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Abstract

Full Text

Physics

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Absorption of Light by CdS Crystals*

(Presented by Academician G. S. Landsberg, 7 I 1957)

It was shown in the work of E. F. Gross and co-workers ([1]) that in the absorption spectrum of CdS single crystals at 4.2° K a series of narrow bands is observed. These bands are located at the edge of continuous absorption and are interpreted in ([1]) as exciton bands.

In studying the influence of temperature stresses on the absorption spectra of CdS crystals ([2]), a strong variability of the spectral bands was found in passing from one specimen to another. In connection with this, a study was undertaken of the spectra of single crystals of the hexagonal modification of CdS at 20.4° K in polarized light. The specimens studied were obtained by evaporation of Cd and S in an Ar atmosphere, by evaporation of Cd in an atmosphere of $H_2S + H_2$, and by evaporation of CdS (recrystallization)**.

The specimens had different faceting and surface quality. Among them were perfect smooth platelets, crystals with fine and coarse striation, and twins with oblique extinction relative to the twin axis. The thickness of the specimens varied from 1 to 100 μ. The spectra were studied with the aid of a polarization microprojector ([3]), owing to which an image of the crystal, magnified 20 times, was projected onto the slit plane of the spectrograph. Using an Iceland spar crystal, two components of the spectrum were simultaneously recorded on the photographic plate, corresponding to polarization of the light vector along the hexagonal axis c and perpendicular to it. By moving the cryostat, it was possible to bring any point of the crystal onto the slit and to obtain the spectrum from a region of dimensions 0.1×0.0015 mm^2.

Figure 1a gives a scheme of the absorption spectrum of a CdS crystal in polarized light. The spectrum contains 10 comparatively narrow bands, situated in the interval 20400–20600 cm^{-1}, and several broader bands situated on a continuous background, from which they are separated only in the thinnest crystals. In the shorter-wavelength region, near 21100 cm^{-1}, continuous absorption begins***.

First of all, attention is drawn to the weak polarization of bands Nos. 9 and 10, which are clearly manifested in both components of the spectrum. Indeed, in a crystal with symmetry C_{6v}^4, all narrow-band intrinsic absorption should be strongly polarized, i.e., each line may be present either in the component parallel to c, or in the component perpendicular to c; moreover, the absorption should also be strongly polarized

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Fig. 1

Figure 1: Fig. 1

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** All the specimens studied were prepared in the laboratory of I. B. Mizetskaya. We take this opportunity to express our gratitude to I. B. Mizetskaya and V. D. Fursenko, who kindly placed them at our disposal.

*** In the strong absorption near 21100 cm^{-1} it has not yet been possible to detect structure, and it appears continuous. However, it is possible that study of still thinner specimens at 4.2° K will permit this absorption region to be studied in greater detail.

near vacant sites and any other defects,* having an axis of symmetry of the third order. Therefore bands Nos. 9 and 10 can be associated only with the absorption of atoms of the near-surface layer or with asymmetric defects located in the bulk. The second important feature is that, not only in different specimens, but also in different regions of one and the same specimen, the intensity of all bands Nos. 1–10, their polarization, and their position in the spectrum may be different. Therefore in Fig. 1a the frequency limits are indicated within which, in different cases, the most characteristic bands Nos. 4 and 10 are located; the satellites of these bands usually shift together with them.

Fig. 1

It is especially convenient to observe changes in the group of bands Nos. 8–10, which is distinctly located in the *c*-component; this makes it possible, from photographs, to judge their changes more confidently. Figure 1b shows a typical change of bands Nos. 9 and 10 in the *c*-component within a single specimen; the relative intensity was estimated visually. Next to it is a diagram of the crystal, indicating the points for which the spectra are given. Similar changes occur with bands Nos. 8–10 in the component perpendicular to *c*, and also with other bands (Nos. 4, 6, 7), but the strong background makes their study difficult without objective photometry. Bands Nos. 1, 2, 3, 8 are completely absent in the spectra of a number of crystals. It was possible, for example, to project a region of the crystal in whose spectrum band No. 2 appeared "dotted," disappearing and reappearing along the height of the slit.

Although all bands, except Nos. 9 and 10, are as a rule strongly polarized and are absent in the *c*-component, in several specimens the strongly weakened bands Nos. 4 and 8 were observed in the *c*-component of the spectrum; in the spectrum of individual regions of one of the specimens the depolarization of band No. 8 was extremely strong.

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The sharp variability of bands Nos. 1–10 in going from crystal to crystal, and especially within a single crystal, in our opinion serves as evidence that they cannot be connected with the intrinsic absorption of the ideal lattice (including exciton absorption) and are due to the presence of defects.

In order to establish the localization of the defects responsible for bands Nos. 9 and 10, various specimens were etched for 2 minutes in HCl 1 : 1. After etching, the intensity of band No. 10 increased sharply, and it proved to be very strong even in those parts of the crystal

* By defects are understood any disturbances of the regular crystalline structure, including vacancies, impurity centers, dislocations, etc.

in whose spectra it had been absent before etching. Consequently, the absorption in this case is, at least in part, localized in the surface layer, which is distorted by etching. It may be thought that the defects responsible for absorption in the remaining, strongly polarized bands are distributed in the bulk.

It is natural to suppose that the proximity of bands Nos. 1–10 to continuous absorption is due to the fact that they are caused not by excitation of weakly bound electrons belonging to defects, but by transitions of electrons belonging to ions of the host substance adjacent to the defects (i.e., that they are to a certain degree analogous to the α- and β-bands in the spectra of alkali-halide crystals ([4])). If this is in fact the case, then one may expect the existence, in the infrared region of the spectrum, of impurity-absorption bands with which some of bands Nos. 1–10 are associated and of which they are "shadows." It is also of interest to determine whether there exists a correlation between the photoelectric sensitivity of the crystals and the intensity of bands Nos. 1–10 in the spectra. Work in this direction is now being carried out.

As was noted by S. I. Pekar in discussing the present work, the absorption experiments are also not contradicted by the supposition that some of bands Nos. 1–10 are due to transitions into exciton states which are forbidden in an ideal lattice and become allowed as a result of the disturbance of its regularity by internal deformations. Consideration by the method of group theory shows that only a few types of deformation in the lattice C_{6v}^4 can lead to the polarization of absorption bands Nos. 1–7 observed experimentally in the plane perpendicular to the c axis; the simplest of these is bending of the plane perpendicular to c, with preservation of the second-order axis. However, in view of the irregular change in the relative intensities of individual bands in going from crystal to crystal, and especially within a single specimen, the entire set of bands Nos. 1–10 cannot be attributed to the allowance of forbidden transitions.

The luminescence spectra of CdS single crystals were also investigated at a temperature of 20.4° K*. Both green and blue luminescence were observed ([5]). Without dwelling on a detailed description of all details of the spectrum and its temperature dependence, we shall note one important feature of it. In the spectrum of a very thin crystal (the choice of small thickness was dictated by the need to reduce reabsorption), narrow bands of blue luminescence were observed,

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corresponding in frequency to absorption bands 1, 2, 4, 8-10; this agrees with the work ([6]), which points to the coincidence of a number of luminescence bands at 77° K and absorption at 4.2° K. In addition, longer-wavelength bands of blue luminescence ([7]) and a series of broader bands of green luminescence are visible in the spectrum. In our opinion, the simultaneous presence in the luminescence spectrum of the six indicated bands is evidence that they are connected not with exciton states, but with electronic transitions near defects. Indeed, at 20.4° K ($kT = 14$ cm^{-1}), luminescence can occur simultaneously from the system of levels (1-10), situated in an interval reaching ~ 200 cm^{-1}, only in the event that an equilibrium distribution does not have time to be established in the excited state, which appears extremely improbable for exciton states. This is possible only if these levels belong to spatially separated defects.

In conclusion it is necessary to note that, in the broad bands Nos. 11-17, present in both components of the spectrum, we were unable to find differences, and they proved to be practically unchanged in all specimens and under

* The luminescence light, excited through a black Wood' s filter, before reaching the slit of the spectrograph, passed through the specimen (so-called photography in transmitted light).

in all regions. We hope that subsequent intensity measurements will make it possible to separate completely the absorption due to the presence of defects from the absorption of the ideal lattice and to elucidate the structure of the edge of intrinsic absorption in a CdS crystal.

We take this opportunity to express our sincere gratitude to Academician of the Academy of Sciences of the Ukrainian SSR V. E. Lashkarev and Corresponding Member of the Academy of Sciences of the Ukrainian SSR A. F. Prikhot' ko for discussion of the present work and for a number of valuable comments during its execution.

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