Self-assembled InAsSb quantum dots on (001) InP substrates

Yueming Qiu[a] and David Uhl
Jet Propulsion Laboratory, California Institute of Technology, Pasadena, California 91109

(Received 29 September 2003; accepted 6 January 2004)

Self-assembled InAsSb quantum dots (QD) on (001) InP substrates have been grown using metalorganic vapor phase epitaxy. The dot density and size are found to be strongly dependent on the presence of arsine. Direct deposition of InSb on InP and GaSb substrates formed large islands of InSb with low density of less than $5\times10^9/\mathrm{cm}^2$, however, InAsSb QDs of density as high as $4\times10^{10}/\mathrm{cm}^2$ could be self-assembled by alternating group III and group V precursors, and high density almost pure InSb QDs were achieved on $In_{0.53}Ga_{0.47}As$/InP. The formation of high density InAsSb QDs is a result of a local nonequilibrium process and a reduction in mobility of In adatoms on the growth surface due to the presence of arsenic atoms, and in the case of high density almost pure InSb QDs on InGaAs/InP, the InAs interface layer is believed to be responsible. Photoluminescence shows that InSb QDs emit light at room temperature in the range of 1.7–2.2 μm.
 [DOI: 10.1063/1.1655690]

Quantum-dot (QD) nanostructures have attracted considerable interest due to their superior physical properties expected from three-dimensional confinement. Most work has focused on the InAs/GaAs material system at wavelength of 1.3 μm (Refs. 1–3) and InP-based InAs QDs to obtain light emitters in the telecom wavelength region ($\sim$1.55 μm).[4] Recently, attempts to use InAs nanostructures based on InP substrates to extend the wavelength further into the infrared region of 1.8–2.3 μm have received more attention, because lasers are attractive for application in molecular spectroscopy, remote sensing of atmospheric and planetary gases, as well as lidar atmospheric detection and ranging. InAs QDs and quantum-dash lasers have been demonstrated recently at various wavelengths from 1.60 to 2.04 μm.[4–7] The emission wavelength of InAs QDs on InP seems to be limited to 2.05 μm, which is very similar to that of highly strained InGaAs quantum wells on InP due to the limit in critical thickness.[8]

In(As)Sb and related compounds based on GaSb substrates have been investigated extensively using metalorganic vapor phase epitaxy (MOVPE) to obtain mid infrared emission.[9] There are only limited reports on In(As)Sb QDs so far, but photoluminescence (PL) at wavelengths as long as 3.5 μm was reported from InSb QDs in an InAs matrix,[10] and InSb QDs with density as high as $4\times10^{10}/\mathrm{cm}^2$ on InP have been achieved by molecular-beam epitaxy (MBE).[11] Nevertheless, typical InSb QDs self-assembled using MOVPE have area density of usually less than $5\times10^9/\mathrm{cm}^2$. QD nucleation in MOVPE is basically an equilibrium process, while in MBE it is a nonequilibrium process. Additionally, the InSb QD growth kinetics are quite different in the two cases, in MOVPE where high growth temperature is needed to crack the precursors but the InSb bond is very weak, In adatoms have high mobility, resulting in big InSb islands of low density. In MBE however, an In sticking coefficient of near unity is expected due to the low growth temperature. Therefore, the key to obtaining self-assembled high density InSb QDs by MOVPE is to use a local nonequilibrium process and reduce the mobility of In adatoms on the growth surface.

In this letter, we present the growth of InAsSb and InSb QDs on InP, where the dot density and size are found to be strongly dependent on the presence of arsine. InAsSb QDs with density as high as 4×10^{10} cm^2 were self-assembled by alternating group III and group V precursors, and high density almost pure InSb QDs were achieved on InGaAs/InP. PL measurement shows that the almost pure InSb QDs emit light at room temperature in the range of 1.7–2.2 μm.

The QDs were self-assembled using low-pressure metalorganic vapor phase epitaxy. Trimethylindium (TMIn), triethylantimony (TMSb), triethylgallium, AsH_3, and PH_3 were used as precursors, and H_2 as the carrier gas. Growth temperatures were typically in the range of 480–550 °C for the QD layers, and 650 °C for the rest of the structures at reactor pressure of 100 Torr. First, InSb QDs were self-assembled directly on (001) InP substrates using a conventional method so that both TMIn and TMSb precursors were supplied simultaneously for InSb growth, and then cooled down under TMSb protection. Atomic force microscopy (AFM) scans revealed that even at growth temperature as low as 480 °C, the InSb islands that formed are very large in size (lateral width of 130 nm, height of 10 nm) with typical area density of less than $3\times10^9/\mathrm{cm}^2$, as shown in Fig. 1. For comparison, InSb QDs were also grown on a (001) GaSb

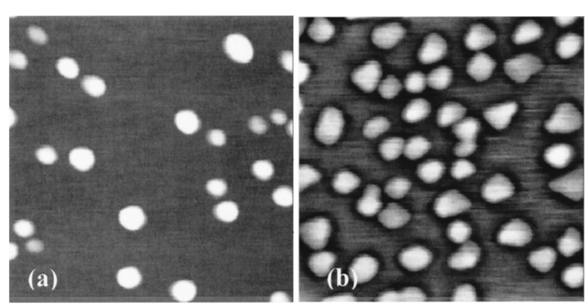

FIG. 1. 1 μm$\times$1 μm AFM scans of InSb islands self-assembled directly on (a) (001) InP and (b) (001) GaSb.

[a]Electronic mail: yueming.qiu@jpl.nasa.gov

substrate, where the lattice mismatch was about 6.3% compared to the 10.4% between InSb and InP. The density of InSb islands increased to about $5\times10^9/\mathrm{cm}^2$ with their average size similar to that of InSb on the InP substrate, however, the shapes of the islands show some degree of irregularity. The degree of QD nucleation is not well defined and thus the transition between coherent and incoherent is not known here. However, it is clear that InSb directly grown either on InP or GaSb results in large islands of InSb with low density of less than $5\times10^9/\mathrm{cm}^2$. This phenomenon suggests that the mobility of In adatoms is sufficiently high to enable In adatoms to migrate easily on the growth surface, and this can be characterized by the mean adatom diffusion length estimated from the average island distance.[12]

Unlike MBE where the QD growth temperature can be significantly low, the growth temperature of 480 °C is near the low limit for MOVPE growth. Although an even lower growth temperature is possible if one uses trisdimethylaminoantimony,[13] considering our purpose, i.e., future growth of an InAsSb QD laser structure on a InP substrate rather than on an InSb QD layer, QD growth temperature above 500 °C is preferred in our case. An effective way in which to reduce the mobility of In adatoms is then to introduce arsenic atoms on the growth surface, since the In–As bond is about 30% stronger than the In–Sb bond. With the use of alternate precursors to separate group III and group V species, therefore forming a local nonequilibrium process, two slightly different approaches were used to grow InAsSb QDs on a lattice matched $In_{0.53}Ga_{0.47}As$ buffer layer on the (001) InP substrate. The basic valve on and off sequences of TMIn and AsH_3 are the same as those for InAs QD growth described previously.[14] In the first approach only the TMSb valve opens for 1–30 s after the AsH_3 valve is closed to allow Sb/As atoms to exchange (we call this antimonidation) and form an InAsSb layer. In the second approach the TMSb valve has the same on and off sequences as those of the AsH_3 valve, thus it can allow both As and Sb atoms to bond to the In-atom terminated surface and form an InAsSb layer. Similar to InAs growth, the grown InAsSb layer thickness per cycle is not exactly 1 monolayer (ML), actually it does increase almost monotonously with the TMIn valve open time as well as with the TMIn flow rate. The samples for PL measurement were embedded in an $In_{0.53}Ga_{0.47}As$ well with total thickness of 7 nm and a cap layer of InP. Figure 2 shows room temperature PL spectra of samples obtained by these two approaches. In both cases, by either increasing the antimonidation time or the TMSb/AsH_3 flow ratio, a very surprising blueshift from 2.0–2.05 to 1.80–1.83 μm was observed accompanied by improved luminescence efficiency. Because the luminescence that peaked near 2.0–2.05 μm was from InAs QDs,[14] it is reasonable to conclude that the luminescence that peaked at 180–1.83 μm was from InAsSb QDs, but ones of smaller size, which will be further confirmed by AFM scans.

The uncapped samples were cooled down under TMSb/AsH_3 protection and examined with the AFM. Figure 3 shows AFM scan images of InAs QDs and InAsSb QDs. The dot densities in both cases are about the same, $4\times10^{10}/\mathrm{cm}^2$ for InAs QDs and $3.7\times10^{10}/\mathrm{cm}^2$ for InAsSb dots, with the InAs QDs having a typical lateral size of 35 nm and height of 6–7 nm, but the InAsSb QDs have a typical lateral size of 37 nm but height of only 4 nm. Although the capped and uncapped QDs may differ in size, the difference in size of InAs QDs and InAsSb QDs agrees with the blueshift observed in PL spectra.

It has not been possible to determine the compositions of the InAsSb QDs in these approaches so far; however, by

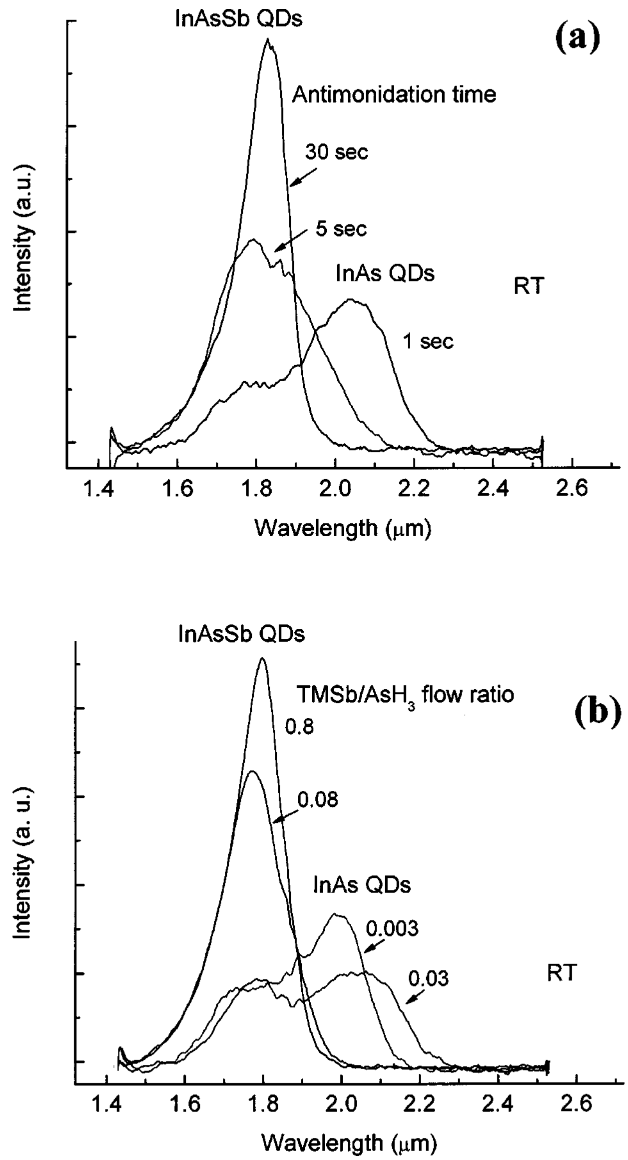

FIG. 2. Room temperature PL spectra of InAs and InAsSb QDs, showing the transition from InAs QDs to InAsSb QDs resulting from (a) antimonidation and (b) the alternating supply for direct growth.

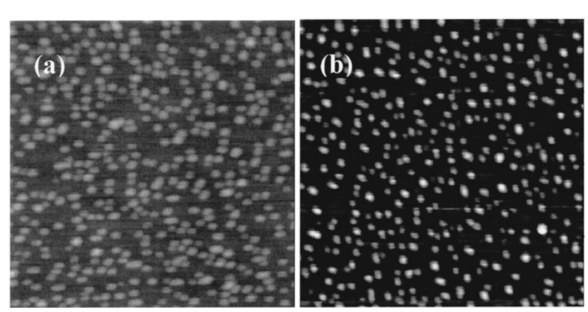

FIG. 3. 1 μm$\times$1 μm AFM scans of InAs and InAsSb QDs self-assembled on InGaAs/(001) InP.

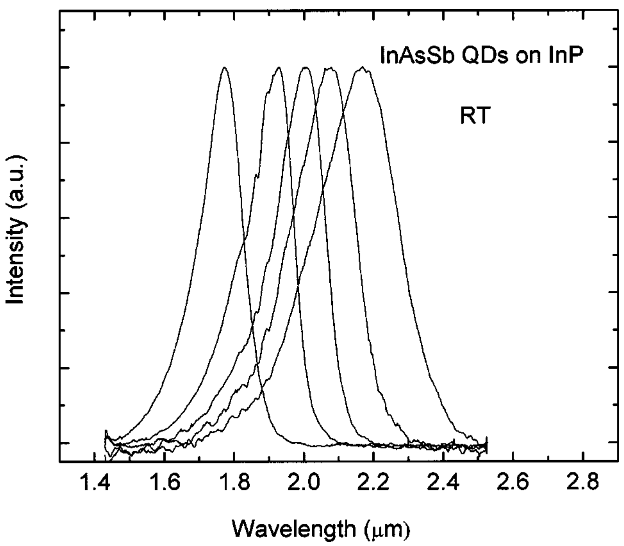

FIG. 4. Normalized room temperature PL spectra of almost pure InSb QDs self-assembled on InGaAs/(001) InP.

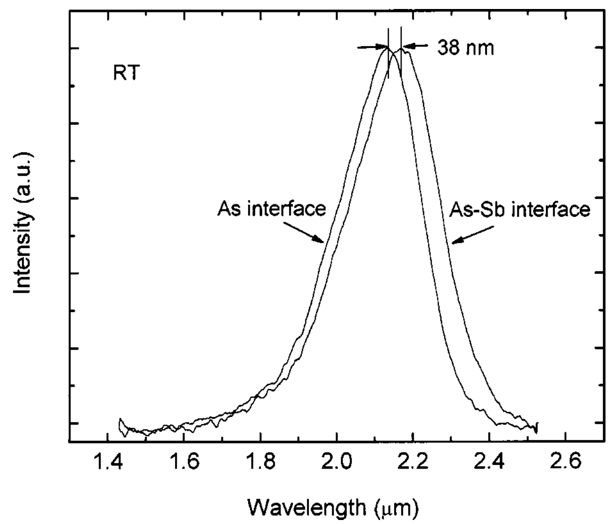

FIG. 5. Room temperature PL spectra of almost pure InSb QDs grown on (1) unmodified and (2) modified by antimonidation InGaAs buffer layers.

reducing the AsH_3 flow in the growth loop, almost pure InSb QDs can be grown using the second approach. These almost pure InSb QDs emit luminescence at room temperature in the wavelength range of 1.7–2.2 μm, as shown in Fig. 4. Given the fact that only little more than 2 ML of InSb is needed to form InSb QDs on the $In_{0.53}Ga_{0.47}As$/InP substrate due to the huge lattice mismatch (10.4%), the interface layer that is InAs formed during TMIn impinging on the As-atom terminated surface plays a key role in the nucleation of QDs. Therefore, we call them almost pure InSb QDs rather than InSb QDs, even though there were only TMIn and TMSb precursors supplied alternately during QD growth. Without this InAs interface layer, the dots that formed would be similar to those shown in Fig. 1(a).

The existence of this InAs interface layer can be further confirmed by antimonidation of the InGaAs layer to convert the interface from As atom terminated to As–Sb atom terminated before InSb QD growth. Such an experiment would result in a noticeable redshift in the PL spectra of the QDs, as shown in Fig. 5; a redshift in the range of 34–38 nm was observed after antimonidation of the interface at the growth temperature for only 2 s under TMSb flow of 50 sccm.

In summary, high density self-assembled InAsSb and InSb QDs on $In_{0.53}Ga_{0.47}As$/InP substrates were demonstrated using MOVPE. The dot density and size are found to be strongly dependent on the presence of arsine. The formation of high density InAsSb QDs is a result of a local nonequilibrium process and reduction in the mobility of In adatoms on the growth surface due to the presence of arsenic atoms, and, in the case of high density almost pure InSb QDs on InGaAs/InP, the InAs interface layer is believed to be responsible. PL measurement shows that the almost pure InSb QDs emit light at room temperature in the range of 1.7–2.2 μm, which can be further extended by antimonidation to the As-atom terminated InGaAs surface.

The authors are grateful to Dr. Nikzad Toomarian for his encouragement and support. The research described in this letter was carrier out at the Jet Propulsion Laboratory, California Institute of Technology, and was sponsored by the Bio-Nano Program and the National Aeronautics and Space Administration.

[1] D. L. Huffaker, G. Park, Z. Zou, O. B. Shchekin, and D. G. Deppe, Appl. Phys. Lett. **73**, 2564 (1998).
[2] K. Mukai, Y. Nakata, K. Otsubo, M. Sugawara, N. Yokoyama, and H. Ishikawa, IEEE Photonics Technol. Lett. **11**, 1205 (1999).
[3] Y. Qiu, P. Gogna, S. Forouhar, A. Stintz, and L. Lester, Appl. Phys. Lett. **79**, 3570 (2001).
[4] R. H. Wang, A. Stintz, P. M. Varangis, T. C. Newell, H. Li, K. J. Malloy, and L. F. Lester, IEEE Photonics Technol. Lett. **13**, 767 (2001).
[5] R. Schwertberger, D. Gold, J. P. Reithmaier, and A. Forchel, IEEE Photonics Technol. Lett. **14**, 735 (2002).
[6] Y. Qiu, D. Uhl, R. Chacon, and R. Q. Yang, Appl. Phys. Lett. **83**, 1704 (2003).
[7] T. Rotter, A. Stintz, and K. J. Malloy (unpublished).
[8] S. Forouhar, A. Ksendzov, A. Larsson, and H. Temkin, Electron. Lett. **28**, 1431 (1992).
[9] R. M. Biefeld, Mater. Sci. Eng., R. **36**, 105 (2002).
[10] A. Norman, N. Mason, M. Fisher, J. Richardson, A. Krier, P. Walker, and G. Booker, Microsc. Semicond. Mater. **157**, 353 (1997).
[11] T. Utzmeier, P. A. Postigo, J. Tamayo, R. Garcia, and F. Briones, Appl. Phys. Lett. **69**, 2674 (1996).
[12] M. Zinke-Allmang and S. Stoyanov, Jpn. J. Appl. Phys., Part 2 **29**, L1884 (1990).
[13] D. L. Partin, M. Pelczynski, P. Cooke, L. Green, J. Heremans, and C. M. Thrush, J. Cryst. Growth **195**, 378 (1998).
[14] Y. Qiu and D. Uhl, J. Cryst. Growth **257**, 225 (2003).