PROC. 15TH INT. CONF. PHYSICS OF SEMICONDUCTORS, KYOTO, 1980
J. PHYS. SOC. JAPAN **49** (1980) SUPPL. A p. 523–526

OPTICAL STUDIES ON THE EXCITON ELECTRON-HOLE PLASMA SYSTEM IN HIGHLY EXCITED GaAs

K.M. Romanek, E.O. Göbel[+], H. Conzelmann and H. Nather

Physikalisches Institut, Teil 4, Universität Stuttgart,
D-7000 Stuttgart 80, Fed. Rep. Germany

[+]now: Max-Planck-Institut für Festkörperforschung, D-7000 Stuttgart 80

We have made optical studies of the thermodynamical behaviour of the electron-hole plasma (EHP) in highly excited GaAs. For this purpose, we employed luminescence, gain and Raman spectroscopy in order to determine carrier temperatures and densities. For very low effective carrier temperatures, we observe no phase separation into an exciton gas and electron-hole droplets, which is in agreement with recent calculations for direct gap semiconductors. At higher carrier temperatures, an insulator-metal transition should take place due to entropy, thermal and Mott dissociation of the excitons. We find, however, that up to carrier temperatures of 100 K, the plasma density is a linear function of temperature, hardly depending on excitation intensity, which is neither in agreement with Mott nor thermal dissociation.

I. Introduction

In many indirect gap materials a phase separation of the carrier system into a gaseous and a liquid phase can occur at sufficiently high optical excitation and below a critical temperature [1]. For direct gap materials such a phase transition has not yet been proven definitely. Especially in the case of GaAs, where a low critical temperature is expected [2,3] , the thermodynamic properties of the carrier system at various densities are not known in detail. The aim of this paper is to prove whether a phase separation occurs at low temperatures as well as to obtain information of the composition of the carrier system at temperatures above the critical value.

II. Experimental

The determination of EHP densities in GaAs from a fit of the band-to-band recombination lineshape is difficult, since a contribution from stimulated transitions cannot be excluded. Therefore, we chose two different, independent approaches: i) Optical gain measurements by probing the transmission of an optically excited thin layer of GaAs [4] or by measuring the amplification of spontaneous emission along an excited stripe on an epitaxial GaAs sample [5]. From gain spectra the Fermi energy and, hence, the carrier density can be directly deduced [6]. Since the carrier system ceases to be degenerate at higher temperatures, gain and amplification measurements cannot be employed to determine the density at high temperatures ($T_{eff} \simeq 100$ K). ii) As an alternative and novel method we employed inelastic light scattering from electronic excitations - i.e. from single particle excitations and from collective mixed plasmon-LO-phonon modes - as a means to determine carrier densities. The light pulses from a cavity dumped Kr^+-ion laser were used to create the EHP and simultaneously for Raman scattering. A similar scattering experiment from mixed plasmon-LO-phonon modes for the indirect gap semiconductor GaP has been reported recently [7].

The effective carrier temperature in GaAs is mainly a function of the excess energy of the exciting photons with respect to the ground state energy of the EHP [8]. The carrier temperature can thus be varied by tuning the exciting laser or by changing the lattice temperature. Both methods yielded identical results. The carrier temperature was estimated from a fit of the exponential high energy slope of the EHP luminescence line.

III. Results and Discussion

Fig. (1) shows transmission spectra of GaAs at a carrier temperature of 50 K. The excitation intensity has been varied from $I_o =$ 160 kW/cm^2 to $7 \cdot I_o = 1{,}12$ MW/cm^2 . For intensities below I_o, the optical gain vanishes and at the same time the EHP-line disappears in the luminescence spectrum. Above I_o, the lineshape of the gain curve remains nearly constant over a wide range in excitation intensities.

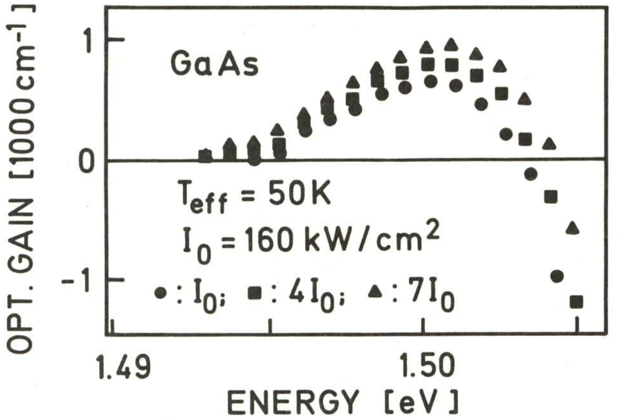


Fig.(1) Optical gain spectra of GaAs as obtained with a two laser transmission experiment at a carrier temperature of 50 K and at three different excitation intensities

The resulting densities only vary from $1.5 \cdot 10^{17}$cm^{-3} to $1.6 \cdot 10^{17}$cm^{-3}. This behaviour did not depend on carrier temperature (in the range from 4 K to 70 K). Gain spectra obtained with a different experimental method yielded identical results [9].

A Raman spectrum as obtained from a photocreated plasma is depicted in Fig. (2). The peak intensity of the incident laser was 150 kW/cm^2 at a photon energy of 1.9155 eV. At this intensity the EHP band was clearly identified in the luminescence spectrum. For comparison, Fig. (2) also depicts a Raman spectrum at relatively low excitation intensity, where no plasma luminescence is seen. This spectrum only shows the well known LO-phonon line which is allowed in the chosen backscattering configuration. The number of incident photons per second was kept the same for both spectra. The upper trace (high excitation) shows the broad scattering from single particle excitations around the laser line with a smeared cutoff near $\hbar q v_F$. The LO-phonon can still be seen, however, less intense as compared to the low intensity spectrum and slightly shifted to higher energies by approximately .5 meV. Possibly these LO-phonon contributions are connected with scattering from the outer laser spot. At 39 meV another peak

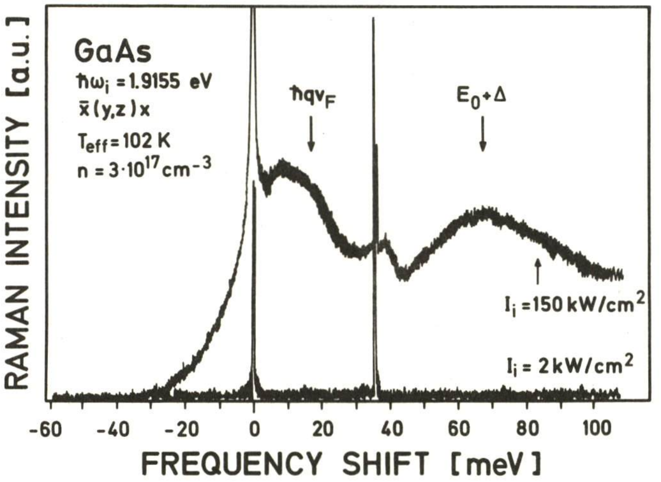
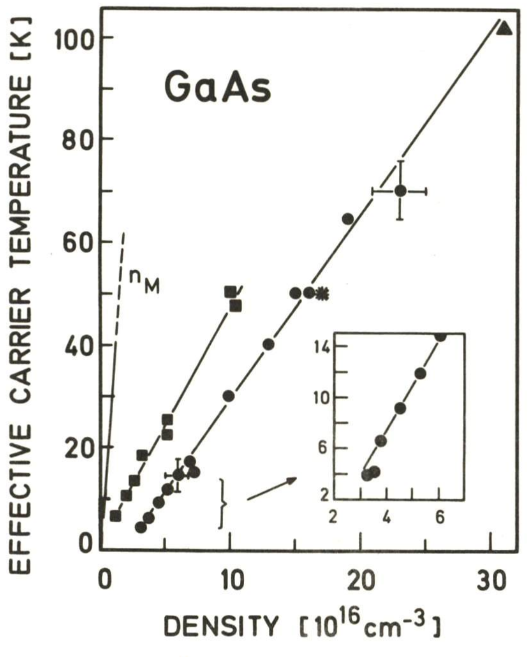


Fig. (2) Raman scattering from an optically excited EHP in backscattering geometry on a (100) surface

appears which is assigned to the coupled plasmon-LO-phonon mode ω_+. The corresponding ω_- is more difficult to observe because it seems to be obscured by the free particle spectrum. Finally, at an absolute energy of 1.847 eV the luminescence from the $E_o+\Delta$ transition appears. The details of our scattering experiments will be reported elsewhere [10]. From the energetic position of the ω_+ line and from the cutoff of the free particle spectrum we consistently derive a carrier density of $3\cdot10^{17}$cm^{-3}. This is a new and independent value for the density of an EHP in GaAs.

Fig. (3) summarizes the results on the density-temperature dependence from gain (■) and transmission (●) measurements. The (▲) in the upper right corner represents the Raman data from Fig. (2) and the (*) stands for a luminescence measurement by Tanaka et al. [11], which agrees very well with our results.

Over the entire temperature range from 4 K to ≃100 K, we find that the density is a linear function of temperature ($n-n_o \simeq 2,80\cdot10^{15}$ cm$^{-3}\cdot T_{eff}/K$) and hardly at all depends on excitation intensity. The gain data yield slightly lower densities since in this case the density is averaged along the excited stripe [12] and the gain spectra are not corrected for internal losses.



The insert in Fig. (3) shows the low temperature transmission data in a blown-up scale. At temperatures in between 6.5K to 10K (critical temperature) a phase separation would be expected from RPA-calculations [2] and from scaling relations [3], respectively. Although the mea-

Fig. (3) Density of the EHP at different temperatures: The ● represent data from transmission experiments and the ■ stand for gain measurements. ▲ was derived from the Raman spectrum of Fig.(2)

sured densities hardly depend on excitation intensity - which would be expected for an electron-hole liquid - their temperature dependence is in contrast to this assumption: For an electron-hole liquid in thermodynamic equilibrium with the exciton gas a decrease of the density is expected for increasing temperature ($n-n_o \sim -T_{eff}$). Therefore, we conclude from our results that no first order phase transition can be observed in GaAs. This is in agreement with recent calculations by Koch and Haug [13], who suggest that a first order phase separation should be impossible for direct gap materials since due to short carrier lifetimes no nucleation of electron-hole droplets can occur.

For temperatures above the critical point, initially a linear density-temperature dependence of the EHP would be expected due to free carrier screening (Debye -Hückel) of the excitons (Mott transition; see straight line n_M in Fig. (3)). However, the experimentally determined densities are by a factor of 10 higher than the Mott density for static screening. Although dynamical screening should generally be considered, static screening becomes effective again near the Mott transition [14]. Furthermore, no effect of thermal dissociation of excitons on the observed EHP density can be found, though this would be expected if the EHP density would be determined by a dissociation equilibrium between excitons and free carriers [15].

The actual mechanisms that determine the quasistationary (i.e. laser pulse long compared to EHP lifetime) density of the EHP are presently not quite clear. Since the EHP is initially under a very high Fermi pressure, it will certainly expand and thus deminish its density. Including this dynamical behaviour in a theoretical description in order to determine the EHP density under quasistationary excitation conditions must be the aim of future work. At present our experiments prove, that the EHP density is not determined by a dissociation equilibrium between excitons and free carriers. This linear density-temperature relation seems to be a more general result, since similar data are reported for Si above the critical point [16].

Acknowledgements - The authors wish to thank M.H. Pilkuhn and M. Cardona for valuable discussions, and E. Bauser for providing her high quality GaAs crystals.

References

1) see e.g. E.O. Göbel, G. Mahler: Festkörperprobl. XIX (1979) 105.
2) M.Rösler, R.Zimmermann: phys.stat.sol.(b) 83 (1977) 85.
3) T.L.Reinecke, S.C.Ying: Phys.Rev.Lett. 43 (1979) 1054.
4) O.Hildebrand, B.Faltermaier, M.Pilkuhn:Sol.Stat.Comm.19 (1967) 841.
5) K.Shaklee, R.Nahory, R.Leheny: J.of Luminesc.7 (1973) 284.
6) O.Hildebrand, E.Göbel, K.Romanek, H.Weber, G.Mahler:Phys.Rev.B 17 (1978) 4775.
7) J.Kardontchik, E.Cohen: Phys.Rev.Lett. 42 (1979) 669.
8) E.Göbel, O.Hildebrand: phys.stat.sol.(b) 88 (1978) 645.
9) K.Romanek, E.Göbel, O.Hildebrand: J.of Luminesc. 18/19 (1979) 317.
10) K.Romanek, H.Nather, E.Göbel: to be published.
11) S.Tanaka, H.Kobayashi, H.Saito, S.Shionoya: Sol.Stat.Comm.33 (1980) 167.
12) E.Göbel, O.Hildebrand, K.Löhnert: IEEE J.of Qu.El. QE-13 (1977) 848.
13) S.W. Koch, H. Haug: Phys.Lett. 74 A (1979) 250.
14) R. Zimmermann, K.Kilimann, W.D. Kraeft, D. Kremp, G. Röpke: phys.stat.sol.(b) 90 (1978) 175.
15) M.Combescot: phys.stat.sol.(b) 86 (1978) 349.
16) A.Forchel, B.Laurich, W.Schmid, M.Pilkuhn: This conference.