Comparative study of the negatively and positively charged excitons in GaAs quantum wells

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We compare the photoluminescence spectra of the negatively and positively charged excitons in GaAs quantum wells. We use a structure which enables us to observe both complexes within the same sample. We find that their binding energy and Zeeman splitting are very similar at zero magnetic field, but evolve very differently at high fields. We discuss the implications of these observations on our understanding of the charge excitons structure in high magnetic fields. [S0163-1829(99)51516-6]

The negatively charged exciton X^-, which is a bound state of two electrons and a hole, was recently observed in the emission and absorption spectra of a depleted two-dimensional electron gas (2DEG).[1–3] It appears in the photoluminescence (PL) spectrum of GaAs quantum wells as a narrow line, approximately 1 meV below the neutral exciton line. This energy is the binding energy of an extra electron to an exciton. The observation of X^- triggered intensive experimental and theoretical studies of its energy spectrum and structure. A special effort was devoted to understanding its behavior in strong magnetic fields, when the cyclotron diameter becomes smaller than the X^- Bohr diameter, and the internal structure is expected to be modified. Indeed, a significant increase in binding energy and appearance of another bound state, in which the two electrons are in a triplet state, were observed in the optical spectrum in high magnetic fields.[4,5]

Very soon after the observation of the X^-, its positive counterpart, the positively charged exciton X^+, was observed.[4] The X^+ consists of two holes and an electron, and is a semiconductor analogue of the hydrogen molecule ion $H_2{}^+$. It was found that the X^+ line emerges from the two-dimensional hole gas (2DHG) PL as the hole density is decreased, very similarly to the X^- appearance in a 2DEG.[6] In this paper we investigate the X^+ spectrum in GaAs/Al$_x$Ga$_{1-x}$As QW at high magnetic fields (0 to 9 T) and compare it to that of X^-. To obtain a meaningful comparison between the two bound complexes we design a structure where we can control the density and type of the excess carriers in the QW by changing the illumination conditions. This structure allows us to alter the carrier gas in the well from 2DHG to 2DEG, and study the X^+ and X^- spectra *within the same sample*.

We find that the binding energies of the two charged excitons are nearly identical at zero magnetic field. However, our results show a profound difference between the spectra of X^+ and that of X^- at a high magnetic field which is applied in a direction normal to the layers. We observe a very different dependence of the binding energy in the two complexes on the magnetic field strength: while X^- exhibits a significant increase in binding energy with increasing field (more than 60% at 7 T), X^+ *binding energy remains nearly constant*. A large difference is observed also between the Zeeman splitting of the X^+ and that of X^-. We discuss the implications of these observations on our understanding of the charge excitons structure in high magnetic fields.

Two nominally undoped samples were studied. Their structure is schematically described in the inset of Fig. 1. It consists of a buffer superlattice made of 25 periods of 10 nm Al$_{0.25}$Ga$_{0.75}$As and 1 nm of GaAs followed by a 20 nm GaAs well, an Al$_{0.37}$Ga$_{63}$As layer denoted as the Spacer, and 15 nm of GaAs cap layer. The thickness of the Spacer layer is 25 and 125 nm in samples 1 and 2, respectively. The samples were grown on (100) oriented semi-insulating GaAs substrates. Under very weak excitation (<10 μW/cm^2) the observed spectrum is of neutral exciton, indicating that the well is nearly empty of carriers. However, when the intensity of the laser is increased the carrier density in the well grows, and charged excitons are observed in the spectrum. The carrier type is determined by the laser photon energy. When this energy is slightly above the GaAs gap ($h\nu_1$), carriers are created in the well and the cap layer. The built-in electric field that is due to the unintentional p-type background doping causes electrons which are excited in the well to tunnel into the cap layer. This process gives rise to electrons deficiency in the well. Similarly, the photoexcited holes in the cap layer may tunnel to the well and accumulate there. These processes are much more effective in the thinner Spacer
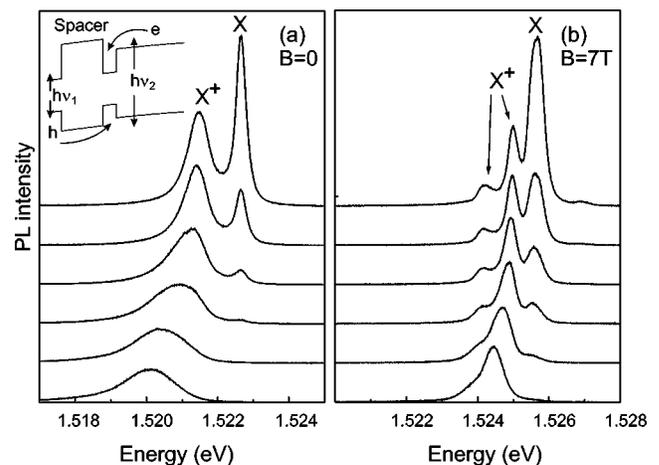

FIG. 1. The PL spectra of sample 1 for increasing He:Ne intensity, between 0 (bottom curve) and 10 (top curve) mW/cm^2. The Ti:S intensity is 100 mW/cm^2. (a) and (b) correspond to measurements at 0 and 7 T, respectively. Inset: The generic structure of samples 1 and 2.

sample 1, resulting in a higher hole density than in sample 2. On the other hand, when the laser is tuned to a higher energy ($h\nu_2$), slightly above the gap of the low $Al_xGa_{1-x}As$ superlattice, most carriers are created at that region. In this case the built-in electric field causes the electrons to be swept into the well and create an excess electron density there. Thus, by changing the excitation energy we can change the carrier gas in the well from 2DHG to 2DEG. In our experiment we use a Ti:S laser at 780 nm and a He:Ne laser at 632.8 nm as the low ($h\nu_1$) and high ($h\nu_2$) energy excitation sources, respectively. The polarized PL measurements are conducted in a 7 T immersion cryostat. The PL is collected through a birefringence free optical window and analyzed through a circular polarizer. Unpolarized PL measurements are carried out in a 9 T magnet, using a fiber optic collection setup. The signal is dispersed in a 0.75 m spectrometer and detected in a cooled charge-coupled device detector.

Figure 1(a) shows the measured PL spectrum of sample 1 at zero magnetic field. The different spectra are measured with a constant Ti:S intensity of ~ 100 mW/cm^2, and with an increasing He:Ne intensity, from 0 to ~ 10 mW/cm^2. Under Ti:S excitation only (bottom curve) the well contains an excess hole density, and the PL spectrum is a broad line, typical of the recombination of a free carrier gas. The effect of the He:Ne laser is to reduce the hole density by supplying electrons to the well. We can see that as the He:Ne intensity is increased the width of the line decreases and it shifts to higher energies. Since the width of the line is related to the holes' Fermi energy, this narrowing of the line reflects the decrease in the hole density. Similarly, the shift to higher energy is due to a reduced band-gap renormalization and a lower built-in electric field in the well. At the largest He:Ne intensity the spectrum consists of two lines, which we identify as the neutral exciton (X) and the X^+ lines. It should be noted that this spectral signature, of two narrow peaks separated by ~ 1 meV which evolve from a broad line, is characteristic of both the $X-X^-$ and the $X-X^+$ doublets, and by itself cannot serve as a criterion for identifying the type of charged exciton. We show in the following that the PL spectrum in a magnetic field enables us to distinguish between the X^+ and the X^-.

The spectral signature of the X^- in a high magnetic field which is applied normal to the layers is rather unique.[4,5] It consists of several lines, associated with the singlet and triplet states of the two electrons in the X^-. These singlet and triplet states have different spatial wave functions, symmetric and antisymmetric, respectively, and hence different binding energies. It is found that these lines move away from the exciton with increasing magnetic field, reflecting an increase in binding energy of the corresponding states. In addition, a series of weak satellite peaks are observed at the low-energy tail of the emission spectrum.[7] These peaks result from shakeup processes, in which a recombination of one of the electrons in the X^- with the hole is accompanied by an ejection of the remaining electron to a high Landau level. This unique spectral signature, consisting of singlet, triplet, and shakeup lines, enables a clear identification of the X^-.

Figure 1(b) shows the PL spectra in a magnetic field of 7 T applied in the direction normal to the layers, for similar excitation conditions as in Fig. 1(a). The measured spectral signature is different than that reported for X^-.[4,5] This is
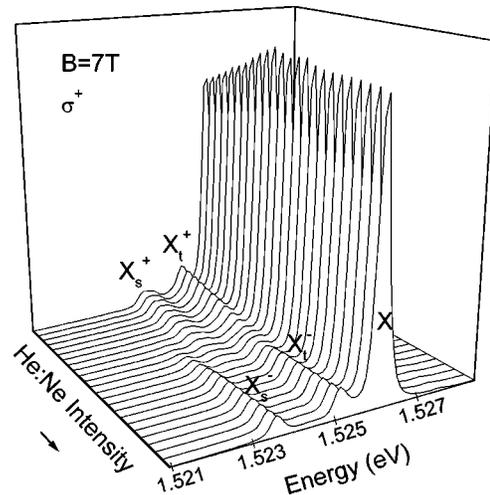

FIG. 2. Evolution of the σ^+ polarized PL spectra at 7 T with increasing He:Ne intensity for sample 2. The evolution from a X^+ to X^- spectrum through a neutral exciton gas is clearly seen. Each spectrum is normalized by its integrated intensity.

especially manifested in the energies of the two low energy lines, which evolve from the charged exciton line. These differences will be discussed in detail later in this paper. A supporting evidence for the identification of the X^+ is provided by measurements of p-type modulation doped samples which contain a 2DHG in the QW. These samples are grown on $(311)A$ oriented GaAs substrates. The generic structure is described in Ref. 4. Different samples with different spacer widths (between 25 to 50 nm) and doping levels (5×10^{17} to 2×10^{18} cm^{-3}) were studied. The density of holes in the QW is controlled by applying a positive voltage to a semitransparent gate with respect to the 2DHG, and the exciting photon energy is kept below the barrier energy gap. The spectral signature, which is observed in these samples, is the same as in Fig. 1(b), only with broader lines. We therefore conclude that this is indeed the spectrum of X^+.

Let us now turn to sample 2. The fact that the Spacer layer is much thicker than in sample 1 causes the initial density of holes in the well to be lower. Figure 2 describes the PL spectrum at 7 T for similar illumination conditions as in Fig. 1. To clarify the evolution of the spectrum we present a circularly polarized spectrum of σ^+. The change from 2DHG to 2DEG spectrum through a neutral exciton gas is clearly visible. This change is manifested by the change of the PL spectrum from that associated with X^+ to that of X^-. Both the X^+ and X^- spectra consist of singlet and triplet lines. We can see that the strength of the X^+ lines decreases gradually with increasing He:Ne intensity, until they disappear and new lines, associated with X^- appear. These changes are accompanied by a dramatic change in the low-energy part of the spectrum, which is too weak to be seen in the figure. Together with the X^- lines we observe the familiar fan of weak shakeup satellite peaks, separated from one another by $\hbar\omega_c^e$, the *electron* cyclotron energy.[7] The low-energy spectrum of X^+ is very different: the electron shakeup lines are not present and a complicated spectrum of impurity related faint lines appears. Remarkably, the energy of the exciton line is fixed in all spectra, providing a constant reference energy.
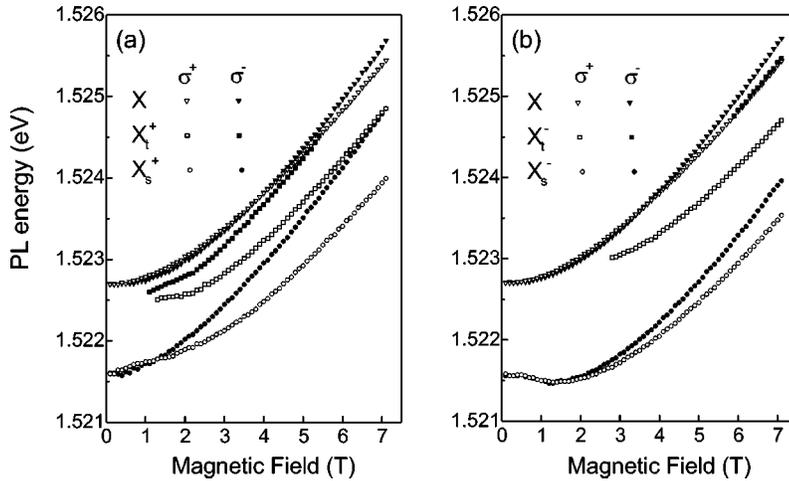
FIG. 3. The energy dispersion of the (a) X^+ and (b) X^- PL peaks as a function of magnetic field. The full and empty symbols stand for the σ^- and σ^+ polarizations, respectively.

Figures 3(a) and 3(b) show the energy dispersion of the X^+ and X^- lines as a function of magnetic field. The Zeeman split neutral exciton state appears at the highest energies, and is characterized by a change of the g factor sign at 3.5 T. The general structure of the energy spectrum is the same for X^- and X^+: they both consist of two pairs of Zeeman split circularly polarized lines. The lowest energy pair is due to a recombination of the singlet X^- (or X^+), and the higher energy pair is due to the recombination from the triplet state (this triplet state becomes bound at a magnetic field and emerges below the exciton line).

In Fig. 4 we compare the binding energy and Zeeman splitting of the singlet state of these two complexes. The binding energy is measured relatively to the exciton state and the mean energy is taken for each Zeeman pair. It can be seen that the binding energy of the singlet X_s^- state increases between 0 and 4 T and then begins to saturate. Between 7 and 9 T it remains nearly constant. The overall increase is from 1.1 meV at 0 T to 1.8 at 9 T, more than 60%. This saturation of the X_s^- was found to persist up to 20 T.[8] The singlet X_s^+ behavior is very different: *the binding energy remains nearly constant* between 0 and 9 T. This behavior of the X_s^+ is observed also in the modulation doped samples. It should be noted here that this observation contradicts an earlier report which found that the behavior of the X^+ binding energy is very similar to that of X^-.[9]

Let us discuss the behavior of the binding energy in the two complexes, starting with the zero magnetic-field behavior. There were several theoretical attempts to compare the zero-field binding energies of X^+ and X^- using a two-dimensional model for the structure of the charged excitons.[10] These calculations yield an X^+ binding energy that exceeds that of X^- by a value which depends on the electron-hole mass ratio. Figure 4(a) clearly shows that the binding energies of X^+ and X^- *are identical at zero field*, with an accuracy of 0.05 meV, namely 5% of the binding energy. We have confirmed that this is not an accidental degeneracy: a similar behavior was observed in a sample with 15 nm well width. We wish to remark that the accuracy of these measurements stems from the fact that both complexes are observed in the same sample, with the exciton line appearing at a constant energy. To explain this observation within the present two-dimensional models one would have to assume a very light in-plane hole mass, close to the elec-

tron mass, which is very unlikely. For example, according to the calculations of Ref. 10 an equal binding energy implies $m_e/m_h > 0.8$. We note, however, that these models assume identical electron-electron and hole-hole interactions in the plane, i.e., $V_{ee}(r) = V_{hh}(r)$. This assumption is invalid for a triangular well, in which the 2DEG or 2DHG is confined along the growth direction z. The different electron and hole masses along z give rise to $V_{hh}(r) > V_{ee}(r)$, and hence to a larger repulsion between the holes in X^+ and lower binding energy.

Let us turn now to the behavior at high magnetic fields. Charged excitons are characterized by a delicate balance between a large Coulomb attraction and a slightly smaller repulsion. This balance is strongly affected by a high magnetic field, which squeezes the in-plane motion to a cyclotron diameter. This has a larger impact on the X^+, where the repulsion interaction V_{hh} is larger. This can explain the fact that the X^- binding energy becomes larger than that of the X^+ at high magnetic fields. Calculations of the structure and binding energy in a magnetic field were conducted only for X^-,
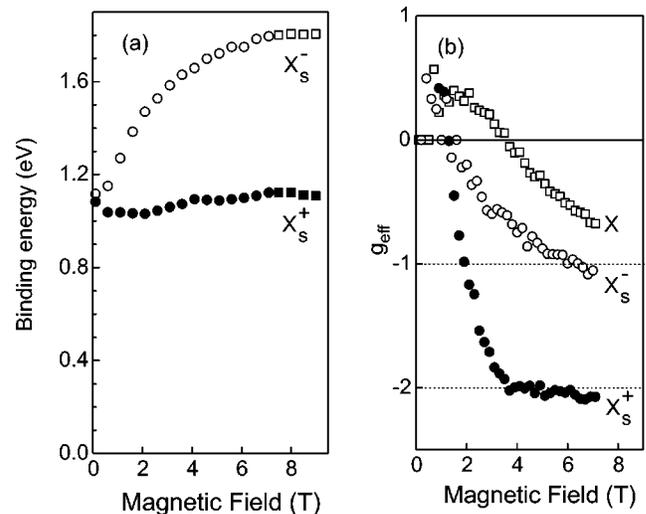

FIG. 4. (a) The binding energies of the X_s^+ and X_s^- states as a function of magnetic field. The circles and squares correspond to the polarized and unpolarized measurements, respectively. (b) The effective g factor g_{eff} of the neutral exciton; the X_s^- and X_s^+ as a function of magnetic field. Note that the neutral exciton Zeeman splitting is the same for both X^+ and X^- spectra.

using a numerical approach.[11,8] The results agree qualitatively with the measured spectra, and show a trend of increased binding energy with magnetic field. However, the values obtained for the binding energy are smaller than those measured experimentally.

In Fig. 4(b) we show the magnetic field dependence of the g factor of the X, X_s^+, and X_s^- recombination. The observed Zeeman splitting of a charged exciton is a sum of the electron and hole contributions, similarly to a regular exciton. Thus, one can define an effective g factor for X_s^+ and X_s^-, in the same way it is done for a regular exciton,[12] by $\Delta E = \mu_B B_z g_{X^\pm}$, where μ_B is the Bohr magneton (the sign of g is determined using the conventions of Refs. 12 and 13). The behavior of g_X in the X^+ and X^- spectra is very similar, and we show only the values obtained from the $X - X^-$ spectrum. It is seen that all exciton complexes have the same g factor value of $\sim +0.5$ at very low fields, but evolve differently as the magnetic field is increased. The behavior of g_{X^+} is especially interesting: it decreases very rapidly, changes sign, and then saturates at a value of -2 at 4 T.

The difference in g factors between the $X^\pm$ and X indicates that at high magnetic field the electron and hole wave functions in a charged exciton are not the same as in a neutral exciton. The magnetic-field dependence of g_X is commonly understood as due to mixing with light-hole states.[14]

This mixing cannot explain the strong field dependence of $g_{X^\pm}$: it gives rise to a relatively weak field dependence of g_X, and its effect is expected to be even weaker for $X^\pm$, which are farther away in energy. On the other hand, the modification of the charged exciton structure in a magnetic field which was discussed above implies mixing of the electron and hole wave functions with higher Landau levels and higher subbands. This mixing gives rise to a magnetic-field dependence of $g_{X^\pm}$. Such mixing was indeed found to play an important role in determining the binding energy in the calculations of Ref. 11. The observed saturation in both Zeeman splitting and binding energy of X^+ can be viewed as an indication of a formation of a stable spatial structure.

In conclusion, we have provided an unambiguous identification of the X^+ spectrum at high magnetic field, and have shown that this spectrum is different from that of the X^-. Understanding the structure and the spectrum of charged excitons in this regime is still an open problem. We believe that the experimental data presented here, which compares the spectrum of X^- and X^+ within the same sample, can serve as a basis for theoretical studies of this problem.

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