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N incorporation into InGaAs cap layer in InAs self-assembled quantum dots

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This study presents the results of incorporating N into self-assembled InAs quantum dots (QDs) capped with an InGaAs cap layer. Experimental results indicate that such incorporation can redshift the QD ground state and decrease the energy spacing between the QD ground and first excited states. However, this incorporation reduces the potential barrier of the cap layer, increasing the electron escape from the QDs. Capacitance-voltage profiling shows that a broad shoulder corresponding to the electron emission from the QD ground to first-excited state cannot be resolved from the peak related to the electron emission from the excited states upon this incorporation. This finding implies that this incorporation reduces the energy spacing between the QD ground and first-excited states in the conduction band, thus correlating well with the photoluminescence data. In contrast, incorporating N directly into the InAs QD produces no redshift of the emission wavelength but introduces a deep trap at ~0.21 eV that depletes the electrons in the QDs. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2140891]

I. INTRODUCTION

InAs/GaAs self-assembled quantum dots (QDs) (Refs. 1–11) have attracted considerable interest in fundamental physics and promising technological applications. Optical fiber communication requires an increased emission wavelength of 1.3 or 1.55 μm. Self-assembled GaInNAs QDs (Ref. 12) have been shown to increase the wavelength to this range but significantly reduce the optical intensity. The emission wavelength can also be increased by capping the InAs QD with a strain-reducing InGaAs layer.[13] Incorporating N atoms into the InGaAs cap layer can further reduce the potential barrier of the cap layer, leading to a redshift in the emission wavelength.[14,15] However, the detailed properties have seldom been investigated, especially, how the optical and electrical properties are related. Therefore, this work undertakes photoluminescence (PL), current-voltage (I-V), capacitance-voltage (C-V), and deep-level transient spectroscopy (DLTS) studies on the effects of N incorporation into the InGaAs cap layer and into InAs QDs. According to those results, incorporating N into the cap layer can redshift the QD ground state and reduce the energy spacing between the ground and first-excited states. This finding is compared with the variation of the carrier distribution around the QDs. Conversely, incorporating N into the InAs QD introduces a deep trap that depletes the electrons in the QDs.

II. EXPERIMENTS

The QD samples were grown on n^+-GaAs (100) substrates by molecular beam epitaxy. The QD structure, grown at 480 °C, consisting of 2.4 ML InAs layer and a 60 Å $\mathrm{In_{0.15}Ga_{0.85}As}$ cap layer, was sandwiched between two 0.3-μm-thick Si-doped GaAs layer (6×10^{16} cm^{-3}) layers. The indium and gallium were supplied from Knudsen cells, and the As in the form of $\mathrm{As_2}$ was supplied from a cracker source. N atoms were incorporated by using an EPI-Unibulb radio frequency (rf) plasma source to supply active nitrogen species from ultrapure $\mathrm{N_2}$ gas. Three samples, with 0.4% and 1% N incorporation into the InGaAs cap layer and with N incorporation into the InAs QD, called the InAsN sample, were grown and compared with a reference sample (without any N incorporation). The N flux used for the N incorporation into the InAs QD was kept the same as that for the sample with 1% N incorporation into the cap layer, resulting in a much higher N composition (more than 10%) in the InAsN sample because the InAs growth rate (0.26 Å/s) was significantly smaller than that of GaAs (2.78 Å/s). Schottky diodes were realized by evaporating Al on the samples with a dot diameter of 1500 μm. PL measurements were carried out using a frequency-doubled yttrium-aluminum-garnet (YAG):Nd laser at 532 nm. The C-V profiling was performed using an HP4194A gain phase analyzer.

III. RESULTS AND DISCUSSIONS

A. PL spectra

Figure 1 shows the 50 K PL spectra of the QD related peaks from the studied samples. The reference sample showed a QD ground state at 1215 nm and a first-excited state at 64 meV away. Incorporating 0.4% N into the cap layer caused a negligible shift on the ground state but redshifted the first-excited state. This incorporation decreased the energy separation between the ground and first-excited states from 64 to 45 meV. Increasing the N incorporation to

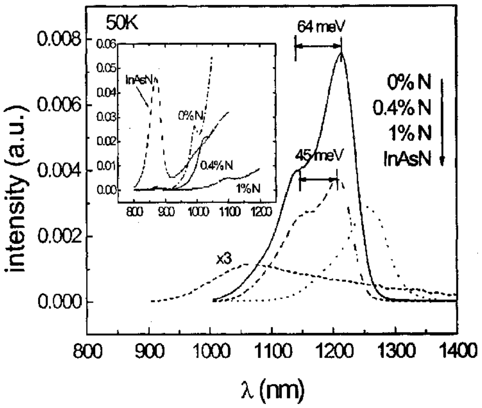

FIG. 1. 50 K PL spectra of the QD samples. Incorporating N into the InGaAs cap layer can redshift the QD ground state and decrease the energy separation between the ground and first-excited states. By contrast, incorporating N into the InAs QD severely degrades the intensity. The 300-K spectra due to the GaAs barrier layers and InGaAs(N) cap layers are shown in the inset. The GaAs barrier layers emit at around 870 nm and the InGaAs cap layer emits at 992 nm which shifts to 1031 nm for 0.4% N incorporation and then to 1100 nm for 1% N incorporation.

1% can redshift the ground state to 1256 nm with the first-excited state not clearly seen. These results show that incorporating N into the cap layer can reduce the energy separation between the ground and first-excited states as well as redshift the ground state. No clear increment in the dot size was observed in the 1% N incorporation relative to the reference sample. A previous investigation[17] showed that incorporating N into GaAs (or InGaAs with small In content) introduces a large conduction band bowing. The 300-K spectra due to the GaAs barrier layers and InGaAs(N) cap layers are shown in the inset. The GaAs barrier layers emit at around 870 nm and the InGaAs cap layer emits at 992 nm which shifts to 1031 nm for 0.4% N incorporation and then to 1100 nm for 1% N incorporation. This feature indicates that the band gap of the cap layer is reduced by 50 meV for 0.4% N incorporation and 123 meV for 1% N incorporation, assuming the band gap reduction is mainly in the conduction band. A comparison of the intensities of these peaks shows that the quality of the cap layer is not degraded by 0.4% N incorporation but significantly degraded by 1% N incorporation. Since the energy levels of the QD are related to the potential barrier of the cap layer, this energy level modulation can be explained by the reduced potential barrier as a result of the N incorporation into the InGaAs cap layer. Conversely, Fig. 1 shows that incorporating N directly into the InAs QD cannot redshift the emission wavelength but results in very weak broad spectra centered at around 1060 nm. Due to the very low growth rate of the InAs coverage, keeping the same N flux as the growth of the cap layer causes a high N composition in the InAs layer. The estimated N composition is as high as 17% and probably because of this high N composition, the optical quality of the QD is significantly degraded as compared with that of a similar InAsN QD sample with an N composition of 2% in Ref. 12. According to our results, this incorporation introduces a deep trap at ~0.21 eV that severely degrades the PL intensity.

To investigate the thermal stability, Fig. 2 shows the temperature-dependent PL integrated intensities of the QD ground states for different N incorporation into the cap layer. The intensity of the reference sample is strongest at 50 K, remains constant from 50 to 80 K, and then decreases as temperature increases. The 0.4% N sample shows a similar temperature dependence except that the intensity is slightly lower than that of the reference sample at low temperatures. The 1% N sample shows that the intensity decreases with increasing temperature from 50 K. By fitting the data to a simplified thermally activated equation (shown as lines), the activation energies were obtained as 45, 41, and 31 meV for the reference, 0.4% and 1% N samples, respectively. The decreased activation energy suggests that the carriers escape easily from the QDs. These thermal energies were much smaller than the electron confinement energy for the QD ground state relative to the GaAs conduction band, which was estimated as 0.25–0.3 eV from the dot size and PL emission wavelength. Therefore, the electrons in the QD ground state did not directly escape to the GaAs. As shown later, DLTS detected no trapping defects in these three samples. Therefore, this degraded PL intensity is not caused by defects. Previous investigations[16] have shown that capping the QDs with an InGaAs cap layer results in poor temperature stability. Since incorporating N reduces the potential barrier of the cap layer, the electrons in the QD escape most easily to the GaAs via the cap layer. According to a two-state emission process proposed by Kapteyn *et al.*,[18] the electrons were thermally activated from the QD ground to first-excited state, and escape to the cap layer due to the reduced potential barrier.

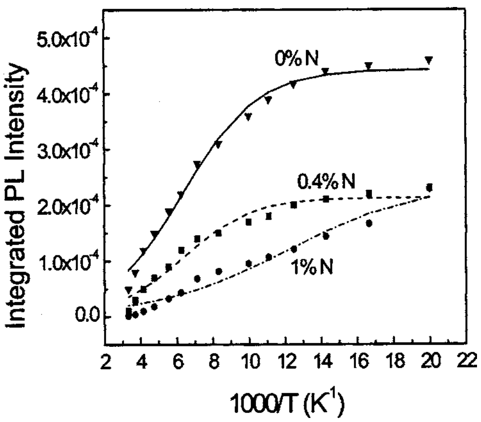

FIG. 2. Temperature dependence of the PL integrated intensities of the ground states for 0%, 0.4%, and 1% N incorporation into the cap layer. The N incorporation enhances the carrier escape from the QDs when temperature increases.

B. *I-V* characteristics

Figure 3 shows the room-temperature current-voltage (I-V) characteristics of the Schottky-contact studied samples. All samples show typical rectified characteristics. The reference, 0.4% and 1% N samples display good ideality factors less than 1.4 but the InAsN sample displays an ideality factor of about 1.9 and a large series resistance of about 1.8 kΩ, estimated from the current bending at large forward voltages. At reversed voltages, the reference sample had the largest reverse current, probably because the QDs was close to the

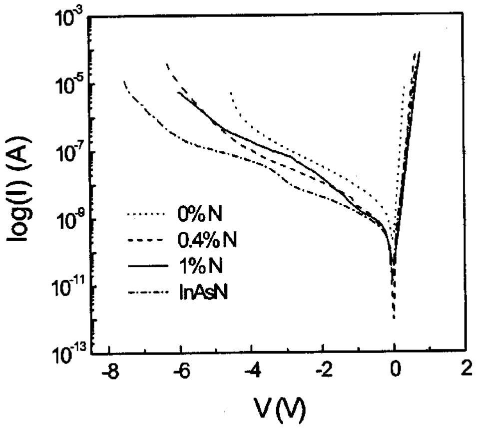

FIG. 3. 300-K I-V characteristics of the Schottky-contact QD samples. The three samples with N incorporation showed nearly the same leakage current from 0 to -1 V. The samples with 0.4% and 1% N incorporation into the cap layer exhibited a strong current rise around -1 to -2 V but the InAsN QD sample exhibited a strong current rise at around -3 V due to a deep trap.

sample front surface due to a chemical etching before the Al evaporation. The 0.4%and 1% N samples show nearly same current from 0 to -1 V, then a strong rise in current between -1 and -2 V. The C-V data show that this current rise corresponds to the edge of the Schottky depletion region sweeping to the edge of the QDs depletion region, indicating that the electrons are being sweeping out of the QDs. The current of the InAsN sample was similar as that of the 0.4% and 1% N samples from 0 to -1 V but rose strongly at around -3 V, due to the formation of a deep trap that depletes the electrons in the QD. Due to the deep energy and very long emission time, the trapped electrons could not be swept out of the trap until more negative dc voltage was provided to move the Fermi level well below this trap. The trap-induced carrier depletion caused the large series resistance, suppressing the reverse leakage current.

C. *C*-*V* profiling

Figures 4(a) and 4(b), respectively, show the 300 K C-V spectra at 100 kHz and their converted concentration profiles for the studied samples. The reference sample had an accumulation peak at 0.23 μm, which is slightly different from that of the other samples (0.3 μm), probably due to chemical etching before Al evaporation. Similar profiles are observed in 0.4% and 1% N incorporation. Conversely, the InAsN sample exhibited significant carrier depletion in the QD region. To understand the detailed properties, Fig. 5(a) shows the temperature-dependent carrier profiles for the reference sample, indicating that the accumulation peak consists of a sharp peak E_1 and a broad shoulder E_0 to the right. The increase in the intensity of the E_1 peak with lowering temperature was due to a Debye-length effect in a quantum structure. This peak did not attenuate up to 1 MHz at 10 K, indicating that the emission time was too fast to be estimated. Based on previous references,[18–21] this peak was assigned to a direct tunneling for the electrons from the QD excited states to the GaAs conduction band. The shoulder E_0 was found to exhibit frequency-dependent attenuation. The emission time could be obtained by measuring its conductance as a function of temperatures. The activation energy (capture cross section) of the emission time was found to be about 60 meV (1.4×10^{-14} cm^2), which is comparable to the predicted energy spacing between the ground and first-excited states in the conduction band and, thus, the shoulder is attributed to the thermal excitation of the electrons from the QD ground to the first-excited state. The area under the shoulder had a density of about 3.8×10^{10} cm^{-2}, which is close to the QD density of $\sim3\times10^{10}$ cm^{-2} estimated from atomic force microscopy (AFM). After thermally excited to the first-excited state, the electrons were subsequently tunneled to the GaAs conduction band. This assignment is consistent with the two-stage emission process first proposed by Kapteyn *et al.*[18]

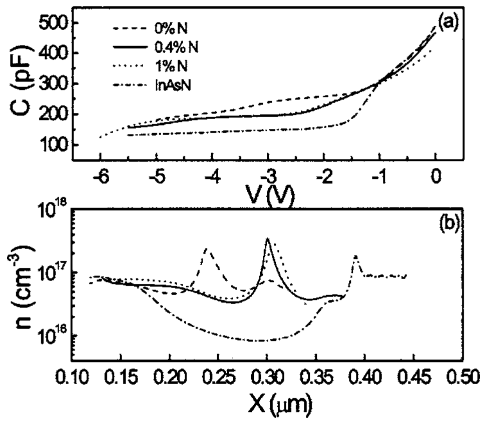

FIG. 4. (a) 300 K C-V spectra at 100 kHz and (b) their converted concentration profiles for the studied samples. The reference, 0.4% and 1% samples exhibited an accumulation peak but the InAsN sample showed significant carrier depletion in the QD region.

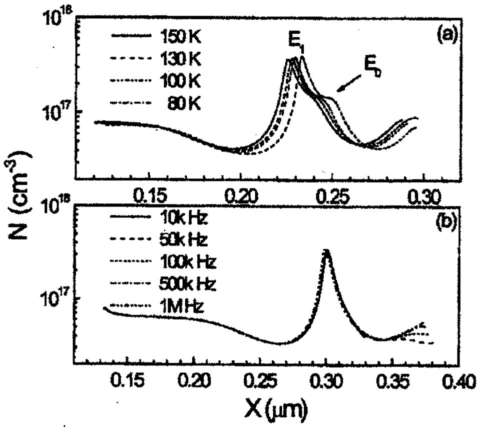

FIG. 5. (a) Carrier profiles at several temperatures for the reference sample. The peak E_1 is attributed to electron tunneling from the QD excited states to GaAs conduction band and the shoulder E_0 is attributed to the thermal excitation from the QD ground to first-excited state; (b) 80 K carrier profiles for 0.4% N incorporation, showing the absence of the shoulder.

No such shoulder was observed when N was incorporated into the cap layer. Figure 5(b) shows the 80 K carrier profiles observed on the 0.4% N incorporation at several frequencies. A similar profile was observed in the 1% N incorporation. This profile shows a sharp accumulation peak which displays no attenuation with frequency. Notably, the

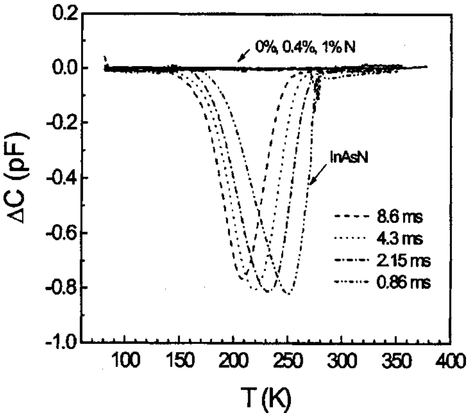

FIG. 6. DLTS spectra of the QD samples. No trapping signals were visible in the reference, 0.4% and 1% N incorporation samples. However, a dominant peak around 200–250 K appeared in the InAsN sample.

shoulder E_0 observed in the reference sample is not visible in this diagram. This feature is not caused by poor sample quality since the peak was narrower than the reference sample nor by different background concentration, since both samples had nearly the same background concentrations. The absence of the shoulder indicates that the energy spacing between the QD ground and first-excited states in the conduction band decreased. This result is consistent with the PL data, which show a reduction in the energy spacing between the QD ground and first-excited states, indicating that the energy decrement is significant in the conduction band. Conversely, Fig. 4(b) shows that incorporating N into the QD causes significant carrier depletion in the QD region, implying the presence of a deep trap in the QD region. The irregular peak (at 0.4 μm) following the carrier depletion reflects the effect of this trap. This peak is visible only when the trapped electrons cannot follow the ac modulating signal and, is thus due to the emission of the trapped electrons when the dc bias moves the Fermi level well below the deep trap. DLTS was performed on this sample along with the other samples to reveal this trap.

D. DLTS measurement

Figure 6 shows the DLTS spectra of the studied samples. Except in the InAsN sample, no trapping signals were detected, indicating that incorporation N into the cap layer introduces no traps. However, a dominant peak from 200 to 250 K was visible in the InAsN sample. The intensity of this trap was dominant in the QD region. Figure 7 shows the intensity variation of this peak for decreasing sweeping biases from −0.5 to −3 V in steps of 0.5 V. The rate windows were 8.6, 4.3, 2.15, and 0.43 ms from left to right of the curves. The intensity decreased considerably when the sweeping biases were away from the QD region (−1 to −2 V), indicating that this trap was in the QD region with strong rate window dependence. This may be due to incomplete filling of the trap. The peak intensity was maximum and independent of rate window in sweeping biases (−1/−1.5 V and −1.5/−2 V) corresponding to the QD region. As the filling pulse time was increased, the peak intensity was saturated at $\Delta C \sim 1$ pF, giving the sheet density of the electrons of 1.8×10^{10} cm^{-2} using the simplified equation $N_d \Delta C \varepsilon A / C^2$, where $N_d = 7.2 \times 10^{16}$ cm^{-3}, $C = 150$ pF, $A = 5 \times 10^{-3}$ cm^2, and $\varepsilon = 1.14 \times 10^{-10}$ F/m. This density is comparable to the QD density of $\sim 3 \times 10^{10}$ cm^{-2} estimated by AFM, implying that the defect trap correlates with the QD. The Arrhenius plots in Fig. 8 show that the activation energy of this trap increased from 0.20 to 0.32 eV as the sweeping biases decreased from −0/−0.5 V to −2.5/−3 V. The activation energy was 0.205–0.21 eV for the biases (−1/−1.5 V and −1.5/−2 V) corresponding to the QD region. Since this trap lies in the QD region, it must have caused the observed depletion in the QD region. As shown in Fig. 3, this sample showed a strong current rise at around −3 V, which is much smaller than the voltages (−1 to −2 V) for the samples with 0.4% and 1% N incorporation into the cap layer, indicating that this trap lies much deeper in energy than the QD ground state. Therefore, this trap could not lie at 0.205–0.21 eV below the GaAs conduction-band edge, and was tentatively assigned to be at 0.205–0.21 eV below the QD ground state. During the DLTS measurements, the elec-

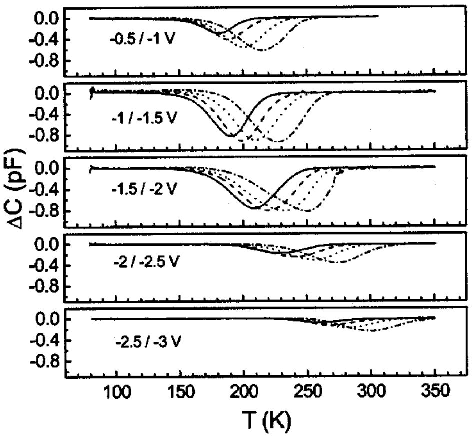

FIG. 7. DLTS spectra of the InAsN sample, showing the variation of the intensity of the trap for different sweeping biases from −0.5 to −3 V with each step of 0.5 V. The intensity variation suggests that this trap is located in the QD region. The rate windows are 8.6, 4.3, 2.15, and 0.43 ms from the left to the right of the curves.

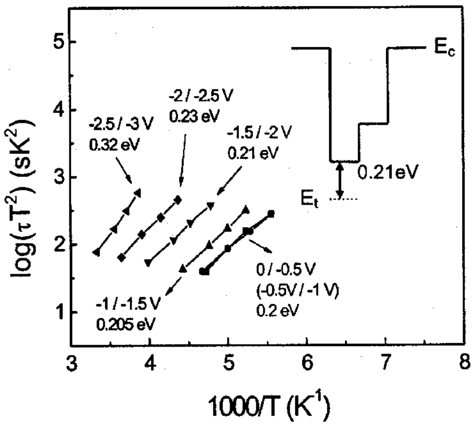

FIG. 8. Arrhenius plots of the emission times of the trap observed in the InAsN sample. The activation energy of this trap increases from 0.20 to 0.32 eV as the sweeping voltages change from 0/−0.5 V to −2.5/−3 V. The energy position of this trap is shown in the inset.

trons were thermally activated from the trap to the QD ground state and subsequently to the GaAs bottom electrode probably via the states of the wetting layer. The latter process was faster than the former process and thus was not detected. A schematic diagram showing the position of this trap is included in the inset of Fig. 8, in which the wetting layer is neglected. The capacitance-time measurement on this trap reveals that the capacitance displays an exponential function with time. Therefore, the trap was not caused by dislocations exhibiting logarithmic function.[22] By comparison, the trap was found to be similar to a trap found in InGaAsN/GaAs quantum-well structures. The activation energy of the trap is independent of the In and Ga compositions. Thus, the trap was tentatively attributed to an N-induced localized state. Further investigation is needed to understand this point in detail.

IV. CONCLUSIONS

The effects of the N incorporation in the InAs QDs capped with an InGaAs layer were studied. Incorporating N into the cap layer was found to redshift the QD ground state and decrease the energy separation between the ground and first-excited states. This decrement in the energy separation enhanced the carrier escape from the QD, degrading the PL intensity as the temperature increased. C-V profiling shows that the peak corresponding to the electron emission from the QD ground to first-excited state could not be resolved from the excited-state peak by this incorporation. This finding correlates with the reduced energy separation observed in PL. Conversely, incorporating N into the QD can introduce a carrier-depletion deep trap at ~0.21 eV in the QD region.

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