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JOURNAL OF APPLIED PHYSICS **97**, 083511 (2005)

Characterization of self-assembled InAs quantum dots with InAlAs/InGaAs strain-reduced layers by photoluminescence spectroscopy

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The optoelectronic characteristics of self-assembled InAs quantum dots (QDs) with strain-reduced layers (SRLs) were investigated using photoluminescence (PL) spectroscopy. Various SRLs that combine $In_{0.14}Al_{0.86}As$ and $In_{0.14}Ga_{0.86}As$ with the same total thickness were examined to ascertain their confining effect on carriers in InAs QDs. The emission wavelength is blueshifted as the thickness of InAlAs is increased. The energy separation between the ground state and the first excited state of QDs with InAlAs SRLs greatly exceeds that of QDs with InGaAs SRLs. Atomic force microscopic images and PL spectra of the QD samples demonstrated that high-quality InAs QDs with long emission wavelengths and a large energy separation can be generated by growing a low-temperature, thin InAlAs SRL onto self-assembled QDs. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1886278]

I. INTRODUCTION

Self-assembled semiconductor quantum dots (QDs) fabricated by the Stranski–Krastanow (S–K) method have attracted much attention over recent years.[1–3] Their electronic characteristics have been comprehensively investigated because of their potential applications in optoelectronic devices.[4,5] High-quality 1.3-μm InGaAs QD lasers have been successfully fabricated to provide an output power of 2.7 W at room temperature.[6]

InAs/InGaAs self-assembled QDs have also been considered to be a candidate for 1.3-μm laser diodes (LDs). However, the performance of InAs/InGaAs QD lasers declines substantially as the temperature increases.[7,8] The thermal excitation of carriers from their ground state to higher energy states, with a high nonradiative recombination rate,[9] has been attributed to the deterioration of the operation characteristics of the InAs/InGaAs QD lasers.[7,8] Although InAs/InGaAs QDs can be tuned to support an energy separation from 66 meV at an emission wavelength of 1.33 μm to one of 82 meV at an emission wavelength of 1.28 μm,[10–12] this range of energy separation does not prevent carriers in InAs QDs from thermally escaping. Alternatively, the use of GaAs as a confining layer of InAs QDs has yielded an energy separation of up to 104 meV.[13] However, considerable InAs/GaAs intermixing occurs at the interface and QDs shrink to reduce the emission wavelength below 1.24 μm.[14]

Recently, many groups have reported approaches to attain self-assembled InAs QDs with a favorable energy separation and emission wavelengths, using InAlAs/InGaAs as the strain-reduced layers (SRLs).[15–17] In these works, self-assembled QDs with SRLs were grown at high temperatures. In segregation and interface intermixing can be effectively suppressed by inserting the InAlAs layer. InAs QDs that emit at long wavelengths and have a large energy separation have since then been successfully fabricated.[17]

This investigation elucidates the dependence of the energy separation and the emission wavelengths on the SRL structures of self-assembled InAs QDs. Various SRL structures that combine $In_{0.14}Al_{0.86}As$ and $In_{0.14}Ga_{0.86}As$ with the same total thickness were used. The optoelectronic characteristics of InAs QDs were studied by photoluminescence (PL) spectroscopy. The variations of the emission wavelength, the energy separation, and the luminescence intensity with the temperature of the samples were investigated to elucidate the behavior of carriers in InAs QDs.

II. EXPERIMENT

Semiconductor thin films were grown by molecular-beam epitaxy (Epineat) using solid sources. Four SRL structures were designed and their confinement effects on the carriers were investigated along with the associated optoelectronic characteristics of InAs QDs. A 0.3-μm buffer GaAs layer was first grown onto the (100) GaAs substrate at 600 °C. The samples were cooled to 480 °C to grow a nominally 2.6-monolayer (ML)-thick InAs QD layer at a growth rate of 0.26 Å/s, as determined by reflection high-energy electron diffraction (RHEED). The self-assembled InAs QDs were then covered with composite $In_{0.14}Al_{0.86}As/In_{0.14}Ga_{0.86}As$ SRLs. Each sample had a total thickness of 54 Å SRLs. The thicknesses of the InAlAs layers in samples A, B, C, and D, respectively were 0, 10, 20, and 54 Å. The samples were finally heated to 600 °C to grow a 0.3-μm GaAs cap layer. Figure 1 schematically depicts the vertical structures of the samples in band diagrams.

The samples were characterized by PL spectroscopy. A diode-pumped solid-state laser with a wavelength of 532 nm

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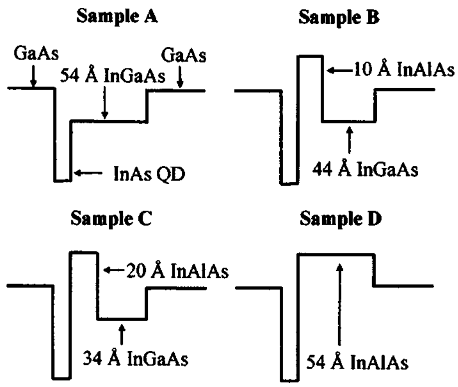

FIG. 1. Energy band diagrams of InAs QDs with various SRL structures for samples A to D.

was used for photon excitation. Luminescence from the sample was dispersed through a monochromator and detected using an InGaAs detector.

III. RESULTS AND DISCUSSION

Samples grown under the same conditions but without a GaAs cap layer were examined by atomic force microscopy (AFM). Figure 2 displays a $1.0\times1.0\ \mu m^2$ AFM image of the self-assembled InAs QDs covered with 10-Å InAlAs/44-Å InGaAs SRLs. The QD density is about $7\times10^{10} cm^{-2}$. The AFM image reveals that the uniformity and the density of the InAs QDs were satisfactory.

Figure 3 presents the PL spectra of the InAs QDs at 300 K. Each sample has two main features. The low-energy PL feature is related to the ground states of InAs QDs. The high-energy PL feature may result from the wetting layer, small InAs QDs with carriers in the ground states and the originally sized InAs QDs with carriers in the excited states. The smooth form of the high-energy PL feature excludes the possibility of emission from the thin wetting layer, which would yield a zigzaglike PL feature associated with the variation in the thickness of the monolayer. The PL intensity as a function of the excitation power indicated that the ratio of the intensity of the high-energy peak to that of the low-energy peak declined as the pumping power was increased at a low pumping power, but increased with pumping power at high pumping power. This dependence of the intensity ratio on the pumping power, determined by the distribution of excess carriers over various energy levels in a quantum-confined structure,[8] reveals that the excited-state transition of carriers in InAs QDs is responsible for the high-energy PL feature. That is, the two PL features are associated with the ground-state and the first excited-state transitions.

FIG. 2. $1.0\times1.0\ \mu m^2$ atomic force microscopic image of a sample with 2.6 ML InAs QDs covered with 10 Å InAlAs/44 Å InGaAs SRL.

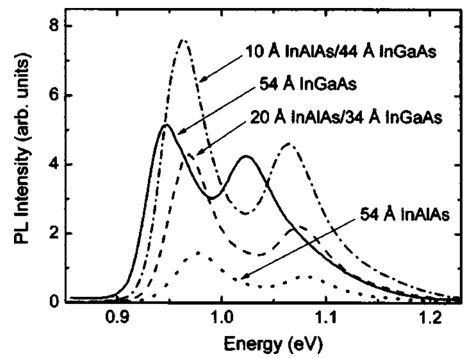

FIG. 3. PL spectra of samples A to D obtained at room temperature.

Figures 4(a)–4(c) plot the peak PL energies, the energy

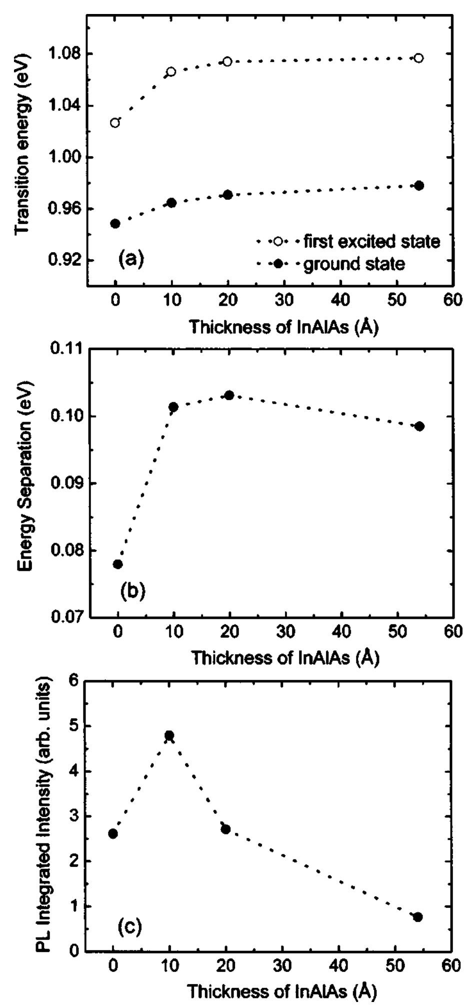

FIG. 4. (a) Energies of PL features associated with the ground state and the first excited state, (b) energy separations between PL features, and (c) ground-state PL-integrated intensities of InAs QDs in samples A to D, in terms of InAlAs thicknesses. The lines are guides for the eye.

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separations between the PL peaks, and the integrated ground-state PL intensity of samples A to D, respectively, in terms of InAlAs thickness, at room temperature. The ground-state transition for sample A with a 54-Å InGaAs SRL occurs at 0.946 eV or a wavelength of 1.31 μm; it has a full width at half maximum of 36 meV and an energy separation of 78 meV. The ground-state transition of sample B with 10-Å InAlAs/44-Å InGaAs SRLs is blueshifted by 16 meV from that of sample A. Further increasing the thickness of InAlAs, as in samples C and D, magnifies the blueshift. The blueshift of the InAs QD samples herein, with InAlAs SRLs grown at low temperature, is inconsistent with the reported redshifts of InAs QDs with InAlAs/InGaAs SRLs grown at high temperature.[15,16]

The structural dimensions and the confinement potentials determine the energy levels of quantum-confined structures. In an InAs/InAlAs/InGaAs system, the strain in InAs and the intermixing at the interface may resize the QD. However, the lattice constants of $In_{0.14}Ga_{0.86}As$ and $In_{0.14}Al_{0.86}As$ are within 0.2% of each other,[18] so the strain in InAs QDs with any combination of InAlAs/InGaAs SRLs is almost the same across all of the samples. Hence, the structural size and the energy levels, influenced by the strain effect, should vary very slightly among the InAs QDs covered with InGaAs, InAlAs, or InAlAs/InGaAs SRLs. Unlike the reported redshift caused by the segregation of indium and intermixing at the QD boundary,[15,16] the blueshift exhibited by the samples reveals that little indium segregation and interface intermixing occurred in the low-temperature-grown InAs QDs with SRLs. Therefore, the energy levels of the InAs QDs herein are dominated by the confinement potentials.

Figures 4(a) and 4(b) indicate that the ground-state and the first excited-state transitions in sample D are blueshifted by 29 and 50 meV, respectively, from those in sample A. The energy separation between the first excited state and the ground state is increased from 78 meV in sample A, which has a low confining InGaAs barrier, to 99 meV in sample D, which has a high confining InAlAs barrier. Although sample D has a large energy separation, its optoelectronic properties, including emission wavelength and intensity, are poorer than those of the other samples from the perspective of potential application to optoelectronic devices. The efficiency of luminescence and the optoelectronic properties can be improved by incorporating the composite InAlAs/InGaAs SRLs into the InAs QDs. The ground-state energy and the energy separation of sample B were 0.965 eV and 101 meV, respectively. As the thickness of the InAlAs layer was increased to 20 Å, as for sample C, the ground-state transition energy and the energy separation increased to 0.971 eV and 103 meV, respectively. These results are consistent with the theoretical analysis, which indicated that the higher energy level is blueshifted or redshifted farther than the lower energy level, as the quantum-confinement structure parameters are varied.

Figure 4(c) presents the ground-state PL-integrated intensities of samples A to D at room temperature. Sample B had the greatest integrated intensity. The PL intensities of samples B, C, and D declined as the thickness of the InAlAs layer increased. The PL intensity is well known to be proportional to the number of carriers in the QDs. The carriers in the QDs are photogenerated therein or are injected from the bottom buffer layers, the peripheral wetting layers, and the top cap layers.[19] In a reverse process, the carriers may thermally escape from the QDs. Basically, the number of carriers photogenerated inside a QD is small and approximately the same for each sample because the QDs are small and have about the same volume. Similarly, these four samples have an identical QD/buffer-layer structure, so the number of carriers transferred between the buffer layers and the QDs is almost the same for each sample. The InAs wetting layer has been demonstrated to serve as a channel through which carriers can be transferred among QDs and the barrier layers located above or under the wetting layer.[19] However, the InAs wetting layer in the samples in this study is so thin that its contribution to the carrier transfer is insignificant. Therefore, the variation among the emission spectra of the samples is related primarily to the differences among the top-side structures of the samples.

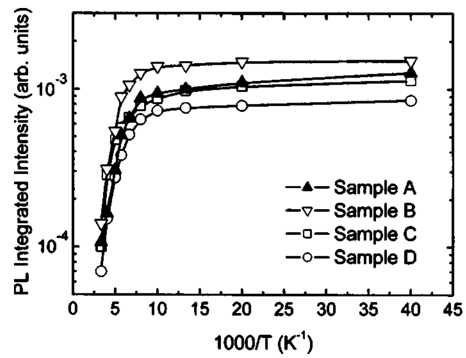

FIG. 5. Temperature dependence of the ground-state PL-integrated intensity of samples A to D. The lines are guides for the eye.

When the samples were illuminated, numerous carriers were photogenerated in the samples with thick cap layers. The sandwiched SRLs limit carriers entering the QD region. However, more carriers are injected into QDs by tunneling, so the PL intensity is higher for a sample with a thinner InAlAs layer. This assertion is consistent with the experimental results obtained for samples B–D. With respect to the band structures, QDs in sample A can be reasonably assumed to incorporate more injected carriers than the QDs in sample B. However, the carriers in sample A are more likely to escape from the QDs, because the difference between the energy of the ground state of the QDs and the band edge of the SRL is smaller for sample A than for sample B. Fewer carriers in QDs of sample A results in a lower PL intensity emitted from sample A than from sample B.

Figure 5 plots the ground-state PL-integrated intensity against the inverse of temperature from 25 to 300 K for samples A to D. The dependence of integrated PL intensity $I_{PL}(T)$ on temperature T can be expressed as[20]

$$I_{PL}(T) = C \exp(E_A/kT), \tag{1}$$

where C is a constant associated with the sample, E_A is the thermal activation energy of the loss mechanism, and k is the Boltzmann constant. The fitted activation energies for samples A to D are 54, 70, 77, and 69 meV, respectively. The fact that the activation energy of sample A is smaller than

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that of sample B is responsible for the stronger thermal quanching for the former, and this causes the PL intensity of sample A to be weaker. At low temperature, the action of thermal quenching declines more noticeably for sample A. The integrated PL intensity of sample A increases as the temperature decreases, whereas that of sample B remains almost invariant. Cooling samples to a sufficiently low temperature of 10 K (Refs 20 and 21) may cause the PL intensity of sample A to exceed that of sample B. This supports the argument that there are more carriers injected into sample A than into sample B.

The above results reveal that laying InAlAs/InGaAs composite layers over InAs/GaAs QDs promotes three-dimensional confinement and preserves the homogeneity of the islands. The sample of QDs with 10-Å InAlAs/44-Å InGaAs SRLs yielded the greatest PL intensity and a large energy separation of 101 meV. Hence, an InAs QD laser operated at a wavelength of 1.3 μm with a high characteristic temperature may be realized.

IV. CONCLUSION

This study investigated the optoelectronic characteristics of InAs QDs with a high-potential InAlAs/InGaAs SRL barrier using PL spectroscopy. The carrier transitions and optical characteristics of InAs QDs depend strongly on the thickness and composition of the SRLs. The ground state in QDs with InAlAs/InGaAs SRLs is blueshifted as the InAlAs thickness is increased. InAs QDs with 20 Å InAlAs/34 Å InGaAs SRLs exhibit a large energy separation of up to 103 meV between the ground-state transition and the first excited-state transition. The low-temperature growth of SRLs reduces the segregation of indium and interface intermixing during epitaxial process. Moreover, introducing a high potential SRL barrier into the QD structures enhances the effects of carrier confinement and the size uniformity of QDs, as verified by the PL spectra and AFM images of the samples.

ACKNOWLEDGMENT

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