SYNTHESIS AND PROPERTIES OF FERROMAGNETIC Mn-DOPED AlN FILMS

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Abstract: Mn-doped AlN semiconductor (AlMnN) films are a promising material for spintronics because of high temperature ferromagnetism. AlMnN films were synthesized by DC sputtering technique. The crystal structure of samples was characterized by XRD. The concentration of Mn was determined by EDS. Using the optimal growth process, we have obtained the single phase AlMnN films with the Mn content up to 13.6 at %. The saturated magnetization and energy band gap were found to decrease with increasing Mn concentration. The values of activation energy were derived from the temperature dependence of resistance, which exhibits clearly a semiconductor characteristic.

1. Introduction

The combination of several discoveries in magnetic semiconductors has opened up the possibility of a new research field that is so-called spintronics. A subset of this field involves diluted magnetic semiconductors (DMS). The main focus of the DMS area is to effectively incorporate magnetic ions into a semiconductor lattice and create a ferromagnetic material. Since Dietl et al. [1] predicted that cubic GaN doped with 5 at % of Mn should exhibit a Curie temperature (Tc) exceeding room temperature, a number of works have focused on wide band gap semiconductors as being the most promising ones for achieving high Tc. Among them, the works on Mn-, or Cr-doped AlN films [2,3] based on AlN semiconductor with the band gap of 6.2 eV are of particular important. Room temperature magnetism has been reported for Mn-doped films grown by molecular beam epitaxy [2].

This paper presents the synthesis of Mn-doped AlN films using reactive DC sputtering technique, along with the effect of Mn concentration on the properties such as crystal structure, magnetization, optical band gap, and resistivity.

2. Experiments

Samples of composition $Al_{1-x}Mn_xN$, where x is the atomic fraction of Mn substituted for Al, were deposited simultaneously on quartz substrates in a reactive DC magnetron sputtering system. The composite target includes a high purity (99.999%) aluminum disk and a number of square Mn pieces 5×5 mm, which were placed symmetrically on the surface of the Al disk. An investigation of deposition process has been done on undoped films. The deposition conditions were optimized and fixed as following: the partial pressure of Ar and N_2 gas mixture (67% Ar, 33% N_2) was 6 mTorr, the DC voltage was 240V, the DC current was 330mA, the substrate temperature during deposition was 300°C. The Mn contents were controlled by varying the number of Mn pieces. The XRD measurements were carried out in grazing mode using Cu-K_α radiation. The magnetization of the films was measured by a SQUID magnetometer. The band gaps were obtained from optical absorption measurement over the wavelength from 200 to 900 nm in transmittance mode.

3. Results and discussion

The atomic concentrations of Mn determined by EDS were from x = 0.00 to 0.136, corresponding to the number of Mn pieces varying from 0.0 to 3.0.

Fig. 1 shows XRD pattern for one particular 0.40 μm thick film with x = 0.075. All peaks are identified with the hexagonal AlN structure. The X-ray data provide evidence that the present Mn-doped AlN films are single phase. The second phases were found by X-ray for the films with Mn content exceeding 0.136.

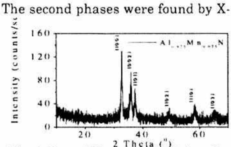

Fig.1. X-ray diffraction (XRD) data for the 0.4 μm thick $Al_{0.925}Cr_{0.075}N$ films.

Fig. 2, shows magnetization versus field data for 0.4 μm thick with x = 0.136. The values of saturated magnetization indicate that 20% of Mn is magnetically active in AlN samples compared with $4\mu_B$/Mn atom [4]. We found that Ms at room temperature decreased with increasing Mn concentration, namely, 9.6, 8.3 and 7.7 emu/cm³ for x = 0.045, 0.075 and 0.136, respectively. The origin of this behavior is unclear. It may result from the compensation by interstitial Mn donors and/or from changes in the local spin configurations [5].

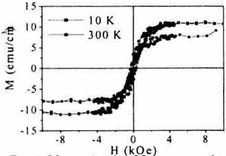

Fig. 2: Magnetization M versus static external magnetic field H measured at 10 K and 300 K for the 0.40 μm thick film. The field was applied in plane

The UV optical absorption was performed on the films grown on quartz substrates with the typical thickness of 400 nm. Inset in Fig. 3 shows α^2 vs hν plots (α is absorption coefficient) for a $Al_{0.955}Mn_{0.045}N$ films obtained by Tsuc's method. The band gap was determined by extrapolating the linear portion of the absorption edge to zero value. The band gap with Mn concentration is shown in main graph of Fig. 3. It is clear that the band gap decreases exponentially with increasing Mn concentration. These band gaps correspond to the transition from the top of the impurity band to the bottom of the conduction band. We are unaware of an electronic structure calculation for transition-metal-doped AlN systems. Some recent reports have theoretically produced the electronic structure of transition-metal-doped GaN, which is rather related to our cases [4,6]. According to Sanyal et al. [6], the system behaviors like a ferromagnetic metal with a reduced magnetic moment at Mn concentration of 5 %. In this point of view, the exponential decrease of optical band gap in Mn-doped is related with band structure change and strongly correlated with magnetic behavior with Mn concentration exceeding 5 %.

Transport property was measured by four-probe method. Fig. 4 shows the temperature dependence of resistance, R(T), for the 13.6 % Mn-doped AlN film. R(T) plots follow the expression $R = R_o T^{1/2}\exp(E_a/k_B T)$, which describes the transport property in a semiconductor material. By fitting the R(T) plot, we could obtain the value of activation energy E_a = 145 meV. Further studies have pointed out that transport characterization depends not only on the Mn concentration but also on the crystalinity of samples.

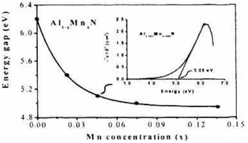

Fig.3. Optical band gap as a function of Mn concentration for $Al_{1-x}Mn_xN$ (x = 0.0 - 0.136) films. Inset is α^2 vs photon energy plots at x = 0.045 to get optical band gap

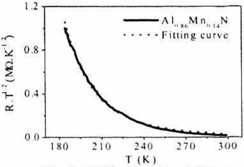

Fig.4. $R.T^{1/2}$ vs T plot and fitting curve for $Al_{0.86}Mn_{0.14}N$ film

In summary, we have obtained the single-phase $Al_{1-x}Mn_xN$ films with the Mn concentration up to a value of x = 0.136. The saturated magnetization and band gap decreased with increasing Mn concentration. The exponential decrease of optical band gap in Mn- doped is strongly correlated with magnetic behavior. Temperature dependence of resistance exhibits clearly a semiconductor characteristic.

Acknowledgements. The work at Hanoi University was supported by the Vietnamese-Italy cooperation program (8BS3). The work in Korea was supported by the Korea Research Foundation Grant (KRF-2003-005-C00018).

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