Properties of the electron-hole plasma in GaAs-(Ga,Al)As quantum wells: The influence of the finite well width

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Photoluminescence spectra of the electron-hole plasma confined in GaAs-(Ga,Al)As quantum wells are presented. The spectra have been successfully fitted using a single-plasmon pole approximation. The results clearly show that the band-gap reduction $\Delta E_g(w)$ depends on the width w of the well and is smaller than in a strictly two-dimensional plasma. Further, theoretical estimations of $\Delta E_g(w)$ are in good agreement with our experimental results.

INTRODUCTION

The properties of the electron-hole (e-h) plasma in bulk semiconductors are well known at present (for a review, see, e.g., Ref. 1). Exchange and correlation effects in the metallic plasma induce a band-gap reduction. This reduction, expressed in terms of the exciton Rydberg and Bohr radius, is universal, i.e., it does not depend on the peculiarities of the band structure.[2,3] Further, the random-phase approximation (RPA) simplified by a single-plasmon pole approximation (SPPA) has proven to be useful for the metallic e-h plasma.[1,4,5] As in the bulk, the analysis of the luminescence spectra of an e-h plasma confined by a quantum well (QW) can be done using a SPPA.[6] Our results as well as theoretical considerations[6,7] indicate that the electronic properties of the e-h plasma depend on the width w of the QW, in contrast to recent claims.[8]

In this letter we propose a new analysis of the photoluminescence spectra of highly excited QW's having different widths. The unperturbed continuum edges are evaluated with the help of recent experimental[9,10] and theoretical[11] results. The line shape is fitted with a model which neglects excitonic enhancement and plasmon sidebands, but takes into account the renormalized single-particle energies and the e-h collision lifetime at finite temperatures.[4,5] We find a band-gap reduction $\Delta E_g(w)$ which agrees well with theory. Introducing an effective two-dimensional (2D) exciton Rydberg $E_0^{(2)\mathrm{eff}}(w)$ that is consistent with our SPPA, we show below that $\Delta E_g(w)/E_0^{(2)\mathrm{eff}}(w)$ does not depend on the properties of the QW.

EXPERIMENTAL

For our photoluminescence experiments we used three different multiple-quantum-well structures grown by molecular-beam epitaxy: their properties are listed in Table I. The exciton energies were measured by optical transmission at a temperature of 2 K. In sample (b) even the transitions to the first-excited exciton states could be detected (see Fig. 1). The luminescence measurements were performed in the usual way (see, e.g., Ref. 12). The samples were immersed in superfluid He. The e-h plasma was excited directly inside the QW by a pulsed dye laser; its photons had an energy in the range between 1.68 and 1.77 eV. For these excitation energies the light absorbed by 25 wells amounts to about 30% and, thus, the decrease of the carrier density with depth is negligible. The laser-pulse duration was about 50 ns and therefore the e-h pairs were in a quasistationary state.

In Fig. 1 plasma-emission spectra of sample (b) for high pump intensities are shown. With respect to the exciton spectra at low intensities, the luminescence lines are much wider (width 30–40 meV) and a low-energy tail appears. This indicates the formation of a dense e-h plasma. With increasing excitation power, the luminescence emission broadens because of band filling; however, the spectra do not show the sharp edge which characterizes the 2D density of states. This is a consequence of the collision broadening of the single-particle states, which is caused by the high carrier temperature and density. Such high temperatures are typical for e-h plasmas confined in a QW.[8]

THEORY

The luminescence spectra have been analyzed, making the following assumptions. (i) The electrons and holes are in quasithermodynamic equilibrium (they both have the same temperature). (ii) For the effective masses we use the "bulk" values proposed in Ref. 8. (iii) For the line-shape fit, the single-plasmon pole model described in Ref. 1 has been used. This model includes collision broadening of the single-particle states, but neglects excitonic enhancement. This latter approximation is justified by the high temperature and density of the carriers; further, in another paper[13] we showed that the Coulomb enhancement does not significantly influence the luminescence line shape. For bulk GaAs and (Ga,Al)As alloys, this model provided a satisfactory explanation of the luminescence spectra.[4] (iv) The one-particle spectral function $A_a(k,E_a)$ is a Lorentzian of width Γ whose low-energy tail is cut off one plasmon energy below the reduced gap.[5] (v) Only the lowest electron, heavy-hole and light-hole subbands have been considered; higher subbands are not significantly populated. Thus, for the luminescence spectrum of an e-h plasma confined in a QW, we write

TABLE I. Properties of the multiple quantum well structures used in the experiment. The excitonic data are for electron-hole pairs of the lowest conduction and the highest valence subbands. Numbers given in parentheses have not been used for the determination of the energy of the lowest continuum pair state.

Sample	(a)	(b)		(c)
Wells:				
number	20	25		29
width (Å)	50	80		122
Barriers:				
width (Å)	220	120		180
Al content	0.38	0.25		0.23
Exciton energies	fundamental	fundamental	excited	fundamental
from optical transmission				
heavy (eV)	1.619	(1.575)	1.585	1.542
light (eV)	1.640	(1.593)	1.603	1.549
binding energies				
heavy (meV)	13	(10)	1.7	9
light (meV)	14	(11)	2.8	10
references	9	9	11	9
lowest continuum edges				
heavy (eV)	1.632	(1.586)	1.587	1.551
light (eV)	1.654	(1.605)	1.606	1.559

$$I(\hbar\omega) \propto \sum_{h=\mathrm{HH,LH}} |M_{e\text{-}h}|^2 \int d^2k \int dE_e\, A_e(k,E_e)F_e(E_e) \int dE_h\, A_h(k,E_h)F_h(E_h)\delta(\hbar\omega - E_e - E_h) \ . \quad (1)$$

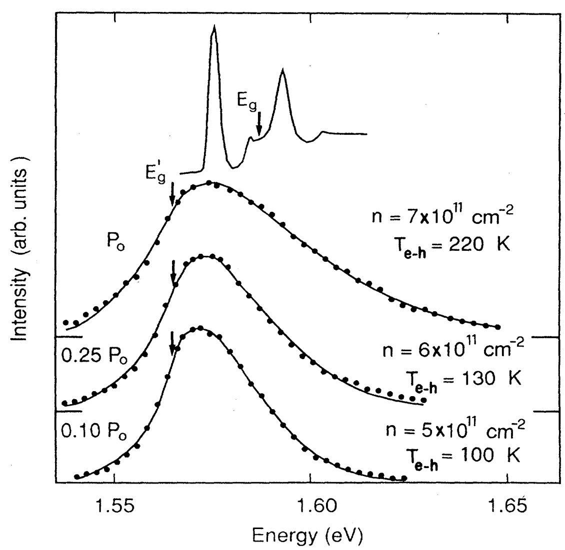

FIG. 1. Photoluminescence spectra of a strongly excited GaAs-$Ga_{0.75}Al_{0.25}$As multiple-quantum-well structure. The 25 wells have a width of 80 Å each. The maximum absorbed light intensity is $P_0 = 100\ \mathrm{kW\,cm^{-2}}$ per well. At the top, an experimental low-density transmission spectrum is also shown. The dots are experimental data. The three lower solid curves are fitted as explained in the text. The renormalized gap E_g', the electron-hole–pair density n and the carrier temperature $T_{e\text{-}h}$ have been extracted from the fit.

Here, $\hbar\omega$, E_e, and E_h are the energies of the emitted photon, the e and the h, F_a is the Fermi-Dirac distribution function, and $M_{e\text{-}h}$ are the interband matrix elements (taken as k independent). The latter have been determined from the low intensity transmission spectra.[13]

As can be expected from theoretical considerations, our experimental results suggest that the finite well width affects the gap renormalization in a non-negligible way. We estimated this dependence theoretically following Ref. 6, and considering only the lowest electron and heavy-hole subbands. In this approximation the calculations can be done as in the pure 2D case, provided an effective (mean) 2D interaction potential is introduced:

$$\overline{V}_{a\text{-}b}(r_\parallel) = \int V_{a\text{-}b}(r_\parallel, z - z')|\phi_a(z')|^2 |\phi_b(z)|^2 dz\, dz' \ . \quad (2)$$

Here, a and $b = e$ or h, the z direction is perpendicular to the well plane, $r_\parallel = (x^2 + y^2)^{1/2}$ is the projection of the interparticle distance onto the well plane,

$$V_{a\text{-}b}(r_\parallel, z - z') = (e_a e_b/\epsilon)[r_\parallel^2 + (z - z')^2]^{-1/2}$$

is the 3D Coulomb potential, ϵ is the dielectric function, and $\phi_a(z)$ is the wave function which describes the motion in the z direction of a single particle in the unperturbed QW with finite barriers. The gap reduction $\Delta E_g(w)$ has been computed for zero temperature and equal e and h masses in the dynamical RPA and in the Hubbard modification[14] (to include the short-range correlation effects in a better approximation). As shown in Fig. 2. the two approximations yield practically equivalent results; thus short-range correlations seem to be negligible. The difference between the wells of finite width and the 2D limit is quite important; in any case the 2D limit is never reached for finite barriers. As for the widths of the single-particle spectral functions $A_a(k,E_a)$ at $k_\parallel=0$, calculated in the RPA at zero temperature, for a plasma of 10^{12} e-h pairs per cm^2, we obtain 5.7 meV for the electrons and 0.6 meV for the holes.

To fit our experimental spectra, we used a dynamical SPPA.[15] Here, following Ref. 6, we introduced directly the shift of the 2D plasma frequency caused by the effective interaction potential:[2]

$$\Omega_p^2(k_\parallel)=(2\pi e^2/\epsilon)k_\parallel \sum_a (n_a/m_a) \times \int dz\, dz' |\Phi_a(z)|^2 \times |\Phi_a(z')|^2 e^{-k_\parallel|z-z'|} , \quad (3)$$

where $k_\parallel$ is the wave vector of the 2D plasmon, and m_a and n_a are the effective mass and the 2D density of the carriers of species a. For the effective plasmon dispersion in our SPPA, we used[15]

$$\Omega_{\rm eff}^2(k_\parallel)=\Omega_p^2(1+k_\parallel/q_{\rm sc})-\Gamma_0^2+k_\parallel^4/16m^2 , \quad (4)$$

where $q_{\rm sc}$ is the bidimensional screening wave number. The so-called plasmon damping constant Γ_0 was adjusted to obtain a good overall agreement of the imaginary part of the self-energies at zero temperature calculated in our

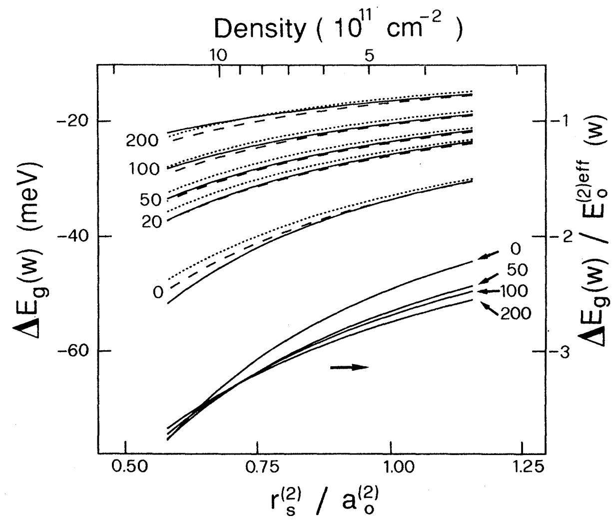

FIG. 2. Calculated band-gap shift $\Delta E_g(w)$ vs 2D interparticle distance $r_s^{(2)}\equiv(\pi n)^{-1/2}$ in a GaAs-Ga$_{0.6}$Al$_{0.4}$As QW. The results of the calculations made at zero temperature and for equal e and h masses in the SPPA, the RPA, and the Hubbard modification are shown as solid, dashed, and dotted curves, respectively. The numbers indicate w in Å. The curves at the bottom are the SPPA results plotted in relative units, $E_0^{(2)\rm eff}(w)$ is the effective binding energy of the exciton in the well of width w (see text), and $a_0^{(2)}$ is the Bohr radius of the strictly 2D exciton.

SPPA with those obtained in the RPA. The Γ_0 determined in this way was used only for the imaginary part of the self-energy; for the real part (which depends only weakly on the plasmon damping constant), Γ_0 was set to a very small value. As in three dimensions,[1] Eq. (4) fulfills the most relevant sum rules.

As mentioned above, in a 3D e-h plasma the band-gap renormalization versus carrier density, measured in excitonic units, is nearly independent of the peculiarities of the band structure.[2,3] In particular, the exciton Rydberg $E_0^{(3)}$ is a measure of the strength of the interaction between quasiparticles in a particular crystal. In a QW the effective 2D interaction [Eq. (2)] depends on the well width w (and weakly on the barrier height). Thus, as unit of energy (and interaction strength) the binding energy $E_0^{(2)\rm eff}(w)$ of an e-h pair interacting via this effective 2D potential has to be used [in order to be consistent with our model, the evaluation of $E_0^{(2)\rm eff}(w)$ must not include conduction-band nonparabolicity or subband mixing]. $E_0^{(2)\rm eff}(w)$ has been calculated numerically by Chan.[16] The plots in Fig. 2 show that $\Delta E_g(w)/E_0^{(2)\rm eff}(w)$ does not depend on w. At first, it might be suprising that as unit of length the Bohr radius $a_0^{(2)}$ of a strictly 2D exciton has to be used [and not $a_0^{(2)\rm eff}(w)$]. Our numerical results, however, show that in our approximation the w dependence of exchange and correlation energy [measured in units of $E_0^{(2)\rm eff}(w)$, of course] cancels out to a large degree. Finally, we mention that also the gap reduction given by Klingshirn *et al.*[17] is quite higher than ours.

DISCUSSION

For the least-squares fit of the luminescence spectra, energy gap, plasma density, and temperature have been taken as free parameters. The energy renormalizations of the heavy- and light-hole subbands have been assumed to be equal. Some of the obtained fits are shown in Fig. 1; they are quite good. The wide low-energy tail is well accounted for by the higher values of the e and h collision broadening parameter Γ_0: for $n=1\times10^{12}$ cm^{-2} and $T_{e\text{-}h}=100$ K our SPPA yields 8.6 meV for the electrons and 5.0 meV for the holes.[18] This is much more than the values used in Ref. 17. Plasma temperatures $T_{e\text{-}h}$ up to 250 K have been found. $T_{e\text{-}h}$ increases with carrier density and with the difference between photon and gap energy. Further, it depends on crystal quality; the highest $T_{e\text{-}h}$ has been reached in the high-quality sample, (b). Thus, as in bulk samples, the presence of defects seems to accelerate carrier-lattice relaxation.[4]

In Fig. 3 the band-gap shift $\Delta E_g(w)$ is reported as a function of the e-h –pair density n. For all three samples, the values extracted from the fit agree with those calculated for $T_{e\text{-}h}=100$ K using the SPPA described above. In particular, the experimentally determined $\Delta E_g(w)$ is much smaller than that predicted by a strict 2D calculation; the difference is far beyond the uncertainty of the fit. Moreover, the band-gap renormalization increases with decreasing well width. The dependence of the measured $\Delta E_g(w)$ on n seems to be weaker than the calculated one, especially for sample (b). However, if the strong increase of the temperature $T_{e\text{-}h}$ with carrier density n is account-

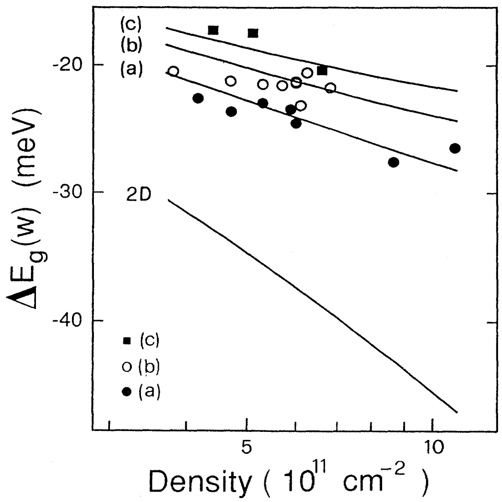

FIG. 3. Reduction of the band-gap width $\Delta E_g(w)$ induced by an *e-h* plasma confined in a GaAs-(Ga,Al)As QW. The curves have been calculated in the SPPA using the "bulk" masses of Ref. 8 for a carrier temperature of 100 K; the labels on the left refer to the samples listed in Table I (2D refers to the two-dimensional case). The dots are experimental data obtained from a numerical analysis of the luminescence spectra as explained in the text.

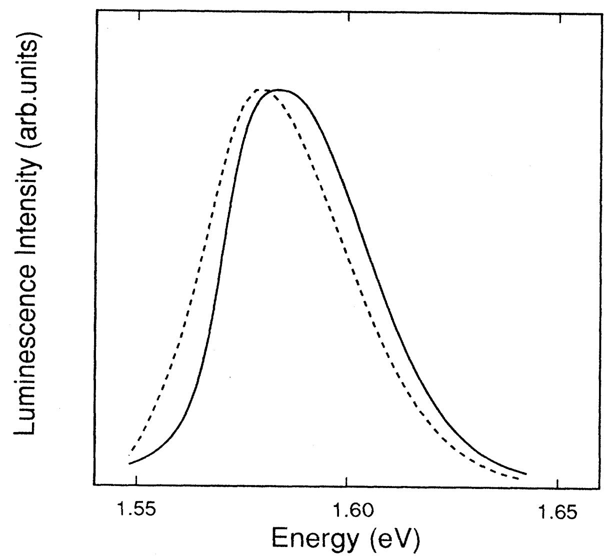

FIG. 4. Calculated luminescence spectra of an *e-h* plasma confined in a well of 80 Å width and having a density of 7.5×10^{11} cm^{-2} and a temperature of 115 K. Dashed line, spectrum computed using Eq. (1); solid line, spectrum computed following Ref. 8 (see text).

ed for in the calculations, the agreement between experiment and theory is better. For example, the calculated gap reduction in a well 80 Å wide [sample (b)] amounts to 20 meV for $n=4\times10^{11}$ cm^{-2} and $T_{e\text{-}h}=50$ K, and also to 20 meV for $n=7\times10^{11}$ cm^{-2} and $T_{e\text{-}h}=250$ K. For the other two samples [(a) and (b)] the temperature increase with n is much lower (about 50 K), and, therefore, the agreement between the theoretical and experimental width dependence of ΔE_g is better. In any case, the deviation between the experimentally and theoretically determined gap reduction is not bigger than 10%.

Tränkle *et al.*[8] reported band-gap reductions for GaAs-(Al,Ga)As multiple-QW (widths of 21, 41, and 83 Å) structures which are in agreement with the 2D limit. This contrast with our results is caused by three differences in the way the optical spectra have been analyzed. (a) The exciton binding energies used in Ref. 8 were obtained from low-field extrapolations of magneto-optical measurements.[19] It has been shown recently that this technique can strongly overestimate the exciton binding energy.[10] (b) To determine the single-particle collision broadening parameter Γ, in Ref. 8 Landsberg's analytic zero-temperature formula is used.[20] In this approximation Γ vanishes at the chemical potential. At finite $T_{e\text{-}h}$ this causes an overestimation of the joint density of states on the high-energy side of the luminescence emission. As pointed out above, the observed plasma temperatures are of the order of 100 K, and thus a 0-K approximation should not be used. (c) Evaluating the luminescence intensities $I(\hbar\omega)$ [Eq. (1)], in Ref. 8, $\Gamma \ll k_B T_{e\text{-}h}$, i.e., $F(E)\simeq F(\hbar^2k^2/2m)$, has been assumed. This again overestimates $I(\hbar\omega)$ on the high-energy side of the emission. As an example, in Fig. 4 we show the luminescence spectrum of an *e-h* plasma ($w=80$ Å, $n=7.5\times10^{11}$ cm^{-2}, $T_{e\text{-}h}=115$ K) calculated once using Eq. (1) and once making the above-mentioned approximations (b) and (c). The two spectra are shifted by 5 meV (for the plasma concentrations discussed in this work, the shift varies between 4 and 7 meV). The overestimation of the exciton binding energy amounts to 5–3 meV.[9] Thus the sum of the three points (a)–(c) leads to an apparent gap reduction which is closer to the 2D limit than the real value. On the other hand, the reasons why the $\Delta E_g(w)$ reported in Ref. 8 is almost independent of the well width are not clear at present.[21]

CONCLUSION

Our investigation of the *e-h* plasma confined in GaAs-(Al,Ga)As QW's yields the following results. (i) Measured in absolute units, the gap reduction $\Delta E_g(w)$ (practically, exchange plus correlation energy in the plasma) depends on the width w of the QW. It is much smaller than it would be in a strictly 2D *e-h* plasma. (ii) Scaled to an appropriate exciton Rydberg $E_0^{(2)\text{eff}}(w)$, the gap renormalization becomes independent of the QW width. (iii) The single-plasmon pole approximation, in spite of its simplicity, reproduces the experimental results well.

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[18]The wide low-energy tail below the band gap observed in the luminescence spectra of the plasma is directly related to the single-particle broadening; therefore it must also appear in the transmission spectra (Ref. 13). On this point there seems to be a fundamental difference between the results presented in Ref. 17 on one hand, and those of Ref. 8 and ours on the other. Actually, we do not understand this difference. In any case, the precision in the determination of the energy-gap reduction depends on the way the collision broadening has been accounted for.

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