Journal of Optoelectronics and Advanced Materials Vol. 4, No. 2, June 2002, p. 311 - 314

TEMPERATURE DEPENDENT INTEGRAL GENERALIZED DeltaM CURVES

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The effect of inter-particle interaction in samples of Co nanoparticles dispersed in different volume fractions in a wax matrix was studied. The evolution of interaction intensity in the Co nanoparticle systems as a function of temperature was determined by measuring the Integral Generalized DeltaM curves. These experimental curves do not require AC demagnetized states of the sample, which is an important simplification of the experimental procedure. The results are analyzed with a Generalized Moving Preisach Model.

(Received February 9, 2002; accepted May 15, 2002)

Keywords: Cobalt nanoparticles, Generalized Moving Preisach model

1. Introduction

The dynamics of magnetic nanoparticle systems is a subject of considerable interest due to their fundamental and technological interest. The properties of such systems are strongly modified compared to the bulk, due to granular texture and the small size of the grains. On one hand, thermal relaxation of the magnetic moment of the particles, i.e. superparamagnetic relaxation, can occur. On the other hand, magnetic inter-particle interactions always exist in fine particle systems, and they can be best probed in particles dispersed in a matrix where the volume concentration can be systematically varied. The demand for high-density recording media requires high packing densities that make the role of inter-particle interactions very important. Consequently, many theoretical and experimental works were devoted to understanding the role of interactions on the magnetic properties of fine particle systems.

2. Experimental setup. Samples. Experimental results

A number of time and temperature dependent experimental techniques have been used in order to study the magnetic properties of nanoscale systems [1]. Among them, the techniques involving small ac magnetic field excitations are extremely useful [2]. In order to investigate the role of the interactions, we performed experiments at various temperatures on samples of Co nanoparticles, dispersed in different concentrations in a wax matrix. The degree of dilution in the wax controls the average particle distance and therefore the strength of interactions.

The synthesis of cobalt nanoparticles was carried out using standard organometalic reaction procedures with airless/moisture-less devices and commercially available reagents. The common approach for the synthesis of cobalt nanocrystallites is to reduce organo-cobalt salt in a non-polar solvent at relatively high temperature, capped/stabilized by some organic compounds. The experimental procedure is similar to that reported by S. Sun et al. [3]. The dispersion of Co nanoparticles in the wax matrix was carried out by ultrasonically mixing the concentrated cobalt nanocrystallites in hexane with wax-hexane solution, followed by increasing temperature of the system to remove the solvent. The ratio of cobalt was determined based on the real amount of cobalt used and amount of wax. The size and size distribution of each filtrated sample in as-prepared cobalt

colloidal solution was monitored using light scattering technique (DynaPro 99 Molecular Sizing Instrument). The obtained Co nanoparticles are spheroidal with an average diameter of 6 nm as determined by transmission electron microscopy. In order to study the effect of interparticle interactions two samples were considered for this study: a sample of $C_V = 0.011$ volume fraction of Co nanoparticles dispersed in wax (Co10) and a sample containing only Co nanoparticles (Co100). The nanoparticles in both samples were from the same batch of Co nanoparticles, to make sure the same size distribution in both samples.

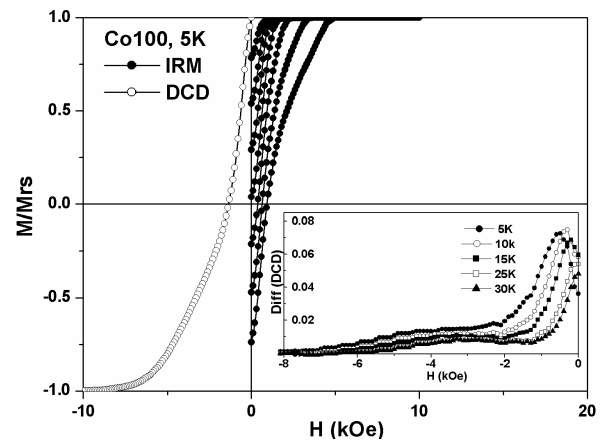


Fig. 1 The forward IRM and the DCD curve for the Co100 sample at T=5K. The inset shows the Switching Field Distribution for the same sample at various temperatures.

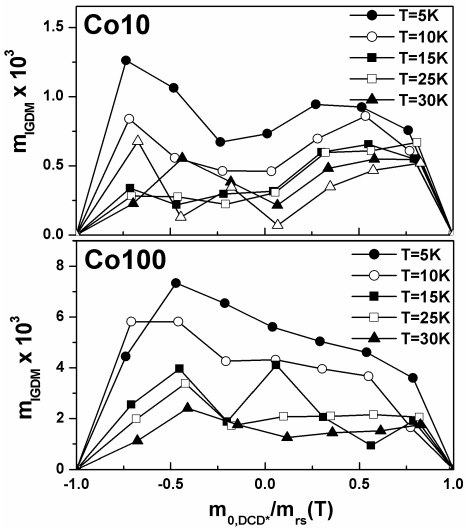


Fig. 2 The IGDM's for the Co100 (a) and Co10 (b) samples at various temperatures.

An analysis of the inter-particle interactions in the two samples was done by measuring the remanent curves, dc demagnetization (DCD) and forward isothermal remanent magnetization curves (IRM). First of all in order to evaluate the evolution of interaction intensity in the systems as a function of temperature, we have measured the Integral Generalized DeltaM (IGDM) at 5 K, 10 K, 15 K, 20 K, 25 K and 30 K [4][5]. We used this methodology to avoid the necessity to produce experimentally an accurate AC demagnetized state. The DCD and the forward IRM curves were measured for seven initial remanent states. The data for C100 at 5K are presented in Fig. 1. For each forward curve the IGDM is calculated. In order to compare the IGDMs at different temperatures they were translated and normalized on the abscissa at the saturation remanent moment for each temperature. The translated IGDM curve was given by the formula:

$$m_{IGDM} = \int_0^{H_m} [m_{DCD*}(H) - m_{DCD}(H)]dH \qquad (1)$$

where H_m is the maximum field applied to the sample in the experiments, m_{DCD} is the magnetic moment in the well known DCD process and the m_{DCD*} is the DCD-like curve calculated using the forward IRM curve, as described in detail in [4]. The moment m_{IGDM} is represented as a function of the initial remanent state used in the IRM forward curve. In Fig. 2 one presents the IGDM curves for the Co100 and Co10 samples as a function of temperature. The Co100 sample IGDMs have a clear central maximum which is characteristic to a system with important static statistic interactions [6]. The static statistic interactions intensity is decreasing with the increase of temperature, as expected. The asymmetry of the IGDM curve could be explained by the asymmetric coercive field distribution. The Switching Field Distributions (SFD), calculated as derivatives of the DCD curves, for different temperatures, confirm this asymmetry (see inset Fig. 1). The Co10 sample IGDMs are significantly different of the Co100 sample. They show a central minimum that separates two maxima. This is characteristic for systems in which the mean field or exchange interactions are more important than the static statistic interactions. It is easy to understand that the statistic interactions intensity in the

Co10 sample is less important than in the Co100 sample. The presence of exchange-type interactions in this sample is probably linked with the presence of clusters in the system. As in the other case, the static interaction intensity, measured by the IGDM, is decreasing with the temperature increase.

3. Preisach-type model. Simulations

The Generalized Moving Preisach (GMP) model we used to simulate the temperature dependent IGDM's is completely described by two distributions, (the irreversible Preisach distribution $P_i(H_\alpha, H_\beta)$, and a singular reversible Preisach distribution $P_r(H_\alpha)$ that is non zero only on the first bisector of the Preisach plane) and the "moving" parameter [7]. We used instead of the switching fields (H_α, H_β) coordinate system the system denoted by (h_c, h_i) and rotated through 45° with respect to the first set where,

$$h_c = \left(\sqrt{2}/2\right)(H_\alpha - H_\beta) \quad \text{and} \quad h_i = -\left(\sqrt{2}/2\right)(H_\alpha + H_\beta). \tag{2}$$

We assume that the coercive and interaction field distributions are statistically independent and that the coercive field distribution is log-normal with a Gaussian interaction field distribution. The irreversible part of the Preisach distribution is given by:

$$p_i(h_c, h_i) = \frac{S}{2\pi h_{c\sigma} h_{i\sigma}} \left\{ \frac{1}{(h_c/h_{c0})} \exp\left[-\frac{\ln^2(h_c/h_{c0})}{2(h_{c\sigma}/h_{c0})^2} \right] \right\} \left\{ \exp\left[-\frac{h_i^2}{2h_{i\sigma}^2} \right] \right\} \tag{3}$$

and the reversible part,

$$p_r(h_i) = \frac{(1-S)}{2h_{r\sigma}} \left\{ \exp\left[-\frac{|h_i|}{h_{r\sigma}} \right] \right\} \tag{4}$$

where the following notations are used:

- S is the weight of the irreversible part;
- $h_{c\sigma}, h_{i\sigma}, h_{r\sigma}$ are parameters describing the dispersions of the coercivity, interaction and reversible part distributions, respectively;
- h_{c0} is related to the position of the maximum of the coercivity distribution (in fact, the position of the maximum, $h_{c,max}$, is given by: $h_{c,max} = h_{c0} \exp(-h_{c\sigma}^2/h_{c0}^2)$).

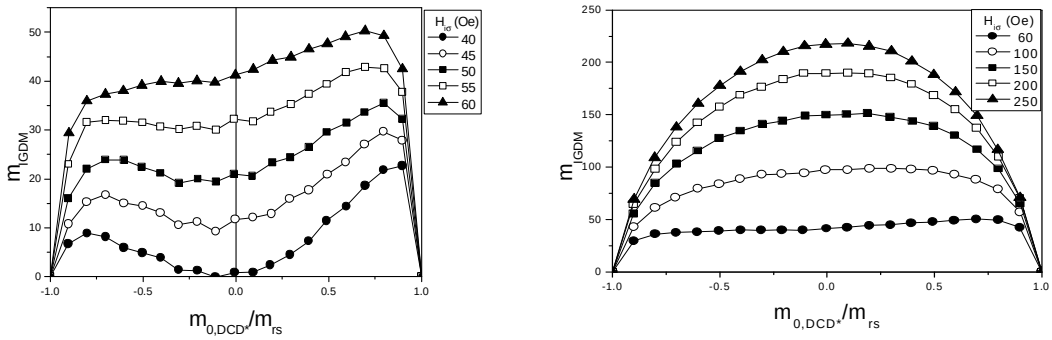


Fig. 3. Simulated IGDM with the GMP model with S= 0.85; α= 90 Oe (the moving parameter); h_{c0}= 1160 Oe; $h_{c\sigma}$ = 300 Oe; $h_{r\sigma}$ = 2262 Oe; and for different values of the h_{is} parameter describing the interactions distribution (a) 40, 45, 50, 55 and 60 Oe., (b) 60, 100, 150, 200 and 250 Oe.

With this GMP model we have simulated the IGDM set of experimental data for the two samples, in a simplified manner. We wished to check if our assumption that the change in the statistical interactions due to blocked particles in the system can explain qualitatively the shape of the experimental data described in the previous section.

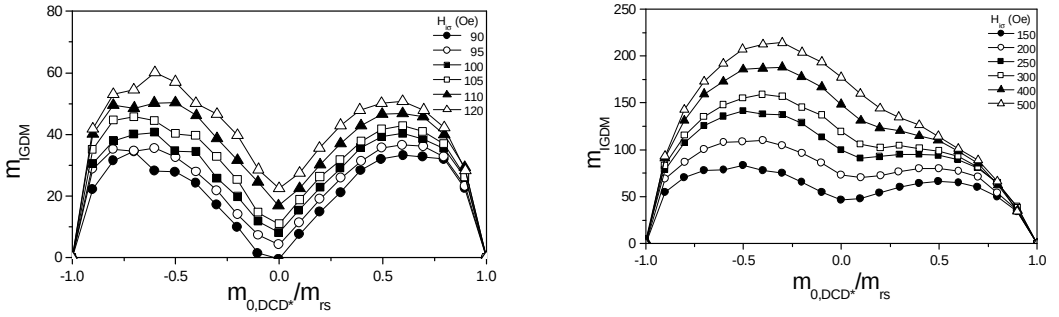
First we have simulated the IGDM's changing only the interaction field distribution dispersion.

The two sets of simulated IGDM's presented in Figs. 3 (a and b) show the same general shape as the experimental IGDM's from Fig. 2 (Fig. 3(a) for the Co10 sample and Fig. 3(b) for the Co100 sample). The main difference is that the simulated IGDMs have systematically greater values for positive values of the initial remanent moment than for the negative values while in the experimental IGDMs the curve has greater values for negative initial remanent states than for the positive ones.

This is usually considered to be due to the asymmetry of the critical field distribution (log-normal in the simulations). As we can see on the SFD obtained by making the derivative of the DCD curve, our system seems to have a peculiar critical field distribution with two maxima. We have changed the critical field distribution in the GMP model with the distribution:

$$P(h_c) = \frac{1}{2}\left[\frac{1}{\sqrt{2\pi h}_{c\sigma 1}}\exp\left(-\frac{(h_c - h_{c01})^2}{2h_{c\omega 1}{}^2}\right) + \frac{1}{\sqrt{2\pi h}_{c\sigma 2}}\exp\left(-\frac{(h_c - h_{c02})^2}{2h_{c\sigma 2}{}^2}\right)\right] \quad (5)$$

With the new critical field distribution extracted from the experimental SFD the IGDMs as a function of the static statistical field distribution dispersion, the simulated set of IGDMs is in a much better qualitative agreement with the experiment (see Figs. 4).



a b

Fig. 4. Simulated IGDM with the GMP model with distribution of the critical fields as in the formula (5), with S = 0.85; α = 90 Oe; h_{c01} = 450 Oe; h_{c02} = 2000 Oe; $h_{c\sigma 1}$ = 150 Oe; $h_{c\sigma 2}$ = 800 Oe. $h_{r\sigma}$ = 2262 Oe; $h_{i\sigma}$ = 150 Oe; and for different values of the h_{is} parameter describing the interactions distribution (a) 90, 95, 100, 105, 110 and 120 Oe; (b) 150, 200, 250, 300, 400 and 500 Oe

4. Conclusions

In this paper we have presented a new method of experimentally evaluating the interactions evolution as a function of temperature and system packing factor in nanoparticulate ensembles. IGDM simulated curve shows the diminishing of the static interactions due to blocked particles as the temperature increases and the packing factor decreases. The GMP model is able to describe this evolution of the IGDMs when the critical field distribution is taken as a function similar to the experimental SFD.

Acknowlwdgement

Work at AMRI was supported through DARPA grant No. MDA 972-97-1-0003 and work in Iasi was supported by Romanian CNCSIS under the grants A/2001 and D42.

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