Spin-triplet negatively charged excitons in GaAs quantum wells

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We observe magnetic-field-induced transitions in the interband optical spectra of GaAs quantum wells with a small excess electron density. Their strengthening with excess electron density, in addition to their light polarization dependence, demonstrate that these correspond to (excited) spin-triplet states of the negatively charged exciton. The second-electron binding energy of both singlet and triplet X^- strengthens with field.

Two-electron atoms, such as helium (He) or the negatively charged hydrogen ion (H^-) have long been of interest for the study of electron correlation and exchange.[1,2] H^- is also of interest, because of its role in determining the opacity of stars such as our sun.[1] The negative donor center (D^-) is a semiconductor analog that facilitates the application of very large effective magnetic fields, which atoms can encounter in the atmosphere of white dwarfs.[3–10] Recently another two-electron system, the negatively charged exciton (X^-), has received renewed attention, because of its enhanced binding energy in quasi-two-dimensional (2D) semiconductors.[11–15] We report here an interband magneto-optical study of GaAs quantum wells (QW's) with a variable excess electron density, where we observe the stabilization of spin-triplet X^- states under an applied magnetic field. Although this has been predicted for D^-,[2,7–10] it has not yet been unambiguously observed.[10]

The Pauli exclusion principle requires systems with two identical fermions to be anti symmetric upon interchange of the particles. This results in the spin states of a two-electron system arranging into an antisymmetric singlet of zero total spin and a symmetric triply degenerate state with a total spin quantum number of 1. In X^- the additional spin of the hole results in a twofold degenerate ground state, which we refer to as singlet X^- or X_s^-, while the first excited state is a sixfold degenerate triplet X^- or X_t^-. The degeneracies are lifted in a magnetic field by the Zeeman splitting of the electron and hole levels, producing the eight exciton energy levels shown in Fig. 1(c). In our experiments, we excite X^- from one electron initial states, using circularly polarized light, e^-+photon$\rightarrow x^-$. Selection rules require the z component of the total spin to change by -1 ($+1$) for σ^- (σ^+) polarization, producing the transitions indicated by the vertical lines in Fig. 1(c).

The *spatial* wave function of X^- can be regarded as a symmetric (for X_s^-) or antisymmetric (for X_t^-) combination of two one-electron orbitals with different extents. This approach allows much physical insight, although it should be noted that each electron has equal probability of being found in the inner or outer orbital.

H^- and D^- have only one bound state, a singlet, at zero magnetic field.[1,16] However, the confining potential introduced by an applied magnetic field binds additional higher-energy levels. Variational calculations find an infinite number of bound states for the 3D D^- center at any magnetic field,[2] while only four bound states, comprising the lowest singlet and triplet, exist in the 2D and high field limits.[7] Another variational calculation[8] of 2D D^- centers estimated the lowest-energy triplet to bind for a small, though finite, field of $\gamma \sim 0.2$, expressed in the dimensionless unit $\gamma = \hbar\omega_c/2R$, where $\hbar\omega_c/2$ is the ground electron cyclotron energy and R the donor Rydberg, corresponding to $\sim$1.3 T for GaAs.

Although X^- has an analogous energy level structure to H^- and D^-, there is a fundamental difference in the spectroscopic techniques that can be applied to it. For He, H^-, or D^-, a photon causes transitions *between* its energy levels. Observation of the triplet states of D^- is hampered by the rule forbidding transitions between singlet and triplet levels.[1] Hence, the triplet transitions are only significant at temperatures where the lowest triplet state is populated. Furthermore, these triplet transitions are obscured by their energetic coincidence with those of the neutral donors.[10] In contrast, an *interband* photon *creates* X^- by exciting a conduction band electron and a valence band hole, which bind a second excess electron. Thus, it measures the ground and excited X^- energy states directly, rather than the energy difference between them, with both singlet and triplet transitions allowed at low temperature. Furthermore, the simultaneous observation of neutral excitons (X) in the spectra, rather than obscuring the X^- transitions, allows the binding energy of the second electron to be estimated directly. Another useful aspect of the system that we study is the ability to vary the excess electron density in order to determine whether a spectral feature derives from X or X^-.

We have studied remotely doped GaAs/$Al_{0.33}Ga_{0.67}As$ single QW's grown by MBE on GaAs substrates. We illustrate our arguments with spectra taken on a 300 Å GaAs QW, topped with 600 Å undoped $Al_{0.33}Ga_{0.67}As$ and 2000 Å Si doped (10^{17} cm^{-3}) $Al_{0.33}Ga_{0.67}As$. Similar behavior was also observed for a

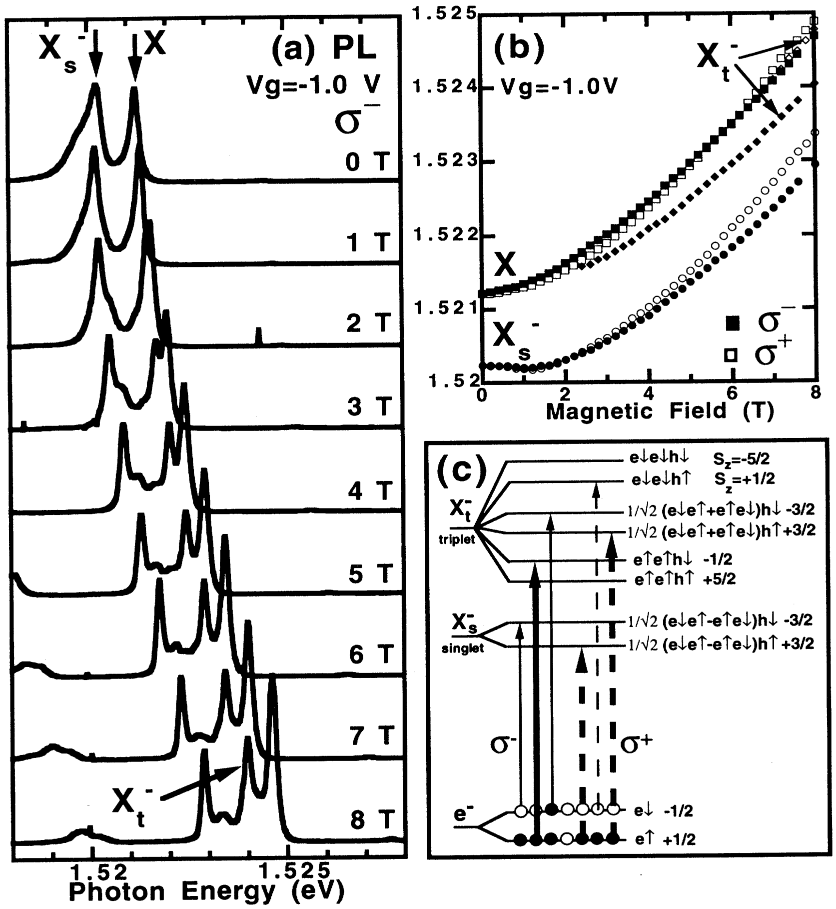

FIG. 1. (a) PL spectra recorded with different applied magnetic fields, emitted in σ^- polarization, at 2.0 K and with a Schottky bias of -1.0 V. (b) Evolution of the PL peak energies (vertical axis) with field, for both σ^- (solid) and σ^+ (open symbols) polarizations. Notice in (a) and (b) the emergence of the triplet X_t^-, (diamonds) near 2.4 T in σ^- polarization. (c) Schematic of X^- transitions, due to absorption of a circularly polarized photon in a magnetic field, e^-+photon$\rightarrow x^-$. The photon changes the total z component spin (S_z) by $+1$ (-1) for σ^+ (σ^-).

220 Å QW sample. The excess electron density is varied by applying a voltage (V_g) between a Au Schottky layer evaporated on the top surface of the sample and Ohmic contacts to the QW layer. The QW has a minimal electron density (of order 10^{10} cm^{-2}) for $V_g \leq -0.8$ V. The 300 Å QW sample displays excitonic linewidths of $\sim$0.3 meV and an ungated 4 K mobility after illumination of 2.9×10^6 cm^2 V^{-1} s^{-1}. Photoluminescence (PL) and electroreflectance (ER) spectra were recorded as a function of the excess electron density and magnetic field applied perpendicular to the QW. ER is a differential, excitation technique, where the alternating reflectivity induced by a small modulation superimposed on the Schottky bias is measured. The circular light polarizations (σ^+ or σ^-) referred to here relate to the incident light for the ER and that emitted for PL.

Figure 1(a) plots σ^- polarized PL spectra taken with different applied magnetic fields for the minimal excess electron density in the QW ($V_g = -1.0$ V). At zero field, two PL peaks are observed corresponding to the neutral (X) and (singlet) negatively charged (X_s^-) excitons of the lowest electron and heavy-hole subbands. The separation of these peaks implies a binding energy for the second electron of about 1.0 meV, which, as expected, lies between the calculated 2D and 3D limits.[11] The X and X_s^- transitions are also observed in ER spectra of Fig. 2(a). Both ER and PL spectra show a dramatic quenching of X with increasing electron density, with the concurrent strengthening of X_s^-.[13,14] This observation, in addition to the magnetic-field dependence described below, confirms unambiguously our assignment of the X and X_s^- transitions.

Under an applied magnetic field, the X peak undergoes a diamagnetic shift [see Figs. 1(a) and (b)] very similar to that reported for neutral excitons in undoped QW's.[17] The change of sign of the X Zeeman splitting near 6 T, apparent in Fig. 1(b), is also observed for undoped QW's,[17,18] and is due to valence band mixing. X_s^- displays a rather different behavior, showing an initial small redshift and then increasing in energy less rapidly than X. The increasing energy separation of X and X_s^- with field indicates a large enhancement of the second-electron binding energy. This derives from the relatively large spatial extent of X^-, which at zero field is roughly twice that of a neutral exciton.[8] Hence, even weak magnetic fields produce significant confinement of the outer (one-electron) orbital in the QW plane and a strong enhancement of its Coulomb interaction with the core of the exciton.

Around 2.4 T, a shoulder is resolved on the low-energy side of the X PL peak in σ^- polarization. With further increasing field this develops into a distinct, lower-energy peak [marked X_t^- in Figs. 1(a) and (b)], whose separation from X increases to about 0.6 meV in the 8 T σ^- spectrum. As discussed below, this magnetic-field-induced transition is a spin-triplet state of X^- and the focus of this paper. A corresponding feature is observed in ER spectra, which tracks the position of the PL peak closely, as can be seen in Fig. 2(b), which compares PL and ER spectra taken at 8 T. The observation of X_s^- and X_t^- in the ER spectra is significant, since it demonstrates that they cannot have an extrinsic origin, such as an impurity or defect bound exciton. In contrast, the weak PL peak with a photon energy between that of X_s^- and X_t^- in the 8-T spectra of Fig. 2(b) shows no corresponding ER feature, and may therefore derive from an impurity bound exciton.

Further information regarding this magnetic-field-induced transition can be gleaned from Fig. 3, which plots PL and ER spectra recorded with different applied offset gate biases at 8 T. At $V_g = -0.8$ V, corresponding to the lowest excess electron densities, a PL peak is observed, due to this transition (marked X_t^-), as well as X_s^- and X, the latter being the dominant feature in the spectrum. As the gate bias (and hence excess electron density) is increased, however, the X intensity is quenched in both the PL and ER spectra, while the X_s^- feature strengthens, as at zero magnetic-field.[14] Notice that the intensity of X_t^- also grows initially with increasing excess electron density. This demonstrates that *X_t^- requires the presence of an excess electron and must, therefore, originate from an excited state of the negatively charged exciton.*

Another magnetic-field-induced transition appears around 6.8 T in σ^+ polarization. This can be seen in the 8 T σ^+ polarized PL (where it appears as a shoulder on the low-energy side of X) and ER spectra of Fig. 2(b) and as open diamonds in Fig. 1(b). This transition shows a similar dependence upon excess electron density to that discussed above for σ^- polarization, indicating that it corresponds to another excited state of X^-.

We now compare the expected interband X^- transitions induced by circularly polarized light, indicated by vertical arrows in Fig. 1(c), to the experimental spectra. A transition to the singlet X_s^- is expected for both polarization senses. The transition in *excitation* spectra taken with σ^+ polarization will be enhanced over that in σ^- by the larger occupancy of $e\uparrow$ initial state than $e\downarrow$ at low temperature.[12] This explains why X_s^- is stronger in σ^+ polarization than σ^- in the 8 T ER spectra plotted in the lower part of Fig. 2(b). In fact, the ratio of the ER intensity in the two polarizations, $I(\sigma^-)/I(\sigma^+)$, follows the relation $\exp(-\Delta E/kT)$, where ΔE $(=|g|\mu_B B)$ is the Zeeman splitting of the ground state. The agreement of the fitted g factor of $|g|=(0.42\pm0.02)$, in our wide QW's with the GaAs conduction band value of (0.44 ± 0.02),[19] is a powerful confirmation that the transition is due to X_s^-.

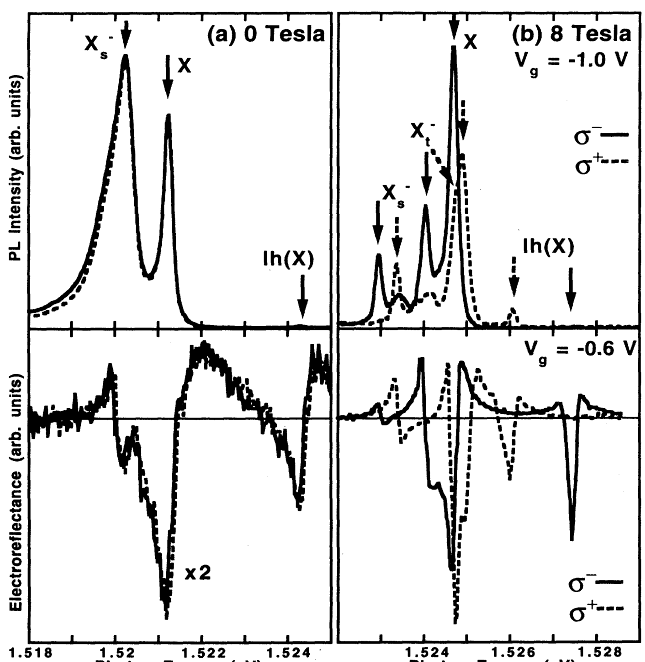

FIG. 2. PL and ER spectra measured at 2.0 K with a magnetic field of (a) 0 and (b) 8 T applied perpendicular to the QW. The PL spectra were recorded with a Schottky bias of −1.0 V, while the ER was measured with an offset of −0.6 V and a modulation of 10 mV amplitude.

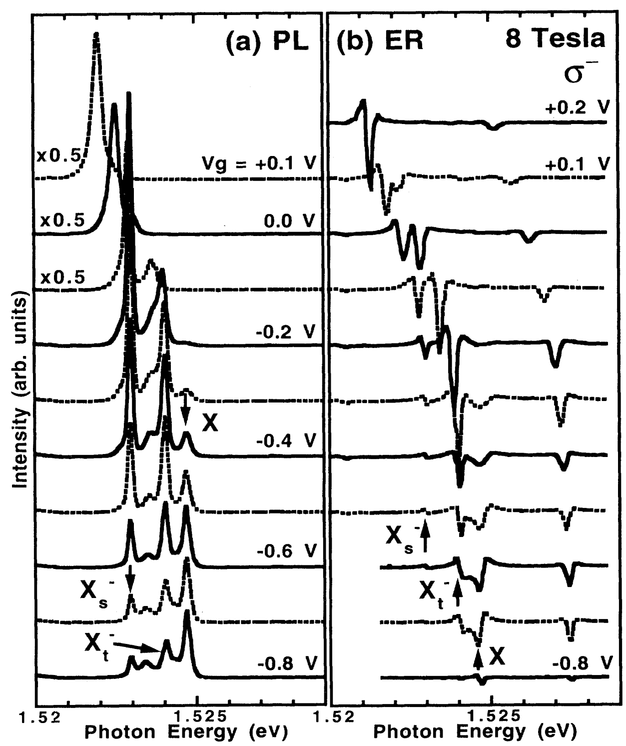

FIG. 3. Schottky bias dependence of (a) PL and (b) ER spectra taken at 8 T and 2.0 K. The electron density increases moving up the curves, which are offset for clarity. At the lowest excess electron densities, the PL and ER spectra are dominated by X, but as the electron density is increased, both X_s^- and X_t^- strengthen, while X is quenched.

Notice in Fig. 1(c) that the selection rules for transitions to the triplet X_t^- differ radically from those for the singlet. The triplet transition with lowest photon energy is expected to give a strong contribution in σ^-, while another strong triplet transition is expected in σ^+ with a higher photon energy. These selection rules for triplet X_t^- agree with the magnetic-field-induced X^- transitions observed in our experimental spectra. As discussed above, we observe the first excited X^- state to emerge near 2.4 T in σ^- polarization and the second near 6.8 T in σ^+ polarization, consistent with these excited states being spin triplet.

Spectra taken on the 220 Å QW display very similar dependences on electron density and magnetic field to those discussed above: the separation of X and X_s^- increases with magnetic field, due to the enhanced binding of the outer orbital; the X_s^- transition strengthens in σ^+ polarized ER, relative to σ^-, due to the spin polarization of its ground state; and the triplet transition X_t^- transition is observed to emerge with increasing field in σ^- polarization. At 8 T, the separations of X_s^- and X_t^- from X are similar to those observed for the 300 Å QW, as one would expect, since the magnetic field, rather than the well width, is the dominant influence on the in-plane motion.

The observed excited X^- transitions could not correspond to higher angular momentum singlet states of X^-, as these would require the same light polarization as the ground state singlet X_s^-. The polarizations of the magnetic-field-induced transitions are also inconsistent with the X_s^- states of the light-hole exciton. Furthermore, these magnetic-field-induced transitions are also observed for the 220 Å QW, with very similar binding energies at 8 T, despite the light-heavy-hole separation being significantly larger in this narrower QW. The X^- states of the light-hole exciton are indeed observed in the ER spectra at higher photon energy and densities. Under applied magnetic field, we again observe the lowest triplet X_t^- for the light hole, but this time in σ^+ polarization, as one would expect from the equivalent diagram to Fig. 1(c) for the X^- light-hole excitons.

Closer inspection of the ER spectra in Fig. 3(b) shows that the X_t^- transition strengthens at lower electron densities than X_s^-. This is due to the greater spatial extent of X_t^-, as implied by its smaller second-electron binding energy. Hence a larger number of excess electrons will be captured by a photoexcited electron/hole pair into X_t^- (than X_s^-) at the lowest electron densities. As the gate bias is increased further, the X_t^- transition eventually weakens and merges into X_s^- in both the ER and PL spectra of Fig. 3. This is presumably due to screening and exclusion effects upon X_t^- at densities where there is more than one excess electron is the vicinity of each photoexcited or recombining exciton.

In conclusion, by applying a magnetic field, we induce transitions in the optical spectra of GaAs QW's containing a small density of excess electrons. The excess-electron-density dependence of these features demonstrates that they derive from excited states of the negatively charged exciton. Furthermore, the polarization dependence of the spectra shows these excited states correspond to spin-triplet combinations. The triplet states become bound at a finite magnetic field, due to the magnetic confinement of the outer (one-electron) orbital increasing its Coulombic interaction with the core. This enhancement in the binding energy of the second electron is also observed for the ground (singlet) negatively charged exciton.

Note added in proof. We have recently observed spin-triplet *positively* charged excitons in the magneto-optical spectra of *p*-type remotely doped QW's; see Ref. 20.

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