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Effects of the spectral inhomogeneity on the optical properties in the four-wave mixing signal

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Abstract

Effects of the inhomogeneous broadening on the optical responses of a two-level system in the four-wave mixing signal (FWM) were studied in this work. In this model, we introduced in the optical Bloch equations a stochastic Bohr frequency, generated as a consequence of the solute–solvent molecular collisions. These expressions are averaged in the statistical ensemble over all realizations of the random variables. Using the convolution theorem, it is possible to incorporate the saturation effects of the electromagnetic field treated at all orders of perturbation. For the averages, we have considered two distributions of linewidth. From the analytical expressions, for both linear and nonlinear responses, numerical calculations were carried out to obtain surfaces as a function of the magnitude of the pump field and the detuning factor. Saturation effect is shown in the linear and the coupled nonlinear responses. In this work, we have already considered the inhomogeneous spectral line and studied the cases in which all the fields propagate through the medium.

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1. Introduction

In the literature, the stochastic Bohr frequency $\xi(t) = \lambda(t) + \omega_0$ on the optical Bloch equations (OBE) has been introduced to obtain the optical stochastic Bloch equations (OSBE) [1–4]. In that expression, $\lambda(t)$ comprises all the fluctuation over the conventional Bohr frequency ω_0, due to different collisions that occur with the environment, so the average $\langle\lambda(t)\rangle = 0$ is valid [5]. Various methods have been used to resolve this system of equations [6–12]. In previous works, we have presented different formalisms to do it. Particularly, in [13,14] the OSBE have been used to describe the interaction of a two-level system in the presence of a four-wave mixing (FWM) signal, that is, the interaction of three laser fields with the wave vectors $\vec{k}_1$, $\vec{k}_2$, and $\vec{k}_3$ and frequencies ω_1, ω_2, and ω_3, where the subscripts 1, 2, and 3 refer to the pump, probe, and signal beams, respectively. Specifically, this technique implies that two-pump beams are focused onto the resonant medium generating a signal with a frequency of works $\omega_3 = 2\omega_1 - \omega_2$ and wave vectors $\vec{k}_3 \approx 2\vec{k}_1 - \vec{k}_2$. In these publications, the average of the coherence over all realizations of the stochastic variable was calculated from a gaussian probability density. At present, we extend that model to consider lorentzian probability distribution and to compare its results with those obtained with the first distribution. From the averaged coherence, we obtained analytical expressions for the components of susceptibility, both linear and nonlinear, and carrying out numerical calculations we obtain surfaces of the optical responses. The behavior of the susceptibility as a function of the parameters of the probability densities was

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explored and an important connection between those and the system–environment interaction was found. In Section 2, we explain in detail the theoretical considerations and methodology used by us. The numerical calculation is discussed in Section 3. Results are shown and analyzed in Section 4 and finally, in Section 5 we give the concluding remarks.

2. Theoretical considerations

The dynamics of a two-level molecular system defined by the states $|a\rangle$ and $|b\rangle$ in the presence of electromagnetic fields are obtained by solving the optical stochastic Bloch equations (OSBE) given by:

$$\dot{\rho}(t) = A(t)\rho(t) + R(t), \tag{1}$$

where $\rho(t)$ is the density matrix that represents the states of the system; $R(t)$ represents the nonradiative relaxation vectors, respectively,

$$\rho(t) = \begin{pmatrix} \rho_{ba}(t) \\ \rho_{ab}(t) \\ \rho_D(t) \end{pmatrix}; \quad R(t) = \begin{pmatrix} 0 \\ 0 \\ \rho_D^{(\mathrm{eq})}/T_1 \end{pmatrix} \tag{2}$$

and $A(t)$ is the radiative matrix that includes the stochastic variable given by:

$$A(t) = \begin{pmatrix} -(\mathrm{i}\xi(t) + 1/T_2) & 0 & \mathrm{i}\Omega \\ 0 & (\mathrm{i}\xi(t) - 1/T_2) & -\mathrm{i}\Omega^* \\ -2\mathrm{i}\Omega^* & 2\mathrm{i}\Omega & -1/T_1 \end{pmatrix}, \tag{3}$$

Here, the dipole-electric approximation has been considered; we have defined that $\rho_D = \rho_{aa}(t) - \rho_{bb}(t)$ is the difference in state population and the superscript "eq" denotes the equilibrium value in the absence of radiation; the interaction radiation–matter is defined by the Rabi frequency, given by $\Omega = \vec{\mu}_{ba} \cdot \vec{E}(t)/\hbar$, where $\vec{\mu}_{ba}$ is the transition dipole moment for the two-level system. In this model, we have considered the permanent dipole moments ($\vec{\mu}_{aa}$ and $\vec{\mu}_{bb}$) in the two different electronic molecular states equal to zero; the total field is given by $\vec{E}(t) = \vec{E}_1(t) + \vec{E}_2(t) + \vec{E}_3(t)$ with each field expressed classically according to $\vec{E}_j(t) = \vec{E}_j(\omega_j)\exp(-\mathrm{i}\omega_j t) + \mathrm{c.c.}$ with $\vec{E}_j(\omega_j) = (\vec{E}_{0j}/2)\exp[\mathrm{i}(\vec{k}_j \cdot \vec{r} + \phi_j)]$ representing the Fourier component that oscillates at frequency ω_j with a phase angle ϕ_j. In the present model, we use the semiclassical approximation, so our formulation does not include spontaneous relaxation channels.

We have assumed that in steady-state the elements of the stochastic density matrix satisfy the relations [15]:

$$\rho_{ba}(t) = \sum_{l=1} \rho_{ba}(\omega_l)\exp(-\mathrm{i}\omega_l t), \tag{4a}$$

$$\rho_D(t) = \rho_D^{\mathrm{dc}} + \sum_{m=1} \rho_D(m\Delta)\exp(-\mathrm{i}m\Delta t), \tag{4b}$$

$$\rho_{ba}(t) = \rho_{ab}^*(t), \tag{4c}$$

where $\Delta = \omega_1 - \omega_2$ is the detuning factor and ρ_D^{dc} is the direct current term of the population difference component that oscillates at zero frequency.

In the rotating wave approximation and in steady-state, the following relationships are obtained:

$$\begin{aligned}\Psi_{2n+1}\rho_{ba}(\omega_1 + n\Delta, \xi) &= \mathrm{i}\Omega_1\rho_D(n\Delta, \xi) + \mathrm{i}\Omega_2\rho_D[(n+1)\Delta, \xi] \\ &\quad + \mathrm{i}\Omega_3\rho_D[(n-1)\Delta, \xi],\end{aligned} \tag{5}$$

$$\begin{aligned}\Xi_n(n\Delta)\rho_D(n\Delta, \xi) &= [\delta_{n,0}\rho_D^{(\mathrm{eq})}/T_1] + 2\mathrm{i}\Omega_1^*\rho_{ba}(\omega_1 + n\Delta, \xi) \\ &\quad - 2\mathrm{i}\Omega_1\rho_{ab}[-(\omega_1 - n\Delta), \xi] \\ &\quad + 2\mathrm{i}\Omega_2^*\rho_{ba}[\omega_1 + (n-1)\Delta, \xi] \\ &\quad - 2\mathrm{i}\Omega_2\rho_{ab}\{-[\omega_1 - (n+1)\Delta], \xi\} \\ &\quad + 2\mathrm{i}\Omega_3^*\rho_{ba}[\omega_1 + (n+1)\Delta, \xi] \\ &\quad - 2\mathrm{i}\Omega_3\rho_{ab}\{-[\omega_1 - (n-1)\Delta], \xi\},\end{aligned} \tag{6}$$

where $\Psi_{2n+1} = \mathrm{i}(\xi - \omega_{2n+1}) + 1/T_2$ represents the resonant term that includes the stochastic molecular frequency ξ, for only the values of $n = 0, 1, -1$, defined for pump, signal, and probe, respectively, and where $\Psi_{-1} \equiv \Psi_2$; $\Xi_n(n\Delta) = 1/T_1 - \mathrm{i}n\Delta$; $\rho_{ba}(\omega_k, \xi)$ and $\rho_D(n\Delta, \xi)$ are the Fourier components of the coherence and population difference that oscillate at the frequencies ω_k and $n\Delta$, respectively. The identities $\rho_{ba}(\omega_k, \xi) = \rho_{ab}^*(-\omega_k, \xi)$ and $\rho_D(n\Delta, \xi) = \rho_D(-n\Delta, \xi)$ are valid.

2.1. Averaged coherence

Starting from Eqs. (5) and (6), and considering all orders of perturbation in the pump field, but only first order in probe and signal fields, the coherences at the pump, probe, and signal frequencies can be expressed as:

$$\rho_{ba}(\omega_1, \xi) = \frac{\mathrm{i}\Omega_1\rho_D^{\mathrm{dc}}}{\Psi_1}, \tag{7}$$

$$\begin{aligned}\rho_{ba}(\omega_2, \xi) = \frac{\mathrm{i}\Omega_2\rho_D^{\mathrm{dc}}}{\Psi_2} - \frac{2\mathrm{i}\Omega_2}{\Psi_2 Z_1^*}\left(\frac{1}{\Psi_1^*} + \frac{1}{\Psi_2}\right)|\Omega_1|^2\rho_D^{\mathrm{dc}} \\ - \frac{2\mathrm{i}\Omega_1^2\Omega_3^*}{\Psi_2 Z_1^*}\left(\frac{1}{\Psi_3^*} + \frac{1}{\Psi_1}\right)\rho_D^{\mathrm{dc}},\end{aligned} \tag{8}$$

$$\begin{aligned}\rho_{ba}(\omega_3, \xi) = \frac{\mathrm{i}\Omega_3\rho_D^{\mathrm{dc}}}{\Psi_3} - \frac{2\mathrm{i}\Omega_3}{\Psi_3 Z_1}\left(\frac{1}{\Psi_1^*} + \frac{1}{\Psi_3}\right)|\Omega_1|^2\rho_D^{\mathrm{dc}} \\ - \frac{2\mathrm{i}\Omega_1^2\Omega_2^*}{\Psi_3 Z_1}\left(\frac{1}{\Psi_2^*} + \frac{1}{\Psi_1}\right)\rho_D^{\mathrm{dc}},\end{aligned} \tag{9}$$

for the zero-frequency component of ρ_D we have:

$$\rho_D^{\mathrm{dc}} = \rho_D^{(\mathrm{eq})}\left[\frac{|\Psi_1|^2 T_2^2}{|\Psi_1|^2 T_2^2 + 4S}\right], \tag{10}$$