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Synthesis and enhanced light-emission of Si nanocrystals embedded in silicon oxide nanowires

Cham-Sol Choi [a], Joon-Young Yoon [b], Jong-Hwan Yoon [a,*]

[a] Department of Physics, Kangwon National University, Chuncheon, Gangwon-do 200-701, Republic of Korea
[b] Department of Chemistry, California Institute of Technology, 1200E, California Blvd, Pasadena, CA 91125, USA

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ABSTRACT

In this paper, we report the synthesis of silicon oxide nanowire-embedded silicon (SONW-Si) nanocrystals (NCs) and their light emission characteristics. The SONW-Si NCs were formed by thermally annealing silicon-rich oxide nanowires (NWs) grown at low temperatures. The well-defined Si NCs were found to have grown in (111) direction and clearly displayed a higher density distribution at the center of the NWs. Photoluminescence (PL) measurements of the SONW-Si NCs showed an enhanced PL intensity compared to the silicon oxide film-embedded Si NCs, which may be related to the difference between the Si diffusion characteristics of SiO_x NWs and films.

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1. Introduction

Luminescent silicon materials have attracted great interest due to their potential for creating optoelectronic, biological, and sensing nanoscale devices [1–5]. With respect to this potential, rather than porous silicon [6], which emits strong visible/near infrared light at room temperature, but exhibits a certain structural fragility and instability in the emission intensity [7–9], Si nanocrystals (NCs) embedded in a silicon oxide matrix have received considerably more attention due to their stable luminescence and robust structure. Si NCs embedded in a silicon oxide matrix can be simply formed by thermally annealing silicon-rich oxide (SiO_x) films grown at low temperatures, which may be produced by various techniques, including sputtering [10], ion implantation [11], and chemical vapor deposition [12]. Many previous studies, however, have shown that silicon oxide films containing Si NCs do not emit light with sufficient intensity for practical applications. As a consequence, attempts to improve the light emission intensity are necessary in order that Si NCs are to be used in a variety of applications.

In this paper we present the synthesis and enhanced light emission of silicon oxide nanowire-embedded Si (SONW-Si) NCs. To synthesize the SONW-Si NCs, SiO_x NWs were grown by plasma-enhanced chemical vapor deposition (PECVD) at a low temperature ($\sim$320 °C), and then these SiO_x NWs were thermally annealed at 1100 °C for 1 h to form the SONW-Si NCs. The PL intensity was higher than that of the silicon oxide film-embedded Si (SOF-Si) NCs by a factor of at least 2. The enhanced PL intensity is suggested to be due to the enhanced formation of Si NCs in the NWs.

2. Experimental

SONW-Si NCs were formed by thermally annealing silicon-rich oxide (SiO_x) NWs. The SiO_x NWs were grown by annealing Si wafers coated with 300 nm of Zn as a catalyst at 320 °C for 10 min in silicon and oxygen chemical vapors produced by plasma enhanced chemical vapor deposition (PECVD) using mixtures of SiH_4 and N_2O gases. The SiO_x NWs with different compositions of Si and O were prepared by varying the gas flow ratio of N_2O and SiH_4, $[N_2O]/[SiH_4]$. The flow ratio was varied by changing only the N_2O flow rate at a fixed flow rate (200 sccm) of SiH_4. The procedural details of growing SiO_x NWs are described in our previous work [13]. To precipitate the Si NCs, the SiO_x NWs were annealed at 1100 °C for 1 h in a quartz-tube furnace using high-purity nitrogen as an ambient gas. For the formation of SOF-Si NCs, SiO_x films were simultaneously deposited with NWs using Si wafer substrates without a Zn film.

The microstructure characteristics of the SONW-Si NCs were analyzed by field-emission scanning electron microscopy (SEM), transmission electron microscopy (TEM), energy dispersive x-ray spectroscopy (EDX), and x-ray photoemission spectroscopy (XPS). The light emission properties were characterized by PL measurements. The PL was excited by the 325 nm line of a He–Cd laser and measured with either an electrically cooled charge-coupled device array camera (CCD) or a photomultiplier tube (PMT)

* Corresponding author. Tel.: +82 33 250 8466; fax: +82 33 257 9689.
E-mail address: jhyoon@kangwon.ac.kr (J.-H. Yoon).

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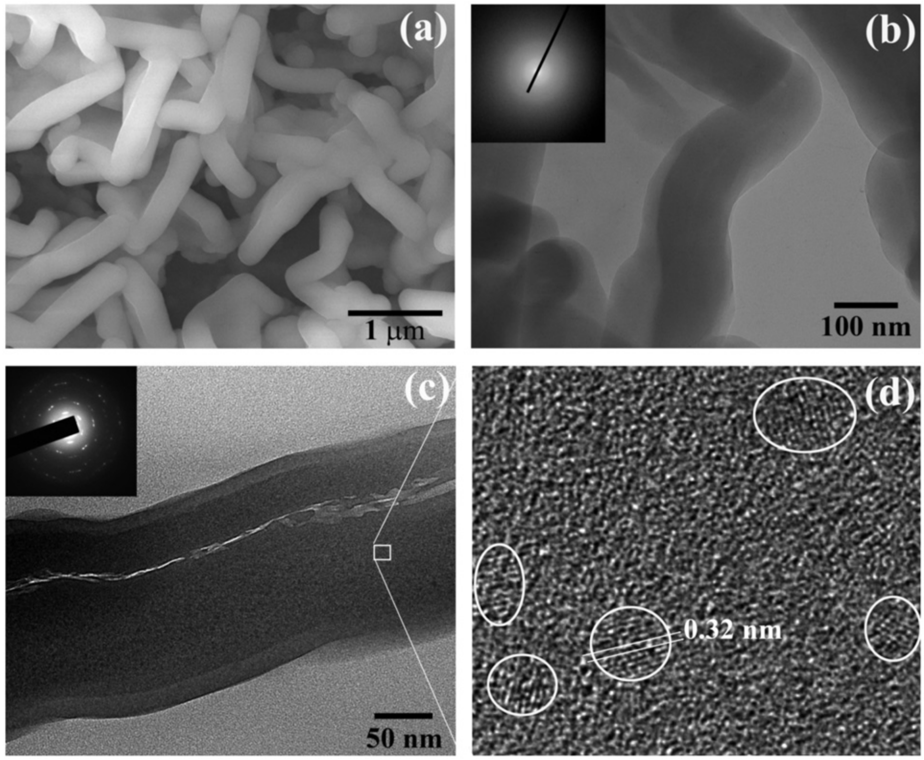

Fig. 1. Morphological images of silicon-rich oxide nanowires grown with a gas flow ratio of $[N_2O]/[SiH_4]=0.3$ before and after annealing: (a) SEM image of as-grown NWs, (b) TEM image and ED pattern (inset) of as-grown NWs, (c) TEM image and ED pattern of annealed NWs, and (d) high-magnification TEM image of the selected area (white square) in (c).

detector attached to a single-grating monochromator at room temperature.

3. Results and discussion

Fig. 1 presents the morphologies of SiO_x NWs grown using a gas flow ratio of $[N_2O]/[SiH_4]=0.3$ before [Fig. 1(a) and (b)] and after [Fig. 1(c) and (d)] annealing. As shown in Fig. 1(a) and (b), the as-grown NWs have a smooth morphology without any particles inside the NWs and exhibit highly diffusive ring electron diffraction (ED) patterns (inset of Fig. 1(b)), supporting the notion that the NWs have an amorphous structure. In contrast, as shown in Fig. 1(c) and (d), the annealed NWs clearly show the presence of a number of dark dots, unlike the images shown in Fig. 1(b). The dark dots are considered to be nanocrystallites from the ED pattern [inset of Fig. 1(c)], which reveals discontinuous rings composed of individual diffraction spots, consistent with the ED pattern from a low concentration of randomly oriented crystallites. In particular, the high-magnification TEM image shown in Fig. 1(d) clearly shows the presence of crystalline NCs (marked with white circles), which reveal a regular lattice structure confirming a single crystal phase. The spacing between two neighbor lattice planes is approximately 0.32 nm, which is in well agreement with the [111] lattice plane of a Si crystal. This conclusion can be further confirmed by the PL spectrum, which shows a peak near 815 nm and a full width at half maximum (FWHM) of 185 nm (not shown), consistent with the PL data usually observed for Si NCs [14,15].

Fig. 2 shows the integrated PL intensities obtained from the SiO_x NWs grown at different gas flow ratios of $[N_2O]/[SiH_4]$, along with those of the SiO_x films fabricated simultaneously with the NWs and annealed simultaneously at 1100 °C for 1 h. For both the NWs and the films, the PL intensity is strongly dependent on

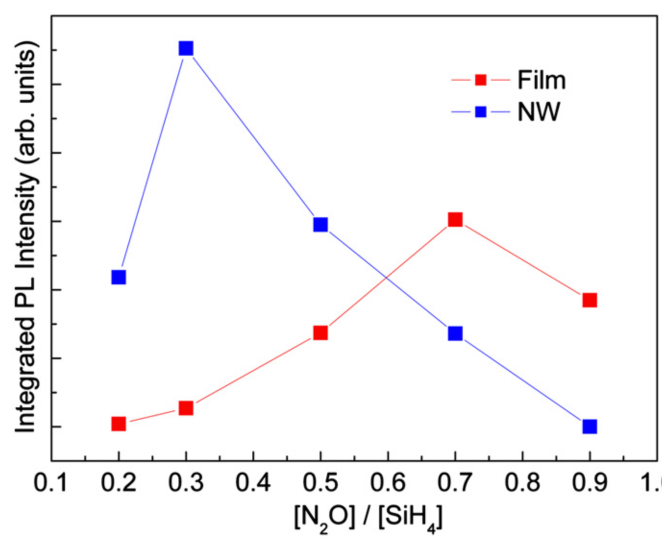

Fig. 2. Integrated PL intensities of silicon-rich oxide NWs and films grown at different gas flow ratios after annealing at 1100 °C for 1 h plotted as a function of flow ratio, $[N_2O]/[SiH_4]$.

the gas flow ratio, increasing and then decreasing as the ratio increases, with a maximum PL intensity observed at the flow ratios of 0.3 and 0.7 for the NWs and films, respectively. The data show that the maximum PL intensity of the NWs is higher over that of the films by a factor of 2. On the other hand, the TEM analysis on the NW and film prepared with the flow ratio of 0.3 and 0.7, respectively, showed that the areal density of the NCs in the NW (1.7×10^{12} cm^{-2}) was larger than the density (7.5×10^{11} cm^{-2}) in the film by a factor of 2, demonstrating that the different maximum PL intensities observed stem from

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