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Stationary properties of dispersive optical bistability in a dense exciton–biexciton system: copper chloride

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A semiclassical exciton–biexciton model for CuCl is used to obtain the stationary solutions for optical bistability (o.b.) by numerical integration of the second-order Maxwell equation in the limit of large Fresnel numbers. The results obtained from the calculation, which we call exact (ex.), are compared with corresponding numerical results by using the slowly varying envelope approximation (s.v.e.a.) as well as corresponding analytical results obtained by using the mean-field approximation (m.f.a.). The results obtained with the s.v.e.a. are shown to be in close quantitative (within 5%) agreement with the ex. results for each point in the parameter space considered, whereas the m.f.a. gives reasonably good qualitative results only (more than 20% quantitative discrepancy with ex.). Furthermore, o.b. is predicted from these calculations for incident laser field detunings on either side of the two-photon biexciton resonance as well as in the neighbourhood of the resonance. The nonlinearity in the dielectric function that causes o.b. is seen to be very nearly of the Kerr medium type, and consequently, the two-photon biexciton resonance contributes only weakly to the o.b. characteristics.

Introduction

Previous calculations for optical bistability (o.b.) in CuCl assumed the dielectric function to be constant throughout the cavity; this version of the so-called 'mean-field approximation' (m.f.a.) has the advantage of yielding analytical results. Results from the m.f.a. were obtained by several authors (Koch & Haug 1981; Sarid *et al.* 1983; Sung & Bowden 1984). Other examples can be found in Bowden *et al.* (1984). Since the m.f.a. limit is idealized, there are always questions concerning the accuracy and reliability of the results. Therefore, we solve here, in addition, the second-order Maxwell equation, which requires numerical integration (ex. results). A large Fresnel number is assumed so that transverse contributions can be neglected. Bearing in mind that numerical integration of the second-order equation is expensive, we also calculate results from the so-called slowly varying envelope approximation (s.v.e.a.) (Icsevgi & Lamb 1969; Fleck 1970). It is found that, with the exception of rather esoteric conditions, the s.v.e.a. can be used to produce results from our model that are within 5% agreement with the ex. results, whereas the m.f.a. yields results that differ quantitatively by more than 20%. Thus, cost-effectively, the s.v.e.a. will be very useful for our future work; for example, the calculation of the dynamics and switching without recourse to adiabatic elimination (Geigenmüller *et al.* 1983).

1. Model Hamiltonian

The model Hamiltonian for CuCl, which has been widely used to calculate the nonlinear interaction with light for an incident laser field tuned near the exciton or two-photon biexciton resonance, is given in the rotating wave and electric dipole approximations, by

$$H = H_0 + H', \tag{1.1}$$

where
$$H_0 = \omega_x b^+ b + \omega_m B^+ B, \tag{1.2a}$$

and
$$H' = \mathrm{i}g_1 E^+ b^+ + \mathrm{i}g_2 E^+ B^+ b + \mathrm{h.c.} \tag{1.2b}$$

Here, $B^+(B)$ and $b^+(b)$ are the creation (annihilation) operators for the biexciton and excitons, respectively, and ω_m and ω_x are their respective transition energies (units such that $\hbar = 1$ are used); g_1 and g_2 are the coupling constants, whose numerical values are inferred from experiment. The electric field E with positive and negative frequency components, E^+ and E^-, respectively, is assumed to be the superposition of rightward and leftward propagating monochromatic plane waves in the dielectric medium and is understood to have both spacial and temporal dependence in general. The exciton and biexciton operators are correspondingly understood to have k-vector dependence consistent with the electric field. We take the electric field E to be classical and proceed with the semiclassical model calculation where the field and dielectric are coupled by the Maxwell equation.

2. Numerical integration of the second-order Maxwell equation at steady state: ex. results

The field-dependent nonlinear dielectric function obtained by solving the equations of motion derived from (1.2) at steady state, and to lowest order in the field–exciton–biexciton interaction is (Koch & Haug 1981; Sung & Bowden 1984*a*)

$$\epsilon^+(E^+) = \epsilon_\infty + 4\pi g_1^2/(\delta' - g_2^2 |E|^2/\Delta'), \tag{2.1}$$

where unity has been replaced in the first term in (2.1) on the right side by the high-frequency dielectric constant ϵ_∞. Here, $\delta' = \omega_x - \omega - \mathrm{i}\gamma_x$, $\Delta' = \omega_m - 2\omega - \mathrm{i}\gamma_m$ and $|E|^2 = E^+E^-$. In these relations ω is the frequency of the incident electric field, where we have used

$$E^+(x,t) = \mathrm{e}^{-\mathrm{i}\omega t} E^+(x) \tag{2.2}$$

and x is the longitudinal direction of propagation in the dielectric medium of length L. Also γ_x and γ_m are the exciton and the biexciton relaxation rates, respectively, which have been added phenomenologically.

The stationary solution of the Maxwell equation based upon (2.1) and (2.2) is obtained by solving (we omit the superscripts on ϵ and E from here on)

$$\partial^2 E(x)/\partial x^2 + \epsilon(E)\, k_0^2 E(x) = 0, \tag{2.3}$$

where $k_0 = \omega/c$ is the wavenumber in vacuum. If the incident laser beam of amplitude E_I is given by

$$E_\mathrm{INC} = E_\mathrm{I} \exp(-\mathrm{i}\omega t + \mathrm{i}k_0 x) \tag{2.4}$$

for $x \leqslant 0$, then the boundary conditions are represented by the following set of equations at $x = 0$ for a given surface reflective dielectric film mirror of reflectivity r_m,

$$(1-r_m)^{\frac{1}{2}} E_{INC} + (1+r_m^{\frac{1}{2}}) E_1 = E, \tag{2.5a}$$

$$ik_0[(1-r_m)^{\frac{1}{2}} E_{INC} - (1-r_m^{\frac{1}{2}}) E_1] = \partial E/\partial x, \tag{2.5b}$$

where E_1 is the amplitude of the reflected light at $x = 0$, i.e. between the material (CuCl) and the cavity mirror. Similar boundary conditions prevail at $x = L$,

$$(1+r_m^{\frac{1}{2}}) E_3 = E, \tag{2.6a}$$

$$ik_0(1-r_m^{\frac{1}{2}}) E_3 = \partial E/\partial x, \tag{2.6b}$$

where E_3 is the amplitude of the forward-propagating wave at $x = L$ between the material (CuCl) and the mirror. The transmitted wave intensity I_T is given by

$$I_T = |E_T|^2 = (1-r_m)\,|E_3|^2. \tag{2.7}$$

These boundary conditions can be combined to solve (2.3) by numerical integration. The results we call ex. results.

3. Mean-field approximation

The m.f.a. is often used in the literature to describe o.b. in a Fabry–Perot cavity filled with a Kerr medium, and avoids the necessity for numerical integration by imposing the ansatz

$$E(x) = E_R^{(0)}\, e^{ikx} + E_L^{(0)}\, e^{-ikx}, \tag{3.1}$$

where $E_R^{(0)}$ and $E_L^{(0)}$ are rightward and leftward propagating components of the electric field E and are taken to be independent of x. Here k is the complex wavevector defined by

$$k = k_0\, \epsilon^{\frac{1}{2}}, \tag{3.2}$$

where ϵ is given by (2.1). If (3.1) is used in conjunction with the boundary conditions (2.5) and (2.6), the result is (Sung & Bowden 1984*b*)

$$\tau \equiv |E_T|^2/|E_I|^2 = |4(1-r_m)\,\epsilon^{\frac{1}{2}}\{[(1+r_m^{\frac{1}{2}})\,\epsilon^{\frac{1}{2}} + 1 - r_m^{\frac{1}{2}}]^2\, e^{\alpha} - [(1+r_m^{\frac{1}{2}})\,\epsilon^{\frac{1}{2}} - (1-r_m^{\frac{1}{2}})]^2\, e^{-\alpha}\}^{-1}|^2, \tag{3.3}$$

where $\alpha = k_0 L \epsilon^{\frac{1}{2}}$ is the complex phase.

4. Slowly varying envelope approximation solution

The s.v.e.a. has been extensively used in the literature (Icsevgi & Lamb 1969; Fleck 1970) and we will not elaborate on its construction here. It essentially results in a linearization of the Maxwell second-order partial differential equation by removal of the rapidly varying temporal and spacial components of the field. The novel aspect of our calculation is that we remove the spacially varying part according to the approximate wavevector in the material

$$\hat{k} = k_0\, \mathrm{Re}\, (\epsilon(0))^{\frac{1}{2}}, \tag{4.1}$$

rather than k_0 in free space, as is usually the procedure. In (4.1) $\epsilon(0) \equiv \epsilon(E=0)$. The details of our s.v.e.a. calculation will be presented elsewhere (Sung *et al.* 1984).

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

From the s.v.e.a. and the solution of the equations of motion from (1.2) in steady state, we have

$$\frac{\partial \epsilon_F^+}{\partial z} = \frac{2\pi i \omega \Delta' g_1^2}{cS}(\delta'\Delta' - g_2^2|\epsilon_F^+|^2)\,\epsilon_F^+ - \frac{i\omega}{2}\frac{(\epsilon_R(0)-\epsilon_\infty)}{\epsilon_R(0)}\epsilon_F^+, \tag{4.2a}$$

$$\frac{\partial \epsilon_B^+}{z} = -\frac{2\pi i \omega \Delta' g_1^2}{cS}(\delta'\Delta' - g_2^2|\epsilon_B^+|^2)\,\epsilon_B^+ + \frac{i\omega}{2}\frac{(\epsilon_R(0)-\epsilon_\infty)}{\epsilon_R(0)}\epsilon_B^+. \tag{4.2b}$$

In these equations ϵ_F^+ and ϵ_B^+ are the slowly varying forward and backward propagating field amplitudes, respectively, and

$$S = (\delta'\Delta')^2 - 2g_2^2\,\delta'\Delta'(|\epsilon_F^+|^2 + |\epsilon_B^+|^2) + g_2^4(|\epsilon_F^+|^4 + |\epsilon_B^+|^4 + |\epsilon_F^+|^2\,|\epsilon_B^+|^2). \tag{4.3}$$

It is to be noted that if the last term in (4.3) on the right side were a perfect square, equations (4.2) could be integrated immediately, by using the boundary conditions (2.5)–(2.7). However, this is not the case, so we proceed with numerical solution of (4.2) with the boundary conditions already mentioned.

5. Results and discussion

The ex., s.v.e.a. and m.f.a. results for a sample length $L \approx 10$ μm are presented in figure 1. We have fixed the frequency $\omega = 3.177$ eV and varied L to obtain a well-defined o.b. condition in m.f.a. This is why $L = 9.98165$ μm in these results. We have chosen a value for ω far from resonance, as was done by Sarid *et al.* (1983). Since γ_x is small compared to γ_m, we neglect it altogether to proceed more easily. The value for γ_m varies considerably in the literature and may well vary from sample to sample. It is also claimed to be field-dependent (Sarid *et al.* 1983). It has been pointed out by Abram (1983), that at least part of the apparent field dependence of the shape of the absorption spectrum for CuCl can arise from spacial 'chirp' due to the field-dependent shift of the peak of the absorption spectrum dictated by (2.1). Since this condition is obviously present in the ex. and s.v.e.a. calculations from our model, we do not impose further conditions extraneous to our model.

The remarkable agreement between the s.v.e.a. and ex. results is due largely to the choice of the wavevector $\hat{k}$, (4.1) in the s.v.e.a. The m.f.a. results give only qualitatively reliable results. The comparison between the s.v.e.a., ex. and m.f.a. depicted in figure 1 is consistent throughout the parameter space tested, including the cases at $L = 1$ μm and $L \approx 30$ μm.

Displayed in figure 2 are results for the incident field frequency tuned to a value approximately as far above the two-photon resonance as the value chosen in figure 1 was below resonance. This shows at least as strong a hysteresis condition as for the previous case below resonance. The s.v.e.a. and m.f.a. obviously compare in the same way as before and the ex. result is not plotted since the agreement is not different from that depicted in figure 1. Also shown in the box on the right in figure 2 is the s.v.e.a. at $L = 30$ μm. The change in length produces a change in intensity threshold by two orders of magnitude. Hence it is possible to reduce the threshold by searching for optimal values of the length and detuning.

A major conclusion from these results is that the nonlinearity that causes o.b. in excitonic CuCl is largely of the Kerr medium type. We have tested the o.b. conditions near the two-photon

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

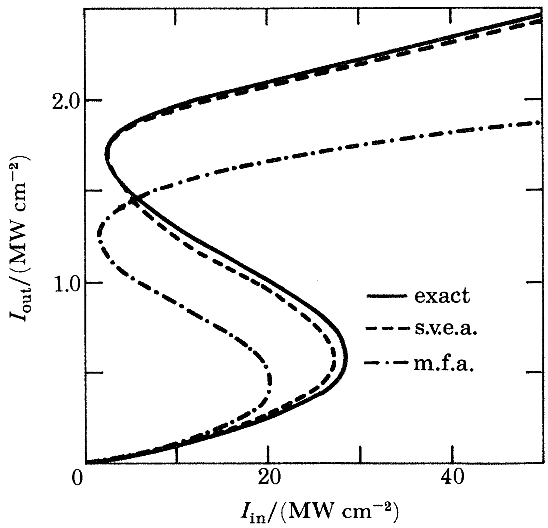

FIGURE 1. Input against output intensities for the laser frequency $\omega = 3.177$ eV and sample length $L = 9.98165$ μm. Other relevant parameters are: $r_m = 0.9$, $\gamma_m = 0.3$ meV, $\gamma_x = 0$, $4\pi g_1^2 = 27.5$ meV^2, $|M|^2 = 1.57 \times 10^{-16}$ meV^2 cm^3 and $g_2^2 = |\epsilon|\,|M|^2/\omega$, $\omega_x = 3.2027$ eV, $\omega_m = 6.3725$ eV and $\epsilon_\infty = 5.0$.

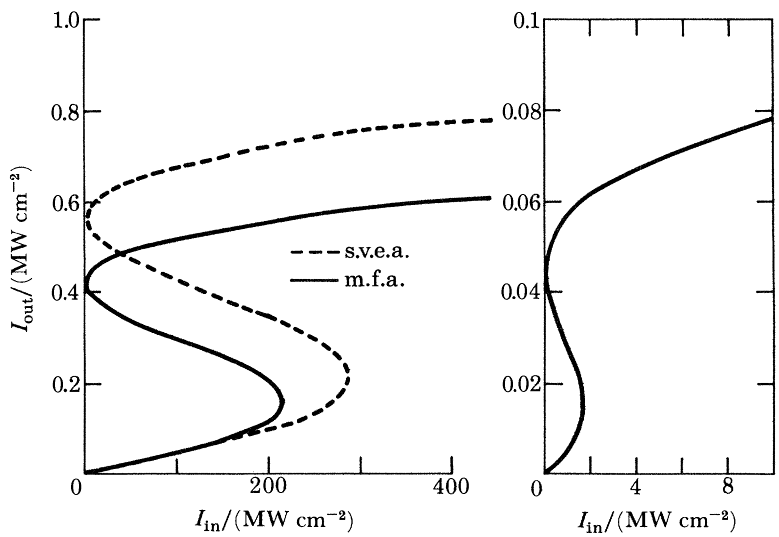

FIGURE 2. Input against output intensity for the laser frequency $\omega = 3.1956$ eV and $L = 10$ μm. The box on the right is the s.v.e.a. for a sample length $L = 30$ μm; note that the output intensity scale is reduced by a factor of 10 and the input intensity scale has been changed. All other parameters are the same as for figure 1 except that $\gamma_m = 0.1$ meV.

resonance and the o.b. is relatively weak. The fact of the Kerr medium behaviour and the choice of (4.1), explains the close agreement between our s.v.e.a. and ex. results.

Based upon these results, we are now in the process of calculating the dynamics and switching times for this system by using the s.v.e.a. and without using adiabatic elimination, which is not valid for CuCl.

J.W.H. is a National Research Council research associate.

References

Abram, I. 1983 *Phys. Rev.* B **28**, 4433.
Bowden, C. M., Gibbs, H. M. & McCall, S. L. (eds) 1984 *Optical Bistability* 2. New York: Plenum Press.
Fleck, J. A. 1970 *Phys. Rev.* B **1**, 84.
Geigenmüller, U., Titulaer, U. M. & Felderhof, B. U. 1983 *Physica* A **119**, 411.
Icsevgi, A. & Lamb, W. E. 1969 *Phys. Rev.* **185**, 517.
Koch, S. W. & Haug, H. 1981 *Phys. Rev. Lett.* **46**, 450.
Sarid, D., Peyghambarian, N. & Gibbs, H. M. 1983 *Phys. Rev.* B **28**, 1184.
Sung, C. C. & Bowden, C. M. 1984*a* *Phys. Rev.* A **29**, 1957.
Sung, C. C. & Bowden, C. M. 1984*b* *J. opt. Soc. Am.* B **1**, 395.
Sung, C. C., Bowden, C. M., Haus, J. W. & Chiu, W. K. 1984 *Phys. Rev* A **30**. (In the press.)

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