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Two-photon intraband absorption of polarized light and its linear-circular dichroism in diamond-like semiconductors

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The theory of interband two-photon absorption of a strong light wave in semiconductors with a complex band is developed. An analytical expression is obtained for the probability of a two-photon interband transition, which contains a dependence on the intensity, polarization vector, and frequency of the absorbed light, as well as on the band parameters of the semiconductor, where the contribution of the Rabi effect to the coefficients of two-quantum absorption of light is taken into account. It is shown that the shape of the absorption edge and linear-circular dichroism depends on the degree of light polarization and on the Rabi parameter.

Keywords: two-quantum absorption of light, Rabi effect, linear-circular dichroism, complex band semiconductor, matrix elements and optical transition probabilities.

1. Introduction

The discovery of intense quantum generators of laser radiation stimulated the development of optical phenomena with nonlinear intensity in condensed matter, in particular in semiconductors [1–3]. In this aspect, it seems necessary to study the nonlinear optical properties of both narrow-gap and wide-gap semiconductors, which is associated with the unusualness and complexity of the properties of this effect, the possibility of obtaining new information about the band properties of crystals, and the prospect of wide practical application.

Two-photon nonlinear optical phenomena caused by intraband transitions in semiconductors were considered in [1–3], where two-photon matrix elements were calculated using perturbation theory by the field of unpolarized light in the two-band Kane approximation [2,3]. In [4–7], two-photon and three-photon linear-circular dichroisms (LCD) were studied[1] in crystals of a cubic lattice. In [8–22], the authors investigated multiphoton absorption of polarized radiation in semiconductors with a complex band, caused by vertical optical transitions between subbands of the valence band with light and heavy mass. In [18], nonlinear single-photon absorption of light in Weyl semimetals was studied depending on the degree of polarization.

In the above-listed studies, it is assumed that the nonlinearity in the intensity of the single-photon light absorption coefficient arises due to the Rabi effect [19], and in intraband [9,10,16–18] light absorption it is due to the photoinduced addition to the distribution function of electrons and holes determined by the relaxation time from the pulse energy using Dirac δ-function, i.e. the law of conservation of energy.

In [8,11,14] the LCD in hole-conducting germanium was studied in the case when comparable contributions to absorption are made by n-photonic processes with $n = (1-5)$. In [16,17], the contribution of Rabi effect, the effect of coherent saturation, to the four-photon absorption of polarized light caused by intraband optical transitions was studied.

In this study, calculations are carried out of the spectral-polarization dependences of the intraband two-photon, i.e. two-quantum light absorption (TQLA) and its LCD in semiconductors of tetrahedral symmetry in the three-band Kane model. In this case, the contributions are taken into account to the multiquantum process of virtual states located both in the subbands of the valence band and in the spin-orbit splitting band, and in the conduction band, where the contribution of the Rabi effect is taken into account.

2. Two-photon intraband absorption of polarized light in narrow-gap semiconductors

Following the studies of [10,13–17], in the below calculations of the spectral and temperature dependence of the TPLA coefficient $K^{(2)}$, where the wave vector of light is neglected, it is assumed that $k' = k_{1,2} \cong k$ (k', k, and $k_{1,2}$) is the wave vector of current carriers in the final (initial and intermediate) state. Then

$$K^{(2)}_{V,\pm 1/2;V,\pm 3/2} = \frac{2\pi}{\hbar}\, 2\hbar\omega\, \frac{1}{I}\, \rho(2\hbar\omega) F(\beta, 2, \omega) \times \sum_{m'=\pm 1/2, m=\pm 3/2} \left\langle \frac{|M^{(2)}_{m'm}(\mathbf{k})|^2}{\sqrt{1 + 4\frac{\alpha_\omega}{\hbar^2\omega^2}|M^{(2)}_{m'm}(\mathbf{k})|^2}} \right\rangle \quad (1)$$

[1] Two-photon linear-circular dichroism caused by interband optical transitions of electrons was predicted by E.L. Ivchenko in [4].

or

$$K_{hh,lh}^{(2)} = \frac{2\pi}{\hbar} 2\hbar\omega \frac{1}{I} \rho(2\hbar\omega) F(\beta, 2, \omega)$$

$$\times \left(\left\langle \frac{|M_{V,\pm 1/2;V,\pm 3/2}^{(12)}(\mathbf{k})|^2}{\sqrt{1 + 4\frac{\alpha_\omega}{\hbar^2\omega^2} |M_{V,\pm 1/2;V,\pm 3/2}^{(2)}(\mathbf{k})|^2}} \right\rangle \right.$$

$$\left. + \left\langle \frac{|M_{V,\pm 1/2;V,\mp 3/2}^{(2)}(\mathbf{k})|^2}{\sqrt{1 + 4\frac{\alpha_\omega}{\hbar^2\omega^2} |M_{V,\pm 1/2;V,\mp 3/2}^{(2)}(\mathbf{k})|^2}} \right\rangle \right), \quad (2)$$

where $\rho(2\hbar\omega)$ is density of states of current carriers involved in the two-photon optical transitions where the law of conservation of energy is taken into account, $F(\beta, 2, \omega)$ is current carrier distribution function in the initial state, $\beta^{-1} = k_B T$, k_B is Boltzmann's constant, T is sample temperature:

$$F(\beta, 2, \omega) = \exp\left[\beta\left(\mu - E_{L=hh}(k_{lh,hh}^{(2\omega)})\right)\right],$$

$$E_{hh}(k_{lh,hh}^{(2\omega)}) = \frac{m_{lh}}{m_{hh} - m_{lh}} 2\hbar\omega,$$

$$\rho(\hbar\omega) = \frac{m_{lh}}{m_{hh} - m_{lh}} k_{lh,hh}^{(2\omega)} / (\pi^2\hbar^2)$$

and it is taken into account that $2\hbar\omega \gg k_B T$.

Now it is necessary to perform angular averaging of moduli of squares of the matrix elements under consideration:

$$\left\langle \frac{|M_{V,\pm 1/2;V,\pm 3/2}^{(2)}(\mathbf{k})|^2}{\sqrt{1 + 4\frac{\alpha_\omega}{\hbar^2\omega^2} |M_{V,\pm 1/2;V,\pm 3/2}^{(2)}(\mathbf{k})|^2}} \right\rangle$$

$$+ \left\langle \frac{|M_{V,\pm 1/2;V,\mp 3/2}^{(2)}(\mathbf{k})|^2}{\sqrt{1 + 4\frac{\alpha_\omega}{\hbar^2\omega^2} |M_{V,\pm 1/2;V,\mp 3/2}^{(2)}(\mathbf{k})|^2}} \right\rangle, \quad (3)$$

where the contribution of the coherent saturation effect to the TPLA coefficient is made by these particular radicals of the latter relationships. Thus, calculations without taking into account the contribution of the coherent saturation effect to the TPLA coefficient are described as follows:

$$\left\langle |M_{V,+1/2;V,+3/2}^{(2)}(\mathbf{k})|^2 \right\rangle + \left\langle |M_{V,\pm 1/2;V,\mp 3/2}^{(2)}(\mathbf{k})|^2 \right\rangle,$$

so in further calculations we will take both cases into account and analyze the obtained theoretical results for each type of optical transitions.

3. Two-photon optical transitions between subbands of heavy and light holes

If we believe that optical transitions occur from the branch of heavy holes to the branches of light holes where intermediate states of current carriers are located in subbands of the valence band, then the matrix element of the two-photon optical transition is determined by the following relationship:

$$|+3/2\rangle \to |m\rangle \to |+1/2\rangle = \frac{M_{+1/2;+3/2}^{(1)} M_{+3/2;+3/2}^{(1)}}{E_{hh} - E_{hh} - \hbar\omega}$$

$$+ \frac{M_{+1/2;+1/2}^{(1)} M_{+1/2;+3/2}^{(1)}}{E_{lh} - E_{hh} - \hbar\omega} + M_{+1/2;+3/2}^{(2)}$$

$$= \frac{M_{+1/2;+3/2}^{(1)} M_{+3/2;+3/2}^{(1)}}{(-\hbar\omega)} + \frac{M_{+1/2;+1/2}^{(1)} M_{+1/2;+3/2}^{(1)}}{(\hbar\omega)}$$

$$+ M_{+1/2;+3/2}^{(2)} = -5\sqrt{3}\left(\frac{eA_0}{c\hbar}\right)^2 B e'_+ e_{z'}, \quad (4)$$

where $M^{(1)}(m \to m') = M_{m'm}^{(1)}(\mathbf{k})$ is matrix element of a single-photon (simultaneously absorbing two photons) optical transition, from which we derive the expression for the squared modulus of optical transition

$$|\pm 3/2\rangle \to |m\rangle \to |\pm 1/2\rangle$$

and we have $75\left(\frac{eA_0}{c\hbar}\right)^4 B^2 |e'_+ e_{z'}|^2$, and for the optical transition $|\pm 3/2\rangle \to |m\rangle \to |\mp 1/2\rangle$ we have $\frac{3}{4}\left(\frac{eA_0}{c\hbar}\right)^4 B^2 (36 e_{z'}^2 |e'_+|^2 + |e'^2_-|^2)$.

If intraband optical transitions occur between the subbands of light and heavy holes, then intermediate states are found both in the conduction band and in the spin-orbit splitting band. Then the matrix elements of optical transitions are determined as follows:

$$a)\ |+3/2\rangle \to |c, m\rangle \to |+1/2\rangle + |+3/2\rangle \to$$

$$|\Delta, m\rangle \to |+1/2\rangle = \frac{M_{V,+1/2;c,+1/2}^{(1)} M_{c,+1/2;V,+3/2}^{(1)}}{E_c - E_{hh} - \hbar\omega}$$

$$+ \frac{M_{V,+1/2;c,-1/2}^{(1)} M_{c,-1/2;V,+3/2}^{(1)}}{E_c - E_{hh} - \hbar\omega}$$

$$+ \frac{M_{V,+1/2;SO,+1/2}^{(1)} M_{SO,+1/2;V,+3/2}^{(1)}}{E_{SO} - E_{hh} - \hbar\omega}$$

$$+ \frac{M_{V,+1/2;SO,-1/2}^{(1)} M_{SO,-1/2;V,+3/2}^{(1)}}{E_{SO} - E_{hh} - \hbar\omega}$$

$$= \left(\frac{eA_0}{c\hbar}\right)^4 \sqrt{\frac{1}{3}}$$

$$\times \left(\frac{p_{cV}^2}{E_c - E_{hh} - \hbar\omega} - \frac{3B^2k^2}{E_{SO} - E_{hh} - \hbar\omega}\right) e'_z e'_-; \quad (5)$$

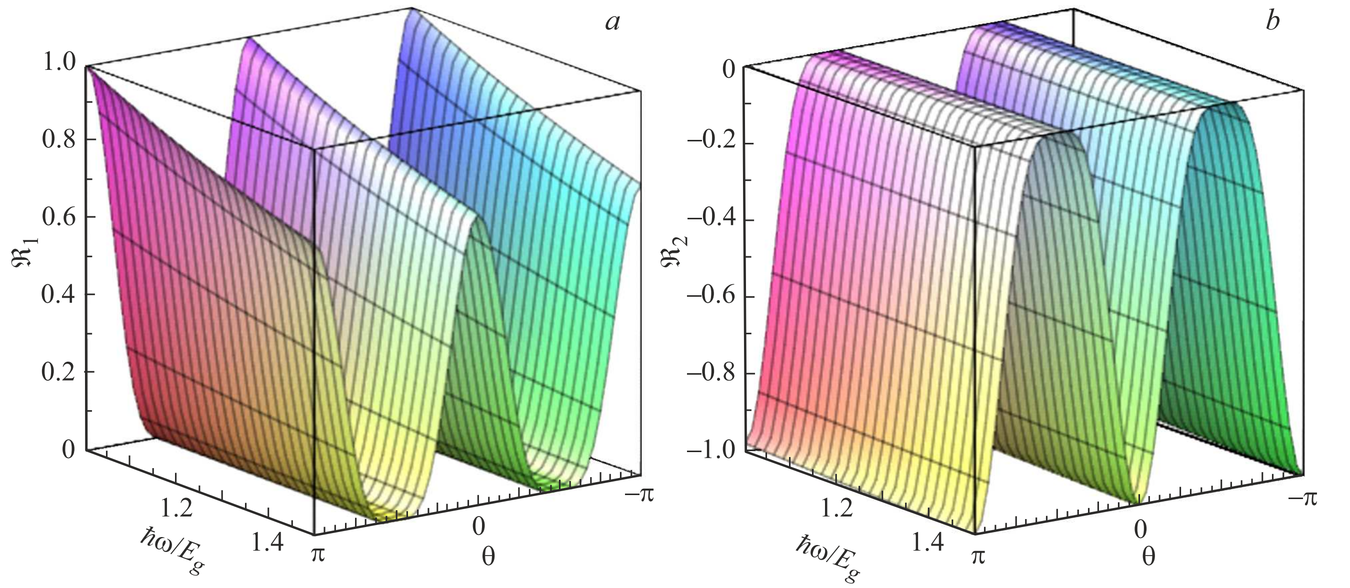

Figure 1. Spectral-polarization dependence of $\tilde{\mathfrak{R}}_1$ and $\tilde{\mathfrak{R}}_2$, which are used to determine spectral-polarization dependence of the TPLA coefficient in GaAs illuminated with linearly polarized light, without taking into account the contribution of the coherent saturation effect. (The colored version of the figure is available on-line).

$b)\ |-3/2\rangle \to |c, m\rangle \to |+1/2\rangle + |-3/2\rangle \to$

$$|SO, m\rangle \to |+1/2\rangle = \frac{M^{(1)}_{V,+1/2;c,+1/2} M^{(1)}_{c,+1/2;V,-3/2}}{E_c - E_{hh} - \hbar\omega} + \frac{M^{(1)}_{V,+1/2;c,-1/2} M^{(1)}_{c,-1/2;V,-3/2}}{E_c - E_{hh} - \hbar\omega} + \frac{M^{(1)}_{V,+1/2;SO,+1/2} M^{(1)}_{\Delta SO,+1/2;V,-3/2}}{E_\Delta - E_{hh} - \hbar\omega} + \frac{M^{(1)}_{V,+1/2;SO,-1/2} M^{(1)}_{SO,-1/2;V,-3/2}}{E_\Delta - E_{hh} - \hbar\omega}$$
$$= \left(\frac{eA_0}{c\hbar}\right)^4 \frac{1}{2\sqrt{3}} \times \left(\frac{p^2_{cV}}{E_c - E_{hh} - \hbar\omega} - 9\frac{B^2k^2}{E_{SO} - E_{hh} - \hbar\omega}\right) e'^2_+. \quad (6)$$

The remaining matrix elements are determined in a similar way. Thus, matrix elements of these optical transitions can be represented as the following matrix:

$$\tilde{M}^{(2)} = \frac{1}{2}\sqrt{\frac{1}{3}}\left(\frac{eA_0}{c\hbar}\right)^4 \frac{p^2_{cV}}{E_c - E_{hh} - \hbar\omega} \begin{bmatrix} 2e'_z e'_- & e'^2_- \\ -e'^2_+ & 2e'_z e'_+ \end{bmatrix} - \frac{\sqrt{3}}{2}\frac{1}{E_{SO} - E_{hh} - \hbar\omega} B^2k^2 \begin{bmatrix} 2e'_z e'_- & 3e'^2_- \\ -3e'^2_+ & 2e'_z e'_+ \end{bmatrix}.$$

Due to the fact that the two-photon linear circular dichroism coefficient and the TPLA coefficient are determined by the square of moduli of the composite matrix elements, for the above mentioned optical transitions the matrix elements can be represented as

$$|\tilde{M}^{(2)}|^2 = \begin{bmatrix} \mathfrak{R}_1 e'^2_z |e'_-|^2 & \mathfrak{R}_2 |e'_-|^4 \\ -\mathfrak{R}_2 |e'_+|^4 & \mathfrak{R}_1 e'^2_z |e'_+|^2 \end{bmatrix} - \begin{bmatrix} \mathfrak{R}_1 e'^2_z |e'_-|^2 & \mathfrak{R}_2 |e'_-|^4 \\ -\mathfrak{R}_2 |e'_+|^4 & \mathfrak{R}_1 e'^2_z |e'_+|^2 \end{bmatrix} \quad (7)$$

where

$$\mathfrak{R}_1 = \left(\frac{eA_0}{c\hbar}\right)^4 \frac{1}{3} \times \left(\frac{p^2_{cV}}{E_c - E_{hh} - \hbar\omega} - \frac{3B^2k^2}{E_{SO} - E_{hh} - \hbar\omega}\right)^2 e'^2_z |e'_-|^2, \quad (8)$$

$$\mathfrak{R}_2 = \frac{1}{12}\left(\frac{eA_0}{c\hbar}\right)^4 \times \left(\frac{p^2_{cV}}{E_c - E_{hh} - \hbar\omega} - 9\frac{B^2k^2}{E_{SO} - E_{hh} - \hbar\omega}\right)^2 |e'^2_z|^2. \quad (9)$$

Matrix elements of two-photon transitions occurring between the spin split-off band and the conduction band are determined in a similar way as in the above cases. It should be noted that in this case virtual states of current carriers are located in the subbands of the valence band, in the conduction band, and the spin-orbit splitting band.

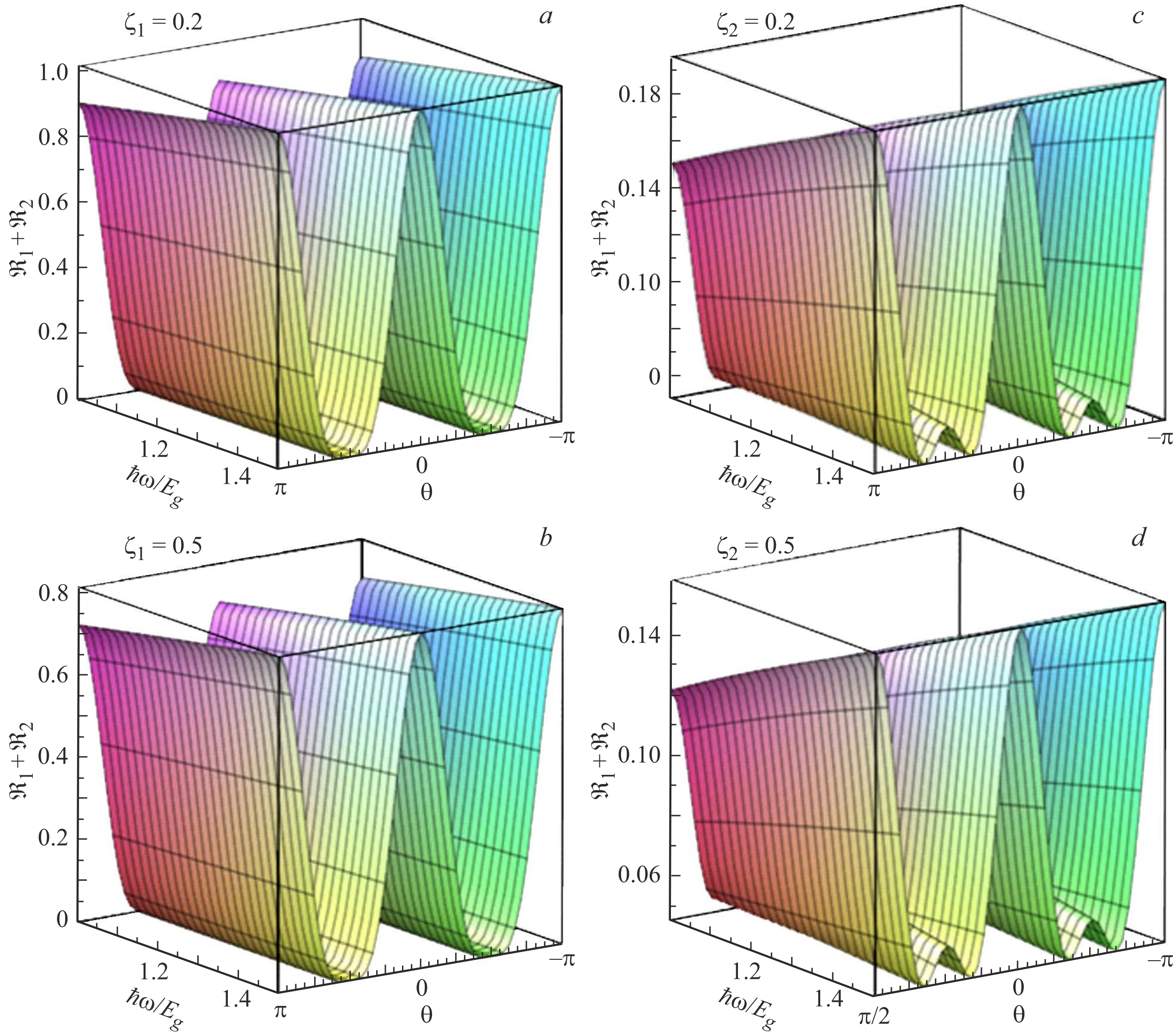

Figure 2. Spectral-polarization dependence of the $\mathfrak{R}_1 + \mathfrak{R}_2$ sum coefficient for both linearly (a, b) and circularly (c, d) polarized light, which is used to determine the coefficient of two-photon absorption of polarized light in InSb, taking into account the contribution of the coherent saturation effect for two values of the Rabi parameter.

4. Spectral-polarization dependences of the TPLA coefficient and LCD

Now let us analyze the spectral-polarization dependence of the TPLA coefficient, which is determined using $\mathfrak{R}_1$ and $\mathfrak{R}_2$ functions. For this purpose, let us rewrite expressions (2) taking into account (7). Spectral-polarization dependence of the TPLA coefficient of a GaAs crystal both without the contribution of the coherent saturation effect (see Figure 1) and taking into account this contribution (Figure 2), as well as two-photon LCD (see Figure 2, 3), caused by optical transitions between subbands of the valence band with light and heavy mass, where intermediate states are in the conduction bands and spin-orbit splitting band (see Figure 1), have an oscillatory behavior relative to the angle between the polarization vectors and current carriers for a linearly polarized light and relative to the angle between the photon wave vectors and current carriers for circularly polarized light. It can be seen in Figure 1 that the amplitude, period, and phase of the oscillations are different. Also, it should be noted that in GaAs the coefficient of two-photon LCD < 1.

When taking into account the contribution of the coherent saturation effect to the coefficient of two-photon LCD, expressions (8) and (9) take the following form:

$$\tilde{\mathfrak{R}}_1 = \left\langle \frac{\xi_1 e_z'^2 |e_-'^2|^2}{\sqrt{1+\xi_1 e_z'^2 |e_-'|^2}} \right\rangle, \quad \tilde{\mathfrak{R}}_2 = \left\langle \frac{\xi_2 |e_-'^2|^2}{\sqrt{1+\xi_2 |e_-'^2|^2}} \right\rangle, \tag{10}$$

where Rabi parameters are

$$\zeta_1 = 4\,\frac{\alpha_\omega}{\hbar^2\omega^2}\,\xi_1, \quad \zeta_2 = 4\,\frac{\alpha_\omega}{\hbar^2\omega^2}\,\xi_2, \tag{11}$$

$$\xi_1 = \left(\frac{eA_0}{c\hbar}\right)^4 \frac{1}{3}\left(\frac{p_{cV}^2}{E_c - E_{hh} - \hbar\omega} - \frac{3B^2k^2}{E_\Delta - E_{hh} - \hbar\omega}\right)^2,$$

$$\xi_2 = \frac{1}{12}\left(\frac{eA_0}{c\hbar}\right)^4 \left(\frac{p_{cV}^2}{E_c - E_{hh} - \hbar\omega} - 9\frac{B^2k^2}{E_\Delta - E_{hh} - \hbar\omega}\right)^2 |e_-'^2|^2.$$

Comparing the data in Figure 1, we find that taking into account the contribution of the coherent saturation effect leads to a decrease in the oscillation amplitude as the light frequency increases, regardless of the angle between the polarization vectors and the wave vector of current carriers for both linearly polarized and circularly polarized light. This is due to the fact that the spectral-polarization dependence for linearly polarized light is described by the angle between the polarization vectors and current carriers, and for circularly polarized light it is described by the angle between the photon vectors and current carriers. Quantitative calculations were carried out for $\xi_{1,2} = 0.2$.

Calculations show that for GaAs, with increasing light frequency, the contribution to the total coefficient of the two-photon LCD of the term proportional to $\mathfrak{R}_1$ decreases relative to the contribution of the term proportional to $\mathfrak{R}_2$, regardless of the angle between vectors **e**, **k**, **q**: if the frequency of light increases by 1.4 times, then this contribution decreases by 2.5 times, where **e** is light polarization vector, **k**(**q**) is wave vector of current carriers (photon). This is due to the fact that the first contribution does not depend on the frequency of light, and the second contribution depends on the frequency as $\propto (\hbar\omega)^{1/2}$. Therefore, the second contribution increases with increasing frequency of light.

To compare the theoretical results, further calculations were carried out using the spectral-polarization dependence of the $\mathfrak{R}_1$ and $\mathfrak{R}_2$ coefficients, which are used to determine the coefficient of two-photon absorption of linearly and circularly polarized light in InSb caused by optical transitions between subbands of the valence band with light and heavy mass for two values of the Rabi parameters $\xi_1 = 0.2, 0.5$ and $\xi_2 = 0.2, 0.5$, where intermediate states are in the conduction band and spin-orbit splitting band, taking into account the contribution of the coherent saturation effect (see Figure 2). It can be seen from Figure 2 that in the spectral-polarization dependence of the $\mathfrak{R}_1 + \mathfrak{R}_2$ sum coefficient for both linearly (a, b) and circularly (c, d) polarized light, which is used to determine the coefficient of two-photon absorption of linearly and circularly polarized light in InSb caused by optical transitions between subbands of the valence band with light and heavy mass, where intermediate states are in the conduction band and spin-orbital splitting band, taking into account the contribution of the coherent saturation effect for two values of the Rabi parameter. It follows therefrom that: firstly, all spectral-polarization dependences are oscillatory in nature; secondly, as the frequency of light increases, the amplitude of the oscillation increases; thirdly, for circularly polarized light the oscillation becomes aperiodic; fourthly, with increasing values of the Rabi parameter, the amplitude values of the oscillatory dependences decrease. In the calculations, the maximum value of the spectral-polarization dependence of the two-photon LCD coefficient at $\xi_{1,2} = 0.2$ was chosen equal to unit.

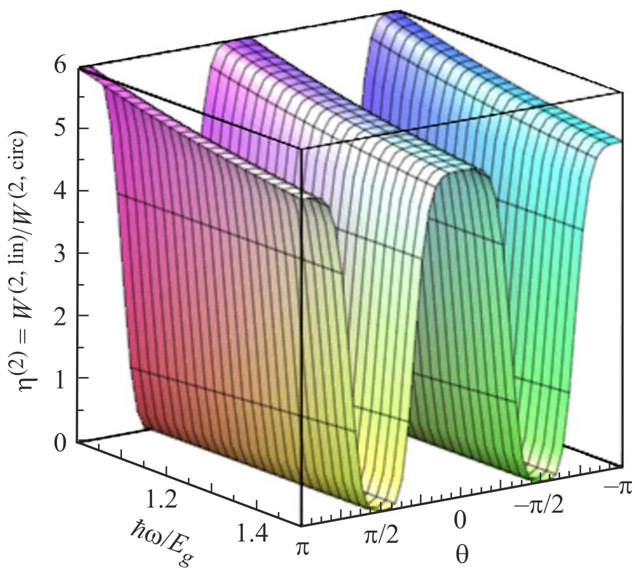

Figure 3. Spectral-polarization dependence of the two-photon LCD in InSb caused by optical transitions between the subbands of light and heavy holes, taking into account the contribution of the coherent saturation effect.

It can be seen from Figure 2, c that with an increase in the Rabi parameter from $\xi_\omega = 0.2$ to $\xi_\omega = 0.5$, the maximum value of the spectral-polarization dependence of the two-photon LCD coefficient in InSb increases by 23%. Therefore, the spectral-polarization dependence of two-photon linearly circular dichroism in InSb, taking into account the contribution of the coherent saturation effect, is almost independent of the value of the Rabi parameter and its amplitude is < 6.

Figure 3 shows the spectral-polarization dependence of the two-photon LCD in InSb caused by transitions between the subbands of light and heavy holes, where intermediate states are in the conduction band and spin-orbit splitting band, taking into account the contribution of the coherent saturation effect at $\xi_\omega = 0.2$. Also, it should be noted that in contrast to the wide-gap GaAs crystal, in the narrow-gap InSb semiconductor the two-photon LCD coefficient is > 1.

Quantitative values of the band parameters were selected from [23].

5. Conclusion

In conclusion, the following should be noted.

1. Expressions are derived for the spectral and polarization dependences for matrix elements of intraband optical transitions occurring with the absorption of two photons, which have taken into account that virtual states are located not only in subbands of the valence band (see, for

example, [15 –17]) but also in the conduction band and spin split-off band.

2. The spectral-polarization dependences of the coefficients of two-photon absorption of light and its LCD are calculated taking into account the contribution of the coherent saturation effect to the absorption. It is shown that, in contrast to the wide-gap GaAs semiconductor, in the narrow-gap InSb semiconductor the two-photon LCD coefficient is > 1 and is almost independent of the Rabi parameter.

3. The theory of LCD coupled by intraband two-photon optical transitions in diamond-like semiconductors has been developed.

Conflict of interest

The authors declare that they have no conflict of interest.

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