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Electroreflection of light and the Franz–Keldysh effect in monoclinic zinc diphosphide crystals

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The results of the study of electroreflectance and the Franz–Keldysh effect in zinc diphosphide monoclinic crystals are presented. The obtained spectra are compared with theoretical positions in the approximations of strong and weak electric fields. The dependences of electro-optical constants on electric field strength and temperature have been obtained. The energies of optical transitions in the energy range from the fundamental absorption edge to 4 eV have been determined.

Keywords: electroreflectance (ER) technique, Franz–Keldysh Effect, semiconducting materials, β -ZnP₂ (Monoclinic Zinc Diphosphide).

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1. Introduction

It is important to investigate the surface and interface properties of semiconductors and semiconductor-based structures is important due to the role of semiconductors in modern electronic and optoelectronic devices [1–3]. Modulation techniques, in particular, electroreflection and electroabsorption, are used to explore electronic phenomena in thin films and near-surface material layers and, thus, to study material properties [4–7]. Electroreflection and electroabsorption methods used to examine semiconductors, in which a wide range of interband transitions are implemented, might be suitable for development of the modulation spectroscopy theory [8]. Such semiconductors are represented by zinc and cadmium diphosphides and diarsenides. Unique optical properties of monoclinic crystals in this group that take the form of allowed and forbidden optical transitions, strongly pronounced exciton states, and considerable values of bond energy of excitons and biexcitons, and of spin-orbital and crystalline splitting [9,10] are of interest for study of these properties by modulation methods. Moreover, semiconductor crystals of group A²B⁵ have good prospects of use in instrumentation technology due to a range of unique properties suitable for radiation converters, optical elements and sensors [11]. Deep knowledge of optical and optoelectrical properties, in particular, energy band structure, is important for practical application.

Modulation spectroscopy methods, including electroabsorption and electroreflection, are successfully used to investigate optical constants and physical and chemical properties of semiconductors and semiconductor-based structures [4,5,12–17]. In particular, λ -modulation and electroabsorption were used to study optical properties [16],

electroreflection in α -ZnP₂ was used to study phase transitions in these crystals [17]. Electromodulation methods were not used to study monoclinic β -ZnP₂ crystals.

This work describes the investigation of optical properties of β -ZnP₂ crystals using the electroreflection method.

2. Experimental procedure

Electronic conductivity type zinc diphosphide crystals were used for the investigation. Electroreflection spectra were measured on active ITO — crystal structures, to which modulating AC and DC voltages were applied simultaneously. Optical radiation was recorded by synchronous detection method at the electric field modulation frequency of 275 Hz. Spectral resolution of the instrument was $\sim 1.45 \cdot 10^{-4}$ eV in the fundamental absorption edge region of the β -ZnP₂ and $< 3.25 \cdot 10^{-4}$ eV in the remaining spectrum region. A tungsten-halogen lamp and DDG-30 gas-discharge lamp were used as visible and near IR radiation and in the UV-region sources, respectively. S1336-VK precision silicon photodiode for IR and visible spectra and FEU-39A multiplier phototube for UV spectrum were used as radiation receivers. ITO– β -ZnP₂ active surface-barrier structure was made by magnetron sputtering of a transparent conducting layer onto the (100) natural faces of the crystal. Crystal thickness was 0.8 mm. A ~ 0.6 eV barrier layer is formed at the heterostructure boundary, the maximum electric field strength $F_{\max}$ in which was $\sim 10^7$ V/m and could be changed by an external voltage from puncture values to zero. Photovoltaic and electric properties of the ITO– β -ZnP₂ surface-barrier structures are described in [18–20]. Electroreflection spectra of ITO– β -ZnP₂ were measured at (283–365) K.

3. Findings and discussion

Figure 1 shows polarized light electroreflection spectra in the energy range of (1.4–3) eV. Oscillations typical of the Franz–Keldysh effect appear in the fundamental absorption edge region in the electroreflection spectra in the $\mathbf{E} \parallel \mathbf{c}$ polarization and in the direct allowed transition region in the $\mathbf{E} \perp \mathbf{c}$ polarization (Figures 2–4). Low-intensity oscillations associated with other interband transitions appear deep in the absorption band.

The electroreflection spectra are characterized by high dependence of signal strength on the modulation voltage U_m and DC voltage U_{cm} applied to the structure. Modulation voltage on the structure was set to a minimum value equal to 0.2–0.7 V. As U_{cm} increases with constant U_m , extrema of the electroreflection spectra shift into a higher energy region (except the extrema denoted as a_1 and e_3) and the energy difference between extremum positions increases.

As the barrier shifts in the forward direction, a fine structure occurs in the electroreflection spectra (Figure 2) and can be represented as oscillations with a lower amplitude and shorter period overlaid on the Franz–Keldysh oscillations on the direct allowed transition a_1 (referred to as E_0 in electromodulation studies). A better resolution of this structure is observed as the resulting electric field in the barrier decreases. As the electric field strength grows (Figure 3), extrema denoted as $n_1^* - n_4^*$ are not exhibited due to broadening of the oscillation bands associated with the transition a_1 . Fine structure in the spectra might display the Franz–Keldysh effect on the absorption lines of an inverse hydrogen-like series (IHS) in a bielectron-impurity complex (BIC) detected in the β -ZnP₂ crystals with n -type conductivity [21]. $n_1^* - n_4^*$ in the electroreflection spectra in this spectrum region keep their position as the voltage applied to the structure varies. IHS BIC absorption lines

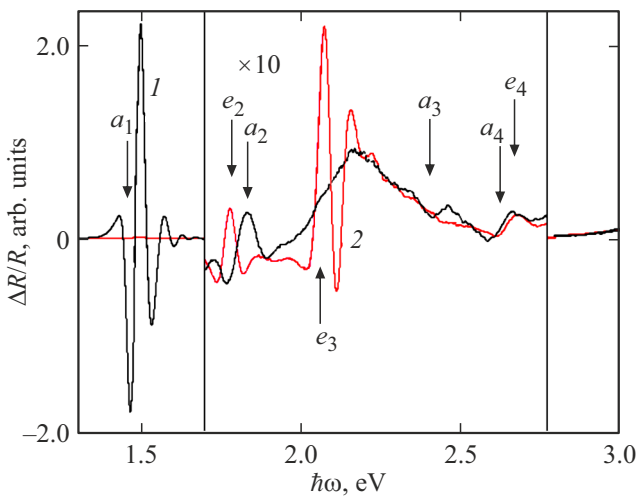


Figure 1. Electroreflection spectra of β -ZnP₂ in the energy range of (1.4–3) eV in the radiation polarization: 1 — $\mathbf{E} \parallel \mathbf{c}$, 2 — $\mathbf{E} \perp \mathbf{c}$.

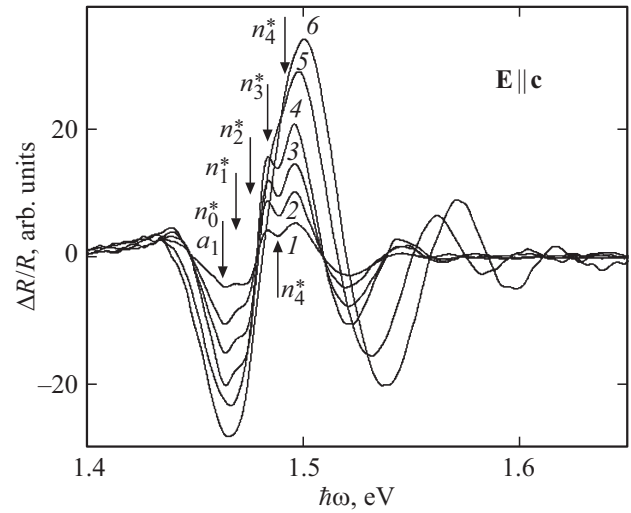


Figure 2. Electroreflection spectra in the fundamental absorption edge region of ITO- β -ZnP₂ at $U_m = 0.25$ V and constant bias voltage U_{cm} , V: 1 — +0.2, 2 — +0.15, 3 — +0.1, 4 — +0.05, 5 — -0.1, 6 — -0.2.

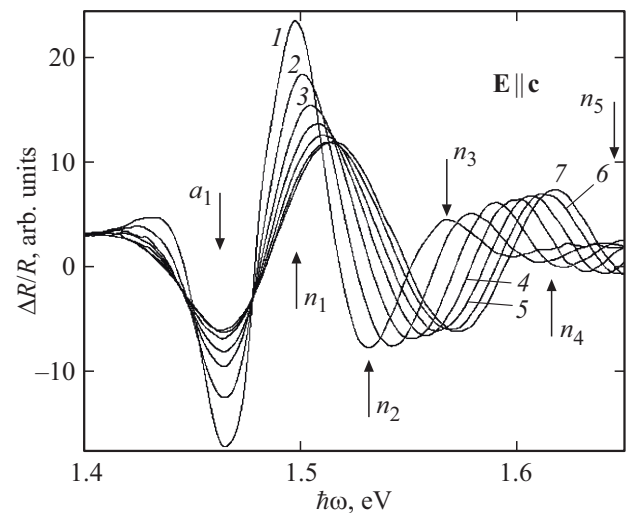


Figure 3. Electroreflection spectra in the fundamental absorption edge region of ITO- β -ZnP₂ at $U_m = 0.7$ V and constant bias voltage U_{cm} , V: 1 — 0, 2 — -2, 3 — -4, 4 — -6, 5 — -8, 6 — -10, 7 — -14.

have the same behavior in an electric field [22]. Energy position of the BIC lines with respect to the fundamental absorption edge of the crystal — (9.5–12) meV, and the energy band occupied by the BIC spectrum (~ 20 meV) don't contradict with this assumption. Interpretation on the assumption that the Franz–Keldysh effect appears on the most intense IHS $n_0 = 4$ BIC line [22] is described below. Similar structures in electroreflection spectra discussed in [4,8,14,17] cannot be reduced to the observed group of energy position extrema in the spectrum or to the implementation conditions.

Electroreflection oscillations in the absorption edge region in the $\mathbf{E} \parallel \mathbf{c}$ polarization (optical transition —

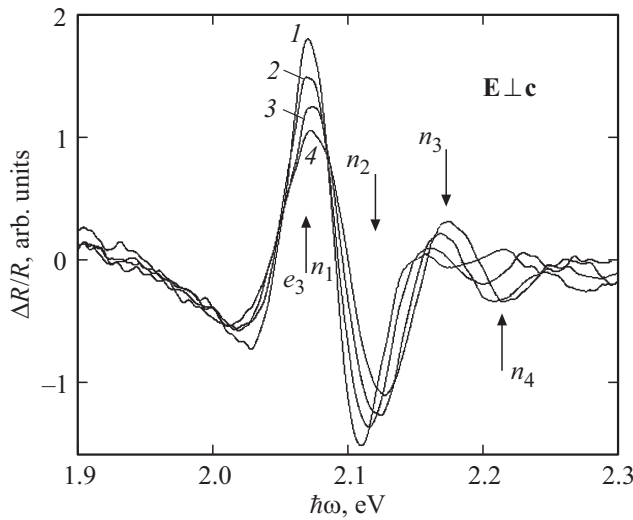


Figure 4. Electroreflection spectra of β -ZnP₂ in the allowed direct transition region in the $\mathbf{E} \perp \mathbf{c}$ polarization at $U_m = 0.7$ V depending on U_{cm} , V: 1 — 0, 2 — -2, 3 — -4, 4 — -6.

$\Gamma_{15}(V_1) - \Gamma_1(C_1)$), as noted above, are typical of the Franz–Keldysh effect, and the oscillation length is defined by quantum particle parameters and electric field strength [7,8]. Energy position of the first negative peak is associated with the allowed direct transition and exciton states destroyed by the electric field. As it follows from the exciton electrooptical effect theory, electric field causes exciton level shift and broadening, and merging of exciton levels with continuum. In β -ZnP₂ in the $\mathbf{E} \parallel \mathbf{c}$ light polarization, exciton absorption is defined by the electric dipole nS -series of a singlet exciton with the excitonic Rydberg energy $R = 35.9$ meV (10 K). Broadening of exciton light absorption bands due to the field exciton ionization in the electric field of surface-barrier structures forms a light absorption and photocurrent spectrum edge at temperatures higher than 80 K [18,22].

In $\mathbf{E} \perp \mathbf{c}$, transitions $\Gamma_{15}(V_1) - \Gamma_1(C_1)$ are forbidden. Direct forbidden transition edge is not exhibited in the electroreflection spectra. Triplet excitons with symmetry $\Gamma_1^- + \Gamma_2^-$ [10] allowed in this spectrum region are characterized by a low oscillator force, ionized and merged with continuum at room temperatures, and not identified in the electroreflection spectrum either (Figure 1).

Theory of electroreflection and Franz–Keldysh effect in semiconductors with the Schottky barrier applicable to GaAs is described in [5]. In the electroreflection spectra in β -ZnP₂ for transitions a_1 in $\mathbf{E} \parallel \mathbf{c}$, and for e_3 in $\mathbf{E} \perp \mathbf{c}$ without constant bias and with inverse bias voltage, oscillations are typical for light reflection in a strong electric field. Spectra are described using the Airy functions [4–7], for which relation for oscillation extremum energies vs. oscillation number is satisfied [8]:

$$n \cdot \pi = \psi + 3/4 [(E_n - E_0)/\hbar\theta]^{3/2}, \quad (1)$$

where n is the oscillation number, ψ is the relative phase factor, E_n is the extremum energy position, E_0 is the electronic transition energy, $\hbar\theta$ is the electrooptical energy or the Franz–Keldysh effect parameter.

Variation of the slope of dependences $(E_n - E_0)^{3/2}$ on the electroreflection oscillation extremum number n is associated with factors defining the wave function oscillation length of a quantum-mechanical particle. Dependences $[(E_n - E_0)/\hbar\theta]^{3/2} = f(n)$ are linear for ITO- β -ZnP₂ for all voltages applied to the structure (Figure 5). $\hbar\theta$ (see the Table) and ψ are determined by slopes of these dependences and cutoff on the n axis.

Phase factor derived from extrapolation of $[(E_n - E_0)/\hbar\theta]^{3/2}$ to the extremum reference axis is equal to $\pi/2$. In this case, critical transition energy can be determined by the energy position of the first negative extremum [6]. Electric field strength (see the Table) was calculated using the following equation:

$$F = [2 \cdot \mu_{\parallel}^* \cdot (\hbar\theta)^3 / q^2 \cdot \hbar^2]^{1/2}, \quad (2)$$

where q is the electron charge.

According to the data of [8], relations between the Franz–Keldysh oscillation periods and electrooptical parameters were calculated using the extrema phase shift: $\Delta E_1 = 1.1\hbar\theta$, $\Delta E_2 = 1.2\hbar\theta$, $\Delta E_3 = 0.9\hbar\theta$, $\Delta E_4 = 0.8\hbar\theta$, $1E_5 = 0.7\hbar\theta$, $1E_6 = 0.7\hbar\theta$. These values agree well with theoretical provisions [14,15].

Oscillation components of spectra shown in Figure 3 and curves 5, 6 in Figure 2, dependences in Figure 4 with some comments given below are described quite correctly by the following equations [4,7]:

$$\begin{aligned} \Delta R/R(E, F) = & \alpha_s(\varepsilon_1 \varepsilon_2) [C_1 \cdot \hbar\theta^{1/2} G(\eta)/E^2] \\ & + \beta_s(\varepsilon_1, \varepsilon_2) \cdot [C_2 \cdot \hbar\theta^{1/2} \cdot F(\eta)/E_2], \end{aligned} \quad (3)$$

$$G(\eta) = \pi \cdot [Ai'(\eta)Bi'(\eta) - \eta \cdot Ai(\eta) \cdot Bi(\eta) + U(\eta) \cdot \eta]^{1/2}, \quad (4)$$

$$F(\eta) = \pi \cdot [Ai'^2(\eta) - \eta \cdot Ai^2(\eta) - U(-\eta) \cdot (-\eta)]^{1/2}, \quad (5)$$

$$\eta = (E_0 - \hbar\omega + i\Gamma)/\hbar\theta, \quad (6)$$

where $F(\eta)$ and $G(\eta)$ are electrooptical functions of the first and second kind, $U(\eta)$ is the Heaviside function, $Ai(\eta) \cdot Bi(\eta)$ and $Ai'(\eta)Bi'(\eta)$ are the Airy functions and the derivatives, respectively, Γ is the broadening parameter of the electronic transition E_0 , $\alpha_s(\varepsilon_1, \varepsilon_2)$ and $\beta_s(\varepsilon_1, \varepsilon_2)$ are the Serafin coefficients.

Compared with the experimental spectra, small deviations in energy positions are observed in the theoretical spectra calculated using equations (3–6) for the $n > 5$. Moreover, it is not possible in some cases to describe quick decay of the Franz–Keldysh oscillations with increasing Γ . According to the authors of [23], full coincidence between the theory and experiment can be achieved provided that nonuniformities of the electric field and energy dependence Γ are taken into account.

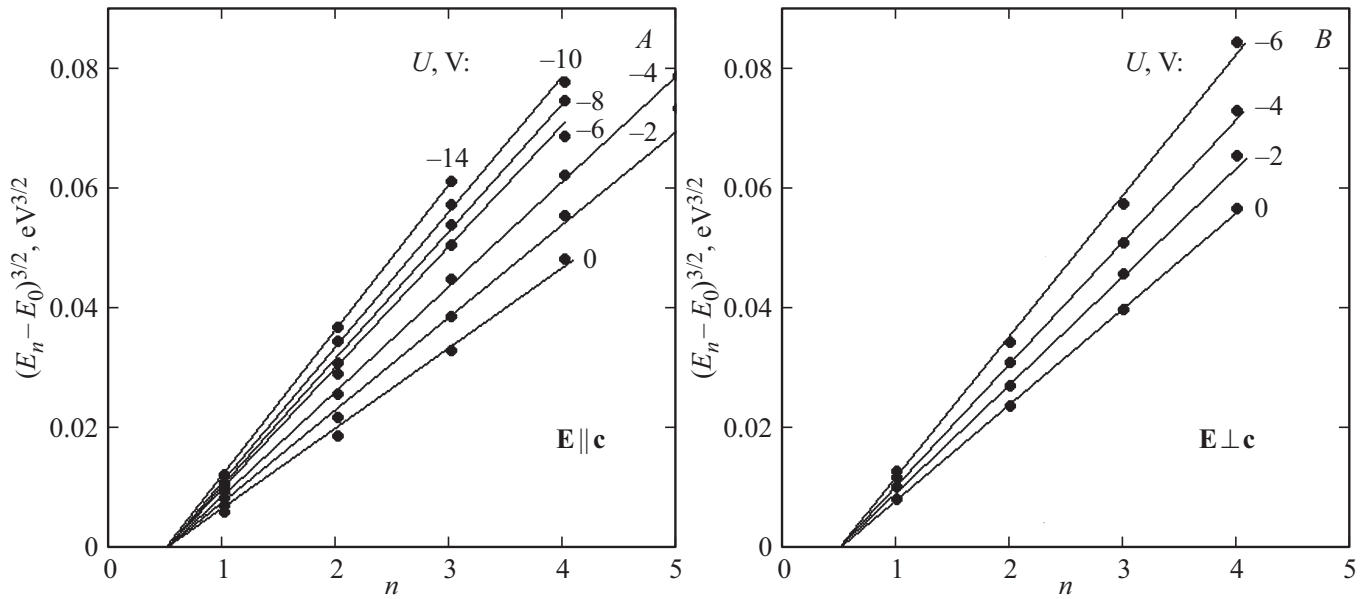


Figure 5. Dependence of the extremum energy difference on the oscillation number for the ITO– β -ZnP₂ structure for voltage series: *A* — for transition *a*₁, *B* — for transition *e*₃.

Values of the electrooptical energy $\hbar\theta$ and electric field F from the applied bias voltage U_{cm}

U_{cm}, V	0	–2	–4	–6	–8	–10	–12	–14
$F \cdot 10^7, V/m$	1.06	1.38	1.58	1.80	1.98	2.16	2.32	2.55
$\hbar\theta, eV$ $a_1 (E \parallel c)$	0.0273	0.0325	0.0389	0.0414	0.0439	0.0460	0.0474	0.0491
$\hbar\theta, eV$ $e_3 (E \perp c)$	0.0334	0.0359	0.0386	0.0430	–	–	–	–

Dependences of the broadening parameters Γ_w and Γ_c determined by the FWHM of the first extremum of the Franz–Keldysh oscillations and oscillation length, respectively, electron wavelength λ_{FK} with $\hbar\omega$ and voltage applied to the structure on the electric field strength F are shown in Figure 6. Calculations of F , Γ_c and λ_{FK} use the effective reduced mass for the allowed transition $\Gamma_{15}(V1) - \Gamma_1(C1)$ in β -ZnP₂ $\mu_{||}^* = 0.21m_0$ [25].

Γ_w was determined from the experimental data concerning the first extremum FWHM, Γ_c was calculated from the relation between the Franz–Keldysh oscillation length $\Delta E = (E_0 - E_{nmax})$ and $\hbar\theta$ and Γ [5,14]:

$$\Gamma_c = [(\hbar\theta)^3 / \Delta E]^{1/2} \quad (7)$$

where E_{nmax} is the extremum energy position of the last oscillation.

Values of Γ calculated by two methods within the measurement errors coincide when the field strengths are $\sim 1 \cdot 10^7$ V/m. As the electric field strength grows, dependences of broadening parameters differ, which is indicative of a more complex form of relation between the

broadening parameter and the oscillation form and photon energy.

Parameter defining the electron wavelength λ_{KF} with $\hbar\theta$ and effective mass μ_e^* was evaluated using the following equation, neglecting possible electric field nonuniformity at the light reflection depth [6]:

$$\lambda_{KF} = \hbar\theta / qF. \quad (8)$$

Relaxation time of photo-excited electrons and holes τ , determined from the oscillation properties is equal to $(3.9 - 2.1) \cdot 10^{-14}$ s in the electric field strength range of $(1.06 - 2.74) \cdot 10^7$ V/m.

Uncertainties in the values shown in Figure 6 and used in the calculations for $\hbar\theta$, Γ_w and Γ_c don't exceed 0.5% of their values and are equal, for example, 0.0273 ± 0.0002 , 0.0169 ± 0.0002 , 0.0173 ± 0.0002 eV, respectively, for $T = 27.5^\circ C$ and $F = 1.06 \cdot 10^7$ V/m.

For all bias voltages applied to the structure in the inverse direction, $\Gamma < \hbar\theta$ is satisfied taking into account the contribution of the modulation voltage component.

Dependences of the electric field strengths calculated using equation (2) and from capacitance-voltage character-

istics [14,18] offer additional opportunities for investigation of electrooptical effects and electronic parameters of semiconductors. Dependence of the bias voltage on the electric field strength on scale $U_{cm}^{1/2} = f(E)$ as shown in Figure 6 is linear and is used to determine the band bending in the heterojunction ITO– β -ZnP₂ structure.

The Franz–Keldysh oscillation behavior in the direct allowed optical transition $e_3(L_3(V1)–L_1(C1))$ in $\mathbf{E} \perp \mathbf{c}$ corresponds to the electrooptical effect case in the M_1 type critical point for $1/\mu_{\perp}^* > 0$ and $\hbar\omega > E_0$. Reduced effective mass $\mu_{\perp}^*$ of this transition, using as reference the reduced effective mass $\mu_{\parallel}^*$ for transition a_1 from the electroreflection spectra measured with the same bias voltages in both polarizations, is estimated as $0.16m_0$. Calculations were conducted on the assumption that the electric field acted in the same way on the electrooptical processes in these transitions and band structure interpretation corresponded to the case in [25].

The same calculations for $\hbar\theta$, Γ_w and Γ_c were conducted on the absorption line $n_0 = 4$ IHS BIC (Figure 2) on the assumption that $n_1^*–n_2^*$ are the Franz–Keldysh oscillations. It follows from the simulation data that the electrooptical energy of a quantum particle exhibited as electroreflection is 0.0077 eV, reduced effective mass $\mu_b^* \sim 6.5m_0$, phase factor $\psi = 1/2\pi$, broadening parameter $\Gamma = 0.0044$ eV. Such high effective mass is typical for the effective mass of bielectron μ_b^* [21]. The calculated μ_b^* in [21], on the assumption that the dielectric constant is equal to the continuum constant, is equal to $5.4\epsilon_0$. Difference in the values of reduced effective masses calculated by the optical and electroreflection spectra might be associated with the fact that the dielectric constant can differ from the continuum constant at short distances in the order of bielectron and BIC radius. Oscillations on the absorption line $n_0 = 4$ of IHS BIC are described by electrooptical functions for 3D critical point M_3 with reduced negative effective mass in the field direction and by the energy spaced away from the critical point a_1 on -11.5 meV.

Besides the above-mentioned Franz–Keldysh oscillations, typical resonance type structures responsible for optical transitions allowed in $\mathbf{E} \parallel \mathbf{c}$ and $\mathbf{E} \perp \mathbf{c}$ appear in the electroreflection spectra (Figure 1).

Energies of these transitions E_n were evaluated and broadening parameters Γ were calculated by the three-point method [6]. In $\mathbf{E} \parallel \mathbf{c}$, besides structures in the spectra in the energy region closest to the fundamental absorption edge as shown in Figure 1, a_2 (1.7600 eV), a_3 (2.3990 eV) and a_4 (2.6004 eV) appear. In $\mathbf{E} \perp \mathbf{c}$ in the direct forbidden optical transition region, there are no any peculiarities in the electroreflection spectrum that could be attributed to weakened prohibition on optical transitions due to the spin-orbit interaction [25]. Resonance structures typical of electroreflection, except that described above e_3 , e_2 (1.7446 eV), e_4 (2.3997 eV) and e_5 (2.6125 eV), appear in this polarization. a_2 and e_2 , a_3 and e_4 , a_4 and e_5 appear in both light polarizations, but are slightly shifted in energy.

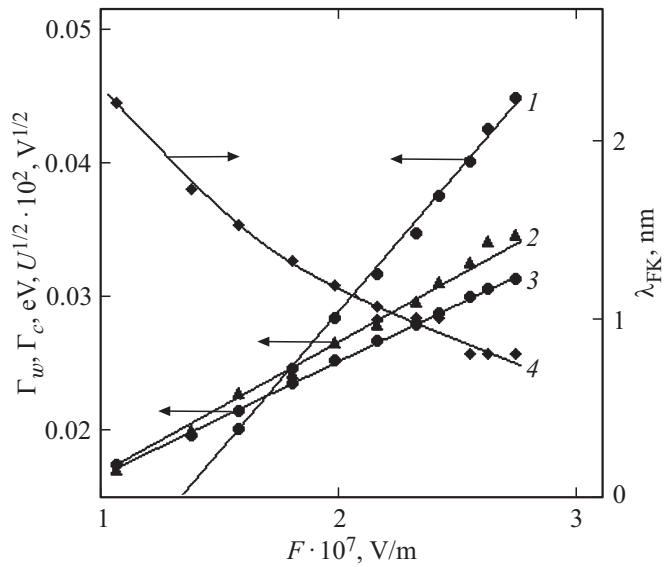


Figure 6. Dependences $U_{cm}^{1/2}$ (1) of the broadening parameters Γ_w and Γ_c determined by the FWHM of the first extremum of the Franz–Keldysh oscillations (2) and oscillation length (3), respectively, λ_{FK} (4) on the electric field strength F .

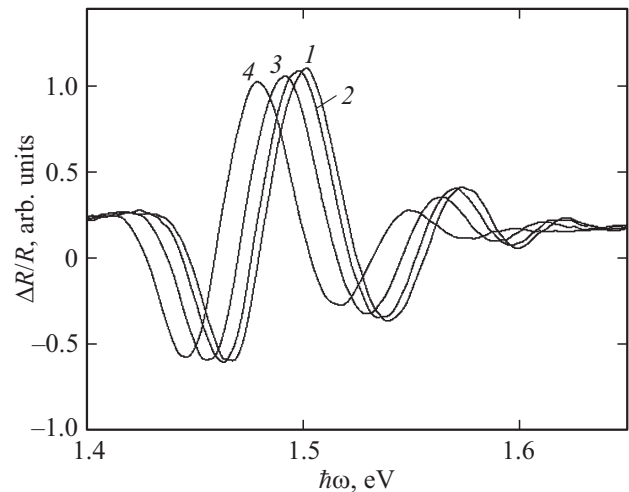


Figure 7. Electroreflection spectra in $\mathbf{E} \parallel \mathbf{c}$ from temperature T , K: 1 — 291, 2 — 339, 3 — 352, 4 — 365.

Figure 7 shows the temperature dependences of electroreflection spectra for transition a_1 related to the β -ZnP₂ band gap in $\mathbf{E} \parallel \mathbf{c}$. The temperature coefficient of absorption edge shift within this temperature range is $\rho = 3.8 \cdot 10^{-4}$ eV/K on average. $\hbar\theta$, λ_{KF} and F at the contact boundary vary insignificantly within $\sim 4\%$.

4. Conclusion

The electroreflection method was used to investigate the electric field impact on the optical properties of β -ZnP₂

and the Franz–Keldysh effect in critical points of optical transitions.

It was found that the Franz–Keldysh effect is exhibited in light polarizations of the dipole-allowed direct electronic transition $\Gamma_{15}(V_1) - \Gamma_1(C_1)$ and $L_3(V_1) - L_1(C_1)$. Electrooptical energy, wave function oscillation length of a quantum-mechanical particle depending on the electric field strength and temperature were determined. Reduced effective mass $\mu_{\perp}^*$ equal to $0.16m_0$ was determined for the optical transition e_3 ($L_3(V_1) - L_1(C_1)$) allowed in the $\mathbf{E} \perp \mathbf{c}$ polarization.

Peculiarities were detected in the electroreflection spectra and can be interpreted within the Franz–Keldysh effect on states in the critical point M_3 typical of the photo-excited charge carrier pairing effects, and formation of IHS absorption lines in crystals [21,27,28].

Optical transition energies in β -ZnP₂ were specified in polarized light within the energy range from the fundamental absorption edge to 4 eV.

The findings show that monoclinic zinc diphosphide is a promising material, experimental investigations of which using the optical differential spectroscopy methods might facilitate the development of theory of electroreflection and electroabsorption in crystals with complex energy structure.

Conflict of interest

The author declares no conflict of interest.

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