In situ tuning biexciton antibinding-binding transition and fine-structure splitting through hydrostatic pressure in single InGaAs quantum dots

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received 21 April 2014; accepted in final form 4 July 2014
published online 22 July 2014

PACS 78.67.Hc – Quantum dots
PACS 07.35.+k – High-pressure apparatus; shock tubes; diamond anvil cells
PACS 78.55.Cr – III-V semiconductors

Abstract – Exciton and biexciton emission energies as well as excitonic fine-structure splitting (FSS) in single (In,Ga)As/GaAs quantum dots (QDs) have been continuously tuned using hydrostatic pressure up to 4.4 GPa. The blue shift of excitonic emission and the increase of FSS are 320 meV and 150 \( \mu \)eV, respectively, which are significantly greater than those that could be achieved by previously reported techniques. We successfully produce a biexciton antibinding-binding transition along with a detailed polarization-resolved emission spectra. It is shown that the biexciton binding energy linearly increases with increasing pressure and tends to be sublinear at high pressure. We have performed atomistic pseudopotential calculations on realistic QDs to understand the pressure-induced effects.

Introduction. – Self-assembled semiconductor quantum dots (QDs) have considerable potential for the use as fundamental building blocks in future quantum information applications. Even though site-controlled growth techniques have been used to control the properties of epitaxial QDs [1,2], it is still very difficult to precisely tune the QD optical properties using growth techniques alone, which are essential for such applications. Therefore, externally tuning of the QD properties after the growth is extremely important. One of the most prominent examples is the polarization-entangled photon pair emission through a bieexciton cascade process in QDs, which requires that different polarized photons are energetically indistinguishable [3]. However, the underlying asymmetry of the self-assembled (In,Ga)As/GaAs QDs leads to an energy splitting of the bright exciton states, known as fine-structure splitting (FSS), which is typically tens of \( \mu \)eV [4–7], and much larger than the radiative linewidth \( \sim 1.0 \mu \)eV [8]; therefore, photon entanglement is destroyed [3,9]. Different tuning techniques to erase the FSS, such as thermal annealing [10], electric fields [11–13], magnetic fields [14], and strain fields [15–22] have been explored. Alternatively, it is also possible to generate the entangled photon pairs via a “time reordering” scheme [23], which requires the biexciton binding energies to be zero. Uniaxial and biaxial stresses have been used to tune the QD structural symmetry, exciton binding energies and FSS. However, the stress that can be generated using these techniques is limited to approximately tens of MPa, which corresponds to a maximum spectral shift by approximately 20 \( \mu \)eV [16–18]. Herein, we report a high-pressure research for individual QDs using the diamond anvil cell (DAC), which has been widely used to study metal-semiconductor transitions, electronic structures, and optical transitions in bulk crystals and microstructures [24–27]. A reported exciton emission line shift in ensemble of InAs/GaAs QDs is as large as approximately 500 \( \mu \)eV at 8 GPa [25], which is much larger than the QD peak shift induced by a piezoelectric actuator PMN-PT [16]. However, till now tuning the QD structural symmetry, exciton transition states and FSS for an individual QD using the DAC has not been reported.
In this work, we demonstrate that the exciton (X) emission energy ($E_X$), bieexciton (XX) binding energy ($E_{BB}(XX)$) and FSS can be continuously tuned using hydrostatic pressure in situ in an optical cryostat up to 4.4 GPa, thanks to the improvement of the DAC pressure device driven by a piezoelectric actuator. With increasing pressure, the $E_X$ increases linearly and the FSS also shows an approximately linear increase. The $E_{BB}(XX)$ increases monotonically with increasing pressure, and the increase tends to be sublinear at high pressure. The bieexciton antibinding-binding transition is observed in some dots. The maximum $E_X$ shift and FSS change can reach to 320 meV and 150 μeV, respectively, which are considerably greater than what can be reached by other techniques. We also perform atomistic pseudopotential calculations on realistic (In,Ga)As/GaAs QDs under hydrostatic stress to discern the physical mechanisms underlying the effects induced by hydrostatic pressure.

Experimental details. – The investigated (In,Ga)As/GaAs QD samples with low QD density ($\sim 4 \times 10^7 \text{cm}^{-2}$) were grown using molecular beam epitaxy (MBE) on a semi-insulating GaAs substrate. Six typical QDs (QD1–QD6) with $E_X = 1.35–1.43 \text{eV}$ were chosen in this study for demonstrating the pressure-induced change of the QD exciton and bieexciton transitions and FSS. Figure 1(a) shows the DAC pressure device used for tuning QD photoluminescence (PL). To fit the QD samples into the DAC chamber (indicated in fig. 1(b)), the samples were mechanically thinned to a total thickness of approximately 20 μm and then cut into pieces of approximately 100 × 100 μm$^2$. Condensed argon was used as the pressure-transmitting medium. The initial pressure can be adjusted at room temperature by driving screws and can be determined in situ using the ruby R1 fluorescence line shift [28], where the ruby is located in the sample chamber. To tune the X and XX transition energies and FSS by pressure at low temperature, an electrically driven DAC (shown in fig. 1(a)) was developed by combining the well-known DAC with a piezoelectric actuator. The QD sample in the DAC is cooled to 20 K through a continuous-flow liquid-helium cryostat and excited by a He-Ne laser with an excitation laser is focused to an approximately 2 μm diameter spot on the sample using a microscope objective (NA: 0.35). The PL is collected using the same objective, spectrally filtered through a 0.5 mm monochromator, and detected using a silicon-charge coupled device (CCD) with a spectral resolution of 90 μeV. The QD linewidth is approximately 8 μeV measured at zero pressure by a scanning Fabry-Perot interferometer with a spectral resolution of 0.5 GHz and free spectral range of 15 GHz. To measure polarization-resolved PL and FSS, a λ/2 wave plate and a linear polarizer are used to find two maximum intensity of X emission through rotating the λ/2 wave plate in front of the polarizer. We notice that even at zero pressure the linearly polarized plane of the QD emission is not always aligned with the crystal direction of GaAs [110] or [110] orientation. Furthermore, we also found in the experiment that different QDs have a different rotation degree of the linearly polarized plane corresponding to the [110] direction with increasing pressure. Thus, FSS is determined by calculating the energy difference between the PL peaks of two maximum intensities of cross-polarized X emission. For simplicity in the discussion below, two linear polarization emissions of PL are still named as the horizontal (H) and vertical (V) components. Figure 1(c) displays the measured pressure values and excitonic emission energies at 20 K as a function of voltage applied on the piezoelectric actuator, showing the blue shift of the PL peak energy as large as 320 meV. The shift is much larger than the previously reported shift ($\sim 20 \text{meV}$) from uniaxial or biaxial stress [15–20].

Theoretical methods. – To understand the experimental results, we calculate the electronic and optical properties for the In$_{1-x}$Ga$_x$As/GaAs QDs under hydrostatic pressure using an atomistic empirical pseudopotential method (EPM). The InGaAs/GaAs QDs are modeled by embedding the InGaAs dots in a 60 × 60 × 60 8-atom GaAs supercell. The QD structures are optimized by the valence force field (VFF) method [29]. The supercell is large enough to assume that the supercell itself deforms under hydrostatic pressure as a bulk material. However, the strain changes in and around the QDs are not homogeneous, and the effects are taken account of by the following VFF relaxation. Indeed, the hydrostatic pressure not only change the isotropic strain, but also the biaxial and shear strains in the QDs. Here we neglect the piezo-field in our calculations since it has been shown that it is not important because the second-order piezoelectric effects tend to cancel in part the first-order ones [30]. We then
calculate the electron/hole single-particle energies and wave functions using the linear combination of the bulk bands (LCBB) method [31]. The exciton and biexciton energies are calculated via the configuration interaction (CI) method [32]. We present the results mainly for three types of QDs: i) QD-A: a lens-shaped In_{0.8}Ga_{0.2}As/GaAs QD with height \( h = 1.5 \text{ nm} \), base \( b = 12 \text{ nm} \); ii) QD-B: a lens-shaped In_{0.8}Ga_{0.2}As/GaAs QD with \( h = 1.5 \text{ nm} \), \( b = 15 \text{ nm} \); and iii) QD-C: an elliptical based In_{0.8}Ga_{0.2}As/GaAs QD with \( h = 2.0 \text{ nm} \) and the elliptical major (minor) axis \( a = 10 \text{ nm} \) (\( b = 7.5 \text{ nm} \)) along the [110] ([110]) crystal direction.

**Results and discussion.** – Figure 2(a) depicts the exciton emission energies as a function of the pressure from \( P = 0 \) to 4.4 GPa for QD1–QD5. When there is no external pressure, the QD exciton emission energies are 1.402, 1.432, 1.394, 1.388 and 1.401 eV, respectively. This means increase linearly with the applied pressure. The pressure coefficients obtained by linearly fitting to the data are 90, 81, 85, 94, and 82 meV/GPa for QD1–QD5, respectively. Figure 2(b) shows the biexciton binding energies for QD1–QD5 as a function of pressure. The biexciton binding energy is defined as \( E_B(XX) = E_X - E_{XX} \), where \( E_{XX} \) is \( XX \) emission energy. When \( E_B(XX) > 0 \), the biexciton is in the “binding” state, and the two excitons are attracted. When \( E_B(XX) < 0 \), the biexciton

Fig. 2: (Color online) (a) Exciton emission energies as a function of pressure for QDs (QD1–QD5). (b) Biexciton binding energies as a function of pressure for QDs (QD1–QD5), showing biexciton antibinding-binding transitions under pressure for QD1, QD4 and QD5. (c) FSS of QDs (QD2, QD3 and QD6) as a function of pressure.

Fig. 3: (Color online) (a)–(e) Polarization-resolved PL spectra for QD5 under different pressures, where red lines (black lines) correspond to the horizontal (H) and vertical (V) polarized photons, respectively. At 1.62 GPa (b) and 2.07 GPa, (d) color-indistinguishable photon pairs are generated for V- and H-polarized photons, respectively. At 1.97 GPa (c), across generation color coincidence for XX and X transition energies is achieved. (f) Level schemes showing the XX-X cascade.

is in the “antibinding” state, and the two excitons are repulsive. For the QDs studied here, \( E_B(XX) \) increases monotonically and sublinearly with increasing pressure, as observed for QD1–QD5. Such nonlinear behavior of \( E_B(XX) \) as a function of pressure depends on the QD geometry and indium composition. For QD1, QD4 and QD5, the biexcitons are in an antibinding state at zero pressure and gradually progress to the binding state at approximately 0.3, 0.7 and 1.5 GPa, respectively. A similar process of the antibinding-to-binding transition was reported [33,34].

To demonstrate the biexciton antibinding-binding transitions under pressure in more detail, we plot the polarization-resolved PL spectra for the QD5 under different pressures, as shown in fig. 3(a)–(e), where the red and black lines correspond to the horizontal (H) and vertical (V) polarized photons, respectively. We denote \( E_{XX}(H2) \) and \( E_{XX}(V2) \) as transition energies from XX to two bright XX states, and \( E_{XX}(H1) \) and \( E_{XX}(V1) \) as two transition energies from two bright X states to ground (G) states (see the schematic diagram of fig. 3(f)). At zero pressure, both \( E_{XX}(H2) \) and \( E_{XX}(V2) \) are larger than \( E_{XX}(H1) \) and \( E_{XX}(V1) \) (see fig. 3(a)), respectively, which means that the biexcitons are in the “antibinding” states. In addition, \( E_{XX}(H2) \) is slightly larger than \( E_{XX}(V2) \) because of the existence of FSS. Under pressure, the blue shift rate of \( E_{XX} \) is bigger than that of \( E_{XX} \). Therefore, with increasing pressure, the V-polarized XX and X emission lines first overlap at 1.62 GPa (see fig. 3(b)), and then the H-polarized XX and X emission lines overlap at 2.07 GPa (see fig. 3(d)). Remarkably, at 1.97 GPa, across generation, color coincidence for XX and X transition energies is generated (i.e., \( E_{XX}(H2) = E_{XX}(V1) \) and \( E_{XX}(V2) = E_{XX}(H1) \)). This is a key condition for generating entangled photon pairs via the “time reordering” scheme [23].

FSS tuning by anisotropic biaxial stress has been studied experimentally [15,19,35,36] and
theoretically [21,22,37], which has shown that the maximum tuned FSS value is approximately $50\mu eV$. However, it is interesting that a much larger FSS change under hydrostatic pressure can be observed in our experiment. Figure 2(c) depicts the FSS for QD2, QD3 and QD6 as a function of pressure. The figure clearly demonstrates that increasing pressure from 0.5 to 4 GPa produces an approximately linear increase in FSS with the slope of 44, 28 and 42 meV/GPa for QD2, QD3 and QD6, respectively, which generates a total FSS shift as large as 150, 100 and 150 meV for QD2, QD3 and QD6, respectively. Similar results were observed from other investigated (In,Ga)As/GaAs QDs, which indicates that such a large shift is typical for FSS under high hydrostatic pressure. We also note that the FSS value at initial pressure of 0.5 GPa for QD6 has not shown any obvious increase compared to the FSS value at $P = 0$ GPa (here, an FSS change was obtained from 0.5 to 4 GPa when the QD sample was in the DAC, whereas FSS the value at $P = 0$ GPa was measured when the QD sample was out of the DAC). The possible causes are still unclear.

The calculated exciton emission energies under pressure are shown in fig. 4(a) and the blue shift rate of 76 meV/GPa is produced for the three In$_{0.8}$Ga$_{0.2}$/GaAs QDs, which is slightly smaller than the experimental value. In fact, the exciton emission energy blue shift rate depends on the Ga composition of the QDs. For the pure InAs dot, the blue shift rate is around 65 meV/GPa. With the increase of the Ga composition to 0.3, the blue shift rate increases to around 85 meV/GPa, because GaAs has larger (in absolute value) band gap deformation potential, $-7.88eV$, than that of InAs, $-6.79eV$ [38]. To understand the emission energy blue shift, we analyze the band offset values and confinement potentials for the QDs under pressure, which strongly depend on the strain distribution in the dots and matrix. When hydrostatic pressure is applied, the lattice constant for the matrix material GaAs decreases, which effectively increases the lattice mismatch between the dot material InAs and GaAs matrix. Therefore, in addition to the change of isotropic strain, there is also a change of biaxial strain in the QDs [39]. As a result, both (absolute values of) isotropic and biaxial strains inside the dots increase. The averaged isotropic strain $I = -0.072 - 0.011 P$ and biaxial strain $\epsilon_{zz} - \epsilon_{xx} = 0.12 + 0.0014 P$ are obtained from VFF calculations, where $P$ is the applied pressure in GPa. Figure 4(b) depicts the strain-modified band offsets for the conduction band (e), heavy-hole (HH), light-hole (LH) and spin-orbit (SO) bands through the dot center under $P = 0$, 2.0 and 4.0 GPa. Whereas the band offset change is small for holes, the band offset changes dramatically for the conduction band. Under pressure, the electron bands move significantly toward a higher energy. The confinement potential also increases dramatically with increasing pressure, which is the major reason for the observed experimental results.

Because the electron-hole Coulomb energy change is relatively small (see fig. 4(d)), the change in exciton emission energy can be estimated using the electron-hole single-particle gap $E_g$, which can be written as follows:

$$E_g(\epsilon) = E_g(0) + a_g I + b_0(\epsilon_{zz} - \epsilon_{xx}),$$

where $\epsilon$ is the strain tensor inside the InAs dots, $a_g = -6.79 eV$ is the hydrostatic deformation potential for the band gap, and $b_0 = -1.62 eV$ is the biaxial deformation potential for the valence band maximum [38]. Therefore, we estimate that the exciton PL blue shift under pressure is 72 meV/GPa for pure InAs/GaAs QDs, which is consistent with the EPM calculations. We note that applying hydrostatic pressure is a much more efficient way to tune the exciton emission energy than uniaxial stress due to the fact that the band gap shift mostly depends on the hydrostatic strain [17,35,40].

![Fig. 4: (Color online) The calculated results of: (a) The exciton emission energies of three QDs as a function of hydrostatic pressure. (b) The band offsets of a pure InAs/GaAs dot at $P = 0$, 2.0 and 4.0 GPa hydrostatic pressure. The black, green, blue and red lines indicate conduction (e), heavy-hole (HH), light-hole (LH) and spin-orbit (SO) bands, respectively. (c) The biexciton binding energies as a function of hydrostatic pressure in the QDs. (d) The changes of direct Coulomb integrals of the lowest electron and hole states in QD-A as a function of the applied pressure. The solid purple line is the change of $E_B(XX)$ calculated by the direct Coulomb energies using eq. (2), whereas the dashed purple line is the change of $E_B(XX)$ calculated from EPM. (e) The FSS and $\Delta_{bd}$ in QD-C as a function of the applied pressure. (f) The wave functions of the lowest electron (e0) and hole (h0) states (the red circles) at $P = 0$ and 4.0 GPa hydrostatic pressure in the QD-A. The outermost circles indicate the QD cross-section.
The calculated $XX$ binding energies $E_B(XX)$ are presented in fig. 4(c). We find that the biexcitons tend toward antibinding in small QDs under zero pressure. When the pressure increases, the calculated $E_B(XX)$ for the dots increases. The increase of binding energy tends to be sub-linear at high pressure, as observed typically for QD1–QD5 in the experiment shown in fig. 2(b). Especially, the biexciton antibinding-binding transition pressure and $XX$ binding energy of QD4 as functions of pressure are in good agreement with calculated values for QD-B.

In the calculations, we find that it is important to include many electron/hole energy levels to obtain the correct $XX$ binding energies using the CI method. However, the change of $XX$ binding energy as a function of pressure can be obtained only using the lowest energy conduction and valence bands (i.e., Hartree-Fock approximation), suggesting that the $XX$ binding energies do not change because of the change of correlation energies. Instead the binding energy changes are mainly due to changes in the direct Coulomb integrals between the lowest electron and hole states [16], as follows:

$$\Delta E_B(XX) \approx 2\Delta J_{eh} - \Delta J_{ee} - \Delta J_{hh}, \quad (2)$$

where $J_{ee}$, $J_{hh}$ and $J_{eh}$ are the direct electron-electron, hole-hole and electron-hole Coulomb integrals, respectively. As shown in fig. 4(d), whereas $J_{ee}$ and $J_{eh}$ increase rapidly with pressure, $J_{hh}$ is approximately flat. The solid purple line describes the changing biexciton binding energy calculated using eq. (2), which is consistent with the dashed purple line from the EPM calculations. To understand how the Coulomb integrals change under pressure, we compare the lowest electron and hole wave functions in fig. 4(f) at $P = 0$ GPa and 4.0 GPa for QD-A. We find that, whereas the hole wave function shape almost does not change under pressure, the electron becomes much more localized due to the band offset changes shown in fig. 4(b), which explains the change of Coulomb integrals under pressure. Both $J_{ee}$ and $J_{eh}$ show nonlinear dependence on the pressure, because of the competition between the kinetic energy and the confinement potential for the electron, resulting in a nonlinear dependence of the biexciton binding energy on pressure.

To examine FSS changes under hydrostatic pressure, FSS calculated as a function of pressure is shown in fig. 4(e), which displays an approximately linear increase in FSS below 3 GPa with the slope of 24 $\mu$eV/GPa close to the experimental result of 28 $\mu$eV/GPa for QD3 in fig. 2(c). It is interesting that FSS changes under hydrostatic pressure, while the latter does not change the QD symmetry. However, because the electron wave functions are more localized under pressure, an electron-hole wave function would have a larger effective overlap under pressure, which increases the electron-hole exchange energies, i.e., the dark-bright splitting $\Delta_{bd}$, also shown in fig. 4(e). It has been shown that FSS can be roughly estimated as $\sim 2\eta\Delta_{bd}$ [40], where $\eta$ is the HH-LH mixing parameter, and therefore, as $\Delta_{bd}$ increases, FSS increases, as clearly demonstrated in fig. 4(e).

**Conclusion.** – We have experimentally and theoretically investigated the effects of hydrostatic pressure on the exciton and biexciton transition energies as well as FSS in single InGaAs QDs. The observed exciton emission energy blue shift and FSS change are as large as up to 320 meV and 150 $\mu$eV, respectively, which are much greater than the values reached by other strain-adjusting techniques. Tuning the QD optical properties over such a larger spectral range yields great advantages for future QD applications, such as generating entangled photon pairs via a time-reordering scheme, two-photon interference of Hong-Ou-Mandel (HOM) experiments [41,42] using one photon emitted from tuned QDs and the other from the GaAs QDs, nitrogen vacancy (NV) center in diamond, and even some atomic systems.

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