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Patterned horizontal growth of ZnO nanowires on SiO₂ surface

Jinwoong Kim, Huiseong Jeong, Ji-Yong Park*

Department of Physics and Division of Energy Systems Research, Ajou University, Suwon, Ggyeonggido 443-749, Republic of Korea

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ABSTRACT

We report patterned horizontal growth of ZnO nanowires on SiO₂ surface for the study of electrical and luminescent characteristics of individual nanowires and for device applications. Patterns of gold catalytic seed islands with barrier layers which suppress vertical growth were employed to facilitate horizontal growth on SiO₂ surface. After the growth, ZnO nanowire devices are fabricated by patterning electrodes aligned over the seed islands and their device characteristics are investigated. We could also investigate history of synthesis conditions by obtaining local luminescence characteristics along individual nanowires.

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1. Introduction

ZnO nanowires (NW) have garnered active researches in the last decade for their unique and exceptional material properties such as large bandgap (\sim 3.6 eV in the bulk), large exciton binding energy (\sim 60 meV) at room temperature, and large piezoelectricity [1]. Proposed and demonstrated applications of ZnO NWs show such diverse devices as field effect transistors (FETs) [2], energy harvesting devices [3], light emitting diodes, lasers [4], photodetectors [5], photovoltaic devices [6], chemical or biological sensors [7]. For the synthesis of ZnO NWs, various methods such as low temperature hydrothermal method [8], metalorganic chemical vapor deposition [9], and high temperature vapor transport [10] with or without catalyst have been reported. After the growth, usually powders of ZnO NWs or vertically arrays of ZnO NWs are obtained by these methods. Most planar ZnO NW devices such as FETs or photodetectors so far have been obtained from randomly dispersed or transferred ZnO NWs on target substrates after the separate growth processes. However, for device integrations and viable applications, deterministic fabrication method is required for planar devices. There have been some efforts for patterned horizontal growth of ZnO NWs using various growth methods to this end [11–18]. But the reports of horizontal growth, especially using high temperature vapor transport method are still limited [13] and properties of such ZnO NWs are not well studied.

In this contribution, we report patterned horizontal growth of ZnO NWs on SiO_2 surface by suppressing vertical growth with a barrier layer for device application in mind. By patterning metal electrodes aligning to the catalyst islands, we could fabricate ZnO NWFETs at the pre-determined positions. Horizontal growth also allows full access to the whole length of individual ZnO NWs. We could study local luminescent properties of individual ZnO NWs from the root to the top, which provides glimpse of history of growth condition.

2. Material and methods

We grow ZnO NWs by thermal vapor phase transport method with carbothermal reduction process in a dual tube low pressure chemical vapor deposition (LPCVD) system as previously reported [19]. We use a mixture of pure ZnO and graphite powder (0.5 g each) as a source material and employ Au as catalytic seed layer for the ZnO growth. Seed patterns are prepared on Si substrate (with 200 nm-thick SiO₂) by photolithography and e-beam evaporation of 1 nm-thick Cr (adhesion layer), Au (seed layer), and Ti or Mo (barrier layer) in order followed by lift-off process. Thickness of Au seed layer ranges from 30 nm to 100 nm and that of barrier layer was from 100 nm to 150 nm. The substrate with seed patterns is loaded into the center of a furnace with the source. The furnace is first pumped down to 10 mTorr and the source is heated to 600 °C. Then, the pressure inside the furnace is maintained at 10 Torr while the source is heated to the reaction temperature (900–970 °C) under high purity nitrogen (carrier gas, 150 sccm) and oxygen gas flows (80 sccm) and kept at the reaction temperature for 40 min.

^{*} Corresponding author. Tel.: +82 31 219 2573; fax: +82 31 219 1615. *E-mail address*: jiyong@ajou.ac.kr (J.-Y. Park).

For device fabrications, patterns of electrodes with gap sizes of $2-6~\mu m$ are defined by photolithography aligned such that they cover seed patterns, and metal electrodes consisting of Ti (100 nm)/Au (50 nm) are prepared by e-beam evaporation and lift-off processes.

The sizes and shapes of the samples are examined after the growth and fabrication with a field emission scanning electron microscope (SEM). Local luminescence properties of ZnO NWs with high spatial resolution are obtained by cathodoluminescence (CL) measurements at room temperature in an SEM as in our previous report [20]. All the CL measurements in this work were carried out with the acceleration voltage of 10 kV. *IV* characteristics of ZnO NWFETs were acquired in a probe station with a semiconductor parameter analyzer in the ambient environment.

3. Results and discussions

Fig. 1(a) shows schematics of our proposed process for horizontal patterned growth of ZnO NWs on SiO₂ surface with a barrier layer. From previous experiments, we found that long vertical arrays of ZnO NWs using thin Au seed layer on SiO₂ surface could be grown with vapor phase transport method [19]. In order to find materials that can suppress vertical growth of ZnO NWs and are compatible with high temperature growth condition, we tested various metal layers for catalytic properties for the growth. After confirming that metal layers such as Ti, Cr, Mo do not act as seed for ZnO NW growth, we employed these metal layers as a barrier layer, which is schematically shown in Fig. 1(a). After depositing Au, Ti or Mo was subsequently deposited on top to form a barrier layer. Both Ti and Mo worked for our purpose. Since the barrier layer inhibits growth of ZnO NWs, NWs mostly grow from the sidewall where Au is exposed as in Fig. 1(b). By employing the barrier layer, we could effectively suppress vertical growth of ZnO NWs and obtain horizontal NWs which can be as long as 100 µm or more as can be seen in Figs. 3 and 4. The growth mode in our setup was identified as vapor—solid (VS) mode in which the formation of ZnO film on Au seed layer precedes subsequent NW growth [19,21]. In many cases, horizontal ZnO NWs in this case were found to originate from ZnO aggregations on the sidewall of the Au layer as shown in the inset of Fig. 1(b).

Since the ZnO NWs originate from patterned seed islands, we can incorporate ZnO NWs into devices by patterning metal electrodes over the Au seed islands. We fabricated Ti/Au (thickness 100 nm and 50 nm, respectively) electrodes so that the seed islands

are completely covered by new electrodes. Then, by patterning counter electrode with the specific gap distance we could fabricate ZnO NW devices in FET geometry as shown in Fig. 2. Although we could control the position of NWs by seed layer patterns and suppress vertical growth of NWs, it is difficult to control direction of horizontal growth. Therefore, we used large counter electrode to increase probability of contacting horizontal NWs. IV characteristics of the devices are also measured and results for corresponding ZnO NW devices are shown in Fig. 2(c) and (d). All the measured devices show typical n-type semiconducting behavior expected for ZnO NWs as shown in Fig. 2(c) and (d) in the dark state. The field effect mobilities extracted from these measurements are mostly in the range of $1-10 \text{ cm}^2/\text{V}$ s, which is typical for ZnO NWs measured in the ambient condition and also similar to other ZnO NW devices fabricated in our group using vertically grown NWs. These results indicate that horizontally grown NWs possess similar electrical characteristics as vertically grown ones.

Our growth strategy also allows us to investigate the local luminescent properties of individual NWs along their full length. Luminescent properties of ZnO NWs are of great importance and interests since they are often employed as luminescent material in optoelectronic devices [4]. Luminescent characteristics also indicate the quality of ZnO NWs. Stronger emission at ~380 nm, due to excitonic recombinations near band edge, relative to one at visible ranges such as at \sim 510 nm which is often associated with oxygen deficiencies [22] implies ZnO NWs with little defects and good crystallinity [23]. Although luminescent characteristics are usually obtained from ensemble of ZnO NWs especially for the case of photoluminescence. CL is well suited for local investigation with its high spatial resolution. Recently, we showed that there can be inhomogeneities in the luminescent characteristics among ZnO NWs even though they are grown in the same batch [20]. In order to measure CL from individual ZnO NWs, we usually deposit NWs horizontally on a substrate from dispersion solution or by mechanical rubbing. However, during transfer, vertically grown ZnO NWs break in the middle so that information from the full length of the individual ZnO NWs is difficult to obtain. Also the direction of the growth is usually difficult to tell after NWs are transferred to another substrate. However, we have access to the whole length of the ZnO NWs from the root for the horizontal ZnO NWs grown from Au seed patterns in our study. Therefore, we can obtain luminescence characteristics of individual ZnO NWs along their full length from the root to the top with CL. We present two

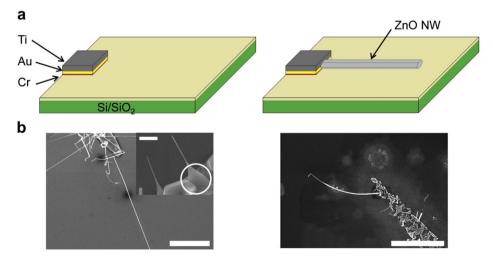


Fig. 1. (a) Schematics of horizontal growth of a ZnO NW. A seed island pattern consists of Cr (1 nm-thick, adhesive layer), Au (100 nm-thick, seed layer), and Mo or Ti (100 nm-thick, barrier layer). ZnO NWs grow from the sidewall of Au layer in the schematic. (b) SEM images of horizontally grown ZnO NWs from seed island patterns. Scale bars are for 10 μm (left) and 20 μm (right). Inset shows ZnO NWs near the root on the sidewall of a Au seed layer, where ZnO is aggregated as marked by a circle. Scale bar in the inset is for 500 nm.

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