

Negatively and positively charged excitons in GaAs/Al_xGa_{1-x}As quantum wells

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We report the observation of the positively charged exciton and of the triplet state of the negatively charged exciton in modulation doped GaAs quantum wells. Applying a gate voltage at high magnetic fields we find that the photoluminescence line of the two-dimensional electron gas smoothly transforms into a negatively charged exciton and not into a neutral exciton. The Zeeman splitting of this line exhibits an abrupt change at the metal-insulator transition.

Bound complexes of electrons and holes dominate the optical spectra of semiconductors close to the band gap. Excitons and biexcitons, which are the analogs of hydrogen atoms and molecules, respectively, are the best known examples for such complexes. The trions, predicted by Lampert in 1958,¹ are much less familiar.² The two types of trions are the negatively charged exciton (X^-), which consists of two electrons bound to a hole, and the positively charged exciton (X^+), which consists of two holes bound to an electron. These complexes are analogous to the well known hydrogen ions H^- and H_2^+ . The trions binding energy is expected to increase substantially in two dimensions. Indeed, X^- was observed in CdTe/Cd_xZn_{1-x}Te quantum wells (QW's),³ and recently in GaAs/Al_xGa_{1-x}As QW's.⁴⁻⁶

It is clear that a large density of electrons, which could bind to the photoexcited electron-hole pairs, is needed for the X^- to be observed. However, a dense electron gas might screen the Coulomb interaction between the hole and the electrons and lead to unbinding of X^- . These considerations lead to the choice of a two-dimensional electron gas (2DEG) near the metal-insulator transition as the appropriate system for the observation of X^- .⁴

In this work we use optical spectroscopy to study the properties of two-dimensional electron and hole gases at magnetic fields from 0 to 9 T. By applying a gate voltage we are able to investigate the transition from the metallic to the insulating state using the same sample. In the insulating state we identify the *positively* charged exciton (X^+) and the *triplet* state (X_t^-) of the negatively charged exciton. The triplet state is found to be bound only at high magnetic fields. An important observation is that the photoluminescence (PL) line of the 2DEG in the metallic state smoothly transforms into X^- at the insulating state and not to an exciton. On the other hand, the Zeeman splitting of this PL line exhibits an abrupt change at the metal-insulator transition. We discuss the implications of these findings on the relations between the system of a 2DEG plus a hole and X^- .

The samples we used are single-sided modulation-doped QW's. The 2DEG and two-dimensional hole gas (2DHG) were grown on (100) and (311)A oriented GaAs wafers, respectively. The generic structure consists of a buffer superlattice, a 200-Å QW, an undoped spacer layer, a Si doped region, and a cap layer. The *n*-type samples have a spacer width of either 500 or 1500 Å and are δ doped (an additional uniform doping is introduced near the surface). We have also

measured several other samples, with different well and spacer widths. They exhibited the same qualitative behavior and will not be discussed here. The *p*-type sample has a 500-Å spacer followed by a 200-Å uniformly doped layer. The electron densities at zero gate voltage are $\sim 6 \times 10^{10} \text{ cm}^{-2}$ and $\sim 2 \times 10^{11} \text{ cm}^{-2}$ for the 1500- and 500-Å samples, respectively. The density of the 2DHG is $\sim 1 \times 10^{11} \text{ cm}^{-2}$.

We first look at the PL spectrum of the *p*-type sample. When depleting the 2DHG we observe a change in the PL, which is very similar to that observed in the *n*-type samples.⁴ The PL line shifts to higher energies and at the critical density of the metal-insulator transition another line appears at an energy 1.25 meV above. The higher energy line dominates the PL spectrum when the 2DHG is further depleted. We associate this line with a neutral exciton and the lower energy one with X^+ [Fig. 1(a)]. This behavior was observed in three samples with different doping levels.

The observed binding energy of X^+ (1.25 meV) is only slightly larger than that of X^- (1.15 meV) in a similar 2DEG structure [Fig. 1(b)]. This is in contrast to the large difference in binding energies between H_2^+ and H^- (2.7 vs 0.75 eV). The binding energies of bound complexes are generally a function of the electron and hole mass ratio. Unlike the large electron-proton mass ratio in hydrogen, the in-plane heavy-hole mass is only about three times larger than the electrons mass in the GaAs QW. Thus, a much smaller difference in binding energy is expected. Indeed, the numerical calculations of Ref. 7 give a difference in the binding energy of only 30%.

To substantiate the assignment of the PL peaks let us examine the expected polarization of the emitted light from a trion as compared to that of a neutral exciton. Consider a resonant excitation at the light-hole exciton energy with a circularly polarized light. We get electrons with a spin projection along the growth direction $s_z^{\text{el}} = +1/2$ and light holes with a total angular momentum projection $s_z^{\text{lh}} = +1/2$ ($\Delta s = s_z^{\text{el}} + s_z^{\text{lh}} = 1$). Within a few tens of picoseconds the hole population relaxes into the heavy-hole states and is equally distributed between the $s_z^{\text{hh}} = \pm 3/2$ states. On the other hand, the electron spin relaxation is relatively slow and a substantial number of electrons preserve their spin projection until they recombine. These different relaxation rates for holes and electrons determine the polarization of the emitted light.

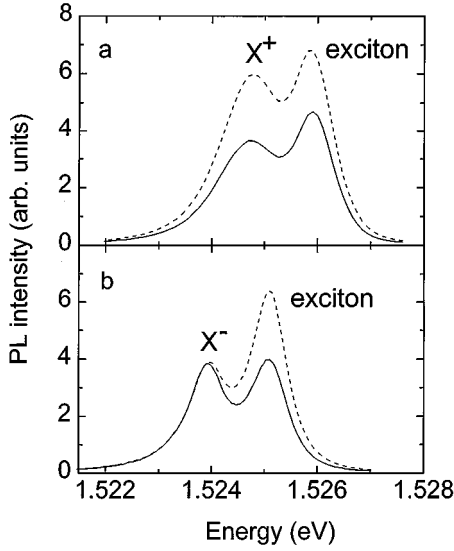


FIG. 1. The PL spectra of (a) the 2DHG sample and (b) the 1500-Å spacer 2DEG at the insulating state ($T=4$ K). Excitation is at the light-hole exciton energy using circularly polarized light. The solid line is copolarized with the excitation and the dashed is counterpolarized.

Since only the $s_z^{\text{hh}} = -3/2$ heavy hole can recombine with the $s_z^{\text{el}} = +1/2$ electron ($\Delta s = s_z^{\text{el}} + s_z^{\text{hh}} = -1$), the neutral exciton emission is predominantly counterpolarized relative to the incident light. The same behavior is expected in X^+ since its electron is polarized and can recombine with only one of the heavy-hole states. On the other hand, the two electrons in X^- are in a singlet state with opposite s_z^{el} , and therefore can recombine with both heavy-holes states. Consequently, the X^- emission should be unpolarized.

Figure 1 compares the polarization of the PL spectra of (a) p -type and (b) n -type samples at the insulating state. The illumination is performed by a circularly polarized light at resonance with the light-hole exciton absorption line. The solid and dotted curves represent the two circularly polarized components of the PL: copolarized with the incident light and counterpolarized, respectively. It is clearly seen that both the neutral exciton and X^+ are substantially polarized in a direction opposite to that of the illumination. On the other hand, X^- is unpolarized. We repeated the same measurement with the laser tuned to a higher heavy-hole line. It was found that the exciton and X^+ lines were copolarized with the incident light, while X_s^- remained unpolarized. This behavior is in good agreement with our assignment of the observed PL peak as X^+ . It also confirms that the two electrons in X^- form a singlet state, which will be labeled in the following as X_s^- .

A careful examination of the PL spectra [Fig. 1(a)] reveals that X^+ is even more polarized than the exciton. This is probably due to the fact that X^+ being charged is localized in the potential fluctuation of the remote ionized acceptors, and its electron can preserve its spin longer than the exciton. It should be noted that the polarization of the exciton line in the 500-Å spacer 2DEG was less pronounced than in the 1500-Å spacer 2DEG. Excitons are most probably formed in the

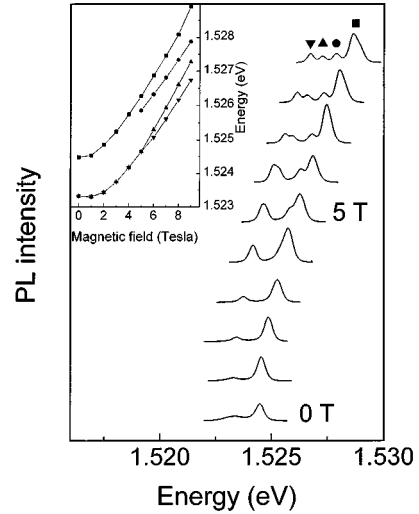


FIG. 2. The PL spectra of the 500-Å spacer 2DEG in the insulating state at different magnetic fields from 0 to 9 T ($T=4$ K). The inset shows the peaks positions as a function of the magnetic field. The squares denote excitons, circles, X_t^- , and triangles, X_s^- .

higher density sample by a photoexcited hole and one of the host electrons, which does not possess any information about the polarization of the light.

The second part of this paper focuses on the properties of the negatively charged exciton in high magnetic fields. The behavior of trions in a magnetic field had been recently addressed theoretically for quantum dot structures.⁸ Figure 2 describes the evolution of the PL spectra of a 2DEG in the insulating state from 0 to 9 T. At low magnetic fields $B < 4$ T, the spectra consist of a neutral exciton and X_s^- lines. For $B > 4$ T a new peak, denoted as X_t^- , appears between the exciton and X_s^- . This peak is observed also at the PL excitation spectra, indicating its intrinsic nature. Approximately at the same range of fields the X_s^- line starts to reveal a clear Zeeman splitting. The energy positions of all emission lines as a function of magnetic field are summarized in the inset of Fig. 2. We attribute the X_t^- line to X^- with its two electrons forming a *triplet* state. This state is not expected to be bound at zero magnetic field, but could become bound at high magnetic field, due to the quenching of the kinetic energy. This should occur when the magnetic length becomes comparable with the exciton diameter, at ≈ 4 T.

To prove the assignment of the X_t^- peak we have analyzed the polarization of the PL spectrum at 7 T and a temperature of 2 K. Under these conditions most of the host electrons are spin polarized $s_z^{\text{el}} = +1/2$. On the other hand, the photoexcited electrons are unpolarized and can pair with a host electron to form either a singlet or a triplet state, depending on the electron involved: an $s_z^{\text{el}} = +1/2$ electron will form X_t^- ($s_z^{\text{el}} = +1$) while an $s_z^{\text{el}} = -1/2$ electron can form both X_t^- and X_s^- ($s_z^{\text{el}} = 0$). We observed that X_t^- peak exhibits a strong circular polarization. It is copolarized with the lower energy component of the Zeeman-split X_s^- doublet, and counterpolarized to the higher energy component of X_s^- . This behavior is consistent with the assignment of the X_t^- line to a negatively charged exciton, in which the electrons are paired in an $s_z^{\text{el}} = 1$ triplet state. The strong polar-

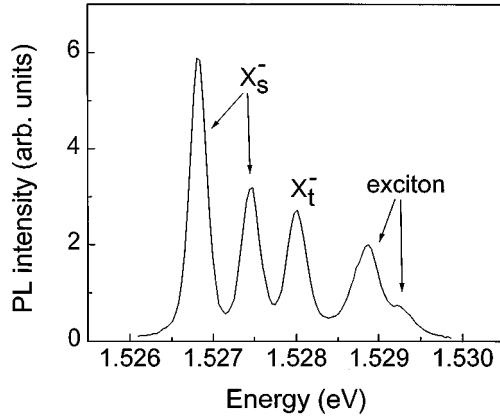


FIG. 3. The PL spectrum of the 1500-Å spacer 2DEG in the insulating state at 9 T ($T=2$ K).

ization of the X_t^- line is then due to the only possible recombination of the $s_z^{\text{el}} = +1/2$ electron with an $s_z^{\text{hh}} = -3/2$ heavy hole.

To further substantiate this assignment we have measured the PL excitation spectrum of the insulating state at strong magnetic field, where the 2DEG is spin polarized. Under these conditions a photoexcited electron can pair with a host electron to form either a singlet or a triplet state, depending on its polarization: $s_z^{\text{el}} = +1/2$ electron will form X_t^- ($s_z^{\text{el}} = +1$) while $s_z^{\text{el}} = -1/2$ electron can form X_s^- . Consequently, only the X_t^- and the high energy component of the X_s^- doublet should be observed in the excitation spectrum. This behavior was indeed observed in the excitation spectrum at 9 T and 2 K. The X_t^- peak was clearly observed and it was seen that the high energy component of the X_s^- doublet was much stronger than the low energy one. Kheng *et al.*³ used similar arguments to assign the singlet negatively charged exciton in the CdTe/Cd_xZn_{1-x}Te QW.

As we apply a gate voltage beyond the metal-insulator transition the relative strength of X_s^- and X_t^- lines varies

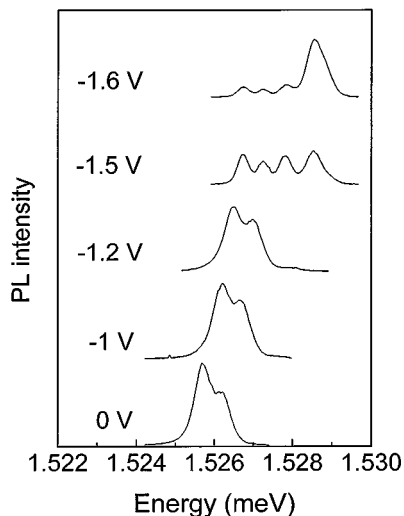


FIG. 4. The PL spectra of the 500-Å spacer 2DEG at 9 T at different gate voltages ($T=4$ K). The lower and upper curves correspond to the metallic and insulating states, respectively.

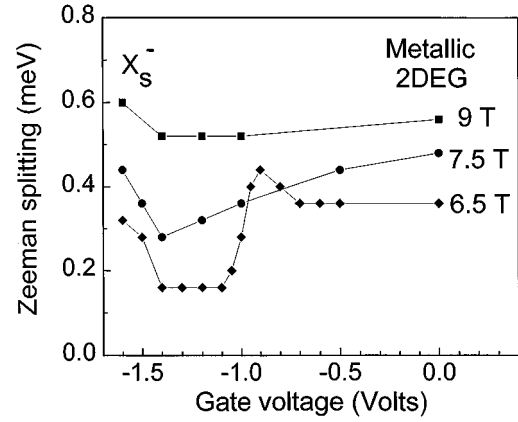


FIG. 5. The Zeeman splitting of the metallic 2DEG and X_s^- lines as a function of the gate voltage at different magnetic fields ($T=2$ K). The enhanced splitting at -1 V and 6.5 T occurs when $\nu=1$ (Ref. 13).

with respect to the neutral exciton, but they keep the same relative strength among themselves (see, for example, the spectra taken at -1.5 and -1.6 V, Fig. 4). This indicates that X_s^- and X_t^- lines indeed have a similar origin and their intensity scales with the electron density.

Figure 3 shows the PL spectrum of the 1500-Å spacer 2DEG in the insulating state at 9 T. It is seen that the Zeeman splitting of the X_s^- peak is significantly ($\sim 50\%$) larger than that of the neutral exciton. The origin of this difference can be attributed to the strong mixing between the heavy- and light-hole states at wave vectors of the order of the inverse exciton radius. As a result of this mixing the heavy-hole exciton Zeeman splitting has a large light-hole contribution. On the other hand, the X_s^- spatial extent is larger than that of the exciton and therefore is less affected by the light-hole admixture. Since the effect of the light-hole admixture is to decrease the effective Zeeman splitting, X_s^- exhibits a larger splitting.

Figure 4 shows the PL spectra of the 500-Å spacer 2DEG at 9 T at several different applied gate voltages. Small absolute gate voltage values correspond to a metallic state of the 2DEG, while large absolute values correspond to an insulating state. One could see that the PL spectrum of the metallic 2DEG consists of two lines, which smoothly transform into the two components of X_s^- . The exciton line appears at a distinct energy ~ 2 meV above. This observation implies that the physics of a 2DEG plus a hole system in a high magnetic field could be better described as X^- dressed by the 2DEG rather than an exciton.⁹

A close examination of X_s^- and the metallic 2DEG PL lines (Fig. 4) shows that their Zeeman splitting varies with gate voltage. This behavior is summarized in Fig. 5, which shows the change of the Zeeman splitting as we go all the way from a metallic to an insulating state. Data for lower magnetic fields are also shown. It can be seen that the Zeeman splitting of the metallic 2DEG line decreases with gate voltage and reaches a minimum at ~ -1.4 V. This behavior is more pronounced at the lower magnetic fields. Beyond this voltage the Zeeman splitting jumps to the X_s^- value. Thus, the measurement of the Zeeman splitting tells us where the crossover between X_s^- and a metallic 2DEG recombination

occurs in the smoothly transforming PL line. Notice that at the same range of gate voltage the neutral exciton first appears in the PL spectra (Fig. 4). Transport measurements show that this transition voltage coincides with that of the metal-insulator transition.⁴

Let us discuss the origin of the changes in the Zeeman splitting with gate voltage. It was shown that an electric field in the growth direction lifts the spin degeneracy of the heavy-hole state and produces an effective Zeeman splitting at zero magnetic field.^{10,11} It was also shown that this mechanism gives the dominant contribution to the Zeeman splitting in heterojunctions at magnetic fields < 10 T.¹¹ Indeed, a very large built-in electric field is present in our single-sided modulation-doped QW samples at zero gate voltage. Applying negative gate voltages this electric field decreases, which results in a reduction of Zeeman splitting. This interpretation is supported by the behavior of the 1500-Å spacer 2DEG. In this sample the built-in electric field is much smaller and indeed the Zeeman splitting of the metallic state exhibits a much weaker dependence on gate voltage. The important observation is that the Zeeman splitting values in the metallic state of that sample almost coincide with the minimum Zeeman splitting of the 500-Å sample at the three magnetic fields. One can conclude that these values correspond to the

bare (vanishing electric field) Zeeman splitting of the metallic 2DEG line. These bare values are smaller than the Zeeman splitting of X_s^- but larger than those of the neutral exciton. This observation may imply that the spatial extent of the valence hole wave function in the metallic 2DEG plus hole system is intermediate between that of X_s^- and a neutral exciton. Clearly, a further theoretical work is needed to clarify this behavior.

In conclusion, we have used optical spectroscopy to study trions in GaAs QW's, emphasizing their polarization properties. The observation of X_t^- binding at high magnetic fields and of the similar binding energies of X_s^- and X^+ provides an opportunity to extend our understanding of ionized bound complexes in semiconductors. Finally, the observed similarities and differences between X_s^- and the metallic 2DEG recombination lines can serve in gaining a deeper insight into the physics of the 2DEG.

Recently Shields *et al.* have reported the observation of positively charged excitons and of the triplet state.¹² The reported binding energies and properties in high magnetic field are consistent with our observations.

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