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Nonlinear optical responses due to exciton-phonon interactions in strongly coupled exciton-phonon systems

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Abstract. The excitonic nonlinear optical responses due to exciton-phonon interactions in strongly coupled exciton-phonon systems are investigated theoretically. It is shown that the influence of exciton-phonon interactions on the nonlinear optical absorptions and Kerr nonlinear coefficients is significant as the signal field frequency detuning from the exciton frequency approaches to the optical phonon frequency. How to manipulate the nonlinear optical responses by using the control fields is also presented.

PACS. 42.50.Gy Effects of atomic coherence on propagation, absorption, and amplification of light; electromagnetically induced transparency and absorption – 42.50.Hz Strong-field excitation of optical transitions in quantum systems; multiphoton processes; dynamic Stark shift – 42.25.Bs Wave propagation, transmission and absorption – 72.80.Le Polymers; organic compounds (including organic semiconductors)

1 Introduction

The control of linear and nonlinear optical properties of a material by using resonant electromagnetic fields is becoming more and more important in recent years. Electromagnetically induced transparency (EIT) are excellent technique that can be used to suppress a linear absorption of a resonant multilevel media, while keeping the nonlinear optical properties at a very high level [1–4]. Recent remarkable applications include ultraslow light pulse propagation [5–8] and light storage [9–11]. The effects of EIT has been used by Schmidt and Imamoglu [12] to devise a scheme involving four level atoms which produces a giant cross-Kerr nonlinearity with no noise. Recently, Kuang et al. [13] develop a fully quantum treatment of EIT in three-level Λ-type atoms and find that the atomic medium with EIT exhibits giant Kerr as well as higher order nonlinearities. Xiao and coauthors [14] have experimentally studied the Kerr-nonlinear index of refraction in a three-level Λ-type atomic systems for several coupling powers and found that the Kerr nonlinearity is greatly enhanced because of atomic coherence in the three-level atomic system compared with that a two-level atomic system. Many other studies of nonlinear optical effects utilizing EIT have been published elsewhere [15–18]. On the other hand, in the recent years there has been an increasing interest in the nonlinear optical properties of excitons confined in low-dimensional molecular geometries such as crystalline organic superlattices, molecular aggregates, conjugate polymers and molecular monolayers [19–32]. These organic materials have many potential applications. In particular, the ordered films of organic conjugated polymers are of strategic relevance for novel optoelectronic devices such as polymer LEDs [31]. In general these organic systems are characterized by a strong exciton-phonon interaction which often leads to large nonlinear optical phenomena. Greene et al. [20] have experimentally shown that the anomalous optical nonlinearity of polydiacetylene-toluene sulfonate (PTS) results from phonon-mediated interactions between virtual excitons. Liu et al. [29] in a recent paper discussed the effects of the detuning between the cavity field and the exciton on the radiation spectra of the high-density excitons in a quantum well. More recently, Zhu and Li [30] have predicted the occurrence of EIT due to strong exciton-phonon interactions in the strongly coupled exciton-phonon systems. In the present article, we will further investigate the influence of exciton-phonon interactions on the nonlinear optical absorption and Kerr coefficients in these strongly coupled exciton-phonon systems. How to manipulate the nonlinear optical responses by using the control fields is also discussed in detail.

2 Theory

In what follows, we consider an interacting exciton system including the coupling of exciton-phonon and external radiation fields. For the sake of simplicity, we only

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$$\chi^{(3)}(\omega_s) = \frac{i(\alpha_1 - \alpha_2\eta(\omega_s))A(\omega_s)}{\{1 + i[\delta_0 + \Delta_c + (2\alpha_1 - \alpha_2)w_0 - \alpha_2 w_0\eta(\omega_s)]\}\{1 + i[\Delta_c + (\alpha_1 - \alpha_2)w_0]\}^2}, \tag{9}$$

$$\text{with} \qquad A(\omega_s) = \frac{-i + [\Delta_c + \delta_0 + (2\alpha_1 - \alpha_2)w_0 - \alpha_2\eta(\omega_s)w_0]}{[\Delta_c + (2\alpha_1 - \alpha_2)w_0 - \alpha_2\eta(\omega_s)w_0]^2 - (\alpha_1 - \alpha_2\eta(\omega_s))^2 w_0^2 + (1 + i\delta_0)^2}, \tag{10}$$

assume the existence of single phonon mode and the linear exciton-phonon interaction. It is anticipated that this is sufficient to illustrate the main physics for the impact of strong exciton-phonon coupling on the nonlinear optical responses in the strongly coupled exciton-phonon systems, although the couplings between exciton and multimode phonons are also important. An extension of this work to the case of the multimode phonons will be presented elsewhere. Then the Hamiltonian in a rotating frame at the control field frequency ω_c reads as follows [19–21]

$$H = \hbar(\omega_{ex} - \omega_c)a^+a + \hbar\omega_{ph}b^+b + \hbar\beta a^{+2}a^2 - \hbar\lambda a^+aQ - \hbar\Omega(a^+ + a) - \mu\left(a^+E_s e^{-i\delta t} + aE_s^* e^{i\delta t}\right), \tag{1}$$

where $\hbar\omega_{ex}$ and $\hbar\omega_{ph}$ are the exciton binding and phonon energies,respectively. β is the exciton-exciton interaction constant assumed as a positive-real number, which means that the biexcitons are not stable and only excitons are presented in the systems, λ is the exciton-phonon coupling constant, $\Omega = \mu E_c/\hbar$ is the Rabi frequency of the control field, E_c is the slowly varying envelope of the control field, μ is the electric dipole moment of the exciton, assumed to be real, a^+a and b^+b are the exciton and phonon populations, respectively. Further, $Q = b^+ + b$ is the phonon amplitude, E_s is the slowly varying envelope of the signal field, and a and b are the exciton coherence and phonon annihilation operators, respectively. $\delta = \omega_s - \omega_c$ is the detuning of the signal and the control field, ω_s is the frequency of the signal field and ω_c is the frequency of the control field. It is shown by Bosma et al. [32] that for the strongly localized excitons in strongly coupled exciton-phonon systems, the linear and nonlinear optical response can be modeled according to the single exciton and its coupling to the phonons, so the polariton effects due to the wavevector dependence of the exciton are negligible in this article.

The temporal evolution of the exciton coherence a and the phonon amplitude Q are determined by the Heisenberg equation of motion, and are given by

$$\frac{da}{dt} = -i\Delta a - 2i\beta a^+aa + i\lambda Qa + i\Omega + i\frac{\mu E_s}{\hbar}e^{-i\delta t} \tag{2}$$

$$\frac{d^2Q}{dt^2} + \omega_{ph}^2 Q = 2\omega_{ph}\lambda a^+a, \tag{3}$$

where $\Delta = \omega_{ex} - \omega_c$. In what follows we ignore the quantum properties of a and Q [19–21], then the semiclassical equation for a and Q will be

$$\frac{da}{dt} = -(i\Delta + \Gamma)a - 2i\beta a^+aa + i\lambda Qa + i\Omega + i\frac{\mu E_s}{\hbar}e^{-i\delta t} \tag{4}$$

$$\frac{d^2Q}{dt^2} + \gamma_{ph}\frac{dQ}{dt} + \omega_{ph}^2 Q = 2\omega_{ph}\lambda a^+a, \tag{5}$$

where Γ and γ_{ph} are phenomenological exciton dephasing and phonon decay rates, respectively.

In order to solve equations (4) and (5) we make the ansatz

$$a(t) = a_0 + a_+e^{-i\delta t} + a_-e^{i\delta t}, \tag{6}$$

$$Q(t) = Q_0 + Q_+e^{-i\delta t} + Q_-e^{i\delta t}. \tag{7}$$

On substituting (6) and (7) in (4) and (5) and on working to the lowest order in E_s but to all orders in Ω, we can obtain a_- in the steady state and then yield the nonlinear optical susceptibility as follows [33]

$$\chi_{eff}^{(3)}(\omega_s) = \frac{N\mu a_-}{3E_c^2E_s^*} = \frac{N\mu^4}{3\hbar^3\Gamma^3}\chi^{(3)}(\omega_s), \tag{8}$$

where N is the number density of excitons and the dimensionless nonlinear optical susceptibility

see equations (9) and (10) above

where $\Delta_c = \Delta/\Gamma$, $\delta_0 = \delta/\Gamma$, $w_0 = |a_0|^2$, $\alpha_1 = 2\beta/\Gamma$, $\alpha_2 = 2\lambda^2/(\omega_{ph}\Gamma)$, $\omega_{ph0} = \omega_{ph}/\Gamma$, $\gamma_{ph0} = \gamma_{ph}/\Gamma$ and the auxiliary function

$$\eta(\omega_s) = \frac{\omega_{ph0}^2}{\omega_{ph0}^2 - \delta_0^2 + i\delta_0\gamma_{ph0}}. \tag{11}$$

The population w_0 of the exciton is determined by the cubic equation

$$w_0\{1 + [\Delta_c + (\alpha_1 - \alpha_2)w_0]^2\} = \Omega_c^2, \tag{12}$$

where $\Omega_c = \Omega/\Gamma$. The cubic equation (12) has either a single or three real roots. The latter case just corresponds to the intrinsic optical bistability which arises from exciton-exciton interactions and exciton-phonon interactions. From the nonlinear optical susceptibility $\chi^{(3)}(\omega_s)$ we can obtain the nonlinear optical absorptions ($\mathrm{Im}\chi^{(3)}(\omega_s)$) and the Kerr coefficients ($\mathrm{Re}\chi^{(3)}(\omega_s)$) of strongly coupled exciton-phonon systems as functions of the detuning $\Delta_s = (\omega_s - \omega_{ex})/\Gamma$ and the other parameters. In our calculations very strong exciton-phonon coupling is assumed, it is so strong that its effective ac-Stark

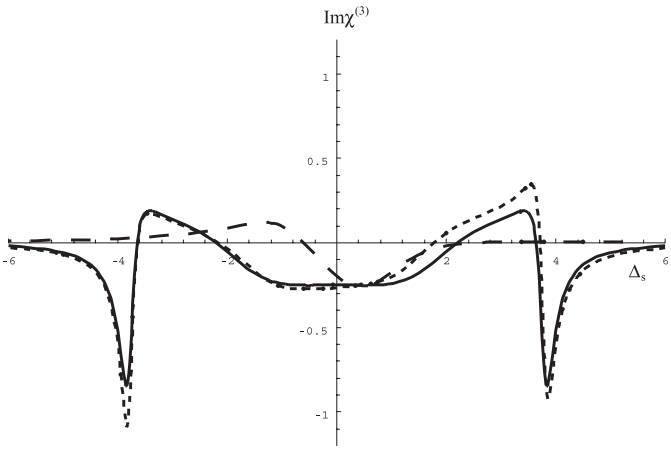

Fig. 1. The dimensionless nonlinear optical absorption as a function of the detuning Δ_s for $\alpha_1 = 0.5$ and $\alpha_2 = 2$ (dotted curve), $\alpha_1 = 0$ and $\alpha_2 = 2$ (full curve), $\alpha_1 = 0.5$ and $\alpha_2 = 0$ (broken curve). The parameters used are $\Omega_c^2 = 1$, $\Delta_c = 0$, $\omega_{ph0} = 4$ and $\gamma_{ph0} = 0.04$.

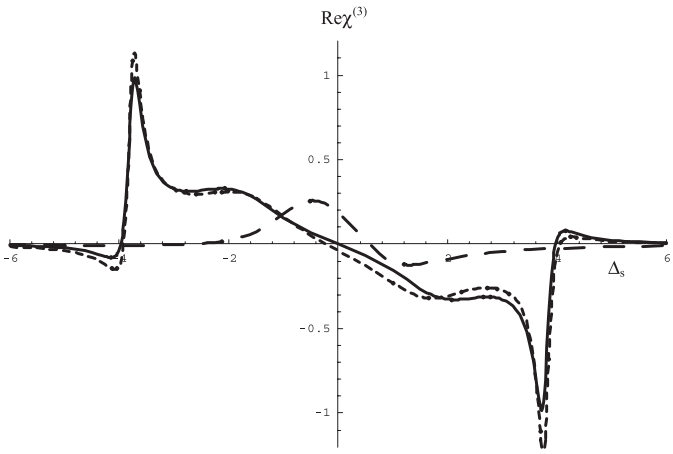

Fig. 2. The dimensionless Kerr coefficient as a function of the detuning Δ_s for $\alpha_1 = 0.5$ and $\alpha_2 = 2$ (dotted curve), $\alpha_1 = 0$ and $\alpha_2 = 2$ (full curve), $\alpha_1 = 0.5$ and $\alpha_2 = 0$ (broken curve). The other parameters used are the same as in Figure 1.

shift is comparable to the exciton linewidth, even when a detuning equal to the phonon frequency is assumed. However, in the experiment [20] Greene et al. observed the distinct nonlinear optical spectra in polydiacetylene-toluene sulfonate (PTS) films, although the exciton-phonon coupling ($\lambda = 0.1$ eV) is so strong in this real PTS material. Therefore in the following calculations we also choose PTS as an example, for which $\omega_{ph} = 0.2$ eV, $\lambda = 0.1$ eV, $\gamma_{ph} = 2$ meV, $\omega_{ex} = 2$ eV and $\Gamma = 50$ meV [20], and then $\alpha_2 = 2$, $\omega_{ph0} = 4$ and $\gamma_{ph0} = 0.04$. But there are no experimental values for the exciton-exciton interaction constant β, so without loss of generality we assume $\alpha_1 = 2\beta/\Gamma = 0.5$.

3 Numerical results and discussion

Figures 1 and 2 show the nonlinear optical absorption ($\mathrm{Im}\chi^{(3)}$) and the Kerr coefficient ($\mathrm{Re}\chi^{(3)}$), respectively, as a function of the detuning $\Delta_s = (\omega_s - \omega_{ex})/\Gamma$ for the case $\Omega_c^2 = 1$ and $\Delta_c = 0$. As the detuning of the signal field approaches to the optical phonon frequency and exciton-phonon interaction is considered, the nonlinear optical absorptions and Kerr coefficients are greatly enhanced (as shown in full and dotted curves of Figs. 1 and 2). However when the exciton-phonon interactions are neglected, the nonlinear optical responses are reduced greatly and nearly become zero (the broken curve), so the influence of the exciton-phonon interaction on the nonlinear optical responses is significant in the strongly coupled exciton-phonon systems, especially for $\Delta_s = \omega_{ph}$. Figures 3 and 4 illustrate the nonlinear absorption and Kerr coefficient, respectively, for various detunings (Δ_c) of the control field from exciton frequency as a function of Δ_s for $\Omega_c^2 = 1$. We see from the figures that varying the detuning of control field from the exciton frequency, one can obtain the different nonlinear absorptions and Kerr coefficients.

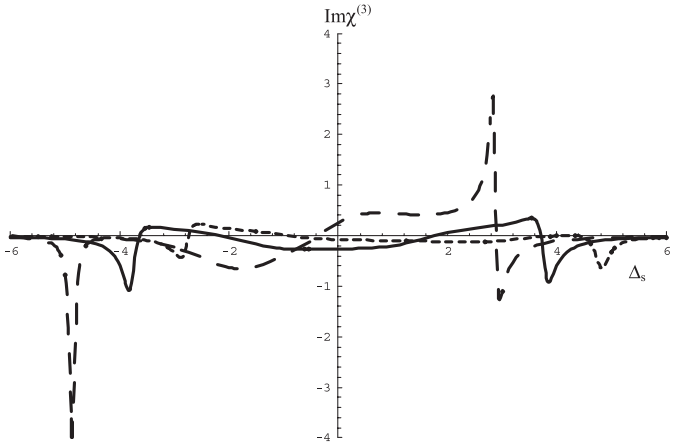

Fig. 3. The dimensionless nonlinear optical absorption as a function of the detuning Δ_s for $\Delta_c = 0$ (full curve), $\Delta_c = 1$ (broken curve) and $\Delta_c = -1$ (dotted curve). The parameters used are $\Omega_c^2 = 1$, $\alpha_1 = 0.5$, $\alpha_2 = 2$, $\omega_{ph0} = 4$ and $\gamma_{ph0} = 0.04$.

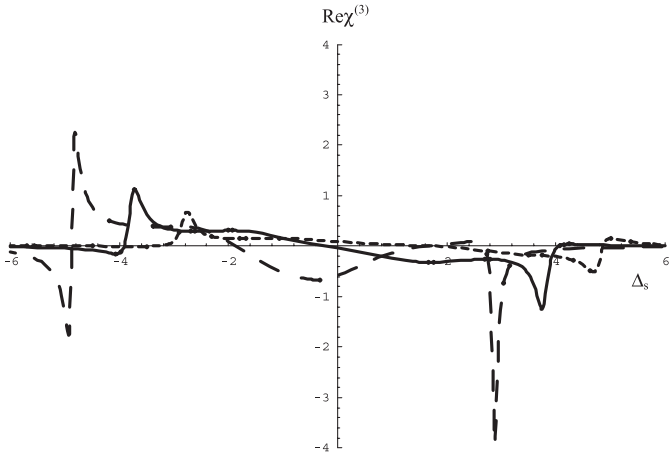

Fig. 4. The dimensionless Kerr coefficient as a function of the detuning Δ_s for $\Delta_c = 0$ (full curve), $\Delta_c = 1$ (broken curve) and $\Delta_c = -1$ (dotted curve). The other parameters used are the same as in Figure 3.

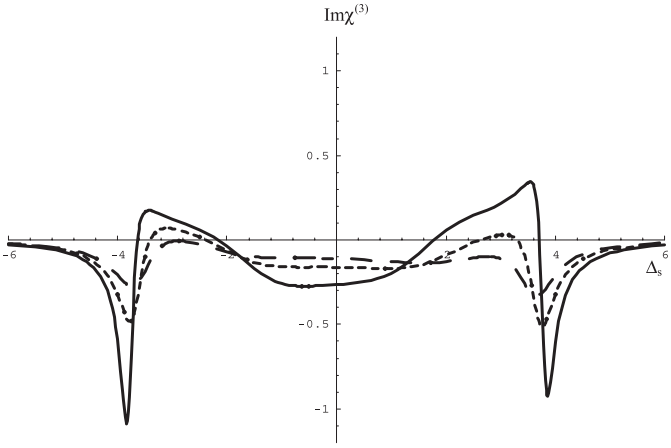

Fig. 5. The dimensionless nonlinear optical absorption as a function of the detuning Δ_s for $\Omega_c^2 = 1$ (full curve), $\Omega_c^2 = 1.5$ (dotted curve) and $\Omega_c^2 = 2$ (broken curve). The parameters used are $\Delta_c = 0$, $\alpha_1 = 0.5$, $\alpha_2 = 2$, $\omega_{ph0} = 4$ and $\gamma_{ph0} = 0.04$.

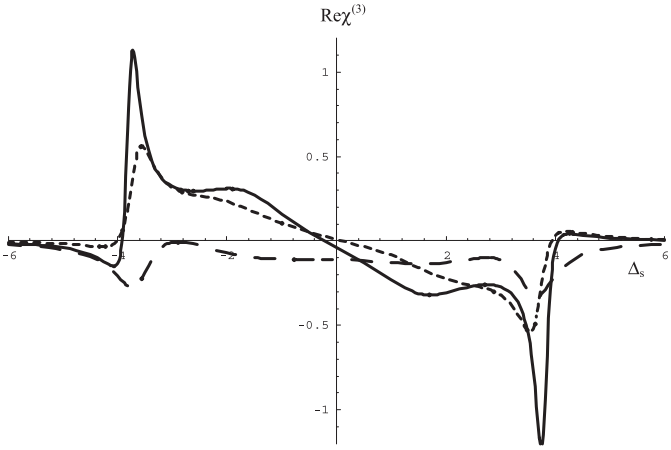

Fig. 6. The dimensionless Kerr coefficient as a function of the detuning Δ_s for $\Omega_c^2 = 1$ (full curve), $\Omega_c^2 = 1.5$ (dotted curve) and $\Omega_c^2 = 2$ (broken curve). The other parameters used are the same as in Figure 5.

The nonlinear optical absorptions and Kerr coefficients as a function of Δ_s for different Rabi frequencies of the control field are shown in Figures 5 and 6, respectively. The results also present that modification of the Rabi frequency of the control field can alter the nonlinear optical absorption and Kerr coefficients significantly. As a result, the control field can obviously manipulate the nonlinear optical responses of the strongly coupled exciton-phonon systems.

4 Conclusions

In summary, we have studied the excitonic nonlinear optical responses due to the strong exciton-phonon interactions in strongly coupled exciton-phonon systems. It is shown that the influence of the exciton-phonon interaction on the nonlinear optical effects is significant in the exciton systems with strong exciton-phonon interactions. The results also present that the nonlinear optical absorptions and the Kerr coefficients are greatly enhanced as the signal field frequency detuning from the exciton frequency approaches to the optical phonon frequency. Further we show that the control fields can manipulate the nonlinear optical response of the systems by tuning the frequency of the control field from the exciton frequency or varying the control field power. It should be noted that the phonons are treated as a single mode in this paper, but if the spread of phonon energies is larger than its inverse lifetime, then this broadening must be taken into account and will significantly reduce the nonlinear optical responses, we will treat this case in a forthcoming paper. Finally, we hope that this work will stimulate more theoretical and experimental works which will be helpful for a better understanding of the strongly coupled exciton-phonon systems.

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