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Pressure behaviour of self-assembled $In_{0.55}Al_{0.45}As/Al_{0.5}Ga_{0.5}As$ quantum dots with multi-modal distribution in size

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Abstract. The pressure behaviour of $In_{0.55}Al_{0.45}As/Al_{0.5}Ga_{0.5}As$ self-assembled quantum dots (QDs) has been studied at 15 K in the pressure range of 0–1.3 GPa. The atomic force microscopy image shows that the QDs have a multi-modal distribution in size. Three emission peaks were observed in the photoluminescence (PL) spectra, corresponding to the different QD families. The measured pressure coefficients are 82, 94 and 98 meV GPa^{-1} for QDs with average lateral size of 26, 52 and 62 nm, respectively. The pressure coefficient of small QDs is about 17% smaller than that of bulk $In_{0.55}Al_{0.45}As$. An envelope-function calculation was used to analyse the effect of pressure-induced change of barrier height, effective mass and dot size on the pressure coefficients of QDs. The Γ–X state mixing was also included in the evaluation of the reduction of the pressure coefficients. The results indicate that both the pressure-induced increase of effective mass and Γ–X mixing respond to the decrease of pressure coefficients, and the Γ–X mixing is more important for small dots. The calculated Γ–X interaction potentials are 15 and 10 meV for QDs with lateral size of 26 and 52 nm, respectively. A type-II alignment for the X conduction band is suggested according to the pressure dependence of the PL intensities. The valence-band offset was then estimated as 0.15 ± 0.02.

1. Introduction

Recent progress in epitaxy growth of coherent islands using Stranski–Krastanow (SK) mode allows the fabrication of high quality quantum dots (QDs). InAs/GaAs, $In_xGa_{1-x}As/GaAs$ QDs with emission in the infrared range have been studied extensively [1–3]. In recent years people have exploited $In_xAl_{1-x}As/Al_yGa_{1-y}As$ QDs with emission in the red visible range [4–6]. Investigation of the optical properties of QDs has revealed that the band structure of QDs differs drastically from those of bulk materials. The spectroscopic investigation under pressure may provide more information on the band structure especially for X-related electron states [7–9]. Iteskevich *et al* [10] have studied self-assembled InAs/GaAs quantum dots under applied pressure and found evidence for the Γ–X crossover and type-II band alignments. They have also found that the pressure coefficient of QDs is about 20% smaller than that of the Γ-band gap in bulk GaAs. Phillips *et al* [11] have presented a study of the pressure dependence of photoluminescence (PL) from $In_{0.5}Al_{0.5}As/Al_{0.25}Ga_{0.75}As$ QDs. A pressure induced energy

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level crossover and a narrowing of PL linewidth were found. However, there are few reports about the dot size dependence of pressure coefficients in the QDs.

In this work we report a study of the PL spectra of $In_{0.55}Al_{0.45}As/Al_{0.5}Ga_{0.5}As$ self-assembled QDs under hydrostatic pressure. The multi-modal distribution in size of the QDs has been confirmed by atomic force microscopy (AFM). We found that the pressure coefficients of QDs decrease with decreasing dot size, especially the pressure coefficient of a small QD is much smaller than that of bulk material. A theoretical calculation indicates that the change of effective mass under pressure and the Γ–X mixing are the main reasons for reduction of pressure coefficient and the Γ–X mixing is more important for small dots. A type-II band alignment was also obtained for the X band in the $In_{0.55}Al_{0.45}As/Al_{0.5}Ga_{0.5}As$ QD system.

2. Experiment

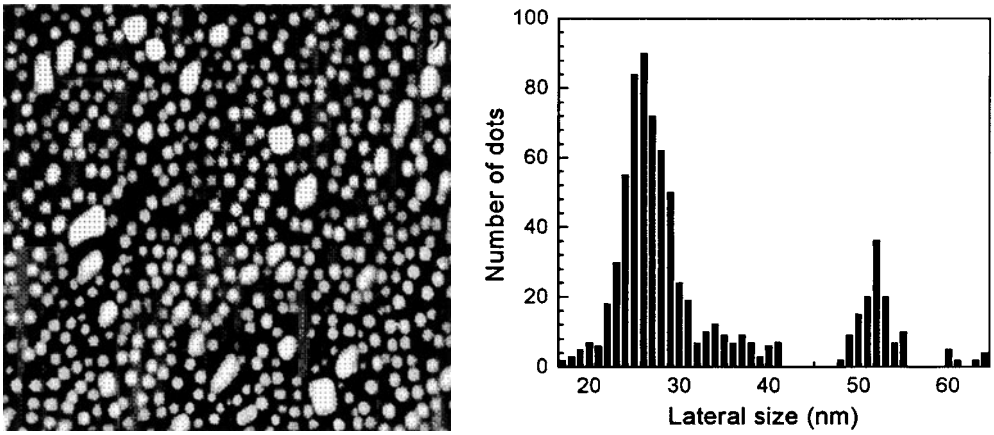
The samples were grown with SK mode on semi-insulating (001) GaAs substrate by a molecular beam epitaxy system. The sample structure consists of an undoped GaAs buffer layer, an $In_{0.55}Al_{0.45}As$ QDs layer sandwiched in two thick $Al_{0.5}Ga_{0.5}As$ cladding layers and a GaAs cap layer. The nominal thickness of $In_{0.55}Al_{0.45}As$ layer is ten monolayers. The formation of three-dimensional island was monitored by the reflection high-energy electron diffraction (RHEED) pattern. The details of the sample growth have been published elsewhere [12]. A Nanoscope III AFM was used to measure the lateral size distribution of QDs on an uncapped sample grown at the same condition.

For pressure experiments the sample was mechanically thinned from the backside to about 20 μm and then cut into pieces of about 100×100 μm^2 in size. The hydrostatic pressure was obtained by using a diamond anvil cell (DAC) with condensed argon as the pressure-transmitting medium. The DAC was put in an APD closed-cycle cryogenic refrigeration system for low temperature PL measurements. The pressure was monitored by the red shift of the ruby lines. The PL spectra were measured at 15 K by using the 488 nm line of an Ar+ laser as the source of excitation. The emitted light was dispersed by a JY-HRD1 double grating monochromator and detected by a GaAs photomultiplier.

3. Results

Figure 1(a) shows the AFM image of the uncapped sample. The statistical distribution of the lateral size of QDs obtained from the image is shown in figure 1(b). There are two main families of QDs. The average lateral sizes are about 26 ± 5 and 52 ± 3 nm, and the densities are about 5.9×10^{10} and 1.4×10^{10} cm^{-2}, respectively.

The PL spectra measured at 15 K and at different pressures are shown in figure 2. At atmospheric pressure two main peaks D1 and D2 are observed, which are attributed to the exciton transition in small and medium QD families as discussed in [12]. The weak band D3 at the lower energy probably comes from some larger QDs with the average lateral size of about 62 nm (see figure 1(b)). With increasing pressure all the peaks shift to higher energy. At the same time, the PL intensity of peak D1 decreases gradually and the peak D3 becomes more and more clear. At about 0.9 GPa peak D1 disappears. D2 and D3 become the dominant peaks. In figure 3 we summarize the pressure dependence of the PL peak energies of three peaks. The solid lines in figure 3 represent the results of least-squares fits to the experimental data using linear relation. The corresponding pressure coefficients are listed in table 1.



(a) (b)

Figure 1. (a) AFM image of the uncapped $In_{0.55}Al_{0.45}As/Al_{0.5}Ga_{0.5}As$ QD sample; (b) the statistical distribution of the lateral size of QDs obtained from $1 \times 1\ \mu m^2$ area.

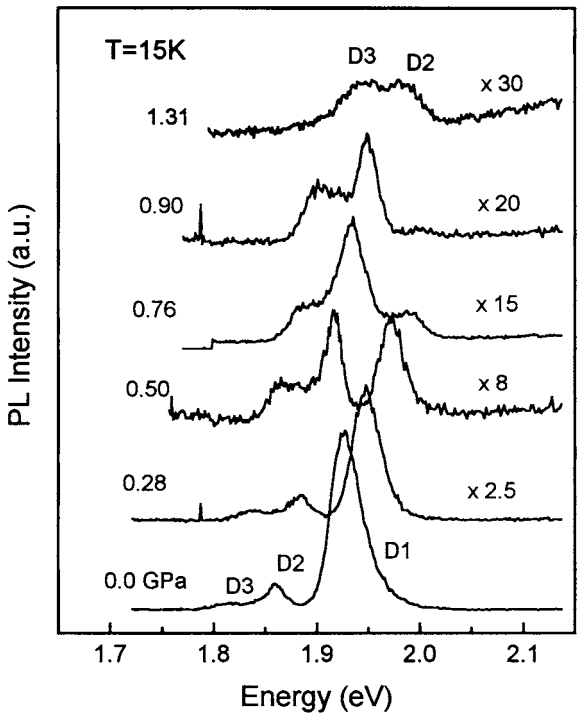


Figure 2. Photoluminescence spectra of $In_{0.55}$ $Al_{0.45}As$ QDs with multi-modal distribution under various pressures.

4. Discussion

4.1. Valence band offset

The pressure dependence of the PL intensities of three peaks is shown in the inset of figure 3. It can be seen that the PL intensity of peak D1 decreases significantly at the pressure about

Table 1. Energies and pressure coefficients of PL peaks in an $In_{0.55}Al_{0.45}As/Al_{0.5}Ga_{0.5}As$ QD sample obtained from least-squares fits to the experimental data using $E(p) = E(0) + a_1 P$. The calculated pressure coefficients a_{PL} by envelope-function approximation and the interaction potential V of Γ–X mixing are also given.

Peaks	$E(0)$ (eV)	a_1 (meV GPa^{-1})	a_{PL} (meV GPa^{-1})	V (meV)
D1	1.928	82 ± 2	95.4	15 ± 1
D2	1.862	94 ± 2	95.7	10 ± 2
D3	1.819	98 ± 2	96.0	

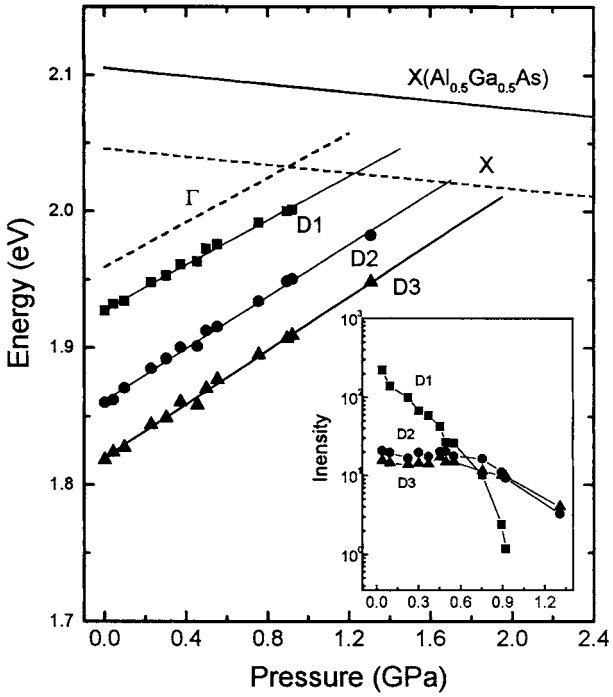


Figure 3. The pressure dependence of the energy position of three PL peaks. The solid lines correspond to the results of least-squares fits. The pressure dependence of the X-valley in $Al_{0.5}Ga_{0.5}As$ is also shown with a straight line. The band gaps corresponding to peak D1 and the X-related level are drawn by dashed lines. The inset shows PL intensity as a function of pressure.

0.9 GPa, This behaviour indicates that the Γ-related conduction band in small QDs crosses with an X-related level. Since peak D1 corresponds to the exciton transition, the Γ-related band gap in QDs can be obtained by adding an exciton binding energy (30 meV [13]) to the PL peak energy. Its pressure dependence is shown in figure 3 by a dashed line. The pressure dependence of the X-band gap of bulk $Al_{0.5}Ga_{0.5}As$ [14, 15] is also drawn in figure 3 by a dotted line. Then we can draw the X-related transition, which follows the pressure dependence of the X-band of bulk $Al_{0.5}Ga_{0.5}As$ and crosses with the Γ-related gap in small QDs at 0.9 GPa, as shown in figure 3. It is clear that the energy position of the X-related transition is lower than the indirect gap of bulk $Al_{0.5}Ga_{0.5}As$, and also than the transition energy between X-valley and heavy-hole states in small $In_{0.55}$ $Al_{0.45}As$ QDs. This suggests that the X-related transition corresponds to the transition from the X-valley of $Al_{0.5}Ga_{0.5}As$ to the heavy hole in $In_{0.55}$

$Al_{0.45}As$ QDs and that the X conduction bands have a type-II band alignment in our sample. The valence-band offset is then determined from:

$$\Delta E_V = E_g^X(Al_{0.5}Ga_{0.5}As) - E^X + E_{1h} \tag{1}$$

where $E_g^X(Al_{0.5}Ga_{0.5}As)$ is the indirect gap of $Al_{0.5}Ga_{0.5}As$. E^X is the transition energy of the X-related transition at atmospheric pressure. E_{1h} is the confinement energy of the heavy hole, about 40 meV for the small QDs according to the model calculation later. From equation (1) we obtain $\Delta E_V = 95 \pm 10$ meV. The corresponding valence-band offset of the $In_{0.55}Al_{0.45}As/Al_{0.5}Ga_{0.5}As$ system is about 0.15 ± 0.02.

There have been only a few reports on the band offset in the $In_yAl_{1-y}As/Al_xGa_{1-x}As$ system. Wang *et al* [13] have calculated the valence-band offset as 0.4 for the $In_{0.55}Al_{0.45}As/Al_{0.35}Ga_{0.65}As$ QD system. It is somewhat larger than our results. The real observation of the X-related emission in the QD system will improve the experimentally determined band offset further.

4.2. Size dependence of the pressure coefficients

The pressure coefficients are 82, 94 and 98 meV GPa^{-1} for peaks D1, D2 and D3, respectively. These are smaller than that of $In_{0.55}$ $Al_{0.45}As$ bulk materials, 99.3 meV GPa^{-1} as interpolated from the values of InAs and AlAs [16]. Furthermore, the pressure coefficients decrease with decreasing dot size. Such a phenomenon is similar to that in $GaAs/Al_xGa_{1-x}As$ quantum wells [17]. Lefebvre *et al* [18] have calculated the well-width dependence of the pressure coefficients for $GaAs/Al_xGa_{1-x}As$ quantum wells taking into account the pressure-induced modifications of the well width, barrier height and effective masses. We performed a similar calculation for the size dependence of the pressure coefficients in $In_{0.55}$ $Al_{0.45}As$ QDs. The PL peak energy of a QD can be expressed as:

$$E_{PL} = E_g + E_{1e} + E_{1h} - E_{ex} \tag{2}$$

where E_g is the band gap of the $In_{0.55}$ $Al_{0.45}As$, E_{1e} and E_{1h} are the confined energies of electrons and heavy holes, respectively and E_{ex} is the exciton binding energy. We calculated the confined energies E_{1e} and E_{1h} using the envelope function approach, assuming a flat quantum box model with the growth direction along the Z-axis. The lateral size of the box is determined from the AFM image. The size in the direction Z is obtained through fitting the calculated PL peak energy to the experimental data at ambient pressure. In the fitting we have used the band offset obtained above and the exciton binding energy of 30 meV.

The pressure-induced change of E_{ex} can be neglected as argued by Lefebvre *et al* [18, 19]. Both E_{1h} and its pressure modification are small due to the large effective mass of the heavy holes. So the main factor influencing the pressure coefficients in QDs is the pressure-induced change of E_{1e}. By using the envelope function approximation we calculated the electron confined energy at various pressures taking into account the changes of dot size, barrier height and effective mass induced by pressure. The pressure dependence of dot size $L_i(p)$ ($i = x$, y and z) can be obtained from the elasticity theory:

$$L_i(p) = L_i(0)[1 - (S_{11} + 2S_{12})p] \tag{3}$$

where S_{11} and S_{12} are the elastic constants of the $In_{0.55}$ $Al_{0.45}As$ bulk material. $L_i(0)$ is the dot size at ambient pressure determined above. The change of effective mass with pressure $m(p)$ is taken from Kane's three-band model [20]:

$$\frac{m(p)}{m(0)} = \frac{E_g + ap + E_{1e} + E_{1l}}{E_g} \frac{E_g + ap + E_{1e} + E_{1l} + \Delta}{E_g + \Delta} \frac{2\Delta + 3E_g}{2\Delta + 3(E_g + ap + E_{1e} + E_{1l})} \tag{4}$$

where E_g is the band gap of $In_{0.55}Al_{0.45}As$, a is its pressure coefficient. Δ is the spin–orbit splitting energy. E_{1l} is the confined energy of light holes. The pressure-induced change of the barrier height $V(p)$ can be expressed as

$$V(p) = V(0) + (a_B - a_D)p \tag{5}$$

where a_B and a_D are pressure coefficients of the $Al_{0.5}Ga_{0.5}As$ barrier and $In_{0.55}$ $Al_{0.45}As$ material.

The pressure coefficient of $Al_{0.5}Ga_{0.5}As$ (102 meV GPa^{-1} [21]) is larger than that of $In_{0.55}$ $Al_{0.45}As$ (99.3 meV GPa^{-1} [16]). Therefore the barrier height will increase with increasing pressure, which results in the increase of the confined energy of electrons. The decrease of the dot size with pressure will also cause the increase of the confined energy. Only the increase of the effective mass responds to the decrease of the confined energy in QDs.

After numerical calculation of the confined energy for electrons at $p = 0$ and 1 GPa, we obtained the pressure coefficients of the PL peak energy from

$$a_{PL} = a_D + \frac{E_{1e}(p) - E_{1e}(0)}{p}. \tag{6}$$

The calculated pressure coefficients for different QDs are listed in table 1. As expected, the pressure coefficients of QDs are smaller than that of bulk material and decrease with decreasing dot size. But the calculated reduction is much smaller than the experimental results. Especially, the measured pressure coefficient for small dots is about 17% smaller than that of bulk $In_{0.55}$ $Al_{0.45}As$, while the calculated reduction is only about 4%. This implies that there may exist other effects, which also respond to the decrease of pressure coefficients. Itskevich *et al* [10] have suggested that the contribution of the X-valley to the wavefunction of the confined electron states would also cause the decrease of the pressure coefficients. This will be discussed in the next section.

Though only a simple model was used in our calculation, the calculated pressure coefficients are still reliable since we obtain the confined energy for electrons at normal pressure through a fitting process. Furthermore, the change of pressure coefficients for QDs with different size is related to $E_{1e}(p) - (E_{1e}(0)$. This will reduce further the approximation in the calculation of E_{1e}.

4.3. Γ–*X mixing*

Now we turn to discuss the contribution of Γ–X mixing to the pressure behaviour of QDs with different size. As show above, the Γ-related conduction band in QDs will cross with the X-valley of $Al_{0.5}Ga_{0.5}As$ under pressure. The interaction or mixing between the Γ and X conduction band states is expected to occur near the Γ–X crossover due to the potential step at the interfaces. In first order perturbation theory the energy position of two interacting states can be expressed as

$$E_\pm = \tfrac{1}{2}\{(E_\Gamma + E_X) \pm [(E_\Gamma - E_X)^2 + 4V^2]^{1/2}\} \tag{7}$$

where E_Γ and E_X are the energies of the Γ and X states without interaction, and V is the interaction potential. If we write the pressure dependence of E_Γ and E_X:

$$E_\Gamma = E(P_c) + a_\Gamma(P - P_c) \tag{8}$$

$$E_X = E(P_c) + a_X(P - P_c) \tag{9}$$

the pressure dependence of $E_\pm$ will be:

$$E_\pm(P) = E(P_c) + \tfrac{1}{2}(a_\Gamma + a_X)(P - P_c) \pm \{[\tfrac{1}{2}(a_\Gamma - a_X)(P - P_c)]^2 + V^2\}^{1/2} \tag{10}$$

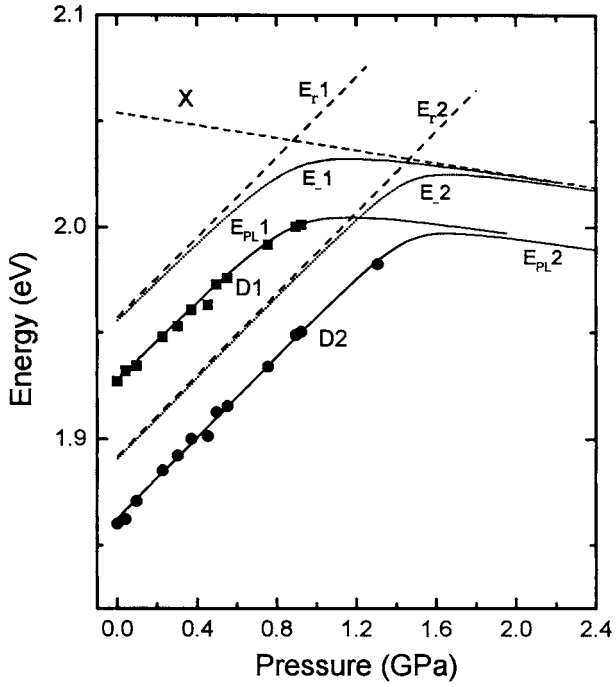


Figure 4. Calculated results for the Γ–X mixing. The solid and dotted lines represent the calculated PL peak energies, E_{PL}, and the corresponding band gaps, E_-, for the Γ–X coupled states. The experimental data of peaks D1 and D2 are shown by the full symbols. The energies of the Γ and X states without interaction, E_Γ and E_X, are also drawn with dashed lines for comparison.

where P_C and $E(P_C)$ are pressure and energy at the crossover point, a_Γ and a_X are the pressure coefficients of E_Γ and E_X, respectively. Since the observed PL peaks correspond to the recombination of excitons, the PL peak position is

$$E_{PL}(P) = E_-(P) - R(P) \tag{11}$$

where $R(P)$ is the exciton binding energy. By using equations (10) and (11) we calculated the pressure dependence of the PL peaks of QDs. In the calculation of the pressure coefficients obtained in section 4.2, a_{PL} was used for a_Γ. The pressure coefficient of the X-valley in bulk $Al_{0.5}Ga_{0.5}As$ was used for a_X. The P_C for peak D1 was taken as 0.9 GPa according to the experimental results. The P_C for peak D2 was obtained from the crossover point of the D2-related band gap and the X-related level. The exciton binding energy was still taken as 30 meV and its pressure dependence is neglected. The only fitting parameter is the interaction potential V. The calculated energy E_- (dotted line), E_{PL} (solid line) for two main QD families are presented in figure 4 together with the experimental data of peaks D1 and D2. For comparison we also draw in figure 4 (dashed line) the energies of the Γ- and X-states without interaction, E_Γ and E_X, as a function of pressure. The obtained interaction potentials V are listed in table 1. We do not fit peak D3 because of its weak intensity and weak Γ–X mixing in large dots. The calculated results agree well with the experimental data. This indicates that the Γ–X mixing can also cause the decrease of pressure coefficient especially for small QDs. On the other hand, the Γ–X mixing and increase of effective mass are nearly equivalent for dots D2. The interaction potentials for the quantum dots D1, D2 are 15 and 10 meV, which are comparable in magnitude to the result of Li *et al* [8]. The larger interaction potential in smaller QDs suggests that the Γ–X mixing is more pronounced in the QDs with stronger localization.

5. Conclusions

We have investigated the low temperature PL of $In_{0.55}Al_{0.45}As/Al_{0.5}Ga_{0.5}As$ self-assembled QDs with multi-modal distribution under hydrostatic pressure. Two main peaks and one weak peak were observed, which are attributed to the emissions from different dot families. With increasing pressure all the peaks shift to high energy. The pressure coefficients are smaller than that of bulk $In_{0.55}$ $Al_{0.45}As$ material and decrease with decreasing dot size. The PL intensity of peak D1 decreases rapidly at about 0.9 GPa, indicating the crossover between the corresponding band and an X-related level. A type-II configuration for X-valleys is suggested according to the energy position of the X-related level. The valence-band offset of 0.15 ± 0.02 was obtained. An envelope-function model was used to calculate the pressure coefficients for QDs with different size, taking into account pressure-induced modifications of barrier height, effective masses and dot size. The obtained results indicate that the pressure-induced increase of effective mass is not enough to explain the 17% difference between the pressure coefficients of peak D1 and bulk $In_{0.55}Al_{0.45}As$. After considering the contribution of Γ–X mixing in QDs, the calculated results then well describe the experimental pressure behaviour. The interaction potentials for Γ–X mixing were estimated to be 15 and 10 meV for dot families D1 and D2, respectively.

Acknowledgment

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