

# Hysteresis reflection and refraction by a nonlinear boundary—a new class of effects in nonlinear optics

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(Submitted June 7, 1976)

Pis'ma Zh. Eksp. Teor. Fiz. **24**, No. 3, 132–137 (5 August 1976)

A new effect is predicted—hysteresis jumps of the reflection and refraction of a strong wave at the boundary of a nonlinear medium. The jumps are due to transitions of the wave to a regime of total internal reflection and back. In the case of small nonlinearity, they should be observed at a small difference between the dielectric constants of the two media and at small grazing angles.

PACS numbers: 42.65. –k

1. We report here the possible existence of a new group of effects in nonlinear optics and electrodynamics, wherein, by varying the incidence angle or the intensity of a plane wave incident on the boundary of a nonlinear medium it is possible to observe hysteresis jumps from conditions of nonlinear total internal reflection (TIR) to a transmission regime, and vice versa. At a definite field intensity, the boundary can become fully transparent, regardless of the angle of incidence. In this case the amplitude of the hysteresis jumps ranges from total reflection to total transmission and back.

The hysteresis is due to multiple-valued character of the regimes in the system in the presence of nonlinearity. Multiple values are possessed by the transmission regime at  $\Delta\epsilon_{nl} < 0$  (two possible states), and by the nonlinear TIR at  $\Delta\epsilon_{nl} > 0$  (up to four states). These effects are not the consequence of a definite shape of the light-beam cross section (in contrast to self-focusing<sup>[1]</sup> or self-bending<sup>[2]</sup>). Therefore the principal results can be obtained by investigating the case of a homogeneous incident plane wave. We consider the incidence of such a wave with amplitude  $E$  from a linear medium with dielectric constant  $\epsilon_0$  at a grazing angle  $\psi$  onto the boundary of semi-infinite nonlinear medium having a dielectric constant  $\epsilon_1 = \epsilon_0 + \Delta\epsilon_l + \Delta\epsilon_{nl}$ , where  $\Delta\epsilon_{nl} = \epsilon_2 |E_1|^2$ ,  $E_1$  is the field amplitude in the nonlinear medium, and  $\Delta\epsilon_l$  is a field-independent increment to the dielectric constant. For the transmission regime, when a homogeneous plane wave travels also from the interface into the interior of the nonlinear medium,<sup>[1]</sup> at a transmission angle  $\psi_1$ , we have from Snell's law  $\cos\psi/\cos\psi_1 = [\epsilon_1(|E_1|^2)/\epsilon_0]^{1/2}$  under the condition

$$|\Delta\epsilon_l + \Delta\epsilon_{nl}| \ll \epsilon_0; \quad \psi \ll \pi/2, \tag{1}$$

(since usually  $|\Delta\epsilon_{nl}| \ll \epsilon_0$ ), and with allowance for the fact that under the condition the Fresnel formulas coincide for both polarizations of the incident wave,

$$\frac{E_1}{E} = p \approx \frac{2\psi}{\psi + \psi_1}; \quad r = p - 1 \approx \frac{\psi - \psi_1}{\psi + \psi_1}, \tag{2}$$

(where  $r$  and  $p$  are the amplitude reflection and transmission coefficients), substituting Eq. (2) in Snell's formula and leaving in it the first-order terms of smallness, we obtain the "nonlinear Snell's formula" for the transmission angle  $\psi_1$ :

$$(\psi + \psi_1)^2 \left( \psi^2 - \psi_1^2 + \frac{\Delta\epsilon_l}{\epsilon_0} \right) + 4 \frac{\epsilon_2 |E|^2}{\epsilon_0} = 0, \quad (3)$$

or the "nonlinear Fresnel formula" for the reflection coefficient  $r$

$$4r\psi^2 + \frac{\Delta\epsilon_l}{\epsilon_0} (1+r)^2 + \frac{\epsilon_2 |E|^2}{\epsilon_0} (1+r)^4 = 0, \quad (4)$$

which are valid for arbitrary polarization of the incident light.

3. For negative nonlinearity ( $\epsilon_2 < 0$ ), Fig. 1 shows in accordance with (4), in particular, plots of  $r(\psi)$  and  $r(E^2)$  in the simplest case  $\Delta\epsilon_l = 0$ , when the boundary is completely transparent to the weak field (i.e., in the linear regime  $r \equiv 0$  at arbitrary  $\psi$ —Fig. 1(a), line 1). In the nonlinear regime, hysteresis jumps from the transmission regime (curves 2) to the TIR regime (lines 3) and back appear. The curves for  $\psi_1$  can be plotted with the aid of formula (2), whence  $\psi_1 = \psi(1-r)/(1+r)$ .

In the general case, as seen from (3) and (4), the transmission goes over into TIR (i.e.,  $r \rightarrow 1$ ,  $\psi_1 \rightarrow 0$ ) either at  $\psi = 0$  or at  $\psi = \psi_{tr}$ , where

$$\epsilon_0 \psi_{tr}^2 + \Delta\epsilon_l + 4\epsilon_2 |E|^2 = 0. \quad (5)$$

In the case  $\epsilon_2 < 0$ , the angle  $\psi_{tr}$  is also the maximum possible TIR angle,  $\psi_{tr} = \psi_{TIR}$ , at which the jump from the TIR to the transmission takes place. At  $\Delta\epsilon_l = 0$  we have  $\psi_{tr} = 2|E| |\epsilon_2/\epsilon_0|^{1/2}$ , and the inverse jump takes place at  $\psi = \psi_{cr} = 4\psi_{tr}/3\sqrt{3}$ , in which case  $\psi_1 = \psi_{cr}/2$  and  $r = 1/3$ . For the  $\text{CdS}_{0.1}\text{Se}_{0.9}$  crystal, for example,  $n_2 = (\epsilon_2/2\sqrt{\epsilon_0}) = -1.1 \times 10^{-10}$  cgs esu,<sup>[4]</sup> in a field  $E \sim 10^6$  v/cm we obtain  $\psi_{tr} \approx 4.6^\circ$  and  $\psi_{cr} \approx 3.5^\circ$  at  $\sqrt{\epsilon_0} \sim 1.5$ .

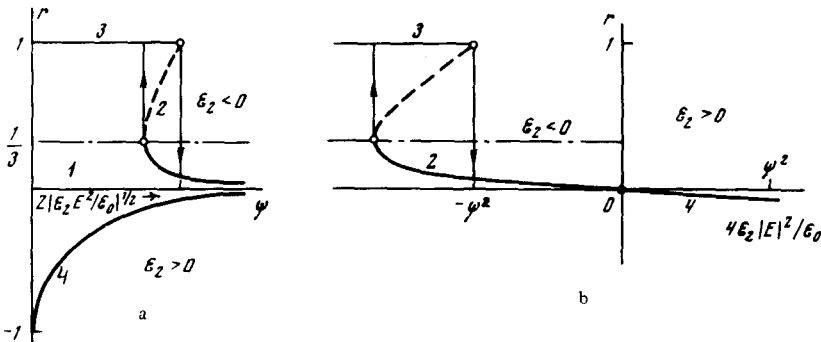


FIG. 1. Amplitude of reflection coefficient  $r$  vs. the grazing angle  $\psi$  at a fixed intensity of the incident field  $|E|^2$  (a), and vs.  $E^2$  at a fixed  $\psi$  (b), in the case  $\Delta\epsilon_l = 0$  (line 1 in Fig. 1(a),  $r \equiv 0$ , corresponds to total transparency in the linear care, curves 2 correspond to transmission at  $\epsilon_2 < 0$ , curves 3 to the TIR regime at  $\epsilon_2 < 0$ , and 4 to transmission at  $\epsilon_2 > 0$ ). All the dashed curves denote unstable branches, and the arrows show the directions of the hysteresis jumps.

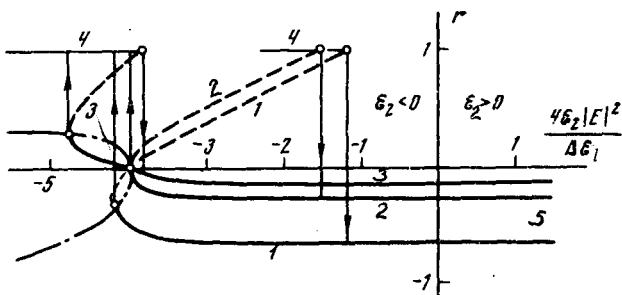


FIG. 2. Dependence of the reflection  $r$  on the intensity  $|E|^2$  at different angles  $\psi$  in the case  $\Delta\epsilon_1 > 0$ . The left-hand side of the figure corresponds to  $\epsilon_2 < 0$  (curve 1—transmission at  $\psi < \psi_0 \equiv (\Delta\epsilon_1/\epsilon_0)^{1/2}$ , 2—at  $\psi = \psi_0$ , 3—at  $\psi > \psi_0$ , 4—TIR regime), the right-hand part corresponds to  $\epsilon_2 > 0$  (curves 5—transmission at the same angles as on curves 1—3).

If  $\epsilon_2 < 0$ , the hysteresis appears at any sign of the linear mismatch. If  $\Delta\epsilon_1 > 0$ , then in sufficiently strong fields ( $|\epsilon_2 E^2| > \Delta\epsilon_1$ , meaning  $E > 0.8 \times 10^6$  V/cm at  $\Delta\epsilon_1 \sim 10^{-3}$  and  $n_2 \sim 10^{-10}$ ), the plot of  $r(\psi)$  has two hysteresis loops. If  $|\epsilon_2 E^2| = \Delta\epsilon_1$ , i. e., when the linear and nonlinear mismatches cancel each other completely,  $\Delta\epsilon_1 = -\Delta\epsilon_{nl}$ , total bleaching of the system takes place<sup>2)</sup> ( $r=0$  in Fig. 2, the point  $|\epsilon_2 E^2| = \Delta\epsilon_1$  on curves 1 and 2) at all angles  $\psi > \psi_0 = (\Delta\epsilon_1/\epsilon_0)^{1/2}$  amounting to  $\psi_0 \sim 1.2^\circ$  at  $\Delta\epsilon_1 \sim 10^{-3}$ ). Therefore jumps from  $r=0$  to  $r=1$  and back take place when  $\psi$  changes. Similar jumps from TIR to transmission occur also when  $E^2$  varies (if the angle  $\psi = \psi_0$  is fixed), at  $|\epsilon_2 E^2| = 3\Delta\epsilon_1/8$ , and back at  $|\epsilon_2 E^2| = \Delta\epsilon_1$  (Fig. 2, curve 2). The  $r(E^2)$  plot always has one hysteresis loop. The cases of transmission hysteresis at  $\epsilon_2 < 0$  and  $\Delta\epsilon_1 < 0$  are shown by curves 2 of Fig. 3.

The TIR regime ( $|r|=1$ ) in the case  $\epsilon_2 < 0$  and  $\psi < \psi_{\text{TIR}}$  corresponds to a near-surface linear wave with a uniquely defined profile, which decreases exponentially to zero at a sufficiently large distance from the boundary at  $\psi^2 < -\Delta\epsilon_1/\epsilon_0$ ,

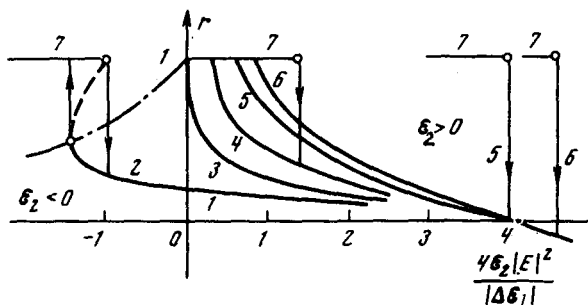


FIG. 3. Dependence of the reflection  $r$  on  $|E|^2$  at different angles  $\psi$  in the case  $\Delta\epsilon_1 < 0$ . Left— $\epsilon_2 < 0$  (curve 2—transmission at  $\psi > \psi_0 \equiv (|\Delta\epsilon_1|/\epsilon_0)^{1/2}$ , 7—TIR regime), on the right— $\epsilon_2 > 0$  (curve 1—transmission at  $\psi > \psi_0$ , 3—at  $\psi = \psi_0$ , 4—at  $1 > \psi > \psi_0 > \sqrt{2}/2$ , 5—at  $\psi = \psi_0/2\sqrt{2}$ , 6—at  $\psi < \psi_0/2\sqrt{2}$ , and 7—TIR).

and to a constant value

$$|E_{1\infty}| = \left[ \left( \psi^2 + \frac{\Delta\epsilon_1}{\epsilon_0} \right) \frac{\epsilon_0}{|c_2|} \right]^{1/2},$$

which does not depend on the amplitude of the incident field  $E$ , at  $\psi_{\text{TIR}}^2 > \psi^2 > -\Delta\epsilon_1/\epsilon_0$ .

4. In the case of positive nonlinearity ( $\epsilon_2 > 0$ ), the transmission regime also changes strongly in the region of small  $\psi$  (curve 4 on Fig. 1(a) at  $\Delta\epsilon_1 = 0$  and curve 5 on Fig. 2 at  $\Delta\epsilon_1 > 0$ ). Now, however, the hysteresis jumps are located only in the region of negative linear mismatches,  $\Delta\epsilon_1 < 0$  (Fig. 3, curves 1 and 3-7).

At  $\epsilon_2 > 0$ , the hysteresis jumps are due to the ambiguity of the TIR regime. The jumps of  $\psi_1$  and  $r$  occur here only on going from TIR to transmission; in the opposite direction, there is only a jump in the phase of the reflection. By solving the nonlinear wave equation for the surface wave we find that the break from the TIR to the transmission regime occurs at

$$\begin{aligned} \psi = \psi_{\text{TIR}_1} &= [ (|\Delta\epsilon_1| - 2\epsilon_2 E^2) / \epsilon_0 ]^{1/2}, & \text{if } \epsilon_2 E^2 < \frac{1}{4} |\Delta\epsilon_1|, \\ \psi = \psi_{\text{TIR}_2} &= \frac{1}{2} |\Delta\epsilon_1| / E (2\epsilon_2 \epsilon_0)^{-1/2}, & \text{if } \epsilon_2 E^2 > \frac{1}{4} |\Delta\epsilon_1|. \end{aligned} \quad (6)$$

Comparison with (5) shows that at  $\epsilon_2 > 0$  we always have  $\psi_{\text{TIR}} > \psi_{\text{tr}}$ , in contrast with  $\epsilon_2 < 0$ , when  $\psi_{\text{TIR}} = \psi_{\text{tr}}$ . The wave profile in the nonlinear medium at  $\epsilon_2 > 0$  is given by

$$\begin{aligned} |E_1(z)| &= \gamma (2\epsilon_0 / \epsilon_2)^{1/2} / \text{ch}(k_0 \gamma z + C); & \gamma &= \left( \frac{|\Delta\epsilon_1|}{\epsilon_0} - \psi^2 \right)^{1/2}; \\ k_0 &= \omega \sqrt{\epsilon_0} / c, \end{aligned} \quad (7)$$

where  $z$  is the coordinate normal to the boundary and  $C$  is determined from the boundary conditions and can have from one value ( $C=0$  at  $\psi = \psi_{\text{TIR}_1}$ ) for four values (at  $\psi_{\text{TIR}_1}^2 < \psi^2 < \psi_{\text{TIR}_2}^2 < |\Delta\epsilon_1| / 2\epsilon_0$ ). Those solutions of (7) which have maxima inside the nonlinear medium are unstable. At  $\epsilon_2 > 0$  and  $\Delta\epsilon_1 < 0$  there exists also a field in which  $\Delta\epsilon_1 = -\Delta\epsilon_{nl}$  (i. e.,  $E_0 = |\Delta\epsilon_1 / \epsilon_2|^{1/2}$ ), and the system becomes completely transparent at all angles (the intersection point of curves 1 and 3-6 at  $E^2 = E_0^2$  on Fig. 3). The jump from TIR to transmission (from  $r=1$  to  $r=0$ ) takes place in this case at  $\psi_{\text{TIR}_2} = (1/2) |\Delta\epsilon_1 / 2\epsilon_0|^{1/2}$ . For  $\text{CS}_2$ , where  $n_2 = 9 \times 10^{-12}$  cgs esu, we have  $E_0 \sim 1.8 \times 10^6$  V/cm at  $\Delta\epsilon_1 \sim -10^{-3}$  in which case  $\psi_{\text{TIR}} \approx 0.54^\circ$ .

5. If the boundary is illuminated by a beam bounded in the transverse direction, then its threshold power  $P_{\text{thr}}$  can be estimated by starting from the condition that the angular diffraction of the beam over the length of its interaction with the boundary should be smaller than the difference between the angles of the hysteresis jumps. At  $\Delta\epsilon_1 = 0$  and  $\epsilon_2 < 0$  this yields

$$P_{\text{thr}} \approx \frac{27}{11} (c\sqrt{\epsilon_0} / 4 |\epsilon_2| k_0^2), \quad (8)$$

which is independent of the beam radius  $a$ . For  $n_2 \sim -1.1 \times 10^{-10}$  cgs esu and  $\lambda$

$\sim 0.7 \mu$  we have  $P_{\text{thr}} \sim 0.46 \text{ kW}$ , and the average angle near the threshold must be chosen to be of the order of  $\psi \sim (k_0 a)^{-1}$ .

6. The effects considered above can be widely used to investigate nonlinear properties of media and in laser technology.

1) They can be used to measure the nonlinearity coefficients  $\epsilon_2$  (or, conversely, the field intensity) with high accuracy, by recording the jumps, for example those observed on the oscillograms of the pulses.

2) An optical hysteresis cell constitutes in essence an optical trigger. When the light intensity is increased under certain conditions (Secs. 3 and 4) the trigger switches over at the instant of the jump from  $r=0$  to  $r=1$  and can therefore serve as an ideal shutter in a laser when giant pulses are generated (a—in the course of lasing itself, once  $r=1$  is reached, it absorbs practically no energy; b—it is not resonant and has furthermore a high operating speed, limited only by the nonlinearity of relaxation; c—the switched-over light intensity is registered simply by choosing the angle of incidence of the beam or by the value of  $\Delta\epsilon_1$ ).

3) In contrast to other self-action effects, the field in the TIR regime penetrates a small depth into the nonlinear medium,  $h \sim (\lambda/2\pi)(|\Delta\epsilon_1| - \epsilon_0\psi^2)^{-1/2}$  (7) ( $h_{\text{min}} \sim 5 \mu$  at  $\lambda \sim 1 \mu$  and  $\Delta\epsilon_1 \sim 10^{-3}$ ), making it possible to investigate the nonlinearity in substances with strong absorption (for example, this would make it possible to observe nonlinear effects at low powers in certain semiconductors near the edge of the absorption band).

4) For the same reason it is possible to study relaxation and fast oscillations of the polarization and of the population in a two-level system<sup>[5]</sup> over a small thickness of matter (i. e., in a given field) upon application of a short strong pulse that is at resonance with the transition.

5) The effects considered could possibly lead to realization of a nonlinear-spectroscopy modification, analogous to linear spectroscopy of internal reflection,<sup>[6]</sup> and possessing its principal advantages.

I am grateful to B. Ya. Zel'dovich, N. F. Pilipetskiĭ, and V. B. Sandomirskiĭ for a discussion of the results.

<sup>1</sup>If a plane homogeneous wave is incident, there can be no regimes other than transmission and TIR in the system; This is the consequence of the field-momentum conservation law generalized in<sup>[3]</sup> to include nonlinear media.

<sup>2</sup>This phenomenon cannot be the analog of Brewster bleaching, since it occurs at any angle  $\psi$  and does not depend on the polarization.

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# **Erratum: Hysteresis reflection and refraction by a nonlinear boundary—a new class of effects in nonlinear optics**

**[JETP Lett. 24, No. 3, 114–119 (5 August 1976)]**

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PACS numbers: 99.10.+g, 42.65.-k

Insert  $\psi^2$  in formula (3) in front of the equal sign.