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INTERBAND ONE- AND TWO-PHOTON ABSORPTION OF POLARIZED LIGHT IN NARROW-GAP CRYSTALS

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МЕЖЗОННОЕ ОДНО- И ДВУХФОТОННОЕ ПОГЛОЩЕНИЕ ПОЛЯРИЗОВАННОГО СВЕТА В УЗКОЗОННЫХ КРИСТАЛЛАХ

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ТОР ЗОНАЛИ КРИСТАЛЛАРДА ҚУТБЛАНГАН ЁРУҒЛИКНИНГ ЗОНАЛАРАРО БИР ВА ИККИ ФОТОНЛИ ЮТИЛИШИ

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Abstract: Interband two-photon optical transitions are analyzed, and expressions are obtained for the matrix elements in a narrow-gap crystal depending on the band parameters, degree of polarization, and light frequency. It is shown that the main contribution to two-photon linear-circular dichroism in narrow-gap semiconductors is made by optical transitions proceeding from the subband of light holes to the conduction band.

The partial coefficients of interband one- and two-photon absorption of light are calculated, which differ from each other in the types of optical transitions in the Kane model.

Key words: initial, virtual and final states, between band two-photon absorption of light, Kane's approximation, optical transition.

Аннотация: Проанализированы межзонные двухфотонные оптические переходы и получены выражения для матричных элементов в узкозонном кристалле в зависимости от параметров зоны, степени поляризации и частоты света. Показано, что основной вклад в двухфотонный линейно-циркулярный дихроизм в узкозонных полупроводниках вносят оптические переходы, идущие из подзоны легких дырок в зону проводимости.

Вычислены парциальные коэффициенты межзонного одно- и двухфотонного поглощения света, которые отличаются друг от друга типами оптических переходов в модели Кейна.

Ключевые слова: начальное, виртуальное и конечное состояния, межзонное двухфотонное поглощение света, приближение Кейна, оптический переход.

Аннотация: Тор зонали кристаллда зоналараро икки фотонли оптик ўтишлар таҳлил қилинган ва матрицавий элементлар учун зонавий параметрларга, қутбланиши даражасига ва ёруғлик частотасига боғлиқ муносабатлар олинган. Тор зонали яримўтказгичларда валент зонаси енгил коваклар тармоғидан ўтказувчанлик зонасига оптик ўтишлар икки фотонли чизиқли-циркуляр дихроизмга асосий улуш берилиши аниқланган, оптик ўтиши турлари билан фарқ қилувчи зоналараро ёруғликнинг бир ва икки фотонли парциал ютилиши коэффициентлари ҳисобланган.

Калит сўзлар: бошланғич, виртуал ва якуний ҳолатлар, тармоқлараро ёруғликнинг икки фотонли ютилиши, Кейн яқинлашиши, оптик ўтиш.

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Introduction

Nonlinear optical phenomena occurring in crystals are now widely used in practice [1-3]. In this aspect, the research of nonlinear absorption of polarized light is relevant both from a physical point of view and from the point of view of practical application. Note that in the case of one-photon absorption of light, optical transitions do not occur through virtual states. Therefore, linear-circular dichroism is not observed in single-photon optical transitions in crystals of cubic and tetrahedral symmetry.

The first works on two-photon interband transitions in crystals were carried out in the early 1960s, soon after the appearance of lasers. In calculating the matrix elements of two-photon transitions in crystals, perturbation theories in the field of an unpolarized electromagnetic wave were used, where the two-band Kane model was used.

In [1-2], both theoretically and experimentally, linear-circular dichroism (LCD) of two- and three-photon absorption of light in crystals of cubic symmetry was investigated.

Multiphoton absorption of light in a semiconductor with a complex valence band, caused by direct optical transitions between the subbands of heavy and light holes and depending on the degree of polarization of light, was investigated in [3-9]. Nonlinear interband one-photon absorption of polarized light in Weyl semimetals was studied in [9]. In these works, it is believed that the nonlinearity in the dependence of the one-photon absorption coefficient on the light intensity arises due to resonant absorption saturation [10]. This saturation in both interband [10] and intraband [4, 7, 9] absorption of light is due to a photoinduced change in the distribution functions of current carriers in the momentum space region near the surface, determined by the energy conservation law, as well as by the relaxation time, the inverse value which is equal to the reciprocal values of the relaxation times in energy and momentum.

In [3], multiphoton linear-circular dichroism (LCD) in p-Ge was investigated in the developed nonlinearity regime, when n-photon processes with a comparable contribution to the absorption. In [4], four-photon processes in semiconductors, caused by optical transitions between subbands of the valence band, were researched, taking into account the effect of coherent saturation.

Note that in [3] a theory of linear-circular dichroism of multiphoton interband absorption of different frequencies and polarization of light in semiconductors near the center of the Brillouin zone was constructed in the three-band approximation, when the condition is

satisfied $\frac{2\pi e^2 I |\vec{e}\vec{p}_{cv}|^2}{cn_\omega \omega^2 m_0^2 (\hbar\omega)^2} \ll 1$, where \vec{e} and I are the polarization vector and light intensity, $\vec{e}\vec{p}_{c\vec{k},v\vec{k}}$ is

the interband matrix element of the momentum operator, is the refractive index of light of the medium at the frequency ω , m_0 is the mass of a free electron, the rest of the quantities are well known.

In this work, in contrast to [3], we calculate the LCD of the interband two-photon absorption of light (TPA), as well as the spectral dependence of the TPA coefficient in semiconductors of the InSb type in the Kane model, which takes into account the contributions to the multi-quantum process of intermediate states in the subbands of light and heavy holes and in the valence band split off due to the spin-orbit interaction, as well as in the conduction band, taking into account the effect of coherent saturation. Note here that in semiconductors of the InSb type, the energy distance between the adjacent lower and upper conduction bands is much larger than the widths of the forbidden or spin-split bands [10], which allows further investigations in the two-band approximation.

Single-photon interband absorption of polarized light in narrow-gap semiconductors

According to [1,2], the coefficient of one-photon absorption of light is determined as follows:

$$K^{(1)}(\omega, T) = \frac{2p}{h} \frac{\hbar\omega}{I} \sum_{l,l\vec{k},r} \dot{a}_r (f_{l\vec{k}}^r - f_{l\vec{k}}^r) \left| M_{l\vec{k},l\vec{k}}^r \right|^2 d(E_{l\vec{k}}^r - E_{l\vec{k}}^r - \hbar\omega), \quad (1)$$

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where $M_{l\phi, lk}^r$ is the composite matrix element of the $|l\phi\rangle^r \otimes |lk\rangle^r$ type optical transition, where $|l\phi\rangle^r (|lk\rangle^r)$ describes the final (initial) state of charge carriers, E_{lk}^r is the energy spectrum of charge carriers in the band number l , where $l = c$ is for the conduction band, $l = V$ for the valence band, and $l = SO$ for the spin split off zone.

To simplify the calculations, we will use the spherically symmetric approximation in the energy spectrum of current carriers (two-band Kane approximation, see, for example, [7,8]), ie,

$$E_c(k)|_{k \rightarrow 0} = E_c(k=0) + \frac{2P_{CV}^2}{3E_g} k^2 \equiv E_c(k=0) + \frac{\hbar^2 k^2}{2m_{el}},$$

$$E_{lh}(k)|_{k \rightarrow 0} = E_c(k=0) - E_g - \frac{2P_{CV}^2}{3E_g} k^2 \equiv E_c(k=0) - E_g - \frac{\hbar^2 k^2}{2m_{lh}} \quad (2)$$

(see Appendix), where it is assumed that the effective masses of electrons and light holes are the same, and the effective mass of heavy holes is equal to infinity. E_g is the band gap, P_{CV} is the Kane parameter determined by the relation $-iP_{CV}\delta_{\alpha\beta} = \int dx dy dz S(\vec{r}) \hat{P}_\alpha X_\beta(\vec{r})$, $X_\beta(\vec{r}) = | \Gamma_6, S \rangle, | \Gamma_8, m \rangle, | \Gamma_7, m' \rangle$ is one of the basis functions in the Luttinger-Kohn approximation (see, for example, formula (13.18) in [8]). Note here that the Luttinger-Kohn parameters [7,8] are expressed by the Kane parameter as follows:

$$D = -\hbar^2 P_{CV}^2 / (\sqrt{3} m_0^2 E_g), \quad A - B = \hbar^2 / (2m_0), \quad A + \frac{B}{2} = \frac{\hbar^2}{2m_0} - \frac{\hbar^2 P_{CV}^2}{2m_0^2 E_g}, \quad (3)$$

Then the polarization dependence of the coefficient of one-photon absorption of light caused by vertical optical transitions between the conduction and valence bands of the crystal is determined by the expression

$$K_0^{(1)} = \frac{4\pi^2 e^2}{c\omega m_0^2 n_\omega} \sum_{s,l,m;\vec{k}} \left| e\vec{p}_{cs,V_l m}(\vec{k}) \right|^2 \delta(E_{c\vec{k}} - E_{V_l \vec{k}} - \hbar\omega) =$$

$$= \frac{\pi^2 e^2}{c\omega n_\omega m_0^2} P_{CV}^2 \left[2 \left(\left| \vec{e}_+ \right|^2 + \left| \vec{e}_- \right|^2 \right) \rho_{c,V_1}^{(0)}(\omega) + \frac{1}{3} \left(\left| \vec{e}_+ \right|^2 + \left| \vec{e}_- \right|^2 \right) + 8 \left| \vec{e}_z \right|^2 \rho_{c,V_2}^{(0)}(\omega) \right], \quad (4)$$

where n_ω is the refractive index of the medium at a frequency of ω

$$\rho_{c,V_l}^{(0)}(\omega) = \sum_{\vec{k}} \delta(E_c(\vec{k}) - E_{V_l}(\vec{k}) - \hbar\omega) = \frac{1}{2} \pi^{-2} \hbar^{-2} \mu_l k_l^{(0)}, \quad k_l^{(0)} = \left[2\mu_l (\hbar\omega - E_g) / \hbar^2 \right]^{1/2},$$

$\mu_l = (m_c^{-1} + m_l^{-1})^{-1}$ is the reduced effective mass of current carriers, in particular at $l = lh(hh)$: $\mu_{hh} = m^*$, $\mu_{lh} = m^* / 2$, the index (0) means that the uncertainty arising from the $\vec{k}\vec{p}$ - interaction was not taken into account in the intermediate calculations.

Then, after averaging in (4) over the solid angles of the wave vector and in approximation (2), i.e. disregarding nonparabolicity in the energy spectrum of current carriers, we have

$$K_0^{(1)} = \frac{2e^2}{cn_\omega} \frac{P_{CV}^2}{\omega m_0^2} \frac{2}{3} \sum_{l=1,2} \mu_l k_l^{(0)} \hbar^{-2} = \frac{e^2}{cn_\omega \hbar} \frac{E_g}{\hbar\omega} k_1^{(0)} \left(1 + \frac{1}{\sqrt{2}} \right), \quad (5)$$

where $k_2^{(0)} = k_1^{(0)} / \sqrt{2}$ was taken into account for crystal *InSb* and it was assumed that the initial state of the current carriers is completely occupied, and the final state is completely free. If we take into account the nonparabolicity in the energy spectrum, then (5) takes the form

$$K^{(1)} = K_0^{(1)} \frac{E_g}{\hbar\omega} \left(\frac{\hbar\omega}{E_g} - 1 \right)^{1/2} \left[\left(\frac{\hbar\omega}{E_g} \right)^{3/2} + \frac{1}{12} \left(1 + 2 \left(\frac{E_g}{\hbar\omega} \right)^2 \right) \cdot \frac{\hbar\omega}{E_g} \cdot \left(\frac{\hbar\omega}{E_g} + 1 \right)^{1/2} \right]. \quad (6)$$

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where $K_0^{(1)} = e^2 \sqrt{2m^* E_g} / (\hbar^2 c n_\omega)$.

The spectral dependence of the light absorption coefficient without taking into account the effect of coherent saturation of one-photon optical transitions in narrow-gap crystals is shown in Fig. 1, where $\xi = \hbar\omega / E_g$. As can be seen from Fig. 1, in the region of low frequencies, the dependence $K^{(1)}(\xi)$ is determined by a power function with a degree less than unity, and in the region of high frequencies, the spectral dependence $K^{(1)}(\xi)$ will be linear. In particular, if the frequency is doubled, then for an InSb crystal, $K^{(1)}(\xi)$ it increases 1.6 times. Such an inadequate dependence of the quantity is described with the complexity of the band structure of the crystal, i.e. nonparabolicity of the energy spectrum of current carriers.

If we take into account the contribution of the coherent saturation effect to the absorption, then (4) has the form

$$K^{(1)}(\omega, T) = \frac{4\pi}{\hbar} \hbar\omega \frac{1}{I} \sum_{k,s=\pm 1/2, m=\pm 1/2, \pm 3/2} (f_{hh} - f_c) \delta(E_{hh} - E_c + \hbar\omega) \left\langle \frac{|M_{C,s;V,m}^{(1)}(\vec{k})|^2}{\sqrt{1 + 4 \frac{\alpha_\omega}{\hbar^2 \omega^2} |M_{C,s;V,m}^{(1)}(\vec{k})|^2}} \right\rangle, \quad (7)$$

where $f_{hh}(f_c)$ and $E_{hh}(E_c)$ are the distribution functions and energy spectra of holes (electrons), respectively, the sign $\langle \dots \rangle$ means averaging over the solid angles of the wave vectors of current carriers, the rest are well-known values.

It can be seen from (7) that the coefficient of interband one-photon absorption of light $K^{(1)}(\omega, T)$ consists of partial components that differ from each other in the types of optical transitions. In particular, for a $|V, \pm 3/2\rangle \rightarrow |C, \pm 1/2\rangle$ -type optical transition is expressed as

$$K^{(1)}(\omega, T) = \frac{16e^2}{3c\omega \hbar^2 n_\omega} \mu_{c,L}^{(+)} \cdot k_{c,L}^{(\omega)} \cdot P_{cv}^2 \cdot F(\beta, 1, \omega) \cdot \mathfrak{Z}(\omega) \cdot \left[f_{hh}(E_{hh}(k_{c,L}^{(\omega)})) - f_c(E_c(k_{c,L}^{(\omega)})) \right], \quad (8)$$

here $F(\beta, 1, \omega) = [1 - \exp(\beta\hbar\omega)] \exp[\beta(\mu - E_{hh}(k_{c,L}^{(\omega)}))]$, $\zeta_\omega = \frac{8\pi e^2 \alpha_\omega I}{c n_\omega (\hbar\omega)^4} P_{CV}^2$,

$$k_{c,L}^2 = \frac{2\mu_{c,L}^{(+)}}{\hbar^2} (\hbar\omega - E_g), \quad \frac{1}{\mu_{c,L}^{(+)}} = \left(\frac{1}{m_c} + \frac{1}{m_L} \right), \quad \beta^{-1} = k_B T, \quad J(\zeta_\omega, \omega) = \left\langle \frac{|e'_\pm|^2}{\sqrt{1 + \zeta_\omega |e'_\pm|^2}} \right\rangle.$$

It can be seen from (8) that the linear-circular dichroism of one-photon absorption of light is determined by a quantity $J(\zeta_\omega, \omega)$ that depends on the frequency and degree of polarization of the light, the band parameters of the sample, and on the relaxation times of photoexcited current carriers.

Note that if we do not take into account the effect of coherent saturation ($\zeta_\omega = 0$), then $K^{(1)}(\omega, T)$ it does not depend on the quantities mentioned above, in particular, on the degree of polarization of light, i.e. is a constant number: $J(\zeta_\omega = 0) = \frac{4}{3}$, i.e. in this case, one-photon linear-circular dichroism is not observed. However, if we take into account the effect of coherent saturation, then what does this mean in this case there is one-photon linear-circular dichroism. This is due to the fact that for linearly polarized light

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$$J_{lin} = \int_{-1}^{+1} d\mu \frac{1 - \mu^2}{\sqrt{1 + \zeta_\omega (1 - \mu^2)}}; \tag{9}$$

for circularly polarized light

$$J_{circ} = \int_{-1}^{+1} d\mu' \frac{\frac{1}{2}(1 + \mu'^2) \mp P_{circ} \mu'}{\sqrt{1 + \zeta_\omega \left[\frac{1}{2}(1 + \mu'^2) \mp P_{circ} \mu' \right]}}, \tag{10}$$

where P_{circ} is the degree of circular polarization of light, sign " \pm " refers to σ_{\pm} polarized light, $\phi(\phi')$ is the angle between vectors \vec{e} and \vec{q} , $\mu' = \cos \phi'$, $\mu = \cos \phi$, \vec{q} is the photon wave vector.

For example, in the case $P_{circ} = 1$
for linearly polarized light

$$J_{lin} = \zeta_\omega^{-5/2} \left\{ \zeta_\omega^{3/2} + \zeta_\omega^2 \cdot \arcsin \left(\frac{\zeta_\omega}{1 + \zeta_\omega} \right)^{1/2} - \zeta_\omega \cdot \arcsin \left(\frac{\zeta_\omega}{1 + \zeta_\omega} \right)^{1/2} \right\}, \tag{11}$$

for circularly polarized light

$$J_{circ} = \frac{2 \left(\zeta_\omega^{3/2} \sqrt{\zeta_\omega + 1} - \zeta_\omega \arcsin \sqrt{\zeta_\omega} \right)}{\zeta_\omega^{5/2}}. \tag{12}$$

In fig. 2 it is shown the graphs of functions $J_{lin}(\zeta_\omega)$ and $J_{circ}(\zeta_\omega)$ versus

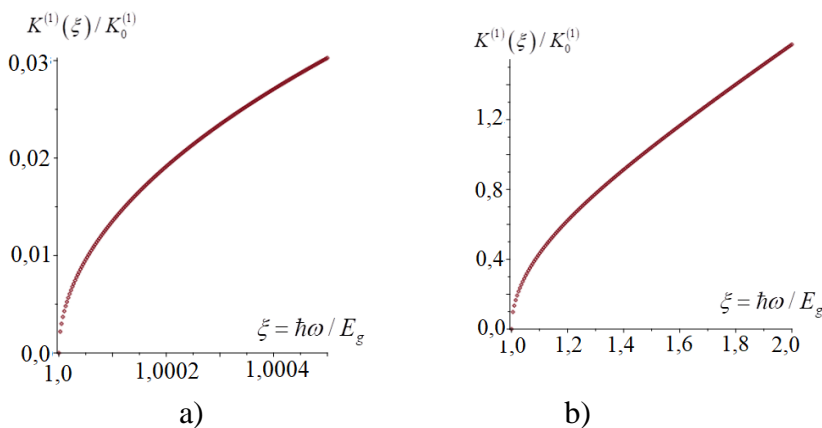


Fig. 1. Spectral dependence of the coefficient of one-photon interband absorption of light in a narrow-gap crystal without taking into account the effect of coherent saturation: a) the region of low frequencies; b) high frequency region, where.

$$\zeta_\omega \propto \left(\frac{eA_0}{c\hbar} \right)^2 \propto I. \text{ As can be}$$

seen from Fig. 2, as the light intensity increases, the coefficient of interband single-photon linear-circular dichroism

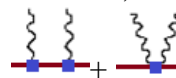
$$\eta = J_{lin}(\zeta_\omega) / J_{circ}(\zeta_\omega)$$

increases and tends to saturation, i.e. at very high values of the intensity ($\zeta_\omega \gg 1$), this value does not depend on the intensity and $\eta \approx 1.1$. In quantitative

calculations, we used the data from [7].

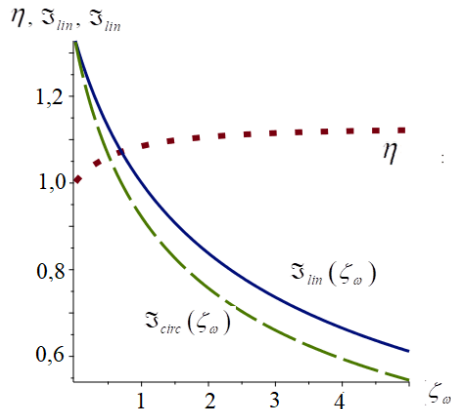
Classification of two-photon interband optical transitions

It is known [4,5-10,15-22] that the probabilities of one- or multi-photon OTs and the corresponding LCD light absorption coefficients are determined using the composite matrix elements of the OT under consideration. Therefore, in the future, we will analyze the matrix

elements associated with two-photon OTs characterized by  type Feynman diagrams, which differ from each other in the choice of the initial states of the current carriers:

- a) let the initial states of current carriers be located in the subband of

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b) c) Fig. 2. Graphs of functions $J_{lin}(\zeta_\omega)$, $J_{circ}(\zeta_\omega)$ and η (linear-circular dichroism factor) versus $\zeta_\omega \propto I$ (on light intensity) in the Kane model in a narrow-gap crystal.

d) heavy holes ($|V, \pm 3/2\rangle$) with the energy E_{hh} , and the virtual states - in the subband of heavy and light holes ($|V, \pm 1/2\rangle$) of the valence band of the semiconductor. In this case, in the Kane model, the matrix element of interband OTs of type $|V, \pm 3/2\rangle \rightarrow |m\rangle \rightarrow |c, \pm 1/2\rangle$, $|V, \pm 3/2\rangle \rightarrow |m\rangle \rightarrow |c, \mp 1/2\rangle$ is determined by the matrix in the order of $(c, +1/2), (c, -1/2)$ and $(V, +3/2), (V, -3/2)$

$$\|M_{m',m}^{(2)}\| = \left(\frac{eA_0}{c\hbar}\right)^2 P_{cV} k \left\| \begin{array}{c} \frac{2(A-B)e'_+e'_{z'}}{-\hbar\omega} + \frac{e'_-B}{E_{lh} - E_{hh} - \hbar\omega} \\ \frac{i\sqrt{2}e'_+e'_-B}{E_{lh} - E_{hh} - \hbar\omega} \\ \frac{\sqrt{2}Be'_+e'_-}{E_{lh} - E_{hh} - \hbar\omega} \end{array} \right\|, \quad (13)$$

where A_0 is the amplitude of the potential vector of an electromagnetic wave ($I = \frac{n_w w^2 A_0^2}{2pc}$),

$M_{c,m';V,m}^{(1)}$ is the matrix element of a single-photon OT of type $|V, m\rangle \rightarrow |c, m'\rangle$, P_{cV} is the Kane parameter [7,8], $E_{lh}(E_{hh})$ is the energy of light (heavy) holes, A, B are the band parameters of the crystal, corresponds to the states of electrons with an energy of $|c, \pm 1/2\rangle$ in the conduction band. The energy conservation law for the indicated OTs is expressed using the function $\delta(E_c - E_{hh} - 2\hbar\omega)$, E_{hh} is the energy of heavy holes;

b) let the initial states of current carriers are located in the subband of light holes of the valence band, and virtual states - in the are subbands of heavy and light holes of the valence band. In this case, in the Kane model, the matrix element of interband OTs of type $|V, \pm 1/2\rangle \rightarrow |m\rangle \rightarrow |c, \pm 1/2\rangle$, $|V, \pm 1/2\rangle \rightarrow |m\rangle \rightarrow |c, \mp 1/2\rangle$ is determined by the matrix in the order of $(c, +1/2), (c, -1/2)$ and $(V, +1/2), (V, -1/2)$

$$\|M_{m',m}^{(2)}\| = \left(\frac{eA_0}{c\hbar}\right)^2 P_{cV} k \left\| \begin{array}{c} \left(\frac{\sqrt{3}Be'_+{}^2}{E_{hh} - E_{lh} - \hbar\omega} - \frac{2(A+B)e'_-e'_{z'}}{\sqrt{3}\hbar\omega} \right) \\ -i2\sqrt{\frac{2}{3}} \frac{(A+B)e'_+{}^2}{\hbar\omega} \\ 2\sqrt{\frac{2}{3}} \frac{(A+B)e'_-{}^2}{(-\hbar\omega)} \end{array} \right\|, \quad (14)$$

The law of conservation of energy for these OT is expressed using the function $\delta(E_c - E_{lh} - 2\hbar\omega)$, E_{lh} - the energy of light holes.

c) let the initial states of current carriers are located in the subband of light holes of the valence band, and virtual states - in the conduction band. In this case, in the Kane model, the matrix

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element of interband OTs of type $|V, \pm 1/2\rangle \rightarrow |m\rangle \rightarrow |c, \pm 1/2\rangle$, $|V, \pm 1/2\rangle \rightarrow |m\rangle \rightarrow |c, \mp 1/2\rangle$ is determined by the matrix in the order of $(c, +1/2), (c, -1/2)$ and $(V, +1/2), (V, -1/2)$

$$\|M_{m',m}^{(2)}\| = \frac{1}{\sqrt{3}} \left(\frac{eA_0}{c\hbar} \right)^2 \frac{1}{\hbar\omega} \frac{\hbar^2 k}{m_c} P_{cV} k \begin{vmatrix} e'_+ & \sqrt{2}e'_z \\ -i\sqrt{2}e'_z & ie'_- \end{vmatrix}, \quad (15)$$

where $|m\rangle = |c, \pm 1/2\rangle$, E_g is the band gap, and the energy conservation law for the indicated OTs is expressed using the function $\delta(E_c - E_{lh} - 2\hbar\omega)$.

d) let the initial states of current carriers be located in the subband of light holes of the valence band, and the virtual states - in the zone of spin-orbital splitting. In this case, in the Kane model, the matrix element of interband OTs of type $|V, \pm 1/2\rangle \rightarrow |m\rangle \rightarrow |c, \pm 1/2\rangle$, $|V, \pm 1/2\rangle \rightarrow |m\rangle \rightarrow |c, \mp 1/2\rangle$ is determined by the matrix in the order of $(c, +1/2), (c, -1/2)$ and $(V, +1/2), (V, -1/2)$

$$\|M_{m',m}^{(2)}\| = \frac{1}{\sqrt{6}} \left(\frac{eA_0}{c\hbar} \right)^2 \frac{P_{cV}}{E_\Delta - E_{hh} - \hbar\omega} \begin{vmatrix} -\sqrt{3}e'_+ H'^* - ie'_z (G' - F') & -ie'_- (G' - F') + e'_z H'^* \\ ie'_z H' - e'_+ (G' - F') & e'_z (G' - F') + i\sqrt{3}e'_- H' \end{vmatrix}. \quad (16)$$

where $|m\rangle = |SO, \pm 1/2\rangle$, E_Δ is the energy spectrum of current carriers in the spin-orbit splitting zone, G', F', H' is the first derivative with respect to the wave vector of current carriers from the quantities G, F, H , which are determined by formula (24.20) [7], (*) is the sign of complex conjugation. The energy conservation law for this case is described by the function $\delta(E_c - E_{lh} - 2\hbar\omega)$.

e) let the initial states of current carriers be located in the subband of heavy holes in the valence band, and virtual states - in the zone of spin-orbit splitting. In this case, in the Kane model, the matrix element of interband OTs of type $|V, \pm 3/2\rangle \rightarrow |m\rangle \rightarrow |c, \pm 1/2\rangle$, $|V, \pm 3/2\rangle \rightarrow |m\rangle \rightarrow |c, \mp 1/2\rangle$ is determined by the matrix in the order of $(c, +1/2), (c, -1/2)$ and $(V, +3/2), (V, -3/2)$

$$\|M_{m',m}^{(2)}\| = \frac{1}{\sqrt{6}} \left(\frac{eA_0}{c\hbar} \right)^2 \frac{P_{cV}}{E_\Delta - E_{hh} - \hbar\omega} \begin{vmatrix} -\sqrt{3}e'_+ H'^* - ie'_z (G' - F') & -ie'_- (G' - F') + e'_z H'^* \\ ie'_z H' - e'_+ (G' - F') & e'_z (G' - F') + i\sqrt{3}e'_- H' \end{vmatrix}. \quad (17)$$

The law of conservation of energy is expressed using the function $\delta(E_c - E_{hh} - 2\hbar\omega)$.

Note that the wave vector in the final state of electrons participating in the interband OT is

determined using the expression $k_{c,L}^{(2\omega)} = \sqrt{\frac{2\mu_+^{(c,L)}}{\hbar^2} (2\hbar\omega - E_g)}$, where m_c is the effective mass of electrons in the conduction band, m_L is the effective mass of holes in the L subband, $L = lh$ ($L = hh$) is for light (heavy) holes, $\mu_+^{(c,L)} = \frac{m_c m_L}{m_c + m_L}$ is the reduced effective mass of current

carriers. Then the following relations are valid for the energies of light and heavy holes: a) if the OT comes from the subband of heavy holes, then

$$E_{L=hh}(k_{c,L=hh}^{(2\omega)}) = \frac{m_c}{m_c + m_{hh}} (2\hbar\omega - E_g), \quad E_{lh}(k_{c,L=hh}^{(2\omega)}) = \frac{m_c \cdot m_{hh}}{m_{lh} (m_c + m_{hh})} (2\hbar\omega - E_g);$$

b) if the OT comes from the subband of light holes, then

$$E_{L=hh}(k_{c,L=hh}^{(2\omega)}) = \frac{m_c \cdot m_{lh}}{m_{hh} (m_c + m_{lh})} (2\hbar\omega - E_g), \quad E_{lh}(k_{c,L=hh}^{(2\omega)}) = \frac{m_c}{m_c + m_{lh}} (2\hbar\omega - E_g).$$

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The matrix elements of two-photon transitions originating from the spin-split band into the conduction band, where the virtual states of the current carriers are located in the subbands of the valence band, in the conduction band, and also in the spin-orbit splitting band of a semiconductor are determined in a similar way as in the above cases .

Interband two-photon absorption of polarized light and its linear-circular dichroism

In this section, we obtain an expression for the spectral dependence of the coefficient of interband TPA of light in narrow-gap semiconductors in the Kane model. In further calculations, we use the calculation method proposed in [1,2,9]).

Note that the multiphoton light absorption coefficient consists of partial components, which by their nature depend on the zone in which the current carriers are located both in the initial and in the virtual state.

In further (intermediate) calculations, instead of $\sum_k (f_L - f_c) \delta(E_c - E_L - 2\hbar\omega) F(k)$, we use the expression $\frac{1}{(2\pi)^3} F(k_{c,L}) k_{c,L}^2$, where $k_{c,L}$ is the wave vector determined from the energy conservation law: $E_c - E_L - 2\hbar\omega = 0$. In particular, in the spherical approximation in the energy spectrum of current carriers, i.e. in the case of $E_L = E_L^{(0)} + \frac{\hbar^2 k^2}{2m_L}$, the wave vector of the current carriers participating in the interband OT is defined as $k_{c,L}^2 = \frac{2\mu_+^{(c,L)}}{\hbar^2} (2\hbar\omega - E_g)$, where $\mu_+^{(c, hh)} = \frac{m_c m_L}{m_c + m_L}$ is the reduced effective mass,

m_L is the effective mass of the current carriers in the zone (or subband) with number L . In particular, $L = c$ for the conduction band, then $E_L^{(0)} = E_g$, and $L = lh (hh)$ for the subband of light (heavy) holes in the valence band, then $E_L^{(0)} = 0$.

Note that the frequency dependence of the denominators in the matrix elements of the OT is determined by the energy conservation law, the type of the considered OT and virtual states. For example, if the virtual states are in the valence band, and the initial one is in the subband of heavy holes, then the denominator in the matrix element of this transition is determined by the expression

$$E_{hh} - E_{lh} - \hbar\omega = \frac{m_c}{m_{hh}} \frac{m_{hh} - m_{lh}}{m_c + m_{lh}} (2\hbar\omega - E_g) + \hbar\omega, \text{ if this transition occurs from the subband of light holes, then the denominator in the matrix element of this transition is determined as}$$

$$E_{lh} - E_{hh} - \hbar\omega = \frac{m_c}{m_{lh}} \frac{m_{hh} - m_{lh}}{m_{hh} + m_c} (2\hbar\omega - E_g), \text{ where the ratios are taken into account:}$$

$$A - B = \frac{\hbar^2}{2m_{hh}}, A + B = \frac{\hbar^2}{2m_{lh}}.$$

In the future, we calculate the partial two-photon absorption coefficients that differ from each other from the types of OT, i.e. from the initial, intermediate and virtual states:

a) if the initial state is in the subband of heavy holes in the valence band, then in this case the coefficient between the two-photon light absorption is determined by the expression

$$K_{C, \pm 1/2; V, \pm 3/2}^{(2)} = \frac{8\pi^2}{\hbar} \hbar\omega \frac{1}{I} \frac{(\mu_+^{(c, hh)})^{3/2}}{(2\pi)^3 \hbar^3} \sqrt{2} \sqrt{2\hbar\omega - E_g} f_{hh} \left[\frac{m_c}{m_c + m_{hh}} (2\hbar\omega - E_g) \right] \left[\left(\frac{eA_0}{c\hbar} \right)^2 P_{cV} k \right]^2 \mathfrak{R}_{C, \pm 1/2; V, \pm 3/2}^{(2)}, \quad (18)$$

here $\Xi_{C,L}^{(2)} = \sum_k (f_L - f_c) \delta(E_c - E_L - 2\hbar\omega)$, $e'_\pm = e'_x \pm ie'_y$ and here (and further) it was assumed that

$$O_z \parallel \vec{k},$$

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$$\mathfrak{R}_{C,\pm 1/2;V,\pm 3/2}^{(2)} = \frac{1}{4\pi} \left\langle \left| \frac{2(A-B)e'_+e'_-}{(-\hbar\omega)} + \frac{e'_-\mathcal{B}}{(E_{lh} - E_{hh} - \hbar\omega)} \right|^2 \right\rangle + \left\langle \left| \sqrt{2}B \frac{e'_+e'_-}{(E_{lh} - E_{hh} - \hbar\omega)} \right|^2 \right\rangle =$$

$$= \frac{B^2}{15(\hbar\omega)^2} \left[\left(2 \frac{A-B}{B} \right)^2 a_{C,\pm 1/2;V,\pm 3/2}^{(2)} + \left(\frac{\hbar\omega}{E_{lh} - E_{hh} - \hbar\omega} \right)^2 b_{C,\pm 1/2;V,\pm 3/2}^{(2)} \right], \quad (19)$$

which in the spherical approximation in the energy spectrum of current carriers takes the form

$$\mathfrak{R}_{C,\pm 1/2;V,\pm 3/2}^{(2, sfer)} = \frac{B^2}{15(\hbar\omega)^2} \left[\frac{16m_{lh}^2}{(m_{hh} - m_{lh})^2} a_{C,\pm 1/2;V,\pm 3/2}^{(2)} + \left(\frac{\hbar\omega(m_{hh} + m_c)m_{lh}}{m_c(m_{hh} - m_{lh})(2\hbar\omega - E_g) - \hbar\omega} \right)^2 b_{C,\pm 1/2;V,\pm 3/2}^{(2)} \right] \quad (20)$$

where for linearly (circularly) - polarized light $a_{C,\pm 1/2;V,\pm 3/2}^{(2)} = 2$ ($a_{C,\pm 1/2;V,\pm 3/2}^{(2)} = 9$), $a_{C,\pm 1/2;V,\pm 3/2}^{(2)} = 3$ ($b_{C,\pm 1/2;V,\pm 3/2}^{(2)} = 13$), $b_{C,\pm 1/2;V,\pm 3/2}^{(2)} = 3$ ($b_{C,\pm 1/2;V,\pm 3/2}^{(2)} = 13$). In this case, the LCD coefficient for these OTs depends on the frequency of the light and the zone parameters;

b) if the initial state is in the sub band of light holes, then we get

$$K_{C,\pm 1/2;V,\pm 1/2}^{(2)} = \frac{32\pi^2}{\hbar} \hbar\omega \frac{1}{I} \Xi_{c, lh} \left(\frac{eA_0}{c\hbar} \right)^2 \frac{P_{cV}^2 k^2}{3} R_{C,\pm 1/2;V,\pm 1/2}^{(2)}, \quad (21)$$

here

$$R_{C,\pm 1/2;V,\pm 1/2}^{(2)} = \frac{1}{4\pi} \left\langle \left| \frac{3Be_+^2}{(E_{hh} - E_{lh} - \hbar\omega)} + 2 \frac{(A+B)e'_+e'_-}{(-\hbar\omega)} \right|^2 \right\rangle + \left\langle \left| 2\sqrt{2}(A+B) \frac{e'_+e'_-}{(-\hbar\omega)} \right|^2 \right\rangle =$$

$$= \frac{B^2}{15(\hbar\omega)^2} \left[4 \left(\frac{A+B}{B} \right)^2 + \left(\frac{3\hbar\omega}{E_{hh} - E_{lh} - \hbar\omega} \right)^2 \right] a_{C,\pm 1/2;V,\pm 1/2}^{(2)}, \quad (22)$$

which in the spherical approximation of the energy spectrum of current carriers takes the form

$$R_{C,\pm 1/2;V,\pm 1/2}^{(2, sfer)} = \frac{\hbar^4 (m_{hh} - m_{lh})^2}{15(4\hbar\omega m_{hh} m_{lh})^2} \left[4 \left(\frac{2m_{hh}}{m_{hh} - m_{lh}} \right)^2 + \left(\frac{3\hbar\omega}{\frac{m_c}{m_{hh}} \frac{m_{hh} - m_{lh}}{m_c + m_{lh}} (2\hbar\omega - E_g) + \hbar\omega} \right)^2 \right] a_{C,\pm 1/2;V,\pm 1/2}^{(2)}, \quad (23)$$

where for linearly (circularly) polarized light $a_{C,\pm 1/2;V,\pm 1/2}^{(2)} = 8$ ($a_{C,\pm 1/2;V,\pm 1/2}^{(2)} = 7$), the LCD coefficient for these OTs does not depend on the light frequency and is equal to 8/7.

Now let the virtual states of the current carriers be in the conduction band. Then:

a) if the initial state is in the heavy hole subband of the valence band, then:

$$K_{C,\pm 1/2;V,\pm 3/2}^{(2)} = \frac{2\pi}{\hbar} 2\hbar\omega \frac{1}{I} \Xi_{c, hh} \left(\frac{eA_0}{c\hbar} \right)^4 \left(\frac{P_{cV} k \hbar^2}{\hbar\omega m_c} \right)^2 \frac{1}{15} a_{C,\pm 1/2;V,\pm 3/2}^{(2)}, \quad (24)$$

where for linearly (circularly) - polarized light $a_{C,\pm 1/2;V,\pm 3/2}^{(2)} = 2$ ($a_{C,\pm 1/2;V,\pm 3/2}^{(2)} = 3$), the LCD coefficient for these OTs is constant and equal to 2/3;

b) if the initial state is in the subband of light holes of the valence band, then

$$K_{C,\pm 1/2;V,\pm 1/2}^{(2)} = \frac{2\pi}{\hbar} 2\hbar\omega \frac{1}{I} \Xi_{c, lh} \left(\frac{eA_0}{c\hbar} \right)^4 \left(\frac{P_{cV} k \hbar^2}{\hbar\omega m_c} \right)^2 J_{C,\pm 1/2;V,\pm 1/2}^{(2)}, \quad (25)$$

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$$J_{C,\pm 1/2;V,\pm 1/2}^{(2)} = \left\langle \frac{|e'_z e'_+|^2}{\sqrt{1 + 4 \frac{\alpha_\omega}{\hbar^2 \omega^2} \left[\frac{1}{\sqrt{3}} \left(\frac{eA_0}{c\hbar} \right)^2 \frac{1}{\hbar \omega} \frac{\hbar^2}{m_c} P_{cV} k \right]^2 |e'_z e'_+|^2}} + \frac{|\sqrt{2} e_z'^2|^2}{\sqrt{1 + 4 \frac{\alpha_\omega}{\hbar^2 \omega^2} \left[\frac{1}{\sqrt{3}} \left(\frac{eA_0}{c\hbar} \right)^2 \frac{1}{\hbar \omega} \frac{\hbar^2}{m_c} P_{cV} k \right]^2 |\sqrt{2} e_z'^2|^2}} \right\rangle, \quad (26)$$

from which, without taking into account the contribution of the effect of coherent saturation in $K_{C,\pm 1/2;V,\pm 1/2}^{(2)}$, we obtain that for light with linear (circular) polarization $\mathfrak{S}_{C,\pm 1/2;V,\pm 1/2}^{(2)} = 8/15$ ($\mathfrak{S}_{C,\pm 1/2;V,\pm 1/2}^{(2)} = 7/15$), and the LCD coefficient is 7/8.

Now let the virtual states of charge carriers be in the extended spin-orbital zone:

a) if the initial state is in the subband of heavy holes of the valence band, then we get that

$$K_{C,\pm 1/2;V,\pm 3/2}^{(2)} = \frac{2\pi}{\hbar} 2\hbar\omega \frac{1}{I} \Xi_{c, hh} \left[\left(\frac{eA_0}{c\hbar} \right)^2 \frac{1}{\sqrt{2}} \frac{B k P_{cV}}{(E_\Delta - E_{hh} - \hbar\omega)} \right]^2 \Phi_{C,\pm 1/2;V,\pm 3/2}^{(2)}, \quad (27)$$

here

$$\Phi_{C,\pm 1/2;V,\pm 3/2}^{(2)} = \left\langle \frac{|e'_z e'_-|^2}{\sqrt{1 + 4 \frac{\alpha_\omega}{\hbar^2 \omega^2} \left[\left(\frac{eA_0}{c\hbar} \right)^2 \frac{1}{\sqrt{2}} \frac{B k P_{cV}}{(E_\Delta - E_{hh} - \hbar\omega)} \right]^2 |e'_z e'_-|^2}} + \frac{|e'_\perp|^2}{\sqrt{1 + 4 \frac{\alpha_\omega}{\hbar^2 \omega^2} \left[\left(\frac{eA_0}{c\hbar} \right)^2 \frac{1}{\sqrt{2}} \frac{B k P_{cV}}{(E_\Delta - E_{hh} - \hbar\omega)} \right]^2 |e'_\perp|^2}} \right\rangle, \quad (28)$$

from which, without taking into account the contribution of the effect of coherent saturation in $K_{C,\pm 1/2;V,\pm 3/2}^{(2)}$, we obtain that for light with linear (circular) polarization, the LCD coefficient is 2/3;

b) if the initial state is in the subband of light holes of the valence band, then the coefficient of two-photon absorption of polarized light is defined as

$$K_{C,\pm 1/2;V,\pm 1/2}^{(2)} = \frac{2\pi}{\hbar} 2\hbar\omega \frac{1}{I} \Xi_{c, lh} \times \left[\left(\frac{eA_0}{c\hbar} \right)^2 \frac{1}{\sqrt{6}} \frac{B k P_{cV}}{(E_\Delta - E_{lh} - \hbar\omega)} \right]^2 \Phi_{C,\pm 1/2;V,\pm 1/2}^{(2)}, \quad (29)$$

$$\Phi_{C,\pm 1/2;V,\pm 1/2}^{(2)} = \left\langle \frac{|3e_\pm'^2 + 4e_z'^2|^2}{\sqrt{1 + 4 \frac{\alpha_\omega}{\hbar^2 \omega^2} \left[\left(\frac{eA_0}{c\hbar} \right)^2 \frac{1}{\sqrt{6}} \frac{B k P_{cV}}{(E_\Delta - E_{lh} - \hbar\omega)} \right]^2 |3e_\pm'^2 + 4e_z'^2|^2}} + \frac{|e'_z e'_+|^2}{\sqrt{1 + 4 \frac{\alpha_\omega}{\hbar^2 \omega^2} \left[\frac{1}{\sqrt{3}} \left(\frac{eA_0}{c\hbar} \right)^2 \frac{1}{\hbar \omega} \frac{\hbar^2}{m_c} P_{cV} k \right]^2 |e'_z e'_+|^2}} \right\rangle, \quad (30)$$

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from which, without taking into account the contribution of the effect of coherent saturation in $K_{C,\pm 1/2;V,\pm 3/2}^{(2)}$, we obtain that the LCD coefficient is $3/2$.

Note that the total coefficient of TPA ($K_{c,V}^{(2)}$) is determined by the sum of the above partial coefficients of TPA.

Thus, the main contribution to the LCD TPA is made by the OTs flowing from the subband of light holes into the conduction band.

Next, we calculate the spectral dependence of the total TPA coefficient in the Kane model and use the following expressions for the energy spectra of current carriers in the parabolic approximation

$$E_c(\vec{k}) = E_g + \frac{\hbar^2 k^2}{2m_0} + \frac{k^2 P_{cV}^2 \left(E_g + \frac{2}{3} \Delta \right)}{E_g (E_g + \Delta)}, \quad E_{hh}(\vec{k}) = \frac{\hbar^2 k^2}{2m_0}, \quad E_{lh}(\vec{k}) = \frac{\hbar^2 k^2}{2m_0} - \frac{2k^2 P_{cV}^2}{3E_g},$$

$$E_{so}(\vec{k}) = -\Delta + \frac{\hbar^2 k^2}{2m_0} - \frac{k^2 P_{cV}^2}{3(\Delta + E_g)}, \quad (31)$$

$E_g(\Delta)$ is the width of the forbidden (spin-orbital) band. Then the spectral dependence of the coefficient of two-photon absorption of linearly polarized light in the region of small values of the wave vector of current carriers has the form

$$K_{c,V}^{(2)}(\omega) = K_{c,V}^{(0)} J_{c,V}^{(2,l)} \left(\frac{2\hbar\omega}{E_g} \right), \quad (32)$$

here $K_{c,V}^{(0)} = \frac{4\pi e^2 P_{cV}}{\hbar c^2 n_\omega^2 E_g^3}$, $E_g \square E_{so}$ for the case $l = 1$, $E_g \square E_{so}$ for the case $l = 2$,

$$J_{c,V}^{(2,1)}(\xi) = \frac{4\xi^{3/2}}{15\sqrt{6}(\xi+1)^3} \left[480 \frac{(\xi+1)^{1/2}}{(3\xi+1)^2} + \frac{(\xi+2)^{3/2}}{(\xi+1)^5} \left(9(\xi+1)^4 + 40(\xi+1)^2 + 96 \right) \right], \quad (33)$$

$$J_{c,V}^{(2,2)}(\xi) = \frac{32\xi^{3/2}}{15(\xi+1)^3} \left\{ 36 \frac{(\xi+1)^{1/2}}{(3\xi+1)^2} + \frac{(\xi+2)^{3/2}}{(\xi+1)^5} \left((\xi+1)^4 + 2(\xi+1)^2 + 6 \right) \right\}, \quad (34)$$

$\xi = (2 \cdot \hbar\omega - E_g) / E_g$. In fig. Figures 3 and 4 show the dependence of $K_{c,V}^{(2)}(\omega)$ for InSb in two cases: $E_g \square E_{so}$ and $E_g \square E_{so}$. In the calculations, it was assumed that the initial states of current carriers are completely occupied, and the final states are completely empty. These results show that under illumination of InSb with linearly polarized light, both in the case of $E_g \square E_{so}$ and $E_g \square E_{so}$, the spectral dependence of $K_{c,V}^{(2)}(\omega)$ increases with increasing frequency, reaches a maximum, and then decreases (Figs. 3 and 4 (3, 4-curves)). This is due to the complexity of the band structure of the semiconductor in the Kane model, which is reflected in the matrix elements and in the energy spectra. This gives rise to complex dependences of the density of states and energies of both the final and initial states of photoexcited current carriers on the frequency of light. If we restrict ourselves to the spherical approximation in the energy spectrum, then $K_{c,V}^{(2)}(\omega)$ will increase with increasing frequency under the condition $E_g \square E_{so}$ (Fig. 3, 1-curve).

The quantitative values of the band parameters were taken from [7].

In conclusion, we note that:

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1. One-photon linear-circular dichroism, caused by interband optical transitions in a narrow-gap crystal, arises when the effect of coherent saturation is taken into account. However, with interband multiphoton absorption of polarized light, linear-circular dichroism is observed regardless of whether the effect of coherent saturation is taken into account or not.

2. Interband two-photon OTs are classified and expressions for the matrix elements are obtained depending on the band parameters, the degree of polarization, and the frequency of light in a narrow-gap crystal.

3. In the Kane model, both with and without taking into account the effect of coherent saturation, the polarization and spectral dependences of the partial coefficients of TPA and their LCDs, differing from each other by the type of OT, are calculated.

4. On the basis of the golden rule of quantum mechanics, it is shown that under illumination of InSb by linearly polarized light, both in the case of $E_g \ll E_{SO}$ and $E_g \approx E_{SO}$, the spectral dependence of $K_{c,v}^{(2)}(\omega)$ increases with increasing frequency, reaches a maximum, and then decreases, and this case is explained by the complexity of the dependences of the density of states and energies both final and initial states of photoexcited carriers of current from the frequency of light, which arise by the peculiarity of the band structure of the semiconductor in the Kane model. If we restrict ourselves to the spherical approximation in the energy spectrum, then $K_{c,v}^{(2)}(\omega)$ will increase with increasing frequency under the condition $E_g \ll E_{SO}$ (Fig. 4, 1-curve).

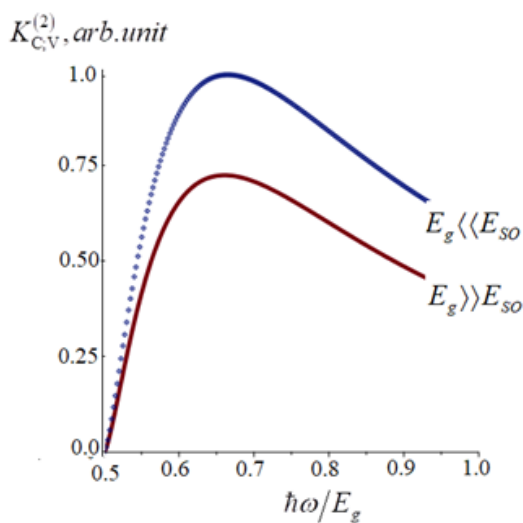


Fig. 3. Spectral dependence of the coefficient of interband two-photon absorption of linearly polarized light in an InSb crystal, corresponding to cases $E_g \ll E_{SO}$ and $E_g \approx E_{SO}$.

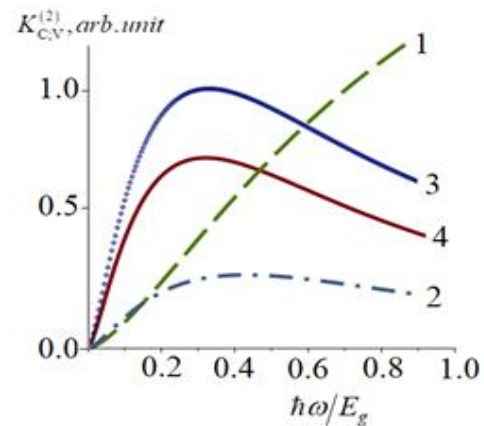


Fig. 4. Spectral dependence of the coefficient of interband two-photon absorption of linearly polarized light in an InSb crystal, corresponding to cases $E_g \ll E_{SO}$ (curves 1 and 3), $E_g \approx E_{SO}$ (curves 2 and 4). Curves 1 and 2 correspond to the parabolic, and curves 3 and 4 to the nonparabolic approximation in the energy spectrum of current carriers.

Application

According to [12, 13], the effective carrier Hamiltonian for the three-band Kane model is expressed as follows:

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$$\hat{H}^{eff}(k_z) = \begin{pmatrix} E_c & -i\sqrt{2/3}P_{cv}k_z & -iP_{cv}k_z/\sqrt{3} \\ i\sqrt{2/3}P_{cv}k_z & E_v & 0 \\ iP_{cv}k_z/\sqrt{3} & 0 & E_v - \Delta \end{pmatrix}, \quad (\text{Ap. 1})$$

whose eigenvalue is determined by the equation:

$$(E_c - E)(E_v - E)(E_v - \Delta - E) - P_{cv}^2 k^2 (E_v - E - 2\Delta/3) = 0, \quad (\text{Ap. 2})$$

where $k_z = k = |\vec{k}|$. This equation has three solutions: $E_{el}(k), E_{lh}(k), E_{so}(k)$. From (Ap. 2) we

have $k^2(E) = \frac{1}{P_{cv}^2} \frac{(E_c - E)(E_v - E)(E_v - E + \Delta)}{(E - E_v + 2\Delta/3)}$ and with the help of the last dependence a graph

$k^2(E)$ is constructed in Fig. 5. It can be seen from Fig. 5 that all energy bands are nonparabolic, and the quadratic dependence of the energy on $k = |\vec{k}|$ is observed in the region of small values of the wave vector.

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