

The features of light reflection from the interface of a semi-infinite nonlinear crystal upon two-photon excitation of biexcitons

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Abstract. The features of light reflection from the interface of a semi-infinite semiconductor are studied upon two-pulse, two-photon excitation of biexcitons from the crystal ground state. It is shown that the reflectivity is a complex multistable function of the amplitudes of the fields of incident pulses. The appearance of solitary closed domains or horn-shaped structures is predicted.

Keywords: self-reflection, exciton, biexciton, dielectric function, distributed feedback.

1. Introduction

A plane-parallel plate of a nonlinear semiconductor is characterised under certain conditions by the multistable functions of reflection and transmission of laser radiation [1–4]. This is caused by the generation of a backward wave reflected from the rear end of the plate, which interferes nonlinearly with the forward wave to produce conditions for multistable light transmission (reflection). At the same time, it was shown in papers [1, 5–9] that, in the approximation of the slowly varying spatial envelope of the amplitude of a propagating wave, the interface of a semi-infinite semiconductor is characterised in most cases by the one-valued nonlinear or bistable reflection function. As a rule, a backward wave does not arise in a semi-infinite optically homogeneous nonlinear medium in this approximation.

Note that the reflection and refraction of light beams with a finite cross section also reveal some additional features. In particular, it has been shown in [10] that narrow dips are formed in the transverse cross section of the reflected beams. The dips are caused by a nonlinear breakdown produced by a narrow 'stream', which comes off the interface of a semi-infinite medium, in the region of the maximum intensity of the incident beam, resulting in the hysteresis reflection. It was shown in [11] that in the region of the total reflection from a nonlinear medium, the transmitted beam is divided into several independent self-focused beams, whose number and propagation direction are determined by the incident radiation intensity.

Hysteresis phenomena in distributed nonlinear systems have been studied in most detail in [12], where the effects of the longitudinal and transverse distributions of systems, the kinetics of the spatial hysteresis, and the formation of spatiotemporal light structures have been investigated. A bistability and hysteresis in the reflection of a plane monochromatic wave from the semiconductor surface have been predicted theoretically in [13]. It was found that at high excitation intensities, the field in a semiconductor exhibits oscillations, which change to the aperiodic spatial decay at a large distance from the semiconductor end. As a result, a multiloop dependence of the reflectivity on the incident wave intensity arises.

It was shown in papers [14–20] that a breakaway from the approximation of slowly varying envelopes results in the additional features in the reflection function of radiation from a semi-infinite optically homogeneous nonlinear medium. Physically, this is caused by the fact that in this case it is possible to take into account the reflection from abrupt gradients of the nonlinear reflective index in a crystal at high excitation intensities, which leads to the generation of a backward wave accompanied by the nonlinear interference between the forward and backward waves, resulting in the multistability of the reflection function. This possibility has been also noted in [10–13].

The generation of the backward wave from abrupt gradients of the refractive index in the system of two-level atoms has been studied in detail in papers [14–18] and was called self-reflection. It was shown in [19, 20] that a similar effect also takes place in a system of excitons and biexcitons, if exciton–photon interaction, optical exciton–biexciton conversion and single-pulse two-photon excitation of biexcitons from the crystal ground state are taken into account.

2. Formulation of the problem and basic equations

Consider a nonlinear reflection function for the interface of a semi-infinite optically homogeneous isotropic semiconductor upon two-photon, two-pulse excitation of biexcitons from the ground state of a crystal. It is known [2, 21] that the oscillator strength for two-photon excitation of biexcitons is giant compared to that for the exciton transition, and the two-photon light absorption band has a narrow delta-like shape. For this reason, the contribution from nonresonance interactions is negligibly small compared to that from the resonance interaction, and the Stark effect in the Hamiltonian of interaction between excitons and photons can be neglected.

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Let two monochromatic laser pulses with the electric field envelopes E_{i1} and E_{i2} and the photon frequencies ω_1 and ω_2 , respectively, be incident normally on the vacuum–semiconductor interface. We assume that the photons of each of the pulses are resonant neither with the transition in the exciton spectral region nor with the transition from the exciton state to the biexciton one in the region of the M band [22]. However, we assume that the sum of the photon energies coincides with the excitation energy of a biexciton from the crystal ground state.

In the general case, when $\omega_1 \neq \omega_2$, two-photon excitation of biexcitons is possible only simultaneously by photons of both pulses, but not by photons of each pulse individually. A part of the radiation of the incident pulses enters into the medium and propagates in it, exciting biexcitons and interacting with them. The other part is reflected. The problem is to determine the amplitudes E_{r1} and E_{r2} of the reflected pulses or the reflectivities from the crystal end as functions of the amplitudes E_{i1} and E_{i2} of the incident pulses.

The Hamiltonian of interaction of biexcitons with the fields of both pulses has the form [2, 21]

$$H_{\text{int}} = -\hbar\mu(b^+ E_1^+ E_2^+ + b E_1^- E_2^-), \quad (1)$$

where μ is the constant of two-photon excitation of biexcitons [19, 25]; b is the amplitude of the biexciton wave; $E_{1,2}^+(E_{1,2}^-)$ is the positive (negative) frequency component of the amplitudes of the pulses propagating in the medium. Using (1), we can easily obtain the Heisenberg (constitutive) Eqn of motion for the amplitude b :

$$ib = \Omega_0 b - i\gamma b - \mu E_1^+ E_2^+, \quad (2)$$

where Ω_0 is the eigenfrequency of the biexciton excitation from the crystal ground state and γ is a phenomenological constant describing the decay of a biexciton state.

Solutions for all the waves in the stationary regime can be written in the form $E_1^+ \sim \exp(-i\omega_1 t)$, $E_2^+ \sim \exp(-i\omega_2 t)$, $b \sim \exp[-i(\omega_1 + \omega_2)t]$. Then, one can find from (2) the expression for the stationary amplitude b of the biexciton wave, and then the polarisations can be determined. After this, the complex dielectric functions ε_1 and ε_2 for each of the waves will be described by the expressions

$$\varepsilon_1 = \varepsilon_1' + i\varepsilon_1'' - \frac{4\pi\hbar\mu^2}{\Delta + i\gamma} |E_2|^2, \quad (3)$$

$$\varepsilon_2 = \varepsilon_2' + i\varepsilon_2'' - \frac{4\pi\hbar\mu^2}{\Delta + i\gamma} |E_1|^2, \quad (4)$$

where $\varepsilon_{1,2}'$ and $\varepsilon_{1,2}''$ are the real and imaginary parts of the background dielectric functions at the frequencies of each of the pulses; $\Delta = \omega_1 + \omega_2 - \Omega_0$ is the detuning between the sum of the pulse frequencies and the transition frequency. It follows from (3) and (4) that the dielectric function at the frequency of the first pulse is determined by the field of the second pulse, and vice versa, i.e., the dielectric functions at the frequency of each of the pulses contain Kerr cross-modulation nonlinear corrections.

For the sake of simplicity, we introduce the normalised quantities

$$F_{1,2} = \alpha E_{1,2}, \quad F_{i1,2} = \alpha E_{i1,2}, \quad F_{r1,2} = \alpha E_{r1,2}, \quad (5)$$

where $\alpha^2 = 4\pi\hbar\mu^2/\gamma$. Then, the spatial distribution of the fields F_1 and F_2 in the medium in the stationary regime can be determined by solving the wave equations

$$\frac{d^2 F_1}{dx^2} + \left(\varepsilon_1' + i\varepsilon_1'' - \frac{\delta - i}{\delta^2 + 1} |F_2|^2 \right) F_1 = 0, \quad (6)$$

$$\frac{d^2 F_2}{dx^2} + s^2 \left(\varepsilon_2' + i\varepsilon_2'' - \frac{\delta - i}{\delta^2 + 1} |F_1|^2 \right) F_2 = 0, \quad (7)$$

where $x = k_1 z$; $k_1 = \omega_1/c$; $\delta = \Delta/\gamma$; $s = \omega_2/\omega_1$; and z is the pulse propagation axis. As boundary conditions at the point $z = 0$ (the end of a semi-infinite crystal), we will use the continuity conditions for the tangential components of the electrical and magnetic fields of both pulses, which can be written as

$$F_{i1} + F_{r1} = F_1|_{x=0}, \quad F_{i1} - F_{r1} = -i \frac{dF_1(x)}{dx} \Big|_{x=0}, \quad (8)$$

$$F_{i2} + F_{r2} = F_2|_{x=0}, \quad s(F_{i2} - F_{r2}) = -i \frac{dF_2(x)}{dx} \Big|_{x=0}. \quad (9)$$

Because the crystal is semi-infinite and absorbs light, the only physical solutions of Eqns (6) and (7) are the solutions for which $F_1(x) \rightarrow 0$ and $F_2(x) \rightarrow 0$ at $x \rightarrow \infty$.

Because it is impossible to obtain exact analytical solutions of the system of nonlinear Eqns (6) and (7) in the general case, we will use numerical methods. It follows from (8) and (9) that it is impossible to begin numerical integration of (6) and (7) from the point $x = 0$, because the amplitudes of the reflected waves are unknown.

We assume that at some point $x = x_0$ inside the crystal the normalised amplitudes of the fields are vanishingly small, i.e., $|F_{1,2}(x_0)| \ll 1$. Such a point always exists, because the light is absorbed during its propagation. Then, nonlinear terms in (6) and (7) vanish, and the solutions represent only the forward waves:

$$F_1(x) = F_1(x_0) \exp [i(\varepsilon_1' + i\varepsilon_1'')^{1/2}(x - x_0)], \quad (10)$$

$$F_2(x) = F_2(x_0) \exp [is(\varepsilon_2' + i\varepsilon_2'')^{1/2}(x - x_0)], \quad (11)$$

where $x = x_0$ is the point from which we begin the integration of Eqns (6) and (7), moving backward.

In accordance with (8)–(11), the fields and their derivatives at the point $x = x_0$ are known, and they determine the solution of Eqns (6) and (7), beginning from the point $x = x_0$. As x decreases from x_0 to zero, the solutions give the spatial distribution of the complex functions $F_1(x)$ and $F_2(x)$ and the amplitudes of the incident (F_{i1} and F_{i2}) and reflected (F_{r1} and F_{r2}) fields. One can see from (10) and (11) that in the linear limit, the moduli of the field amplitudes of both waves decrease exponentially with the distance, the rates of the spatial changes in the field profiles being much greater at $\varepsilon_{1,2}' < 0$ than at $\varepsilon_{1,2}' > 0$. The fast exponential decrease in the field amplitudes at $\varepsilon_{1,2}' < 0$ is caused both by purely dissipative absorption and nondissipative non-transmission.

3. Discussion of the results

Consider the results of numerical integration of the system of Eqns (6)–(9). The reflectivities $R_1 = |F_{r1}|^2/|F_{i1}|^2$ and $R_2 = |F_{r2}|^2/|F_{i2}|^2$ calculated as functions of the amplitudes F_{i1} and F_{i2} of the fields of the incident radiation are pre-

sented in Fig. 1. One can see that the reflectivities R_1 and R_2 depend substantially on the parameters $\varepsilon'_{1,2}$, $\varepsilon''_{1,2}$, s and the detuning δ from the resonance. For vanishingly small excitation intensities, when the nonlinear corrections to ε_1 and ε_2 in (3) and (4) vanish, we obtain

$$R_{1,2} = [(n_{1,2} - 1)^2 + \kappa_{1,2}^2][(n_{1,2} + 1)^2 + \kappa_{1,2}^2]^{-1}, \quad (12)$$

where

$$n_{1,2} = \left(\frac{r_{1,2} + \varepsilon'_{1,2}}{2} \right)^{1/2}; \quad \kappa_{1,2} = \left(\frac{r_{1,2} - \varepsilon'_{1,2}}{2} \right)^{1/2};$$

$$r_{1,2} = [(\varepsilon'_{1,2})^2 + (\varepsilon''_{1,2})^2]^{1/2}. \quad (13)$$

In this limit, the reflectivities do not depend on the excitation level.

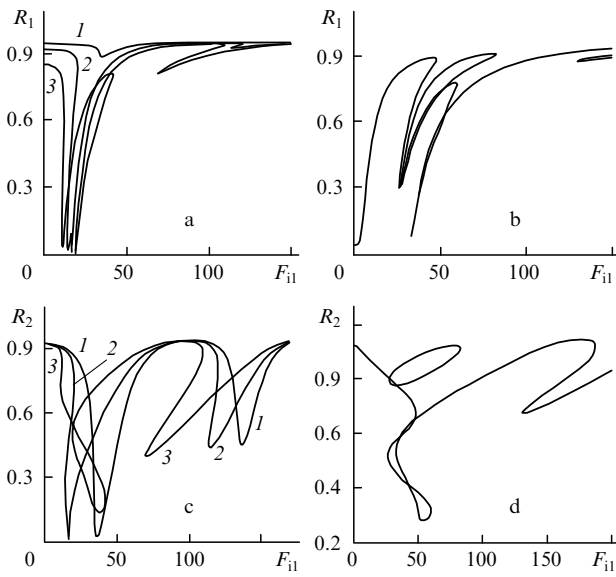


Figure 1. Dependences of the reflectivities R_1 (a, b) and R_2 (c, d) on the field amplitude F_{11} of the incident radiation for $\varepsilon'_1 = -15$, $\varepsilon''_1 = 1.5$, $\varepsilon'_2 = -10$, $\varepsilon''_2 = 1.5$, $s = 1.1$, $\delta = -15$, and the field amplitude of the incident radiation $F_{12} = 10$ (1), 20 (2), 30 (3) (a, c) and 50 (b, d).

Generally speaking, the dependence $R_1(F_{11}, F_{12})$ or $R_2(F_{11}, F_{12})$ represents a complex surface in the space of the variables F_{11} and F_{12} , the surfaces being a plane for small F_{11} and F_{12} , which acquires an increasing curvature outwards from the origin of coordinates. Because it is impossible to present this dependence graphically, we will show only few plots $R_1(F_{11})$ and $R_2(F_{11})$ for $F_{12} = \text{const}$, i.e., the curves that represent cross sections of this complex surface for a number of values $F_{12} = \text{const}$.

One can see in Fig. 1 the characteristic features of these curves. The function $R_1(F_{11})$ at different $F_{12} = \text{const}$ has different values at $F_{11} = 0$ (Fig. 1a), whereas functions $R_2(F_{11})$ (Fig. 1c) at $F_{11} = 0$ have the same value. This is explained by the fact that for $F_{11} > 0$, the dielectric function ε_1 is determined by the field amplitude F_2 , which is nonzero, whereas F_1 is almost zero, and, hence, $\varepsilon_2 = \text{const}$. The reflectivity R_2 in this case is given by the expression (12). The square of the field amplitude F_2 in the medium near to the crystal end is approximately equal to $(1 - R_2)|F_{12}|^2$. Then, the reflectivity R_1 from the crystal end can be written in the form

$$R_1 = [(N_1 - 1)^2 + Q_1^2][(N_1 + 1)^2 + Q_1^2]^{-1},$$

where

$$N_1 = \left(\frac{\tilde{\rho}_1 + \tilde{\varepsilon}'_1}{2} \right)^{1/2}; \quad Q_1 = \left(\frac{\tilde{\rho}_1 - \tilde{\varepsilon}'_1}{2} \right)^{1/2};$$

$$\tilde{\rho}_1 = [(\tilde{\varepsilon}'_1)^2 + (\tilde{\varepsilon}''_1)^2]^{1/2};$$

$$\tilde{\varepsilon}'_1 = \varepsilon'_1 - \frac{\delta(1 - R_2)|F_{12}|^2}{\delta^2 + 1}; \quad \tilde{\varepsilon}''_1 = \varepsilon''_1 + \frac{\delta(1 - R_2)|F_{12}|^2}{\delta^2 + 1}.$$

Thus, for $F_{11} \rightarrow 0$, the reflectivity R_2 is constant, whereas R_1 depends on the amplitude F_{12} of the incident wave and on the detuning δ from the resonance. For small F_{12} ($F_{12} = 10$), the function $R_1(F_{11})$ exhibits a weak dip (Fig. 1a, curve 1). As F_{12} increases ($F_{12} = 20$), the plot of the function $R_1(F_{11})$ changes substantially – two multistability loops arise (Fig. 1a, curve 2).

As F_{12} further increases ($F_{12} = 30$), this structure becomes even more complicated – the number of multistability loops increases, and moreover, an additional structure appears that has the form of self-crossing of the curve R_1 in the region of the first loop (F_{11}) (Fig. 1a, curve 3). In this region of F_{11} , the function $R_1(F_{11})$ reveals a complicated hysteresis behaviour upon cyclic changes in F_{11} . This function exhibits jumps only when F_{11} decreases, whereas the jumps are virtually absent when F_{11} increases. In the region of the second loop (and further), the jumps in $R_1(F_{11})$ upon cyclic changes in F_{11} are the same as the jumps in the multistable reflection curve of a usual Fabry–Perot cavity [1–3].

A qualitatively new complication in the behaviour of the function $R_1(F_{11})$ arises at $F_{12} = 50$ – in the region of the first loop, an oval of a complicated shape appears, which is separated from the main multistable curve (Fig. 1b). As F_{12} increases, the region of existence of this oval decreases rapidly, and then it disappears. However, a new oval or several ovals appear simultaneously at other values of F_{12} , which disappear gradually, making room for other ovals. The ovals, produced sequentially, are located in the vicinity of the loops of increasingly higher orders.

As for the dependences $R_2(F_{11})$ (Fig. 1c, d), one can see that as F_{12} increases, they also become more complicated, featuring multistable loops, self-crossing of loops, and the formation of one or several ovals.

The appearance of independent oval-like curves along with the multistable curves in $R_1(F_{11})$ and $R_2(F_{11})$ at $F_{12} = \text{const}$ has a simple physical explanation. In the F_{11}, F_{12} -space, the functions $R_1(F_{11}, F_{12})$ and $R_2(F_{11}, F_{12})$ represent complicated multivalued surfaces, which are characterised by the appearance of sharp horn-shaped structures. The horn-shaped extensions are located in the regions of F_{11} and F_{12} where the traditional (flat) multistable curves have multivalued loops. In the spatial version of multistable reflection, these loops are converted into the horn-shaped extensions. The cross sections of such a surface at different $F_{12} = \text{const}$ give not only multistable regions, but also the regions of horn-shaped extensions, which are mapped as ovals, which are gradually separated from rather complicated profiles of multistable curves with increasing F_{12} .

Fig. 2 shows the dependence of the reflectivity R_1 on the amplitudes F_{11} and F_{12} of incident fields in a simpler case. One can see that the surface corresponding to this depend-

ence has one- and three-valued regions. In the three-valued region, cross sections of this surface have the bistable regions of the reflectivity at $F_{i1} = \text{const}$ or $F_{i2} = \text{const}$. If we set $F_{i2} = \text{const}$, then a jump from the lower branch of the hysteresis curve $R_1(F_{i1})$ to the upper one takes place with increasing F_{i1} , while for $F_{i1} = \text{const}$, a jump occurs from the upper branch of the hysteresis curve $R_1(F_{i1})$ to the lower one with increasing F_{i2} .

Note also that for $F_{i1} = \text{const}$, the jump with increasing F_{i2} occurs at much smaller values than the jump with increasing F_{i1} at $F_{i2} = \text{const}$, which is related to the values of the parameters $\varepsilon'_{1,2}$, $\varepsilon''_{1,2}$, s , and δ . For the values of parameters chosen, the horn-shaped structures are absent, although a region with sharp bend of the surface $R_1(F_{i1}, F_{i2})$ is observed for small values of F_{i1} and F_{i2} .

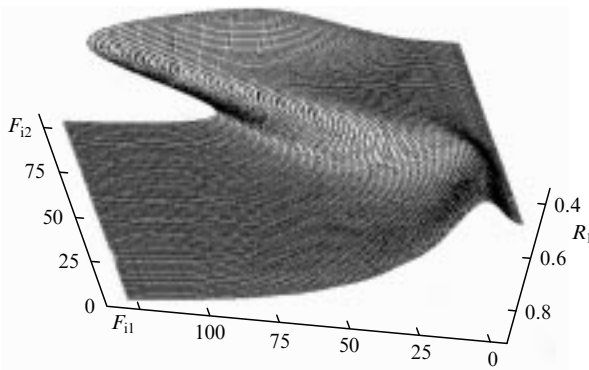


Figure 2. Dependence of the reflectivity R_1 on the field amplitudes F_{i1} and F_{i2} of the incident radiation for $\varepsilon'_1 = 1$, $\varepsilon''_1 = 15$, $\varepsilon'_2 = -2$, $\varepsilon''_2 = 1$, $\delta = -15$, and $s = 1.1$.

The above features of the dependence of reflectivities from the crystal end on the excitation intensity are caused by the renormalisation of the semiconductor energy spectrum at high excitation intensities, which is also manifested in the spatial distribution of the field amplitudes $|F_1|$ and $|F_2|$ in the medium.

Fig. 3 shows the dependences of $|F_1|$ and $|F_2|$ on the coordinate. One can see that both distributions have exponential tails inside the crystal, which are transformed into non-exponential functions as the crystal end is approached. These functions oscillate in certain spatial regions. The exponential tails are caused by non-transmission and absorption of light in the medium. For large field amplitudes, the effect of non-transmission is absent, and only nonlinear absorption takes place.

The oscillatory structure of the spatial distribution of the fields is caused by the nonlinear dispersion. A strong inhomogeneity of the field distribution in space causes the spatial inhomogeneity of nonlinear refractive indices and the extinction and internal reflection coefficients. As a result, narrow regions with large gradients of the refractive index are formed in the medium, where the backward waves arise.

The complicated nonlinear interference between forward and backward waves results in the stationary structure of the spatial field profiles, which was discussed above. The narrow region with the large gradient of the nonlinear refractive index and the corresponding abrupt peak of the internal reflection coefficient suggests the appearance of the Fabry–Perot cavity, induced by the pump field. The ref-

lection from this cavity causes the multistability. The absence of such cavity leads only to the one-valued reflectivity.

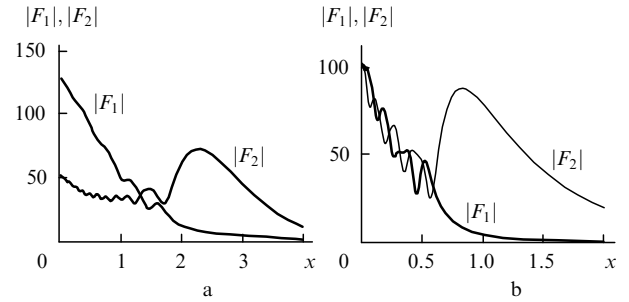


Figure 3. Spatial distribution of the field amplitudes $|F_1|$ and $|F_2|$ in the medium for $s = 1.1$ and $\varepsilon'_1 = 1$, $\varepsilon''_1 = 15$, $\varepsilon'_2 = -2$, $\varepsilon''_2 = 1$, $\delta = -15$ (a) and $\varepsilon'_1 = -2$, $\varepsilon''_1 = 15$, $\varepsilon'_2 = -1$, $\varepsilon''_2 = 17$, $\delta = -5$ (b).

4. Conclusions

Thus, if the self-reflection of light is directly taken into account, i.e., the generation of backward waves during the propagation of two pump waves is taken into account, the reflectivities from the interface of a semi-infinite medium depend on the pump wave intensity in a very complicated way. In particular, we predict the existence of the complicated, multivalued surface of the reflectivity $R_{1,2}$ in the $F_{i1}, F_{i2}, R_{1,2}$ space, whose projections on the $F_{i1}, R_{1,2}$ plane at $F_{i2} = \text{const}$ lead to the appearance of closed solitary oval-shaped curves of the reflection function.

In relation to the results obtained, consider the possibility of the experimental observation of the effect studied. In [23], the self-reflection was found in a system of two-level atoms by observing the Doppler shift of the frequency of a self-reflected wave caused by the motion of the interface between the regions with high and low absorption. Not being experimenters, we would like to propose nevertheless an additional method for experimental observation of a new physical effect — two-beam reflection.

Let a high-power electromagnetic pump wave be incident normally on a semi-infinite crystal, by creating a narrow region with an abrupt gradient of the refractive index at some distance from the crystal end. Let a weak probe beam be incident on the crystal at some angle to the normal. This beam is partially reflected from the interface (at the point of incidence), and partially passes into the medium after refraction. The angle of incidence can be chosen such that the probe beam, which has passed into the medium, is incident on the interface between the regions with high and low absorption, i.e., on the region with the abrupt gradient of the refractive index. The beam is partially reflected from this region and comes from the crystal into vacuum at certain distance from the point of incidence. This longitudinal displacement between the incident and outgoing beams can substantially exceed the known Goos–Hanchen displacement [24], which will prove that the reflection takes place from the internal region in the crystal.

By changing the pump beam power, one can display the internal interface with the abrupt gradient of the refractive index, which will result in a change in the longitudinal displacement of the probe beam leaving the medium. Because the probe beam is ‘reflected’ at two widely separated points, this phenomenon can be called ‘two-beam reflection’.

Depending on whether the peak of the internal reflectivity is narrow (sharp) or broad (spatially distributed in the propagation direction of a high-power beam), one can observe two reflected beams or a broad spatial distribution of the reflected beam over total length of the longitudinal displacement.

It was shown in [25] that the probability of two-photon absorption accompanied by the generation of a biexciton is higher than the probability of exciton absorption when the photon density of the incident radiation exceeds 10^{15} cm^{-3} . The direct two-phonon generation of biexcitons in CuCl has been first observed in [26] upon excitation of the crystal by 25-ps laser pulses with the peak intensity of 1 GW cm^{-2} .

Let us estimate the intensity required to observe the self-reflection from the expression

$$P = \frac{c^2 E}{8\pi} = \frac{c F^2 \gamma}{2(4\pi\mu)^2 \hbar}.$$

Assuming that the decay rate of the biexciton state is $\gamma = 10^{10} \text{ s}^{-1}$, and taking $\mu = 10^{17} \text{ CGSE units}$ [22, 25], we obtain $P = 5 \text{ kW cm}^{-2}$ for the normalised field $F = 1$. Then, the normalised amplitude of pump field, for instance, $F = 30$ corresponds to the intensity $P = 4.5 \text{ MW cm}^{-2}$, which can be achieved in the experiment. The probe pulse intensity should be lower by one-two orders of magnitude.

A competing mechanism in the experimental observation of self-reflection in the case of large detunings from the resonance is the exciton–photon interaction, which is not considered in this paper. This means that the detuning from the resonance should be smaller than the half binding energy of a biexciton, which is 15–20 meV for CuCl. In addition, the biexciton–biexciton interaction should be taken into account at high biexciton densities in the medium. However, for a CuCl crystal with the exciton radius of the order of 7 \AA (the biexciton radius is 14 \AA), the density at which this interaction becomes important is approximately $3 \times 10^{20} \text{ cm}^{-3}$.

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