Electron-hole plasma in direct-gap semiconductors

Autor(en): **Capizzi, M. / Frova, A. / Modesti, S.**

Objekttyp: **Article**

Zeitschrift: **Helvetica Physica Acta**

Band (Jahr): **58 (1985)**

Heft 2-3

PDF erstellt am: **13.09.2024**

Persistenter Link: https://doi.org/10.5169/seals-115597

Helvetica Physica Acta, Vol. **58** (1985) 272–280 0018–0238/85/020272–09$1.50 + 0.20/0

Electron-hole plasma in direct-gap semiconductors

By M. Capizzi, A. Frova, S. Modesti, and A. Selloni, Dipartimento di Fisica, Universita' degli Studi di Roma "La Sapienza", Piazzale A. Moro 2, I-00185 Roma, Italy

J. L. Staehli, Institut de Physique Appliquee, Ecole Polytechnique Federale, PH-Ecublens, CH-1015 Lausanne, Switzerland

M. Guzzi, Dipartimento di Fisica, Universita' degli Studi di Milano, I-20133 Milano, Italy

(1. VII. 1984)

In honor of Emanuel Mooser's 60th birthday

Abstract. Electron-hole plasma luminescence in direct gap $Ga_{1-x}Al_xAs$ has been studied in 2- and 3-dimensionally confined samples. An upper limit of $\sim 25\ \mu m$ to the plasma diffusion length has been determined. Luminescence spectra of uniform, non expanding plasmas have been successfully analyzed in terms of a k-conserving many-body model, in which single-particle energy renormalization and lifetime broadening are included. The values of the gap renormalization are in very good agreement with current theoretical estimates.

I. Introduction

Since the experimental evidence for the existence of an electron-hole liquid (EHL) in Ge and Si was firmly established [1], much work has been devoted to determine whether a similar phase occurred also in highly excited direct-gap semiconductors [2–3]. Theoretical calculations – similar to those which had been very successful in the case of Si and Ge – did in fact predict a bound EHL in several materials, e.g. GaAs and CdS, even though quite different values for the EHL binding energy have been estimated [4–5]. Experimentally, however, no conclusive evidence for droplet formation has been found in direct-gap semiconductors (DGS). The failure of theoretical predictions is even more surprising, because DGS – due to their simpler band structure – should be in principle better described by current theoretical schemes.

The key quantity which determines the different behaviour of Si and Ge or GaP versus GaAs, CdS, etc. is the recombination time τ_r. It is well known that this is of the order of microseconds in Si and Ge [1], while it is typically about one nanosecond or less in direct-gap materials [6]. Most probably such a carrier lifetime is too short to permit phase separation [7]. In addition, plasmas can be

generated only at very high excitation intensities, with effective carrier temperatures sensibly above both the critical and the lattice temperature.

EHP properties in DGS have been most frequently studied by means of gain and luminescence spectroscopy [2, 3, 8]. It has been often pointed out [2] that the values of the band gap renormalization ΔE_G obtained from experiments were substantially higher (up to 1.5 effective Rydbergs) than the theoretical estimates for the corresponding plasma density n. Recently, these discrepancies have been attributed to: the use of unjustified phenomenological models to fit the experimental lineshapes [9]; and/or the neglect of non-equilibrium effects in the plasma (i.e. the Fermi pressure [10] and the drift velocity [11]). Because of the high excitation intensities which are usually employed to overcome the short carrier lifetime, the plasma density in the optically pumped region exceeds the so-called liquid density n^*, i.e. the density which minimizes the plasma free energy. As a consequence, the Fermi pressure causes a rapid plasma expansion. Some aspects of this problem will be discussed in Section II. For excitation pulses longer than τ_r, a stationary condition is reached, characterized by plasma density and temperature profiles. A number of papers have recently appeared in which the problem of the EHP expansion is studied. A few fluidodynamical models have been proposed [10, 12], but at present none of them seems quite satisfactory. The stationary density profile strongly depends on model parameters whose values are uncertain. The difficulties of the fluidodynamical approach makes the analysis of experimental spectra ambiguous. In particular, it becomes difficult to quantitatively study the effects of many-body interactions in the plasma.

It has been recently suggested that the EHP can be 3-dimensionally confined by sandwiching the active region within a material of wider gap, and by photolythographically etching a mesa island [9]. If the dimensions of the active region are appropriately chosen, it is possible to obtain a uniform density. Luminescence from an EHP has been thus extensively studied in direct gap $Ga_{1-x}Al_xAs$. Fitting of experimental spectra has been achieved within the Random Phase Approximation (RPA) [13]. The main results are summarized in Section III. A very good lineshape fitting is obtained using simple spectral functions in which plasmon replicas are neglected. At the same time the corresponding values of ΔE_G are found to be in excellent agreement with the most reliable estimates presently available.

II. EHP expansion

Several techniques have been used in order to quantitatively measure plasma expansion in DGS. By direct photographic recording of the carrier recombination luminescence, an expansion of about 10–20 microns has been determined in direct gap $GaAs_{1-X}P_X$ at pumping power density of ~ 1 MW/cm^2 [10]. At this excitation level the plasma density at the center of the pumped area ($\sim 20\ \mu$m wide) was estimated to be about ten times n^*. This technique, however, does not provide a direct experimental density profile and does not discriminate between plasma and exciton luminescence.

Spectra of the light emission from the whole excited carrier system and from the pumped area only were analyzed using a simplified fluidodynamical model for the plasma expansion [10]. A reasonably good agreement between calculated and

experimental luminescence lineshapes required using a rather long carrier-lattice scattering time, $\tau_c \sim 10^{-11}$ s. The calculated value for the maximum diffusion velocity v_D was $\sim 3 \times 10^6$ cm/s, and the diffusion length L_D was ~ 20 μm.

An experimental plasma density profile was obtained by Romanek et al. in GaAs by means of spatially resolved luminescence spectroscopy [14]. The luminescence intensity, integrated over the energy and depth dependence, was used to measure the carrier density. The EHP was detected at distances as large as 250 μm from the exciting laser spot. Such an expansion would correspond to an average velocity $v_D \geqslant 10^7$ cm/s, a value close to the saturated drift velocity in high electric fields, and of the order of the excited-carrier Fermi velocity. The integrated luminescence emission, however, is not a very reliable measure of the local plasma density: light emitted from the pumped area and scattered from the surface roughness can indeed give rise to spurious contributions far away from the excited region.

Two contrasting results have been reported on plasma expansion in CdS and CdSe [15, 16]. By means of time and space resolved picosecond luminescence, Cornet et al. [15] measured an ambipolar diffusion coefficient D as high as 10^6 cm^2/s. This corresponds to a velocity v_D in the range $10^7 \div 10^8$ cm/s and to a diffusion length L_D of several tens of microns. Using spatially resolved gain and reflectivity measurements, Kempf and Klingshirn [16] obtained instead $v_D \sim 2 \times 10^6$ cm/s and $L_D \leqslant 5$ μm.

In order to discriminate between the strong expansion ($v_D \sim 10^7$ cm/s, $L_D \sim 100$ μm) and the moderate expansion ($v_D \leqslant 10^6$ cm/s, $L_D \leqslant 20$ μm) picture, we performed spatially resolved luminescence measurements in $Ga_{1-X}Al_XAs$ samples tailored to insure a 'one-dimensional' density gradient. These samples were obtained by photolithographically etching small filamentary islands (~ 15 μm $\times$ 400 μm) from a liquid phase epitaxy-grown structure. The latter consists of a 0.5 μm thick 'active' layer of $Ga_{1-X}Al_XAs$ ($0 \leqslant x \leqslant 0.42$) sandwiched between wider-gap ternary material ($x \sim 0.5$). The samples were immersed in superfluid helium and pumped by a pulsed dye laser ($\lambda_p = 5720$ A, pulse duration 150 ns) focused to a spot of 15–20 μm diameter. A spatial resolution of ~ 20 μm was achieved focusing the image of the excited island on the plane of the monocromator slit.

Figure 1 shows the plasma density distribution away from the center of the exciting laser spot in $Ga_{0.81}Al_{0.19}As$, for two different levels of the exciting power density. The local density is extracted from fitting the luminescence lineshape, using the model discussed in Section III. The scattering in the data (see error bars) is mainly due to some stimulated emission and to critical sample positioning. All the plasma profiles are consistently described by a diffusion length equal to about 25 μm. However, this must be taken as an upper limit, because of the finite laser spot dimensions and of the presence, at low density, of an underlying bound excition band, roughly 5 meV broad.

Another estimate of the diffusion length can be derived by measuring the spatially integrated luminescence from small circular $Ga_{1-X}Al_XAs$ islands, obtained from the same structure used for the unidimensional study. In order to reduce stimulated emission effects [9], a sample with $x = 0.39$ has been chosen for these measurements. Figure 2 shows the power density dependence of the full width at half height ΔE of the luminescence spectra for different values of the island diameter. ΔE is related to the spatially averaged density of the plasma.

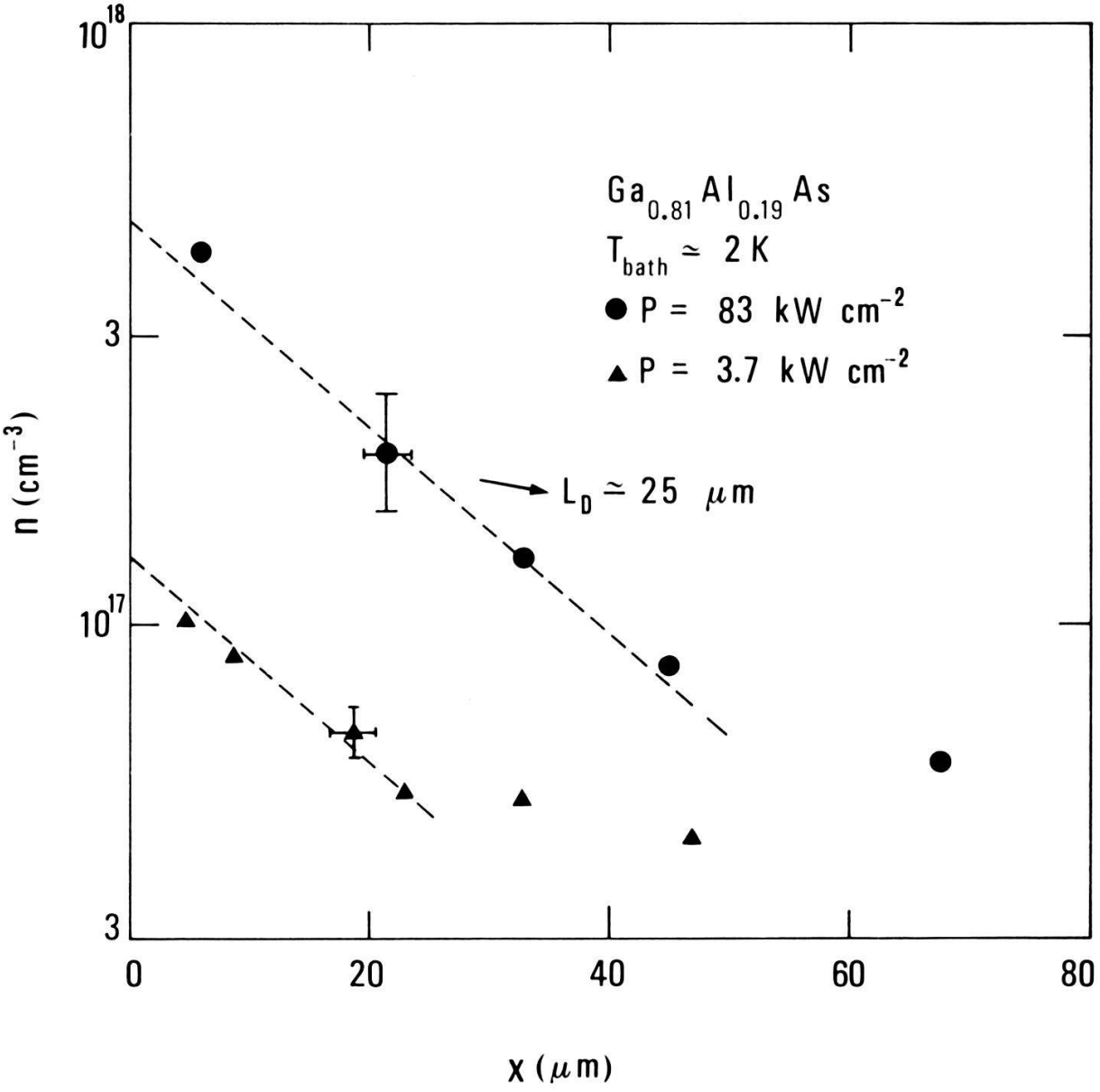

Figure 1
Density profiles of the EHP in $Ga_{0.81}Al_{0.19}As$ at two different levels of the exciting power density. The abscissa X gives the distance from the center of the exciting laser spot (20 μm in diameter). Error bars are indicative of scattering in the data, as discussed on the text.

Plasma expansion starts at power density of the order of $\sim 10\,kW/cm^2$, which corresponds to an EHP density of about $10^{17}\,cm^{-3}$, roughly a factor 2 above the estimated EHL density. At higher power, unless the island is smaller than the laser spot (20 μm), the EHP expands, resulting in a smaller ΔE. The luminescence bandwidth does not show any appreciable dependence on the island dimension as soon as this exceeds 50 μm. The plasma expansion, therefore, does not exceed this value in our experiment, consistently with the upper limit for the diffusion length obtained from the data of Fig. 1.

Even if the expansion in GaAs and GaAlAs seems to be less dramatic than that sometimes reported [14], it strongly influences the luminescence and gain spectra of the EHP. The energy distribution of the luminescence spectra, as measured by restricting the observation to the pumped region only, can be up to 50% wider than that from the whole emitting region [10]. One should therefore

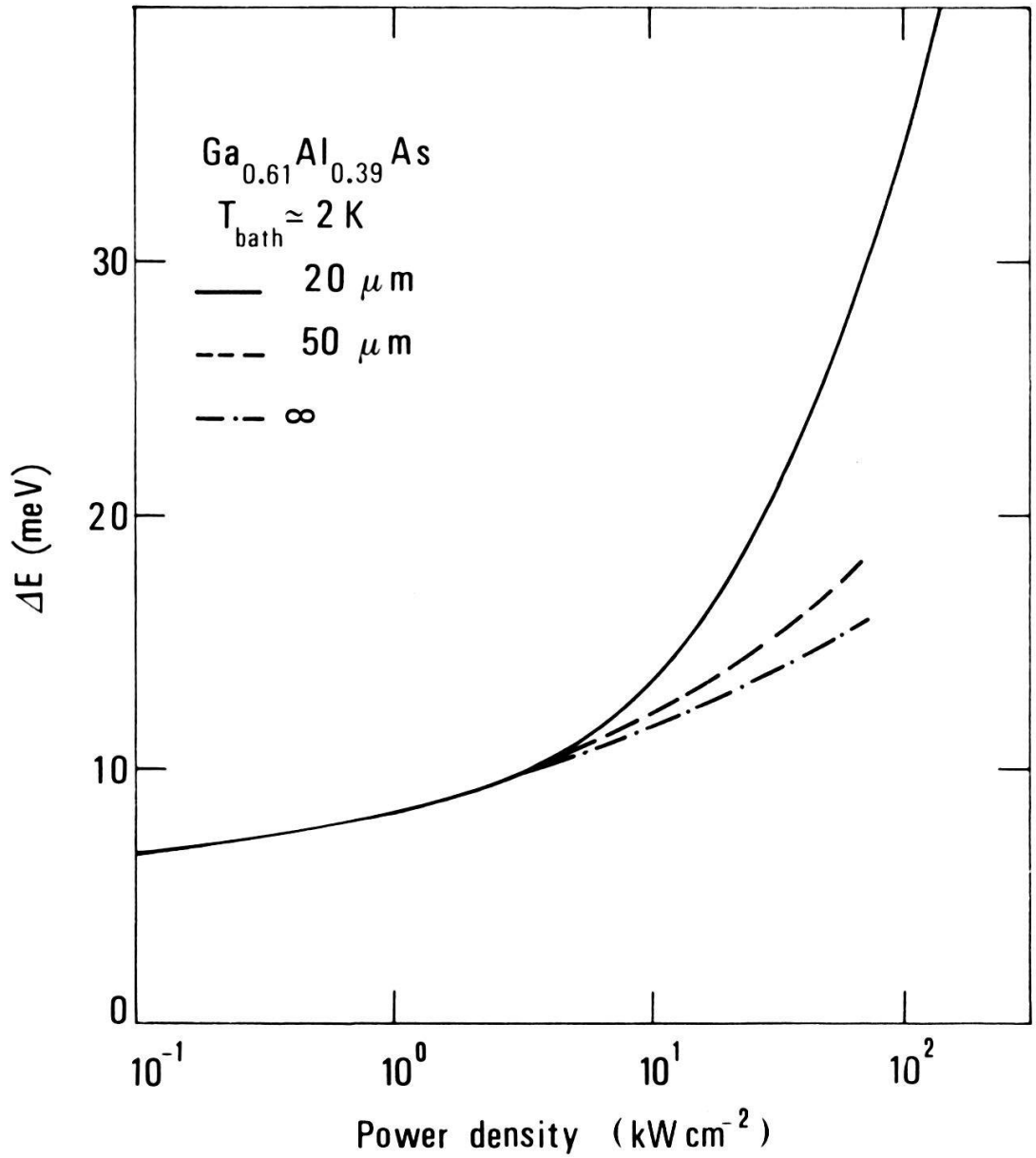

Figure 2
EHP luminescence full width at half maximum is shown versus incident light power density in $Ga_{0.61}Al_{0.39}As$. Different curves refer to circular islands of different diameter.

be very cautious in deducing plasma parameters from luminescence, unless the EHP density distribution is known by solving a fluidodynamical model of the EHP expansion, or the plasma is confined in a small fixed volume. Only the spectra obtained from islands with diameter smaller than or equal to the exciting laser spot – where the EHP density can be safely assumed to be uniform – will be considered in the following section.

III. Fitting of luminescence spectra for uniform EHP

EHP luminescence and gain spectra in direct gap semiconductors have been often described in terms of a one-electron model in which the k-selection rule was allowed to break down because of many-body interactions [17]. While this model

yields a quite good lineshape fitting, the resulting values of ΔE_G turn out to be significantly larger than the theoretical values. It has been pointed out that the procedure of neglecting k-conservation is purely phenomenological and lacks adequate justification [9].

Our analysis of the EHP luminescence is based on the RPA. The energy renormalization and lifetime broadening of single particle states (self-energy corrections) as well as plasmon-assisted transitions are accounted for. Taking into account k-conservation and assuming constant optical matrix element M, we express the luminescence intensity in the form

$$I(\omega) = |M|^2 \int d^3k \int d\omega_e \, d\omega_h A_e(k, \omega_e) A_h(k, \omega_h) \times F_e(\omega_e) F_h(\omega_h) \, \delta(\omega - \omega_e - \omega_h) \tag{1}$$

where A_e and A_h are the spectral functions for the electrons and holes respectively, and F_e and F_h are the corresponding Fermi functions. $A_e(k, \omega_e)$ (alternatively $A_h(k, \omega_h)$) represents the probability that the creation of a hole (electron) of momentum k leaves the system of the electrons (holes) i an excited state of energy $\omega_e(\omega_h)$. In equation (1) these two probabilities are independent, which means that e–h interactions (vertex corrections) are ignored. These terms are difficult to calculate in a simple, yet consistent scheme. We therefore choose to evalute them indirectly, by comparison of our calculated spectra with experiments. It is indeed known that the main effect of vertex corrections is to cause a strong reduction of plasmon replicas with respect to the case where only self-energy corrections are included [18]. In fact coupling to plasmons is weaker for e–h pairs – as involved in recombination processes – than for single electrons or holes.

We applied equation (1) using different forms for the spectral functions. We started with the 'true' expression

$$A_{e,h}(k, \omega) = \pi^{-1} \operatorname{Im} [\omega - e_{e,h}(k) - \Sigma_{e,h}(k, \omega)]^{-1} \tag{2}$$

where $e_{e,h}(k)$ are the electron and hole energies in the absence of interactions, while $\Sigma_{e,h}(k, \omega)$ are the corresponding self energies. According to the RPA, Σ is evaluated to lowest order in the dynamically screened interaction. Light-heavy hole coupling [19] and temperature effects [20] have been included in the calculation of Σ. With the full ω-dependence of Σ taken into account, equation (2) corresponds to the selfconsistent solution of Dyson's equation

$$E(k) = e(k) + \operatorname{Re} \Sigma(k, E(k)) \tag{3}$$

for single particle energies. Fitting of luminescence spectra in 20 μm islands obtained using the spectral functions (2) has been reported in Ref. 13. Although the overall shapes and positions of the experimental spectra are reproduced reasonable well, the weights of plasmon replicas are found to be significantly overestimated. Electron-hole interactions – neglected in our calculations – are thus important in the EHP luminescence. This is quite surprising, at least for the highest EHP densities and temperatures, $r_s \sim 0.5$ and $kT \sim 0.2 E_F$, where r_s is the interparticle distance measured in effective Bohr radii. We also find that our calculated values of ΔE_G are – at all densities – significantly lower than commonly accepted theoretical values [4, 19, 21].

In order to quantitatively evaluate the influence of plasmon replicas on luminescence lineshapes and band positions, we next considered a simplified version of equation (2), in which the full ω-dependent $\Sigma_{e,h}(k, \omega)$ is replaced by its value at the independent particle peak $\omega = e_{e,h}(k)$. The spectral functions become then simple Lorentzians,

$$A(k, \omega) = \pi^{-1} \frac{\Gamma(k)}{[\omega - e(k) - \Delta(k)]^2 + \Gamma^2(k)} \tag{4}$$

i.e. plasmon replicas are completely suppressed. Here we defined $\Sigma(k, e(k)) = \Delta(k) + i\Gamma(k)$. Single particle energies are given by

$$E(k) = e(k) + \Delta(k) \tag{5}$$

According to Rice [22], equation (5) is a better estimate of single particle energies than that provided by the selfconsistent solution (3), if a low order approximation – like RPA – is used for the selfenergy. Large cancellations would indeed occur between higher order selfenergy terms – not accounted for by RPA – and terms involved in the iterative selfconsistent solution of Dyson's equation.

For the calculation of luminescence spectra a slight modification of the spectral functions (4) was introduced. Functions (4) actually lead to luminescence lineshapes having an unphysical low energy tail extending deep into the gap. We therefore introduce a low energy cut-off starting at

$$\bar{\omega}(k) = e(k) + \Delta(k) - \omega_p \tag{6}$$

where ω_p is the plasmon energy. The choice of this cut-off, although somewhat arbitrary, is suggested by the behavior of the 'true' spectral functions (2), which rapidly decay at energies below the plasmon replica [13].

Fitting of experimental spectra obtained using these modified functions is shown in Fig. 3. These fits are as good as the best obtained for the EHL in Ge [23]. In addition, the values of ΔE_G given by equation (5) closely agree with those predicted by very sophisticated theories [4, 19, 21]. This suggests that – besides e–h correlations – also 'beyond RPA' selfenergy corrections are important in luminescence.

Conclusions

In summary, in the present paper we have made an experimental determination of the diffusion length of the e–h plasma in $Ga_{1-X}Al_XAs$ under heavy pumping. For a uniform EHP, as obtained with three dimensional confinement, we have shown that an excellent fit of the luminescence lineshape over a wide range of densities and temperatures can be achieved using a k-conserving many-body model which accounts for the energy renormalization and broadening of single particle states. Correspondingly, good agreement between theory and experiment is found also for ΔE_G. Detailed analysis shows that the weight of plasmon replicas in the experiment does not exceed 10%, thus suggesting that e–h correlations in luminescence are important also at high densities ($r_s \sim 0.5$).

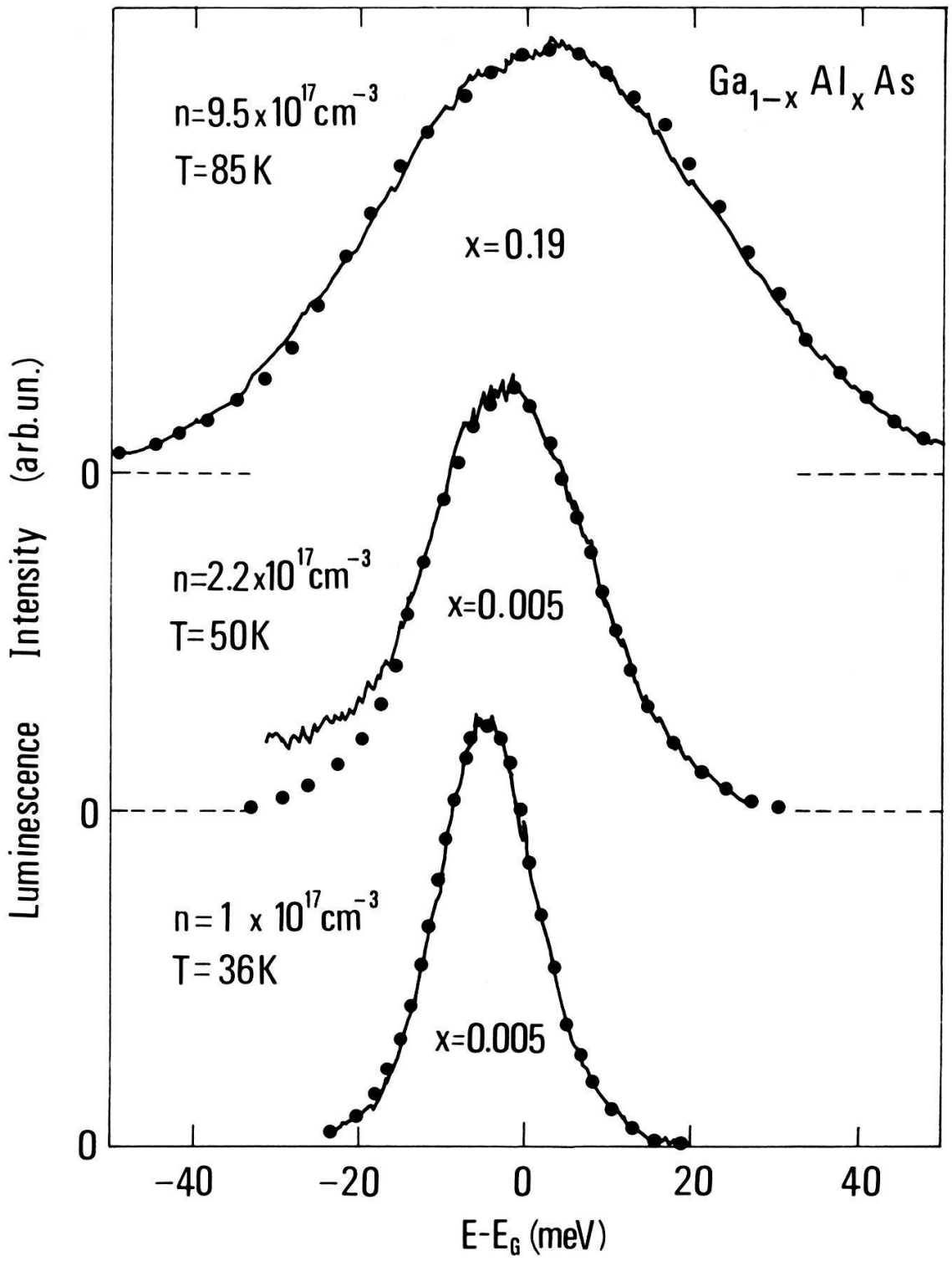

Figure 3
Fitting of the experimental plasma luminescence spectra (solid lines). Calculated spectra (dots) are obtained in the static phonon screening approximation. The energy zero is referred to the independent particle gap E (inclusive of the electron and hole polaron shifts). Results for three different sets of plasma parameters are shown.

Acknowledgements

We are very grateful to Ralph Logan of A.T. & T. Bell Laboratories for custom growing the epitaxial samples and to Luigi Chiaretti of Italtel for photo-

lithographically processing them. It is a pleasure to express our gratitude to Prof. E. Mooser for continuous support, encouragement and discussion in the course of a many-year long and fruitful interaction in the field of high-excitation spectroscopy of semiconductors.

REFERENCES

[1] For a review see J. C. HENSEL, T. G. PHILLIPS and G. A. THOMAS, Solid State Physics *32*, 87 (1977).
[2] O. HILDEBRAND, E. O. GOEBEL, K. M. ROMANEK, H. WEBER and G. MAHLER, Phys. Rev. *B17*, 4775 (1978); and references therein.
[3] S. TANAKA, H. KOBAYASHI, H. SAITO and S. SHIONOYA, J. Phys. Soc. Japan *49*, 1051 (1980); R. F. Leheny and J. Shah, Phys. Rev. Letters *37*, 871 (1976).
[4] G. BENI and T. M. RICE, Phys. Rev. Letters *37*, 874 (1976); Phys. Rev. *B18*, 768 (1978).
[5] M. ROESLER and R. ZIMMERMANN, Physica Status Solidi *B83*, 85 (1977).
[6] E. O. GOEBEL, P. H. LIANG and D. VON DER LINDE, Sol. St. Commun. *37*, 609 (1981).
[7] H. HAUG and F. F. ABRAHAM, Phys. Rev. *B23*, 2960 (1981).
[8] For a review see: C. KLINGSHIRN and H. HAUG, Phys. Reps. *70*, 315 (1981).
[9] M. CAPIZZI, S. MODESTI, A. FROVA, J. L. STAEHLI, M. GUZZI and R. A. LOGAN, Phys. Rev. *B29*, 2028 (1984).
[10] S. MODESTI, A. FROVA, J. L. STAEHLI, M. GUZZI and M. CAPIZZI, Phys. Status Solidi *108*, 281 (1981).
[11] A. FORCHEL, H. SCHWEIZER and G. MAHLER, Phys. Rev. Letters *51*, 501 (1983).
[12] G. MAHLER, G. MAIER, A. FORCHEL, B. LAURICH, H. SANWALD and W. SCHMID, Phys. Rev. Letters *47*, 1855 (1981).
[13] A. SELLONI, S. MODESTI and M. CAPIZZI, Phys. Rev. *B30*, 821 (1984).
[14] K. M. ROMANEK, H. NATHER, J. FISCHER and E. O. GOEBEL, J. Lumin. *24/25*, 585 (1981).
[15] A. CORNET, T. AMAND, M. PUGNET and M. BROUSSEAU, Solid State Commun. 43, 147 (1982); A. Cornet, M. Pugnet, J. Collet, T. Amand, and M. Brousseau, Int. Conf. on Hot Carriers, Montpellier (France), July 1981; J. Physique, Colloque C7, Suppl. au No. 10, 42, p. C7-471, (1981).
[16] K. KEMPF and C. KLINGSHIRN, Sol. St. Commun. *49*, 23 (1984).
[17] E. O. GOEBEL and G. MAHLER, Adv. Solid State Phys. *19*, 105 (1979).
[18] W. F. BRINKMAN and P. A. LEE, Phys. Rev. Letters *31*, 237 (1973); M. Roesler and R. Zimmermann, Phys. Status Solidi *B67*, 525 (1975).
[19] M. COMBESCOT and P. NOZIERES, J. Phys. *C5*, 2369 (1972).
[20] H. HAUG and D. B. TRAN THOAI, Phys. Status Solidi *B98*, 581 (1980).
[21] W. F. BRINKMAN and T. M. RICE, Phys. Rev. *B7*, 1508 (1973); P. Vashista, P. Bhattacharyya, and K. S. Singwi, ibid. *10*, 5108 (1974).
[22] T. M. RICE, Ann. Phys. *31*, 100 (1965).
[23] R. W. MARTIN and H. STOERMER, Solid State Commun. *22*, 523 (1977).