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（博士課程）
Doctoral Program

論 文 要 約

THESIS OUTLINE

系・コース： Department of, Graduate major in	材料	系 コース	
学生氏名： Student's Name	Xu Xinxing		

申請学位（専攻分野）： Academic Degree Requested	博士 Doctor of （ 工学 ）
指導教員（主）： Academic Supervisor(main)	中村 吉男
指導教員（副）： Academic Supervisor(sub)	史蹟

Study on the structural and magnetic properties of FeCoN thin films

FeCo based thin films are attractive functional materials due to their large saturation magnetization, high permeability and high curie temperature properties, which are widely used in electronic devices, such as magnetic recording head and planar inductor. However, the coercivity of the FeCo thin film is relatively large, and the magnetostriction is strong, the electrical resistivity is low, which limits its application. Nitrogen doping is supposed to resolve these problems. In this work, we prepared a series of FeCoN thin films sandwiched by AlN layer using direct current reactive magnetron sputtering method. The nitrogen content in the FeCoN thin films is controlled by adjusting the N_2 gas flow ratio. First, the formation of iron cobalt nitrides, and the effect of nitrogen contents on film structure and film properties are systematically investigated. Second, in order to decrease the coercivity of FeCoN films at high temperature, the exchange coupling behavior between soft/hard FeCo(N) layer with different nitrogen contents are studied. Third, an abnormal angle-dependent inverted hysteresis loop in the FeCoN films are observed, and the corresponding mechanisms are discussed.

The organizations and contents of the thesis are summarized as follows:

Chapter 1 Introduction: The research background of the FeCoN films and some fundamental concepts of magnetism are introduced. The objectives of this study and the organization of the thesis are included.

Chapter 2 Preparation and characterizations of FeCoN magnetic thin films: The

preparation of FeCoN thin films is described in detail. The characterization techniques used in the experiments are introduced. The typical results of film microstructure, magnetization hysteresis loops, element depth distribution, film electric resistivity and magnetostriction are shown. In addition, glass/AlN/FeCo(N)/AlN is selected as the optimized film structure.

Chapter 3 Evolution of microstructure and magnetic properties of FeCoN films sandwiched by AlN layers: In this part, a series of FeCoN films sandwiched by AlN layers have been deposited using reactive sputtering method on glass substrate. The nitrogen content in the FeCoN film is controlled by adjusting the nitrogen gas flow ratio of the Ar/N_2 gas mixture, and the nitrogen gas flow ratio ($N_2/Ar+N_2$) is set from 0%-50%. The formation of iron cobalt nitrides are studied. **Figure 1** shows the XRD results of as-deposited FeCoN films with different nitrogen gas flow ratios. With the increases of nitrogen content, the main phase of FeCoN films transforms from polycrystalline α-FeCo(N) to amorphous, polycrystalline ε-$(Fe,Co)_3N$ and ξ-$(Fe,Co)_2N$. The magnetic properties and electric resistivity results of FeCoN films are summarized in **Fig.2(b)**. It can be found that the saturation magnetization of films decreases with increases of nitrogen content, which is related to the phase transition. The coercivity of the films initially decreases with increasing N_2 flow ratio, reaching the lowest value of 8-10 Oe at N_2 flow ratio around 20% to 30%. In addition, the resistivity of films increases with increases of nitrogen contents. **Figure 3** shows the microstructure of FeCoN films. It revealed that the main phase of the amorphous films (20% N_2) is γ'-$(Fe,Co)_4N$ phase based on the corresponding SAED patterns. The low coercivity of amorphous films is attributed to the fine particles according to the Random anisotropies model.

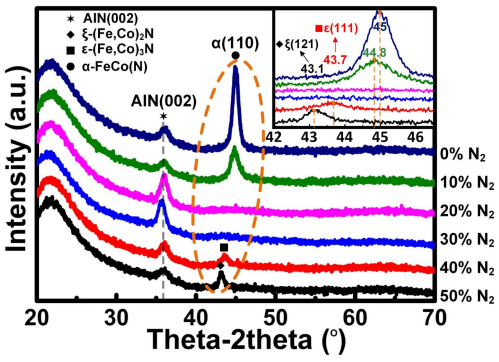

Fig.1 XRD results of as-deposited films with FeCoN layer sputtered at different N2 /(Ar+N2) ratios.

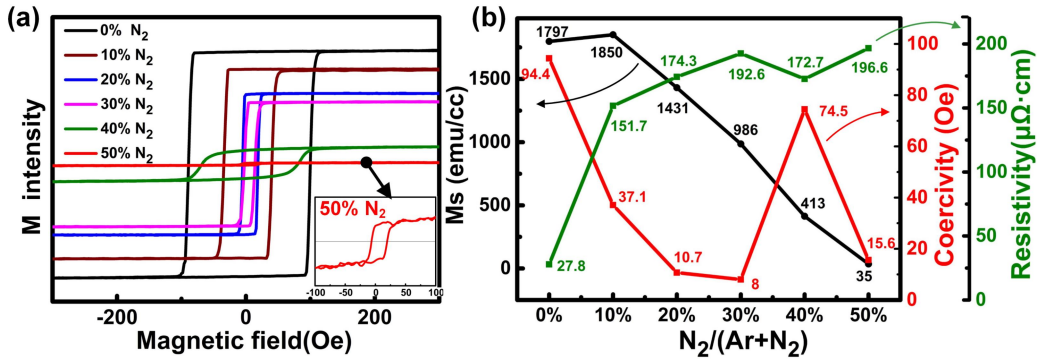

Fig.2 (a) Magnetic hysteresis loop and (b)Changes in Ms, Hc and ρ of the as-deposited Fe-Co-N films.

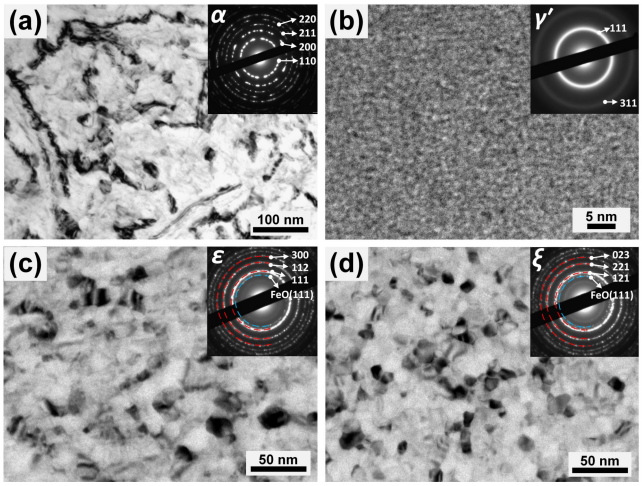

Fig.3 TEM images and the corresponding SAED patterns of 20 nm FeCoN single layer deposited at (a) 0% N_2, (b) 20% N_2, (c)40% N_2, and (d)50% N_2 flow ratio.

Systematic annealing experiments have also been carried out from 100 °C to 600 °C for FeCoN films with different N_2 flow ratios. The magnetic property results are shown in **Fig. 4**. FeCo film shows good thermal stability since its saturation magnetization and coercivity basically unchanged with increase of films. However, for 20-40% N_2 films, the *Ms* value suddenly increases after annealing at 350 °C for 1 hour, which is related to the release of nitrogen. The coercivity of amorphous films (20-30% N_2) shows strong annealing temperature dependence, and it increases at first and then decreases. The changes in film stress and the formation of new phase during annealing account for the variation of coercivity.

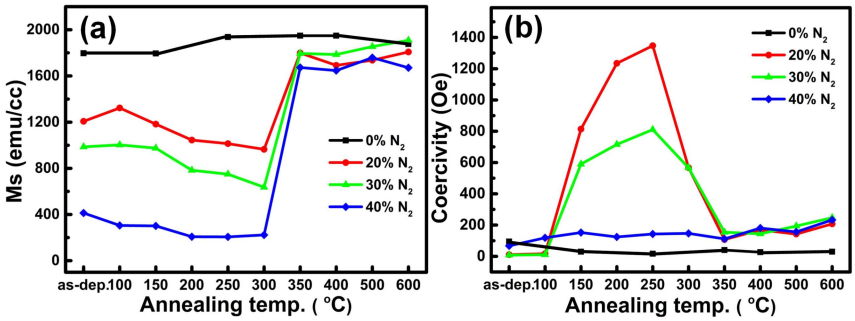

Fig.4 Annealing temperature dependence of (a) *Ms* and (b) *Hc* in 0% N_2 and 20% - 40% N_2 films.

Chapter 4 Investigation of the exchange coupling behavior of the hard/soft FeCoN thin films: To decrease the large coercivity of amorphous-like FeCoN films at high annealing temperature, we have investigated the exchange coupling behavior between the FeCo layer and the FeCoN layer. The FeCoN layer is deposited with 20-30% N_2 gas flow ratio, and the film structure includes FeCo/FeCoN bilayer and FeCo/AlN/FeCoN trilayer structures. The magnetic properties of these multilayer films are characterized. **Figure 5** shows the magnetic property results of bilayer structure annealed at 200-250 °C/1 h with different FeCo layer thickness. It can be found that The coercivity of annealed films largely decreases as the FeCo layer thickness is greater than 3 nm.

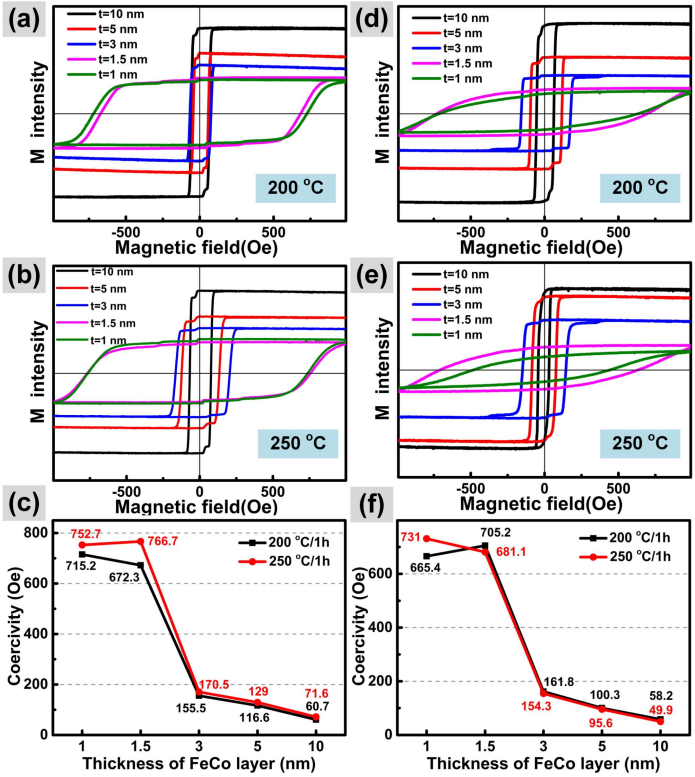

Fig.5 VSM results of the 20% N_2 bilayer films annealed at (a) 200 °C and (b) 250 °C for 1 hour, (c) summarized coercivity results. (d-f) The corresponding VSM results for 30 % N_2 bilayer films.

Figure 6 shows the magnetic properties of the trilayer films with the different thickness of the AlN spacer layer. For the AlN spacer layer thickness is 5~10 nm, FeCo layer and FeCoN layer are decoupled according to the two-stage M-H curves. Exchange spring-like hysteresis loops can be observed with AlN thickness of 1~2 nm. When the AlN spacer layer thickness decreases to 0.5 nm, two layers are strongly exchange coupled.

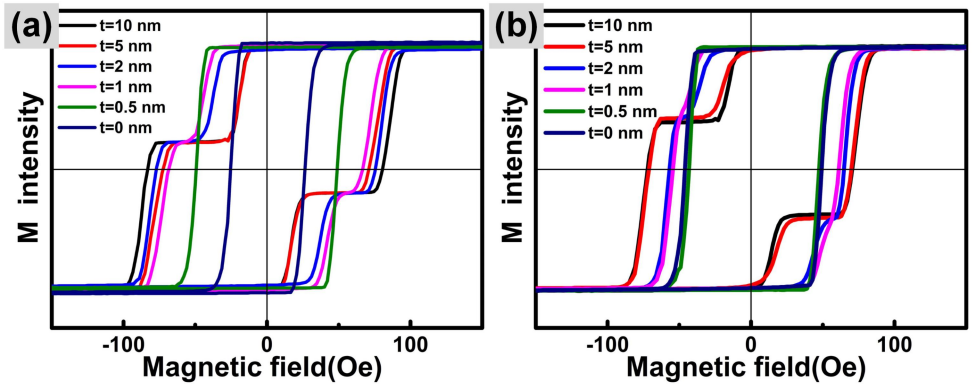

Fig.6 Magnetic properties of the FeCo (10 nm)/ AlN (t nm)/FeCoN (10 nm) trilayer films with the FeCoN layer deposited with (a) 20% N_2, (b) 30% N_2 gas flow ratios.

Chapter 5 Observation of angular dependent inverted magnetic hysteresis loop in FeCoN thin films with two competitive magnetic anisotropies: an abnormal phenomenon of inverted hysteresis loop (IHL) has been observed when we measuring the out-of-plane magnetic hysteresis loop of FeCoN amorphous films(20% N_2) using VSM equipment. In addition, IHL shows angle dependence. The sample plane deviates from the center plane by a small angle during VSM measurement, causing the differences in magnetic properties when rotate the sample around film normal direction. **Figure 7** shows the anisotropies distribution of the FeCoN film. It can be found that uniaxial planar-anisotropy and biaxial anisotropies co-exist in the FeCoN film. The uniaxial anisotropy of the FeCo film is induced by the sputtering field, and the bi-axis anisotropy is originated from the inclined deposition in the facing-targets-sputtering system as shown in **Fig.7(d)**.

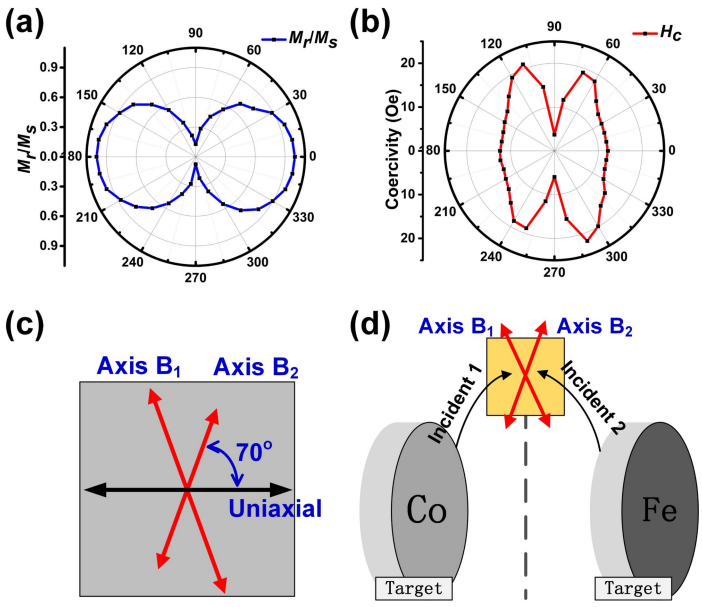

Fig. 7 Angular-dependent (a) normalized remanence *Mr*/*Ms*, (b) coercivity field *Hc* of FeCoN films. Schematic of (c) anisotropy distribution in the FeCoN film, (d) the origin of bi-axes B_1 and B_2 that generated from the deposition.

The out-of-plane magnetic hysteresis loops are also measured by VSM at intervals of 10 degree as shown in **Fig.8**. We found the normal hysteresis loop and inverted hysteresis loops periodically appear in the specific angle range according to the M_r results in **Fig.8(b)**, and the typical *M*-*H* curves at φ=0° and 90° are shown in **Fig.8(c)** and **(d)**, respectively. The inverted hysteresis loop is caused by the anti-ferromagnetic coupling of two different magnetization components at low applied field, and the two components originate from the inhomogeneous compositions in the film. The angular dependence is determined by the rotation of in-plane magnetization component under the competition of the anisotropies.

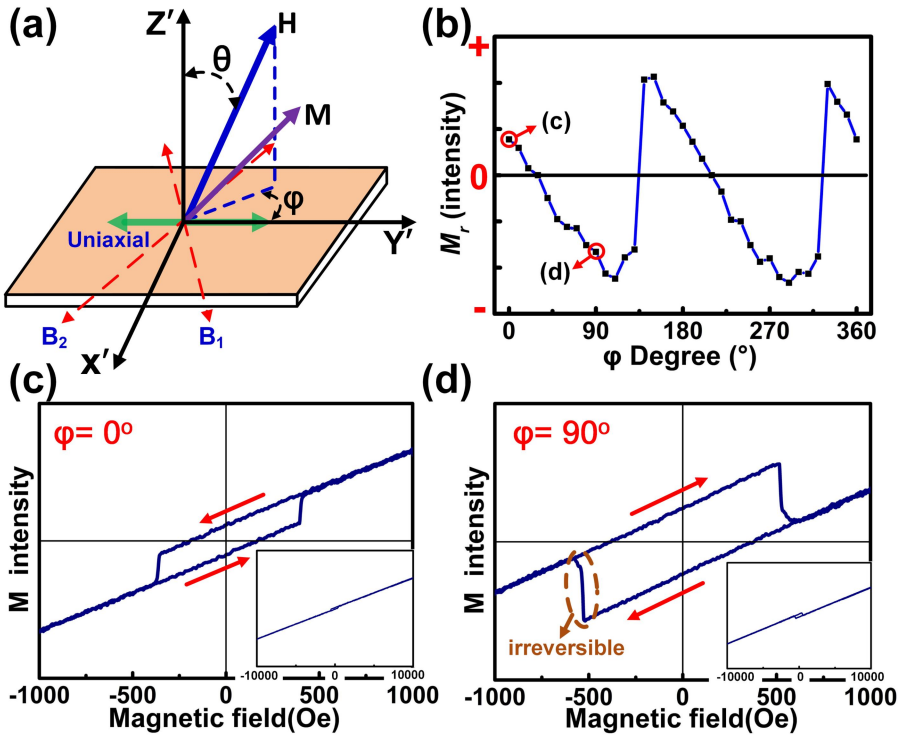

Fig. 8 (a) Schematic of applied field and film anisotropy, φ is referred to the angle between the projection of applied field H and uniaxial direction in the film plane, θ is defined as the angle between applied field and film normal. (b) Out-of-plane remanent magnetization of FeCoN layer as a function of applied field projection angle φ. Typical out-of-plane hysteresis loop at (c)φ=0° and (d) φ=90°, the inserted figures are the corresponding M-H loops measured at 10000 Oe.

Chapter 6 General conclusions: The general conclusions of the thesis are given.