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Letter

Microstructure and magnetic properties of iron nitride granular thin films obtained by oblique RF reactive sputtering

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ABSTRACT

A series of iron nitride nano-crystalline granular thin films with different thickness and different angle of oblique sputtering have been fabricated on Si(100) by oblique RF reactive sputtering. The microstructure and magnetic properties of these samples have been investigated. Iron nitride thin films are composed of irregular spherical particles of 3–18 nm diameters with disordered face-centered-cubic structure. These particles are embedded in the amorphous iron nitride matrix with average spacing of $\sim$3 nm, and the films exhibit an in-plane uniaxial magnetic anisotropy. The results show that both thickness of iron nitride thin films and oblique angle have important effects on the magnetic properties of the films and magnetic resonant frequency of the films can also be tailored by these two factors. The in-plane uniaxial magnetic anisotropy fields of samples deposited for 30 min can be adjusted from 156 to 360 Oe by increasing the oblique angle from 3° to 32°. As a consequence, the resonance frequencies of films continuously increase from 2.87 to 6.05 GHz.

1. Introduction

Recently, soft magnetic thin film materials have attracted much attention [1,2] and are strongly desired for high-frequency applications such as soft magnetic underlayers in recording media [3], high-density recording head cores [4,5], planar inductors, and film transformers in integrated circuits [1,6]. The basic demands for soft magnetic films operated in a few MHz to GHz range include high resistivity (ρ), high permeability (μ), high saturation magnetization ($4\pi Ms$) and appropriate large anisotropy field (H_k) so as to effectively suppress eddy current loss and to achieve high ferromagnetic resonance (FMR) frequency f_r (which determines the cut-off frequency for the high frequency application). Granular films consisting of nano-ferromagnetic metallic particles distributed uniformly in an insulator matrix are one of the best candidates for satisfying above demands, because these films have the advantages of high M_s and high μ of magnetic metals as well as the high resistivity of insulators.

Iron nitride thin films have been widely studied due to their surface hardness, high corrosion/wear resistance, high thermal stability and excellent magnetic properties [7–11]. Iron nitrides are known to form various phases, such as α-Fe(N), γ'-Fe_4N (fcc), ε-$Fe_{2\text{-}3}N$ (hcp) and α''-$Fe_{16}N_2$ (bct) which show ferromagnetic properties [12–14]. In addition, paramagnetic ξ-Fe_2N and nonmagnetic iron nitride phases also have received much attention in these years [15,16]. Compared with the crystalline structures, amorphous magnetic iron nitride thin films had also cause of concern because of the isotropic nature of the amorphous structure [17–19].

In this work, a series of different iron nitride granular thin films, which have a fcc iron nitride ferromagnetic particles embedded in amorphous iron nitride matrix, were fabricated by oblique radio frequency (RF) reactive sputtering. We chose the method of oblique deposition [20,21] to acquire a high in-plane uniaxial magnetic anisotropy (IPUMA) field H_k. According to the formula [22] $f_r = (\gamma/\pi)\sqrt{H_k(4\pi M_s + H_k)}$, where γ is the gyromagnetic ratio and M_s is the saturation magnetization, the resonance frequency of iron nitride thin films can be tuned by IPUMA field H_k which was adjusted by oblique deposition angle θ and deposition time t.

2. Experimental

Iron nitride thin films were prepared by RF reactive magnetron sputtering on Si(100) substrates at room temperature with a 3-in. diameter iron target (purity 99.99%) in the N_2 and Ar gas mixture. The N_2 and Ar flow rates were accurately controlled by a mass flow controller, where the flow rate ratio of $N_2/N_2 + Ar$ was fixed at 5%. The total pressure of $N_2 + Ar$ mixture was 0.5 Pa. The base pressure was below 3×10^{-5} Pa and the sputtering power was kept constant at 110 W. The distance between the target and the substrate was about 110 mm. The diagram of experimental facility is shown in Fig. 1. During deposition, the target of Fe faced the substrate with an incident angle θ without an external magnetic field. Silicon substrates were cut into rectangles of 2.5 cm*7.5 cm and its long side parallel to the sputtering direction [23]. An IPUMA can be acquired by inclined incidence of the sputtering and the direction of in-plane easy axis (EA) is perpendicular to the incidence direction of the sputtered particles.

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The thickness of the film was determined by a surface profilometer (VEECO Dektak8 ADP-8). The microstructures of the films were examined by a FEI Tecnai G2 F30 transmission electron microscope (TEM) and X-ray diffraction (XRD) with Cu Kα radiation. The static magnetic properties of iron nitride thin films were measured using vibrating sample magnetometer (Lakeshore model 7304). The permeability spectra of the samples were obtained by the one-terminal microstrip transmission-line method [24], from 100 MHz to 9 GHz. All the above measurements on as-deposited samples were performed at room temperature. f_r and damping parameter α were determined by fitting the permeability spectrum with Landau-Lifshitz-Gilbert equation [25].

3. Results and discussion

Fig. 2 shows the XRD patterns of iron nitride thin films with sputtering time t varying from 10 to 30 min. No evident diffraction peaks from the iron nitride phase or Fe phase are observed for all samples, which suggests as-deposited samples may include iron nitride amorphous phase. An evident broad bump between $2\theta = 40°$ and $2\theta==45°$ can be observed with increasing t from 20 min to 30 min. It indicates that the nano-crystalline iron nitride may have appeared in the samples except the amorphous iron nitride, and the results also suggest that small granular particles may have existed [26].

To reveal the structure of these films more clearly, we performed Transmission Electron Microscopy (TEM) observation on our iron nitride thin films produced at $t = 30$ min. TEM is an ideal tool for the investigation of the size and shape distributions of the particles in a granular sample [26]. The TEM images shown in Fig. 3(a and b) indicate that irregular randomly oriented fcc $FeN_{0.056}$ fine particles uniformly disperse in the amorphous iron nitride matrix according to the standard powder diffraction files (PDF752129). The nano-particles have a very small diameter of 3–18 nm and are separated by mean distance of ~3 nm from each other. The HRTEM image in Fig. 3(c) shows the crystallized $FeN_{0.056}$ grains with the (1 1 1) lattice spacing of ~2.082 Å. Fig. 3(d) exhibits the diffraction rings from (1 1 1), (2 0 0), (2 2 0) and (3 1 1) planes of fcc $FeN_{0.056}$ (PDF752129), and there is no other diffraction rings corresponding to crystalline iron nitride except $FeN_{0.056}$, which indicates that iron nitride matrix is amorphous.

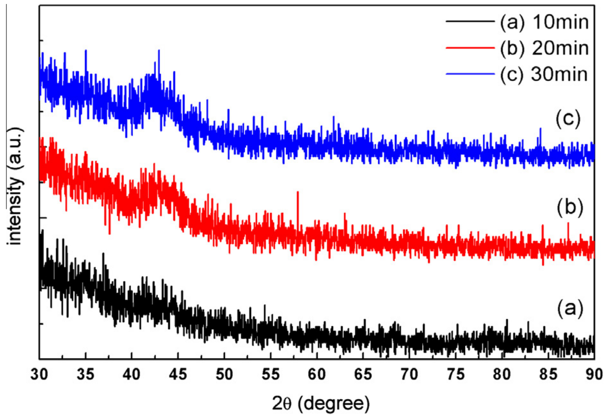

Fig. 2. The XRD pattern of the iron nitride thin films with different deposition time t of 10 min (a), 20 min (b), and 30 min (c).

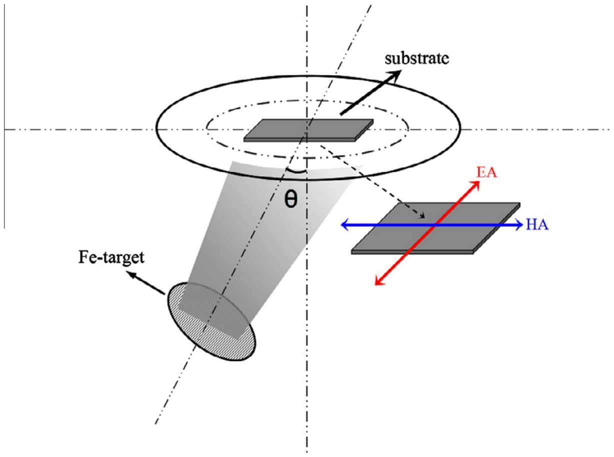

Fig. 1. The schematic illustration of oblique sputtering system.

According to the Herzer's statement [27], if the average size D of magnetic particles and the distance S between particles become smaller than a characteristic length, namely, the exchange length L_{ex}, the exchange interaction between particles will tend to align the magnetic moments of neighboring particles parallel and compete with magneto-crystalline anisotropy and the demagnetization effect of individual particles. As a result, the effective magnetic anisotropy of materials is reduced significantly, which leads to the decrease in coercivity, and then good soft magnetic properties are obtained. For our samples discussed above, it can be seen that the exchange length $L_{ex} > (D + S)$. Hence, exchange coupling can be realized in these films, which is responsible for the good soft magnetic properties.

The static magnetic properties of $t = 30$ min iron nitride thin films with different sputtering angles θ from 3° to 32° are investigated. The in-plane hysteresis loops of iron nitrides thin films with $\theta = 3°$ and $\theta = 32°$ are shown in Fig. 4(a and b), respectively. It has been reported that the thickness of the films deposited by this method decreases with increasing the oblique angle at the same deposition time t [23]. That means, in our case, the film at the oblique angle of 3° (about 126 nm) is thicker than that of 32° (about 70 nm). It can be deduced that the change of hysteresis loop from Fig. 4(b–a) can be attributed to the increase in the thickness of the film [28]. For all samples except $\theta = 3°$ the loops along the easy axis (HA) are basically a rectangle with the remanent magnetization ratio Mr/Ms~1(not shown here for $\theta = 11°, 19°, 26°$). The calculated magnetic anisotropy fields corresponding to Fig. 4(a and b) are 156 Oe and 360 Oe, respectively. The high frequency performances of both samples are shown in Fig. 4(c and d), where

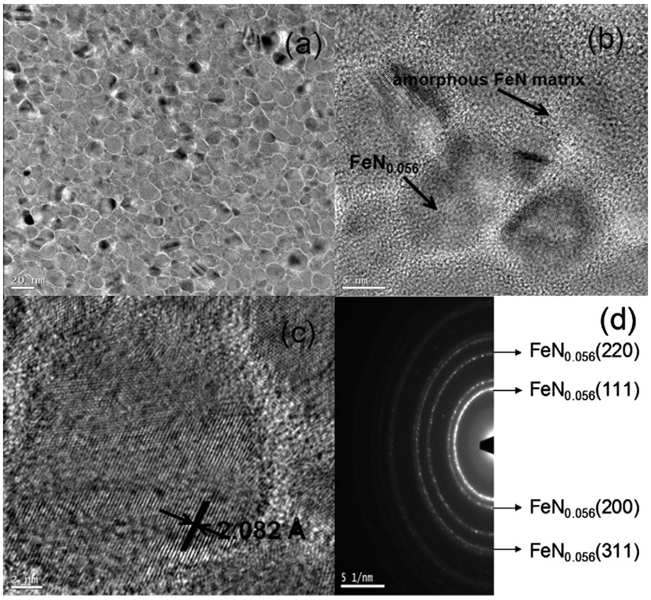

Fig. 3. TEM image (a), HRTEM image (b) and (c), SAED pattern (d) of iron nitride film with deposition time $t = 30$ min.