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Structural and optical evolution of Ga_2O_3/glass thin films deposited by radio frequency magnetron sputtering

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ABSTRACT

We investigated the structural and optical evolution of Ga_2O_3 thin films on glass substrates deposited using radio frequency magnetron sputtering. Initially, amorphous Ga_2O_3 thin film is grown, and then, surface humps and nanowire (NW) bundles are gradually formed as the film thickness increases. The surface humps are Ga-rich and provide nucleation sites for NWs through a self-catalytic vapor–liquid–solid mechanism with self-assembled Ga droplets. Both the surface humps and the NWs induce variation of the optical properties such as the optical bandgap and refractive index by absorbing light in the ultraviolet region.

1. Introduction

Gallium oxide (Ga_2O_3) has attracted considerable attention due to its potential for applications such as field effect transistors and gas sensors [1,2]. With a wide bandgap of ~4.9 eV at room temperature, Ga_2O_3 thin films have also been widely investigated for use as transparent conducting oxides in ultraviolet (UV) optoelectronic devices [3–5]. The optical properties of Ga_2O_3 thin films such as photoluminescence, transparence, and optical bandgap are strongly coupled with the microstructure as well as the chemical inhomogeneity. It was postulated that either excess gallium or an oxygen deficiency in Ga_2O_3 thin films could change the bandgap because they both make the shallow levels near the band-edge [6–8]. Many studies have shown, in particular, that the bandgap of Ga_2O_3 thin films can be tuned by doping Ga_2O_3 with other elements [9–11] or by annealing Ga_2O_3 under different gas environments at high temperatures [12,13]. Annealing facilitates the production of unintentional n-type semiconductors with hydrogen or oxygen vacancies [7,8,14]. Metal dopants such as Cu, Ti, and W result in a variation of the bandgap in the range of 4.2–5.23 eV [9–13]. It is also well known that the anisotropy of the crystalline orientation in Ga_2O_3 is one of the crucial factors in determining the bandgap of Ga_2O_3 thin films [15].

We very recently reported on the self-catalytic growth of monoclinic β-Ga_2O_3 nanowires (NWs) by radio frequency (RF) magnetron sputtering, in which the growth mode was gradually transformed from initial epitaxial films to NW bundles [16,17]. Here, we report how a change in growth mode affected the optical properties with increasing film thicknesses. The UV–visible (UV–vis) spectroscopy results revealed the gradual decrease of both the optical bandgap and the refractive index with increasing film thickness. This might be attributed to the increased amount of self-assembled Ga droplets and the non-stoichiometric β-Ga_2O_3 NWs, which absorbs a significant amount of light in the UV region.

2. Experimental

The Ga_2O_3 thin films were deposited by RF powder sputtering on glass substrates (Corning, EAGLE 2000). Prior to deposition, the substrates were cleaned with a standard chemical process in an ultrasonic bath. A sputtering power of 100 W was applied for a 2 in. diameter powder target. High purity Ar gas (99.999%) with a flow rate of 20 sccm was introduced into the sputtering chamber using a mass flow controller. The working pressure was maintained at 5×10^{-3} Torr, and the growth temperature was set to 550 °C. In order to obtain a series of samples with different thicknesses, the growth time was controlled from 1 to 60 min resulting in a change in the film thickness from 15 nm to 1.35 μm, respectively.

The structure of the as-grown samples was examined using synchrotron x-ray diffraction (XRD) at the 5D beamline of the Pohang Light Source in Korea. An x-ray energy of 10 keV was selected by a double-bound Si (111) monochromator. The standard XRD profiles (θ–2θ scan) were measured. The evolution of the surface morphologies was examined using scanning electron microscopy (SEM) and atomic force microscopy (AFM). The optical

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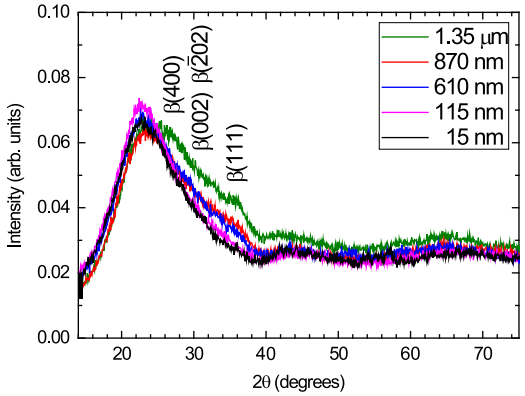

Fig. 1. XRD patterns of Ga_2O_3 thin films with different thicknesses. The broad profile near $2\theta=23^\circ$ originates from the glass substrate. The peak denoted as β(111) appears and becomes dominant with increasing film thickness, while the broad profile represents the short-range-ordered polycrystalline β-Ga_2O_3 thin films.

transmittance curves were measured at room temperature using UV–vis spectroscopy.

3. Results and discussion

Fig. 1 shows the XRD profiles of the Ga_2O_3 thin films. For a 15-nm-thick sample, no noticeable peaks were observed with the exception of the broad profiles originating from the glass substrates. This indicates that the Ga_2O_3 thin film is mostly amorphous. On the other hand, a broad peak at $2\theta=35.9^\circ$ appeared and became dominant as the film thickness increased. This peak corresponds to the (111) plane of β-Ga_2O_3. The XRD profile for the 1.35-μm-thick film clearly shows a distinct profile with broad tails that might be assigned to the Bragg reflections of short-range-ordered polycrystalline β-Ga_2O_3. Indeed, the XRD profile of NW sample shows clearly the Bragg peaks of β-Ga_2O_3, as presented in Supplementary materials. The XRD results suggest that

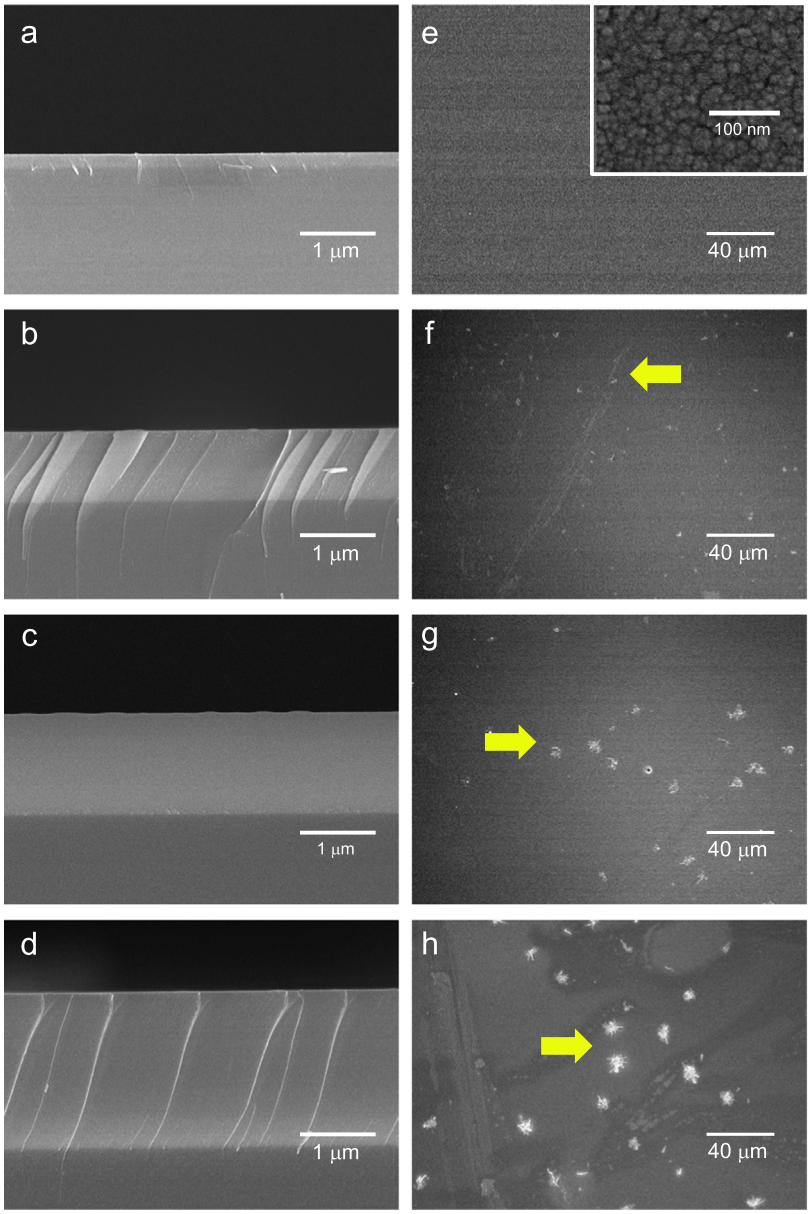

Fig. 2. Top-view and cross-sectional SEM images of Ga_2O_3 thin films. (a, e) 115-nm-, (b, f) 610-nm-, (c, g) 870-nm-, and (d, h) 1.35-μm-thick samples. The inset of Fig. 2(e) represents the high-magnification SEM image to highlight the surface morphology of 115-nm-thick sample. The arrows indicate the evolution of the surface humps and NWs.