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Electron-hole plasma in photoexcited indirect-gap $Al_xGa_{1-x}As$

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We report luminescence studies of pulse photoexcited, indirect-gap $Al_xGa_{1-x}As$ in the temperature range of 2–150 K. Two spectral features are observed: excitons scattered by random potential fluctuations, and the electron-hole plasma (EHP). For $0.44 \leq x \leq 0.53$ we find a plasma density between 0.5 and 2×10^{19} cm^{-3}. The plasma binding energy, relative to the free exciton, is very small and may be negative. A strong no-phonon band is observed, which we attribute to scattering by potential fluctuations, as well as momentum-conserving phonon-assisted transitions. The lifetime of the EHP below 20 K is about five times shorter than that of the "intrinsic" exciton and is predominantly radiative. There is good, if not definitive, evidence that plasma-exciton phase separation occurs. Stimulated emission from the EHP is observed in nominally undoped, indirect-gap material, for $x \leq 0.46$. Further spectral features are present at moderate to high excitation, which may result from multiexciton complexes.

I. INTRODUCTION

Following the discovery and detailed study of the electron-hole liquid[11] in Ge, similar results have been reported in other indirect-gap semiconductors, Si,[1] SiC,[2] and GaP.[3] Calculations of the liquid density and binding energy have been made which agree quite well with experiment in all these cases.[4] In all these materials, when undoped, recombination is through phonon-assisted transitions. In binary semiconductors the overlap between transitions involving different phonons precludes accurate measurements of line shape, from which the density is deduced.

Recently it has been found that in highly excited nitrogen-doped GaP there is a strong no-phonon band with the line shape characteristic of the electron-hole liquid or plasma (EHP).[5] Thus the density can be determined relatively accurately and it is found to depend on excitation level and on time delay after the exciting pulse. These results have been confirmed by Raman scattering measurements of the coupled plasmon-phonon modes.[6] These studies show, furthermore, that the EHP persists with essentially constant density up to room temperature.

These results on GaP:N are quite different from those obtained in Ge and Si, and suggest that the model of a liquid phase in equilibrium with an exciton gas is not necessarily correct for a binary semiconductor under the conditions of these experiments. For this reason we prefer to use the more general term "plasma" rather than "liquid" to describe the dense electron-hole phase.

In this paper we report a photoluminescence study of intensely excited $Al_xGa_{1-x}As$, with $0.44 \leq x \leq 0.53$ in which the minimum gap is indirect. We have studied the dependence of the Fermi energy and electron-hole pair density on temperature, time, and excitation intensity for various x. While the particle energy distribution in these materials is similar to the binary semiconductors mentioned above, the EHP radiative recombination processes are peculiar to a random alloy with the direct-band minimum (Γ) lying close to the indirect-band minimum (X). This results in an allowed no-phonon band, in a strong enhancement of the radiative recombination process relative to impurity bound excitons and in the appearance of stimulated emission in the indirect gap.

II. EXPERIMENTAL PROCEDURE AND RESULTS

The samples used in this study were $Al_xGa_{1-x}As$ epilayers, 5–10 μm thick, grown on GaAs substrates by liquid phase epitaxy. The samples were nominally undoped but had a net background impurity level of $N_D - N_A \sim 10^{16}$–10^{17} cm^{-3}, as determined by capacitance measurements at room temperature. This impurity content should not affect the EHP, whose density is of order 10^{19} cm^{-3}.

Crystal composition was determined from the position of the Γ peak in the absorption or excitation spectrum, using the calibration of Dingle *et al.*[7] These spectra were obtained with a cw tunable dye laser. The samples were found to have an Al composition parameter x which varies with depth by approximately 0.005 per μm, the lowest x being at the outer surface.[8] This is the value of x quoted throughout this paper, since the penetration depth of the excitation never exceeded 1 μm.

Photoluminescence data were obtained by excitation between 2.0 and 2.3 eV with a pulsed dye laser pumped by an N_2 laser. The dye-laser peak power was 5 kW, its pulse width was 5 nsec at half maximum and its linewidth was less than 0.1 Å. The peak excitation intensity at the sample

surface varied between 10^4–10^7 W/cm^2, depending on beam focusing and attenuation. The photoluminescence was monitored with a double monochromator (resolution of 1 to 4 Å), equipped with a cooled photomultiplier which had a rise time of 3 nsec. The pulses were analyzed with a boxcar integrator employing a 1-nsec or a 10-nsec gate. The samples were either immersed in liquid helium (for 2 and 4.2 K runs) or mounted in a temperature-controlled cryostat (for $T > 4.2$ K). The samples were in contact with the cold helium gas in order to provide heat dissipation from the illuminated surface. The actual crystal temperature was slightly higher than that measured by a thermocouple attached to the sample holder, owing to heating by photoexcitation. Measurements on the bound exciton, whose intensity is strongly temperature dependent, show that this heating effect is only a few degrees. However, the effective electron temperature can be considerably higher. This is discussed further below.

The present study is limited to strictly indirect-gap $Al_xGa_{1-x}As$, with $x \geq 0.44$. Figure 1 shows the photoluminescence spectra of $Al_{0.45}Ga_{0.55}As$ obtained under various excitation conditions. The cw spectrum shows the "intrinsic" exciton (BX) emission, which is believed to arise from excitons scattered by random potential fluctuations,[9] and part of the donor-acceptor pair band. When low-intensity pulse excitation (~10^4 W/cm^2 corresponding to 10^{14} photons/cm^2 per pulse) is used, only the bound exciton line is observed. The donor-acceptor pairs are long lived and escape detection when the time delay after the exciting pulse is less than a few hundred nsec. As the excitation intensity is increased to ~10^{16} photons/cm^2, a broad emission band (designated EHP in Fig. 1) appears. Associated with this band is a phonon sideband whose intensity relative to the no-phonon band increases with increasing x. For crystals with $x \leq 0.46$ (i.e., well into the indirect-gap range of composition), a further increase in excitation intensity results in stimulated emission (SE) on top of the EHP band. The data shown in Fig. 1 are typical of all indirect-gap $Al_xGa_{1-x}As$ (except for the SE which does not appear for $x > 0.46$).

Figure 2 shows the EHP band of $Al_{0.46}Ga_{0.54}As$ observed at 17 and 110 K. Also shown are parts of the absorption spectra at these temperatures.

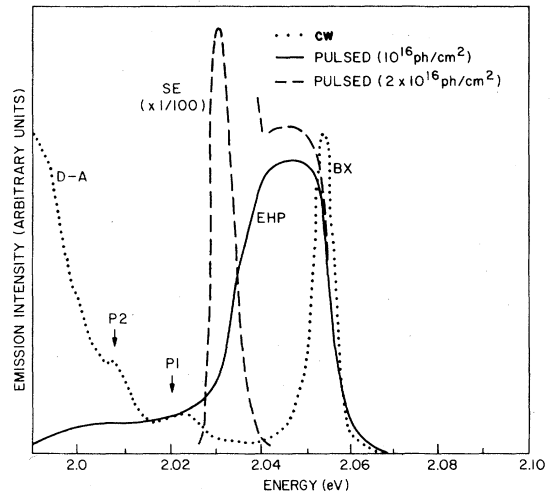

FIG. 1. Photoluminescence spectra of undoped $Al_{0.45}Ga_{0.55}As$ at 4.2 K. The cw spectrum shows the intrinsic exciton scattered by potential fluctuations (BX), its phonon sidebands ($P1$, $P2$) and the high-energy part of the donor-acceptor pair spectrum (D-A). The pulse spectrum shows the electron-hole plasma emission (EHP) below and above the threshold for stimulated emission (SE).

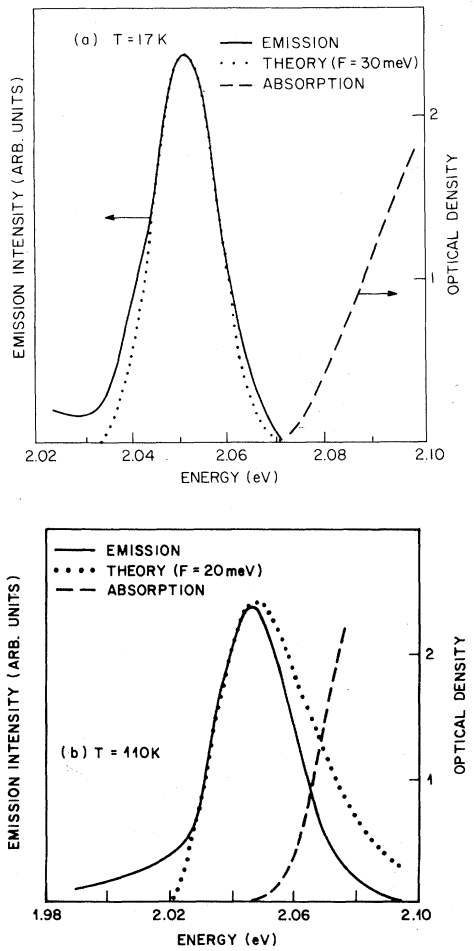

FIG. 2. EHP radiative recombination of undoped $Al_{0.46}Ga_{0.54}As$ observed at (a) 17 K and (b) 110 K, with no delay after the exciting pulse. The excitation energy used was 2.1 eV. Also shown are the cw absorption spectra at the same temperatures. The theoretical spectra are calculated from Eq. (1), taking T as the lattice temperature.

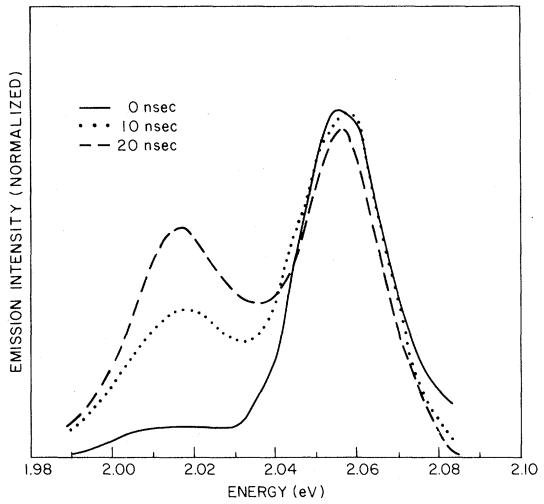

FIG. 3. EHP radiative recombination of undoped $Al_{0.46}Ga_{0.54}As$ at 35 K observed at various times after the excitation pulse. The gate used was 10 nsec wide. Excitation energy: 2.1 eV.

Note that both absorption and emission bands show the usual shift to lower energy as the temperature increases. Figure 3 shows the emission spectrum of $Al_{0.46}Ga_{0.54}As$ at 35 K under intense pulse excitation and for various time delays after the exciting pulse (the lifetime of the EHP at this temperature is 20 nsec). All three spectra shown are normalized at the EHP band peak, and it is clear that the relative intensity of the phonon sideband increases with elapsed time.

The EHP and BX both decay exponentially with time but with different lifetimes. Figure 4 shows typical logarithmic decay curves for $x = 0.48$ at 4.8 K. Care has been taken to separate out the

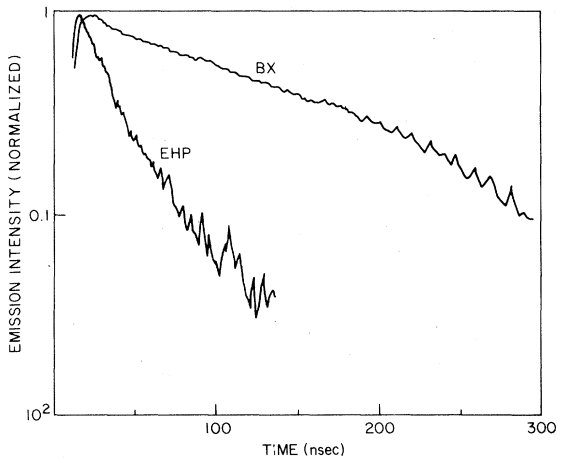

FIG. 4. Time dependence of the radiative combination of the exciton (BX) and the electron-hole plasma (EHP) of $Al_{0.48}Ga_{0.52}As$ at 4.8 K. The long-lived tail on the EHP curve is due to background BX emission.

spectrally overlapping BX and EHP contributions. The BX lifetime drops off at low excitation level, as does its luminescent efficiency. This is presumably because of nonradiative decay paths via impurities and defects, which become saturated at high excitation. We take the lifetime at an excitation level just below the threshold for formation of the EHP to be the best approximation to the true BX lifetime.

III. ANALYSIS

In this section the subjects to be treated are: (1) Determination of the electron-hole pair density in the EHP as a function of temperature. (2) Proof that the EHP forms a separate phase, and the determination of its binding energy. (3) The recombination processes of the EHP. (4) The possibility that other multiexciton entities contribute to the photoluminescence spectrum. We shall also show the dependence of each of the measured quantities on the composition parameter x.

The no-phonon band of the EHP radiative recombination is fitted to the following expression[10]:

$$I(h\nu) = I_0 \int_0^{h\bar{\nu}} dE\, D_e(E) D_h(h\bar{\nu} - E) f(E, F_e, T) \times f(h\bar{\nu} - E, F_h, T) . \quad (1)$$

F_e, F_h are the electron and hole Fermi energies, respectively, D_e and D_h are the respective densities of states, $h\nu$ is the emitted photon energy, and $h\bar{\nu} = h\nu - E'_{BB}$, where E'_{BB} is the renormalized band gap in the regions occupied by the EHP. This energy, E'_{BB}, is obtained from the observed data and is taken as the intersection of the extrapolated low-energy part of the no-phonon band with the energy axis. The f's are the Fermi functions for the electrons and holes. The density-of-states masses are obtained from the interpolation expressions summarized in Ref. 11: $m_h(x) = (0.48 + 0.31x)m_0$ for the hole and $m_e(x) = (0.85 - 0.07x)m_0$ for the electron at the X conduction-band minimum. It is assumed that any "camel's-back" effects[12] on the conduction-band minimum can be neglected. Composition broadening of the EHP emission is presumably no greater than that of the BX (7 meV) and was neglected. Equation 1 could be satisfactorily fitted to the data for $T \gtrsim 15$ K. Below that, except at the highest excitation levels, BX emission obscures the high-energy side of the EHP band.

A good fit can be obtained for all data up to 80 K. For most of the data we could assume that the EHP was at the lattice temperature, so that $F = F_e + F_h$ is the only adjustable parameter. However, at high excitation levels and low lattice temperatures, it is necessary to assume some heating

of the EHP to get a good fit.

We can obtain the electron-hole pair density from F by performing the integration $D_e(E)f(E,F_e)$. The results are shown in Fig. 5. The density is not very sensitive to the assumed mass ratio, m_e/m_h, and its accuracy is limited to ±20% at 20 K primarily by the quality of the fit to the spectrum. This is confused on the low-energy side by the presence of the phonon sidebands, and also by emission from radiative Auger transitions in which part of the energy of recombination is given up to a third particle or a plasmon. On the high-energy side, the fit is affected by the choice of temperature, which leads to errors in F at high excitation levels where the EHP temperature exceeds that of the lattice by up to 20 K. (This is apparently only important for $T \lesssim 40$ K.)

Above 80 K, the fit is confused by overlap with absorption due to direct transitions to the Γ minimum of the conduction band. The effect of this absorption is probably somewhat reduced by emission from the Γ minimum, and also by resonant re-emission. Because of the large number of unknowns (the renormalized direct bandgap, the relative strength of the Γ emission, the re-emission efficiency, and the depth of penetration of the excitation) it is impossible to make a meaningful fit to the high-energy side of the emission curve. We attribute the deviation of the experimental curve from theory [see Fig. 2(b)] to these causes. It is possible to make a reasonable estimate of the Fermi energy from the low-energy side of the curve alone. However, above about 80 K the EHP ceases to be degenerate and the fit becomes progressively less sensitive to F. (In the classical limit, the line shape is independent of F, and only an upper limit on n is possible.) This insensitivity, and other uncertainties of the fit, is reflected in the large error bars for the high temperature points in Fig. 5.

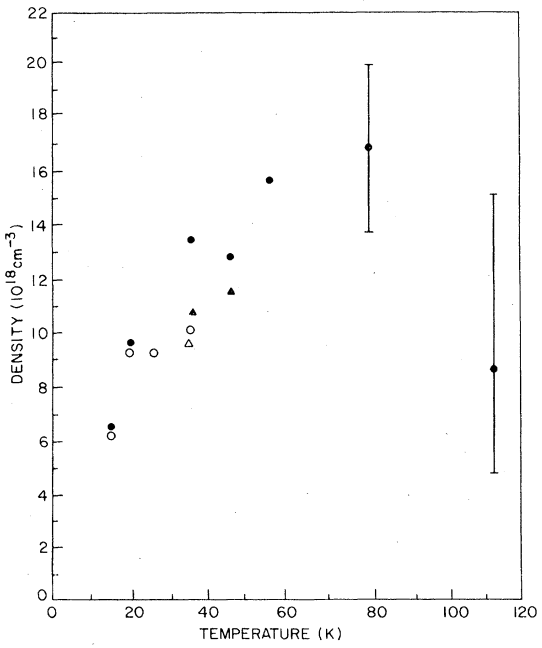

FIG. 5. Electron-hole pair density for $Al_{0.46}Ga_{0.54}As$ as a function of temperature. The density is deduced from E_F by using Eq. (1). The excitation energy was 2.1 eV. The excitation intensity for the open symbols is approximately 20 times weaker than that for the solid ones. Circles—zero delay; triangles—20 nsec delay. The gate width was 10 nsec in both cases.

The triangles in Fig. 5 show the density observed 20 nsec after the exciting pulse, by which time the integrated intensity has fallen by a factor of 20. The EHP density does not change significantly with delay, except at very high excitation levels. This result strongly suggests that the EHP density observed corresponds to its quasiequilibrium value. Furthermore, the EHP appears to coexist with the exciton gas. However, it is possible that the exciton emission that we observe simultaneously with the EHP emission comes from regions of the crystal where the excitation level is below the EHP threshold.

That the EHP is in quasiequilibrium is confirmed by the fact that the EHP density, as obtained from the line shape, varies little, if at all, with excitation in the range over which the EHP can be observed. Since the luminescent efficiency is found to vary only slowly with excitation over this range, it follows that either the excited volume, or the fraction occupied by the EHP, must increase with excitation intensity. In the former case, the EHP would penetrate the full thickness of the layer at high excitation level. Since the GaAs substrate acts as a perfect sink for exctiation, we would expect a marked falloff in luminescent efficiency, which is not observed. (A low luminescent efficiency *is* observed in layers less than about 1.5 μm thick.)

We have checked this point further with a sample in which a 1-μm layer with $x = 0.5$ is separated from the GaAs substrate by a sublayer with $x = 0.65$. In this case the EHP is confined (by the wider band gap of the sublayer) to the 1-μm-deep excited region, and thus the volume of the plasma cannot increase nor can excitons escape. We find that the EHP density is independent of excitation, as in thick layers, up to about 10^{17} photons/cm^2. (If the sublayer is not present, the luminescent efficiency is low, and it is difficult to establish the EHP at all with the excitation intensity we have available.)

It follows from the constancy of the density that the fraction of the excited volume occupied by EHP must increase with excitation, and it is

therefore less than unity. Thus there is a separation in the excited volume, into separate regions of EHP and (presumably) of exciton gas. From the present data we have no way of telling what the scale of these inhomogeneities is, but since the EHP is established at its final density within the 3 nsec exciting pulse, the scale cannot exceed 10^{-5} cm and may be much less.

The fitting parameter F and the density n deduced therefrom is plotted as a function of temperature, for the case $x=0.46$, in Fig. 5. A general increase in n with temperature is observed for all $x>x_c$, while n is independent of x. However, as x increases, the luminescent efficiency drops off at high temperature, so that for $x=0.51$ the EHP band has virtually disappeared by 70 K.

The renormalized bandgap, E'_{BB}, is shown as a function of x in Fig. 6. It depends somewhat on excitation level, as indicated by the vertical bars. E'_{BB} follows the bound exciton energy (E_{BX}) dependence on x. This means that for all indirect-gap $Al_xGa_{1-x}As$ crystals the electrons in the EHP are all confined to the X band minimum.

Another quantity related to the energetics of the EHP is the plasma binding energy relative to the free exciton, $\phi = E_{gx} - (E_{BB}+F)$. E_{gx}, the free-exciton energy, is difficult to measure in the indirect-gap $Al_xGa_{1-x}As$, since our layers are too thin for absorption across this gap to be detectable. The exciton moves in the random potential due to composition fluctuations. This results in inhomogeneous broadening of the exciton states and in a shift between emission and absorption. In direct-gap material E_{gx} can be determined directly from absorption measurements, and the emission peak E_{BX} is found to be about 10 meV lower. The emission band is slightly narrower in indirect material, so we assume its displacement from E_{gx} also to be 10 meV or less. Since $E'_{BB}+F$ is found to exceed E_{BX} by about 10 meV, it is clear that ϕ must be very small, and may even be negative, in which case the EHP at its equilibrium density will not be the stable phase at 0 K. The case of negative ϕ has been discussed by Combescot and by Reinecke and Ying,[13] who point out that the EHP can still exist at $T=0$, but only at sufficiently high excitation density. This is in agreement with our observation of a threshold excitation for the EHP even at low temperatures. However, they predict a critical temperature given by $kT_c \sim \frac{1}{10}\epsilon_g$, where $\epsilon_g = E_g - (E'_{BB}+F)$, E_g being the band gap. In our case $\epsilon_g \sim 20$ meV, so that T_c is expected to be about 20 K. We observe no critical behavior in this region; if there is a critical temperature it must be above 80 K (see Fig. 5).

The radiative recombination of the EHP is predominantly in the no-phonon band rather than in the momentum-conserving phonon sidebands, as it is in the other indirect-gap semiconductors, such as Ge, Si, GaP, and SiC. The origin of this no-phonon band is peculiar to a random alloy. As in the case of the exciton,[9] the transition becomes allowed through electron scattering from X to Γ band minima by the random potential fluctuations. Of interest here is the short plasma lifetime τ_p, rela-

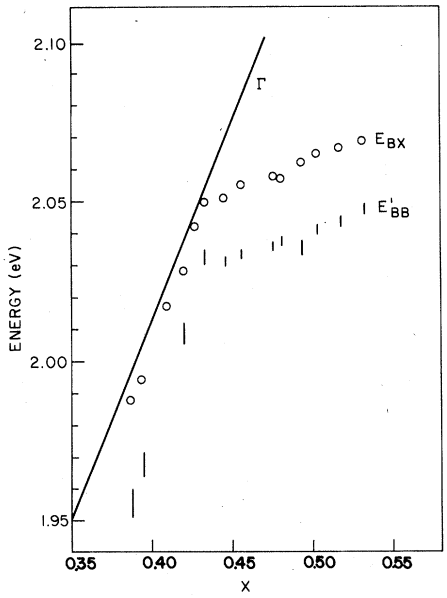

FIG. 6. Dependence on Al content x of the energy of the conduction-band minimum Γ, the intrinsic exciton E_{BX}, and the renormalized band gap E_{BB} ("bottom of the band") when the EHP is present.

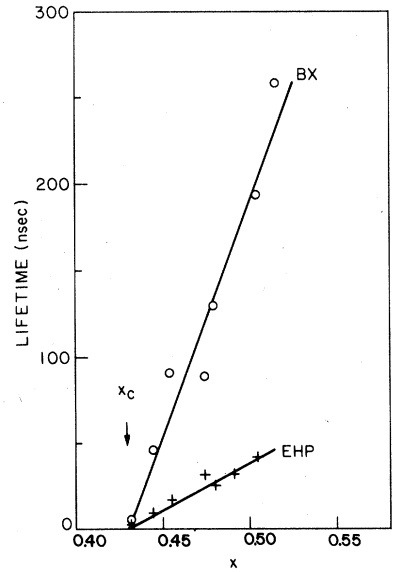

FIG. 7. Dependence on x of the exciton and EHP lifetimes at 4–5 K. For $x \leq x_c$ both lifetimes are less than our resolution (3 nsec).

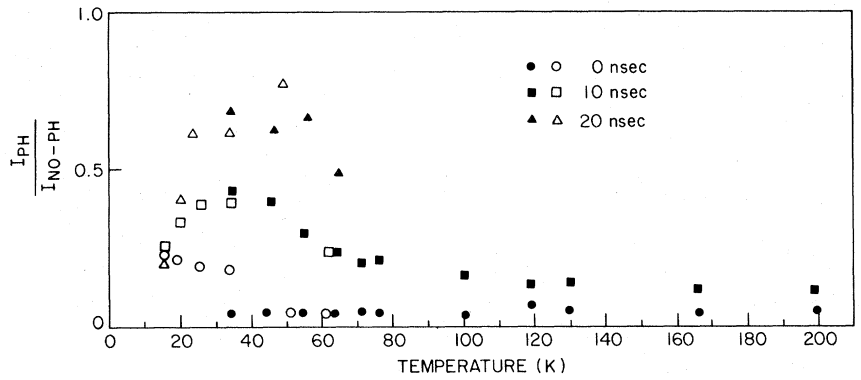

FIG. 8. Ratio of the peak intensity of the phonon sideband to that of the no-phonon EHP band as a function of temperature for various time delays (10 nsec gate). The excitation energy was 2.1 eV. The excitation intensity for the open symbols is 20 times weaker than that for the solid ones. $x = 0.46$.

tive to that of the "intrinsic" exciton, τ_{ex}. While we can expect a reduction in τ_p due to the nonradiative Auger process, several observations indicate that τ_p is mainly radiative at low temperatures. First, the integrated emission intensity is approximately linear in excitation density right through the evolution from BX emission to EHP emission, while the lifetime changes by a factor of 5. It seems unlikely that the nonradiative decay rate will change in precisely the same proportion as the radiative rate, particularly if it is an Auger process, so the nonradiative rate must be a small fraction of the whole. Secondly, as has been mentioned in Sec. II, stimulated emission is obtained even in the indirect material, and, while the threshold is a factor of 10–100 higher than in direct material, this can be accounted for qualitatively by the decrease in gain. Finally, τ_p increases with x, roughly proportionally to $E_1 - E_{BX}$ for $x \lesssim 0.52$, as shown in Fig. 7. This increase is qualitatively what is expected for a radiative rate arising from Γ-X admixture by a potential independent of x. It is not expected for an Auger process, which presumably limits τ at larger x.

If we assume that both lifetimes are purely radiative, we can deduce the "enhancement factor" g_{eh} from Eq. (16.3) of Ref. 1:

$$g_{eh} = \tau_x/(\tau_p n_0 \pi a_x^3).$$

Here n_0 is the plasma density and a_x the exciton radius. This factor is a measure of the electron-hole correlation, which makes the radiative rate exceed that expected from the average density. Taking $n_0 = 6\times10^{18}$ cm^{-3}, $a_x = 30$ Å, we find $g_{eh} = 10$, with an uncertainty of perhaps a factor 2. This is consistent with the values 5 to 7 obtained for Ge (Ref. 1) by less direct methods. The theoretical values for Ge and Si are 2.3 and 3.5, respectively.[14]

The other process which contributes to the radiative recombination under intense photoexcitation is the phonon-assisted transition. As can be seen in Figs. 2 and 3, the phonon sideband has roughly the same width as the no-phonon EHP band and is shifted by about 40 meV to lower energies. Thus, it would be natural to assume that this band is due to phonon-assisted transitions of the EHP, and consists of two overlapping bands, corresponding to the two phonons observed with the BX (see Fig. 1).[15] These phonons are close to the known optic phonons of $Al_xGa_{1-x}As$: $\hbar\omega$ (LO_1) = 37 meV and $\hbar\omega$ (LO_2) = 43 meV for $x = 0.5$.[16] However, the phonon sideband has a different time evolution when compared with the no-phonon band; its intensity grows relative to the no-phonon band as time elapses (within the short lifetime of the EHP). The temperature dependence of I_{PH}/I_{NP} for various delays and two excitation intensities for $Al_{0.46}Ga_{0.54}As$ is shown in Fig. 8. When we compare the results at low temperatures obtained with two excitation intensities we find that I_{PH}/I_{NP} is larger for the weaker excitation. The intrinsic exciton BX for $x \lesssim 0.47$, has no perceptible phonon sidebands under pulsed excitation. It follows that there is some other luminescent entity, present only at intermediate excitation levels, which has a relatively strong phonon sideband and a lifetime longer than the EHP, though shorter than the BX.

Although the spectroscopic data are insufficient to determine the properties of this entity, we can speculate that it may consist of multiexciton complexes. These presumably appear in regions in which the density of photoexcited electron-hole pairs is too small to condense into a plasma. Bound multiexciton complexes, such as the biexciton bound to isoelectronic nitrogen in GaP, have been extensively studied.[10,17] The potential fluctuations in a random alloy provide a wealth of binding sites for these complexes; however, it is not clear why the no-phonon transitions of these complexes should be so much weaker, relative to

the phonon-assisted transitions, than those of the BX. Until this point is settled, this interpretation can only by very tentative.

VI. CONCLUSIONS

We have shown that an electron-hole plasma phase of a definite density exists in highly excited indirect-gap $Al_xGa_{1-x}As$ over a wide range of excitation. Below 20 K, exciton and plasma emission can be observed simultaneously. While this probably implies that the excitons and plasma coexist in quasiequilibrium, it could be due to nonuniform sample excitation. The binding energy of the plasma with respect to the free exciton, determined spectroscopically, is small or negative. The plasma also differs from the liquid observed in Ge and Si in that its density increases, rather than decreases, with temperature, and no critical point is detectable up to 100 K. This difference may be related to the bandgap fluctuations present in a random alloy, or perhaps to the short time scale of our experiment.[18] Near Γ-X crossover the decay is predominantly through no-phonon radiative recombination, and in consequence stimulated emission can be obtained in this indirect-gap material.

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