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Self-reflection effect in semiconductors in a two-pulse regime

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***Abstract*. Peculiarities of reflection at the end face of a semi-infinite semiconductor in a two-pulse regime are studied. The reflection functions behave in a complex and ambiguous manner governed by the amplitudes of the fields of incident pulses. The possibility of a complete bleaching of the medium for the field in the M-band is predicted.**

Keywords: *exciton, biexciton, self-reflection.*

It was shown in Refs [1 – 4] that the self-reflection effect can exist in a semi-infinite medium simulated by a system of two-level atoms. Self-reflection means the appearance of a backward wave in an optically homogeneous semi-infinite medium in the spatially inhomogeneous distribution of the nonlinear refractive index induced by the field of the forward wave. Self-reflection also occurs in a system of excitons and biexcitons in semiconductors when the exciton – photon interaction, optical exciton – biexciton conversion, and the one-pulse and two-pulse two-photon excitation of biexcitons from the ground state of a crystal are taken into account [5 – 9]. It was shown that self-reflection is a consequence of the optical Stark effect responsible for renormalisation of the energy spectrum of the system at high-power laser excitation.

In this study, we present the main results of investigation of self-reflection of laser radiation in a semi-infinite CuCl crystal under conditions when one pulse with the field amplitude E_1 and frequency ω_1 excites excitons from the ground state of the crystal and another pulse with the field amplitude E_2 and frequency ω_2 converts excitons into biexcitons. The energies $\hbar\omega_1$ and $\hbar\omega_2$ of pulse photons differ by the binding energy of the biexciton. The parameters of both pulses remain unchanged during their propagation. The problem is to determine the coefficient of reflection at the end face of the crystal as a function of the excitation level taking self-reflection into account.

Nonlinear interaction of laser radiation with excitons and biexcitons causes renormalisation of the energy spectrum of a semiconductor, which leads to the dependence of the dielectric function of the crystal on the intensity of propagating radiation pulses. The interaction of the pulse fields with excitons and biexcitons in the crystal is described by the Hamiltonian [10]

$$H_{\text{int}} = -\hbar g(a^+E_1^+ + aE_1^-) - \hbar\sigma(b^+aE_2^+ + a^+bE_2^-), \quad (1)$$

where a and b are the amplitudes of exciton and biexciton polarisation waves; $E_{1,2}^+$, $E_{1,2}^-$ are the positive and negative frequency components of the wave fields; g is the constant of interaction of the field with excitons; and σ is the optical exciton – biexciton interaction constant. Using Eqn (1), we can write the Heisenberg equation for the amplitudes a and b in the steady-state regime and determine the polarisation of the medium and dielectric functions ε_1 and ε_2 of the crystal for each wave, which have the form

$$\varepsilon_1 = \varepsilon_{1\infty}\Bigg(1 - \psi \times\left\{\delta_1 - \frac{(\delta_1+\delta_2)|f_2|^2}{(\delta_1+\delta_2)^2+s^2} - \mathrm{i}\left[1 + \frac{s|f_2|^2}{(\delta_1+\delta_2)^2+s^2}\right]\right\} \times\left\{\left[\delta_1 - \frac{(\delta_1+\delta_2)|f_2|^2}{(\delta_1+\delta_2)^2+s^2}\right]^2 + \left[1 + \frac{s|f_2|^2}{(\delta_1+\delta_2)^2+s^2}\right]^2\right\}^{-1}\Bigg),$$

$$\varepsilon_2 = \varepsilon_{2\infty}\Bigg(1 - \psi r|f_1|^2(\delta_1+\delta_2-\mathrm{i}s)[(\delta_1+\delta_2)^2+s^2]^{-1} \times\left\{\left[\delta_1 - \frac{(\delta_1+\delta_2)|f_2|^2}{(\delta_1+\delta_2)^2+s^2}\right]^2 + \left[1 + \frac{s|f_2|^2}{(\delta_1+\delta_2)^2+s^2}\right]^2\right\}^{-1}\Bigg), \quad (2)$$

where $\varepsilon_{1\infty}$ and $\varepsilon_{2\infty}$ are the background dielectric functions of the semiconductor for each wave; $r = \varepsilon_{2\infty}/\varepsilon_{1\infty}$; $\psi = \omega_{\text{LT}}/\gamma_1$ is the longitudinal – transverse splitting frequency normalised to γ_1; $s = \gamma_2/\gamma_1$; γ_1 and γ_2 are phenomenologically introduced decay constants of the exciton and biexciton states, respectively; $\delta_{1,2} = \Delta_{1,2}/\gamma_1$ is the normalised resonance detuning for frequencies of each pulse; and $f_{1,2} = \sigma E_{1,2}/\gamma_1$.

The spatial distribution of fields in a medium under steady-state conditions is determined from the solution of wave equations, which have the following form for normalised field amplitudes $f_{1,2}$:

$$\frac{\mathrm{d}^2 f_1}{\mathrm{d}z^2} + \varepsilon_1 f_1 = 0, \quad \frac{\mathrm{d}^2 f_2}{\mathrm{d}z^2} + \alpha\varepsilon_2 f_2 = 0, \quad (3)$$

where $z = (\omega_1/c)x$; c is the velocity of light in vacuum; x is the coordinate in the direction of propagation of pulses; and $\alpha = \omega_2^2/\omega_1^2$. For the boundary conditions at the point

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$z = 0$, we use the condition of continuity of tangential components of the electric and magnetic fields:

$$f_{1i} + f_{1r} = f_1|_{z=0}, \quad f_{1i} - f_{1r} = -i\frac{\partial f_1}{\partial z}\bigg|_{z=0}, \tag{4}$$

$$f_{2i} + f_{2r} = f_2|_{z=0}, \quad \sqrt{\alpha}(f_{2i} - f_{2r}) = -i\frac{\partial f_2}{\partial z}\bigg|_{z=0}, \tag{5}$$

where f_{1i}, f_{2i} are normalised amplitudes of the incident waves; and f_{1r}, f_{2r} are the corresponding amplitudes of the reflected waves.

Let us now discuss the results of numerical solution of the system of equations (3) using expressions (2) and (4), (5), as well as the results of calculation of the reflection coefficients $R_1 = |f_{1r}/f_{1i}|^2$ and $R_2 = |f_{2r}/f_{2i}|^2$. Figure 1 shows the dependence of the reflection coefficients R_1 (Figs 1a–c) and R_2 (Figs 1d–f) on the amplitude f_{1i} of the incident pulse for various fixed values of f_{2i} under the conditions of exact resonance $\delta_1 = \delta_2 = 0$. One can see that for small amplitudes f_{2i} of the second wave which is responsible for the exciton–biexciton conversion, the reflection coefficient at the frequency ω_1 of the first wave increases monotonically, tending to a certain limiting value (Fig. 1a). The reflection coefficient at the frequency ω_2 of the second wave asymptotically tends to unity for large values of f_{1i} (Fig. 1d) since its dielectric function (2) is determined by the amplitude of the first wave. With increasing f_{2i}, the situation starts becoming complicated: first a three-valued hysteresis loop emerges for $f_{2i} = 10$, after which the number of loops starts increasing rapidly. Note that the multivalued nature of the reflection coefficient appears in a narrow range of values of f_{1i} whose threshold value is approximately equal to f_{2i}. Investigations show that the density of biexcitons attains its peak in this case, which is the reason behind the multiple values. It is also interesting to note that the reflection coefficients R_1 and R_2 are constants for small values of f_{1i} but for $f_{2i} \gg 1$ (Figs 1b, c, e, f). This is so because the dielectric functions ε_1 and ε_2 under these conditions practically do not depend on the wave intensities for $\delta_1 = \delta_2 = 0$ and are equal to $\varepsilon_{1\infty}$ and $\varepsilon_{2\infty}$.The vanishingly small imaginary corrections do not contribute towards the reflection coefficients.

One can also see that for $f_{1i} \gg f_{2i}$, the dependences $R_1(f_{1i})$ and $R_2(f_{2i})$ tend asymptotically to certain values that do not depend on the degree of excitation. This is due to the fact that in this limit ε_1 assumes a constant value that does not depend on f_{1i} and f_{2i}, while ε_2 turns out to be a purely imaginary quantity proportional to $i|f_{1i}|^2$. A purely

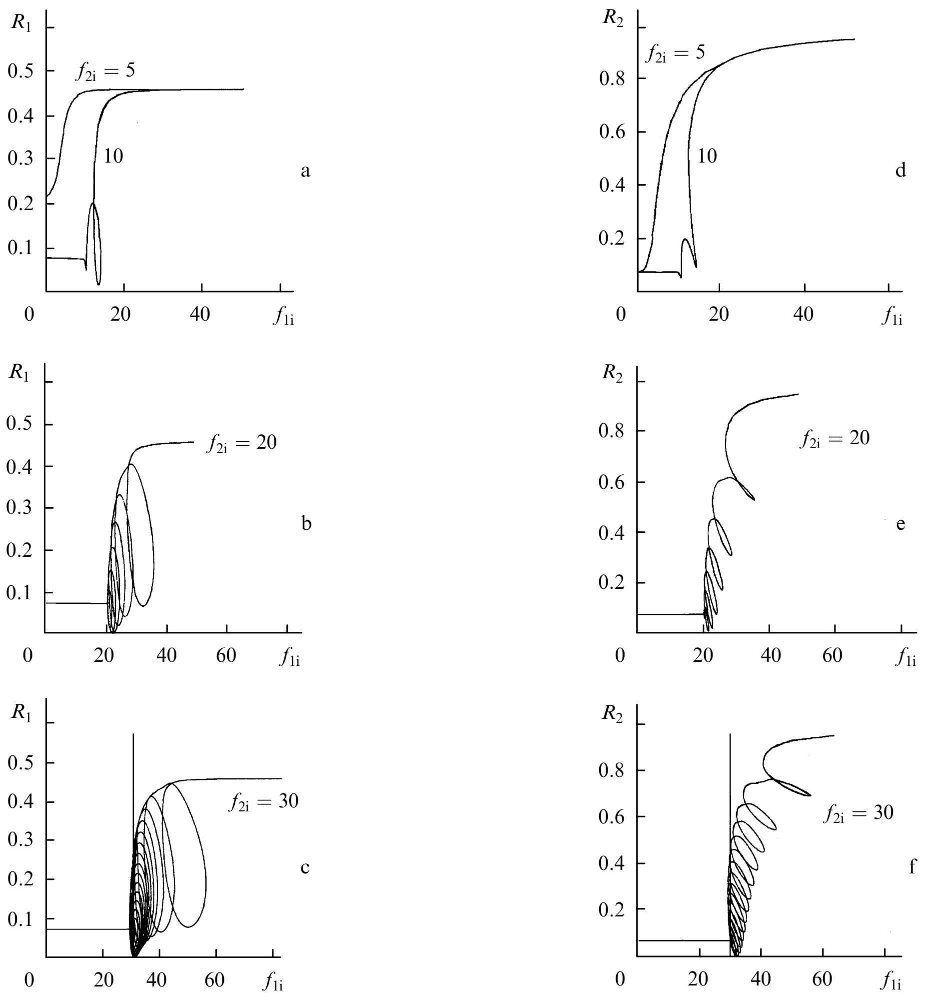

Figure 1. Dependences of the reflection coefficients R_1 (a–c) and R_2 (d–f) on the normalised field amplitude f_{1i} of incident radiation for $\varepsilon_{1\infty} = \varepsilon_{2\infty} = 3$, $\psi = 5$, $\alpha = 0.99$, $s = 2$, $\delta_1 = \delta_2 = 0$ and for various values of the amplitude f_{2i}.

imaginary dielectric function makes R_2 tend asymptotically to unity, while a complex but constant ε_1 leads to a value $R_1 < 1$.

Figure 2 shows the spatial profiles of the field amplitudes $f_{1,2}$, nonlinear refractive indices $n_{1,2}$, extinction factors $\kappa_{1,2}$, and the internal reflection coefficients $\rho_{1,2}$ under conditions of exact resonance ($\delta_1 = \delta_2 = 0$), corresponding to three different sets of values of R_1 and R_2 in Figs 1c and f. The cases considered below correspond to the energy reflection coefficients R_1, R_2 marked by vertical straight lines in Figs 1c and f. The normalised amplitudes of the incident waves are $f_{1i} = 32$, $f_{2i} = 30$. The first case (Figs 2a, d) corresponds to coefficients R_1 and R_2 of reflection at the end face of the crystal, which are indicated at the beginning of the unfolding spirals in Figs 1c and f. In this case (as well as in all the remaining cases), regions (domains) with high and low densities of quasiparticles exist in the medium. In the first of these regions (Figs 2 a, d), i.e. in domains with a high density of quasiparticles, the amplitudes of fields decrease monotonically, while the extinction coefficients and the nonlinear refractive indices remain constant. Upon a transition from the first to second domain, i.e., to a domain with a low density, the field amplitudes decrease exponentially, while the values of n_1 and κ_1 increase (Fig. 2a) and peaks are formed for n_2 and κ_2 (Fig. 2d). Peaks of total internal reflection factors are formed near the domain boundaries. The behaviour of fields in the second domains is associated with the absence of excitons without which biexcitons cannot be formed. Hence, while the first wave attenuates to zero, the second tends to a certain limit, propagating without attenuation as in an absolutely transparent medium. In this region, the value of κ_1 does not change, assuming nonzero values while κ_2 tends to zero, which also points towards the absence of absorption of the second wave.

The second case (Figs 2b, e) corresponds to reflection coefficients R_1 and R_2 that are further removed from the beginning of the unfolding spiral (Figs 1c, f). Weak oscillations of fields are observed in the domain with a high density of excitons and biexcitons, and auxiliary peaks of internal reflection coefficients are observed, attaining higher

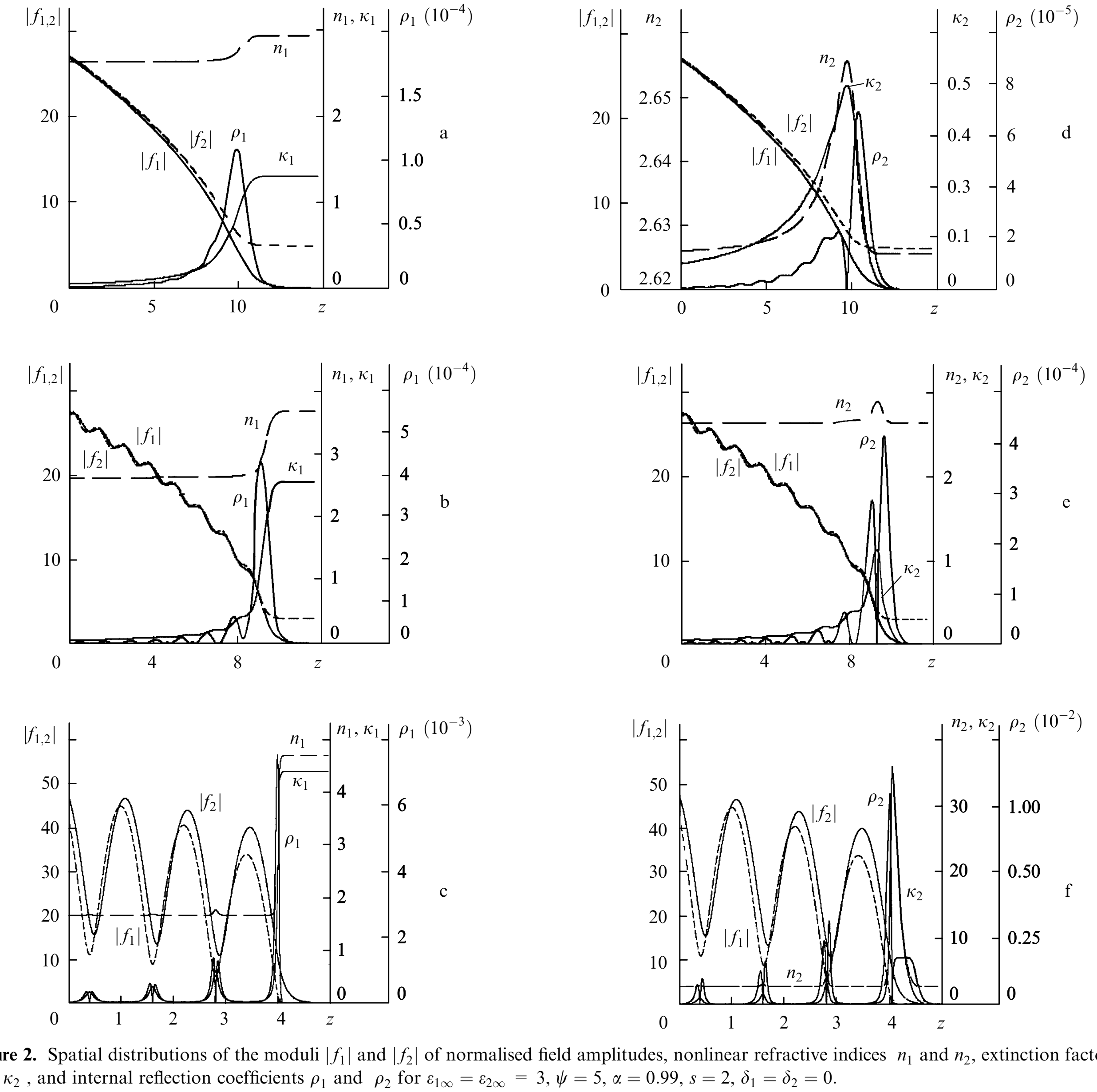

Figure 2. Spatial distributions of the moduli $|f_1|$ and $|f_2|$ of normalised field amplitudes, nonlinear refractive indices n_1 and n_2, extinction factors κ_1 and κ_2, and internal reflection coefficients ρ_1 and ρ_2 for $\varepsilon_{1\infty} = \varepsilon_{2\infty} = 3$, $\psi = 5$, $\alpha = 0.99$, $s = 2$, $\delta_1 = \delta_2 = 0$.

values than in the preceding case. The second wave decays more strongly, and the medium continues to be transparent to it.

In the third case (Figs 2c, f), the reflection coefficients R_1 and R_2 are farthest from the beginning of the unfolding spiral (Figs 1c, f). The fields in the first domain are characterised by a well-defined oscillatory behaviour in space (Figs 2 c, f), and the optical functions also oscillate, while the internal reflection coefficients attain quite large values. In the second domain, the field with amplitude f_2 completely decays.

In the case of departure from exact resonance conditions, the behaviour of the reflection coefficients does not change much. As before, there are some regions of less significance. However, hornlike structures described in [8, 9] are formed for some values of parameters. Similar peculiarities are observed in the spatial distribution of fields, extinction coefficients and nonlinear refractive indices. Note that the peaks of reflection coefficients assume colossal values for $\delta_1 \neq 0$ and $\delta_2 \neq 0$.

The observed peculiarities in the behaviour of coefficients of reflection from the end face of a crystal as functions of the level of excitation and spatial distribution of the field amplitudes are connected with renormalisation of the energy spectrum of the semiconductor for high levels of excitation. The oscillatory structure of the spatial distribution of fields is due to nonlinear dispersion. A considerable inhomogeneity in the field distribution determines the spatial inhomogeneity of nonlinear refractive indices, extinction factors and internal reflection coefficients. Consequently, narrow regions with high gradients of the refractive index of the medium are formed, where the backward waves are produced. The complex nonlinear interference of forward and backward waves leads to the above-mentioned steady-state structure of spatial field profiles. The narrow region with a large gradient of nonlinear refractive index and a corresponding sharp peak of internal reflection coefficient is nothing but a pump-field-induced Fabry–Perot resonator, the reflection at whose end faces leads to the emergence of multistability. The absence of such a resonator leads only to a single-valued nonlinear reflection function. The longitudinal size of the resonator is determined by the amplitude of fields in the medium.

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