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## Materials Research Bulletin

journal homepage: www.elsevier.com/locate/matresbu

# Anisotropic magnetoresistance in facing-target reactively sputtered epitaxial $\gamma'$ -Fe<sub>4</sub>N films



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#### ARTICLE INFO

Article history: Received 11 October 2014 Received in revised form 19 January 2015 Accepted 26 January 2015 Available online 28 January 2015

*Keywords:* A. Magnetic materials A. Nitrides B. Epitaxial growth D. Electrical properties

#### ABSTRACT

The negative anisotropic magnetoresistance that comes from the spin-down conduction electrons are observed in facing-target sputtered epitaxial  $\gamma'$ -Fe<sub>4</sub>N films with different film thicknesses, substrates and orientations. Anisotropic magnetoresistance of  $\gamma'$ -Fe<sub>4</sub>N films on LaAlO<sub>3</sub>(100) is larger than other substrates. The magnitude of anisotropic magnetoresistance in (100)-oriented films is always larger than (110)-oriented films. The anisotropic magnetoresistance is intimately related to the magnetocrystalline anisotropy. Fourier coefficient  $C_{2\theta}$  and  $C_{4\theta}$  of cos  $2\theta$  and cos  $4\theta$  terms strongly depend on the measuring temperature. No significant influence of magnetic field on  $C_{2\theta}$  and  $C_{4\theta}$  appears. The marked change of  $C_{2\theta}$  and appearance of  $C_{4\theta}$  at low temperatures are from crystal field splitting of *d* orbitals induced by the lattice change due to the tensile stress from substrate and the compressive stress from decreased temperatures.

A. Magnetic materials; A. Nitrides; B. Epitaxial growth; D. Electrical properties

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#### 1. Introduction

Ferromagnetic iron nitrides have attracted much attention for spintronic devices due to their excellent magnetic properties. Among them,  $\gamma'$ -Fe<sub>4</sub>N was theoretically predicted to have a negative spin polarization of –100% [1]. The high spin polarization was experimentally determined by point-contact Andreev reflection and spin-resolved photoelectron spectroscopy [2,3]. The negative spin polarization was also confirmed via inverse tunnel magnetoresistance and inverse current-induced magnetization switching effect in magnetic tunnel junctions with a  $\gamma'$ -Fe<sub>4</sub>N electrode [4–6]. Because of the negative spin polarization,  $\gamma'$ -Fe<sub>4</sub>N films may show various special magnetoransport properties.

The anisotropic magnetoresistance (AMR) is one of the most fundamental characteristics involving magnetic and electronic transport properties, where the resistivity of the sample is dependent on the relative angle between the magnetization and applied current directions [7–20]. AMR effects have been studied in different ferromagnetic materials, such as Fe [17], Co [17], Ni [17], Fe<sub>3</sub>O<sub>4</sub> [18–20], La<sub>1-x</sub>Ca<sub>x</sub>MnO<sub>3</sub> [21,22], Ga<sub>1-x</sub>Mn<sub>x</sub>As [23]. Generally, AMR ratio is defined as

$$\frac{\Delta\rho}{\rho} = \frac{\rho_{//} - \rho_{\perp}}{\rho_{\perp}} \tag{1}$$

where  $\rho_{ll}(\rho_{\perp})$  represents resistivity for the case of the current parallel (perpendicular) to the magnetization. The negative AMR was observed in the polycrystalline, pseudo-single-crystal and epitaxial  $\nu'$ -Fe<sub>4</sub>N films, and the negative AMR was considered to be clear evidence of spin-down conduction electron in v'-Fe<sub>4</sub>N films [24–28]. However, in the triode-sputtered epitaxial  $\gamma'$ -Fe<sub>4</sub>N films deposited on SrTiO<sub>3</sub>(001) substrates, the positive AMR was reported [29]. The sign of AMR is different from other reports and the origin of difference remains unclear. Recently, Kokado et al. theoretically analyzed the relationship between the AMR sign and *s*–*d* scattering process [30]. The result was drawn that the negative AMR ratio of  $\gamma'$ -Fe<sub>4</sub>N was due to the dominant s-d scattering process of  $s \downarrow \rightarrow d \downarrow [30]$ . In addition, the large stepwise change in the AMR ratio of  $\gamma'$ -Fe<sub>4</sub>N appeared below 50K [24,25]. The appearance of an anomalous  $\cos 4\theta$  term and the enhancement of  $\cos 2\theta$  term at low temperatures were also observed [25,27,28]. Crystal field effect was considered as the possible reason of these behaviors [27]. The reported AMR in the  $\gamma'$ -Fe<sub>4</sub>N films presents different results. So, it is necessary to investigate the AMR in the  $\gamma'$ -Fe<sub>4</sub>N films.

To clarify the physical origin, this work investigates the AMR effect in epitaxial  $\gamma'$ -Fe<sub>4</sub>N films by changing film thickness, substrate and orientation. In the previous work, we have reported the surface morphology, microstructure, magnetic and electrical

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transport properties of epitaxial  $\gamma'$ -Fe<sub>4</sub>N films with different thicknesses on various substrates [31]. In this paper, the epitaxial  $\gamma'$ -Fe<sub>4</sub>N films with different lattice orientations and film thicknesses are fabricated on different substrates by facing-target reactive sputtering. AMR effect of epitaxial  $\gamma'$ -Fe<sub>4</sub>N films has been investigated in details.

#### 2. Experimental details

Epitaxial  $\gamma'$ -Fe<sub>4</sub>N films with different thicknesses were fabricated on MgO(100), SrTiO<sub>3</sub>(100), SrTiO<sub>3</sub>(110) and LaAlO<sub>3</sub> (100) substrates by a DC reactive facing-target sputtering method. MgO(100), SrTiO<sub>3</sub>(100), SrTiO<sub>3</sub>(110) and LaAlO<sub>3</sub>(100) substrates were defined as MgO(100), STO(100), STO(110) and LAO(100), respectively. Details for the epitaxial growth of  $\gamma'$ -Fe<sub>4</sub>N were referred to our previous work [31]. For comparison, the polycrystalline  $\gamma'$ -Fe<sub>4</sub>N films were also grown on glass substrate. The film thickness was determined using a Dektak 6M surface profiler, and confirmed by transmission electron microscopy (TEM). The structure was analyzed by X-ray diffraction (XRD) with Cu Ka radiation (wavelength 1.5406 Å) and high-resolution TEM (HRTEM). The AMR effect was measured with a standard four-terminal method by a Quantum Design physical property measurement system (PPMS-9) at temperatures ranging from 5 to 300 K. The applied magnetic field was parallel to the film plane. At 5 and 300K, different magnetic fields from 0.2 to 50 kOe were applied for AMR measurements. At other selected temperatures, a 10-kOe field was applied.

#### 3. Results and discussion

Fig. 1a shows the XRD  $\theta$ -2 $\theta$  pattern of epitaxial  $\gamma'$ -Fe<sub>4</sub>N films on MgO(100). Except for the diffraction peaks from face-centered cubic MgO(200) and (400), only peaks from  $\gamma'$ -Fe<sub>4</sub>N(100) and

(200) that located at 23.3° and 47.9° appear. No other peaks from  $\gamma'$ -Fe<sub>4</sub>N can be observed, suggesting that the film grows with a preferred (100) orientation. Fig. 1d gives the XRD  $\theta$ -2 $\theta$  pattern of epitaxial  $\gamma'$ -Fe<sub>4</sub>N films on STO(110). Only a peak at 70.1° from  $\gamma'$ -Fe<sub>4</sub>N(220) is observed, showing the film grows with a preferred (110) orientation. The preferred orientation can not completely account for the epitaxial growth of the films. In general,  $\phi$  scan and pole figure are used to characterize the epitaxial structure by identifying the symmetry of a diffraction peak. For confirming the preferred oriented films are epitaxial, X-ray  $\phi$  scan is performed. X-ray is collected at  $2\theta = 41.22^{\circ}$ , where no peaks from MgO and STO substrates appear, and only a peak from  $\gamma'$ -Fe<sub>4</sub>N(111) is detected. Fig. 1b and e shows the X-ray  $\phi$  scan of epitaxial  $\gamma'$ -Fe<sub>4</sub>N films on MgO(100) and STO(110). For  $\gamma'$ -Fe<sub>4</sub>N films on MgO(100), sharp 4-fold symmetric diffraction peaks of  $\gamma'$ -Fe<sub>4</sub>N(111) with identical intervals appear. According to XRD results, the epitaxial relationship of  $\gamma'$ -Fe<sub>4</sub>N films on MgO(100) can be confirmed as  $\gamma'$ -Fe<sub>4</sub>N (100) [001]|| MgO(100) [001]. For  $\gamma'$ -Fe<sub>4</sub>N films on STO(110), 2-fold symmetric peaks with identical intervals appear, suggesting that epitaxial relationship of  $\gamma'$ -Fe<sub>4</sub>N films on STO(110) is  $\gamma'$ -Fe<sub>4</sub>N (110) [110]]| STO(110) [110]. The above X-ray  $\theta$ -2 $\theta$  and  $\phi$  scan suggest that the  $\gamma'$ -Fe<sub>4</sub>N films on MgO(100) and STO(110) are epitaxial and cubic. Fig. 1c and f gives the X-ray pole figures of the  $\gamma'$ -Fe<sub>4</sub>N films on MgO(100) and STO(110), showing cubic structure and in-plane rotational symmetry of  $\gamma'$ -Fe<sub>4</sub>N films. On the (111) plane, the (100)-oriented films have four symmetrical diffraction peaks, while (110)-oriented films show two symmetrical diffraction peaks. These relevant strong diffraction peaks further suggest that all the films are fully epitaxial.

Fig. 2 shows the cross-sectional HRTEM images of epitaxial  $\gamma'$ -Fe<sub>4</sub>N films on MgO(100) and STO(110). In Fig. 2a, the HRTEM image at  $\gamma'$ -Fe<sub>4</sub>N/MgO(100) interface shows the epitaxial growth of the film even though the lattice mismatch rate is 10%, which further confirms the X-ray  $\theta$ -2 $\theta$ ,  $\phi$  scan and pole figures results.



**Fig. 1.** X-ray  $\theta$ -2 $\theta$ , $\phi$  scan and pole figure of epitaxial  $\gamma'$ -Fe<sub>4</sub>N films on (a)–(c) MgO(100) and (d)–(f) STO(110) substrates.

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