Origin of enhanced coercivity in $(\text{Fe}_{65}\text{Co}_{35})_x(\text{Al}_2\text{O}_3)_{1-x}$ granular thin films

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The magnetic behaviors of granular $(\text{Fe}_{65}\text{Co}_{35})_x(\text{Al}_2\text{O}_3)_{1-x}$ films have been examined. The results show that the coercivity at 5 K first increases sharply from 80 Oe for $x = 0.1$ to 700 Oe for $x = 0.3$ and then drops with increasing x . This behavior differs from the system of Fe–SiO₂. Considering the interfacial area, we can theoretically explain the difference between these systems. It is concluded that the peak value of coercivity becomes smaller and occurs at a lower value of x for a more rapidly increasing granular size with volume fraction.

1. Introduction

Artificially structured magnetic solids have attracted much attention in recent years [1]. Their development has led to several new research areas in magnetism and has also provided potential technological applications, especially in high-density magnetic recording. Granular film is a typical example. With adequate process conditions, granular films have proved to offer much promise as high-density recording media with superior magnetic characteristics, stable chemical properties and good wear resistance [1].

Granular film consists of ultrafine metal granules, usually a few nanometers in size, embedded in an immiscible medium that may be insulating or metallic. Due to the small number of atoms in the granules, and their large surface to volume ratios, granular films reveal many unusual mechanical, chemical and physical properties.

Recently, Chien et al. have observed a giant enhancement of H_c (≈ 2500 Oe) in the granular Fe–SiO₂ system [2], but this anomalous behavior has not been found in the granular Fe₅₀Ni₅₀–Al₂O₃ system [2]. In order to explore the origin of this peculiar coercivity enhancement, which is not yet clear, we have investigated the magnetic properties of granular $(\text{Fe}_{65}\text{Co}_{35})_x(\text{Al}_2\text{O}_3)_{1-x}$ films and have compared them with those of other systems using a proposed model.

2. Experiment

Granular $(\text{Fe}_{65}\text{Co}_{35})_x(\text{Al}_2\text{O}_3)_{1-x}$ films, where x denotes the relative volume fraction of the Fe₆₅Co₃₅

alloy, were prepared by a rf magnetron sputtering from composite targets. The kapton foils used as substrates were not temperature controlled during sputtering. But due to the heating effect of deposition onto substrate, the substrate temperature was raised above ambient temperature and quickly reached a final equilibrium temperature, which was determined to be about 150°C. The thicknesses of granular films are several μm .

After sputtering, the granular film structure was determined by X-ray diffraction and the shapes of granules were investigated by TEM. The magnetic measurements were performed using SQUID and vibrating sample magnetometer.

3. Results and discussion

Figure 1 indicates that the structures of all granular films are body-centered cubic, the same as $\alpha\text{-Fe}$. No other discernible phase has been detected. The width of diffraction peaks becomes larger as the alloy content

Fig. 1. Representative X-ray diffraction patterns for granular $(\text{Fe}_{65}\text{Co}_{35})_x(\text{Al}_2\text{O}_3)_{1-x}$ films.

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Fig. 2. Coercivity versus volume fraction for granular Fe-SiO₂ [2] and (Fe₆₅Co₃₅)_x(Al₂O₃)_{1-x}. The solid and dash lines are the theoretical results for different values of q . Here h_c is normalized by 2500 Oe.

is reduced. From the Scherrer formula [3], the grain size can be determined from X-ray diffraction patterns; the size of granules becomes larger with increasing alloy content.

The coercivity of granular (Fe₆₅Co₃₅)_x(Al₂O₃)_{1-x} films versus volume fraction is shown in fig. 2 as well as the results from ref. [2] for Fe-SiO₂ films. The coercivity of (Fe₆₅Co₃₅)_x(Al₂O₃)_{1-x} rises smoothly as the alloy content increases and reaches 700 Oe at $x = 0.3$. As x exceeds 0.45, H_c drops off sharply. The small value of H_c for $x \geq 0.5$ is due to the formation of a connecting network of granules so that the magnetic particles in the film no longer behave as single-domain particles [1]. Therefore, the magnetic closure patterns can be easily formed and its magnetic properties are similar to the soft magnets. For small alloy volume fractions, the magnetic granular particles are isolated and protected by the nonmagnetic matrix. These ultra-fine particles can be considered as a noninteracting assembly of single-domain particles or superparamagnetic particles. Almost all the existing models assume free-standing particles dispersed in a nonbonding medium and can not explain the anomalously high H_c for the moderate value of volume fraction. Furthermore, the dipolar interactions between particles can even reduce the coercivity in the those models [4].

In our case, however the granules are strongly bonded to the insulating matrix and should be subject to very large stresses. Presumably, the interfacial ef-

fects can dominate the coercivity in such systems. Because of the lattice mismatch on the particle surfaces, strains within the film may induce some anisotropy. This mismatch may be relaxed either by a homogeneous strain or through the introduction of interfacial dislocations [5]. The energy cost of a homogeneous strain is proportional to the volume of the granules, whereas the interfacial dislocation energy is proportional to its surface area. Recently, a rigorous treatment for the elastically symmetric system suggested that the critical dimension (d_c) is less than 10 Å [4]. Therefore, for particle size larger than d_c , the misfit strain anisotropy energy per volume is $K_v = AK_s/V_t$, where A is the interfacial area, V_t is the total volume, and K_s is surface anisotropy induced from the dislocation. Hence, the discrepancy in the coercivity for the above systems can be due to the magneto-elastic anisotropy. This possibility is investigated in details as follows.

We first calculate the average contacting surface of two different materials. Without loss of any generality, the granules are assumed to be small cubes for simplicity. The total expectational surface of N single ferromagnetic particles in contact with the nonmagnetic material is

$$\langle A \rangle = N \frac{a_F}{6} f(x), \quad (1)$$

where N is the particle number, a_F is the surface of a single ferromagnetic particle, $f(x)$ represents the probability of adjacent positions occupied by the nonmagnetic materials, and

$$\begin{aligned} f(x) = & 6(1-x)^6 + C_1^6 5(1-x)^5 x + C_2^6 4(1-x)^4 x^2 \\ & + C_3^6 3(1-x)^3 x^3 + C_4^6 2(1-x)^2 x^4 \\ & + C_5^6 (1-x)x^5. \end{aligned} \quad (2)$$

From eqs. (1) and (2), we can easily derive $\langle A \rangle/V_t$ after some algebraic manipulations. Therefore, the enhancement of coercivity is

$$\Delta H_c = \frac{2K_v}{M_s} = \frac{2K_s}{M_s} \frac{f(x)x}{d(x)}, \quad (3)$$

where M_s is the magnetization and $d(x)$, which is a function of volume fraction, is the length of a small cube.

In order to fit the experimental data, we assume that the granular size increases exponentially with the volume fraction, $d(x) = C \exp(qx)$, where C and q are the adjusting parameters to fit the experimental data. If $d(x)$ is independent of the volume fraction, the expectational value of total contacting surface shows a

peak at $x = 0.5$. However, if the particle size increases with the volume fraction, the peak value will decrease and occurs at a lower volume fraction. In fig. 2, if we arbitrarily normalize the calculated coercivity by the peak value of Fe-SiO₂ data at $x = 0.45$, the peculiar coercivity enhancement for the intermediate range of x can then be explained by the proposed model. For different behaviors of the coercivity between Fe-SiO₂ and (Fe₆₅Co₃₅)_x(Al₂O₃)_{1-x} systems, a possible explanation may be the difference of contacting surfaces in those systems. For large x , the coercivity of the experimental data falls faster than the calculated results. It should be the result of the formation of multi-domain regions when $d(x)$ is larger than a certain critical value. However, the initial increase in H_c is proportional to x in our model, but the experimental data seem to show a x^2 behavior (fig. 2). It is probably because the granules are actually more like spheres than cubes. We also did not consider the size distribution of $d(x)$ here. Our present goal is to use the simplest possible model that can describe the most important features. For future work, the accuracy of

the assumption of $d(x)$ should be justified. The size and shape effects of the granules will be taken into account in more detailed work.

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