Effect of Sputtering Power on the Structural and Optical Properties of InN Nanodots on Al₂O₃ by Magnetron Sputtering

Ziming Zhang *, Jingjie Li *, Yijian Zhou *, Hongyuan Fu *, Zixu Zhang *, Guojiao Xiang *, Yang Zhao ***, Shiwei Zhuang †, Fan Yang ††, and Hui Wang *

*Henan University of Science and Technology, School of Physics and Engineering, Henan Key Laboratory of Photoelectric Energy Storage Materials and Applications, 471003, Luoyang, China
†Zhengzhou University, Department of Physics and Engineering, Key Laboratory of Materials Physics of Ministry of Education, 450052, Zhengzhou, China
††Jilin Jianzhu University, School of Electrical Engineering and Computer, Jilin Provincial Key Laboratory of Architectural Electricity & Comprehensive Energy Saving, 130118, Changchun, China

Received: June 13, 2019; Revised: August 31, 2019; Accepted: November 25, 2019

In this work, we reported the effects of sputtering power on the structure, optical and electrical properties of InN nanodots prepared on Al₂O₃ substrate by magnetron sputtering. The results showed that the as-grown InN films exhibited uniform nanodot morphology and the size of the InN nano grains increased with the sputtering power was increased. The InN nanodot exhibited highly c-axis preferred orientation with mainly InN (002) diffraction. The optical band gap of InN samples showed an decreasing trend with the increase in sputtering power. Moreover, the electrical properties of the InN samples were discussed in detail by hall effect and the carrier concentration and mobility could be adjusted from 3.233×10¹⁹ to 1.655×10²⁰ cm⁻³ and 1.151 to 10.101 cm²/v•s, respectively. These results will lay a good foundation for the application of InN material in the field of gas sensors and light emitting diodes.

Keywords: InN nanodots, magnetron sputtering, highly preferred orientation, electrical characteristic.

1. Introduction

Indium nitride, as a kind of novel material among the III-V group nitrides, has been broadly applied in the fields of high-speed electronic devices, high efficiency solar cells, infrared light emitting diodes and laser diodes. It has lower effective electron mass, higher saturation electron drift rate and higher electron mobility, which make it ideal for the development of the above optoelectronic devices. However, the growth of high-quality InN films is difficult due to the lower decomposition temperature of InN (~550°C). So far, InN films could be prepared by using a variety of techniques. It has been confirmed that the bandgap of single crystal InN grown by molecular beam epitaxy (MBE) or metal organic vapor phase epitaxy (MOVPE) is around 0.7 eV. It is noted that the initial band gap value of the InN films prepared by magnetron sputtering is about 2.0 eV due to the Burstein-Moss effect and oxygen impurity incorporation. Nevertheless, from the point of experiment cost and low temperature of growth condition, the sputtering has the advantages over the other methods such as MBE or MOVPE. At the same time, the material prepared by magnetron sputtering usually exhibited highly textured structure like nanodots, nanowires and nanopillars, which would be affected relatively lighter by the lattice mismatch between the epilayer and substrate. This would be benefit for the application of InN-based gas sensors and photodevices. Recently, Chen et al. have reported the synthesis of self-organized InN nanodots on Si substrate by droplet epitaxy method. Li et al. have reported the growth of well-aligned InN nanorods on glass substrate by metal-organic chemical vapor deposition. In this work, we reported the growth of well-oriented InN nanodots on sapphire substrate by magnetron sputtering. The structure, morphology, optical and electrical characteristics of InN nanodots were systematic investigated as a function of the sputtering power.

2. Experiments

InN films were prepared on sapphire substrate by radio frequency (RF) vacuum magnetron sputtering system. The substrate was cleaned by ultrasonic cleaning in acetone (CH₃COCH₃), alcohol (C₂H₅OH) and deionized water respectively for 5 minutes. Then the substrates were dried with nitrogen and placed 5 cm from the target in the reaction chamber. The substrate temperature and pressure were kept at 200 °C and 1 pa, respectively. The nitrogen flux added into the chamber were maintained at 20 sccm. Before formal sputtering, the target was under presputter for 5 minutes to remove the contaminants on the target surface. Then the sputtering was controlled at 1 hour. Under the above experimental conditions, InN samples marked A-D were prepared by setting the sputtering power at 80, 90, 100 and 110 W, respectively.
The crystallization of InN samples were analyzed by X-ray diffraction (XRD; D8 Discover Gadds) measurement. The surface morphology of InN samples were investigated by atomic force microscope (AFM; Veeco Dimension 3100). Moreover, the absorption and electrical characteristics of InN samples were analyzed by UV-2700 spectrophotometer and Accent HL5500PC Hall effect system, respectively.

### 3. Results and Discussion

To analyze the crystal structure of InN films prepared under different sputtering power, XRD measurements were performed as shown in Fig. 1. As seen from Fig. 1, all InN samples exhibited mainly InN (002) diffraction, which indicated that all InN samples grew preferentially along the c-axis direction and the (002) orientation. Besides, the intensity of the InN (002) diffractions increased with the sputtering power was increased from 80 to 110 W. And the full width at half maximum (FWHM) of InN (002) diffractions showed a decreasing trend as the sputtering power was increased. These results indicated that the increase of sputtering power is beneficial to the growth and crystallization of InN grains. This was probably attributed to the increase of kinetic energy for InN grains to nucleation as increasing the sputtering power. While the intensity ratio of InN (002)/(101) for the InN samples grown at sputtering power of 80, 90, 100 and 110 W were 1.59, 1.655×10^20 cm^-3 and 1.151 to 10.101 cm^2/v•s, respectively. As can be seen from Fig. 5, all InN samples exhibited mainly InN (002) diffraction, which was usually due to the fact that the materials prepared by sputtering usually exhibited polycrystal qualities.

The optical properties of InN samples were determined by optical absorption measurement. These experimental results were then combined with theoretical calculations. The value of optical band gap could be determined following equation derived independently by Tauc et al.:

\[ a h \nu = A (h \nu - E_g)^{2} \]  

Where A is a constant and \( h \nu \) is the photon energy. The value of the \( E_g \) can be calculated by the extrapolation of linear part to the horizontal axis as shown in Fig. 4. The \( E_g \) for the InN samples marked A-D sputtered at the power of 80, 90, 100, and 110 W, respectively, was found to be 1.83, 1.82, 1.78 and 1.77 ev, respectively. These values of bandgap indicated that the as-grown InN materials could be used in the field of near infrared photodetectors and light emitting diodes. Moreover, the value of the bandgap was in according with the reported results (1.60-1.90 eV) obtained by Felip et al. using the radio frequency sputtering. However, the results was more larger than the established bandgap values of 0.7-1.0 eV reported for high quality single-crystalline InN grown by MBE or MOVPE. The larger optical band gap could be attributed to the high free electron concentration, which usually induced the Burstein-Moss effect.

Fig. 5 shows the relationship between the electrical properties of InN samples and the sputtering power. As can be seen from Fig. 5, all InN samples exhibited strong n-type conductivity characteristics with high carrier concentrations. And the carrier concentration and mobility could be adjusted from 3.233×10^19 to 1.655×10^20 cm^-3 and 1.151 to 10.101 cm^2/v•s, respectively.
Effect of Sputtering Power on the Structural and Optical Properties of InN Nanodots on Al₂O₃ by Magnetron Sputtering

Figure 2. 3D morphology of the AFM images (5×5 um²) of the InN nanodots grown at different sputtering powers: (a) 80W, (b) 90W, (c) 100W and (d) 110 W.

Figure 3. SEM images of the InN samples grown at different sputtering powers: (a) 80W, (b) 90W, (c) 100W and (d) 110 W.
These values were in good agreement with previous results reported in Wang’s et al. research work. One can observe that the mobility of the InN samples were significantly increased with the increase in sputtering power, which was mainly attributed to the increased surface migration energy of the adatoms. Besides, the resistivities of the InN samples exhibited an increasing trend when the sputtering power was increased as shown in Fig. 5(b). This was in accordance with the trend of surface roughness, which was mainly due to the influence of the changes in InN crystal qualities. These results indicated that the electrical properties of InN nanodots can be improved by the selection of appropriate sputtering power.

4. Conclusion

InN nanodots were prepared by magnetron sputtering on sapphire substrates at various sputtering powers. We have presented a detailed research on the influence of sputtering power on the physical properties of InN nanodots. It was found that the InN nanodots grew preferentially along the c-axis direction with InN (002) orientation. It had a homogeneous surface and the size of InN nanodots increased as the sputtering power was increased. Moreover, the optical band gap of the InN nanodots were found to be around 1.77–1.83 eV. And the Hall test results showed that all InN nanodots exhibited n-type conductivity characteristics with higher carrier concentration. It was believed that the InN nanodots can be used as a good n-type semiconductor material in the field of photovoltaic devices.

5. Acknowledgment

This work was supported by the National Natural Science Foundation of China (Grant Nos. 61674052 and 11404097), the Key Scientific Research Projects of Higher Education Institutions of Henan Provinc, China (Grant No. 20A140012), the Innovation and Entrepreneurship Training Program for Provincial Undergraduates of Henan Province (Grant No. S201910464024), the Student Research Training Program of Henan University of Science and Technology (Grant No. 2019208), and the Student Research Training Program of School of Physics and Engineering of Henan University of Science and Technology (Grant Nos. WLSRTP201910 and WLSRTP201802).
6. Reference

1. Reilly CE, Lund C, Nakamura S, Mishra UK, DenBaars SP, Keller S. Infrared luminescence from N-polar InN quantum dots and thin films grown by metal organic chemical vapor deposition. Applied Physics Letters. 2019;114(24):241103.

2. Zhao Y, Wang H, Li XZ, Li JJ, Shi ZF, Wu GG, et al. Parametric study on the well-oriented growth of InxAl1-xN nanodots by magnetron sputtering. Materials Science in Semiconductor Processing. 2019;102:104583.

3. Shi ZF, Li Y, Zhang YT, Chen YS, Li XJ, Wu D, et al. Nano Letters. 2017;17:313-21.

4. Zhao Y, Wang H, Gong XY, Li QZ, Wu GG, Li WC, et al. Near infrared electroluminescence from p-NiO/n-InN/n-GaN light-emitting diode fabricated by PAMBE. Journal of Luminescence. 2017;186:243-6.

5. Wang H, Zhao Y, Li XZ, Zhen ZQ, Li QZ, Li HH, et al. Dominant near infrared light-emitting diodes based on p-NiO/n-InN heterostructure on SiC substrate. Journal of Alloys and Compounds. 2018;735:1402-5.

6. Wang H, Zhao Y, Li Z, Li JJ, Zhang ZM, Wan S, et al. Growth of well-oriented InN nanodots by magnetron sputtering with varying sputtering temperature. Journal of Vacuum Science and Technology: B. 2018;36:041204.

7. Shi ZF, Li Y, Li S, Li XY, Wu D, Xu TT, et al. Localized surface plasmon enhanced all-inorganic perovskite quantum dot light-emitting diodes based on coaxial core/shell heterojunction architecture. Advanced Functional Materials. 2018;28(20):1707031.

8. Koukitu A, Takahashi N, Seki H. Thermodynamic study on metalorganic vapor-phase epitaxial growth of group III nitrides. Japanese Journal of Applied Physics. 1997;36(Pt 2):L1136.

9. Bhuiyan AG, Hashimoto A, Yamamoto A. Indium nitride (InN): a review on growth, characterization, and properties. Journal of Applied Physics. 2003;94(5):2779.

10. Wu J, Walukiewicz W, Shan W, Yu KM, Ager JW, Haller EE, et al. Effects of the narrow band gap on the properties of InN. Physical Review: B. 2002;66(20):201403.

11. Chen HJ, Yang D, Huang TW, Yu IS. Formation and temperature effect of inn nanodots by PA-MBE via droplet epitaxy technique. Nanoscale Research Letters. 2016;11:241.

12. Li HJ, Zhao GJ, Wei HY, Wang LS, Chen Z, Yang SY. Growth of well-aligned InN nanorods on amorphous glass substrates. Nanoscale Research Letters. 2016;11(1):270.

13. Zhao Y, Wang H, Yang F, Wang ZY, Li JJ, Gao YT, et al. Materials Research-Ibero-American Journal. 2018;21:e20170836.

14. Wang H, Zhao Y, Li XZ, Zhen ZQ, Li HH, Wang JG, et al. Vacuum. 2017;144:199-202.

15. Wu J, Walukiewicz W, Li SX, Armitage R, Ho JC, Weber ER. Effects of electron concentration on the optical absorption edge of InN. Applied Physics Letters. 2004;84:2805.

16. Biju KP, Jain MK. The effect of RF power on the growth of InN films by modified activated reactive evaporation. Applied Surface Science. 2008;254(22):7259-65.

17. Tauc J, Grigorovici R, Vancu A. Optical properties and electronic structure of amorphous germanium. Physica Status Solidi: B. 1996;15(2):627.

18. Chowdhury AM, Pant R, Roul B, Singh DK, Nanda KK, Krupanidhi SB. Double Gaussian distribution of barrier heights and self-powered infrared photoresponse of InN/AlN/Si (111) heterostructure. Journal of Applied Physics. 2019;126(2):025301.

19. Valdueza-Felip S, Naranjo FB, González-Herráez M, Lahourcade L, Monroy E, Fernández S. Influence of deposition conditions on nanocrystalline InN layers synthesized on Si(1 1 1) and GaN templates by RF sputtering. Journal of Crystal Growth. 2010;312(19):2689-94.

20. Kumar M, Bhat TN, Rajpalke MK, Roul B, Kalghatgi AT, Krupanidhi SB. Indium flux, growth temperature and RF power induced effects in InN layers grown on GaN/Si substrate by plasma-assisted MBE. Journal of Alloys and Compounds. 2012;513:6-9.