



# NONLINEAR ABSORPTION OF POLARIZED RADIATION IN CRYSTALS WITH A COMPLEX BAND

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## ABSTRACT

The matrix elements of one- and two-photon optical transitions from the subbands of the valence band and the spin-orbital splitting band to the conduction band are calculated, expressions are obtained for them depending on the polarization vector and light frequency, as well as on the band parameters of the crystal.

Expressions are obtained for the spectral-polarization and spectral-temperature dependences of the coefficients of interband two-photon absorption of light and linear-circular dichroism in semiconductors of tetrahedral symmetry in the three-band Kane model.

It is shown that in the spectral-polarization dependence of the total coefficient of two-photon absorption of linearly and circularly polarized light in InSb: firstly, all spectral-polarization dependences have an oscillatory character; secondly, as the light frequency increases, the oscillation amplitude increases; thirdly, for circularly polarized light, the oscillation becomes aperiodic.

**KEYWORDS:** spectral, temperature and polarization dependence, optical transitions, linear-circular dichroism, two-photon absorption of linearly and circularly polarized light.

## INTRODUCTION

At present, nonlinear optical phenomena occurring in crystals are widely used in practice [1–3]. In this context, the study of nonlinear absorption of polarized light is relevant both from the physical point of view and from the point of view of application. We note that in the case of single-photon absorption of light, optical transitions do not occur through virtual states. Therefore, in single-photon optical transitions in crystals of cubic and tetrahedral symmetry, linear-circular dichroism is not observed.

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

In [4–7], linear circular dichroism (LCD)<sup>1</sup> of two- and three-photon absorption of light in crystals of cubic symmetry was studied both theoretically and experimentally.

Multiphoton absorption of light in a semiconductor with a complex valence band, which is due to direct optical transitions between heavy and light hole subbands and depends on the degree of light polarization, was studied in [8–17]. Nonlinear interband single-photon absorption of polarized light in Weyl semimetals was studied in [18]. In these works, it is assumed that the nonlinearity in the dependence of the single-photon absorption coefficient on the light intensity arises due to resonant absorption saturation [19]. This saturation in both interband [18] and intraband [9, 10, 16, 17] light absorption is due to the photoinduced change in the distribution functions of current carriers in the region of momentum space near the surface, which

<sup>1</sup> Two-photon linear-circular dichroism due to interband optical transitions of electrons was predicted by EL Ivchenko in [4].

is determined by the law of conservation of energy and the relaxation time, the reciprocal of which is equal to reciprocal relaxation times in energy and momentum.

In [8, 11, 14], multiphoton linear-circular dichroism (LCD) in p-Ge was studied in the regime of developed nonlinearity, when n-photon processes with  $n = (1, 5)$  make a comparable contribution to absorption. In [16, 17], four-photon processes in semiconductors due to optical transitions between subbands of the valence band were studied with allowance for the effect of coherent saturation.

In the present work, in contrast to [7], we carry out LCD calculations of interband two-photon light absorption (TPLA), as well as on the spectral dependence of the TPLA coefficient in semiconductors of tetrahedral symmetry in the three-band Kane model, where we take into account the contributions to the multi-quantum process of intermediate states in the subbands of light and heavy holes and in the zone of spin-orbit splitting, as well as in the conduction band, taking into account the effect of coherent saturation.

### Two-Photon Interband Absorption of Polarized Light in Narrow-Gap Semiconductors

Following [14, 16] in further calculations of the spectral and temperature dependence of the two-photon light absorption coefficient  $K^{(2)}$ , where the light wave vector is neglected, i.e. we assume 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

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

or

$$K_{hh, hh}^{(2)} = \frac{2\pi}{\hbar} 2\hbar\omega \frac{1}{I} \rho(2\hbar\omega) F(\beta, 2, \omega) \times \left[ \left\langle \frac{|M_{V,\pm 1/2;V,\pm 3/2}^{(12)}(\vec{k})|^2}{\sqrt{1 + 4 \frac{\alpha_\omega}{\hbar^2 \omega^2} |M_{V,\pm 1/2;V,\pm 3/2}^{(2)}(\vec{k})|^2}} \right\rangle + \left\langle \frac{|M_{V,\pm 1/2;V,\mp 3/2}^{(2)}(\vec{k})|^2}{\sqrt{1 + 4 \frac{\alpha_\omega}{\hbar^2 \omega^2} |M_{V,\pm 1/2;V,\mp 3/2}^{(2)}(\vec{k})|^2}} \right\rangle \right], \quad (2)$$

where  $\rho(2\hbar\omega)$  is the density of states of current carriers involved in two-photon optical transitions, where the law of conservation of energy is taken into account,  $F(\beta, 2, \omega)$  is the distribution function of current carriers in the initial state,  $\beta^{-1} = k_B T$ ,  $k_B$  is the Boltzmann constant,  $T$  is the temperature of sample:  $F(\beta, 2, \omega) = \exp[\beta(\mu - E_{L=hh}(k_{lh, hh}^{(2\omega)}))]$ ,

$$E_{hh}(k_{lh, hh}^{(2\omega)}) = \frac{m_{lh}}{m_{hh} - m_{lh}} 2\hbar\omega, \quad \rho(\hbar\omega) = \frac{m_{lh}}{m_{hh} - m_{lh}} k_{lh, hh}^{(2\omega)} / (\pi^2 \hbar^2) \text{ and it is also taken into account that } 2\hbar\omega \gg k_B T.$$

Now it is required to perform the angular averaging of the modules of squares of the considered matrix elements

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

here is the contribution of the coherent saturation effect to the coefficient of two-photon absorption of light by precisely these radicals of the last relations. That. calculations without taking into account the contribution of the coherent saturation effect to the coefficient of two-photon light absorption is described by the expression:  $\left\langle |M_{V,+1/2;V,+3/2}^{(2)}(\vec{k})|^2 \right\rangle + \left\langle |M_{V,\pm 1/2;V,\mp 3/2}^{(2)}(\vec{k})|^2 \right\rangle$ , so in

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

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

If we assume that optical transitions occur from the heavy hole branch to the light hole branch, where the intermediate states of the current carriers are in the subbands of the valence band, then the matrix element of the two-photon optical transition is determined by the relation

$$\begin{aligned}
 | +3/2 \rangle \rightarrow | m \rangle \rightarrow | +1/2 \rangle &= \frac{M_{+1/2;+3/2}^{(1)} M_{+3/2;+3/2}^{(1)}}{E_{hh} - E_{hh} - \hbar\omega} + \frac{M_{+1/2;+1/2}^{(1)} M_{+1/2;+3/2}^{(1)}}{E_{lh} - E_{hh} - \hbar\omega} + M_{+1/2;+3/2}^{(2)} = \\
 &= \frac{M_{+1/2;+3/2}^{(1)} M_{+3/2;+3/2}^{(1)}}{(-\hbar\omega)} + \frac{M_{+1/2;+1/2}^{(1)} M_{+1/2;+3/2}^{(1)}}{(\hbar\omega)} + M_{+1/2;+3/2}^{(2)} = -5\sqrt{3} \left( \frac{eA_0}{c\hbar} \right)^2 B e'_+ e'_-, \quad (4)
 \end{aligned}$$

where  $M^{(1)}(m \rightarrow m') = M_{m'm}^{(1)}(\vec{k}) [M^{(2)}(m \Rightarrow m') = M_{m'm}^{(2)}(\vec{k})]$  - is the matrix element of one (simultaneously absorbing two) photonic optical transition (OT), from which we obtain the expression for the square of the modulus of the optical transition of the  $| \pm 3/2 \rangle \rightarrow | m \rangle \rightarrow | \pm 1/2 \rangle$  type, we have  $75 \left( \frac{eA_0}{c\hbar} \right)^4 B^2 |e'_+ e'_-|^2$ , and for the optical transition of the  $| \pm 3/2 \rangle \rightarrow | m \rangle \rightarrow | \mp 1/2 \rangle$  type, we have  $\frac{3}{4} \left( \frac{eA_0}{c\hbar} \right)^4 B^2 (36e_z'^2 |e'_+|^2 + |e_-'^2|^2)$ .

If intraband optical transitions occur between the subbands of light and heavy holes, then the intermediate states are both in the conduction band and in the spin-orbit splitting band. Then the matrix elements of these optical transitions are described by the expressions:

$$\begin{aligned}
 \text{a) } | +3/2 \rangle \longrightarrow | c, m \rangle \longrightarrow | +1/2 \rangle + | +3/2 \rangle \longrightarrow | \Delta, m \rangle \longrightarrow | +1/2 \rangle &= \\
 &= \frac{M_{V,+1/2;c,+1/2}^{(1)} M_{c,+1/2;V,+3/2}^{(1)}}{E_c - E_{hh} - \hbar\omega} + \frac{M_{V,+1/2;c,-1/2}^{(1)} M_{c,-1/2;V,+3/2}^{(1)}}{E_c - E_{hh} - \hbar\omega} + \frac{M_{V,+1/2;SO,+1/2}^{(1)} M_{SO,+1/2;V,+3/2}^{(1)}}{E_{SO} - E_{hh} - \hbar\omega} + \\
 &\frac{M_{V,+1/2;SO,-1/2}^{(1)} M_{SO,-1/2;V,+3/2}^{(1)}}{E_{SO} - E_{hh} - \hbar\omega} = \left( \frac{eA_0}{c\hbar} \right)^4 \sqrt{\frac{1}{3}} \left( \frac{p_{cv}^2}{E_c - E_{hh} - \hbar\omega} - \frac{3B^2 k^2}{E_{SO} - E_{hh} - \hbar\omega} \right) e'_z e'_-; \quad (5)
 \end{aligned}$$

$$\begin{aligned}
 \text{b) } | -3/2 \rangle \rightarrow | c, m \rangle \rightarrow | +1/2 \rangle + | -3/2 \rangle \rightarrow | SO, m \rangle \rightarrow | +1/2 \rangle &= \\
 &= \frac{M_{V,+1/2;c,+1/2}^{(1)} M_{c,+1/2;V,-3/2}^{(1)}}{E_c - E_{hh} - \hbar\omega} + \frac{M_{V,+1/2;c,-1/2}^{(1)} M_{c,-1/2;V,-3/2}^{(1)}}{E_c - E_{hh} - \hbar\omega} + \frac{M_{V,+1/2;SO,+1/2}^{(1)} M_{\Delta SO,+1/2;V,-3/2}^{(1)}}{E_{\Delta} - E_{hh} - \hbar\omega} + \\
 &+ \frac{M_{V,+1/2;SO,-1/2}^{(1)} M_{SO,-1/2;V,-3/2}^{(1)}}{E_{\Delta} - E_{hh} - \hbar\omega} = - \left( \frac{eA_0}{c\hbar} \right)^4 \frac{1}{2\sqrt{3}} \left( \frac{p_{cv}^2}{E_c - E_{hh} - \hbar\omega} - 9 \frac{B^2 k^2}{E_{SO} - E_{hh} - \hbar\omega} \right) e_+'^2. \quad (6)
 \end{aligned}$$

The remaining matrix elements are defined in a similar way. That. the matrix elements of these optical transitions can be represented as the following matrix

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

Since both the coefficient of two-photon linear-circular dichroism and the coefficient of two-photon light absorption are determined by the square of the modules of the composite matrix elements, the form of which for the above optical transitions is

$$\left| \tilde{M}^{(2)} \right| = \begin{bmatrix} \Re_1 e_z'^2 |e'_-|^2 & \Re_2 |e'_-|^4 \\ -\Re_2 |e'_+|^4 & \Re_1 e_z'^2 |e'_+|^2 \end{bmatrix} - \begin{bmatrix} \Re_1 e_z'^2 |e'_-|^2 & \Re_2 |e'_-|^4 \\ -\Re_2 |e'_+|^4 & \Re_1 e_z'^2 |e'_+|^2 \end{bmatrix}, \quad (7)$$

where

$$\Re_1 = \left( \frac{eA_0}{c\hbar} \right)^4 \frac{1}{3} \left( \frac{p_{cv}^2}{E_c - E_{hh} - \hbar\omega} - \frac{3B^2 k^2}{E_{SO} - E_{hh} - \hbar\omega} \right)^2 e_z'^2 |e'_-|^2, \quad (8)$$

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

Matrix elements of two-photon transitions occurring from the spin-split band to the conduction band, where the virtual states of current carriers are located in subbands of the valence band, in the conduction band, as well as in the zone of spin-orbit splitting of the semiconductor, which are shown in Fig. 3 and are defined in the same way as in the above cases.

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

### Spectral-Polarization Dependences of the Coefficient of Two-Photon Light Absorption and Linear-Circular Dichroism

Let us now analyze the spectral-polarization dependence of the coefficient of two-photon absorption of light, which is determined using the functions  $\mathfrak{R}_1$  and  $\mathfrak{R}_2$ . To do this, we rewrite expressions (2) taking into account (7). Then it is easy to verify that for a *GaAs* crystal the spectral-polarization dependence of the coefficient of two-photon absorption of light both without taking into account (see Fig. 1) and taking into account the contribution of the coherent saturation effect (see Fig. 2), as well as the two-photon linear circular dichroism (see Fig. 2.3), caused between the subbands of light and heavy holes, where the intermediate states are in the conduction bands and spin orbital splitting (see Fig. 1) and has an oscillatory character with respect to the angle between the polarization vectors and current carriers for linearly polarized light and relative to the angle between the wave vectors of the photon and current carriers for circularly polarized light. It can be seen from Fig. 1 that the amplitude, period, and phase of the oscillations are different in this case. Note here that the coefficient of two-photon linear-circular dichroism in *GaAs* is less than unity.

When the contribution of the coherent saturation effect to the coefficient of two-photon linear-circular dichroism is taken into account, expressions (8) and (9) take the form

$$\tilde{\mathfrak{R}}_1 = \left\langle \frac{\xi_1 e_z'^2 |e'_-|^2}{\sqrt{1 + \zeta_1 e_z'^2 |e'_-|^2}} \right\rangle, \quad \tilde{\mathfrak{R}}_2 = \left\langle \frac{\xi_2 |e'_-|^2}{\sqrt{1 + \zeta_2 |e'_-|^2}} \right\rangle, \quad (10)$$

where the Rabi parameters are

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

$$\xi_1 = \left( \frac{eA_0}{c\hbar} \right)^4 \frac{1}{3} \left( \frac{p_{cv}^2}{E_c - E_{hh} - \hbar\omega} - \frac{3B^2 k^2}{E_\Delta - E_{hh} - \hbar\omega} \right)^2, \quad \xi_2 = \frac{1}{12} \left( \frac{p_{cv}^2}{E_c - E_{hh} - \hbar\omega} - 9 \frac{B^2 k^2}{E_\Delta - E_{hh} - \hbar\omega} \right)^2 |e'_-|^2.$$

Comparing the data in fig. 1 and 2, we find that taking into account the contribution of the coherent saturation effect leads to a decrease in the oscillation amplitude with increasing light frequency, 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 vectors of polarization and current carriers, and for circularly polarized light - the angle between the vectors of a photon and current carriers. Quantitative calculations were carried out at  $\zeta_{1,2} = 0, 2$ .

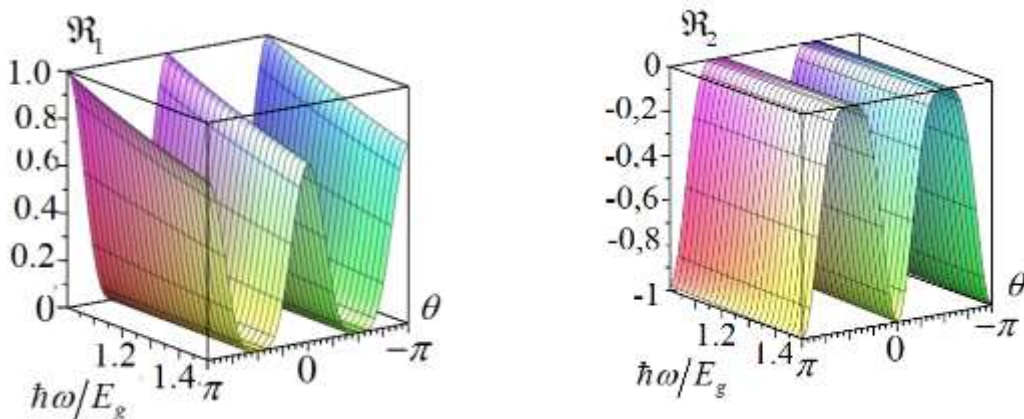


Fig.1. Spectral polarization dependence of the quantities  $\tilde{\mathfrak{R}}_1$  and  $\tilde{\mathfrak{R}}_2$ , which are used to determine the spectral polarization dependence of the two-photon light absorption coefficient in *GaAs* under illumination with linearly polarized light, without taking into account the contribution of the coherent saturation effect.

Calculations show that for GaAs, as the light frequency increases, the contribution to the total coefficient of two-photon linear-circular dichroism of the term proportional to  $\mathfrak{R}_1$  decreases with respect to the contribution of the term proportional to  $\mathfrak{R}_2$  regardless of the angle between the vectors  $\vec{e}$ ,  $\vec{k}$ ,  $\vec{q}$ : if the light frequency increases by a factor of 1,4, then this the contribution decreases by a factor of 2,5 where,  $\vec{e}$  is the light polarization vector,  $\vec{k}$  ( $\vec{q}$ ) is the wave vector of the current carriers (photon). This is due to the fact that the first contribution does not depend on the frequency of the light, while the second contribution depends on the frequency as  $\propto (\hbar\omega)^{1/2}$ . Therefore, the second contribution increases with frequency of the light.

To compare the theoretical results, below we performed calculations on the spectral-polarization dependence of the coefficients  $\mathfrak{R}_1$  and  $\mathfrak{R}_2$ , which are used to determine the coefficient of two-photon absorption of linearly and circularly polarized light in InSb, which is caused between the subbands of light and heavy holes for two values of the Rabi parameters  $\zeta_1 = 0, 2; 0, 5$  and  $\zeta_2 = 0, 2; 0, 5$ , where the

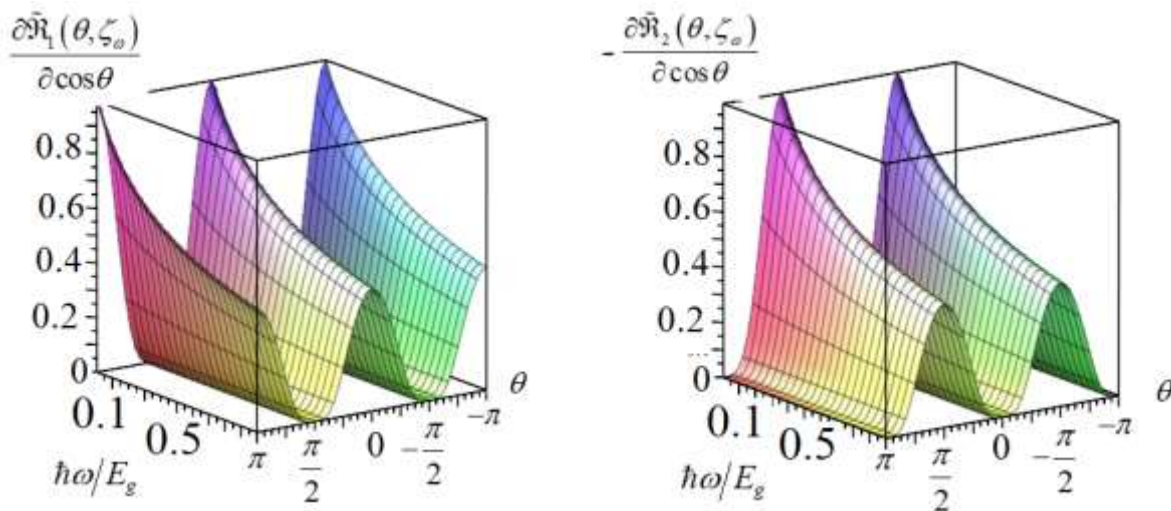
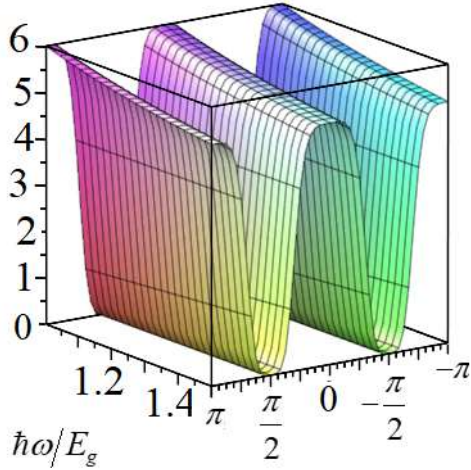


Fig.2. Spectral-polarization dependence of the coefficients  $\mathfrak{R}_1 = \frac{\partial \tilde{\mathfrak{R}}_1}{\partial \cos \theta}$  and  $\mathfrak{R}_2 = \frac{\partial \tilde{\mathfrak{R}}_2}{\partial \cos \theta}$  for two-photon absorption of linearly polarized light in GaAs with allowance for the contribution of the coherent saturation effect, where  $\zeta_1 = 0, 2$ .

Intermediate states are in the conduction and spin-orbital splitting bands, taking into account the contribution of the coherent saturation effect (see Fig. 3). Fig. 3 shows that in the spectral-polarization dependence of the total

$$\eta^{(2)} = W^{(2, lin)} / W^{(2, circ)}$$



of two-photon linear-circular dichroism in *InSb*, caused between the subbands of light and heavy holes, taking into account the contribution of the coherent effect.

coefficient  $\nu \mathfrak{R}_1 + \mathfrak{R}_2$  for both linearly (a, b) and circularly polarized light (c, d) by polarized light, which is used to determine the coefficient of two-photon absorption of linearly and circularly polarized light in *InSb*, due to between the subbands of light and heavy holes, where the intermediate states are in the conduction and spin-orbital splitting bands, taking into account the contribution of the coherent saturation effect for two values of the Rabi parameter: first, all the spectral-polarization dependences have an oscillatory character; secondly, as the light frequency increases, the oscillation amplitude increases; thirdly, for circularly polarized light, the oscillation becomes aperiodic; fourthly, as the values of the Rabi parameter increase, the amplitude values of the oscillation dependences decrease. In calculations, the maximum value of the spectral-polarization dependence of the two-photon coefficient of linear-circular dichroism at  $\zeta_{1,2} = 0, 2$  was taken to be unity.

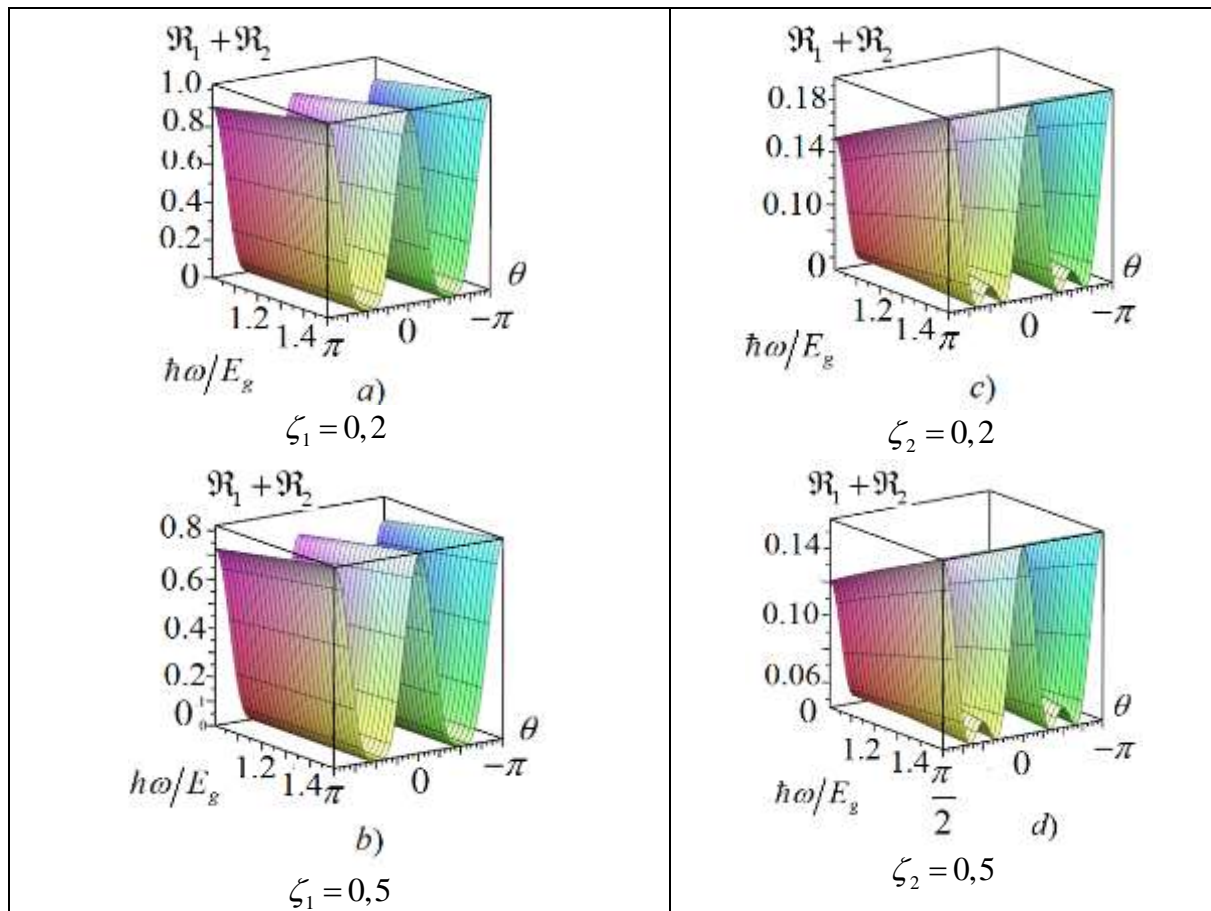


Fig.3. Spectral-polarization dependence of the total coefficient  $\mathfrak{R}_1 + \mathfrak{R}_2$  for both linearly (a, b) and circularly polarized light (c, d), which is used to determine the coefficient absorption of two-photon polarized light in *InSb*, taking into account the contribution of the coherent effect saturation for two values of the Rabi parameter.



On fig. 4 shows the dependence of the two-photon linear-circular dichroism in *InSb*, which is caused between the subbands of light and heavy holes, where the intermediate states are in the conduction bands and the spin-orbital splitting, taking into account the contribution of the coherent saturation effect, is equal to 6 at  $\zeta_{\omega} = 0,2$ . We note here that, in contrast to the wide-gap *GaAs* crystal in the narrow-gap semiconductor *InSb*, the coefficient of two-photon linear-circular dichroism is greater than unity. From fig. 2.6 c shows that as the Rabi parameter increases from  $\zeta_{\omega} = 0,2$  to  $\zeta_{\omega} = 0,5$ , the maximum value of the spectral-polarization dependence of the coefficient of two-photon linear-circular dichroism in *InSb*, both for linear and circular polarization, increases by 23%. Therefore, the spectral polarization dependence of two-photon linear-circular dichroism in *InSb*, taking into account the contribution of the effect of coherent saturation, is almost independent of the value of the Rabi parameter and the amplitude value of the two-photon linear-circular dichroism coefficient is no more than 6.

The quantitative values of the band parameters were taken from [24].

## CONCLUSION

In conclusion, we note that in the three-zone Kane approximation:

1. The matrix elements of interband two-photon optical transitions in a semiconductor are classified depending on the light polarization vector.
2. Both with and without allowance for the effect of coherent saturation, the polarization and spectral dependences of the coefficients of two-photon linear-circular dichroism and light absorption, which differ from each other by the type of intermediate states, are calculated.
3. A theory has been developed for linear-circular dichroism coupled by interband two-photon optical transitions in narrow-gap semiconductors in the Kane approximation.

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