非均一なナノワイヤ成長速度の時間依存性 Nonuniform growth rate of nanowires with growth time in the self-assisted vapor-liquid-solid mode

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Semiconductor nanowires (NWs) have become the next-generation building blocks in various fields [1]. Despite the progress that has been made on NW applications, it has been insufficient to reveal the NW growth mechanism, which is fundamentally different from that of the conventional film growth [2]. Conventional film growth occurs on a flat two-dimensional surface, while NW growth occurs in a three-dimensional space. The decomposition and migration of source gases occur on both the substrate surface and the NW side face. This may induce different growth rate dependence on growth time from the film growth mode, which normally shows constant growth rate. Here we provide direct evidence indicating that the NW growth rate is not uniform with growth time in the vapor-liquid-solid (VLS) mode.

The NWs were grown in a low-pressure (76 Torr) MOVPE system. Instead of the widely used Auassisted VLS synthesis approach that has been employed for various NWs, we used the self-assisted VLS growth to avoid the incorporation of Au impurity atoms into the NWs during growth [3,4]. We have established a technique for growing InP and InAs NWs using indium particles in the self-assisted VLS mode [5,6]. This enables us to grow InP/InAs hetero NWs in the indium-particle-assisted VLS mode [7].

The thin InAs layer can function as markers and enable us to measure the thickness of an InP segment grown in a different period from the intensity profile of the HAADF-STEM image. There were 60 InAs layers with a 2-s growth time for each segment and 59 InP layers with a 20-s growth time for each segment (Fig. 1). We plot the thickness of the single InP layer as a function of the layer number (Fig. 2). Theoretically, there are two paths in the source supply for the axial growth; one is that the vapor source materials directly intrude into the droplet, and the other is the adsorbed source materials on the NW side face, which can migrate on the NW side and finally reach the droplet. Assuming the decomposition of sources is fast at the droplet, the former does not change with the NW height because the droplet particle size is constant. However, the latter depends on the diffusion length of the source material on the side and migrate into the droplet. This induces increasing growth rate with growth time. This growth model accounts for the experimental result shown in Fig. 2.

References: [1] M. D. Birowosuto, et al. Nature Mater. 13 (2014) 279-285; [2] Borgstrom, et al. Nature Nanotech. 2, 541-544 (2007) [3] J. Kasahara, et al J. Cryst. Growth 38, 23-28 (1977); [4] C. J. Novotny and P. K. L. Yu Appl. Phys. Lett. 87, 203111 (2005). [5] G. Zhang, et al. Appl. Phys. Exp. 5, 055201 (2012); [6] G. Zhang, et al. AIP Advances 3, 052107 (2013); [7] G. Zhang, et al. submitted.



Fig. 1. HAADF-STEM images of a single NW. There are 59 InP segments between the adjacent InAs segments with a 20-s growth time for each segment. The numbers on the left represent the growth sequence of the InP layers in (b). The vertical white arrow indicates the growth direction.



Fig. 2. Plot of the thickness of each single InP layer as a function of the layer number. The vertical line is the error bar of the thickness, induced by the determination of the interface between the InP and InAs segments.