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Relaxation-induced lattice misfits and their effects on the emission properties of InAs quantum dots

J F Chen¹, Y Z Wang¹, C H Chiang¹, R S Hsiao¹, Y H Wu², L Chang³, J S Wang³, T W Chi⁴ and J Y Chi⁴

¹ Department of Electrophysics, National Chiao Tung University, Hsinchu, Taiwan, Republic of China
² Department of Materials Science and Engineering, National Chiao Tung University, Hsinchu, Taiwan, Republic of China
³ Department of Physics, Chung Yuan Christian University, Chung-Li, Taiwan, Republic of China
⁴ Industrial Technology Research Institute (OES/ITRI), Hsinchu, Taiwan, Republic of China

E-mail: jfchen@cc.nctu.edu.tw

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Abstract
Strain relaxation in InAs/InGaAs quantum dots (QDs) is shown to introduce misfits in the QD and neighboring GaAs bottom layer. A capacitance–voltage profiling shows an electron accumulation peak at the QD with a long emission time, followed by additional carrier depletion caused by the misfits in the GaAs bottom layer. The emission-time increase is explained by the suppression of tunneling for the QD excited states due to the additional carrier depletion. As a result, electrons are thermally activated from the QD states to the GaAs conduction band, consistent with observed emission energies of 0.160 and 0.068 eV which are comparable to the confinement energies of the QD electron ground and first-excited states, respectively, relative to the GaAs conduction band. This is in contrast to non-relaxed samples in which emission energy of 60 meV is observed, corresponding to the emission from the QD ground state to the first-excited state.

1. Introduction
Recently, InAs/GaAs self-assembled quantum dots (QDs) [1–5] have attracted considerable attention because of their promising technological applications [6–8] and for scientific studies [9–13]. One of the important issues is experimentally determining the electronic band structure of the QD [9–14]. Kapteyn et al [9] have proposed a two-step emission process for electrons in the QD: a thermal activation from the QD ground state to the first-excited state and then tunneling to the GaAs conduction band. This suggests a strong tunneling for electrons emitting from the QD excited states. Since the tunneling probability can be affected by varying the depletion width, introducing additional carrier depletion may suppress the tunneling process and enable the observation of the thermal emission from the QD ground state to the GaAs conduction band. Coherent QDs can be formed by partial strain relaxation. However, when the InAs thickness is increased beyond the critical thickness (∼3 ML), the strain is relaxed by generating misfit dislocations [13]. Uchida et al [15] have observed a perfect confinement of misfit dislocations at the relaxation interface in InGaAs/GaAs heterostructures. In previous work [14], misfit dislocations were shown to be electron-trapping centers. The misfits in the bottom GaAs layer may cause carrier depletion and suppress the tunneling probability. This may significantly modify the emission properties of the QD. Therefore, in this work, the relaxation-induced misfit dislocations and their effects on the electron emission in InAs QDs are investigated by transmission electron microscopy (TEM), capacitance–voltage (C–V) profiling and deep-level-transient spectroscopy (DLTS).

The samples studied are InAs QDs capped with an InGaAs layer. With this capping layer, relaxation-induced misfit
dislocations are found in the QD and neighboring GaAs bottom layer. The carrier depletion caused by the misfit dislocations in the neighboring GaAs bottom layer can suppress the electron emission from the QD excited states, leading to a longer emission time and larger emission energy. Evidence of this tunneling suppression is provided by another QD sample without an InGaAs capping layer. Strain relaxation does not produce misfit dislocations in the bottom GaAs layer. Without additional carrier depletion behind the QD, the emission time remains very short.

2. Experiments

The QD structures were grown on n⁺-GaAs(100) substrates by solid source molecular beam epitaxy in a Riber machine. On top of a 0.3 μm thick Si-doped GaAs (6–10 × 10¹⁰ cm⁻²) barrier layer, an InAs layer with different thickness from 2 to 3.3 ML was deposited at 490 °C to form the QDs. Then the QDs were capped with a 60 Å In₀.₁₅Ga₀.₈₅As layer and a 0.2 μm thick Si-doped GaAs (6–10 × 10¹⁰ cm⁻²) layer to terminate the growth. Detailed growth conditions can be found elsewhere [16]. A typical QD sheet density about 3 × 10¹⁰ cm⁻² was observed by atomic force microscopy (AFM). For C–V profiling, Schottky diodes were realized by evaporating Al on the samples. The apparent-carrier concentration is obtained by converting the C–V curve using the depletion-layer approximation: 

\[ N(w) = \frac{A_{sample} \cdot C}{d^2}, \]

where \( W \) is the width of the space-charge region and \( A \) is the contact area. Photoluminescence (PL) measurements were carried out using a double-frequency yttrium–aluminum–garnet (YAG): Nd laser at 532 nm.

3. Measurement and results

3.1. TEM characterization of misfit dislocations

In contrast to there being no dislocations in the non-relaxed samples, misfit dislocations are observed in the relaxed InAs/InGaAs QDs samples. Figure 1(a) shows a large-scale cross-sectional TEM picture of a QD sample with a 3.3 ML thick InAs layer. The QD is relaxed since the InAs thickness exceeds the critical thickness of ∼3 ML [13]. A line of QDs is visible. No threading dislocations are observed in the top GaAs layer. Figure 1(b) shows the TEM picture around a dot whose contrast is similar to that of a non-relaxed dot. The shape of the dot looks more like a trapezium with a height ∼10 nm and a base width ∼20 nm. Figure 1(c) shows a high-resolution TEM picture of a typical dot (dashed ellipse). As a guide to the eyes, the wetting layer is indicated by a line. Figure 1(d) shows the Fourier transformed image of figure 1(c). The area around the GaAs bottom layer near the QD is emphasized in figure 1(e) for clarity. Several (about ten) dislocations, as indicated by loops, can be seen in the QD. No dislocations are found in the intervening GaAs region between adjacent QDs. About eight dislocations are observed in the bottom GaAs layer near the dot: two in each of the two small loops on the sides, and four in the large middle loop. From a dot density of ∼3 × 10¹⁰ cm⁻² observed by AFM, the total density of the dislocations is ∼5.4 × 10¹¹ cm⁻² on the average. These dislocations do not propagate into the GaAs layers but are confined near the QD lower interface. Therefore, there are misfit dislocations induced by strain relaxation, rather than threading dislocations generated from the sample surface or substrate through a gliding process. These misfit dislocations bend toward the interface. It should be noted that, besides these misfit dislocations, the sample reveals no other defects. Hence, relaxation-induced misfit dislocations are confined in the QD and in the neighboring GaAs bottom layer. (e) Part of figure (d), showing more clearly the misfit dislocations in the large middle loop in (d).

Figure 1. (a) Cross-sectional TEM picture of the 3.3 ML InAs/InGaAs QDs sample, showing a line of QDs and no threading dislocations in the top GaAs layer. (b) The TEM image of a typical QD, showing a height ∼10 nm and a base width ∼20 nm. (c) The HRTEM picture of a dot (dashed ellipse). As a guide to the eyes, the wetting layer is indicated by a line. (d) The corresponding Fourier transformed image, showing a number of misfit dislocations in the QD and in the neighboring GaAs bottom layer. (e) Part of figure (d), showing more clearly the misfit dislocations in the large middle loop in (d).
The characteristic of a Debye-length effect in a quantum structure. The intensity of the peak increases with decreasing temperature, is defined as the distance from the sample surface. The should be noted that the carrier peak at by traps presumably associated with the misfits in the QD. It relaxed QD. Hence, some electrons in the QD are depleted suggests a smaller number of electrons accumulated in the and its converted carrier-accumulation peak in the QD region 

\[
\frac{1}{2} < \mu < 3
\]

Figures 2(a) and (b) show a C plateau (from \(-2\) to \(-3\) V) and its converted carrier-accumulation peak in the QD region (\(\sim 0.2\ \mu m\)), respectively. The \(x\)-coordinate in figure 2(b) is defined as the distance from the sample surface. The intensity of the peak increases with decreasing temperature, characteristic of a Debye-length effect in a quantum structure. The C plateau appears at nearly the same dc voltage as in a non-relaxed 2.3 ML QD sample (in the inset of figure 2(a)), and thus it is ascribed to electron emission from the QD states. The smaller voltage width (from \(-2\) to \(-3\) V) for the C plateau suggests a smaller number of electrons accumulated in the relaxed QD. Hence, some electrons in the QD are depleted by traps presumably associated with the misfits in the QD. It should be noted that the carrier peak at \(\sim 0.2\ \mu m\) cannot be interpreted by the depopulation of the traps associated with the misfits in the QD [14]. Recently, in work on relaxed InAsSb QDs [17], the traps associated with the misfits in the QD were found to emit electrons to the GaAs conduction band with emission energy of 0.35 eV, in a way similar to the misfits in the GaAs layer. Therefore, due to its deeper energy than the QD electron ground state (about 0.18 eV), electron emission from this trap would appear at a much deeper depth than observed. From a simple band diagram simulation, a trap located at 0.2 \(\mu m\) and at 0.35 eV below the GaAs conduction band would appear at about 0.3 \(\mu m\), rather than the \(\sim 0.2\ \mu m\) observed. Thus, the carrier peak at 0.2 \(\mu m\) is attributed to electron emission from the QD. As shown in figure 2(b), the carrier peak at 0.2 \(\mu m\) is followed by a large peak at around 0.33 \(\mu m\). Due to the long emission time, the electrons trapped

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\text{Figures 2. (a) Frequency-dependent C–V spectra and (b) converted concentration profiles of the relaxed 3.3 ML sample, showing carrier accumulation in the dots and additional carrier depletion in the neighboring GaAs bottom layer. The peaks at 0.2 and 0.33 \(\mu m\) are considered as electron emission from the QD and from the traps related to the misfits, respectively. The C–V spectra of the non-relaxed 2.3 ML sample are shown in the inset of (a) for comparison.}
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3.2. Electron emission from a QD

The electron-emission properties of a relaxed QD can be seen from the C–V spectra measured on the 3.3 ML sample. Figures 2(a) and (b) show a C plateau (from \(-2\) to \(-3\) V) and its converted carrier-accumulation peak in the QD region (\(\sim 0.2\ \mu m\)), respectively. The \(x\)-coordinate in figure 2(b) is defined as the distance from the sample surface. The intensity of the peak increases with decreasing temperature, characteristic of a Debye-length effect in a quantum structure. The C plateau appears at nearly the same dc voltage as in a non-relaxed 2.3 ML QD sample (in the inset of figure 2(a)), and thus it is ascribed to electron emission from the QD states. The smaller voltage width (from \(-2\) to \(-3\) V) for the C plateau suggests a smaller number of electrons accumulated in the relaxed QD. Hence, some electrons in the QD are depleted by traps presumably associated with the misfits in the QD. It should be noted that the carrier peak at \(\sim 0.2\ \mu m\) cannot be interpreted by the depopulation of the traps associated with the misfits in the QD [14]. Recently, in work on relaxed InAsSb QDs [17], the traps associated with the misfits in the QD were found to emit electrons to the GaAs conduction band with emission energy of 0.35 eV, in a way similar to the misfits in the GaAs layer. Therefore, due to its deeper energy than the QD electron ground state (about 0.18 eV), electron emission from this trap would appear at a much deeper depth than observed. From a simple band diagram simulation, a trap located at 0.2 \(\mu m\) and at 0.35 eV below the GaAs conduction band would appear at about 0.3 \(\mu m\), rather than the \(\sim 0.2\ \mu m\) observed. Thus, the carrier peak at 0.2 \(\mu m\) is attributed to electron emission from the QD. As shown in figure 2(b), the carrier peak at 0.2 \(\mu m\) is followed by a large peak at around 0.33 \(\mu m\). Due to the long emission time, the electrons trapped

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\text{Figures 3. Temperature-dependent PL spectra of the relaxed 3.3 ML and non-relaxed 2.3 ML samples. The QD ground state emits at 1158 nm in the relaxed sample. A relatively strong increase in the linewidth of the QD emission can be seen with increasing temperature.}
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Figure 4. The $G/F$–$F$ spectra of the 3.3 ML sample measured at $-2.6$ V, corresponding to the electron emission from the QD. The frequency corresponding to the conductance peak is taken as the carrier emission rate.

Figure 5. Arrhenius plots of the emission times in the 3.3 ML sample obtained from the $G/F$–$F$ spectra at different dc voltage. The plots at high temperatures yield emission energy from 0.068 to 0.182 eV from $-2$ to $-3.2$ V. The decreased temperature dependence at low temperatures suggests a tunneling effect.

$10^3$ Hz, but cannot at $2 \times 10^5$ Hz, as shown by the low-$C$ plateau of 180 pF. The emission rate is between the two frequencies. By taking the inflexion frequency as the inverse of the emission time, the emission times (at 110 K) are $10^{-5}$ s at $-1.9$ V, $2 \times 10^{-5}$ s at $-2$ V, $10^{-4}$ s at $-2.2$ V, and $10^{-3}$ s at $-2.4$ V, respectively. This suggests an increased emission energy as the Fermi level is shifted downward. Detailed emission time and energy as a function of voltage are obtained from the conductance/frequency–frequency ($G/F$–$F$) spectra, as shown in figure 4 for $-2.6$ V. The conductance displays a peak at a frequency comparable to carrier emission rate. Figure 5 shows the obtained Arrhenius plots from $-2$ to $-3.3$ V. The emission times at high temperatures can be connected by a straight line from which emission energy is obtained, which increases from 0.068 to 0.182 eV from $-2$ to $-3.2$ V. Figure 5 shows a decrease in the emission energy with lowering temperature, suggesting some tunneling effect at low temperatures. Since a tunneling effect is usually observed for QDs [9–11], this decreased temperature dependence further supports the assignment of the peak at $\sim 0.2 \mu$m as the electron emission from the QD states. The highest-bound emission energy of 0.182 eV is comparable to the confinement energy of the QD electron ground state with respect to the GaAs conduction band. Kapteyn et al [9] reported a value of 0.190 eV for the confinement energy of the InAs QD ground state for an emission at 1.12 eV which is close to our QD emission at 1.07 eV (at 50 K) in figure 3. Thus, the emission energy 0.182 eV is attributed to a thermal activation from the QD electron ground state to the GaAs conduction band. As regards the lowest-bound emission energy of 0.068 eV, it can be due to the depopulation of the QD first-excited state. As discussed above, from the area under the electron-accumulation peak, each QD contains about three electrons. Thus, the QD is filled up to the first-excited state and electron emission from this state can occur. This is indeed the case. When the temperature is lowered below 120 K, the emissions from the QD electron ground and first-excited states are well separated, as shown in figure 6(a), which shows a splitting of one $C$ plateau into two plateaus, as indicated by arrows. The corresponding electron-accumulation peak also splits into two well-separated peaks, as shown in figure 6(b), which displays the depth profile at 93 K. The peak at 0.25 (0.275) $\mu$m corresponds to the QD electron first-excited (ground) state. The smaller peak height for the first-excited state is consistent with the filling of only one electron in the first-excited state, relative to two electrons in the ground state. As illustrated in figure 5, tunneling is unavoidable at low temperatures. The emission energies for these two states are simply estimated from the high-temperature $G/F$–$F$ spectra at the voltages corresponding to the two $C$ plateaus in figure 6(a), which are 0.068 and 0.160 eV, respectively, as indicated in figure 5. These two values are comparable to 0.086 and 0.177 eV calculated for QDs [19] and 0.096 and 0.190 eV experimentally determined by Kapteyn et al [9], and 0.060 and 0.14 eV by Brunkov et al [20] from the relative voltage positions of the...
C plateaus. This comparability is a good indication that the emission energies of 0.160 and 0.068 eV are the confinement energies of the QD electron ground and first-excited states, respectively, with respect to the GaAs conduction band. Their energy difference (0.092 eV) is comparable to the energy difference between the PL ground and first-excited emissions of the QD, consistent with a very small energy separation between the hole ground and first-excited states [19, 20]. By subtracting the GaAs band gap of 1.50 eV from the electron ground-state energy of 0.160 eV and the ground-state PL emission energy of 1.079 eV (at 50 K), we obtain the confinement energy of the hole ground state to be 0.261 eV, a value close to that of 0.205 eV previously determined by Brunkov et al [20]. These results suggest that the observed emission processes (at high temperatures) are from the QD electron and first-excited states to the GaAs conduction band. Thus, the confinement energies of these states with respect to the GaAs conduction band are directly determined from the temperature dependence of the emission times, rather than from the relative voltage positions of the C plateaus which can be strongly affected by the sample resistance. As discussed above, the electron emission from the QD ground and first-excited states can be distinguished when the temperature is lowered to ~120 K. This feature is related to the PL linewidth of the QD ground-state emission in figure 3, which shows a relatively small linewidth of ~60 meV from 50 to 150 K. However, the linewidth increases strongly to ~100 meV when the temperature is increased to 300 K. Since the energy spacing between the ground and first-excited states is only about 0.092 eV, the strongly increased linewidth of the QD ground state would cause the ground and first-excited states to be indistinguishable, consistent with the observation of a single electron-accumulation peak at high temperatures. This comparative study of the PL and the depth profile further confirms that the observed emission is related to the QD states.

So far, we have compared our results with those obtained from capacitance spectroscopy. We now turn to the comparison with the reported data from optical absorption measurements. From intraband absorption, Pal et al [21] have reported an energy separation of 0.10 eV between the electron ground and first-excited states for a QD ground state at 1.19 eV at 77 K, which is very close to our observed value of 0.092 eV. On the other hand, from intraband transmission studies, Adawi et al [22] have observed an absorption peak with 0.113 eV due to the transition from the QD electron ground state to the second-excited state. From photocurrent studies, the same authors also reported transitions with 0.17 and 0.22 eV for the transitions from the QD electron ground state to wetting layer and to GaAs continuum states, respectively. Since the lowest-lying state in the wetting layer is at ~50 meV below the GaAs conduction band, we cannot exclude the possibility that our 0.16 eV emission is due to the transition from the electron ground to wetting layer, rather than directly to the GaAs conduction band. If this is true, the QD electron ground state is at about 0.21 eV below the GaAs conduction band edge. This value is more consistent with the analysis by Kim et al [23] who claimed that the confinement energy for the QD electron ground state should be larger than 0.19 eV for a QD ground emission at 1.127 eV. Furthermore, based on an argument that the binding energy of the electron ground state is higher than that for the hole ground state, Raghavan et al [24] have reported a minimum binding energy of 0.21 eV for the QD electron ground state. These values are all larger than our observed value of 0.16 eV. Thus, there is a possibility that the observed emission processes by time-resolved capacitance spectroscopy is relative to the wetting layer, rather than the GaAs conduction band, leading to a reduction of about 50 meV in the confinement energy. Further investigation on the effect of the wetting layer is needed to make a more conclusive argument on this matter.

For comparison, figure 7 shows a typical depth profile for a non-relaxed InAs/InGaAs QD sample with a 2.4 ML thick InAs layer. Detailed discussions on this sample can be found elsewhere [14]. The strong carrier peak at 0.305 μm is attributed to the electrons tunneling [9–11] from the QD excited states to the GaAs conduction band. The emission time is too short to be resolved since no attenuation of this peak is seen up to 1 MHz at 10 K. The weak peak at 0.325 μm (as indicated by an arrow) shows frequency-dependent dispersion whose emission energy is determined to be ~60 meV [14]. Since this energy is comparable to the PL energy spacing between the ground and first-excited peaks, the weak peak is attributed to a thermal excitation from the QD ground state to the first-excited state. After being thermally excited to the first-excited state, the electron subsequently tunnels to the GaAs conduction band, in a two-stage emission process previously described [9]. This shows a marked tunneling effect for the QD excited states in the non-relaxed QD sample.

3.3. Additional carrier depletion due to misfit dislocations

A comparison between figures 2(b) and 7 reveals that strain relaxation considerably lengthens the emission time for the QD. This effect can be explained by the suppression of tunneling due to the additional carrier depletion in the GaAs bottom layer near the QD. Figure 2(b) shows an asymmetrical depth profile with additional carrier depletion in the bottom GaAs layer (0.25–0.32 μm). This depletion has a valley concentration of 1 × 10^{10} cm^{-3}, compared to that of 5 × 10^{16} cm^{-3} in the front side of the QD. Furthermore, the width
of the additional carrier depletion is about 0.1 μm, which is more than three times broader than that in the front side of the QD. Since carriers are emitted to the bottom GaAs electrode, such a broad depletion layer in the back of the QD would significantly reduce the tunneling probability. As a result, the electrons in the QD ground state would have to be thermally activated to the GaAs conduction band. This is consistent with the observed emission energy of 0.16 eV, which is close to the energy spacing between the QD ground state and the GaAs conduction band, in contrast to the observed emission energy of 60 meV for activation from the QD ground state to the first-excited state in the non-relaxed sample.

In view of the TEM data, the misfit dislocations in the bottom GaAs layer can be the reason for the additional carrier depletion. Due to the long emission time, the related traps are revealed by the DLTS spectra as shown in figure 8 for a rate window of 0.86 ms. In contrast to those in the non-relaxed 2 and 2.3 ML samples, the relaxed 3.3 ML sample displays a trap around 275 K for the sweeping voltage of −1.5 V/−3 V, corresponding to the QD and neighboring GaAs bottom layer. The top GaAs layer is free of traps, as is evident from the inset, which shows no trapping signals for 0 V/−0.5 V and −0.5 V/−1.5 V. The continuous broad background signal at low temperatures is thought to be due to the electron emission from the QDs.

Figure 8. The DLTS spectra at a rate window of 0.86 ms for the non-relaxed 2 and 2.3 ML samples and the relaxed 3.3 ML sample. A trap at 0.35 eV is detected in the 3.3 ML sample for −1.5 V/−3 V, corresponding to the QD and neighboring GaAs bottom layer. The top GaAs layer is free of traps, as is evident from the inset, which shows no trapping signals for 0 V/−0.5 V and −0.5 V/−1.5 V. The continuous broad background signal at low temperatures is thought to be due to the electron emission from the QDs.

The carrier distribution in the back of the QD is normal (with a valley concentration of $5 \times 10^{16}$ cm$^{-3}$), consistent with there being no misfits in the bottom GaAs layer. Without the additional carrier depletion in the back of the QD, the emission time for the QD is too short to be resolved even up to $10^3$ Hz at 20 K.

Figure 9. The 20 K depth profile of the 2.8 ML InAsSb relaxed QD sample, showing a weak carrier peak at the QD (indicated by QE) and drastic carrier depletion in the front of the QD. Without the additional carrier depletion in the back of the QD, the emission time for the QD is too short to be resolved even up to $10^3$ Hz at 20 K.
related to the misfits. Due to the suppression of tunneling for the QD excited states, the electrons in the QD ground and first-excited states are thermally activated to the GaAs conduction band, allowing for the determination of the QD electronic band structure.

4. Conclusions

Strain relaxation is shown to induce additional carrier depletion in the GaAs bottom layer which can lengthen the emission time from the QD. The TEM data show the misfits in the QD and neighboring GaAs bottom layer. Thus, the misfits in the GaAs bottom layer may cause additional carrier depletion which can lengthen the emission time by the suppression of tunneling. As a result, the electrons in the QD states are thermally activated to the GaAs conduction band. This can explain the observed emission energies of 0.160 and 0.068 eV which are comparable to the confinement energies of the QD ground and first-excited states, respectively, with respect to the GaAs conduction band. DLTS reveals a trap at 0.35 eV in the bottom GaAs layer, which is attributed to the misfits. Its intensity is about two orders of magnitude less than the misfit intensity, suggesting that most of the misfits are not active traps.

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