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ARTICLE

Preparation of Mg-doped $In_xGa_{1-x}N$ Thin Films by Magnetron Sputtering and Their Characterization

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Abstract: $In_xGa_{1-x}N$ and Mg-doped $In_xGa_{1-x}N$ thin films were deposited on Si substrate by magnetron sputtering using In_2O_3, Ga_2O_3 and Mg as In, Ga and Mg sources, respectively. The results show that the In component in the film decreases with the doping of Mg, because Mg doping suppresses the formation of In-N bonds and increases the chance of Ga entering the film. The EDS analysis of as-prepared Mg-doped $In_xGa_{1-x}N$ film shows that 1.4% of Mg content is successfully doped into the $In_xGa_{1-x}N$ film. The electrical performances of $In_{0.84}Ga_{0.16}N$ and Mg-doped $In_{0.1}Ga_{0.9}N$ thin films reveal that the conduction type of $In_xGa_{1-x}N$ thin films is transformed from n-type to p-type conduction, and the hole concentration and mobility of Mg-doped $In_{0.1}Ga_{0.9}N$ thin film are 2.65×10^{18} cm^{-3} and 3.9 $cm^2/(V\cdot s)$, respectively.

Key words: $In_xGa_{1-x}N$ thin film; magnetron sputtering; Mg doping; electrical properties

Wide band gap semiconductor has become a core project of sophisticated military and energy conservation industries and has been widely applied to military, new energy, electric cars and other fields because of its high breakdown electric field strength, and thermal conductivity etc, which can significantly promote the performance of electronic devices[1-4]. In $In_xGa_{1-x}N$ alloy material, the band gap of $In_xGa_{1-x}N$ can be adjusted in the range of 0.7~3.4 eV by changing the value of x, which almost covers the whole spectrum of sunlight[5,6]. Therefore, $In_xGa_{1-x}N$ is considered as a promising candidate for solar spectrum materials, such as the multi-junction solar cell white light emitting device and a polishing detector[7, 8].

In recent years, the indium component of $In_xGa_{1-x}N$ thin films as-prepared is lower, and $In_xGa_{1-x}N$ with different indium components can give off light with different colors in the visible range, which has great influence on the light-emitting devices[9,10]. At the same time, the indium localization effect of $In_xGa_{1-x}N$ film plays a major role in the high brightness of light-emitting device[11,12], resulting in that the position of emission peak shifts to long wavelength direction with increasing the indium components. At the present stage, most of $In_xGa_{1-x}N$ materials of the light-emitting diodes are n-type semiconductors with higher electron concentration and lower mobility[13,14], which leads to the reduced hole injection efficiency of n-type $In_xGa_{1-x}N$ material for light-emitting diode (LED). Kumakura[15] et al reported the growth and electric property of the Mg-doped $In_xGa_{1-x}N$ film prepared by MOCVD method and the indium component of Mg-doped $In_xGa_{1-x}N$ thin films was less than 0.2. The results demonstrate that the increase of indium components causes the decreased activation energy (EA) and higher hole concentration of Mg-doped $In_xGa_{1-x}N$ thin film[16,17]. Therefore, it is necessary to dope Mg element into the $In_xGa_{1-x}N$ film, and then the film becomes p-type, and the semiconductor type also transforms from n-type to p-type[18]. The high hole concentration of p-type $In_xGa_{1-x}N$ material will increase the breakdown voltage of the device, while reducing the threshold voltage and leakage current of the device[19,20].

In the present work, $In_xGa_{1-x}N$ and Mg-doped $In_xGa_{1-x}N$ thin films were prepared by radio-frequency magnetron sputtering.

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The structure, morphology, composition and electrical properties of the $In_xGa_{1-x}N$ films were investigated.

1 Experiment

$In_xGa_{1-x}N$ and Mg-doped $In_xGa_{1-x}N$ thin films were deposited on Si substrate by magnetron sputtering. In_2O_3 target and Ga_2O_3 target, with 50.8 mm in diameter and as indium and Ga sources, respectively, were kept at a distance of 80 mm from substrate, and the sputtering powers of them were used at constant RF power of 100 W. The flow rate of nitrogen was kept at 10 $mL·min^{-1}$ and the sputtering pressure was fixed at 1.0 Pa. The deposited temperature and time were 650 ºC and 30 min, respectively. For Mg-doped $In_xGa_{1-x}N$ thin films, Mg target as an Mg source was used at an RF power of 30 W. All the sputtered $In_xGa_{1-x}N$ and Mg-doped $In_xGa_{1-x}N$ thin films were then annealed at 700 ºC in nitrogen atmosphere for 1 h to improve the crystal structure and electrical properties.

The crystal structures of $In_xGa_{1-x}N$ and Mg-doped $In_xGa_{1-x}N$ thin films were analyzed by the X-ray diffraction pattern (XRD, 6100, SHIMADZU) equipped with Cu Kα X-ray source at an incidence angle of 2.0°. The surface microstructures of the two samples as-prepared were observed by scanning electron microscopy (SEM, Zeiss ΣIGMA/VP) and atomic force microscopy (AFM, S-3700N, Hitachi, Japan). The binding energy level and the chemical structure of Mg-doped $In_xGa_{1-x}N$ thin films were investigated by X-ray photoelectron spectroscopy with a monochromatic Al Kα radiation source (XPS, PHI-5400). In order to measure the electrical properties of $In_xGa_{1-x}N$ and Mg-doped $In_xGa_{1-x}N$ thin films, variable temperature Hall system (Phystech RH-2035) was applied to characterize the carrier concentration and electron mobility at room temperature.

2 Results and Discussion

2.1 XRD analysis of thin films

Fig.1 shows the XRD patterns of $In_xGa_{1-x}N$ and Mg-doped $In_xGa_{1-x}N$ thin films. As can be seen from Fig.1a, (100), (002) and (101) peaks of as-deposited thin film are located between the hexagonal wurtzite of InN phase (JCPDS card No. 65-3412) and the hexagonal wurtzite of GaN phase (JCPDS card No. 50-0792), which correspond to the hexagonal wurtzite $In_xGa_{1-x}N$. XRD patterns show no diffraction peaks of the indium and InN, suggesting that the as-prepared thin film does not exhibit indium drops and phase separation phenomenon. Beyond that, it is very interesting that diffraction peak of $In_xGa_{1-x}N$ thin films is close to that of the InN phase, which is attributed to the more indium components. In the XRD patterns for the as-deposited Mg-doped $In_xGa_{1-x}N$ thin films (Fig.1b), the corresponding peaks of (100), (002) and (101) are located at 32.100°, 34.123° and 36.540°, respectively, which red-shift compared to the diffraction peaks of $In_xGa_{1-x}N$ thin films and are closes to the diffraction peaks of GaN phase, because Mg doping suppresses the formation of In-N bond and increases the chance of Ga into the film.

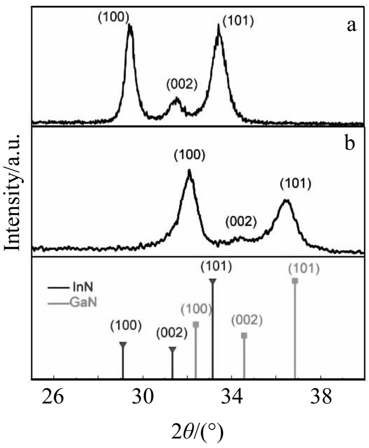

Fig.1 XRD patterns of $In_xGa_{1-x}N$ (a) and Mg-doped $In_xGa_{1-x}N$ (b) thin films

2.2 SEM analysis of thin films

The SEM images of $In_xGa_{1-x}N$ and Mg-doped $In_xGa_{1-x}N$ thin films are shown in Fig.2. As shown in Fig.2a, the surface of $In_xGa_{1-x}N$ thin films is dense with a few sheet particles of regular shapes. Fig.2b shows that Mg-doped $In_xGa_{1-x}N$ thin film exhibits the irregular shape texture and fleeciness of surface morphology due to the introduction of Mg doping.

The energy-dispersive X-ray spectroscopy results in Fig.3 show that the surfaces of the thin films have the expected composition of In, Ga, N, O and Si for the $In_xGa_{1-x}N$ while In, Ga, N, Mg, O and Si for the Mg-doped $Zr_xGa_{1-x}N$. As can be

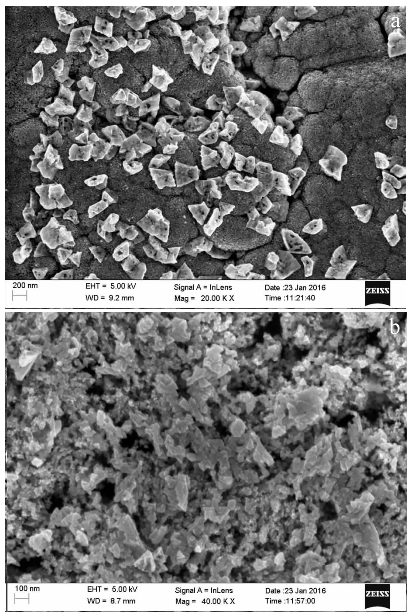

Fig.2 SEM images of the $In_xGa_{1-x}N$ (a) and Mg-doped $In_xGa_{1-x}N$ (b) thin films

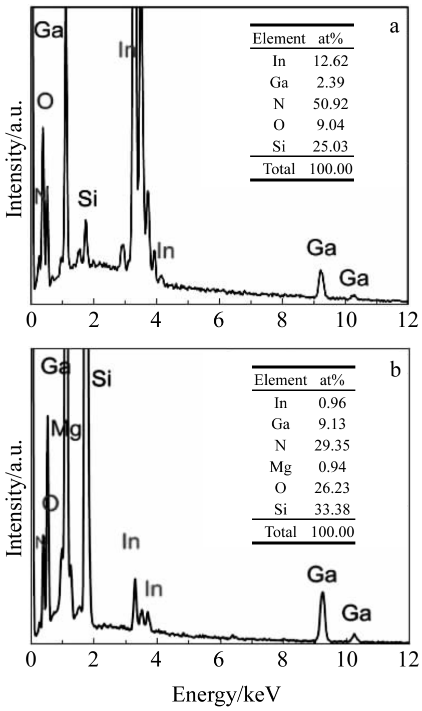

Fig.3 EDS spectra of the $In_xGa_{1-x}N$ (a) and Mg-doped $In_xGa_{1-x}N$ (b) thin films

seen from Fig.3a, the ratio of In:(In+Ga) in $In_xGa_{1-x}N$ thin films is 84%, indicating that the component of thin film is $In_{0.84}Ga_{0.16}N$. The ratio of In:(In+Ga) in Mg-doped $In_xGa_{1-x}N$ thin films is 0.1 and the content of Mg is 0.94 at%, as shown in Fig.3b, which illustrates that the file is $In_{0.1}Ga_{0.9}N$ with 1.4% Mg. It is proved that the Mg elements are incorporated into the films. The indium component in Mg-doped $In_{0.1}Ga_{0.9}N$ thin film decreases to 10% compared with that in $In_{0.84}Ga_{0.16}N$ thin film, which is corresponded to the results of XRD.

2.3 AFM analysis of thin films

Surface morphologies of the $In_xGa_{1-x}N$ and Mg-doped $In_xGa_{1-x}N$ thin films were analyzed by AFM, as shown in Fig.4. It can be seen that the surface of the film presents the island growth model. The root-mean-square (rms) roughness values are 67.15 and 63.28 nm for the $In_xGa_{1-x}N$ (Fig.4a) and Mg-doped $In_xGa_{1-x}N$ (Fig.4b) thin films, respectively, which suggests that the roughness is not a strong function of the Mg content, as the content is not largely changed[21].

2.4 XPS analysis of thin films

In order to study the chemical states, the as-deposited Mg-doped $In_{0.1}Ga_{0.9}N$ thin films were analyzed by XPS. The overall XPS survey scans of the investigated Mg-doped $In_{0.1}Ga_{0.9}N$ thin films deposited after Ar^+ etching are shown in Fig.5, which detects six kinds of elements, including In, Ga, N, C, O and Mg. As can be seen from Fig.5, the electron binding energy peaks at 445.251 and 452.634 eV correspond to In 3d, which is derived from the In-N bond[22]. The electron binding energy peak at 1118.70 eV corresponds to Ga 2p, which is derived from the Ga combined-state[23]. C 1s is the calibration element. The electron binding energy peak at 532.328 eV corresponds to O 1s, which is derived from the oxygen desorption of film surface. The electron binding energy peaks located at 397.900, 395.857 and 393.625 eV correspond to N 1s, which is derived from the In-N and Ga-N bonds, illustrating

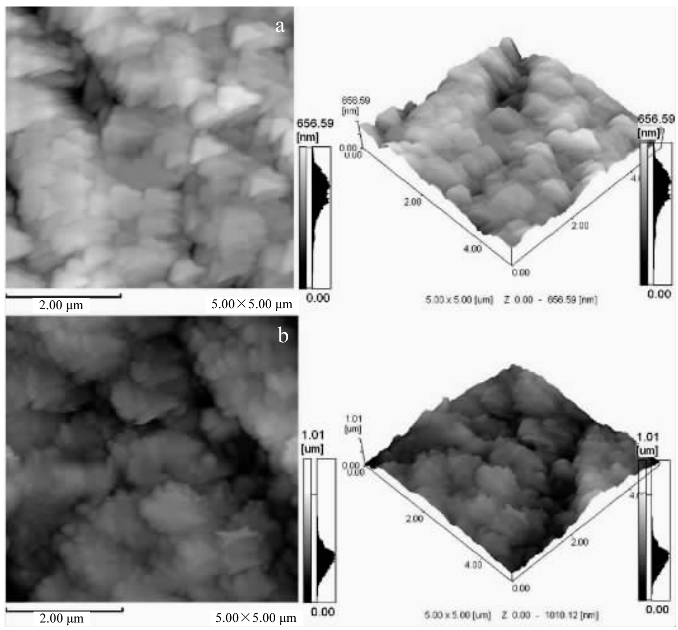

Fig.4 AFM surface images of the $In_xGa_{1-x}N$ (a) and Mg-doped $In_xGa_{1-x}N$ (b) thin films

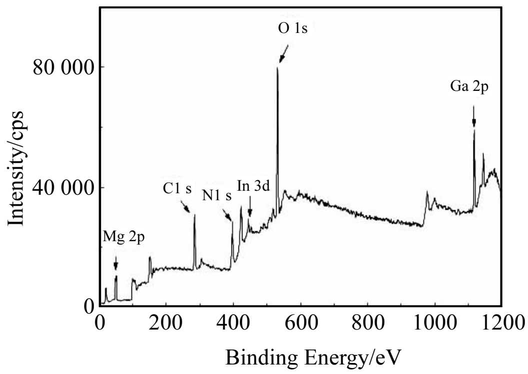

Fig.5 XPS spectrum of Mg-doped $In_xGa_{1-x}N$ thin films

that the N element exists in the form of compounds[22]. The electron binding energy peak at 50.657 eV corresponds to Mg 2p, which is derived from the Mg element, proving that some Mg dopants are successfully incorporated into the $In_xGa_{1-x}N$ films[24].

2.5 Electrical properties of thin film

The carrier concentration, mobility, and electrical conduction type of the $In_xGa_{1-x}N$ and Mg-doped $In_xGa_{1-x}N$ thin films are shown in Table 1.

The as-deposited undoped $In_xGa_{1-x}N$ thin film exhibits the n-type conduction. The electron concentration and mobility of $In_{0.84}Ga_{0.16}N$ thin film are found to be 4.53×10^{20} cm^{-3} and 10.2 $cm^2/(V\cdot s)$, respectively. Furthermore, the as-deposited Mg-doped $In_xGa_{1-x}N$ thin film exhibits the p-type conduction. The hole concentration and mobility of Mg-doped $In_{0.1}Ga_{0.9}N$ thin film are found to be 2.65×10^{18} cm^{-3} and 3.9 $cm^2/(V\cdot s)$, respectively. Lee[4] reported that the Mg-doped p-$In_{0.04}Ga_{0.96}N$ thin films were prepared by the metal organic chemical vapor deposition (MOCVD). The results show that the hole concentration of Mg-doped p-$In_{0.04}Ga_{0.96}N$ thin films is 1.6×10^{18} cm^{-3} and the improvement of hole concentration is caused by the reduction in ionization of the Mg acceptor activation energy in $In_xGa_{1-x}N$ films. Kuo[21] et al reported that the Mg-$In_{0.05}Ga_{0.95}N$ thin films were prepared by the radio frequency magnetron sputtering method, and the carrier concentration and mobility of the film were 1.3×10^{17} cm^{-3} and 4.1 $cm^2/(V\cdot s)$, respectively. And when the In component increased from 0.05 to 0.075, the carrier concentration increased and the mobility decreased due to the increase in the carrier scattering. Therefore, in this work, when In component is 0.1, the values of the carrier concentration and mobility of the Mg-doped $In_{0.1}Ga_{0.9}N$ films are consistentwith the previous study.

Table 1 Holzer test results of undoped and Mg-doped $In_xGa_{1-x}N$ thin film

Sample	Type	Carrier concentration/cm^{-3}	Mobility/ $cm^2\cdot(V\cdot s)^{-1}$
$In_{0.84}Ga_{0.16}N$	n	4.53×10^{20}	10.2
Mg-doped $In_{0.1}Ga_{0.9}N$	p	2.65×10^{18}	3.9

3 Conclusions

1) $In_{0.84}Ga_{0.16}N$ and 1.4%Mg-doped $In_{0.1}Ga_{0.9}N$ thin films are successfully deposited on Si substrates by magnetron sputtering at 660 ºC. The as-deposited thin films are the hexagonal wurtzite $In_xGa_{1-x}N$.

2) The In component in the film decreases with the doping of Mg, because Mg doping suppresses the formation of In-N bonds and increases the chance of Ga entering the film.

3) The conduction type of $In_xGa_{1-x}N$ thin films is transformed from n-type of $In_{0.84}Ga_{0.16}N$ to p-type of Mg-doped $In_{0.1}Ga_{0.9}N$. The hole concentration and mobility of Mg-doped $In_{0.1}Ga_{0.9}N$ thin film are 2.65×10^{18} cm^{-3} and 3.9 cm^2/Vs, respectively.

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Mg 掺杂 $In_xGa_{1-x}N$ 薄膜的磁控溅射法制备和表征

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摘　要：采用磁控溅射法，用 In_2O_3 靶、Ga_2O_3 靶、Mg 靶在 Si 片上制备出 $In_xGa_{1-x}N$ 薄膜和 Mg 掺杂的 $In_xGa_{1-x}N$ 薄膜。薄膜中的 In 组分随着 Mg 的掺杂而减少，因为 Mg 的掺杂抑制了 In-N 键的形成，并增加了 Ga 进入薄膜的机会。通过 EDS 对 Mg 掺杂的 $In_xGa_{1-x}N$ 薄膜的分析表明，有 1.4%的 Mg 组分被成功地掺入 $In_xGa_{1-x}N$ 薄膜。电学性能分析表明 $In_{0.84}Ga_{0.16}N$ 和 Mg 掺杂的 $In_{0.1}Ga_{0.9}N$ 薄膜导电类型由 n 型转变为 p 型，而且 Mg 掺杂的 $In_{0.1}Ga_{0.9}N$ 薄膜的空穴浓度和电子迁移率分别为 2.65×10^{18} cm^{-3} 和 3.9 $cm^2/(V\cdot s)$。

关键词：$In_xGa_{1-x}N$ 薄膜；磁控溅射法；Mg 掺杂；电学性能

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