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Transient Dynamics in Excitonic Optical Bistability in Polymers

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Transient dynamics in excitonic optical bistability in polymers

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Abstract

Transient behavior in optical bistability mediated by phonon mode and virtual exciton coupling is studied for an electron-phonon coupling system such as a polydiacetylene-toluene-sulfonate. Numerical calculations are used to analyze the evolution between bistable states, and novel switching behavior and Rabi oscillations are found.

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1. Introduction

Since its first observation in sodium vapor in 1976,[1] optical bistability has become an important phenomenon in optics, and there has been much interest in the promise of optical bistability as a mean of producing fast all-optical switching devices. Therefore, good nonlinear optical materials and more efficient device structures have been sought for practical applications, and attention is being paid to reducing sizes and decreasing the switching time and operating power for such devices.

Optical bistability means that there are two stable output optical states for a given input optical state. In general, optical bistability occurs by means of the joint effects of the nonlinear optical response of the material and the external feedback typically provided by an optical resonator. However, there are also some systems exhibiting optical bistability without the need for external feedback. For example, absorption of light tuned just below the band gap of some semiconductors can lead to increasing absorption optical bistability;[2] reflection at an interface between a linear and nonlinear medium[3] or transmission through a nonlinear thin film[4] have been proposed to exhibit optical bistability; and mutually induced self-focusing by interacting laser beams[5] and light beams interacting through four-wave mixing[6] have been predicted and observed, respectively, to display optical bistability. Very recently, we have found surface-induced optical bistability of polymer chains near a metal surface,[7] accompanied by reduced vacuum field fluctuations. In

fact, certain classes of organic molecules have been found to exhibit extremely large optical nonlinearities with fast response.[8] Besides, organic materials offer other attractive advantages for trying to alter the molecular structure to optimize the nonlinear optical and bulk physical properties. An example is the second-order hyperpolarizability: here the dipole of the molecular structure of a polymer is created by substituting electron-donating and electron-accepting groups at either end of the conjugated length of the molecule, where a proper choice of the donor and thus freezing in the alignment can lead to a large second-order hyperpolarizability. This flexibility in structure makes organic materials well suited for fabrication and device engineering.

Among the polymers, polydiacetylene (PDA) is a good candidate for future applications because of its giant optical nonlinearity and small transmission loss. There have been reports[9] about the observation of optical bistability in PDA quasi-waveguides. In the past few years, there has also been much interest in the nonresonant nonlinear optical responses of PDA-toluene sulfonate (PTS) excited by laser fields tuned well below the absorption edge.[10-15] Phonon-mediated bleaching[11] and excitonic Stark shift[12] have been observed, and optical nutation,[13] dispersive optical bistability[14] and splitting in pump-probe spectra[15] have been predicted.

In this paper, we shall theoretically analyze the transient dynamics of a PTS material irradiated by a laser field. Special attention will be paid to optical bistability and how the system

switches from one state to the other. In contrast to Ref. 14 where external feedback by an optical cavity was examined, here we shall consider another kind of bistability without external feedback. While studies of optical bistability are usually focused on the behavior of the steady-state output field intensity as a function of input intensity,[2] here we shall study the transient behavior of the system, through which we can extract knowledge about how the output intensity grows from zero and how fast the system evolves from one state to another,[16] and about possible unstable states and optical nutation. Our work is also close to experimental situations, where measurements are usually made in the transient time regime.

In Sec. II, we shall describe our model, where we treat excitons and phonons as damped oscillators and use Dekker's quantization procedure for dissipative systems[17] to obtain the equations of motion for the excitons and the phonons. These equations will be solved numerically for the scattered field intensity by virtual excitons, and in Sec. III we shall display the numerical results and provide a discussion and some physical conclusions.

II. Model

It is known that there are elementary excitations of excitons and phonons in PTS[18] and only a few phonon modes coupled very strongly to the excitons. We consider a light beam with its frequency near the exciton resonance falling on a PTS material, and

thus the excitons interact with both the light and the lattice vibrations. The light can excite the system to creat excitons and, inversely, the electrons and holes composing excitons can recombine to create radiation. However, because of the much lower frequencies of phonon modes, the number of excitons remains unchanged in their coupling with phonon modes, namely, the phonons can only play the role of mediation.[11-14]

It is known that both the excitons and the phonons have finite lifetimes. We may model them here as damped oscillators. For the PTS system exposed to an external electric field with frequency ω and amplitude $\underline{E}$, we can write a nonHermitian Hamiltonian for this system[7,15] as

$$H = (\omega_x - i\gamma_x)a^\dagger a + \sum_i (\omega_i - i\gamma_i) b_i^\dagger b_i + \sum_i \lambda_i\, a^\dagger a (b_i^\dagger + b_i) - (\mu_x^* a^\dagger E e^{-i\omega_L t} + \mu_x a E^* e^{i\omega_L t}) \quad , \tag{1}$$

where $a^\dagger$ (a) and $b_i^\dagger$ (b_i) stand for the creation (annihilation) operators for the exciton and i-th phonon modes with corresponding frequencies ω_x and ω_i, respectively, γ_x and γ_i are the damping rates for the exciton and i-th phonon mode, respectively, λ_i is the coupling constant for the interaction between the exciton and i-th phonon mode, and μ_x is the component of the dipole matrix element for the exciton in the $\vec{E}$-field direction. As in Refs. 11-15, we neglect the momentum dependence of the exciton. With this

nonHermitian Hamiltonian characterizing energy dissipation, we can use $a^\dagger$, a, $b_i^\dagger$ and b_i as canonical variables for this dissipative PTS system and extend Dekker's quantization procedure[17] for damped oscillators to write down the quantum Liouville equation for the density operator ρ:

$$\dot{\rho} = -i[a^\dagger, [a,H]\rho] + i[\rho[H^\dagger,a^\dagger],a] - i\sum_i [b_i^\dagger,[b_i,H]\rho] - i\sum_i [\rho[H^\dagger,b_i^\dagger],b_i] \quad . \tag{2}$$

In this paper, we are interested neither in any quantity that is sensitive to the *quantum number counting*, nor in any thermal noise present in the driving field so that quantum fluctuations can generally be neglected in our consideration.[2] Thus, instead of the operators, we deal with their mean values[2,14] $\alpha = \langle a \rangle$ and $\beta_i = \langle b_i \rangle$. Such replacement simply means that we are taking the semiclassical approximation.[19] It has already been shown[14] that for excitonic optical bistability in the PTS system, there is no qualitative change in physics if we only consider a single phonon mode coupled most strongly to the excitons. Therefore, for simplicity we consider here only one phonon mode and thus delete the subscript i on β, ω, γ and λ in the following discussion. With the equation of motion for the density operator (2), we obtain the following equations of motion for the mean values of the dynamical variables in question:

$$\dot{\alpha} = -(i\omega_x+\gamma_x)\alpha - i\lambda(\beta+\beta^*)\alpha + i\mu_x^* E e^{-i\omega_L t} \quad , \tag{3a}$$

$$\dot{\beta} = -(i\omega+\gamma)\beta - i\lambda|\alpha|^2 \quad . \tag{3b}$$

For convenience, in what follows we use the rotating frame. Namely, we replace α in Eqs. (3) by $\alpha e^{-i\omega_L t}$ to obtain

$$\dot{\alpha} = -(i\Delta+\gamma_x)\alpha - i\lambda(\beta+\beta^*)\alpha + i\Omega \quad , \tag{4a}$$

$$\dot{\beta} = -(i\omega+\gamma)\beta - i\lambda|\alpha|^2 \quad , \tag{4b}$$

where we have defined the detuning $\Delta = \omega_x - \omega_L$ and the Rabi frequency $\Omega = \mu_x^* E$.

When the external laser field with its frequency near the exciton resonance falls on the PTS material, the radiation field is scattered by the excitons. In the scattering region where the incident external field vanishes, the relation between the positive-frequency part of the scattered field and the excitonic dipole moment can be written as[20]

$$E_{sc}^{(+)}(\vec{r}) = \psi(\vec{r})a(t-\frac{r}{c}) \quad , \tag{5}$$

where $\psi(\vec{r}) = -\frac{\omega_x^2}{4\pi c^2 r^3}(\vec{\mu}_x \times \vec{r}) \times \vec{r}$, and $\vec{\mu}_x a$ is the excitonic dipole moment. Hence, the mean value of the dimensionless scattered field intensity is given by

$$I_{sc}(t) = |\langle E_{sc}^{(+)}(\vec{r},t)\rangle|^2/\psi^2(\vec{r}) = |\alpha|^2 \quad . \tag{6}$$

In the steady state, it is easy to find, by setting the terms on the left-hand side of Eqs. (4) equal to zero, that this scattered intensity, $I_{sc}(t\to\infty)$, is determined by the cubic equation

$$\lambda_p^2 I_{sc}^3 - 2\Delta\lambda_p I_{sc}^2 + (\Delta^2+\gamma_x^2)I_{sc} - I_{in} = 0 \quad , \tag{7}$$

where we have defined the incident field intensity $I_{in} = |\Omega|^2$, and $\lambda_p = 2\lambda^2\omega/(\omega^2+\gamma^2)$. However, in the transient regime, we have to solve the nonlinear Eqs. (4) numerically.

III. Results and discussion

In this paper we use the following parameters for PTS:[11,14] $\gamma_x = 0.05$ eV, $\gamma = 0.002$ eV, $\lambda = 0.1$ eV and $\omega = 0.258$ eV. For the first step, it is helpful to solve for the steady-state solution using Eq. (7). Through analyzing this equation, we have found that

cnly when the incident field is tuned sufficiently below the exciton resonance, say, $\omega_x - \omega_L > 0.4\omega$, does there exist optical bistability, indicating that the virtual exciton is essential for this bistability. We have also found that the phonon-exciton coupling is indispensible to this bistability. This is similar to the case where PTS is placed in an optical cavity.[14]

Figure 1(a) is an example showing the scattered field intensity plotted vs the incident field intensity when the system is in steady state and $\Delta = 0.5\omega$. Optical bistability is clearly shown in this figure. Generally speaking, optical bistability occurs only when the system exhibits nonlinear responses to the external field and a feedback of the field or other agent exists. In semiconductors, it is well known that[10,11] virtual excitons play almost the same role as do real excitons in response to external fields. In the present case, as the incident field intensity changes, the dynamics of virtual excitons changes accordingly, leading to a subsequent change in the phonon mode. As a consequence, the exciton-phonon coupling changes the properties of the virtual excitons, resulting in a change in the scattered light. This internal process of "feedback" provided by the phonon mode permits possible multivalued solutions to the output intensity of the nonlinear optical scattering problem. This means that within a certain range, there can be three distinct scattered intensities for each incident intensity. We have found, through numerical simulation, that the middle solution is unstable, and hence only the upper and lower solutions can be reached. As observed in Fig.

1(a), when I_{in} increases continuously from zero, I_{sc} increases continuously until it reaches point b where it becomes unstable. Further increase of I_{in} leads to an abrupt jump of I_{sc} to the upper branch. Similarly, when I_{in} decreases continuously, I_{sc} jumps to the lower branch at point a where the solution is unstable.

Through numerical calculations of Eqs. (4) and (6), we have studied the time evolution of the scattered intensity after a sudden intensity change or sudden switching on of the incident field. Figures 1(b), 1(c) and 2(a)-(c) show some interesting results. Figure 1(b) describes the situation that when I_{sc} reaches steady state (point a) with $I_{in} = 1.58\gamma_x^2$, then we suddenly increase I_{in} from $1.58\gamma_x^2$ to $2.29\gamma_x^2$. How I_{sc} evolves from the lower state (point a) to the upper state (point c) is clearly shown. We see that the switching time is approximately one order of magnitude longer than the lifetime of the virtual exciton but the same order of magnitude as the lifetime of the phonon mode. Comparing Fig. 1(a) with 1(b), one can find that in the course of I_{sc} going from the lower state to the upper one (from point a to c), it goes along the line of the lower branch, and then gradually goes upward, and finally, after oscillating a bit, reaches point c. Figure 1(c) describes the reverse case of Fig. 1(b). By comparing Figs. 1(b) and 1(c), one can see that I_{sc} goes along a different path from point c to a. By comparing Figs. 1(a) and 1(c), one can see that I_{sc} goes along the upper branch until it reaches point d, then it goes gradually down to point a. Therefore, interestingly enough, through our study in the transient regime, we can obtain detailed

information about not only the evolution of I_{sc} between bistable states, but also how it completes a hysteresis loop.

In addition to the switching behavior of the scattered field intensity I_{sc}, we have also studied how I_{sc} builds up from zero through a switching-on by an incident field at intial time $t = 0$. Some numerical results calculated from Eqs. (4) and (6) are shown in Fig. 2, in which we have chosen three different input intensities in the region that bistability occurs. For fixed detuning Δ there is a corresponding threshold, and smaller Δ leads to lower threshold as long as bistability occurs. If I_{in} lies below the threshold, I_{sc} eventually evolves to reach the lower state. Otherwise, I_{sc} eventually reaches the upper state. In Fig. 2 we choose $\Delta = 0.5\omega$, and the subsequent threshold for I_{in} is found to be approximately $2.21\gamma_x^2$. By comparing Fig. 1(a) with Fig. 2, we see that in Figs. 2(a) and (b) where the I_{in}'s are smaller than this threshold value, I_{sc} goes into the lower branch, while in Fig. 2(c) where I_{in} is beyond the threshold, I_{sc} goes into the upper branch. From Fig. 2 we can also see Rabi oscillations. However, these oscillations are modulated, i.e., the centers of oscillation change with respect to time. When I_{in} is below the threshold, I_{sc} first goes up then goes down to reach the lower state [see Figs. 2(a) and (b)]. By contrast, when I_{in} is above the threshold, I_{sc} first reaches the lower state then goes up to reach the upper state [see Fig. 2(c)]. In addition, through numerical analysis, we have found that no matter what initial condition is used, I_{sc} cannot evolve into the middle branch [see the line between points b and d in Fig.

1(a)]. So, we can conclude that the middle or third solution of I_{sc} is not stable. Meanwhile, the peaks in Figs. 2(a) and (b) show the possibility for I_{sc} to temporarily stay in the middle state, which means that the middle state can only be reached temporarily.

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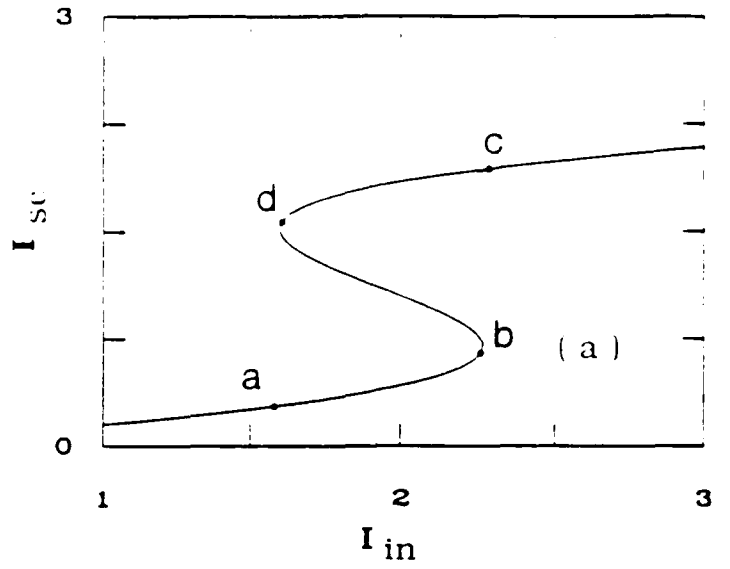
Figure Captions

1. (a) Scattered field intensity I_{sc} vs driving field intensity I_{in} (in the unit of γ_x^2) for steady-state operation. (b) Transition of I_{sc} from the lower state, corresponding to $I_{in} = 1.58\gamma_x^2$, to the upper state, corresponding to $I_{in} = 2.29\gamma_x^2$. (c) Transition of I_{sc} from the upper state corresonding to $I_{in} = 2.29\gamma_x^2$, to the lower state, corresponding to $I_{in} = 1.58\gamma_x^2$.

2. Time (t) evolution of the scattered field intensity I_{sc} for different driving intensities, where (a) $I_{in} = 1.93\gamma_x^2$, (b) $I_{in} = 2.20\gamma_x^2$ and (c) $I_{in} = 2.21\gamma_x^2$.

Fig. 1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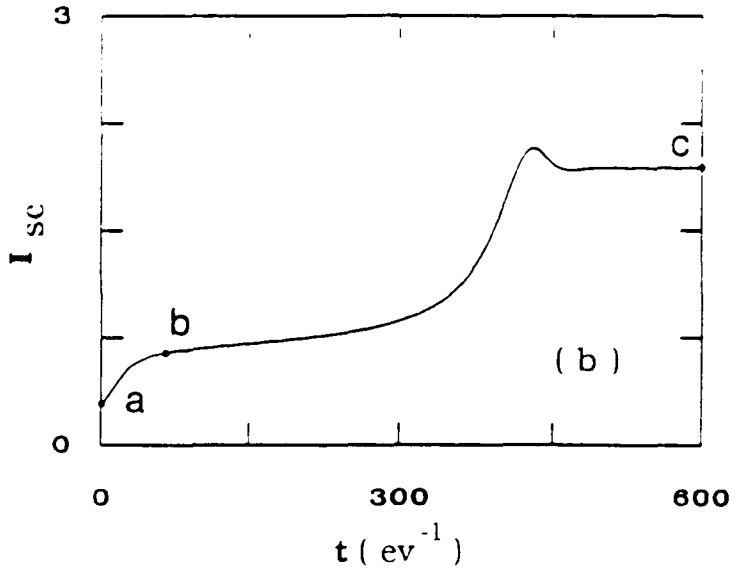
I_{sc}
I_{in}
a
b
c
d
(a)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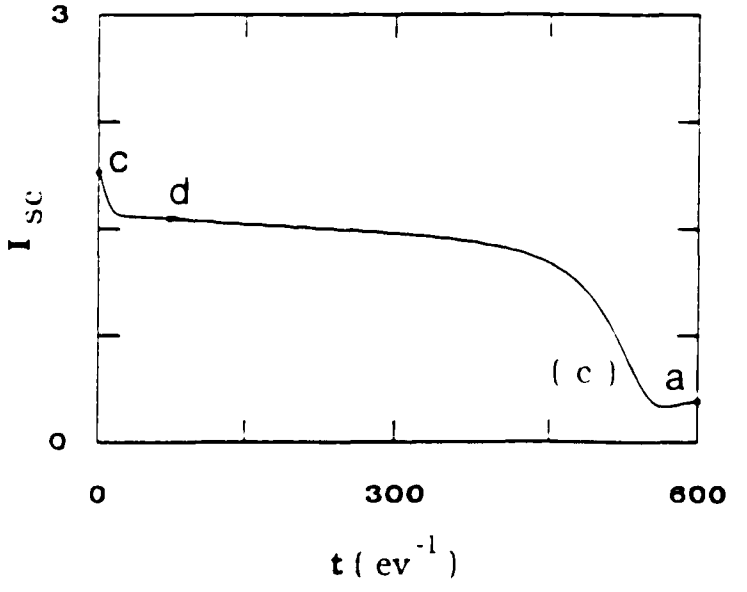
(b)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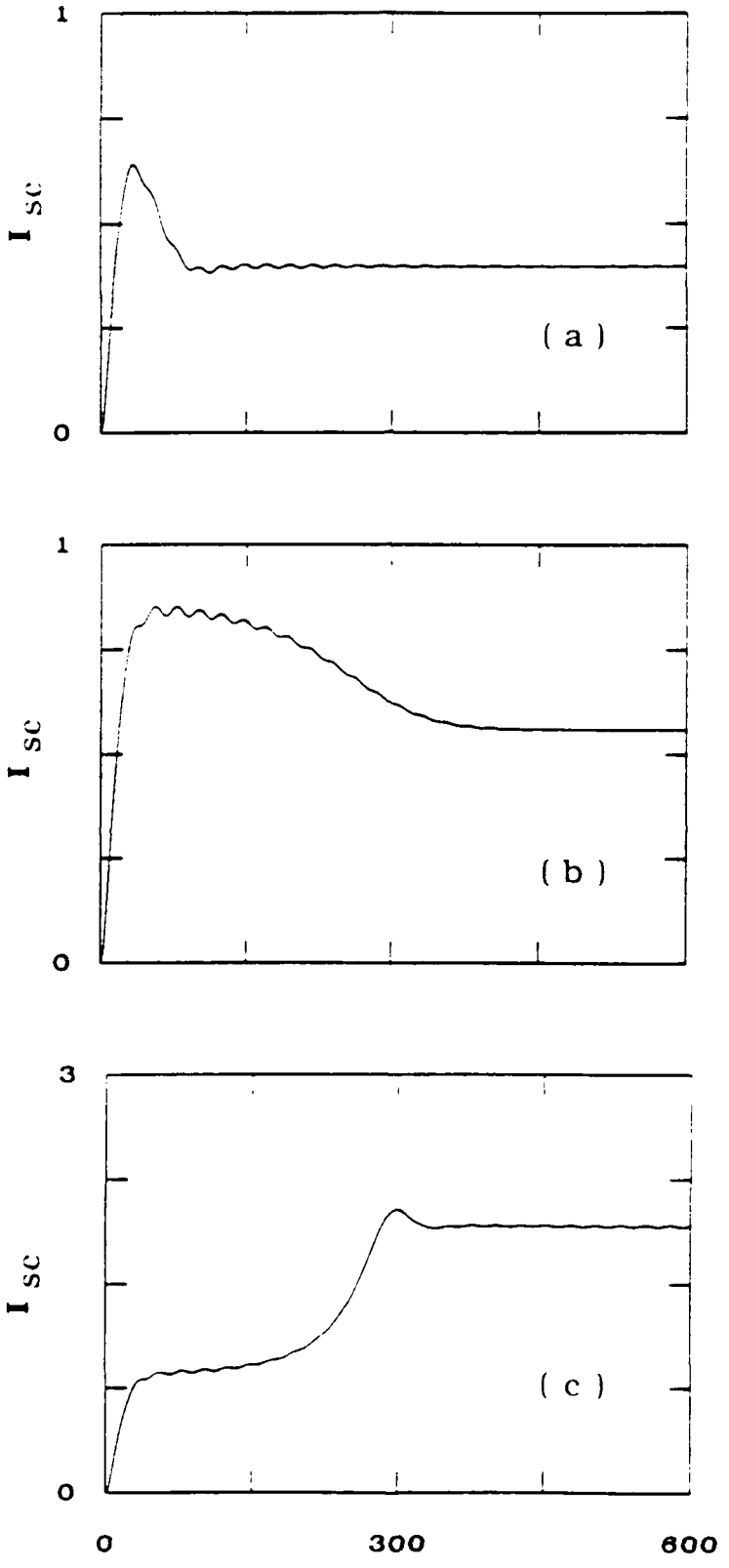
(c)

Fig. 2



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