Polarization-spectral dependences of three-photon interband absorption of light and linear-circular dichroism in semiconductors with cubic symmetry

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The polarization and frequency-polarization dependences of the linear-circular dichroism and light absorption coefficients in semiconductors of cubic symmetry, caused by vertical three-photon optical transitions between the states of the spin-orbit splitting and conduction bands, are calculated in narrow-gap semiconductors in the Kane model.

Keywords: initial, virtual and final states, interband three–photon absorption of light, Kane's approximation.

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1. Introduction

Currently, the main research in the field of multiphoton light absorption is carried out in wide-band semiconductors, since a number of their physicochemical properties have been studied in depth and in more detail. In this respect, the multiphoton effects occurring in narrow-band crystals have been poorly studied both in theoretical and experimental aspects. The main reason for this is that the theoretical study of a number of photonic-kinetic phenomena in narrow-band crystals requires the use of not only the Luttinger–Cohn approximation, but also the multi-band Kane approach. In the latter case, theoretical calculations are performed using matrices of at least 6×6 or 8×8 in size.

The first studies on two-photon interband transitions in crystals were carried out in the early 1960s shortly after the appearance of lasers [1–3]. When calculating the matrix elements of two-photon transitions in crystals, the theories of perturbations in the field of an unpolarized electromagnetic wave [2,3] were used, where the two-band Kane model was used.

In [4–11], linear-circular dichroism (LCD) of two- and three-photon light absorption in crystals has been studied both theoretically and experimentally. Non-linear interband single-photon absorption of polarized light in Weyl semimetals was examined in the article [12]. However, the question of the polarization and frequency-polarization dependence of the three-photon interband absorption of light in cubic symmetry crystals, due to the states of the spin-orbit splitting zone and the conduction band in the Kane approximation, remained open, the study of which is covered in this paper.

2. Polarization dependence of three-photon linear-circular dichroism

In order to calculate the polarization dependences of interband three-photon optical transitions, which are used to determine the coefficients of light absorption and linear-circular dichroism, we will consider in more detail three-photon optical transitions originating from the state of the spin-orbital splitting subzone ($|\mathrm{SO}, -1/2\rangle$) to the conduction bands ($c, -1/2\rangle$), which consist of the following different transitions

$$\sum_{m,m'} |\mathrm{SO}, -1/2\rangle \to |V, m\rangle \to |V, m'\rangle \to |c, -1/2\rangle,$$

$$\sum_{m,m'} |\mathrm{SO}, -1/2\rangle \to |c, m\rangle \to |c, m'\rangle \to |c, -1/2\rangle,$$

$$\sum_{m,m'} |\mathrm{SO}, -1/2\rangle \to |\mathrm{SO}, m\rangle \to |\mathrm{SO}, m'\rangle \to |c, -1/2\rangle,$$

$$\sum_{m,m'} |\mathrm{SO}, -1/2\rangle \to |c, m'\rangle \to |V, m\rangle \to |c, -1/2\rangle,$$

$$\sum_{m,m'} |\mathrm{SO}, -1/2\rangle \to |V, m\rangle \to |c, m'\rangle \to |c, -1/2\rangle,$$

$$\sum_{m,m'} |\mathrm{SO}, -1/2\rangle \to |V, m\rangle \to |\mathrm{SO}, m'\rangle \to |c, -1/2\rangle,$$

$$\sum_{m,m'} |\mathrm{SO}, -1/2\rangle \to |\mathrm{SO}, m\rangle \to |V, m'\rangle \to |c, -1/2\rangle,$$

$$\sum_{m,m'} |\mathrm{SO}, -1/2\rangle \to |c, m\rangle \to |\mathrm{SO}, m'\rangle \to |c, -1/2\rangle,$$

$$\sum_{m,m'} |\mathrm{SO}, -1/2\rangle \to |\mathrm{SO}, m\rangle \to |c, m'\rangle \to |c, -1/2\rangle, \quad (1)$$

Matrix elements of three-photon optical transitions

№	Virtual states	Matrix element of optical transition type $\|SO, -1/2\rangle \to \|c - 1/2\rangle$
1	Virtual states lie in the valence band	$\frac{i}{2\sqrt{3}}\frac{\hbar^2}{m_c}\frac{P_{cV}Bk^2}{\hbar\omega}e'_z\left(\mathscr{R}_3\|e'_+\|^2+\mathscr{R}_4e'^2_z\right)$.
2	Virtual states lie in the subzone spin-orbit splitting	$\frac{-i}{2\sqrt{3}}\left(\frac{\hbar^2}{m_c}\right)^2\frac{P_{cV}k^2}{(\hbar\omega)^2}e'^3_z$.
3	Virtual states lie in the subzone spin-orbital splitting takes the form	$\frac{i}{\sqrt{3}}\frac{P_{cV}A^2k^2}{(\hbar\omega)^2}e'^3_z$.
4	The first virtual state lies in the valence, and the second in the conduction band	$\frac{i}{2\sqrt{3}}\frac{\hbar^2}{m_c}\frac{P_{cV}Bk^2}{\hbar\omega}e'_z\left(\mathscr{R}_5\|e'_+\|^2+\mathscr{R}_6e'^2_z\right)$.
5	The first virtual state lies in the conduction band, a second — in the valence band	$\frac{i}{4\sqrt{3}}\frac{P_{cV}B^2k^2e'_z}{\hbar\omega}e'_z\left(\mathscr{R}_7\|e'_+\|^2+\mathscr{R}_8e'^2_z\right)$.
6	The first virtual state lies in the valence band, and the second in the spin-orbit splitting subzone	$\frac{-i}{12\sqrt{3}}\frac{P^3_{cV}}{\hbar\omega}e'_z\left(\mathscr{R}_9\|e'_+\|^2+\mathscr{R}_{10}\|e'_z\|^2\right)$.
7	The first virtual state lies in the conduction band, and the second in the spin-orbit splitting subzone	$\frac{i}{12\sqrt{3}}\frac{P^3_{cV}}{(\hbar\omega)^2}e'_z\left(\|e'_+\|^2+\|e'_z\|^2\right)$.
8	The first virtual state lies in the subzone spin-orbital splitting, and the second in the conduction band	$-\frac{2i}{\sqrt{3}}\frac{\hbar^2}{m_c}\frac{P_{cV}Ak^2}{(\hbar\omega)^2}e'^3_z$.
9	The first virtual state lies in the subzone spin-orbital splitting, and the second in the valence band	$\frac{-i}{12\sqrt{3}}\frac{P^3_{cV}}{\hbar\omega}e'_z\left(\mathscr{R}_{11}\|e'_+\|^2+\mathscr{R}_{12}\|e'_z\|^2\right)$.

Here $\mathscr{R}_3E^{-1}_{hh,\mathrm{SO},2}\left(\left(\frac{A}{B}-1\right)E^{-1}_{hh,\mathrm{SO},1}-E^{-1}_{lh,\mathrm{SO},1}\right)+3E^{-1}_{lh,\mathrm{SO},2}E^{-1}_{hh,\mathrm{SO},1}$, $\mathscr{R}_4=4\left(\frac{A}{B}-1\right)E^{-1}_{lh,So,2}E^{-1}_{hh,\mathrm{SO},1}$, $\mathscr{R}_9=3E^{-1}_{hh,\mathrm{SO},2}+E^{-1}_{lh,\mathrm{SO},2}$, $\mathscr{R}_{10}=4E^{-1}_{lh,\mathrm{SO},2}$, $\mathscr{R}_{11}=3\left(E^{-1}_{hh,\mathrm{SO},2}+E^{-1}_{lh,\mathrm{SO},2}\right)$, $\mathscr{R}_{12}=4E^{-1}_{lh,\mathrm{SO},2}$.

and differ from each other in the arrangement of virtual states and their order, where $|A, m\rangle \to |B, m\rangle$ describes the optical transition from the state of $|A, m\rangle$ to the state of $|B, m\rangle$, occurring during the absorption of a single photon, $m, m' = \pm 3/2$ for heavy hole subzone ($|hh, m\rangle$), $m, m' = \pm 1/2$ — for light hole subzones ($|lh, m\rangle$) and spin-orbit splitting ($|\mathrm{SO}, m\rangle$) and conduction bands ($|c, m'\rangle$) [13,14]. Further calculations are made according to the golden rule of quantum mechanics [15]. Then each term of the equation (1) has its own composite matrix element. In particular, when both virtual states lie in the heavy hole subzones, then the matrix element of a three-photon optical transition in the Kane model is defined as

$$-\frac{i}{\sqrt{3}}P_{cV}B^2k^2e'_z\left[3|e'_-|^2\mathscr{R}_1+\mathscr{R}_2\left(3|e'_-|^2+4e'^2_z\right)\right], \quad (2)$$

where

$$\mathscr{R}_1 = E^{-1}_{hh,\mathrm{SO},2}\left[\left(\frac{A}{B}-1\right)E^{-1}_{hh,\mathrm{SO},1}-E^{-1}_{lh,\mathrm{SO},1}\right] + E^{-1}_{lh,\mathrm{SO},2}E^{-1}_{hh,\mathrm{SO},2},$$

$$\mathscr{R}_2 = \left(\frac{A}{B}+1\right)E^{-1}_{lh,\mathrm{SO},3}E^{-1}_{lh,\mathrm{SO},1},$$

E_a — energy spectrum of current carriers in the $|a\rangle$ zone, N — number of absorbed photons, P_{cV} — Kane parameter, A, B — band parameters of the crystal, with the help of which the effective masses of light (m_{lh}) and heavy holes (m_{hh}) are determined or the parameters of the Luttinger–Cohn γ_1, γ_2 by equalities

$$\frac{\hbar^2}{2m_{lh}} = A + B = \frac{\hbar^2}{2m_0}(\gamma_1 + 2\gamma_2),$$

$$\frac{\hbar^2}{2m_{hh}} = A - B\frac{\hbar^2}{2m_0}(\gamma_1 - 2\gamma_2)\ [13,14],$$

e'_x, e'_y, e'_z — polarization vector projections $\mathbf{e}$ along the axes x', y', z' associated with the direction of the electron wave vector $\mathbf{k}(\mathbf{k} \parallel z')$, $e'_\pm = e'_x \pm ie'_y$. In particular, for linearly polarized light $|e'_z|^2 = \cos^2\alpha$, $|e'_\pm|^2 = 1 - |e'_z|^2$, and for circularly polarized light

$$|e'_z|^2 = \frac{1}{2}\sin^2\beta,$$

$$|e'_\pm|^2 = 1 - |e'_z|^2 \mp P_{circ}\cos\beta\ [4,8,9],$$

where $\alpha(\beta)$ — the angle between the polarization vectors $\mathbf{e}$ (photon wave vector $\mathbf{q}$) and the wave vector of current carriers ($\mathbf{k}$), P_{circ} — the degree of circular polarization of light. Matrix elements of the remaining three-photon optical transitions are given in the following table.

It can be seen from the data in the table that there are seven different optical transitions, differing from each other in intermediate states, giving different contributions to the

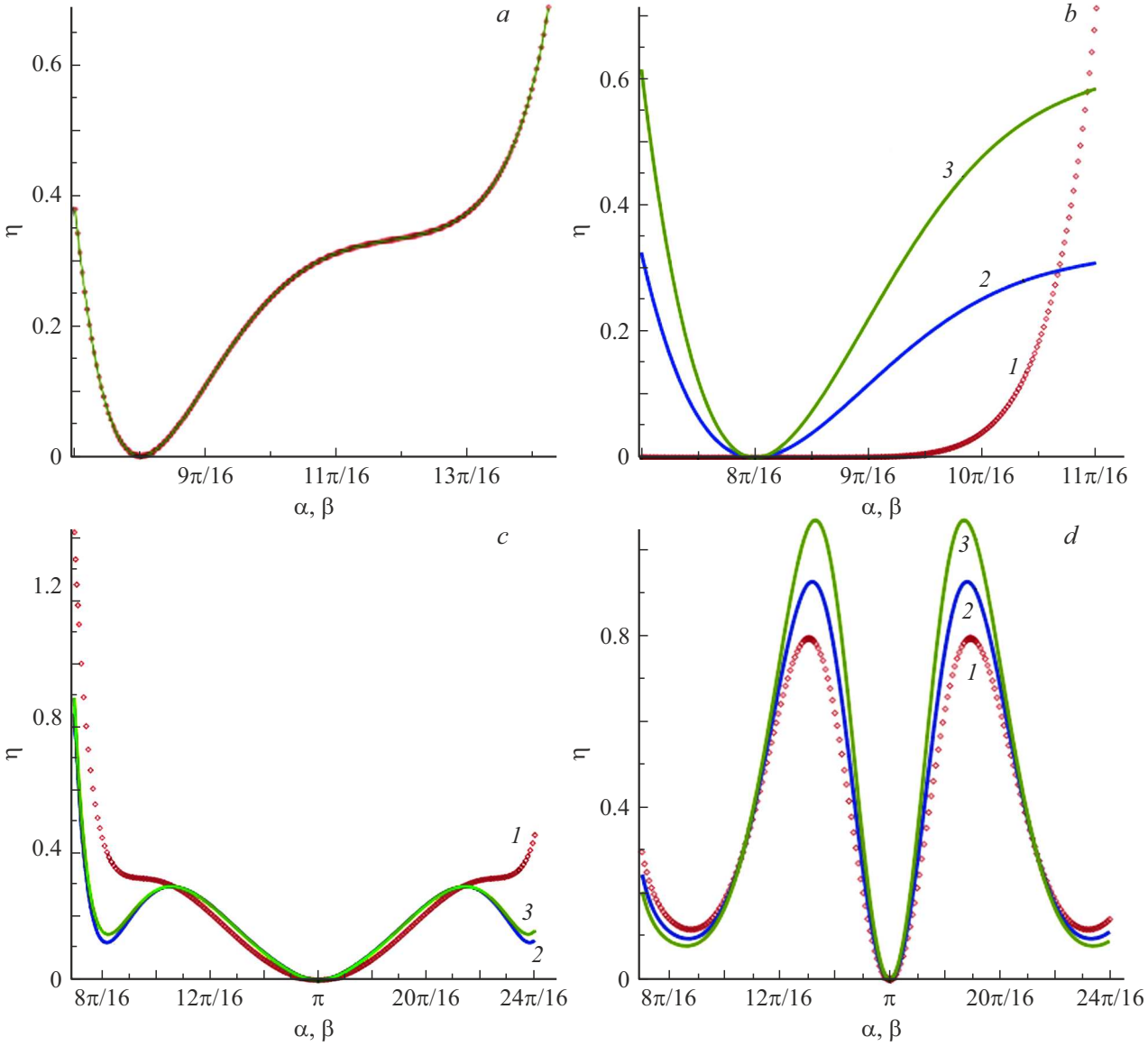

Figure 1. The polarization dependences of the three-photon linear-circular dichroism in InSb. Curves *1* correspond to the frequency of light $\hbar\omega = 0.6$ eV, curves *2* — $\hbar\omega = 0.5$ eV, and curves *3* — $\hbar\omega = 0.44$ eV.

resulting coefficient of three-photon light absorption (linear-circular dichroism) for both linear and circular polarization of light in cubic symmetry crystals with a complex band structure due to the difference in their dependences on the components of the polarization vector, band parameters and energy denominators.

Similarly, matrix elements of three-photon interband optical transitions are determined from $|\mathrm{SO}, -1/2\rangle$ to $|c, +1/2\rangle$ and from $|\mathrm{SO}, +1/2\rangle$ to $|c, \pm 1/2\rangle$.

Calculations carried out according to the golden rule of quantum mechanics [11] for InSb show that in three-photon transitions of the type $|\mathrm{SO} \pm 1/2\rangle \Rightarrow |c, \pm 1/2\rangle$ a graph of the polarization dependence of the coefficient of three-photon linear-circular dichroism, determined by the ratio of the probabilities of optical transitions $\eta = W^{(lin)}_{c,m'_c;\mathrm{SO},m}/W^{(lcirc)}_{c,m'_c;\mathrm{SO},m}$, shown in Fig. 1, *a*, if two virtual states lie in the subzones of the valence band, and Fig. 1, *b*, if both virtual states lie in the conduction band, as well as Fig. 1, *with*, if the initial virtual states lie in the conduction band, and the following — in the valence band, where the probability of N photonic optical transition

$$W^{(N)}_{c,m_c;SO,m_{\mathrm{SO}}} = \frac{2\pi}{\hbar}\sum_{\varsigma,m_c}|M_{c,m_c;\varsigma,m_\varsigma;SO,m_{\mathrm{SO}}}(\mathbf{k})|^2$$

$$\times\,[f_c(\mathbf{k}) - f_{\mathrm{SO}}(\mathbf{k})]\cdot\delta\big(E_c(\mathbf{k}) - E_{\mathrm{SO}}(\mathbf{k}) - N\hbar\omega\big), \quad (3)$$

$|\varsigma, m'_\varsigma\rangle$ — describes virtual states: $|V_{hh}, m'_{hh}\rangle (m'_{hh} = \pm 3/2)$ and for the subzones of heavy and light holes; $|\mathrm{SO}, m'_{\mathrm{SO}}\rangle (m'_{\mathrm{SO}} = \pm 1/2)$ — for the spin-orbit splitting subzone; $M^{(N)}_{c,m'_c;\varsigma,m'_\varsigma;V,m'_V}(\mathbf{k})$ — composite matrix element of

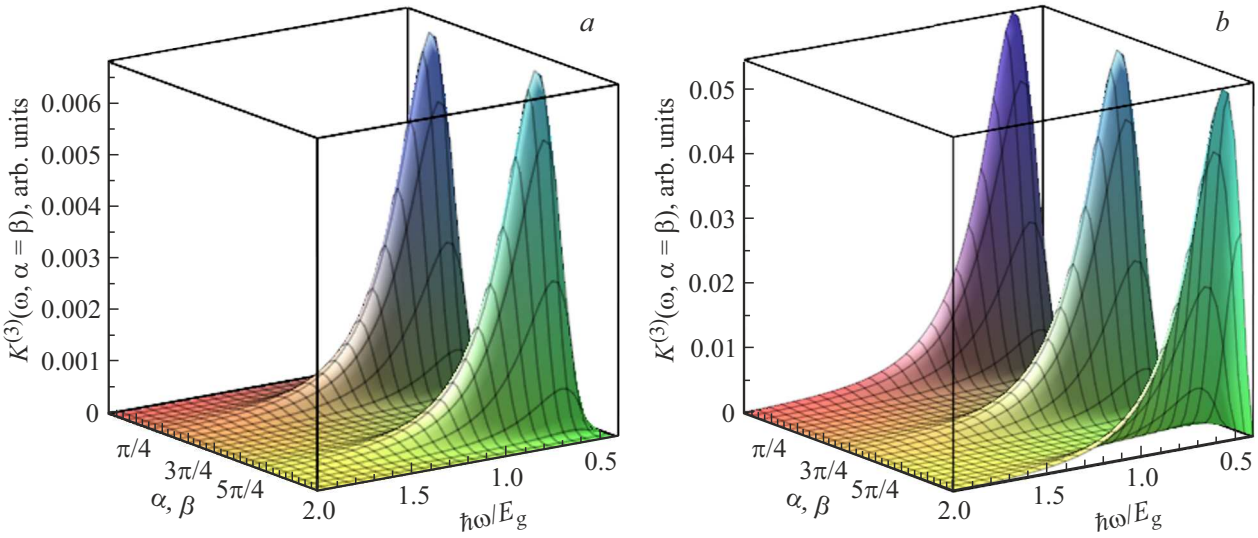

Figure 2. Frequency-polarization dependences of the three-photon light absorption coefficient in InSb. *a* — corresponds to circular polarization, *b* — linear polarization.

the optical transition under consideration, $E_c\mathbf{k}[E_{\mathrm{SO}}(\mathbf{k})]$ and $f_c(\mathbf{k})[f_{\mathrm{SO}}(\mathbf{k})]$ — the energy spectrum of current carriers in the conduction band (in the spin-orbit splitting subzone) and the distribution function of current carriers, m'_ς — the eigenvalue of the operator of the full moment, $\varsigma = c, V_l$, SO. The matrix element of the electron-single-photon interaction operator is defined as

$$H_{ll'} = \frac{e}{im_0\omega}\left(\frac{2\pi I}{n_\omega c}\right)^{1/2}(\mathbf{e}\cdot\mathbf{p})_{ll'},$$

where $\mathbf{p}$ — pulse operator, I — light intensity, refractive index of the crystal at a frequency of ω.

Since $M^{(N)}_{c,m'_c;\varsigma,m'_\varsigma;V,m_V}(\mathbf{k}$ has several components (see the table), then in calculations it is necessary to use (for example, for A and B types of optical transition) the following relation:

$$\begin{aligned}\left|M^{(N)}_{c,m'_c;\varsigma,m_\varsigma;Vm'_V}\left(k^{N_\omega}_{c,m_{V_l}}\right)\right|^2 &= \left|M^{(A)}_{c,m'_c;\varsigma,m'_\varsigma;V,m'_V}\left(k^{(N\omega)}_{c,m_{V_l}}\right)\right|^2 \\ &+ \left|M^{(B)}_{c,m'_c;\varsigma,m'_\varsigma;V,m'_V}\left(k^{(N\omega)}_{c,m_{V_l}}\right)\right|^2 \\ &+ \left[M^{(A)}_{c,m'_c;\varsigma,m'_\varsigma;V,m'_V}\left(k^{(N\omega)}_{c,m_{V_l}}\right)\right]^* M^{(B)}_{c,m'_c;\varsigma,m/_\varsigma;V,m'_V}\left(k^{(N\omega)}_{c,m_{V_l}}\right) \\ &+ M^{(A)}_{c,m'_c;\varsigma,m'_\varsigma;V,m'_V}\left(k^{(N\omega)}_{c,m_{V_l}}\right)\left[M^{(B)}_{c,m'_c;\varsigma,m'_\varsigma;V,m'_V}\left(k^{(N\omega)}_{c,m_{V_l}}\right)\right]^*, \end{aligned} \quad (4)$$

where the wave vector of the current carriers in the final state, determined from the law of conservation of energy for the N photon transition, is equal to

$$k^{(N\omega)}_{c,m_{\mathrm{SO}}} = \left[\frac{2m_c m_{\mathrm{SO}}}{m_c + m_{\mathrm{SO}}}\right](N\hbar\omega - E_g - \Delta_{\mathrm{SO}}),$$

Δ_{SO} — spin-orbit splitting energy. Note that the energy spectra of current carriers in all zones are assumed to be parabolic (if the three branches of the band structure of a semiconductor are non-parabolic, then the quadratic dependence of energy on the wave vector is approximately restored only near the edges of the Brillouin zones), i.e.

$$E_c(\mathbf{k}) = E_g + \frac{\hbar^2K^2}{2m_c}, \quad E_{\mathrm{SO}}(\mathbf{k}) = -\Delta_{\mathrm{SO}} + \frac{\hbar^2k^2}{1m_{\mathrm{SO}}},$$

where $m_c(m_{\mathrm{SO}})$ — effective masses in the zone $c(\mathrm{SO})$.

For three-photon transitions of the type $|\mathrm{SO}, \pm1/2\rangle \Rightarrow |c, \mp1/2\rangle$: if both virtual states lie in the subzones of the valence band, then the graph of the polarization dependence of the coefficient of three-photon linear-circular dichroism is described in Fig. 1, *d*. Similarly, the polarization dependences of the linear-circular dichroism coefficient of three-photon light absorption are determined, which differ from the above cases by the type and order of virtual states through which three-photon optical transitions occur. In quantitative calculations, the numerical values of the zone parameters are taken from the work [16].

Thus, the polarization dependence of the linear-circular dichroism of three-photon light absorption on the type of optical transitions was shown. Since the matrix elements of some optical transitions in the numerator have differences in the energy of current carriers, which tend to zero in a certain region of the light frequency, which leads to an abnormal increase in the spectral dependence of the three-photon light absorption coefficient. But this phenomenon does not lead to a noticeable change in the spectral dependence of linear-circular dichroism, since the linear-circular dichroism coefficient is determined by the ratio of the probabilities of optical transitions occurring during the absorption of light of linear and circular polarizations.

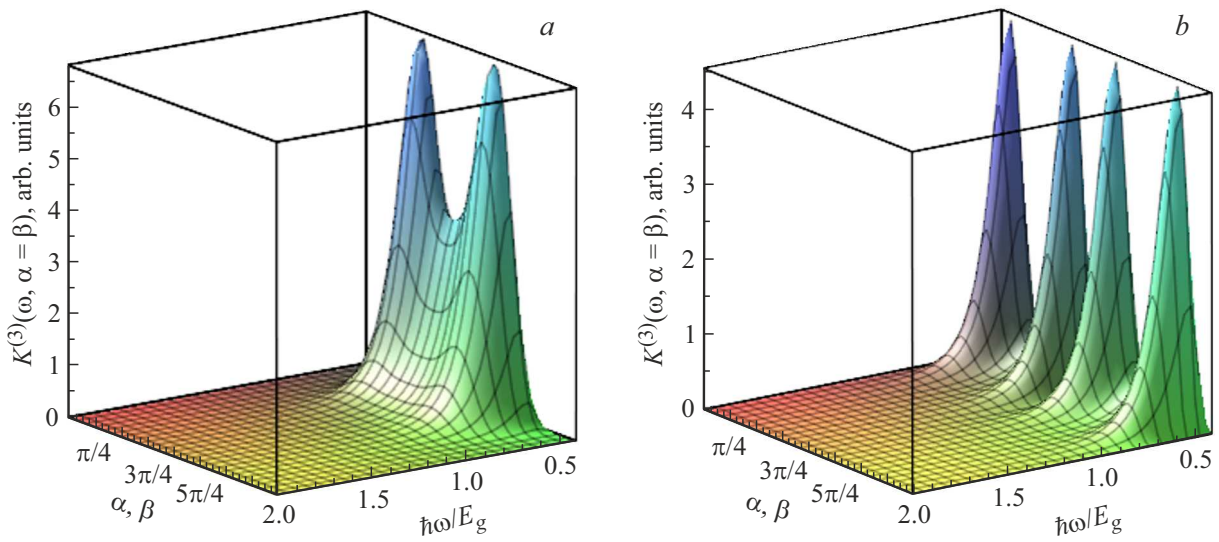

Figure 3. Frequency-polarization dependences of the three-photon light absorption coefficient in InSb. *a* — corresponds to circular polarization, *b* — linear polarization.

3. Polarization-spectral dependences of the three-photon light absorption coefficient

Now let us analyze the polarization-spectral dependences of the light absorption coefficient for InSb due to three-photon transitions of the type $|\mathrm{SO}, \pm 1/2\rangle \to |c, \pm 1/2\rangle$ and $|\mathrm{SO}, \pm 1/2\rangle \to |c, \mp 1/2\rangle$, calculated by the formula

$$K^{(N)}(\omega, T) = \frac{N\hbar\omega}{I} \sum_{c, m'_c; \varsigma, m'_\varsigma; \mathrm{SO}, m'_{\mathrm{SO}}} W^{(N)}_{c, m'_c; \varsigma, m'_\varsigma; \mathrm{SO}, m'_{\mathrm{SO}}}. \quad (5)$$

Calculations show that the graph of the polarization dependence of the three-photon light absorption coefficient in the InSb semiconductor for the above three-photon transitions is shown in Fig. 2, when two virtual states lie in the valence band subzones, and Fig. 3, if the initial virtual states lie in the conduction band, and the following — in the valence band.

Calculations show that the main contribution to the coefficient of three-photon light absorption is made by optical transitions, in which the initial virtual states lie in the conduction band, and the following — in the valence band.

4. Conclusion

From the above results and Fig. 1–3, it can be seen that both the polarization and frequency-polarization dependences of the coefficients of three-photon light absorption and linear-circular dichroism have several extremes. This is explained by the specifics of the Kane and Luttinger–Cohn models used in the study of the band structure in narrow-band crystals [13,14]. In particular, in the Kane model, some non-diagonal matrix elements of the momentum operator do not depend, and in the Luttinger–Kohn approximation depend on the wave vector of the current carriers. Therefore, each type of optical transitions gives an unequal contribution to the frequency and frequency-polarization dependences of the three-photon light absorption coefficient and linear-circular dichroism, since the energy denominators in the composite matrix element of the optical transition under consideration depend on the type of virtual states through which single-photon transitions pass.

In conclusion, we note that it is possible to analyze the frequency-polarization dependences of interband optical transitions occurring from the subzones of light and heavy holes of the valence band to the conduction band. This case requires separate consideration, to which a separate work will be devoted.

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Conflict of interest

The authors declare that they have no conflict of interest.

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