



Growth of vertically aligned ZnO nanowire arrays on ZnO single crystals

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ABSTRACT

Vertically aligned ZnO nanowire arrays were grown on ZnO single crystals using a chemical vapor deposition method. Nucleation sites were defined by patterning Au nanodot catalysts fabricated through anodized aluminum oxide (AAO) templates. Morphology and structure characterizations showed that the as-grown nanowires had the single-crystal hexagonal wurtzite structure with a $\langle 0001 \rangle$ growth direction. By changing the pore distance of AAO, the density of nanowires could be well controlled. A sharp peak in the UV region and a weak broad peak in the visible region were shown in the room temperature photoluminescence spectra, suggesting the high-quality ZnO nanowire arrays were obtained.

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1. Introduction

Vertically aligned ZnO nanowires grown on large-area substrates have been studied extensively due to their practical applications such as solar cells [1], nanowire lasers [2], and field emitters [3]. Among the growth methods of ZnO nanowires, chemical vapor deposition (CVD) using Au as catalyst is most widely used [2]. The uniformity, density and ordering can be controlled if a catalyst pattern created by a mask is applied [4,5]. One promising approach is to fabricate patterned Au catalyst arrays via anodic aluminum oxide (AAO) templates. Nevertheless, the growth of ZnO nanowires via patterned Au arrays fabricated through AAO template is rarely investigated. Chik et al. reported Au catalysts formed in AAO to grow periodic array of ZnO nanowires [5]. However, these ZnO nanowires were grown on heterogeneous substrates such as GaN. Low crystallinity region exists at the nanowire–substrate interface due to lattice mismatch. This layer has a bad effect on performance of electronic device such as field emitters because electrons have to pass through the interface. Therefore, ZnO nanowires grown on ZnO single crystals are expected to have good performance due to the absence of the interfacial defects and grain boundaries. However, the study of

ZnO nanowires on ZnO single crystals via patterned Au catalyst fabricated by AAO template has not been reported.

In this paper, we demonstrate the growth of vertically aligned ZnO nanowire arrays on ZnO single crystals by a conventional CVD method. Catalyst Au nanodot arrays are produced via AAO-patterning technique followed by thermal annealing. The density of the nanowires is controlled by the interpore distance of AAO templates.

2. Experimental

Ga-doped ZnO single-crystals were provided by Prof. Huang's group [6]. AAO templates were prepared in oxalic and sulfuric acid solutions under anodization voltages of 50 V and 25 V, respectively, which was reported in detail elsewhere [7]. After the transfer of AAO onto +C face of the substrate, 5 nm Au film was deposited over the masks using electron-beam evaporation at a rate of 0.1 nm s^{-1} . Then, AAO was removed and the samples were annealed at 825°C for 2 h in Ar. ZnO nanowires were grown by the CVD method in a horizontal tube furnace. The source (1:1 mass ratio of ZnO/C powders) and substrates were held at 910°C and 825°C , respectively, with a flow of 1 sccm O_2 and 200 sccm Ar under a pressure of 4000 Pa for 10–20 min.

The morphology and structure of samples was studied by SEM Hitachi S4800 and FEI Tecnai G2 F30 (300 kV) TEM. Room temperature photoluminescence (PL) spectrum was acquired with a He–Cd laser (325 nm) as an excitation source. The pump beam was

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focused on nanowires at an incidence angle 10° to the symmetric axis of the nanowire.

3. Results and discussion

Fig. 1a shows the schematic of the fabrication process of catalyst Au nanoparticle and ZnO nanowire arrays. The first three steps of the process are similar to previous report [5]. But in this work, low Au deposition thickness (5 nm) and a thermal annealing step (825°C for 2 h in Ar) were taken in order to turn each Au nanopad into only one Au nanodot. Catalytic Au arrays shown in Fig. 1b were achieved through an AAO mask prepared in oxalic solution under anodization of 50 V. The in-plane diameter and interparticle distance are ~ 27 nm and 125 nm. Before annealing, Au nanopads had a larger in-plane size. But each pad became one nanodot after annealing due to coalescence and Ostwald ripening [8]. We found that if this step was missed, Au pads would split into several nanodots during the fast heating before nanowire growth, and then each tiny particle would guide the growth of one thin ZnO nanowire [9].

Fig. 1c presents the top view SEM image recorded from the ZnO nanowire arrays obtained by using the Au nanodot array in Fig. 1b. Each white spot represents one ZnO nanowire. The arrangement of

the Au pattern was preserved during the growth process. Hexagon end planes of the nanowires can also be clearly identified, providing that these nanowires are of hexagonal wurtzite crystal structure, grow along the $\langle 0001 \rangle$ direction. A 30° side view of the arrays (Fig. 1d) shows clearly the well-aligned growth of the ZnO nanowires. They are oriented perpendicular to the substrate surface as a result of homogeneous epitaxial growth of ZnO. All of the ZnO nanowires have about the same height, of about $1.5\ \mu\text{m}$ and their diameters range between 35 and 50 nm. The height of nanowires could be controlled by changing the growth time. It is noteworthy that there was a pyramidal base at the root of the each wire. This is a typical phenomenon in the vapor phase epitaxial growth of nanowires [10]. The base forms in an early stage and serves as nucleation sites for subsequent growth of nanowires.

By changing the electrochemical anodization parameters, AAO templates with different interpore distances can be obtained. Here, another AAO template was fabricated in sulfuric acid solution under anodization voltage of 25 V. The interpore distance is ~ 65 nm. By using this mask for Au deposition, nanowires with higher density can be fabricated (Fig. 1f and g).

Catalyst Au nanodots can be clearly observed in Fig. 1e. Au dots at the edge of the base and on the nanowire sidewalls are indicated by dotted circles and a solid white arrow, respectively. In fact, there are also some Au nanodots located at the tips of nanowires, which

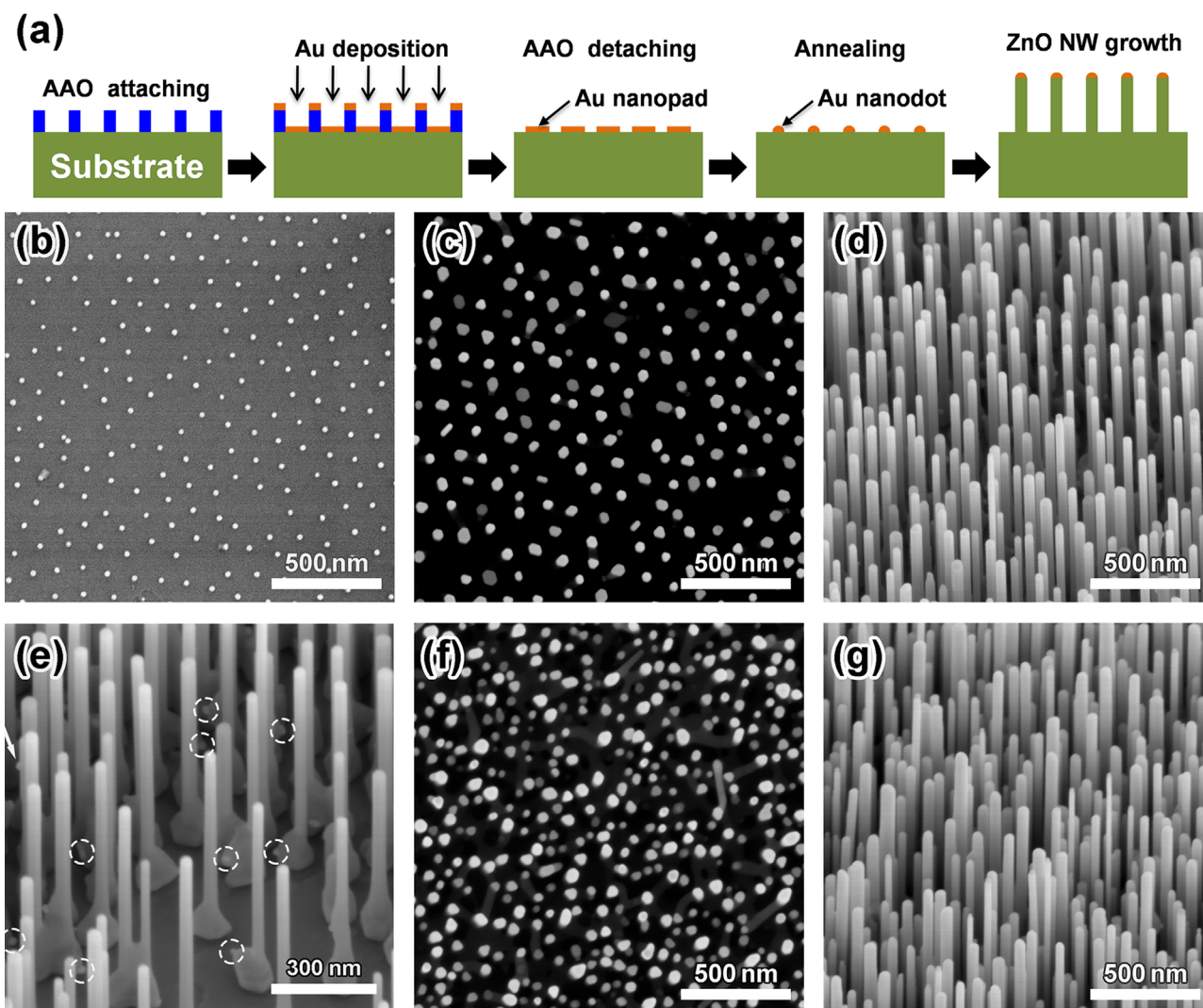


Fig. 1. (a) Schematic of the fabrication process of Au nanodot and ZnO nanowire arrays. (b) SEM image of Au array obtained using AAO as a shadow mask followed by thermal annealing. (c, d) Top and a 30° view of aligned ZnO nanowires. (e) A high magnification SEM image. (f, g) Top and a 30° view of aligned ZnO nanowires with a higher distribution density.

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