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Effect of nanohole size on selective area growth of InAs nanowire arrays on Si substrates

Xiaoye Wang[a,d,*], Wenyuan Yang[c], Baojun Wang[c], Xianghai Ji[a], Shengyong Xu[c], Wei Wang[c], Qing Chen[c], Tao Yang[a,b,*]

[a] *Key Laboratory of Semiconductor Materials Science, Institute of Semiconductors, Chinese Academy of Sciences, 100083 Beijing, China*
[b] *College of Materials Science and Opto-Electronic Technology, University of Chinese Academy of Sciences, 100049 Beijing, China*
[c] *School of Electrical Engineering and Computer Science, and Key Laboratory for the Physics and Chemistry of Nanodevices, Peking University, 100871 Beijing, China*
[d] *National Key Laboratory of Science and Technology on Vacuum Technology and Physics, Lanzhou Institute of Space Technology and Physics, P.O. BOX 94, 730000 Lanzhou, China*

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ABSTRACT

We have investigated the influence of nanohole size on selective-area growth (SAG) of InAs nanowire (NW) arrays on Si(111) substrates by metal-organic chemical vapor deposition. The growth of well-defined and position-controlled InAs NW arrays with united vertical orientation can be achieved on the patterned substrates with a certain range of nanohole size, which paves the way for the fabrication of high-electron-mobility and surrounding-gate transistor arrays using NWs as channels. Moreover, it is found that more than one NW are increasingly likely grown per nanohole as the nanohole size increases, and the NWs become increasingly thin and short. This is considered to be due to the supersaturation of adsorbed species in the nanohole and the intense competition for adatoms among multiple NWs per nanohole.

1. Introduction

One-dimensional semiconductor nanowires (NWs) are promising for the use in nanostructure electronic and optical devices. As a kind of technologically important semiconductor material, II-VIII-V semiconductor NWs have attracted much attention because of their advantages, such as unique optoelectronic property, small diameter of NW, large surface-to-volume ratio of NW, radial relaxation of strain between the lattice-mismatched materials as well as easy integration with Si directly. In the last ten years, various types of planar devices based on III-V NWs have been fabricated [1–5]. In particular, vertical III-V NWs grown on silicon create a promising opportunity in the integration of 3D nanodevices with modern CMOS technology [6,7]. In order to achieve functional NW devices with predictive performance, it is necessary to control over position, directionality, size and crystal structure of the NWs. So far, foreign metals, such as gold, are employed as catalysts for the growth of NWs. However, gold may form unwanted deep-level recombination centers in Si band gap [8,9] and degrade the electronic properties of NWs [10,11]. Therefore, it is highly desirable to develop the site-controlled growth of III-V NWs without any foreign catalysts.

To meet these demands, selective-area growth (SAG) of III-V NWs on patterned Si substrates, which involves a combination of bottom-up (epitaxial growth) and top-down (mask by electron beam lithography) approaches, has been investigated by many groups [12–17]. Some groups have demonstrated good control of NW position [12,13] and vertical directionality [14,15]. The dependences of NW size and growth rate on pitch between nanoholes have also been studied in details [16,17]. Besides, it was found that the performances of NW devices, such as electron field-effect mobility, threshold voltage, I-V characterization and I_{on}/I_{off} ratio, are primarily affected by the diameter of NWs [18–20], so it is necessary to study the influence of nanohole size on the diameter of NW arrays.

In this paper, we report on the influence of nanohole size on SAG of vertical InAs NW arrays on lithographically defined Si(111) substrates by metal-organic chemical vapor deposition (MOCVD). The growth was carried out under identical growth conditions, it is observed that the diameter of the NWs decreases and more and more NWs are grown in each hole as the nanohole size increases. InAs NW arrays with different nanohole sizes exhibit very different morphologies.

* Corresponding authors at: Key Laboratory of Semiconductor Materials Science, Institute of Semiconductors, Chinese Academy of Sciences, 100083 Beijing, China.
E-mail addresses: ywangx06@semi.ac.cn (X. Wang), tyang@semi.ac.cn (T. Yang).

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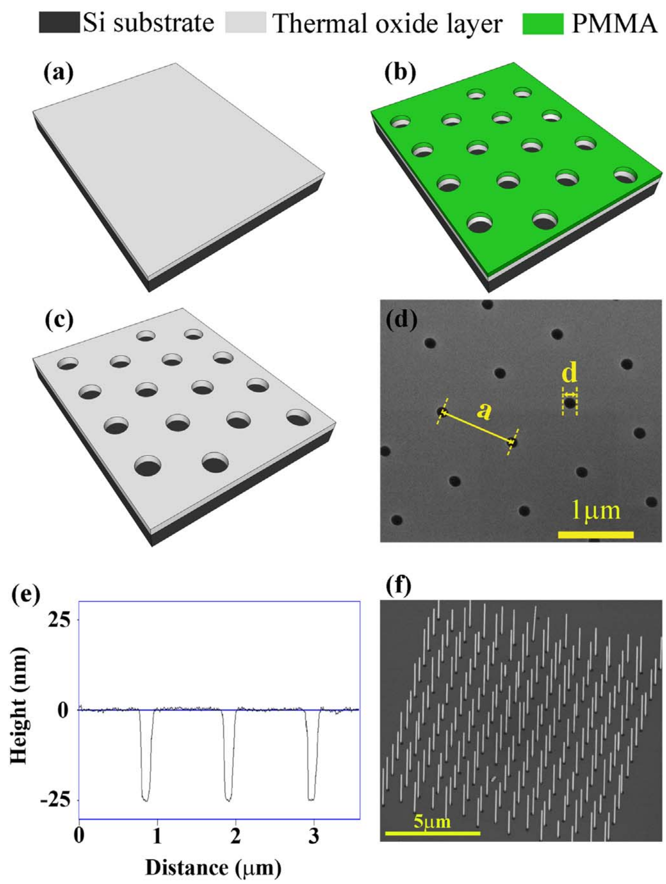

Fig. 1. The fabrication process of patterned substrates. (a) Thermally oxidized Si substrate. (b) Substrate with PMMA after EBL exposure, development and wet-chemical etching. (c) Substrate with nanoholes in the oxide layer after removing PMMA. (d) SEM image of nanoholes. (e) Depth of nanoholes measured by AFM. (f) SEM image of InAs NW arrays grown on patterned substrate.

2. Experimental details

The fabrication process of patterned Si substrates is schematically illustrated in Fig. 1. First, as shown in Fig. 1a, a 25-nm-thick SiO_2 layer was thermally oxidized onto the p-type Si(111) substrate. Then, the substrates were experienced coating PMMA, electron beam lithography (EBL) exposure, development and wet-chemical HF etching, a pattern of nanoholes was transferred into the SiO_2 layer, as shown in Fig. 1b. Finally, a patterned substrate was obtained after removing the PMMA, as shown in Fig. 1c. All of the nanoholes are arranged in a shape of regular triangle with a pitch of 1 µm. Fig. 1d and e show a typical scanning electron microscopy (SEM) image of the nanoholes and their depth measured by atomic force microscopy (AFM), respectively.

InAs NW arrays were grown on patterned Si(111) substrates by a close-coupled showerhead, low-pressure MOCVD system (AIXTRON Ltd., Germany). Before the growth of NWs, substrates were cleaned in a solution of acetone using an ultrasonic bath in order to remove organic impurity and contaminant, then etched in a 1.9% aqueous hydrofluoric (HF) acid solution for short time to remove native oxide layer within the nanoholes and expose the Si(111) surface. After etching, the substrates were rinsed in deionized water for 20 s, dried with N2 and then loaded into the MOCVD reactor as quickly as possible. Ultra-high purity H_2, trimethylindium (TMIn) and AsH_3 were used as carrier gas, In precursor and As precursor, respectively. The total flow rate of H_2 was 12 slm and the chamber pressure was 133 mbar during the growth. The substrates were first heated in situ to 635 °C for 5 min and then cooled down to 400 °C. Next, the substrates were heated to the growth temperature of 565 °C in an AsH_3-rich (2.0×10^{-4} mol/min) atmosphere. After a short stabilization time, InAs NWs began to grow as introducing TMIn (0.79×10^{-6} mol/min). The growth time of the NWs was 3 min. Finally, the substrates were cooled down in an AsH_3 ambient to prevent the decomposition of NWs. Fig. 1f shows a SEM image of InAs NW arrays grown on patterned substrates.

3. Results and discussion

Fig. 2 shows SEM images of InAs NWs via SAG during a same growth run. From Fig. 2a to f, the nanohole sizes are 160 nm, 170 nm, 185 nm, 205 nm, 250 nm and 300 nm, respectively. It can be seen that well controlled growth (one NW per nanohole) of NW arrays with high yield can be achieved on patterned Si substrates with the nanohole sizes of 160 nm and 170 nm, as shown in Fig. 2a and b. NWs have a constant diameter from base to top and a hexagonal cross-section. However, in the Fig. 2c, two NWs are grown in some nanoholes with the size of 185 nm. As the nanohole size further increases, multiple NWs and parasitic islands form in the nanoholes. For the nanohole with the size of 300 nm, most of the nanoholes are filled with multiple short and thin NWs.

With a few exceptions, most of the NWs grow along the vertical [111] direction. There are at least two reasons for this: First, symmetry considerations indicate the existence of four equivalent [111] directions on a group-IV (111) substrate, including one vertical and three tilted 19.6° with respect to the substrate plane [21]. But very small area of Si surface within the small nanoholes exposes the vertical [111] direction, leading to anisotropic [111] crystal growth of NWs. Second, the substrate surface is exposed to the group-V precursor flow prior to NW growth, so passivation of the Si surface dangling bonds takes place with group-V atoms [22]. This forms an As-incorporated, -reconstructed (111)B plane on the exposed Si surface areas, lowering the symmetry of the system so that only the vertical [111] direction growth is preferential.

Fig. 3, as a detailed statistics, shows the fractions of single NW, two NWs, three NWs and over three NWs per nanohole with different sizes, respectively. Here, the fraction of single NW per nanohole is defined as the number of nanoholes which only produce one NW per nanohole divided by the total number of nanoholes in one array. The fraction of multiple NWs per nanohole is defined in a same way as the number of nanoholes which form multiple NWs per nanohole divided by the total number of nanoholes in one array. By summarizing Fig. 2 and Fig. 3, it can be seen that the small size of nanoholes have high probability of growing one NW per nanohole while the large size of nanoholes have high probability of growing more than one NW per nanohole. There should be two reasons for this phenomenon. First, both SAG regime that is suitable for smaller nanoholes [23] and the strain-mediated, direct-heteroepitaxial growth regime that takes place for larger nanoholes all exist. It has to be proven whether the strain-mediated, direct-heteroepitaxial growth regime [24–26] or In particle-catalyzed growth regime [27] is applied in larger nanoholes. One way to confirm which growth regime plays a role in the growth of InAs NWs in larger nanoholes is to compare InAs NWs on oxide-templated InAs (111)B wafers with the already shown growth results using Si wafers. Because the growth of InAs NWs on InAs (111)B is homoepitaxy, the diameter of NWs is same to or larger than the radius of nanoholes on InAs (111) B, multiple NWs cannot grow in larger nanoholes on InAs substrates [28]. This compare between the growths on Si (111) and InAs (111)B can be explained by strain-mediated, direct-heteroepitaxial growth regime. Therefore, single InAs NW is grown in smaller nanoholes due to SAG regime and multiple InAs NWs are grown in larger nanoholes due to strain-mediated, direct-heteroepitaxial growth regime.

Second, the adsorption efficiency of species on the exposed Si within the nanoholes would determine the available material for growth. Assuming that the surface diffusion length on the exposed Si is larger than the nanohole size, the supersaturation of adsorbed