Charged and neutral exciton phase formation in the magnetically quantized two-dimensional electron gas

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We report on a spectroscopic study of the lowest-energy electron-hole transitions of the two-dimensional electron gas (2DEG), with a density varied by photoexcitation in GaAs/Al_{0.1}Ga_{0.9}As quantum wells and under a perpendicularly applied magnetic field *B*. The transition into the phase consisting of a charged exciton singlet (X_s^-) ground state and neutral exciton (X) excited state occurs at a filling factor $\nu=1$. The relative intensities of the X_s^- and X transitions measured by photoluminescence excitation at $T=2$ K, as a function of $\nu \le 1$ and of *B*, are shown to depend on the relative area occupied by the magnetic-field–localized 2DEG and on the reduced orbit of the additional electron bound to X_s^- . [S0163-1829(96)08539-6]

It is well known that in the presence of a two-dimensional electron gas $(2DEG)$ the exciton (X) binding is significantly reduced by screening and phase space filling. $1-3$ Recent spectroscopic investigations⁴⁻⁷ in GaAs/Al_xGa_{1-x}As quantum wells $(QW's)$ have revealed that as the 2DEG density (n_e) decreases, the (neutral) exciton appearance is preceded by the formation of a negatively charged exciton (X^-) : two electrons and one (heavy-) hole bound complex. Under the application of a perpendicular magnetic field (*B*), the diamagnetic energy shift of the singlet (X_s^-) and triplet (X_t^-) transitions was observed^{8,9} in circularly polarized optical spectra.

In the magnetically quantized 2DEG the strength of the electron-hole interaction is controlled by the magnetic phase space filling factor (v) . At high v (low *B*), the interband transitions are between a (free or localized) hole and the 2DEG Landau levels with a negligible electron-hole interaction.³ The purpose of this work is to study the nature of the X^{-}/X phase formation, using high magnetic fields (low ν) in order to restore the electron-hole binding. We study interband photoluminescence (PL) and its excitation (PLE) spectra in modulation *n*-type doped GaAs/ Al_{0.1}Ga_{0.9}As multiple QW's (MQW's) at $2 \le T \le 25$ K and under magnetic fields $(0 \le B \le 5 \text{ T})$, applied perpendicularly to the QW plane (and parallel to the light direction). The structure under study¹⁰ has ten 300-Å-wide GaAs wells with 3500-Å-thick $Al_{0.1}Ga_{0.9}As barriers. It is symmetrically$ δ -doped in the middle of each barrier in order to produce a QW confined 2DEG with a density $n_e^0 = 7.1 \times 10^{10}$ cm⁻² and high mobility (\sim 4 \times 10⁶ cm²/V s). The QW's are resonantly excited with a laser at $E_{L_1} \le 1.53$ eV and $i_{L_1} = 1$ mW, modulated at a frequency of \sim 1 kHz. The 2DEG density is optically reduced¹¹⁻¹³ in the range $7.1 \ge n_e [10^{10} \text{ cm}^{-2}] \ge 0.8 \text{ by }$ above-barrier bandgap uniform illumination of an area \approx (0.5 mm)² with another laser ($E_{L_2} \approx 1.7 \text{ eV}$) of various intensities 1.5 mW $\ge i_{L_2} \ge 0$. The n_e dependence on i_{L_2} [Fig. 1(a)] was determined for $n_e > 10^{10}$ cm⁻² (and $\nu \ge 2$) by measuring the *B* dependence of the PL Landau transitions integrated intensity ratio (at $T=2 K$) and equating it to $(I_{n=1}/I_{n=0}) = \nu/2 - 1$, where

$$
\nu = \frac{2\pi\hbar c n_e}{eB} = \frac{0.41 n_e [10^{10} \text{ cm}^{-2}]}{B[T]}
$$

[see Fig. 1(b)]. Lower values of n_e corresponding to the X^{-}/X phase at *B*=0, were determined by measuring the *T* dependence of the PL intensity ratio between the X^- and X transitions^{5,6} [as in Fig. 1(c)] and relating it (at $T \ge 10$ K) to that measured in the PLE spectrum by

$$
\left(\frac{I_{X_s^-}}{I_X}\right)_{\rm PL} = \left(\frac{I_{X_s^-}}{I_X}\right)_{\rm PLE} \frac{E_F}{2kT} \exp\left(\frac{\varepsilon_b}{kT}\right).
$$

Here $E_F = \pi \hbar^2 n_e / m_e = 0.36 n_e$ [10¹⁰ cm⁻²] meV is the Fermi energy ($E_F \le kT$) and $\varepsilon_b \approx 0.85$ meV is the X_s^- binding energy measured with respect to that of *X*. The values of n_e obtained by these two procedures are interpolated by the fitting function¹¹ (Fig. 1(a), solid line):

$$
i_{L_2} = A \left(1 - \frac{n_e}{n_e^0} \right) \exp \left\{ -B \left(\frac{n_e}{n_e^0} \right) + C \left(\frac{n_e}{n_e^0} \right)^2 \right\},\tag{1}
$$

with $A \approx 6.0$ mW, $B \approx 13.4$, and $C \approx 10.1$. We found that for a given i_{L_2} , the PL spectral line shape measured under the L_2 excitation only is virtually the same as under simultaneous excitation with both L_1 and L_2 . Since L_1 penetrates the entire MQW structure, this proves that there is a nearly uniform $(within 5\%)$ depletion of all the 2DEG layers.

Figure 2 shows the energy dependence of the observed PL transitions (solid dots) on the magnetic field at $T=2$ K and for i_{L_2} = 20 μ W, corresponding to n_e = 5 × 10¹⁰ cm⁻². For $\nu \geq 2$ the $n=0$ and $n=1$ Landau transitions are observed with fitted slopes (solid lines) corresponding to an electron effective mass $m_e^* = 0.096m_0$. It is significantly larger than the conduction-band mass (m_e =0.067 m_0). We found that m_e^* increases from 0.069 m_0 to 0.12 m_0 as n_e decreases from

FIG. 1. (a) The measured 2DEG density n_e as a function of above-barrier bandgap laser intensity i_{L_2} and the fitting curve given by Eq. (1). The n_e values are obtained by the *B* dependence of the $n=1/n=0$ intensity ratio such as shown in (b), or by the *T* dependence of the X_s^-/X intensity ratio for $i_L \ge 950 \mu$ W as shown in (c). The observed PL spectra (thick solid lines) are fitted by multiple Lorentzian functions for each transition (dotted line). The fitting curves are indicated by thin solid line (virtually identical to the experimental curves).

7.1 to 3.2×10^{10} cm⁻². This is consistent with the predicted¹⁴ increase of the electron effective mass at the Fermi surface with decreasing density as a result of electron-electron interactions. At $\nu=2$ there is a discontinuity in the derivative of the PL energy (with respect to *B*) for the $n=0$ Landau transition. This is due to the electron-hole interaction ''freeze out" resulting from the depletion of the $n=1$ Landau level. It results in the formation of a many electron-bound hole complex at $1<\nu<2$. However, contrary to previously suggested models,^{15,16} this complex is far from being an exciton. As will be shown below, it transforms exactly at $\nu=1$ into the X_s^- ground state with the subsequent appearance of the higher-energy X_t^- and X excited states.¹⁷ In order to confirm the assignment of the transitions that appear at $\nu \leq 1$ we show in the inset of Fig. 2 the σ^- polarized (dashed line) and σ^+ polarized (solid line) PL spectra measured at $B=4$ T. The appearance of the X_t^- transition in σ^- polarized spectra^{8,9} occurs at a finite magnetic field (it can be clearly identified at $B \approx 2.5$ T), provided that $\nu < 1$. For comparison, we also show in Fig. 2 (by dashed lines) the measured diamagnetic energy shift of the X_s^- , X_t^- , and X PL transitions for the substantially depleted $2DEG$ ($n_e \approx 0.8 \times 10^{10}$ cm⁻²). We find that the *B* dependence of the observed PL transition energies becomes nearly independent of n_e at $\nu \leq 1$.

In Fig. 3 we show the PLE $(solid line)$ and PL $(dashed)$

FIG. 2. The PL transition energies (solid dots) measured as a function of magnetic field *B* at $T=2$ K and for $i_{L_2} = 20 \mu W$. The values of *B* corresponding to filling factors $\nu=2$ and $\nu=1$ are shown by arrows. The solid lines are fittings to the $n=1$ and $n=0$ Landau transitions dependence with $m_e = 0.096m_0$. The inset shows the PL spectra measured at $B=4$ T for σ^+ -polarized (dashed line) and σ^- -polarized (solid line) light. For comparison, also shown (by dashed lines) is the B dependence of the PL transition energies measured for i_{L_2} = 1.5 mW, corresponding to the substantially depleted 2DEG ($n_e = 0.8 \times 10^{10}$ cm⁻²).

line) spectra measured for i_{L_2} = 400 μ W corresponding to $n_e = 1.6 \times 10^{10}$ cm⁻² and at different $\nu < 1$ (0.7 \le 4 T). The relative intensities I_{X} and I_{X} (indicated by bars) of the corresponding transitions observed in the PLE spectra are measured by line-shape fitting (shown by a thin line) to a multiple Lorentzian function. We observe that scanning the monitored energy (E_m) throughout the low-energy PL tail does not change the ratio I_X - $/I_X$. However, decreasing E_m results in an appreciable PLE transition broadening.¹⁸ It can be seen that $I_{X_s^-}/I_X$ strongly increases with v: it is ≈ 0.55 for ν =0.16 [Fig. 3(d)] and it becomes too high for measuring at ν =0.94 [Fig. 3(a)].

We explain the transition into the X^{-}/X phase by a magnetic-field–induced spatial localization of the 2DEG at $\nu \leq 1$, when only the lowest $(n=0)$ spin-polarized Landau level is filled. A large splitting between the lower spin-up (\uparrow) and higher spin-down (\downarrow) levels is known to be due to electron-electron exchange enhanced magnetic interaction.¹⁹ Magnetotransport measurements show²⁰ that at $\nu=1$ the energy gap is $\approx 0.5B[T]$ meV. Thus, it is sufficiently large to assume that the ↑ and ↓ levels are resolved in our range of applied *B* and at $T \approx 2$ K. The X ⁻ PLE transition intensity, being proportional to the absorption cross section, is sensitive to the available QW area occupied by the 2DEG, where a photocreated *e*-*h* pair can bind an additional electron. The *X* PLE transition intensity, on the other hand, is proportional to the unoccupied QW area. Assuming a slowly varying, weak disorder potential that localizes the independent electrons in Landau states, the relative area occupied by the 2DEG is given exactly by the filling factor. Then, the ratio I_X - $/I_X$ is expected to be given by

FIG. 3. The PLE (thick solid lines) and PL (dashed lines) spectra measured at $T=2$ K for $i_{L_2}=400 \mu W$ with different values of magnetic field *B* (yielding the filling factor ν). E_m is the PL monitored energy. The relative PLE intensities (shown by bars) of the $X⁻$ and *X* transitions are measured by fitting the PLE spectra to multiple Lorentzian functions. The fitting curves are plotted by thin solid lines.

$$
\frac{I_{X^-}}{I_X} = \frac{\nu}{1-\nu} \left(\frac{f_{X^-}}{f_X} \right)_B, \tag{2}
$$

where f_{X} - and f_{X} are the corresponding oscillator strengths.

These simple arguments are valid when the ν -dependent localization length is much larger than the magnetic length $\lambda \equiv \sqrt{\hbar c/eB}$, the *X*⁻ and the *X* Bohr radii. This is the case for a long-range disorder potential²¹ that arises from random fluctuations of the donor position in modulation doped QW's with large undoped spacers. Such a potential is effective in our sample as evidenced by the Landau transition widths $(\Gamma \approx 0.6 \text{ meV})$ that are almost independent of *B*.^{18,21}

In Fig. 4(a) we plot $I_{X_s^-}/I_X$ (by error bars) and $I_{X_t^-}/I_X$ (by open dots) that are measured in the PLE spectra for $B=4.7$ T as a function of ν . The solid line is a plot of Eq. (2) with $(f_X - f_X)_{B=4.7 \text{ T}} = 2$ (see below). We find that it fits well the experimental values for X_s^- , but not those for X_t^- , since the triplet state has a much larger spatial extent than the singlet state.

In order to calculate the ratio $f_{X_s^-}/f_X$ we assume twodimensional 1*S* hydrogenic orbitals $\Phi_{1s}(r)$ and $\Phi'_{1s}(r')$ for the first and the second electrons in their relative motion with respect to the center of mass of the charged exciton. Their effective Bohr radii (denoted by *a* and a' , respectively) are *B* dependent. The X_s^- state is described by a symmetric

FIG. 4. (a) The X_s^{-}/X (error bars) and X_t^{-}/X (open dots) PLE intensity ratios measured at $B=4.7$ T as a function of filling factor v. The model curve (shown by a solid line) is given by $2\nu/(1-\nu)$. (b) The X_s^-/X PLE intensity ratio measured for two values of the 2DEG density as a function of filling factor ν . The corresponding model curves for n_{e1} and n_{e2} are shown by thick solid and dashed lines, respectively. The solid and dashed arrows at $v_1=0.20$ and v_2 =0.34 correspond to $B_0 \approx 3.3$ T at which the magnetic length equals the exciton Bohr radius for n_{e1} and n_{e2} , respectively. The two thin lines are given by $2\nu/(1-\nu)$ and $\nu/(1-\nu)$.

wave function of the form²²
$$
\Psi_{X_s^-}(r,r')
$$

= $\Phi_{1s}(r)\Phi'_{1s}(r') + \Phi_{1s}(r')\Phi'_{1s}(r)$. Then,

$$
f_{X_s^-} \sim \frac{2\int_0^\infty |\Psi_{X_s^-}(0,r)|^2 d^2 \mathbf{r}}{\int_0^\infty |\Psi_{X_s^-}(r,r')|^2 d^2 \mathbf{r} d^2 \mathbf{r}'} = \frac{2}{\pi} \frac{(a^{-2} + a'^{-2} + 2\overline{a}^{-2})}{\left(1 + \frac{a^2 a'^2}{\overline{a}^4}\right)}.
$$
\n(3)

Here $\overline{a} = (a + a')/2$. The two-dimensional hydrogenic 1*S*exciton oscillator strength is proportional to

$$
f_X \sim |\Phi_{1s}^0(0)|^2 = \frac{2}{\pi} a_0^{-2},\tag{4}
$$

where a_0 is the exciton variational (Bohr) radius. All the radii that enter Eqs. (3) and (4) are calculated numerically as a function of *B* in Ref. 22 by a variational method. The results of this calculation show that $a \approx a_0$ and they are weakly dependent on *B* varying in our experimental range. Their value can be approximated by the bulk GaAs (at zero field) exciton Bohr radius $a_B \approx 140$ Å (since the QW width is $L=300$ Å). On the other hand, *a'* being much larger than *a* and a_0 at $B=0$, shows a strong decrease with increasing *B*,

approximately as λ . At magnetic fields larger than the value $B_0 \approx 3.3$ T (as determined by the condition $a_B \approx \lambda$), the ex- $\frac{1}{2}$ (second) electron is eventually pushed towards the $X_s^$ center of mass and all the variational radii become nearly equal, i.e., $a \approx a' \approx a_0$. Consequently, according to Eqs. (3) and (4) as $B \rightarrow 0$ the ratio $f_{X_s^-}/f_X$ approaches 1, while for $B \ge B_0$ it nearly equals 2.

Fig. 4(b) presents $I_{X_s^-}/I_X$ measured in the PLE spectra as a function of ν for two different values of the 2DEG density: $n_{e1} = 1.6 \times 10^{10}$ cm⁻² (solid dots) and $n_{e2} = 2.7 \times 10^{10}$ cm⁻² (open dots). The corresponding model curves (denoted by a thick solid and a dashed line, respectively) were calculated using Eqs. $(2-4)$ for various values of *B*. The values of B_0 corresponding to n_{e1} and n_{e2} are indicated by solid and dashed arrows at $v_1=0.20$ and $v_2=0.34$, respectively. Note that the experimental points lie between the two thin lines calculated using Eq. (2) with $f_{X_s^-}/f_X = 1$ and $f_{X_s^-}/f_X = 2$, except the sharp cusplike behavior of the measured values near $B \approx B_0$. This is simulated by an abrupt slope change of the model curves (shown in the insets for clarity).

We emphasize that the 2DEG localization at $\nu=1$ is due to the existence of a gap in the energy spectrum. Extrapolating this result to lower temperatures $(T \ll 2 K)$ one should expect a similar divergence of the $I_{X_s^-}/I_X$ ratio at energy gaps of all fractional filling factors corresponding to incompressible Laughlin states. 23 One of the main factors preventing such a spectroscopic observation at fractional ν values might be an effective 2DEG heating by the optical excitation.

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Finally, we comment on the essential difference between the nature of the transition into the X^{-}/X phase at $B=0$ and that under a high magnetic field. In the former case it was shown⁴ to occur near the metal-insulator transition of the 2DEG, since the extended electron states (in the form of plane waves) result in the 2DEG occupying the entire QW area. In the presence of a strongly quantizing magnetic field, on the other hand, the extended states in real space are thin ''bands'' that stretch but *do not* cover the entire QW plane. These leave parts of the QW plane available for exciton absorption and therefore, the X^{-}/X phase can be formed in the magnetically quantized metallic 2DEG.

In summary, we studied spectroscopically the transition of the lowest-energy electron-hole states in the magnetically quantized 2DEG into the bound X^{-}/X phase. We measured the X_s^-/X PLE intensity ratio as a function of ν and *B* and showed that the transition occurs at $\nu=1$ due to the 2DEG spatial localization. We also found, that the X_s^- oscillator strength increases by a factor of \approx 2 as *B* increases from zero to $B_0 \approx 3.3$ T, when the external electron approaches the $X_s^$ center of mass, and its Bohr radius equals the magnetic length.

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