Phase diagram of a two-dimensional liquid in GaAs/$Al_xGa_{1-x}As$ biased double quantum wells

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Photoluminescence (PL) and PL excitation (PLE) measurements have been performed in GaAs/$Al_xGa_{1-x}As$ biased double quantum well heterostructures. The recombination of electrons, *e*, with holes, *h*, located in the same or in two adjacent wells, has been investigated for different exciting power densities, *P*, and temperatures, *T*. For increasing *P* or decreasing *T*, a sharp transition from two gases of photoexcited electrons and holes, spatially separated and confined in the two wells, to two two-dimensional (2D) liquids has been observed. The gas-to-2D-liquid transition is evidenced by a strong screening of applied biases and by major changes in the optical spectra. The phase diagram in the (P,T) plane of the *e*-*h* system has been determined. Time-resolved PL, cw PL, and PLE in the presence of a magnetic field normal to the quantum wells support the presence of *e*- and *h*-liquid phases in the two wells with a critical density equal to 8.8×10^{10} cm^{-2} and a binding energy of 2.5 meV.

I. INTRODUCTION

Symmetric and asymmetric double quantum wells (DQW's) have been the object of extensive study in the last decade,[1–6] since their properties are very attractive on purely fundamental grounds. In particular, in biased DQW's excitons can be photoexcited where electrons and holes are confined in adjacent QW's and, therefore, spatially separated (interwell excitons). These excitons should be characterized by radiative decay times much longer than those of intrawell excitons, where both carriers are in the same well. Interwell excitons, therefore, can attain a high density at low *T*, even in a two-dimensional (2D) system, and condensate as theoretically predicted.[7–12] Some claims of the observation of collective phenomena in a gas of interacting interwell excitons have been reported, indeed, in the last years.[3–6]

In the present paper, low-*T* photoluminescence (PL) measurements are performed in biased GaAs/$Al_xGa_{1-x}As$ double quantum well heterostructures. These measurements provide a strong evidence that interwell excitons undergo a phase transition from a gas phase to a 2D liquid phase. This transition is sharp, both in exciting power density, *P*, and temperature, *T*. It is characterized by clear changes in the PL spectra and by a strong screening of the electrical field inside the *p*-*i*-*n* DQW structure. The presence of a liquid has been confirmed by time-resolved experiments and by an analysis of PL and PL excitation (PLE) spectra under a magnetic field applied along the growth direction. Characteristic energy and thermodynamic parameters of the liquid have also been evaluated.

II. EXPERIMENTAL DETAILS

p-*i*-*n* GaAs/$Al_xGa_{1-x}As$ ($x=0.35$) DQW heterostructures have been grown by molecular beam epitaxy on *n*-type GaAs(001) substrates (2×10^{18} cm^{-3} Si doping). In all structures, the sequence of layers is the following: 0.5 μm *n*-type GaAs buffer layer (10^{18} cm^{-3} Si doping); 0.3 μm $Al_xGa_{1-x}As$ insulating barrier; 8 nm GaAs well; 5 nm AlGaAs barrier; 8 nm GaAs well; 0.3 μm $Al_xGa_{1-x}As$ insulating barrier; 100 nm *p*-type GaAs top contact layer (10^{18} cm^{-3} Be doping), and finally a 5 nm GaAs cap layer. Ohmic Au contacts have been made to *n* and *p* regions in 0.1×0.1 cm^2 lithographically etched mesa. In dark as well as at low photoexcitation power with photon energies smaller than the $Al_xGa_{1-x}As$ barrier band gap, the samples exhibit a *p*-*i*-*n* diode I-V characteristic ($I \lesssim 10$ nA for applied biases ranging from $+0.8$ to -1 V). Steady-state excitation conditions were always obtained by a tunable Ti-sapphire laser. Time-resolved experiments were carried out with the use of a picosecond laser ($\lambda=625$ nm) and a time-correlated photon counting system. The emitted light was dispersed by a double monochromator and detected by a charge coupled device camera (PL) and/or a cooled GaAs photomultiplier (PLE and time-resolved PL spectra). For magneto-optical studies, an optical cryostat with a superconducting split-coil solenoid ($B\leqslant7$ T) has been used, with the magnetic field parallel to the heterostructure growth direction. All magneto-optical measurements were carried out in a Faraday geometry with circular light polarization of the PL and PLE spectra.

III. EXPERIMENTAL RESULTS

First, we present PL spectra taken at different temperatures and fixed excitation power, always near the gas-to-2D liquid phase transition. PL measurements have been performed with homogeneous *resonant* excitation of intrawell light hole (1*s*LH) excitons. Crossed slits have been used in

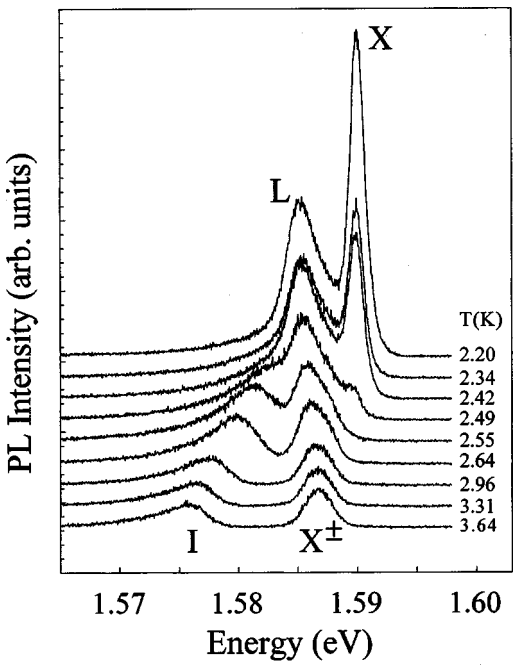

FIG. 1. Photoluminescence spectra under resonant excitation ($E_{exc}=1.613$ eV) of the intrawell 1*s*LH-exciton state measured at different temperatures in the range (3.64–2.2) K. $P=1\times10^{-2}$ W cm^{-2}, applied direct bias equal to 0.7 V.

order to select the radiation emitted by a 100×100 μm^2 spot at the center of the photexcited region of the mesa. PL spectra have been measured between 3.64 and 2.2 K in steps of ~0.1 K. Some of these spectra are displayed in Fig. 1 in the case of a low excitation-power intensity, $P=1\times10^{-2}$ W cm^{-2}. Between 3.64 and 2.55 K, only interwell (I) and charged ($X^{\pm}$) excitons are observed. The *I*-band energy depends linearly on the applied bias (not shown here) as expected for interwell excitons which are formed by electrons and holes confined in different, adjacent QW's. It has been shown that, in the present case of low *P* and *T*, these interwell excitons are localized by random potential fluctuations, source of an inhomogeneous broadening.[13,14] Moreover, free excitons (X) may bind to localized carriers and form three-particle exciton complexes, or trions, which are responsible for the $X^{\pm}$-band emission in Fig. 1. Between 3.64 and 2.55 K, at low *P*, both interwell excitons and localized trions are more stable than free excitons, whose emission is observed typically at $T\gtrsim10$ K. In that case, indeed, the thermal energy becomes comparable to the trion binding energy and trions are ionized [$X^{\pm}\rightarrow X+e(h)$]. Interwell excitons and trions in this regime of intermediate-to-high temperatures are discussed in detail elsewhere.[15]

The main object of interest of the present paper is the phase transition from a gas of interwell excitons to an interwell exciton condensate, as discussed in the following. In a very narrow interval of *T*, namely, for *T* going from 2.55 to 2.42 K, the PL spectrum undergoes a series of major changes. At $T=2.49$ K the interwell *I* band virtually vanishes, while an intrawell exciton band abruptly appears, the intensity of which increases for decreasing *T*. At the same time, a new PL band (*L* in Fig. 1) gradually replaces the trion band from which it differs with respect to spectral position, shape, and linewidth. In the *T* range where PL spectra exhibit those drastic changes, the photocurrent in the *p-i-n* DQW circuit smoothly varies by a small amount (from 8 nA at 6 K to 3 nA at 2 K). There is no relation, therefore, between the PL changes observed in the photoexcited *e-h* system across the critical temperature $T_c=2.49$ K and the current flowing in the circuit.

For $T\geqslant T_c$ and $P=1\times10^{-2}$ W cm^{-2}, the photoexcited electron-hole system is rather dilute, with an upper limit to the average *I*-exciton density $n_c\leqslant8\times10^{8}$ cm^{-2}. This value has been estimated by assuming that the interwell-exciton decay time is of the order of 2×10^{-8} s and that resonant photoexcitation at 1.61 eV is fully converted into *I* excitons in the QW's. Such a diluted, homogeneous gas cannot screen significantly the electric field applied to the *p-i-n*-structure. In fact, at $T=3.64$ K and for a forward bias of $+0.7$ V, the interwell *I* band is lower than the free exciton band *X* by as much as 14 meV, see Fig. 1, which corresponds to a Stark shift produced by an electric field in the DQW equal to 16 kV/cm. The disappearance of the *I* band below T_c clearly indicates, therefore, a strong screening of the electric field in the *p-i-n* structure. This is possible only in the presence of a rather dense system ($\sim10^{11}$ cm^{-2}) of photoexcited carriers where, most likely, electrons and holes are spatially separated between adjacent QW's.

From a theoretical point of view, the system of two homogeneous, rather dilute gases of photoexcited electrons and holes localized each in a different QW becomes unstable at T_c toward an inhomogeneous redistribution of electrons and holes.[9] In particular, below T_c the system divides into two phases, the former with a low, the latter with a rather high carrier density. Some indications of such an instability can be found in the PL spectra reported in Fig. 1 where, for $3>T>2.5$ K, the *I* band of interwell excitons begins to move toward high energies and to broaden for *decreasing T* before vanishing below T_c. In the following, we will demonstrate that the new band *L* observed below T_c is due to two 2D liquids of electrons and holes where electrons and holes are spatially separated and confined in adjacent QW's.

The phase boundary in the (P,T) plane of the gas-to-2D-liquid transition has been defined by determining at different *P*'s the abrupt changes in the PL spectra which characterize T_c. The resulting phase diagram is shown in Fig. 2 for an applied bias of $+0.7$ V. The critical temperature increases fast with *P* at low power density, then it asymptotically saturates at ~8.5 K at the highest *P*. In the upper part of the phase diagram, namely, for $T>T_c$, all bands are due to charged excitonic complexes, to interwell recombination of spatially separated *e-h* pairs, or, at higher *T*, to intrawell excitons. In the lower part of the phase diagram, namely, for $T<T_c$, bands due to two condensed carrier liquids and free excitons dominate the PL spectra. A sharp transition in the PL spectra, similar to that reported in Fig. 1 for decreasing *T* at fixed *P*, has been observed also when crossing the phase boundary by increasing *P* at fixed *T* (the transition takes place for a change of *P* of the order of $\pm15\%$ at high-power density, of $\pm5\%$ at low-power density). Preliminary *C-V* measurements performed in the same DQW's simultaneously with PL measurements show that the capacitance of the system changes sharply by roughly 25% at the gas-to-2D-liquid transition. No sharp changes have been observed,

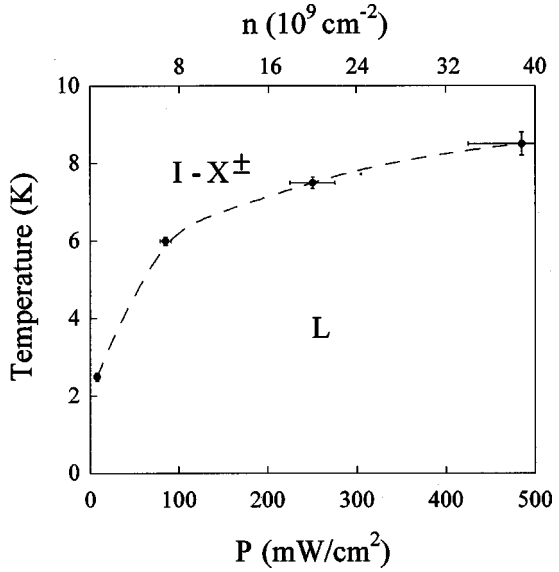

FIG. 2. Phase boundary dividing a diluted exciton phase (interwell excitons, I, and charged exciton complexes, $X^{\pm}$) from a dense 2D liquid phase with electrons and holes separately confined in adjacent QW's. The applied direct bias is 0.7 V. The upper scale provides an estimate of the upper limit of the I-exciton density. These values have been obtained by assuming that the decay time of the I excitons is of the order of 2×10^{-8} s and that photoexcitation at 1.61 eV, resonant with the 1sLH, is fully converted into I excitons in the QW's.

instead, when C-V measurements have been performed in the dark. This behavior confirms the presence of a phase transition in the system when excess carriers are photoexcited.

The critical density for the transition to a 2D liquid, as estimated from the excitation intensity P at the phase boundary at fixed T, is roughly independent of the applied forward bias, while it increases with an applied reverse bias. This supports the transition of the two diluted gases of e and h to two 2D liquids where electrons and holes are spatially separated and confined in adjacent QW's, against the formation, e.g., of a *single*, neutral e-h liquid in one or both the QW's. In fact, a reverse bias lowers the energy of interwell excitons, I. This requires a lower energy of the interband liquid before condensation can take place and therefore a higher critical density (and excitation power), as experimentally found. On the other hand, a reverse bias should not much affect a neutral liquid in one well, as it does not much affect the free excitons, X.

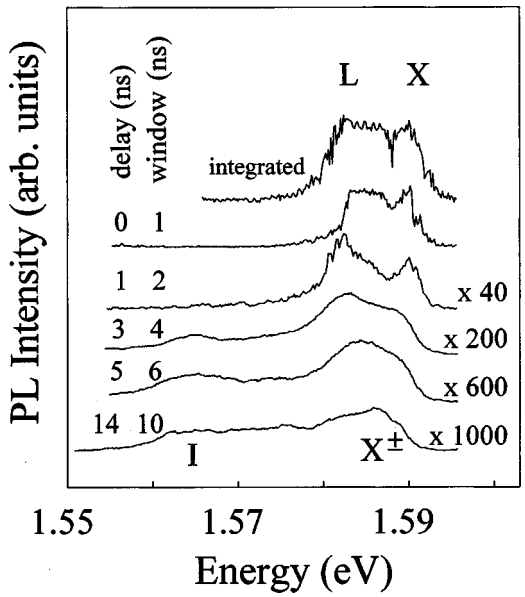

FIG. 3. Time-resolved PL spectra taken at 2 K under pulsed picosecond laser excitation ($E_{exc}=1.983$ eV, 30 ps pulses, $P=1\times10^5$ W cm^{-2}, $V=0$). The upper spectrum is integrated, lower spectra are measured with different windows and at different time delays with respect to the excitation laser pulse.

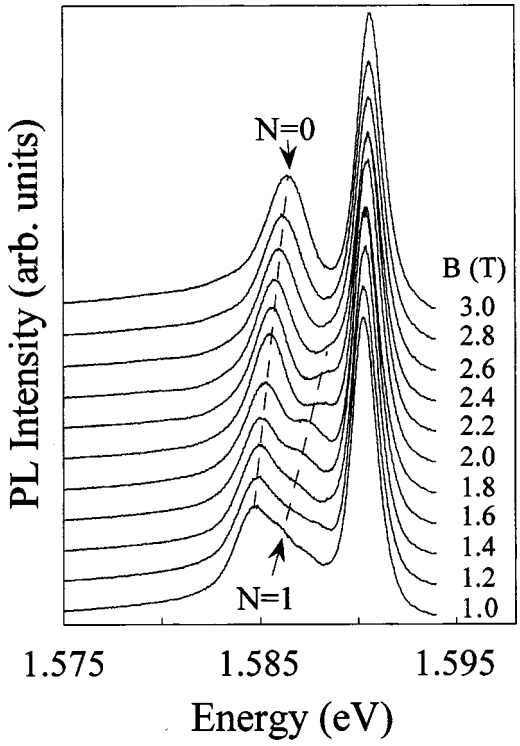

FIG. 4. PL spectra at $T=2$ K measured for different magnetic fields B from 1 to 3 T. ($E_{exc}=1.613$ eV, $P=1\times10^{-2}$ W cm^{-2}, applied direct bias equal to 0.7 V). The magnetic field is applied parallel to the sample growth direction. Dashed lines join different $N=0$ and $N=1$ occupied Landau level states.

The results of time-resolved PL experiments (laser pulses of a few picoseconds and wavelength $\lambda=625$ nm) are reported in Fig. 3 for different delay times, together with a cw, integrated spectrum. Emission from the L and X band dominates the time-resolved spectra at high excitation intensity, up to a 4 ns delay, where it exhibits a roughly constant spectral width. For delays greater than 4 ns, the trion line $X^{\pm}$ and interwell line I appear and begin to increase in intensity. Preliminary time-decay measurements at the emission energy of the L band have also been performed. After an initial fast decrease, with a characteristic time of the order of 1 ns, the signal decreases quite slowly, with a lifetime of about 20 ns, much longer than decay times typical of intrawell free excitons (100 ps). The above features confirm that, for decreasing excitation intensity, the system undergoes a drastic transition from a 2D-liquid phase to a gas phase.

The behavior of the L band in a magnetic field B perpendicular to the QW planes has been then studied. Low-

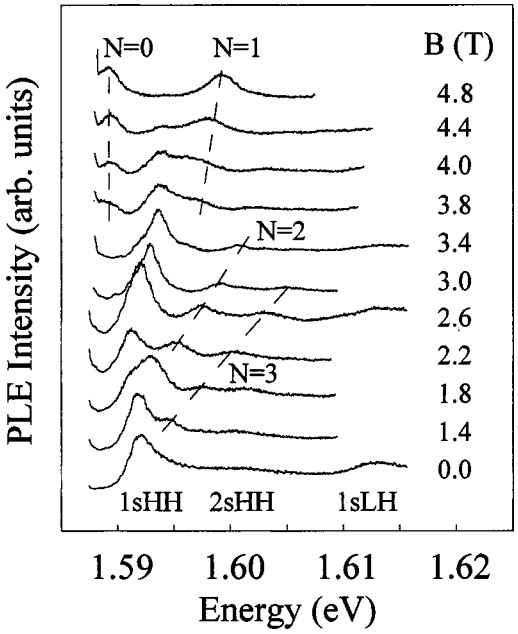

FIG. 5. PLE spectra measured at $T=2$ K for different magnetic fields B from 1.4 to 5 T ($P=1\times10^{-2}$ W cm^{-2}, applied direct bias equal to 0.7 V). The magnetic field is applied parallel to the sample growth direction. Different Landau level structures corresponding $N=0$, 1, 2, and 3 states of the liquid phase are joined by dashed lines. A PLE spectrum at $B=0$ T showing 1*s*HH, 2*s*HH, and 1*s*LH exciton bands is also reported.

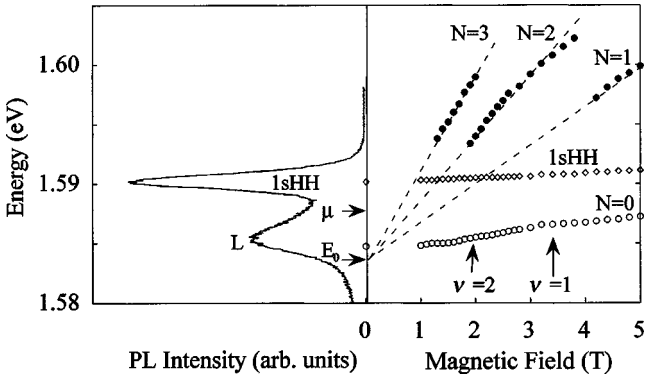

FIG. 6. The Landau fan diagram of unfilled (full dots) and occupied (open circles) Landau states of the liquid phase and the behavior of 1*s*HH exciton (diamonds) in a magnetic field are shown in the right side of the figure. This diagram has been obtained from PL (open symbols) and PLE spectra (full dots) at $T=2$ K, like those reported in Figs. 4 and 5. In the left side, the PL spectrum of intrawell excitons and 2D liquid measured at $B=0$ T and $T=2$ K is reported. E_0 is the bottom of the L band, and μ is the chemical potential of a pair in the liquid state. $\nu=1$ and $\nu=2$ are filling factors.

temperature PL spectra measured in the liquid phase for σ^- circular polarization are reported in Fig. 4 for $1\leqslant B\leqslant 3$ T. The spectra exhibit well-resolved $N=0$ and $N=1$ occupied Landau levels (just below the Fermi energy of the liquid) which are typical of electrons and holes in adjacent QW's. Low-temperature PLE spectra ($T=2$ K) have been taken, then, in the same heterostructures in order to identify the first unoccupied liquid states (namely, those just above the Fermi energy). These spectra have been measured in the case of σ^- circular polarization in steps of 0.2 T and are reported in Fig. 5 in steps of 0.4 T for $1.4\leqslant B\leqslant 5$ T, as well as for $B=0$. The detection energy was that of the $N=0$ state. While at $B=0$ T only the exciton states of heavy and light holes (1*s*HH, 2*s*HH, and 1*s*LH) can be detected, unfilled Landau levels of the liquid begin to be resolved above $B=1.4$ T, starting from the $N=3$ level, and to move to higher energy with increasing B. The $N=2$ level is resolved for $B\gtrsim 2$ T, while the $N=1$ level is resolved only for $B\gtrsim 3.4$ T. Finally, when the magnetic field exceeds 3.6 T, the $N=0$ ground Landau state begins to appear in the spectra. The anticrossing between the 1*s*HH-intrawell exciton level and each different Landau level can be noticed from the spectra reported in the figure. A reduced cyclotron mass of $0.052m_0$ is deduced from the dependence on B of the Landau levels, which implies that free holes are involved in the optical transitions. By taking an electron mass $m_e=0.07m_0$, one gets $m_h=0.2m_0$ for the heavy-hole mass.

The results obtained from PL and PLE spectra are summarized in the Landau fan diagram reported in the right side of Fig. 6. This allows us to determine some energy and thermodynamic parameters of the liquid. The linear extrapolation to zero magnetic field of the energies of the different Landau levels provides the bottom of the L band ($E_0=1.5835$ eV). The quasiequilibrium density of the liquid has been also determined, in two independent ways. First, an upper limit of the liquid density, n_s, can be estimated from the relation $n_s\rightarrow eB^*/\hbar$, where B^* is the magnetic field for which the lowest spin states begin to appear in the PLE spectra. In fact, only the emitted radiation with σ^- polarization is measured in the present PLE spectra. Therefore, only the lowest spin sublevels are detected, namely, sublevels where electron spin is $+1/2$ and hole spin is $-3/2$. For the Pauli exclusion principle, the $N=0$ Landau state becomes observable in PLE spectra only for filling factor $\nu=n_s\hbar/eB<1$, wherefore the above relation. Second, a different estimate of n_s can be obtained from the analysis of the discontinuities in the dependence on B of the peak PL energy of the L band (the first spectral moment).[16] Such discontinuities are related to cyclotron and spin gaps in the energy spectrum of the liquid and can be observed in Fig. 6 for the filling factors $\nu=2$ and $\nu=1$. Thus, the quasiequilibrium density n_s is evaluated equal to 8.8×10^{10} cm^{-2}, which leads to a Fermi energy $E_F=E_F^e+E_F^h=\pi\hbar^2 n_s/\mu$ equal to 4.1 meV [for the above deduced value ($0.052m_0$) of the reduced effective mass μ]. This value is independent of the applied forward bias and provides an upper estimate of the mobility edge (metal-insulator transition) in the electron and hole 2D layers. In the case of a reverse bias, instead, the critical density increases with the applied field, reaching the value of 1.5×10^{11} cm^{-2} for $V=-2.5$ V.

The line shape of the liquid PL band has an asymmetric triangular form. This is typical of the radiative recombination band in a 2D system of degenerate electrons and holes under the constraint of conservation of the e and h quasimomenta and of an e degeneracy higher than the h degeneracy. We have approximated the L emission band, with no adjustable parameter and no broadening of quantum states, with the following simple formula: $I(\hbar\omega)\propto f_e(\hbar\omega-E)f_h(E)\delta[\hbar\omega-E_0-E_e(\mathbf{K}_e)-E_h(\mathbf{K}_h)]$, given by the dashed line in Fig. 7.

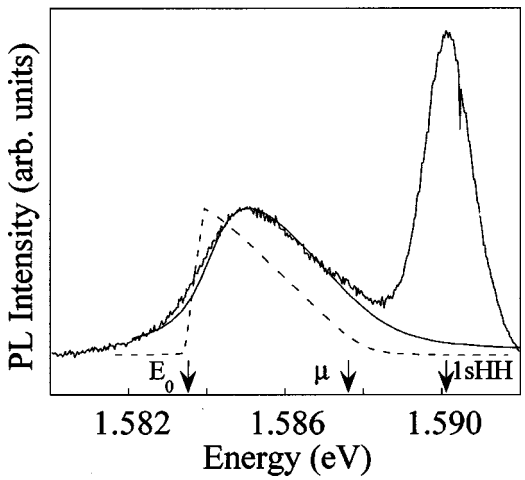

FIG. 7. Simulation (dashed line) of the *L*-band line shape at $T=0$ K with no adjustable parameter and null broadening. Best fit (thin solid line) of the 2D *L*-band line shape at $T=2$ K ($P=1\times10^{-2}$ W cm^{-2}, applied direct bias equal to 0.7 V) for a Gaussian broadening $\Gamma=0.7$ meV.

If we account also for a Gaussian broadening of the quantum states, the 2D liquid band intensity is given by $I(\hbar\omega)\propto\int_0^\infty f_e(\hbar\omega-E)f_h(E)\varphi(E)dE$, where $\varphi(E)\propto\exp[(-E/\Gamma)^2]$. By introducing a single adjustable parameter, Γ, the simulation of the liquid band much improves, as shown by the full line in Fig. 7 ($\Gamma=0.7$ meV). Since the mobile carriers in the liquid screen the random potential fluctuations which give rise to exciton localization, the value of the broadening Γ estimated for the *L* band is much less than the inhomogeneous broadening of the *I* line in the dilute regime (where the fluctuations are unscreened). The binding energy of the 2D liquid, as estimated on the basis of the above fit, is 2.5 meV. Finally, it should be noticed that the absence of sharp changes in the photocurrent in correspondence with the sharp changes observed, instead, in PL and *C-V* measurements suggests that gas and liquid phases coexist in each of the two wells, where a separation of a metal, liquid phase from an insulating gas phase should take place.

IV. CONCLUSIONS

In conclusion, we have shown that at low *T* a system of photoexcited electrons and holes in GaAs/Al_xGa_{1-x}As *p-i-n* DQW undergoes a phase transition from a gas phase to two 2D liquid states where electrons and holes are spatially separated and confined in adjacent QW layers. This transition is accompanied by major and sharp changes in the PL and PLE spectra, as well as in *C-V* measurements, with no corresponding abrupt change in the sample photoconductivity. The phase boundary between the diluted excitonic phase and the 2D liquid phase has been determined in the exciting power density vs carrier-temperature plane for different applied bias. The main parameters of the 2D liquid were determined.

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