Formation and optical characteristics of strain-relieved and densely stacked GaSb/GaAs quantum dots

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The authors report the formation and optical characteristics of type-II, strain-relieved, and densely stacked GaSb/GaAs quantum dots (QDs) using an interfacial misfit (IMF) growth mode. A moderate V/III ratio during the growth of GaSb QDs produces strain-relieved QDs facilitated by the IMF array without Sb segregation associated with defects and threading dislocations. In contrast, a low V/III ratio establishes conventional Stranski-Krastanov QDs. The strain-free nature of the IMF QDs allows densely packed, multistacked ensembles which retain very high crystalline quality demonstrated by x-ray diffraction, room-temperature photoluminescence, and electroluminescence. The possibility for dense stacking enabled by the strain-relieved growth mode may prove beneficial for QD sensors, emitters, and solar cells.
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GaSb quantum dots (QDs) in a GaAs matrix have drawn recent attention for their potential ability to demonstrate QD-based emitters at a technologically important wavelength of 1.55 μm along with their unique electronic and optical properties caused by their staggered (type-II) band alignment and large valence band offset.[1] The type-II staggered active region can be designed to work at a variety of wavelengths by varying the composition of the matrix surrounding GaSb QDs. These types of active regions have been shown to work very effectively in the mid-IR (Ref. 2) and the GaSb/GaAs QDs can be used for demonstrating similar results in the near-IR,[3–10] where no type-II devices have yet been demonstrated. In these type-II QDs, holes can be confined within QDs, while electrons produce quantum shell around QDs by the Coulomb interaction. Thus, they could be also useful for a single carrier, even unipolar storage devices such as optical memory owing to their longer radiative recombination lifetime.[3,5] So far, several groups have reported formation and optical properties of type-II GaSb/GaAs QDs using the Stranski-Krastanov (SK) growth mode with demonstrations of light emission from QDs and their wetting layers.[3–10] By definition, the SK growth mode of GaSb/GaAs produces highly strained QDs which is somewhat of a hindrance in device realization since it limits both peak wavelength (up to $\cong$1.2 μm) and dense stacking. Indeed, there have been few reports of the formation of multistacked GaSb QDs.[9]

In contrast to the SK QDs, it is also possible to form strain-relieved GaSb QDs using an interfacial misfit (IMF) based growth mode.[11] The two-dimensional IMF array, localized to the heterointerface, relieves strain energy due to lattice mismatch and has been noted to form a wide range of surface phenomena from strain-free islands to highly planar strain-free, defect-free bulk material.[11–13] The strain-free nature of the IMF growth mode is extremely attractive for QDs as it enables dense QD stacking with a spacer thickness of $t \sim 15$ nm, compared to SK QDs ($t \sim 30$ nm). The growth of IMF GaSb QDs has not been as straightforward as that of conventional InAs/GaAs QDs due to the rather narrow V/III ratio window and therefore not as well studied.[14,15] To date, the IMF GaSb QDs have primarily been investigated in crystal growth and materials characterization studies,[13] with only one report of photoluminescence[16] (PL) and there are no reports of electroluminescence (EL). Our group has recently demonstrated high optical quality in both SK and IMF growth modes with PL indicating quantized energy levels and blue shifting of the type-II QD emission peak.[16]

In this letter, we report the formation of IMF QDs, within a narrow V/III ratio, along with electrical and optical characteristics of strain-relieved, densely stacked IMF GaSb QDs. Room-temperature (RT) luminescence at a wavelength of 1.3 μm is achieved using both photopumping and current injection. We note that the strain-free nature of the IMF growth mode permits us to stack the ensembles with a spacer thickness of $t \sim 15$ nm while not propagating threading dislocations. The excellent crystalline quality of these densely packed stacked IMF QD ensembles is demonstrated using x-ray diffraction (XRD) analyses.

Samples are grown on semi-insulating GaAs (001) substrates by MBE with a 100 nm GaAs buffer. The GaSb QDs are formed at 510 °C on the As-rich GaAs surface with a growth rate of 0.32 ML/s with V/III ratios, $6.5 > \mathrm{V/III} \geqslant 2$ and $\cong$1, for IMF and SK modes, respectively. The samples for microscopic characterization are immediately cooled to RT. As for the PL characterization, the GaSb QDs are immediately capped with a 100 nm GaAs layer without growth interruption. Figures 1(a)–1(d) contrast QD shape and density for the IMF and SK growth modes. Figure 1(a) shows the atomic force microscopy (AFM) image of IMF QDs. The IMF QDs are elongated slightly along the [1–10] direction with a average length, width, and height of 50, 30, and 6 nm, respectively. On the other hand, the SK QDs display typical characteristics for more uniform QDs, as shown in Fig. 1(b), with an average width and height of 10 and 5 nm, respectively. The densities of IMF and SK QDs are 6×10^{10} and $3 \times 10^{10}/\mathrm{cm}^2$, respectively. Figures 1(c) and 1(d) show the high-resolution transmission electron microscope (TEM) images of a single surface QD using IMF and SK growth

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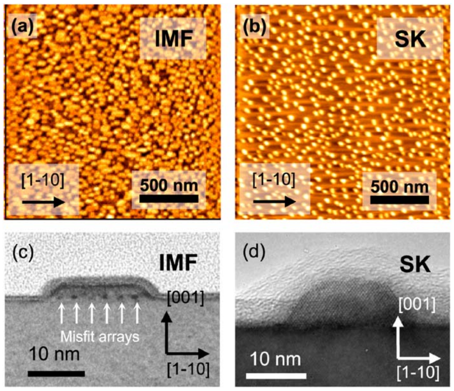

FIG. 1. (Color online) AFM images of GaSb QDs with a total coverage of 3 ML under (a) the V/III ratio of 10 resulting in IMF growth mode and (b) the V/III ratio of 1 resulting in SK QDs, and cross-sectional TEM images of (c) IMF GaSb QDs with strain relieving misfit dislocations that can be observed at the interface and (d) SK GaSb QDs. The densities of IMF and SK QDs are 6×10^{10} and $3\times10^{10}/\mathrm{cm}^2$, respectively.

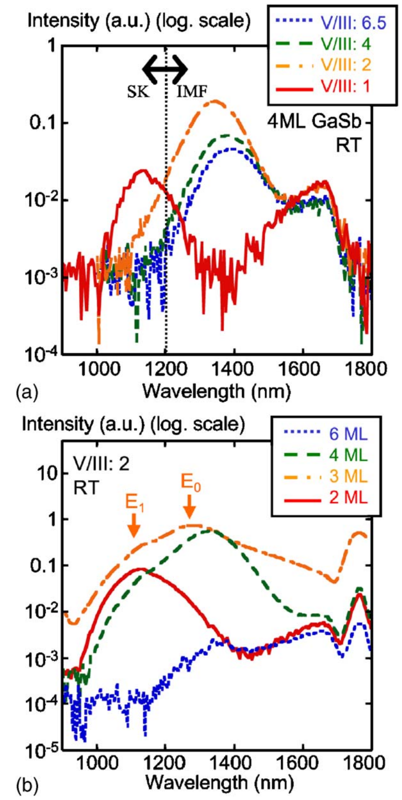

FIG. 2. (Color online) (a) RT PL spectra of single layer of GaSb QDs at various V/III ratios of the growth of GaAs QDs ranging from 1 to 6.5. The PL peak and FWHM of GaSb QDs at V/III ratios of 1, 2, 4, or 6.5 are 1.14 μm and 64 meV; 1.34 μm and 53 meV; 1.38 μm and 56 meV; and 1.39 μm and 53 meV, respectively. (b) RT PL spectra of single layer of GaSb QDs with the V/III ratio of 2 at a total coverage ranging from 2 to 6 ML. The PL peaks of GaSb QDs at a total coverage of 2, 3, 4, and 6 ML are 1.12, 1.28, 1.33, and 1.35 μm, respectively.

modes, respectively. The shape of the IMF QD is very flat and broad compared with that of the SK QD which displays a domelike shape typical of the SK growth mode. The dark line in Fig. 1(c) shows the native oxide formed on the sample between growth and analysis. The characteristic array of misfits can be observed at the GaSb/GaAs interface, where the space between the misfit dislocations is ~56 Å, as expected from the $\Delta a_o/a_o=7.8\%$.[13,17]

The RT PL spectra for both ensembles with a total coverage of 4 ML are collected using a conventional PL setup with an excitation by either a He–Ne or an Ar^+ laser with a 2 mm spot size. Figure 2(a) shows the RT PL spectra of GaSb QDs with an excitation by a He–Ne laser at V/III ratios of ranging from 1 to 6.5. When the V/III ratio of GaSb growth is 1, PL spectra indicates weak light emission near 1.14 μm with a full width at half maximum (FWHM) of 64 meV from highly strained SK QDs. Emission from large bulklike GaSb islands at 1.66 μm is also measured due to the coalescence of neighboring dots by an excess of GaSb materials. When the V/III ratio is more than 2, the PL peak of GaSb QDs shifts abruptly towards 1.3 μm (FWHM of $\cong$53 meV) because of the transition from strained SK to unstrained IMF growth mode. Moreover, by increasing the V/III ratio from 2 to 6.5, the PL peak shifts towards longer wavelengths from 1.34 to 1.39 μm, and the PL intensity decreases dramatically. An excess of Sb atoms during the formation of GaSb QDs increases Sb intermixing from GaSb QD layers into GaAs matrices, resulting in the extended wavelength, increased lattice mismatch, and strain in the barriers associated with defect formation and reduced PL intensity.

Figure 2(b) shows the PL spectra of GaSb QDs with a V/III ratio of 2 with an excitation by Ar^+ lasers (0.58 W) at total GaSb coverage ranging from 2 to 6 ML at RT. When the GaSb coverage is 2 ML, very weak PL can be observed at 1.12 μm. By increasing the GaSb coverage, the PL peak shifts towards longer wavelengths over 1.3 μm, and the peak intensity increases dramatically. Moreover, we can observe the excited state of GaSb QDs at 1.13 μm which grows with higher pump powers as the lower states fill.[16] On the other hand, light emission from large bulklike GaSb islands at 1.66 μm is also observed here. We also note a second order GaAs peak that is visible at 1.75 μm at higher excitation by Ar^+ lasers. However, increased GaSb coverage results in planar growth and semi-two-dimensional GaSb layers using the IMF.

Optical characteristics and XRD analyses of ten stacked GaSb QDs separated by 15 nm GaAs spacer layers are also studied at RT with an excitation by Ar^+ laser. Figure 3(a) shows the PL spectra of ten stacked GaSb QDs with V/III ratios of (i) 2 and (ii) 6.5. The PL spectrum (i) with a higher V/III ratio shows very low PL intensity because an excess of Sb adatom within stacked ensembles causes the formation of large defects, resulting in surface undulations after stacking QDs or the formation of threading dislocations. On the other hand, the PL spectrum (ii) displays an improved PL intensity due to the improved crystalline quality of stacked QD ensembles by the suppression of Sb segregation, reduced defect formation, and threading dislocations. Strong PL at 1.34 μm from ten stacked GaSb QDs is obtained at RT with a FWHM of 104 meV. Figure 3(b) shows XRD spectra from ten stacked GaSb QDs with V/III ratios of (i) 2 and (ii) 6.5 using symmetric scans around the (004) reflection in $\omega/2\theta$ geometry. Improved crystalline quality is evident in the narrowing FWHM of the zero-order peak from (i) 330 arc sec to (ii) 103 arc sec by decreasing the V/III ratio during the growth of GaSb QDs, which elucidates the reduction of the formation of threading dislocations or defects. The zero-order peak ($\Delta\theta$) in the XRD spectrum (i) is located closer to the peak of GaAs substrates ($\Delta\theta=-539$ arc sec) compared with the XRD spectrum (ii) ($\Delta\theta=-472$ arc sec). In addition, these values

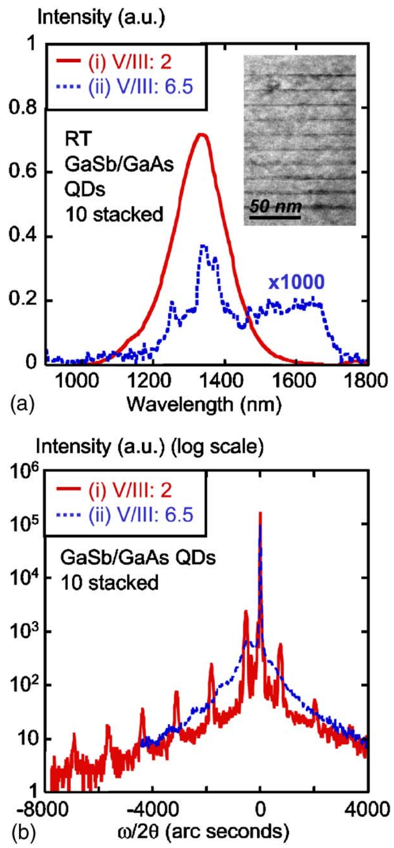

FIG. 3. (Color online) (a) RT PL spectra of ten stacked GaSb QDs at various V/III ratios of GaSb growth of (i) 2 and (ii) 6.5. The spacer thickness between each QD layer is 15 nm. The inset is a cross-sectional TEM image of ten stacked GaSb QDs. (b) Symmetric 004 x-ray diffraction pattern for ten stacked GaSb QDs at V/III ratios of (i) 2 and (ii) 6.5.

are much lower than those of conventional stacked InAs/GaAs QDs although InAs on GaAs has a lattice-mismatched interface similar to GaSb on GaAs.[18] This indicates that the overall compressive strain from GaSb QDs can be relaxed via strain relieving misfit at the interface between GaSb QDs and GaAs, resulting in excellent crystalline quality of densely stacked QD ensembles.

The QD emitters consisting of six stacked GaSb/GaAs IMF QDs as a active layers separated by $t=15$ nm GaAs layers are grown on a (100) n-GaAs substrate followed by a 1.46 μm n-$Al_{0.3}Ga_{0.7}As$ cladding layer grown at 560 °C, the active layer, a 1.46 μm p-$Al_{0.3}Ga_{0.7}As$ cladding layer grown at 510 °C, and a 50 nm p^+-GaAs contact layer. GeTe and Be are used as n- and p-type doping materials, respectively. Broad area edge emitters, 50 μm wide with a cavity length of 2 mm, are fabricated for measurements of the device characteristics. Figure 4 shows RT EL spectra of the QD emitters that have as-cleaved facets on both sides at various injected current densities under pulsed conditions (10% duty cycle) ranging from 200 A/cm^2 to 1.5 kA/cm^2. We can observe a single peak at a wavelength of 1.28 μm at RT from six stacked strain-relieved GaSb QDs. EL peaks show almost the same wavelength as that of PL spectra in Fig. 2(b), which indicates that observed PL and EL spectra originate from the GaSb QDs.

In summary, we report the formation and optical characteristics of strain-relieved and densely stacked GaSb QDs by using the IMF growth mode. In the growth of GaSb/GaAs QDs, a moderate V/III ratio ($2 \leq V/III < 6.5$) produces

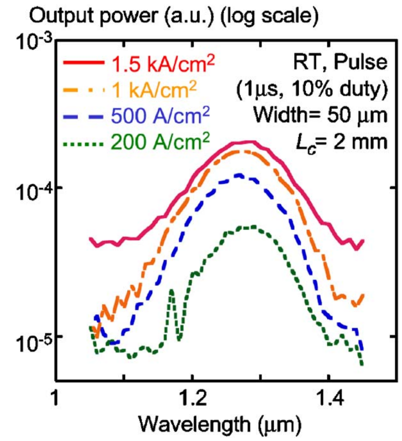

FIG. 4. (Color online) RT EL spectra of QD emitters consisting of six stacked strain-relieved GaSb QDs at various injected current densities ranging from 200 A/cm^2 to 1.5 kA/cm^2.

strain-relieved QDs by IMF emitting at over 1.3 μm and the low V/III ratio of $\cong 1$ establishes highly strained QDs by SK emitting at 1.14 μm. Further increases in the V/III ratio lead to Sb segregation with associated defects and threading dislocations. Densely packed stacked IMF QD ensembles demonstrate excellent crystalline quality along with RT EL at 1.3 μm. The possibility for dense stacking ($t \sim 15$ nm) of GaSb QDs enabled by the strain-relieved IMF growth mode can prove beneficial for QD sensors, emitters, and solar cells.

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