Ultrahigh-frequency ferromagnetic properties of FeCoHf films deposited by gradient sputtering

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Nanocrystalline FeCoHf films with Hf composition gradient were prepared by gradient sputtering method at room temperature. A uniaxial magnetic anisotropy with high anisotropy field H_k up to 547 Oe was achieved after magnetic annealing with external field along the gradient direction. Ultrahigh ferromagnetic resonance frequency over 7 GHz was obtained in magnetic annealed gradient sputtered films. The origin of ultrahigh ferromagnetic properties in gradient sputtered films is discussed. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2889447]

Recently, there has been an increasing demand for magnetic films in high-frequency applications including magnetic recording write heads, soft underlayers for perpendicular media, and thin film wireless inductor cores.[1–10] Ferromagnetic thin films covered on the microinductors give rise to an increase of inductance. Therefore, the magnetic microinductor is an effective approach to improve the property of inductors and to miniaturize the electromagnetic devices.[11–14] Operating frequency has reached gigahertz band to increase the data transition rate in modern technology, e.g., 2.4 and 5.8 GHz are typical frequencies for bluetooth technology.[14] FeCo alloys are promising candidates due to their high saturation magnetization. However, it is difficult to prepare FeCo ferromagnetic films with resonance frequency in excess of 3 GHz due to a relatively low resistivity and anisotropy field.[15–17] In this letter, a gradient sputtering (GS) method was employed to enhance the anisotropy field of FeCoHf films. Very high anisotropy field and resonance frequency were achieved in GS films.

A rf GS device was designed as illustrated in Fig. 1(a).[18] Si (100) substrates (25×10 mm^2) were put on a sample turn plate with the length along the radial direction. The main target $Fe_{70}Co_{30}$ was faced to the geometric center of samples, while the geometric center of the doping target Hf was outside of the samples. Thus, a geometrically uniform $Fe_{70}Co_{30}$ composition is doped with a radially increasing Hf content. The result is a Hf composition gradient (HCG) along the *AR* direction [see the inset in Fig. 1(a)]. The substrate end close to the doping target is referred to as the *S* point, and the opposite end as the *E* point. The test position was defined as the distance from the test point to *E*. The base pressure was less than 3×10^{-6} Torr, and the working pressure was 2.8 mTorr with an Ar floating rate of 20 SCCM (SCCM denotes cubic centimeter per minute at STP). GS films with average thickness of 100 nm were deposited at rf powers of 60 W for both targets for 30 min. The GS film was annealed at 350 °C for 1 h in the magnetic field of 1.2 kG along *AR* (the composition gradient direction). The microstructure of the films was characterized by a microarea x-ray diffractometer (MA-XRD). The composition distribution of GS films was detected with a field emission electron probe microanalyzer (FE-EPMA). The magnetic properties were measured by a vibrating sample magnetometer. The magnetic domain structure was observed by a magnetic force microscope (MFM). The high-frequency ferromagnetic properties (HF-FMPs) of the magnetic films were measured by a permeameter with a maximum frequency of 9 GHz.[19] The stress distribution of the magnetic films was detected by an optical deflectometer.[20]

FE-EPMA results, shown in Fig. 1(b), reveal that the Hf composition increases from 8.66 at position *E* to 12.36 at position *S*, with the Fe/Co ratio of around 2.73 for all the test positions. This fact demonstrates a gradient composition distribution for Hf and a homogeneous one for Fe and Co in films along *AR*. On the other hand, the compositions of Fe, Co, and Hf were all homogeneous for a test line along *PR*. These composition distribution characteristics were also verified by MA-XRD results. As shown in Figs. 1(c) and 1(d), distinct left shift of the (110) peak was observed for test positions from five to one along *AR*. However, the peak shift was not observed for test positions along *PR* (e.g., 6-3-7). The left shift of XRD peaks is attributed to the entrance of

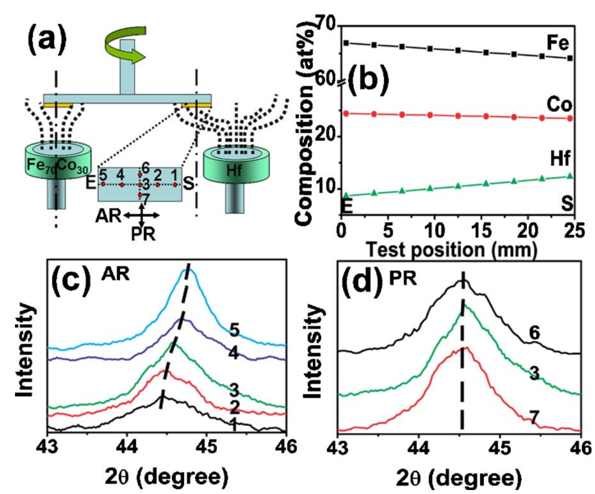

FIG. 1. (Color online) (a) Schematic drawing of GS device; (b) Composition distribution along *AR*; [(c) and (d)] (110) peaks of GS films along *AR* and *PR* directions, respectively. The numbers in (c) and (d) stand for the test positions shown in the inset of (a).

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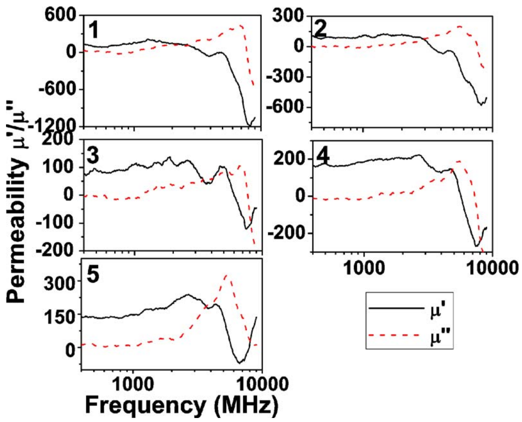

FIG. 2. (Color online) Frequency dependence of permeability along *AR* at various positions.

Hf into FeCo alloy lattice since the atomic radius of Hf (2.16 Å) is much larger than that of Fe (1.72 Å) and Co (1.67 Å). FE-EPMA and MA-XRD results reveal that the composition and microstructure along the *AR* direction are gradually changed, while those along *PR* for individual test lines (e.g., 6-3-7) are homogeneous. In addition, the crystal size of the GS films was estimated by the Scherrer equation as about 6 nm, which is small enough to obtain good soft magnetic properties.[21]

A very high ferromagnetic resonance frequency f_{FMR} (up to 7.18 GHz) and cutoff frequencies f_{cutoff} (between 4.35 and 4.93 GHz) were achieved in magnetic annealed GS films, as shown in Fig. 2 and Table I. It is significant that the metallic FeCoHf GS films exhibit better HF-FMPs compared with the conventional metallic ferromagnetic films, suggesting that the promising HF-FMPs are related to the HCG.

The position dependences of hysteresis loop, stress, and magnetic domain were investigated for exploring the origin of HF-FMPs in GS films. As illustrated in Figs. 3(a) and 3(b), hysteresis loops along *PR* are similar at various positions, while evident enhancement of anisotropy field H_k for positions from five to one along *AR* was observed, suggesting a distinct uniaxial magnetic anisotropy (UMA) with a hard axis along *AR*. The position dependence of magnetic properties and resonance frequency of the GS films are summarized in Table I. The calculated f_{FMR} fits well the experimental one. Figure 3(c) reveals that the compressive stress along *AR* increases with the increase of Hf content. The increase of stress can be attributed to the partial substitution of larger atoms of Hf for small atoms Fe and Co.

When the direction of the annealing magnetic field is parallel to that of the HCG, the magnetic annealing favors the enhancement of H_k in GS films. If they are perpendicular, the magnetic annealing deteriorates the HF-FMPs. Therefore,

TABLE I. Magnetic and HF-FMPs of FeCoHf GS films at various positions.

Test position	M_s (kG)	H_k (Oe)	f_{cutoff} (GHz)	f_{FMR} (GHz) Expt.	f_{FMR} (GHz) Calc,
1	18.01	547	4.93	7.18	8.79
2	18.41	407	4.79	7.08	7.66
3	19.72	282	4.95	6.70	6.60
4	20.17	191	4.65	5.47	5. 50
5	22.11	167	4.35	5.34	5.38

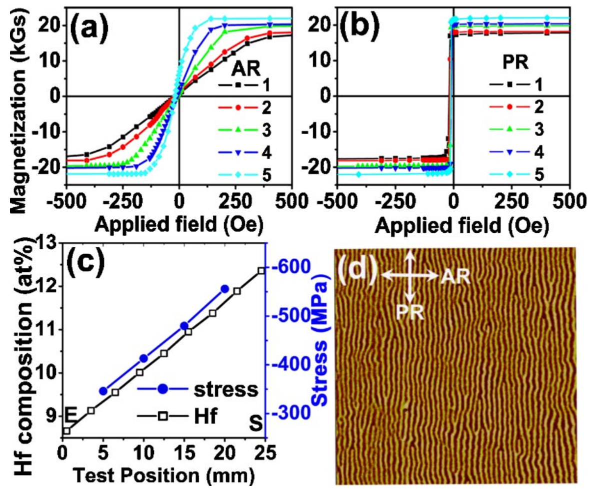

FIG. 3. (Color online) Position dependence of half-hysteresis loops along the (a) *AR* and (b) *PR*, and (c) Hf composition and stress; (d) *a* representative MFM image for the test point three (10×10 μm^2).

the ultrahigh H_k in GS films can be attributed to two effective mechanisms: (1) induced by uniaxial stress and (2) atoms redirection. (1) According to the theory of ferromagnetism, magnetostriction energy is expressed as

$$E = -\frac{3}{2}\lambda_S \sigma \cos^2 \theta. \qquad (1)$$

Here, λ_S is the saturation magnetostriction coefficient and σ is the stress. Positive and negative signs of stress are taken for tensile and compressive stresses, respectively. θ is the angle between stress and magnetization. Based on Eq. (1), for a film with a positive λ_S, the compressive stress results in the arrangement of magnetic moments perpendicular to the compressive stress direction, and vice versa.[22,23] Generally, the intrinsic stress is randomly dispersed in the sample, giving rise to a high damping constant, which goes against the enhancement of the resonance frequency. However, if the intrinsic stress orientation is arranged along one direction, a UMA will be formed, accompanied by an enhancement of HF-FMPs. For the investigated GS films, a uniaxial stress along *AR* is induced by the HCG. Due to the FeCoHf GS films with a positive λ_S and compressive stress direction along *AR*, the magnetic moments are preferentially arranged parallel to the *PR* direction,[24,25] as demonstrated by MFM observation. Stripe domains were parallel to *PR* for all test positions. The representative MFM image at test position three was shown in Fig. 3(d). The domain structure reveals that the magnetic moments in the case of GS films arranged along the *PR* direction, giving rise to magnetically hard axis along *AR*. The formation of UMA induced by magnetic field annealing can be caused by atoms rearrangement over the possible sites in a Boltzmann distribution.[26,27] The Hf composition dependence of H_k along *AR* suggests that Hf atoms are responsible for the UMA. Since the magnetic field and HCG are parallel, the magnetic annealing is further prone to form UMA in GS films than in random dispersed films. On the contrary, no UMA in Hf-free FeCo films implies that the UMA results from HCG. For cosputtered FeCoHf film with homogeneous Hf composition, small H_k (<100 Oe), and deteriorated HF-FMPs with f_{FMR} (<2.5 GHz) demonstrate that the HCG plays an important role to improve HF-FMPs.

In summary, the ultra-HF-FMPs of FeCoHf GS films were obtained under the condition of magnetic annealing with magnetic field along the Hf gradient direction, and can be attributed to the high H_k induced by uniaxial stress and magnetic field. GS is an effective method to prepare high-frequency ferromagnetic films more than conventional cosputtering.

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