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Magnetic Softness of Permalloy Granular Films Produced by Co-Evaporation

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Permalloy granular films were fabricated by co-evaporation. In this paper, MgF_2 and CaF_2 are used as matrices. The films exhibit good magnetic softness and perpendicular or inplane uniaxial anisotropy that determines high frequency characteristics. The anisotropy is considered to originate from alignment of nanosized granules connected with exchange interaction. Magnetization curves, susceptibility and transmission electron microscopic images support speculation of microscopic granular structure and origin of anisotropy. Very high deposition speed is possible without serious deterioration of magnetic softness. High controllability of anisotropy and high speed deposition are advantageous from the industrial standpoint.

***Index Terms*—Anisotropy, evaporation, granular, softness, thin film.**

I. Introduction

THE granular films produced by sputtering exhibit high permeability and high resistivity [1]–[3]. These magnetic properties indicate their high potential as high frequency magnetic cores to be used as thin film cores in integrated inductive elements. Recently the films have been studied intensively from the standpoint of electromagnetic noise suppressors in integrated circuits [4]. Magnetic softness of the films originates from inter granular exchange interaction of ferromagnetic granules embedded in oxides or fluorides. This exchange interaction is optimized by proper annealing after deposition.

On the other hand, studies of magnetically soft granular films by vapor deposition are very rare. The reason may be that in the vapor deposition microscopic voids and stress due to low energy injection to the substrate surface are unavoidable.

We have been studying magnetic properties of granular films by vapor deposition for the reason that high speed and low temperature deposition is attractive and, by co-evaporation from separate sources of ferromagnetic metal and matrix, phase separation is enhanced in the as-deposited state and may lead to unique properties of magnetic softness. In the past we studied $N_{I80}Fe_{20}$(Py)–B_2O_3 films by co-evaporation and reported that the films exhibit relatively strong uniaxial anisotropy with the easy axis perpendicular to the film plane [5]. This anisotropy gives rise to the high frequency property that is quite different from the conventional granular films by sputtering. So, we believe that magnetic anisotropy of co-evaporated films is worth detailed investigation for development of new granular film material with high performance at very high frequencies. In this study granular films of Py–CaF_2 and Py–MgF_2 were fabricated. Magnetic and geometrical microstructures are investigated in correlation with magnetic anisotropy and magnetic softness.

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II. Experiments

The films were fabricated by an evaporation system equipped with two electron beam guns. They are charged with Py alloy and CaF_2 pressed powders or MgF_2 chips. The reason why fluorides such as CaF_2 and MgF_2 are used as matrices is that their low melting points are great advantage in the evaporation process. The base pressure before evaporation depends on baking time of the chamber. The base pressure is a dominant factor to determine strength and direction of the uniaxial anisotropy. Deposition rate is usually about 0.2 μm/min, but to see dependence of magnetic properties on the deposition rate the rate was rose up to 10 μm/min tentatively. The substrate temperature is lower than 80°C for the rate 0.2 μm/min and rose up to 250°C for 5 min deposition at 10 μm/min. The magnetic properties were measured mainly by a vibrating sample magnetometer. Magnetic force microscopic (MFM), transmission electron microscopic (TEM) images and low temperature susceptibility were studied for samples with perpendicular anisotropy. The saturation magnetization of Py deposited by evaporation was about 805 emu/cc. The packing fraction p (volume ratio of Py particles to the total film volume) was determined from the saturation magnetization.

III. Results and Discussion

Fig. 1 shows magnetization curves of Py–CaF_2 as a function of p. For $p = 0.44$, magnetic anisotropy with an easy axis perpendicular to the film plane is observed. From the curve anisotropy field H_K is estimated to be about 480 Oe and H_C to be 2.5 Oe. For $p = 0.40$ residual magnetization is very low and $H_C = 2.0$ Oe. It should be noted that H_K seems almost unchanged. For further decrease of P the curves indicate superparamagnetic behavior of Py granules. Susceptibility at low temperatures was measured for these samples to clarify inclusion of superparamagnetism. Fig. 2 shows the results.

Susceptibility was measured by well-known field cooling (FC) and zero field cooling (ZFC) process. As clearly seen in Fig. 2 the critical point is above room temperature for $p = 0.40$

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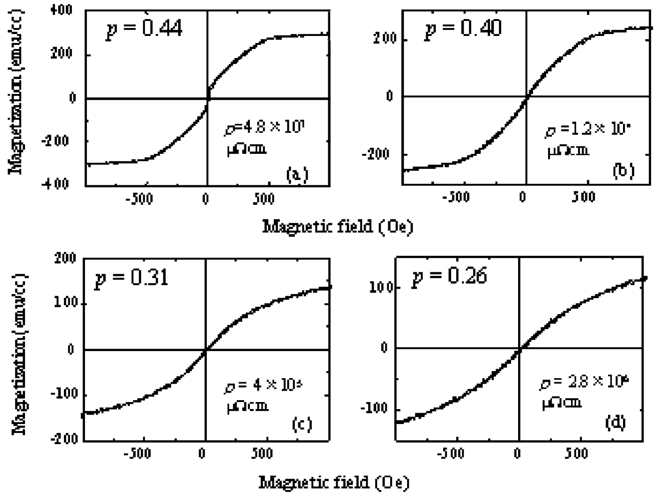

Fig. 1. Magnetization curves measured in the inplane direction of Py–CaF_2 films with a variety of p. These are isotropic in plane. Thickness: $\sim 0.5\ \mu$m.

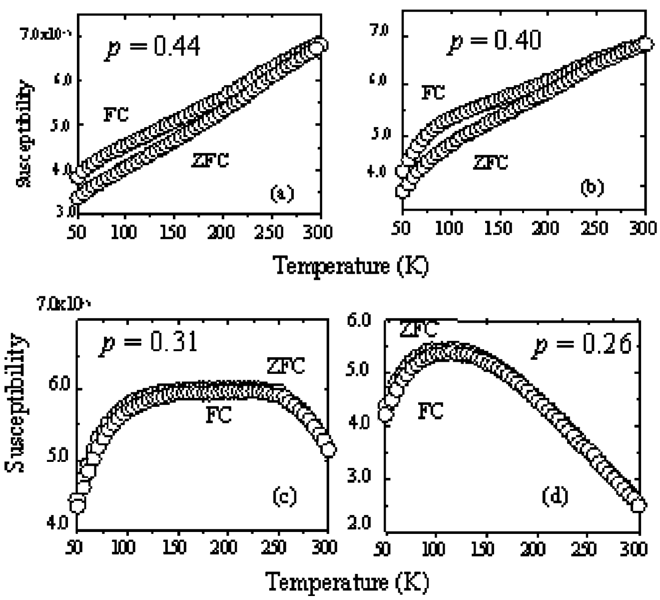

Fig. 2. FC and ZFC curves for Py–CaF_2 films. The peak of $\chi - T$ curves is above room temperature for $\mathrm{P} = 0.40$ and 0.44 and it is lower than room temperature for $\mathrm{P} = 0.31$ and 0.26. This means that the samples for $\mathrm{P} = 0.40$ and 0.44 are ferromagnetic.

and 0.44. It is lower than 300 K for $p = 0.31$ and 0.26. Usually susceptibility for an assembly of ferromagnetic particles the FC curve is positioned well above ZFC curve. This is because in FC process spins in particles are frozen in the field direction and susceptibility increases. But for $p = 0.31$ and 0.26 the curves tend to rest in the same position at the lowest temperature. This is plausible if magnetization curves analogous to that of $p = 0.44$ are reproduced at low temperatures for $p = 0.31$ and 0.26.Thus the susceptibility measurement confirms that the samples for $p = 0.40$ and 0.44 are ferromagnetic.

Transmission electron microscope images are given in Fig. 3(a) and (b). In Fig. 3(a) granules with a diameter of 3–4 nm are clearly observed and it seems that they are partly connected with each other in the perpendicular direction to the film plane. This is similar to the image of Py–B_2O_3 in [5]. The dark field image in Fig. 3(b) also suggests alignment of the granules. A cross section image of Lorentz microscopy was also observed. In Fig. 4(a) a regularly aligned domains with width of about 160 nm are observed for $p = 0.40$. This regularity is in agreement with the MFM image in Fig. 4(b). Where the domain width of about 250 nm is in good agreement with that of Fig. 4(a).

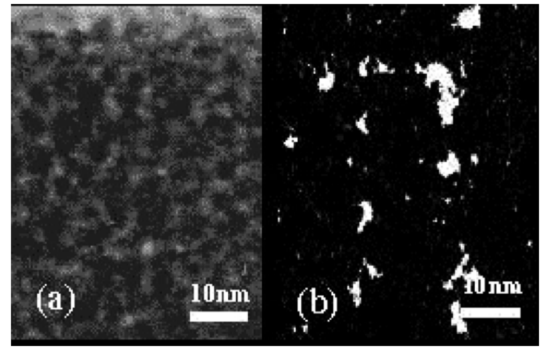

Fig. 3. Bright and dark field image of cross sectional area of a Py–CaF_2 film. $p = 0.40$.

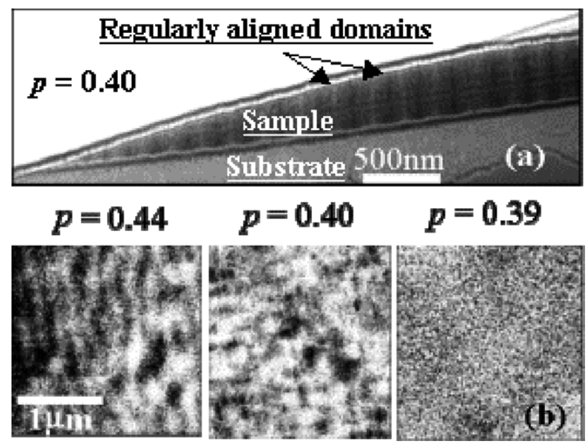

Fig. 4. Lorenz image of a (a) cross sectional area and (b) surface domain structure by MFM. (a) Regularly aligned domains are observed in sample. The lower part is substrate.

The origin of anisotropy is to be ascribed to this alignment for the following reasons. Usually magnetic anisotropy of thin films is caused by crystalline anisotropy, induced anisotropy, magnetoelastic effect or microscopic anisotropic texture (microscopic shape anisotropy). For the present samples none of these can explain the strong anisotropy of Py granules except for the microscopic texture.

In the past we reported that Py–B_2O_3 films by co-evaporation have an analogous microstructure and exhibit an explicit ferromagnetic resonance in permeability measurement where μ'' is up to 100 with resistivity of about $10^4 \mu\Omega$ cm. This high frequency property is suitable for electromagnetic noise suppression in high density integrated circuits. In this study the microscopic magnetic structure associated with this property has been made clearer. Namely, we proposed a model of fiber structure in which Py granules are aligned with strong exchange interaction in the perpendicular direction, but in this experiment clusters of relatively large number of granules rather than individual fibers behave as a unit of ferromagnet. As seen in Fig. 4 dispersion of anisotropy is very small although the granules themselves seem

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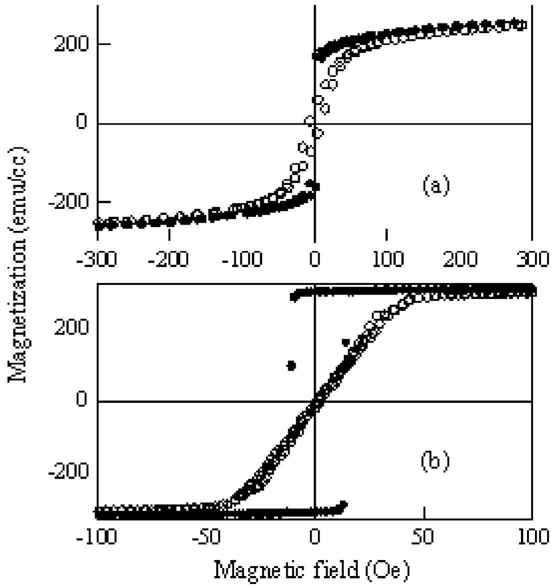

Fig. 5. Magnetization curves of (a) Py–CaF_2 $(p \sim 0.40)$ and (b) Py–MgF_2 $(p \sim 0.44)$ films. The magnetization curves are measured inplane. The open and closed circles indicate magnetic hard and easy axis direction, respectively.

to be fairly dispersed. Permeability measurement of the present films will be reported in future.

Here, it should be stressed that the films have inplane anisotropy also. The easy axis of inplane anisotropy is in the direction vertical to the injection plane including the substrate and two evaporation sources. The magnetization curves of evaporated films in our study reflect these two anisotropies. The curves for Py–CaF_2 and Py–MgF_2 represent typical cases where either perpendicular or inplane anisotropy dominates. It was generally observed that inplane anisotropy overcomes perpendicular anisotropy when the base pressure and the pressure during evaporation are high. This can be attributed to surface mobility of atoms during deposition. If it is high, phase separation of Py and CaF_2 is enhanced and alignment of granules is in more progress. This speculation is supported by the fact that by annealing at 250°C inplane anisotropy is replaced by perpendicular anisotropy.

Experimental evidences for these discussions will be reported in future and only demonstration of magnetic softness of inplane anisotropy films is given in this report. Fig. 5(a) and (b) show typical magnetization curves for Py–CaF_2 $(p \sim 0.40)$ and Py–MgF_2 $(p \sim 0.44)$. In the former case H_C in the hard axis direction is relatively high. This may be due to dispersion of inplane anisotropy in Py–CaF_2 for unknown reasons. As mentioned above, the film changes its direction of anisotropy from inplane to perpendicular after annealing. The anisotropy field can be determined accurately and its permeability exhibits resonance at 1.5 GHz in good agreement with resonance frequency predicted by the Landau–Lifshitz equation.

From the experimental results described above, it can be said that magnetic uniaxial anisotropy which dominates high frequency characteristics of granular films is controllable in the as-made state by adjusting substrate position, packing fraction and vacuum environment. The last has not been investigated fully but seems to play a dominant role to determine the easy axis direction.

The great advantage of evaporation to sputtering is speed of deposition. In this experiment deposition rate was raised up to 3 μm/min to see change of magnetic softness. Surprisingly, H_C is lower than 3 Oe in the hard axis direction for a 2 μm thick sample made with the above rate. The easy axis tends to rotate from inplane to perpendicular as the rate increases. This result suggests very high potential of co-evaporated granular films from the industrial standpoint.

Uniaxial anisotropy that determines permeability profiles in the GHz range has its origin in the granular microstructure and is controllable in the as-made state rather than by post annealing process. The advantage of co-evaporation is its high speed deposition together with low substrate temperature process. In this report the composition is limited to Py-fluoride granular structure. Co-evaporation of other transition metals with oxides as matrix will be of great interest because of the advantage mentioned above.

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