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DEGENERACY OF EXCITON GAS FOLLOWING POWERFUL OPTICAL EXCITATION IN CdS CRYSTAL

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Submitted 3 August 1972
ZhETF Pis. Red. 16, No. 5, 291 - 295 (5 September 1972)

The question of producing non-equilibrium exciton gas of high and controllable density is still a pressing one. We have attempted to determine by direct measurement the occupation numbers of the excitons produced in CdS crystals by powerful single-photon excitation. At the maximum pumps attained in our experiment ($P \sim 3 \times 10^6$ W/cm^2), these numbers turned out to be $n \gtrsim 1$.

To determine the exciton occupation numbers, we used a method consisting of measuring the gain in the region of the induced exciton-phonon luminescence [1]. The exciton occupation numbers were then estimated directly from the values of this coefficient.

It was shown earlier [1] that an important factor limiting the accumulation of the exciton in the band during optical pumping in CdS crystals is their induced radiative decay. This decay can be limited by decreasing the dimensions of the excited region of the crystal. In our experiment, one of the dimentions of this region was determined by the absorption coefficient for the exciting light ($L_1 \sim 1/\alpha \sim 10^{-4} - 10^{-5}$ cm), while the other two dimensions, which determined the area of surface excitation, could be controlled under the conditions of the experiment ($L_2 \lesssim 10^{-2}$ cm, $L_3 \gtrsim 10^{-3}$ cm). Using a tunable pulsed dye laser [2] delivering a maximum power density $\sim 10^6$ W/cm^2 at a generation line width 2 - 3 cm^{-1} and a monopulse duration 10 nsec, we excited "monochromatic" excitons in a wide spectrum of their motion energy $\varepsilon(\vec{k})$ (up to $\varepsilon(\vec{k}) \sim 3\hbar\omega_{LO}$, where $\hbar\omega_{LO}$ is the energy of the optical phonon). We used high-grade CdS crystals in which the donor and acceptor densities did not exceed 5×10^{14} cm^{-3}, and the edge-dislocation density was less than 10^2 cm^{-2}. The investigations were performed at T = 77°K, when radiative decay processes of only free excitons were manifest in the spectrum [3]. Figure 1 shows the exciton spontaneous emission spectrum at a low excitation power level, $\sim 10^{-2}$ W/cm^2 (A corresponds to direct radiative annihilation of the excitons and A - LO corresponds to the "red" boundary of the exciton-phonon transitions with emission of LO phonons). The induced exciton-phonon luminescence corresponding to a maximum linear dimension of the excited region of the crystal on the order of 10^{-2} cm occurs at single-photon excitation power densities close to 10^4 W/cm^2 (curve 2 of Fig. 1). Up to 10^6 W/cm^2, processes of radiative decay of free excitons predominate in the induced-luminescence spectrum (curves 3 - 7). There was no generation of light, since the induced luminescence had a single-pass character[1)].

[1)] Lasing on exciton-phonon transitions in CdS was first observed in [4,5].

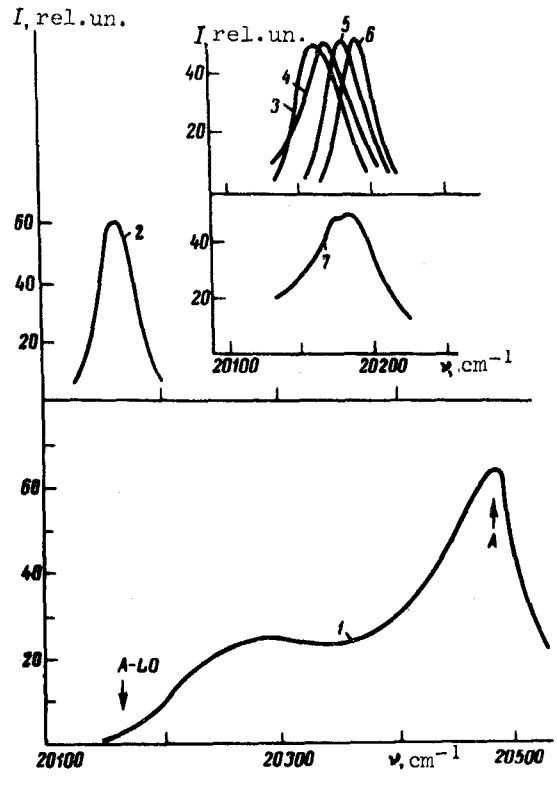

Fig. 1

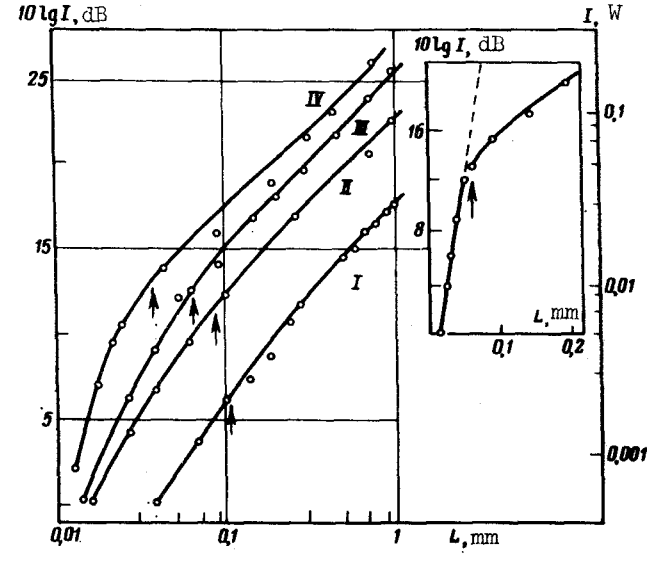

Fig. 2

Fig. 1. Spectra of spontaneous (curve 1) and induced (curves 2 - 7) exciton-phonon luminescence. Curves 3 - 6 correspond to different motion energies of the excitons generated with the aid of a tunable laser (tuning range 4 meV, pump power 5×10^5 W/cm^2).

Fig. 2. Intensity at the maximum of the induced exciton-photon luminescence vs. length of excited region of the crystals. Curves I - IV correspond to excitation powers 4×10^4, 1.5×10^5, 5×10^5, and 3×10^6 W/cm^2, respectively.

The intensity I of the exciton-phonon luminescence can be written in the form

$$I \sim \{[n(\varepsilon)(\nu + 1)]/[n(\varepsilon) - \nu]\}\{\exp(\alpha L) - 1\}, \qquad (1)$$

where ν and $n(\varepsilon)$ are the occupation numbers of the LO-phonons and of the excitons, L is the length of the homogeneously excited part of the sample in the observation direction, and α is the gain (the negative coefficient of exciton phonon absorption) and is equal to

$$\alpha = \alpha_0 \frac{\varepsilon^{3/2}}{(\varepsilon - \hbar\omega_{LO})^2}[n(\varepsilon) - \nu], \qquad (2)$$

where ε is the energy reckoned from the "red" boundary of the exciton-phonon spectrum. By varying the length L of the pumped sections we can plot $I = I(L)$, from which, using (1), we determine the gain α at $\alpha L > 1$. Figure 2 shows the corresponding plots, in coordinates logI and logL, obtained at different levels of ruby-laser second-harmonic excitation (the luminescence spectrum is curve 7 of Fig. 1). The figure shows also the scale of absolute intensity values in watts, corresponding to measurements of the luminescence powers within the limits of its diffraction divergence. By way of an example, the I(L) plot for curve III is shown in Fig. 2 in coordinates logI and L. At small values of L this plot is a straight line agreeing well with (1), and at large L the

dependence becomes sublinear. The arrows in Fig. 2 indicate the lengths at which the dependence deviates from linearity. The experimentally determined criterion of applicability of expression (1) turned out to be $\alpha L \lesssim 5 - 7$. The physical reason for such a sublinear dependence is understandable and is connected with the saturation of the gain in the strong induced-luminescence field. Indeed, the growth of the gain slows down when the rate of the induced radiative decays of the excitons becomes equalized, and the rate of their nonradiative annihilation begins to predominate (cf., e.g., [6, 7]). We note that the density of the power of the induced exciton-phonon luminescence, referred to the end face of the crystal, amounts to 10^4 W/cm^2 for the regions marked by arrows in Fig. 2, yielding a value $\tau_{exc} \sim 10^{-9}$ sec which agrees with the nonradiative lifetime of the exciton in CdS (T = 77°K) [8, 9].

The gains for each of the curves of Fig. 2 were determined from the slopes of the linear sections of the plots of logI against L, and turned out to be $\alpha_I = 2.8 \times 10^2$, $\alpha_{II} = 5 \times 10^2$, $\alpha_{III} = 1.3 \times 10^3$, and $\alpha_{IV} = 4 \times 10^3$ cm^{-1}.

Using these values, we can determine from (2) the occupation numbers of excitons having in the band a motion energy close to the maximum of the luminescence curve. For the case of the maximum gain ($\alpha = 4 \times 10^3$ cm^{-1}, curve IV of Fig. 2), this energy was assumed to be 3 meV, since the maximum of the induced luminescence is located in this case 2 meV away from the boundary of the exciton-phonon spectrum, and is determined with accuracy ±1 meV. (It is also assumed that the position of the red boundary does not depend on the pump level.) The lower bound obtained in this manner for the occupation numbers is $n \sim 1$. The gain α can also be estimated independently by determining experimentally, from the power of the induced exciton-phonon luminescence, the photon occupation numbers q_{ph}, which are in turn connected with the gain α by the relation $q_{ph} \simeq \exp(\alpha L)$. At a maximum pump 3×10^6 W/cm^2 and a length $L \sim 2 \times 10^{-3}$ cm of the excited region, the luminescence power density measured in a solid angle 8×10^{-3} rad and in a spectral interval $\Delta\nu \sim 40$ cm^{-1} amounted to $P \sim 2 \times 10^3$ W/cm^2. This corresponds to photon occupation numbers $q_{ph} \simeq 50$, whence $\alpha \simeq 2 \times 10^3$ and $n \gtrsim 1$, which agrees with the estimate given above.

Thus, the exciton occupation numbers at maximum pumps turned out to be $n \gtrsim 1$, indicating that the exciton gas is degenerate. A very important question here is the actual exciton energy distribution. Occupation numbers $n \gtrsim 1$ at a distance $\varepsilon \sim 3$ meV from the bottom of the band do not contradict a quasi-equilibrium exciton distribution with a chemical potential $\mu \sim 0$.

Unfortunately, the true exciton distribution established in the band under the conditions of the maximally realized pumps remains unknown. Special experiments on the excitation of excitons with different motion energies in the band did not yield unambiguous information on the character of the exciton distribution. An argument favoring the assumption that the distribution does not deviate greatly from equilibrium is the fact that, independently of the motion energy of the excitons excited by the tunable laser, the induced exciton-phonon luminescence occurred at frequencies corresponding to exciton localization near the bottom of the band. However, the presence of a certain shift of the luminescence maximum, within a range 4 meV (see Fig. 1, curves 3 - 6), in accord with the variation of the exciting-light frequency, indicates that the distribution may deviate from equilibrium.

We are continuing research on the distribution of the excitons under degeneracy conditions.

The authors thank V.L. Broude, I.B. Levinson, and E.I. Rashba for useful discussions and valuable remarks.

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EXPERIMENTAL DETERMINATION OF THE ANGULAR DEPENDENCE OF THE ELECTRON REFLECTION COEFFICIENT

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Submitted 24 July 1972
ZhETF Pis. Red. 16, No. 5, 295 - 298 (5 September 1972)

The dependence of the conductivity σ of filamentary whiskers and plates on the temperature T has been measured in a number of experiments (cf., e.g., [1 - 3]). The measurement results were interpreted on the basis of the assumed temperature dependence of the coefficient q of specular electron reflection from the surface of the metal. Yet it is difficult to expect a significant dependence of q on T at the low temperatures used in the cited studies.

We shall show that the results of these investigations can be naturally attributed to the well-known appreciable dependence of q on the angle ϕ of incidence of the electrons on the surface of the plate (see the figure) in the region of small ϕ, when the electron reflection is close to specular. Moreover - and this is particularly important - from the form of $\sigma(T)$ we can reconstruct the function $q(\phi)$ (see [4] on this subject; $q(\phi)$ was investigated theoretically in [5 - 7]).

To simplify the exposition, let us examine in greater detail the case of a plate that is thin in comparison with the electron mean free path ℓ. If the electrons moving at an angle ϕ were not to experience any collisions in the volume, they would be scattered in each collision with the surface with a probability $1 - q(\phi)$ and would continue their path without scattering with a probability $q(\phi)$. The path negotiated by them without scattering is (see the figure, d is the plate thickness)

$$\lambda(\phi) \equiv \frac{d}{\phi} + \frac{d}{\phi} q(\phi) + \frac{d}{\phi} q^2(\phi) + \ldots = \frac{d}{\phi(1 - q(\phi))}. \qquad (1)$$

Collisions inside the volume limit the average path ℓ_{eff} traversed without scattering to a value on the order of ℓ, so that it can be assumed that ℓ_{eff} is of the order of $\lambda(\phi)$ if $\lambda(\phi) < \ell$, and $\ell_{eff} \sim \ell$ if