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OPTICAL PROPERTIES OF PLASTICALLY DEFORMED $\text{A}^{\text{II}}\text{B}^{\text{VI}}$ CRYSTALS


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Résumé - On décrit la diffraction et la propagation anormale de la lumière dans une structure périodique de dislocations formées dans un cristal CdS, l'activité optique des dislocations ainsi que les changements brusques de la polarisation de la photoluminescence qui accompagnent les lignes de glissement. Les effets ainsi mis en évidence peuvent être interprétés en termes d'interactions collectives dans des sous-ensembles de dislocations et de défauts ponctuels.

Abstract - Diffraction and anomalous propagation of light in the periphery of dislocation structure of CdS, optical activity of dislocations and abrupt changes in the polarization of the photoluminescence of dislocation traces have been described. The effects revealed can be explained in terms of the collective interactions in the subsystems of dislocations and point defects.

At the related conference in 1978 in our talk on optical properties of II-VI compounds with dislocations we reported on the light focusing by dislocations, the appearance of dislocation bands in the absorption and luminescence spectra, the redistribution of luminescence intensity of bound excitons in the vicinity of the dislocations. Principally, said phenomena can be explained in terms of the properties of isolated dislocations and point defects under assumption that no collective interactions occur between them. The further investigations carried out in our Institute have shown that the light propagation and the photoluminescence in plastically deformed $\text{A}^{\text{II}}\text{B}^{\text{VI}}$ single crystals have some features which testify in favour of strong collective interactions in the system of defects introduced by plastic deformation.

The study of elastic stresses in plastically deformed cadmium sulphide specimens by the method of photoelasticity /2/ and the observation of dislocation glide traces on lateral crystal faces pointed to the fact that the distribution of dislocation glide bands was close to periodic in the direction normal to the shear, the distance between neighbouring glide bands being several microns.

Taking into account that dislocations in their vicinity produce considerable local changes in the optical characteristics of refraction and absorption /2,3/, one could expect the appearance of effects of light diffraction similar to those of the X-ray diffraction by periodic, atomic crystal lattice /4/.

Indeed, we have revealed the expected diffraction effects when exposing plastically deformed CdS specimens to a parallel monochromatic light beam (the angular beam divergence did not exceed 3 minutes,
the spectral width was 10 Å). At certain (rather small) angles between the wave vector of light and the glide bands on the exit surface of the specimen we observed a periodic distribution of light intensity in the form of fringes aligned parallel to the direction of shear. Within the experimental error the light pattern period of 4 Å was in accord with the distance between glide bands determined from the dislocation glide traces on lateral faces of the specimen. For the wavelength 560 nm the most distinct periodic distribution of light intensity was observed at two angular positions between the normal to the crystal face and the light flux, namely at 0° and 90°. The periodic modulation of intensity disappeared in the intermediate positions (the normal to the light entrance surface was deviated from the 6th-order axis by 20°).

In cadmium sulphide in the spectral region of 560 nm a band of light absorption by prismatic dislocations is seen /3/. It turned out that in this spectral region the degree of crystal transmittance oscillates as the angle between the wave vector and the glide bands is changed. In the above specimen the luminous transmittance was maximal at 0° and 90° (i.e. when the distinct periodic modulation of intensity was observed), and a step in the transmission spectrum corresponding to the dislocation absorption was not seen. In other angular positions when the intensity modulation vanishes the transmittance drops and the step of dislocation absorption is clearly seen in the spectrum (Fig. 1). Angular oscillations of dislocation absorption are well seen in Fig. 2, where the slope of transmission spectrum is plotted as a function of the angle of light incidence for λ = 560 nm.

By increasing the angle between the wave vector and the light flux one more diffraction effect is observed - the appearance of a secondary beam deflected from the incident one by some fractions of a degree and polarized normally to the latter. Figures 3 and 4 show the rays geometry and the angular distribution of emergent light

![Fig. 1 - Transmission spectra of prismatic dislocated CdS at various angles of light incidence. The light polarization is parallel to the glide bands. a) Ψ = 0°10'; b) 3°20'; c) 9°10'; d) 13°20'.](image-url)
Fig. 2 - The slope of transmission spectrum of cadmium sulphide with prismatic dislocations at 560 nm as a function of the angle of light incidence.

Fig. 3 - Diagram for secondary beam formation at great angles between the light flux and the glide band.

Fig. 4 - The angular distribution of the light flux emerging from plastically deformed specimen.

fluxes (the angle between the incident light flux and the glide bands was about 45°). The spectral dependence of the secondary beam intensity in the red part of the spectrum is shown in Fig. 5, where quasiperiodic oscillations are observed. In order to gain some insight into the mechanism of the secondary beam origination we should emphasize that the intensity minimum at 562 nm resembles in form the photoelastic minimum observed at 555 nm in the transmission spectrum of the system of two crossed polarizers with a plastically deformed cadmium sulphide specimen between them, but is shifted from
Fig. 5 - Secondary beam intensity oscillations in the red part of the spectrum.

the latter to the red side by 7 nm.

The described diffraction effects result from collective regularization in the space distribution of dislocations. Below we shall give the experimental evidence for the deformation-induced regularization in the arrangement and orientation of point defects.

In earlier papers we have treated the complicated structure of spectra of light focusing and scattering by dislocations in the region approaching the absorption edge of cadmium sulphide /4–6/ in the polarization parallel to the glide bands. Further investigations have shown that in the perpendicular polarization the scattering and focusing spectra have minima in the region lying far away from the absorption edge /5/. For basal dislocations this minimum lies at 580 nm, for prismatic ones at 610 nm (Fig. 6,7). The sign of the focusing contrast changes in these points, i.e. the regions of minimum and maximum light intensities interchange their positions near the glide bands. In the cadmium sulphide specimens studied the excitation of green luminescence by red light was also observed. This can be accounted for by two-step optical transitions through the levels of deep centres /7/. It is worth noting that in the vicinity of the glide bands on which the said minima of light scattering and focusing are observed the two-step excitation of green luminescence also gained a distinct polarization anisotropy: when the red light is polarized normally to the glide bands the green luminescence near them is much more intensive than far from them or than in the case of parallel polarization in the region of 700 nm. The two-step excitation spectra have broad maxima correlating with the spectra of dislocation scattering.

The photoluminescence of dislocation traces observed in the green part of the spectrum in cadmium sulphide single crystals deformed plastically at low temperatures (77 K and below) exhibits a number of interesting features /8/. For specimens with basal slip the dislocation traces are perpendicular to the 6th-order axis, whereas their luminescence is polarized in a direction unusual for CdS; at
Fig. 6 - Low-angle scattering spectra in CdS with basal dislocations. The light polarization is perpendicular to the glide bands.

± 45° to the above axis. In this case some portions of traces emit in the polarization +45°, some in the polarization -45°. The emission polarization is unstable. From time to time the polarization of separate portions changes abruptly from one angle to another. In a polarizer oriented at 45° this process manifests itself as scintillation of separate portions of traces (Fig. 8). The jumps occur quicker, the higher the specimen temperature or excitation intensity.

Fig. 7 - Low-angle scattering spectra in CdS with prismatic dislocations. The light polarization is perpendicular to the glide bands.

Fig. 8 - Fragment of the time dependence of the polarized luminescence intensity of a dislocation trace portion at 6 K.

Our experimental data show that in the subsystems of dislocations and point defects arising in the course of plastic deformation there take place collective interactions between them resulting in the regularization of their space distribution and, in some instances,
in the cooperative behaviour of defects with time. These factors materially affect the optical properties of plastically deformed $A_{11}P_{11}$ crystals. The angular and spectral oscillations of the wave field pattern as well as of the intensity of emergent light fluxes point rather conclusively toward a high periodicity in the distribution of dislocation density in the direction normal to the glide bands. In this case the observed diffraction effects may be easily explained on the basis of the dynamical theory of X-ray diffraction in medium with periodic distribution of dielectric constant /9/. When the angle between wave vector and glide bands satisfies the Bragg condition

$$2d \sin \theta = m \lambda$$

(where $d$ is the structure period, $\lambda$ - the wavelength, $m$ - the integer), the waves scattered by individual glide bands are in phase and reinforce each other. As a result, the periodic wave field is formed in a plastically deformed crystal at the expense of the interference between incident and scattered waves. In the first order of diffraction ($m=1$) two standing waves are formed in the crystal. The first has its amplitude peak in the maximum of dislocation density, the second in the minimum. At the Bragg condition the crystal transmittance increases in the spectral region of a high dislocation absorption owing to the second wave which propagates in the regions of reduced dislocation density. This is similar to the anomalous transmission of X-rays, i.e. to the Borrman effect /9,10/. The distances between glide bands measured experimentally ($4\mu m$) and the angles at which the periodic wave field and anomalous light transmission are seen ($\pm 4.5^0$) fit the Bragg condition well.

Spectral oscillations of the secondary beam intensity (Fig. 5) are also well described by the Bragg condition when the structure period is about $4\mu m$ (the order of diffraction $m=25$). This makes possible the assumption that the secondary beam is formed by waves that originate in separate glide bands and then interfere. The appearance of this beam is easily comprehended if to assume that the like screw components of dislocations fill up rather closely and homogeneously the layer along the glide band. On different sides of this layer the long-range shear stresses are of opposite sign /11/ and this involves an abrupt change in the orientation of principal axes of the indicatrix of refractive index due to photoelasticity /12/. Moreover, in the layer of like screw dislocations the plane of light polarization may rotate on account of the screw structure of the dislocation core. Owing to these two factors the polarization of the light flux behind the glide band does not coincide with the principal axes of the indicatrix of refractive index. Therefore, the light flux decomposes into two ones polarized along the new principal axes of the indicatrix. One of them polarized parallel to the axis of the former refractive index runs along the original beam while the other polarized along the second principal axis is refracted because of the difference in the refractive indexes and thus forms a secondary beam. The deviation of the secondary beam intensity minimum from the null point of photoelastic constants ($555\ nm$) is indicative of a considerable value of the polarization plane rotation by the cores of screw dislocations. This in turn points to a high spatial dispersion in the dislocation core /13/.

The reasons for the formation of periodic distribution of dislocation density are not clear yet. Mutual repulsion of dislocations of like sign in the direction normal to the slip planes is acknowledged to be one of the possible reasons of static nature /11/. Since the dislocations are forced to occupy a limited volume, the elastic energy of the system of mutually repulsing dislocations approaches its
minimum at a periodic distribution of the latter. This fact will primarily affect the distribution of screw dislocations that can glide in any direction.

Undoubtedly, the reasons of dynamic nature, for example, the formation of an extremum of the shear stresses at a specified distance from the developing dislocation row may be of great importance too.

The minima of dislocation focusing and scattering at 580 nm and 610 nm, the polarization anisotropy of the two-step exitation of green luminescence as well as temporal and orientation features of the photoluminescence of dislocation traces evidence that plastic deformation involves a regularization of spatial and angular distributions of anisotropic point defects. Thus, the redistribution and reorientation of deep centres in the vicinity of the dislocation rows give rise to the anisotropic inhomogeneity of dielectric constant whose maximum corresponds to the peak in the polarizability of deep centres (700 nm). If these centres are concentrated near edge dislocations in the region of tension, their contribution to polarizability compensates that of elastic strains, and with their absolute values equal, one would expect the minimum of scattering as well as the inversion of focusing contrast in one of the spectrum points. This, actually, was observed in the experiment.

The polarization anisotropy of green photoluminescence of dislocation traces indicates that in the low-temperature motion of dislocations clustering and orientation of donor-acceptor pairs takes place. The cooperative forces (probably, of dipole character) acting between them make all the pairs adopt either of two equivalent orientations. The light excitation shifts part of the pairs to the neutral state, this may increase the frequency of the dipole jumps from one orientation into another.

Thus, on account of collective interactions in the subsystems of dislocations and point defects a number of rather interesting and unexpected features of the optical properties of plastically deformed cadmium sulphide crystals has been revealed.

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