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Exciton fine structure and biexciton binding energy in single self-assembled InAs/AlAs quantum dots

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The exciton and biexciton emissions of a series of single quantum dots of InAs in an AlAs matrix have been studied. These emissions consist of linear cross polarized doublets showing large values of both the biexciton binding energy and the fine-structure splitting. At increasing exciton emission energy, corresponding to decreasing dot size, the biexciton binding energy of 9 meV decreases down to zero, reflecting a possible crossover to an antibinding regime. Simultaneously the fine-structure splitting diminishes from a value of 0.3 meV down to zero, at the same energy, suggesting a common origin for the two effects. © *2006 American Institute of Physics*.
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INTRODUCTION

Semiconductor quantum dots (QDs) are important for production of single photons[1] and entangled photon pairs[2,3] for quantum information processing, due to the discrete and tunable electronic structure of zero-dimensional systems. Confinement of carriers in QDs at distances smaller than the Bohr radius increases electron interactions, which are crucial to determine the electronic structure and the optical properties of the QD. Electron interactions determine the relative alignment and the eventual coherent coupling of excitons and biexcitons,[4] which has been proposed as a suitable system for quantum gate operations.[5] The biexciton binding energy E_b^{XX} and the fine-structure splitting Δ_{FS} are basic features of QDs caused by Coulomb interactions, which depend on the QD shape, size, and strain. The biexciton binding energy is the difference in energy needed to create a second electron-hole pair in a QD with already one pair present. E_b^{XX} depends on the details of Coulomb interaction and it can become negative when confinement is reduced in small dots, leading to biexciton antibinding.[6–8] The origin of Δ_{FS} is the asymmetry of the electron-hole exchange along the (110) and (1 – 10) crystallographic directions, which leads to the splitting of the degenerate bright exciton states. This asymmetry has been initially attributed to a QD lateral elongation along these directions.[9–12] In addition to that, the asymmetric piezoelectric potential associated to shear strain[13–15] and the intrinsic atomistic asymmetry of the zinc-blende lattice[8,16] can also produce a splitting even in a cylindrical QD. The control of Δ_{FS} is important because the possibility to make it smaller than the homogeneous emission linewidth of the QD is a crucial step to obtain entangled pairs of photons. Recent results reveal a sign reversal of Δ_{FS} for increasing photoluminescence (PL) emission energy[17] in InAs/GaAs dots.

While the vast majority of systems studied so far are InAs QDs with GaAs or AlGaAs (Ref. 11) barriers, the use of AlAs barriers has some advantages: (i) stronger carrier confinement leading to visible emission and enhanced electron interactions, (ii) longer lifetimes[18] due to the poorer phonon coupling to the barriers,[19] and (iii) strong reduction of intermixing between the QD and the barriers.[20,21] In addition, the higher confinement leads to a different mixing of heavy and light holes, which in turn affects the electronic and optical properties of the QD.

In this paper we report a PL study of the biexciton binding energy and the fine-structure splitting in single InAs/AlAs QDs. Both quantities have larger values than in InAs/GaAs dots and decrease monotonously with increasing QD emission energy,[22] i.e., with decreasing QD size. Both E_b^{XX} and Δ_{FS} vanish for emission energy around 1.63 eV, suggesting a possible sign reversal in both magnitudes. These results suggest a common origin for the size dependent reduction and vanishing of E_b^{XX} and Δ_{FS}. They are tentatively interpreted in terms of the changes in the exciton extension and shape as the confinement is reduced when the dot size is decreased. Our results give support to the possibility to create entangled photon pairs in the visible region using InAs/AlAs QDs.

EXPERIMENT

The InAs/AlAs QD samples were grown by molecular beam epitaxy (MBE) on GaAs (100) substrates as described in Ref. 23. The structure consists of a single layer of InAs QDs confined by 20 nm thick AlAs barrier layers, as shown in Fig. 1(a). A 10 nm GaAs cap layer was grown on top of the sample. Interruption of sample rotation during QD deposition produced an InAs coverage gradient across the wafer. The coverage varied between 1.6 and 2.0 monolayers, corresponding to different dot sizes and densities, as measured by

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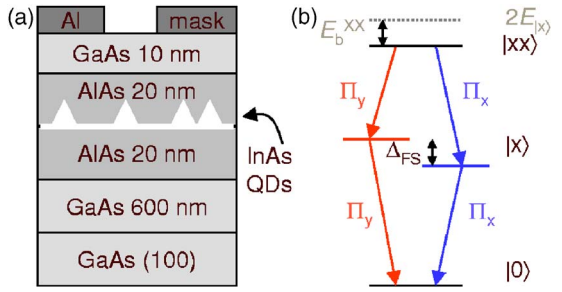

FIG. 1. (Color online) (a) Layer structure of the sample with InAs quantum dots embedded in an AlAs matrix. (b) Exciton level scheme: Fine-structure splitting of the $|X\rangle$ state results in linear cross polarized doublets for the X and XX transitions.

atomic force microscopy.[23] Typical values are QD densities of 10^{10} cm^{-2}, dot diameters between 25 and 30 nm, and dot heights around 3 nm. The samples were covered by an aluminum mask with squared apertures of different sizes fabricated by electron beam lithography. The aperture sizes range from 10 to 0.2 μm, allowing for detection of single QD luminescence. The PL emission was excited with an argon-ion laser with a typical power density on the sample of 10^4 W/cm^2. This rather large excitation power was necessary to compensate for the low intensity of the light collected from the dots. The sample excitation and the light collection were done with a microscope setup using a long working distance objective ($50\times$) and a double grating spectrometer with a charge-coupled device detector. The accumulation time in the detector was typically between 1 and 30 min. The excitation spot size was 5 μm. The sample was cooled in a continuous flow He cryostat for microscope application. All the measurements were taken at about 9 K.

RESULTS AND DISCUSSION

Selected openings of the sample metal mask, as well as larger areas (10×10 μm^2) outside the mask, were initially mapped by PL to identify exciton (X) and biexciton (XX) peaks belonging to the same QD. The identification required three conditions: (A) parallel random time evolution of the PL energies (jitter), which is due to random changes in the electrostatic potential by carrier trapping processes near the QD;[24] (B) linear and quadratic PL intensity dependences on excitation power for X and XX, respectively, in the low excitation regime;[25] and (C) linear counterpolarization of the X and XX split emission lines along the (110) and (1−10) directions. The spin-singlet biexciton ground state does not split; however, the biexciton transition inherits its fine structure from the final exciton state[10] [Fig. 1(b)], which is split due to structural in-plane asymmetry of the QDs or piezoelectric effects. Therefore the X and XX emission lines consist each of two orthogonally linear polarized transitions with identical splitting but reversed polarization.

A typical result showing (A) and (C) criteria is presented in Fig. 2. Figures 2(c) and 2(d) show details of the PL spectra [Fig. 2(b)] for the two orthogonal linear polarizations (continuous and dashed lines, respectively) showing the X and XX emissions around 1.566 and 1.574 eV, respectively. The quantities E_b^{XX} (~8 meV) and Δ_{FS} (~0.2 meV) are indicated. Figure 2(a) shows the time evolution of the PL emission energies for unpolarized light collection. The figure contains a series of 30 spectra of 1 min integration time each. Criterion (B) is shown in Fig. 3, where the PL intensity of the X and XX emission lines is plotted as a function of excitation power in logarithmic scale. The straight lines with slopes 1 and 2 unambiguously identify exciton and biexciton emissions, respectively.[26]

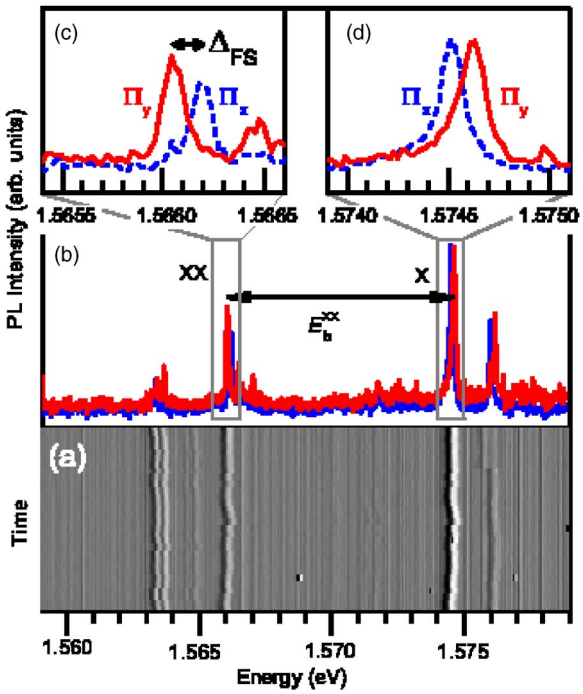

FIG. 2. (Color online) (a) Time evolution of the spectrum shown in (b). (b) Photoluminescence spectra of the exciton (X) and biexciton (XX) of a single quantum dot showing linear cross polarization along the main crystal axes [110] and [1−10] shown in detail in (c) and (d).

Eleven QDs with different emission energies have been investigated to determine the dependence of the biexciton binding energy and the fine-structure splitting on the QD exciton emission energy. The results for E_b^{XX} and Δ_{FS} are shown in Figs. 4(a) and 4(b), respectively. The main result in Fig. 4(a) is the decrease of the biexciton binding energy with the exciton emission energy and its vanishing at the highest exciton energy measured (1.63 eV). This trend is the same as in InAs dots with GaAs barriers,[6,7] although the observed values of E_b^{XX} are essentially larger (up to 9 meV) than the

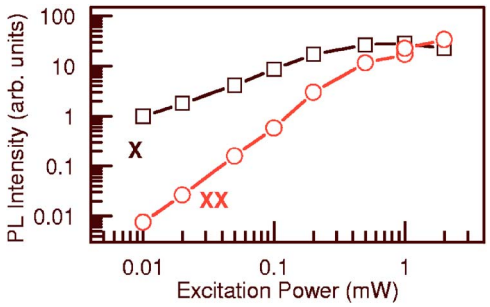

FIG. 3. (Color online) Excitation power dependence of the exciton (X) and biexciton (XX) emission of a single quantum dot. The intensities I grow with the excitation power P linearly and quadratically, respectively.

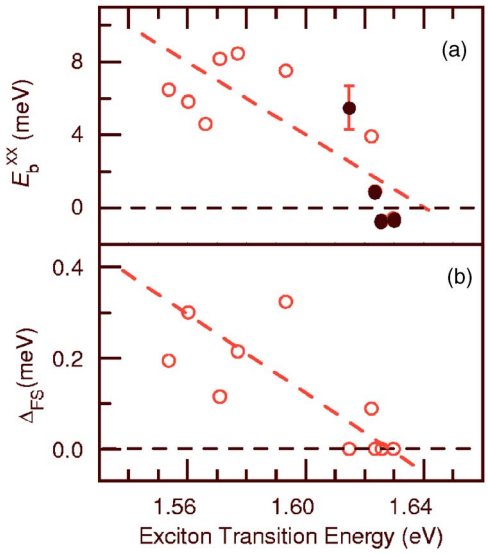

FIG. 4. (Color online) (a) Biexciton binding energy and (b) fine-structure splitting of the bright exciton state vs exciton emission energy. For the dots marked with filled circles no splitting could be measured, making the reliable assignment of the biexciton peak difficult, which then was only determined by the excitation dependence. For the data point with an error bar, several peaks were candidates for being biexcitons. Dashed lines serve as guide to the eye.

common values reported for GaAs barriers (~2–3 meV). The three black points at the higher emission energies correspond to QDs whose fine-structure splitting was too small to be resolved. In these cases only criteria (A) and (B) were used to identify X and XX lines of a single dot. The value of the binding energy E_b^{XX} is determined by the Coulomb interaction and more specifically by correlation between electrons and holes. It has been shown experimentally[6,7] and theoretically[6–8] that E_b^{XX} can vanish and change its sign depending on the dot size. A state with negative binding energy (antibinding) can exist because of the three dimensional confinement. Antibinding of the biexciton ground state is expected to occur in small dots when the number of bound states decreases, thus reducing the correlation energy.[7] In our case the high AlAs barriers result in a stronger exciton confinement and more bound states in the QD. The higher biexciton binding energy observed in our dots can be thus attributed to increased correlation effects.

The variation of the fine-structure splitting Δ_{FS} with the QD exciton emission energy is shown in Fig. 4(b). We observe large values of Δ_{FS} up to 0.3 meV similar to InAs/AlGaAs dots [up to 1 meV (Ref. 11)] and to recently reported values for InAs/GaAs QDs [up to 0.52 meV (Ref. 15)]. This is larger than the previous reported values (0.05–0.2 meV) for In(Ga)As/GaAs QDs.[9,26] The value of Δ_{FS} decreases with the exciton emission energy and vanishes at roughly the same energy as E_b^{XX}. This suggests a possible splitting reversal for emission energies above 1.63 eV. Such sign reversal has been predicted for InGaAs/GaAs QDs (Ref. 27) of changing size and recently reported experimentally in InAs/GaAs QDs.[17] No effect of the QD density on Δ_{FS} has been observed in our sample, so that we can exclude in our case any influence of interdot interactions on Δ_{FS}.

Three different possibilities have been given as the origin of the exciton fine-structure splitting, which are not mutually excluding: (a) electron-hole exchange anisotropy induced by lateral dot elongation,[10,11] (b) potential anisotropy due to strain-induced piezoelectric fields,[13–15] and (c) intrinsic anisotropy of the atomic structure.[8,16,27] The dot elongation mechanism predicts larger Δ_{FS} in smaller dots[10] and is unlikely to account alone for its sign reversal. The piezoelectric mechanism implies elongation of the electron and hole wave functions along (110) and (1−10) directions, respectively, for pyramid-shaped QDs.[14] As this elongation is hindered in small QDs, Δ_{FS} is expected to decrease for decreasing dot size. A mixture of the mechanisms of (a) and (b) has been invoked to explain the experimental results of Ref. 17. An atomistic calculation also predicts a decrease of Δ_{FS} for decreasing dot heights in lens-shaped circular dots.[16] The similar trend of Fig. 4(b) to previous results in InAs/GaAs dots,[15,17] where sign reversal of Δ_{FS} has been found, allows us to assume that piezoelectric and intrinsic symmetry mechanisms are at the origin of our results. Moreover, the simultaneous vanishing of Δ_{FS} and E_b^{XX} around 1.63 eV strongly suggests a common origin: The spread of carriers out of small dots due to reduced confinement seems to account for a simultaneous reduction of exchange and correlation terms, leading to the decrease of Δ_{FS} and E_b^{XX}, respectively.

CONCLUSION

In photoluminescence spectra of InAs/AlAs quantum dots, emission lines of excitons and biexcitons of individual dots have been observed. Both the fine-structure splitting of the exciton level and the biexciton binding energy decrease for increasing exciton emission energy (decreasing dot size), disappearing around 1.63 eV. These effects possibly originate in the spread of carriers out of small dots due to reduced confinement. In comparison with InAs/GaAs quantum dots, a larger biexciton binding energy (up to 9 meV) and a larger fine-structure splitting of the bright exciton state (up to 0.3 meV) are observed and may be attributed to a larger confinement due to the higher AlAs barriers.

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