Electron-hole plasma in direct-gap $Ga_{1-x}Al_xAs$ and k-selection rule

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Uniformly excited electron-hole plasma under three-dimensional confinement has been studied in $Ga_{1-x}Al_xAs$ by photoluminescence at 2 K. With this approach, a satisfactory fitting of the experimental data has been obtained over a wide range of densities and Al concentrations using a single value of density and temperature throughout the excited region. The theoretical model used is based on the preservation of k-selection rules with a built-in broadening in the single-particle density of states. The fitting renormalized gap closely agrees with existing many-body theories, contrary to recent claims. The possibilities of plasmon-replica contributions and of thermalization processes by impurity or disorder-activated scattering are discussed. A new estimate of the radiative-recombination coefficient is reported.

I. INTRODUCTION

Since Keldysh[1] first suggested that electrons and holes in optically excited semiconductors may undergo a gas-to-liquid transition at low temperature, a great deal of effort has been spent in the investigation of highly excited materials. Many theoretical and experimental review papers[2] give a full description of the state of the art in the field of the electron-hole plasma (EHP) in semiconductors. Many-body theories are the key to quantitative understanding of the subject.[3–6] In indirect-gap semiconductors the agreement between theoretical prediction and spectroscopical data is very good. Such an agreement in most cases is not found for direct-gap materials, although they have a much simpler band structure. Since the work of Goebel *et al.*[7] in GaAs, several papers have been published where EHP gain and luminescence spectral line shapes have been fitted allowing the k-selection rule to break down.[8,9] It was pointed out[10] that the "simple one-electron k-selection rule" line shape cannot be expected to describe the EHP recombination properly, because in this model the only effect of carrier interaction taken into account is the band-gap reduction. Instead, both lifetime broadening[11] and screened Coulomb interaction—enhancement factor, carrier-carrier multiple scattering, plasmon sidebands—should be included.[12,13] The most conspicuous result of these effects is the development of a low-energy tail in the optical spectra. The procedure of neglecting k conservation[7–10] is a phenomenological approach that need not take into account all the above phenomena. A reasonable line-shape fitting is indeed obtained at the expense of a correct determination of the renormalized gap E_G', which is found one effective Rydberg unit or more below the theoretical estimate. A systematic fitting of gain and luminescence including correctly all many-body effects is therefore important. On the other hand, from the theoretical standpoint, at very high excitation levels the EHP system enters a regime where the Fermi pressure becomes competitive with recombination, so that the plasma expands far beyond the optically pumped region.[14] The ensuing density and temperature gradients make gain and luminescence line-shape analyses unreliably dependent on the hydrodynamical model chosen to describe the particle flow.[14] Moreover, high velocities affect the luminescence and gain line shape through a modified distribution function, even for a single valued velocity, density, and temperature.[15]

In a previous short communication[16] we have shown how to overcome expansion problems. By photolithographic etching of $Ga_{1-x}Al_xAs$, grown by liquid-phase epitaxy, we prepared micrometric samples whose area and thickness matched, respectively, the incident laser spot dimension and the excitation depth at low levels (see Fig. 1). This allowed the investigation of the properties of EHP in the bulk of a direct-gap material in the condition of a nearly uniform carrier density and temperature. On the other hand, it permitted extension of the use of photoluminescence (PL) to a range of excitation levels, where gain measurements are usually preferred, due to the early insurgence of stimulated emission. The range covered was further extended up to ~ 20 times the "liquid" density n_0 by the fact that in a ternary alloy approaching crossover

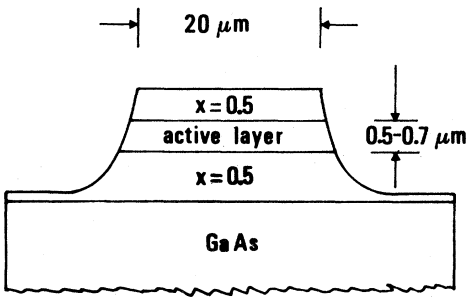

FIG. 1. Small mesa islands obtained by photolithographic etching a Ga-Al-As structure grown by LPE. The active-layer composition ranges from $x=0$ to 0.42. The top and bottom layer with wider bandgap ($x=0.5$) minimize surface recombination and provide carrier confinement, respectively. The dimensions of the active layer match the exciting laser spot size and the penetration depth at low level of excitation.

concentration x_c—for which the indirect and direct energy gaps are equal—the onset of gain appears to be shifted upwards in carrier density.

In this paper we present in greater detail PL measurements taken at 2 K over an extended range of aluminum concentration x. In the second part we discuss the theoretical model used to analyze the PL results. The third part is devoted to the presentation of the experimental details. In the last part we show that the theoretical model introduced accounts very well for our PL data of a three-dimensional (3D) confined EHP, for all values of alloy composition. It also yields values of E_G' which coincide within experimental uncertainty with current many-body theories only if k-selection rules are preserved.

II. THEORETICAL MODEL

Gain spectra in GaAs and CdS have been successfully explained by Haug and Tran Thoai[17] in terms of a many-body theory, which takes into account k-selection rules, collision broadening in the single-particle states, e-h correlation in the plasma, and plasmon-assisted transitions. The resulting line-shape fitting is as good as that obtained with the simple, unbroadened, no k-conservation model, with E_G' values in smaller but still appreciable disagreement with theory.

We fit our PL spectra with a model similar to Haug's, with the introduction of two simplifying assumptions: The energy dependence of the enhancement factor due to e-h correlation[5,6] and the plasmon replicas are neglected. The former is expected to be weak due to the relatively high plasma temperatures[17] and densities[6] attained in the present work; the latter will be discussed later on.

Haug *et al.*[17] give the expression for the imaginary part of the particle (e or h) self-energy as a function of momentum k, energy $E(k)$, temperature T, and density n. We take this quantity as the broadening parameter Γ of the Lorentzian function, which approximates the particle distribution of states.[18–21] To avoid tailing of states deep into the gap, due to the peculiar asymptotic behavior of the Lorentzian distribution, we truncate it at an energy $E=2E(k)-E_F$, where E_F is the Fermi energy.[22] We have checked that lowering of this cut-off energy is not critical for the determination of E_G', which is displaced upwards by at most two-tenths of a Rydberg. The electron distribution is therefore

$$G_e(E_e,k,n,T)=\frac{\Gamma_e}{[E_e-\tilde{E}_e(k)]^2+\Gamma_e^2}\times\Theta(E_e-[2\tilde{E}_e(k)-E_F])\ ,$$

where we recall that $\Gamma_e=\Gamma_e(k,\tilde{E}(k),n,T)$, Θ is the unit step function, $\tilde{E}(k)$ is equal to $\tilde{E}(0)+\hbar^2k^2/2m_e^*$, m_e^* being the electron effective mass, if one neglects the weak k dependence of the real part of the self-energy.[23] The plasmon-pole damping constant Γ_0, needed to calculate Γ_e as in Ref. 17, is taken equal to $0.45\hbar\omega_p$, where $\hbar\omega_p$ is the plasmon energy. $\tilde{E}_e(0)-\tilde{E}_h(0)$ gives the renormalized gap E_G'. The luminescence intensity is then fitted with the expression

$$I(\hbar\omega)\propto\int d^3k\int dE_e dE_h G_e G_h F_e F_h \delta(E_e-E_h-\hbar\omega)\ ,$$

where F_e,F_h are the usual Fermi functions for electrons and holes. The values of the effective masses of electrons and holes and of the static dielectric permittivities ϵ_0 used in the fitting procedure are given in Table I for different compositions, together with the Bohr radii and Rydbergs evaluated in the effective-mass approximation. They have been derived from Ref. 24, taking $\epsilon_0=10.06$ in Al-As.[25] The adjustable parameters used to fit the experimental line shape are the plasma density, temperature, and renormalized gap, which will be compared in the fourth section with values derived by current many-body theories.

TABLE I. Values of the effective masses of electrons, heavy and light holes, static dielectric permittivities, effective Bohr radii, and Rydbergs used in this work.

x	m_e^*	m_{lh}^*	m_{hh}^*	ϵ_0	a_x^* (Å)	Ry* (meV)
0.00 0.005	0.0665	0.0863	0.470	12.6	146	3.90
0.05	0.0695	0.0896	0.473	12.5	139	4.15
0.19	0.0785	0.100	0.480	12.1	120	4.95
0.40 0.42	0.0902	0.114	0.488	11.6	100	6.20

III. EXPERIMENTAL DETAILS AND RESULTS

Micrometric samples, such as that shown in Fig. 1, have been prepared by photolithographically etching small mesa islands on a liquid-phase epitaxy (LPE)—grown structure, consisting of a (0.5—0.7)-μm-thick "active" layer of $Ga_{1-x}Al_xAs$, sandwiched between two similar layers with wider bandgap ($x \simeq 0.5$). The active layers, having an aluminum concentration ranging from 0 (pure GaAs) to the crossover value $x_c \simeq 0.43$ (Refs. 21 and 26)—in particular $x = 0$, 0.005, 0.05, 0.19, 0.40, and 0.42—have been investigated over 5 orders of magnitude in excitation intensity.[27] A cw He-Ne laser or a pulsed dye laser ($\lambda_{peak} = 5750$ Å) were focused to ~ 20 μm onto an island of similar size, thus permitting the achievement of nearly-uniform excitation both sidewise and depthwise with a maximum intensity of $\simeq 10^2$ kW cm^{-2}. The dye laser was driven by a frequency-doubled neodymium-doped yttrium aluminum garnet (Nd-YAG) laser (pulse duration ~ 150 ns, pulse repetition ~ 1 kHz). The samples were immersed in superfluid helium. Quasistationary conditions were achieved, as shown by the fact that the emission pulse closely followed the shape of excitation.

A luminescence spectrum taken with a cw He-Ne laser at very low excitation (~ 1 W cm^{-2}) is shown in Fig. 2 for $x = 0.05$. Compositional inhomogeneity, impurity density, and alloy disorder in this sample are sufficiently low that it is possible to resolve and identify the free-exciton (FE) contribution. The arrows give, relative to FE, the energy position of several other typical contributors to the spectrum, as taken from the literature for GaAs. Their good matching to the features resolved in the spectrum allows the establishment of an absolute energy scale and therefore the determination of the crystal composition x (± 0.005), with reference to the calibration of Dingle *et al.*[26] A series of spectra taken with the pulsed dye laser in an $x = 0.19$ sample for various peak power densities is shown in Fig. 3. (See Table II.) Also shown (dots) are the fits obtained on the basis of the theoretical model previously discussed. The fit is quite good, except for some mismatch on the low-energy side, which becomes more apparent at high excitation. The gap shift to lower energy for increasing carrier density is quite evident, and so is the increase of *e-h* temperature far above the bath temperature (~ 2 K), already reported in the literature.[8,10] The spectrum for $P_0/200$ is typical of the low-excitation limit. We assign it to bound-exciton recombination. This is used to determine the crystal composition x (± 0.01) for samples $x = 0.40$ and 0.42. These samples do not show the FE in the cw very-low-excitation spectra.

A series of spectra have been taken and fitted for the full set of samples. Figure 4 shows spectra taken for different values of x at relatively similar interparticle average

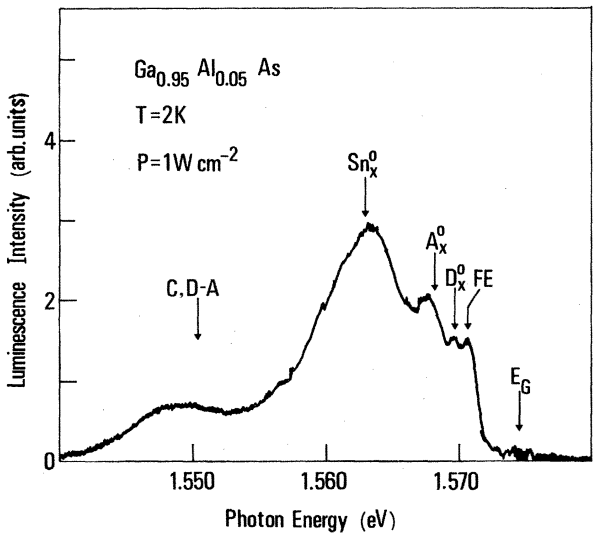

FIG. 2. Shows a photoluminescence spectrum taken with a cw He-Ne laser at low level of excitation. The free-exciton peak is identified by an arrow. The position of other typical PL peaks, taken from the literature for GaAs, and rigidly shifted to higher energy to take into account the difference in the energy gaps, are shown for comparison. D_x^0 (A_x^0) is an exciton bound to a neutral donor (acceptor), while Sn_x^0 is an exciton bound to a neutral Sn impurity. Finally, *C,D-A* includes transitions from carbon levels and/or donor-acceptor transitions.

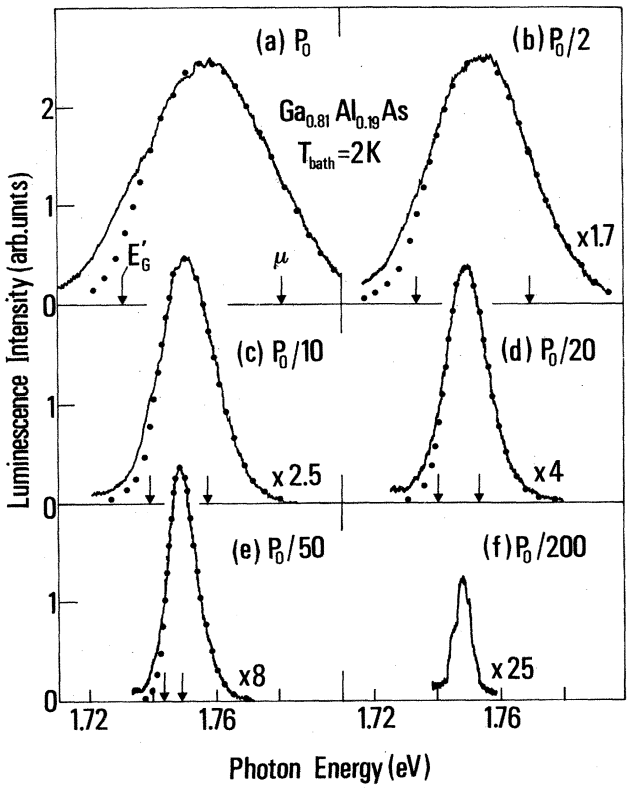

FIG. 3. A series of spectra, taken with a pulsed dye laser ($\lambda_{peak} \simeq 5750$ Å) at various excitation levels, is shown. The maximum intensity absorbed by the active layer is $P_0 = 79$ kW cm^{-2}. Fits obtained on the basis of the theoretical model discussed in the text are given by dots. Renormalized gap E_G', as derived from the fitting, and the chemical potential $\mu = E_G' + E_{Fe} + E_{Fh}$ are shown by arrows. The fitting parameters $n_{e\text{-}h}$ and $T_{e\text{-}h}$ and the values of $\Gamma_e(0)$, $\Gamma_{hh}(0)$ and $\Gamma_{lh}(0)$ (the electron, heavy-hole, and light-hole broadening parameter at $k = 0$) are given in Table II.

TABLE II. Values of the *e-h* density and temperature obtained by fitting the curves shown in Fig. 3. Also given are the values of the broadening parameter $\Gamma(0)$, at $k=0$, for electrons and heavy and light holes. The broadening goes to zero, for $T=0$, at the Fermi energy (Ref. 17).

	$n_{e\text{-}h}$ (10^{16} cm^{-3})	$T_{e\text{-}h}$ (K)	$\Gamma_e(0)$ (meV)	$\Gamma_{hh}(0)$ (meV)	$\Gamma_{lh}(0)$ (meV)
(a)	102	75	5.7	2.3	1.7
(b)	65	70	5.0	2.3	1.7
(c)	24	45	3.6	1.8	1.3
(d)	15	37	3.0	1.7	1.2
(e)	6	30	2.4	1.6	1.2

separation $r_s=(\frac{3}{4}\pi n a_x^3)^{1/3}$. The data are plotted as a function of $E-E_G'$, with arrows showing the chemical potential. The fitting (solid dots) is good for all x values, again with some deviation in the low-energy tail. The possible causes for this misfit will be discussed in the following section.

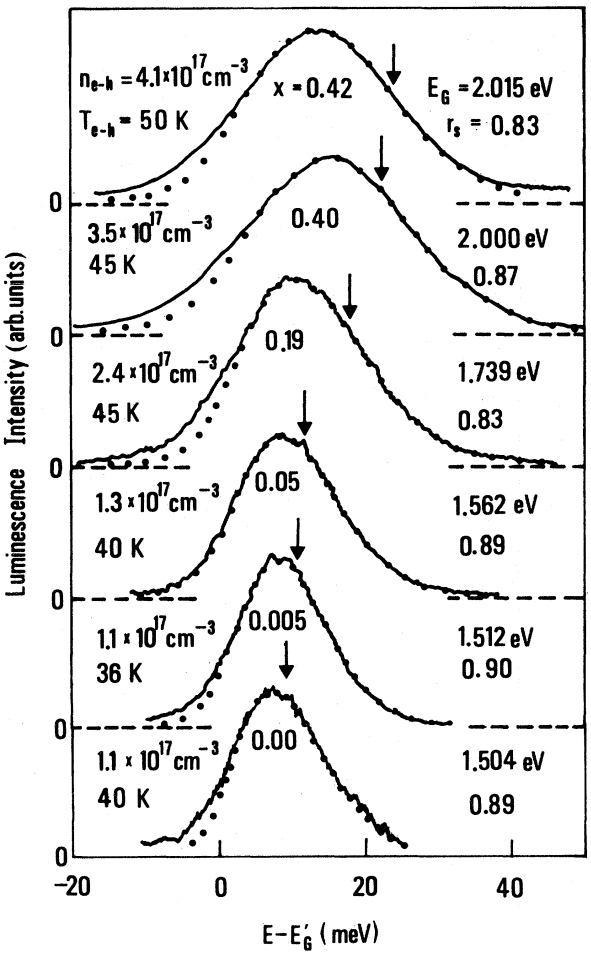

FIG. 4. Photoluminescence spectra, taken for different values of composition x at similar interparticle average separation r_s, are plotted as a function of the energy measured from E_G'. The dots give the fits by the theoretical model discussed in the text. The fitting parameters $n_{e\text{-}h}$, $T_{e\text{-}h}$, and E_G' are given, while arrows show the chemical potential.

IV. DISCUSSION

A. Plasma renormalized gap

Very recently Vashista and Kalia[28] (VK) and Thuselt[29] have demonstrated that the sum of exchange and correlation energy, E_{xc}, as measured in effective Rydbergs (R_y^*), is a nearly universal function of the normalized interparticle distance r_s. Both papers, which give full theoretical support to a number of previous theoretical and experimental claims,[5,30] arrive at an analytical expression for E_{xc} which results in nearly identical values and can be employed to obtain $E_G'=E_{xc}+n\partial E_{xc}/\partial n+E_G$.[5] Values of the difference between $E_G'(n)$ and the gap value E_G [equal to $E_G'(0)$], derived from fitting the experimental spectra with the theory presented in Sec. II, are plotted in Fig. 5

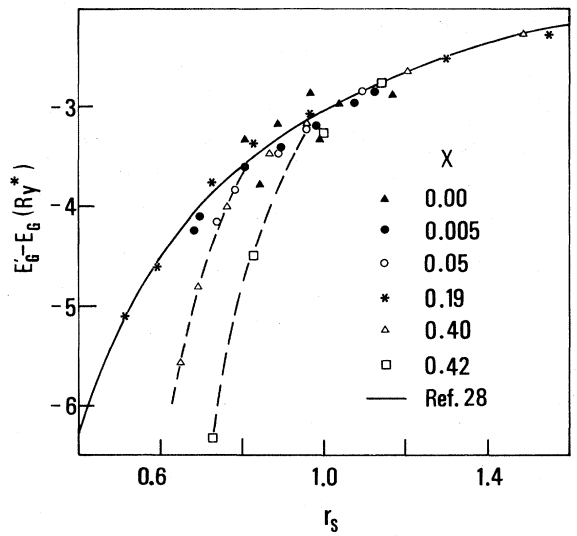

FIG. 5. Difference between the values of the renormalized gap E_G', deduced from the fitting as explained in the text, and the gap value in the diluted limit E_G, is plotted vs the normalized interparticle distance r_s. All data reported here refer to spectra where stimulated emission was not detectable. Solid line gives the behavior expected from the many-body theory presented in Ref. 28. Dashed lines are a guide for the eye. The values for $x=0$, 0.005, 0.05, and 0.19 samples have been reported without any adjustment, because in these cases an absolute energy scale can be deduced from the low excitation spectra (Fig. 2). The data for $x=0.40$ and 0.42 have been instead shifted rigidly along the energy scale for best matching of the $x=0$, 0.005, 0.05, and 0.19 samples data at low densities (high r_s).

versus r_s. The agreement is quite good save for samples $x=0.40$ and 0.42, which deviate at densities such that the EHP Fermi-energy values (28 and 17 meV, respectively) agree with the Γ-X separation, equal to 36 ± 13 and 13 ± 13 meV, respectively,[26] thus suggesting that both minima are populated. In the following, we shall set these data aside, because a specific theory would be required to allow correct extraction of E_G' and n values from these PL spectra. We compare now our results with the $T=0$ universal function by VK (Ref. 28):

$$E_{xc}=-\frac{4.83+5.09r_s}{0.0152+3.04r_s+r_s^2},$$

from which the full line in Fig. 5 has been calculated. Comparison is possible, although our carrier temperature can be as high as 75 K at the highest excitation (see Fig. 8), because E_{xc} is almost temperature independent as long as kT is smaller than $\hbar\omega_p$,[31] as is always verified in our data. A comparably good agreement would be found, within experimental uncertainty, with other many-body approaches.[4]

In order to explain the discrepancy between the above conclusion and previously reported disagreement between theoretical and experimental E_G' values, we focus the attention on the two novel aspects of our approach: k conservation and 3D confinement. The role of the former factor can be weighed by fitting the data for the 3D-confined sample $x=0.19$, allowing now for full k relaxation. We maintain instead the broadening in the single-particle density of states. A typical fit is shown in Fig. 6 (solid line) together with a fit conserving k-selection rule (dashed line). Both models fit the PL spectrum quite well, but for values of E_G' which differ by $\sim$1Ry*. This is a consequence of the different energy dependence of the unbroadened transition rates in the two cases. The discrepancy betwen E_G', evaluated from the fitting with k relaxation, and the VK theory becomes even more pronounced at smaller interparticles separation, as illustrated by Fig. 7. It is concluded that k conservation is therefore crucially important.

The role of the second factor, carrier confinement, is difficult to ascertain, because fitting of the experimental data must rely on some kind of hydrodynamical model for the plasma expansion.[14,32] Aside from objections that have been raised[33] against a particular model,[32] it is true in general that in an expanding plasma the quasi-equilibrium carrier distribution near the Fermi energy is modified by density-gradient-dependent drift velocities.[15] We therefore argue that fitting spectra from an expanding plasma may at most test a model of expansion, once the validity of the many-body theory is well established, or vice versa. In conclusion many of the high-excitation spectra reported in the literature cannot be plainly related to the relevant parameters of the EHP.

B. Plasma temperature

Values of the e-h quasiequilibrium temperatures $T_{e\text{-}h}$, as derived from fitting the photoluminescence, are reported in Fig. 8 versus plasma density in a log-log plot. The data are consistent with previous reports. The temperature increases for increasing density.[32,34,35] The linear dependence of $T_{e\text{-}h}$ vs $n_{e\text{-}h}$ shown by the 3D-confined plasma at low and medium density agrees with that found in GaAs for an expanding plasma[34,35] (solid squares). Thus it is very unlikely that such linear behavior be related to the expansion mechanism, contrary to a recent proposal.[32,35] Moreover, at high density $T_{e\text{-}h}$ saturates with $n_{e\text{-}h}$ (see sample $x=0.19$), as also previously reported for an expanding plasma in GaAs (Shah and Leite[36,37]). Shah and Leite interpreted their data in terms of thermalization through scattering processes with LO phonons, giving

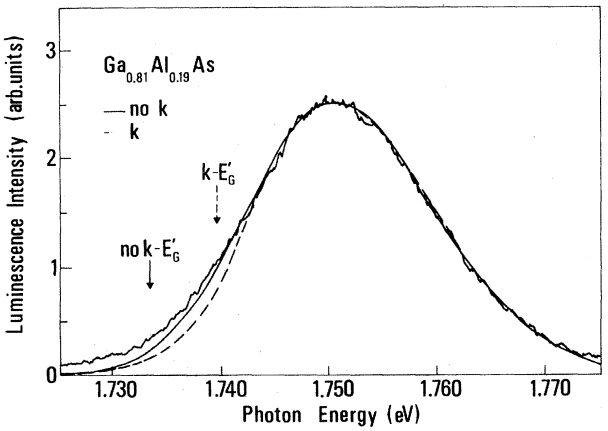

FIG. 6. Line-shape fitting of photoluminescence in a $x=0.19$ sample under two different assumptions: k-selection rule is relaxed (solid line) or preserved (dashed line). In both cases the broadening in the single-particle density of states is included. The fitting parameters are $E_G'=1.7330$ eV, $n_{e\text{-}h}=2.45\times10^{17}$ cm^{-3}, and $T_{e\text{-}h}=45$ K in the first case, $E_G'=1.7393$ eV, $n_{e\text{-}h}=2.42\times10^{17}$ cm^{-3}, and $T_{e\text{-}h}=45$ K in the second case.

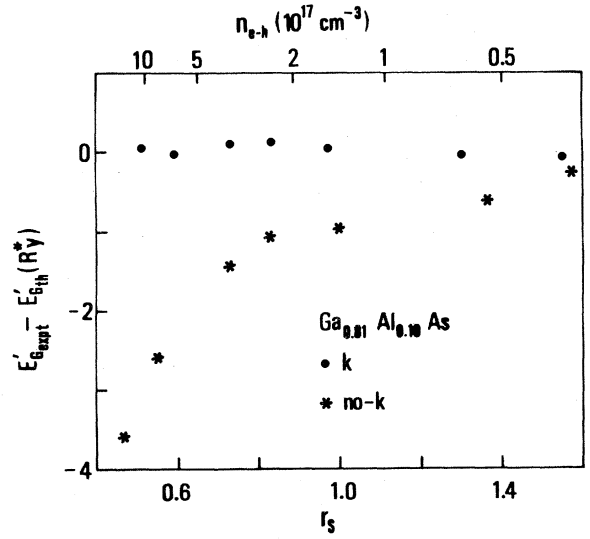

FIG. 7. Deviation of E_G' values from theory (Ref. 28) as obtained by fitting photoluminescence with k conservation (circles) and without (stars).

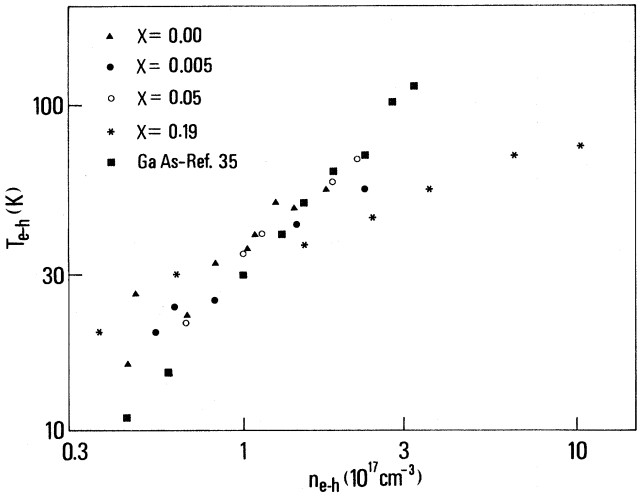

FIG. 8. Log-log representation of plasma temperature vs plasma density.

$$P \propto \exp(-\hbar\omega_0/kT)$$

where P is the absorbed power per unit volume and $\hbar\omega_0$ is the LO-phonon energy (36.8 meV in GaAs). This relationship is shown in Fig. 9 (dashed line) for comparison with our data. It is seen that the model possibly accounts for the results only at high temperatures. Below 60 K, the plasma seems to cool down more efficiently than expected on the basis of this scattering mechanism alone. This may be due to other processes, such as nonpolar optical scattering, deformation potential, and piezoelectric scattering. However, the latter are expected to become dominant only below $\sim$40 K,[38–40] as is experimentally verified for GaAs. This is evidence that, in a ternary alloy, other processes—e.g., phonon emission without k conservation—are activated by disorder and impurities, resulting in an efficient thermalization channel which is not available in high-quality GaAs.

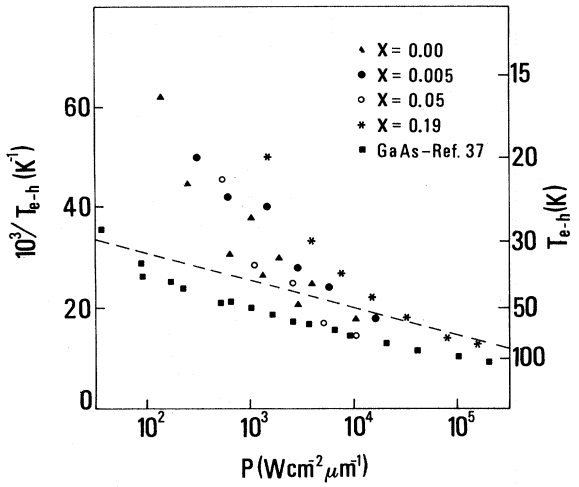

FIG. 9. Reciprocal plasma temperature vs volume power density. The dashed line shows the theoretical slope for thermalization via LO-phonon scattering, given by $P \simeq \exp(\hbar\omega_0/kT)$, with $\hbar\omega_0 = 36.8$ meV.

C. Plasma lifetime

Values of the total lifetime τ, given by the ratio between the e-h density and the generation rate per unit volume, are plotted in Fig. 10 versus n_{e-h}. The full lines are our best fit to the expression

$$\tau^{-1} = A + Bn_{e-h} + Cn_{e-h}^2 .$$

The coefficients A,B,C and the range of variation of the measured lifetime are given in Table III, together with values previously measured at liquid-nitrogen (LN) temperature. The A term corresponds to recombination associated with impurities and disorder and is therefore sample dependent. It is seen that its contribution is important only for sample $x = 0.19$. The band-to-band radiative coefficient B can be theoretically calculated, but is critically sensitive to a number of conditions.[41–44] For a degenerate system, a constant B value may cease to be meaningful.[45] Southgate[43] has shown that in a region of moderate degeneracy, where $1 < E_F/kT < m_h^*/m_e^*$, B can still be considered independent of n_{e-h} and very close to the classical value.[42] By applying his model to the analysis of absorption data, he gets for B at LN a value 7.6×10^{-9} cm^{-3}s^{-1} for undoped material, and 0.5–16×10^{-9} cm^{-3}s^{-1} for highly doped material. The two extrema correspond to maximum and zero estimated gap shrinkage.

The ratio E_F/kT always satisfies the above condition in our experiment, so we can estimate B from our data.[46] The value of B that we get is almost the same for the three aluminum concentrations investigated. It is important to stress that our results are in very good agreement (see Fig. 10) with those reported by Dapkus *et al.*[47] from the Burstein shift in GaAs. This is the only determination done,

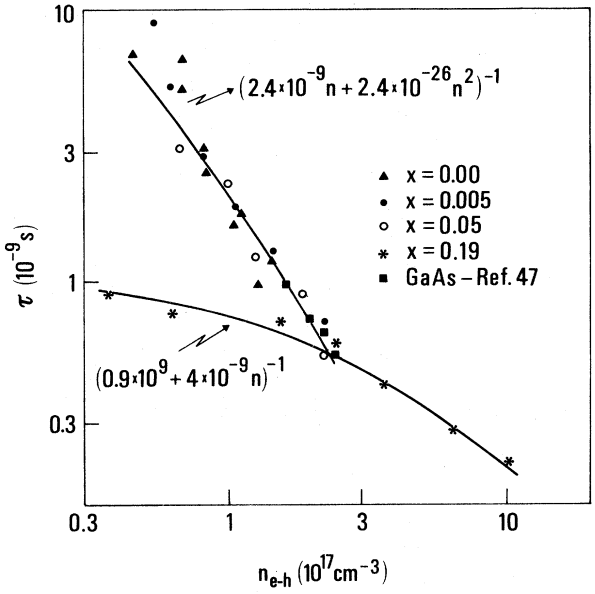

FIG. 10. Plasma lifetime vs density. A best fitting in terms of the expression $\tau = (A + Bn_{e-h} + Cn_{e-h}^2)^{-1}$ is given by the solid line. In all cases, values in the region where stimulated emission becomes clearly detectable are not shown.

TABLE III. Values of the lifetime coefficients A, B, and C, as obtained in the present work, compared with other available data and with theoretical calculations.

Material	A (10^9 s^{-1})	B (10^{-9} $cm^3 s^{-1}$)	C (10^{-26} $cm^3 s^{-1}$)	τ (10^{-10} s)	Reference
GaAs $Ga_{0.95}Al_{0.05}As$		2.4±1	2.4±1	4–60	This work
$Ga_{0.81}Al_{0.19}As$	0.9±0.5	4±2		2–10	This wo
GaAs		5.5		6–10	47
GaAs		16		1	48
$GaAs_{0.62}P_{0.38}$		36		5	14
GaAs		50		10	49
GaAs			10^{-5}		50
GaAs		2–20			40
GaAs		0.5–16			43
GaAs			5×10^{-4}		51

as in our case, in a uniformly excited plasma, volume confined, without losses due to surface recombination. Divergent results from other sources shown in Table III are possibly related to nonuniform excitation,[14,48,49] and to plasmon and/or stimulated emission effects.[49]

The Auger coefficient C, not measurable in the $x=0.19$ sample, takes instead, for the $x=0$, 0.005, and 0.05 samples, much larger values than previously measured by experiment[50] or theoretically estimated.[51] The reason for such behavior is not clear and further investigation is necessary. In particular one should explore the possibility that some stimulated emission, although not experimentally detected, may be present and affect the lifetime with a strongly density-dependent component.

D. Plasmon replica

An analysis of Fig. 3 shows that the theory does not fit the low-energy tail of the experimental line shape very well. This misfit is present in all the investigated samples, except for $x=0.05$ (see Fig. 4). By subtracting the theoretical line shape from the experimental curve, one gets a bell-shaped structure, peaking at an energy ΔE below the photoluminescence maximum. This quantity falls on a straight line with slope $\frac{1}{2}$, suggesting that the observed misfit can be related to a plasmon replica. This misfit could also be due to having neglected, in our fitting, the broadening due to alloy disorder and/or to the cutting procedure used for the Lorentzian smearing. Further measurements and the use of a theory which properly includes plasmon replicas are therefore needed to rule out the possibility that this square-root dependence of ΔE on $n_{e\text{-}h}$ is just accidental.

V. CONCLUSIONS

By studying Ga-Al-As alloys of various compositions, we have shown that photoluminescence can be the right tool to investigate EHP properties in direct-gap materials over a wide density range, provided the plasma is properly confined to warrant uniform excitation and to reduce optical amplification effects. The use of k conservation and of a built-in many-body broadening allows measuring values of the renormalized gap which closely agree with existing many-body theories over a wide range of densities and of Al concentrations. From the relationship between the plasma temperature and absorbed power density, thermalization processes by impurity or disorder activated scattering have been inferred. Analysis of the results has also enabled measurements of the radiative recombination coefficient B to be done with a novel procedure.

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