# Magnetic-field dependence of the spin states of the negatively charged exciton in GaAs quantum wells

T. Vanhoucke and M. Hayne*
*Laboratorium voor Vaste-Stoffysica en Magnetisme, Katholieke Universiteit Leuven, Celestijnenlaan 200 D, B-3001 Leuven, Belgium*

M. Henini
*School of Physics and Astronomy, University of Nottingham, Nottingham NG7 2RD, United Kingdom*

V. V. Moshchalkov
*Laboratorium voor Vaste-Stoffysica en Magnetisme, Katholieke Universiteit Leuven, Celestijnenlaan 200 D, B-3001 Leuven, Belgium*
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We present high-field ($<50$ T) photoluminescence measurements of the binding energy of the singlet and triplet states of the negatively charged exciton in a 200-Å quantum well. Comparing our data with those of other groups and with theoretical predictions we clearly show how the singlet, "bright" and "dark" triplet states may be identified according to the high-field dependence of their binding energies. We demonstrate that a very consistent behavior of the binding energy in a magnetic field has been observed in quantum wells of different widths by different groups and conclude that the triplet state found in this, as well as nearly all other experiments, is undoubtedly the bright triplet. By combining our data with that in the literature we are able to present the generic form of the binding energy of the spin states of the charged exciton in a magnetic field, which reveals the predicted singlet to dark triplet ground state transition at about 20 T.

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The bound state of a hole and two electrons, the negatively charged exciton or trion ($X^-$), has been the subject of intense investigation since its first observation in 1993.[1] In particular, the character of the spin states and the dependence of their binding energies on a magnetic field have been the subject of much debate.[2–9] It is widely accepted that in zero magnetic field the spin singlet is the only bound state of the charged exciton, while the triplet state is stabilized by the application of a magnetic field.[2] The experimental observation of a bound triplet at finite field was nonetheless somewhat controversial, as it was expected that the triplet state should be "dark,"[3] due to the fact that the $z$ component of its orbital angular momentum $L_z=-1$, making an optical recombination, in which an electron is left in the lowest Landau level, forbidden. A relaxation of this selection rule is possible when translational invariance is broken, e.g., by well width fluctuations.[3] However, this could not explain why the theoretically predicted singlet-triplet crossing, in which the triplet becomes the ground state at experimentally accessible magnetic fields, was not observed.[2,4] An answer to both these questions was found by Wójs, Quinn, and Hawrylak (WQH),[5] who showed the existence of a second, higher-energy "bright" triplet state with $L_z=0$. They proposed that it was the bright triplet that was seen in experiments, thereby explaining the observation of a triplet state and also why it never crossed the singlet to become the ground state in high fields. The theory of WQH was rapidly confirmed by new experiments, which showed a very good agreement with the predicted high-field binding energies of the singlet, bright triplet (and dark triplet) in a series of narrow quantum wells (QW's) for which it is applicable.[6] Meanwhile, at much lower fields Yusa *et al*. were able to observe three $X^-$ states in a single sample for the first time, which they identified as the singlet, bright triplet and dark triplet.[7] On the other hand, the calculations of Riva *et al*. showed that the bright triplet was barely bound in a 100 Å QW and totally unbound in a 300-Å QW.[8] They found good agreement between their calculated dark triplet binding energy and the experimental data of Whittaker and Shields[2] in low fields. Thus, the final identification of the triplet states observed in experiment is still an open question.

Here we report photoluminescence (PL) measurements on a 200-Å-wide GaAs QW in magnetic fields up to 50 T. In accordance with our earlier data on narrower QW's,[6] we observe recombination from the neutral and negatively charged excitons in the high-field regime and determine the singlet and (bright) triplet binding energies at fields between 15 and 45 T. Our data match up remarkably well with those of Yusa *et al.*[7] and Glasberg *et al.*,[9] who studied a QW of the same width in low fields, and show that in the high-field regime the singlet and bright triplet binding energies are essentially field invariant. We further show that a brief examination of the data in the literature reveals that the high-field $X^-$ binding energies are rather independent of the QW width and therefore that a generic form of the binding energies of the $X^-$ spin states can be drawn. Doing this clearly reveals the singlet to triplet ground state transition at $B_c \approx 20$ T.

The sample used in this study is a single asymmetrically modulation-doped $Al_xGa_{1-x}As/GaAs/Al_xGa_{1-x}As$ QW grown by molecular beam epitaxy. The sample structure is identical to those studied previously,[6] but with a well width of 200 Å. A second sample with the same structure and a well width of 300 Å was also investigated, but its PL lines were too broad to be well resolved for most of the field range. This sample will not be discussed in detail here, but the data are entirely consistent with the results and conclu-

 

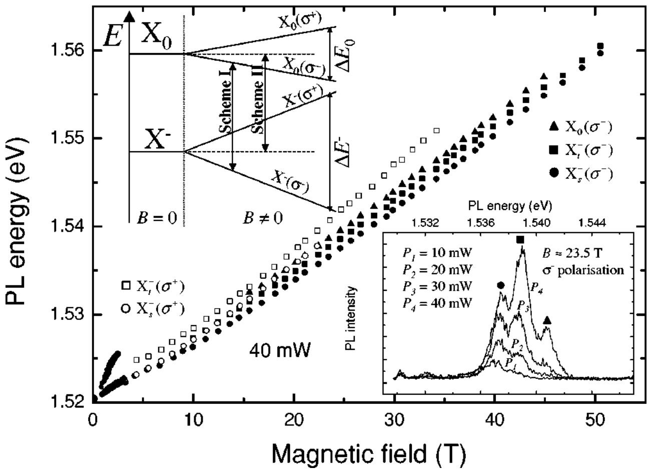


FIG. 1. Magnetic field dependance of the PL peak energies at a bath temperature of 4.2 K and a laser power of 40 mW. The lower inset shows the evolution of the recombination with incident laser power. A power of 40 mW corresponds to a power density of 1700 $\mathrm{mW\,cm^{-1}}$. The upper inset shows the two schemes for determining the charged exciton binding energy, either including (scheme I) or excluding (scheme II) the Zeeman interaction, indicated as $\Delta E_0$ and $\Delta E^-$ for $X_0$ and $X^-$, respectively. For details see Ref. 11.

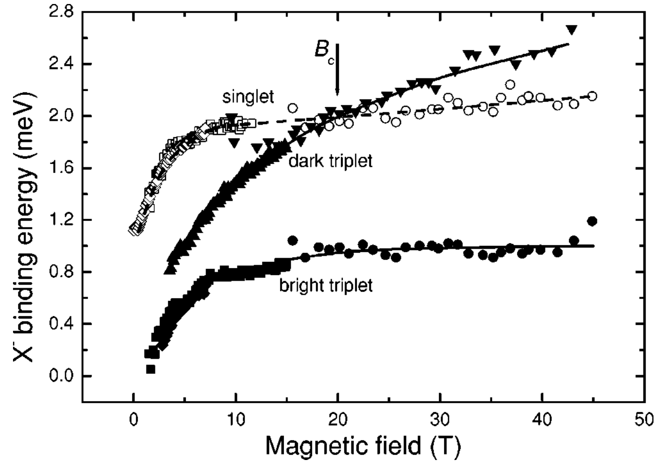


FIG. 2. Binding energy of the negatively charged exciton as a function of magnetic field. The singlet binding energy is plotted as open symbols, and the bright and dark triplet binding energies as solid symbols. The present data are plotted as circles; the squares and up triangles are from Ref. 7, the diamonds are from Ref. 9 and the down triangles from Ref. 6. The lines are a guide to the eye.

sions presented here. PL experiments were carried out at 1.2 and 4.2 K in a He bath cryostat placed in the center of a nitrogen-cooled pulsed field magnet. At 1.2 K the (high-energy) PL peaks were considerably weaker in intensity, so we present data for 4.2 K. Indeed, in order to observe recombination from the neutral exciton it was necessary to increase the incident laser power density from 400 $\mathrm{mW\,cm^{-2}}$ used previously[6] to 1700 $\mathrm{mW\,cm^{-2}}$, as shown in the lower inset of Fig. 1. The 514.5-nm line of an $Ar^+$ laser was used for this purpose. The laser light and photoluminescence were transmitted to and from the sample by a bundle of optical fibers with core diameters of 400 $\mu$m. The light was dispersed in a 0.275-m spectrometer and collected by an intensified charge-coupled-device detector. An *in situ* polarizer in combination with reversing the direction of the magnetic field allowed us to distinguish between left- ($\sigma^-$) and right- ($\sigma^+$) handed circularly polarized light. The spectral resolution was better than 0.13 meV, and the field resolution was $\pm 1\%$.

The lower inset of Fig. 1 shows typical PL spectra with $\sigma^-$ polarization for this sample for a variety of incident laser powers. At low power two peaks can barely be resolved in the spectrum, but as the laser power is increased they become clearly visible, and a third peak appears. Going from low to high energy we attribute the three peaks to recombination from the singlet state of $X^-$, the triplet state of $X^-$, and the neutral exciton. We shall show later that the triplet can be clearly identified as the bright triplet. The increase in relative intensity of the triplet PL and the appearance of the neutral exciton peak are for the most part a result of moderate sample heating, sufficient to give some thermal occupation of these higher-energy states which, as we will show, lie $\leqslant 2$ meV above the singlet. A further decrease in the electron density in the QW as a result of illumination increased above $Al_xGa_{1-x}As$ band-gap,[10] which would also enhance the relative intensity of the neutral exciton, is not excluded, but there is no reason why this mechanism should enhance the intensity of triplet PL over that of the singlet.

The main part of Fig. 1 shows the evolution of the peaks as a function of magnetic field. It is very similar to that reported previously in 120- and 150-Å QW samples,[6] already giving an indication that the properties of the charged exciton in a high magnetic field have a weak dependence on well width and therefore that a very general description of $X^-$ in high fields can be achieved. At low field ($<2.5$ T) free-carrier recombination from two Landau levels is seen, but as the field is increased it gives rise to an effective dilution of the two-dimensional electron gas, allowing the observation of neutral and charged excitons. Over the entire field range the PL is dominated by the $\sigma^-$ peaks, which arise from the lowest-energy Zeeman-split states of excitons in GaAs QW's in a magnetic field.[6] For a broad range of magnetic fields $\sigma^+$ recombination from the singlet and then the triplet state is observed, but it is always very weak and disappears in high fields as the spin polarization of the holes increases. The $\sigma^+$ recombination from the neutral exciton is not present because of the high energy of its upper spin state.

Figure 2 shows the experimental binding energies for the singlet (open circles) and triplet (solid circles) states obtained from Fig. 1 by taking the difference between the neutral exciton and singlet recombination peaks and the neutral exciton and triplet recombination peaks, respectively, for the $\sigma^-$ recombination, i.e., according to scheme I shown in the upper inset of Fig. 1.[11] The alternative method by which the binding energy is determined, scheme II, involves averaging the Zeeman contribution of the states. In the case where the neutral and charged exciton $g$ factors are the same, the two schemes give the same result. In our experiment the neutral exciton is not observed at low fields, but over the range of fields for which it is seen, 15–45 T, we find that both the singlet and triplet binding energies are essentially field independent. This is entirely consistent with our results for 120- and 150-Å QW's.[6] Also shown in Fig. 2 are binding energies determined by Glasberg *et al*.[9] (open and solid diamonds) and Yusa *et al*.[7] (squares and up triangles). It is worth noting that we have reanalyzed the data of Glasberg *et al*. and found the binding energy according to scheme I, whereas scheme II was used in their report. Doing this gives an impressive agreement with their later measurements reported in Ref. 7, which were also analyzed using scheme I.

We now go on to discuss the identification of the states, which can be achieved through the (calculated) dependence of their binding energies on the magnetic field. The identification of the singlet state is not disputed. It is the only bound state at zero field and is known, according to recent theories[2,5,8] and experiments,[2,6,7,9] to show a strong increase in binding energy from 0 to about 10 or 15 T, after which it flattens off to become almost field independent. This behavior can be clearly seen in the data of Fig. 2 and has been observed and/or calculated for well widths of 100, 115, 120, 130, 150, 200, and 300 Å in Refs. 2 and 5–9. However, as discussed in the introductory paragraph, the same consensus has not been reached for the triplet, where the remaining argument centers on which triplets (i.e., bright or dark) have been observed in which experiment.

We first turn to the dark ($L_z=-1$) triplet, whose field dependence is quite different from the singlet. It is not bound in zero field, and according to all recent theories its binding energy increases monotonically as the field increases.[2,5,8] Such a characteristic behavior has been calculated for $X^-$ in QW's with widths of 100, 115, 130, 200, and 300 Å. The bright ($L_z=0$) triplet is also unbound in zero field, but the same calculations show that in high fields it behaves like the singlet: i.e., its binding energy is field independent.[5,8] Note that up to the highest fields for which it has been investigated (65 T), the bright triplet has a binding energy which is always lower than that of the singlet. It is also worth noting that even though Riva *et al*. report the bright triplet to be only marginally bound in a 100-Å QW and unbound in a 300-Å QW, they also find the binding energy to be field independent in high field. Thus, the way to distinguish between the bright and dark triplets is their field dependence above about 15 T. Moreover, since both bright and dark triplets are unbound in zero field and their binding energies both increase with field at low fields, it is easy to confuse the two in the low-field regime.

With this in mind we can immediately say that the triplet observed in our experiment (solid circles in Fig. 2) is the bright triplet, that the same state was observed in the experiments of Glasberg *et al*. and Yusa *et al*., and that it was correctly identified by Yusa *et al*. as such. The other state from Ref. 7, shown as up triangles in Fig. 2, is the dark triplet. We do not observe the dark triplet, as would be expected, and we note that it was only detected by Yusa *et al*. as a weak line at very low temperatures. *Indeed, we believe that the only other observation of the dark triplet was in our experiments on a* 100-Å *QW reported in Ref. 6*. In this case the dark triplet is most likely rendered visible by quantum well width fluctuations.[3] In all other cases, at least where high-field data are taken, the triplet binding energy is found to saturate at high fields and so is the bright triplet. This has been experimentally observed for QW widths of 120, 150, 200, and 300 Å.[2,6,7,9]

Having established the high-field character of the $X^-$ spin-state binding energies, we now go on to discuss their behavior in more general terms. At zero field only the *singlet state* is bound, and its binding energy increases rapidly with field up to 10 or 15 T, where it saturates at about 2 meV. As can be seen from Fig. 2 and Refs. 2 and 6, this behavior is rather independent of the QW width. The *bright triplet*, which is unbound at zero field, has the same qualitative behavior, but saturates at about 1 meV, again for a wide range of well widths.[2,6,7,9] The *dark triplet* is also unbound in zero field, but its binding energy increases monotonically with magnetic field, such that it eventually becomes the ground state. Indeed, working on the basis that the high-field binding energies are rather independent of QW width we also plot the dark triplet binding energy for a 100-Å QW in Fig. 2 (down triangles).[6,12] It can be seen that despite the large difference in QW width the data match up very well with those of Yusa *et al.* and reveal the predicted singlet–to–dark-triplet ground-state transition $B_c$ at about 20 T. Overall, it can be said that the combined data of Fig. 2 represent the generic form of the binding energies of the spin states of $X^-$ in a magnetic field.

To summarize, we report photoluminescence measurements on the negatively charged exciton in a 200-Å QW in magnetic fields up to 50 T. We determine the binding energy of the singlet and bright triplet states in the high-field regime and use the characteristic field dependence of the singlet, bright triplet, and dark triplet states to distinguish between bright and dark triplets, showing that high-magnetic-field PL data are needed to make such a distinction. We find that our data and the body of experimental results in the literature are very consistent, and conclude that a generic picture of the high-field binding energy of the charged exciton has been obtained, which is rather independent of the QW width. We assert that, except for two particular instances, the triplet observed in experiment is the bright triplet and show that the singlet–to–dark-triplet ground-state transition occurs at $B_c \approx 20$ T.

*Note added in proof.* Schüller *et al.*[13] recently also observed the dark triplet in photoluminescence at low temperatures, but it was found to be absent in the absorption spectrum, thereby demonstrating its dark character.

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*Electronic address: manus.hayne@fys.kuleuven.ac.be

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