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Physica B 384 (2006) 290–293

www.elsevier.com/locate/physb

Effects of magnetic interparticle coupling in the blocking temperature of granular Co multilayers

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Abstract

In order to study the influence of magnetic interactions on the blocking temperature of magnetic nanoparticles, magnetization measurements were carried out on a discontinuous Co/SiO_2 multilayer. The observed field dependence of the blocking temperature does not fit any of the non-interacting laws commonly used to describe this behavior. We applied a generalized model which considers a field-dependent magnetic correlation length. This model explains well the experimental results and can be used in other nanoparticulate systems.

PACS: 75.50.Tt; 75.20.−g; 75.75.+a

Keywords: Magnetic nanoparticles; Dipolar interactions

1. Introduction

The synthesis of magnetic nanoparticle systems with controlled morphology is very important in view of their interest for device and storage information applications. Moreover, ordered arrays of magnetic nanoparticles deposited on two-dimensional substrates constitute ideal geometry for device applications [1]. The study of the magnetic properties of such systems, with controlled sizes of particles, is particularly interesting because the effect of size distribution is minimized and the effect of magnetic interactions can be controlled. The development of a detailed understanding of the effects of interparticle interactions represents one of the most important challenges to the renewed interest in magnetic nanoparticles [2–4]. The problem is extremely complex because these systems display a rich variety of magnetic configurations, resulting from different contributions to their total energy. The interparticle interactions compete with the magnetic anisotropy in determining the orientation of the particle moments. If strong enough, the interactions can make a collection of individual superparamagnetic moments that behaves like a collective magnetic system. A disordered collective state, or spin cluster glass, is expected when the system is dominated by dipolar interactions. Also, a ferromagnetic state can be formed in the case that the interactions are dominated by exchange coupling, for example, which appears in systems with particles that are in direct contact or dispersed in ferromagnetic amorphous matrix. However, many recent works have indicated the presence of a ferromagnetic correlation length larger than the size of the particles in systems with concentration of particles lower than the percolation, both in insulating [5,6] and metallic (paramagnetic) matrix [7], leading to the so-called superferromagnetism. This fact opens the possibility to analyze the interaction-induced variations of the superparamagnetic properties using the random anisotropy model (RAM). In this paper, we present a quantitative analysis of the field dependence of the blocking temperature in magnetic nanoparticles immersed in an insulating matrix. We apply a phenomenological model that takes

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doi:10.1016/j.physb.2006.06.013

into account particle coupling effects [8]. The model explains the experimental results with accuracy and, in principle, can be used in other nanostructured magnetic systems.

2. Experiment

The study of the influence of magnetic interactions on the blocking temperature was carried out in samples of discontinuous Co/SiO$_2$ multilayers, grown by DC (Co) and RF (SiO$_2$) sputtering on Si substrates held at room temperature. The sample consists of almost periodically arranged Co nanoparticles immersed in a SiO$_2$ matrix. More details on the production method can be found elsewhere [9]. Cross-section transmission electron microscopy (TEM) characterization was performed using a Jeol JEM-3010 ARP microscope. Magnetization properties were measured in the Quantum Design MPMS XL7 system in the temperature range 5–300 K.

3. Results and discussion

The TEM image of Fig. 1 shows that the sample is composed of almost periodically arranged Co nanoparticles. The particle size distribution (see also Fig. 1) was obtained by measuring the particle sizes in images of several different regions of the sample, and it is well fitted by a Gaussian distribution, leading to an average diameter of 3 nm. Fig. 2 shows the zero-field cooling/field cooling curves (ZFC/FC), measured for different applied DC magnetic fields H. One can observe that the maximum of the ZFC curves, which is related to the mean blocking temperature, displaces toward lower temperatures for increasing H, as expected.

The effect of external magnetic field on the blocking temperature has been considered for uniaxial magnetic systems by several authors [10,11]. A simple analytical expression is usually employed

$$T_B(H) = \frac{KV}{k_B \ln(\tau_m/\tau_0)}\left[1 - \left(\frac{H}{H_k}\right)\right]^{\alpha}, \quad (1)$$

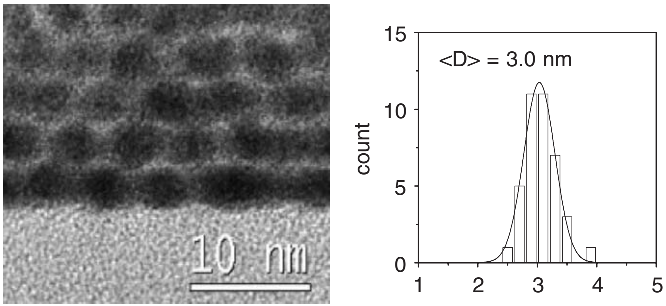

Fig. 1. Cross-section TEM image of the granular multilayer (left). The average diameter of the particles is 3 nm, as shown in the size distribution (right).

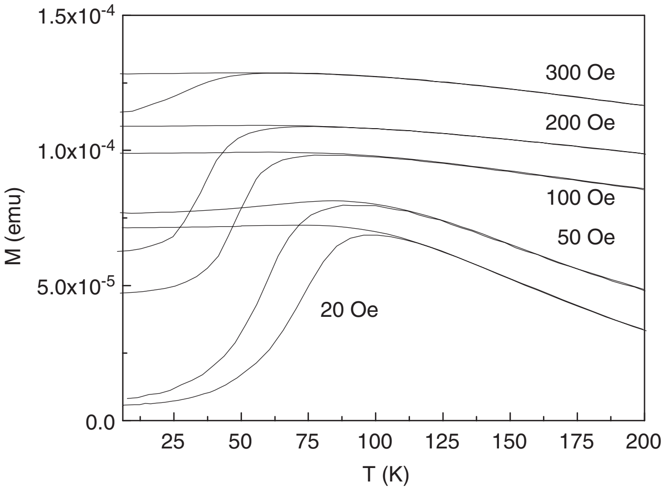

Fig. 2. ZFC, FC magnetization curves, measured at five different fields H.

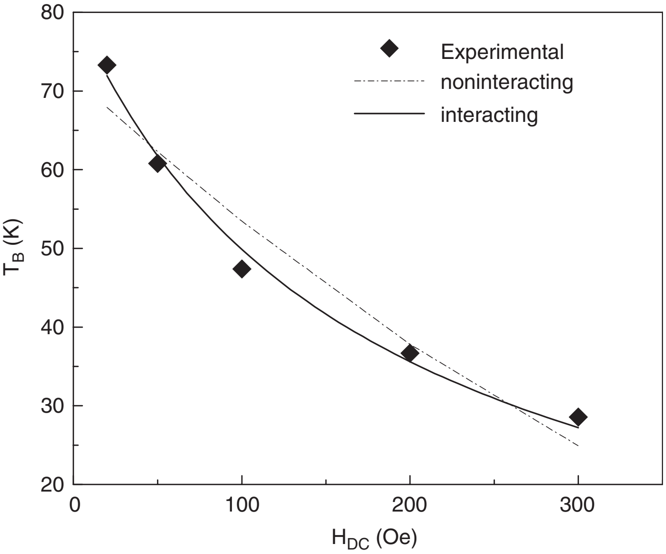

Fig. 3. The field dependence of the blocking temperature. Fits by using Eq. (1) (dashed-dotted line) and the modified RAM expression, given by Eq. (2) (solid line).

where K is the uniaxial anisotropy constant, V the particle volume, k_B is the Boltzmann's constant, and τ_0 is a constant related to gyromagnetic precession, and may be considered to be of the order of 10^{-9}–10^{-10} s. The relaxation time $\tau = \tau_0 \exp(KV/k_BT)$ is a fundamental quantity of these systems that describes how rapidly the magnetization reversal of the particles occurs by thermal activation [12], and the blocking temperature T_B, for a system of particles with mean volume V, is defined as the temperature at which $\tau = \tau_m$, the measurement time (typically in the order of 100 s for DC measurements). H_k is the anisotropy field ($H_k = 2K/M_S$, M_S being the saturation magnetization) and the exponent α is close to 1.5 [13].

Table 1
Experimentally measured particle size (TEM) and fitting parameters obtained by means of the uncoupled particles model (Eq. (1)) and the modified RAM (Eq. (2))

	Results obtained by different studies			
TEM	Uncoupled particles model		Coupled particles model	
D(nm)	D (nm)	K (erg/cm^3)	L_0 (nm)	K (erg/cm^3)
3.0	10.3	0.4×10^6	15.5	2.7×10^6

Fig. 3 shows the measured blocking temperature T_B as a function of magnetic field H for the sample. We tried to fit the experimental data to the non-interacting model described above. The dashed line in Fig 3 is the result of the best fit to Eq. (1), by using $\alpha = 1.5$, K and the particle diameter D as free parameters and the M_S of bulk Co (1420 emu/cm^3). As can be observed in Table 1 (uncoupled particles model) the D values obtained from the fits are much larger than the D values obtained by the TEM analysis; on the other hand, the K values estimated through the fit turn out to be well below those of bulk Co. This disagreement between the experimental points and the theoretical prediction is usually imputed to the presence of interparticle interactions [8].

A different approach that takes into account the interaction effects on the field dependence of $\langle T_B\rangle$ can be carried out in the framework of the RAM. According to this model, the anisotropy is averaged to an effective value $K_{\rm eff}$ within the correlation length due to the magnetic interactions, whose value decreases with increasing correlation length due to a statistical random-walk effect [14].

The relevant parameters for the magnetization processes of a system of ferromagnetically correlated particles, with correlation length L, are the effective anisotropy and the effective volume of the particles (Λ) in the correlation volume (L^3). Two simple modifications of the RAM expressions for $K_{\rm eff}$ and Λ that account for nanoparticles in a non-magnetic matrix are

$$K_{\rm eff} = K/\sqrt{N}; \quad \Lambda = \frac{\pi}{6}[D + x(L^3 - D^3)],$$

where N is the number of correlated particles, i.e.,

$$N = [1 + x(L^3 - D^3)/D^3].$$

Both expressions tend, respectively, to the anisotropy and volume of an individual particle when interactions are very weak and $L\to D$. On the other hand, when $L \gg D$, they tend to the usual relations used in the study of correlated particles systems (see Refs. [15,16]).

By substituting the anisotropy and volume of individual particles in Eq. (1) by the effective anisotropy and particle volume, $K_{\rm eff}$ and Λ, respectively, one can calculate the field dependence of the blocking temperature for coupled particles in terms of the structural parameters of nanoparticulate systems

$$T_B = \frac{K\pi\left[D^3 + x(L_H^3 - D^3)\right]}{6k_B\ln(\tau_m/\tau_0)\left[1 + x(L_H^3 - D^3)/D^3\right]^{1/2}} \times \left[1 - \left(\frac{H_{DC}M_S\left[1 + x(L_H^3 - D^3)/D^3\right]^{1/2}}{2K}\right)\right]^{1.5}, \tag{2}$$

where $L_H = D + [2A_{\rm eff}/(M_sH_{DC} + C)]^{1/2}$ is the experimentally observed [17] correlation length expressed as a function of the applied field, and $A_{\rm eff}$ represents the interaction intensity which, for nanocrystalline alloys, is the intergranular exchange constant A [18].

We have used this expression to fit our experimental data of Fig. 3. The fits, represented by the solid lines, were carried out by using the experimental D values (estimated by TEM), $x = 0.35$ (estimated from sputtering parameters), $M_S = 1420\,{\rm erg/cm^3}$ (the bulk Co value) and $A = 3.1\times10^{-7}\,{\rm erg/cm}$, and K and C as free parameters. We expect that the parameter C approach zero for systems of particles clustered together and increase with the progressive dilution of particles, reaching a value $C\approx 2A_{\rm eff}-M_SH$ for systems composed of independent particles.

Expression (2) provides a very good description of the field dependence of T_B for our sample by employing $K_{\rm eff}$ values of the order of magnitude of the bulk anisotropy of Co. Also, the zero applied field correlation length value (L_0), calculated using the fitted C parameter ($L_0 = D + (2A_{\rm eff}/C)^{1/2}$), is much larger than the diameter of the nanoparticles, as shown in Table 1. In the case of granular systems with just dipole–dipole interactions, there are strong indications of the presence of correlations among the particles with a characteristic length longer than the particle size [5,6,8,19].

In conclusion, we have presented a quantitative analysis of the field dependence of the blocking temperature of Co nanostructured multilayers. We applied a phenomenological model that takes into account particle coupling effects in superparamagnetic properties, based on a modified RAM. The model has been also employed in co-sputtered granular systems, explaining the experimental results with accuracy [8]. In principle, this model could be used in any magnetic nanoparticulate system, because it does not depend on the specific coupling mechanism.

Acknowledgments

This work was financially supported by FAPESP and CNPq (brazilian agencies), and Millennium Science Nucleus "Condensed Matter Physics" P02-054F of Chile. The TEM images were performed at the *Laboratório de Microscopia Eletrônica* (LME—LNLS), Campinas, Brazil. The samples were produced at LMMM in collaboration with L.S. Dorneles and L.F. Schelp.

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