^{eff}(\omega_3) + i \chi_{\text{Im}[\chi]}^{eff}(\omega_3)$.

Once the expression for the complex macroscopic polarization has been obtained, the intensity of nonlinear signal associated to the FWM process in the local regime, is calculated from:

$$I(\omega_3) = \frac{c\epsilon}{8\pi} |P(\omega_3)|^2 \quad [24]$$

Eq. [24] is formally exact. However, their numerical application will be practically only if the summation, Eq. [5] converges after considering a few terms, and is valid the optical resonance condition between the two beams.

Final comments

In this work we have presented a formalism to study high-order effects of the pump-beams in FWM signal for a two-level system immersed in a thermal bath. The interaction solute-solvent induces frequency shifts that lead to a stochastic modulation of the transition Bohr frequency. The present formalism has practical applicability only when the expression $4S / T_2^2 |D_{\xi}|^2$ is less than one, and for the resonant case, given by $\omega_1 \sim \omega_2$. Unfortunately, when is not valid this condition, the solution of the problem becomes more complicated, by considering products of different lorentzian curves, of same spectral width (of the order of $1/T_2$)

but centered at different optical frequencies. It is important to notice that, though the previous equation for the nonlinear intensity, is valid only in the region of maximum resonance, it offers the experimentalist the condition of maximum sensitivity if the saturation effects of the field are under study. This condition is related with a maximum of the population pulsation effect (10). It is convenient to stress also that the equation we obtained allows for the identification of two effects in a separable form. The first one is identified by the $\Lambda_n(S)$, which determines the effects of the field in the FWM signal. The second effect is identified by the function $\xi^{(n)}$ which governs the different contributions from the effects of the solvent on the processes in FWM. The previous results are applicable in the local limit since we did not take into account the absorption of the signal with frequency ω_3 in the optical length traversed.

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