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Suppression of surface nanocrystal nucleation in growth of GaAs nanowire on Si(111) by molecular beam epitaxy

Jinkwan Kwoen[1], Naoto Kumagai[2], Katsuyuki Watanabe[2], Shunsuke Ohkouchi[2], Satoshi Iwamoto[1, 2], and Yasuhiko Arakawa[1, 2]

[1] Institute of Industrial Science (IIS), The Univ. of Tokyo
4-6-1 Komaba, Meguro, Tokyo 153-8505, Japan
Phone: +81-3-5452-6291 FAX: +81-3-5452-6247 E-mail: jkkwoen@iis.u-tokyo.ac.jp
[2] Institute for Nano Quantum Information Electronics (NanoQuine), The Univ. of Tokyo
4-6-1 Komaba, Meguro, Tokyo 153-8505, Japan

1. Introduction

III-V compound semiconductor nanowires have been attractive for applications in photonics [1-3] and quantum information technology [4] due to their small diameter of less than a few hundred nm and height of several μm. To date, many reports have been made on nanowire growth of homo- and hetero-epitaxial growth systems, for example, GaAs nanowires on GaAs substrate [5], GaAs nanowires on silicon (Si) substrate [6], InAs nanowires on Si substrate [7], and so on.

Among these growth systems, growth of III-V compound semiconductor nanowires on Si substrates [2-3] is of great interest in the Si nano-photonics field, because it enables the monolithic integration of the optical devices based on III-V compound semiconductor with Si based devices. Also, nanowire growth on lattice-mismatched substrates can overcome the problem of dislocations generated from the interface due to the lattice constant mismatch. Nanowire growth can reduce the interface areas between the lattice-mismatched materials so that high quality crystals without dislocations may be obtained.

Self-catalyzed vapor-liquid-solid (VLS) growth, in which complicated pre-growth processes such as electron beam lithography or metal catalyst deposition are not necessary, is one of the promising methods to obtain nanowires. In most cases, nanowires are grown on the substrates, on which oxide layers are formed. In these cases, ground structures comprising the nanocrystals are commonly incorporated with the nanowire growth. These nanocrystal structures cause indefinite emissions and prevent exact characterization of the fabricated nanowires.

In this paper, we report on the suppression of nanocrystal structure formation on the Si substrate for the fabrication of GaAs nanowires using the self-catalyst VLS growth method. This is achieved by controlling the As pressure before and during nanowire growth. The generation of nano crystals was drastically reduced by thermal cleaning without As flux and following supply of Ga before nanowire growth.

2. Experimental

Nanowire samples are grown by molecular beam epitaxy (MBE) equipped with an As valved cracking cell which gives good controllability of As flux. We used n-type Si (111) substrates without any surface treatment (as-received substrate). Therefore, a native oxide layer (~2nm) exists on the substrate surface. We prepared three kinds of samples grown by different conditions.

The first one (Sample I) was grown using a conventional condition with a self-catalyzed VLS growth method [6]. The substrate was thermally cleaned at 580 °C for 600 s under an As flux of 1.20×10^{-5} Torr. Then, we supplied a Ga flux (6.0×10^{-7} Torr) with As flux for 300 s in order to grow the nanowires. The growth procedures of the second one (Sample II) are the same as those of the sample I except for the Ga flux during the nanowire growth. The Ga flux for the sample II was 3.0×10^{7} Torr. The third one (Sample III) was prepared as follows. First, a Si substrate was heated up to 580°C without As flux and kept for 600s at the same temperature for surface cleaning. Then, only a Ga flux of 3.0×10^{-7} Torr was supplied for 60 s. After that, the nanowires were grown for 300 s with the Ga and an As flux. All the samples were cooled to room temperature in the As flux after the nanowire growth, and the surface structures were evaluated by scanning electron microscopy (SEM).

3. Results and discussions

Figure 1 shows an SEM image of Sample I. GaAs nanowires of ~40 nm radius and ~2.2 μm height with the surface density of 4.2×10^{7} cm^{-2} are formed. As shown in the top view in Fig. 1, almost the entire surface was covered with ground structures. The coverage was ~81%. These ground structures may be composed of nano crystals such as poly and/or amorphous GaAs. When the Ga flux during the nanowire growth was reduced, the densities of the nanowires and nano crystals were changed as shown in Fig. 2. That is, the surface density of the nanowires was increased to 1.1×10^{8} cm^{-2} and that of nano crystals was reduced to approximately one third compared to Sample I. In addition, the area covered by nanocrystal structures was decreased to ~13%. This change in the densities was caused by the reduced Ga flux during the nanowire growth. In other words, the supplied Ga was consumed to form the nanowires instead of the nucleation of the nano crystals by reducing the V/III ratio during the nanowire growth.

Figure 3 shows an SEM image of Sample III. The densities of both GaAs nanowires and nano crystals are drastically reduced compared to Sample I and II. The typical density of the nanowires was 8.3×10^{6} cm^{-2} and nano crystals were seldom observed on the surface. In addition, the area covered by nanocrystal structures

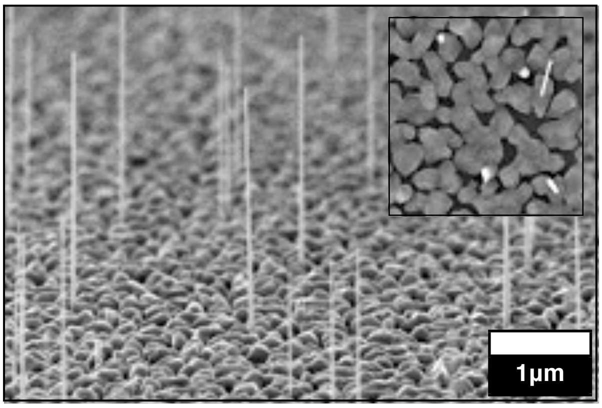

Fig. 1 Typical SEM image of sample I. The inset is top view of the surface and its area is 2μm×2μm square.

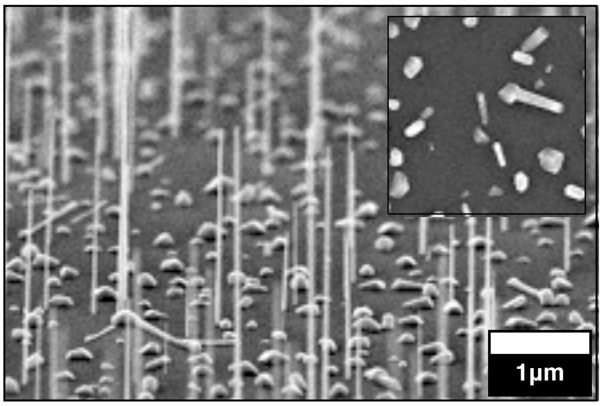

Fig. 2 Typical SEM image of sample II. The inset is top view of the surface and its area is 2μm×2μm square.

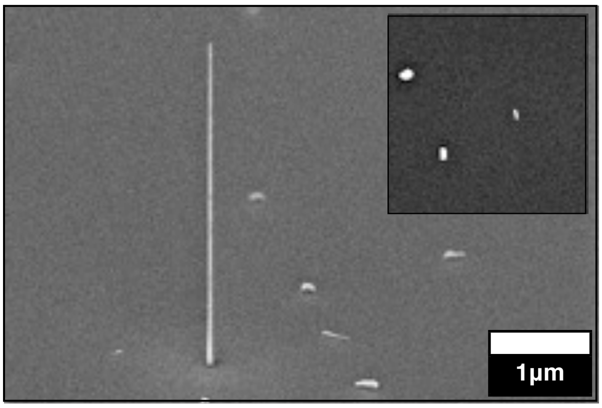

Fig. 3 Typical SEM image of sample III. The inset is top view of the surface and its area is 2μm×2μm square.

was decreased less than 1%. The difference in the procedures between Sample I, II and III is the surface treatment before the nanowire growth in the growth chamber of MBE system. In the cases of the sample I and II, As ambient pressure remains in the growth chamber before the nanowire growth, because the As flux was irradiated for the surface cleaning. On the other hand, in the case of Sample III, there is no As ambient pressure before the nanowire growth, because the thermal cleaning was performed without As flux. Furthermore, in this case, Ga was supplied without As pressure before nanowire growth, resulting in Ga droplet formation on the surface. Although the precise mechanism is not clear, this Ga droplet formation and/ or termination with Ga on the surface may be related to the drastic change in the surface densities of the GaAs nanowires and nano crystals.

4. Conclusion

We have achieved the suppression of nanocrystal formation on the Si substrate surface in self-catalyzed VLS growth of GaAs nanowires on Si substrates by reducing the V/III ratio during the nanowire growth. In addition, we realized the formation of low density GaAs nanowires of 8.3×10^{6} cm^{-2} with little incorporation of nano crystals by eliminating the As flux and following Ga supply before nanowire growth. The fabricated sample using this new method can be used for the exact evaluation of the optical properties of single nanowires.

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