Excitonic molecules in InGaAs/GaAs quantum dots

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Optical studies of semiconductor quantum dots (QDs) open new possibilities for investigating many-particle atoms consisting of electrons (e) and holes (h) or of multiexcitons. In bulk and in quantum wells (QW) the two excitons in a biexciton are spatially localized by their effective Coulomb interaction alone. Molecules of three or more excitons in semiconductors with simple conduction and valence band structures are not stable because of the strong Pauli repulsion between electrons (holes) with the same spin. In QDs the geometric confining potential localizes excitons in the same spatial region. In addition, it provides a discrete level structure. The e–h interaction results in a renormalization of the transition energies and of the transition matrix elements of the confined multiexcitons. The effect of the spatial confinement on the biexciton binding energy has recently been investigated both experimentally and theoretically in QDs [1–5].

In the present paper we discuss the influence of an external magnetic field on multiexciton states in InGaAs/GaAs QDs. In the two-dimensional (2D) case a magnetic field normal to the QW plane transforms the continuous energy spectrum of electrons and holes into a discrete one. As a consequence, the optical spectra demonstrate a well pronounced Landau level structure. In contrast, in QDs with their discrete energy level structure (caused by the geometric confining potential) the magnetic field lifts the degeneracies of levels and, to some extent, leads the system back to a 'quasicontinuous' energy spectrum [6–8]. In small QDs with diameters comparable with a_X, the exciton spectrum has been found to retain the features of the electron spectrum but to be influenced by the e-h interaction [9, 10]. Recent high excitation studies of the QD photoluminescence (PL) have revealed that the effect of many-particle interactions in small QDs is also relatively small [11, 12]. These studies were carried out on arrays containing a large number of QDs with relatively large inhomogeneous PL line broadening by dot size fluctuations. In the present paper we discuss the PL spectra of single InGaAs/GaAs QDs when inhomogeneous broadening effects are strongly suppressed [5, 13] that allows us to extract information on e-h interaction effects. In addition, we discuss the PL studies of QD arrays fabricated from shallow InGaAs/GaAs QW structures which also demonstrate a relatively small inhomogeneous broadening of the QD emission line. However the cost for that is the weak geometric confinement which does not allow us to realize states with more than two excitons in the QD. Therefore shallow QD arrays were used to study spin-induced effects in two exciton states in a magnetic field.

The InGaAs/GaAs single QW dots were fabricated from a 5-nm thick $In_{0.13}Ga_{0.87}As/GaAs$ QW using low voltage electron beam lithography [14]. This technique allows us to avoid proximity effects and to prepare quantum dots with a well defined size. For the optical investigations a spacing of 50 μm between adjacent single dots was chosen. This permits the investigation of individual QDs in an optical cryostat with a superconducting solenoid. The QDs were excited by an Ar^+-ion laser. The excitation power was varied over the range 0.1 to 200 μW and the laser spot was focused down to a diameter of 20 μm. The single dot emission was dispersed by a monochromator and detected by a liquid nitrogen cooled CCD camera. The luminescence was integrated over times ranging from 10 s at high excitation densities to 2 min at low excitation densities. The arrays of shallow dots were fabricated from a 5-nm thick $In_{0.03}Ga_{0.97}As/GaAs$ QW. The technique was similar to that for the single QDs, but the distances between the dots in an array were within 200–500 nm.

For our studies we chose QDs with lateral sizes $L_{x,y}$ of 50–80 nm which slightly exceed the exciton radius, a_X. In this case, on the one hand, there is a balance between the Coulomb interaction and the QD confining potentials and, on the other, the magnetic length l_B is comparable both with $L_{x,y}$ and a_X already at $B = 3–6$ T. Hence, the magnetic field strongly influences the QD multiexciton states.

Figures 1 and 2 display PL spectra from single $5 \times 50 \times 70$ and $5 \times 45 \times 55$ nm^3 boxes at $B = 0$. At low excitation the emission line labeled by X corresponds to the recombination of a single e–h pair (exciton) in the QD. Its full width at half maximum is about 2 meV which is larger than the width in natural QDs [13]. This may be due to time fluctuations of the QD potential related to photoexcited surface charges.

With increasing excitation density a well pronounced shoulder X_2 appears at the low energy side of the exciton line which has to be assigned to biexciton emission (recombination of one of two excitons confined in the dot, see Fig. 1). In the smaller QD (see Fig. 2) the X_2 line is well resolved and is located more than 2 meV below the exciton emission line. This low energy shift arises from the effective

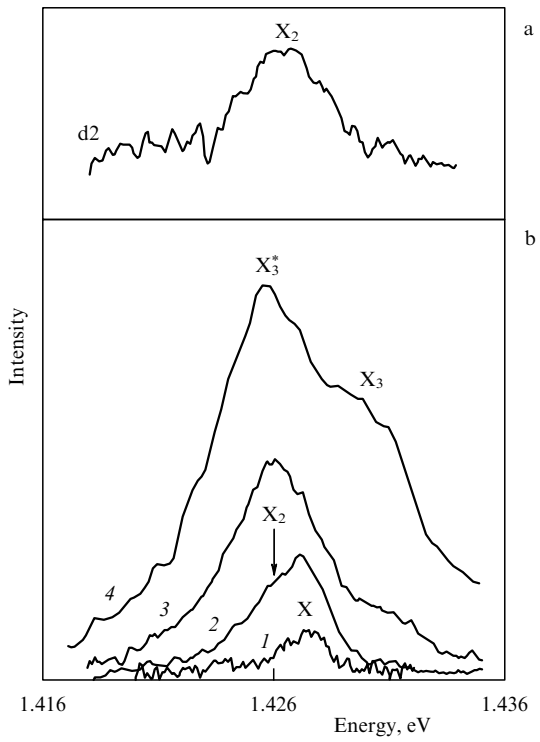

Figure 1. (a) PL spectra from a single $5 \times 50 \times 70$ nm^3 QD at $B = 0$ for different excitation powers P [μW] = 10 (*1*), 25 (*2*), 50 (*3*), and 200 (*4*); (b) the differential spectrum d2 was recorded at $P = 50$ μW. The diameter of the excited spot was 20 μ.

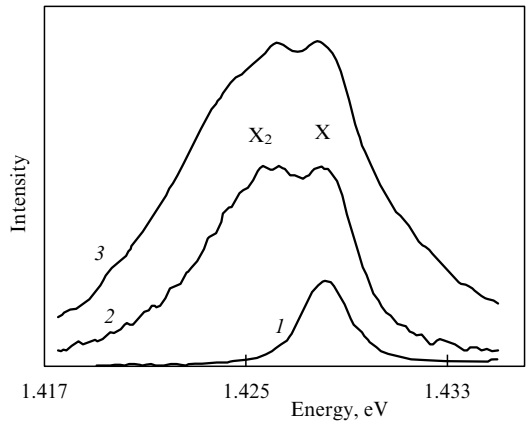

Figure 2. PL spectra from a single $5 \times 45 \times 55$ nm^3 QD at $B = 0$ for different excitation powers P [μW] = 100 (*1*), 400 (*2*) and 1000 (*3*).

exciton – exciton Coulomb interaction in the dot and therefore it can be related to the QD 'biexciton' binding energy Δ_{XX}. In the larger dot (Fig. 1b) the X_2 line is located closer to the X line and is only weakly resolved in the spectrum. However, it is well pronounced in the differential spectrum shown in Fig. 1a which is the difference of the spectra recorded at $P = 25$ μW and at $P = 10$ μW. It shows that the line X_2 is shifted by about 1.2 meV to lower energies compared to the exciton emission. Figure 3 displays the dependence of Δ_{XX} on the lateral QD size for cylindrical dots over a wide range of $2R = 50-200$ nm. Δ_{XX} increases monotonically with reducing QD size, in agreement with theoretical predictions for dots with $R > a_X$ [2, 3].

The comparison of Figs 1 and 2 shows that in order to observe two exciton complex emission from the smaller QD we have to use significantly higher excitation. This difference is due to the larger surface recombination in the smaller QD. High excitation results in a marked heating of the QD and does not allow us to fill it with many excitons even at the highest excitation power of 1 mW (see Fig. 2, trace *3*). In contrast, Fig. 1 shows that in case of the larger QD well pronounced shoulder labeled by X_3 appears at the high energy side of spectrum at $P = 50-100$ μW whereas the sharp single exciton line disappears from the spectrum. In addition, the low energy peak shifts markedly to the low energies. To explain these changes we should suppose that the QD is filled with three e – h pairs. Two of them completely fill the doubly spin degenerate ground states of the electron and hole in the dot and the third e – h pair occupies their first excited states and therefore results in a new emission line X_3 at a higher energy. Note, that this line cannot be assigned to the excited exciton state connected with quantization of exciton motion in the QD. Indeed, the calculated energy of such state in the 50×70 nm^2 QD is 0.8 meV whereas the experimental value exceeds 2.5 meV and corresponds to the sum of electron and hole quantization energies.

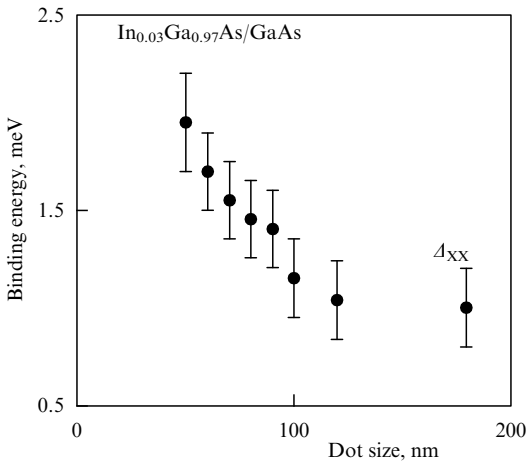

Figure 3. Dependence of the QD biexciton binding energy on the QD size.

Thus the three exciton states in the QD with $R = (3-6)a_X$ should be described within the framework of a shell model which uses an electron and hole rather than exciton shells to classify multiexciton states. A similar model was used in bulk semiconductors to describe bound multiple exciton complexes [15]. The Pauli exclusion principle leads to the scheme of optical transitions shown in Fig. 4. The ground states with one (X) and two (X_2) excitons consist of electrons and holes in the ground e and h shells, whereas the three exciton state (X_3) includes two electrons and two holes in the ground shells and one electron and one hole in the excited e* and h* shells, respectively. Figure 4 shows that, in agreement with the experiment, the shell model predicts one emission line, X_2, for the two exciton state and two lines, X_3 and X_3^*, for the three exciton complex. The final (two exciton) state of the transition X_3^* is excited; it consists of electrons and holes from the ground (e, h) and excited (e*, h*) shells.

The shell model predicts that an external magnetic field splitting electron and hole states should result in a rearrangement of the multiexciton states. Figures 5a and 5b display the depolarized QD PL spectra recorded under the same excitation conditions as those in Figs 2 and 1b, respectively, but at $B = 6$ T. The spectral features in a magnetic field are strongly different. Figure 5a shows that in the small QD the X_2 line appears only as a weakly resolved shoulder indicating a strong decrease of Δ_{XX}. In the larger, $5 \times 50 \times 70$ nm^3 dot the lines X and X_2 are not even resolvable. The first pronounced shoulder appears on the high energy side of the X-line at $P = 50$ μW, as in the case of zero magnetic field. Therefore we can connect this shoulder with the appearance of the third exciton in the QD. At smaller excitation powers we observe only the broadening of the X line.

A comparison of the high excitation spectra in Figs 1b and 5b shows that the splitting of the emission lines X_3^* and X_3 corresponding to the recombination of three exciton complexes decreases strongly in a magnetic field. As can be seen from the transition scheme in Fig. 4 the splitting of these lines

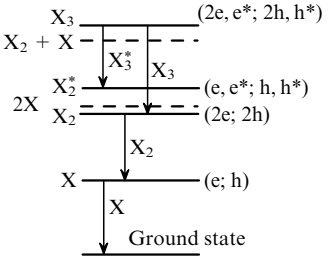

Figure 4. Scheme of the optical transitions of QD multiexcitons. The dashed lines 2X and $X_2 + X$ indicate the sum of the energies of two excitons and of a biexciton and exciton, respectively.

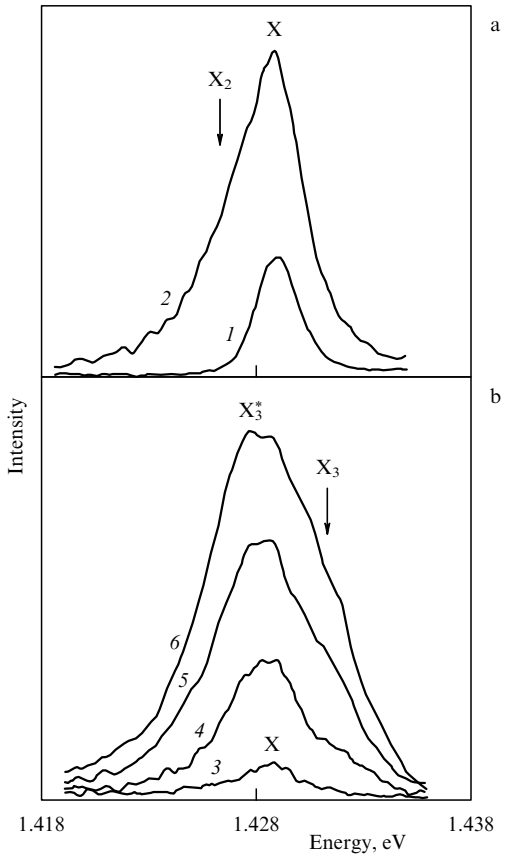

Figure 5. PL spectra from single $5 \times 45 \times 55$ (a) and $5 \times 50 \times 70$ nm^3 (b) QDs at $B = 6$ T for different excitation powers P [μW] = 100 (*1*), 400 (*2*), 10 (*3*), 25 (*4*), 50 (*5*), and 200 (*6*).

reflects the energy of the first excited two exciton state δ_1. The decrease of δ_1 means that the magnetic field suppresses the repulsion of excitons with one e–h pair in the ground and one e–h pair in the excited shells. This behavior correlates with the magnetic field induced lowering of the energy of the first excited electron state in the QD [6, 8] and is in accordance with results of previous studies of the QD PL on dot arrays [11].

The magnetic field dependence of the binding (repulsion) energies for a few two and three exciton states in the $5 \times 50 \times 70$ nm^3 QD is shown in Fig. 6. The three exciton state energy in the small dots is larger than the triple single exciton energy and, hence, such a state is confined only due to the QD confining potential. This is obviously associated with the strong Pauli repulsion of electrons (holes) with identical spins which appears with the third exciton. This repulsion pushes the third exciton into the next shell. Figure 6 shows that the repulsion energy of a third exciton to the two exciton complex $\Delta_{XXX} = \hbar\omega(X_3) - \hbar\omega(X)$ decreases with magnetic field. This occurs due to localization of electrons and holes within the magnetic length. As a consequence the Pauli repulsion is reduced when l_B becomes comparable with $L_{x,y}$. Figure 6 shows that Δ_{XXX} changes from -4 meV at $B = 0$ to -3 meV at 6 T. It is obvious that Δ_{XXX} should tend to zero for L_x and/or $L_y \gg l_B$, i.e. when the excitons in the QD are separated by several l_B. Indeed, we have found that at $B = 8$ T ($l_B = 9$ nm) the lateral quantization effects disappear in the dots with $L_{x,y} = 150-200$ nm.

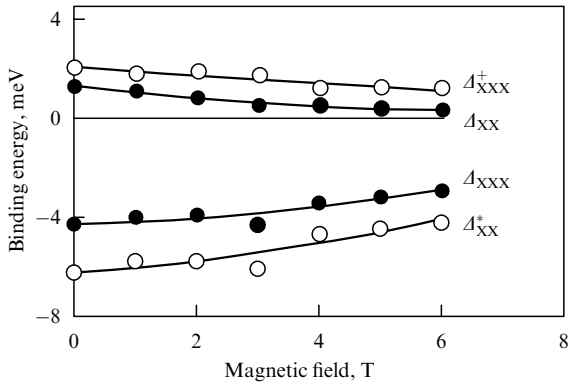

Figure 6. Magnetic field dependence of binding (repulsion) energies for two and three exciton complexes in the $5 \times 50 \times 70$ nm^3 QD.

Similar arguments also explain the behavior of the excited two exciton state in the QD. Figure 3b shows that the net coupling energy for this state $\Delta^*_{XX} = \Delta_{XX} + \hbar\omega(X_3) - \hbar\omega(X_3^*)$ changes from -5 meV at $B = 0$ to -3.5 meV at 6 T. This change is mainly determined by the splitting of the single particle states which decreases with magnetic field.

In the case of a third exciton added to the excited two exciton complex its binding energy can be determined as $\Delta^+_{XXX} = \hbar\omega(X_3^*) - \hbar\omega(X)$. Figure 6 shows that the effective interaction in this case is attractive and Δ^+_{XXX} even exceeds Δ_{XX}. This is natural because the third e–h pair fills the empty places in the inner shells occupied with one particle. The Pauli repulsion is absent whereas the correlation energy increases with the number of particles in the QD. As expected, Fig. 6 shows that a magnetic field reduces Δ_{XX} and Δ^+_{XXX} in a similar way.

The decrease of the binding energy of the two exciton complex $\Delta_{XX} = \hbar\omega(X) - \hbar\omega(X_2)$ with magnetic field observed in Fig. 6 can occur both due to Zeeman splitting of electron and hole states and due to magnetic field induced rearrangement of their wave functions. To separate these two contributions we recorded polarized emission spectra. To avoid the problem with the signal intensity, samples with QD arrays fabricated from shallow $In_{0.03}Ga_{0.97}As/GaAs$ QW structures with a relatively small inhomogeneous broadening of the QD emission line were used. Figure 7 displays σ^+ and

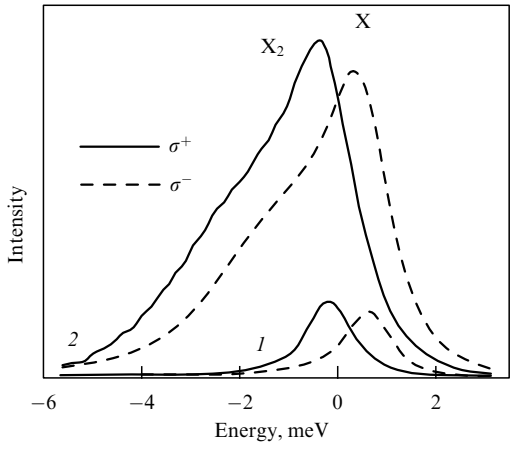

Figure 7. Polarized PL spectra from an array of QDs with $R = 80$ nm at $B = 8$ T at low (*1*) and high (*2*) excitation densities.

σ^- emission spectra from an array of QDs with $R = 80$ nm at $B = 8$ T. At small P the both σ^+- and σ^- spectra display the single exciton line (X^+ or X^-) in accordance with the allowed transition scheme in Fig. 8. The splitting of the X^+ and X^- lines, Δ_s, connected to the Zeeman splitting of electron and hole states, reaches 0.7 meV at $B = 8$ T. A larger intensity of the σ^+ component reflects a higher filling of the ground exciton state. At high P the new lines appear at the low energy side of the X line. They are labeled X_2^+ and X_2^- in the σ^+- and σ^- polarization, respectively, and correspond to the emission of two exciton states. The energy gap between these lines is equal to that between lines X^+ and X^-. This is in agreement with the transition scheme in Fig. 8 because the splitting of the X_2 line is completely determined by the splitting of the final exciton state.

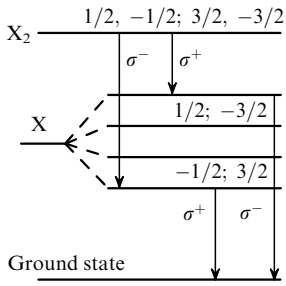

Figure 8. Scheme of the optical transitions for QD excitons and biexcitons with and without magnetic field.

As follows from Fig. 8 Δ_{XX} should be defined as the energy gap between the lines X_2^- and X^+ whereas the gap between the exciton and two exciton complex emission lines in the same polarization corresponds to the binding energy of two excitons with opposite spins of both the electrons and the holes, Δ_{XX}^0. Figure 9 shows that the magnetic field decreases Δ_{XX} strongly whereas the change of Δ_{XX}^0 is within experimental error. This means that the decrease of Δ_{XX} is mainly due to the Zeeman splitting of electron and hole states, whereas magnetic field induced rearrangement of electron and hole wave functions has a very small effect.

To conclude, the experimental studies of multiexciton complexes confined in QDs have shown that an exciton-exciton interaction in the two exciton complex enhances its confinement and that this effect increases with decreased dot size. The magnetic field normal to the QW plane has small influence on the binding energy of two excitons with opposite spins of two electrons and holes. The three exciton complex is confined only by the QD geometric confining potential. The magnetic field strongly reduces the exciton – exciton repulsion in such a complex when the magnetic length becomes smaller than $L_{x,y}$.

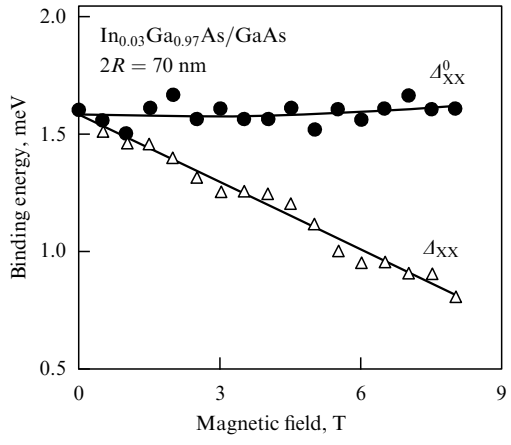

Figure 9. Magnetic field dependence of binding energy for two excitons with similar (Δ_{XX}) and opposite (Δ_{XX}^0) spins of both electrons and holes.

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Anomalous transport and luminescence of indirect excitons in coupled quantum wells

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The electron – hole (e – h) interaction in a neutral e – h system can result in the condensation of bound e – h pairs (excitons) [1]. In case of a dilute exciton gas ($na_B^d \ll 1$, where a_B is the exciton Bohr radius, n is the e – h density, and d is the dimensionality) the excitons can be considered as point Bose particles and the condensation is analogous to the Bose – Einstein condensation of bosons, while for a dense e – h system ($na_B^d \gg 1$) the condensed state is analogous to the BCS superconductor state.

The condensation conditions can be achieved only if the temperature of the excitons is below a critical temperature, T_c. This criterion is hard to achieve experimentally. Due to e – h recombination the excitons in semiconductors have a finite lifetime which results in an increase of the exciton temperature. Therefore, to search for exciton condensation, semiconductors with a long exciton lifetime are selected. Degenerate Bose – Einstein statistics have been observed for excitons in Cu_2O [2] and in Ge [3]. Recently the passing of the exciton condensate phase boundary for excitons in Cu_2O was reported [4].

The semiconductor quantum well (QW) structures provide an opportunity for the experimental realization of a quasi-two-dimensional exciton condensate. It has been