Relaxation effects in interacting nanostructured particulate systems

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(Produced on 1 November 2005; published online 26 April 2006)

In this paper we have systematically studied the effect of interparticle dipolar interaction on the magnetic relaxation of a system of interacting uniaxial, single domain spherical particles lying in a regularly network formed within one layer. We have generated structured samples, with narrow distributions of the dimension and orientation of the particles, and we have analyzed the evolution of the interaction field distribution (IFD) during the magnetic relaxation at different temperatures. It is showed that IFD is state dependent and that the magnetic viscosity does not monotonically increase with the temperature. © 2006 American Institute of Physics. [DOI: 10.1063/1.2176332]

INTRODUCTION

The study of nanoparticulate assemblies is important from both fundamental and technological points of view. In these systems thermally activated reversal of magnetization is essential. When an applied magnetic field is changed, magnetic materials present a characteristic time dependence of the magnetization and this phenomenon is generally called magnetic viscosity.

Néel\textsuperscript{1} assumed that the particles are subjected to thermal fluctuations which can be represented by a fictitious magnetic field. Under constant environmental conditions, the fictitious field occurs randomly with the passage of time. Street and Wooley adopted a slightly different but essentially equivalent point of view, using the concept of activation energy.\textsuperscript{3,4} They considered that within a magnetic material there is a distribution of energy barriers which must be overcome by the magnetization in order to change its direction. These energy barriers can be overcome through energy supplied by thermal fluctuations and are a function of the applied field. For a uniform distribution of energy barriers at a fixed field the magnetization $M$ decays with time $t$ as\textsuperscript{5}

\begin{equation}
M(t) = M_0 + S \log t,
\end{equation}

where $M_0$ and $S$ are constants dependent on the initial conditions. The magnetic viscosity coefficient $S$ is given by

\begin{equation}
S = S_{\text{irr}} \chi_{\text{irr}},
\end{equation}

where $\chi_{\text{irr}}$ is the irreversible magnetic susceptibility and is associated with an irreversible jump over an energy barrier. Also it is shown that $S$ depends linearly with temperature. Assuming the same change in magnetization for each jump it is showed that $S_{\text{irr}}$ is connected with the energy barriers:

\[ S_{\text{irr}} = -k_B T (dE/dH), \]

where $T$ is the temperature, $k_B$ Boltzmann’s constant, $E$ the energy barrier, and $H$ the magnetic field.

Magnetic relaxation is a rather complex phenomenon and many models have been proposed to explain it. In these systems the particle interactions are also essential. When the relaxation is important the interaction field is fluctuating at a high rate. These interactions are qualitatively different from the static ones, and they are called dynamic interactions. In a previous study\textsuperscript{5} we have observed that in Langevin-type simulations made on nanostructured materials the decreasing of the temperature determines a multiple peak structure of the interaction field distribution (IFD), similar to the one observed in the two-dimensional (2D) Ising-type simulations.\textsuperscript{6,7} In this paper we are extending the analysis of IFD during the relaxation phenomena at a given temperature starting on different points of field cooled (FC) magnetization curve at the field removal.

MICROMAGNETIC MODEL

We have made simulations on systems of a few thousands of interacting (magnetostatic interactions) uniaxial single-domain spherical particles lying in a regular network formed within one layer in the $xz$ plane so that the fast Fourier transform (FFT) technique can be applied in order to compute the field due to all particles. We have generated samples with the easy axes uniformly distributed within a cone of semiangle $5^\circ$ with axis into the sample plane, along the $Oz$ axis, with the volume dispersion $\sigma_v=0.1$ in regard to the mean value $V_{\text{mean}}=1.5 \times 10^{-26}$ m$^3$ and with the same uniaxial anisotropy constant $K=700$ kJ/m$^3$. The saturation magnetization is $M_s=1440$ kA/m. The distance $D$ between particles was varied in order to observe the variation of interaction intensity. Periodic boundary conditions are used in order to avoid the effect of the demagnetization field. No temperature dependence of anisotropy constant and satu-
RATION magnetization is taken in our simulations and only a fluctuating stochastic field is added to the effective field. The stochastic field changes the deterministic motion of the magnetization into a random walk. The stochastic Landau-Lifshitz-Gilbert equation is numerically integrated using the Heun method. Garcia-Palacios and Lazaro showed that the equation has to be interpreted in the sense of Stratonovich, in order to obtain the correct thermal equilibrium properties. The magnetic properties follow from averages over many numerical realizations of the dynamic process.

RESULTS

In FC process the magnetic moment is measured while cooling the sample in an external field. In our simulation the magnetic field is applied in the plane of the sample along the $Oz$ axis and is equal to 5% of the anisotropy field. Starting from different points of FC curves we have suppressed the magnetic field and we have simulated the relaxation curves in zero fields for two values of the distance between particles. Also we have simulated the relaxation curves started on a thermally demagnetized state, when a field $h_z=0.05$ is applied along the $Oz$ axis, like in a zero field cooled (ZFC) process. One observes that the time dependence of the magnetization cannot be treated simple with a relation of the type (1), that the relaxation rate (magnetic viscosity) is time dependent. However, one can identify an approximately linear regime in the initial and in the final regions of semilog plot from Figs. 1 and 2. For each temperature we have determined the slope of these linear portions in order to obtain the magnetic viscosity. In the final portions of the relaxation curves the magnetic viscosity practically does not depend on the temperature. Otherwise in the initially portions the magnetic viscosity is temperature dependent (Fig. 3), but not

FIG. 1. The normalized magnetization decay after switching off the external field, starting from different points of FC curve, for two values of the distances between particles: $D=5$ nm (a) and $D=7$ nm (b).

FIG. 2. The normalized magnetization increase in the field $h_z=0.05$, starting on a thermally demagnetized state.

FIG. 3. The absolute values of the magnetic viscosity coefficient as a function of temperature.

FIG. 4. The distribution of the interaction field projection on the $Ox$ and $Oz$ axes at the initial moment ($t=0$) of the curves obtained in zero field.

$T(K)$
through a linear dependence, as the Street-Wooley theory predicts.

Also in each magnetization state of the system, we have calculated the interaction field which acts on each particle and then we have determined the distribution of the interaction field. The interaction field projections on the $Oy$ axis are very small because the particles’ magnetic moments are predominantly oriented in the $xz$ plane. At the initial moment ($t=0$) of the curves obtained in zero field we have Gaussian distributions of the interaction field projections on the $Ox$ and $Oz$ axes (see Fig. 4) for all values of the temperature, but the distributions’ dispersions decrease with the temperature because the amplitude of the thermal fluctuations increases with the temperature.

During the relaxation important changes of the IFD occur. If the distribution on the $Ox$ axis has a Gaussian allure always, one observes the appearance of a multiple peak structure in the distribution of interaction field projections on the $Oz$ axis (see Figs. 5 and 6). The variations of interaction distribution are more important for higher temperature that can be associated to a lower order in the system.

CONCLUSIONS

The magnetization variation during the relaxation processes does not follow a universal logarithmic law as the Street-Wooley theory predicts. In the initial portion of the relaxation curves the magnetic viscosity increases with the temperature by a nonlinear law, while in the final portion the magnetic viscosity is temperature independent. Also the distribution of the interaction magnetic fields is strongly dependent on the sample temperature and it is drastically changing during relaxation processes.

ACKNOWLEDGMENTS

Work at AMRI was supported by DARPA under Grant No. HR0011-05-1-0031. The support from Romanian CNC-SIS under the Grants A and Ac NANOCONS is also acknowledged.