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Four-wave mixing in molecular magnets via electromagnetically induced transparency

Ying Wu and Xiaoxue Yang
Department of Physics, Huazhong University of Science and Technology, Wuhan 430074, People's Republic of China

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We perform a time-dependent analysis of four-wave mixing (FWM) in molecular magnets via electromagnetically induced transparency and obtain the analytical expressions of pulsed electromagnetic waves, including the FWM-generated pulse, group velocities, phase shifts, and absorption coefficients. We have also investigated analytically the time-dependent electromagnetically induced transparency in molecular magnets.

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I. INTRODUCTION

Recently, there has been considerable interest in investigating systems of molecular magnets because such systems are shown to have many interesting features both of fundamental importance, such as quantum magnetic phenomena at macroscopic scale, and wide applications in magnetic memory, quantum computing, and as powerful sources of coherent microwave radiation.[1–11]

On the other hand, there also exist intensive investigations on the oscillations and wave propagations in systems of molecular magnets such as the phonon super-radiance and phonon laser effect,[12] the nonstationary behavior of a high-spin molecule in a bifrequency ac magnetic field[13] or in an acoustic wave and an ac magnetic field,[14] the parametric interaction of two acoustic waves in the presence of a strong ac magnetic field,[15] the electromagnetically induced transparency,[16] and the nonlinear propagation of acoustic wave via electromagnetically induced transparency.[17]

The existence of the electromagnetically induced transparency (EIT) in systems of molecular magnets[16] is of particular significance because the EIT and the EIT-related phenomena in cold atom media have been proven to have a vast number of important applications and have hence received considerable attention in the past decades.[18–49] It opens up, therefore, an avenue to explore new possibilities for nonlinear optics and quantum information processing via the EIT by means of systems of molecular magnets.

In this paper, we analyze the four-wave mixing (FWM) in molecular magnets via electromagnetically induced transparency by a time-dependent theory. We shall derive the analytical expressions of pulsed electromagnetic waves including the FWM-generated pulse, group velocities, phase shifts and absorption coefficients. We have also investigated analytically the time-dependent electromagnetically induced transparency in molecular magnets, which goes beyond the previous results with a continuous-wave (cw) approximation.[16]

The paper is organized as follows. In Sec. II, we describe the FWM configuration and the corresponding model. We shall also discuss how to put the Hamiltonian into the convenient form for dealing with the FWM and derive the differential equations governing the dynamics of the molecular magnets and the propagations of the pulsed electromagnetic waves. In Sec. III, we present analytical solutions to these governing equations and the analytical expressions of pulsed electromagnetic waves including the FWM-generated pulse, group velocities, phase shifts, and absorption coefficients. Then, we investigate analytically the time-dependent electromagnetically induced transparency in molecular magnets and compare our results with the previous results with a cw approximation. Lastly, we discuss how to greatly diminish the Doppler effects in the FWM via EIT. Section IV concludes the paper with a summary and discussions.

II. MODEL AND GOVERNING EQUATIONS

We consider FWM in a system of noninteracting molecular magnets, as shown in Fig. 1, where a four-state molecule interacts with two continuous electromagnetic waves (1 and 4) and a weak-pulsed electromagnetic wave (2), and a pulsed FWM electromagnetic wave (3) can then be generated efficiently. Below, we describe the features of the system of noninteracting molecular magnets and derive the corresponding model and governing equations.

A. Hamiltonian and its simplification

In describing a system of noninteracting molecular magnets (for example, Mn_{12} acetate or Fe_8) subject to a dc magnetic field H_0 along the x axis and four electromagnetic waves with their magnetic fields of the form $(\mathbf{H}_j/2)e^{-i\omega_j t+i\mathbf{k}_j\cdot\mathbf{r}}+\text{c.c.}$ $(j=1,2,3,4)$, we can use the following Hamiltonian for one molecule[16] $\hat{\mathcal{H}}=\hat{\mathcal{H}}_0+\hat{\mathcal{V}}$ with $(\hbar=1)$

$$\hat{\mathcal{H}}_0=-D\hat{S}_z^2+\hat{\mathcal{H}}_{tr}-g\mu_B\hat{S}_xH_0, \tag{1a}$$

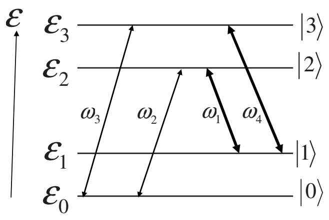

FIG. 1. Schematic of FWM in a system of molecular magnets. A four-level molecule (its states and the corresponding energies are denoted as $|j\rangle$ and $\mathcal{E}_j$, $j=0,1,2,3$) interact with two continuous electromagnetic waves (1 and 4) (with frequencies ω_1 and ω_4, respectively) serving as the pump fields and a weak-pulsed electromagnetic wave (2) of frequency ω_2 serving as a probe pump to generate a FWM-generated pulsed electromagnetic wave (3) of frequency ω_3.

$$\hat{\mathcal{V}} = -\frac{g\mu_B}{2}\sum_{j=1}^{4}\hat{\mathbf{S}}\cdot\mathbf{H}_j e^{-i\omega_j t + i\mathbf{k}_j\cdot\mathbf{r}} + \text{H.c.}, \tag{1b}$$

where z is assumed to be the easy anisotropy axis, $\hat{S}_x$, $\hat{S}_y$, and $\hat{S}_z$ are the x, y, and z projections of the spin operator, and $\hat{\mathcal{H}}_{tr}$ is the operator of the transverse anisotropy energy; D, g, and μ_B are the longitudinal anisotropy energy constant, the Landé factor, and the Bohr magneton, respectively.

Let φ_n or $|n\rangle$ and $\mathcal{E}_n$ $(n=0,1,2,\ldots)$ denote the eigenstates and eigenenergies of the Hamiltonian $\hat{\mathcal{H}}_0$, i.e., $\hat{\mathcal{H}}_0|n\rangle=\mathcal{E}_n|n\rangle$ so that $\hat{\mathcal{H}}_0=\Sigma_n\mathcal{E}_n|n\rangle\langle n|$. As shown in Fig. 1, we need to consider only the four lowest energy levels $|j\rangle$ $(j=0,1,2,3)$ relevant for the investigation of the four-wave mixing considered here and, hence, we can take $\hat{\mathcal{H}}_0=\Sigma_{j=0}^{3}\mathcal{E}_j|j\rangle\langle j|$ for our purpose. Besides, the wave magnetic fields $\mathbf{H}_2$ and $\mathbf{H}_4$ are chosen to be polarized along the x axis while $\mathbf{H}_1$ and $\mathbf{H}_3$ are chosen to be polarized along the y axis with their frequencies chosen to satisfy the relevant resonance or near-resonance conditions, i.e., $\omega_1\sim\omega_{21}$, $\omega_2\sim\omega_{20}$, $\omega_3\sim\omega_{30}$, and $\omega_4\sim\omega_{31}$, with $\omega_{jn}=|\mathcal{E}_j-\mathcal{E}_n|/\hbar$ denoting the corresponding transition frequencies. The interaction Hamiltonian in the interaction picture, $\hat{\mathcal{H}}_{int}=e^{i\hat{\mathcal{H}}_0 t/\hbar}\hat{\mathcal{V}}e^{-i\hat{\mathcal{H}}_0 t/\hbar}$, under the rotating-wave approximation reads as

$$\begin{aligned}\hat{\mathcal{H}}_{int}/\hbar = &-\Omega_1 e^{-i\delta_1 t+i\mathbf{k}_1\cdot\mathbf{r}}|2\rangle\langle 1| - \Omega_2 e^{-i\delta_2 t+i\mathbf{k}_2\cdot\mathbf{r}}|2\rangle\langle 0| \\ &- \Omega_3 e^{-i\delta_3 t+i\mathbf{k}_3\cdot\mathbf{r}}|3\rangle\langle 0| - \Omega_4 e^{-i\delta_4 t+i\mathbf{k}_4\cdot\mathbf{r}}|3\rangle\langle 1| + \text{H.c.},\end{aligned} \tag{2}$$

where δ_j are detunings defined as $\delta_1=\omega_1-\omega_{21}$, $\delta_2=\omega_2-\omega_{20}$, $\delta_3=\omega_3-\omega_{30}$, and $\delta_4=\omega_4-\omega_{31}$, and the Rabi frequencies Ω_j are defined as[16]

$$\Omega_1=\frac{g\mu_B H_1}{2\hbar}\langle 2|\hat{S}_y|1\rangle, \quad \Omega_2=\frac{g\mu_B H_2}{2\hbar}\langle 2|\hat{S}_x|0\rangle,$$

$$\Omega_3=\frac{g\mu_B H_3}{2\hbar}\langle 3|\hat{S}_y|0\rangle, \quad \Omega_4=\frac{g\mu_B H_4}{2\hbar}\langle 3|\hat{S}_x|1\rangle, \tag{3}$$

with H_j characterizing the magnitude of the wave magnetic field $\mathbf{H}_j$. Notice[16] that in the situation where the z and x axes coincide with the easy anisotropy axis and the dc magnetic field, respectively, all the expectation values $\langle j|\hat{S}_x|i\rangle$ in Eq. (3) are nonzero due to the symmetric feature of the eigenstates $|j\rangle$, i.e., $|0\rangle$ and $|2\rangle$ are symmetric states while $|1\rangle$ and $|3\rangle$ are antisymmetric states.

B. Description of the molecule's dynamics

In describing the system's dynamics, there usually exist two formalisms: the Shrödinger formalism, where the state is described by a state function or a state ket satisfying the Shrödinger equation with decay rates inclusive, and the density operator formalism, where the state is described by a density operator satisfying the master equation. It has been shown that the Shrödinger formalism is equivalent to but much simpler than the density operator formalism in describing the phenomenon of the EIT and the EIT-related multiwave mixing and soliton phenomena,[20,27,29,35–39,43–45] in particular, see Ref. 39.

We, therefore, adapt the much simpler Shrödinger formalism but we shall show that our results reduce to those of the EIT in molecular magnets[16] when the same conditions are considered.

Denoting the state of one molecule as $|\Psi\rangle=C_0(t)|0\rangle + C_1(t)e^{-i\Delta_1 t+i(\mathbf{k}_2-\mathbf{k}_1)\cdot\mathbf{r}}|1\rangle+C_2(t)e^{-i\Delta_2 t+i\mathbf{k}_2\cdot\mathbf{r}}|2\rangle+C_3(t)e^{-i\Delta_3 t+i\mathbf{k}_3\cdot\mathbf{r}}\times|3\rangle$, with $\Delta_1=(\omega_2-\omega_1)-\omega_{10}$ denoting a two-photon detuning and $\Delta_k=\omega_k-\omega_{k0}$ $(k=2,3)$ being single-photon detunings, and noting the phase matching conditions of $\mathbf{k}_3-\mathbf{k}_4=\mathbf{k}_2-\mathbf{k}_1$ and $\omega_3-\omega_4=\omega_2-\omega_1$ from the Schrödinger equation $i\hbar\partial|\Psi\rangle/\partial t=\hat{\mathcal{H}}_{int}|\Psi\rangle$ in the interaction picture, we then obtain the evolution equations for the probability amplitudes $C_j(t)$ as follows:

$$\frac{\partial C_1}{\partial t}=i(\Delta_1+i\gamma_1)C_1+i\Omega_4^*C_3+i\Omega_1^*C_2, \tag{4a}$$

$$\frac{\partial C_2}{\partial t}=i(\Delta_2+i\gamma_2)C_2+i\Omega_2C_0+i\Omega_1C_1, \tag{4b}$$

$$\frac{\partial C_3}{\partial t}=i(\Delta_3+i\gamma_3)C_3+i\Omega_3C_0+i\Omega_4C_1, \tag{4c}$$

where $2\gamma_k$ $(k=1,2,3)$ is the decay rate of the state $|k\rangle$.

C. Equations for propagation of electromagnetic waves

We consider the four-wave mixing where electromagnetic waves 1 and 4 are two strong cws and electromagnetic waves 2 and 3 are weak-pulsed electromagnetic waves. Consequently the envelopes $\mathbf{H}_1$ and $\mathbf{H}_4$ are independent of the space-time variables z and t while the slowly-varying envelopes $\mathbf{H}_2(z,t)$ and $\mathbf{H}_3(z,t)$ depend on the space-time variables z and t. The differential equations for $H_2(z,t)$ and $H_3(z,t)$ can be readily derived from Maxwell's equations and they read as

$$\left(\frac{\partial}{\partial z}+\frac{1}{v}\frac{\partial}{\partial t}\right)H_2(z,t)=i\frac{2\pi\omega_2\chi_2}{v}H_2(z,t), \tag{5a}$$

$$\left(\frac{\partial}{\partial z}+\frac{1}{v}\frac{\partial}{\partial t}\right)H_3(z,t)=i\frac{2\pi\omega_3\chi_3}{v}H_3(z,t), \tag{5b}$$

where χ_j $(j=2,3)$ is the magnetic susceptibility defined by the relation $\mathbf{M}_j(z,t)=\chi_j\mathbf{H}_j(z,t)$ for the magnetization $\mathbf{M}_j(z,t)e^{-i\omega_j t+ik_j z}/2+\text{c.c.}$ produced by the wave magnetic field $\mathbf{H}_j(z,t)e^{-i\omega_j t+ik_j z}/2+\text{c.c.}$ and v is the wave speed of the electromagnetic wave without taking into account the back action of the magnetization $\mathbf{M}(z,t)$.

It is pointed out that the static magnetization of the noninteracting molecular magnets has been included in the wave speed v, and the static magnetization should not be confused with the magnetization $\mathbf{M}(z,t)$ produced by the electromagnetic wave. Besides, in writing Eq. (5), we have approximately taken $v=v(\omega_2)\equiv\omega_2/k_2\approx v(\omega_3)\equiv\omega_3/k_3$, which is a good approximation if $v(\omega)$ varies slowly in the range of

$[\omega_2,\omega_3]$, a small frequency interval due to the fact that $\omega_2 \sim \omega_{20}$, $\omega_3 \sim \omega_{30}$, and $(\omega_{30}-\omega_{20})/\omega_{30} \ll 1$ for the system at hand.

The magnetic susceptibilities χ_j $(j=2,3)$ can be expressed in terms of the probability amplitudes $C_j(t)$ and $C_0(t)$ by the formulas[16]

$$\chi_2 = \frac{g\mu_B N\langle 0|\hat{S}_x|2\rangle C_2C_0^*}{H_2} = \frac{N(g\mu_B|\langle 0|\hat{S}_x|2\rangle|)^2}{2\hbar}\frac{C_2C_0^*}{\Omega_2},$$

$$\chi_3 = \frac{g\mu_B N\langle 0|\hat{S}_y|3\rangle C_3C_0^*}{H_3} = \frac{N(g\mu_B|\langle 0|\hat{S}_y|3\rangle|)^2}{2\hbar}\frac{C_3C_0^*}{\Omega_3}, \quad (6)$$

where N is the number of molecules per unit volume.

Using Eqs. (3) and (6), we can set Eq. (5) into the following form:

$$\left(\frac{\partial}{\partial z} + \frac{1}{v}\frac{\partial}{\partial t}\right)\Omega_j = i\eta_j C_j C_0^*, \quad j=2,3, \quad (7)$$

where $\eta_2 = N\pi\omega_2(g\mu_B|\langle 0|\hat{S}_x|2\rangle|)^2/\hbar v$ and $\eta_3 = N\pi\omega_3 \times (g\mu_B|\langle 0|\hat{S}_y|3\rangle|)^2/\hbar v$. This set of equations, together with Eq. (4), describes the propagation of pulsed electromagnetic waves 2 and 3 in the system with the Rabi frequencies Ω_j $(j=2,3)$ characterizing the corresponding wave magnetic fields.

III. SOLUTIONS AND DISCUSSIONS

In this section, we solve differential Eqs. (4) and (7) under the initial conditions $C_j(t=0)=\delta_{j0}$, i.e., the molecules are all in their ground state $|0\rangle$ at $t=0$, and the boundary conditions where weak-pulsed electromagnetic wave 2 is illuminated at the boundary $z=0$ while pulsed FWM electromagnetic wave 3 is not injected at the boundary $z=0$ and it is only generated efficiently via the four-wave mixing within the system of molecular magnets. It is noted that the two continuous electromagnetic waves (1 and 4) are always on and, hence, Ω_1 and Ω_4 are time- and space-independent constants. Besides, throughout this section, we shall make the nondepleted ground state approximation $[C_0(t) \simeq 1]$ which is always adapted in describing the phenomenon of the EIT and the EIT-related multiwave mixing and soliton phenomena.[18–40,43–45]

A. Analytical solution for four-wave mixing

In order to obtain the analytical solutions, we use the Fourier transform technique with respect to the time t. Taking the Fourier transform of Eqs. (4) and (7) and using the nondepleted ground state approximation $(A_0 \simeq 1)$, we obtain

$$(\omega + \Delta_1 + i\gamma_1)\beta_1 + \Omega_4^*\beta_3 + \Omega_1^*\beta_2 = 0, \quad (8a)$$

$$(\omega + \Delta_2 + i\gamma_2)\beta_2 + \Omega_1\beta_1 = -\Lambda_2, \quad (8b)$$

$$(\omega + \Delta_3 + i\gamma_3)\beta_3 + \Omega_4\beta_1 = -\Lambda_3, \quad (8c)$$

$$\frac{\partial\Lambda_j}{\partial z} - i\frac{\omega}{v}\Lambda_j = i\eta_j\beta_j, \quad j=2,3, \quad (9)$$

where β_k and Λ_j are the Fourier transforms of C_k and Ω_j $(j=2,3;\ k=1,2,3)$, respectively, and ω is the Fourier variable.

The solution to Eq. (8) is

$$\beta_1 = -\frac{(\omega + \Delta_3 + i\gamma_3)\Omega_1^*\Lambda_2 + (\omega + \Delta_2 + i\gamma_2)\Omega_4^*\Lambda_3}{S(\omega)}, \quad (10a)$$

$$\beta_2 = -\frac{S_2(\omega)}{S(\omega)}\Lambda_2 + \frac{\Omega_1\Omega_4^*}{S(\omega)}\Lambda_3, \quad \beta_3 = \frac{\Omega_1^*\Omega_4}{S(\omega)}\Lambda_2 - \frac{S_3(\omega)}{S(\omega)}\Lambda_3, \quad (10b)$$

with

$$S(\omega) = |\Omega_1|^2(\omega + \Delta_3 + i\gamma_3) + |\Omega_4|^2(\omega + \Delta_2 + i\gamma_2) - (\omega + \Delta_1 + i\gamma_1)(\omega + \Delta_2 + i\gamma_2)(\omega + \Delta_3 + i\gamma_3), \quad (11a)$$

$$S_2(\omega) = |\Omega_4|^2 - (\omega + \Delta_1 + i\gamma_1)(\omega + \Delta_3 + i\gamma_3), \quad (11b)$$

$$S_3(\omega) = |\Omega_1|^2 - (\omega + \Delta_1 + i\gamma_1)(\omega + \Delta_2 + i\gamma_2). \quad (11c)$$

Substituting Eq. (10b) into Eq. (9) and making use of the initial condition for the FWM-generated field, i.e., $\Lambda_3(0,\omega)=0$, we obtain

$$\Lambda_2(z,\omega) = \frac{\Lambda_2(0,\omega)[U_+e^{izK_-} - U_-e^{izK_+}]}{U_+ - U_-}, \quad (12a)$$

$$\Lambda_3(z,\omega) = \frac{U_+U_-\Lambda_2(0,\omega)[e^{izK_-} - e^{izK_+}]}{U_+ - U_-}, \quad (12b)$$

where $K_\pm = K_\pm(\omega)$, $U_\pm = U_\pm(\omega)$, and

$$K_\pm(\omega) = \frac{\omega}{v} + \frac{-[\eta_3 S_3(\omega) + \eta_2 S_2(\omega)] \pm \sqrt{G(\omega)}}{2S(\omega)} = K_\pm(0) + K_\pm^{(1)}\omega + \mathcal{O}(\omega^2), \quad (13a)$$

$$U_\pm(\omega) = \frac{\eta_2 S_2(\omega) - \eta_3 S_3(\omega) \pm \sqrt{G(\omega)}}{2\eta_2\Omega_1\Omega_4^*} = W_\pm + \mathcal{O}(\omega), \quad W_\pm = U_\pm(0), \quad (13b)$$

with $G(\omega) = [\eta_3 S_3(\omega) - \eta_2 S_2(\omega)]^2 + 4\eta_2\eta_3|\Omega_1|^2|\Omega_4|^2$.

Figure 2 shows the generated FWM field intensity $\Lambda_3(z,\omega)$ versus the detuning Δ_3 for several different values of the decay rate γ_1. Figure 3 illustrates the absorption coefficients $\alpha_\pm = 2\,\mathrm{Im}[K_\pm]$ of the two modes characterized by the subscripts ± in Eqs. (12) and (13) versus the Rabi frequency Ω_4.

These two figures clearly demonstrate how the generated FWM field intensity is affected by the detunings and the decay rate or dephasing rate γ_1 originated from[50] those broadenings such as collisions, disorder, and the inhomoge-

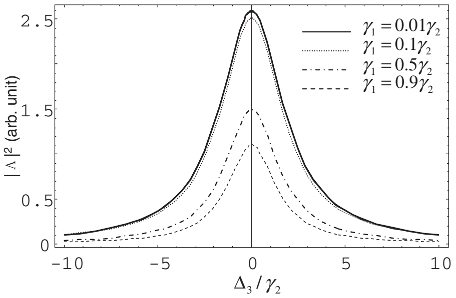

FIG. 2. Generated FWM field intensity $|\Lambda_3|^2$ versus the detuning $\Delta_3=\omega_3-\omega_{30}$ for several different values of the decay rate γ_1. The parameter values are $\Omega_1=\Omega_4=\gamma_2$, $\eta_2=10\eta_3=0.1\gamma_2/z$, $\omega=\Delta_1=\Delta_2=\Delta_3=0$, and $\gamma_3=\gamma_2$.

neous broadening of levels due to dipolar fields and disorder. On the other hand, the limitations on the inhomogeneous broadening of levels due to dipolar fields and disorder not accounted for by the detunings and the decay rates are discussed in detail in part B of Sec. IV of Ref. 16

The analytical expressions of the pulsed fields are still complicated in order to perform inverse Fourier transform, but much physical insight can be gained by seeking their approximated inverse Fourier transform with the approximation of neglecting both $\mathcal{O}(\omega)$ terms in $U_\pm$ and $\mathcal{O}(\omega^2)$ terms in $K_\pm$. Then, it is straightforward to obtain

$$\Omega_2(z,t)=\frac{W_+\Omega_2(\xi_-)e^{izK_-(0)}-W_-\Omega_2(\xi_+)e^{izK_+(0)}}{W_+-W_-},\tag{14a}$$

$$\Omega_3(z,t)=\frac{W_+W_-[\Omega_2(\xi_-)e^{izK_-(0)}-\Omega_2(\xi_+)e^{izK_+(0)}]}{W_+-W_-},\tag{14b}$$

where $\Omega_2(t)\equiv\Omega_2(z=0,t)$ is electromagnetic field 2 at $z=0$, $\xi_\pm=t-z/V_{g\pm}$, $W_\pm=U_\pm(0)$, the group velocities $V_{g\pm}$ are determined by $1/V_{g\pm}=\mathrm{Re}[K_\pm^{(1)}]$, $\mathrm{Re}[K_\pm(0)]$ denote the phase shifts per unit length, and $2\,\mathrm{Im}[K_\pm(0)]\equiv\alpha_\pm$ are absorption coefficients.

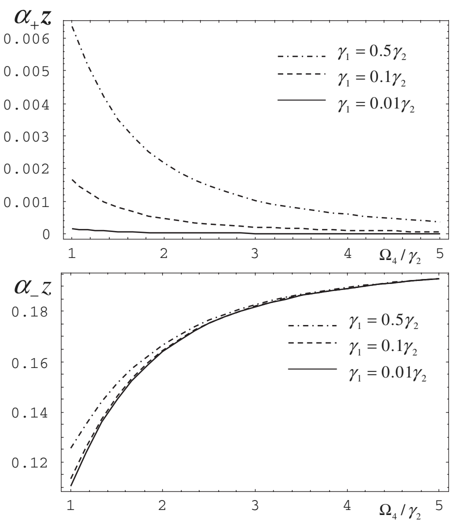

FIG. 3. Absorptions $\alpha_\pm z=2z\,\mathrm{Im}[K_\pm]$ versus the Rabi frequency Ω_4/γ_2 for several different values of the decay rate γ_1. The parameter values are $\Omega_1=\gamma_2$, $\eta_2=10\eta_3=0.1\gamma_2/z$, $\omega=\Delta_1=\Delta_2=\Delta_3=0$, and $\gamma_3=\gamma_2$.

B. Analytical solution for electromagnetically induced transparency

In this subsection, we consider the time-dependent EIT where electromagnetic waves 3 and 4 do not exist. Our purposes are threefold. First of all, we want to show that all the previous results on the EIT in molecular magnets[16] can simply be obtained by considering Eqs. (4) and (6) by taking $\Omega_3=\Omega_4=0$ because electromagnetic waves 3 and 4 do not exist in the case of the EIT.[16] Secondly, this agreement between our theory with the previous one adapting the density operator formalism[16] serves to illustrate the equivalence of the Shrödinger formalism and the density operator formalism in describing the EIT and the EIT-related phenomena in molecular magnets, although such equivalence in cold atom media has already been shown before.[20,27,29,35–39,43–45] Lastly, but no less importance, in the previous study on the EIT,[16] Shvetsov *et al.* have adapted the steady-state and continuous-wave approximations which amount to consider the situation of taking $\partial/\partial t=0$ in Eq. (4) and that Ω_1 and Ω_2 are time- and space-independent constants. Here, we want to go one step further by considering a time-dependent treatment. In this way, our theory is able to consider the pulse propagation of the probe or signal wave, i.e., electromagnetic wave 2, and to obtain the corresponding group velocity.

Performing the Fourier transform to Eq. (4) with $\Omega_3=\Omega_4=0$, we obtain

$$\beta_2=\frac{\omega+\Delta_1+i\gamma_1}{|\Omega_1|^2-(\omega+\Delta_1+i\gamma_1)(\omega+\Delta_2+i\gamma_2)}\Lambda_2.\tag{15}$$

This equation can also be obtained from Eqs. (10) and (11) simply by taking $\Lambda_3=0$ and $\Omega_4=0$.

In the steady-state approximation, by taking $\partial/\partial t=0$ in Eq. (4) again with $\Omega_3=\Omega_4=0$, we have the steady-state result

$$C_2=\frac{\Delta_1+i\gamma_1}{|\Omega_1|^2-(\Delta_1+i\gamma_1)(\Delta_2+i\gamma_2)}\Omega_2.\tag{16}$$

Once again, this equation can be simply obtained from Eq. (15) by the rule of taking $\omega=0$, $\beta_2\to C_2$, and $\Lambda_2\to\Omega_2$. This rule is equivalent to the steady-state approximation applying to Eq. (4) with $\Omega_3=\Omega_4=0$. Using Eqs. (6) and (16) with $C_0\approx1$, we then have the result

$$\chi_2=\frac{N(g\mu_B|\langle0|\hat{S}_x|2\rangle|)^2}{2\hbar}\frac{\Delta_1+i\gamma_1}{|\Omega_1|^2-(\Delta_1+i\gamma_1)(\Delta_2+i\gamma_2)},\tag{17}$$

where $\Delta_1=(\omega_2-\omega_1)-\omega_{10}$ denotes the two-photon detuning and $\Delta_2=\omega_2-\omega_{20}$ is the single-photon detuning. Obviously, this result is the same as the central result of Ref. 15 in its Eq. (20) except for a difference of a factor of 2. This difference obviously comes solely from the definition difference and does not come from the theory derivation. In our view, there should be no factor of 2 in the first line (defining the susceptibility) of Eq. (20) in Ref. 15.

Let us now discuss the time-dependent EIT by using Eq. (15) and the following equation, i.e., Eq. (7) for $j=2$ with $C_0 \approx 1$:

$$\left(\frac{\partial}{\partial z}+\frac{1}{v}\frac{\partial}{\partial t}\right)\Omega_2 = i\eta_2 C_2, \tag{18}$$

where $\eta_2 = N\pi\omega_2(g\mu_B|\langle 0|\hat{S}_x|2\rangle|)^2/\hbar v$. Performing the Fourier transform to this equation, using Eq. (15b), we readily obtain

$$\Lambda_2(z,\omega) = \Lambda_2(0,\omega)e^{iK(\omega)z}, \tag{19}$$

with

$$K(\omega) = \frac{\omega}{v} + \frac{(\omega+\Delta_1+i\gamma_1)\eta_2}{|\Omega_1|^2-(\omega+\Delta_1+i\gamma_1)(\omega+\Delta_2+i\gamma_2)}. \tag{20}$$

Again, using the approximation $K(\omega)=K(0)+K^{(1)}\omega+\mathcal{O}(\omega^2)$ and performing the inverse Fourier transform, we then have the analytical expression for the propagation of pulsed wave 2 as follows:

$$\Omega_2(z,t) = \Omega_2(t-z/V_g)e^{iK(0)z}, \tag{21}$$

where $\Omega_2(t) \equiv \Omega_2(z=0,t)$ is electromagnetic field 2 at $z=0$,

$$K(0) = \frac{(\Delta_1+i\gamma_1)\eta_2}{|\Omega_1|^2-(\Delta_1+i\gamma_1)(\Delta_2+i\gamma_2)} \equiv \Delta k + i\frac{\alpha}{2}, \tag{22}$$

and the complex group velocity $V_g = 1/K^{(1)}$ can be exactly obtained but we only give its approximate expression as follows:

$$V_g \approx \frac{v}{1+\zeta}, \quad \zeta = \frac{N\pi\omega_2(g\mu_B|\langle 0|\hat{S}_x|2\rangle|)^2}{[|\Omega_1|^2-(\Delta_1+i\gamma_1)(\Delta_2+i\gamma_2)]\hbar}, \tag{23}$$

where $\Delta_1=(\omega_2-\omega_1)-\omega_{10}$ and $\Delta_2=\omega_2-\omega_{20}$.

In Eq. (22), the two real parameters Δk and α denote the phase shift per unit length and the absorption coefficient, respectively, if the group velocity takes nearly a real value in some parameter regimes. In the two-photon resonance condition or Raman resonance condition, $\Delta_1=(\omega_2-\omega_1)-\omega_{10}=0$, and the conditions of the strong control field, $|\Omega_1|^2 \gg \gamma_1|(\Delta_2+i\gamma_2)|$ and $|\Omega_1|^2 \gg \gamma_1\eta_2$, the absorption coefficient α is very small and, hence, the probe wave or electromagnetic wave 2 can go nearly freely through an optically thick medium due to the presence of the strong control field.

C. Discussions on Doppler Effect

Before ending this section, we briefly discuss the effects of Doppler broadening due to the molecule's thermal velocity. So far, we have only considered the motionless molecules but the results can be readily generalized to those when a molecule moves with a velocity $\mathbf{V}$ by the replacement $\omega_j \rightarrow \omega_j - \mathbf{k}\cdot\mathbf{V} \equiv \omega_j - k_{jz}V_z$ (all the waves are assumed to propagate along the positive or negative z directions in this paper) or the replacements $\Delta_1=(\omega_2-\omega_1)-\omega_{10} \rightarrow (\omega_2-\omega_1)-\omega_{10}+(k_{1z}-k_{2z})V_z$ and $\Delta_j=\omega_j-\omega_{j0} \rightarrow \omega_j-\omega_{j0}-k_{jz}V_z$ ($j=2,3$). The V_z-dependent quantities thus obtained are then averaged over a given thermal velocity distribution $f(V_z)$, which results in the so-called Doppler broadening. From the above discussions, we see that the velocity-dependent effect in the two-photon detuning Δ_1 in the copropagating case $k_{1z}k_{2z}>0$ is much smaller than the one in the counterpropagating case $k_{1z}k_{2z}<0$. This difference is significant when considering $|k_{1z}| \approx |k_{2z}|$ because $|\omega 20-\omega 10|/\omega 21 \ll 1$ in the usual EIT configuration. Consequently, the velocity-dependent or the Doppler effect in the two-photon detuning Δ_1 can be usually neglected compared with the Doppler effect in the single-photon detunings if we choose the waves to propagate in the same direction.

On the other hand, in studying the EIT or the EIT-related phenomena, one can take the strong control field(s) to suppress the Doppler effect in the single-photon detunings. For instance, from Eq. (22), it is seen that in the two-photon resonance condition, $\Delta_1=0$ (or $\Delta_1 \approx 0$ when the small velocity-dependent effect is taken into account in the copropagating case), and the condition of the strong control field, $|\Omega_1|^2 \gg \gamma_1|(\Delta_2+i\gamma_2)|$, the absorption coefficient $\alpha \approx \gamma_1\eta_2/|\Omega_1|^2$ is nearly independent of the velocity-dependent effect. Therefore, we see that the EIT or the EIT-related phenomena can occur not only in the cold media but also in media of relatively high temperatures, as the experiments showed before.[41]

IV. CONCLUSIONS

In conclusion, we have analyzed the four-wave mixing in molecular magnets via electromagnetically induced transparency by a time-dependent treatment. We have explicitly obtained the analytical expressions of pulsed electromagnetic waves (2 and 3), their group velocities, the corresponding phase shifts, and absorption coefficients. We have investigated analytically the time-dependent electromagnetically induced transparency in molecular magnets as well, which goes beyond the previous results with the cw treatment. Our results may be used to produce coherent radiation of new frequencies and with certain tunability at low pump intensities in applications such as nonlinear laser spectroscopy and quantum nonlinear optics.

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