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Optical bistability in $\chi^{(2)}$ nonlinear media due to cascaded process

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We present a model calculation of bistable light transmission through nonlinear Fabry–Perot cavity filled with media without a centre of symmetry. The susceptibility tensor of the third rank is assumed as the dominant nonlinear quantity. The model is based on the cascade-process. Two special cases are considered: interaction of two waves (incoming and its second harmonic) and that of three waves (two incoming beams of different frequencies and the third beam of difference frequency). The formulas for the intensity-dependent refractive indices are derived. With appropriately chosen parameters we obtain formulae for the transmitted intensity, which are analogous to the customary ones for Kerr-like media with symmetry centre.

1. Introduction

Optical bistability was first discovered in media with the third-order nonlinearity [1]–[3]. With such a nonlinearity the refractive index depends on the light intensity. The second-order bistability has focused much less attention [4]–[6]. The mechanisms leading to it are different from those of the third-order bistability. To explain second-order bistability it seems necessary to consider the so-called cascade-process.

In the simplest case such process is connected with the second-harmonic generation. In the situation of sufficiently large wave-vector mismatch, the generated wave of frequency 2ω is of a weak intensity and does not essentially change the amplitude of the incoming wave of frequency ω. However, its influence on the phase of the incoming wave is crucial. This phase change, together with an appropriate feedback is sufficient for bringing about the intensity bistability.

In the case when the sample is illuminated by two waves of different frequencies ω_1 and ω_2, the role of the second-harmonic wave is taken over by the wave with frequency $\omega_3 = \omega_1 - \omega_2$. This wave changes the phases of the two incoming waves in such a way that their intensities may exhibit bistability.

2. Two-waves interaction in the second-harmonic generation regime

We assume that the sample of nonlinear crystal is illuminated by a strong beam normal to the surface of the film. The wave is characterized by frequency ω, wave vector $\mathbf{k}_\omega = (0,0,k_\omega)$ parallel to the z-axis of Cartesian coordinate system. The second-order nonlinearity of the material is described by the susceptibility tensor of third rank $\chi^{(2)}$.

In order to simplify calculations, we assume that this tensor reduces to one component $\chi^{(2)}(2\omega;\omega,\omega) = \chi^{(2)}(\omega;2\omega,-\omega) \equiv \gamma$ Due to the nonlinearity the fundamental wave with electric field vector $\mathbf{E}_\omega$ and its second harmonic $\mathbf{E}_{2\omega}$ are coupled and there appears a mutual energy transfer. This transfer may exhibit periodicity along the z-axis and was named a cascaded process. It was suggested in [7]–[9] that the cascaded process may lead to nonlinear phase shifts of the fundamental beam. Since the phase shift arising from optical Kerr nonlinearity is one of the causes of dispersive optical bistability a bistable behaviour based on nonlinear phase shift in the cascading process is expected.

In the slowly-varying-envelope approximation, Maxwell's equations applied to a noncentrosymmetric and lossless crystal lead to coupled amplitude equations, which govern the second-harmonic generation process [3]

$$\frac{dE_\omega}{dz} = -i\frac{\omega\mu_0 c}{2n_\omega}\chi^{(2)}(\omega;2\omega,-\omega)E_{2\omega}E_\omega^*\exp(-i\Delta kz),$$

$$\frac{dE_{2\omega}}{dz} = -i\frac{\omega\mu_0 c}{n_{2\omega}}\chi^{(2)}(2\omega;\omega,\omega)E_\omega^2\exp(i\Delta kz) \tag{1}$$

where $\Delta k = 2\omega(n_\omega - n_{2\omega})/c$ is the wave-vector mismatch and n_ω, $n_{2\omega}$ are (linear) refractive indices of the material for both frequencies.

Our main assumption is that of a small conversion efficiency $|E_{2\omega}|^2/|E_\omega|^2 \ll 1$ which demands the condition on the sufficiently large wave-vector mismatch $|\Delta k|^2 \gg \frac{(2\mu_0 c\omega\gamma)^2}{n_\omega n_{2\omega}}|E_\omega(z)|^2$. This implies the absence of depletion of the incident wave amplitude, $|E_\omega(z)| = \text{const}$. However, its phase φ_ω defined by the equality $E_\omega(z) = |E_\omega(z)|\exp(i\varphi_\omega(z))$ depends on z. The solution of the system (1) for the intensity-dependent phase shift $\delta\varphi_\omega(z) = \varphi_\omega(z) - \varphi_\omega(0)$ of the fundamental beam is

$$\delta\varphi_\omega(z) = -\frac{\omega^2\mu_0^2c^2\gamma^2 z}{4n_\omega n_{2\omega}\Delta k}\left\{1 - \frac{\sin(\Delta kz)}{\Delta kz}\right\}|E_\omega|^2. \tag{2}$$

This function demonstrates an oscillating length-of-path dependence of the intensity of the second harmonic wave.

In order to draw consequences from the result obtained above, we refer to the case of media with the third-order nonlinearity, for which the effective refractive index may be written in the form

$$n_\omega^{\text{eff}} = n_\omega + n_\omega^{(2)}(z)|E_\omega(z)|^2. \tag{3}$$

Considering the relation between the phase shift of the fundamental wave and the intensity-induced change of the refractive index $\delta\varphi_\omega(z) = \omega z(n_\omega^{\text{eff}} - n_\omega/c$, we find that

$$n_\omega^{(2)}(z) = -\frac{\omega\mu_0^2c^3\gamma^2}{2n_\omega n_{2\omega}\Delta k}\left\{1 - \frac{\sin(\Delta kz)}{\Delta kz}\right\}. \tag{4}$$

From these it follows that in media with second-order nonlinearity and with an appreciable wave-vector mismatch Δk, parallel with the dependence on the intensity of the wave, there appears dependence of the refractive index on the path length of the beam. An appreciable z-dependence of $n_{\omega}^{(2)}$ appears only for a sufficiently large phase-mismatch Δkz. We also observe that the intensity $|E_{2\omega}|^2$ of the second beam does not appear in the expression for the phase of the first wave.

3. Three-waves interaction in the regime of difference-wave generation

The method of calculation of the previous section may be extended to cover the case of interaction of three waves. We now assume that two waves of frequencies ω_1 (pump beam) and ω_2 (probe beam) enter a nonlinear medium. The nonlinear interaction between them leads to waves with frequencies combined from ω_1 and ω_2, in particular to the wave with frequency $\omega_3 = \omega_1 - \omega_2$. In the slowly-varying-envelope approximation. Maxwell's equations applied to this system lead to coupled amplitude equations:

$$\begin{aligned} \frac{dE_1}{dz} &= -i\,\frac{\mu_0 c\omega_1}{n_1}\,\chi^{(2)}(\omega_1;\omega_2,\omega_3)E_2E_3e^{i\Delta kz}, \\ \frac{dE_2}{dz} &= -i\,\frac{\mu_0 c\omega_2}{n_2}\,\chi^{(2)}(\omega_2;\omega_1,-\omega_3)E_3^*E_1e^{-i\Delta kz}, \\ \frac{dE_3}{dz} &= -i\,\frac{\mu_0 c\omega_3}{n_3}\,\chi^{(2)}(\omega_3;\omega_1,-\omega_2)E_2^*E_1e^{-i\Delta kz} \end{aligned} \tag{5}$$

where $\Delta k = (\omega_1 n_1 - \omega_2 n_2 - \omega_3 n_3)/c$ and $\chi^{(2)}(\omega_1) = \chi^{(2)}(\omega_2) = \chi^{(2)}(\omega_3) = \gamma$. The assumption of a small ω_1-wave depletion ($|E_1(z)|^2 \approx |E_1(0)|^2$) demands the condition

$$\left(\frac{(\Delta k)^2}{(\Delta k_{\mathrm{eff}})^2} - 1\right)\frac{|E_2(0)|^2}{E_1(0)|^2} \ll 1 \tag{6}$$

where $(\Delta k_{\mathrm{eff}})^2 = (\Delta k)^2 - \dfrac{4\mu_0^2c^2\gamma^2\omega_2\omega_3}{n_2n_3}|E_1|^2$.

Below we consider two special cases of the realisation of this condition

a) $|E_2(0)|^2 \sim |E_1(0)|^2$, $|\Delta k_{\mathrm{eff}}| \simeq |\Delta k| \gg \mu_0 c\gamma\sqrt{\dfrac{\omega_2\omega_3}{n_2n_3}}|E_1(z)|$. In this case we assume large wave-vector mismatch, which implies that the intensities of both two waves remain practically unchanged. Under this condition, we can derive the expressions for the phase shifts:

$$\delta\varphi_1(z) = \frac{\mu_0^2c^2\gamma^2\omega_1\omega_3 z}{n_1n_3\Delta k}\left(1 - \frac{\sin(\Delta kz)}{\Delta kz}\right)|E_2(0)|^2, \tag{7a}$$

$$\delta\varphi_2(z) = \frac{\mu_0^2c^2\gamma^2 z\omega_2\omega_3}{n_2n_3\Delta k}\left(1 - \frac{\sin(\Delta kz)}{\Delta kz}\right)|E_1(0)|^2. \tag{7b}$$

These can be related (similarly to Eqs. (2)–(4)) to intensity-dependent parts of refractive indices:

$$n_1^{(2)}(z) = \frac{\mu_0^2 c^3 \gamma^2 \omega_3}{n_1 n_3 \Delta k}\left(1 - \frac{\sin(\Delta kz)}{\Delta kz}\right), \tag{8a}$$

$$n_2^{(2)}(z) = \frac{\mu_0 c^3 \gamma^2 \omega_3}{n_2 n_3 \Delta k}\left(1 - \frac{\sin(\Delta kz)}{\Delta kz}\right), \tag{8b}$$

which are defined by the relations:

$$n_1^{\text{eff}} = n_1 + n_1^{(2)}(z)|E_2(z)|^2, \tag{9a}$$

$$n_2^{\text{eff}} = n_2 + n_2^{(2)}(z)|E_1(z)|^2. \tag{9b}$$

We observe that the refractive indices of both two waves are not functions of the corresponding intensity but they depend on the complementary one. This cross interdependence represents a new effect in comparison with (2). However, such behaviour is not contrary to (2), since in the second-harmonic generation regime the fundamental wave E_ω plays the role of both incoming waves E_1 and E_2.

b) $|E_2(0)|^2 \ll |E_1(0)|^2$, $|\Delta k| > \mu_0 c\gamma \sqrt{\frac{\omega_2\omega_3}{n_2 n_3}}|E_1(z)|$. In this case we assume that the intensity of the pump beam is much larger than that of the probe beam. Following the discussion leading to Eqs. (4), (8a), (8b) we obtain expressions for the intensity-dependent parts of refractive indices:

$$n_1^{(2)}(|E_1|, z) = \frac{\mu_0^2 c^3 \gamma^2 \omega_3}{n_1 n_3 \Delta k}\left(\frac{\Delta k}{\Delta^2 k_{\text{eff}}}\right)\left(1 - \frac{\sin(\Delta k_{\text{eff}} z)}{\Delta k_{\text{eff}} z}\right), \tag{10a}$$

$$n_2^{(2)}(z) = \frac{\mu_0 c^3 \gamma^2 \omega_3}{n_2 n_3 \Delta k}\left(1 - \frac{\sin(\Delta kz)}{\Delta kz}\right). \tag{10b}$$

We observe that within this approximation the formulas for intensity-dependent refractive indices (Eqs. (10a), (10b)) have similar structure to (8a) and (8b) derived within the former approximation. However, an important difference is that now $n_1^{(2)}$ depends also on the corresponding pump beam intensity $|E_1|^2$.

4. Optical feedback in a Fabry–Perot cavity

The basic assumption of our calculation is that of a large wave-vector mismatch Δk. In consequence, the amplitude of the incoming wave $|E_\omega|$ is practically constant. However, the phase φ_ω of the incoming wave exhibits a dependence on the path length and a linear dependence on the intensity $|E_\omega|^2$. The role of the second-harmonic wave $E_{2\omega}$ is to bring about the path-length dependence of the phase of the fundamental wave. The cascaded process is required for the appearence of the second-harmonic wave, without which intensity and path-length dependence of the phase of the fundamental wave cannot appear. The dependence of the nonlinear refractive index $n_\omega^{\rm eff}$ in (3) on the path length represents a new effect which is a direct consequence of the phase change $\delta\varphi_\omega(z)$ (Eq. (2)).

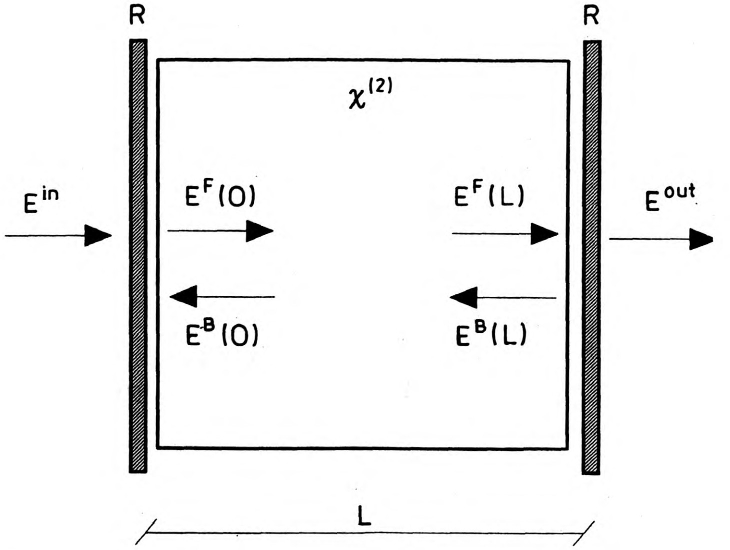

Fig. 1. Fabry–Perot optical resonator considered in this paper

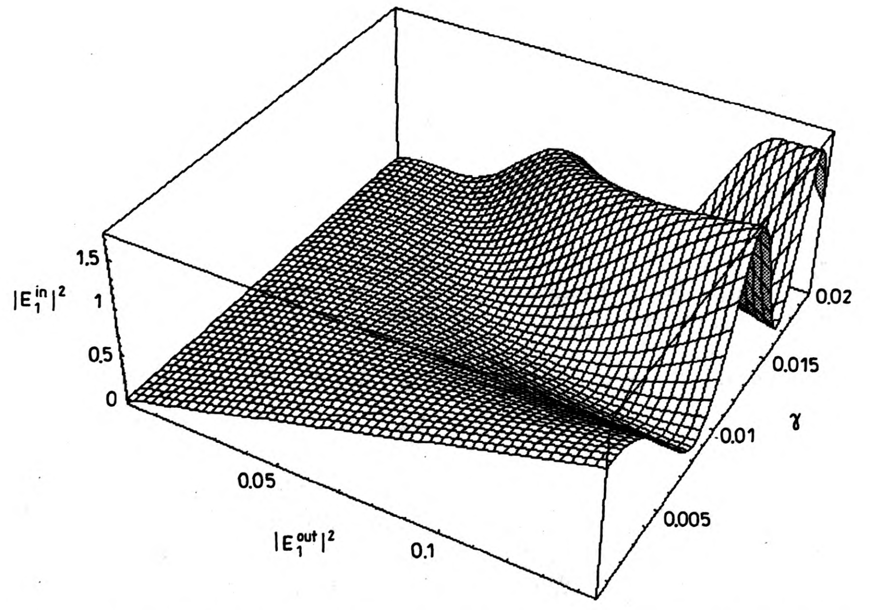

Fig. 2. Influence of nonlinearity parameter γ on the input–output cavity characteristics. Reflection coefficient of the mirrors is taken as $R = 50\%$, the length of the cavity is $L = 2\cdot10^{-4}$ m. The following units are used: $[E^2] = 10^{20}$ V^2/m^2, $[\gamma] = 10^{-20}$ C/V^2. These units apply for all the figures

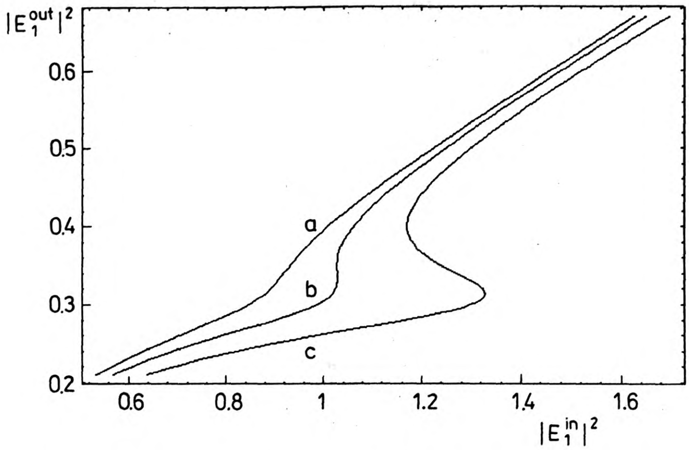

Fig. 3. Outgoing versus incoming intensity of the ω_1-wave for different values of the intensity of the incident wave with frequency ω_2: a — $|E_2^{in}|^2 = 0.01$, b — $|E_2^{in}|^2 = 0.02$, c — $|E_2^{in}|^2 = 0.04$

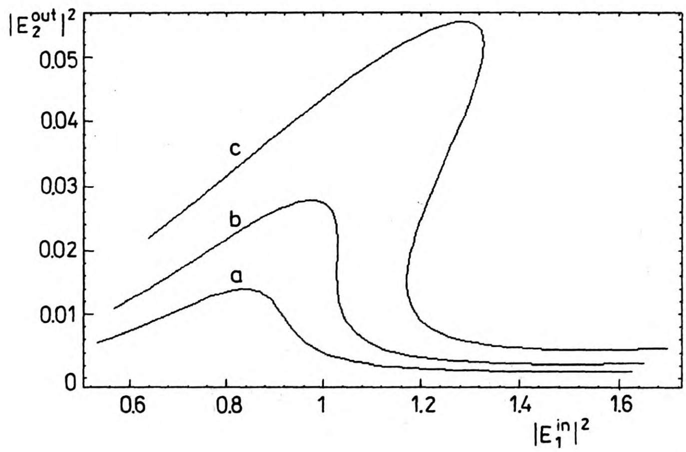

Fig. 4. Intensity of the outgoing ω_2-wave versus intensity of the incoming ω_1-wave for different values of the intensity of the incident wave with frequency ω_2: a — $|E_2^{in}|^2 = 0.01$, b — $|E_2^{in}|^2 = 0.02$, c — $|E_2^{in}|^2 = 0.04$

Now we consider the Fabry—Perot resonator filled with the $\chi^{(2)}$-nonlinear medium and illuminated with the field intensity E_ω (Fig. 1). As the intensity of the second-harmonic wave does not appear in (2), the presence of this wave in the Fabry—Perot cavity can be neglected. The mode of reasoning developed for lossless dispersive Kerr-like media with third-order nonlinearity [1], [2], can be directly applied in our case. The feedback process in the cavity can be described by considering only the forward and backward waves of frequency ω. In Figure 2, we plotted (in general multistable) transmittance characteristics of the Fabry—Perot cavity. The parameter of nonlinearity γ is taken as the control parameter of the phenomenon of bistability.

Bistability resulting from three-waves interaction was described in an analogous way. The wave with frequency $\omega_3 = \omega_1 - \omega_2$ takes over the role of the second-harmonic wave. This wave is indispensable for the appearance of the phase changes $\delta\varphi_1$ and $\delta\varphi_2$, however, due to its small intensity it can be neglected in this discussion of the feedback in Fabry–Perot cavity.

To obtain Figures 3 and 4, we assumed that the cavity is illuminated by a strong pump beam of frequency ω_1, the intensity of which on the input is varied, and the probe beam of frequency ω_2, which on the input remains constant. In Figure 3, we observe that changing the value of input probe beam intensity we influence the input–output characteristics of the Fabry–Perot cavity for the pump beam, from linear to bistable. We also observe (Fig. 4) that the intensity of the probe beam at the output is strongly influenced by the change of input pump beam intensity, and that the shape of this relation can exhibit bistability.

5. Conclusions

As a consequence of a sufficiently large wave-vector mismatch, the phase of the incoming wave (two waves in the case of non-degenerated three waves-interaction, respectively) exhibited a dependence on the path length and a linear dependence on the intensity of the fundamental beam. The dependence of the nonlinear refractive index on the intensity is a direct consequence of the phase change. As the intensity of the second-harmonic wave (difference wave, respectively) does not appear in the formula for the phase of fundamental wave (waves), the presence of this wave in the Fabry–Perot cavity can be neglected.

We thus have constructed a model mechanism of optical bistability in media without space-inversion centre, which exhibit second-order polarization. The number of such materials is large, however, their bistable properties were as yet not examined in a satisfactory manner. An experimental verification of the conclusions of this paper for several organic materials is reported by Pura *et al.* (to be published).

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References

[1] LUGIATO L., BONIFACIO R., [In] *Progress in Optics*, Vol. XXI, [Ed]. E. Wolf., North-Holland, Amsterdam 1981.
[2] GIBBS H. M., *Optical Bistability*, Academic Press, New York 1985.
[2] WHERRETT B. S., HUTCHINGS D. C., [In] *Nonlinear Optics in Signal Processing*, [Ed.] R. W. Eason, A. Miller, Chapman & Hall, London 1993.
[4] TRUTSCHEL J., *et al.*, IEEE J. Quantum Electron. **21** (1986), 1693.
[5] PURA B., ZAGÓRSKI A., PETYKIEWICZ J., Opt. App. **24** (1994), 229.
[6] REINISCH R., NEVIERE M., POPOV E., Phys. Lett. **20** (1995), 2472.
[7] DESALVO R., HAGAN D. J., SHEIK-BAHAE M., STEGEMAN G., VAN STRYLAND E. W., VANHERZEELE H., Opt. Lett **17** (1992), 28.

[8] STEGEMAN G., SHEIK-BAHAE M., VAN STRYLAND E. W., ASSANTO G., Opt. Lett. **18** (1993), 13.
[9] SCHIEK R., J. Opt. Soc. Am. B **10** (1993), 1848.

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