Journal of Alloys and Compounds 506 (2010) 109–114

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom

Thickness dependence of magnetic properties in $(Fe)_{0.73}(Sm_2O_3)_{0.27}$ nano-granular films

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ARTICLE INFO

Article history:
Received 9 March 2010
Received in revised form 17 June 2010
Accepted 22 June 2010
Available online 16 July 2010

Keywords:
Nano-granular film
Stripe domains
High-frequency property

ABSTRACT

$(Fe)_{0.73}(Sm_2O_3)_{0.27}$ nano-granular films with thicknesses varying from 46 nm to 220 nm were fabricated by a RF magnetron sputtering method. The static and dynamic magnetic properties of these films on thickness dependence have been investigated in detail. High-resolution transmission electron micrograph shows that the films consist of Fe granules and crystalline Sm_2O_3 matrix. The results reveal that there is a critical thickness t_c around 65 nm in the $(Fe)_{0.73}(Sm_2O_3)_{0.27}$ nano-granular films. Below t_c, the films possess an in-plane uniaxial anisotropy and lower H_c. Above t_c, the films exhibit a small perpendicular anisotropy, higher H_c and stripe domains, and the stripe period increases with increasing film thickness. The dynamic permeability spectra measured over the frequency range of 0.1–7 GHz display one resonance peak for all thicknesses. The spectrum of the film with thickness below t_c can be ascribed to the uniform coherent spin procession. Above t_c, the increasing film thickness leads to an monotonously increase of the permeability (imaginary part), and a shift of resonance frequency towards the low frequency and a decrease of resonance linewidth for the films with thickness larger than 120 nm.

1. Introduction

With the rapid improvement and miniaturization of electromagnetic devices, soft magnetic films with excellent high-frequency characteristics used in magnetic components, such as inductive devices and magnetic heads, are increasingly in demand [1–3]. The basic demands for soft magnetic material worked in the GHz range include high resistivity ρ, high permeability μ, high saturation magnetization M_S and appropriate large anisotropy field H_K so as to effectively suppress eddy current loss and to acquire high cut-off frequency. Metal-insulator granular films (MIGFs) consisting of magnetic metal nano-granules uniformly distributed in insulator matrix are one of the best candidates for satisfying these demands. These films have the advantages of high M_S and high μ of magnetic metal and high ρ of insulator, modulated magnetic properties and transport properties by adjusting the volume fraction x of magnetic metal and granule size.

So far, many kinds of granular films, such as the films composed of magnetic metals (and their alloys) and various X-(oxide, nitride and fluoride) where X is nonmagnetic elements such as Hf, Al, Si, Zr, etc. have been investigated [4–8]. However, the granular films composed of ferromagnetic (FM) metals granules and rare earth (RE) oxides were less investigated. Meanwhile, most of studies for soft magnetic films were performed on the films with small thickness (e.g. smaller than 100 nm). However, for application point of view, the thicker films have more advantages. Therefore, it is necessary to investigate if the thickness influences the magnetic properties. But there is little research in this field up to now.

In this work, $(Fe)_{0.73}(Sm_2O_3)_{0.27}$ nano-granular films were deposited on water-cooled Si substrates by a RF magnetron sputtering method. Magnetic properties, microstructure and domain structure of the films with different thicknesses were investigated.

2. Experiment

$(Fe)_{0.73}(Sm_2O_3)_{0.27}$ nano-granular films were deposited on water-cooled Si substrates by a RF magnetron sputtering system from a composite target consisting of an Fe disk with Sm_2O_3 chips placed on it. The base pressure is lower than 5×10^{-5} Pa. The sputtering gas Ar was kept at 0.4 Pa and sputtering power was 50 W during deposition. The thickness of these films is 46 nm, 79 nm, 120 nm, 175 nm, 220 nm, respectively.

Saturation magnetization, coercivity and in-plane magnetic anisotropy field were measured by a vibrating sample magnetometer (VSM) (Lakeshore 7304 model). The domain structure was observed by a Magnetic Force Microscope (MFM). The structure and microstructure were characterized by an X-ray diffraction (XRD) spectrometer (Philips X'Pert model) with Cu-K radiation and a high-resolution transmission electronic microscope (HRTEM), respectively. The complex permeability was obtained by a shorted microstrip transmission-line perturbation method [9], which works from 100 MHz to 7 GHz. The film thickness was measured by a surface profile-meter (Dektak 8). The composition of samples was determined by energy-dispersive X-ray spectroscopy (EDX). All the above measurements were performed for as-deposited samples without any post heat treatment.

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doi:10.1016/j.jallcom.2010.06.133

3. Result and discussion

According to the composition of samples determined by EDX, the volume fraction x of Fe in the film samples was determined to be 0.73 and hence the film composition can be expressed as $(Fe)_{0.73}(Sm_2O_3)_{0.27}$. Fig. 1 shows the XRD spectrum (a), the HRTEM image (b) and the corresponding electron diffraction (ED) pattern (c) for the thickest film with $t = 220$ nm. The XRD spectrum exhibits diffraction peaks from the (1 1 0) plane of bcc Fe and (2 2 2) plane of Sm_2O_3, respectively. The average grain sizes calculated by the full width at half maximum (FWHM) of the diffraction peaks in XRD spectrum through Scherrer's equation are approximately 14.3 nm for Fe and 10.1 nm for Sm_2O_3. It is seen from HRTEM image that the films consist of Fe granules and crystalline Sm_2O_3 matrix where the dark part represents the Fe granules and the bright part the Sm_2O_3. The average granule sizes from HRTEM image are approximately 12 nm for Fe and 9 nm for Sm_2O_3, respectively. The corresponding ED pattern exhibits diffraction peaks from (1 1 0) and (2 0 0) planes of bcc Fe, and diffraction peaks from (2 2 2) and (4 0 0) planes of Sm_2O_3. Thus the results from XRD and HRTEM are consistent with each other.

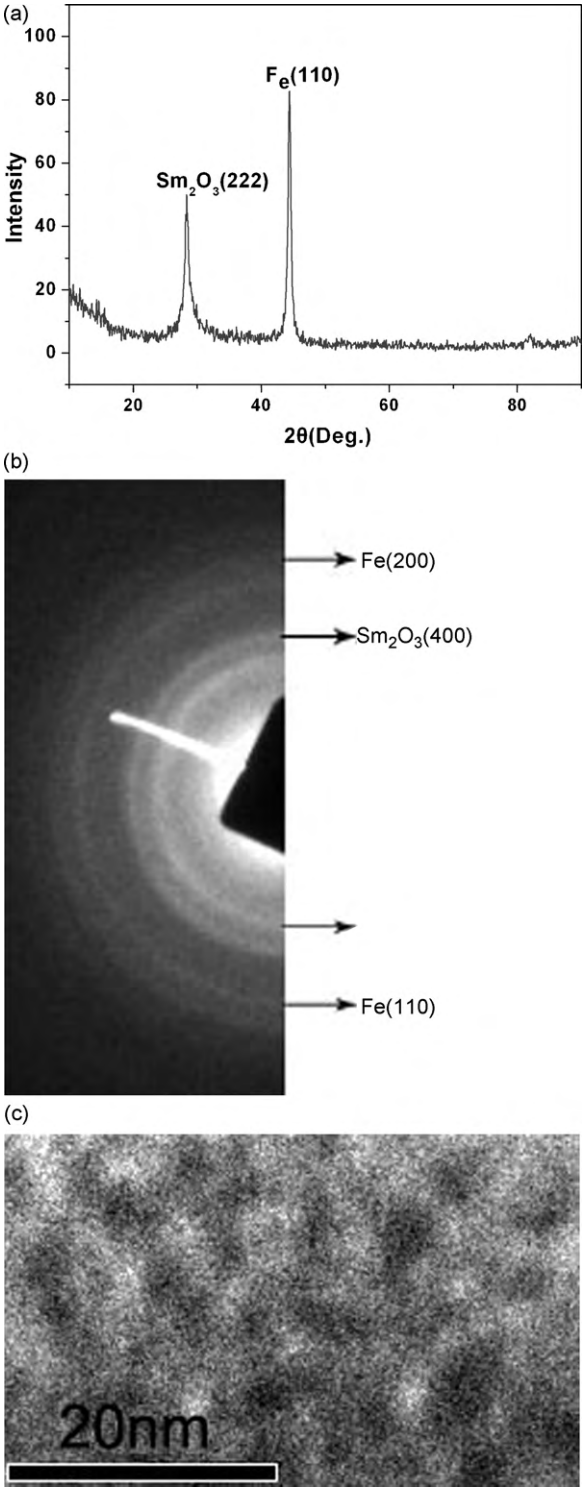

Fig. 1. XRD spectrum (a), HRTEM image (b) and the corresponding ED pattern (c) for the thickest film of $t = 220$ nm.

Fig. 2 displays the in-plane hysteresis loops of $(Fe)_{0.73}(Sm_2O_3)_{0.27}$ nano-granular films with different thicknesses. From which the saturation induction $4\pi M_S$ of the films was calculated to be 1.5 T. For the thinnest film ($t = 46$ nm), a square loop is observed along the easy axis (EA) while a slightly tilted loop with the remanence close to zero appears along the hard axis (HA), indicating that the film possesses a good in-plane uniaxial anisotropy. The anisotropy field H_K determined from the saturation field along the HA is 6360 A/m. With the increase of film thickness, the difference between the EA and HA loops reduces gradually. For the thicker films ($t > 79$ nm), the EA and HA hysteresis loops were superimposed, and the hysteresis loop shape was unchanged whatever the orientation of the in-plane applied magnetic field. These hysteresis loops are characterized by a linear decrease of magnetization from its saturation value and a moderate remanence. The shape of these loops is similar to those of the thin films with small perpendicular anisotropy exhibiting a weak stripe domain structure [10].

From the above change of the in-plane hysteresis loop shape, it is found that the magnetization of the films re-orientates from in-plane to out of the film plane with the increasing film thickness. The critical thickness t_c for this transition obtained by experiments should be between 46 and 79 nm. Below t_c, the films exhibit an in-plane uniaxial anisotropy. Above t_c, the films possess a small perpendicular anisotropy, and the in-plane remanence reduces with the increasing film thickness. Fig. 3 displays the in-plane M_R/M_S and H_c of the films as a function of film thicknesses. Below t_c the film has a higher M_R/M_S (>0.9) and lower H_c, while the M_R/M_S reduces with increasing film thickness and H_c becomes larger above t_c.

The zero-field magnetic domain structures of these films were imaged by MFM. Fig. 4 displays the zero-field parallel stripe domain structure of $(Fe)_{0.73}(Sm_2O_3)_{0.27}$ nano-granular films with different thicknesses. These domain structures of weak stripe type are induced by first saturating the films in an in-plane dc magnetic field then reducing the field to zero. It is found by our experiments that the films appear stripe domain structure when film thickness is above the critical thickness t_c, and the stripe period increases with the increasing film thickness.

From the magnetic force microscopy images and the in-plane hysteresis loops, the critical thickness for stripe domain of these $(Fe)_{0.73}(Sm_2O_3)_{0.27}$ nano-granular films is approximately $t_c = 65 \pm 15$ nm.

Fig. 5 shows the dependence of complex permeability $\mu = \mu' - j\mu''$ (μ' and μ'' are the real and imaginary parts of μ respectively) on frequency f for the films with different thicknesses. The permeability variations below 300 MHz are affected by the experimental noise. For the thinnest film ($t = 46$ nm), the film possesses an in-plane uniaxial anisotropy. It is seen from Fig. 5 (a) that for this film μ' is more than 140 below 1.5 GHz, then gradually decreases with frequency, while the imaginary part μ'' gradually increases to a maximum at $f = 2.6$ GHz, which can be ascribed to the ferromagnetic resonance (FMR) [11]. The measured curves $\mu \sim f$ data were fitted well with the solution of Landau–Lifshitz equations

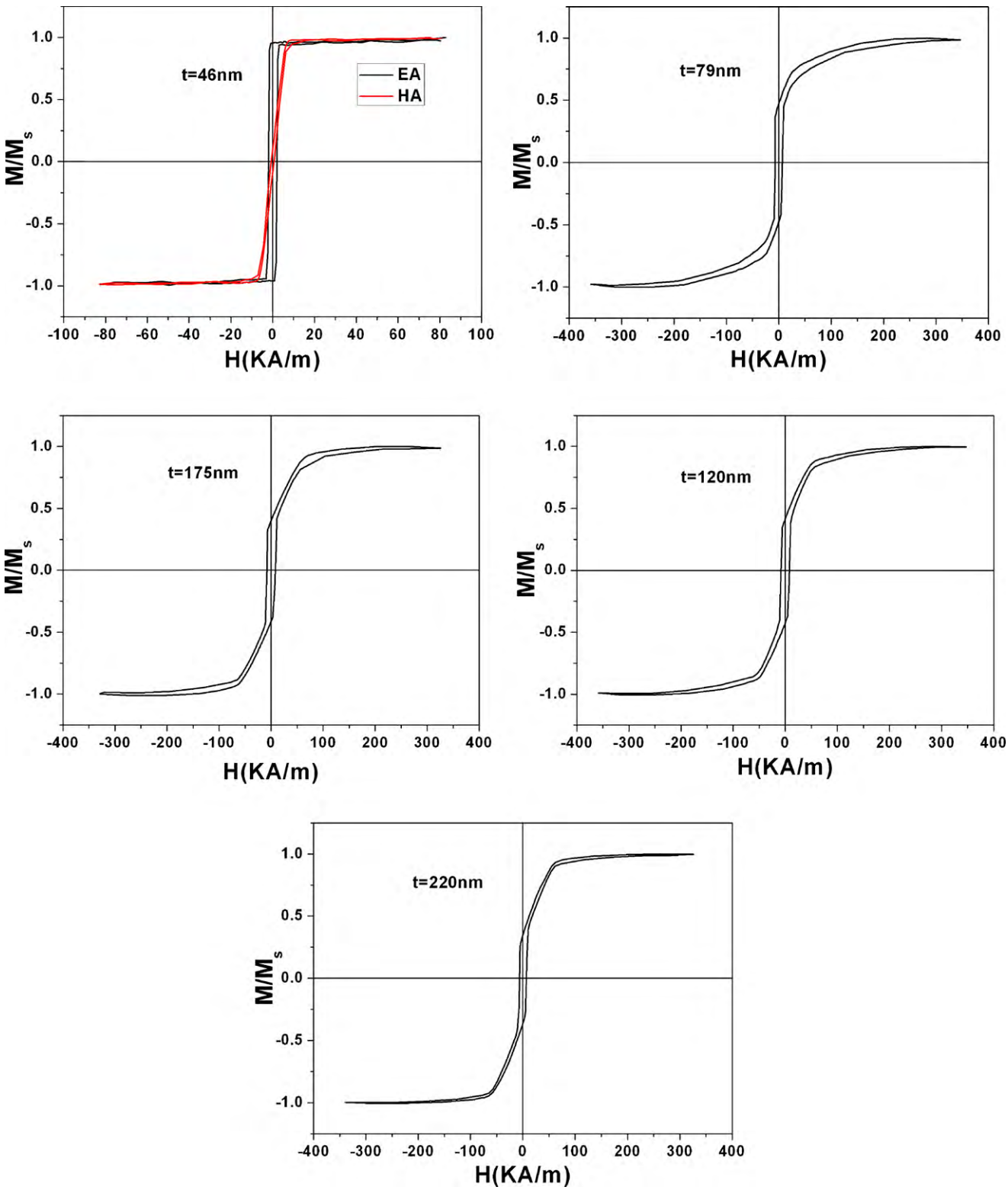

Fig. 2. Experimental in-plane hysteresis loops of $(Fe)_{0.73}(Sm_2O_3)_{0.27}$ films with different thicknesses.

taking account of the coherent spin procession [12,13], as shown by the solid lines in Fig. 5(a). For the thicker films ($t > 79$ nm), the samples possess stripe domain structure. The dependences of complex permeability μ on frequency f of these films are showed in Fig. 5(b)–(e). One resonance peak is observed over the measured frequency range for each spectrum. The increasing film thickness leads to an increase of the permeability (imaginary part), and a shift of resonance frequency towards the low frequency and a decrease of resonance linewidth for the films with thickness larger than 120 nm. These characteristics of $\mu \sim f$ spectra above t_c are related with the stripe domain structure, a study on the mechanism in detail is in progress.

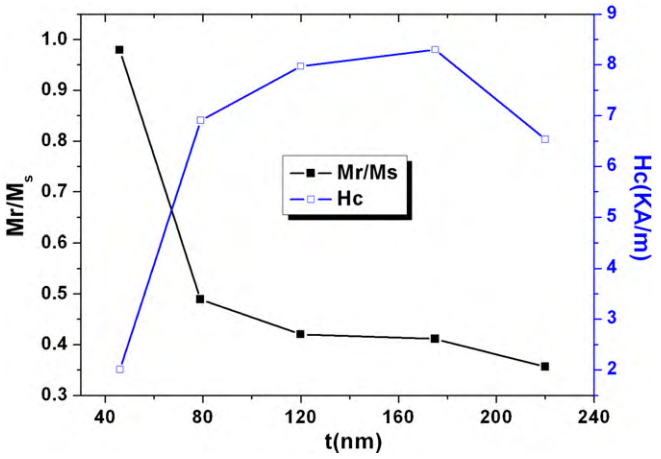

Fig. 3. Dependence of in-plane M_R/M_S and H_c on thicknesses for $(Fe)_{0.73}(Sm_2O_3)_{0.27}$ films.

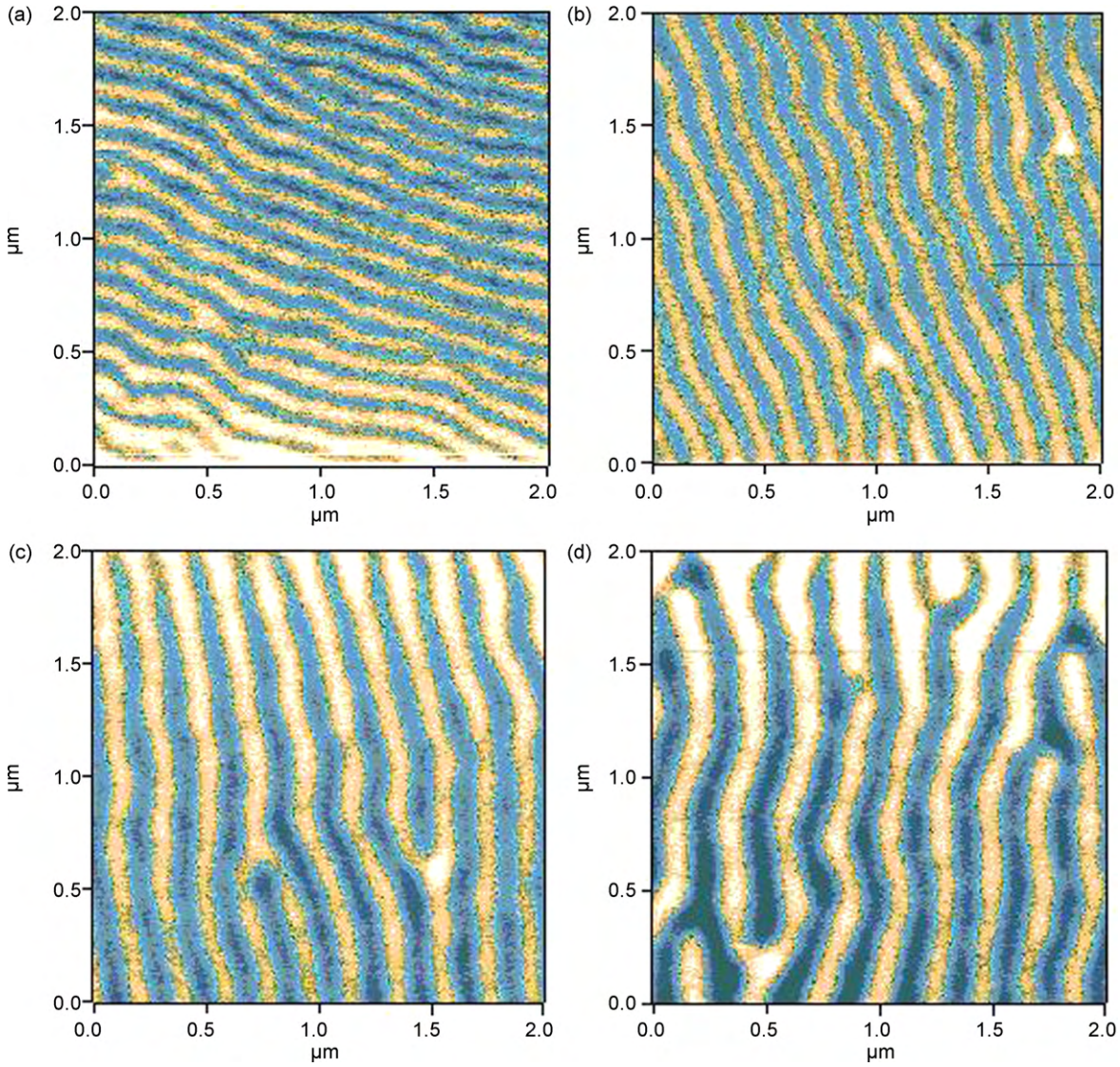

Fig. 4. Zero-field MFM images of the weak stripe domain structure as a function of film thickness: (a) t = 79 nm, (b) t = 120 nm, (c) t = 175 nm, (d) t = 220 nm.

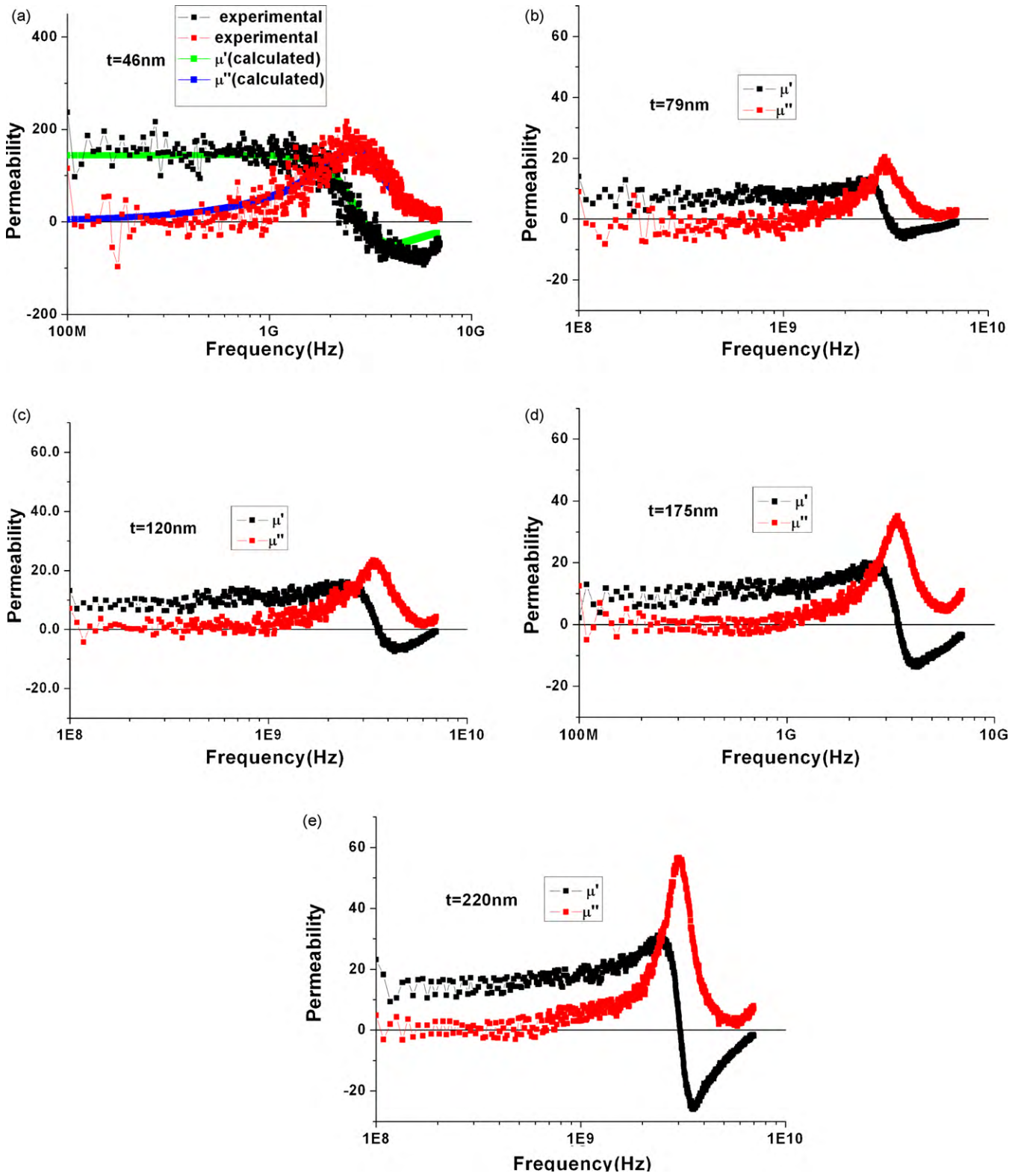

Fig. 5. Experimental dependences of complex permeability $\mu = \mu' - j\mu''$ on frequency f for $(Fe)_{0.73}(Sm_2O_3)_{0.27}$ film with different thicknesses.

4. Conclusion

The microstructure and magnetic properties of $(Fe)_{0.73}(Sm_2O_3)_{0.27}$ nano-granular films with thicknesses varying from 46 nm to 220 nm fabricated by RF magnetron sputtering were investigated. The films consist of Fe granules and crystalline Sm_2O_3 matrix. The $(Fe)_{0.73}(Sm_2O_3)_{0.27}$ nano-granular films exist a critical thickness of $t_c = 65 \pm 15$ nm. Below t_c, the films show an in-plane uniaxial anisotropy, low H_c and one resonance peak corresponding to the in-plane uniform coherent spin procession. The films with thickness above the t_c show a small perpendicular anisotropy, higher H_c and stripe domain structure,

and the increasing film thickness leads to an monotonously increase of the permeability (imaginary part), and a shift of resonance frequency towards the low frequency and a decrease of resonance linewidth for the films with thickness larger than 120 nm.

Acknowledgment

This work was supported by National Natural Science Foundation of China (Grant No. 10774061).

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