Self-formation of ultrahigh-density (10^{12} \text{cm}^{-2}) InAs quantum dots on InAsSb/GaAs(001) and their photoluminescence properties

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Ultrahigh-density quantum dots (QDs) have attracted considerable interest with regard to optoelectronic device applications. In particular, intermediate-band solar cells (IBSCs) have a high power conversion efficiency and could represent the next generation of solar devices.\(^1\)

Using a detailed balance model,\(^2,3\) we have recently calculated the QD density dependence of IBSCs based on InAs QDs on GaAsSb.\(^4\) A total QD density of $3 \times 10^{13} \text{cm}^{-2}$ is required to have a high-power conversion efficiency of at least 40% under concentrated sunlight. However, normal self-assembled QD densities are about $5 \times 10^{10} \text{cm}^{-2}$, thus requiring over 600 stacked layers. Hence, there has been much effort in the growth of closely stacked QD layers for IBSCs.\(^5-12\) There have been a few reports concerning ultrahigh-density in-plane QDs.\(^13-15\) Recently, we have reported InAs QDs with an ultrahigh density of $5 \times 10^{11} \text{cm}^{-2}$ on GaAsSb buffer layers on GaAs(001) substrates, fabricated by Stranski–Krasnov (SK)-mode molecular beam epitaxy (MBE).\(^16\) The QD density was increased by suppressing the coalescence and ripening of neighboring QDs.\(^16,17\) The maximum density was approximately $5 \times 10^{11} \text{cm}^{-2}$, and little is known about their fundamental properties at these densities. For example, they may have different optical and electronic properties because of in-plane coupling. Therefore, the characterization of carrier dynamics in laterally coupled QDs is very important for device applications.

To increase the QD density, the initial three-dimensional (3D) nucleation should be enhanced on the two-dimensional (2D) wetting layer (WL) just before QD formation. Here, an InAsSb WL deposited on a GaAs(001) buffer layer was precisely controlled to maximize the density of subsequently deposited InAs QDs. The QD photoluminescence (PL) provided insight on lateral coupling and carrier dynamics.

The samples were grown on semi-insulating GaAs(001) substrates by MBE. After a GaAs buffer layer was grown at 590 °C, the temperature was reduced to 450, 460, or 470 °C. An InAsSb WL was then grown, followed by InAs. The InAsSb coverage was varied from 0.6 to 1.5 monolayers (ML), keeping the total InAsSb and InAs coverage constant at 2.2 ML. The InAs growth rate was 0.035 ML/s, and 3D islands spontaneously formed at approximately 1.6 ML through the SK growth mode. This critical thickness

was approximately 8.6% smaller than that for InAs growth on InAsSb/GaAs(001). After the growth of InAs QDs, the sample was annealed at the growth temperature for 15 s under arsenic pressure. Next, either a 60 nm GaAs or 57 nm GaAs/3 nm GaAsSb capping layer was grown. The GaAsSb capping layer forms a type-II band alignment and reduces the compressive strain of the InAs QDs. From X-ray diffraction data, the Sb content was 12–14% in the GaAsSb capping layer, which is very close to the type-II band alignment of the GaAsSb/InAs heterointerface.\(^18\)

Figures 1(a) and 1(b) respectively show the density and average height of InAs QDs grown at 470 °C as a function of InAsSb WL coverage. The dotted lines represent QDs grown on GaAsSb buffer layers at 470 °C. AFM images show InAs QDs grown on (c) an InAsSb WL (1.25 ML), (d) a GaAsSb buffer layer, and (e) a GaAs buffer layer.
10^{11} \text{ cm}^{-2}$, and the average height was approximately 2.2 nm. From this result, an InAsSb coverage of 1.25 ML was selected in the following experiments.

Figure 2(a) shows the growth temperature dependence of the InAs QD density on a 1.25 ML InAsSb WL, for Sb pressures of $2.1 \times 10^{-7}$ and $3.5 \times 10^{-7}$ Torr. The QD density increases with increasing Sb pressure and decreasing growth temperature. Note that a QD density of $10^{12} \text{ cm}^{-2}$, which is the highest density on GaAs, was obtained at 460 and 450 °C. Figure 2(b) shows AFM images of the InAs QDs at this density. The average lateral size of the QDs was 11 nm, as measured from a large-area AFM image. Figure 2(c) shows the height distribution of the InAs QDs in Fig. 2(b). The average QD height was 1.7 nm. Despite the extremely high QD density, the formation of coalesced giant dots was suppressed by the Sb surfactant effect.\(^{17}\)

To obtain a better understanding of high-density nucleation, the InAsSb WL surface structure was imaged by AFM. Figures 3(a) and 3(b) show AFM images of 1.25 ML InAs WLs grown on GaAs buffer layers at 500 and 460 °C, respectively. When the InAs coverage was 2.2 ML, the InAs QD densities were $4 \times 10^{10} \text{ cm}^{-2}$ at 500 °C and $2 \times 10^{11} \text{ cm}^{-2}$ at 460 °C. As shown in Figs. 3(a) and 3(b), the 2D InAs islands are smaller, and the density is higher at 460 °C than at 500 °C. The 2D layer structure is a result of the short surface migration at the lower growth temperature. The 2D islands frequently induce 3D nucleation. Figure 3(c) shows an AFM image of a 1.25 ML InAsSb WL grown on GaAs(001) at 460 °C. Many small 2D InAs islands are formed along the [110] direction and provide long step edges. The ratio of step edge lengths in Figs. 3(a)–3(c) is 1.0 : 1.5 : 2.0. The anisotropic shape of 2D islands is caused by the surface reconstruction and by the anisotropy of adatom migration. Typically, the migration length is larger in the [\(\overline{1}10\)] direction than in the [110] direction.\(^{19}\) A large lattice mismatch between InAsSb and GaAs increases the compressive strain of InAsSb islands, as compared with that of InAs islands. Thereby, the lateral expansion of InAsSb islands is suppressed to reduce the strain energy. It is considered that such stepped surfaces and Sb surface atoms prevent the adatom migration because of the modified surface potential barrier. In addition, step edges are 3D nucleation sites for strained islands because they relieve strain.\(^{20,21}\) Therefore, the InAsSb WL structure enhances the in-plane density of 3D islands. Furthermore, segregated Sb surface atoms suppress the coalescence of neighboring 3D islands,\(^{17}\) and the small islands prevent ripening.\(^{15}\) Therefore, the ultrahigh-density InAs QDs were derived from multiple effects.

Low-temperature PL spectra (Fig. 4) were acquired for ultrahigh-density ($10^{12} \text{ cm}^{-2}$) InAs QDs embedded in either a GaAs capping layer (sample B) or a GaAsSb layer (sample C). The PL spectra were compared with that for low-density ($5 \times 10^{10} \text{ cm}^{-2}$) InAs QDs embedded in a GaAs capping layer (sample A). Figure 4 shows the optical excitation power dependences of the normalized PL spectra at 15 K, where the PL intensities increased with excitation power. For sample A [Fig. 4(a)], the peak PL wavelength based on ground states remained constant at 1167 nm. A subpeak at 1100 nm from excited states was enhanced at a higher excitation power, which is a characteristic of the separated low-density QDs. The broadened PL spectra of samples B and C [Figs. 4(b) and 4(c), respectively] did not feature these subpeaks based on the excited state transitions. Instead, the PL peak from sample B shifted continuously to shorter wavelengths with increased excitation power, and the short-wavelength shoulder shifted more than the long-wavelength shoulder. The blue shift of the PL peak was also observed for sample C [Fig. 4(c)]. However, the long- and
short-wavelength shoulders had similar shifts. The continuous blue shift of the short-wavelength shoulder in the PL spectra of samples B and C can be understood as follows. The distance between neighboring QDs is a few nanometers, which is smaller than the carrier diffusion length. Hence, the carriers can tunnel between neighboring QDs. Because the optical excitation power density per QD was not high for samples B and C, the photoexcited carriers are sequentially occupied from the lower ground states of the larger QDs to the higher ground states of smaller QDs. This ground-state-filling effect due to tunneling results in the continuous blue shift of the short-wavelength shoulder in the PL spectra.

Figure 5 shows time-resolved PL spectra of samples B and C at 15 K. The excitation wavelength was 785 nm, and the optical power of the 300 ps laser pulse was 300 mW. Sample B has a type-I band alignment at the GaAs/InAs QD heterointerface, while sample C has a type-II band alignment at the GaAsSb/InAs QD. PL from sample B [Fig. 5(a)] was detected at 1000 nm. The biexponential PL decay curve was fitted with 0.7 and 1.9 ns decay times. PL from sample C was detected at 1140 [Fig. 5(b)], 1216 [Fig. 5(c)], and 1250 nm [Fig. 5(d)]. At longer wavelengths, the fast decay component can be neglected in Figs. 5(c) and 5(d); the long decay times are 4.6 ns at 1216 nm [Fig. 5(c)] and 5.3 ns at 1250 nm [Fig. 5(d)]. The PL transitions at long wavelengths and decay times can be attributed to an indirect transition of the type-II band alignment of GaAsSb/InAs. However, at short PL wavelengths of 1040–1080 nm, fast decay components are also observed. For example, in Fig. 5(b), the short decay time is 0.81 ns, and the long decay time is 3.31 ns. The 3.31 ns time is attributed to the indirect transition of the type-II band structure, while the 0.81 ns time is attributed to a direct transition in the InAs QDs, and corresponds to the decay component of sample B [Fig. 5(a)]. Thus, the short-wavelength PL components for sample C include contributions from direct and indirect transitions, as discussed below.

Schematic energy-band structures for small and large InAs QDs in sample C are shown in Fig. 6(a). When the QD size decreases, the holes are confined to the GaAsSb valence band because of the type-II band structure and the higher energy levels of InAs QDs. Therefore, indirect transitions occur only for small QDs. However, in large QDs, holes in the valence band are distributed over the GaAsSb capping layer and InAs QDs with lower energy levels. Therefore, both indirect and direct transitions occur in large QDs. For example, the PL spectrum of sample C has three components: indirect transitions of small QDs and indirect and direct transitions of large QDs. The indirect transitions of the small QDs appear at the intermediate and short wavelengths, whereas the indirect and...
direct transitions of the large QDs appear respectively at long and short wavelengths. Hence, the width of the PL spectrum depends on the energy difference between the indirect and direct transitions of the large QDs. The continuous PL blue shift at the long wavelengths with higher excitation powers in Fig. 4(c) for sample C is thus a result of the ground-state filling of the InAs conduction band and the higher energy distribution of holes in the GaAsSb valence band.

Figure 6(b) shows the excitation power dependences of the full width at half maximum (FWHM) of the PL peak for samples A, B, and C. The FWHM for sample A, based on ground states, was almost constant (31 meV) under a relative excitation power below 200. It increased only slightly above 200 because of the excited-state subpeak. For sample B, with a type-I band structure, the PL FWHM increased with excitation power because of the ground-state filling discussed above. The PL FWHM for sample C decreased for a relative excitation power of above 30. This happened because the continuous PL blue shift occurred for both short and long wavelengths, and the blue shift is larger for the long wavelengths than for the short wavelengths. Thus, the decrease in FWHM is attributed to the type-II band indirect transitions of large QDs.

In conclusion, ultrahigh-density (10\(^{12}\) cm\(^{-2}\)) InAs QDs were grown on a 1.25-ML-thick InAsSb WL that was grown at 460 °C on GaAs(001) by MBE. The high density was attributed to the presence of a large number of 2D InAsSb islands on the GaAs buffer layer. The PL peak energy of these QDs shifted continuously to higher values at increased optical excitation intensities. This was attributed to the filling of inhomogeneous ground states via tunneling between QDs. The ultrahigh-density InAs QDs were embedded in a GaAsSb capping layer. Indirect PL transitions in a type-II band structure were observed for small QDs. Large QDs exhibited both indirect and direct transitions at high optical excitation powers. As a result, the PL FWHM decreased with increasing excitation.

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