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Soft magnetism and microwave magnetic properties of Fe-Co-Hf films deposited by composition gradient sputtering

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A composition gradient sputtering method was employed to deposit a series of $(Fe_xCo_{1-x})_{1-y}Hf_y$ alloy films with different Fe/Co atomic ratios and Hf-doping. At Fe/Co = 70/30, the magnetically annealed $(Fe_{0.7}Co_{0.3})_{1-y}Hf_y$ films with y = 0.087 – 0.124 showed a very high uniaxial anisotropy and an ultrahigh ferromagnetic resonance frequency over 7 GHz after the films were annealed at 350 °C, while at Fe/Co = 50/50, the as-deposited $(Fe_{0.5}Co_{0.5})_{1-y}Hf_y$ films with y = 0.074 – 0.168 showed an excellent in-plane uniaxial anisotropy and high ferromagnetic resonance frequency over 3 GHz. These Fe-Co-Hf films deposited by the composition gradient sputtering method exhibited a high saturation magnetization of 1.8 – 2.2 T, a large uniaxial anisotropy field of 200 – 500 Oe, and a high ferromagnetic resonance frequency over 7 GHz, which provides great opportunities for integrated magnetic devices. © *2011 American Institute of Physics*. [doi:10.1063/1.3549584]

Soft magnetic thin films show great promise for integrated RF/microwave magnetic devices, which, however, exhibit limited operation frequency of <1 – 2 GHz due to their low in-plane uniaxial magnetic anisotropy (UMA) fields.[1] High-frequency soft magnetic films working in the GHz band require high ferromagnetic resonance frequencies, $f_{FMR} = \frac{\gamma}{2\pi}\sqrt{H_K 4\pi M_S}$, where γ is the gyromagnetic ratio close to 2.8 MHz/Oe, M_S is the saturation magnetization, and H_K is the in-plane UMA.[2] Obviously high M_S and H_K are needed for achieving a high f_{FMR} and, therefore, a high operation of the RF magnetic films. The research of RF magnetic films has mostly been focused on achieving enhanced UMA through different methods since H_K is easy to control and its magnitude can be changed by one or two orders.[3–21]

Magnetron sputtering of soft magnetic films in *in situ* magnetic field or through a subsequent magnetic field annealing have been widely employed for inducing the UMA.[5–7] However, the induced UMA fields are usually in the range of <50 Oe, which leads to limited f_{FMR} of <3 GHz for most of the magnetic films. Several different approaches have been investigated for achieving RF magnetic films with high UMA, such as oblique sputtering (OS), facing target sputtering (FTS), exchange coupling, magnetoelectric coupling, etc. OS[8–10] and FTS[11] can generate magnetic films with high UMA field values of up to 200–300 Oe. However, these magnetic films suffer severely from nonuniform thickness, particularly across a large wafer size, which is associated with the deposition configuration of OS and FTS. Exchange coupling provides another way of achieving enhanced UMA fields, such as antiferromagnetic (AFM)/ferromagnetic (FM) exchange coupling[4,12,13] and FM coupling between magnetically soft and hard layers.[14] However, the high UMA fields are limited in very thick films through exchange coupling, and it is difficult to achieve thick magnetic films with high UMA through exchange coupling. Magnetoelectric coupling in FM/ferroelectric (FE) multiferroic heterostructures could lead to dramatically enhanced and electric field tunable UMA fields in the range of 0 ~ 1000 Oe, which could lead to many novel tunable RF and microwave devices.[15–18]

In this work, an alternative method, namely the composition gradient sputtering (CGS) method, was employed to achieve high UMA in a series of $(Fe_xCo_{1-x})_{1-y}Hf_y$ alloy films with different Fe/Co atomic ratios and Hf-doping. At Fe/Co = 70/30, the CGS $(Fe_{0.7}Co_{0.3})_{1-y}Hf_y$ films with y = 0.087 – 0.124 showed a very high UMA field, up to 547 Oe, and an ultrahigh, over 7 GHz, after magnetically annealing at 350 °C for 1 h.[19] At Fe/Co = 50/50, the as-deposited $(Fe_{0.5}Co_{0.5})_{1-y}Hf_y$ films with y = 0.074 – 0.168 exhibit an excellent in-plane UMA and high f_{FMR} over 3 GHz without any annealing.

Figure 1 (upper) shows the schematic top-view of the CGS system. The main target of the $Fe_{50}Co_{50}$ directly faces the center of the substrate, which sits on a rotating turntable, while the doping target of Hf is offset radially from the

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sample center for a certain distance. This geometrical structure ensures that the composition from the $Fe_{50}Co_{50}$ target is distributed homogeneously across the sample, while the Hf atoms have a composition gradient that gradually decreases along the radial orientation, from outer (O) to inner (I), as shown in Fig. 1. A detailed description of the CGS system is also available in previous works.[19–21]

Si (100) substrates with the dimensions of 20 × 50 mm were pasted on the sample turntable [Fig. 1 (upper)]. The CGS soft magnetic films with thicknesses of 100 nm were deposited at room temperature under 2.8 mTorr Ar atmosphere with a floating rate of 20 sccm, along with a RF power of 60 W for both targets of $Fe_{50}Co_{50}$ and Hf. For comparison, the co-sputtered (CS) films were prepared by the conventional co-sputtering method under the condition of a fixed RF power of 60 W for $Fe_{50}Co_{50}$ and varied RF power from 10 to 60 W for the Hf target. Composition distribution of the films was detected by a field emission electron probe microanalyzer (FE-EPMA). Magnetic properties were measured by vibrating sample magnetometer (VSM). The high-frequency behaviors of the magnetic film were evaluated by a permeameter. Residual stress of the films was measured by an optical deflectometer. The resistivity of films was measured by four-probing method.

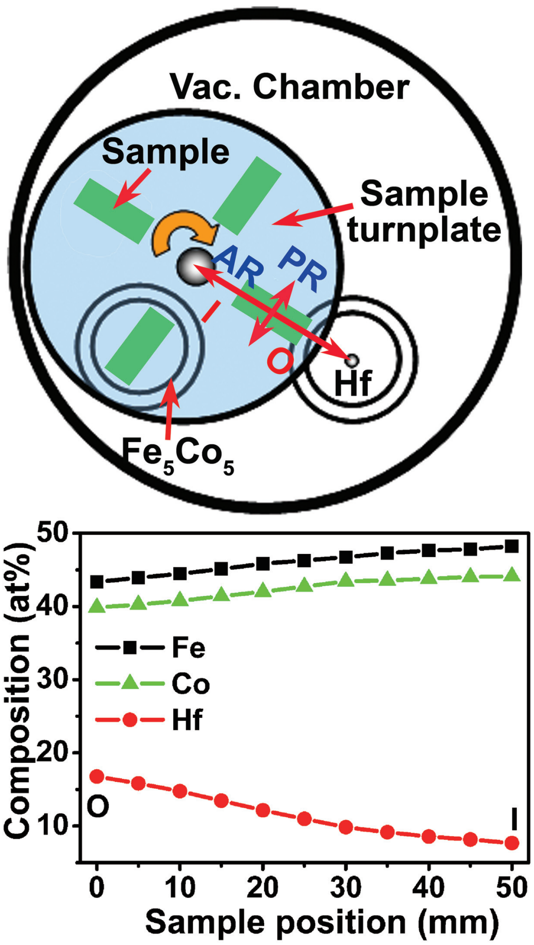

FIG. 1. (Color online) The schematic top-view of the CGS device (upper) and the composition distribution in the CGS film along the AR direction, from O to I (lower).

Figure 1 showed a gradient distribution of Hf along the radial direction (AR) of the CGS films, which decreases monotonously from 16.8 to 7.4 at. % from O toward I. However, Fe and Co compositions showed a fixed Fe/Co atomic ratio of 1.1. These facts suggest that the compositions of FeCo are homogeneous, while that of Hf varies with a radial gradient. In comparison, the compositions of Fe, Co, and Hf in CS films were homogeneous without a composition gradient.

Figure 2 shows representative hysteresis loops for CGS and CS films along the AR and perpendicular to radius (PR) directions. The as-deposited CGS films, without any annealing, exhibit large M_S, ranging from 1.5 to 2.2 T; a strong Hf-dependent UMA field H_K up to 400 Oe [Fig. 2(b)] with a hard axis along the AR direction; and coercivity (H_C) reducing from 69.1 Oe to the minimum of 0.5 Oe (at Hf = 10.4 at. %) and then increasing a little to 1.9 Oe. The H_K decreases with Hf composition. The UMA disappears when the Hf composition is less than 7.9 at. % [Fig. 2(e)]. Figure 2(f) reveals that although the CS film of $Fe_{46.0}Co_{41.4}Hf_{12.6}$ has almost the same composition as the CGS film of $Fe_{45.6}Co_{41.6}Hf_{12.8}$ [Fig. 2(c)], it exhibited isotropic hysteresis loops, while a very large UMA field of 329 Oe was observed in the latter. Moreover, the H_C of the CS films are larger than the corresponding CGS ones. These facts indicate that the Hf composition gradient in CGS films is responsible for the origin of the UMA field and the reduction of H_C. However, excessively doping Hf ruins the H_K and H_C and,

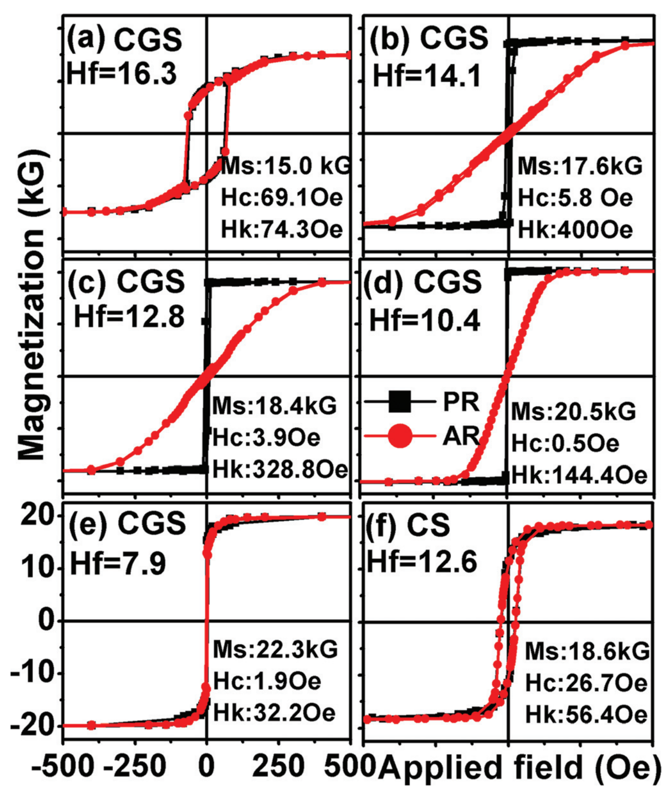

FIG. 2. (Color online) Representative hysteresis loops for CGS and CS films along AR and PR directions. All the subfigures use the same scales of magnetization (±22.5 kG) and applied field (±500 Oe). The detailed magnetic properties of films along the hard axis are marked on the figure.

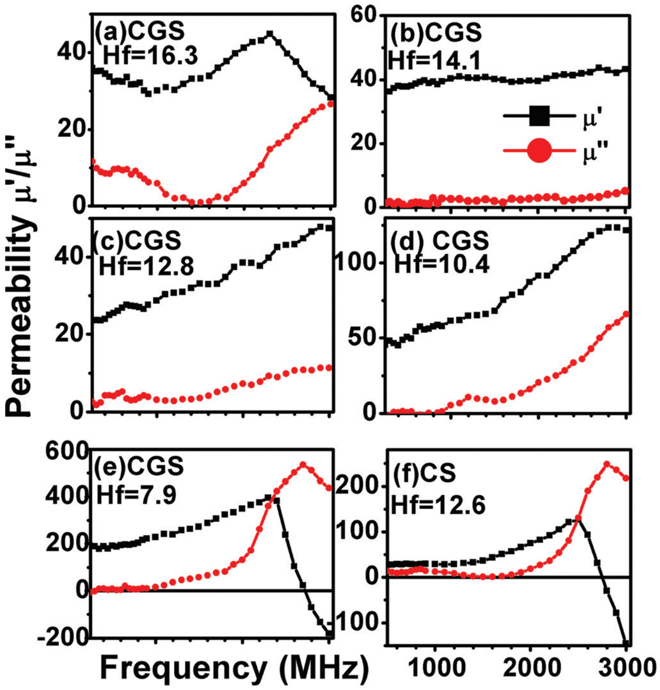

FIG. 3. (Color online) Hf composition dependence of permeability for representative CGS and CS films.

therefore, the high-frequency performance [Fig. 2(a) and Fig. 3(a)].

Since the atomic radius of Hf (2.16 Å) is greater than that of Fe (1.72 Å) or Co (1.67 Å), introducing gradient-dispersed Hf composition into Fe-Co alloy results in the presence of an additional stress along the AR. Based on the relationship between magnetostriction energy and stress, $E = -\frac{3}{2}\lambda_S \sigma \cos^2\theta$ (where λ_S is the saturation magnetostriction coefficient, σ the stress, and θ the angle between stress and magnetization), for a film with a positive λ_S, the compressive stress results in the arrangement of magnetic moments perpendicular to the compressive stress direction. The residual compressive stress σ in the CGS films varies along the AR from −386 to −155 MPa from O to I positions. The variation trend of stress is similar to that of UMA fields; they are all consistent with that of Hf composition. Therefore, the UMA in $(Fe_{0.5}Co_{0.5})_{1-y}Hf_y$ CGS films can also be attributed to the stress-induced uniaxial anisotropy, as described in Ref. 19 [Fig. 3(c)] in the case of $(Fe_{0.7}Co_{0.3})_{1-y}Hf_y$.

The high UMA field exhibited in as-deposited CGS films could lead to good RF magnetic properties in CGS films. As expected, the as-deposited CGS films exhibit a high f_{FMR} [Figs. 3(a)–3(e)]. The f_{FMR} for the most part of a CGS film with Hf composition in the range of 16.3–8.7 at. % was over 3 GHz, which can be attributed to the high UMA field in CGS films [Figs. 2(a)–2(d)]. The CGS films located at the inner site with a narrow Hf composition region from 8.7 to 7.9 at. % showed relatively lower f_{FMR}, but still more than 2 GHz, which is attributed to the lower anisotropy field [Fig. 2(e)]. By comparison, the CS film exhibited a far lower f_{FMR} than the CGS film, although they have almost the same composition [Figs. 3(c) and 3(f)]. The permeability μ increases from several tens to several hundreds along the AR, from inner to outer, which can be attributed to the Hf-dependent M_S and H_K. For instance, the μ of the sample Hf = 14.1 with $M_S = 17.6$ kG and $H_K = 400$ Oe can be estimated as 44, which agrees well with the experimental value of 40. In the case of Hf = 8.7, M_S and H_K are 22.1 kG and 71.3 Oe, so the estimated permeability is 310, which is close to the measured value of 245.

It should be mentioned here that the high UMA in these FeCoHf films is induced by a small Hf composition gradient with a negligible thickness difference of about 5% between two terminals, leading thick magnetic films with good film uniformity, which is crucial for real applications. In addition, the CGS films exhibit a relatively higher resistivity of 334-421 μΩ·cm due to the doping of Hf atoms, which reduces the eddy current loss to sustain the good high-frequency performance of $(Fe_{0.5}Co_{0.5})_{1-y}Hf_y$ CGS films.

In summary, a series of $(Fe_xCo_{1-x})_{1-y}Hf_y$ films deposited by the composition gradient sputtering method exhibit excellent high-frequency performances, which provide great opportunities for integrated magnetic devices working in the GHz frequency range.

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