Optical bistability and switching dynamics in an exciton-biexciton model for CuCl

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The dynamical evolution of the electromagnetic field amplitudes is numerically calculated using an exciton-biexciton model of CuCl. For a bistable system we determine the switch-down and switch-up times and find them to be well explained by the polariton escape time using the group velocity for the polaritons. Our switching times, about 80 psec at photon energies of 3177 meV, are at least an order of magnitude larger than a previous theoretical estimate and we predict the switching times will become longer as the exciton resonance is approached.

In this paper we consider results for optical bistability in the nonlinear optical medium, CuCl, in a Fabry-Perot geometry and report results for the transient dynamics of the transmitted intensity.[1] The model we use for CuCl is based on the production of excitons and of excitonic molecules, the so-called biexcitons. The biexcitons are strongly bound and have been observed, for instance, in two-photon absorption experiments.[2] Biexcitonic states have served, therefore, as a mechanism of optical nonlinearities in CuCl and, in particular, they provide the possibility of using CuCl as an optically bistable device. This was proposed by Hanamura[3] and Koch and Haug,[4] and their results paved the way for further, more detailed theoretical calculations[5,6] and for experiments, which successfully demonstrated optical bistability in this medium.[7,8]

The experiments have not yet accurately reported the switching times, although the switching is known to be faster than 500 psec.[8] The only theoretical statement about the switching times was by Hanamura[3] in which he states that the switching should be of the order of a picosecond. His results were based on a mean-field treatment of the Maxwell and Heisenberg equations of motion, as well as an adiabatic elimination of the exciton and biexciton dynamics. We find that both of these approximations are unwarranted for CuCl and we treat the problem in a more precise manner; this technique should also be useful for other systems.

We have developed a slowly varying envelope approximation[9] (SVEA), appropriate for the CuCl system. Our theory provided an accurate reconstruction of the steady-state field values obtained from a numerical treatment of the second-order Maxwell equation in the large Fresnel number limit. Employing the mode expansion appropriate for slowly varying amplitudes,[10] we express our dynamical equations in terms of forward- and backward-propagating electric fields $E_F(x,t)$, $E_B(x,t)$, exciton fields $b_F(x,t)$, $b_B(x,t)$, biexciton fields $B_F(x,t)$, $B_B(x,t)$, and a complex biexciton amplitude $B_0(x,t)$. The propagation direction is along the x axis. Previous publications by Bishofberger and Shen[11] and Goldstone and Garmire[12] have studied the dynamic response of a Fabry-Perot cavity filled with a Kerr medium. Our work differs from those by the inclusion of propagation in the equations and, as we shall see, by the large dispersion of phase velocity in this system.

The equations of motion in the SVEA are written in the following compact form:

$$\frac{\partial}{\partial t}E_{\binom{F}{B}} \pm v\frac{\partial}{\partial x}E_{\binom{F}{B}} = -\frac{2\pi\omega}{\epsilon_R(0)}\left\{g_1 b_{\binom{F}{B}} + i\frac{[\epsilon_R(0)-\epsilon_\infty]}{4\pi}E_{\binom{F}{B}}\right\} , \quad (1)$$

$$\frac{\partial}{\partial t}b_{\binom{F}{B}} = -i(\delta - i\gamma_x)b_{\binom{F}{B}} + g_1 E_{\binom{F}{B}} - g_2(B_0 E^*_{\binom{B}{F}} + E^*_{\binom{F}{B}} B_{\binom{F}{B}}) , \quad (2)$$

$$\frac{\partial}{\partial t}B_{\binom{F}{B}} = -i(\Delta - i\gamma_m)B_{\binom{F}{B}} + g_2 E_{\binom{F}{B}} b_{\binom{F}{B}} , \quad (3)$$

and

$$\frac{\partial}{\partial t}B_0 = -i(\Delta - i\gamma_m)B_0 + g_2(E_F b_F + E_B b_B) . \quad (4)$$

The negative sign of the left-hand side of Eq. (1) is taken for the backward (B) field amplitude. The laser frequency is ω; $\delta = (\omega_x - \omega)$ is the detuning of the laser from the exciton transition frequency ω_x, and $\Delta = (\omega_m - 2\omega)$ is the detuning of the laser from the biexciton transition frequency ω_m. The exciton and biexciton decay rates are γ_x and γ_m, respectively, and g_1 and g_2 are coupling constants between the exciton and field and between the biexciton-exciton and field. ϵ_∞ is the high-frequency dielectric constant of the medium, i.e., it contains the optical modes not including the exciton or biexciton contributions. The exciton coupling g_1 is strong and is treated exactly; this alters the low-field value of the dielectric constant significantly, $\epsilon(0) = \epsilon_\infty + 4\pi g_1^2/(\delta - i\gamma_x)$; $\epsilon_R(0)$ is the real part of this expression. v is the phase velocity of light in the medium, $v = c/\sqrt{\epsilon_R(0)}$, where c is the velocity of light in vacuum.

Equation (1) needs to be supplemented by the appropriate boundary conditions. For a medium of length L and external dielectric mirrors of reflectivity R, the boundary conditions are

$$E_F(0)=\{E_B(0)[(1+\sqrt{R})\sqrt{\epsilon_R(0)}-1+\sqrt{R}]+2\sqrt{1-R}E_{\rm IN}\}/[(1+\sqrt{R})\sqrt{\epsilon_R(0)}+1-\sqrt{R}] \quad (5)$$

and

$$E_B(L)=E_F(L)e^{2ikL}[(1+\sqrt{R})\sqrt{\epsilon_R(0)}-1+\sqrt{R}]/[(1+\sqrt{R})\sqrt{\epsilon_R(0)}+1-\sqrt{R}] \; . \quad (6)$$

$E_{\rm IN}$ is the electromagnetic field amplitude of the injected photons and k is the wave number of the photons in the medium. All numerical results quoted below use the values summarized in Table I.

In Fig. 1 the SVEA steady-state solution is plotted for the output intensity versus input intensity and the corresponding average biexciton density

$$n_B=\frac{1}{L}\int_0^L (|B_0|^2+|B_F|^2+|B_B|^2)\,dx$$

is also shown in the same plot. The biexciton density is proportional to the square of the internal field intensity at low powers, since it is a two-photon process. These curves were generated using 100 points along the x axis and a predictor-corrector integration method. We have chosen the laser frequency far enough off resonance so that any resonance enhancement of the biexciton linewidth may be neglected.[2,4,13] The biexciton density is multivalued and at least an order of magnitude smaller than the exciton density,

$$n_b=\frac{1}{L}\int_0^L (|b_F|^2+|b_B|^2)\,dx \; ,$$

at the largest input intensity in the figure. We found the exciton density to be a monotonic function of the input field and at low intensities it is proportional to the internal intensity.

We note that multistability is also obtained at higher intensities within the present theory. The mean-field theory used by Hanamura[3] follows the development of Bonifacio and Lugiato.[14] This theory has only a single bistable regime and we find, using our equations for the exciton and biexciton densities, switching intensities which are three orders of magnitude greater than demonstrated in Fig. 1.[9] We remark that the steady-state fields generated from Eqs. (1)–(4) vary across the cavity only by 10%–20% at the largest input intensities shown in Fig. 1, and at low intensities the variation is much less. Therefore, we expect that an averaging technique performed on these equations, similar to Ref. 14, will describe the essential features found here.

The switch-up and switch-down times were determined by starting the system in a stable steady state very close to the turning points in Fig. 1. The fields in both directions were ramped at a constant rate 0.01 MW/cm^2 per round trip, where the round trip was calculated from the *phase* velocity

TABLE I. The physical parameters used in the numerical work. These values are identical to those in Ref. 9.

$\omega=3177$ meV	$\omega_x=3202.7$ meV
$\omega_m=6372.5$ meV	$\gamma_x=0.03$ meV
$\gamma_m=0.3$ meV	$4\pi g_1^2=27.5$ meV
$g_2^2=1.875\times10^9\,\dfrac{\text{meV}^2\,\text{cm}^3}{\text{MJ}}$	$\epsilon_\infty=5$
$L=9.98165\ \mu$m	$R=0.9$

$\tau_R=2L/v\simeq0.165$ psec. For the switch-up curve in Fig. 2 our initial input intensity is 32.09 MW/cm^2 and the switch-down curve starts with an input intensity 15.75 MW/cm^2. The total reflection coefficient after accounting for the index of refraction changes at the surfaces is $R_T\simeq0.96$; therefore, the photon escape time is about $\tau_E=\tau_R/(1-R_T)\simeq4.1$ psec. This is comparable to the exciton and biexciton relaxation times (see Table I), $\tau_{\rm exc}=\gamma_x^{-1}=2.2$ psec and $\tau_{\rm biexc}=\gamma_m^{-1}=0.22$ psec. Since the exciton and photon relaxation times are the same order of magnitude, this incidates that the dynamics of the electric field alone do not dominate the time behavior and the exciton modes cannot be adiabatically eliminated.[15]

Our numerical results for the dynamics were obtained by solving Eqs. (1)–(4) using the method of characteristics. As the photon-exciton interaction is strong, in this case, and the exciton resonance is close to the biexciton resonance, the fields have a strong frequency-dependent dispersion curve. Using Eqs. (1) and (2), the group velocity is ($\gamma_x=0$, $g_2=0$) (Ref. 16)

$$v_g=v\Bigg/\left[1+\frac{2\pi g_1^2\omega}{\epsilon_R(0)\delta^2}\right] \; . \quad (7)$$

At $\omega=3177$ meV, $v_g\approx v/12$; hence for the strongly interacting system, i.e., polariton modes, the signals propagate with a reduced velocity, namely, the escape time for polari-

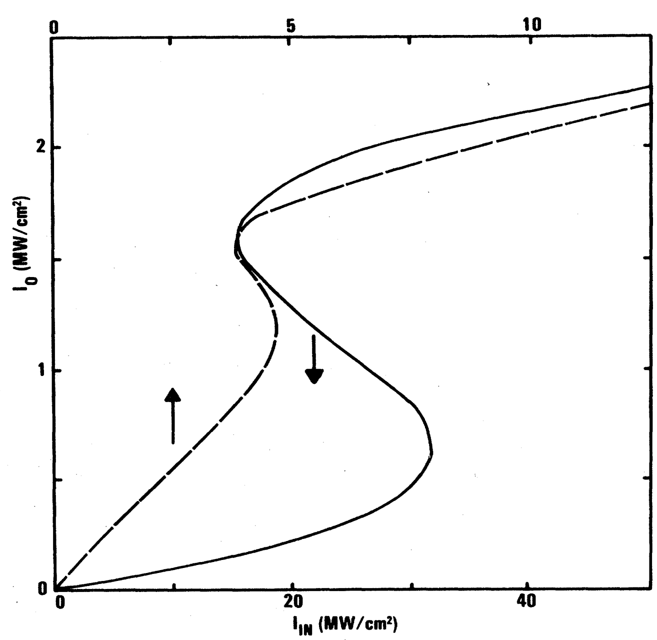

FIG. 1. Steady-state values of the input intensity vs the output intensity (solid line) and the biexciton density vs the output intensity (— — —). Values chosen are displayed in Table I.

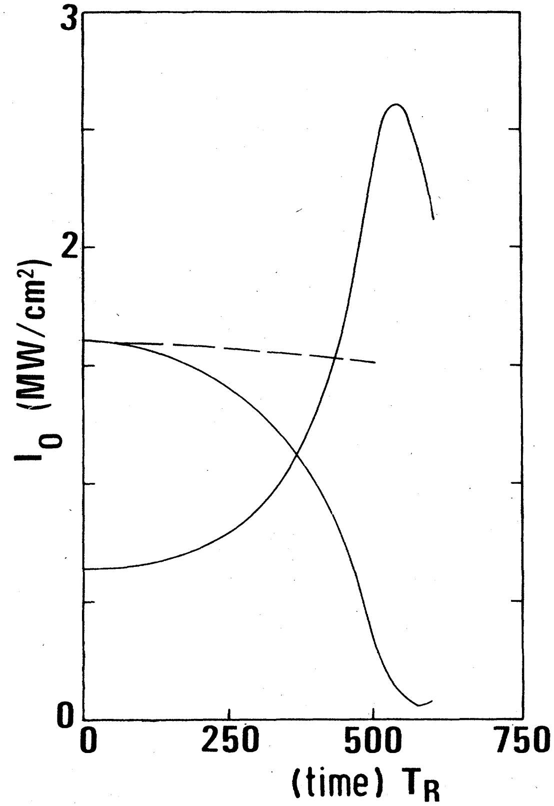

FIG. 2. The output intensity vs time in units of τ_R. The switch-up and switch-down times are nearly equal. The dashed curve is generated by holding the input intensity constant at $I_{IN}=15.4$ MW/cm^2.

tons which is $\tau_{E,\text{pol}} \simeq 300\tau_R$. This result is in agreement with our numerical results plotted in Fig. 2. We obtain from that figure a value for the switching times, defined as the time required for the output intensity to attain an extremum value. This time is about $500\tau_R \simeq 80$ psec and it is the same for both the switch-up and switch-down curves. We note in the figure that there is a ringing of the output intensity which is quite pronounced in the switch-up case. Also shown in Fig. 2 is a run with a constant input field shifted 2% below the upper branch turning point in Fig. 1. This curve dramatically demonstrates the critical slowing-down phenomenon in the unstable region near the turning points.

The group velocity of the linearized exciton-photon system provides the major contribution to the cavity relaxation time and therefore the polariton is the dominant slow mode of the system. This suggests that an adiabatic elimination procedure should be possible based on the polariton modes. Furthermore, we can infer that the switching times will become dramatically longer as the laser frequency approaches the exciton resonance. The group velocity, Eq. (7), is approximately inversely proportional to δ^2; therefore, provided that the energy velocity is close to the group velocity,[16] we expect that at $\omega = 3186$ meV, the switching times would be about four times longer. The actual magnitude of the switching times would depend on the cavity quality and cavity length as well as the material parameters. At this point no optimal set of parameters has been determined; however, it would be an important test of the theory if the trends in the switching times were experimentally confirmed.

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