Radiation of electron-hole condensate in CdS single crystals

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We have observed, for the first time ever, resonant interaction between states responsible for the *P*–*Q* and I_1–*LO* luminescence lines. This serves as additional confirmation of the plasma origin of the lines *P* and *Q*. The concentration of the electron-hole pairs is estimated at 1.8×10^{18} cm^{-3} (*P* line) and 1.2×10^{19} cm^{-3} (*Q* line).

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When single-crystal CdS is strongly excited at a temperature 4.2 K, there appears in its photoluminescence spectrum the so-called *P* line, which is attributed in[1] to recombination radiation of a non-equilibrium electron-hole plasma (EHP). The same mechanism was proposed also to explain the *Q* line.[2]

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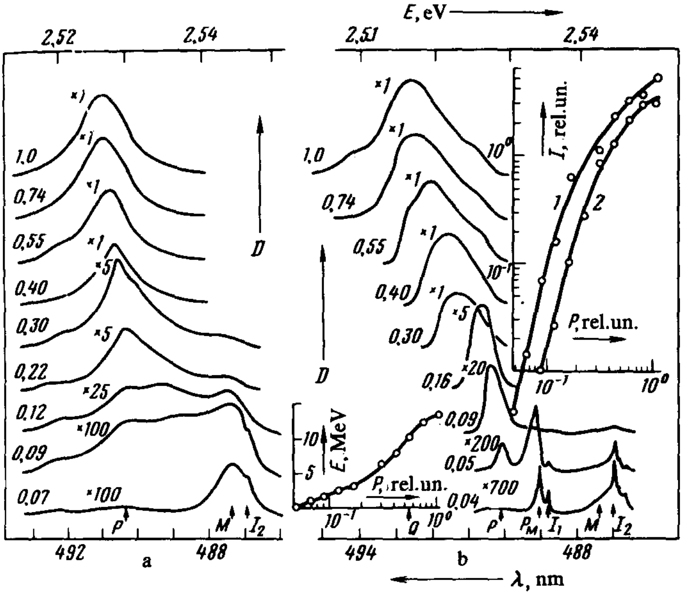

FIG. 1. Microphotographs of the luminescence spectra of CdS single crystals. The pump intensities in unit of P_{max} are marked on the left for each spectrum. Upper insert–dependence of the radiation intensity of th P-Q lines (for b) and the P line (for a) on the pump. Lower insert–shift of energy position of the maximun radiation of the P-Q lines as a function of the pump.

In this paper we present additional proof of the plasma origin of the lines P and Q the behavior of which can be described by the electron-hole condensate (EHC) model

We investigated strip-like CdS single crystals grown from the gas phase. Th impurity density was 10^{14}–10^{15} cm^{-3}. The photoluminescence was excited with an LGI 21 laser at a maximum pump intensity $P_{max}=2$–3 MW/cm^2 and a sample temperatur 4.2 K. The photoluminescence spectra were photographed with a spectrograph havin a linear dispersion 7 Å/mm. Microphotographs of spectra of various samples ar shown in Fig. 1.

For the purest samples, in which the donor density determined from the impurit photoconductivity does not exceed 2×10^{14} cm^{-3}, and there is no radiation from exci ton-impurity complexes (EHC) that are connected with the neutral acceptors, th radiation of the P line predominates in the photoluminescence spectrum at pum intensities $>0.2P_{max}$. In the pump range $(0.1$–$0.6)P_{max}$, the maximum radiation of th P line is shifted approximately 2 meV towards longer wavelengths, and the line itse broadens insignificantly. The dependence of the intensity of the radiation of the line P-Q [for Fig. 1(b)] and of the line P [for Fig. 1(a)] on the pump are shown on th upper insert of Fig. 1(b) by curves 1 and 2, respectively.

In samples whose photoluminescence spectra obtained at low excitation level contain rather intense EIC radiation due to neutral acceptors (I_1 line), the dependenc

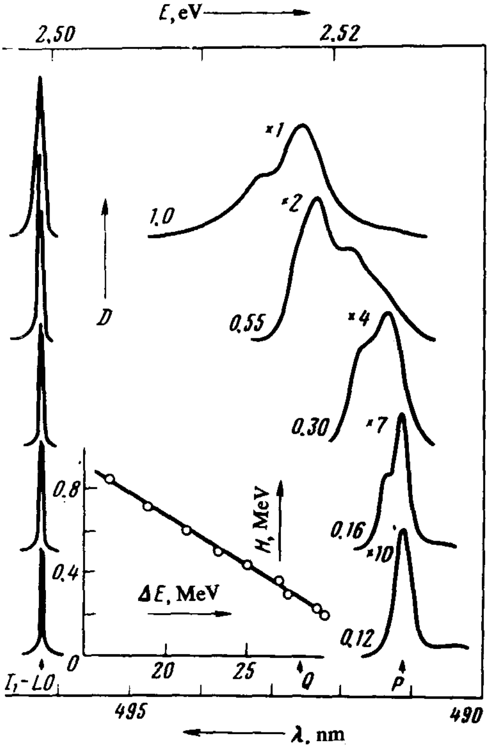

FIG. 2. Microphotographs of the luminescence spectra of CdS in the presence of resonant interaction. Lower insert–dependence of the half-width of the I_1-LQ line (H) on the distance between the lines I_1-LQ and P-Q.

f the intensity of the radiation of the phonon replica (the line I_1-LO) on the pump as an anomalous character. Prior to the appearance of the P line in the photolumıescence spectrum, the intensity of I_1-LO increases with increasing pump. Once the P ne appears, the intensity of the I_1-LO line increases sharply (by several orders of ıagnitude), duplicating the growth of the intensity of the P-Q lines, although the EIC adiation of I_1 is already saturated at these pump intensities. It should be noted that in ome samples the intensity of the I_1-LO line even exceeds the intensity of the P-Q lines Fig. 2). When the Q line approaches I_1-LO, the half-width of the latter increases ecause of the shift of the long-wave wing towards lower energies (see Fig. 2 and its ısert).

This behavior of the I_1-LO line points to the presence of resonant interaction etween the states corresponding to the lines P-Q and I_1-LO, an interaction similar to ıe plasmon-phonon resonance in the IR absorption spectra.[3] Such a resonant interction can exist only if the lines P-Q and I_1-LO correspond to real energy levels in the rystal. Neither exciton-exciton nor exciton-electron collisions, which were proposed ɔ explain the lines P[4] and Q,[5] can produce such a resonant interaction, for in this ase the lines P and Q correspond to polariton states. In addition, the smoothness of ıe transition of the radiation of the P line into the Q line both with respect to their nergy position and with respect to their intensity [see the lower and upper inserts of 'ig. 1(b)], as well as the identical character of their interaction with the I_1-LO, indi-

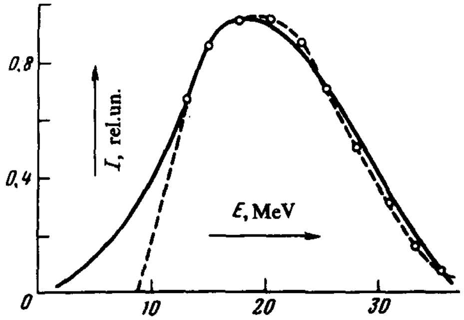

FIG. 3. Line shape of EHC emission in CdS. Solid curve–experiment results, the points and the dashed lines show the results of calculations for $n_c=0.9\times10^{18}$ cm^{-3} and $T=30$ K.

cate that these lines are due to similar energy states of the crystal. The first real state of the crystal, with a broad continuous spectrum, which takes part in the resonant interaction, can be due only to EHP. The second real state is due apparently to an exciton-phonon complex.[6] It should be noted that the presence of phonons is a mandatory condition for resonance, since the plasma screens the Coulomb interaction between the electrons and the holes, and therefore the EIC in the plasma are spatially separated.

According to[1,7], the EHP energy in CdS has a miminum at an electron-hole pair concentration $n_c\sim10^{18}$ cm^{-3}. When EHP is produced, the electrons and holes tend to condense in EHC with an electron-hole pair concentration equal to n_c. The electron-hole pair density inside the condensate should not change at constant temperature with increasing excitation level. Raising the pump level can lead only to an increase in the number or dimensions of the EHC. The EHC emission line shape and the energy position of the line depend only on n_c, and should likewise not depend on the pump level. This was actually observed for the case of the purest samples for which the half-width and energy position of the P line, at pump intensities $\geqslant 0.6P_{\max}$, do not depend on the excitation level.

A theoretical analysis of the EHC emission line shape, carried out in accordance with formula (7) of[1] for different concentrations and temperatures, has shown that the best agreement with the experimental results occurs at values $n_c=0.9\times10^{18}$ cm^{-3} and $T=30$ K (Fig. 3).

The strong broadening and shift of the P line towards longer wavelengths, and the transition of this line into the Q line [Fig. 1(b)] point to an increase in the density of the electron-hole pairs inside the condensate. The energy position and half-width of the Q line, obtained at maximum pumps, corresponds to an electron-hole pair concentration $n=1.2\times10^{19}$ cm^{-3}. This increase of the electron-hole pair concentration inside the condensate is apparently due to the fact that the entire pump volume is completely filled with EHC and further increase of the pump should lead to an increase of the density.

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