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Intrinsic optical bistability in a thin layer of nonlinear optical material by means of local field effects

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Abstract

We show that a single thin layer (thickness much less than an optical wavelength) of nonlinear optical material can display optical bistability solely as a consequence of local field effects. Optical bistability is predicted when the phase of the complex third-order susceptibility $\chi^{(3)}$ is nearly opposite that of the linear dielectric constant ϵ and when the incident field amplitude is adjusted so that $|\chi^{(3)}||E_0|^2$ is approximately equal to $10^{-3}|\epsilon|^3$. © 2000 Elsevier Science B.V. All rights reserved.

Following the initial demonstration of optical bistability using Na vapor in a Fabry–Perot resonator [1], optical bistability has received much attention because of its potential uses including optical logic and other optical switching devices [2]. In most configurations, optical bistability is achieved by the combined action of the nonlinear response of a material medium and the feedback of an optical resonator. However, the use of an external resonator is in many ways undesirable, for instance in that it slows down the temporal response of the bistable device and requires careful alignment. For such reasons, it would be desirable to produce optical bistability using internal feedback based on the modification of material properties by the incident optical beam.

Mirrorless or intrinsic optical bistability was first considered by Bowden and Sung [3] who pointed out that a system comprised of a collection of identical two-level atoms interacting with the electromagnetic field can show optical bistable behavior without external feedback. Optical bistability via dipole–dipole coupling was observed experimentally by Hehlen et al. [4] in crystalline $Cs_3Y_2Br_9$:Yb^{3+}. Another approach for achieving intrinsic optical bistability was proposed by Kalyaniwalla et al. [5] and by Bergman et al. [6] based on local field enhancement in a metal and dielectric composite. This approach was later demonstrated successfully by Neuendorf et al. [7] using an aqueous colloidal solution of silver-coated CdS. Bergman et al. [6] found that multiple solutions for the local electric field can occur in the composite if one of the components has an optical nonlinearity. For example, in a composite composed of alternating thin layers of a metal and a dielectric,

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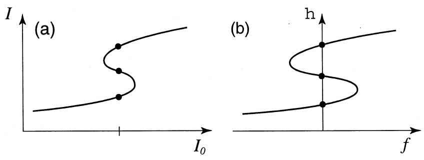

Fig. 1. (a) Optical bistability occurs when the internal intensity I has three possible values for some incident intensity I_0. (b) This in turn means that the auxiliary function $f(\eta)$ (defined in Eq. (3), where η is the dimensionless intensity variable) must have three real, positive roots.

the surface normal components of $\boldsymbol{E}$ and $\boldsymbol{D}$ inside the composite can have three solutions for certain values of incident field E_0 if the optical constants of the constituent materials and their volume fill fractions are selected appropriately. Based on these results, one might suppose that optical bistability could occur even from a single thin film of nonlinear material situated in vacuum. In this paper we report the results of our theoretical investigation of this conjecture. In particular, we find that if the linear and nonlinear susceptibilities of the material have the appropriate relative phase, optical bistability is predicted for some range of incident intensities.

We allow a beam of light to fall at oblique incidence on a thin film, and we consider the component of the electric field E_0 perpendicular to the layer surface. Assuming a continuum description of the response of the film is adequate, for a sufficiently thin film $(d \ll \lambda)$ we can use the electrostatic result that the electric field E inside the film is given by $E = E_0 - 4\pi P$. In this case, if the material has a third-order optical nonlinearity, the induced polarization P becomes $P = \chi^{(1)}E + 3\chi^{(3)}|E|^2E$. Combining the two equations, we obtain the relation

$$E_0 = \left[\epsilon + 12\pi\chi^{(3)}|E|^2\right]E \qquad (1)$$

between E_0 and E. By squaring the absolute values of both sides of this equation, we obtain a cubic equation relating the 'intensity' $I = |E|^2$ inside the film to the incident 'intensity' $I_0 = |E_0|^2$ as follows:

$$I^3 + \frac{\mathrm{Re}\{\epsilon^*\chi^{(3)}\}}{6\pi|\chi^{(3)}|^2}I^2 + \frac{|\epsilon|^2}{144\pi^2|\chi^{(3)}|^2}I - \frac{I_0}{144\pi^2|\chi^{(3)}|^2} = 0. \qquad (2)$$

Therefore, optical bistability can occur if this cubic equation has three real, positive solutions for I for some value of I_0 (Fig. 1(a)). If we define new variables $\eta \equiv |\epsilon|^2 I/I_0$ and $\zeta \equiv 12\pi\chi^{(3)}I_0/(\epsilon|\epsilon|^2)$, Eq. (2) becomes

$$f(\eta;\zeta) \equiv \eta^3 + 2\mathrm{Re}\{1/\zeta\}\eta^2 + 1/|\zeta|^2\eta - 1/|\zeta|^2 = 0. \qquad (3)$$

Since η is proportional to the ratio of the intensity I inside the material to the applied intensity I_0, the existence of three real, positive solutions of the equation $f(\eta) = 0$ signifies the occurrence of optical bistability (Fig. 1(b)). We shall see below that optical bistability is predicted only for certain values of the complex parameter ζ. Since ζ depends on material parameters and on the intensity of the incident light field, optical bistability can occur only for certain incident intensities and certain values of the relative phase of $\chi^{(3)}$ and of ϵ.

Two conditions guarantee that $f(\eta) = 0$ has three real, positive solutions:

1. *The equation $df/d\eta = 0$ has two real, positive solutions η_1 and η_2 $(\eta_2 > \eta_1 > 0)$.*
2. *$f(\eta_1)$ and $f(\eta_2)$ are positive and negative, respectively.*

Using these criteria we were able to determine numerically the requirements on ζ for the occur-

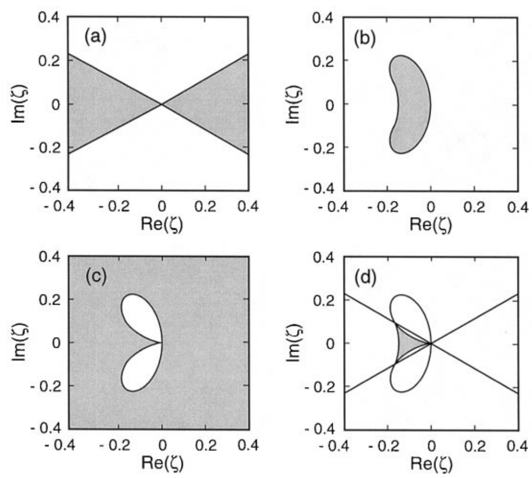

Fig. 2. The shaded region shows the values of ζ for which (a) $\mathrm{d}f(\eta)/\mathrm{d}\eta = 0$ has two positive real solutions, (b) $f(\eta_1) > 0$, (c) $f(\eta_2) < 0$, and (d) $f(\eta) = 0$ has three positive real solutions

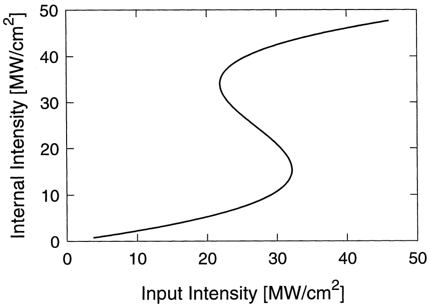

Fig. 3. The predicted bistable behavior of a thin film of DR1 functionalized PMMA ($\chi^{(3)} = -7.8\times10^{-8} - 3.0\times10^{-8}$i esu).

rence of bistability. Fig. 2 shows these results. For values of ζ within the shaded region of Fig. 2(a), $\mathrm{d}f/\mathrm{d}\eta = 0$ has two real, positive solutions. The shaded regions of Fig. 2(b) and Fig. 2(c) correspond to values of ζ where $f(\eta_1) > 0$ and $f(\eta_2) < 0$, respectively. The intersection of these three regions is shown in Fig. 2(d); it is in this region that optical bistability is predicted. Together, the two conditions above place a restriction on the phase of ζ, namely that $150^\circ < \arg\{\zeta\} < 210^\circ$. Therefore, to achieve intrinsic bistability, the phase difference between $\chi^{(3)}$ and ϵ must be between 150° and 210°. That is, the phase of $\chi^{(3)}$ should be nearly opposite to that of ϵ (as it is, for instance, for any nonlinearity resulting from saturation effects). In addition, the magnitude of ζ must lie within a certain range that varies slightly with $\arg(\zeta)$. As a general rule, bistability exists when the ratio of $|\chi^{(3)}||E_0|^2$ to $|\epsilon|^3$ is of the order of 10^{-3}.

Let us now consider the experimental conditions under which optical bistability due to local field effects might be observed. For a lossless dielectric material, the dielectric constant is real and positive, and thus the occurrence of bistability requires that the third-order susceptibility be predominantly real and negative. Although it was once believed that the third order susceptibility had to be positive for a lossless dielectric material [8], there are now several known examples of materials with a negative third-order susceptibility [9–11]. For instance, Rangel-Rojo et al. reported a value of $\chi^{(3)} = (-1.1 - 0.415\mathrm{i}) \times 10^{-15}$ m^2/V^2 $= (-7.8 - 3.0\mathrm{i}) \times 10^{-8}$ esu for a 10% concentration of disperse red one (DR1) functionalized into poly-methyl methacrylate (PMMA). For a thin film of this material, the desired value of ζ can be obtained by supposing an incident intensity of 30 MW/cm^2 and assuming that the index of refraction of DR1-functionalized PMMA is same as that of pure PMMA ($n = 1.49$). For this case $I_0 \equiv |E_0|^2 = 3.8 \times 10^{13}$ V^2/m^2 and ζ is calculated to be $-0.14 - 0.054\mathrm{i}$, which lies in the shaded region of Fig. 2(d). Therefore, three different values of I satisfy the boundary conditions for this value of I_0. Thus this system is expected to show optical bistable behavior over some range of incident intensities about 30 MW/cm^2, as shown in Fig. 3. Other highly nonlinear candidate materials for observing bistability of this type include the noble metals, semiconductors, and atomic vapors. Our analysis predicts that the internal intensity can be a multivalued function of the input intensity. This effect could be observed experimentally either by monitoring the intensity scattered from the interior of the sample or perhaps by monitoring the nonlinear phase shift imposed on the transmitted laser beam.

In summary, we have presented a simple calculation which demonstrates the possibility of optical bistability in a single layer by means of local field effects. To show optical bistability, the phase difference between $\chi^{(3)}$ and ϵ should be larger than 150° and smaller than 210°, while $|\chi^{(3)}||E_0|^2/|\epsilon|^3$ should be on the order of 10^{-3}.

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