

Correlation between photoluminescence and varied growth pressure of well-aligned ZnO nanorods on fused silica substrate

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Abstract

Well-aligned ZnO nanorods with variable diameters were fabricated on fused silica substrates by the simple physical vapor deposition method with and without pre-deposited NiO particles. The diameter control of ZnO nanorods on the fused silica substrates was achieved by varying the growth pressure. The room-temperature photoluminescence spectra of the ZnO nanorods exhibit strong exciton emission at 3.25 eV that reveals good crystal quality. The exciton emission peak shifts toward 3.22 eV for the samples made at the larger growth pressure that is attributed to the influence of the electron-acceptor-level transition. The ZnO nanorods were examined by high resolution transmission electron microscopy and X-ray diffraction to show single crystal quality and preferentially *c*-axis alignment. Furthermore, we used scanning electron microscope to verify not only the size but also the growth mechanisms of ZnO nanorods. The results show a ZnO buffer layer formed between the flat top-facet nanorods and the substrate. In comparison with the samples without pre-deposited NiO particles on the substrate, the pre-deposited NiO particles act as nucleation centers for Zn vapor during the deposition process that improves the quality of the buffer layer.

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1. Introduction

ZnO has some of the greatest potential among semiconductor materials for applications in ultraviolet regions and nanotechnology. It has a wide direct bandgap of about 3.37 eV at room temperature, and large exciton binding energy of about 60 meV. These properties allow it to emit ultraviolet light with high efficiency at room temperature, and give it superiority in many applications. For instance, ZnO thin film structures can be utilized as piezoelectrical device and ultraviolet light emitting diode [1]. Recently, large second order susceptibility (up to 56 pm/V), which

is a crucial technological parameter, had been measured in ZnO films deposited on MgO substrates [2]. The nano-confined effect and interface separating substrate and the nanoparticles play the major role on the large optical non-linearity in these ZnO films. For low-dimensional ZnO nanostructures, ZnO nanoparticles or quantum dots have attracted much research attention in fundamental studies and technical applications because of their unusual characteristics [3]. Besides, ZnO had been made into solar cells using nanorods [4] and nanoparticles [5], nano logic circuit [6], nano laser source [7], and field emission device [8] have been studied using one-dimensional (1D) ZnO nanostructures, which can be fabricated on various substrates such as Si [9–12], sapphire [13,14], Al₂O₃ [15], silica [16], etc.

It is very important for fabricating nano-phonic devices in 1D nanostructures with well control of diameter.

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Yang et al. [9] reported that the diameters of the ZnO nanorods should depend on the size of the catalyst, gold nanoparticles, in a vapor–liquid–solid growth mechanism. Lee et al. [15] reported that the diameter can be controlled by the growth temperatures in a limit range. By varying the growth ambient pressure, diameter-controlled Si nanowires of 6–25 nm were reported [17]. For the potential of photonic application and of merging with the Si-based technologies, in this paper, we fabricated well-aligned ZnO nanorods by using the simple physical vapor-deposition method [15] on silica substrates and showed that the mean diameter of ZnO nanorods can be varied by the ambient pressure inside the furnace tube. The room-temperature photoluminescence (PL) properties of the samples were investigated to realize the growth pressure dependence of the ZnO nanorod quality. Furthermore, the growth mechanism is also discussed.

2. Experimental

The ZnO nanorods were fabricated by the following procedure: The 0.01 M $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ dissolved in ethanol was sprayed on a clean fused silica substrate. After baking for 5 min in air on a hotplate at 60 °C, the substrate was loaded 2 cm above an alumina boat that contains 1 g metal zinc balls (99.999% with 1–3 mm in diameter). Then the boat was placed in the center of a quartz tube furnace. After evacuating the tube by a mechanical pump, argon gas was introduced into the tube with a flow rate of 50 sccm. Then the tube was heated at the rate of 20 °C/min to the reaction temperature of 600 °C, and maintained for 30 min at the ambient pressures of 50 Torr. Finally, the furnace was cooled to room temperature at cooling rate of about 100 °C per hour, and the gray wax-like product was found on the substrate surface.

A 20-mW He–Cd laser (Omnichrome 74 Series), radiated at 325 nm, was used as the excitation source for the PL measurement at room temperature. The emitted light was collected by a Triax 320 spectrometer equipped with a UV-sensitive photomultiplier tube. The morphology and crystal structure of the product were characterized by field emission scanning electron microscopy (FESEM), high resolution transmission electron microscopy (HRTEM), and X-ray diffraction (XRD).

3. Results and discussion

3.1. Photoluminescence

The power of about 20-mW from the He–Cd laser (Omnichrome 74 Series) was used to excite the samples in the PL measurement. Fig. 1a shows a room-temperature PL spectrum of the ZnO nanorods that were grown at the ambient pressures of 50, 100, 110, and 150 Torr, respectively. As the ambient pressure increases over 100 Torr, the strong near bandedge (NBE) peak, which is attributed to the exciton emission, starts to red-shift from 3.25 eV to

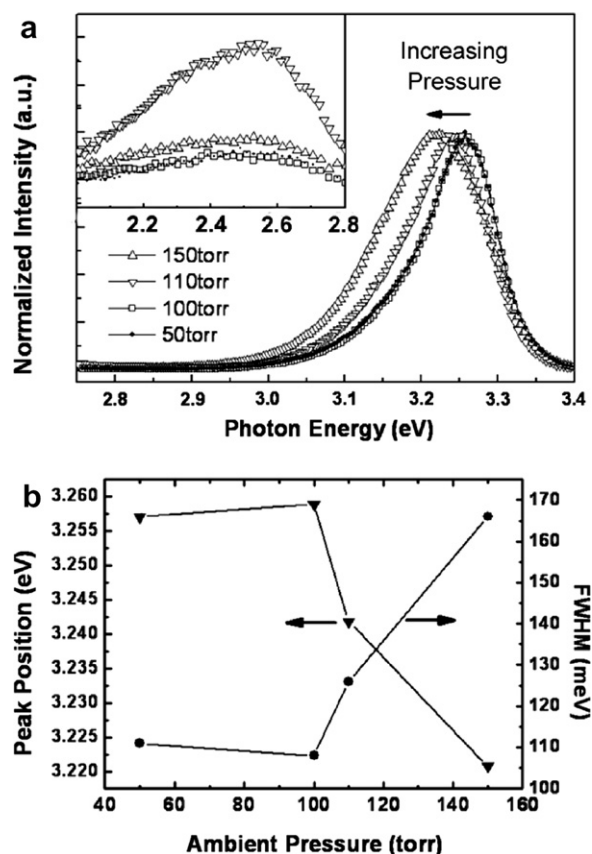


Fig. 1. (a) The room-temperature PL spectrum of the ZnO nanorods grown at the ambient pressure of 50, 100, 110 and 150 Torr, and the weak broadband emission at about 2.54 eV inside the inset picture. (b) The FWHM and the peak position of the strong emission depending on the ambient pressures.

3.22 eV with its FWHM being broadened from 111 meV to 164 meV. The inset of Fig. 1a shows the relatively weak broadband emission at about 2.54 eV with large FWHM of ~770 meV. The FWHM and the peak position of the NBE emission versus the growth pressure are shown in Fig. 1b. The FWHM increases obviously with increasing ambient pressure when the pressure exceeds 100 Torr; meanwhile, the peak position starts to red-shift.

From the PL spectra, the samples formed at less than 100 Torr ambient pressure have a strong NBE emission, with peak position at about 3.25 eV. The red-shifting behavior of the NBE emission at room temperature for the ambient pressure beyond 100 Torr is attributed to the defect influence. The defects in the ZnO produce the electron-acceptor-level transition emission (eA^0) and lead the NBE peak position to vary from 3.22 eV to 3.25 eV. This was attributed to the overlap of the 3.31 eV (due to free exciton emission) and 3.23 eV (due to eA^0) [14,18]. The acceptor could be the Zn vacancy because, in the experiment procedure, there was no any acceptor source except for the Zn vacancy. The red-shifting behavior implies that the acceptor density increases with the growth pressure. The acceptor could be attributed to the turbulent flow

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