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THE ELECTRON-HOLE PLASMA IN DIRECT II-VI COMPOUNDS

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We compare the experimental gain and absorption spectra of an electron-hole plasma (EHP) in CdS with our lineshape calculations and deduce from the fit the plasma temperature T_p and density n_p. The nucleation theory predicts the existence of small clusters only (≈ 20 e-h pairs per cluster). Consistently with this result, the experiments indicate, that no liquid like equilibrium phase is reached in the plasma because of the short lifetime of the e-h pairs.

I. Calculation of the EHP gain spectrum

The gain spectra of highly excited semiconductors with a direct gap are strongly influenced by many-body effects[1,2,3,6].In [4,5] we developed a theory of the gain spectrum, containing the complex self-energy corrections of the single particle energies, i.e. the energetic shifts and the collision broadening of the electron and hole correlations, giving rise to an excitonic enhancement. The gain spectrum is determined by the imaginary part of the dielectric function

$$\varepsilon_2(\omega) = M^2 C \sum_k \mathrm{Im}\, P_{eh}(k,\omega - i\varepsilon) \ , \qquad (1)$$

where P_{eh} is the polarization function. P_{eh} obeys a Bethe-Salpeter-equation (BSE), which describes the multiple scattering of e-h pairs due to their interaction via the screened Coulomb potential [4]. This equation can be rewritten in terms of a BSE for the T-matrix. The screened Coulomb potential which appears in this equation is only weakly momentum and frequency dependent, because for an e-h plasma in CdS the Fermi momentum k_F is smaller than the inverse Thomas-Fermi length k_{TF} . Under these conditions the T-matrix is also a slowly varying function and can approximately be pulled out of the integral in the BSE [5]. The resulting polarization function is given by

$$P_{eh}(k,\omega - i\varepsilon) \simeq \frac{P^o_{eh}(k,\omega - i\varepsilon)}{1 + \sum_{k'} V_s(k-k',0)\, P^o_{eh}(k',\omega - i\varepsilon)} \ . \qquad (2)$$

The polarization bubble P^o_{eh} is

$$P^o_{eh}(k,\omega - i\varepsilon) \simeq \frac{1 - f_e\left(e_e(k)\right) - f_h\left(e_h(k)\right)}{\omega - \varepsilon_e(k) - \Sigma_e\left(k,\omega - e_h(k)\right) - \varepsilon_h(k) - \Sigma_h\left(k,\omega - e_e(k)\right)} \ , \qquad (3)$$

where $\varepsilon_i(k)$ is the free particle energy, while $e_i(k) = \varepsilon_i(k) + \mathrm{Re}\,\Sigma_i(k_F, e_i(k_F))$ is the renormalized RPA self-energy. The excitonic enhancement is given by the denominator in eq.(2) in which for simplicity only the statically screened Coulomb potential is used. Note, that we take here the full excitonic enhancement into account, and not only its real part as in [4], improving the agreement between theory and experiment. The Coulomb potential is screened by the optical phonons and by the electronic excitations. We approximate the latter contribution by plasmon-pole approximation, in which mixed phonon-plasmon eigenmodes appear. For further details we refer to [5].

II. Calculation of the e-h cluster formation kinetics

The growth and decay of clusters which are formed at the inset of the plasma instability at low temperature are determined by the master equation for the concentration F(n,t) of clusters with n e-h pairs:

$$\frac{\partial F(n,t)}{\partial t} = j_{n-1} - j_n \quad ; \quad n > 1 \tag{4}$$

with the current $j_n = g_n F(n,t) - (e_{n+1} + (n+1)/\tau_n) F(n+1,t)$. (5)
g_n is the generation rate, which describes the collection of an exciton by a cluster with n e-h pairs, e_n is the rate with which excitons are evaporated out of this cluster and n/τ_n is the radiative decay rate. The exciton concentration can be obtained from the conservation law:

$$\sum_{n=1}^{\infty} n\, F(n,t) = G(t) - \sum_{n=1}^{\infty} n\, F(n,t)/\tau_n \quad , \tag{6}$$

where G(t) is the laser generation rate. In [7], we have solved eq.(4) - (6) for exciton pulses with various halfwidths in GaAs. In Figs. (1a) and (1b) we show the cluster distribution for CdS for excitation pulses with 7 ns and 10 ps temporal halfwidth, respectively. Obviously the size of the clusters is very small. More detailed information about the temporal evaluation of e.g. the average cluster size or the chemical potential μ will be given elsewhere.

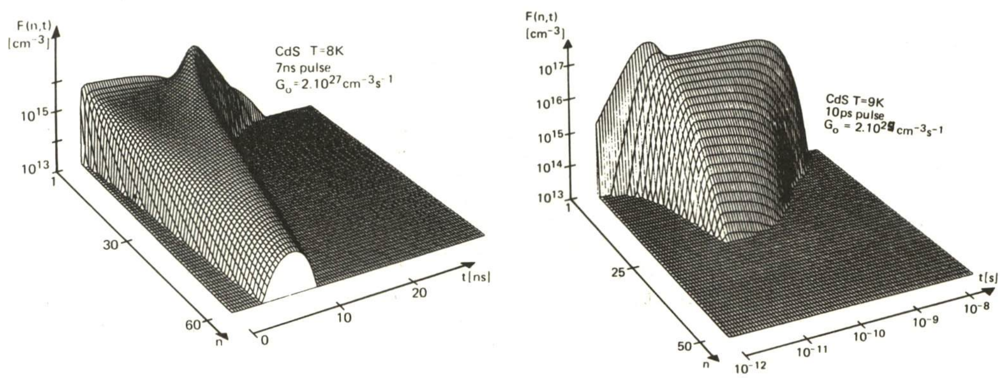
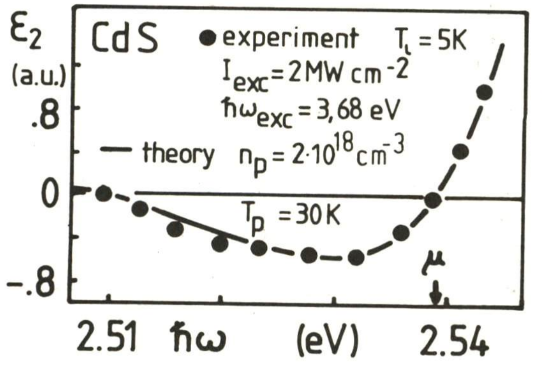


Fig.1 Concentration function F(n,t) for ns and ps excitation pulses, respectively

III. Experimental results and their comparison to theory

We investigate the reflection and transmission spectra of CdS platelets as a function of the excitation intensity I_{exc}, the lattice temperature T_L and the energy of the exciting photons $\hbar\omega_{exc}$. We use the two beam method (TBM). The details of the experiment are described in [8], with the difference that now the excitation source is a narrow, tunable dye-laser. The polarization of the electric field in the exciting and the probe beams is perpendicular to the crystallographic c-axis.

Figure (2) gives an experimentally observed gain spectrum together with a calculated one according to the theory described above. The parameters deduced are $n_p = 2 \cdot 10^{18} cm^{-3}$ and $T_p = 30$ K . Almost identical parameters are obtained from a fit with the empirical "no-k-selection rule" [8]. This justifies to a certain extent the use of this rule for the determination of plasma parameters. However it should be mentioned, that this model yields a wrong behaviour for $\hbar\omega >> \mu$ and in the vicinity of the reduced gap E_g' [6].



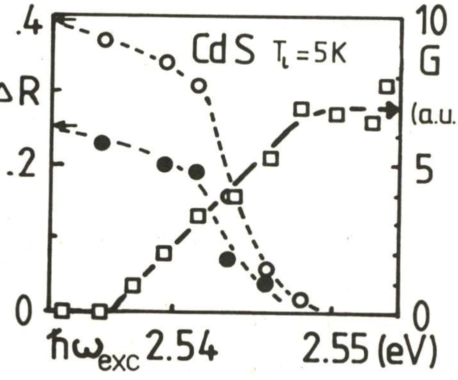


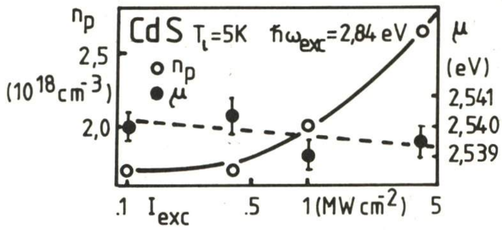
Fig.2 Measured (●●) and calculated (——) gain spectrum of the EHP in CdS

Fig.3 The integral G (□), (I_{exc} = 0.4 MW/ cm^2) over the gain spectrum and the difference ΔR between the reflection maximum and minimum of A(○) and B(●) excitons (I_{exc} = 4 MW/cm^2) as a function of $\hbar\omega_{exc}$

Some gain spectra show long tails which extend up to 30 meV below E_g'. We assume that these tails are due to an overdense plasma produced by the excitation in a thin top layer of the excited volume. It should be pointed out here, that the TBM averages temporally and spatially over this volume. The parameters n_p and T_p deduced from these spectra have therefore to be regarded as average values. Slight variations of the parameters are obtained under identical conditions for different samples and even for different regions of the excitation spot.

The temperature of the plasma depends at constant T_L = 5 K only weakly on I_{exc}. The difference $T_p - T_L$ increases however from about 2 K at almost resonant excitation ($\Delta E = \hbar\omega_{exc} - \mu \approx 40$ meV) to values around 25 K for $E \approx 1.1$ eV.

It is well known, that the excitonic reflection structures disappear if the samples are highly excited with $\hbar\omega_{exc} \geq E_X$ (= exciton energy) e.g. [8]. Fig.(3) shows the correlation between the excitonic reflectivity and the EHP-gain. If $\hbar\omega_{exc}$ is decreased from E_X to μ, the EHP gain disappears and the excitonic reflection structures reappear. If I_{exc} is increased at constant T_L and $\hbar\omega_{exc}$, the gain spectra become broader and higher. The chemical potential μ remains constant or decreases with increasing I_{exc} by about 2 meV.



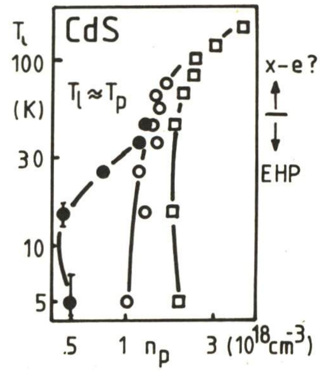


Fig.4 n_p and μ as a function of I_{exc}

Fig.5 The relation between n_p and T_L for various excitation conditions (• I_{exc} = 0.15 MW cm^{-2}, $\hbar\omega_{exc}$ = 2.58 eV; o 1 MW cm^{-2}, 2.58 eV; □ 1 MW cm^{-2}, 2.84 eV)

The plasma density however, increases significantly, as shown in Fig.(4).

Fig.(5) shows the temperature dependence of the plasma density for various I_{exc} and $\hbar\omega_{exc}$, respectively. n_p tends to increase with T_L. For lower I_{exc}, the EHP-gain disappears around 50 K, for higher I_{exc}, gain can be observed for T > 150 K. The shape of the gain spectra at higher temperatures suggests a gradual transition from an EHP-gain to an exciton-electron scattering mechanism. The rules of n_p and T_p in Figs. 4 and 5 are obtained by fitting the gain spectra with the empirical "no-k-selection rule".

The results shown in Figs. 4 and 5 are not compatible with the assumptions, that the EHP reaches a liquid like state as e.g. in Ge. They rather indicate that due to the short e-h pair lifetime only tiny clusters may be formed as predicted by theory. These mini-clusters are subject to strong fluctuations, furthermore μ and n_p are functions of the cluster radius.

The ideas developed above are supported by the fact, that the gain spectra exhibit no Landau-level structures in magnetic fields B up to 16.5 T, where the sum of electron- and hole-cyclotron energies is about 13 meV. Fluctuations in n_p and the monotonous relation between n_p and E_g' [9] result in the superposition of Landau-ladders with fluctuating starting points $E_g'(n_p) + 1/2\hbar(\omega_c^e + \omega_c^h)$ and possible structures are consequently averaged out.

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