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Effect of incorporating an InAlAs layer on electron emission in self-assembled InAs quantum dots

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We perform capacitance-voltage and admittance spectroscopy to investigate the effect of incorporating an InAlAs layer before an InGaAs cap layer on electron emission in self-assembled InAs quantum dots (QDs). We show that this incorporation of a high potential barrier increases the emission time of the electrons thermally activated from the QD ground to the first excited state. The energy separation between the ground and first excited states in the conduction band increases from 57.2 to 79.1, to 89.2, and to 95.6 meV with increasing the thickness of the InAlAs layer from 0 to 10, to 14, and to 20 Å. Combining with photoluminescence (PL) data, the ratios of the energy separation between the ground and first excited states in the conduction band and valence band are determined to be 7.3:2.7 and 7.8:2.2 for 0 and 10 Å InAlAs, respectively. In addition, this incorporation is shown to blueshift the PL first excited state much larger than the ground state.
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I. INTRODUCTION

InAs/GaAs self-assembled quantum dots[1–7] (QDs) have attracted considerable interest in fundamental physics and promising technological applications. In order to achieve useful applications, extensive variation of emission wavelength is required. Previous investigations have shown that embedding the QDs in different layers[8,9] or incorporating different atoms[10] into the QDs can vary the emission wavelength. For example, capping the InAs QDs with a strain-reducing InGaAs layer can shift the emission wavelength to the important telecommunication range.[11] However, this InGaAs cap layer results in a reduction of energy separation between the QD ground and excited states, leading to degraded temperature stability. Capping the QD with a high potential barrier such as an Al(Ga)As or InAlAs layer has been shown to blueshift the emission wavelength and increase dot density.[12–16] However, these works mainly focus on optical properties. Little attention is paid to the effect of an InAlAs layer on the electrical properties of the QDs. In this work, we investigated the effects of an InAlAs layer with different thickness on the electron emission from the InAs QDs by frequency-dependent capacitance-voltage (*C-V*) and admittance spectroscopy. We show that this InAlAs layer can significantly increase the electron emission time due to an increment in the energy separation between the QD ground and first excited states in the conduction band, and the result is compared with photoluminescence (PL) data to obtain the energy-band diagram.

II. EXPERIMENT

The samples under study were grown on n^+-GaAs (100) substrates by solid source molecular-beam epitaxy in a Riber Epineat machine. The QDs were formed by depositing a 2.4 monolayer (ML) InAs layer at a substrate temperature of 490 °C and, then, covered by an InAlAs layer with different thicknesses of 0, 10, 14, and 20 Å and followed by a 44 Å $In_{0.14}Ga_{0.86}As$ layer. The QDs are sandwiched between two 0.2-μm-thick Si-doped GaAs ($6 \sim 10 \times 10^{16}$ cm^{-3}) layers so that the QDs are outside the depletion region at zero bias. A QD sheet density about $(3\text{–}5) \times 10^{10}$ cm^{-2} was observed by atomic field microscopy (AFM) images in samples without the InAlAs layer. Schottky diodes were realized by evaporating Al on the samples with a dot diameter of 1500 μm. PL measurements were carried out using a double-frequency Nd-doped yttrium aluminum garnet (YAG) laser at 532 nm. A HP 4194A gain phase analyzer was used to perform *C-V* profiling and admittance spectroscopy.

III. MEASUREMENT AND RESULTS

A. Carrier distribution

Figures 1(a)–1(d) show the temperature-dependent carrier distribution profiles at 10 KHz, converted from the *C-V* spectra shown in each inset, for 0, 10, 14, and 20 Å InAlAs, respectively. The profile for 0 Å InAlAs shows a strong peak E_1 (at 0.305 μm) whose intensity increases with lowering temperature, characteristic of a Debye-length effect in a quantum structure. No intensity attenuation of this peak is seen up to 1 MHz at 10 K, indicating that the emission time is too short to be obtained. We ascribe this peak to the

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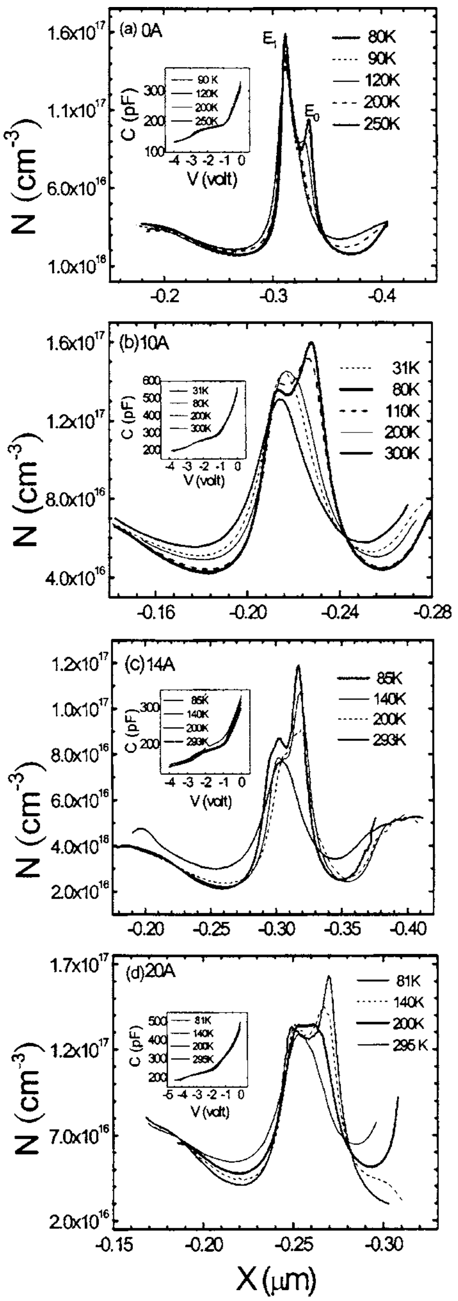

FIG. 1. Temperature-dependent carrier distribution profiles at 10 KHz, converted from the *C-V* spectra shown in the inset, for (a) 0, (b) 10, (c) 14, and (d) 20 Å InAlAs. The E_0 peak in (a) is ascribed to the electron excitation from the QD ground to first excited state. Incorporating the InAlAs layer can increase the intensity and the emission time of the E_0 peak.

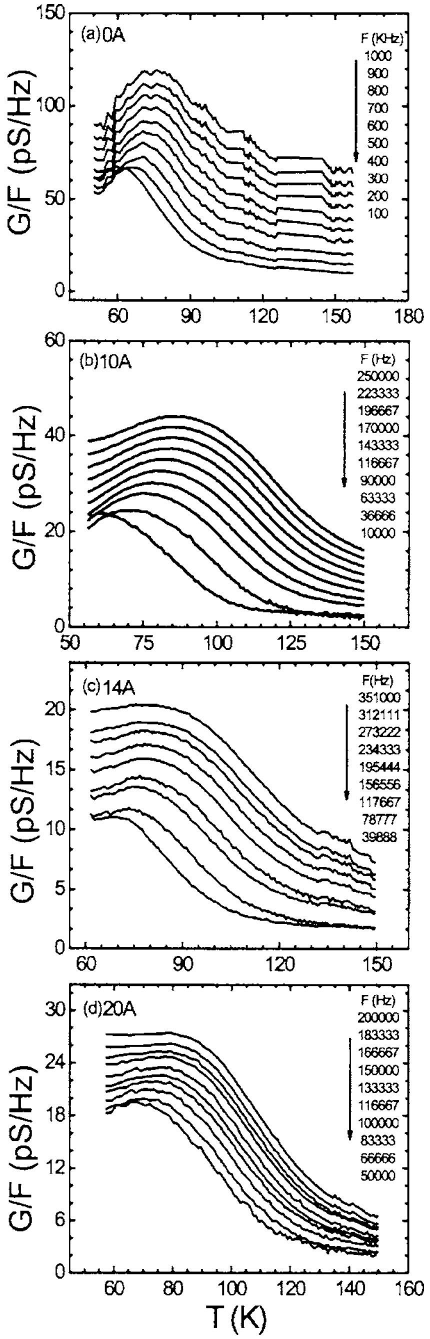

FIG. 2. Experimental conductance/frequency-temperature(G/F-T) spectra of the E_0 peaks for (a) 0, (b) 10, (c) 14, and (d) 20 Å InAlAs. The shift of the peak to higher temperature shows an increase of the emission time due to the InAlAs layer.

electron emission from the QD excited states to GaAs conduction band. Besides the E_1 peak, another weak peak E_0 appears at 0.325 μm when temperature is less than 120 K. This peak displays frequency-dependent attenuation and, thus, we can obtain its emission time by measuring the conductance of the peak. Figure 2(a) shows its conductance/frequency-temperature (G/F-T) spectra under several frequencies. The conductance will exhibit a peak when measuring frequency is comparable to the emission rate. Figure 3 shows its Arrhenius plots, from which we obtained an activation energy (capture cross section) of 57.2±5.1 meV (1.4×10^{-14} cm^2). This energy is close to the energy separation between the QD ground and first excited states in the conduction band, predicted from PL data and dot size, and thus this E_0 peak is ascribed to the electron thermal excitation from the ground to first excited state. After that, the electrons subsequently tunnel to the GaAs conduction band. This assignment is consistent with the two-stage emission process previously proposed.[17,18]

B. Admittance spectroscopy

Capping the QD with the InAlAs layer significantly increases the emission time of the E_0 peak, as illustrated from

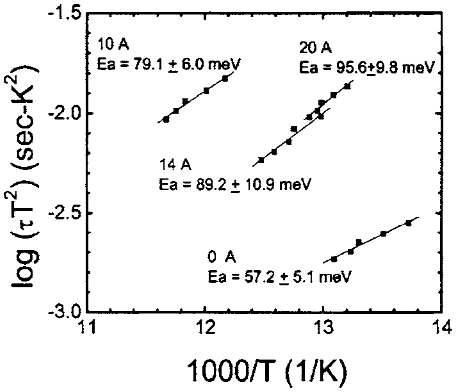

FIG. 3. Arrhenius plots of the emission times of the E_0 peaks for 0, 10, 14, and 20 Å InAlAs. The activation energy increases with increasing the InAlAs layer thickness.

the higher temperature at which the E_0 peak starts to appear in Figs. 1. Figures 2(b)–2(d) show the measured G/F-T spectra of the E_0 peaks for 10, 14, and 20 Å InAlAs, respectively. It can be seen that the conductance peak shifts to higher temperature relative to the 0 Å InAlAs. Figure 3 shows the Arrhenius plots of the emission times. As shown, the activation energy (capture cross section) of the E_0 peak increases from 57.2±5.1 (1.4×10^{-14}) to 79.1±6.0 (2.2×10^{-14}), to 89.2±10.9 (3.2×10^{-13}), and to 95.6±9.8 meV (7.48×10^{-13} cm^2) with increasing the InAlAs layer thickness from 0 to 10, to 14, and to 20 Å. This result indicates that the InAlAs cap layer can increase the potential barrier of the cap layer, leading to an increment in the energy separation between the ground and first excited states in the conduction band $\Delta E_{c(g\text{-}e)}$.

C. PL spectra

For comparison, Fig. 4 shows the 300 K PL spectra, under a power of 90 mW, for the studied samples. In each sample, two peaks can be clearly seen. The high-energy peak belongs to the first excited state. It can be seen that increasing the InAlAs layer can blueshift the ground state from 16 to 22 meV and the first excited state from 39 to 47 meV without any appreciable increase in the linewidth. This blueshift is mainly due to the energy-level modification due to the InAlAs high-energy barrier layer. Notably, the blueshift in the first excited state is much larger than that in the ground state. This difference is slightly too large to be explained by the effect of the high potential barrier only and leads us to suspect an effect of Al intermixing from the InAlAs layer with the QDs, leading to a band-gap increment. Since the first excited state (*p*-*p* transition) is more localized in the QD periphery, relative to the ground state (*s*-*s* transition), this band-gap increment is more significant in the first excited state. Figure 4 shows that the energy separation between the ground and first excited states $\Delta E_{(g\text{-}e)}$ can be increased from 78.0 to 101.4, to 102.6, and to 103.1 meV with increasing the InAlAs thickness from 0 to 10, to 14, and to 20 Å. This result shows that capping the QD with a 10 Å InAlAs layer can significantly increase the energy separation, but further increasing the InAlAs thickness beyond that results in no significant change. Subtracting these values from the energy separation between the ground and first excited states in the conduction band $\Delta E_{c(g\text{-}e)}$, as shown in Fig. 3, we obtain the corresponding energy separations in the valence band $\Delta E_{v(g\text{-}e)}$ to be 20.8, 22.3, 13.4, and 7.5 meV, respectively, for 0, 10, 14, and 20 Å InAlAs. This result shows no increase of $\Delta E_{v(g\text{-}e)}$ by the incorporation of the InAlAs layer. Therefore, the effect of the InAlAs layer on the energy-level modification is mainly in the conduction band. With these results, the ratios of $\Delta E_{c(g\text{-}e)}$ and $\Delta E_{v(g\text{-}e)}$ are 7.3:2.7 and 7.8:2.2 for the 0 and 10 Å InAlAs layers, respectively, and their schematic band structures are shown in Fig. 5.

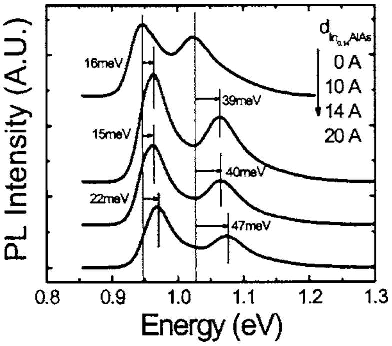

FIG. 4. PL spectra at 300 K for 0, 10, 14, and 20 Å InAlAs. The energy separation between the ground and first excited states increases from 78.0 to 101.4, to 102.6, and to 103.1 meV with increasing the InAlAs thickness from 0 to 10, to 14, and to 20 Å.

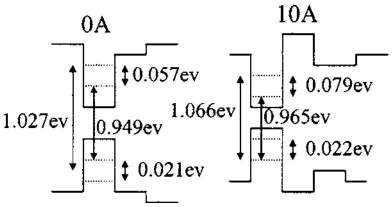

FIG. 5. Schematic band structures for 0 and 10 Å InAlAs.

Figures 1 show that the addition of the InAlAs layer can increase the intensity of the E_0 peak (relative to the E_1 peak). The ratio of the area under the E_0 peak to the total area under the E_0 and E_1 peaks increases from 0.38 to 0.63 with a 10 Å InAlAs layer, suggesting that more electrons are confined in the QD ground state. This result can be explained by an increase in the QD intensity. AFM images show a density of $(3\text{–}5)\times10^{10}$ cm^{-2} without the InAlAs layer and $\sim9\times10^{10}$ cm^{-2} with a 10 Å InAlAs layer. This increase in the dot density can be explained by the suppression of the In segregation[13] from the QDs by the InAlAs layer. However, the density of the QD cannot be further increased by increasing the InAlAs thickness to 14 and 20 Å, as illustrated from the no increase in the ratio of the area and as confirmed by AFM images. This consistency also corroborates the assignment of the E_0 and E_1 peaks in the carrier profiles.

IV. CONCLUSIONS

In summary, we investigate the effect of incorporating an InAlAs layer before an InGaAs cap layer on the electron distribution and emission in self-assembled InAs quantum dots (QDs). We show that this incorporation increases the electron emission time from the dot ground to first excited state due to an increase in the energy separation between the ground and first excited states in the conduction band from 57.2 to 79.1, to 89.2, and to 95.6 meV with increasing the thickness of the InAlAs layer from 0 to 10, to 14, and to 20 Å. By comparison with photoluminescence (PL) data, the ratios of the energy separation between the ground and first excited states in the conduction band and valence band are determined to be 7.3:2.7 and 7.8:2.2 for 0 and 10 Å InAlAs, respectively. This incorporation also causes a larger blueshift of the PL first excited state than that of the ground state.

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