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Dimensional evolution of silicon nanowires synthesized by Au–Si island-catalyzed chemical vapor deposition

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Abstract

This study explores the nucleation and morphological evolution of silicon nanowires (Si-NWs) on Si (0 0 1) and (1 1 1) substrates synthesized using nanoscale Au–Si island-catalyzed rapid thermal chemical vapor deposition. The Au–Si islands are formed by Au thin film (1.2–3.0 nm) deposition at room temperature followed by annealing at 700 °C, which are employed as a liquid-droplet catalysis during the growth of the Si-NWs. The Si-NWs are grown by exposing the substrates with Au–Si islands to a mixture of gasses SiH_4 and H_2. The growth temperatures and the pressures are 500–600 °C and 0.1–1.0 Torr, respectively. We found a critical thickness of the Au film for Si-NWs nucleation at a given growth condition. Also, we observed that the dimensional evolution of the NWs significantly depends on the growth pressure and temperature. The resulting NWs are ~30–100 nm in diameter and ~0.4–12.0 μm in length. For Si (0 0 1) substrates ~80% of the NWs are aligned along the ⟨1 1 1⟩ direction which are 30° and 60° with respect to the substrate surface while for Si (1 1 1) most of the NWs are aligned vertically along the ⟨1 1 1⟩ direction. In particular, we observed that there appears to be two types of NWs; one with a straight and another with a tapered shape. The morphological and dimensional evolution of the Si-NWs is significantly related to atomic diffusion kinetics and energetics in the vapor–liquid–solid processes.
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1. Introduction

The ongoing reduction of electronic device size has led to a transition of technological approach from top–down to bottom–up due to current lithographic limitations. The controlled fabrication of the self-organized nanostructures as a building block in the bottom–up approach has been significant for advanced technological applications. In particular, one dimensional silicon nanowires (Si-NWs) have recently become of interest for potential applications in various technologies such as optics, electronics, and chemical sensors. This is the Si-NWs can offer the possibility of integration with conventional Si integrated-circuit technology [1–3]. The quantum effects in the electronic and optical properties of the nanodevices are strongly related to nanostructure's dimensions [2]. Therefore, good control of the dimensions and alignments of the NWs is required to employ them as elements of nanodevices.

Vapor–liquid–solid (VLS) growth method has been widely employed for the NW growth of various materials [3]. In metal island-catalyzed growth of Si-NWs via the VLS processes, the diameter and alignment of the NWs can be readily controlled by the size of the catalytic metal islands and the substrate orientation [4,5]. However, the diameters of the reported NWs did not correspond to that of the metal islands but were extensively modified with variation in growth conditions [4,6]. Thus, detailed studies on the correlation of the growth parameters with morphological evolution of Si-NWs in the VLS processes are still required for the well-controlled growth. In the VLS growth mechanism, the evolution of the NWs proceeds with three well-known stages: metal alloying process, crystal nucleation, and axial growth [7]. These processes involve mass transport through metal alloying and energetics of the

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system. Thus, these factors will determine the dimension and orientation of the evolving Si-NWs.

In this study, we investigate the morphological evolution of Si-NWs on Si substrates grown by nanoscale Au–Si island-catalyzed rapid thermal chemical vapor deposition (RTCVD). The initial nucleation of the NWs from the Au–Si islands is examined while varying island sizes. Also, we investigate the variation in the morphology and dimension of the NWs depending on the growth pressures, temperatures and times. In particular, preferential growth directions of the NWs are identified for Si (0 0 1) and (1 1 1) substrates. The results are discussed in terms of atomic mass transport and energetics at interfaces of vapor/liquid and liquid/solid in the NW growth via the VLS mechanism.

2. Experimental procedure

P-type Si (0 0 1) and (1 1 1) wafers were employed as substrates. The wafers were cleaned ultrasonically with acetone and methanol for 10 min, and then rinsed under running de-ionized water. In order to remove native oxide, the wafers were dipped into 2% HF ($HF:H_2O = 1:50$) for 3 min and then flushed by dry nitrogen. The cleaned wafers were transferred into an e-beam evaporator chamber to deposit 1.2–3.0 nm thick Au films with a growth rate of 0.01 nm/s at room temperature. Then, the Au-deposited substrates were transferred to the RTCVD chamber, where they were annealed in hydrogen ambient at 0.5 Torr and 700 °C for 10 min to form Au–Si islands. After the island formation, the substrate temperature was reduced to Si-NW growth temperatures and the substrates were exposed to a mixture of SiH_4 (1–4 sccm) and H_2 (50 sccm) for 30–120 min. The growth temperatures and total chamber pressures are 500–600 °C and 0.1–1.0 Torr, respectively. The morphology of the grown Au–Si islands and the Si-NWs were characterized by using a field emission scanning electron microscopy (FESEM) for a 30° tilted view and cross-sectional geometries. The dimensions of the Au–Si islands and the Si-NWs were measured from the obtained SEM images.

3. Results and discussion

To explore initial nucleation of Si-NWs depending on catalysis island size, we initiated the NW growth with varying Au film thicknesses (1.2, 2.0 and 3.0 nm). Fig. 1 shows the substrate morphology before and after exposing the annealed Au films to the silane (SiH_4) mixture gases. Upon annealing at above the Au–Si eutectic temperature (~360 °C), the Au film reacts with Si substrate and dissolves Si to form Au–Si alloy liquid [8]. Further annealing leads to a transformation of the liquid into Au–Si alloy droplet structures, whose shape is determined by minimization of the surface and interface energy of the liquid/substrate. Also, the composition of the Au–Si liquid alloy droplets will follow the liquid at annealing temperatures [8]. Thus, the islands in Figs. 1(a) and (b) were formed from the Au–Si alloy droplets after cooling down to room temperature. For annealing temperature of 700 °C, the composition of the Au–Si alloy islands might be ~9% Si, which can be estimated from Au–Si binary phase diagram [8]. The surface shape of the islands is smooth and circular even though the edge of the islands are irregular in Figs. 1(a) and (b), indicating that the islands were formed from the liquid droplets.

As the Au film thickness was increased, the average size of the islands became larger while the size distribution was broader and the number density of the islands was reduced. For a 1.2 nm thick Au film, the average diameter of the islands was ~8 nm and all islands were smaller than 20 nm (Fig. 1(a)) while for 2.0 nm Au film, the average diameter was ~13 nm and the fraction of the islands smaller than 20 nm was about 85% (not shown in Fig. 1). For further increase in thickness of 3.0 nm, the average diameter was increased to 15 nm and the fraction of the islands smaller than 20 nm was decreased to 77% (Fig. 1(b)). These results indicate that for thicker films the initially nucleated Au–Si alloy droplets would tend to grow larger through droplet coarsening with neighboring droplets [9]. Thus, the diameter of the islands will be more uniform for Au films thinner than 1.2 nm at a given annealing temperature.

After silane exposure of the Au–Si droplets on Si in Figs. 1(a) and (b), we observed the formation of nanostructures on the surface. Figs. 1 (c) and (d) display the resulting SEM images obtained with 30° tilt with respect to the horizon. For the 1.2 nm Au film, the nanostructures are observed to be pillar shaped over all surfaces (Fig. 1(c)). The nanopillars (NP) seem to be grown prior to the nucleation of the NWs [10]. For the 2.0 nm film, similar NP structures were observed. However, for the 3.0 nm film we observed the nucleation of the Si-NWs with average diameter of ~60 nm and length of ~350 nm (Fig. 1(d)). It may indicate that a critical thickness of Au films exists for the Si-NW nucleation in the given growth conditions. In other words, there exists a minimum size of Au–Si droplets to initiate Si-NW nucleation from the droplet catalysis. The bright tips of the NWs seem to be Au–Si droplets in Fig. 1(d). The diameters of the NWs are similar to those of the Au–Si droplets. The existence of a critical Au film thickness for initiation of NW nucleation may be related to competition of Si homogeneous nuclei formation in the Au–Si droplets with the adatom attachment at the Au–Si droplet-substrate interface [11].

To examine the effects of growth pressure on the Si-NW morphology and dimension, a series of samples of 3.0 nm of Au films were exposed to the silane mixture gases at total pressures ranging from 0.1 to 1.0 Torr. For 0.1 Torr, the surface morphology displays an early stage of the nucleation of the Si-NWs (Fig. 2(a)). Most of the nanostructures seem to be NP shaped and some are of NW shape. As the growth pressure increased to 0.5 Torr, more NWs nucleated and the NWs coexisted with the NPs (Fig. 2(b)). For 1.0 Torr, most of the NPs transformed to the NWs and the resulting surface shows randomly

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