Effect of thickness on the structural and optical properties of the niobium-doped β-Ga_2O_3 films

Hao Zhang, Jinxiang Deng ✉, *Le Kong, Zhiwei Pan, Zhiying Bai, Jiyou Wang*

College of Applied Sciences, Beijing University of Technology, Beijing 100124, People's Republic of China
✉ *E-mail: jdeng@bjut.edu.cn*

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To investigate the effect of thickness on the structural and optical properties of niobium-doped beta gallium oxide (β-Ga_2O_3:Nb) thin films, a series of β-Ga_2O_3:Nb thin films with different thicknesses were prepared by radio-frequency magnetron method. The crystalline quality is highly improved when the film thickness exceeds 145 nm. The surface exhibits different morphologies under different film thicknesses. The average transmittance of all the films are over 80% in visible range, and that the ultraviolet absorption edge shifts to longer wavelength indicates the bandgap shrinks with increasing the film thickness. Moreover, the photoluminescence spectrum measurements indicate that fewer defects were formed when the film thickness is around 145 nm.

1. Introduction: With bandgap (E_g) of about 4.9 eV, monoclinic beta gallium oxide (β-Ga_2O_3) has attracted more and more attention as key material for ultraviolet (UV) optoelectronic devices [1–6]. Moreover, it is considered to be a promising candidate for power devices in the near future due to its excellent thermal and chemical properties and suitability for mass production. Till now, many works have been done to investigate the properties of the intrinsic β-Ga_2O_3. Moreover, various preparation technologies have been improved to fabricate high-quality β-Ga_2O_3 films by controlling the growth temperature and pressure [7–9]. Considering the poor conductive properties of the pure β-Ga_2O_3 thin film, many research groups begin to improve the properties of β-Ga_2O_3 thin films by doping technology [10–14]. Recent studies indicate that the memory capacitor with lightly niobium (Nb)-doped Ga_2O_3 shows better charge-trapping characteristics [15]. Moreover, Nb-doped Ga_2O_3 has immensely applied prospects in high-performance memory applications. So it is necessary to explore the properties of β-Ga_2O_3:Nb thin film in detail. We have investigated the influence of Nb doping concentration and annealing atmosphere on the properties of β-Ga_2O_3:Nb thin film in our previous work [16, 17]. The film thickness is also an important factor that affects the properties of various kinds of thin films [18, 19]. Metal/semiconductor/metal structured UV photodetectors based on various thickness β-Ga_2O_3 thin films have been investigated by An [20]. The photoconductive gain of β-Ga_2O_3 photodetectors increases, then decreases with increasing thickness of the β-Ga_2O_3 films. Moreover, the photoconductive gain of the photodetector is also the highest when the thickness of β-Ga_2O_3 film about 200 nm. For a thin film in nanosize, the quantum size effect will play an important role in the physical properties of thin films, and as the result, the behaviour of the device would be tuned as well. Therefore, it is of great significance to investigate the properties of β-Ga_2O_3:Nb thin films with different thicknesses. Moreover, this work has been guiding significance for the application of β-Ga_2O_3:Nb thin films in devices.

In this Letter, a series of β-Ga_2O_3:Nb thin films with different thicknesses were prepared by radio-frequency (RF) magnetron sputtering. The effect of film thickness on the microstructure, morphology and optical properties of β-Ga_2O_3:Nb thin films was investigated in detail.

2. Experiment: The β-Ga_2O_3:Nb thin films were deposited on the silicon (Si) and quartz glass substrates by RF magnetron sputtering method. A Ga_2O_3 disc embedded by an Nb_2O_5 tabletting was used as the target. The base pressure in the sputtering chamber was 1×10^{-4} Pa; then, high-purity argon gas (99.999%) was introduced into the chamber through a mass flow controller and the films were deposited at a working pressure of 0.6 Pa. The RF power applied to the target was set at ~80 W. To obtain a series of β-Ga_2O_3:Nb thin films with different thicknesses, the growth time was controlled from 20 to 60 min, resulting in a change in the film thickness from 95 to 280 nm, respectively. Subsequently, the samples were subjected to annealing at 1000°C for 1 h in flowing N_2 atmosphere.

The elemental composition of the film was measured by energy-dispersive X-ray spectroscopy (EDS). The crystalline structure of the films was examined by X-ray diffraction (XRD) using Purkinje D3 diffractometer. UV–visible absorption spectrums were carried out using a Shimadzu UV-3600 spectrophotometer. The surface morphology was characterised using a DI MULTIMODE atomic force microscopy (AFM). The photoluminescence (PL) spectra were measured at room temperature with an Edinburgh FLSP920 spectrophotometer. The excitation light was the monochromatic light from a xenon short arc lamp with a wavelength of $\lambda = 341$ nm.

3. Results and discussion: The elemental composition and concentration of the β-Ga_2O_3:Nb thin film with a thickness of 190 nm was measured by EDS. As shown in Table 1, elements of oxygen, gallium and Nb are observed. It indicates that the Ga_2O_3:Nb thin films were successfully prepared by RF magnetron sputtering. Moreover, the concentration of Nb element is 0.69 wt% in the β-Ga_2O_3:Nb thin film.

To investigate the influence of film thickness on the crystal structure and crystallinity of β-Ga_2O_3:Nb thin films, the XRD analysis on the β-Ga_2O_3:Nb thin films with different thicknesses was performed. As shown in Fig. 1, the diffraction peaks of (201), (110) and (−111), which are all indexed to the β-Ga_2O_3, are observed in all the β-Ga_2O_3:Nb thin films with different thicknesses. It is worth noting that the diffraction peak intensity of β-Ga_2O_3:Nb thin film with a thickness of 145 nm is stronger than that of 95 nm, but the change of the diffraction peak intensities is no longer evident when the film thickness exceeds 145 nm. Fig. 2 shows the full width at half maximum (FWHM) of XRD (−111) peak and grain sizes of the samples as a function of film thickness. As shown in Fig. 2, the grain sizes of the films show a trend of increasing first and then decreasing with increasing film thickness, and the FWHM reaches the minimum and the grain size reaches the maximum for β-Ga_2O_3:Nb thin film with a thickness of 145 nm in

Table 1 Elemental composition and concentration of the film with a thickness of 190 nm

Element	wt%	wt% Sigma
O	20.79	1.33
Ga	78.51	2.73
Nb	0.69	3.12
total	100.00	—

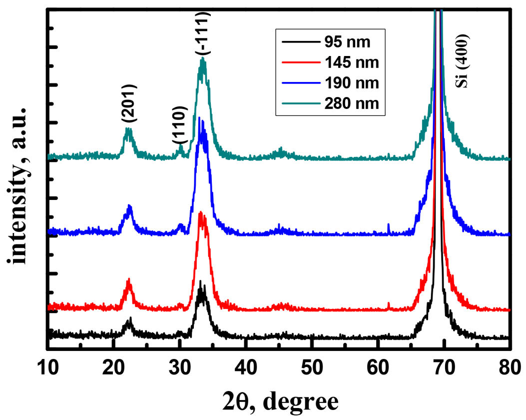

Fig. 1 *XRD spectra of β-Ga_2O_3:Nb thin films with different thicknesses*

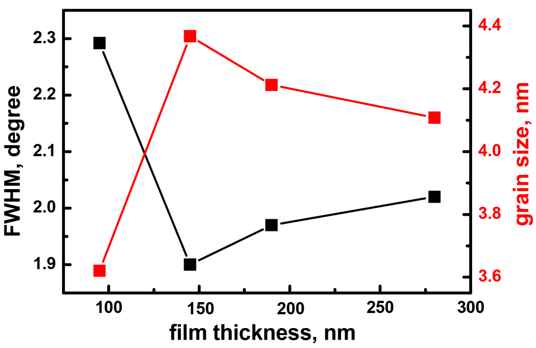

Fig. 2 *FWHM and grain size of (−111) peak as a function of film thickness*

our present Letter. The β-Ga_2O_3:Nb thin film with a thickness of 145 nm possesses the best crystallinity, and it indicates that the crystalline quality of β-Ga_2O_3:Nb thin film can be enhanced by appropriately adjusting the film thickness. This work has guiding significance for the application of β-Ga_2O_3:Nb thin films in devices. The above phenomenon may be mainly related to the lattice mismatch between the β-Ga_2O_3:Nb thin film and Si substrate. In the initial stage of film deposition, a distinct lattice mismatch degrades the crystalline quality. Moreover, the influence of the substrate on the lattice of films decreases to the least when the film thickness exceeds 145 nm.

Fig. 3 displays the 4 × 4 μm AFM images for β-Ga_2O_3:Nb thin films with different film thicknesses. Moreover, the film thicknesses of samples in Figs. 3*a*–*d* are 95, 145, 190 and 280 nm, respectively. In the view of Fig. 3*a*, the surface exhibits hillock morphology. As the film thickness reaches 145 nm, quite a few hillocks merge together and form ridge morphology as shown in Fig. 3*b*. The surface exhibits hillock morphology again after the film thickness exceeds 190 nm. The root-mean-square (RMS) values of the films show a trend of increasing first and then decreasing with an increasing film thickness in this work. Moreover, the RMS value of the 145 nm thick film is the largest, which may be due to the larger grain size of the film.

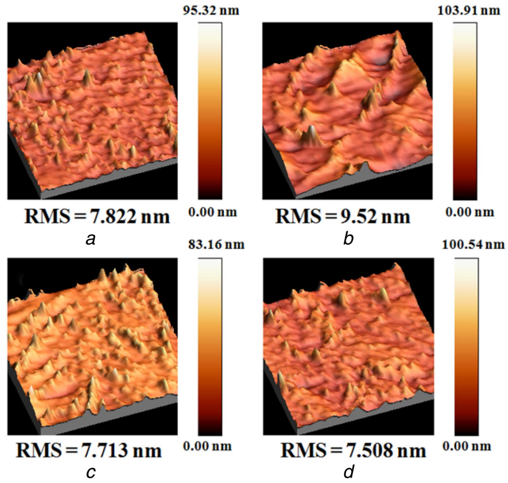

Fig. 3 *AFM images of β-Ga_2O_3:Nb thin films with different thicknesses*
a RMS=7.822 nm
b RMS=9.52 nm
c RMS=7.713 nm
d RMS=7.508 nm

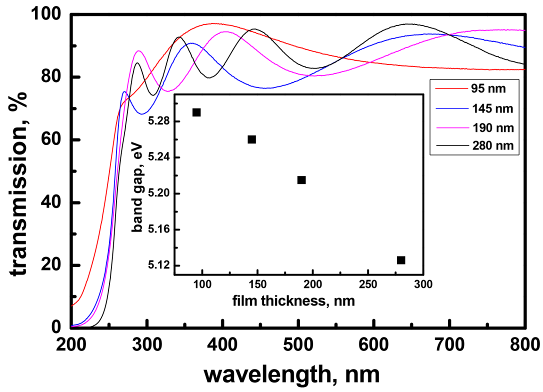

Fig. 4 *Optical transmission spectra of β-Ga_2O_3:Nb thin films. The inset figure shows the optical bandgap of β-Ga_2O_3:Nb thin films with different thicknesses*

The optical transmittance spectra of the β-Ga_2O_3:Nb thin films with different thicknesses in the wavelength range of 200–800 nm are shown in Fig. 4. The average transmittance of all the films in the visible range is over 80%, and it indicates that the thickness of the β-Ga_2O_3:Nb film rarely affects the transmittance of the films. The spectra of the samples show a sharp intrinsic absorption edge at ∼250 nm. With the increase of film thickness, the absorption edge gradually shifts to a long wavelength. For the direct bandgap transition semiconductors, the absorption coefficient α and optical bandgap (E_g) are related by the equation

$$\alpha h\nu = A(h\nu - E_g)^{1/2}$$

where A is a constant, ν is the frequency of the incident photon and h is the Planck's constant. Then, the relation curve of $(\alpha h\nu)^2$ and hν can be plotted, and E_g can be estimated by extrapolating the straight-line portion of this plot to the energy axis. The bandgap E_g for the β-Ga_2O_3:Nb films range from 5.29 to 5.12 eV with increasing the film thickness from 95 to 280 nm, and this is an interesting phenomenon. Previous studies [10–14] indicate that the bandgap of the β-Ga_2O_3 film decreases after the Pr, Nd, W or Sn is incorporated into the film. This is due to the bandgaps of Pr_6O_{11}, Nd_2O_3, WO_3 and SnO_2 are smaller than that of the

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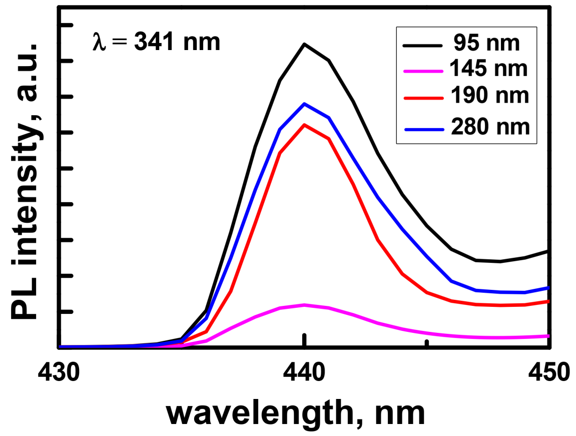

Fig. 5 *Room temperature PL spectra of β-Ga_2O_3:Nb thin films with different thicknesses*

β-Ga_2O_3 film. The bandgap of Nb_2O_5 is 3.4 eV, and it is also narrower than the bandgap of β-Ga_2O_3. While in this work, the bandgap of β-Ga_2O_3:Nb film is larger than 4.9 eV. Moreover, the further systematic study is necessary to reveal the mechanism of this phenomenon. Till now, the β-Ga_2O_3 film has been widely used in the fabrication of photoelectric devices. As is well known, the concentration of impurities and annealing treatment have an influence on the optical properties of the β-Ga_2O_3 film. Moreover, many efforts have been made to improve the photoelectric devices' properties by controlling the concentration of impurities and annealing treatment. While almost no researcher focuses his attention on the influence of film thickness on the device's properties. Our current study shows that the film thickness is also an important factor that influences the film's bandgap, which is closely related to the feature of the device. Therefore, it is of great necessity to consideration of the influence brought along by the film thickness in a future study.

Fig. 5 shows the PL spectra of the β-Ga_2O_3:Nb thin films with different thicknesses in the wavelength range of 430–450 nm, which were recorded at room temperature (300 K) under the excitation of the 341 nm lights. The emissions from β-Ga_2O_3:Nb films are bands located at a wavelength of 440 nm in the blue spectral region, and the peak intensity is a distinct difference between the PL spectra of the four samples. So far as we know, the green emission is related to the radial recombination of a photogenerated hole with the electron in ionised V_O, V_{Ga} and V_{Ga}:V_O [9, 21]. Moreover, the PL intensity of blue luminescence can be reliably used to determine the quality of β-Ga_2O_3 [22]. The PL spectrum is attributed to the radiation recombination caused by the defects. The film with a thickness of 145 nm shows a distinct lower peak intensity than the others, which indicate that this sample possesses fewer defects. It implies that the film with a thickness of 145 nm possesses better crystallinity than the others, and this is consistent with the XRD results. The improved crystallinity decreases the radiation recombination caused by the defects, and further decreases the peak intensity. Similarly, Sun *et al.* [23] also report that the crystalline quality has an effect on the non-radiation recombination and radiation recombination for non-equilibrium photogenerated carriers. There are two factors influencing the PL intensity: one is the defects on the surface of the film and second is defects in the film. At the surface, the crystal lattice ends abruptly, which leaves a large number of dangling bonds. Moreover, the dangling bonds become recombination sites for carriers. Moreover, the amount of defects produced in the film is related to the film thickness, so the film thickness may also have a relationship with the PL intensity. Till now, it cannot be quantitatively evaluated how much the two factors influence the intensity of the PL spectra and which one plays a dominant role in influencing the PL intensity, and further researches are required to investigate it in detail.

4. Conclusions: The β-Ga_2O_3:Nb thin films with different thicknesses were prepared by RF magnetron sputtering method, and the effect of film thickness on the structural and optical properties of β-Ga_2O_3:Nb thin film was investigated. The crystalline quality of β-Ga_2O_3:Nb thin film is significantly improved when the film thickness exceeds 145 nm. The thickness of the β-Ga_2O_3:Nb film rarely affects transmittance of the β-Ga_2O_3:Nb thin film. The bandgap for the β-Ga_2O_3:Nb films range from 5.29 to 5.12 eV with increasing the film thickness from 95 to 280 nm. The improved crystallinity decreases the defects, and further decreases the PL peak intensity.

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6 References

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