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BIEXCITON IN CdS SPECTRUM. INDUCED RADIATIVE DECAY OF EXCITON-IMPURITY COMPLEXES

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It is shown that the emission bands produced by strong optical excitation in CdS crystals at low temperatures, and ascribed by some authors to biexcitons and to collisions between them, are actually connected with induced raidative decay of electron-impurity complexes (EIC) with emission of long-wave acoustic phonons.

A number of recent papers report experimental observation of biexcitons in the recombination emission spectra of CdS and CdSe [1 - 3]. When these crystals are exposed to strong laser radiation at sufficiently low temperatures, several intense emission bands are produced, and two of them, designated M and P_M, were attributed by the authors to radiative decay of biexcitons [1] and to elastic collisions between them [2]. The main arguments favoring the biexciton concept are based on the faster-than-linear dependence of the intensities of the corresponding bands on the laser pumping, and also on an analysis of the band shapes. It must be borne in mind, however, that a significant restructuring of the radiative-recombination spectrum is still not an unambiguous consequence of a bimolecular process in the system of interacting excitons, and may have a more trivial cause, namely induced radiative processes.

The purpose of the present communication is the show that the CdS-spectrum singularities attributed to biexcitons are due to induced radiative decay of exciton-impurity complexes (EIC) on the neutral donors and acceptors, accompanied by emission of long-wave acoustic phonons.

We have investigated different CdS crystals, some of which were highly purified and contained not more than $(1 - 5)\times10^{15}$ cm^{-3} donor and acceptor impurities responsible for the EIC lines I_2 and I_1. We have also investigated a number of crystals with different degrees of compensation for the acceptor impurities, and the intensity ratio of I_2 and I_1 differed as a result of this compensation by an order of magnitude or more.

The luminescence was excited with a pulsed nitrogen laser (peak power $\sim$ 1.6 kW, repetition frequency 100 Hz). The spectral resolution was worse than 0.2 Å (10^{-4} eV).

Figure 1 shows the behavior of the luminescence spectrum of a high-purity CdS sample at T = 1.3 with increasing pumping (observation at 45° to the excited surface, dimension of laser spot on crystal $\sim$ 0.2 mm). Here M and P_M are the bands so designated in [1] and [2], A and A-LO denote the bands of direct radiative exciton annihilation and with emission of an LO phonon, I_2 and I_1 are the phononless EIC lines, and P is the emission band due to inelastic collisions of two free excitons [4, 5]. We confine ourselves to the properties of the bands M and P_M.

1. In uncompensated metals, the P_M band is clearly observed in the spontaneous-luminescence spectra even at low excitation levels (Fig. 1, spectra a and b), and constitutes the Stokes acoustic wing of the phononless line I (transitions with emission of long-wave acoustic

TA phonons) [6, 7]. The maximum of this wing coincides with the P_M line position given in [2]. The faster-than-linear growth of the P_M intensity begins at pumps $\sim 5\times10^4$ W/cm^2. At a definite excitation geometry and at a definite value of the pump (cf. infra) the P_M band intensity may predominate in the spectrum in uncompensated crystals. In crystals in which the acceptors are compensated (by approximately one order of magnitude), the P_M band appears and increases faster than linearly only at high excitation levels ($\sim 10^5$ W/cm^2). In strongly compensated crystals there is no P_M band up to excitations levels capable of damaging the crystal.

2. The M band, in contrast to P_M, is practically nonexistent in the spontaneous-luminescence spectra (Fig. 1, spectrum a). Its appearance, accompanied by a superlinear growth of intensity, occurs in a narrow dynamic range of pumps (10^3 - 10^4 W/cm^2). With further increase of the pump, the intensity of the M line increases linearly, like I_2 and I_1, up to total saturation of the intensity (saturation of the EIC concentration).

We point out first of all the close connection between the lines I_2 and M. Like the lines I_1 and P_M, their polarizations are equal, 95° (dipole-allowed transitions with E C). In a magnetic field H C, the splittings of I_2 and M practically coincide ($\delta E \sim 0.4$ meV at H = 40 kOe).

Furthermore, most importantly, the I_2 and M bands have equal "chemical" shifts of their positions in the spectrum (within 1.5 meV), owing to the different natures of the shallow donors (e.g., I, Cl, and Al) responsible for the EIC of I_2 [8].

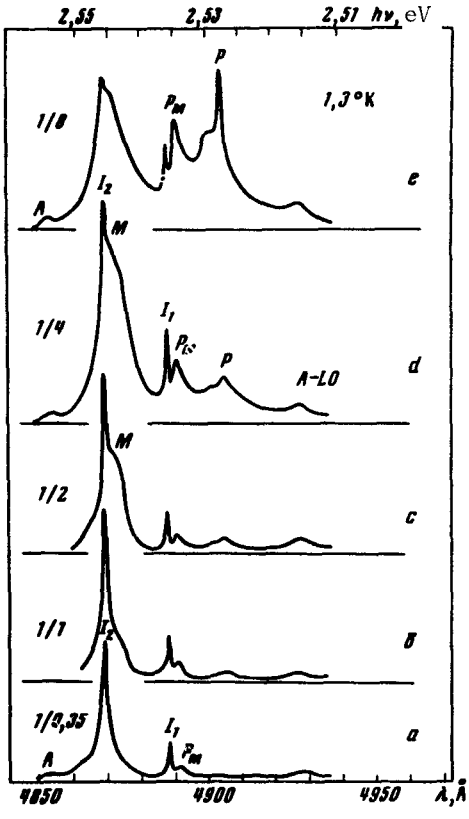

Fig. 1. Luminescence spectrum of CdS at T = 1.3°K and at different optical-excitation power densities (W/cm^2): a) 3.5×10^3, b) 1.4×10^4, c) 4.2×10^4, d) 2.1×10^5, e) 5.4×10^5.

With increasing temperature and at a fixed pump, the M band does not attenuate, as one might expect as a result of thermal dissociation of the biexcitons. Up to 20°K, its maximum intensity remains practically constant, only its width increases (see Fig. 2).

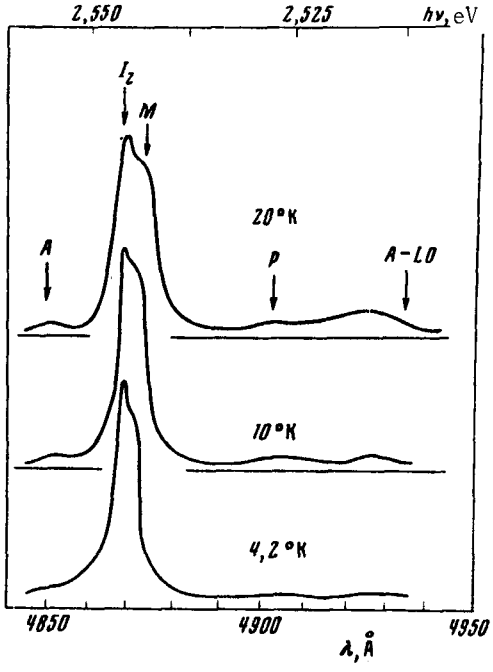

Fig. 2. Luminescence spectrum of CdS in the region of the I_2 - M structure at a fixed pump $\sim 10^4$ W/cm^2 and at different temperatures. Crystal with compensated acceptors.

We note finally that the shape of the M band changes from sample to sample, and the dependence of its intensity on the pump is not a quadratic function of the free-exciton density, as would follow from the concept of the bimolecular process.

Thus, the M-band properties listed above contradict the concept of a biexciton or of a complicated multiexciton-impurity complex (two or more excitons bound to a center). We assume that the M band, like P_M, is of electron-vibrational origin and results from radiative decay of the EIC line I_2 with emission of long-wave acoustic phonons.

3. We show in conclusion that the observed faster-than-linear dependences of the intensities of the bands M and P_M on the pumping are the conseuqence of induced radiative decay of the IEC lines I_2 and I_1, with emission of long-wave acoustic phonons. Figure 3 shows the transformation of the luminescence spectrum at a fixed pump ($G \sim 2\times10^5$ W/cm^2), the only variable being the length ℓ of the strip (of width ~ 20 μ) into which the exciting radiation is focused with a cylindrical lens. The observation is carried out in this case along the direction of the varied length ℓ and in a direction perpendicular to the exciting beam ($\ell \parallel c$, where c is the crystal axis). When ℓ is varied, the intensities of the M and P_M bands vary in faster-than-linear fashion. This is most clearly seen by comparing the P_M band with the phononless line I_1, which increases linearly with ℓ and experiences no reabsorption up to $\ell \sim 200$ μ. We note that under these conditions the I_2 line is strongly reabsorbed already starting with $\ell \sim 75$ μ. At pumps up to $G \sim 2\times10^5$ W/cm^2 we observed no induced luminescence for the phononless lines I_2 and I_1 (i.e., there is still no population inversion in the two-level system comprising the IEC and the neutral donor or acceptor).

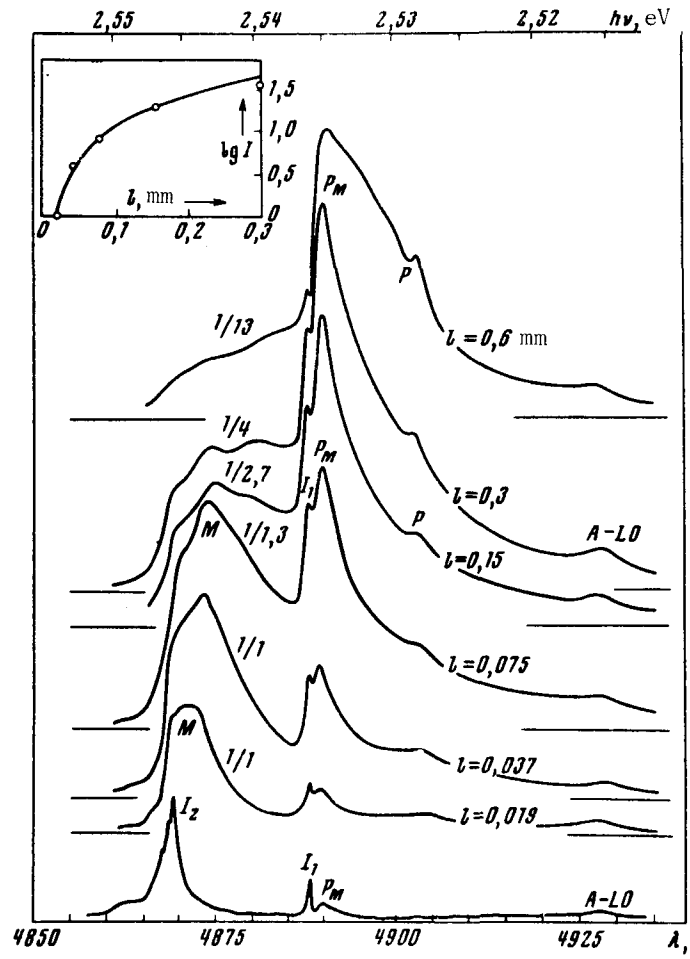

Fig. 3. Distribution in the luminescence spectrum of CdS at T = 1.3°K and at a fixed pump $\sim 2\times10^5$ W/cm^2 vs. the length ℓ of the pumped region of the crystal. The lower figure shows the spontaneous luminescence spectrum produced by excitation with a mercury lamp.

The insert in Fig. 3 shows the dependence of the intensity at the maximum of P_M (λ = 4889.9 Å) in coordinates log I and ℓ, from which it is easy to determine the gain [9, 10]. The continuous curve is the result of an approximation at a gain α = 240 cm^{-1}. In the M-band region (at λ = 4875 Å) the gain reaches $\alpha \sim 300$ cm^{-1}. Such large gains in the M and P_M bands are the result of the gigantic probabilities of the radiative decay of the IEC (on the order of $w_R \gtrsim 10^9$ sec^{-1} [11, 12]. The low induced-luminescence threshold in the M and P_M bands is connected with the fact that the corresponding optical transitions are of electron-vibrational origin and correspond to the known four-level system of laser physics.

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MAGNETIC PARAMETRIC RESONANCE IN CONDUCTORS

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A new resonance effect — magnetic parametric resonance (MPR) — is predicted for semiconductors placed in a magnetic field $\vec{H}$ that varies periodically with time and in a constant electric field $\vec{E}$ perpendicular to $\vec{H}$.

We consider an electron gas placed in an alternating homogeneous magnetic field $\vec{H} \parallel OZ$ and a constant electric field E. The value of H depends periodically on the time, with a frequency γ,

$$H = H_0(1 + a\cos\gamma t), \quad a = \text{const}. \tag{1}$$

The magnetic field is homogeneous in the conductor if the skin-layer depth $\delta\gamma$ at the frequency γ greatly exceeds the mean free path ℓ and the sample thickness d, i.e.,

$$\delta_\gamma = \left(\frac{c^2}{2\pi\sigma\gamma}\right)^{1/2} \gg \ell, d; \qquad \sigma = \frac{Ne^2\ell}{mv}. \tag{2}$$

Here σ is the static conductivity, N the concentration, ℓ the absolute value of the charge, m the mass, and v the velocity of the conduction electrons. In addition, we assume that the amplitude E_γ of the induced electric field is small in comparison with the constant electric field E

$$E_\gamma \sim H_0 a \frac{\gamma\delta_\gamma}{c} \ll E. \tag{3}$$

It is well known that in a constant and homogeneous magnetic field the motion of an electron in a plane perpendicular to the vector $\vec{H}_0$ is analogous to the behavior of a linear oscillator with constant cyclotron frequency $\Omega_0 = eH_0/mc$. The electron trajectory is described in this case by the integrals of motion $\varepsilon = \text{const}$ and $p_z = \text{const}$, where ε is the energy and p_z is the projection of the electron momentum on the magnetic-field direction. In the case of an alternating magnetic field (1) (neglecting the field E_γ (3)), the trajectory is determined by the same conserved quantities, in contrast to the situation considered in [1, 2]. In other words, although the parameter of the system (the cyclotron frequency) varies with time, the system remains conservative. The motion of the electron in a plane perpendicular to $\vec{H}$ is more complicated and contains not one frequency Ω_0, but an entire set of frequencies $\omega_e = \Omega_0 - n\gamma$ (where n is an integer).

It is known that the energy absorbed by electrons in a constant electric field has a maximum when the natural frequency is zero, i.e., when

$$\Omega_0 = n\gamma. \tag{4}$$

In metals and semiconductors, this resonance becomes "smeared out" by collisions between the electrons and scatterers. Resonance is observed if the collision frequency ν is the smallest quantity

$$\nu \ll \Omega_0, \gamma. \tag{5}$$

Thus, if the conditions indicated above are satisfied, a resonance effect should be observed in the electron-hole plasma of a solid; we call it magnetic parametric resonance.

The time-averaged power Q absorbed per unit volume of the conductor equals