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Anomalous Dispersion of Forced Birefringence under Deformation and the Relation of Photoelasticity to Piezospectroscopic Phenomena in Crystals

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In a number of studies carried out in recent years, various reversible phenomena have been discovered and studied in the optical spectra of cubic crystals under oriented elastic deformation of the crystals, which lowers their symmetry. These "piezospectroscopic" phenomena consist in the polarized splitting of spectral bands, changes in the intensity and polarization of bands (see, for example, ([1],[2]) and references therein). The existence of significant anisotropic changes in the absorption spectra of crystals under deformation raises the question of how these changes affect the dispersion of the refractive index associated with the absorption bands. In other words, the question arises of the relation between piezospectroscopic phenomena and the known phenomenon of photoelasticity—forced birefringence of cubic crystals under their mechanical deformation. To clarify this question, it was of interest to investigate experimentally the dependence of the forced birefringence of crystals on the wavelength of light in the spectral region near absorption bands.

To investigate the dispersion of photoelasticity we used an interference method analogous to that previously used in studying, near dispersion lines, birefringence in other optical phenomena—birefringence and rotation of the plane of polarization in gases in a magnetic field (the Voigt and Macaluso-Corbino effects—see ([3])), and birefringence in optically anisotropic noncubic crystals ([4]). In the adopted arrangement (Fig. 1), light from a continuous-spectrum source L passes through a polarizer N_1, which polarizes the light at 45° to the vertical, and is focused by objective O_1 on the single-crystal specimen M. The specimen is mounted in a special press placed in a Dewar with liquid helium (or nitrogen) and is uniaxially compressed in it along the vertical axis P. Next, through objective O_2, the light falls on the slit of spectrograph S; immediately at the slit there are a quartz wedge (or Babinet compensator) K and a polaroid N_2, crossed with N_1. Compression is applied along $\langle 100\rangle$, $\langle 111\rangle$, $\langle 110\rangle$; in this case the principal axes of the ellipsoid of refractive indices in the stressed specimen prove to be directed $\parallel P$ and in the plane $\perp P$.

Fig. 1. Schematic of the setup (side view)

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In the absence of deformation, light polarized at 45° passes through the optically isotropic crystal and, in the birefringent wedge, is resolved into waves of vertical $(E_\parallel)$ and horizontal $(E_\perp)$ polarization. Interference of the waves at the exit from the wedge gives, in the general case, elliptically polarized light transmitted by N_2. However, N_2 blocks the light at those heights of the wedge (slit) h (measured from the apex of the wedge) where the optical path difference accumulated in the wedge is equal to an integer number k of wavelengths:

$(n_0 - n_e)l = (n_0 - n_e)h \tan\alpha = k\lambda$; n_0, n_e are the principal refractive indices of quartz; α is the wedge angle; l is the path in the wedge at height h. Therefore the absorption spectrum of the crystal in the spectrograph is crossed by a series of almost horizontal dark bands corresponding to different orders k of interference. Under deformation, owing to the induced birefringence of the crystal, the waves with $E_\parallel$ and $E_\perp$ acquire in it an additional path difference, and the extinguishing conditions of interference are then fulfilled at other heights h' of the wedge (slit): $(n_0 - n_e)h' \tan\alpha + \Delta n(\lambda)d = k\lambda$, where $\Delta n(\lambda) = n_\parallel(\lambda) - n_\perp(\lambda)$ is the difference of refractive indices in the stressed crystal for light polarized with $E_\parallel(\parallel P)$ and $E_\perp(\perp P)$; d is the specimen thickness. The vertical displacement of the interference bands arising in this case is

$$\Delta H = (h' - h)f = \Delta n(\lambda) df / (n_0 - n_e) \tan\alpha,$$

where f is the magnification of the spectrograph. Thus, the course of the displacement of the bands over the spectrum $\Delta H(\lambda)$ directly reflects the frequency dependence of the induced birefringence

$$\Delta n(\lambda) = n_\parallel(\lambda) - n_\perp(\lambda).$$

Fig. 1. Schematic of the setup (side view)

The investigations were first carried out on several crystals in which the existence of appreciable deformation effects in narrow-line "impurity" absorption spectra had previously been established.

A. Long-wavelength absorption lines of CaF_2—Sm^{2+} (6901 Å) and CaF_2—Eu^{2+} (4130 Å), corresponding to interconfigurational transitions $4f^n \to 4f^{n-1}5d$ in rare-earth ions ([5,6]). From ([7,8]) it is known that these lines split under deformation, owing to splitting of the upper $(4f^{n-1}5d)$ degenerate levels of Sm^{2+} and Eu^{2+} under a deformation distortion of the symmetry of the crystal field (O_h) in which the ions are located. As is seen from Fig. 2a, in the case of the CaF_2—Sm^{2+} 6901 Å line for $P \parallel [110]$, $L \parallel [1\bar{1}0]$, when a completely polarized doublet is observed in absorption (L is the direction of observation), the interference

Figure 2

Figure 2: Figure 2

bands are symmetrically bent on both sides outside the doublet; the direction of the bending shows that outside the doublet the birefringence $\Delta n < 0$. Inside the doublet the bands form symmetric curves with sharp extrema lying in the region $\Delta n > 0$. At points corresponding to the positions of the components of the deformation doublet, the course of the interference bands has a "discontinuous" character. A qualitatively analogous effect, but with the opposite sign of Δn outside and inside the doublet, is also observed near the CaF_2–Eu^{2+} 4130 Å line for $P \parallel \langle 100 \rangle$, $L \perp P$, when splitting of the line into a partially polarized doublet occurs (Fig. 2b).

B. The 391 mμ line in the spectrum of colored LiF, corresponding to a purely electronic transition in R-centers ([9,10]). As established in ([16]), under deformation ($T = 4.2°$ K), along with splitting of the line, there occurs a redistribution of absorption intensity among different polarization states, caused by reorientation of anisotropic R-centers in the stressed crystal. In particular, for $P \parallel \langle 100 \rangle$, $L \perp P$, a single component is observed in the spectrum, polarized predominantly with $E \parallel P$. As is seen from Fig. 2c, in this case the interference bands form characteristic bends of opposite sign on the two sides of the line.

Thus, in the region of the investigated CaF_2–Sm^{2+}, Eu^{2+}, and LiF lines, the induced birefringence $\Delta n(\lambda)$ undergoes sharp changes; it changes both in the sign of Δn and in the sign of the dispersion $d\Delta n / d\lambda$ (anomalous dispersion of photoelasticity). The picture here is outwardly similar to the anomalous dispersion of birefringence in other optical phenomena mentioned in the second paragraph of the article ([3,4]), and, for its explanation, similar qualitative considerations based on the study of ⋯

Fig. 2. Anomalous dispersion of the photoelasticity of crystals. (The photographs were obtained according to the scheme of Fig. 1. Upward displacement of the interference fringes corresponds to an increase in $\Delta n = n_{\parallel} - n_{\perp}$, downward displacement to a decrease in Δn.) **a** –CaF_2–Sm^{2+}, region of the 6901 Å line ($T = 4.2°$K) for $P \parallel [110]$, $L \parallel [110]$ and various compressive stresses P (in kg/mm^2); at the top, the spectrograms in polarized light of the 6901 Å doublet at the same conditions. **b** –CaF_2–Eu^{2+}, region of the 4130 Å line ($T = 4.2°$K) for $P \parallel \langle 100 \rangle$, $L \perp P$; at the top, the 4130 Å doublet in unpolarized light; $P = 13.4$ kg/mm^2. **c** –LiF, region of the 391 mμ line ($T = 4.2°$K) with a short-wavelength vibrational satellite, marked by an arrow, for $P \parallel \langle 100 \rangle$, $L \perp P$; $P = 16.8$ kg/mm^2. **d** –ZnTe, region preceding the edge of fundamental absorption ($T = 77°$K) for $P \parallel \langle 100 \rangle$, $L \perp P$; the position of the edge observed for the sample thickness used ($d \approx 1$ mm) is marked by an arrow.

of the known relation between absorption lines and the refractive index of the medium (Fig. 3). Indeed, in the case of a completely polarized doublet, its

Fig. 3. Scheme of the origin of anomalous dispersion of photoelasticity near lines

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components, polarized with $E \parallel P$ and $E \perp P$, contribute respectively to the dispersion of the refractive indices $n_{\parallel}(\lambda)$ and $n_{\perp}(\lambda)$ near the lines, and, as is easily seen from Fig. 3a, the difference $\Delta n(\lambda) = n_{\parallel}(\lambda) - n_{\perp}(\lambda)$ gives a symmetric curve, in qualitatively good agreement with the experimental picture for 6901 Å CaF_2–Sm^{2+} (Fig. 2a). In the case where deformation causes only polarization of a line, the behavior of $n_{\parallel}(\lambda)$ and $n_{\perp}(\lambda)$ near the line

Fig. 3. Scheme of the origin of anomalous dispersion of photoelasticity near lines

differs only by the scale of the two curves (Fig. 3b), and their difference Δn gives the typical curve of anomalous dispersion, in agreement with that observed experimentally for LiF 391 mμ (Fig. 2c). In the case of a partially polarized doublet (Fig. 3c), for birefringence only the "difference" of the intensities of each component in $E \parallel P$ and $E \perp P$ is essential; this difference is the same in absolute magnitude for both components, which accounts—in agreement with experiment (4130 Å CaF_2+Eu^{2+}, Fig. 2b)—for the symmetry of the resulting pattern, quite analogous to the case of a completely polarized doublet. The opposite sign of Δn at 4130 and 6901 Å is explained by the different order of arrangement of the components with preferential polarization $E \parallel P$ and $E \perp P$.

The discovery of anomalous dispersion of "impurity" photoelasticity and the establishment of a direct connection between impurity photoelasticity and the splitting and polarization of impurity lines under deformation* testify in favor of the view that the "intrinsic" photoelasticity of crystals is also due (at least in part) to deformation-induced splitting and polarization of bands—excitonic and mesonic—in the spectrum of intrinsic absorption of crystals. We note that the connection of forced birefringence with a possible change under deformation of the natural frequencies and oscillator strengths of the dispersion electrons of the atoms forming the lattice, and with the resulting change in the polarizability of the atoms, was assumed earlier in certain phenomenological theories of the dispersion of piezo-optical coefficients of crystals and glasses ([11–13]).

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* "Impurity" photoelasticity of crystals is apparently observed in the present work for the first time. We note also that under conditions where deformation does not cause splitting or polarization of the lines (CaF_2–Sm^{2+}, 6901 Å, with $P \parallel [110]$, $L \parallel [001]$ and with $P \parallel \langle 111 \rangle$ ([7,8])), no anomalies were observed in the interference pattern near the line. The quantity $\Delta n(\lambda)$ of "impurity" photoelasticity was considered above relative to the forced "intrinsic" birefringence of crystals, which in the visible region has no noticeable dispersion and

in experiment gives a general shift of the interference fringes.

According to the scheme of Fig. 1, we photographed the dispersion of induced birefringence near the edge of the fundamental absorption of cubic ZnTe single crystals, for which in (^{15}) deformation doublet splitting of the long-wavelength exciton band (5236 Å) was observed, caused by splitting of the degenerate valence band of ZnTe. For $P \parallel \langle 100 \rangle$, $L \perp P$, the interference bands exhibited a strong bending near the edge (Fig. 2a); here it is significant that the direction of the bending agrees with the character of the polarization of the deformation exciton doublet (the long-wavelength component is polarized predominantly with $E \parallel P$, the short-wavelength one completely with $E \perp P$). This circumstance indicates a large contribution to the photoelasticity of ZnTe in the region near the edge of the splitting effect of the exciton band (and also, possibly, of the bands corresponding to interband transitions).

The results of the present work directly show that the photoelastic properties of crystals are closely connected with the scheme of quantum energy states (intrinsic—exciton and band—and impurity states) and optical transitions in the crystal, and with changes in the energy spectrum under deformation. The photoelastic effect turns out to be just as derivative of the piezospectroscopic effect as the refractive index of crystals is determined by their absorption spectrum. In particular, the numerical photoelastic constants directly depend on the parameters of deformation splitting (^{1}, ^{2}) of bands in the spectra and on the degree of polarization of the split bands. Let us note that the usually considered phenomenological theory of the photoelasticity of crystals (^{14}), in which the birefringence effect is assumed to be small in magnitude ($\Delta n \ll n$) and linearly dependent on stresses σ_{ik} (deformations u_{ik}) in the lattice, is evidently valid only for the spectral region far from absorption bands. Indeed, as is seen from Figs. 2 and 3, the magnitude of the induced birefringence Δn near a band is comparable with the very magnitude of the contribution to the refractive index n caused by the band, i.e., the effect can be very large (“resonance” photoelasticity). In addition, it depends nonlinearly on $\sigma_{ik}(u_{ik})$.

The study of induced birefringence near absorption bands makes it possible to investigate the contribution of bands, in particular impurity bands, to the dispersion of the refractive index of a crystal and to find the oscillator strengths (the numbers Nf) for the bands. It can also be used as a sensitive method for investigating deformation splittings in intrinsic and impurity spectra, which yields valuable information about the electronic levels of the crystal (^{1}, ^{2}). The considerable magnitude of resonance photoelasticity is also of interest for piezo-optical modulation of light.

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Note: Figure translations are in progress. See original paper for figures.