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New Type of Optical Bistability in Polymers Mediated by Phonons

by

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New type of optical bistability in polymers mediated by phonons

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Abstract

A new type of optical bistability mediated by phonon and virtual exciton coupling is suggested for polymers. Numerical results with parameters pertaining to polydiacetylene-toluene-sulfonate are given as an illustration. The stability of the steady-state solution is examined, and the physical mechanism responsible for the bistability is discussed.

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In the transparent region well below the absorption edge of semiconductor quantum wells, a considerable amount of work has been done on nonlinear coherent optical processes due to the excitation of light.¹⁻⁴ Such investigations have been extended to polymers in recent years.⁵⁻⁸ When the frequency of the incident laser beam is tuned below the exciton resonance in the semiconductor, it is known that virtual excitons are generated. These excitons interact nonlinearly with photons in the same way as real excitons.^{2,4-6} This nonlinear interaction, sometimes known as the phase space filling effect, determines the spectral changes associated with the optical excitation of two-dimensional (2D) systems of semiconductor quantum wells or of one-dimensional (1D) systems of polymers. It also predicts a reduction in exciton oscillator strength or a bleaching of the exciton transition.

Several recent studies⁶⁻⁸ have demonstrated that it is the optical phonon modes that mediate the exciton bleaching in polydiacetylene-toluene sulfonate (PTS). Conjugated polymers have attracted much attention because of their large nonlinear optical susceptibility $\chi^{(3)}$. All these point to their tremendous potential of applications in optical signal processing and high-speed computing. It is therefore of great interest to investigate the fundamental mechanism that yields such a giant optical nonlinearity.

We consider in this Communication the special case in which a sample of polymer is contained in an optical cavity, and study its response to a laser beam directed into the cavity. The polymer system in our treatment is described by a Hamiltonian which is obtained by adding the photon-exciton interaction to the model proposed in Ref. 6. This simple model has been proven successful in the explanation of phonon-mediated optical Stark effect observed experimentally.⁷ The purpose of this paper is to investigate the relation between the input optical field intensity and the output intensity

with special interest in the possible existence of any optical multistability in the system. Our approach is semiclassical in the sense that quantum fluctuations are neglected as usual. The phonon and exciton variables are then eliminated adiabatically in our search for the multistability.

The total Hamiltonian can be written as

$$H = \Omega a^\dagger a + \omega_{ph} b^\dagger b + \omega_{ex} c^\dagger c + \lambda c^\dagger c (b + b^\dagger) + ig(a^\dagger c - c^\dagger a) + i(a^\dagger E e^{-i\Omega t} - a E^* e^{i\Omega t}) \quad (1)$$

where we have ignored the spatial variation of the cavity field which creates the virtual exciton. We consider in (1) only one of the many phonon modes coupling most strongly to the exciton and neglect the momentum dependence of the exciton.⁶ Here, $a^\dagger(a)$, $b^\dagger(b)$ and $c^\dagger(c)$ are the creation (annihilation) operators for the cavity field, phonon and exciton, respectively, with corresponding frequencies Ω , ω_{ph} and ω_{ex} . λ is the coupling constant for the phonon-exciton interaction, while g stands for the exciton-field coupling constant. The coherent driving field is taken to have the same frequency Ω , and its amplitude is E .

As mentioned above, our approach is semiclassical. The procedure is as follows. We first find from the Hamiltonian (1) equations of motion for the operators a , b , c in the rotating frame. Then we replace the operators by their mean values defined as classical variables $\alpha = \langle a \rangle$, $\beta = \langle b \rangle$, $\eta = \langle c \rangle$ and $n_c = \langle c^\dagger c \rangle$. In other words, in the limit of zero fluctuations, all correlation functions may factorize and we have the optical Bloch-like equations

$$\dot{\alpha} = E - \kappa\alpha + g\eta \quad (2a)$$

$$\dot{\beta} = -(i\omega_{ph} + \gamma_{ph})\beta - i\lambda n_c \quad (2b)$$

$$\dot{\eta} = -(i\Delta + \gamma_{ex})\eta - i\lambda\eta(\beta + \beta^*) - g\alpha \quad (2c)$$

$$\dot{n}_c = -g(\alpha^*\eta + \eta^*\alpha) - \gamma_{ex}n_c \quad (2d)$$

where we have defined the detuning $\Delta = \omega_{ex} - \Omega$. The damping rates κ , γ_{ph} and γ_{ex} have been introduced phenomenologically.

In the limit of high Q cavity, we have $\kappa \ll \gamma_{ex}, \gamma_{ph}$. Consequently, the excitonic and phononic variables can be eliminated adiabatically.⁹ In other words, the time variation of these variables are assumed to follow that of the fields. By setting the left-hand side equal to zero, it is therefore straightforward to obtain from Eqs. (2b)-(2d)

$$\beta = -\lambda(\omega_{ph} + i\gamma_{ph})n_c/(\omega_{ph}^2 + \gamma_{ph}^2) \quad (3a)$$

$$\beta + \beta^* \approx -2\lambda n_c/\omega_{ph} \quad (3b)$$

$$\eta = -g(1 - i\gamma)\alpha/(1 + Y^2) \quad (3c)$$

$$\lambda_p^2 n_c^3 - 2\lambda_p \Delta n_c^2 + (1 + \Delta^2)n_c = 2g^2 |\alpha|^2 \quad (3d)$$

where we have defined

$$Y = \Delta - \lambda_p n_c \quad (4a)$$

$$\lambda_p = 2\lambda^2/\omega_{ph} \quad (4b)$$

and have made use of the fact that $\omega_{ph} \gg \gamma_{ph}$.^{6,7} All quantities with the dimension t^{-1} are measured in the unit $\gamma_{ex} = 1$. Equation (3c) follows directly from (2c) and (3b), and (3d) is obtained by plugging (3c) in (2d).

For convenience, we now introduce the dimensionless variables $\tau = \kappa t$, $E = E/\kappa$ and $\alpha_c = \sqrt{2g^2}\alpha$. The equation of motion (2a) for the field mode then becomes

$$\frac{\partial}{\partial \tau} \alpha_c = \sqrt{2g^2} E - \alpha_c f(|\alpha_c|^2) \quad (5)$$

where the function f is defined as

$$f(|\alpha_c|^2) = 1 + \left(\frac{g^2}{\kappa}\right) \frac{1 - i[\Delta - \lambda_p n_c(|\alpha_c|^2)]}{1 + [\Delta - \lambda_p n_c(|\alpha_c|^2)]^2} \quad (6)$$

That the number of virtual excitons n_c is a function of the field intensity $I = |\alpha_c|^2$ only is clearly seen from (3d). In the steady state, we set $\alpha_c = \alpha_s =$ constant in (5) and (6). A straightforward calculation then leads to

$$I_s = \frac{1 + Y_s^2(I_{in})}{(1 + g^2/\kappa)^2 + Y_s^2(I_{in})} I_{in} \quad (7)$$

where we have defined the steady-state cavity field intensity $I_s = |\alpha_s|^2$, the driving field intensity $I_{in} = 2g^2|E|^2$ and $Y_s(I_{in}) = \Delta - \lambda_p n_s(I_{in})$. The fact that n_s is a function of the driving field intensity I_{in} is seen from

$$n_s^3 - 2(\Delta/\lambda_p)n_s^2 + [\Delta^2 + (1 + g^2/\kappa)^2]\lambda_p^{-2}n_s - I_{in}/\lambda_p^2 = 0 \quad , \quad (8)$$

which follows from (3d) and (7). Equation (8) may have one or three real roots depending on the parameters chosen. If there are three possible values of n_s for a given I_{in} , Eq. (7) implies that there may exist three possible intensities of the output light since the output intensity is proportional to the cavity field intensity.

It is necessary to determine the stability of the steady-state solutions. This may be done in a similar fashion as discussed in Ref. 11. Consider a general differential equation of the type

$$\dot{\alpha}_c = F(\alpha_c, \alpha_c^*) \quad , \quad (9)$$

which may be linearized by a standard procedure for small perturbations $\delta\alpha_c$ and $\delta\alpha_c^*$ about their steady-state solutions α_s and α_s^* . The resulting linear equation is

$$\frac{\partial}{\partial \tau} \begin{pmatrix} \delta\alpha_c \\ \delta\alpha_c^* \end{pmatrix} = A \begin{pmatrix} \delta\alpha_c \\ \delta\alpha_c^* \end{pmatrix} \quad , \quad (10)$$

where the matrix A is given by

$$A = \begin{bmatrix} \frac{\partial}{\partial \alpha_c} F(\alpha_s, \alpha_s^*) & \frac{\partial}{\partial \alpha_c^*} F(\alpha_s, \alpha_s^*) \\ \frac{\partial}{\partial \alpha_c} F^*(\alpha_s, \alpha_s^*) & \frac{\partial}{\partial \alpha_c^*} F^*(\alpha_s, \alpha_s^*) \end{bmatrix} \quad . \quad (11)$$

The condition for stability is that the eigenvalues of A must have negative real parts, which requires that $\text{Tr}|A| < 0$ and $\det|A| > 0$. More explicitly, we have

$$\frac{\partial}{\partial u} (\text{Re}F) + \frac{\partial}{\partial v} (\text{Im}F) < 0 \quad (12a)$$

$$\frac{\partial}{\partial u} (\text{Re}F) \frac{\partial}{\partial v} (\text{Im}F) - \frac{\partial}{\partial v} (\text{Re}F) \frac{\partial}{\partial u} (\text{Im}F) > 0 \quad , \quad (12b)$$

where $u = \text{Re}(\alpha_c)$ and $v = \text{Im}(\alpha_c)$. Thus the stability conditions for our Eq. (5) are

$$f_1(I_s) + I_s \frac{\partial}{\partial I} f_1(I_s) > 0 \quad (13a)$$

$$\sum_{i=1}^2 f_i(I_s) [f_i(I_s) + 2I_s \frac{\partial}{\partial I} f_i(I_s)] > 0 \quad , \quad (13b)$$

where $f_1(f_2)$ is the real (imaginary) part of f .

On the other hand, since Eq. (8) has only one real root for $\Delta \leq 0$, it is quite clear that there is no multiple output intensity whenever the cavity field is tuned on or above the resonance with the exciton, no matter how strong the input intensity is. The situation is the same when no phonons are involved or when $\lambda_p = 0$. Therefore, the presence of the optical multistability is really mediated by the phonons via their nonlinear interaction with virtual excitons.

It is known that polydiacetylenes possess not only large $\chi^{(3)}$ but also small transmission loss α .¹⁰ An added attraction is their flexibility in the

construction of waveguides.¹¹ We consider in what follows polydiacetylene-toluene-sulfonate (PTS) as a specific example in our numerical calculation. Following Ref. 6, we take only one of the phonon modes which couple most strongly to the excitons. The parameters for PTS are^{6,7} $\gamma_{\text{ex}} = 0.05$ eV, $\omega_{\text{ph}} = 0.258$ eV, $\lambda = 0.1$ eV and $\gamma_{\text{ph}} = 0.002$ eV. The steady-state intensity I_s has been calculated numerically as a function of I_{in} with the energy unit γ_{ex} . Thus $\omega_{\text{ph}} = 5.16$ and $\lambda = 2.0$. The results for different values of Δ and g^2/κ are plotted in Fig. 1. It is found that only when the cavity field is tuned sufficiently below the exciton resonance, say by an amount $\geq \frac{1}{2} \omega_{\text{ph}}$, does there exist optical tristability. When the cavity damping is large or when g^2/κ is small, there is no such nonlinear phenomenon either. This means that the cavity feedback is one of the vital factors to create optical tristability in the present case. On the other hand, such nonlinear phenomenon does not appear in cavities of very high quality, namely, cavities of exceedingly high figure of merit Q , or g^2/κ is large. This implies that enough line width of the cavity is required to create such nonlinear interactions, and hence that this tristability is at least a dispersive one.

Through the linearized stability analysis of the steady-state solutions discussed above, we have found that all the multiple solutions are stable except for those in the vicinity of turning points such as A and B in Fig. 1 (b) and (c). Hence, when the input field intensity increases continuously, passing the turning point, and then decreases, the output field intensity will follow a reversed hysteresis loop. In contrast to the ordinary optical bistability behavior, this novel multistability behaves more like the special hysteresis loop in systems that exhibit increasing absorption optical bistability.¹³ It is also of interest to note that the middle branch as well as the other two as shown in the figure is stable. This is of course very

different from what happens in ordinary cases. However, as long as the input field intensity changes monotonically, the cavity field intensity is not expected to move into the middle branch. For instance, the system follows the upper branch as I_{in} increases until it reaches the turning point A where it is unstable and jumps to the lower branch as I_{in} increases further. Similarly, the system follows the lower branch and jumps to the upper branch at the unstable point B when I_{in} decreases. It is not impossible to reach the middle branch if the input intensity reverses its course of change at the turning points. In addition, we learn from the numerical study that the threshold for the bistability is raised by increasing the detuning Δ . Thus, a proper choice of Δ may be preferable for the realization of optical bistability.

We now attempt to understand physically the mechanism responsible for this novel phenomenon of reversed optical bistability. It is seen from Eq. (3c) that the resonance enhancement of the dipole moment in steady state occurs at $\omega_x - \Omega - \lambda_p n_s = 0$. The term $\lambda_p n_s$ may be regarded as the lattice relaxation energy.⁶ Therefore both the real and imaginary parts of the optical response of the PTS system depend upon this relaxation energy, which is directly proportional to the number of virtual excitons induced by the cavity field. Since both the absorption and dispersion of PTS are known to be intensity dependent, it appears that the bistability discovered in the present work belongs to a mixed type of dispersion and absorption. As we have found in our numerical analysis, when I_{in} increases from zero and reaches a point where n_s can have three different values, the largest n_s always satisfies the relation $\Delta - \lambda_p n_s = 0$. Further increase of I_{in} beyond that point leads to an increase of the refractive index of PTS. This may be understood in the following way. As the input intensity increases beyond a certain level, the number of virtual excitons becomes large enough that the lattice relaxation is

sufficiently high for the refractive index to deviate from the cavity resonance. Consequently, the cavity field switches to the lower branch. The dispersion effect plays a crucial role in this type of optical bistability.

As concluding remarks, we note that the virtual excitons and lattice vibrations in PTS indeed damp much faster than the damping in an ordinary cavity. This enables us to eliminate the excitonic and phononic variables adiabatically for the special kind of bistability discussed above. Furthermore, the novel phenomenon of optical tristability is found by considering only a single phonon mode in PTS. It is therefore expected to find optical multistability if more phonon modes are included.

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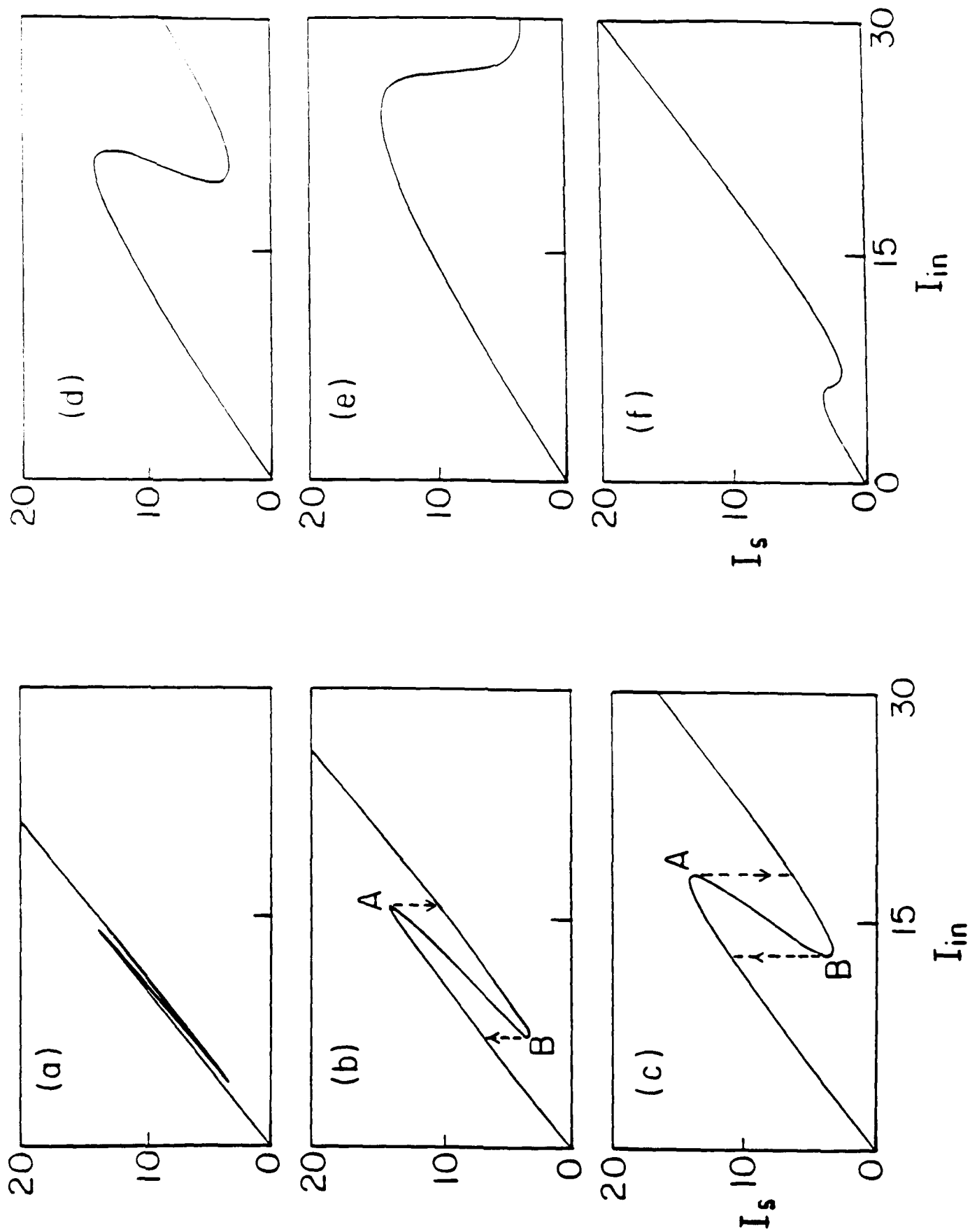
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Figure Caption

1. Intensity of the cavity field I_s versus the input field intensity I_{in} .
 (a) $\Delta = \omega_{ph} = 5.16$, $g^2/\kappa = 0.1$; (b) $\Delta = \omega_{ph}$, $g^2/\kappa = 0.5$; (c) $\Delta = \omega_{ph}$, $g^2/\kappa = 1.0$; (d) $\Delta = \omega_{ph}$, $g^2/\kappa = 1.5$; (e) $\Delta = \omega_{ph}$, $g^2/\kappa = 2.0$; (f) $\Delta = 3.0$, $g^2/\kappa = 1.0$. The dashed lines in (b) and (c) are hypothetical to complete the reversed hysteresis loop.

Fig. 1



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