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Polarized optical transmittance spectra of nonlinear thiogallate crystals near “isotropic point”

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Abstract. The results of researches of the polarized optical transmittance spectra of nonlinear thiogallate crystals near the “isotropic point” are represented and discussed.

Keywords: polarization, optical transmittance spectra, nonlinear optics, thiogallate crystal, isotropic point.

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Wide-gap semiconducting crystals of the thiogallate family are well-known and promising nonlinear materials. Among their optical properties, those occurring in a vicinity of the light wavelength $\lambda = \lambda_i$, where the ordinary and extraordinary refractive indices coincide [the point referred to as the “isotropic point” (IP)] represent a not fully resolved problem. They offer also a possibility for constructing a unique type of ultranarrow-band optical filters [1-3]. There still exist the results that have not been satisfactorily explained, e.g., the unusual spectral behaviors of the polarized light transmittances $J_{\parallel}(\lambda)$ and $J_{\perp}(\lambda)$ detected for the input linear polarizations $E \parallel c$ and $E \perp c$, and the non-polarized transmittance $J_{np}(\lambda)$ near the IP (see [1-3] and Fig. 1). The interpretation of those phenomena has been for the first time done in [2, 3], though it is mainly based on the qualitative arguments rather than on the exact analytic expressions.

The approach adopted in this study is based on the rigorous development of a hypothesis of the influence of the differential absorption [the so-called linear dichroism (LD)] on the transmittance spectra near the IP. We have derived the functions $J_{\parallel}(\lambda)$, $J_{\perp}(\lambda)$, and $J_{np}(\lambda)$, by using the Jones matrix formalism (see [4-6]). Namely, the transmittances in the cases of the initial polarizations “1” ($E \parallel c$ or the azimuth $\varphi = 0^\circ$ – see [2, 3]) and “2” ($E \perp c$ or $\varphi = 90^\circ$) are as follows:

$$J_{\parallel, \perp} = e^{-\alpha d} [\cos^2 a + \sinh^2 b + \frac{\Delta_l^2 + \Delta_c^2 + \delta_l^2}{4A_\Delta^2} (\sin^2 a + \sinh^2 b) \mp \Delta J],$$

$$J_{np} = (J_{\parallel} + J_{\perp})/2, \quad (1)$$

$$\Delta J = \frac{\sqrt{\Delta_l^2 + \delta_l^2}}{2A_\Delta} [\sin(\arctan \frac{\delta_l}{\Delta_l} - \varphi_\Delta) \sin 2a + \cos(\arctan \frac{\delta_l}{\Delta_l} - \varphi_\Delta) \sinh 2b],$$

$$a + ib = A_\Delta \exp(i\varphi_\Delta) = \sqrt{(\Delta_l + i\delta_l)^2 + \Delta_c^2} / 2,$$

$$A_\Delta = \sqrt{(\Delta_l^2 + \Delta_c^2 - \delta_l^2)^2 + 4\Delta_l^2\delta_l^2} / 2,$$

$$\varphi_\Delta = (1/2) \arctan \frac{2\Delta_l\delta_l}{\Delta_l^2 + \Delta_c^2 - \delta_l^2}. \quad (2)$$

In (1) and (2), α is the mean absorption coefficient, $\Delta_l = (2\pi d/\lambda)\Delta n_l$ and $\Delta_c = (2\pi d/\lambda)\Delta n_c$ are, the phase retardations due to the linear ($\Delta n_l = n_e - n_o$) and circular (Δn_c) birefringences, respectively, $\delta_l = (2\pi d/\lambda)\Delta \kappa_l$ is the analogous parameter of LD ($\Delta \kappa_l = \kappa_e - \kappa_o$ with the dichroic absorption difference $\Delta \alpha = \alpha_{\parallel} - \alpha_{\perp} \equiv \alpha_e - \alpha_o = 2\delta_l/d$), and d is the crystal thickness.

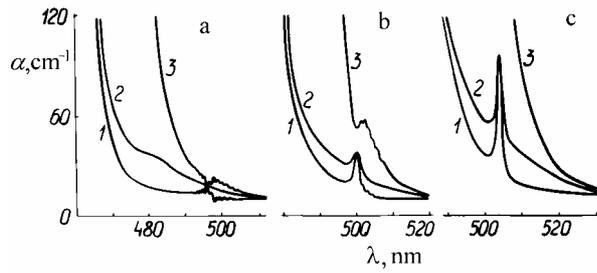


Fig. 1. Spectral dependences of the absorption coefficients α for AgGaS₂ crystals in a vicinity of the IP for the polarizations $E \perp c$ (curves 1), $E \parallel c$ (3), and non-polarized light (2). The thickness of the crystal is $d = 0.35$ mm and the temperatures are $T = 460$ K (a), $T = 460$ K (b) and $T = 460$ K (c). The IP appears at $\lambda_i = 497$ nm at room temperature.

If $\Delta_c = 0$, we have $J_{\parallel\perp} = \exp(-\alpha_{\parallel\perp}d)$. For non-dichroic crystals ($\Delta_c \neq 0$, $\delta_l = 0$),

$$J_{\parallel} = J_{\perp} = e^{-\alpha d}. \quad (3)$$

Let us now concentrate at the polarized transmittance anomalies just at the IP, where the total phase retardation is $\Delta = \sqrt{\Delta_c^2 - \delta_l^2}$. Consider the ideal situation, when Δ_c , δ_l , and α manifest no dispersion and the birefringence near the IP behaves as $\Delta n_l = \gamma(\lambda - \lambda_i)$. Let us label the “apparent” LD at the IP as $\Delta\alpha_{ob}$ and the “true” LD (defined as if the IP does not exist – see Fig. 2) as $\Delta\alpha_i = \Delta\alpha(\lambda_i)$. Using formulae (1) and (2) and the definition $\Delta\alpha_{ob} = \ln[J_{\perp}(\lambda_i)/J_{\parallel}(\lambda_i)]/d$, we get

$$\Delta\alpha_{ob} \approx \Delta\alpha_i \text{ sinc } \Delta \quad (4)$$

in the practical case of a weak LD ($\delta_l \ll \Delta$). Formula (4) yields the inequality $|\Delta\alpha_{ob}| \leq |\Delta\alpha_i|$, a progressive decrease in $\Delta\alpha_{ob}$ with increase in the sample thickness ($\Delta \approx d^{-1}$), and a rapidly achieved limit $\Delta\alpha_{ob} \rightarrow 0$ (i.e., touching the spectral curves $J_{\parallel}(\lambda)$ and $J_{\perp}(\lambda)$ at the IP – see Fig. 2, the insert). Note that the “apparent” LD behavior according to (4) correlates well with the simple, though general, considerations about the spatial evolution of wave polarization and absorption owing to the wave coupling [3].

Quantitative verifications of formula (4) with the use of a large variety of experimental results (ZnO, CdS, CdSiP₂, CdGa₂S₄, AgGaS₂, and AgGa_{0.98}In_{0.02}S₂ crystals, various Δ and d , see, e.g., Fig. 1) testify to its validity. Some disagreement is present only for very thin AgGa_{0.98}In_{0.02}S₂ samples [3]. They might likely originate from a non-zero azimuth φ (see [3]). Then relation (4) should be replaced with

$$\Delta\alpha_{ob} \approx \Delta\alpha_i [\text{sinc } \Delta \cos 2\varphi + (\Delta_c/2) \text{sinc}^2(\Delta/2) \sin 2\varphi], \quad (5)$$

producing a notable “overlapping” effect for the curves $\alpha_{\perp}(\lambda)$ and $\alpha_{\parallel}(\lambda)$.

Let us finally analyze the non-polarized anomaly $\Delta\alpha_{np}$ at the IP detected in [2, 3]. Its value may be defined as $\Delta\alpha_{np} = \alpha_{np} - \alpha_{np}^{(0)}$ (see Fig. 2). Using (1) and (2), along with the relations $\alpha_{np}^{(0)} = -\ln J_{np}^{(0)}/d$ and $\alpha_{np} = -\ln J_{np}/d$, we have

$$\Delta\alpha_{np} \approx \frac{\Delta\alpha_i^2 d}{8} (1 - \text{sinc}^2 \frac{\Delta}{2}) \quad (6)$$

in case of $\delta_l/\Delta \ll 1$. Formula (6) suggests a positive $\Delta\alpha_{np}$ for arbitrary crystal thicknesses ($\Delta\alpha_{np} > 0$), the actual absence of the anomaly for thin crystals ($\Delta\alpha_{np} \ll \Delta\alpha_i$), and its rapid increase ($\propto \Delta\alpha_i^2$) with increase in LD. All of these facts correlate well with the available experimental results [2, 3]. Moreover, we have a quite good quantitative agreement, e.g., $\Delta\alpha_{np}^{\text{th}} \approx 12.8 \text{ cm}^{-1}$ and $\Delta\alpha_{np}^{\text{exp}} \approx 14 \text{ cm}^{-1}$ [2], $\Delta\alpha_{np}^{\text{th}} \approx 0.3 \text{ cm}^{-1}$ and $\Delta\alpha_{np}^{\text{exp}} \approx 0 \text{ cm}^{-1}$ [3], where the superscripts “th” and “exp” mean, respectively, theoretical and experimental values. Formula (6) also predicts the evolution of the type of $\lim_{d \rightarrow \infty} \Delta\alpha_{np} = \Delta\alpha_i/2$, in consonance with the data in [2, 3] for AgGaS₂ and

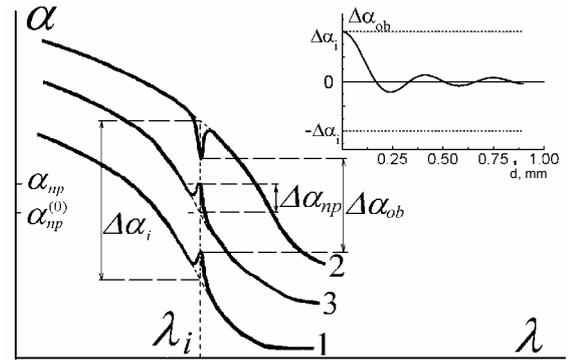


Fig. 2. Schematic representation of the absorption spectra α_{\perp} (curve 1), α_{\parallel} (2), and α_{np} (3) near the IP $\lambda = \lambda_i$ for dichroic crystals and the definitions of parameters $\Delta\alpha_i$, $\Delta\alpha_{ob}$ and $\Delta\alpha_{np}$ (see the text). The insert shows a specific dependence of the “apparent” LD $\Delta\alpha_{ob}$ at the IP on the thickness d according to formula (4) for $\lambda_i = 497$ nm, $\Delta n_c = 1.5 \cdot 10^{-3}$, and $\Delta \kappa_l = 1.5 \cdot 10^{-4}$.

AgGa_{0.98}In_{0.02}S₂ samples with the largest thicknesses ($\Delta\alpha_{np}/\Delta\alpha_i \approx 0.42$ and 0.44 , respectively).

The theory presented above enables one to explain the temperature evolution of $\Delta\alpha_{ob}$ and $\Delta\alpha_{np}$ for AgGaS₂ [3] (Fig. 1). At $T = 300$ K, we have $\Delta\alpha_{ob}^{exp} \approx 0$, $\Delta\alpha_{np}^{exp} \approx 0$, and $\Delta\alpha_{ob}^{th} \approx 0$ (since $\Delta \approx 2\pi$), $\Delta\alpha_{np}^{th} \approx 0.8$ cm⁻¹. The latter value can be hardly detectable in a real experiment. To describe changes in the absorption parameter with increase in the temperature ($\Delta\alpha_{ob}^{exp} \approx 15$ cm⁻¹ and $\Delta\alpha_{np}^{exp} \approx 11$ cm⁻¹ at $T = 460$ K), we have to account for the temperature variations of both the LD [$\Delta\kappa_l(\lambda_i)$ or $\Delta\alpha_i$] and the optical activity $\Delta n_c(\lambda_i)$. Extrapolating the corresponding literature data toward the high-temperature region, we obtain a rough estimation of the gyration at $T = 460$ K (~ 600 deg/mm). Then we have $\Delta\alpha_{ob}^{th} \sim 8$ cm⁻¹ and $\Delta\alpha_{np}^{th} \approx 13$ cm⁻¹, where the highest errors associated with calculations of the oscillating factor $\text{sinc}(\Delta/2)$ affect $\Delta\alpha_{ob}^{th}$ much significantly than $\Delta\alpha_{np}^{th}$. The agreement of theory and experiment should be regarded as a satisfactory one.

Thus, we have improved the theory of the propagation of electromagnetic waves in the crystals possessing the IP with regard for LD. The effect of LD on the spectral phenomena in the vicinity of the IP turns out to have a prominent character. We have explained quantitatively the anomalies at the IP for both the polarized and non-polarized optical transmittance spectra. The further consideration of the effects of LD will be a subject of a forthcoming paper.

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