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Substantially enhancing ferromagnetic resonance frequency via superposition of composition gradient sputtering and magnetoelectric coupling in FeCoAlO/PZN–PT heterostructures

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ABSTRACT

Various methods were proposed to enhance the magnetic anisotropy field H_K and ferromagnetic resonance frequency f_{FMR} of soft magnetic films (SMFs), however, the values of H_K and f_{FMR} were still relatively low for the samples prepared by a single method alone. To further enhance the microwave ferromagnetic performances, in this paper, we proposed a superposition method in which the directions of uniaxial stresses induced by composition gradient and magnetoelectric coupling were designed to be parallel to each other, so that two individual H_K added up. As a result, the H_K was dramatically increased from 100 Oe to 735 Oe, and the f_{FMR} from 3.4 GHz to 9.6 GHz in FeCoAlO/lead zinc niobate–lead titanate (PZN–PT with composition of $0.93Pb(Zn_{1/3}Nb_{2/3})O_3$–$0.07PbTiO_3$) multiferroic heterostructures. This superposition method showed big advantage over any of the single method, therefore was a promising method for preparing high-performance microwave SMFs with f_{FMR} in excess of X-band.

1. Introduction

Modern electronics are being developed to have high-frequency/broad-band, frequency tunability, lightweight, low energy consumption, etc. Microwave soft magnetic films (SMFs) have drawn an increasing attention because they satisfy this developing trend, since SMFs exhibit high saturation magnetization, sufficiently high ferromagnetic resonance frequency f_{FMR}, high permeability μ, large frequency tunability, and to be IC fabrication process compatible [1–3]. FeCo-based metallic SMFs exhibit unique advantages in monolithic microwave integrated circuits (MMICs) devices for the high $4\pi M_S$ and permeability, as well as good compatibility with the IC process [4–6]. The dominate microwave performance in MMICs is ferromagnetic resonance frequency f_{FMR}, while the key approach to enhance f_{FMR} is to increase the anisotropy magnetic field H_K, as described by Kittel equation [7]. Many preparation methods were proposed to obtain a high H_K, such as *in-situ* magnetic field deposition [8,9], post magnetic annealing, oblique sputtering [10,11], exchange coupling between ferromagnetic (FM) layer and antiferromagnetic (AFM) layer or FM layer with higher coercivity [12,13], and magnetoelectric (ME) coupling [14,15]. In our previous work [16–19], a novel composition gradient sputtering (CGS) method was applied to achieve a high uniaxial magnetic anisotropy in SMFs, which dramatically increased the in-plane uniaxial H_K to 547 Oe due to the uniaxial stress distribution induced by composition gradient. As a result, good microwave ferromagnetic properties with f_{FMR} over 7 GHz were obtained in CGS magnetic films [16]. Recently, Ong et al., reported that CGS film exhibits an abnormal temperature characteristic, i.e. the H_K and f_{FMR} is improved with the increase of temperature [20]. This unusual temperature characteristic indicates that CGS-SMFs could overcome the shortcomings of temperature degradation of microwave performance for traditional SMFs. It would be expected to prepare SMFs working at higher temperature, and to fabricate new principle devices based on the abnormal temperature characteristic of CGS-SMFs.

Although many efforts were made in the past few decades, the f_{FMR} of SMFs is still relatively low due to the limitation of preparation methods, as well as the isolation of these methods from each other. In this paper, we proposed an effective method to enhance H_K, in which two H_K enhancing methods were combined together.

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As a result, the H_K was dramatically enhanced from 100 to 735 Oe, and the f_{FMR} was increased from 3.4 to 9.6 GHz in CGS-FeCoAlO/PZN–PT multiferroic heterostructures due to the synergy enhancing effect between the CGS-induced and electric-field-induced stresses.

2. Experimental procedure

The FeCoAlO films were deposited on the (1 0 0) single crystal Si substrate using a CGS method, shown in Fig. 1, at room temperature under 2.8 mTorr Ar atmosphere with a floating rate of 20 sccm, along with a RF power of 80 W for $Fe_{50}Co_{50}$ target and 120 W for Al_2O_3 target. The Si substrate with a dimension of 30 $mm^L \times 5\ mm^W \times 0.5\ mm^T$ was fixed on the sample turntable with the longitudinal direction along the radial direction of the sample turntable. As described in our previous work [16–19], the main target of $Fe_{50}Co_{50}$ directly was facing the center of the substrate which seated on a rotating turntable, while the doping target of Al_2O_3 was with a radial offset from the sample center. This configuration allowed the compositions from $Fe_{50}Co_{50}$ target to be distributed homogeneously across the sample, and that from Al_2O_3 to have a center-to-edge gradient along the radial direction (see Fig. 2). As discussed in previous work, the composition gradient along the radial direction gave rise to a well-defined uniaxial magnetic anisotropy with the magnetic hard axis (HA) along the radial direction and an easy axis (EA) along the tangential direction (see Fig. 3(a)). Moreover, the magnetic and high-frequency ferromagnetic properties showed an obvious position n dependence. For convenience, the radial direction of the sample was named as ***R*** direction, while the perpendicular direction (i.e. tangential direction) was named as ***T*** direction. The sample was divided into 6 length-equal segments along the ***R*** direction, i.e. the dimension of each segment was 5 $mm^L \times 5mm^W \times 0.5mm^T$. The 6 sample segments were subsequently numbered as n = 1, 2, 3, 4, 5, and 6 from inner to outer (see Fig. 1 insert).

As described above, both CGS and ME coupling are good methods to prepare microwave SMFs. In order to further enhance H_K by combining these two effects, we selected a special deposition position, at which the high-frequency ferromagnetic properties were optimized. It was previously verified by the FeCoAlO CGS films deposited on Si substrate which was bonded to a small (0 1 1)-cut single crystal PZN–PT substrate with a dimension of 5 $mm^{[100]} \times 5\ mm^{[01-1]} \times 0.5\ mm^{[011]}$. In this study, the special position is located as n = 6 (see Fig. 1 inset, position 6). Note that the [1 0 0] direction of PZN–PT substrate was along the ***R*** direction, and [0 1 −1] along ***T*** direction (i.e. [1 0 0]//***R***, [0 1 −1]//***T***). After that, the FeCoAlO CGS film was deposited on the PZN–PT substrate under the same deposition conditions. For convenience, the FeCoAlO CGS films deposited on Si and PZN–PT substrates were referred to as S_{CGS} and S_{ME}, respectively.

The magnetic properties were measured by a vibrating sample magnetometer (VSM). The compositions of FeCoAlO films were detected by a field-emission electron probe micro-area analyzer (FE-EPMA) at thin film mode. The microwave frequency performances of the multiferroic composites were evaluated by using the vector network analyzer with a co-plane waveguide fixture and a wideband ferromagnetic resonance (FMR) spectrometer.

3. Results and discussion

The composition of FeCoAlO CGS films detected by FE-EPMA is shown in Fig. 2. The test distance ***x*** refers to the distance from the inner termination of the 30-mm Si substrate. As illustrated,

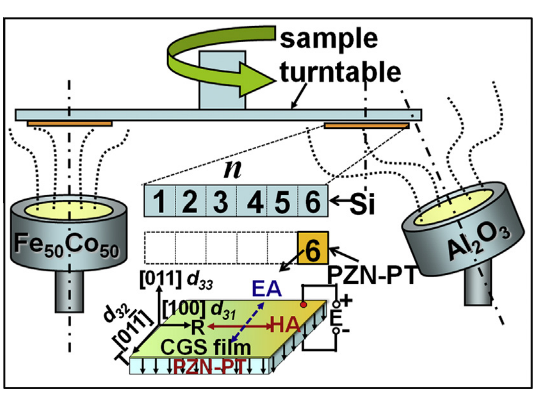

Fig. 1. The schematic drawing of CGS device. The inserts from top to bottom showed the position distribution of S_{CGS} sample on Si, S_{ME} sample on PZN–PT, and the profile of S_{ME} sample.

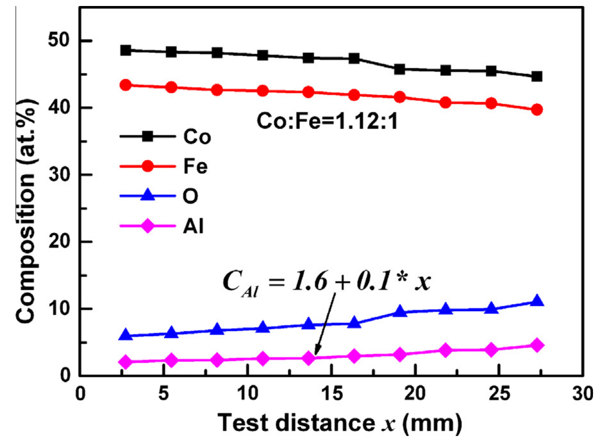

Fig. 2. The test position ***x*** dependency of compositions.

the fixed Co:Fe atomic ratio of 1.12:1 for all the test distance ***x*** demonstrates that the compositions from $Fe_{50}Co_{50}$ target is homogenous. However, the Al content, which was from Al_2O_3 target, almost increased linearly following the equation of $C_{Al} = 1.6 + 0.1 * \boldsymbol{x}$. As discussed below, this composition gradient of doping elements gave rise to a well-defined uniaxial magnetic anisotropy with EA along ***T*** and HA along ***R*** direction (see Fig. 3(a) and (c)).

For a (0 1 1)-cut PZN–PT single crystal substrate, positive voltage would lead to a compressive stress in the SMFs along the [1 0 0] direction and a tensile stress along the [0 1 −1] direction. According to the theory of ferromagnetism, magnetostriction energy is expressed as,

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

where λ_S is the saturation magnetostriction coefficient and σ is the stress. Positive and negative signs of stress correspond to tensile and compressive stresses, respectively. θ is the angle between the stress and magnetization. Based on Eq. (1), for the magnetostrictive material with a positive λ_S (e.g. the FeCoAlO films in this study), the compressive (tensile) stress results in the alignment of magnetic moments perpendicular (parallel) to the compressive (tensile) stress direction [21,22]. Therefore, if the [1 0 0] direction of PZN–PT is along ***R*** direction and the [0 1 −1] direction along ***T*** (i.e. [1 0 0]//***R***, [0 1 −1]//***T***), the EA direction induced by CGS and ME coupling would be parallel to each other. Thus, these two H_K enhancing effects add up, and a significant increase of H_K could be obtained in S_{ME} sample. Fig. 3 showed the position ***n*** and E-field dependency of hysteresis loops for S_{CGS} and S_{ME} samples. As illustrated in Fig. 3(a), a well-defined uniaxial magnetic anisotropy with a HA along ***R*** and EA along ***T*** was obtained in S_{CGS} sample. With the position ***n*** shifted from 1 to 6, the H_K along ***R*** direction increased gradually from 100 Oe to 260 Oe for S_{CGS} samples (see Fig. 3(b)). Compared with S_{CGS} samples, S_{ME} sample prepared under the same conditions at ***n*** = 6, showed a similar hysteresis behavior but larger EA coercivity and slightly smaller H_K of 230 Oe. This slight difference was attributed to the different properties between the Si and PZN–PT substrates. The E-field dependency of hysteresis loops of S_{ME} sample, as shown in Fig. 3(d), indicated that the H_K was increased from 230 to 735 Oe when the E-field increased from 0 to 8 kV/mm. It implied that a large shift of f_{FMR} could be achieved by E-field tuning.

The E-field induced change of H_K can be estimated by doing magnetic hysteresis loop measurements, while accurate H_K shift could be characterized by field sweep FMR measurements [15]. Fig. 4 showed the change of FMR field H_r with the sample position