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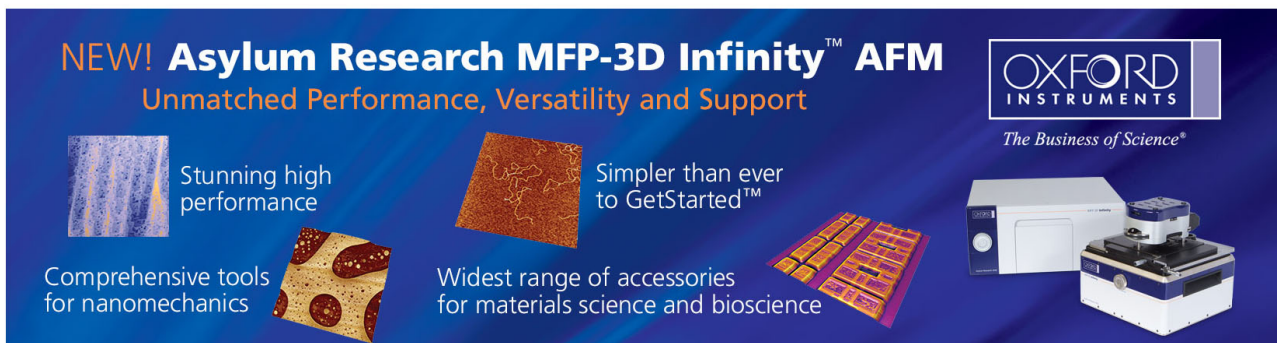
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## Bimodel onset strain relaxation in InAs quantum dots with an InGaAs capping layer

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Capping InAs quantum dots (QDs) with an InGaAs layer allows strain relaxation to induce a low-energy electron state below a set of fine dot family states, which is consistent with photoluminescence (PL) spectra. The evolution of InAs thickness suggests a bimodal onset relaxation, i.e., a fine dot family that is strain-relieved by indium outdiffusion from the QDs, as suggested by transmission electron microscopy, and a low-energy dot family that is strain relaxed by the generation of lattice misfits. The indium outdiffusion can explain an abnormal PL blueshift in 70 meV in the fine dot family at onset of strain relaxation. © 2010 American Institute of Physics. [doi:10.1063/1.3483757]

Understanding the onset of strain relaxation in InAs self-assembled quantum dots (QDs) (Refs. 1–16) is necessary for designing QD devices.<sup>13,14</sup> Strain relaxation in the QDs is accommodated by the generation of threading dislocations in the top GaAs layer and lattice misfits near the front QDs interface,<sup>17</sup> suggesting an occurrence of relaxation toward the top GaAs layer. Growing an InGaAs capping layer on top of the QDs leads to strain relaxation by the generation of the lattice misfits in the bottom GaAs layer near the QDs while the top GaAs layer is dislocation-free.<sup>18</sup> Thus, the InGaAs capping layer can effectively relieve strain in the top GaAs layer and relaxation occurs toward the bottom GaAs layer with an accompanied photoluminescence (PL) blueshift in about 70 meV.<sup>19</sup> This blueshift contradicts an effect of compressive strain reduction in the QDs, which is expected to produce a redshift. This observation suggests a more complicated relaxation than expected. Detailed strain relaxation mechanism has not yet thoroughly established. Therefore, this work describes detailed carrier confinement and PL spectra of the InAs QDs with the InAs deposition thickness over a critical relaxation thickness of 3 ML to understand how the strain is relaxed in the InAs QDs.

The InAs QDs were grown by solid source molecular beam epitaxy. On top of a  $n^+$ -GaAs (100) substrate, a 0.3  $\mu\text{m}$  thick Si-doped GaAs ( $\sim 7 \times 10^{16} \text{ cm}^{-3}$ ) barrier layer, an InAs layer with deposition thickness from 2 to 3.5 ML was deposited at 490 °C (at a rate of 0.26 Å/s) to form the QDs. Following the growth of the QDs layer, a 60 Å  $\text{In}_{0.15}\text{Ga}_{0.85}\text{As}$  capping layer and a 0.2  $\mu\text{m}$  thick Si-doped GaAs barrier layer were grown to terminate the growth. The QDs were relaxed by increasing the InAs deposition thickness slightly above 3 ML, as evident from the induction of lattice misfits<sup>18</sup> and a PL blueshift about 70 meV.<sup>19</sup> The QD sheet density was estimated at about  $3 \times 10^{10} \text{ cm}^{-2}$ . For capacitance-voltage (C-V) profiling, Schottky diodes were realized by evaporating Al with an area of  $5 \times 10^{-3} \text{ cm}^2$ .

Figure 1 show the simulated and experimental 90 K C-V spectra (at a low-frequency 3 kHz and a high-frequency 100 kHz) and converted experimental electron distribution of a

relaxed InAs QDs diode with the InAs deposition thickness of 3.3 ML. Two major C plateaus are visible at the QD layer: a shallow one from  $-1.5$  to  $-2.2$  V, as attributed to a state related to the InGaAs capping layer, and a deep one from

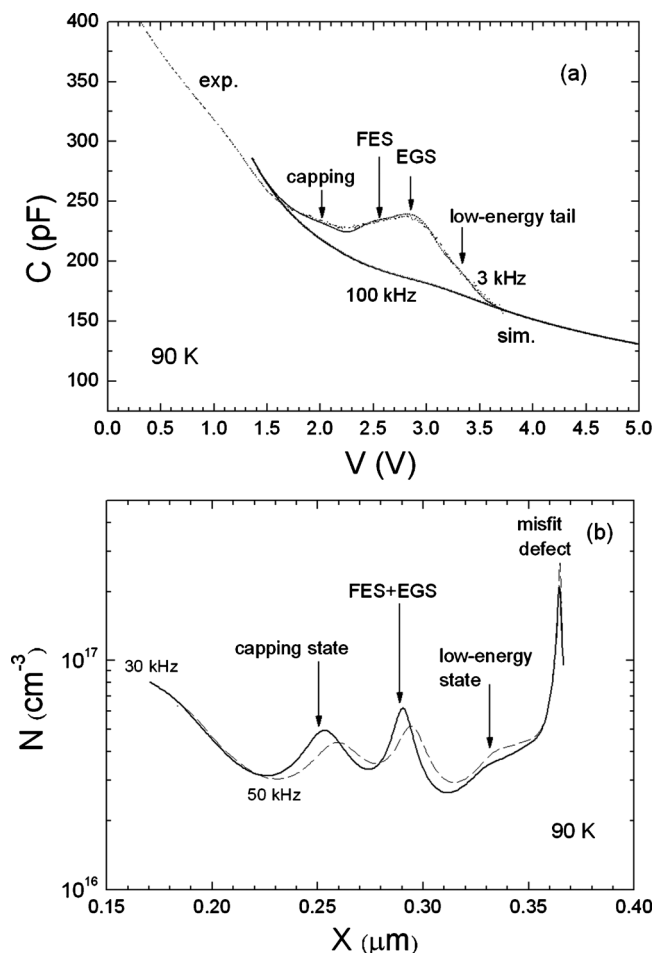


FIG. 1. (a) 90 K experimental and simulated C-V spectra (at a low-frequency of 3 kHz and a high-frequency of 100 kHz) and (b) converted electron distribution of a relaxed InAs QDs diode with the InAs thickness of 3.3 ML. The spectra display a shallow capping-layer state, a fine dot family of the FES and EGS, a low-energy tail and a misfit-related defect state.

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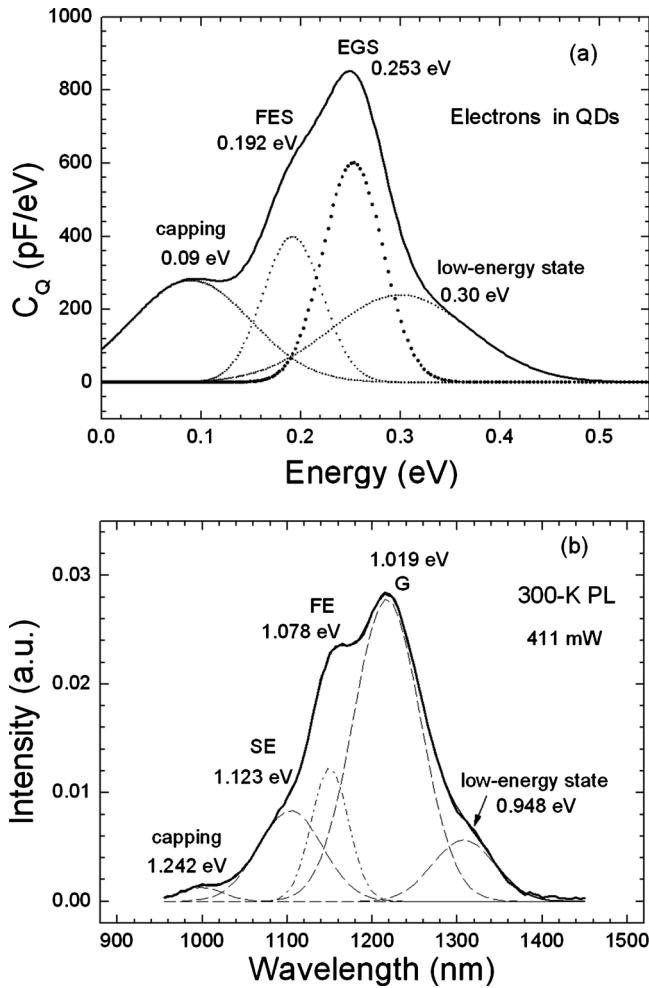


FIG. 2. (a) Gaussian electron distributions of a shallow state, FES, EGS, and a long-tail state used for the simulation in Fig. 1(a). This electron distribution is correlated with the (b) 300 K PL spectra which show a deconvoluted capping-layer emission, the second-excited, first-excited, ground transitions, and a low-energy transition.

–2.2 to –3.5 V, as attributed to a fine dot family consisting of electron ground state (EGS) and first excited state (FES), and an additional low-energy tail. Following these two major plateaus is another carrier confinement related to the misfit-related defect state at 0.35 eV.<sup>18</sup> The

experimental carrier distribution in Fig. 1(b) clearly reveals the capping-layer state, the fine dot family, the low-energy tail and the misfit defect state. The carrier confinement is analyzed by a C-V simulation based on a similar treatment,<sup>20,21</sup> where the low-frequency capacitance is expressed by  $C_L = dQ_1 + dQ_2 / dV = dQ_1 / dV + dQ_2 / dV = dV_1 / dV \epsilon / L_1 + dQ_2 / dV_1 dV_1 / dV = (C_1 + C_Q) C_2 / (C_1 + C_Q) + C_2$ , where  $C_1 = \epsilon / L_1$  and  $C_2 = \epsilon / L_2$  are the geometric capacitance per unit area across  $L_1$  (distance between the QD layer and the edge of the total depletion width) and  $L_2$  (the depth of the QD layer), and  $C_Q = dQ_2 / dV_1$  is the occupied density of states of the QDs. The confinement energy  $E$  (relative to the GaAs CB edge) of the probed QD electrons can be expressed by  $E = V_1 + \phi_n = (q/2\epsilon) N_D' L_1^2 + (kT/q) \ln(N_C / N_D)$ , where  $N_C$  is the effective density of states in the GaAs CB. Additionally,  $L_1$  is related to the reverse voltage  $V_R$  by  $V_R = V_1 + V_2 - V_{bi}$ , where  $V_1 = (q/2\epsilon) N_D' L_1^2$ ,  $V_2 = (q/\epsilon) N_D' L_1 L_2 + (q/2\epsilon) N_D' L_2^2 - (L_2/\epsilon A) \int_{-\infty}^E C_Q dE$  and  $V_{bi}$  denotes the Schottky barrier height of the GaAs and  $A$  is the area of the diode. According to Fig. 1(a), the simulated results (solid curves) correlate well with the experimental data (dotted line) by using Gaussian distributions  $C_Q(E)$  of a shallow state peaked at 0.09 eV, a FES at 0.192 eV, a EGS at 0.253, and a long-tail state at 0.30 eV, as shown in Fig. 2(a). Although a better correlation can be achieved, this work neglects the second-excited state to more clearly visualize other contributions. The fitted  $E = 0.253$  eV for the EGS and the 50 K PL ground emission at 1.074 eV (1.019 eV at 300 K) give  $E$  of a hole ground state (HGS) of  $1.50 - 1.074 - 0.253 = 0.173$  eV and a ratio of EGS to HGS of 0.59 to 0.41, a value close to a previously reported 0.61:0.39.<sup>21</sup>

The fitted carrier distribution in Fig. 2(a) has a one-to-one correlation with the deconvoluted PL spectra (at 300 K) in Fig. 2(b), showing a 1.242 eV shallow transition, transitions from a fine dot family including the second excited (SE), first excited (FE), and ground (G) states, and an additional 0.948 eV low-energy transition. The FES and EGS have a similar full-width-half-maximum broadness of about 60 meV as the PL broadness of the FE and ground transitions. According to our results, the 1.242 eV shallow transition displays a significant redshift with an N incorporation into the InGaAs capping layer, and thus is attributed to a

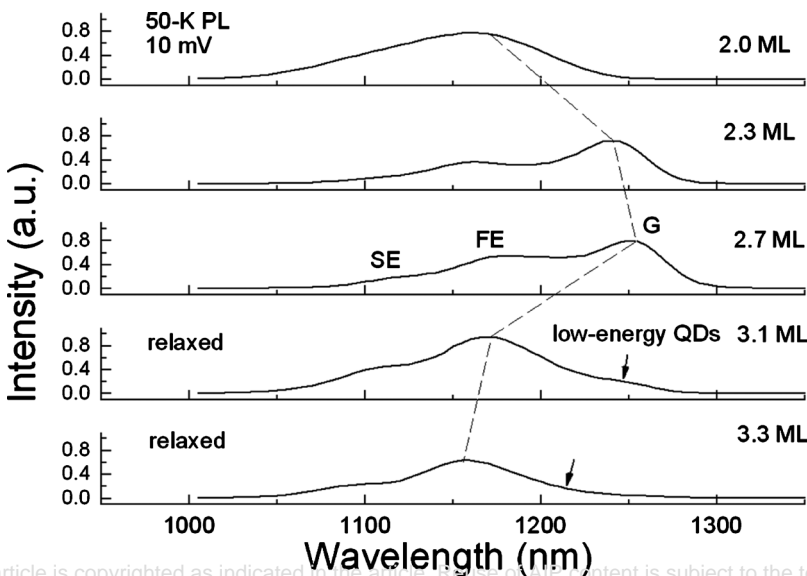


FIG. 3. Evolution of the 50 K PL spectra of the QDs with increasing InAs coverage, under the excitation of 10 mW. For guiding eyes, a dashed line is drawn on the ground transition in the fine dot family. The y-scale is the same for all the spectra.

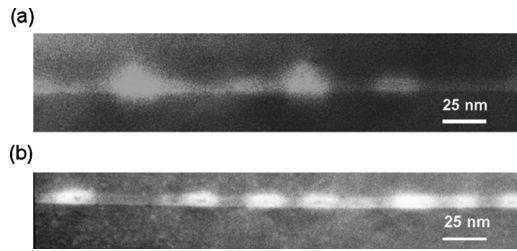


FIG. 4. TEM images of (a) the relaxed 3.1 ML coverage and (b) nonrelaxed 2.7 ML coverage. In the relaxed QDs, bright color emanating out of some QDs can be seen, suggesting indium outdiffusion from the QDs.

state belonging to the capping layer. The above results indicate that onset of strain relaxation does not degrade the feature of the QDs: it displays a fine dot family similar to that observed in nonrelaxed QDs. Onset of strain relaxation mainly induces a low-energy state.

To illustrate the evolution of strain relaxation, Fig. 3 shows the 50 K PL spectra of the InAs QDs with an increasing InAs coverage, under the excitation of 10 mW. A dashed line is drawn to guide the ground emissions from the fine dot family. At the initial formation of the QDs at 2.0 ML, the PL spectra are broad due to large dot-size dispersion at the initial stage. Increasing the InAs coverage increases the dot size and causes a redshift in the ground emission to a maximum of  $\sim 1250$  nm ( $\sim 1300$  nm at 300 K) at 2.7 ML. The DLTS measurements reveal no defect signals, indicating coherent QD formation. However, when the InAs coverage is increased over a critical thickness of 3 ML to 3.1 ML, the fine dot family still maintains but undergoes an abnormal blueshift in about 70 meV, in addition to the emergence of a low-energy state (at  $\sim 1250$  nm). When the temperature is lowered (from 300 to 50 K), the low-energy state increases its PL intensity by only a factor of 2, relative to an improvement of a factor of ten in the fine dot family. Because PL can be degraded by defects through which photogenerated carriers are recombined, the low-energy state is ascribed to a degraded dot family due to the misfit defect state at 0.35 eV.<sup>18</sup> This finding suggests a nonuniform strain relaxation: a family of dislocated dots and a fine dot family less affected by misfit defects, as evaluated from its fine, well-resolved emissions that are stronger and sharper than that of the 2 ML nonrelaxed dots.

The blueshift in 70 meV observed in the fine dot family contradicts to the strain relaxation in the QDs because the reduction in a compressive strain in the QDs is expected to increase the lattice constant perpendicular to the growth direction and extend the emission wavelength. Thus, this blueshift cannot be explained by the generation of lattice misfits. The TEM image of the relaxed 3.1 ML coverage, as shown in Fig. 4(a), leads us to attribute the blueshift to indium outdiffusion from the QDs, in which apparent brightness around the tops of some dots and emanated into the top GaAs layer can be seen, in relation to the TEM image of the nonrelaxed 2.7 ML QDs in Fig. 4(b). Previous studies have observed strain related indium segregation.<sup>22,23</sup> This indium outdiffusion can cause a PL blueshift as postgrowth thermal annealing. Thus, the built-in strain in the fine dot family is

mainly relieved by the migration of indium adatoms to the top GaAs layer, leading to a reduction in the indium content in the dots and the correlated 70 meV blueshift. According to the results of Fourier transformed TEM images, the dots without the brightness in Fig. 4(a) display significant lattice misfits inside and near the dots. Thus, we attribute these dots to the family of the dislocated dots. A compressive strain reduction in the QDs can produce a low-energy transition. This bimodal relaxation is probably related to the InGaAs capping layer. Without the capping layer, strain relaxation occurs when the QD deposition thickness is increased to 2.8 ML by the generation of threading dislocation in the top GaAs layer and lattice misfits near the QD top boundary, leading to degraded electronic and PL spectra of the QDs without the blueshift.<sup>17</sup>

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