The European Journal of Technical
and Natural Sciences 2025, No 6
ISSN 2414-2352

PREMIER Publishing
ppublishing.org

Section 7. Physics

DOI:10.29013/EJTNS-25-6-51-54

INTERBAND ONE-PHOTON LINEAR-CIRCULAR DICHROISM IN NARROW-GaP CRYSTALS. PART 1

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Cite: *Rasulov V.R., Mamatova M.A., Muminov I.A., Urinova K.K., Toshtemirova M.N. (2025). Interband One-Photon Linear-Circular Dichroism in Narrow-Gap Crystals. European Journal of Technical and Natural Sciences 2025, No 6.* https://doi.org/10.29013/EJTNS-25-6-51-54

Abstract

In this paper, from a microscopic point of view, the linear-circular dichroism of interband one-photon absorption of light in the Kane approximation in narrow-gap crystals is investigated.

The linear-circular dichroism of one-photon absorption of polarized light is calculated taking into account the effect of coherent saturation in photoexcited charge carriers.

The matrix elements of one-photon interband optical transitions and the corresponding linear-circular dichroism and the spectral dependence of the light absorption coefficient are calculated.

Keywords: *one-photon absorption; linear–circular dichroism; coherent saturation; Kane approximation; cubic semiconductors; interband transitions; spin–orbit split-off band; polarization degree; intensity-dependent absorption; spectral dependence*

Introduction

At present, nonlinear optical phenomena occurring in crystals are widely used in practice (Rostami A., 2006; Pattanaik H. S., Reichert M., Hagan D. J., and Van Stryland E. W.; Yu J. H., Kwon S.-H., Petrášek Z., Park O. K., Jun S. W., Shin K., Choi M., Park Y. I., Park K., Na H. B., Lee N., Lee D. W., Kim J. H., Schwille P., and Hyeon T., 2013).

In this context, the study of nonlinear absorption of polarized light is relevant both from a physical standpoint and from the perspective of practical applications.

It should be noted that in the case of one-photon light absorption, optical transitions do not occur through virtual states and are generally not observed. Therefore, in one-photon optical transitions within crystals possessing cubic and tetrahedral symmetry, linear-circular dichroism is not observed.

The one- and multiphoton absorption of polarized light in crystals, caused by optical transitions between the subbands of the valence band, have been investigated in (Ivchenko E. L., 1972; Rasulov R. Ya., 1993; Ganichev S. D., Ivchenko E. L., Rasulov R. Ya., Yaroshetsky I. D., and Averbukh B. Ya., 1993; Parshin D. A. and Shabaev A. R., 1987; Rasulov R. Ya., 1988; Rasulov R. Ya., Khoshimov G. Kh., and Kholitdinov Kh., 1996; Rasulov R. Ya., 1993; Leppenen N. V., Ivchenko E. L., and Golub L. E.). However, the contribution of the coherent saturation effect (Ganichev S. D., Ivchenko E. L., Rasulov R. Ya., Yaroshetsky I. D., and Averbukh B. Ya., 1993; Parshin D. A. and Shabaev A. R., 1987) to interband one-photon light absorption has not been taken into account in those studies. This effect arises from the finite lifetime of photoexcited charge carriers in the final state, which is the main focus of the present work.

Interband One-Photon Light Absorption Coefficient: Quantitative Theory

From equation (10) in the first part of this work, it is evident that the interband one-photon light absorption coefficient $K^{(1)}(\omega,T)$ consists of partial components that differ from one another according to the type of optical transitions. In particular, for an optical transition of the type $|V,\pm 3/2\rangle \to |C,\pm 1/2\rangle$, it is expressed as follows:

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

where $\zeta_\omega=4\dfrac{\alpha_\omega}{\hbar^2\omega^2}\left(\dfrac{eA_0}{c\hbar}\right)^2 P_{CV}^2$,

$$F(\beta,1,\omega)=$$
$$=\left[1-\exp(\beta\hbar\omega)\right]\exp\left[\beta\left(\mu-E_{hh}(k_{c,L=hh}^{(\omega)})\right)\right],$$

$$k_{c,L}^2=\frac{2\mu_{c,L}^{(\omega)}}{\hbar^2}\left(\hbar\omega-E_g\right),\quad \frac{1}{\mu_{c,L}^{(+)}}=\left(\frac{1}{m_c}+\frac{1}{m_L}\right),$$

$$\beta^{-1}=k_B T,\ \Im(\omega)=\left\langle\frac{|e'_\pm|^2}{\sqrt{1+\zeta_\omega|e'_\pm|^2}}\right\rangle.$$

From equation (1), it follows that the linear-circular dichroism of one-photon light absorption is determined by the quantity $\Im(\omega)$, which depends on the frequency and degree of light polarization, as well as on the band parameters of the sample. This phenomenon arises due to the complexity of the crystal's band structure.

It should be noted that if the effect of coherent saturation ($\zeta_\omega=0$) is not taken into account, then $K^{(1)}(\omega,T)$ does not depend on the aforementioned quantities — in particular, on the degree of light polarization — and thus represents a constant value $\Im(\zeta_\omega=0)=\dfrac{4}{3}$. In this case, one-photon linear-circular dichroism is not observed. However, if the coherent saturation effect is taken into account, then $\zeta_\omega\neq 0$, which means that one-photon linear-circular dichroism arises. This is due to the fact that:

for linearly polarized light

$$\Im_{lin}=\int_{-1}^{+1}d\mu\frac{1-\mu^2}{\sqrt{1+\zeta_\omega\left(1-\mu^2\right)}}; \quad (12)$$

for circularly polarized light

$$\Im_{circ}=\int_{-1}^{+1}d\mu'\frac{\frac{1}{2}\left(1+\mu'^2\right)\mp P_{circ}\mu'}{\sqrt{1+\zeta_\omega\left[\frac{1}{2}\left(1+\mu'^2\right)\mp P_{circ}\mu'\right]}}, \quad (13)$$

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

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

$$\Im_{lin}=\zeta_\omega^{-5/2}\left\{\zeta_\omega^{3/2}+\zeta_\omega^2\cdot\arcsin\left(\frac{\zeta_\omega}{1+\zeta_\omega}\right)^{1/2}-\zeta_\omega\cdot\arcsin\left(\frac{\zeta_\omega}{1+\zeta_\omega}\right)^{1/2}\right\}, \quad (14)$$

for circularly polarized light

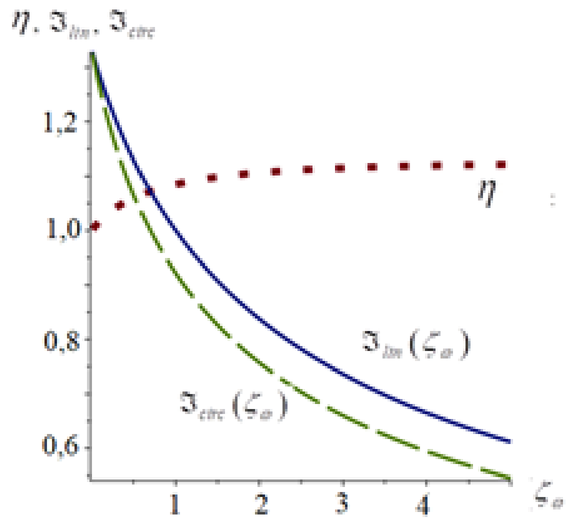
$$\Im_{circ}=\frac{2\left(\zeta_{\omega}^{3/2}\sqrt{\zeta_{\omega}+1}-\zeta_{\omega}\arcsin\sqrt{\zeta_{\omega}}\right)}{\zeta_{\omega}^{5/2}} \quad (15)$$

Figure 3 presents the graphs of the functions $\Im_{lin}(\zeta_{\omega})$ and $\Im_{circ}(\zeta_{\omega})$ as a function of the quantity $\zeta_{\omega}\propto\left(\frac{eA_0}{c\hbar}\right)^2\propto I$, which are used to determine the spectral dependence of the one-photon light absorption coefficient. From Figure 1, it can be seen that as the light intensity increases, the coefficient of interband one-photon linear-circular dichroism $\eta=\Im_{lin}(\zeta_{\omega})/\Im_{circ}(\zeta_{\omega})$ also increases and tends toward saturation; that is, at very high intensity values, ($\zeta_{\omega}>>1$) becomes independent of intensity and $\eta\approx 1.1$. The quantitative calculations were performed using the data from (Vurgaftman I., Meyer J. R. M., and Ram-Mohan J. R., 2001).

Conclusion

Figure 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.

Figure 2.



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

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submitted 31.10.2025;
accepted for publication 14.11.2025;
published 30.12.2025

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