High magnetic field studies of charged exciton localization in GaAs/Al$_x$Ga$_{1-x}$As quantum wells

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We report on low temperature, polarization resolved, high magnetic field (up to 23 T) photoluminescence experiments on high mobility asymmetric GaAs quantum wells. At high magnetic fields, we detect two strong emission lines of the neutral and positively charged excitons (X and X^+) and a series of weaker lines of the excitons bound to ionized acceptors (AX^-). From polarization energy splittings of these lines, we determine the hole Landé factors (g_h) of different complexes. For X and X^+, g_h initially grows with magnetic field and then saturates at $g_h = 0.88$ and 1.55, respectively; for AX^-'s, g_h begins from a high value (from 6 to 11 at zero field) and decreases with the field growth. This contrasting behavior is traced to the structure of valence band Landau levels, calculated numerically in the Luttinger model, beyond axial approximation. This points to the coexistence (in the same well) of mobile X and X^+ with localized and interface-pressed AX^- states. [http://dx.doi.org/10.1063/1.4896158]

Excitons, bound electron-hole pairs ($X = e + h$), are semiconductor counterparts of hydrogen atoms. Charged excitons or "trions" ($X^{\pm} = 2e + h$ or $2h + e$) predicted[1] in analogy to hydrogen ions were also observed experimentally in two-dimensional (2D) structures,[2] aided by strong binding enhancement compared to bulk crystals. Observation of trions naturally raised controversies about their localization. Sanvitto *et al.*[3] reported experiments in favour of mobile nature, demonstrating drift of negative trions, created by laser excitation of a high electron mobility transistor, upon applying voltage between source and drain. However, other authors argued that charged and massive trions cannot avoid localization by ionized impurities of the opposite charge. For example, Solovyev and Kukushkin[4] studied photoluminescence (PL) from quantum well (QW) structures with δ-doped donor layers placed at different distances d from the QW and suggested that the onset of an additional emission line and its energy dependence on d points to trion localization.

In all asymmetric structures, electrons and holes are separated by inherent electric field. Moreover, the field-induced band bending pushes heavy holes closer to the interface than light holes. We have recently shown[5] that in high magnetic fields the g-factors of X in asymmetric wide QWs are governed by excited hole levels of the light-hole character. This is related to larger overlap of electron envelope wave functions with light holes than with heavy holes, and larger overlap yields stronger exciton binding, exceeding Landau level (LL) splitting.

In this paper, we exploit connection between g-factors and wave functions of valence holes to determine localization of different excitonic complexes from polarization splittings of their optical recombination energies. Thus, we report extensive magneto-PL studies of superior quality p-type doped GaAs QWs. From the Zeeman splitting of emission lines recorded in σ^- and σ^+ polarizations we have determined hole Landé factor (g_h) of X, X^+, and the acceptor-bound complex AX^-. By the comparison of experimental results with theoretical calculations of single-hole LLs in the valence band, we found that the measured g_h of X mainly comes from the excited hole levels of a light hole character, g_h of X^+ reveals balanced occupation of ground and excited hole levels, and g_h of AX^- comes predominantly from the ground heavy hole states. These assignments allow us to confidently identify X and X^+ as nearly free objects, whereas the multiple AX^- states are understood as excitons bound by Coulomb interaction to ionized acceptors placed on subsequent crystallographic planes in one of the barriers.

The main two studied samples S1 and S2 were remotely doped GaAs/Al$_{0.33}$Ga$_{0.67}$As asymmetric QWs of width $w = 15$ nm, grown by molecular beam epitaxy on the (001) oriented semi-insulating GaAs substrate. The excess carriers were supplied to the QW by carbon acceptor doping of 200 nm layers in both Al$_{0.33}$Ga$_{0.67}$As barriers. Importantly, different doping dose was used on each side: 5×10^{16} cm^{-3} in the bottom barrier of both samples, and 3×10^{17} and 2.3×10^{17} cm^{-3} in the top barrier of S1 and S2, respectively. Doping was set back from the QWs by undoped 60 nm spacers. Hole mobilities at temperature $T = 4.2$ K were $\mu = 7.67 \times 10^5$ and 8.67×10^5 cm^2/V s, and concentrations measured in the dark were $p = 2.22 \times 10^{11}$ and 1.45×10^{11} cm^{-2} for samples S1 and S2, respectively.

For comparison, we will also refer to another pair of previously investigated samples S3 and S4. They are similar to S1, except for different either doping or width: S3 has the

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same width $w = 15$ nm but is symmetrically doped (Ref. 6), while S4 is also asymmetric but has a larger width $w = 22$ nm (Ref. 7).

All measurements were performed in liquid helium, at $T = 1.8$ K. PL was excited by the 632.8 nm line of a helium-neon laser. Magnetic field (B) was applied in the Faraday configuration, parallel to the QW growth direction, up to the $B = 23$ T with step $\Delta B = 0.05$ T. Fiber optics was used, with a linear polarizer and a quarter-wave plate placed close to the sample. Circular polarizations σ^- and σ^+ were switched by reversing the field direction. The spectra were analyzed using a monochromator, equipped with the liquid-nitrogen cooled charge-coupled Si device.

Let us begin the discussion of our results with the spectra recorded in the absence of magnetic field. At lower laser power densities $P < 15$ mW/cm^2, only one line was observed in the PL spectra of both samples. Based on our previous study[6] of a symmetric 15 nm wide QW, we attribute this line to emission of the spin-singlet positive trion (X_s^+). When magnetic field is applied, several lines emerge in the PL spectra. In Fig. 1, we present the field evolution of the PL spectrum of sample S1 (results for S2 being similar), recorded at $T = 1.8$ K, in both light polarizations. All shown emission lines (related to the recombination of 2D holes) can be divided into two main groups. The main common attribute of the first group of lines located at higher energies in Fig. 1 (SU-AX^+ and above) is strong dependence of their energy position on both width and symmetry of the QW. All these lines shift toward higher energies with the decrease of the well width and also as the well changes profile from asymmetric to symmetric. In contrast, the second group of lines, detected at lower energies (below SU-AX^+), are virtually insensitive to the width or symmetry of the QW.

This contrasting behavior of the two groups of lines is evident in Fig. 2, in which we compare the PL spectrum of sample S1 (blue curve) taken at the particular field $B = 15$ T in the σ^- polarization, with the spectra of two similar QWs, but different in either doping (S3; black curve) or width (S4; red curve). The strong QW width- and symmetry-dependence of energy position of lines from the first group reflects their association with (weakly localized) electron-hole complexes whose wave functions extend over the entire

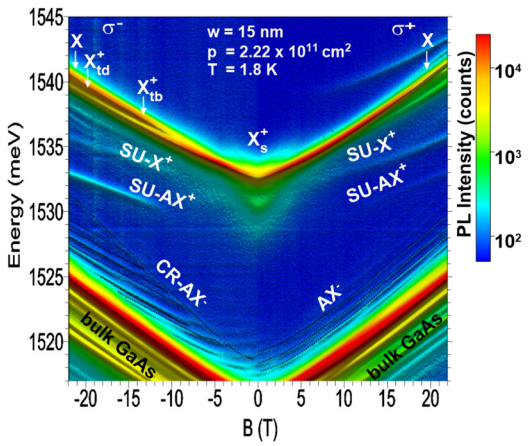

FIG. 1. The magnetic field (B) evolution of the PL spectrum of sample S1 in σ^- and σ^+ polarizations. Two distinct groups of lines are (1) SU-AX^+ and above; (2) lines below SU-AX^+.

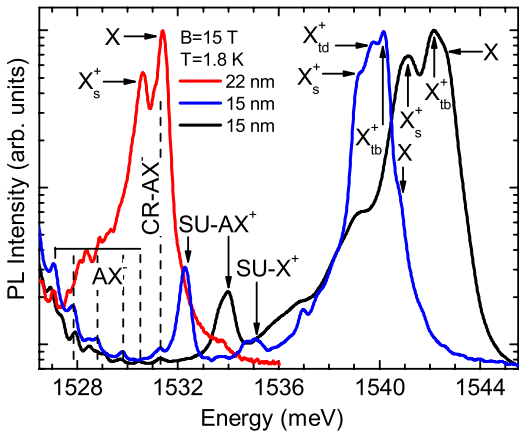

FIG. 2. The PL spectrum of sample S1, recorded at magnetic field $B = 15$ T in polarization σ^- (blue curve) compared with the spectra of two similar QWs, different in either doping (S3; black curve) or width (S4; red curve).

QW width. On the other hand, the virtual insensitivity of energy positions of lines from the second group reveals their association with the radiative complexes which are strongly localized by deep Coulomb potentials of ionized acceptors placed in the barriers and, in consequence, which are also tightly pressed to one of the well-barrier interfaces, and, hence, also insensitive to the opposite interface. (To be precise, unavoidable disorder causes some lateral localization of all excitonic complexes; by weak and strong localization, respectively, we will mean such that has relatively weak or strong effect on the emission energy.)

The electron-hole complexes of the first group are neutral exciton and positive trions. The exciton and spin-singlet trion (X_s^+) are observed in both polarizations σ^- and σ^+, while the "bright" and "dark" spin-triplet trions (X_{tb}^+ and X_{td}^+) are only seen in σ^-. At lower energy, we also find two parallel weaker lines, departing downward from X and X^+, proportionally to the increase of magnetic field. This pair of lines SU-X^+ and SU-AX^+ are the hole shake-up replicas of the nearly free and acceptor-bound trions.[6,8] In a shake-up process, recombination of an e-h pair is accompanied by (and coupled with) excitation of an additional carrier from the surrounding gas to a higher LL, which results in lowering of the emission energy by a cyclotron energy. Comparison of the energy vs. magnetic field slope of the shake-up lines with those of X and X^+ yields a difference of 0.3 meV/T, corresponding to the heavy-hole effective mass[9] $m^* = 0.38\, m_e$.

The second line group contains multiple rather weak, parallel, narrow, almost equidistant, and closely spaced transitions, all with the same energy-field slope as that of X and X^+. We attribute these lines to the recombination of complexes $AX^- = A^- + h + e$ (i.e., the excitons bound to barrier ionized acceptors). In this group we also observe, but only in σ^- polarization, one line with the higher energy-field slope, which in comparison with the slope of X and X^+ yields the difference of 0.3 meV/T, which is the same as for shake-up replicas but with an opposite sign. This suggests a similar origin, but with de-excitation of an additional hole from a higher to the lower LL. We have labeled this transition CR-AX^-.[7]

The arguments for our proposed interpretation of the AX^- lines are as follows: (a) Studied p-type QWs contain (due to diffusion) residual acceptors in the entire structure.

(b) Barrier potentials were almost identical in all studied samples. (c) Energy separation between the AX^- and X lines is fairly large: 14 meV for the lowest AX^- line, matching the AX^- binding energy from previous numerical calculation (for A^- located at an interface of a symmetric 15 nm wide QW[10,11]); when A^- sits in the middle of the QW, binding energy increases to 24.5 meV, close to 22 meV of bulk GaAs.[12] (d) Small distance (0.28 nm) between consecutive crystallographic planes hosting the acceptors yields fine quantization of the AX^- levels, observed in the PL spectrum as a series of parallel lines. (e) The two larger acceptor-bound radiative complexes, AX and AX^+, have much smaller binding energy and hence are detected at higher energies in the PL spectrum.[6]

From the σ^+/σ^- polarization energy splitting of the exciton lines (X, X^+, and AX^-), we have determined their Zeeman splitting and the effective g-factors (called g_{ex}), using the equation

$$\Delta E_{ex} = E(\sigma^+) - E(\sigma^-) = g_{ex}\mu_B B, \tag{1}$$

where μ_B is the Bohr magneton. Then using the following sign convention for the electron and hole g-factors:

$$g_{ex} = g_h - g_e \tag{2}$$

and the well-established[13] empirical formula for g_e

$$g_e(E) = -0.445 + 3.38(E - 1.519) - 2.21(E - 1.519)^2, \tag{3}$$

for transition energy E given in units of eV, we evaluated hole g-factors (g_h). The results are presented in Figs. 3(a) and 3(c). Clearly, the values of g_h determined for X and X^+ demonstrate rather distinct behavior, in terms of both magnitude and magnetic field evolution (g_h of X is smaller and crosses zero).

To understand the difference between g_h determined (experimentally) for different complexes, and also anticipating connection with the problem of X^+ localization, we used the previously developed method[5] to calculate the energies and wave functions of 2D carriers in our structures: Potential distribution $V(z)$ and energies of the hole subbands at $B = 0$ were found from self-consistent solution of Schrödinger and Poisson equations, using Hartree approximation to include contribution of mobile holes to $V(z)$. Exact hole eigenfunctions were taken into account in the Luttinger Hamiltonian. Potential $V(z)$ was used to find wave functions and energies of hole LLs at $B > 0$. The calculations were extended beyond axial approximation by inclusion of the cubic term.

Figure 4(a) presents the high magnetic field evolution of energies of topmost valence-band levels calculated for our structure S1. All shown states belong to the ground heavy-hole subband. They are harmonic oscillator functions labeled by index $n = -2, -1, 0, 1,\ldots$ of the largest axial component of the wave function. For $n \geq 1$, there are two axial levels with the same number n in each subband, distinguished by the letters a and b. Optically active states are marked in colors: σ^- (blue) or σ^+ (red).

The values of g_h obtained from the splitting of ground states $1a$ and -2 and of the first excited states $1b$ and -1 are compared with the experimental values for X and X^+ in Fig. 3(a). Clearly, g_h of X and of excited states $1b/-1$ are similar both in terms of magnitude and field evolution. This is explained as follows. For magnetic fields used in our experiment, the characteristic e-h Coulomb interaction energy exceeds the hole LL splitting. Electron and hole envelope functions are shown in Fig. 4(b). Due to high mass, ground state heavy holes are pressed against the doped barrier. In contrast, the excited hole states have significant light-hole contribution (due to valence subband mixing) and extend over the entire QW width. This yields higher overlap with the electron

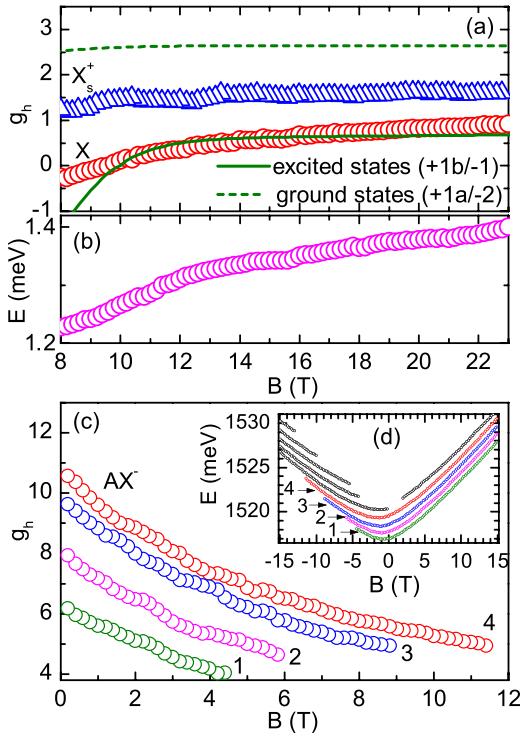

FIG. 3. (a) Hole g-factors of X and X^+ (solid lines show theoretical curves for two lowest hole *eigenstates*; see also Fig. 4). (b) Coulomb binding energy of X^+. (c) Hole g-factor of AX^-. (d) Magnetic field dependence of several AX^- *emission energies in polarizations* σ^+ and σ^-.

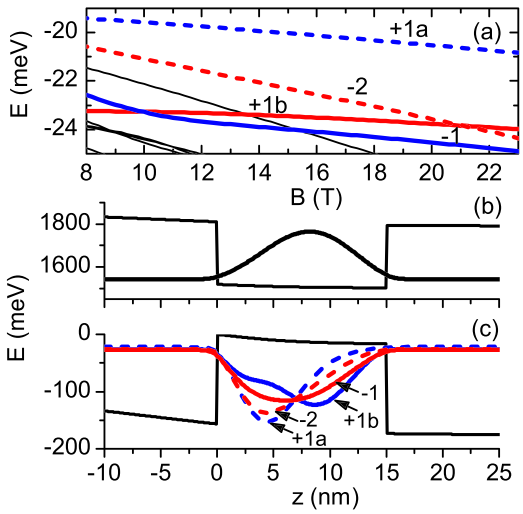

FIG. 4. Results of theoretical calculations: eigenenergies (a) and envelope wave functions (c) of the topmost hole LL; envelope wave function of lowest electron LL (b).

state, resulting in stronger Coulomb attraction and hence large contribution of excited hole states ($1b/-1$) to X.

Still in Fig. 3(a), the g_h of X^+ is approximately equal to an average between ground $1a/-2$ and excited $1b/-1$ hole states, suggesting balanced contribution from the ground and excited hole states. This seems to correspond to the weaker binding of an additional hole to X. Indeed, the experimental X^+ Coulomb binding energy (determined as the energy separation of X and X^+ lines, averaged over both σ^+ and σ^- polarizations to exclude Zeeman effect) is smaller than 1.4 eV (see Fig. 3(b)) and hence comparable with the splitting of hole LLs.

Finally, let us discuss the second group of lines identified as emission from excitons bound to ionized barrier acceptors, AX^-. As seen in Fig. 3(c), the g_h of the AX^- states begins from a high value at zero field and rapidly decreases with the field growth, in striking contrast to X and X^+. Also, g_h increases with the AX^- emission energy, from 6 to 11, at $B=0$. These features are likely related to the interface localization of the recombining state. It is known that electron g-factor in the barrier is much larger than in the well;[14] we are unaware of the relevant experiments for the holes but a similar effect can be expected for them, too. On the one hand, this may cause the observed anomaly, tentatively associated with the significant (and sensitive to the A^-/QW distance) penetration of the exciton envelope function into the barrier, simultaneously affecting emission energy and g_h. On the other hand, we assumed g_e according to Eq. (3) when calculating the plotted g_h from the PL $\sigma^\pm$ splitting, so this g_h would in fact contain some electron contribution. However, we have not explored this problem quantitatively (especially, the dependence on magnetic field).

In conclusion, from the polarization-resolved high-field magneto-photoluminescence spectra of asymmetric GaAs quantum wells we have determined the spin splittings and corresponding valence hole g-factors of various (neutral and charged) excitonic complexes. Taking advantage of two facts: (i) correlation between localization in the plane of the well and at one of its sides in the growth direction, both effects simultaneously caused by Coulomb binding to the ionized acceptors placed in the barriers at different distances from the well; and (ii) different magnitude and field dependence of the hole g-factor in different valence-band Landau-like levels (as we have found from detailed numerical calculations) and therefore also in different excitonic complexes distinguished by hole contribution from these levels—we have been able to characterize all detected emission lines with respect to the localization of the associated recombining complex. As a result, we have demonstrated coincidence of nearly free excitons (X) and positive trions (X^+) in the same sample with the AX^- complexes involving strong binding to the ionized barrier acceptors. Remarkably, we were also able to resolve multiple AX^- lines associated with acceptors placed on subsequent crystallographic planes (at different discrete distances from the well).

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