Supporting Information

Controlling the Morphology and Wavelength of the Self-assembly Coaxial GaAs/Ga(As)Sb/GaAs Single Quantum-well Nanowires

Yubin Kang,[a,b,1] Fengyuan Lin,[b,1] Jilong Tang,[b,*] Qian Dai,[c] Xiaobing Hou,[b] Bingheng Meng,[b] Dongyue Wang,[b] Le Wang[a,**] and Zhipeng Wei[b,***]

[a]*College of Optics and Electronic Science and Technology, China Jiliang University, Hangzhou, 310018, PR China*

[b]*State Key Laboratory of High Power Semiconductor Lasers, Changchun University of Science and Technology, Changchun, 130022, PR China*

[c]*Southwest Institute of Technical Physics, Chengdu, 610041, PR China.*

*Corresponding authors E-mail: *jl_tangcust@163.com (Jilong Tang), calla@cjlu.edu.cn (Le Wang), zpweicust@126.com (Zhipeng Wei).*

S1. Growth of the GaAs/Ga(As)Sb/GaAs SQW NWs

Six GaAs/GaAsSb/GaAs single quantum well (SQW) nanowires (NWs) were grown on Si (111) substrates by MBE system, marked as Sample A, B, C, D, E, and F, respectively. For Samples A-C, the GaAs core was grown on Si (111) substrate, with the growth temperature, Ga flux and V/III ratio of 600 °C, 6.7×10^{-8} Torr and 19.4, respectively. Then, the Ga flux was closed and the Ga droplets were consumed for 10 min under the protection of As flux. After that, for GaAsSb well layer, the Ga and As fluxes remained the same conditions with V/III ratio of 27.3, the growth time of 10 min, and the corresponding growth temperatures of 600, 550 and 530°C, respectively. Finally, the GaAs barrier layer and GaAs core had the same growth conditions (V/III ratio and temperature), and the growth time of 10 min.

For Samples D and E, the GaAs core, GaAsSb well and GaAs barrier layers have the same growth temperature and Ga flux with Sample B. The difference is that the As and Sb fluxes were changed in the growth of the Sample D and E. In Samples D and E, for the growth of the GaAs core and shell layer, the As fluxes are 2.3×10^{-6} Torr and 3.3×10^{-6} Torr, respectively. For Sample D,[1] the fluxes of As and Sb during the growth of the GaAsSb well layer are 2.3×10^{-6} and 1.9×10^{-7} Torr, respectively. For Sample E, the fluxes of As and Sb component in the GaAsSb well layer are 3.3×10^{-6} Torr and 4.3×10^{-7} Torr, respectively.

In contrast to other Samples, the As and Sb fluxes of Sample F are changed and no interrupt growth is used in the growth process. For Sample F, the GaAs core and barrier layer have the same growth temperature of 600 °C, and the growth temperature of the GaSb well layer is 550 °C. In addition, the flux of As remained the same conditions in GaAs core and barrier layer with the corresponding flux about 1.3×10^{-6} Torr. Unlike other Samples, Sample F did not consume Ga droplets after GaAs core growth. The flux of Ga remained unchanged and Sb flux is about 4.3×10^{-7} Torr in GaSb well layer.

After the NWs growth, the Ga flux is switched off and the flux of the As maintained the same as that of the GaAs cores until the substrate is cooled to below 300 °C. Finally, the Samples are cooled to room temperature naturally.

S2. SEM images of the GaAs NWs (V/III=19.4)

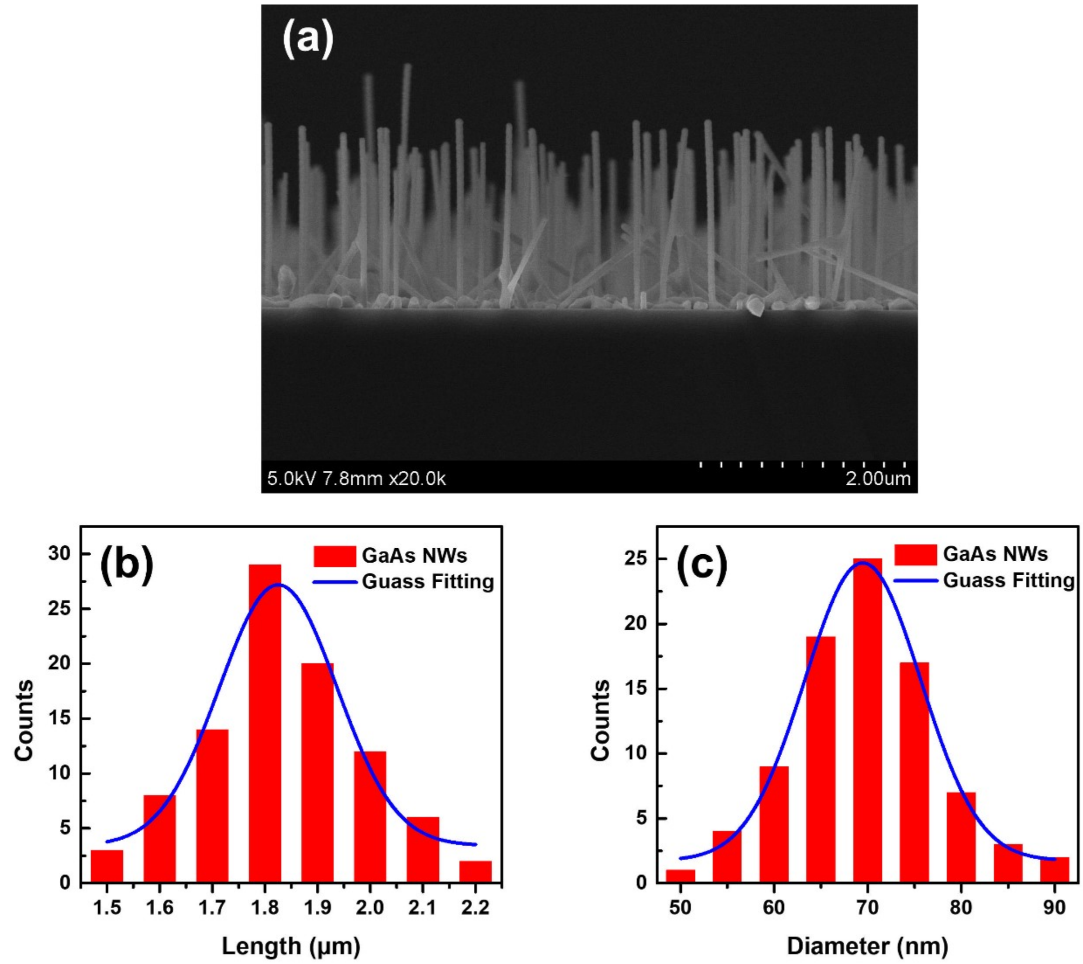

Fig. S1. (a) The side-view SEM image of the GaAs NWs grown on the Si(111) substrate. (b) The length distribution of the GaAs NWs. (c) The diameter distribution of the GaAs NWs. The length and diameter of the GaAs NWs are 1.83 ± 0.3 μm and 65 ± 20 nm, respectively.

S3. Density analysis of GaAs/Ga(As)Sb/GaAs SQW NWs

The top-view SEM images of the GaAs/Ga(As)Sb/GaAs SQW NWs grown on the Si(111) substrate, as shown in Fig. S2. Fig. S2 shows that the density of the six GaAs/Ga(As)Sb/GaAs SQW NWs are ~1.18×10^{8} cm^{-2}, ~1.32×10^{8} cm^{-2}, ~1.12×10^{8} cm^{-2}, ~3.68×10^{7} cm^{-2}, ~3.36×10^{7} cm^{-2}, and ~1.24×10^{8} cm^{-2}.

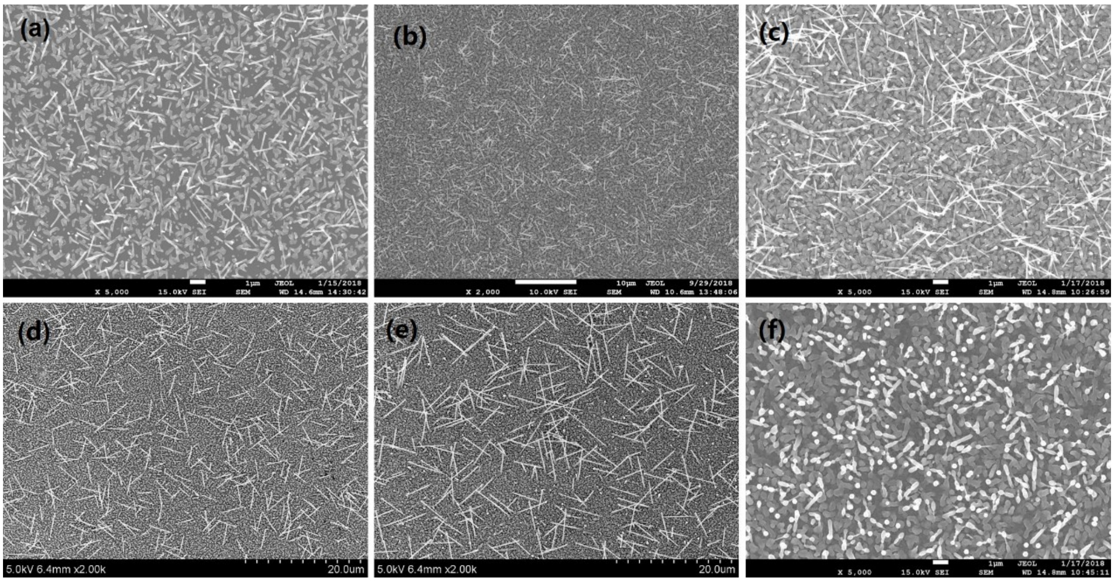

Fig. S2. Low-magnification top-view SEM images of the GaAs/Ga(As)Sb/GaAs SQW NWs.

S4. Length distribution of the GaAs/GaAsSb/GaAs SQW NWs

Statistics of diameter distribution are shown in Fig. S3, the length of the five GaAs/GaAsSb/GaAs SQW NWs are estimated, and the corresponding lengths are 1.9 ± 0.6 μm, 1.95 ± 0.6 μm, 2.0 ± 0.7 μm, 3.7 ± 1.5 μm, and 4.5 ± 1.5 μm, respectively. For Sample F, due to the GaAs/GaSb/GaAs SQW NWs are more inclined during the growth process, the length of the NWs cannot be observed are shown in Fig. 4(a) and S2(f). Moreover, the angle is unfixed between the top view and the plane of the NWs, thus the length of the SQW NWs cannot be accurately obtained.

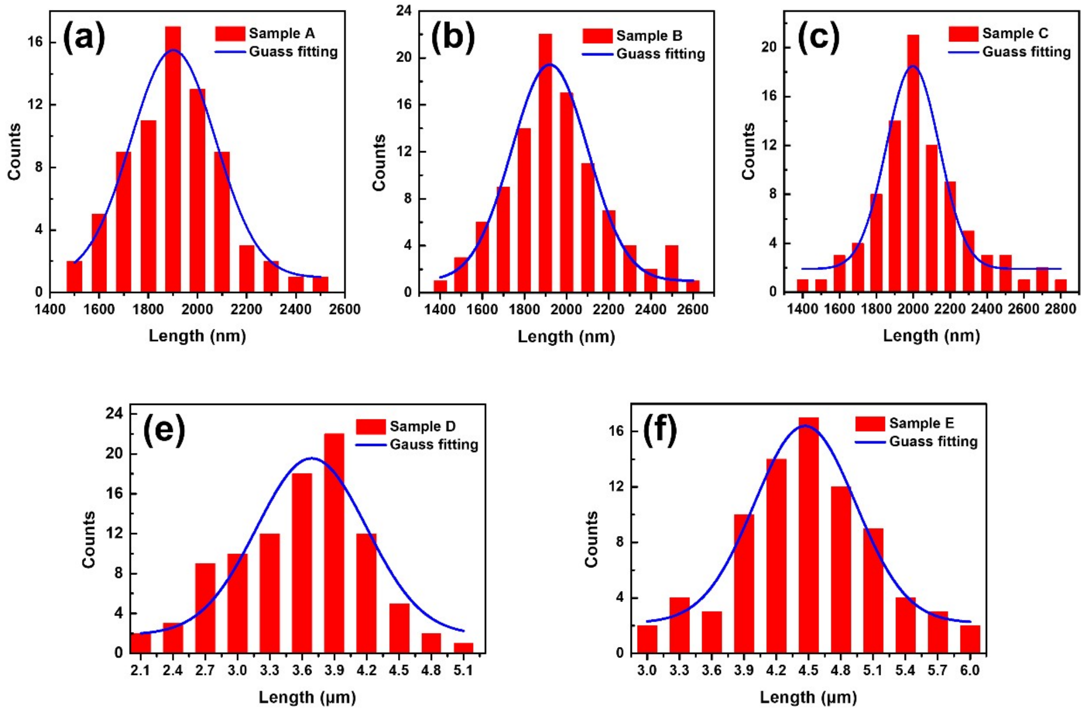

Fig. S3. Statistical of length distribution of the GaAs/GaAsSb/GaAs single QW NWs.

S5. Diameter diatribution of the GaAs/Ga(As)Sb/GaAs SQW NWs

Statistical diagram of diameter distribution is shown in Fig. S4, the diameter of the six GaAs/Ga(As)Sb/GaAs SQW NWs are estimated, and the correaponding diameters are 95 ± 30 nm, 90 ± 20 nm, 110 ± 30 nm, 135 ± 45 nm, 190 ± 70 nm, 125 ± 35 and 200 ± 40 nm, respectively. Meanwhile, two kinds of coarse and fine morphologies in the intermediate position of the Sample F, there are two phenomena of Guass distribution in the obtained diameter, and the corresponding distribution is shown in Fig. S4(f).

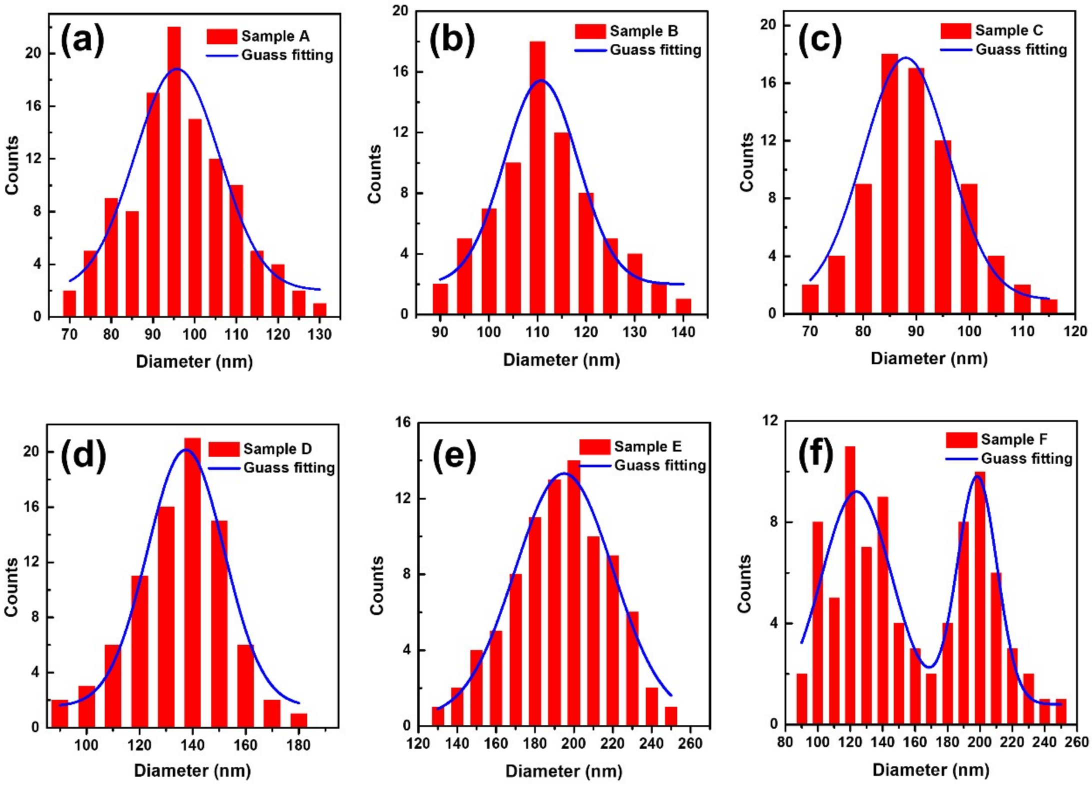

Fig. S4. Statistical of diameter distribution of the GaAs/Ga(As)Sb/GaAs SQW NWs.

S6. Fitting the Raman spectra of the GaSb substrate

The Raman spectra of the GaSb substrate can be well fitted with three Lorentzian peaks which can be assigned as GaSb TO, GaSb SO and GaAs LO, respectively. The corresponding Raman peaks are ~226.16 cm^{-1}, ~232.85 cm^{-1} and ~235.02 cm^{-1}, respectively.

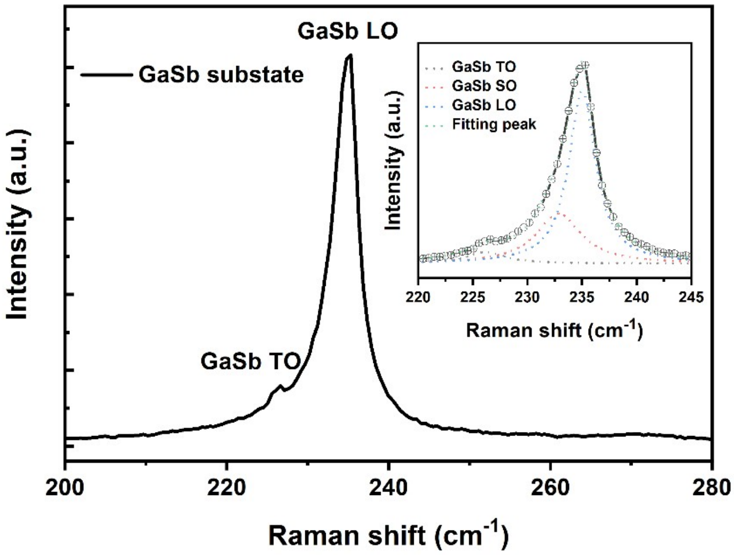

Fig. S5. Raman spectra of the GaSb substrate are fitted by multi-Lorentzian.

S7. Fitting the Raman spectra of the GaAs/GaAsSb/GaAs SQW and GaAs NWs

In order to point out the physical origin of the Raman modes, the Raman spectra were fitted by Multi-Lorentzian. Fix Lorentzian peaks of the GaAs/GaAsSb/GaAs SQW NWs and three Lorentzian peaks of the GaAs NWs are shown in Fig. S6(a)-(f). The Raman spectra of the GaAs/GaAsSb/GaAs SQW NWs can be well fitted with fix Lorentzian peaks which can be assigned as GaSb-like TO, GaAs-like TO and LO, GaAs TO and LO with SO, respectively. The Raman spectra of GaSb-like TO and GaAs-like TO confirmed the presence of the ternary alloy GaAsSb in GaAs/GaAsSb/GaAs SQW NWs.[2,3] In addition, the Raman spectra of the GaAs NWs can be well fitted with three Lorentzian peaks which can be marked as GaAs TO, GaAs SO and GaAs TO, respectively.

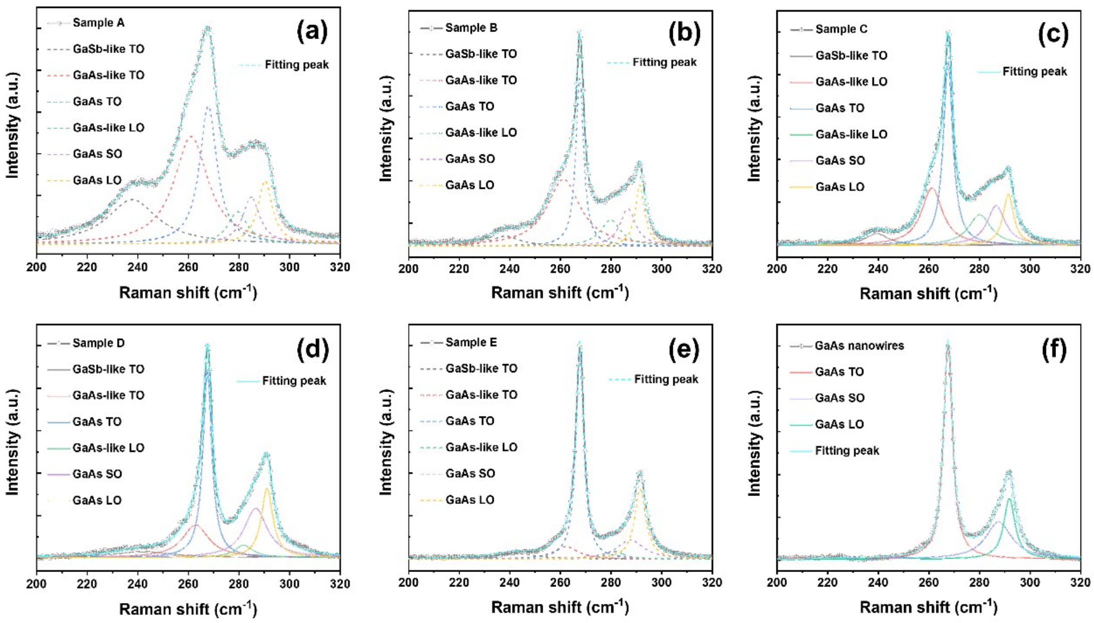

Fig. S6. Raman spectra of the GaAs/GaAsSb/GaAs SQW NWs with different growth conditions and GaAs NWs are fitted by multi-Lorentzian, respectively.

S8. Fitting the Raman spectra of the GaAs/GaAsSb/GaAs SQW and GaAs NWs

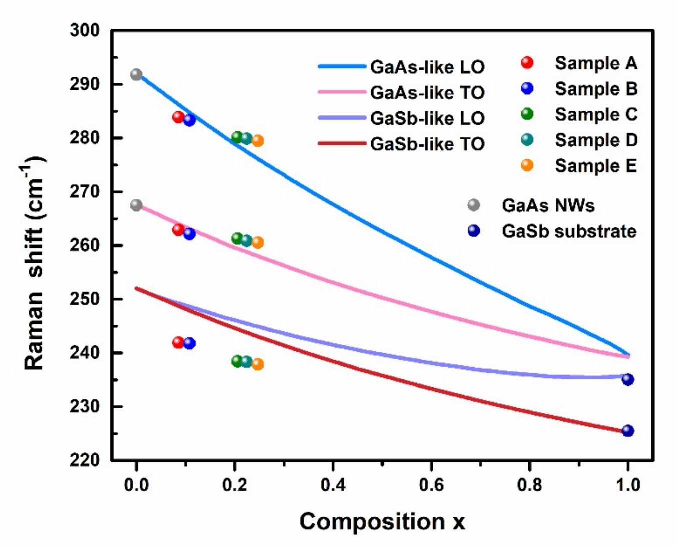

Fig. S7. Phonon mode frequencies of the GaAs/Ga(As)Sb/GaAs SQW NWs as a function of Sb composition *x* (XRD determined) are shown (solid sphere). Experimental Raman frequencies are obtained from fitting the spectra to Lorentzian functions. Calculated results correspond to the expected frequency are shown by solid lines.[2,3]

S9. TEM images of the GaAs/GaSb/GaAs SQW NWs

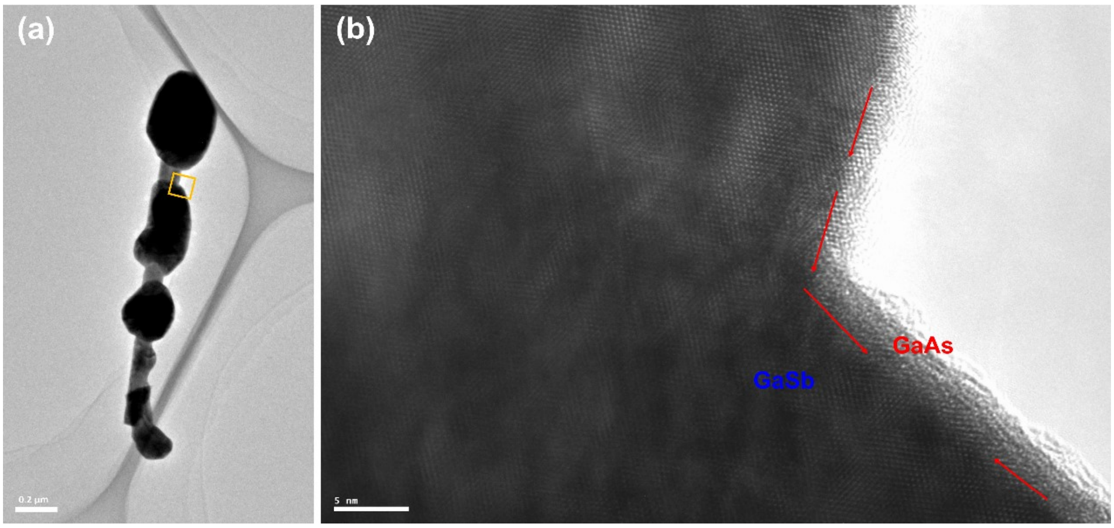

Fig. S8. (a) TEM image of a GaAs/GaSb/GaAs SQW NW. (b) HRTEM corresponding to yellow rectangle regions. The interface between GaSb and GaAs can be clearly observed, which is marked by red arrows. The thickness of GaAs shell layer is very thin of ~ 1-2 nm.

S10. SEM images of the GaAs NWs (V/III=49.5)

The morphology of the GaAs NWs are characterized by SEM, as shown in Fig. S9. Fig. S9 show that the morphology of the GaAs NWs is thicker at the roots, while the other positions are thinner. This is due to the migration length of atoms is limited, the length of the whole NW cannot be well migrated.

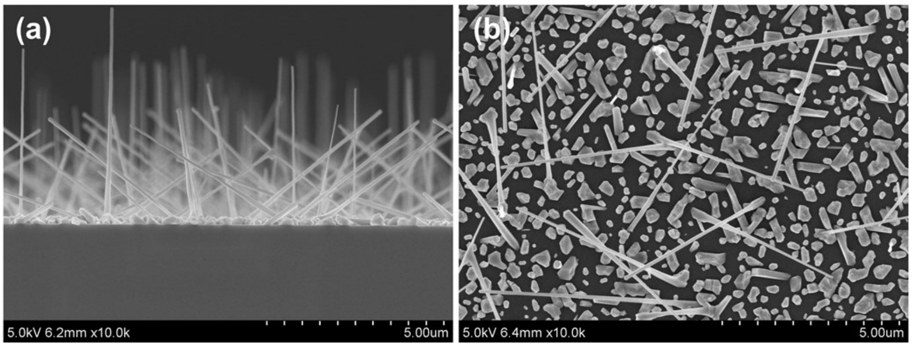

Fig. S9. SEM images of the GaAs NWs. Typical SEM images of the GaAs NWs grown V/III=49.5, side-view (a) and planar view (b).

S11. SEM images of the GaAs NWs (V/III=49.5)

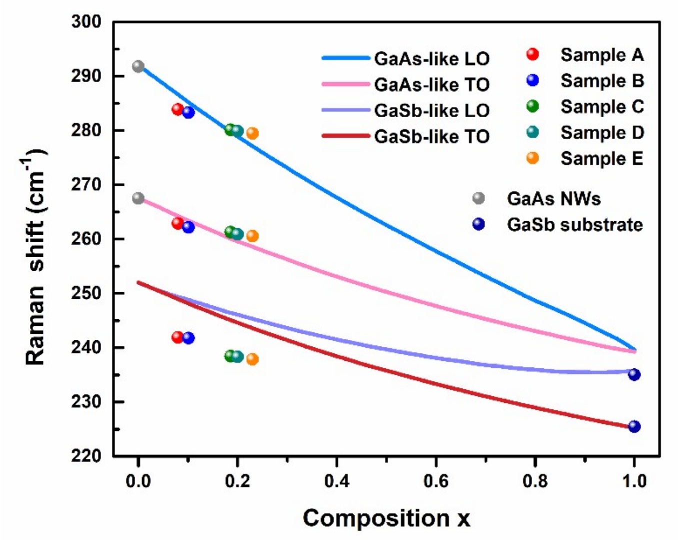

Fig. S10. Phonon mode frequencies of the GaAs/Ga(As)Sb/GaAs SQW NWs as a function of Sb composition *x* (PL spectra determined) are shown (solid sphere). Experimental Raman frequencies are obtained from fitting the spectra to Lorentzian functions. Calculated results correspond to the expected frequency are shown by solid lines.[2,3]

Supporting references

[1] H. Li, J. Tang, Y. Kang, H. Zhao, D. Fang, X. Fang, R. Chen, Z. Wei, Optical properties of quasi-type-II structure in GaAs/GaAsSb/GaAs coaxial single quantum-well nanowires, Appl. Phys. Lett. 113(23) (2018) 233104.

[2] E. Alarcón-Lladó, S. Conesa-Boj, X. Wallart, P. Caroff, A. Fontcuberta i Morral, Raman spectroscopy of self-catalyzed $GaAs_{1-x}Sb_x$ nanowires grown on silicon, Nanotechnology. 24 (40) (2013) 405707.

[3] L. Li, D. Pan, Y. Xue, X. Wang, M. Lin, D. Su, Q. Zhang, X. Yu, H. So, D. Wei, B. Sun, P. Tan, A. Pan, J. Zhao, Near full-composition-range high-quality $GaAs_{1−x}Sb_x$ nanowires grown by molecular-beam epitaxy, Nano Lett. 17(2) (2017) 622-630.