Formation and shape control of InAsSb/InP (001) nanostructures
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Adatom condensation and quantum dot sizes in InGaAs/GaAs (001)
Semicontactor nanostructures, such as quantum dots (QDs), quantum dashes (QDashes), and quantum wires (QWRs), have attracted wide attention due to their applications in devices. \cite{1,2} Most of the work on QDs and QWRs are focused on GaAs- and InP-based In(Ga)As nanostructures with the target at 1.3 and 1.55 μm emission wavelength. \cite{3,4} Recently, much attention has been devoted to extending the emission wavelength of InP-based InAs nanostructures into 1.8–3 μm region due to their use in military, telecommunications, molecular spectroscopy, biomedical surgery, environmental protection, and manufacturing industry applications. However, it is very hard to extend the emission wavelength of InP-based In(Ga)As nanostructures above 2 μm due to the limitation of InAs/InP material system.

Incorporating some antimony (Sb) into InAs nanostructures to form InAsSb nanostructures provides a very promising way to achieve the 2–3 μm emission wavelength. Theoretical calculations showed that the emission wavelength of InP-based InAs QDs can be extended into the 2–3 μm range, even reaching 5 μm.\cite{5} However, because of the difficulty in growing antimony compounds, limited experimental work has been done on the growth of InAsSb QDs on InP substrates.\cite{6,7,8,9,10} In this paper, we investigate the growth and shape control of InAsSb/InP nanostructures on InP (001) substrates. The growth parameters of InAsSb nanostructures such as growth temperature and growth rate are demonstrated to be important factors to achieve the controlled growth of InAsSb/InP nanostructures.

InAs(Sb)/InP nanostructures were grown on semi-insulating InP (001) substrates using a horizontal flow metal-organic chemical-vapor deposition reactor (AIX200/4) at a pressure of 180 mbar. Trimethylindium, trimethylgallium, trimethylantimony (TMSb), PH3, and AsH3 were used as precursors, and ultrahigh purity H2 as the carrier gas. For reference, an InAs/InP QD sample was grown. The InAs/InP QD sample was grown using the following layer sequence: first, a 200 nm InP buffer was deposited at 650 °C, then a 0.6 nm GaAs interlayer was grown at 650 °C to suppress the As–P exchange, then the growth temperature (Rg) was reduced to 520 °C to grow 4 ML InAs QD layer at a growth rate (Rg) of 0.5 ML/s and a V/III ratio of 15,\cite{11} then the InAs QDs were capped immediately without any growth interruption with a 100 nm InP cap layer while the growth temperature is ramped up to 650 °C. The same steps were followed to grow a top layer of InAs QDs for atomic force microscopy (AFM) measurements. The InAsSb/InP nanostructure samples had the same growth sequence as the InAs/InP QD samples. During the growth of InAsSb nanostructures, the valve of TMSb source had the same on and off sequence as that of AsH3 source, which allowed both As and Sb atoms to bond with In atom terminated surface simultaneously, and form an InAsSb layer (different from the “Sb-protected annealing” growth method in Refs. 7 and 8, where the morphology of InAsSb islands is mainly influenced by that of InAs islands formed). The InAsSb nanostructures had a nominal composition of InAs0.94Sb0.06. A typical V/III ratio of 1.5 was applied for the growth of InAsSb nanostructures.\cite{12} To study the effect of Tg and Rg on the morphology of InAsSb nanostructures, Tg of InAsSb layers varied from 500 to 540 °C, and Rg of InAsSb layers varied from 0.33 to 1.17 ML/s. The morphology of top InAs(Sb) nanowires was characterized by using AFM in tapping mode. The photoluminescence (PL) spectra of the samples were measured at 77 K under excitation by a 635 nm laser line. The luminescence signal was collected by a liquid nitrogen cooled extended InGaAs photodetector.

Figures 1(a) and 1(b) show the typical AFM images of the InAs/InP and InAsSb/InP nanostructures grown at a Rg of 0.5 ML/s at 520 °C. It is observed that elongated QDs are formed in the InAs/InP sample. The InAs QDs are, on average, 8 nm in height, 30 nm in width, and 55 nm in length. The areal density of InAs QDs is around 1.34×1010 cm−2. In contrast, flat QDashes with higher density are formed in the InAsSb/InP sample. The InAsSb QDashes are elongated along the [110] direction with a mean height of 1.7 nm, width (along the [110] direction) of 25 nm, and length (along the [110] direction) of 105 nm, respectively. This morphology change after incorporating Sb atoms into InAs QDs has also been noted in previous works.\cite{9,10} The formation of flat InAs SBQDashes can be mainly attributed to the surfactant effect of Sb atoms in the structures.\cite{9,10,13,14,15} According to previous reports,\cite{9,10} some Sb atoms in InAsSb structures deposited will segregate to the surface, and the resultant Sb-stabilized surface prefers to lower its surface energy by
forming flat islands with large areas.\textsuperscript{13,14} Theoretically, to reach minimum energy in the system, an anisotropic shape (wire or dash) is more stable for quantum structures with large areas when the InAsSb island size exceeds its equilibrium size $a_0$. Also, the most stable anisotropic shape requires that the lateral size of the islands equals to the equilibrium size $a_0$.\textsuperscript{13} This leads to the formation of dash or wire structures for InAsSb/InP system. The alignment of InAsSb QDashes along the $[\bar{1}10]$ direction can be ascribed to the large migration length of In adatoms along the $[\bar{1}10]$ direction induced by surface anisotropy. The decreased width in InAsSb QDashes can be mainly attributed to the increased strain in InAsSb/InP system compared with InAs/InP system. In addition, the reduced migration length of In adatoms induced by Sb atoms in wetting layer also contributes to the decreased width in InAsSb QDashes. The 77 PL spectra of the two samples are shown in Fig. 1(c). The PL peak of InAs QDs and InAsSb QDashes are centered at 0.762 and 0.793 eV, respectively. Obviously, the introduction of Sb atoms into InAs QDs induces a 31 meV blueshift for the PL peak, which can be attributed to the small height of InAsSb QDashes.

In addition, the reduced migration length of In adatoms in the InAsSb/InP system compared with InAs/InP system. The 77 PL spectra of the two samples grown under 500 and 540 °C. Combined with Fig. 1(c), it is observed that the PL peaks are centered at 0.803, 0.793, and 0.775 eV with a full width at half maximum (FWHM) of 42, 48, and 52 meV for InAsSb nanostructures obtained at 500, 520, and 540 °C, respectively. With increasing $T_g$, the PL peak of InAsSb nanostructures shifts to longer wavelength side, which can be mainly explained by the size (height) change in InAsSb islands with increasing $T_g$. The FWHM of PL peaks also increases somewhat with increasing growth temperature, which can be attributed to the increased size fluctuation of InAsSb nanostructures with increasing $T_g$, as discussed.

Figure 2 shows the typical AFM images of the InAsSb nanostructures grown under 520 °C at a $R_g$ of 0.33, 0.83, and 1.17 ML/s. All the InAsSb islands obtained have small average heights (1.5, 1.3, and 1.1 nm for the InAsSb islands grown at 0.33, 0.83, and 1.17 ML/s, respectively). Combined
with Fig. 1(b), it is noted that the aspect ratio of InAsSb islands changes a lot with varying InAsSb $R_g$. When the InAsSb $R_g$ increases from 0.33 to 1.17 ML/s, the length of InAsSb islands decreases from 120 to 55 nm, while their widths are all kept at around 25 nm. This decreased aspect ratio with increasing $R_g$ can also be ascribed to the kinetic characteristic of SK growth.\textsuperscript{17,18} At low $R_g$, In adatoms have large migration length and will have much more chances to be incorporated into existing QDs that are more energetically favorable. This leads to the formation of long QDash or QWR structures with relatively large size, large size fluctuation, and low density. However, at high $R_g$, In adatoms will have much shorter migration length. Thus, the In adatoms deposited will have more chances to form new islands, resulting in the formation of InAsSb islands with relatively small aspect ratio, small size, small size fluctuation, and high density. It should be noted that when the growth rate is too large, such as 1.17 ML/s, not all In adatoms deposited have enough time to migrate away to form new islands though their migration length is even shorter. Therefore, some In adatoms deposited will agglomerate together and form very big InAsSb islands, as shown in the inset of Fig. 3(c). Figure 4 shows the 77 K PL spectra of the InAsSb nanostructures grown under different $R_g$. Combined with Fig. 1(c), it is observed that the PL peaks are centered at 0.8, 0.793, 0.803, and 0.811 eV with a FWHM of 47, 48, 45, and 42 meV for InAsSb nanostructures grown at 0.33, 0.5, 0.83, and 1.17 ML/s, respectively. Obviously, with increasing $R_g$ from 0.5 to 1.17 ML/s there is a blueshift for the PL peak, which confirms the island size change with increasing $R_g$ as observed in Fig. 3. However, there is no blueshift for the PL peak when the $R_g$ increases from 0.33 to 0.5 ML/s, the reason of which is not very clear yet.

In conclusion, the formation and shape control of InAsSb/InP nanostructures has been investigated in detail. The incorporation of Sb atoms into InAs QDs leads to a dramatic change in the nanostructure morphology due to the surfactant effect of Sb atoms and large strain in the system. Due to the kinetic characteristics of SK growth, both elongated InAsSb QDs (short QDashes) and QWRs (long QDashes) can be obtained by choosing proper growth parameters. Low growth temperature and high growth rate will be helpful for achieving elongated QDs, while high growth temperature and low growth rate are good for growing QWRs and QDashes.

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