Optical spectroscopy of neutral and charged excitons in GaAs/AlGaAs quantum wells in high magnetic fields

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Abstract

We implement optical spectroscopy to study charged excitons in modulation-doped GaAs/AlGaAs quantum wells. We report the first observation of the positively charged exciton and of the triplet state of the negatively charged exciton. Applying a gate voltage at high magnetic fields, we investigate the transition between metallic and insulating states. We find that while the photoluminescence line of the metallic two-dimensional electron gas transforms smoothly into a negatively charged exciton, the Zeeman splitting of this line exhibits an abrupt change at the metal-insulator transition.

Keywords: Gallium arsenide; Molecular beam epitaxy; Photoluminescence; Quantum wells; Semiconductor-semiconductor heterostructures

1. Introduction

Charged excitons are the semiconductor analog of the $H^-$ and $H^+_2$ hydrogen ions. Their existence was first discussed nearly 40 years ago [1] and they were first observed in bulk Ge and Si [2]. The first observation of charged excitons in confined structures was only recently reported [3]. It is clear that a large density of electrons (holes), which could bind to the photoexcited electron–hole pairs, is needed for the negatively (positively) charged exciton to be observed. However, a dense electron or hole gas might screen the Coulomb interaction and lead to unbinding of the charged exciton. These considerations led us to the choice of a two-dimensional electron gas (2DEG) near the metal–insulator transition as the appropriate system for the observation of the negatively charged exciton $X^-$. Indeed, we have recently shown that under these conditions a new line appears in the optical emission and excitation spectra of a high-quality modulation-doped GaAs/AlGaAs quantum well (QW) [4]. We associated this line, which is $\sim 1.2 \text{ meV}$ below the exciton line, with an $X^-$. A similar lineshape was observed in a resonant tunneling structure, when a low density of electrons is accumulated in the well [5].

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.

2. The negatively charged exciton $X^-$

Fig. 1 describes the evolution of the PL and PLE spectra of a 2DEG sample (500 Å spacer and
Fig. 1. Evolution of the PL (solid line) and PLE (dotted line) spectra of the 500 Å spacer 2DEG with the gate voltage. The arrow represents the PLE detection energy (1.5212 eV).

200 Å QW) with gate voltage. The PLE spectrum at zero gate voltage shows a step-like behavior, typical for absorption in presence of a 2DEG. The PL spectrum is Stokes shifted due to the relatively large 2DEG density of \( \sim 2 \times 10^{11} \text{cm}^{-2} \) (Burstein–Moss shift). At \(-1.5\) V two broad peaks, corresponding to the onset of the heavy and light hole transitions, are seen.

As the gate voltage approaches the metal–insulator transition, the optical spectra change drastically. The Stokes shift decreases and both the PL and PLE peaks narrow to form a sharp resonance, denoted \( X_{\text{hh}} \), at the band edge. At \(-1.6\) V three broad features \( (X_{\text{hh}}, X_{\text{yh}} \text{ and } X_{\text{th}}) \) are seen above this sharp resonance. As the gate voltage is further decreased the \( X_{\text{hh}} \) evolves into the heavy-hole exciton and the \( X_{\text{th}} \) into the light-hole exciton. At \(-1.65\) V the Stokes shift between the PL and the PLE has nearly disappeared. This is despite the presence of a relatively large electron density in the well \( (\sim 1 \times 10^{11} \text{cm}^{-2}) \). At this gate voltage, and more pronouncedly at \(-1.67\) V, there is a unique double-peak structure, consisting of a heavy-hole exciton and \( X^- \). We observe that as the \( X^- \) peak decreases the heavy-hole exciton grows. We identify the lowest-energy peak as the heavy-hole negatively charged exciton, which is formed by binding of a photoexcited electron and heavy-hole pair to one of the host electrons, localized in the potential fluctuations of the remote ionized donors [4].

Examining the light-hole related peaks at the PLE spectra we observe a replica of this behavior. The appearance of the light-hole exciton and the shrinking of the \( X_{\text{th}}^- \) are strongly correlated with the growth of the heavy-hole exciton. One can thus identify this \( X_{\text{th}}^- \) peak as the light-hole negatively charged exciton.

3. The positively charged exciton \( X^+ \)

When depleting the 2DHG (500 Å spacer, 200 Å QW) we observe a change in the PL, which is very similar to that observed in the \( n \)-type samples. The PL line shifts to higher energies and at a certain critical density a neutral exciton line appears at an energy which is 1.25 meV higher. We associate the low-energy line with \( X^+ \). This behavior was observed in three different samples, with different doping levels, and in different pieces of the wafers.

To substantiate the assignment of the PL peaks, let us examine the expected polarization of the emitted light from a charged exciton 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 \( M = \pm 1/2 \) and light-holes with a total angular momentum projection \( s^h = \pm 1/2 \). Within a few tens of ps the hole population relaxes into the heavy-hole states and is equally distributed between the \( s^h = \pm 3/2 \) 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. Since only the \( s^h = -3/2 \) heavy-hole can recombine with the \( s^e = +1/2 \) electron, the neutral exciton emission is predominantly counter-polarized relative to the incident light. The same behavior is expected in \( X^+ \) since its electron is polarized and can recom-
bine with only one of the heavy-hole states. On the other hand, the two electrons in $X^-$ are in a singlet state with opposite $s^d$ and can recombine with both heavy-hole states. Consequently, the $X^-$ emission should be unpolarized.

Fig. 2 compares the polarization of the PL spectra of the (a) $p$-type and (b) the $n$-type samples at the insulating state. The illumination is performed by a circularly polarized light at the light-hole exciton absorption line. The solid and the dotted curves represent the two circularly polarized components of the PL: co-polarized and counter-polarized with the incident light, respectively. It is clearly seen in Fig. 3 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 co-polarized with the incident light, while $X^-$ 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 $X_{s^-}$.

4. The triplet $X_{t^-}$

Fig. 3 shows the evolution of the PL spectra of a 2DEG in the insulating state from 0 to 9 T. At $B > 4$ T a new peak, denoted $X_{t^-}$, appears between the exciton and $X_{s^-}$. Approximately at the same range of fields, the $X_{s^-}$ line starts to reveal a clear Zeeman splitting. We attribute the $X_{s^-}$ line to a negatively charged exciton 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 fields, due to the quenching of the kinetic energy. This should occur when the magnetic length becomes comparable with the exciton diameter, i.e. at $\sim 4$ T.

To prove the assignment of the $X_{t^-}$ peak we examine the PL spectra at high magnetic fields and low temperatures, when the electron gas is spin polarized ($s^d = +1/2$). Fig. 4 shows the PL spectra in the two circular polarizations at 7 T and

![Fig. 2. PL spectra of (a) the 2DHG sample and (b) the 1500 Å spacer 2DEG at the insulating state ($T=4$ K). The solid line is co-polarized with the excitation and the dashed is counter-polarized.](image)

![Fig. 3. PL spectra of the 500 Å spacer 2DEG in the insulating state from 0 to 9 T ($T=4$ K). Inset: the positions of the peaks as a function of the magnetic field. (■) Excitons, (●) $X_{s^-}$, (▼) $X_{t^-}$.](image)
2 K. We observe that the $X_t^-$ peak exhibits a strong circular polarization. It is co-polarized with the lower-energy component of the Zeeman-split $X_t^-$ doublet, and counter-polarized to the higher-energy component of $X_t^-$. This behavior is consistent with the assignment of the $X_t^-$ line to a negatively charged exciton, in which the electrons are paired in $\hat{S}=1$ triplet state. This behavior is consistent with the assignment of the $X_t^-$ line to a negatively charged exciton, in which the electrons are paired in $\hat{S}=1$ triplet state. The strong polarization of the $X_t^-$ line is due to the fact that the only possible recombination process of the $\hat{S}^e=+1/2$ electron is with an $\hat{S}^{hh}=-3/2$ heavy-hole.

To further substantiate this assignment we studied the PLE spectra of a spin-polarized 2DEG. Under this condition a photoexcited electron can pair with a host electron to form either a singlet or a triplet state, depending on its polarization: the $\hat{S}^e=+1/2$ electron will form $X_t^-$ ($\hat{S}^{hh}=+1$) while the $\hat{S}^e=-1/2$ electron can form $X_t^-$. In the first case we get absorption at the $X_t^-$ energy and in the second, we get absorption at the high-energy component of the Zeeman-split $X_t^-$. This behavior is indeed observed in Fig. 5, which shows the PLE spectrum at 9 T and 2 K. The $X_t^-$ peak is clearly visible and it is seen that the high-energy component of the $X_t^-$ doublet is much stronger than the low-energy one. Similar arguments were used by Kheng et al. to prove the assignment of the peak in the PL and absorption of CdTe/CdZnTe QW with a negatively charged exciton [3].

We note that as we apply a gate voltage beyond the metal–insulator transition the relative strength of the $X_t^-$ and $X_t^-$ lines vary with respect to the neutral exciton, but they keep the same relative strength among themselves. This indicates that the $X_t^-$ and $X_t^-$ lines indeed have a similar origin and their intensity scales with the density of electrons in the sample.

The second triplet state, with $\hat{S}^e=0$, is not observed. A possible explanation is that it rapidly transforms into the singlet state, due to interaction of the total electron spin with the spin of the hole. We also do not observe the $X_t^-$ with $\hat{S}^{hh}=-1$ state. Clearly, it cannot be formed in a spin-polarized 2DEG with $\hat{S}^e=+1/2$.

5. Zeeman splitting and the metal–insulator transition

The Zeeman splitting of the $X_t^-$ peak is significantly (~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 $X_t^-$ 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 splitting, $X_t^-$ exhibits a larger splitting.

Fig. 6 shows the PL spectra of the 500 Å spacer 2DEG at 9 T at several different applied gate
Metallic 2DEG

![Diagram of PL spectra and Zeeman splitting](image)

**Fig. 6.** 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. Inset: Zeeman splitting of the metallic 2DEG and X⁻ 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 v = 1 [9].

Small absolute gate voltage values correspond to a metallic state of the 2DEG, while large absolute values correspond to an insulating state. One can see from Fig. 6 that the PL spectrum of the metallic 2DEG consists of two lines, which smoothly transform into the two components of X⁻. The exciton line appears at a distinct energy which is ~2 meV higher. This observation implies that the physics of a 2DEG plus a hole system in a high field could be better described as X⁻ dressed by the 2DEG rather than an exciton [6].

An examination of X⁻ and the metallic 2DEG PL lines (Fig. 6) shows that their Zeeman splitting varies with gate voltage. This behavior is summarized in the inset to Fig. 6, which shows the change of the Zeeman splitting as we go all the way from a metallic state 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 lower magnetic fields. Beyond this voltage, the Zeeman splitting jumps to X⁻ value. Thus, the measurement of the Zeeman splitting tells us where the crossover between X⁻ 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. 6). Transport measurements show that this transition voltage coincides with that of the metal–insulator transition [4].

Let us now 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 [7,8]. This mechanism gives the dominant contribution to the Zeeman splitting in heterojunctions at magnetic fields <10 T [8]. Indeed, a very large built-in electric field is present in our single-side 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 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⁻ 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⁻ and a neutral exciton.

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References