Excitonic binding in coupled quantum wells

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We study excitonic states in the presence of applied electric field in 8-nm GaAs coupled quantum wells (QW's) separated by a 4-nm $Al_{0.33}Ga_{0.67}As$ barrier and in 6-nm $In_{0.1}Ga_{0.9}As$ coupled QW's separated by a 4-nm GaAs barrier in which effects attributed to macroscopically ordered excitonic states have been recently reported. We discuss the differences in the nature of the states and in the origin of confinement which determines the change of excitonic properties with increase in the applied electric field in both structures. We have found that the indirect exciton binding energy for the field amplitude used in the experiment with InGaAs QW's is around 3.5 meV, much less than the previously reported 10 meV value. This suggests that the optically induced ring structure, reported to persist to near 100 K, might not be caused by collective excitonic transport.

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A number of proposed experimental systems to achieve Bose condensation of excitons rely on a long lifetime for recombination. These includes Cu_2O, CuCl and dipolar excitons in coupled quantum wells. A good review of this subject can be found in Refs. 1–3. Recently, systems which attracted the most interest due to the observation of some novel effects are dipolar excitons in coupled quantum wells. A coupled quantum well system consists of two different, adjacent planes with electrons in one layer and holes in the other. An applied electric field keeps both kinds of carriers in separate two-dimensional planes. This arrangement reduces the overlap of the wave functions of the electron and hole and thus increases the lifetime of excitons. While the first set of experiments claiming to see Bose statistics in these systems[4] encountered difficulties with interface roughness which trapped excitons in local energy minima, the more recent work by Butov *et al.*[5–7] concludes that there is evidence of stimulated scattering and therefore densities reaching the quantum regime, but not yet evidence for Bose-Einstein condensation (BEC).

In a last few months new effects which could be attributed to the macroscopically ordered excitonic state have been reported independently by three groups.[8–11] Observation of two bright rings in the spatially resolved photoluminescence separated by a dark region was explained by the long-range transport of superfluid dark excitons. In the experiments of Refs. 8 and 9 the ring structure appears only at very low temperatures up to around 5 K and the external ring is fragmented into circular droplets that form a periodic array while in experiment of Ref. 10 the rings are observed up to 118 K and even at zero current (due to doping even at zero current there is a small built-in field),[12] and the fragmentation into circular structures is not recorded. The estimate of exciton binding energy of 10 meV given in Ref. 10 would be consistent with 100 K threshold for the effect but it was recently withdrawn and a new estimate of 2 meV given.[12]

The aim of this paper is to make quantitative calculations for both ground and excited excitonic states of the exact physical systems under experiment in order to help the experimental analysis. Understanding differences in excitonic states and in the dependence of their properties on the electric field between the two sets of experiments[8–10] is also of interest.

We perform realistic calculations of the ground state and excited states of electron and hole in samples of Refs. 8 and 9 (called structure B) and Ref. 10 (called structure S) in a presence of electric field. We include the single-particle potential and the Coulomb interaction between the electron and hole on an equal footing. Our method is based on an exact numerical solution of the Schrödinger equation in a certain basis within the anisotropic effective mass approximation.[13] Using this method we calculate energies of various states, excitonic binding energies, oscillator strengths, and the two-particle wave functions in both samples as a function of applied electric field. Structure B consist of an 8-nm GaAs coupled quantum well (QW) separated by a 4-nm $Al_{0.33}Ga_{0.67}As$ barrier while structure S contains two 6-nm $In_{0.1}Ga_{0.9}As$ QW separated by a 4-nm GaAs. QW potential in structure B is almost 4 times deeper that in S. We use a static dielectric constant $\epsilon = 13.2$ and a conduction band offset ratio $Q_c = \Delta E_{cond}/\Delta E_g$ of 0.65. For the difference in band gaps on the GaAs/$Al_xGa_{1-x}As$ interface we use the formula $\Delta E_g = 1247 \times x$ meV for $x < 0.45$ while on the $In_yGa_{1-y}As$/GaAs interface the dependence $\Delta E_g = 1070 \times y$ valid for small y. Parameters used for the electron and hole effective masses are discussed later.

Having studied the shape of electron-hole wave function for ground and excited states as well as trends in excitonic binding energies E_b and oscillator strengths for various states with the change in electric field we can identify the differences between excitons in structure B and structure S. For structure B the picture is somewhat simpler and can be understood in terms of usual direct (D) and indirect (I) excitons, in which electron and hole are present in the same or in opposite QW's, respectively. Electrons and holes in structure B are very well confined inside the wells and the amplitude of their wave functions inside the barrier is very small. The confinement originates mainly from a deep potential created by AlGaAs/GaAs interface. The energies and oscillator strengths of direct and indirect excitons do not change very much with electric field, which only induces a switch be-

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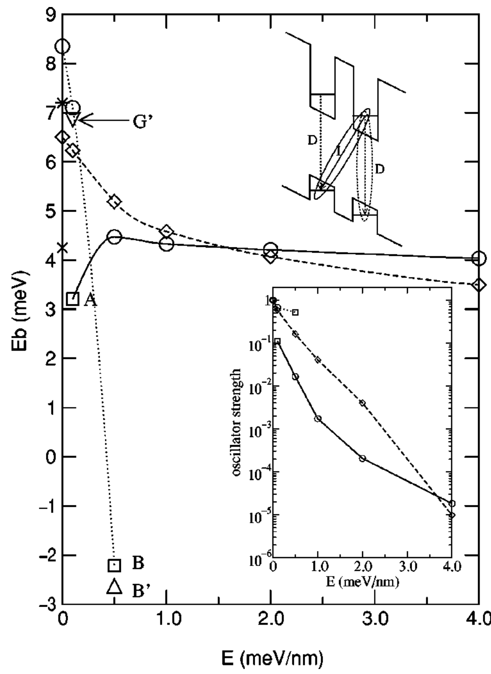

FIG. 1. Binding energy E_b of the ground-state exciton in structures S (diamonds and dashed line) and of various excitonic states in structure B as a function of electric field *E*. For structure B the E_b of an indirect exciton (solid line) as well as of the direct one (dotted line) is shown. The ground state is indicated by circles while squares, triangles, ⋆, and x mark higher-energy states discussed in the text and shown in Figs. 3 and 4. Note that E_b of the direct exciton is calculated with respect to energies of electron and hole in the ground state (electron and hole in separate wells). Inset: Oscillator strength as a function of *E* for various excitonic states. Notation the same as for E_b.

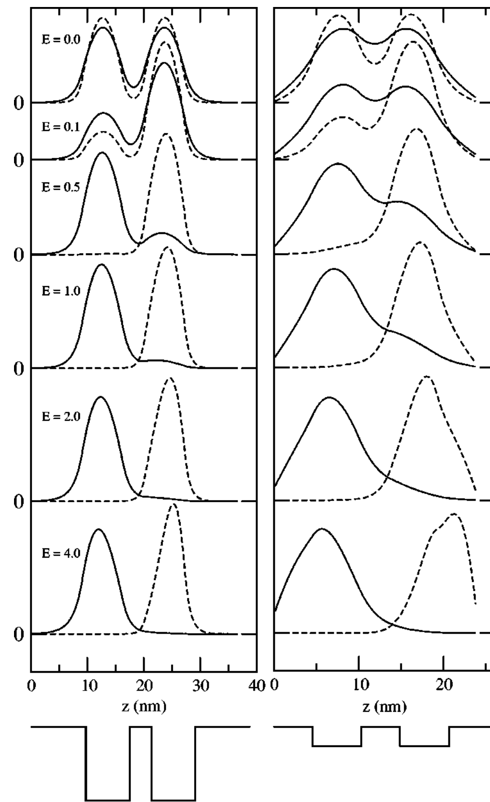

FIG. 2. Ground-state two-particle wave function for different values of applied electric field *E*, structure B (left panel) and structure S (right panel). Electron wave function in the out-of-plane direction *z* is shown after averaging over the hole position (solid line) and the hole wave function after averaging over the electron position (dashed line).

tween the two types of excitons. At zero or very small electric field up to around 0.5 meV/nm the ground state is a direct exciton with electron and hole being in the same QW. As the electric field increases the ground state switches to the indirect exciton, where electron and hole are in opposite QW's. This switching can be observed in a shape of electron-hole wave functions as well as in an abrupt drop of the ground-state exciton binding energy and oscillator strength around 0.5 meV/nm. For structure S the picture is more complicated. Potential wells in structure S are very shallow, resulting in a very large amplitude of the electron-hole wave function in the barrier, giving an almost three-dimensional-like (3D-like) exciton in the absence of an electric field. An external electric field pushes both carriers apart, continuously decreasing the binding energy and oscillator strength. Since the main mechanism of confinement in structure S is an electric field, the exciton binding energy and oscillator strength depend on the amplitude of the electric field more strongly than in structure B. There is no clear distinction between direct and indirect excitons in structure S at moderate electric fields. Electrons and holes smoothly separate in space as the electric field is increased.

Figure 1 shows the binding energy E_b and oscillator strength (in the inset) of the ground-state exciton in sample B (circles) and in sample S (diamonds). An inverse of the oscillator strength is proportional to the excitonic lifetime with respect to a radiative recombination. In Fig. 2 we show ground-state wave functions for structure B (left panel) and S (right panel) at different electric fields. Since the center-of-mass motion in the in-plane direction is a plane wave, the wave functions depend on four spatial coordinates: namely, the center of mass in the out-of-plane direction, *z*, and on three relative coordinates. The solid line is an electron wave function after averaging over the hole position while the dashed line shows the hole wave function after averaging over the electron position. In this work we discuss excitonic states only, for which the wave function in relative coordinates shows a very clear excitonic peak. Since it is very similar for all states, we will not show it here.

For structure B the ground-state binding energy and oscillator strength fall abruptly around $E=0.5$ meV/nm from 8.3 meV for E_b at zero field to around 4 meV at larger fields. Detailed inspection of the ground-state wave functions (see Fig. 2, left panel) reveals that this fall corresponds to a switch between direct (the two upper curves in the left panel of Fig. 2) and indirect excitons (the lower four curves in the left panel of Fig. 2). The binding energy of indirect excitons (solid lines in Fig. 1) does not change much with electric field. The dashed line shows binding energy of direct exciton (electron and hole in the same QW) with respect to the ground-state energy of free electron and hole without Cou-

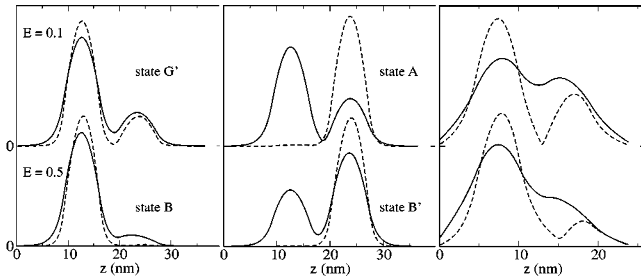

FIG. 3. Some characteristic excited-state excitonic wave functions for structure B (left and middle panels) and structure S (right panel). Notation as in Fig. 2 and the state labels as in Fig. 1.

lomb interaction (electron and hole being in separate QW's due to the presence of electric field). The large drop of this energy even below zero shows that direct excitons become unstable as the electric field is increased. At very low electric field ($E=0.1$ meV/nm) the indirect exciton appears as an excited state (marked A in Fig. 1 and in Fig. 3, upper middle panel) after the ground-state direct exciton in which electron and hole are localized in a well with a potential minimum for the hole and another direct excitonic state, very close in energy to the ground state, in which electron and hole are localized in a well with a potential minimum for the electron (marked G′ in Fig. 1 and Fig. 3, upper left panel). At 0.5 meV/nm electric field these two direct excitons appears as excited states (marked B and B′ in Fig. 1 and Fig. 3, lower left and middle panels) with the ground state being an indirect exciton.

The binding energy (and oscillator strength) of the ground-state exciton in structure S (diamonds and dashed line in Fig. 1) smoothly decreases with electric field from 6.5 meV for E_b at zero field to 3.5 meV at 0.4 meV/nm. The nature of states cannot be explained simply in terms of a direct and indirect exciton picture. At low electric fields the electron and hole wave functions have very large overlap (three upper curves in Fig. 1, right panel) although their maxima are moving into opposite wells, as E increases, decreasing the binding energy and oscillator strength. One can also identify higher-energy states which would correspond in some sense to direct excitons, where the maxima of electron and hole wave functions are in the same well (see Fig. 3, right panel). It can be seen in Fig. 2 (right panel) then the separation and confinement is generated more by electric field than the potential barrier.

In Fig. 4 we show an asymmetric exciton, which is the first excited state at zero electric field. Asymmetric excitons have zero oscillator strength and the binding energy is marked as ⋆ (for structure B) and × (for structure S) in Fig. 1.

Finally, Fig. 5 shows the absolute energies of direct (dashed line) and indirect (solid line) excitons for structure B (upper curves marked with circles) and structure S (lower curves marked with diamonds). As observed in experiment[14] the direct exciton energy remains approximately constant while the indirect exciton energy decreases significantly with the electric field. For structure B we record approximately linear decrease with a slope of around 24 meV per 1

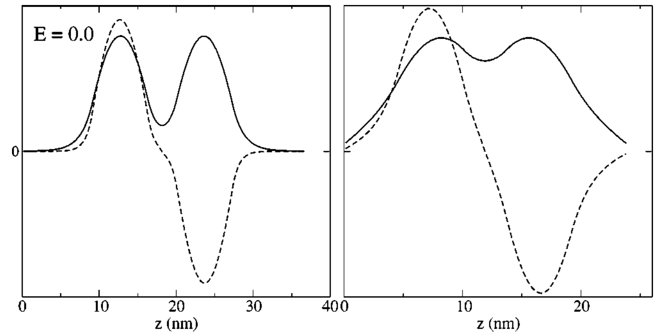

FIG. 4. Asymmetric exciton wave function for structure B (left panel), marked by ⋆ in Fig. 1, and structure S (right panel), marked by × in Fig. 1. Notation as in Fig. 2.

meV/nm applied electric field in comparison with around 20 meV in experiment.[14] Again, switching between the two states in structure B (circles) can be observed as crossing of the two curves.

The numerical method we are using is very accurate and the binding energies are converged to around 0.2 meV,[13] but the accuracy of the results can also be influenced by the uncertainty associated with the input parameters, mainly the hole effective mass. The exciton effective mass in structure B was measured to be $0.22m_0$,[15] which is in a good agreement with calculated mass of heavy hole exciton in GaAs QW of $0.25m_0$,[15] using electron mass $m_e=0.067m_0$ and in-plane heavy hole mass $m_{hx}=0.18m_0$ from Ref. 16. We use these values of parameters together with out-of-plane heavy hole mass of $m_{hz}=0.34m_0$ for our calculations. In other work[17] the Luttinger parameters used for GaAs coupled QW's correspond to the in-plane effective mass of around $0.1m_0$. We have calculated the ground-state exciton binding

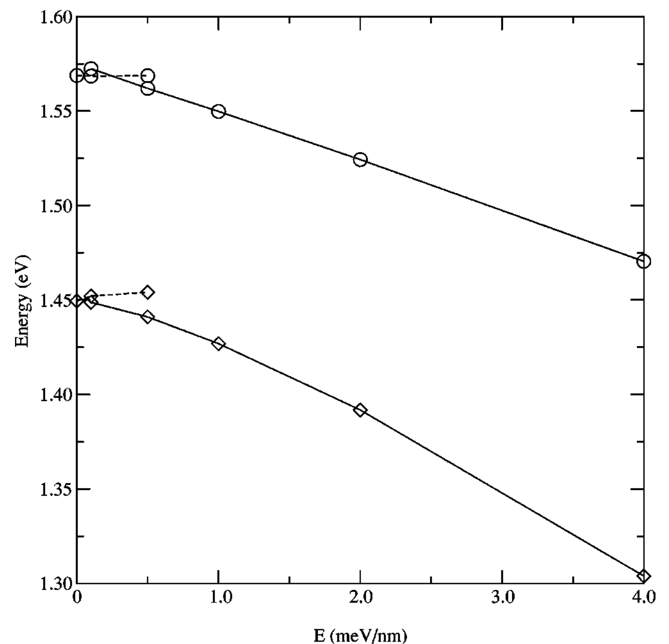

FIG. 5. Energies of the indirect (solid line) and the direct (dashed line) excitons for structure B (upper curves) and structure S (lower curves).

TABLE I. Binding energy E_b in meV of the ground-state exciton at zero electric field in sample S and in sample B for different values of the heavy hole effective mass.

	S		B
$m_{hx}=m_{hy}$	$m_{hz}=0.339$	$m_{hz}=0.7$	$m_{hz}=0.34$
0.07		6.02	
0.1	6.51		7.38
0.18			8.35
0.22	7.31	7.32	

energy at zero electric field using $m_{hx}=0.1m_0$, and we have found the difference of 1 meV with respect to the binding energy calculated using $m_{hx}=0.18m_0$ (see Table I). For structure S we use electron mass $m_e=0.061m_0$, in-plane heavy hole mass $m_{hx}=0.1m_0$, and out-of-plane heavy hole mass $m_{hz}=0.339m_0$. The discrepancy for the hole mass in structure S seems larger than for the structure B. The differences in out-of-plane hole mass are less important as even for almost twice as large out-of-plane heavy hole mass of $0.7m_0$ suggested[12] the difference in the binding energy is negligible (see Table I). The magneto-optical measurements on strained $In_{0.12}Ga_{0.88}As$ well with width between 20 and 5 nm show a variation of the in-plane heavy hole mass between $0.17m_0$ and $0.22m_0$.[18] Another source[19] gives the measured hole mass of $0.14m_0$ in strained $In_{0.2}Ga_{0.8}As/GaAs$ structures. The calculated in-plane heavy hole effective mass is given to be around $0.125m_0$ in a strained 9-nm $In_{0.18}Ga_{0.82}As$ QW.[20] Thus we calculate the ground state exciton binding energy in structure S for an in-plane heavy hole mass of suggested 0.07,[12] 0.1, and the upper bound of $0.22m_0$, which gives a difference of around 1 meV with respect to the value of $0.1m_0$ used in this work (see Table I).

To our knowledge there has been no calculations for InGaAs/GaAs coupled QW's with electric field. For coupled GaAs/AlGaAs QW's we have found two variational calculations of the ground-state exciton binding energy,[17,21] which, however, have not included parameters of structure B. Calculations in Ref. 21 consider only coupled QW's with 1.415-nm barriers, which is 3 times narrower than in structure B. Calculations in Ref. 17, however, consider a wide range of different structures for different values of well width and barrier. The Luttinger parameters used correspond to the in-plane hole mass of $0.1m_0$. The ground-state excitonic E_b at zero electric field of the most similar structures to sample B, which are considered in Ref. 17—namely, 5, and 10-nm coupled QW's with 4-nm barrier—are around 6.7 meV and 5.9 meV, respectively, which are slightly lower than our 7.3 meV (see Table I) for 8-nm QW's with similar value of the hole mass.

For additional check of the method and parameters used we have performed calculations for 10 ML $In_{0.08}Ga_{0.92}As/GaAs$ coupled QW's with 3-nm barriers for which the ground-state exciton binding energy at zero field has been experimentally measured to be 8.4 meV.[22] Our calculations using $0.1m_0$ in-plane heavy hole mass give 7.43 meV for this structure which is in very good agreement taking into account the possible error due to the in-plane hole mass uncertainty of 1 meV (see Table I).

Summarizing, we have studied differences in excitonic states and origins of confinement in structure S and structure B, in which novel effects have recently been reported. We have also studied in detail the binding energy of different types of excitons as the electric field is changed. The binding energy of indirect excitons in structure S for the values of electric field used in the experiment[10] is around 4 meV, and thus much too small for excitons to exist up to 118 K. The ring structure, similar in both experiments, which persist up to 118 K, seems more likely to be attributed to other effects than associated with bound excitons.

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