Ferromagnetism and Structure in Fe-doped GaN Films

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【Abstract】 Fe-doped GaN films have been successfully fabricated on silicon (100) substrates through ammoniating Ga_2O_3:Fe films under flowing ammonia atmosphere at the temperature of 950 °C. The structure of the samples was characterized by X-ray diffraction (XRD). No second phases are found with the increasing of Fe ion concentration from 0% to 7%. Magnetic measurements indicate that all the Fe-doped films were ferromagnetic at room temperature, and the moment per Fe atom decreases with increasing Fe concentration. The largest magnetic moment observed was 1.92μ_B/Fe for $Ga_{1-x}Fe_xN$ (x=0.01) film.

Keywords: Ferromagnetism, Structure, GaN, RF alternate magnetron sputtering, Ammoniation
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Fe 掺杂的 GaN 薄膜的结构和铁磁性

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【摘要】 本文通过高温氨化由磁控溅射方法制备的 Ga_2O_3:Fe 薄膜,成功地在单晶硅(100)基底上制备了 Fe 掺杂的 GaN 薄膜.X 射线衍射结果显示 Fe 掺杂浓度为0%~7%的 GaN 薄膜均未发现第二相.磁性测量表明所有 Fe 掺杂的样品均显示出室温铁磁性,而且每个 Fe 原子的磁矩随 Fe 浓度的增加而减小,Fe 的浓度为 1%时每个 Fe 的磁矩最大,最大值为 1.92μ_B/Fe.

关键词:铁磁性,结构,GaN,磁控溅射,高温氨化
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1 Introduction

Spintronics could lead to new types of devices and circuits. The development of ferromagnetic semiconductors is one of the principal research directions in this field. In recent years, III-V diluted magnetic semiconductors (DMSs) have attracted much attention as materials for application in spintronic devices [1]. In the area of DMS related to III-V compound semiconductors, wide bandgap semiconductor GaN has very recently received great attention as promising host material for making DMS, since its Curie temperature (T_c) higher than room temperature was predicted by theoretical studies [2]. Recently, reports related to GaMnN have been extensively published [3-9], while reports about the GaN:Fe were not studied thoroughly [10-20]. Both ferromagnetism (FM)[11,12,15-20] and paramagnetism (PM)[13] behaviors were observed in (Ga, Fe)N samples. Additionally, Theodoroupoulou *et al.* [16] and Shon *et al.* [12] reported about ferromagnetism in Fe implanted GaN and T_c up to room temperature and even above 350 K, respectively. In these experiments related to (Ga,Fe)N referred above, many experiments were performed by metal-organic chemical vapor deposition method (MOCVD) [12,14] or ion implantation [17,18]. Nevertheless, no one found that (Ga, Fe) N films were prepared by the two-step method using Rf alternate magnetron sputtering and ammoniation at high temperature. So, in this work, Fe-doped GaN films ($Ga_{1-x}Fe_xN$, $x=0.00\sim0.07$) were prepared by this method, and the magnetism and structure properties of the films were studied by various measurements.

2 Experimental procedures

$Ga_{1-x}Fe_xN$ films were fabricated with different concentration ($x=0.00$, 0.01, 0.04, 0.07, 0.10). Firstly, the Ga_2O_3:Fe films were grown by RF alternate magnetron sputtering of Fe (99.99%) and Ga_2O_3 (99.99%) target on silicon (100) substrates. The size of target is ~5 cm in diameter and ~0.3 cm in thickness. The sputtering chamber was evacuated by a molecular pump to a base pressure below 3×10^{-5} Pa. During sputtering, the substrate temperature was kept at 400 ℃. Argon and nitrogen were introduced into the chamber as working gases, and the flow ratio of argon and nitrogen was controlled at 10:1. The chamber pressure was fixed at 2.0 Pa. After sputtering, annealing treatment of the as-grown films was carried out in the sputtering chamber at 600 °C for 10 min. The film thickness was typically from 200 to 300 nm. The concentration of Fe ion in the samples was varied by controlling the sputtering time of Fe target.

Subsequently, the Ga_2O_3:Fe films were ammoniated under flowing ammonia atmosphere with a flow rate of 200 ml/min in a horizontal tube furnace. The temperature and the duration of the ammoniating were 950 °C and 40 min, respectively.

The crystal structure was characterized by X-ray diffraction (XRD), using a CuK_α radiation source. Magnetic measurements were performed using a vibrating sample magnetometer (VSM). The optical properties were studied by photoluminescence spectroscopy (PL).

3 Results and Discussion

XRD patterns of the thin films were shown in Fig. 1. The patterns were indexed to a wurtzite structure GaN. No second phases were detected in the samples with the increasing of Fe ion concentration up to

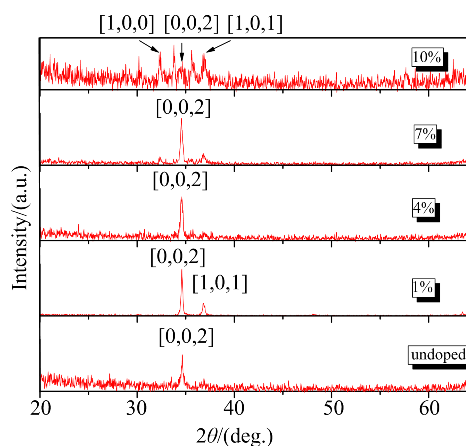

Fig. 1 XRD patterns for the Fe-doped GaN thin films ($Ga_{1-x}Fe_xN$, $x=0.00\sim0.07$)

7%. When Fe ion concentration increased to 10%, the impurity phase ($GaFeO_3$) was observed. It is verified that Fe has substituted Ga or formed interstitial atom in the hexagonal structure with Fe ion concentration from 0% to 7%. The lattice constant c of the samples was calculated from the XRD patterns. Fig. 2 shows the linear increase of c parameters with the increase of Fe concentration. This Vegard's law behavior is evidence of the incorporation of Fe in the Ga site with retention of the wurtzite structure [21].

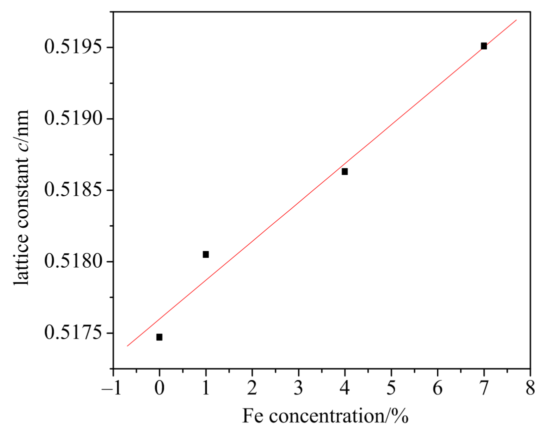

Fig. 2 The linear relation of lattice constant c with Fe concentration

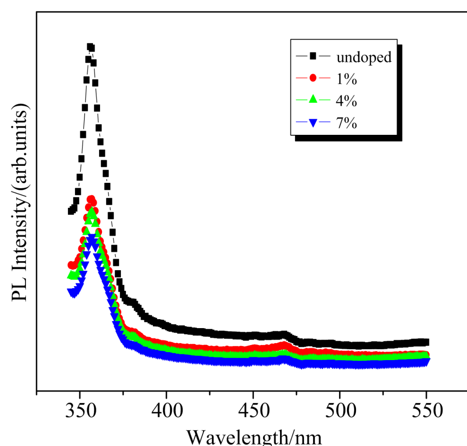

Fig. 3 PL spectra of the Fe-doped GaN thin films ($Ga_{1-x}Fe_xN$, x=0.00~0.07) at RT

Fig. 3 shows photoluminescence (PL) spectra for the samples at room temperature (RT). The excitation source is the 325 nm line of a He-Cd laser with the total power of 50 mW. Two peaks appear at 357 nm and 468 nm in all samples, respectively. The peak at 357 nm is corresponding to 3.48 eV in energy, this is very close to a band gap emission of GaN (3.4 eV) [22]. Another peak at 468 nm is blue luminescence, which corresponds to 2.65 eV energy. This peak is inferred to be mainly caused by the vacancies of N-atoms, which is consistent with the results reported by Z. H. Feng *et al*. [23] It indicates that the doping of Fe does not affect the photoluminescence measurement results of GaN. It proves that the doping of Fe would not change the structure of GaN, and the position of Ga atoms was successfully replaced by Fe atoms. This is also consistent with our previous XRD measurement results.

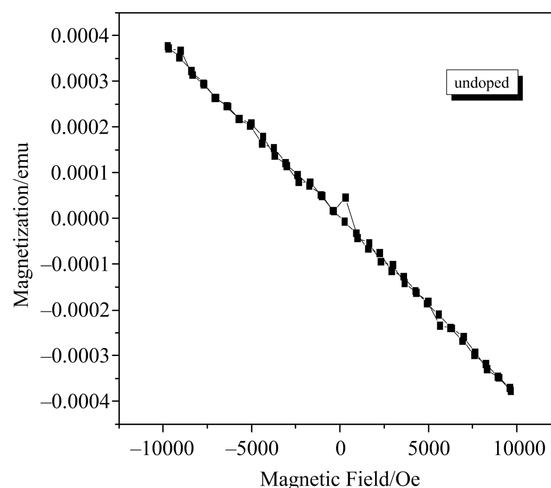

Fig. 4 The M-H curve of undoped GaN sample at RT

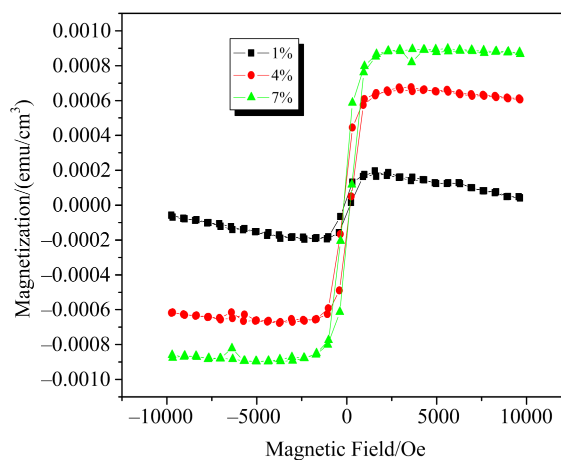

Fig. 5 M-H curves for $Ga_{1-x}Fe_xN$ films (x= 0.01, 0.04, 0.07) at RT

The magnetic properties of the films were determined using a vibrating sample magnetometer. Fig. 4

shows the M-H curve of undoped GaN sample, and it indicates that the undoped GaN sample is diamagnetic. The magnetization versus magnetic field (M-H) curves for Fe-doped GaN films are shown in Fig. 5. In these curves, the diamagnetic background of Si substrate was subtracted. One could see that all the films are ferromagnetic at RT and as the Fe concentration increases, the magnetic moment per Fe ion decreases. The RT saturation moment was calculated with 1.92, 1.65, and 1.26 μ_B per Fe atom for $x = 0.01, 0.04$, and 0.07, respectively. These saturation moments are much lower than the theoretical value of $\mu = g\mu_B[S(S+1)]^{1/2} = 6.7\ \mu_B$ (S represents the total spin quantum number). Combined with the results of XRD and PL measurements, it is concluded that the room temperature ferromagnetism of the films are their intrinsic property originated from the substitute Fe for Ga in the lattice.

The reason why the ferromagnetism of the sample decreased is that with the increase of Fe concentration, the number of Fe ions in the sample increases, resulting in the reduction of the distance between Fe ions. In the competition process of ferromagnetic interaction and antiferromagnetic interaction between Fe ions, the antiferromagnetic interaction gradually increases due to the decrease of the distance between Fe ions, resulting in the decrease of the ferromagnetic interaction of the sample with the increase of Fe concentration.

4 Conclusion

In conclusion, we obtained room-temperature ferromagnetic $Ga_{1-x}Fe_xN$ thin films on Si (100) substrates by ammonification technique under flowing ammonia atmosphere at 950 °C. The XRD patterns of the films were indexed to a wurtzite structure GaN with the Fe concentration from 0% to 7 %. All the Fe-doped GaN thin films are ferromagnetic at room temperature, and the magnetic moment decreases with increasing Fe concentration.

参 考 文 献

[1] Y. Ohno, D. K. Young, B. Beschoten, F. Matsukura, H. Ohno, D. D. Awschalom, *Nature*, **402** (1999), 790

[2] T. Dietl, H. Ohno, F. Matsukura, J. Cibert, D. Ferrand, *Science*, **287** (2000), 1019

[3] S. H. Doo, P. Jeunghee, W. R. Kung, K. Soonkyu, C. Joonyeon, *Appl. Phys. Lett.*, **86** (2005), 032506

[4] C. K. Xu, J. Chun, K. Rho, D. E. Kim, B. J. Kim, S. Yoon, S. E. Han, J. J. Kim, *J. Appl. Phys.*, **99** (2006), 064312

[5] S. Sonoda, I. Tanaka, F. Oba, H. Ikeno, H. Hayashi, T. Yamamoto, Y. Yuba, Y. Akasaka, K. Yoshida, M. Aoki, M. Asari, T. Araki, Y. Nanishi, K. Kindo, H. Hori, *Appl. Phys. Lett.*, **90** (2007), 012504

[6] Y. K. Byeun, K. S. Han, H. J. Choi, S. C. Choi, *Mater. Sci. Eng. A*, **452** (2007), 499

[7] K. Biswas, K. Sardar, C. N. R. Rao, *Appl. Phys. Lett.*, **89** (2006), 132503

[8] Y. Shi, Y. X. Zhang, C. Z. Jiang, D. J. Fu, X. J. Fan, *Physica B*, **388** (2007), 82

[9] M. H. Ham, J. M. Myoung, *Appl. Phys. Lett.*, **89** (2006), 173117

[10] S. Kuwabara, K. Ishii, S. Haneda, T. Kondo, H. Munekat, *Phys. E*, **10** (2001), 233

[11] N. Theodoropoulou, M. E. Overberg, S. N. G. Chu, A. F. Hebard, C. R. Abernathy, R. G. Wilson, J. M. Zavada, K. P. Lee, S. J. Pearton, *Phys. Stat. Sol.* (*b*), **228** (2001), 337

[12] Y. Shon, Y. H. Kwon, Y. S. Park, S. U. Yuldashev, S. J. Lee, C. S. Park, K. J. Chung, S. J. Yoon, H. J. Kim, W. C. Lee, D. J. Fu, T. W. Kang, X. J. Fan, Y. J. Park, H. T. Oh, *J. Appl. Phys.*, **95** (2004), 761

[13] J. Gosk, M, Zajac, M. Byszewski, M. Kaminska, J. Szczytko, A. Twardowski, B. Strojek, S. Podsiadlo, *J. Supercond.*, **16** (2003), 79

[14] H. Przybylinska, A. Bonanni, A. Wolos, M. Kiecana, M. Sawicki, T. Dietl, H. Malissa, C. Simbrunner, M. Wegscheider, H. Sitter, K. Rumpf, P. Granitzer, H. Krenn, W. Jantsch, *Mater. Sci. Eng. B*, **126** (2006), 222

[15] H. Akinaga, S. Nemeth, J. De Boeck, L. Nistor, H. Bender, G. Borghs, H. Ofuchi, M. Oshima, *Appl. Phys. Lett.*, **77** (2000), 4377

[16] N. Theodoropoulou, A. F. Hebard, S. N. G. Chu, M. E.

Overberg, C. R. Abernathy, S. J. Pearton, R. G. Wilson, J. M. Zavada, *J. Appl. Phys.*, **91** (2002), 7499

[17] W. Kim, H. J. Kang, S. K. Noh, J. Song, C. S. Kim, *J. Magn. Magn. Mater.*, **316** (2007), e199

[18] N. Theodoropoulou, A. F. Hebard, S. N. G. Chu, M. E. Overberg, C. R. Abernathy, S. J. Pearton, R. G. Wilson, J. M. Zavada, *Appl. Phys. Lett.*, **79** (2001), 3452

[19] P. Muzammil, S. M. Basha, G. S. Muhammed, *J. Supercond. Nov. Magn.*, **33** (2020), 2767

[20] B. S. Li, D. P. Peng, J. H. Li, L. Kang, T. M. Zhang, Z. X. Zhang, S. X. Jin, X. Z. Cao, J. H. Liu, L. Wu, X. Wang, Z. Q. Fang, C. L. Zhou, Z. Yang, V. Krsjak, *Vacuum*, **184** (2021), 109909

[21] E. Chikoidze, Y. Dumont, F. Jomard, D. Ballutaud, P. Galtier, D. Ferrand, V. Sallet, O. Gorochov, *Mater. Res. Bull.*, **41** (2006), 1038

[22] H. Y. Peng, X. T. Zhou, N. Wang, Y. F. Zheng, L. S. Liao, W. S. Shi, C. S. Lee, S. T. Lee, *Chem. Phys. Lett.*, **327** (2000), 263

[23] Z. H. Feng, B. Liu, F. P. Yuan, J. Y. Yin, D. Liang, X. B. Li, Z. Feng, K. W. Yang, S. J. Cai, *J. Cryst. Growth*, **309** (2007), 8