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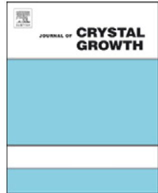

Nucleation and growth of Au-assisted GaAs nanowires on GaAs(1 1 1)B and Si(1 1 1) in comparison

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ABSTRACT

The nucleation and growth of GaAs nanowires fabricated by molecular beam epitaxy (MBE) following the Au-assisted vapor-liquid-solid mechanism were compared on GaAs(1 1 1)B and on Si(1 1 1) substrates. On both substrates, reflection high-energy electron diffraction (RHEED) patterns and scanning electron microscopy (SEM) images of several samples belonging to a growth time series were analyzed. During the nucleation stage, growth on Si(1 1 1) is dominated by horizontally growing traces and coalescing islands, while growth on GaAs(1 1 1)B proceeds instantly in the vertical direction. After this nucleation stage, the Si substrate is covered by a closed, rough GaAs layer, and nanowires of similar shape grow on both substrates with similar axial and radial growth rates. However, the diameter of the nanowires on Si(1 1 1) is different than that on GaAs(1 1 1)B, because the size of the Au droplets, which result from the annealing of a thin Au layer, is different on the two types of substrates.

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1. Introduction

Epitaxially grown GaAs nanowires are being considered for applications in solar cells [1] and in light emitting diodes [2]. Furthermore, GaAs is probably the best studied compound semiconductor material and can be considered as a role model for other (III–V) compound semiconductors. Vertical GaAs nanowires have successfully been fabricated by the Au-assisted vapor-liquid-solid (VLS) mechanism [3,4] on different substrates, most notably by homoepitaxial growth on GaAs(1 1 1)B [5,6] and by heteroepitaxial growth on Si(1 1 1) [7,8].

For a high level of nanowire growth control, it is important to understand the effects imposed by the substrate on nanowire shape and crystal structure. Judging by the knowledge of planar growth, strong differences between homoepitaxial and heteroepitaxial growth can be expected. For example, the crystal quality of planar GaAs layers on Si substrates still has not reached the level achieved by homoepitaxial GaAs, despite decades of intense research [9]. For nanowires, in contrast, the effect of the substrate might be much smaller, since residual strain, which accumulates in heteroepitaxial planar layers, can laterally relax in nanowires at the free sidewalls [10].

Effects of the substrate are strongest during nucleation. At the very beginning of the homoepitaxial nucleation of GaAs nanowires on GaAs(1 1 1)B, pyramids with triangular bases and tilted lateral facets develop, which turn into nanowires in the moment when these facets become vertical [11]. We recently reported about the heteroepitaxial nucleation of Au-assisted GaAs nanowires on Si(1 1 1), where we showed that initially horizontal traces grow, which evolve into vertical nanowires after the Si is covered by coalesced GaAs [12]. Here, we present a detailed comparison of the nucleation and growth of Au-assisted GaAs nanowires on GaAs(1 1 1)B and Si(1 1 1), in order to analyze the effect of the substrate on nanowire formation.

2. Experimental

The GaAs nanowires were grown by molecular beam epitaxy on GaAs(1 1 1)B and Si(1 1 1) substrates. The crystallographic orientation of each substrate was chosen in order to obtain a majority of vertical nanowires.

Homoepitaxial GaAs nanowire nucleation was studied on quartered two-inch, semi-insulating GaAs(1 1 1)B substrates. After oxide desorption at 580 °C, a 200 nm thick buffer layer was grown at 600 °C. Au droplets were prepared on the clean surface by depositing 0.6 nm of Au at 400 °C and annealing at 550 °C for 5 min. For nanowire growth, the substrate temperature was set to 500 °C. The As valve was opened before growth to establish a stable As supply. The Ga shutter opening initiated GaAs growth. The V/III flux ratio, i.e. F_{As}/F_{Ga}, was set to 2.0 and the Ga flux was set to match a planar GaAs(1 1 1)B growth rate of 0.11 nm/s. The Ga and As fluxes had been calibrated using reflection high-energy electron diffraction (RHEED) oscillations on GaAs(0 0 1) substrates. The substrates

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were not rotated during growth, in order to permit the detailed RHEED analysis.

Heteroepitaxial nucleation was studied on quartered two-inch, n-type, Si(1 1 1) $\pm 0.5^\circ$ substrates. The silicon oxide was removed from the substrates by Ga-polishing, i.e., deposition of 3 nm Ga at a substrate temperature of 500 °C followed by desorption of the resulting gallium oxide at 800 °C and formation of Si(1 1 1) terraces [13]. The steps for Au droplet formation and GaAs growth were identical to the homoepitaxial case.

Secondary electron microscopy (SEM) was performed by a Hitachi S4800 field emission microscope. For image analysis, the open source software package ImageJ was employed.

3. Results

RHEED gives in-situ information about the crystal structure present during growth at and near the surface. The patterns were recorded during growth on both substrates with the incident electron beam along the $[1\,\bar{1}\,0]$ azimuthal direction and a selection of images is presented in Fig. 1. Initially, vertical streaks indicate the presence of atomically flat substrate surfaces, while the gradual appearance of spotty patterns shows the increasing dominance of three-dimensional (3-D) structures.

On GaAs(1 1 1)B, initially GaAs in the cubic zinc-blende (ZB) structure can be identified, and by 60 s growth time, GaAs in the hexagonal wurtzite (WZ) structure dominates (Figs. 1b-d). On Si(1 1 1), initially the reflex patterns of ZB and its rotational twin, then additionally WZ spots, and finally only WZ spots are observed (Figs. 1f–h). Thus, in contrast to the nucleation on GaAs, on Si the rotational twin of the ZB structure forms and the transition to the WZ structure is delayed.

For ex situ analysis of the surface morphology by SEM, series of growth experiments with different GaAs growth durations were prepared under otherwise identical conditions on both substrates. SEM images recorded under 45° sample inclination are depicted in Fig. 2. They show that the nucleation of nanowires on both substrates leads to an initially very different surface morphology, but eventually, rather similar nanowires grow.

On GaAs(1 1 1)B, after 5 s (Fig. 2a), 3-D structures are visible that exhibit an aspect ratio of about one. These are actually triangular pyramids, ca. 20 nm in diameter, and have a Au droplet on top. After 60 s (b), there are fully formed nanowires with vertical sidewalls which rest on much larger basal pyramids, whose diameter typically exceeds 200 nm. After 150 s (c), the nanowires as well as the basal pyramids have increased in height. The alignment of a nanowire and its corresponding basal pyramid is not concentric, but offset to one side, which is due to growth without substrate rotation. After 1800 s (d), the basal pyramids have coalesced to a continuous rough layer and the nanowires reach an average length of (1.7 ± 0.3) µm.

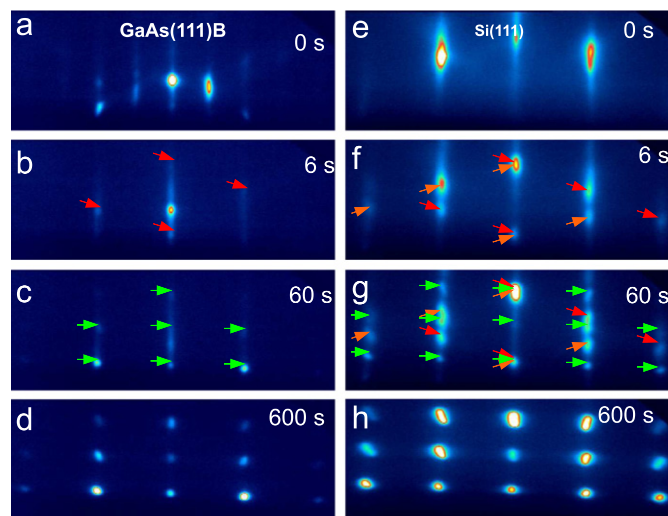

Fig. 1. (Color online) RHEED images of GaAs(1 1 1)B and Si(1 1 1) substrates decorated with Au droplets (0 s), as well as of both substrates after Au-assisted GaAs growth for 6, 60, and 600 s. The reflex positions of ZB GaAs (↘), of its rotational twin (↗), and of WZ GaAs (→) are indicated in selected images.

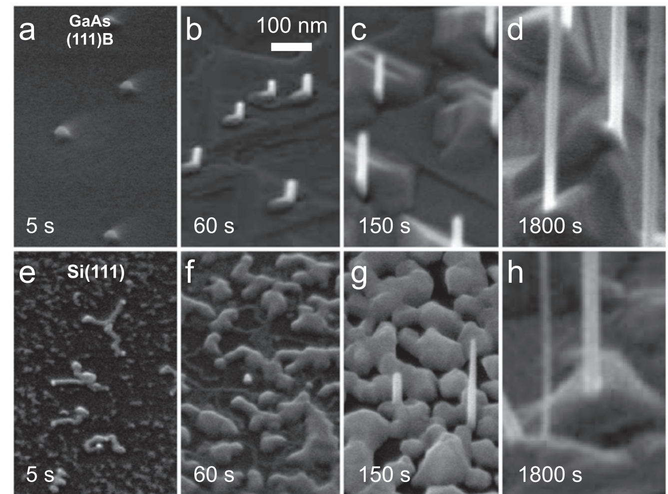

Fig. 2. SEM images of GaAs(1 1 1)B and Si(1 1 1) substrates after Au-assisted GaAs growth for 5, 60, 150, and 1800 s. All images were recorded under 45° sample inclination and are shown with identical magnification as indicated by the scale bar in (b).

On Si(1 1 1), after 5 s (Fig. 2e), horizontal traces with ca. 10 nm diameter and smaller structures are visible. We emphasize that the traces are pronouncedly elongated in the horizontal direction and thus exhibit a completely different shape than the structures found on GaAs(1 1 1)B. After 60 s (f), the horizontal length of the traces has increased, many elongated 3-D islands of irregular shape have appeared, the smaller structures are not visible, and very few vertical nanowires can be observed. After 150 s (g), the islands have coalesced to a connected network and there is a small number of vertical nanowires. After 1800 s (h), a higher number of vertical nanowires is present and the 3-D islands have completely covered the Si substrate.

For a quantitative study, further SEM images were recorded and analyzed using ImageJ. The number density of vertical nanowires was determined using top-view images taken at three different positions on the sample and in two magnifications each. The lengths and diameters of at least 30 nanowires were measured for each sample, except for the 60 and 150 s samples on Si for which at least 8 nanowires were measured. The average results are presented as plots versus growth time in Fig. 3. Error bars represent the standard deviation and linear approximations were obtained by least square fits.

On GaAs(1 1 1)B substrates, the nanowire number densities are essentially constant at $(12 \pm 2)\ \mu m^{-2}$. A slightly decreasing trend for longer growth times is observed but lies within the margin of variation between several images. Nevertheless, this might indicate the dropping out of a small number of nanowires for an unknown reason. For the nanowire lengths, there is no perfect linear fit. During the first 300 s of growth, an average axial growth rate of (0.7 ± 0.1) nm/s was determined as indicated in the plot, but the sample with 1800 s growth time yields (1 ± 0.2) nm/s instead. This may also be related to a small number of nanowires having dropped out after some growth time and then the same amount of

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