



Three-dimensional lattice rotation in GaAs nanowire growth on hydrogen-silsesquioxane covered GaAs (001) using molecular beam epitaxy

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ABSTRACT

We report on crystallographic behaviors of inclined GaAs nanowires (NWs) self-crystallized on GaAs (001) substrate. The NWs were grown on hydrogen-silsesquioxane (HSQ) covered substrates using molecular beam epitaxy (MBE). Commonly, the epitaxial growth of GaAs (111)B (B-polar) NWs is prominently observed on GaAs (001); however, we yielded a remarkable number of epitaxially grown GaAs (111)A (A-polar) NWs in addition to the majorly obtained B-polar NWs. Such NW orientations are always accompanied by a typical inclined angle of 35° from (001) plane. NWs with another inclined angle of 74° were additionally observed and attributed to be (111)-oriented, not in direct epitaxial relation with the substrate. Such 74° NWs' existence is related to first-order three-dimensional (3D) lattice rotation taking place at the very beginning of the growth. It turns out that spatially 60° lattice rotation around (111) directions at GaAs seeds is essentially in charge of A- and B-polar 74° NWs. Transmission electron microscope observations reveal a high density of twinning in the B-polar NWs and twin-free characteristic in the A-polar NWs.

1. Introduction

One-dimensional (1D) structures such as nanowires (NWs) have been developed over the last two decades into a large field and they are believed to be building blocks for a wide range of applications. Especially, III-V semiconductor NWs have been intensively studied because they have potential to show high electron mobility and saturate velocity that are important for future high-speed electronic applications [1]. Among them, GaAs NWs have been widely studied so far for nano-electronic, photonic and photovoltaic applications [2–5]. Up to now, a plenty of work has been done on GaAs NW growth using vapor-liquid-solid (VLS) method in molecular beam epitaxy (MBE) system with the help of metal nanoparticles (NPs) as catalysts [6–8]. Au is the most commonly used catalyst in NW growth using VLS method due to high chemical stability; however, there is a possibility of unintentional incorporation of Au into NWs as impurities forming deep levels, which inherently limit the performance of NW-based devices [9–11]. To avoid such unintentional incorporation of metal catalysts, one of the typical

methods is self-catalyzed VLS-MBE using Ga NPs with SiO₂ [12], native oxide [13], or HSQ-based amorphous silicon oxide (SiO_x) [14]. The use of HSQ-based masks shows many advantages compared to the use of usual oxide masks. For instances, uniform nanometer-thick HSQ layers have been achieved by a simple spin-coating method and the ability of pin-hole self-creation at growth temperature [14]. The self-catalyzed VLS-MBE has been studied mainly on (111)-oriented substrates but less on (001)-oriented ones, which are commonly used in various device applications. Moreover, the growth on GaAs (001) tends to induce a complex NW net [15] due to polar-related or abnormal growth that needs to be systematically investigated.

In this paper, we present the self-catalyzed VLS-MBE growth of GaAs NWs on HSQ-coated GaAs (001) substrates. Through a careful analysis, we show that NWs in optimized conditions are epitaxially grown in not only (111)B but also (111)A directions, and the inclined angles are estimated to be about 35°. Additionally, NWs slanting 74° from (001) plane are also observed along the same projections with the 35° NWs. A model of first-order three-dimensional lattice rotation is comprehen-

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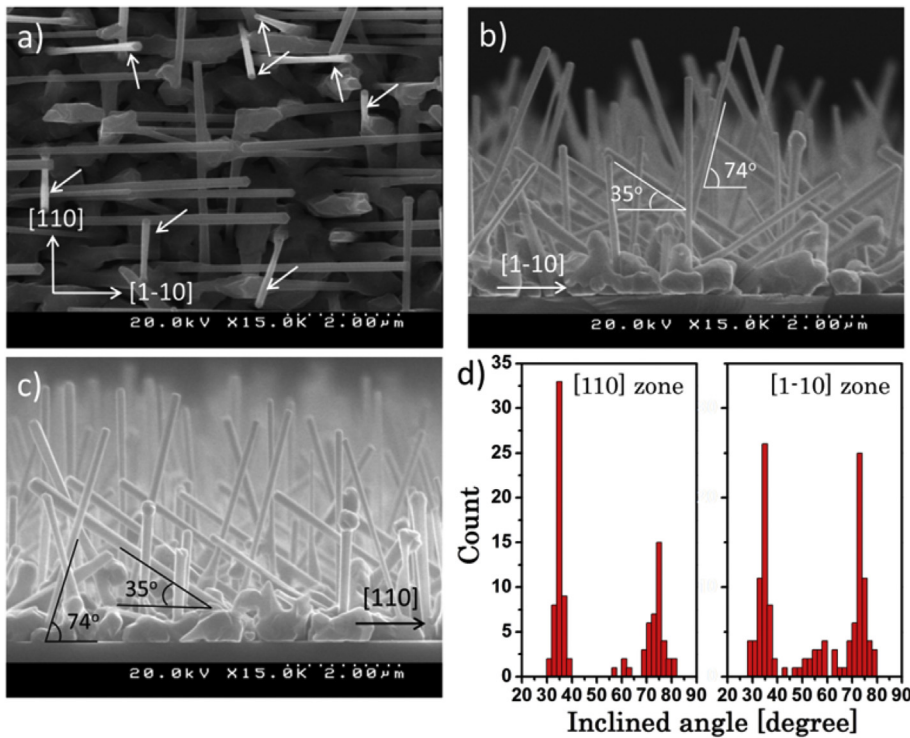


Fig. 1. Scanning electron microscopy (SEM) images taken from (a) top view, (b) side view in [110] zone, (c) side view in [1-10] zone. (d) Distribution of NWs' inclined angles in [110] and [1-10] zones. The angle distribution was calculated from a relatively large number of side-view SEM images at the same magnification and without tilting. The equal numbers of images were taken into account for the two zones [110] and [1-10].

sively discussed to explain the formation of the 74° NWs. Finally, a comparison between $\langle 111 \rangle$ B and $\langle 111 \rangle$ A NWs in terms of morphology and crystallography is conducted.

2. Experimental procedure

After removing native oxides on GaAs surface by an organic-ammonium chemical, the GaAs (001) wafers were spin-coated with an HSQ solution at 5000 rpm in 1 min. The solution consists of Dow Corning Fox 14 and methyl-isobutyl-ketone (MIBK) with the volume ratio of 1:30. Subsequently, the wafers were baked on a hot plate in the air at 150 °C in 2 min and then at 300 °C in 10 min to transform the spin-coated HSQ layer into SiO_x . The obtained SiO_x thickness was typically 20 nm measured with an ellipsometer. Before loading the substrates into MBE system having three chambers, they were successively cleaned with acetone, methanol, and deionized (DI) water for 5 min in each solution. After loading the substrates, they were heated up and kept at 370 °C for 1 h in transfer chamber for degassing and then transferred to the main chamber. After reaching growth temperature, the substrates were kept in 10 min for thermal stabilization. Subsequently, Ga deposition without opening As shutter was carried out in 20 s and followed by Ga NP incubation process in 10 min. The NW growth was then started by simultaneously opening both Ga and As shutters. The optimized growth conditions were substrate temperature of 620 °C, Ga BEP of 0.04 ML/s, As BEP of 1.3×10^{-6} Torr (V/III ratio of 24), and growth time of 4 h. After the growth, Ga NP removal procedure was carried out by closing Ga shutter, cooling the sample down to 490 °C with keeping As exposure for 1 h. At the final step, the As flux was continuously kept open during cooling down process.

The NW morphology, density, and inclined angles were analyzed using a scanning electron microscopy (SEM). NWs were transferred onto a copper mesh, and their crystal structure was investigated by means of transmission electron microscopy (TEM).

3. Results and discussion

Fig. 1 shows SEM images of the sample grown under optimized

conditions. In Fig. 1a, the top-view image shows two typical types of NWs located on the sample including lower contrast NWs presented by longer lines and higher contrast NWs presented by shorter lines. The NWs of both types point along four orthogonal directions ([1-10], [110], [-110], and [-1-10]). Around NW bottoms, polycrystalline GaAs islands were alternately deposited on SiO_x surface during the growth. From the side views as shown in Figs. 1b and c, the NWs incline from (001) plane with two typical angles of 35° and 74°. As a result, the lower and higher contrast NWs observed in Fig. 1a are clarified to be 35° and 74° NWs, respectively. Also, the 74° NWs are marked by the arrows in the top-view image. The NW density of about $0.9 \mu\text{m}^{-2}$ was achieved that is comparable to the highest value induced by the same growth method on HSQ-covered GaAs(111)B substrates [14]. The lengths and diameters of all NWs are relatively uniform and estimated to be $\sim 4 \mu\text{m}$ and $\sim 160 \text{ nm}$, respectively. In addition, they possess the same hexagonal cross-section with side facets belonging to {110}. The two typical inclined angles are quantitatively confirmed by the angle distribution plotted based on NWs observed in [110] and [1-10] zones as shown in Fig. 1d. The angle deviation around 35° seems to be smaller than that around 74° in both two zones.

The 35° NWs in [110] and [1-10] zones could be crystallographically indexed to be in $\langle 111 \rangle$ B and $\langle 111 \rangle$ A orientations, respectively, based on epitaxial relation with the substrate. In terms of NWs with respect to the inclined angle of 74°, their orientations are indexed nearby $\langle 115 \rangle$ A/B if that is the fact, they are impossible to be crystallized into the hexagonal cross-section, which is unique for $\langle 111 \rangle$ orientations. As commonly observed in B-polar GaAs NWs, twin planes separate the NW into many segments, in which two adjacent ones are mutually associated with a typical 60° rotation around growth axis i.e. $\langle 111 \rangle$ B [15–17]. And such rotation shall lead to no change in the growth direction. If we take 3D lattice rotation on the B-polar NWs into account and set rotation axes to be not only $\langle 111 \rangle$ B but also $\langle 111 \rangle$ A, we would observe kinking effect by which the NWs selectively switch to a new direction inclined 74° from (001) plane and observable in both [1-10] and [110] zones. To avoid any confusion in naming $\langle 111 \rangle$ A and $\langle 111 \rangle$ B directions attached to the rotation axes, we define them as directions pointing upwards from (001) plane. Different from the already-mentioned 3D rotation on B-polar NWs,

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