GaAs nanowires on Si substrates grown by a solid source molecular beam epitaxy

Soo-Ghang Ihn and Jong-In Song[a)]
Department of Information and Communications, Gwangju Institute of Science and Technology (GIST), 1 Oryong-dong, Buk-gu, Gwangju 500-712, Korea

Young-Hun Kim and Jeong Yong Lee
Department of Materials Science and Engineering, Korea Advanced Institute of Science and Technology (KAIST), 373-1 Guseong-Dong, Yuseong-Gu, Daejeon 305-701, Korea

(Received 13 March 2006; accepted 13 June 2006; published online 31 July 2006)

High-quality Au-catalyzed GaAs nanowires were grown on Si substrates by vapor-liquid-solid growth in a solid source molecular beam epitaxy system. X-ray diffraction, scanning electron microscopy, and high-resolution transmission electron microscopy reveal that the GaAs nanowires were epitaxially grown on Si substrates with uniform diameters along the nanowires. While GaAs nanowires on Si(111) and (001) substrates were mainly grown along the ⟨111⟩ direction with zinc-blende and wurtzite structures, unusual GaAs nanowires grown along ⟨001⟩ with a pure zinc-blende structure were also observed. Strong photoluminescence was observed from GaAs nanowires grown on a Si(001) substrate at room temperature. © *2006 American Institute of Physics*.
[DOI: 10.1063/1.2245348]

In the past few years, GaAs nanowires have been widely investigated due to their potential as key building blocks for future electronic and photonic devices. Metal-organic chemical vapor deposition (MOCVD),[1] chemical beam epitaxy,[2] and molecular beam epitaxy[3] (MBE) have been usually employed for the growth of GaAs nanowires using vapor-liquid-solid (VLS) growth.[4] In particular, Wu *et al.* reported that MBE-based VLS growth of GaAs nanowires under ultrahigh vacuum condition provided dramatic suppression of lateral growth that was usually observed in CVD-based VLS growth, demonstrating MBE-grown GaAs nanowires on GaAs substrates having homogeneous diameters along the wire axis.[3] GaAs nanowires have been usually grown on GaAs, especially on GaAs(111)B substrates for vertical growth since the preferable VLS growth direction of GaAs nanowires is ⟨111⟩.[5,6] While wide band gap compound semiconductor nanostructures such as GaN and ZnO nanorods synthesized on Si(111) substrates using MOCVD or MBE have been reported frequently,[7–9] epitaxial growth of GaAs nanowires on Si substrates has been rarely reported except for MOCVD growth of GaAs nanowires on Si by Mårtensson *et al.*[10] In this letter, we demonstrate epitaxially grown GaAs nanowires on Si substrates by using a solid source MBE (SSMBE) system.

GaAs nanowires were grown on Si substrates by using a VG V80H-10K MBE system equipped with an arsenic valved-cracker cell. The Si substrates were degreased and then immerged in diluted HF (1%) for native oxide removal and hydrogen passivation. After removing the native oxide, the Si substrates were instantly loaded in an electron beam evaporator for deposition of a thin (~4 Å) Au layer. The Au-deposited Si substrates were then transferred into the MBE growth chamber and annealed at 540 °C for 10 min by using a substrate heater of the MBE to generate Au nanoislands. GaAs nanowires were grown at 580 °C with the V/III ratio of 9 with a fixed Ga beam flux which is required for the homoepitaxial GaAs growth rate of 0.9 μm/h on a GaAs(001) substrate. The growth was terminated by switching off the Ga supply while maintaining the As_2 supply until the substrate temperature fell below 300 °C.

Structural properties of the GaAs nanowires were investigated by using field-emission scanning electron microscopy (FE-SEM), high-resolution transmission electron microscopy (HRTEM), and x-ray diffraction (XRD). HRTEM images were obtained from the GaAs nanowires dispersed on carbon-film-coated Cu grids through sonication in isopropyl alcohol. Optical property of as-grown GaAs nanowires on a Si(001) substrate was simply checked by using room-temperature photoluminescence (RT-PL) measurement.

Figure 1(a) shows a cross-sectional SEM image of the GaAs nanowires grown on a Si(111) substrate for 5 min. Lateral size and height of the wires are approximately 10–40 nm and 0.3–1.1 μm, respectively. The height of the nanowires is reported to be dependent on the lateral size of nanowires or Au seed particles.[11] The SEM image shows that most of the GaAs nanowires on a Si(111) substrate were well grown along the [111] direction with homogeneous diameters along the wire axis. Coincidence of the crystallographic directions of the nanowires and the substrate indicates that the GaAs nanowires are epitaxially grown. As shown in Fig. 1(b), the GaAs nanowires on the Si(111) substrate were analyzed by an x-ray diffraction symmetric $2\theta/\omega$ scan. Wavelength of the incident radiation for the XRD measurements was selected to be 1.540 598 Å, and the incident beam was focused by using hybrid optics that consist of a double crystal Ge(220) monochromator and a graded multilayer mirror. Diffracted signals were collected by a point detector with a receiving slit. There are no significant peaks observed within the whole scan range (from 20° to 80°) except for the GaAs (111) diffraction peak at 27.308° and Si(111) peak at 28.44°.

Figure 2 shows high-resolution TEM images of the GaAs nanowires grown on a Si(111) substrate for 5 min.

[a)]Author to whom correspondence should be addressed; FAX: +82-62-970-2204; electronic mail: jisong@gist.ac.kr

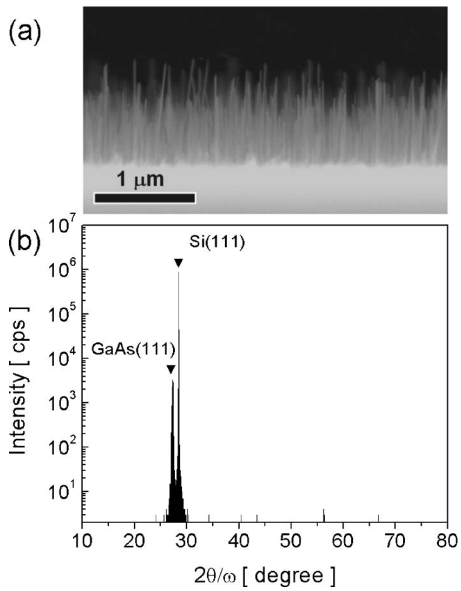

FIG. 1. (a) Cross-sectional SEM image of the GaAs nanowires grown on a Si(111) substrate at 580 °C with V/III ratio of 9 for 5 min and (b) x-ray diffraction symmetric $2\theta/\omega$ scan.

Figure 2(a) is the HRTEM image of the GaAs⟨111⟩ nanowire tip (for convenience, we use "GaAs⟨111⟩nanowires" to describe the GaAs nanowires grown along the ⟨111⟩ direction). As indicated in the figure, while most of the stem region had the hexagonal wurtzite structure, regions having the cubic zinc-blende structure with twin boundaries were observed occasionally, which is usually observed in GaAs and InAs nanowires.[5,12] Figure 2(b) shows the HRTEM image of another GaAs⟨111⟩ nanowire having pure wurtzite structure. The inset is two-dimensional Fourier transforms of the stem region of the GaAs nanowire having the hexagonal wurtzite structure and indicates that the zone axis is $[11\bar{2}0]$ and the growth direction of the GaAs nanowire is [0001]. Although no significant peak was observed in x-ray diffraction and it was very hard to find in SEM images, unusual GaAs⟨001⟩ nanowires (GaAs nanowires grown along the ⟨001⟩ direction) were observed through the HRTEM analysis. Figure 2(c) shows an HRTEM image of the GaAs⟨001⟩ nanowire. The inset is two-dimensional Fourier transforms of the stem region of the GaAs⟨001⟩ nanowire having the cubic zinc-blende structure. It indicates that the zone axis is [110] and the growth direction of the GaAs nanowire is [001]. No twinned structures or stacking faults were observed. It is interesting to note that the GaAs⟨001⟩ nanowire has a smaller diameter (around 10 nm) than those (10–40 nm) of the GaAs⟨111⟩ nanowires, which can be explained by size-dependent growth direction of nanowires. It is well known that the preferable growth direction of a nanowire is determined by surface free energy. The surface free energy of the orientation ⟨111⟩ is generally lower than that of ⟨001⟩.[13] However, the surface free energy of a nanowire is dependent on its diameter.[14] Cai *et al.* reported size-dependent growth direction of ZnSe nanowires and revealed that the surface and interface (between seed particle and nanowire) energies of the ZnSe nanowires change depending upon the diameters of the nanowires or seed particles. It is inferred that for the seed particle smaller than approximately 10 nm, the surface and interface energies of GaAs nanowires on Si(111) might be smaller for growth direction of ⟨001⟩ than that for ⟨111⟩. Further experimental research on the dependence of the GaAs nanowire growth direction on the size of catalytic Au particles is under way. It is also interesting to note that the interfacial plane between the Au seed particle and the GaAs⟨001⟩ nanowire shown in Fig. 2(c) is not perpendicular to the wire axis (growth direction), whereas typical VLS-grown GaAs⟨111⟩ nanowires shown in Fig. 2(a) have the interfacial plane perpendicular to the wire axis.

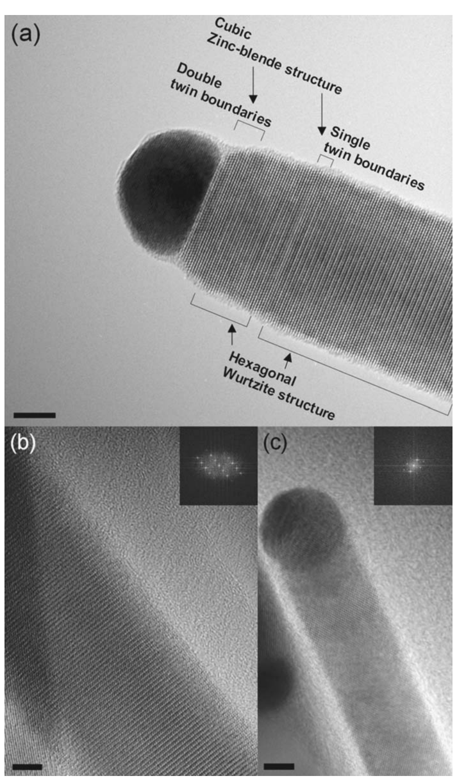

FIG. 2. High-resolution TEM images of (a) the tip region of the GaAs⟨111⟩ nanowire, (b) the stem region of the GaAs⟨111⟩ nanowire, and (c) the GaAs⟨001⟩ nanowire grown on a Si(111) substrate at 580 °C with V/III ratio of 9 for 5 min. The insets are two-dimensional Fourier transforms of the GaAs nanowires at the stem region. All scale bars are 5 nm long.

PL measurements were carried out at room temperature in order to investigate the crystalline quality of the GaAs nanowires on Si substrates grown by using an SSMBE system with an InGaAs detector and a 532 nm neodymium-doped yttrium aluminum garnett laser (optical power of 8.5 mW) as an excitation source. The PL spectrum was obtained from the GaAs⟨111⟩ nanowires grown on a Si(001) substrate at 580 °C with the V/III ratio of 9 for 40 min. The RT-PL shown in Fig. 3 is strong enough to reveal the quality of the GaAs nanowires. The peak at 863.2 nm with the full width at half maximum of 22.8 nm indicates that weak radial quantum confinement effect (~10 nm of blueshift compared to the peak of the bulk GaAs) can be observed.

In summary, high-quality GaAs nanowires epitaxially grown on Si substrates in an SSMBE system were demonstrated. It was observed that MBE-grown GaAs nanowires on Si substrates mainly have hexagonal wurtzite structure with ⟨111⟩ growth direction. However, unusual GaAs nanowires having cubic zinc-blende structure with ⟨001⟩ growth direction were also observed through HRTEM analysis. The results indicate possibility of integration of optical or electrical nanodevices utilizing GaAs nanowires with Si integrated circuits.

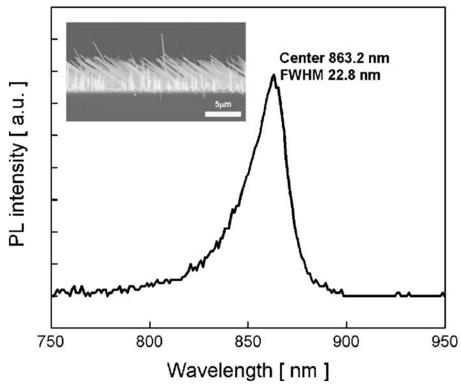

FIG. 3. RT-PL spectrum of the as-grown GaAs nanowires on a Si(001) substrate. The inset shows SEM of the as-grown GaAs nanowires on a Si(001) substrate.

The authors would like to express their gratitude to Sang-Geul Lee (KBSI at Daegu) for the XRD data and to Kyoung-Young Oh (PRFC at GIST) for FE-SEM analysis. This work was supported, in part, by the Ministry of Information and Communication, Korea, under the Information Technology Research Center support program supervised by the Institute of Information Technology Assessment (IITA-2005-C1090-0502-0029).

[1]K. Hiruma, T. Katsuyama, K. Ogawa, G. P. Morgan, M. Koguchi, and H. Kakibayashi, Appl. Phys. Lett. **59**, 431 (1991).
[2]B. J. Ohlsson, M. T. Björk, A. I. Persson, C. Thelander, L. R. Wallenberg, M. H. Magnusson, K. Deppert, and L. Samuelson, Physica E (Amsterdam) **13**, 1126 (2002).
[3]Z. H. Wu, X. Y. Mei, D. Kim, M. Blumin, and H. E. Ruda, Appl. Phys. Lett. **81**, 5177 (2002).
[4]R. S. Wagner and W. C. Ellis, Appl. Phys. Lett. **4**, 89 (1964).
[5]K. Hiruma, M. Yazawa, T. Katsuyama, K. Ogawa, K. Haraguchi, M. Koguchi, and H. Kakibayashi, J. Appl. Phys. **77**, 447 (1995).
[6]X. Duan, J. Wang, and C. M. Lieber, Appl. Phys. Lett. **76**, 1116 (2000).
[7]W. I. Park, J. S. Kim, M. H. Bae, H.-J. Lee, and G.-C. Yi, Appl. Phys. Lett. **85**, 5052 (2004).
[8]Y. S. Park, S.-H. Lee, J.-E. Oh, C.-M. Park, and T.-W. Kang, J. Cryst. Growth **282**, 313 (2005).
[9]R. Calarco, M. Marso, T. Richter, A. I. Aykanat, R. Meijers, A. v.d. Hart, T. Stoica, and H. Lüth, Nano Lett. **5**, 981 (2005).
[10]T. Mårtensson, C. P. T. Svensson, B.A. Wacaser, M. W. Larsson, W. Seifert, K. Deppert, A. Gustafsson, L. R. Wallenberg, and L. Samuelson, Nano Lett. **4**, 1987 (2004).
[11]V. G. Dubrovskii, I. P. Soshnikov, G. E. Cirlin, A. A. Tonkikh, Yu. B. Samsonenko, N. V. Sibirev, and V. M. Ustinov, Phys. Status Solidi B **241**, R30 (2004).
[12]M. Koguchi, H. Kakibayashi, M. Yazawa, K. Hiruma, and T. Katsuyama, Jpn. J. Appl. Phys., Part 1 **31**, 2061 (1992).
[13]W. Braun, V. M. Kaganer, A. Trampert, H. Schonherr, Q. Gong, R. Notzel, L. Daweritz, and K. H. Ploog, J. Cryst. Growth **227-228**, 51 (2001).
[14]Y. Cai, S. K. Chan, I. K. Sou, Y. F. Chan, D. S. Su, and N. Wang, Adv. Mater. (Weinheim, Ger.) **18**, 109 (2006).