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Magnetic anisotropy in multilayer nanogranular films $(Co_{40}Fe_{40}B_{20})_{50}(SiO_2)_{50}$/α-Si:H

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A B S T R A C T

Macroscopic and local magnetic anisotropy of $[CoFeB\text{-}SiO_2/\alpha\text{-}Si:H]_{60}$ multilayer films was studied in comparison with that for the thick $CoFeB\text{-}SiO_2$ granular films. The volume fraction of magnetic component $Co_{40}Fe_{40}B_{20}$ is ranged from 0.34 to 0.70. In the multilayer films the magnetic layer and silicon spacer thicknesses are 2.5÷4.5 nm and 3.5 nm correspondingly. The magnetization curves and ferromagnetic resonance measurements were used to determining magnetic anisotropy energy constants. The main contribution to the local magnetic anisotropy energy constant is found to be from surface magnetic anisotropy of magnetic granules. It is found that the magnetic percolation point of multilayer films (X_C~40) is shifted in comparison with that in granular films (X_C~30). Surface magnetic anisotropy energy constants of multilayer films with different X evaluated from the surface modes of spin-wave resonance are in the range $0.5 < K_s < 0.9$ erg/cm^2.

1. Introduction

Granular magnetic films with the soft magnetic granules and nonmagnetic matrix with low conductivity are promising magnetic nanostructures for high-frequency applications due to their high resistivity and a high magnetic permeability [1–3]. Magnetic anisotropy is one of the most important applied properties of magnetic materials. Depending on the type of application (permanent magnets, information storage media or magnetic cores in transformers and magnetic recording heads etc.) materials with high, medium or low magnetic anisotropy may be required [4–6]. Therefore, it is important to study the magnetic anisotropy localized at different spatial scales. The anisotropy localized at scale much smaller than the sample size determines the hysteresis loop shape, even in the absence of any texture [7]. Macroscopic magnetic anisotropy controls magnetization curve in the functional element using different directions of applied field. Hence, we conduct a detail analysis of both, local and macroscopic magnetic anisotropies. The local magnetic anisotropy field in granular films can be obtained using the law of approach to saturation of magnetization [8,9]. In this paper, we use grain size dependence of local magnetic anisotropy to separate volume and surface contributions to anisotropy. Macroscopic magnetic anisotropy can be determined using measurements of uniform modes of ferromagnetic resonance in transverse and longitude film orientations. The observation of surface mode can be used to determine the surface anisotropy constant K_s[10]. In the paper we will demonstrate that the surface anisotropy gives a significant contribution to magnetic anisotropy for the granular multilayer films.

In spite of the long range magnetic dipolar interaction the microscopic magnetic anisotropy disappears in granular films with non-zero volume fraction of ferromagnetic granules [11,12]. This phenomenon is observed in granular multilayer films as well. The granular layers as thin as granule diameter are of interest due to a sharp change in the value of the percolation threshold and a dramatic change of the magnetic properties [8]. The 3D granular magnetic films composed of magnetic nanoparticles imbedded in a matrix of SiO_2 have been widely studied and their magnetic properties have been published in the literature [9,12–18]. The interesting possibility in tailoring properties of the magnetic granular films arises when using semiconductors in matrix material. The magnetic metallic granules embedded in a matrix with free electrons can experience the exchange interaction via common electronic system of a composite even if the concentration of granules is lower than the percolation threshold [19]. In this paper we investigate the magnetic properties of nanogranular $CoFeB\text{-}SiO_2$

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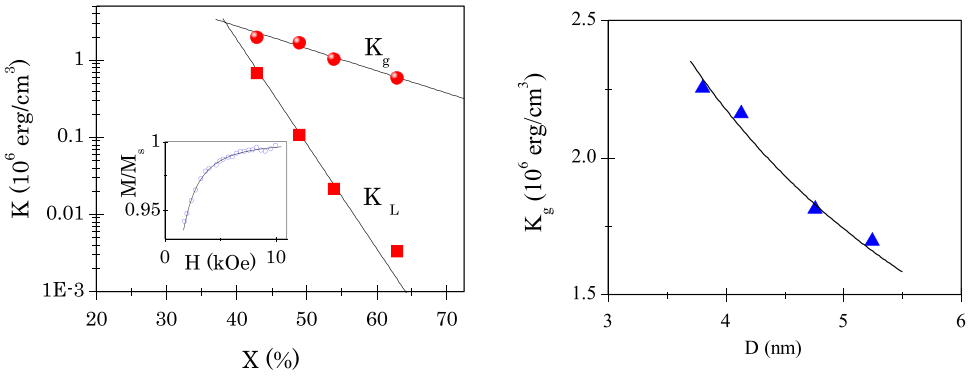

Fig. 1. Anisotropy constant of individual granules K_g and stochastic magnetic domain $\langle K\rangle_L$ vs volume fraction of magnetic granules. Inset, approach to magnetic saturation for the sample x=49% (dots) and the fitting by Eq. (2) (a). Anisotropy constant of individual granules vs it's size. The solid line – eq. $K_g=K_V+6k_s/D$, where $K_V\approx10^5$erg/cm^3 and $k_s\approx1.45$ erg/cm^2 (b).

ultrathin 2D layers sandwiched by a-Si:H, and we focus our attention on the analysis of different types of magnetic anisotropy.

2. Experiment

Nanogranular films with a nominal composition of $(Co_{40}Fe_{40}B_{20})_{100-X}(SiO_2)_X$ $(34 < X < 70)$ and $[(Co_{40}Fe_{40}B_{20})_{100-X}(SiO_2)_X/\alpha\text{-Si:H}]_{60}$ multilayer films were produced by the ion-beam sputtering [15,16]. The details on the films fabrication, the particle size distribution and the microstructure of the composite films as well as the volume fracture dependences of electric and magnetic properties of $(Co_{40}Fe_{40}B_{20})_X(SiO_2)_{100-X}/\alpha$-Si:H films were reported elsewhere [8,9,12,16]. The TEM photographs of the $(Co_{40}Fe_{40}B_{20})_X(SiO_2)_{100-X}$ granular films show a system of particles with sizes varying from 3 to 4–6–8 nm when X increases from 34% to 70%. In the $Co_{40}Fe_{40}B_{20}$-SiO_2 system, the investigated samples do not have well insulated metal particles, due to the Co particles agglomerates forming. Above X > 50 the system shows arrangements of touching metal particles and the formation of the labyrinth-like structure of ferromagnetic granules. Magnetic metallic granules $Co_{40}Fe_{40}B_{20}$ were amorphous. The magnetic percolation threshold in $Co_{40}Fe_{40}B_{20}$-SiO_2 films with the thickness much greater than the granule size was found to be approximately 0.3 according to magnetometric measurements [9]. Below the magnetic percolation threshold the films are superparamagnetic and above it they are ferromagnetic [8]. The thickness of the single layer film $(Co_{40}Fe_{40}B_{20})_{100-X}(SiO_2)_X$ is 300 nm. The magnetic layer thickness in multilayers is in the range of 2.5–4.5 nm and the number of layers is 60. The thickness of nonmagnetic spacers in multilayers is 3.5 nm for $[(Co_{40}Fe_{40}B_{20}\text{-}SiO_2/\alpha\text{-Si:H}]_{60}$. The X-band ferromagnetic resonance spectra at 9.2 GHz were measured at room temperature using a standard EPR spectrometer. The magnetization curves were measured using vibrating sample magnetometer in the fields of up to 14 kOe applied in the film plane.

3. Results and discussion

There is no magnetic anisotropy in the film plane according to magnetization curve studies for all the samples. Approach magnetization to saturation curves $M(H)$ for granular and multilayer films at 77 and 300 K are well fitted by the expression:

$$M(H) = M_s\cdot(1 - 1/15\cdot H_a^2\cdot H^{-1/2}\cdot(H^{3/2} + H_R^{3/2})^{-1}) \quad (1)$$

where $H_a=2k_g/M_s$ is the local magnetic anisotropy field (magnetic anisotropy of granule) and $H_R=2A/M_sR_c^2$ is exchange field, where A is the exchange stiffness, M_s is magnetization and R_c is the granule size. The Eq. (1) is derived for the random anisotropy model [20,21]. According to this model, the easy axis of different granules with the size R_c and magnetization M_s is oriented arbitrary. Exchange induced magnetic correlations are spread out through the numerous granules that results in reduction of effective magnetic anisotropy (stochastic domain anisotropy) averaged within scale of stochastic domain size R_L. The macroscopic anisotropy field in a stochastic domain is defined as $\langle H_a\rangle_L = 2K_L/M_s = 2A/M_sR_L^2$ and can be estimated for given values of H_a and H_R according to $\langle H_a\rangle_L=H_a^4/H_R^3$. The values of local magnetic anisotropy field H_a and anisotropy field of stochastic magnetic domain $\langle H_a\rangle_L$ are found to decrease with increasing volume fraction of the magnetic phase. The magnetic anisotropy energy constant of granule, $K_g= H_aM_s/2$, and anisotropy energy constant of stochastic magnetic domain, $K_L=\langle H_a\rangle_LM_s/2$, are calculated using fitted parameters H_a and H_R. Resulting values of $\langle H_a\rangle_L$ are shown in Fig. 1 for all samples for different granule concentrations, X. The figure shows that for $[(Co_{40}Fe_{40}B_{20})_X(SiO_2)_{100-X}/\alpha\text{-Si:H}]_n$ multilayer films, values of K_L and K_g increase exponentially and converge as X decreases. The extrapolation of these dependences to the low X allows us to estimate the percolation threshold by interparticle exchange as a point of convergence $X_C\approx40\%$. Our earlier studies of $Co_{40}Fe_{40}B_{20}$-SiO_2 granular 3D films have found the percolation threshold, X_C, to be about 30% [9]. An increase of the percolation threshold in comparison with 3D granular films has been predicted by percolation theory and has been observed in the magnetic granular films for 2D granular media [22–24] up to $X_C\approx60\div80\%$. The individual granular layers in our multilayer structure can be considered as 2D granular media as far as granule size is close to the layers thickness. However as we discussed in [8] there is exchange interaction through the 3 nm silicon spacers in our granular multilayer films. As a result, 3D magnetic correlations in the granular multilayer film are present, and therefore, Eq. (1) can be applied to 3D media [20]. Nevertheless, the exchange interaction between granules inside the single layer is stronger, than between granules of different layers. Thus, we have an intermediate situation between 2D and 3D granular media, which can explain a slight increase of X_C to 40%. Fig. 1b shows that magnetic anisotropy of granules K_g is creasing function of granule size D. The following equation that takes into account both, surface and volume anisotropies to the local anisotropy energy of magnetic granule [25], is used to fit the data:

$$K_g = K_V + 6k_S/D \quad (2)$$

The resulting value are found to be $k_s = 1.45$ erg/cm^2 and $K_V\approx10^5$ erg/cm^3.

Ferromagnetic resonance (FMR) spectra of the $(Co_{40}Fe_{40}B_{20})_X(SiO_2)_{100-X}$ nanogranular and $[(Co_{40}Fe_{40}B_{20})_X(SiO_2)_{100-X}/\alpha\text{-Si:H}]_n$ multilayer films were studied for various orientations of the film plane with respect to the external field. The FMR spectrum for the field applied parallel to the film plane consists of a single absorption line for all types of samples. The FMR peaks of the granular and multilayer films with the same composition of metal layer differ by position and line width. Fig. 2 shows the dependencies of resonance fields on volume fraction of magnetic phase.

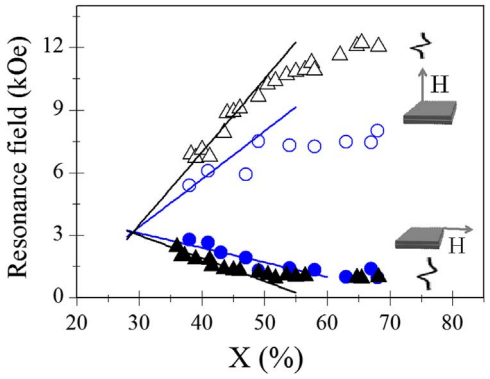

Fig. 2. Ferromagnetic resonance field for the single thick composite layer and multilayer. The difference in transverse and lateral mode of applied filed is the measure of macroscopic magnetic anisotropy.

The difference in transverse and longitude resonance fields can be considered as a measure of macroscopic magnetic anisotropy of the film-shaped granular media. The transverse and longitude resonance fields converge as X decreases. That could have been expected, as the effective magnetic shape anisotropy decreases as the interaction among granules becomes weaker. Extrapolations of transverse and longitude $H_r(X)$ dependencies to the low X values reveal an intersection at H_r =ω/γ=3.0 kOe, that corresponds to a resonance in isolated spherical granules. The volume fraction of magnetic granules at the point of convergence is X_i~33%. The convergence point, X_i, should be distinguished from the percolation threshold, X_C. Magnetic isotropy in the media that consist of ferromagnetic granules in nonmagnetic matrix can be achieved only in the limit X_i=0 [26] as far as macroscopic magnetic anisotropy is result from the long-range dipole-dipole interactions. The observed non-zero value of X_i can be attributed to a higher fraction of superparamagnetic granules when X is lower. These granules can be regarded as non-magnetic, because they do not create a non-zero average DC field that would affect other granules. At the point X=X_i the fraction of these granules becomes close to 100%, and the fraction of ferromagnetic granules becomes negligible. As a result, the macroscopic magnetic anisotropy disappears.

The spectrum of standing spin-wave has been registered in the perpendicular experiment configuration for all types of films with the metal phase content above 48 vol%. A surface resonance mode is observed for all samples (Fig. 3 insert). It means that there was the easy-plane anisotropy in all films, which can be caused by a change of chemical composition of granules on the surface or by an increase of structural defects quantities. The surface anisotropy value is evaluated using the equation [27]:

$$K_s = \left(\frac{(H_s - H_1)AM_{eff}}{2}\right)^{1/2} \quad (3)$$

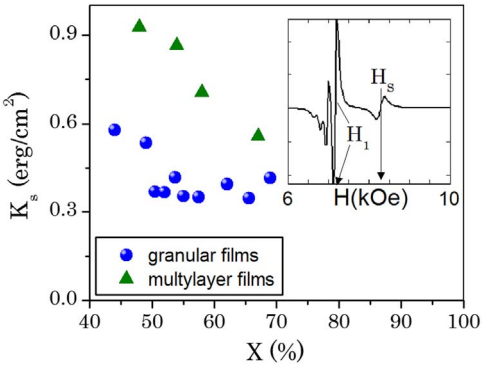

Fig. 3. Surface magnetic anisotropy *vs ferromagnetic phase content* in granular and multilayer $[(Co_{40}Fe_{40}B_{20})_{54}(SiO_2)_{46}/\alpha\text{-Si:H}]_{60}$ films. Inset, SWR spectrum of the multilayer $[(Co_{40}Fe_{40}B_{20})_{54}(SiO_2)_{46}/\alpha\text{-Si:H}]_{60}$ films.

where H_s is the resonance field that corresponds to the surface mode. Fig. 3 shows the dependencies of surface magnetic anisotropy value on volume of magnetic phase for $(Co_{40}Fe_{40}B_{20})_X(SiO_2)_{100-X}$ granular and $[(Co_{40}Fe_{40}B_{20})_X(SiO_2)_{100-X}/\alpha\text{-Si:H}]_{60}$ multilayer films. The K_s magnitudes for multilayer films are larger than that for single layer films. The difference of K_s values for granular and multilayer films diminishes as metal volume fraction increases. Enhanced values of the surface anisotropy ($0.5 < K_s < 0.9$ erg/sm^2) for multilayer films probably arise from the interfacial anisotropy between the $Co_{40}Fe_{40}B_{20}$-SiO_2 and silica layers.

4. Conclusions

The magnetic anisotropy of $[(Co_{40}Fe_{40}B_{20})_X(SiO_2)_{100-X}/\alpha\text{-Si:H}]_{60}$ multilayer films ($34 < X < 70$) produced by the ion-beam sputtering was investigated. Information on local anisotropy field was obtained from investigation of approach to saturation magnetization law. It was found that surface anisotropy of granules is predominant. The magnetic anisotropy energy constant K_g of granule and anisotropy energy constant of stochastic magnetic domain K_L increase exponentially as the volume fraction of magnetic phase decreases for $[(Co_{40}Fe_{40}B_{20})_X(SiO_2)_{100-X}/\alpha\text{-Si:H}]_n$ multilayer films. We showed that the magnetic percolation point (X_C~40) of multilayer films was shifted relative to the granular films (X_C~30). The surface anisotropy value of $[Co_{40}Fe_{40}B_{20}$-$SiO_2/\alpha\text{-Si:H}]_n$ multilayer films was evaluated from the surface resonance modes of SWR spectra as $0.5 < K_s < 0.9$ erg/cm^2.

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