Density of Energy Barriers in Fine Magnetic Particle Systems

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***Abstract*--Using a numerical method based on the minimization of the Onsager-Machlup functional we have studied the energy barrier distributions in systems of fine magnetic particles with the uniaxial anisotropy and the dipolar interaction. In such systems the interplay of the single particle anisotropy and** (ii) **the dipolar interaction strength (particle concentration) determines whether the single-particle or collective remagnetization dominates the system behaviour (including barrier heights).**

Our main conclusion is that the influence of the dipolar interparticle interaction on the energy barrier density $\rho(E)$ depends *qualitatively* on the single particle anisotropy. For *low* anisotropies $\rho(E)$ shifts towards *higher* barriers when increasing interaction strength, whereby for moderate and high anisotropy values the opposite shift takes place. We propose the explanation for this phenomenon.

***Index Terms*--energy barriers, fine magnetic particles, magnetic anisotropy, magnetodipolar interaction.**

I. INTRODUCTION

The problem of evaluating the energy barriers ΔE and transition probabilities p between metastable states in many-particle systems with strong interparticle interaction is probably one of the most challenging tasks in physics of disordered systems [1]. It is especially the case for systems of fine magnetic particles (spin and dipolar glasses, ferrofluids, magnetic nanocomposites) where disorder and frustration make both analytical and numerical calculations extremely difficult [2].

For barriers ΔE comparable with the temperature T direct Langevin-dynamics simulations of the escape over the barrier are possible [1]. However, for high energy barriers $\Delta E >> T$ such simulations are nearly useless because the mean escape time growth exponentially with the barrier height - as for real systems - due to the Arrhenius-Van't Hoff law: escape time over the barrier growths with its height ΔE as $\tau_{esc} \sim \exp(-\Delta E/T)$.

Calculations of the barrier heights for such high barriers in magnetic systems are extremely important from the practical point of view (not to mention the fundamental problems in the phase transition theory). Such calculations are the only way to predict the long-time stability of the information storage both for traditional and novel (MRAM) technologies. In this case we are interested in storage (stability) times of at least several years, so that the energy barriers should be *much* higher than the working temperature in order to make the transition probability p vanishingly small. Fortunately, for $\Delta E >> T$ the task of evaluation p is somewhat simpler because in this limit we do not need to know all the details of the transition trajectory: it is sufficient to find the lowest saddle point between the two metastable states of interest. The height of this saddle gives us the energy barrier ΔE between these states, thus allowing to estimate p using the same Arrhenius law.

The problem of finding such a saddle point can in general *not* be solved analytically for an interacting system. The reason is that a saddle point can be found only as a solution of a nonlinear equation system $\partial E/\partial x_i = 0$. We lack corresponding general methods, and there exist strong arguments that there will never be any [3]. Reliable analytical methods for the saddle point search in magnetic systems are applicable only to a single particle case [4, 5] or when the system remagnetization mode is known and relatively simple [6].

II. NUMERICAL METHOD

Recently [7] we have developed a general and powerful numerical method for the evaluation of the energy barrier between any two given metastable states in interacting many-particle systems, basing on the idea of Onsager and Machlup [8]. The method finds the *most probable* (optimal) *transition path* between these two states by minimizing the thermodynamical action (Onsager-Machlup functional) derived from the path-integral formulation of the problem. The required energy barrier is then calculated as the barrier along this optimal trajectory.

Thermodynamical action S for a fine magnetic particle system is [7]

$$S\left[\Omega(t)\right] = \int_0^{t_f} dt \sum_i \left[\left(\frac{d\theta_i}{dt} + \frac{\partial E\{\Omega\}}{\partial \theta_i} \right)^2 + \left(\sin\theta_i \cdot \frac{d\phi_i}{dt} + \frac{1}{\sin\theta_i} \cdot \frac{\partial E\{\Omega\}}{\partial \phi_i} \right)^2 \right]. \quad (1)$$

Here θ_i, φ_i are spherical coordinates of the unit orientation vector $\mathbf{m}_i$ of the magnetic moment of the i-th particle, $\{\Omega\}$ denotes the set of all angles (θ_i, φ_i) and $E\{\Omega\}$ is the system energy (including all interparticle interactions, single-particle anisotropy and energy due to the external field). The expression (1) is the immediate consequence of (i) stochastic equations of motion with temperature fluctuations taken into account as random fields [9] and (ii) the assumption that the components of these random fields on each particle are uncorrelated random variables with Gaussian distributions. The upper integral limit t_f, being a transition time in the original formulation of

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the problem, can be considered as an adjustable parameter when evaluating the energy barrier by the minimization of (1) (see [7] for detailed explanations).

An analytical minimization of the functional (1) is impossible for any really interesting interacting system. For this reason we have developed an algorithm for a numerical minimization of the discrete representation of the action (1) [7]. For interacting fine magnetic particle systems even this task is highly nontrivial due to the presence of *many 'false' local minima* of the functional (1), i.e., trajectories between the two states which *minimize* (1) but do *not* provide any information about the corresponding energy barrier. The existence of such 'false' minima of the action is due to its following feature: *any* trajectory which goes between the two energy minima *along the gradient lines* of the energy surface provides a local extremum to the action $S\{\Omega\}$ [10]. This means that, e.g., a trajectory between the given energy minima going through the energy *maximum* (along the gradient lines!) would also minimize the action (1). However, such a trajectory obviously contains no information about the height of a saddle separating these two minima.

In [7] we have constructed a special algorithm for distinguishing between the 'true' and 'false' optimal trajectories by analyzing the trajectories themselves (the algorithm employs the energy minimization starting from the points lying in the vicinity of the optimal trajectory and analyzing local energy minima obtained this way). An additional check whether the trajectory found by the action minimization really passes via a saddle point was performed by analyzing the *curvature tensor* of the energy surface at the point where the energy along the optimal trajectory has its *maximal* value. Only if the corresponding matrix has exactly *one* negative eigenvalue, then this energy maximum along the trajectory indeed corresponds to a transition saddle [1] and we have a 'true' optimal path.

III. Results and Discussion

Using this method, we have calculated the distribution of the energy barriers in a system of N (= 200 – 300) single-domain particles (embedded in a non-magnetic matrix) each having the uniaxial anisotropy $E_{an} = -KV\cdot(\mathbf{m}\cdot\mathbf{n})^2$ (V being the particle volume, K – the anisotropy constant and $\mathbf{n}$ – the unit vector of the anisotropy axis). The magnetodipolar interparticle interaction was treated using the extended Lorentz cavity method [11]. Periodic boundary conditions were assumed.

The most intriguing question for this system is the influence of the magnetodipolar interaction on the energy barrier distribution $\rho(E)$ (for the two opposite points of view see [12, 13]), because it controls both the reversible and irreversible thermodynamics of the system. To study this question, we have computed $\rho(E)$ for various particle volume fractions c (thus varying the interaction strength) and reduced single-particle anisotropies $\beta = 2K/M_S^2$. The latter quantity is very convenient for estimating the relative strength of the dipolar interaction: for $\beta >> 1$ the anisotropy plays a dominant role, whereby in the opposite limit the interaction dominates.

Here we present results for systems with relatively high (β = 2.0) and low (β = 0.2) single-particle anisotropies. For each (β,c)-pair the distribution of the energy barriers was accumulated from 8 realizations of the particle disorder; for each realization ≈ 200 transitions between metastable states were analyzed. Fig. 1 and 2 display the distributions of the reduced energy barriers $\varepsilon = E/M_S^2V$ (on the left) and the histograms of the moment changes corresponding to these transitions (right).

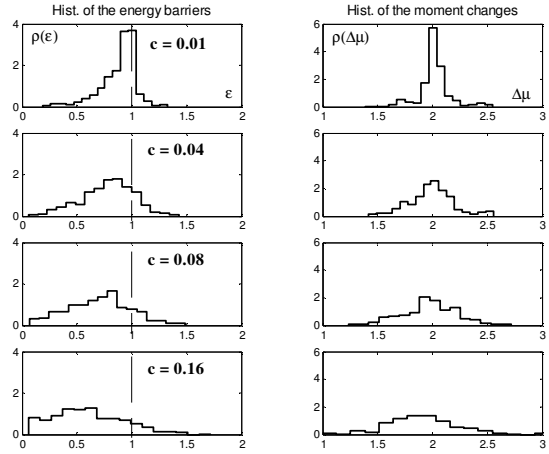

Fig. 1. Density of the energy barriers (left) and magnetic moment changes (right histogram) for transitions between randomly chosen energy minima in a system of magnetic particles with the high anisotropy β = 2.0. Dashed lines on the left histograms represent the position of a single particle energy barrier.

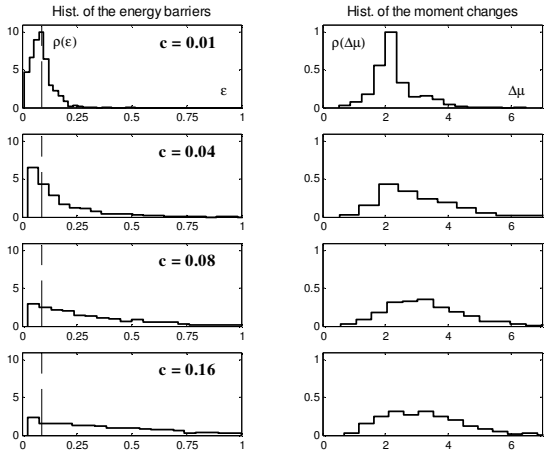

Fig. 2. The same as in Fig. 1 for the low anisotropy case β = 0.2

First of all, it can be seen that for low particle concentrations (≤ 1%) $\rho(\varepsilon)$ consists of a relatively narrow peak positioned at the value corresponding to the single particle energy barrier $\varepsilon_{sp} = \beta/2$, as it should be for a weakly interacting system.

When the concentration increases, the energy barrier density is getting broader, but for systems with the low and high anisotropy this broadening occurs in a *qualitatively different* ways. For the *high*-anisotropy case (Fig. 1) the broadening of $\rho(\varepsilon)$ with the increasing c is accompanied by its shift towards *lower* energy barriers; already for moderate concentration ($c \geq 4\%$) almost all barriers are below the single particle value.

For the system of particles with the *low* anisotropy (Fig. 2)

barriers both higher and lower, than for a single particle, arise. However, the overall energy barrier spectrum clearly shifts towards *higher* energies with increasing particle concentration.

The energy barrier distribution $\rho(E)$, being itself highly interesting from the fundamental point of view, can not be compared directly with the experimental data. The establishing of the relation between $\rho(E)$ and measurable quantities like *ac*-susceptibility and magnetic viscosity is by no means trivial at least for the following reasons: (i) experiments are performed at finite temperatures so that the *free* energy barrier density is required for their interpretation, (ii) different moment changes occur by transitions over different barriers.

Due to the lack of space we shall address here only the second problem. To solve it we have only to keep record of the differences between the magnetic moments $\Delta\boldsymbol{\mu}_{AB} = \boldsymbol{\mu}_A - \boldsymbol{\mu}_B$ of the limiting states A and B for each transition studied. The corresponding histograms of the $\Delta\mu$ -distributions $\rho(\Delta\mu)$ are shown on the right in Fig. 1 and 2. It can be seen, that for weakly interacting systems (single-particle flips) the corresponding distributions exhibit a sharp peak near $\mu = 2$ (the moment change for a single-particle flip $\mathbf{m} \rightarrow -\mathbf{m}$ is $|\Delta\mathbf{m}| = |\mathbf{m} - (-\mathbf{m})| = |2\mathbf{m}| = 2$). With the increasing particle concentration the distribution $\rho(\Delta m)$ broadens signaling the appearance of collective remagnetization processes.

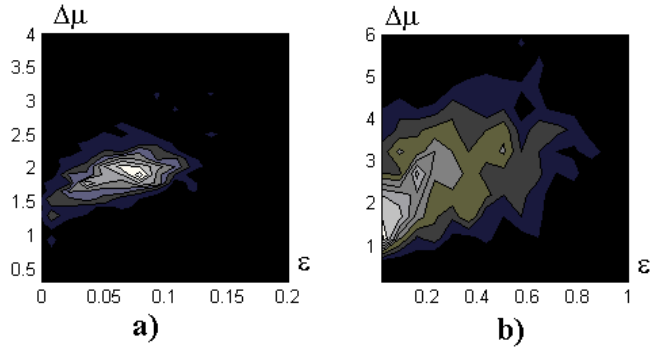

Fig. 3. Mutual 2D distribution density $\rho(\varepsilon,\Delta\mu)$ of the energy barriers and moment changes for dilute (part a), $c = 0.01$) and concentrated (part b), $c = 0.16$) system of fine magnetic particles with $\beta = 0.2$

The key question by the analysis of the moment changes is whether their magnitude is *correlated* with the height of the corresponding energy barriers. If, e.g., the moment change would tend to zero when the energy barrier height for the transition decreases, than small energy barriers would not play any significant role in the system thermodynamics (corresponding magnetization changes would be hardly noticeable). We have found that this is *not* the case by plotting the 2D *mutual* distribution $\rho(\varepsilon,\Delta\mu)$ of the energy barriers *and* moment changes. Corresponding plots for a system with the low anisotropy $\beta = 0.2$ and two different concentrations are shown in Fig. 3. The density $\rho(\varepsilon,\Delta\mu)$ for the lowest concentration $c = 0.01$ consists, as expected, of a single sharp peak positioned at ($\varepsilon \approx 0.1$, $\Delta\mu \approx 2.0$). From the picture for the high concentration $c = 0.16$ it can be seen that, although the moment changes $\Delta\mu$ for the low barriers are (on average) somewhat smaller than for the higher ones, they do *not* tend to zero at all. Hence all transitions provide comparable contributions to the system thermodynamics.

The *qualitative* difference in the behaviour of the energy barriers density with increasing particle concentration for systems with low and high single-particle anisotropies can be understood as follows. For particles with the *high* anisotropy high energy barriers do already exist even in a non-interacting system. In this case the dipolar interaction leads to the decrease (on average) of these barriers, simply because the randomly oriented interaction field will more likely reduce the barrier created by the uniaxial anisotropy than increase it (see [14] for the corresponding theory in the pair approximation). On the other hand, if the barriers due to the single-particle anisotropy are weak ($\beta << 1$) than starting from some concentration the main contribution to the energy barriers height will come from the dipolar interaction. Such barriers will obviously get larger with increasing particle concentration, leading to the shift of $\rho(E)$ towards higher energy values.

IV. Conclusion

Using the rigorous numerical method for evaluating the energy barriers between the two given metastable states, we have proven that the dipolar interaction in fine magnetic particle systems can either *decrease* the energy barriers - if such barriers are *already created* by a relatively high single-particle anisotropy, or *increase* them - if these barriers are mainly due to the *dipolar interaction itself*.

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