

Two-step Fabrication of Self-catalyzed Ga-based Semiconductor Nanowires on Si by Molecular-beam Epitaxy

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1. Optimization of the surface oxide

We varied the dipping duration of Si substrate in a range from 10 s to 25 s which guarantees a highly uniform morphology of GaAs NWs on Si (Figs. S1 (b-d) and (f-h)), in terms of length, diameter and surface density of NWs. However, for the duration below than this range (e.g. $T_2 = 2$ s as in Figs. S1 (a) and (e)), the NWs only appears occasionally, and for too large one, NWs become tilted and few, even disappear (not shown), both of which indicate a failure of forming Ga nano-droplets. In the text, the re-oxidation method is adopted for the generation of uniformly distributed Ga nano-droplets.

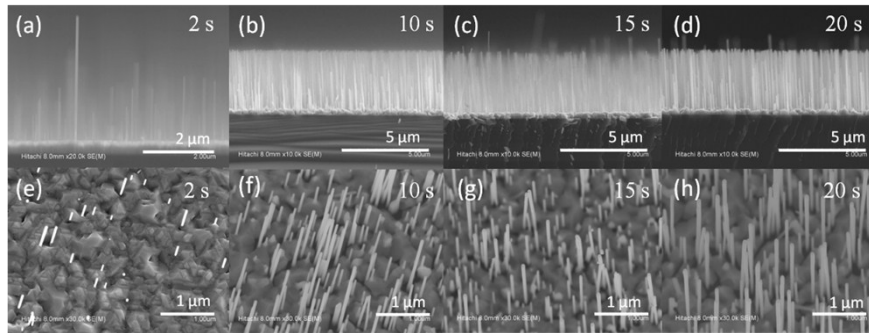


Figure S1. SEM images of GaAs NWs grown on the Si substrates, treated with different T_2 in the re-oxidation method, which is shown at top right corner of each picture. (a-d) Side view images and (e-h) 15° tilted top view of GaAs NWs.

2. Another two-temperature procedure study

We implemented another two samples with the second growth temperature (T_s) of 660 and 680 °C respectively, while other parameters were kept the same as those for the two-temperature-A series. For the sample grown at 660 °C (Fig. S2 (a)), the length is comparable to that of 500 °C (Fig. 3 (c) in the text). However, it shows high quality zinc-blende structure (Fig. S2 (c)). For that grown at 680 °C, the axial growth does not happen (Fig. S2 (b)) and the NWs have been deformed into polycrystalline (Fig. S2 (d) and the inset).

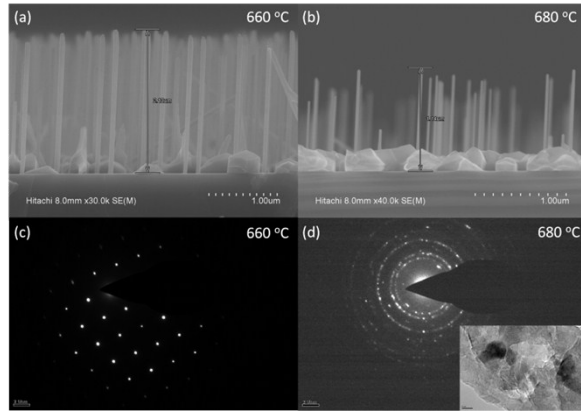


Figure S2. (a-b) SEM images of two GaAs NW samples with the second T_s of 660 and 680 °C. (c-d) SAD images for the two NW samples. The inset of (d) is a HRTEM image of NWs grown at 680 °C.

3. Photoluminescence spectra for Two-temperature-A series

Photoluminescence spectra at 77 K for four samples of GaAs NWs from two-temperature-A series grown at 620, 560, 520 and 500 °C are shown in Fig. S3. A clear peak at 1.51 eV is observed for all the samples except that grown at 500 °C, which is characteristic of free exciton recombination in GaAs. It is worth mentioning that all the GaAs NWs are without AlGaAs shells, thus inevitably resulting in the surface defect states. The PL signals around 1.45eV are attributed to the surface states and the comprehensive study of them is beyond the scope of this work. The PL intensity of GaAs free exciton decreases as the second T_s lowers, which is due to the increased twin-plane density. For NWs grown at 500 °C, the characteristic peak even vanishes.

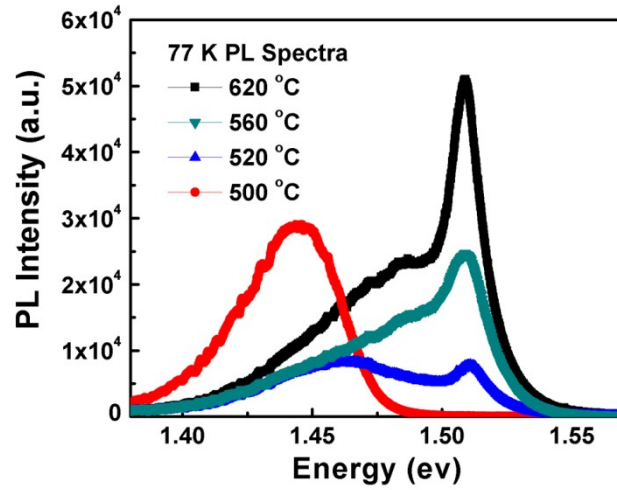


Figure S3. Photoluminescence spectra for GaAs NWs at 77 K under 633 nm excitation for GaAs NWs grown at different T_s .

4. EDX measurements

We also implemented EDX measurement along the axial and cross-axial directions at interface and the results are shown in Figs. S4. We observe that the interface is not as sharp as we expected. The blue color in Fig. S4 (a) shows the range with the length of 100 nm where Sb and As overlapped. Scanning along the cross-axial direction at interface, we see that this section (blue range) shows a typical core-shell structure (Fig. S4 (b)) with GaSb as shell NW. However, for the section above (below) this range, it is pure GaSb (GaAs) section (Fig. S4 (a)). So we obtained a structure as in Fig. S4 (d).

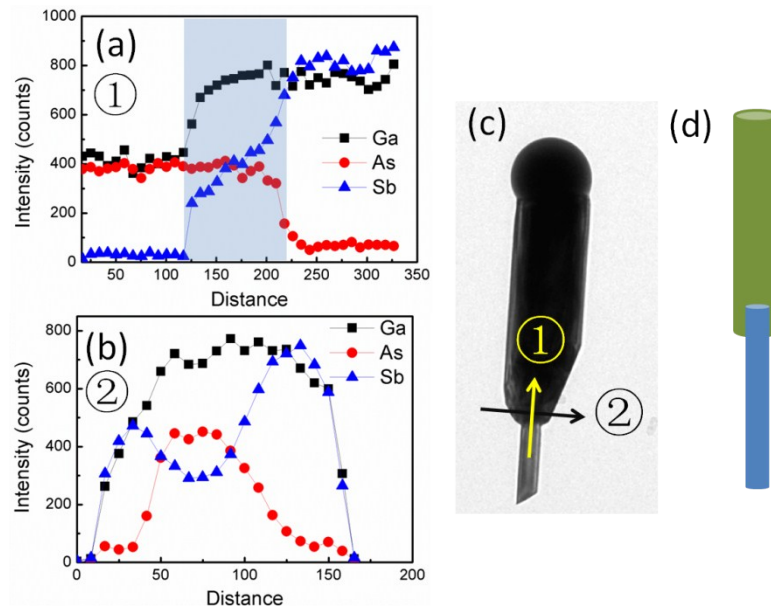


Figure S4. EDX linescans along the (a) axial and (b) cross-axial direction at interface. (c) TEM image of GaSb section showing the directions of EDX linescans. (d) An illustration for GaAs/GaSb NWs we obtained.