

Exciton-exciton interaction and absorption bistability in CdS at low optical excitation levels

V. S. Dneprovskii, V. I. Klimov, E. V. Nazvanova, and A. I. Furtichev
M. V. Lomonosov Moscow State University

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A sharp decrease has been observed in the transmission near the fundamental absorption edge of CdS crystals (80 K) at low levels of the excitation by the beam from an argon laser (below 10 kW/cm²). An absorption bistability has been detected under the same conditions. Analysis of luminescence spectra reveals that the rapid change in the nonlinear absorption which occurs in the crystals during such an excitation is a consequence of an exciton-exciton interaction.

The optical properties of semiconductors change substantially during the resonant excitation of excitons.^{1–6} The broadening of an exciton absorption line during a collective interaction of excitons may lead to an increase in the absorption near the fundamental absorption edge of the semiconductor. The nonlinear increase in the absorption of the exciting light which occurs is of particular interest for the development of bistable optical devices with an intrinsic feedback in which an S-shaped dependence of the absorption coefficient on the particle concentration is caused by a dynamic relation between an increase in the number of excited particles and their recombination.⁷ A resonatorless absorption bistability has been observed in CdS crystals cooled to liquid-nitrogen and liquid-helium temperatures at relatively high excitation levels^{2,3,8,9} (about 1 MW/cm²). The nonlinear increase in the absorption near the fundamental absorption edge of the semiconductor CdS was apparently caused primarily by a renormalization (narrowing) of the band gap during excitation of a dense plasma.

In this letter we report measurements of the transmission and luminescence spectra of CdS crystals (at 80 K) during resonant excitation by pulses from an argon laser near the absorption edge. The nonlinear increase in the absorption of the semiconductor, the rapid switching of the bistable element on and off, and the particular features of the luminescence spectra are explained in terms of a collective exciton-exciton interaction.

In the experiments we use lamellar CdS samples 3–5 μm thick, cooled to liquid-nitrogen temperature. For excitation we use an ILA-120 argon laser ($\lambda = 488 \text{ nm}$), whose cw output is converted into pulses of tunable length by means of a modulator. The polarization of the light is parallel to the optic axis of the crystal. Light with a wavelength $\lambda = 488 \text{ nm}$ excites the samples near the fundamental absorption edge. The diameter of the spot of focused light on the surface of the crystal is 30 μm . We simultaneously measure the shape of the pulse incident on the sample and that transmitted through it at various excitation levels and the luminescence spectra of the crystals, with the help of a DFS-24 diffraction spectrometer. Figure 1a shows oscillo-

scope traces of an incident laser pulse (intensity I) and of the pulse transmitted through a CdS sample (I_T), with a length of $7 \mu\text{s}$, along with the hysteretic $I_T(I)$ curve. The rapid switching of the bistable element (the time required for switching on or off does not exceed $1 \mu\text{s}$: the time resolution of the photodetectors used) is caused by a nonlinear change in the absorption near the fundamental absorption edge of the semiconductor. The rapid nonlinear change in absorption may be caused by the following processes: 1) a broadening of an exciton absorption line upon an increase in the exciton concentration; 2) the formation of excitonic molecules (biexcitons) during two-photon absorption of light; 3) a decrease in the width of the band gap (a renormalization) by an amount determined by the sum of the exchange and correlation corrections to the energy of the interacting electrons and holes of high density. The features of the luminescence spectra (Fig. 2) suggest that an exciton-exciton interaction is the dominant process. At the excitation levels used, a luminescence band P arises. The energy position of this band (below the energy of a free exciton by an amount equal to the binding energy of the exciton), the superlinear dependence of the intensity of this band on the pump intensity, the difference between this lineshape and the lineshape of the first phonon repetition of A excitons (the A -LO band arises at low excitation levels and is asymmetric, having a high-energy tail), and the appearance of stimulated emission (Fig. 2) in the long-wavelength wing of the band allow an explanation of this band in terms of an inelastic exciton-exciton interaction: $ex + ex \rightarrow h\nu + (e + h)$ ($e + h$ is an electron-hole pair). Estimates show that the rapid switching of the element occurs at an exciton concentration $\sim 10^{16} \text{ cm}^{-3}$. It can thus be assumed that, at least in the initial stage of the switching, the increase in absorption is not due to a renormalization of the width of the band gap. The absorption edge must "be shifted" 10–13 meV for the switching of the particular element. Such a shift, caused by a renormalization of the band, can be achieved in CdS at a carrier-density $n \geq 10^{17} \text{ cm}^{-3}$ (Refs. 8, 10, and 11). An absorption component due to the formation of

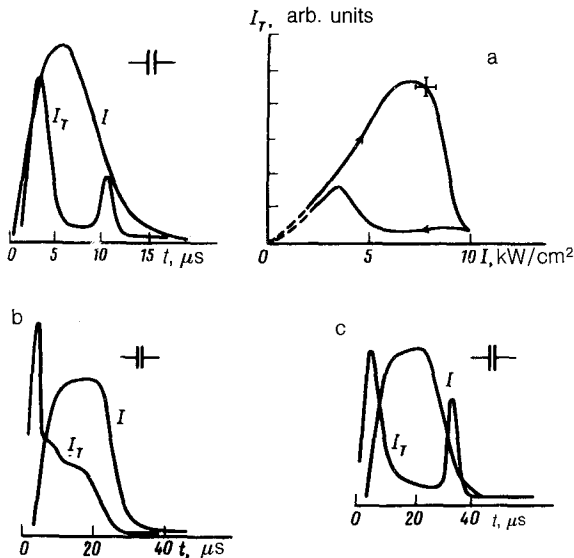


FIG. 1. Oscilloscope traces of the pulse from an argon laser ($\lambda = 488 \text{ nm}$) incident on the CdS crystal and of the pulse transmitted by the crystal, for various pulse lengths and intensities; curve of $I_T(I)$ for a pulse $7 \mu\text{s}$ long (I is the intensity of the incident light, and I_T that of the transmitted light).

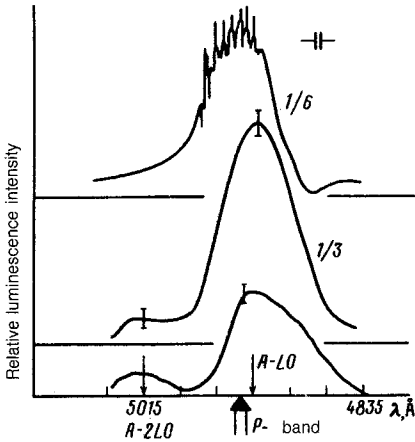


FIG. 2. Luminescence spectra of CdS crystals (80 K) for various levels of the excitation by the beam from an argon laser: 4, 9, and 15 kW/cm².

biexcitons in CdS at liquid-nitrogen temperature is improbable: The binding energy of the exciton is low.

The effect of the thermal shift of the absorption edge on the switching regime can be seen by increasing the length of the exciting pulse by a factor of several units¹⁾ (Fig. 1, b and c). We see a competition between two types of nonlinear changes in the absorption: a "fast" change due to the collective interaction of excitons and a "slow" change due to the heating of the semiconductor by the laser light. The rapid decrease in the transmission (Fig. 1b) is apparently due to an exciton switching mechanism in the sample, which is heated to a temperature higher than for the conditions corresponding to Fig. 1c (at a relatively low pump intensity, a longer time elapses before the light reaches the intensity required for the exciton switching mechanism, and the crystal is heated to a greater extent). As the crystal is heated, there is apparently a change in the absorption, and the spectrum of the nonlinearly absorbing sample shifts into a region in which a feedback arises.²⁾ The element is turned off more rapidly (!) than it is turned on as the excitation level is raised (Fig. 1c). This effect is a consequence of the exciton nonlinearity mechanism. This circumstance can also be explained in terms of a change in the absorption spectrum as the sample is heated to the level at which a feedback occurs (an S-shaped dependence of the absorption coefficient on the exciton concentration).

A fast nonlinear process of an exciton nature can apparently arise at low excitation levels even in microscopic crystals of the semiconductor CdS, in which a quantum size effect occurs.^{12,13}

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¹⁾For the particular CdS crystals used (80 K), we detected a steady-state bistable regime associated with a thermo-optic nonlinearity (with a thermal shift of the absorption edge during excitation of the samples by millisecond pulses or the cw output from an argon laser).

²⁾The transmission spectrum can also be changed (the working point of the nonlinear system with feedback can be chosen) by rotating the polarization vector of the light around the optic axis of the crystal.

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