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Thickness dependence of magnetic properties of granular thin films with interacting particles

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The effect of film thickness on magnetic properties of $\text{Cu}_{80}\text{Co}_{20}$ granular alloy was studied. It was observed that the susceptibility peak temperature, T_M , strongly increases with the film thickness, t , for $t < 100$ nm. The long-range nature of this effect points to magnetic dipole interaction as responsible mechanism. This dependence of T_M can be explained within the framework of Dormann's theory of dipolar interaction between magnetic particles. The coercive field has different thickness dependence and it is related to formation of magnetic domain structure of Co particles in the granular alloy. © 1999 American Institute of Physics. [S0003-6951(99)03032-6]

It is well known that the dc susceptibility of superparamagnetic systems exhibits a peak as a function of temperature. This phenomenon was interpreted by Néel and Brown as a result of freezing magnetic particles moments below a characteristic temperature, T_M .^{1,2} Below T_M , the energy of thermal excitations is too low to overcome energy barriers to rotate the magnetic moments within the characteristic time of measurement. Above the peak temperature the magnetic moments of the particles are subjected to thermal relaxation, which is a time-dependent stochastic process. Therefore, the peak temperatures determined from ac susceptibility measurements vary with the frequency. Néel and Brown's theory on the static and dynamic behavior of superparamagnetic materials is in satisfactory agreement with experimental data for diluted magnetic particles systems with sufficiently large distances between the particles. This theory was further developed to explain the effects of particle size and anisotropy distributions on T_M .³ T_M was also found to be dependent on the magnitude of the applied magnetic field.²

In the last decade, a growing interest has been developed in better understanding of magnetic properties of granular alloys with high concentrations of magnetic nanoparticles, suitable for certain applications. The nanocrystalline alloys with 60%–80% of magnetic particles embedded in amorphous magnetic matrix demonstrate outstanding soft magnetic properties.⁴ On the other hand, granular systems with 10–40 vol. % of magnetic particles (Fe, Fe–Ni, and Co) in nonmagnetic hosts (Au, Cu, or Ag) exhibit giant magnetoresistance effect. In both kinds of alloys interparticle interactions play an essential role and considerably modify magnetic properties compared to noninteracting particle systems. Recent theoretical and experimental works of several groups made marked progress in understanding static and dynamic properties of granular systems with high concentrations of magnetic particles.^{5–11} They showed that dipolar interaction between magnetic particles influences the susceptibility, and its dependence on measuring frequency and magnetic field.

A study of dipole interaction among nanosized magnetic particles dispersed in nonmagnetic medium in less concentrated limit (<4%) indicated that even in such dilute limit, the magnetic behavior is different from normal spin-glass behavior.¹² The study also revealed that as the particle concentration increases T_M increases in the low field range (<100 Oe), demonstrating the importance of interparticle coupling. In our current work, a new aspect of the interaction in superparamagnetic system is studied. While maintaining a constant particle concentration the effects of the dimensional constraint of the interaction is examined. We studied a series of $\text{Cu}_{80}\text{Co}_{20}$ granular thin films and observed a strong dependence of T_M on the film thickness as a result of decreased dimensionality of the sample. This dependence of T_M can be interpreted within the framework of a theory of magnetic dipolar interaction, by taking into account of far neighbor interactions.¹¹ Such understanding is important in searching for viable magnetic recording media with enhanced magnetic stability.

A series of granular films with their thickness ranging from 7 to 400 nm were deposited on (100) Si substrates using magnetron sputtering. Two S-research guns with elemental Co and Cu targets were biased with dc power supplies. The deposition was carried out simultaneously with different deposition rates for the two sources (0.4 nm/s for Cu and 0.1 nm/s for Co) to achieve desired composition ratio. The 3-in. Si wafers were placed on a planetary motion system, which performed cyclical motion passing over the guns. This deposition mode is equivalent to thorough vapor mixing at atomic level and results in heterogeneous granular films from immiscible elements. The thickness of our films was determined from small-angle x-ray reflectivity measurements (SAXR), by Philip X'pert diffractometer, and was verified by profilometer measurements (Tencor) for thicker samples. The magnetic properties were characterized by quantum design SQUID magnetometer. The zero-field-cooled (ZFC) and field-cooled (FC) magnetic susceptibility curves were typically measured in a small field of 50 Oe. The low and high temperature magnetic hysteresis loops were measured to determine the spontaneous magnetization of the

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FIG. 1. Co-particle size (ϕ_{Co}) for different thickness values (circles refer to Langevin fitting results, squares are for Curie-Weiss method). Inset: an example of fitting (solid line) the Langevin function to the magnetization curve (circles) at 300 K.

single domain particles, and to determine the average magnetic particle size.

The microstructure of selected films was studied by x-ray diffraction and transmission electron microscope (JEOL 2010). The TEM samples were prepared using cross-section sample preparation technique by polishing, dimpling, and ion milling. The granular thin film samples were shown to be continuous and have smooth surfaces, even for the thinnest samples examined (12 nm). This result was verified by SAXR measurement showing well-defined interference peaks in the measured spectra. The composition was checked by electron dispersion spectroscopy in our JEOL 2010 TEM, and was found to be accurate within 0.5% of the nominal value. The quantitative analysis of TEM images is less conclusive due to the difficulty to distinguish the nanosized Co grains from the Cu phases, since both elements have almost the same atomic weight.

The mean size of Co particles was estimated by the measured magnetic properties. This can be done either by measuring the susceptibility and magnetization to fulfill the Curie-Weiss law for interacting particles, or by measuring magnetization curve at a high temperature well above T_M , where the magnetic particles are in superparamagnetic state.^{13,14} The former method is more rigorous but involves three separate measurements (susceptibility, magnetization, and volume), thus prone to errors. The latter technique relies on fitting a high temperature magnetization curve to the well-known Langevin function and involves only a single measurement. The two techniques produce results in good agreement with each other.¹³ In Fig. 1 we show Co-particle size determined by both techniques for films with different thickness values, demonstrating such good agreement. The inset gives an example of Langevin function fitting at $T=300$ K to the magnetization curve for 50 nm thick Cu-Co film. The fitted curve reproduces the measurement quite well and gives a Co-particle size of 2.8 nm. A weak variation of the fitted particle size with the film thickness is within measuring error and the mean diameter of Co particles is 2.8 nm. This value is in excellent agreement with the results determined by x-ray diffraction for cosputtered Cu-Co films in a separate study.¹⁵ One may question the validity of superparamag-

FIG. 2. The dependencies of zero-field-cooled and field-cooled susceptibilities on the temperature for samples with different thickness values: (a) $t = 200$ nm and (b) $t = 25$ nm. The corresponding inverse susceptibility is plotted in the inset of Fig. 1(b).

netism in the presence of interparticle coupling. Due to ferromagnetic nature of the single domain particles typically containing hundreds of magnetic ions per particle, in the field range of the measurement (-5 – 5 T) the interaction between the field and the particle dipole moment dominates over other interactions. Thus, one expects the Langevin description to be valid especially for thinner films whose T_M is substantially lower than that of thicker films to be shown below.

The dc ZFC and FC susceptibility data presented in Fig. 2, for films with two different thickness values, demonstrate typical superparamagnetic behavior. The inset in Fig. 2 demonstrates that the high temperature linear extrapolation of the inverse susceptibility curve intersects the temperature axis at a finite temperature. Such behavior is characteristic of superparamagnetic systems with interacting particles. A supporting evidence for interparticle interactions in our films was the lack of a strong frequency dependence of the real and imaginary susceptibilities. The shift of T_M was only a few degrees in the frequency range from 0.9 to 900 Hz, which is much smaller than expected for noninteracting particles with high T_M values. One may relate T_M to the blocking temperature, T_B , according to conventional theory of magnetic blocking due to anisotropy energy: T_B is proportional to the particle volume, V , as $T_B = KV/30k_B$, where K is the anisotropy constant and k_B is the Boltzmann constant. From the known value¹⁶ of K for the bulk Co, 2.7×10^6 erg/cm³, the estimated T_B is about 7 K, in rough agreement with the measured value ~ 20 K for a powder sample composed of loosely bound Co nanoparticles.¹⁴ Thus, interparticle interaction plays a crucial role in enhancing the magnetic stability.

The effect of interparticle interactions is also manifested in the thickness dependence of T_M as shown in Fig. 3, where we observe a dramatic reduction of the temperature T_M with decreasing thickness, t , of the granular films. The peak temperature changes almost two orders of magnitude in a wide

FIG. 3. The dependencies of the susceptibility peak temperature T_M and coercivity H_C on the film thickness.

thickness range between 7 and 100 nm. This is a finite-size effect due to interparticle interactions because the microstructure of the granular films is evidenced to be almost unaffected by the film thickness. The influence of the thickness on the enthalpy energy of a granular film with interacting particles can be qualitatively understood, because the number of the neighboring interacting particles for a specific particle decreases with decreasing thickness. Among long-range effects dipole-dipole interaction is considered to be dominant, although other interactions may contribute. For example, the RKKY interaction responsible for the exchange coupling in Co/Cu multilayers is likely to exist in the Cu-Co granular system; however, due to arbitrary shape and 3D randomness of the particles the strength of the interaction should be drastically reduced compared to multilayered films.^{11,12} Qualitative estimation of the energy barrier due to dipolar interactions is possible using the following relation:¹¹

$$E_{\text{int}} \cong M^2 V \sum_i n_i b_i L \left(M^2 \frac{V a_i}{k_B T} \right), \quad (1)$$

where for $i=1,2,3,\dots$, n_i 's are the numbers of the first and higher nearest neighbors of the interacting particles with magnetization M . The coefficients, a_i , are given by the formula $a_i = V(3 \cos^2 \alpha_i - 1)/d_i^3$ and dependent on the distances d_i and the position angle α_i between particles, and the coefficients, b_i , are comparable to a_i .¹¹ The function $L(x)$ refers to the Langevin function. In the case of thin films with infinite dimensions in the film plane, the formula [Eq. (1)] can be converted to the continuous space variables and expressed in the following form:

$$E_{\text{int}} \cong M^2 V^2 \langle \rho \rangle 2\pi \oint_{-1/2}^{1/2} \oint_{\delta}^{\infty} \frac{r(3 \cos^2 \alpha - 1)}{(r^2 + \zeta^2)^{3/2}} L \left(\frac{M^2 V^2 (3 \cos^2 \alpha - 1)}{k_B T (r^2 + \zeta^2)^{3/2}} \right) dr d\zeta, \quad (2)$$

where $\langle \rho \rangle$ denotes the mean density of magnetic particles and the integration in the film plane starts at a value δ , corresponding to the distance between nearest neighbors. In the cylindrical coordinate ζ is related to the thickness, t , and $\cos \alpha$ is a function of r and ζ : $\cos \alpha = r/(r^2 + \zeta^2)^{1/2}$. Results of integration of Eq. (2) presented in Fig. 4 indicate that around 100 nm, the energy barrier related to dipolar interaction decreases dramatically, which explains the experimental facts. Similar results, also presented, were achieved using com-

FIG. 4. Calculated energy barrier associated with dipolar interactions between Co particles as a function of the granular film thickness: curve A—according to modified Dormann's model and B—computer simulation of particles array.

puter modeling in which mean dipole-dipole interaction energy of a 3D system with 100×100 particles in plane and variable number of particle planes were considered. A random angular distribution of the particle moments was assumed.

The coercive field (H_C) dependence on the thickness, presented in Fig. 3 has more complicated characteristics than that of T_M . The detailed discussion of this problem is out of the scope of this letter. However, it is worth noticing that H_C , measured at 5 K where the nanoparticles are in ordered state, starts to decrease at a lower film thickness (approximately 30 nm) than in the case of T_M . Recent experiments determined that the magnetic particles in granular films form ferromagnetic domains, which suggests that the coercivity dependence on the film thickness may be associated with the collective switching of ferromagnetic clusters rather than the individual particles.^{17,18}

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