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Modeling of Long-Time Thermal Magnetization Decay in Interacting Granular Magnetic Materials

O. Chubykalo-Fesenko[1] and R. W. Chantrell[2], *Member, IEEE*

[1]Instituto de Ciencia de Materiales de Madrid, CSIC, Madrid 28049, Spain
[2]Physics Department, York University, York YO10 5DD, U.K.

We present a general method to evaluate the long-time magnetization decay in granular magnetic systems. The method is based on Arrhenius–Neel kinetics with the evaluation of the energy barriers in a multidimensional space. To establish a possible reversal mode, we suggest the use of Metropolis Monte Carlo and for the mode statistical sampling—the kinetic Monte Carlo criteria. The examples considered include long-time magnetization decay in CoCrPt low-magnetization longitudinal recording media and in a collection of Co particles with different concentrations.

Index Terms—**Energy barrier, magnetic viscosity, micromagnetics, numerical methods.**

LONG-TIME thermal magnetization decay is an important characteristic of granular materials, both from the fundamental and applied points of view. The magnetization decay results from a complex interplay between microstructure and magnetic interactions, which leads to energy barrier distributions. Thermal fluctuations cause a random walk of the system in a multidimensional energy space, which ultimately produces a decay of the initial magnetization state. From the computational point of view, the modeling of such a process without further simplifications is rather complicated, and most authors adopt Arrhenius–Neel kinetics and a simplified way to evaluate the energy barriers based on the local field [1], [2]. Generally speaking, this approach is only valid in the case when no collective reversal takes place and the interactions produce a mere perturbation on the original energy barrier. In the opposite case, the energy barriers should be evaluated in a multidimensional space [3]–[6]. In this paper, we present a method capable of calculating long-time magnetization reversal in strongly interacting magnetic materials.

In our approach, the low barriers in the systems (if they exist) are overcome by integrating the stochastic Landau–Lifshitz–Gilbert equation. For higher barriers, similar to the general kinetic Monte Carlo approach, our method is based on the idea of the infrequent-event approach. As expected in this case, the Arrhenius–Neel kinetics is applied, with the probability of surmounting a particular energy barrier given by

$$f = f_0 \exp(-\Delta E_B / k_B T). \tag{1}$$

where ΔE_B is the energy barrier, k_B the Boltzman constant, and T is the temperature of the system. Differently from all other previous approaches, the energy barrier ΔE_B is evaluated in a multidimensional space [5], [6], thereby taking into account the possible collective character of the reversal mode.

Different stages of the method are outlined as follows.

1) An initial magnetization state is established by minimizing the energy. In the present paper, this is done by following the hysteresis cycle, i.e., starting with the saturation state and slowly decreasing the external field until the remanent magnetization state at zero field is achieved.
2) The stochastic Landau–Lifshitz–Gilbert equation is integrated during 10 000 time steps in order to overcome small energy barriers, which could be present in the system.
3) To evaluate the reversal frequency f_0 in the full harmonic (Venyard) approximation [7], a normal mode analysis is performed in the minimum. f_0 is then given by

$$f_0 = \frac{\alpha\gamma}{(1+\alpha^2)\pi M_s V} \frac{\prod_{i=1}^{N} \omega_i^{\min}}{\prod_{i=2}^{N} \omega_i^{\max}}. \tag{2}$$

Here, α is the damping parameter, γ the gyromagnetic factor, M_s the saturation magnetization, V the particle volume, $\omega_i^{\min}$ the eigenfrequencies of the energy functional evaluated in the minimum, and $\omega_i^{\max}$ the positive eigenfrequencies evaluated at the saddle point. For a similar evaluation of the reversal frequency, see also [8].
4) To force the system to choose a physically reasonable reversal path, we use a thermal acceleration method based on the Metropolis Monte Carlo technique with a slowly increasing temperature. Namely, at each temperature, we perform 10 000 Metropolis Monte Carlo steps. Each 1000 steps, the energy is minimized to determine if the system converges to a new minimum defining a new stationary state of the system. The estimated path is used as an initial guess to evaluate the energy barrier in a multidimensional space. Alternatively, the choice of the initial path could be done using Langevin dynamics with increasing temperature such as in the temperature acceleration method developed by Voter [9].
5) To determine a first approximation to the saddle point separating the two minima, we use the ridge optimization method [10]. Specifically, the roughly estimated maximum is used as an initial guess $E^1_{\max}$. The step Δx_0 is taken in both sides of the maximum along the current trajectory obtaining points $A_1^{(1)}$ and $A_2^{(1)}$. These points are moved by a step Δx_1 in the directions $-\mathrm{grad}\, E(A_1^{(1)})$

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and $-\mathrm{grad}\, E(A_2^{(1)})$, where E is the multidimensional energy function of magnetization vector. Subsequently, the new points are joined by a straight line, and a linear search for the maximum $E^2_{\max}$ is made. Adaptive step size for Δx_0 and Δx_1 is used to ensure that there exists a maximum between the points $A_1^{(i)}$ and $A_2^{(i)}$. The step size is decreased each $N = 20$ steps, provided that the absolute value of the gradient at the maximum is decreased. The sequence of points $E^i_{\max}$ converges to the minimum on the ridge giving a crude approximation to the vicinity of the saddle point.

6) To converge to the saddle point with a given precision, the augmented Hessian method [11] is used. The method is similar to that of the Newton method, but the corresponding Hessian matrix is augmented by gradients. The method converges to the nearest saddle point with a larger convergence radius than that of the Newton method.
7) Finally, a normal mode analysis is performed at the saddle point to ensure that the negative eigenvalue is unique. This check is necessary to be sure that the point found is not a higher order saddle, which may artificially couple two independent energy barriers. In addition, the normal mode analysis provides the means to evaluate the reversal frequency (2) and the reversal probability (1).
8) We have found it necessary to check that the obtained saddle point separates the basin of attraction of two minima, of which one is the initial state. This is done by minimizing the energy starting from the final points $A_1^{(i)}$ and $A_2^{(i)}$.
9) The search for the reasonable trajectory (stage 4) is repeated several times in order to evaluate other possible reversal modes. If those exist, then the reversal probability is a sum of possible reversal probabilities corresponding to each reversal mode.
10) The reversal modes are statistically sampled using the kinetic Monte Carlo criterion [12] and the time—from the exponential distribution with a probability

$$P_i(t)\, dt = f_i \exp(-f_i t)\, dt. \tag{3}$$

11) The total procedure is repeated starting from stage 2 and the new magnetization minimum. The probability to be moved p_i is calculated for each particle as a number of times the particle changed its magnetization over the total number of the kinetic Monte Carlo steps (the number of times the magnetization state is changed). To improve the performance of the method, the particles with the $p_i > 0.1$ are frozen each N_1 kinetic Monte Carlo steps, the particles with $p_i > 0.01$ are frozen each $2N_1$ kinetic Monte Carlo steps, ... the particles with $p_i > 10^{-k}$ are frozen each kN_1 kinetic Monte Carlo steps. The particles are frozen only for the Metropolis Monte Carlo procedure (stage 4) but not for the determination of the multidimensional energy barriers (stages 5–7). We used $N_1 = 5$. This procedure is necessary to avoid the time sampling process from being dominated by the rapidly reversing (superparamagnetic) fraction. The effects of the superparamagnetic fraction are represented by a contribution to the energy barrier from a statistically significant "snapshot" of the magnetization state; however, they do not contribute to the energy barriers involved in the long-timescale relaxation.

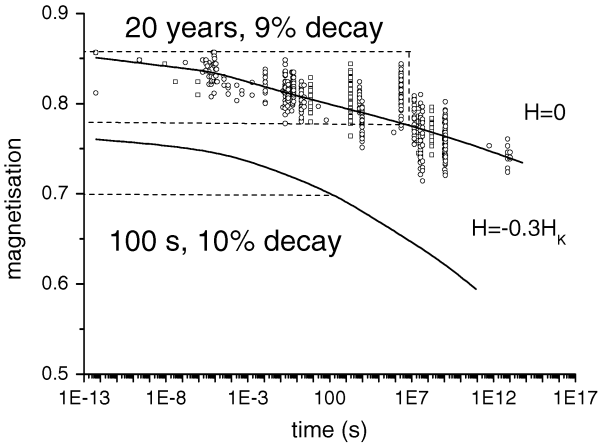

Fig. 1. Calculated thermal magnetization decay in CoCrPt low-magnetization longitudinal recording medium.

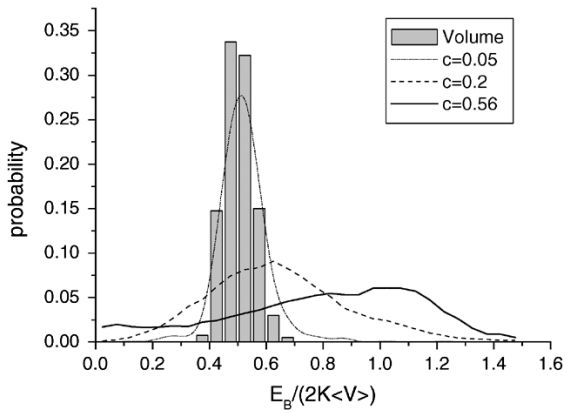

Fig. 2. Volume distribution and initial energy barrier distribution, evaluated at the remanent state, for Co particles with different concentrations.

To illustrate the performance of the method, we present in Fig. 1 an example of the calculation of long-time magnetization decay for low-magnetization CoCrPt longitudinal recording medium. The recording granular medium was modeled by 40 Voronoi-type polyhedra with an average size of 6.5 nm and random in-plane easy axes distribution with anisotropy value of $K = 2.4\ 10^6$ erg/cm^3 and $M_s = 442$ emu/cm^3. As is clearly observed, the recording medium is very stable, showing less than 10% magnetization decay for 20 years. However, if a field $H = -0.3(2K/M_s)$ is applied, a similar magnetization decay is produced during a period of two hours. A small system size results in a statistical scatter of the data, as evident in Fig. 1, so that an additional averaging over multiple curves (solid line in Fig. 1) is necessary.

Another example is presented in Figs. 2 and 3. We considered an ensemble of 400 Co particles with log-normal size distribution of average diameter 4 nm and $\sigma = 0.1$ dispersion. The

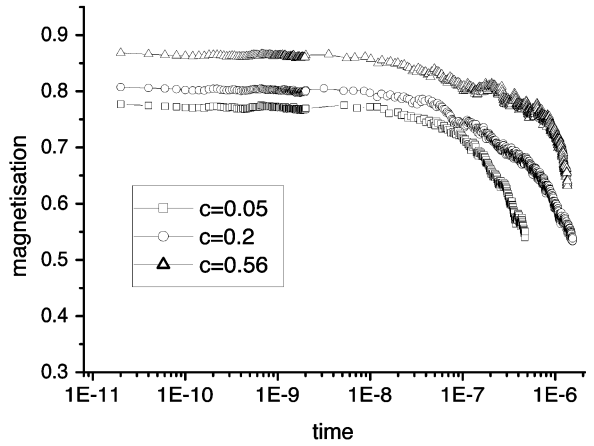

Fig. 3. Calculated magnetization decay for Co particles with different concentrations.

anisotropy value is taken to be $K = 2\ 10^6$ erg/cm^3, the magnetization saturation value $M_s = 1400$ emu/cm^3, and a two-dimensional (2-D) random anisotropy easy axes distribution was supposed. The concentration of particles varied from a low-density $c = 0.05$ structure to a closely packed structure of $c = 0.56$. The particle arrangement was modeled by specially designed Monte Carlo algorithm, as described in details in [13]. Essentially, an initial state is produced under a compression algorithm after the particles are placed on a squared lattice. The particles are then allowed to move under the influence of an attractive central potential, which leads to structures with short-ranged order dependent upon the packing density. The algorithm produces the formation of chains and loops of particles for intermediate concentrations $(c = 0.2)$ and that of the self-organized quasi-crystal hexagonal structure for large particles concentration $(c = 0.56)$.

The initial (remanent state) energy barrier distributions are presented in Fig. 2, and the calculated magnetization decay is shown in Fig. 3. In an interacting system, the relation between the initial energy barrier distribution and the resulting decay has a limited sense since the energy barrier distributions are time- and magnetization-state-dependent characteristics [6]. In addition, the magnetization change corresponding to a particular energy barrier is larger in the case of a collective process. For example, for a concentration of $c = 0.56$, the average cluster size corresponding to one energy barrier is six particles, while the $c = 0.05$ concentrated system has energy barriers corresponding to individual particle rotations.

However, some qualitative understanding could be gained analyzing the energy barriers of Fig. 2. Some initial differences corresponding to very small energy barriers (with the related timescale of the order of nanoseconds) are practically not seen in the total decay since the associated magnetization change is small. However, at the timescale of the order of $K\langle V\rangle$ (corresponding to $2\ 10^{-7}$ s in a noninteracting system), the magnetization change is larger for systems with small concentrations due to the fact that more barriers have values below this energy scale. However, at a larger time scale ($>10^{-6}$ s), the system with higher concentration $(c = 0.56)$ has a faster decay rate than that of $c = 0.2$ due to a collective process that involves larger magnetization changes. Consequently, the influence of magnetostatic interactions on the character of the decay is different at different timescales.

In conclusion, we have presented a general method to evaluate long-time magnetization decay in a strongly interacting system with distributed properties. At the present moment, reasonable performance of the method could be expected for systems of small sizes only, since the method involves the diagonalization of large-size Hessian matrices. The concrete implementation of the method is still subject to some degree of optimization with respect to the use of modern methods of computer algebra. In addition, with the increasing availability of fast computers, more realistic system calculations are expected to become feasible in the near future.

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