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Magnetism modulation in Cu-doped AlN via coupling between AlN thin film and ferroelectric substrate

J.T. Luo [a,*], X.Y. Kang [b], B. Fan [b], F. Zeng [b,*], F. Pan [b], P. Fan [a], Z. Jiang [c], Y. Wang [c]

[a] Institute of Thin Film Physics and Applications, Shenzhen Key Laboratory of Sensor Technology, Shenzhen University, Shenzhen 518060, China
[b] Laboratory of Advanced Materials, Department of Materials Science and Engineering, Tsinghua University, Beijing 100084, China
[c] Shanghai Synchrotron Radiation Facilities, Shanghai Institute of Applied Physics, Chinese Academy of Sciences, Shanghai 201204, China

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ABSTRACT

Cu-doped AlN (Cu:AlN) thin films were deposited on non-ferroelectric substrate (Si (1 1 1)) and ferroelectric substrates ($LiTaO_3$ and $LiNbO_3$) by direct current reactive magnetron sputtering. It was discovered that all the deposited Cu:AlN thin films showed room temperature ferromagnetism and the average magnetic moment (AMM) per Cu atom of the film increased with the decrease of the angle (θ) between AlN film plane and the *c*-axis of ferroelectric substrate. A giant AMM as high as 2.23 μ_B/Cu was obtained when Cu:AlN thin film was deposited on $LiTaO_3$(110) with θ of 0 degree, while a small AMM of about 0.11 μ_B/Cu was observed when Cu:AlN thin film was deposited on $LiNbO_3$ (006) with θ of 90°. It was discovered that the AMM of the film increased with decreasing of θ. The ferromagnetism modulation was ascribed to the coupling between AlN piezoelectric film and ferroelectric substrate, resulting from the change of host lattice structure and spontaneous polarization of AlN thin film. The in-plane anisotropy of AMM and the annealing relaxation results confirmed the anomalous coupling.

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1. Introduction

Just like the permanent magnetic materials [1], as a kind of promising material to integrate the synergetic use of charge and spin dynamics of electron into one component, diluted magnetic semiconductors/insulators (DMS/DMI) have attracted much attention in the last decades [2–4]. Among the potential host materials for DMS/DMI, AlN based DMS/DMI have been extensively studied since they exhibit room temperature ferromagnetism [5–7]. However, some references indicated that the ferromagnetism may come from magnetic precipitates or other clusters [8,9]. Fortunately, Cu-doped AlN (Cu:AlN) has the advantage to avoid the problem of magnetic precipitate because neither Cu nor Cu-related precipitates contributes to ferromagnetism [10–12]. Therefore, room temperature ferromagnetic Cu:AlN has been studied extensively, but most of the studies focused on the hysteresis property and an important effect, electrical field induced modulation of magnetization has been ignored. While this effect was firstly demonstrated by applying an external electric field [13], it was reported later that a polarized piezoelectric substrate could modulate the magnetization of anatase $Co:TiO_2$ film [14].

Moreover, the experimental observation of electrical field induced modulation of magnetization in III–V magnetic materials stimulated the study of magnetoelectric coupling [15]. Recently, artificial multiferroics made of ferroelectric (FE) and ferromagnetic (FM) phases exhibit the coexistence of FE and FM ordering, offering the possibility in search of large magnetoelectric coupling, which sparked the study of FM/FE heterostructures, including FM thin film on FE substrates heterostructures. Most of the electrically induced change in magnetization is limited to the FM/FE interface ranging about several nanometers. And the magnetism modulation in FM thin film with the thickness over several hundred nanometers on FE substrate has rarely been studied.

In this work, electrical field induced modulation of magnetization in Cu:AlN thin films with thickness of ~120 nm was studied by using FE substrate. The large difference of average magnetic moment (AMM) in Cu:AlN was observed on different FE substrate. To interpret the phenomenon, bound magnetic polaron (BMP) mechanism [16] for DMI was developed by considering the effect of internal electric field in Cu:AlN.

2. Experiment

Cu:AlN films were deposited by direct current reactive magnetron sputtering using high purity Al disc (99.99%) target attached with several high purity Cu pieces (99.99%). The base vacuum of the chamber was better than 5.0×10^{-5} Pa. During the deposition, the argon and the nitrogen flowed and the working pressure was a sum of argon 0.75 Pa (99.95% purity) and nitrogen 0.15 Pa (99.95% purity). The

* Corresponding authors. Tel.: +86 755 26538974; fax: +86 755 26538735 (J.T. Luo). Tel.: +86 10 62795373; fax: +86 10 62771160 (F. Zeng).

E-mail addresses: luojt07@hotmail.com (J.T. Luo), zengfei@mail.tsinghua.edu.cn (F. Zeng).

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substrate holder was heated at 260 °C during sputtering. The distance between the substrate and the target was about 40 mm. Ten minute pre-sputtering was performed before deposition to get rid of contaminates on the Al target. For comparison, both non-FE Si (1 1 1) and FE single crystalline substrates including $LiTaO_3$ (1 1 0), $LiTaO_3$ (0 2 4), $LiNbO_3$ (1 0 4), and $LiNbO_3$ (0 0 6) were used. The compositions of the films were determined by induced-coupled-plasma atomic emission spectrum and confirmed by X-ray fluorescence. The content of Cu in all the films was measured to be about 4.0 at.%. The thicknesses of all the films were about 120 nm.

The microstructure of the deposited films was characterized by X-ray diffraction (XRD) using Cu Kα radiation. In order to examine the local structure both of Cu dopant and the AlN host, Cu *K*-edge X-ray absorption fine structure (XAFS) and X-ray absorption spectroscopy (XAS) across the N 1s threshold was measured in the fluorescence mode using synchrotron radiation in Shanghai Synchrotron Radiation Facilities. The magnetic properties at room temperature were characterized by a superconducting quantum interference device magnetometer with the applied field parallel to the film plane.

3. Results and discussion

Typical XRD patterns of Cu:AlN film on Si (1 1 1) and $LiTaO_3$ (1 1 0) are shown in Fig. 1. Only the peaks of Cu:AlN film and substrates can be seen in the spectra. On both substrates, (1 0 0) preferred orientation can be found in Cu:AlN films. The standard diffraction peak of AlN (1 0 0) should be located at ~33.32°. However, the diffraction peak of Cu:AlN (1 0 0) on Si (1 1 1) and $LiTaO_3$ are located at 32.94° and 32.74°, respectively. The radius of Cu^{2+} (0.72 Å) is larger than that of Al^{3+} (0.54 Å), therefore, the incorporation of Cu into AlN wurtzite lattice causes the distortion, which results in the shift of AlN (1 0 0) peak to lower angle. The angle of AlN (1 0 0) peak on $LiTaO_3$ (1 1 0) is lower than that on Si (1 1 1) as can be seen in the inset of Fig. 1(a), which is attributed to substrate induced strain of the films on $LiTaO_3$ (1 1 0). The saturation magnetization of Cu:AlN on $LiTaO_3$ (1 1 0) is about three times larger than that on Si (1 1 1) as shown in the inset of Fig. 1(b).

The difference of magnetic moments between Cu:AlN films on Si (1 1 1) and $LiTaO_3$ (1 1 0) indicates that the magnetic properties of Cu:AlN were significantly affected by the substrate. A similar phenomenon was also observed in Co:ZnO [17]. Generally, the magnetism modulation phenomenon on FE substrates is considered to be the well known magnetoelectric coupling effect. For example, the magnetoelectric coupling effect was reported in a $CoFe_2O_4$–$BaTiO_3$ double layer [18]. The action range of exchange interaction of ferromagnetism is quite short. Therefore, in order to explore the origin of the special effect of $LiTaO_3$ (1 1 0) on the magnetic properties of Cu:AlN films, the local structure of Cu and N in AlN host materials was characterized by Cu *K*-edge XAFS and N 1s XAS.

Fig. 2(a) shows the Cu *K*-edge XAFS of Cu:AlN on Si (1 1 1) and $LiTaO_3$ (1 1 0). It is found that the spectra of Cu:AlN films on Si (1 1 1) and $LiTaO_3$ (1 1 0) are nearly the same, indicating that substrate has little effect on the Cu local structure. If only Cu ions contributed to the FM interaction, then the same Cu local structures should lead to the same magnetic moments in the samples on the two different substrates. But this is opposite to the experimental results. Therefore, the origin of FM interaction of Cu:AlN should be related to some other factors besides Cu ions.

Then, what is the structural difference between the two samples on Si (1 1 1) and $LiTaO_3$ (1 1 0)? The difference is revealed by XAS across the N 1s threshold of the samples as shown in Fig. 2(b). Five peaks are apparently observed in the two spectra, denoted as peak A, B, C, D, and E. The pre-edge peak A is corresponding to the e_g and t_{2g} split up-spin and down-spin states of Cu hybridized with the N 2p states. Comparing the XAS spectra of Cu:AlN on Si (1 1 1) and $LiTaO_3$ (110), the pre-edge peak A is the same for AlN films on both substrates, which implies that Cu ion and N ion in AlN films on both substrates are incorporated into one phase [19]. However peaks B, C and D of the AlN films on $LiTaO_3$ (1 1 0) are lower than that on Si (1 1 1), indicating that N 1s to 2*p* transition is damped in the former sample [19]. Since the electron orbital round N is mainly affected by the crystalline field of the host lattice, this damped transition shows that the FE substrate $LiTaO_3$ (1 1 0) modifies the host lattice more significantly than the non-FE substrate Si (1 1 1). This is coincident to the large strain of the films on $LiTaO_3$ (1 1 0) revealed by XRD. Now, it is clear that the larger AMM of Cu:AlN on FE $LiTaO_3$ (1 1 0) is due to the change of AlN host lattice rather than the local structure of Cu.

To further study the effect of FE substrates on the magnetic properties of Cu:AlN, we prepared Cu:AlN films on other FE substrates, including $LiTaO_3$ (0 2 4), $LiNbO_3$ (1 0 4), and $LiNbO_3$ (0 0 6). The magnetization curves of all the samples show hysteresis loops as can be seen in the inset of Fig. 3. Large difference of AMM was observed on different FE substrate. The largest AMM was 2.23 μ_B/Cu on $LiTaO_3$ (1 1 0) and the smallest AMM was 0.11 μ_B/Cu on $LiNbO_3$ (0 0 6). In this work, all the Cu:AlN films showed (1 0 0) preferred orientation and the *c*-axis of Cu:AlN lied in the film plane. Here, we define the angle between the film plane and the *c*-axis of FE substrate as θ (inset of Fig. 2(a)). The FE substrate including $LiNbO_3$ and $LiTaO_3$ are trigonal system, therefore, based on the tangential direction of the FE substrate, the angle (θ) between AlN film plane and the *c*-axis of FE substrate can be estimated as following:

$$\cos\theta = \frac{a^4 d_1 d_2}{V^2}[\sin^2\alpha(h_1h_2 + k_1k_2 + l_1l_2) + (\cos^2\alpha - \cos\alpha)(k_1l_2 + k_2l_1 + l_1h_2 + l_2h_1 + h_1k_2 + h_2k_1)]$$

where *a* for $LiTaO_3$ and $LiNbO_3$ are 5.474 and 5.494 Å, respectively, α for $LiTaO_3$ and $LiNbO_3$ are 56.17° and 55.87°, respectively. d_1 is the

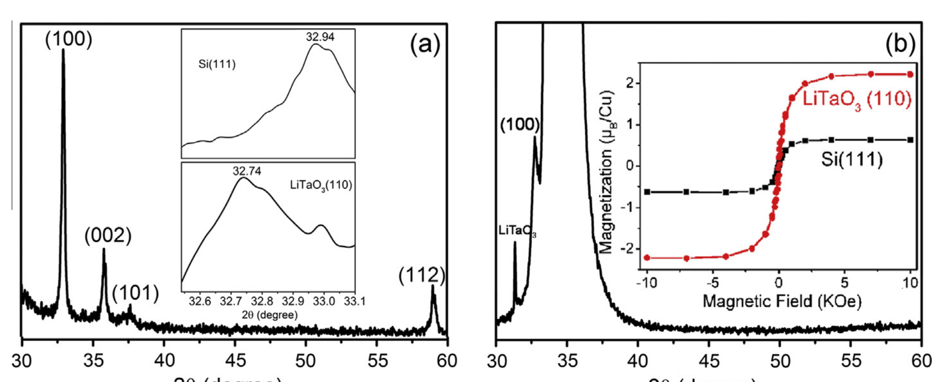

Fig. 1. XRD patterns of Cu:AlN film on (a) Si (1 1 1) and (b) $LiTaO_3$ (1 1 0). The inset of (a) is the enlarged comparative view of two XRD patterns. The inset of (b) is the magnetization versus the magnetic field curves of Cu:AlN films on Si (1 1 1) and $LiTaO_3$ (1 1 0).

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