Singularities of exciton absorption in cadmium sulfide at low temperatures

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Exciton absorption in cadmium sulfide was investigated in the temperature range 6–65 K. A decrease was observed in the integral exciton absorption for picosecond pulses of white light.

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A number of studies[1–3] have revealed certain singularities in the spectra of exciton absorption of semiconductors at low temperatures, namely, a decrease in the integral exciton absorption with decreasing temperature, and a dependence of the absorption coefficient on the sample thickness and on the concentration of the impurities and defects. These phenomena point to an important role played by dissipation processes (scattering of excitons by phonons, by the crystal surface, by impurities, or by defects) in the mechanism of exciton absorption of light and indicate apparently the need for a polariton approach when it comes to describing absorption in the exciton region of the spectrum[1)].[1–4] It can be assumed that there exists one more manifestation of the effect of polariton transparency (nonstationary absorption of light)—pulses of white light of low intensity, but short enough, can propagate with lower losses in the case of resonant excitation of excitons in the semiconductor. In fact, if the pulse duration is shorter than the characteristic relaxation time of the excitons, then a polariton packet of such a duration, excited in the crystal, will propagate apparently with smaller absorption than in the case of longer pulses or continuous radiation.

In the present study we investigated exciton absorption in plate-like CdS crystals 1–50 μm thick following irradiation of the samples with both continuous white light from an incandescent lamp and with ultrashort pulses (USP) of white light, with duration not exceeding 5 psec. To obtain the USP of white light we used the effect of the broadening, in D_2O of the spectrum of high-power USP of radiation of wavelength 1.06 μm (a train of 10–20 pulses of duration 5 psec, total train energy 0.1 J).[5] The energy of the train of the USP of white light was about 0.01 J and was distributed in a very wide spectral band. According to[5], the duration of the white-light pulse was less than the duration of the exciting pulse—5 psec in our case. Control measurements with the aid of an FÉR-2 photoelectronic recorder with a time resolution 30 psec have shown that the duration of the USP of white light does not exceed the resolution time of the apparatus.

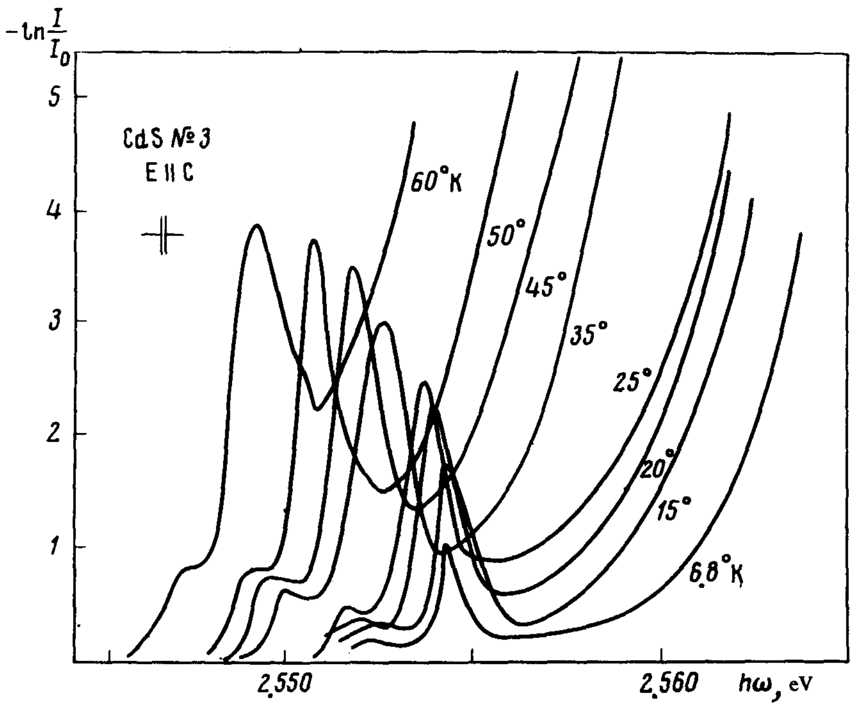

FIG. 1. Absorption spectra of CdS crystal No. 3 for continuous white light.

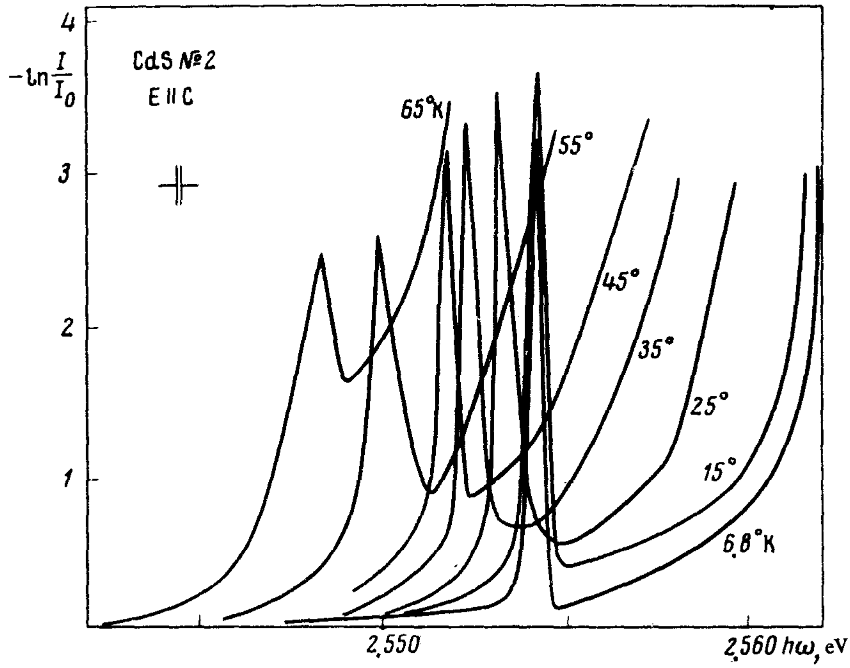

FIG. 2. Absorption spectra of CdS crystal No. 2 for continuous white light.

The linearly polarized white light was aimed on a sample placed in vacuum on the cold finger of a helium cryostat. The radiation passing through the crystal was directed to the entrance slit of the spectral instrument of the DFS-24 spectrometer, with a reciprocal linear dispersion 4.5 Å/mm and was used to study the absorption of the continuous white light; an autocollimation chamber UF-90 with diffraction grating of 1200 lines/mm was used to investigate the absorption of both the pulsed and the continuous white light, the reciprocal linear dispersion of such a spectral instrument was approximately 6 Å/mm.

Figures 1 and 2 show the results of the investigation of the exciton absorption in two CdS samples of different purity at $T=6.8$–65 K using continuous white light. (The investigations of the transmission spectra in the region of the exciton-impurity complex lines have shown that in both crystals there are relatively few acceptor impurities, while crystal No. 3 is much purer with respect to donor impurities that crystal No. 2). It is seen from the absorption spectra that the integral exciton absorption decreases with decreasing temperature in both crystals, but in the purer sample this effect is much more strongly pronounced.

The exciton absorption of USP of white light was investigated in the thinner sample No. 1. Excitation with continuous white light produced in this sample a rather negligible polariton transparency, apparently because of the smaller thickness of the crystal.[3] Figure 3 shows the absorption spectra of the crystal for continuous white light and USP of white light. Comparison of these spectra shows a considerable de-

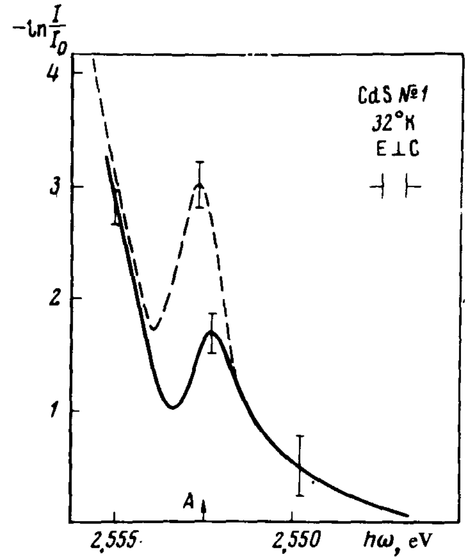

FIG. 3. Absorption spectra of CdS crystal No. 1 for continuous white light (dashed) and USP of white light (solid).

crease of the exciton absorption in the case when the spectrum was plotted with the aid of USP of white light.

The results point to a dependence of the exciton absorption of light in CdS on the temperature in the region 7–65 K. This is apparently due to the decreased role of the phonon channel of exciton relaxation with decreasing temperature. In pure crystals we observed a decrease of the integral exciton absorption by a factor of 10 when the temperature changed from 65 to 7 K. In the samples with impurities and defects, this phenomenon was much weaker, probably because in such crystals a larger role is played in dissipation processes by the scattering of the excitons by impurities and defects. In exactly the same way scattering by the crystal surface becomes more substantial than scattering by phonons in the case of excitons in thinner samples, as is manifest in the weaker dependence of the integral exciton absorption on the temperature. Nonethless, in such thin crystals we were able to observe a decrease of the exciton absorption by using picosecond pulses of white light. This indicates probably that when USP of white light is used it becomes possible to exclude not only the phonon mechanism of the relaxation, but also dissipative processes. At 32 K, the integrated exciton absorption for USP of white light is half as large as for continuous white light. The absorption for the other sectons of the spectrum remains unchanged in this case.

We note that the low intensity of the white-light pulses—to obtain the transmission spectrum in the region of normal density of the photographic film it was necessary to carry out several dozen exposures—makes it possible to exclude effects of saturation[6] and of self-induced transparency[7] as causes of the observed decrease of the exciton absorption.

Further investigations of the polariton-transparency effect in semiconductors, using USP of white light, can help explain the role of different processes in exciton nonradiative recombination and determine the characteristic times of these processes.

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[1)]This effect was called in[3] polariton induced transparency.

[1]F.N. Kreĭngol'd and V.L. Makarov, Fiz. Tverd. Tela (Leningrad) **17**, 472 (1975) [Sov. Phys. Solid State **17**, 297 (1975)].
[2]J. Voight, Phys. Stat. Sol. (b) **64**, 546 (1974).
[3]A. Bosacchi, B. Bosacchi, S. Franchi, Proc. 13th Int. Conf. Phys. Semicond., Rome, 1976, p. 841.
[4]A.S. Davydov and A.A. Sericov, Phys. Stat. Sol. (b) **56**, 351 (1973).
[5]D.K. Sharma, R.W. Yip, D.F. Williams, S.E. Sugamuri, and L.L.T. Bradley, Chem. Phys. Lett. **41**, 460 (1976).
[6]A.F. Dite, V.B. Timofeev, V.M. Fain, and E.G. Yashchin, Zh. Eksp. Teor. Fiz. **58**, 460 (1970) [Sov. Phys. JETP **31**, 245 (1970)].
[7]F. Bruckner, V.S. Dneprovskiĭ, V.S. Fokin, D.G. Koshchug, M.V. Kraevskiĭ, Sh.M. Ok, E.K. Silina, and E.A. Zhukov, Proc. 13th Int. Conf. Phys. Semicond., Rome, 1976, p. 845.