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QUANTUM DOT MULTIEXCITONS IN MAGNETIC FIELD

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Multiexcitons confined in InGaAs/GaAs quantum dots (QD's) with a lateral size slightly exceeding the exciton Bohr radius have been investigated by magnetophotoluminescence spectroscopy at 2 K. The Coulomb correlations in the two-exciton-complex result in an additional confinement, which increases with decreasing dot size, the magnetic field reduces this effect. A three exciton complex is confined only due to the geometric confinement potential of the QD. The exciton-exciton repulsion increases with decreasing dot size, the magnetic field reduces strongly the repulsion when the magnetic length becomes smaller than the lateral QD size. A shell model for the quantum dot multiexciton states has been suggested.

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Optical studies of semiconductor quantum dots (QD's) open new possibilities for investigating many-particle atoms consisting of electrons (e) and holes (h) or of multiexcitons. In bulk and in quantum wells 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 QD's the geometric confining potential localizes excitons in the same spatial region. The $e-h$ interaction results in a renormalization of the transition energies and of the transition matrix elements of the confined multiexcitons. In particular, it has been shown that the spatial confinement increases the biexciton binding energy in QD's [1-5].

In the present paper we investigate the influence of an external magnetic field on multiexciton states in single, free standing InGaAs/GaAs QD's. In contrast to the 2D case, in QD's with their discrete energy level structure (caused by the geometric confining potential) the magnetic field lifts degeneracies of levels and, to some extent, leads the system back to a "quasicontinuous" energy spectrum [6-10]. Recent high excitation studies of the QD magneto photoluminescence (PL) carried out on arrays containing a large number of QD's have revealed that the effect of many-particle interactions is relatively small [11, 12]. The interaction energy was found to be within the inhomogeneous broadening of the luminescence by dot size fluctuations. We investigate PL spectra of single QD's which allow us to obtain excitonic emission lines with narrow linewidths [5, 13] and to extract information on $e-h$ interaction effects.

The InGaAs/GaAs isolated QD's dots have been fabricated from a 5-nm thick quantum well by using low voltage electron beam lithography [14]. For the optical investigations a spacing of 50 μm between adjacent single dots was chosen. This permits the investigation of individual QD's in an optical cryostat with a superconducting solenoid. The QD's were excited by an Ar^+-ion laser. The excitation power was varied in the range between 0.1 and 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 seconds at high excitation densities to 2 minutes at low excitation densities.

For our studies we have chosen QD's with lateral sizes $L_{x,y}$ of 50–80 nm which exceed slightly the exciton radius a_x. In this case, on one hand, there is a balance between the Coulomb interaction and the QD confining potentials. On the other hand, at relatively small magnetic fields of 3–6 T the magnetic length l_B is comparable both with $L_{x,y}$ and a_x and influences strongly the multiexciton states in the QD. We compare the behavior of excitons in such small dots with that in a relatively large dot where the localization of carriers in the exciton due to the Coulomb interaction dominates.

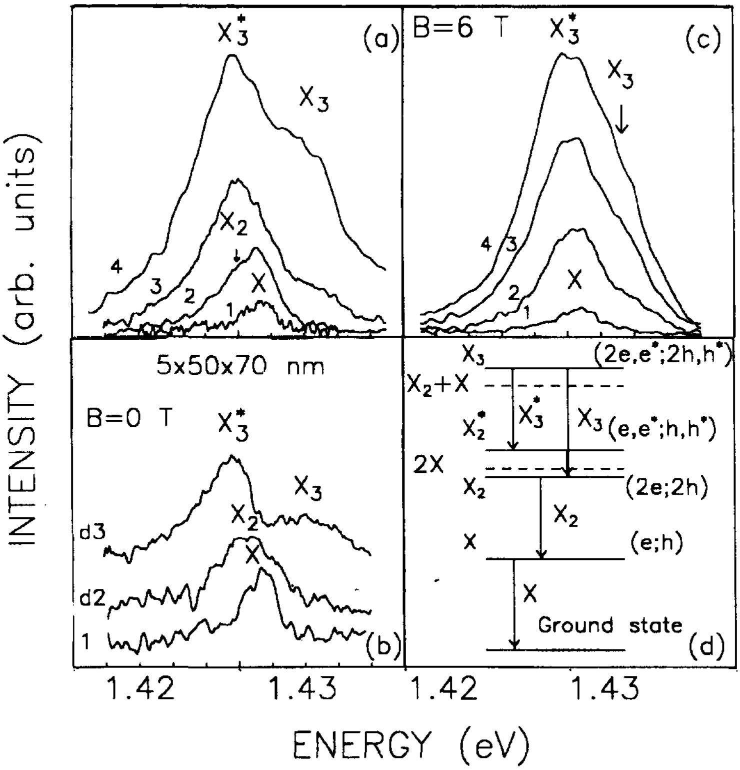

Fig.1. PL spectra from single $5 \times 50 \times 70$ nm^3 QD at $B = 0$ (a,b) and 6 T (c) for different excitation powers. P [μW] = 10 (*1*), 25 (*2*), 50 (*3*), and 200 (*4*); the differential spectra *d*1 and *d*2 are obtained at 25 μW and 50 μW, respectively. The diameter of the excited spot is 20 μ. Scheme of the optical transitions of QD multiexcitons is shown in Fig.1*d*. The dashed lines 2X and X_2+X indicate the sum of energies of two excitons and of biexciton and exciton, respectively

Figs.1*a* and 2*a* display PL spectra from single $5 \times 45 \times 55$ and $5 \times 50 \times 70$ nm boxes at $B = 0$ for different excitation densities. At low excitation the 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. This width is larger than the width in natural QD's [13]. This may be due to elastic scattering processes or due to temporal fluctuations of the QD potential which are related to photoexcited surface charges.

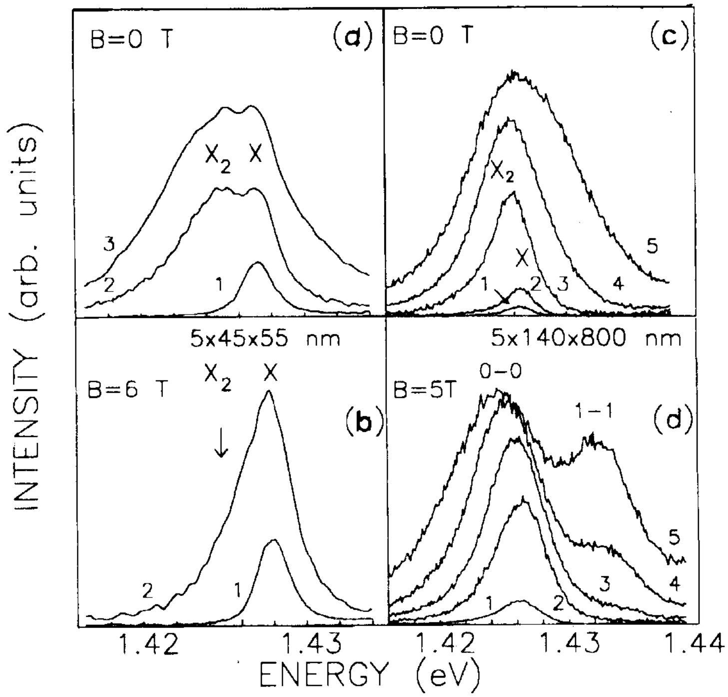

Fig.2. PL spectra from single $5 \times 45 \times 55$ (*a*, *b*) and $5 \times 140 \times 800$ (*c*, *d*) nm^3 QDs for different excitation powers and magnetic fields. In Figs. *a* and *b* P (μW)= 100 (*1*), 400 (*2*) and 1000 (*2*). In Figs.*c* and *d*, P (μW)= 10 (*1*), 20 (*2*), 100 (*3*), 200 (*4*), 400 (*5*) and P (μW)= 20 (*1*), 100 (*2*) 200 (*3*), 400 (*4*), 600 (*5*), respectively

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 emission of a two exciton state (recombination of one of two excitons confined in the dot, Fig.1*d*). In the smaller QD (Fig.2*a*) the X_2 line is well resolved and is located more than 2 meV below the exciton line. This shift arises from the effective exciton-exciton Coulomb interaction in the dot and therefore it can be related to the binding energy of the two exciton state, Δ_{xx}. In the larger dot (Fig.1*a*) the X_2 line is located closer to the X line and is only weakly resolved in the spectrum. To resolve it we have recorded a differential spectrum, *d*2 (Fig.1*b*) which is the difference of the spectra recorded at $P = 25$ μW and 10 μW. It shows that in the $5 \times 50 \times 70$ nm^3 dot Δ_{xx} = 1.2 meV. The decrease of Δ_{xx} is in agreement with theoretical predictions [2, 3].

The comparison of Figs.1*a* and 2*a* shows that the two exciton state emission from the smaller QD appears at a much (nearly 5 times) higher optical excitation. This difference is most likely due to larger surface recombination in the small QD which does not allow us to fill the dot with many excitons even at the excitation power of 1 mW (Fig.2*a*, trace *3*). In contrast, Fig.1*a* shows that in case of the larger QD an increase of the excitation power to 50-100 μW results in a further increase of the number of excitons. The Pauli exclusion principle forbids the relaxation of additional electrons and holes into the ground shells because the latter is only double spin-degenerate. Therefore the third exciton should occupy an excited state as shown in Fig.1*d*. As a result, two lines, X_3 and X_3^*, are expected in the emission spectrum of X_3, which correspond to the transitions into the ground or into the excited two exciton state. Figure 1*a* shows that only the line X_3 appears in the spectrum as a well pronounced shoulder at the high energy side of the X line. The line X_3^* is not resolved at the low energy tail of the line X_2 but it is well pronounced in the differential spectrum shown in Fig.1*b*, trace *d*3. Thus two peaks located 2 meV lower and 3 meV higher than the X transition can be assigned to the recombination of excitons in the inner and the outer shells, respectively. The intensity of the X_3^* is about two times larger than X_3, in accordance with ratio of the numbers of electrons in the inner and outer shells. The differential spectrum recorded at 50 μW is very similar to the QD emission spectrum at 200 μW when the mean number of excitons in the QD exceeds 2.

The magnetic field changes the PL spectra. Figs.1*c* and 2*b* display QD spectra recorded under the same excitation conditions as those in Figs.1*a* and 2*a*, respectively, but at $B = 6$ T. In the small QD (Fig.2*b*) the two exciton state emission appears only as a weakly resolved shoulder indicating a strong decrease of Δ_{xx}. In the larger dot the lines X and X_2 are even not resolvable (Fig.1*c*). The first pronounced high energy shoulder appears at 50 μW, similarly to the case of $B = 0$. Therefore we can conclude that this shoulder is connected with the appearance of the third exciton in the QD. The effect of exciton-exciton interaction in the two exciton complex is observed only in differential spectra recorded at $20-40\,\mu$W which show a relatively small ($\sim$ 0.6 meV) red shift which could be connected to Δ_{xx}.

A comparison of the high excitation spectra in Figs.1*a* and 1*c* shows that the splitting of the emission lines X_3^* and X_3 corresponding to the recombination of three exciton complexes is also markedly smaller at $B = 6$ T than at $B = 0$ T. As can be seen from the transition scheme in Fig.1*d* the splitting of these lines 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 similar electron (hole) spins. This behavior is in accordance with results of previous studies of the QD PL on dot arrays [11].

The limitations originated from the Pauli repulsion should decrease with increasing ratio $L_{x,y}/a_x$. Indeed, Fig.2 illustrates that the PL behavior in a large dot is very similar to that in a quantum well. First, exciton and then biexciton emission appears in the spectrum with increasing excitation (Fig.2*c*). However, no pronounced structure appears at the high energy side of the X line with further increase of excitation density. Instead, the line transforms into the structureless broad emission characteristic of a Fermi system ($e-h$ plasma). Second, in contrast to the case of small dots the new line in the magnetoluminescence spectrum appears at much higher energy which is close to the cyclotron energy and, hence, has to be assigned to the emission of electrons and holes in the next Landau level rather than to an exciton-exciton interaction.

The magnetic field dependences of the transition energies in the $5 \times 50 \times 70$ nm QD are shown in Fig.3*c*. The exciton energies were determined from the PL spectra at the lowest (10 μW) excitation power whereas the transition energies of the two and three exciton complexes were obtained from the differential spectra recorded at $P = 25$ and $P = 100$ μW, respectively. Using these data and the scheme of multiexciton transitions in Fig.1*d*, we have determined the magnetic field dependence of the exciton-exciton interaction energies in the ground state, $\Delta_{xx} = \hbar\omega(X) - \hbar\omega(X_2)$, and in the excited two exciton state, $\Delta^*_{xx} = \Delta_{xx} + \hbar\omega(X_3) - \hbar\omega(X^*_3)$, and also the interaction energy of a third exciton in the three exciton state, $\Delta_{xxx} = \hbar\omega(X_3) - \hbar\omega(X)$. These energies are displayed in Fig.3*b*. Δ_{xx} is positive whereas Δ_{xxx} is negative. This means that the effective exciton-exciton interaction in the QD is attractive only for biexcitons. Δ_{xx} increases with reduction of the lateral size of the QD, from 0.8 meV to 2.2 meV for the smallest, 45×55qnm dot. Magnetic field decreases Δ_{xx}. The decrease depends weakly on the QD size and reaches 0.7–0.8 meV at 6 T. This effect originates mainly from Zeeman splitting of excitons in the magnetic field because the two excitons forms the singlet state and hence the second exciton in the magnetic field must fill the excited spin state.

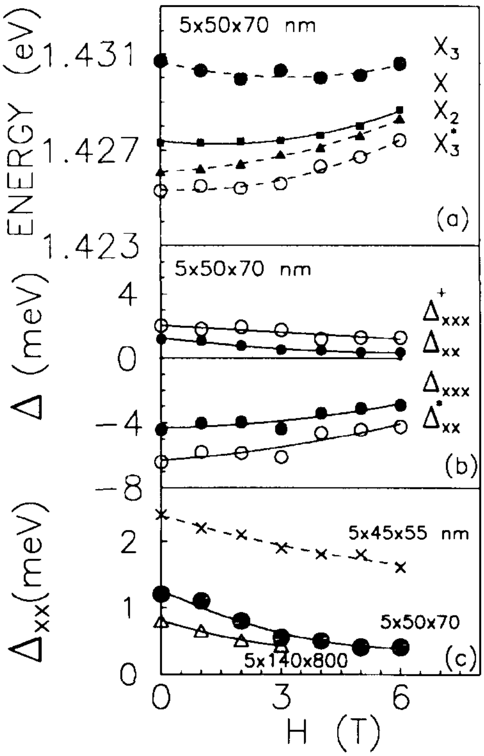

Fig.3. Magnetic field dependence (*a*) of the exciton and multiexciton transition energies and (*b*, *c*) of the multiexciton binding energies in the QDs

The three exciton state energy 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 and pushes the third exciton into the next shell. An increased magnetic field leads to a localization of particles within $2l_B$ and, hence, the reduction of the Pauli repulsion, in agreement with the experimental data in Fig.3*b*. Δ_{xxx} changes from -4meV at B = 0 T to -3meV at 6 T. It is obvious that Δ_{xxx} should tend to zero for L_x and/or $L_y >> l_B$, i.e. when the excitons in the QD are separated by several l_B. This case is realized in the large, $140 \times 800\,\mathrm{nm}^2$ dot. Similar arguments explain also the behavior of the excited two exciton state in the QD. Figure 3*b* shows that Δ^*_{xx} changes from -5meV at $B=0$ T to -3.5meV at 6 T.

Finally, we can consider the case when a third exciton in the ground state is added to the excited two exciton state. Its binding energy can be determined by $\Delta^+_{xxx} = \hbar\omega(X^*_3) - \hbar\omega(X)$. Fig.3*b* shows that the effective interaction in this case is attractive and Δ^+_{xxx} even exceeds Δ_{xx}. This is natural because the third exciton fills the empty place in the inner shell occupied with one particle. The Pauli repulsion in this case has no effect whereas the correlation energy increases with the number of particles in the QD. As expected, Fig.3*b* shows that a magnetic field reduces Δ_{xx} and Δ^+_{xxx} in a similar way.

To conclude, we have investigated experimentally multiexciton complexes consisting of two and three excitons which are confined in a single QD with lateral dimensions slightly exceeding the exciton Bohr radius. The exciton-exciton interaction in two exciton complex enhances the confinement, it is rather strong at zero magnetic field but decreases with increasing magnetic field. The three exciton complex is confined only due to the QD geometric confining potential. The magnetic field reduces strongly the exciton-exciton repulsion in such a complex.

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