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Fabry–Perot bistability with nonlinear absorption and dispersion

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Calculations of light interaction with medium exhibiting both nonlinear absorption and dispersion based on one plane-wave and slowly varying envelope approximations are presented. A two level model of atoms (molecules) has been assumed as a basis for the nonlinear susceptibility which was then determined without expanding it in a series of the electric field. General formulae for the intensity of the transmitted wave are derived and some numerical calculations are performed. It has been shown that under this model the optical bistability is possible and some conditions for its onset have been established.

1. Formulation of the problem and theoretical background

This paper deals mainly with the optical bistability which still belongs to the most important optical nonlinear effects. There exists a huge literature on this subject, concerning its theoretical, experimental and practical aspects. Excellent reviews of this field may be found in many monographs and textbooks (see, for instance, [1]–[3]), where the reader is referred to for more details.

The main effects of optical nonlinearities on the propagation of a monochromatic light beam are modifications of the refractive index and absorption coefficient by the addition of terms that depend on the optical intensity. The light beam itself changes the properties of the material through which it propagates to produce several kinds of self-inflicted alterations of its propagation characteristics. The changes of both the refractive index and absorption coefficient provide mechanisms for optical bistability [4], [5]. Usually, they are considered separately, leading to two classes of bistability – dispersive and absorptive ones.

Purely dispersive bistability is described, for instance, in [4]–[6]. The model used therein is, however, limited to Kerr-like media, *e.g.*, those with linear absorption and quadratic (with respect to electric field) refractive index. On the other hand, purely absorptive bistability was analysed in [4], [5], [7] on the basis of two-level model.

Optical properties are usually analysed in terms of susceptibility tensor χ. The linear relation between field and polarization is generalised for large fields to an expression of the form

$$P = \varepsilon_0\{\chi^{(1)} + \chi^{(2)} + \chi^{(3)}E + \ldots\}$$

where $\chi^{(2)}$, $\chi^{(3)}$, *etc.*, are components of nonlinear susceptibility. For materials with

the centrosymmetric structure only odd-order terms survive. Then the above expressions is essentially an expansion in powers of the beam intensity. An example of such media are glassy polymers, which are model materials for our later calculations.

For two-level saturable systems, one cen derive [8] a closed formula for χ of the form

$$\chi(|E|^2) = \chi_d + i\chi_a = \frac{\alpha(\Delta + i)}{1 + \Delta^2 + \frac{|E|^2}{I_s}} \tag{1}$$

where α is the linear absorption coefficient, I_s – saturation intensity. The quantity

$$\Delta = (\omega - \omega_0)\tau = 2\pi\left(\frac{1}{\lambda} - \frac{1}{\lambda_0}\right)\tau \tag{2}$$

(τ – relaxation time) is a measure of the difference between the incoming frequency ω and the resonant frequency ω_0 of the system. The real part of this tensor is responsible for the dispersion and the imaginary part – for the absorption of the material. The magnetic permeability is taken to be 1.

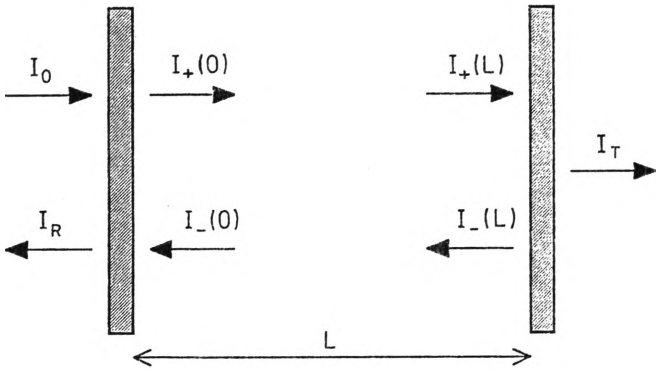

Fig. 1. Geometrical relations in the system considered

Now we consider a monochromatic plane wave propagating along the Oz axis (Fig. 1) whose (one) electric component has the following form:

$$E(z,t) = E_+(z)e^{i(\omega t - kz)} + E_-(z)e^{i(\omega t + kz)}. \tag{3}$$

Here we have one wave propagating forwards (with the amplitude E_+) and one going backwards (with the amplitude E_-).

The electric field fulfils the standard wave equation

$$\nabla^2 E - \mu_0 \frac{\partial^2}{\partial t^2}(\varepsilon_0 E + P_L) = \mu_0 \frac{\partial^2 P_{NL}}{\partial t^2}. \tag{4}$$

We now use the slow-varying amplitude approximation:

$$\left|\frac{d^2E_+(z)}{dz^2}\right| \ll 2k\left|\frac{dE_+(z)}{dz}\right|, \quad \left|\frac{d^2E_-(z)}{dz^2}\right| \ll 2k\left|\frac{dE_-(z)}{dz}\right|. \tag{5}$$

After some algebra we get the following equation for the (complex) amplitudes:

$$e^{-ikz}\left(-2ik\frac{dE_+(z)}{dz}-k^2E_+(z)+\mu_0\varepsilon_0\omega^2E_+(z)\right)$$
$$+e^{ikz}\left(2ik\frac{dE_-(z)}{dz}-k^2E_-(z)+\mu_0\varepsilon_0\omega^2E_-(z)\right)=$$
$$=-\frac{\mu_0\omega^2\alpha(\Delta+i)(E_+(z)e^{-ikz}+E_-(z)e^{ikz})}{1+\Delta^2+\dfrac{|E_+(z)|^2+|E_-(z)|^2+E_+(z)E_-^*(z)e^{-2ikz}+E_+^*(z)E_-(z)e^{2ikz}}{I_s}}. \tag{6}$$

Let us now introduce the following abbreviations:

$$a=\frac{\mu_0\varepsilon_0\omega^2}{k}-k, \quad b=\frac{\mu_0\omega^2\alpha}{k}, \quad c=1+\Delta^2, \tag{7}$$

$$\frac{E_+(z)}{\sqrt{I_s}}=\tilde{E}_+(z), \quad \frac{E_-(z)}{\sqrt{I_s}}=\tilde{E}_-(z). \tag{8}$$

The amplitudes $\tilde{E}_+(z)$ and $\tilde{E}_-(z)$ may be further written as products of real amplitudes ρ and phase factors according to the formulas:

$$\tilde{E}_+(z)=\rho_+(z)e^{i\varphi_+(z)},$$
$$\tilde{E}_-(z)=\rho_-(z)e^{i\varphi_-(z)}, \tag{9}$$

($\varphi_+(z)$, $\varphi_-(z)$ – phases).

Averaging over space and separating real and imaginary parts lead to four new equations:

$$-\frac{d\ln(\rho_+^2)}{dz}(c+\rho_+^2+\rho_-^2)+\frac{d\rho_-^2}{dz}+b=0, \tag{10a}$$

$$2\frac{d\varphi_+}{dz}(c+\rho_+^2+\rho_-^2)+a(c+\rho_+^2+2\rho_-^2)-2\rho_-^2\frac{d\varphi_-}{dz}+b\Delta=0, \tag{10b}$$

$$\frac{d\ln(\rho_-^2)}{dz}(c+\rho_+^2+\rho_-^2)-\frac{d\rho_+^2}{dz}+b=0, \tag{10a}$$

$$-2\frac{d\varphi_-}{dz}(c+\rho_+^2+\rho_-^2)+a(c+\rho_+^2+2\rho_-^2)+2\rho_+^2\frac{d\varphi_+}{dz}+b\Delta=0. \tag{10b}$$

The squared (real) amplitudes may be thought of as intensities of the corresponding waves:

$$I_+=\rho_+^2, \quad I_-=\rho_-^2. \tag{11}$$

From Eqs. (10) we obtain the following relations for the above intensities:

$$-\frac{d\ln I_+}{dz}(c+I_++I_-)+\frac{dI_-}{dz}+b=0, \tag{12a}$$

$$\frac{d\ln I_-}{dz}(c+I_++I_-)-\frac{dI_+}{dz}+b=0. \tag{12b}$$

By substracting them we get immediately

$$\frac{d}{dz}[\ln(I_-I_+)-\ln(c+I_++I_-)]=0. \tag{13}$$

In this way we obtain a conservation low

$$\frac{I_+(z)I_-(z)}{(c+I_+(z)+I_-(z))}=C_1(R,I_0,L)=\text{const.} \tag{14}$$

Now we make use of the boundary conditions which are formulated in the following way:

$$\begin{aligned} &I_+(0)=TI_0+RI_-(0),\\ &I_-(L)=RI_+(0),\\ &I_T=TI_+(L) \end{aligned} \tag{15}$$

where I_0 is the incident intensity, I_T – transmitted intensity. These relations allow us to express backward waves by forward ones:

$$I_-(z)=\frac{RI_+^2(L)(c+I_+(z))}{I_+(z)(c+I_+(L)(1+R))-RI_+^2(L)}, \tag{16}$$

$$\frac{dI_-(z)}{dz}=-\frac{RI_+^2(L)(c+I_+(L))(c+I_+(L)R)}{(cI_+(z)-I_+^2(L)R+I_+(L)I_+(z)+RI_+(L)I_+(z))^2}\frac{dI_-(z)}{dz}. \tag{17}$$

The equation for forward intensities takes the form

$$\begin{aligned} &-\left\{\frac{1}{I_+(z)}\left(c+I_+(z)+\frac{RI_+^2(L)(c+I_+(z))}{I_+(z)(c+I_+(L)(1+R))-RI_+^2(L)}\right)\right.\\ &\left.+\frac{RI_+^2(L)(c+I_+(L))(c+RI_+(L))}{(I_+(z)(c+I_+(L)(1+R))-RI_+^2(L))^2}\right\}\frac{dI_+(z)}{dz}+b=0. \end{aligned} \tag{18}$$

The integration of this equation leads to the general formula

$$\int_{I_+(0)=TI_0+RI_-(0)}^{I_+(L)=I_T/R} f(I_+)dI_+=-bL \tag{19}$$

which gives the relation between two unknowns I_0, I_T

$$F(I_0,I_T)+bL=0. \tag{20}$$

The bistable state of the system is determined by the values of the following parameters:

R – reflection coefficient of the mirror ($R \in [0,1]$),

$c = 1+\Delta^2$ – parameter describing the detuning of frequencies ($c = 1$ in the case of perfect resonance),

the factor bL, where:

L – the length of the Fabry–Perot cavity,

$b = \dfrac{\mu_0 \omega^2 \alpha}{k}$ – low-intensity absorption coefficient of the material.

The above equations are the basis for the numerical analysis of the bistable effects.

2. Analysis of histeresis loops

Figures 2–4 show the results obtained for different values of parameters R and bL. We have divided them into three groups:

1. $c = 1.1$, $R = 0.95$, bL varies in the range from 0.1 to 3.1 (Fig. 2).
2. $c = 1$, $bL = 5$, $R \propto 0.5-0.9$ (Fig. 3).
3. $bL = 5$, $R = 0.95$, $c \propto 1-10$ (Fig. 4).

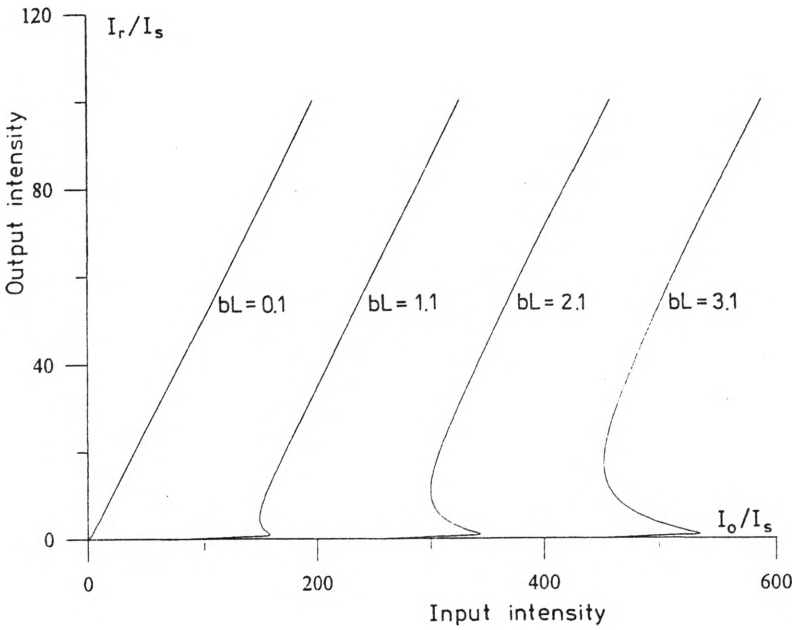

Fig. 2. Dependence of transmitted power on incident one for different values of the parameter bL. Both quantities are related to the saturation power of the cavity ($R = 0.95$, $c = 1.1$)

Figure 2 corresponds to the optimal situation for appearing of the bistability: $c = 1.1$ (the system close to the resonance) and $R = 0.95$. It is easy to see that the critical value of bL, leading to the bistability, is $bL \approx 1$. For lower values this effect is impossible independently of the intensity of incident wave.

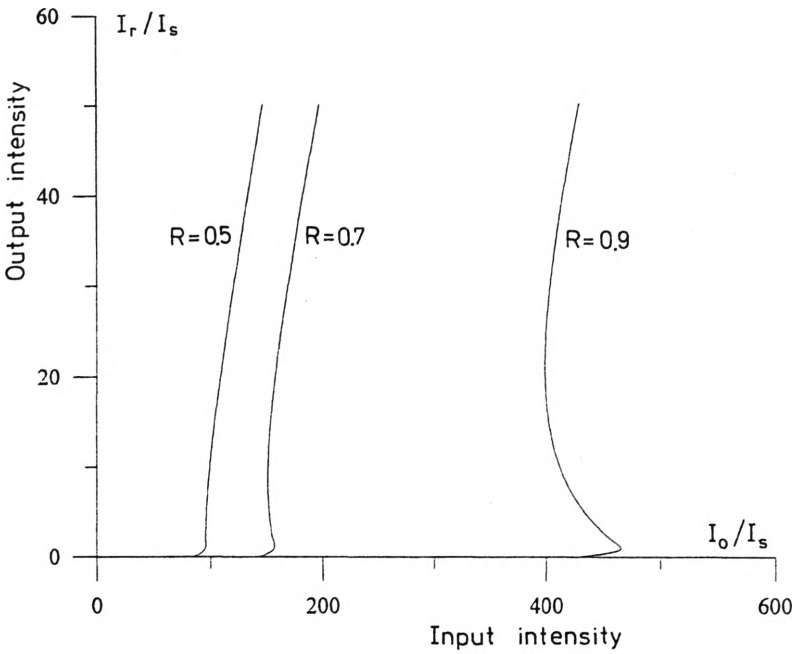

Fig. 3. The same as in Fig. 2, but for R as a parameter ($c = 1$, $bL = 5$)

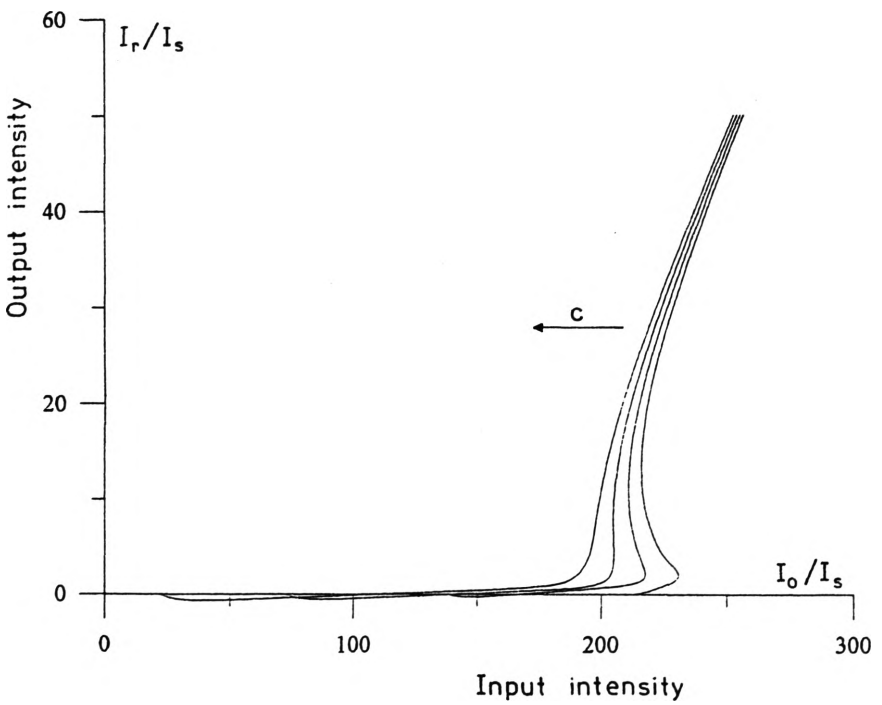

Fig. 4. Influence of c on the shape of bistable curves

Figure 3 corresponds to $c = 1$ (resonance) and $bL = 5$ (large absorption) A critical curve appears already for $R = 0.5$. By increasing this parameter up to $R = 0.9$, the size of the histeresis loop also increases. The jump is relatively high – almost two times for $R = 0.9$.

Figure 4 reflects the tendency to diminishing bistable effects with increasing c. It influences mainly the shape of central parts of the curves.

3. Optical bistability in the polymer SINC

In the paper [9], the results of experimental investigations of optical bistability in a glassy polymer named SINC are reported. It belongs to the noncentrosymmetric materials, thus exhibiting the nonlinearity of the third order, as in our model. The material was placed in the Fabry–Perot cavity (Fig. 1).

The authors of [5] used the following values of physical parameters: $I_S = 4.4 \times 10^9$ W/m^2, $bL = 1.4$, resonant wavelength $\lambda_0 = 810$ nm, laser wavelength $\lambda = 811$ nm, hence $c \simeq 1$.

Coefficient R was not given and will be treated here as a parameter (typically it equals to 0.95 for standard Fabry–Perot cavity).

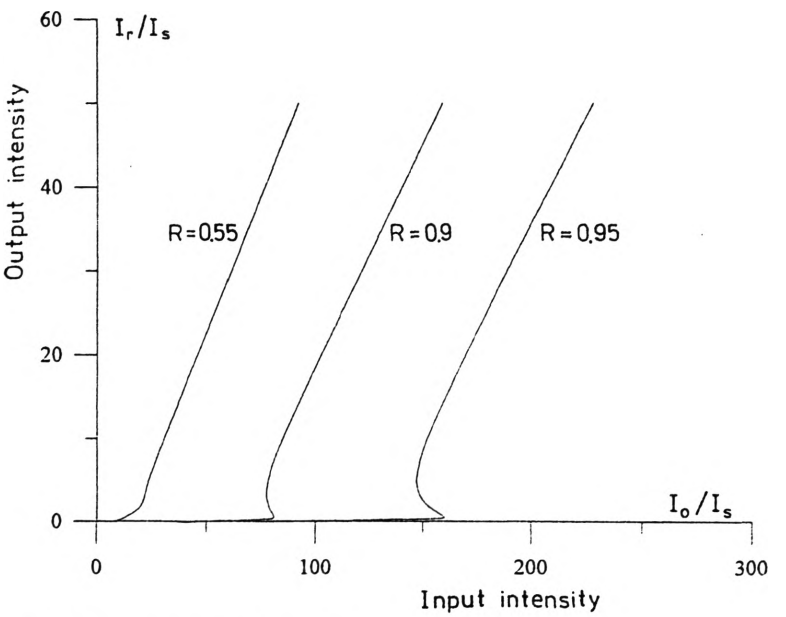

Fig. 5. Bistable curves for polymer SINC

In Figure 5, we present a number of curves $I_T(I_0)$ for $R \propto 0.55-0.9$, which are solutions of Eq. (20) with respect to I_0 (with $I_T \propto 0-50$).

These results confirm the possibility of bistability in such material, provided that $R > 0.7$. Above this value loops grow and the jump gets higher. For the critical curve, $I_0 = 1.32 \times 10^{11}$ W/m^2.

4. Conclusions

Theoretical considerations and numerical calculations based on them show that the optical intensity bistability is possible in systems described by two-level atoms model. Crucial role is played here by such parameters as the difference of frequencies of the resonant and of the incident waves, reflection coefficient of the Fabry–Perot cavity, absorption coefficient and the length of the cavity. In organic materials the values of these parameters may be properly suited in order to create the bistable effects.

Using the approximation of a two-level system may at first seem unreasonable for such a complex entity as a molecule containing many atoms. In practice, it is often the case that one of the molecular excited states has a dominant contribution to the nonlinearity. This state usually corresponds to a major $\pi-\pi^*$ transition, involving a redistribution of the delocalized electrons of the conjugated electron chain. Using this model, principal optical parameters have been successfully calculated (see, for instance, [10]).

In our considerations, we have in fact taken into account two mechanisms of nonlinearity – the dispersion together with the absorption. They may act independently but there is also a contribution arising from their interaction. It seems that our model is more realistic than others discussed in the literature.

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