Exciton-plasmon interaction in nonequilibrium electron-hole plasma of CdS crystals

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A broadening and splitting of the free-exciton line and a shift of the Stokes component toward lower frequencies with an increase in excitation intensity and temperature were observed. The obtained data indicate that an exciton-plasmon interaction occurs in cadium sulfide crystals.

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As shown for the first time in Refs. 1 and 2, the recombination-radiation (RR) spectrum is rearranged as a result of increase of the density of free excitons (FE) and free carriers (FC)—new emission lines, whose origin is attributable to a radiative Auger process due to the interaction of FE, FE, and FC, appear on the long-wave side of the FE line. However, the studies of the behavior of the RR spectra as a function of the excitation intensity and temperature, which were performed by us, do not allow us to interpret the existing anomalies as the result of inelastic interactions of FE and FC.

A N_2 laser with a pulse duration of 10^{-8} sec and up to 100-Hz repetition frequency was used to excite the RR spectra. The spectra were recorded by means of a double monochromator with a dispersion of 4.5 Å/mm and a photoelectric stroboscopic recording system with a resolution time of at least 10^{-9} sec. The range of investigated temperatures T was 1.5–300 K. CdS samples with $N_D \lesssim 10^{15}$ cm^{-3} were used in the experiment.

Figure 1 shows the RR spectra measured at different temperatures in the excitation interval of 2×10^{18} to 5×10^{23} cm$^{-2}\cdot$sec^{-1}. As seen from an analysis of the spectra, when the excitation increases at a constant temperature, the FE line is split and the Stokes component is continuously shifted toward longer wavelengths with increasing excitation. Figure 2 shows the experimental dependences of the shift $\Delta\hbar\omega = E_x - E_-$, where E_x and E_- are the energy of the FE and of the new line, respectively. We can see that as the excitation intensity I increases, the magnitude of the shift is $\Delta\hbar\omega \sim I^{1/4}$, and for a constant excitation level $\Delta\hbar\omega$ increases with increasing temperature. We should point out that it was assumed in Ref. 3 on the basis of a study of the RR spectra at $T = 77$ K that the recombination in the E_- band may be caused by the radiative decay of the FE with energy deficit expended on the excitation of collective oscillations—plasmons—in the electron-hole gas. Our experimental data make it possible to confirm this viewpoint. The plasmon frequency in the case of a semiconductor with a strong electron-phonon interaction is determined from the equations[4]

$$\epsilon(\omega) \equiv \epsilon_\infty + \left(\frac{\epsilon_0 - \epsilon_\infty}{\omega_T^2 - \omega^2}\right)\omega_T^2 - \frac{\omega_P^2}{\omega^2} = 0, \quad (1)$$

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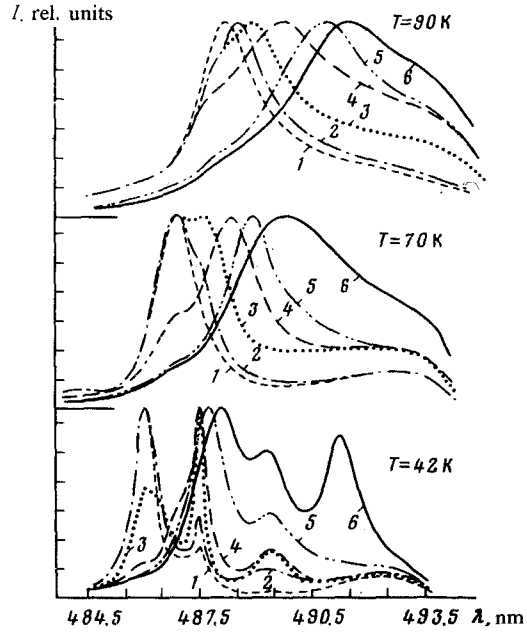

FIG. 1. RR spectra of CdS crystals, polarization $E \perp c$, $t_{del} = 5 \times 10^{-9}$ sec, and $I \times 10^{18}$ cm^{-2}·sec^{-1}: 1, 2; 2, 20; 3, 2×10^2; 4, 5×10^3; 5, 5×10^4; 6, 5×10^5.

$$\omega_{\pm}^2 = \frac{1}{2}\left(\omega_L^2 + \bar{\omega}_P^2\right) \pm \frac{1}{2}\left[(\omega_L^2 + \bar{\omega}_P^2)^2 - 4\bar{\omega}_P^2\omega_T^2\right]^{1/2},$$
$$\omega_L^2 = \omega_T^2 \epsilon_0/\epsilon_\infty, \quad \bar{\omega}_P^2 = \frac{4\pi n_e e^2}{\epsilon_\infty m_e^*}, \quad \omega_P^2 = \frac{4\pi n_e e^2}{m_e^*}, \tag{2}$$

where ω_L and ω_T are the frequencies of the longitudinal and transverse phonon, n_e is the density of free carriers, ϵ_∞ and ϵ_0 are the high-frequency and low-frequency dielectric constants, respectively, and m_e^* is the effective mass of FC, thus

$$\Delta\hbar\omega = E_x - E_- \equiv \hbar\omega_- . \tag{3}$$

To compare the observed $\Delta\hbar\omega$ values and those calculated from Eq. (2), it is necessary to determine the density of FC obtained in the experiment. Assuming that thermodynamic equilibrium exists between the FC and the FE with allowance for the law of mass action, the functional dependence between n_x—the density of FE—and n_e can be determined for each point of the investigated temperature interval. The change in the FE binding energy because of screening effects was taken into account in the expression for the static plasma screening.[5,6] The value of n_x was determined from the generation rate for $k = 10^5$ cm^{-1}—the absorption coefficient—and $\tau_x = 2 \times 10^{-9}$ sec—the FE lifetime (provided that the exciton recombination is the primary particle-destruction channel). It must be noted that the FE lifetime was not determined in our

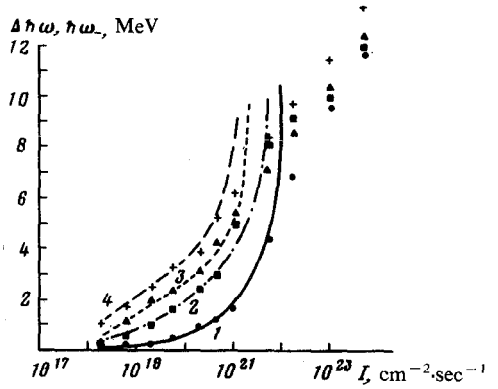

FIG. 2. Dependence of $\Delta\hbar\omega = E_x - E_-$ on excitation intensity I: 1, 42 K; 2, 60 K; 3, 70 K; 4, 90 K.

experiments; therefore, the value $\tau_x = 2\times10^{-9}$ sec was obtained for the best matching of the curve, which was calculated from Eq. (2), with the experimental value of $\Delta\hbar\omega$ for a specific temperature; it was assumed that τ_x does not depend on the excitation intensity, which can be validated by the experimental fact that no change is observed in the slope angle of the $I_x \sim I$ dependence, where I_x is the intensity of the FE line. It must be noted that the value $\tau_x = 2\times10^{-9}$ sec determined in this paper is in satisfactory agreement with the value $\tau_x = (0.5\text{–}2.5)\times10^{-9}$ sec, which was determined experimentally[7] in a study of the decay of exciton luminescence due to excitation by picosecond laser flashes. The τ_x value was assumed constant in the calculations of the other temperatures. The results of the calculation are shown in Fig. 2. The discrepancy between the theory and the experiment observed at $I = I_{cr}(T)$ is attributable to the radiation in the L band. We can see in Fig. 2 that the experimental variation of $\Delta\hbar\omega$ as a function of the excitation intensity and temperature occurs as predicted by Eq. (2). Because of the absence of a theory for the exciton-plasmon interaction process, it is much more complicated to explain the ratio of the intensities in the spectrum. The presence of emission bands of different shape and width makes it impossible to draw a conclusion about the relative intensity of the RR bands in the spectrum from an estimate of the amplitudes of the RR at the maxima of the corresponding lines. An additional experimental difficulty in determining the relative intensity of the RR lines is attributable to the sharp change in the absorption coefficient near the FE line.

In conclusion, we must note that the spectral location of the line for $I \sim 10^5$ W/cm^2 coincides with the spectral location of the exciton-electron interaction line.[2] However, since the one-particle response of the plasma is suppressed at large FC densities in the electron-hole plasma,[4] we cannot regard the E_- line as a result of an exciton-electron interaction. Moreover, the line of exciton-electron interactions at the outset must be separated from the FE line by an amount $\Delta\hbar\omega_{x-e} = \frac{1}{2}\frac{m_x^*}{m_e^*}K_B T$, where m_x^* is the FE mass, whereas we can observe experimentally that $\Delta\hbar\omega$ varies from zero as $I^{1/4}$.

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