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Thickness effect of catalyst layer on silicon nanowires morphology and features

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ABSTRACT

Silicon nanowires (SiNWs) have been synthesized on gold layer-coated silicon substrates via plasma enhanced chemical vapor deposition method (PECVD). Various thicknesses of Au layers were coated on Si (111) substrates using radio frequency magnetron sputtering. Field emission scanning electron microscopy (FESEM), X-ray diffraction (XRD), energy dispersive X-ray spectroscopy (EDX), and transmission electron microscopy (TEM) were used to characterize the morphology, compositions, and structures of the samples. The results show that the sample consisted of single-crystalline SiNWs with the diameters ranging from 40 to 160 nm and length up to 3 μm. It was observed that the diameter of SiNWs increases with increasing of Au catalyst layers thickness. Raman spectra display peaks with narrow and asymmetric shape at 518 cm^{-1} for the SiNWs, indicating the high crystalline nature of the samples. A possible growth mechanism is proposed for the formation of nanowires (NWs). It has been found that the features of SiNWs depend on the thickness of Au layers.

1. Introduction

One-dimensional nanomaterials with specific features, structure, morphology, and size have attracted intensive research interests due to their potential application in nonoscale electronics, optoelectronic devices, and building blocks. Many applications of SiNWs in devices such as photonic devices, solar cells, sensors, field-effect transistors have been demonstrated so far [1–4]. Several synthetic methods such as thermal vapor deposition, laser ablation, chemical vapor deposition (CVD) and plasma enhanced chemical vapor deposition (PECVD) have been used for synthesis of SiNWs which allow synthesis of crystalline SiNWs with controllable features and sizes [5–8]. These methods often lead to grow NWs with various qualities and features. Among the mentioned methods, PECVD with different frequencies is one of the most advanced techniques which is classified as a direct deposition method. In the PECVD process a precursor gas is dissociates by the plasma into highly active radicals, which can be operated to grow SiNWs.

In addition, SiNWs are fabricated through various mechanisms with different growth catalysts, but they are still under investigation. The vapor–liquid–solid (VLS) mechanism that was first suggested by Wagner and Ellis appears to become the main method used to synthesize SiNWs [9]. This mechanism requires a metal in liquid state as a catalyst. Moreover, among the most commonly metal catalysts, Au is an appropriate catalyst to grow SiNWs. Compared with other conventional catalysts, Au has several advantages such as chemical inertness, high resistant to oxidation, good thermal stability and can be deposited easily as liquid droplets or thin film layers [10,11]. Since the morphology and features of NWs depend on size of the catalyst, it is important to control diameters and thickness of Au catalyst.

In this work, Au nanolayers as a catalyst were deposited on silicon substrate to grow SiNWs by PECVD (13.5 MHz) technique. We report the role of Au films thickness on morphology and features of SiNWs.

2. Experimental details

SiNWs were synthesized on Si (111) substrates, deposited by various thickness of Au layer, using a PECVD technique. Si substrates were cleaned by immersing them in a mixture of HCl, DI water and H_2O_2 solution. At the next step, the cleaned substrates were immersed in 10% hydrofluoric acid to remove the surface oxide layer. Then, it was rinsed with DI water and dried by nitrogen. The cleaned substrate was deposited with different Au films thickness ranging from 5 to 20 nm at room temperature by a radio frequency magnetron sputtering system at a base pressure of 2.5×10^{-6} Torr. The Au-deposited substrates were heated to 400 °C in a vacuum chamber to produce Au catalyst islands. Finally, the Au

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deposited Si substrates were placed in a PECVD chamber to grow SiNWs. For the synthesis of the SiNWs, silane (SiH_4) gas with purity of 99.9995% as the Si source with 10 sccm was introduced to vacuum reactor operated at the pressure of 80 mTorr and at deposition times of 10 min. During the growth, the RF power was kept at 10 W and the substrate was heated up 450 °C. Microscopic structure of the NWs was taken by a field emission scanning electron microscope (FESEM, JEOL, JSM-6701F) and high-resolution transmission electron microscopy (HRTEM) with a JEOL, JEM-2100 microscope. The optical properties were analyzed by an NIR, FT-Raman spectrum (Perkin Elmer Spectrum GX System) with a Nd crystal laser source.

3. Results and discussion

At the first step, samples were prepared according to described methodology in Section 2. At the next step, different Au layers ranging from 5 to 20 nm were deposited on the Si surface and annealed at 400 °C in pure Ar atmosphere. After annealing of the as-deposited Au film (see Fig. 1), Au nanoparticles (NPs) begin to form on the surface of the substrate with different sizes.

Table 1 represents the specifications of samples with different thickness of Au layers deposited on the Si substrates and also various sizes of obtained Au NPs from annealing process. Fig. 1 shows FESEM images of performed Au catalysts on the Si substrate after heat treatment at 400 °C. As is clear from Fig. 1, the sizes of formed NPs on Si substrates were increased with increasing of Au layer thickness. The mean diameter of Au NPs in Fig. 1(a) is about 5 nm, and their areal density is about 1.7×10^{12} cm^{-2}. When the Au film thickness is about 10 nm NPs with diameters about 10 nm were formed on the surface of the substrate (see Fig. 1(b)). According to Fig. 1(c), as the Au films thickness is increased to 15 nm of the number of NPs decreased while their sizes are increased. As shown in Fig. 1(d), further increase in the Au film thickness to 20 nm caused formation of bigger NPs. Moreover, Fig. 1 reveals that with an increase of Au films thickness ranging from 5 to 20 nm, the average diameter of NPs enlarges to about 35 nm while their areal density decreases to 4.0×10^{10} cm^{-2}. The insets in Fig. 1 exhibit the statistical analyses of NPs related to their size distribution histograms. It has been seen that most of the created NPs from Au films thickness of 5, 10, 15 and 20 nm have the diameter of about 5, 10, 15 and 25 nm, respectively. These results clarifies that the size of Au NPs increases when the Au film thickness increases, while their areal density decreases.

We use these results to discuss about mechanism of Au NPs formation. It is found that with annealing of the Au layer at 400 °C, the Au atoms are thermally activated and flowed around at the Si surface and nucleated into NPs. With increasing of the Au films thickness, more Au atoms diffused at the Si surface and lead to form bigger Au NPs via the coalescence of smaller ones. In fact, several Au NPs touched with each other or located very closely, to reach to lower their interface and surface energy and thus the total matrix energy.

Fig. 2 demonstrates FESEM images of SiNWs grown on Au-deposited substrates with Au films thickness ranging from 5 to 20 nm, respectively. These images reveal that the morphology and areal density of SiNWs depend on Au NPs sizes, resulting in catalyst films thickness. According to Fig. 2, the average diameters of SiNWs increased with increasing of Au film thickness while its areal density was decreased. In addition, Fig. 2 exhibits that morphology and areal density of SiNWs can be controlled by the catalyst morphology and density. Fig. 3 shows the diameter distribution of the obtained SiNWs from various thickness of Au film. These histograms estimated from corresponding images of Fig. 2. The mean diameter of SiNWs grown for Au film with thickness of 5, 10, 15 and 20 nm are found to be about 40, 80, 100 and 145 nm, respectively. Furthermore, the average lengths of SiNWs grown with Au film thickness of 5, 10, 15 and 20 nm are 0.7, 1.2, 1.9 and 2.8 μm,

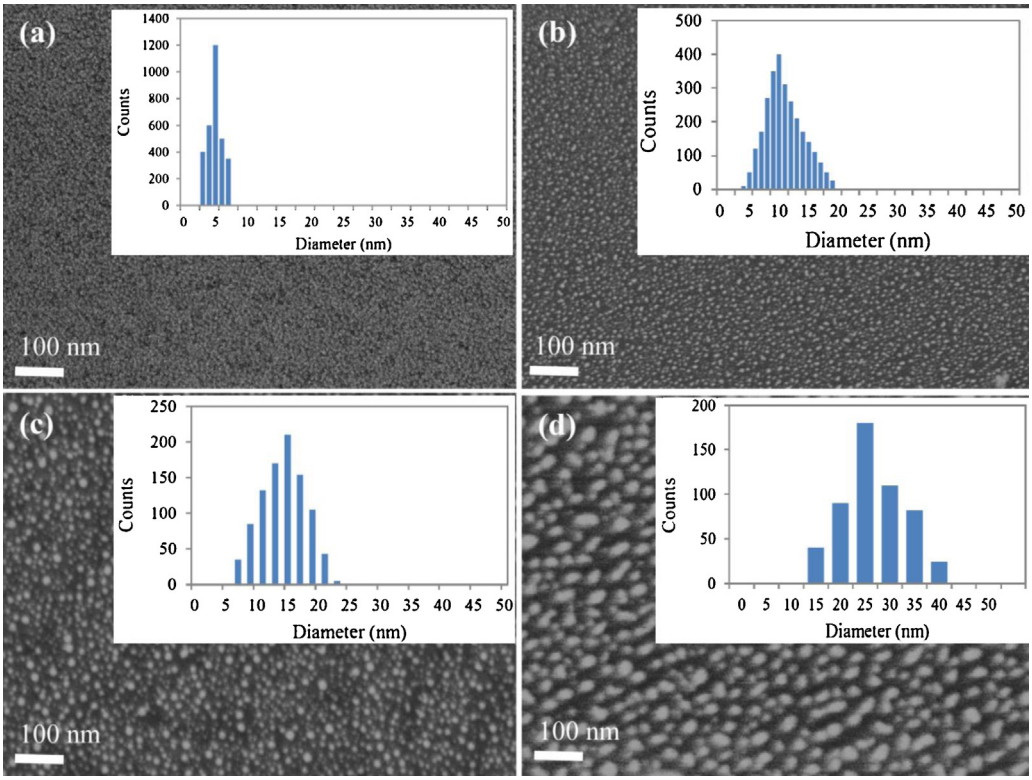

Fig. 1. FESEM images of generated Au NPs from Au film with thickness of (a) 5 nm, (b) 10 nm, (c) 15 nm, and (d) 20 nm on the Si substrate after annealing treatments at temperature of 400 °C. Insets show corresponding size distribution histograms.