Supporting Information

Unexpected Formation of Hierarchical Structure in Ternary InGaAs Nanowires via "One-Pot" Growth

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Fig. S1a shows the side-view SEM image of the 60 min grown nanowires and **Fig. S1b,c** is the TEM images of the cross sections taken from the nanowires middle and bottom regions, respectively. Since the cross sections were cut from nanowires bottom to top with a step of ~ 50 nm, the small-size cross sections in **Fig. S1** indicate that they are from the nanowires top (with lower height), while the larger-size cross sections indicate that they are from the nanowire middle regions (as shown in **Fig. S1a**). Since only one group-V element in the InGaAs system, the composition of As is constant, which is confirmed by the EDS As maps of the cross sections at varied nanowires regions (see **Fig. S1d**)

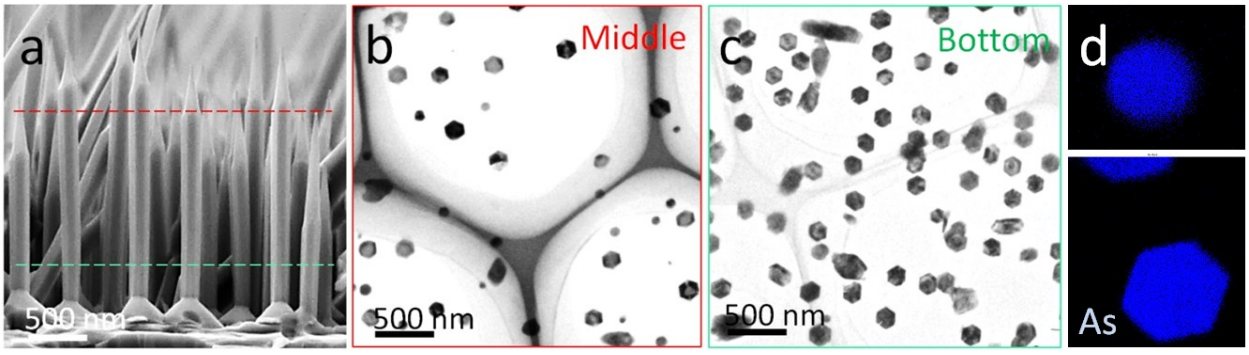

Fig. S1 (a) Side-view SEM image of the 60 min grown InGaAs nanowires. Typical TEM images of the cross sections taken from the nanowires (b) middle and (c) bottom regions marked in (a). (d) EDS As maps of the typical cross section from nanowire top and body.

Fig. S2a is a typical TEM image of the pencil-shape nanowire top and **Fig. S2b-d** is typical TEM images of the cross sections taken from the varied regions of the pencil top with lateral dimensions of ~27 nm, 70 nm and 100 nm respectively (marked in **Fig. S2a**). **Fig. S2e-g** is corresponding EDS line scan profiles of the cross sections, in which nanowires keeps the core-shell structure of In-poor core and In-enriched shell until there is no shell growth. This result is consistent with the core-shell structure observed at nanowires middle regions (see **Fig. 2f**).

Fig.S2h-i is corresponding [0001] zone-axis SAED pattern, indicating that nanowires keep the $\{11\bar{2}0\}$ side-facets during the growth. **Fig. S2j** shows the corresponding composition measurements of the cores composition taken from the cross sections, indicating that the In concentration ranging from 32 ± 2 at.%, 30 ± 2 at.% to 28 ± 2 at.% with the decreased diameter of the cross section. On the other hand, **Fig. S2k** is a typical TEM image of the 15 min grown nanowire and **Fig. S2m** is TEM images of the cross sections taken from the nanowires middle and top regions, respectively. The corresponding EDS elements maps **(Fig. S2n**) confirm that the grown nanowires form core-shell structure with the In-enriched core and In-poor shell along the nanowire (expect the region close to the catalyst). **Fig. S2l** is corresponding [0001] zone-axis SAED patterns, indicating the $\{11\bar{2}0\}$ side-facets of the nanowires.

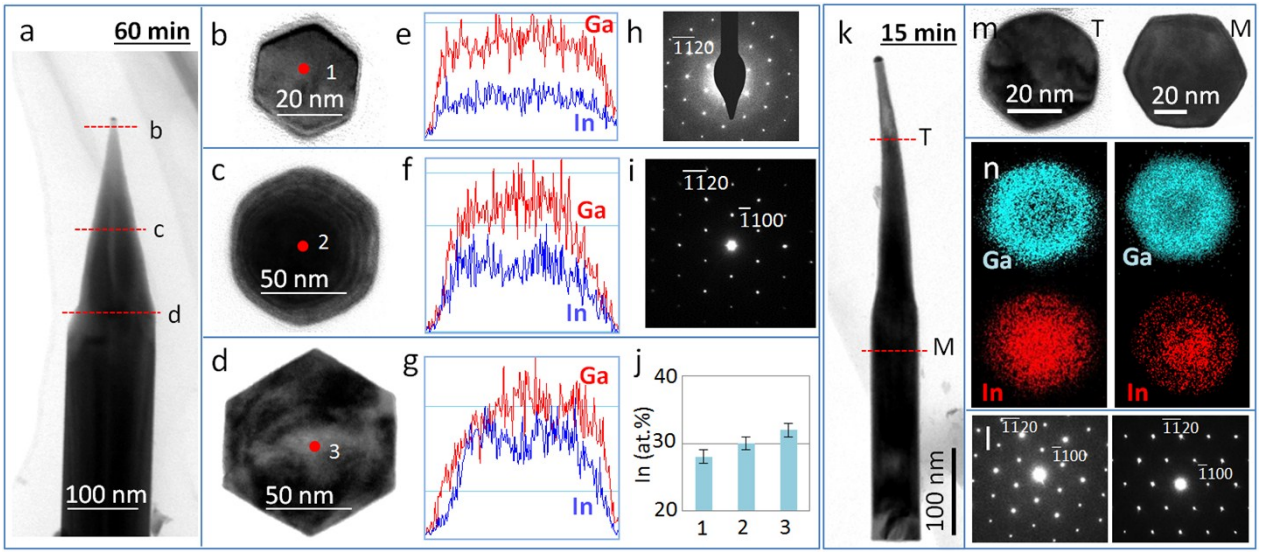

Fig. S2 (a) Typical TEM image of the 60 min grown nanowire top. (b-d) Typical TEM images of the cross sections taken from the pencil-shape top as marked in (a). Corresponding (e-g) EDS line scan profiles and (h-i) SAED patterns. (j) Compositional measurements of the cores compositions marked in (b-d). (k) Typical TEM image of the 15 min grown nanowire. (m) Typical TEM images of the cross sections taken from the middle and top regions marked in (k). Corresponding (n) EDS elements maps and (l) SAED patterns.