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The optical properties of $Mg_xZn_{1-x}O$ thin films*

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$Mg_xZn_{1-x}O$ thin films have been prepared on silicon substrates by radio frequency magnetron sputtering at 60°C. The thin films have hexagonal wurtzite single-phase structure and a preferred orientation with the *c*-axis perpendicular to the substrates. The refractive indices of $Mg_xZn_{1-x}O$ films are studied at room temperature by spectroscopic ellipsometry over the wavelength range of 400–760 nm at the incident angle of 70°. Both absorption coefficients and optical band gaps of $Mg_xZn_{1-x}O$ films are determined by the transmittance spectra. While Mg content is increasing, the absorption edges of $Mg_xZn_{1-x}O$ films shift to higher energies and band gaps linearly increase from 3.24. eV at x=0 to 3.90 eV at x=0.30. These results provide important information for the design and modelling of ZnO/ $Mg_xZn_{1-x}O$ heterostructure optoelectronic devices.

Keywords: $Mg_xZn_{1-x}O$ films, optical properties, sputtering
PACC: 6855, 4225B, 0760F

1. Introduction

In the past several years, a great deal of research efforts on wide band gap semiconductors have resulted in the commercialization of GaN-based blue lasers, light emitting diodes, and ultraviolet photodetectors.[1,2] As an alternative to the GaN material system, ZnO and its alloys are of substantial interest.[3] The exciton binding energy of ZnO is ∼60 meV and results in extremely efficient emission with room temperature optically pumped lasing reported in thin films and microcrystallites.[4,5] In optoelectronic device applications modulation of the optical constant and band gap energy of the constituent layers while keeping their lattice constant close to each other is important for constructing appropriate heterostructures. $Mg_xZn_{1-x}O$ has been considered as an appropriate ternary alloy system that can construct efficient heterojunction with ZnO.[6] Alloying ZnO films with MgO facilitate band gap engineering for heterostructure device design and the replacement of Zn by Mg is not expected to induce significant change of the lattice constants.

There are only few studies on the refractive index and absorption coefficient, which are essential for design and analysis to ZnO-based optoelectronic devices. Spectroscopic ellipsometry (SE) has widely been used as a nondestructive technique to investigate the optical response of semiconductors. Refractive indices can be obtained accurately by fitting SE data (ψ, Δ) to a theoretical model. In this paper, we describe the results of SE experiments performed on $Mg_xZn_{1-x}O$ films prepared on silicon substrates over the wavelength range of 400–760 nm. Excellent fitting of SE measurement data was accomplished by using the first-order Sellmeier dispersion equation, which led to the determination of the dispersions of refractive indices. The dependence of absorption coefficients and optical band gaps of $Mg_xZn_{1-x}O$ films on the Mg content are also investigated by the transmittance spectra.

2. Experimental details

The $Mg_xZn_{1-x}O$ films were deposited on silicon substrates in a JPGF-450 model radio frequency magnetron sputtering system with a base pressure of 5×10^{-3} Pa. A power supply operated at a

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crystal-controlled frequency of 13.56 MHz. The sintered ceramic targets with a mixture of ZnO (purity 99.99%) and MgO (purity 99.99%) were employed as source material. A mixture of Ar (99.999%) and O_2 (99.999%) was used as sputtering gas. The separation between the substrate and target was 5 cm. During film deposition, the argon and oxygen partial pressure maintained at 0.5 Pa, the substrate temperature was kept at 60°C.

The structural properties of the $Mg_xZn_{1-x}O$ films were determined with a Rigaku D/Max-γB x-ray diffractometer (XRD), which used a Cu$K\alpha$ radiation. The SE measurements were carried out in the spectral range 400–760 nm using a Woollam Co. model M88 spectroscopic ellipsometer at the incident angle of 70°. The optical transmittance measurements were performed with a TV-1900 UV double beam spectrophotometer.

3. Results and discussion

The crystal structure of $Mg_xZn_{1-x}O$ films was investigated. The x-ray diffraction spectra for four samples with different Mg contents deposited on silicon substrates are shown in Fig.1. Up to x=0.30 in $Mg_xZn_{1-x}O$ films, $\theta - 2\theta$ scan data exhibited only (002) peaks of $Mg_xZn_{1-x}O$ films. The observation of only (002) peaks revealed that all of the obtained films were sing-phase hexagonal wurtzite structure without any significant formation of a separated MgO phase, and had a preferred orientation with the c-axis perpendicular to the substrate. It is seen that the (002) peak of the alloys shifts to higher angles with increasing Mg content, leading to a decrease of the c-axis lattice constant. The c-axis lattice constants of the films can be calculated by the formula

$$c = \frac{\lambda}{2\sin\theta}\sqrt{\frac{4}{3(a/c)^2}(h^2+hk+k^2)+l^2}, \qquad (1)$$

where h, k and l are Miller indices, λ is the wavelength of x-ray and θ is Bragg diffraction angle.[7] The results indicated that the c-axis lattice constant of the $Mg_xZn_{1-x}O$ films for x=0.30 decreased 1.9% than pure ZnO. Because the ionic radius of Mg^{2+} (0.57Å: coordination number is four) is similar to that of Zn^{2+} (0.60Å: coordination number is four), the replacement of Zn^{2+} by Mg^{2+} does not induce a significant change in lattice constant,[8] which can facilitate heteroepitaxial growth of ZnO/ $Mg_xZn_{1-x}O$.

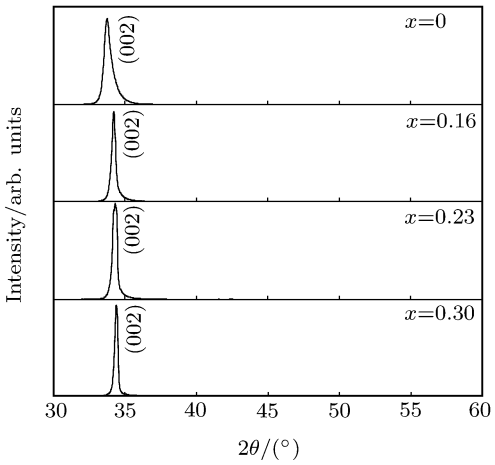


Fig.1. XRD spectra of $Mg_xZn_{1-x}O$ films with different Mg contents.

Two parameters $\psi(\lambda)$ and $\Delta(\lambda)$ shown in SE measurements are related to the complex reflection coefficient $\rho(\lambda)$ and to the ratio of Fresnel coefficients $R_p(\lambda)$ and $R_s(\lambda)$ by the formula

$$\rho(\lambda) = R_p(\lambda)/R_s(\lambda) = \tan(\psi)e^{i\Delta}, \qquad (2)$$

where $\psi(\lambda)$ is the amplitude change between the p- (parallel to the plane of incidence) and s- (perpendicular to the plane of incidence) components of the polarized light that occurs during reflection, and $\Delta(\lambda)$ is difference in the phase. The refractive indices were obtained by fitting the measured ellipsometric parameters $\psi(\lambda)$ and $\Delta(\lambda)$ to an optical model using Woollam Inc. analysis software (WVASE32). Fig.2 showed the refractive indices of $Mg_xZn_{1-x}O$ films obtained by SE technique. As can be seen, (a) the refractive indices were decreased with the increasing of wavelength for different $Mg_xZn_{1-x}O$ films; (b) the refractive indices were decreased with the increasing of Mg content at any certain wavelength over the range of 400–760 nm. The data at each Mg content were fitted by least-square method to the first-order Sellmeier dispersion relationship

$$n(\lambda)^2 = 1 + \frac{A\lambda^2}{\lambda^2 - B^2}, \qquad (3)$$

where A and B are fitting parameters. The fitted values for each Mg content are listed in Table 1, which are similar to the results reported by Teng using a prism-coupling technique.[9] The refractive index of an $Mg_xZn_{1-x}O$ thin film below the band gap can also be determined by analysing the interference fringes of optical transmission spectra. The technique has been

outlined by Swanepoel.[10] We have achieved the refractive indices of $Mg_xZn_{1-x}O$ films by this method and the results, which will be published in another paper, agree with the data gained by SE technique.

Table 1. Fitting parameters in the first-order Sellmeier dispersion formula.

Mg content (x)	0	0.16	0.23	0.30
A	2.57	2.34	2.29	2.17
B	199	202	181	193

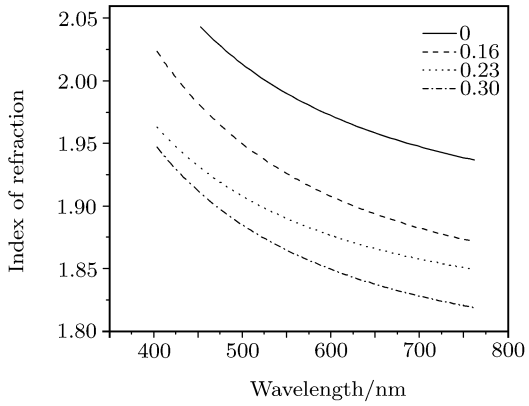


Fig.2. Refractive indices of $Mg_xZn_{1-x}O$ films with different Mg content.

Figure 3 shows the transmittance spectra of the $Mg_xZn_{1-x}O$ films deposited on sapphire substrates with different Mg contents. Transmittances of all $Mg_xZn_{1-x}O$ films are highly transparent in the visible region from 400 to 800 nm and have a sharp absorption edge in the UV region. The absorption edge shifted to shorter wavelength as the Mg content increased, which indicated that the band gap of $Mg_xZn_{1-x}O$ films became wider as the Mg content increased.

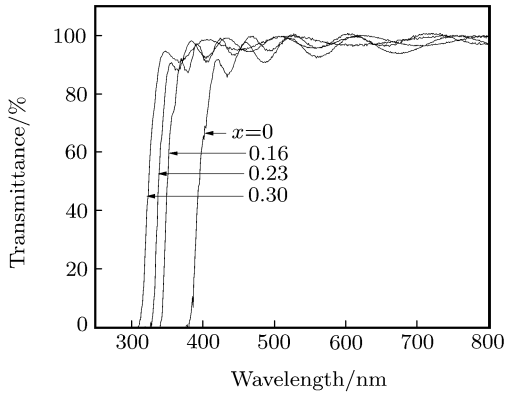


Fig.3. Transmittance spectra of $Mg_xZn_{1-x}O$ films deposited on sapphire substrates.

Figure 4 shows absorption coefficients of $Mg_xZn_{1-x}O$ films as a function of photon energy near the absorption edge. The absorption coefficient was calculated from the transmittance spectra as shown in Fig.3. As can be seen, the absorption coefficients are remarkably dependent on Mg content. The dependence of the band gap energy on the Mg content in these films is shown in Fig.5. It can be seen that the band gap energy of $Mg_xZn_{1-x}O$ films widened linearly from 3.24 to 3.90 eV when the Mg content of $Mg_xZn_{1-x}O$ films increased from x=0 to x=0.30, which is in agreement with the results previously reported,[11,12] demonstrating that the band gap can be modulated within this region. Because the $Mg_xZn_{1-x}O$ films ($0 \leq x \leq 0.30$) have the same structure as ZnO films with a wider band gap, $Mg_xZn_{1-x}O$ is a suitable material for potential barrier layers in ZnO based device.

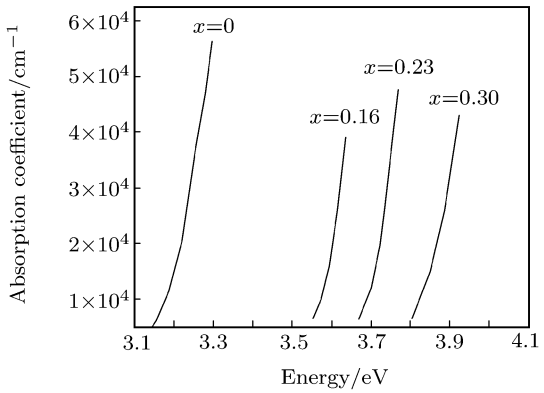


Fig.4. Absorption coefficients of $Mg_xZn_{1-x}O$ films as a function of energy.

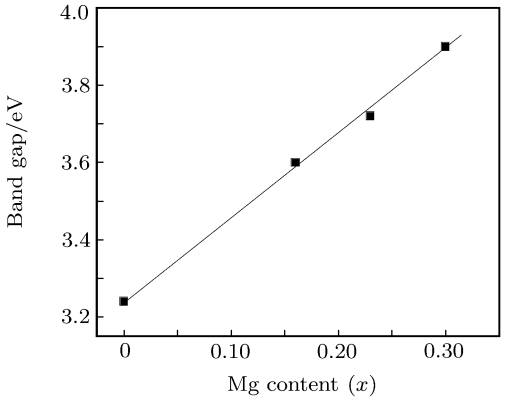


Fig.5. Dependence of the band gap energy of the $Mg_xZn_{1-x}O$ films on the Mg content.

4. Conclusions

In summary, we reported the results of XRD, SE and transmittance spectra of $Mg_xZn_{1-x}O$ films. The $Mg_xZn_{1-x}O$ alloy has the same hexagonal wurtzite structure as ZnO. The c-axis lattice constant of the

$Mg_xZn_{1-x}O$ films decreased slightly than pure ZnO while the refractive indices, band gap energy and absorption coefficients can be modulated by changing Mg content. These results facilitate the design and modelling of ZnO/ $Mg_xZn_{1-x}O$ heterostructure optoelectronic devices.

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