



# Memory and aging effects in interacting sub-10 nm nanomagnets with large uniaxial anisotropy

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## ABSTRACT

Using a nonequilibrium Monte Carlo method suitable to nanomagnetism, we investigate representative systems of interacting sub-10 nm grained nanomagnets with large uniaxial anisotropy. Various magnetization memory and aging effects are found in such systems. We explain these dynamical effects using the distributed relaxation times of the interacting nanomagnets due to their large anisotropy energies.

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## 1. Introduction

Nanomagnets attract huge interest because of their amazing properties and promising applications [1–6]. For well-separated nanomagnets (including single-molecule magnets), quantum tunnelling, interference, and coherence can be observed at extra-low temperatures [6–10], and the magnetization behaviours at some higher temperatures can be described by Neel–Brown law [11,12]. When inter-particle distances become small enough, the dipolar-dipolar interaction will modify the magnetization behavior leading to some super-spin-glasses behaviors [13,14], similar to conventional spin glasses [15–18]. Grained nanomagnets with large uniaxial anisotropy are essential to modern magnetic data storage. For typical CoCrPtB media, usual average grain sizes must be about 10 nm to keep magnetic stability of 10 years at room temperature [19]. In the case of CoCrPt-oxide media for perpendicular recording, dominant inter-grain interactions are weak antiferromagnetic (AFM) couplings and average grain size can be 8 nm or smaller for the same stability [19]. Such average size can even

be reduced down to 3 nm or smaller when FePt in the L1<sub>0</sub> phase is used as data storage media, because its magnetocrystalline anisotropy reaches to 44 meV/nm<sup>3</sup> [19]. Such nanomagnets with large uniaxial anisotropy, especially when composing special systems, can yield various dynamical phenomena waiting for exploration.

Here we explore dynamical magnetic properties of representative systems of sub-10 nm grained nanomagnets with large uniaxial anisotropy. We use the giant spin approach for the nanomagnet because the magnetic interactions between electronic spins in it are strong. We assume that the magnetic anisotropy energies satisfy a Gaussian distribution to consider their fluctuations due to different shapes, sizes, and interfacial environments, and the inter-nanomagnet interactions, including magnetic dipolar interaction, are AFM [19]. We use a giant-spin model and a dynamical spin Monte Carlo (DSMC) method [20] to correctly simulate dynamical magnetization of the giant-spins of the component nanomagnets. Through systematical DSMC simulations, we find various field-cooling (FC) and zero-field-cooling (ZFC) magnetization memory and aging effects in such systems. We explain these dynamical effects uniformly in terms of the continuously-distributed relaxation times of the component nanomagnets due to the various uniaxial anisotropy energies. More detailed results will be presented in the following.

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## 2. Model, method and parameters

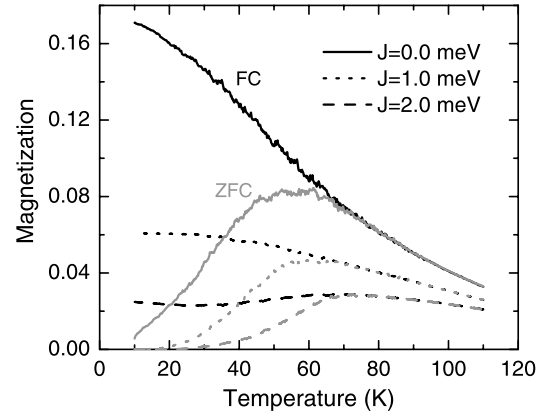
We consider a finite two-dimensional lattice of grained nanomagnets. Each nanomagnet actually includes many magnetic atoms, but the magnetic interactions between the magnetic atoms are much stronger than those among different nanomagnets. The inter-nanomagnet magnetic interactions are described by an anti-ferromagnetic coupling according to actual materials for modern magnetic data storage [19]. We take the giant spin approach and use one spin variable  $\vec{S}_i$  to describe the magnetic property of each nanomagnet. Because the spin value  $S_i$  of such a nanomagnet is typically  $10^2$ – $10^3$ , large anisotropy energies of such nanomagnets are mainly dependent on their shapes and interfacial environments [19], and thus can be reasonably assumed to satisfy a Gaussian distribution. This simplification keeps the main physics of these nanomagnet systems. Because  $S$  is large enough, we treat the spin operator  $\vec{S}_i$  as  $S_i\vec{s}_i$ , where  $\vec{s}_i$  is a classical unit vector. Generally speaking,  $S_i$  should vary from one nanomagnet to another, but for our systems all the  $\{S_i\}$  are uniform enough to be let have the same average value  $\bar{S}$  because the deviations from  $\bar{S}$  make little differences. Therefore, our model can be described by the Hamiltonian,

$$H = - \sum_i k_{ui} \vec{s}_i^2 + \sum_{i,j} J_{ij} \vec{s}_i \cdot \vec{s}_j - \gamma \vec{B} \cdot \sum_i \vec{s}_i, \quad (1)$$

where  $k_{ui}$  describes the uniaxial anisotropy energy satisfying a Gaussian distribution with the average value  $k_u$  and width  $\sigma_u$ ,  $\gamma$  is defined as  $g\mu_0\mu_B\bar{S}$ , and the field  $B$  is in the easy axis. Here  $J_{ij}$  describes the inter-nanomagnet AFM interactions. For actual perpendicular media for modern magnetic data storage, we assume that the easy axis is perpendicular to the plane of the  $N \times N$  nanomagnet lattice. In such a setup, the magnetic dipolar interaction is reduced to an AFM inter-nanomagnet interaction, which has been included in the  $J_{ij}$  parameters in Eq. (1).

Each of the spins has two meta-stable orientations along the easy axis and needs to overcome an energy barrier to achieve a reversal. We use the DSMC method to simulate the spin dynamics of these nanomagnet systems [20]. This method originates from the kinetic Monte Carlo method for simulating atomic dynamics during epitaxial growth [21]. Using  $\theta_i$  to describe the angle deviation of  $\vec{s}_i$  from the easy axis, we express the energy increment of the  $i$ th nanomagnet as  $\Delta E_i = k_{ui} \sin^2 \theta_i - h_i (\cos \theta_i - 1)$  to leading order, where  $h_i = (\gamma B - \sum_j J_{ij} s_j) s_i$  and the reduced variable  $s_i$  takes either 1 or  $-1$  [20]. As a result, an energy barrier  $\Delta E_i = (2k_{ui} + h_i^2)/4k_{ui}$  must be overcome to achieve the reversal of the  $i$ th spin. The rate for the spin reversal obeys Arrhenius law  $R_i = R_0 \exp(-\Delta E_i/k_B T)$ , where  $k_B$  is Boltzmann constant and  $T$  is temperature.

In our simulation, we use a typical value  $1.0 \times 10^9 \text{ s}^{-1}$  for the characteristic frequency  $R_0$ . As for the anisotropy energy parameters, we reasonably assume that the average value  $k_u$  of  $\{k_{ui}\}$  is 80.0 meV and the Gaussian width  $\sigma_u$  is 44.7 meV. We change temperature  $T$  by a step 0.2 K and set the sweeping rate to  $1 \text{ K s}^{-1}$  in most of the following cases. Special cases will be explicitly described otherwise. We take  $N = 40$  and use periodic boundary condition. This lattice size is appropriate considering the actual situation in the media for the modern magnetic data storage. It almost does not matter which boundary condition is chosen with the lattice size. Further simulations with larger  $N$  are done for confirmation. Each of our data is obtained by calculating the average value over 500–1000 independent simulation runs.



**Fig. 1.** Simulated FC (black) and ZFC (gray) magnetization curves under a field 100 Oe for  $J = 0.0, 1.0,$  and  $2.0$  meV. These curves are calculated in the same way as corresponding experimental magnetization curves are measured.

## 3. Main simulated results

### 3.1. FC and ZFC magnetization curves

We simulate the FC curves by calculating the average magnetizations under 100 Oe at every  $T$  point when cooling from 110 to 10 K. The ZFC curves are simulated by letting the system cool under zero field from 110 to 10 K and then calculating the average magnetizations under 100 Oe at every  $T$  point when warming from 10 to 110 K. Our simulation realizes what happens in measuring experimental FC and ZFC magnetization curves. Our simulated results are presented in Fig. 1. For zero interaction or  $J = 0$ , the FC magnetization increases monotonously with decreasing  $T$ , and the ZFC magnetization increases first with increasing  $T$ , reaches its maximum at the blocking temperature  $T_B = 55$  K, and then decreases. The FC and ZFC magnetization curves follow the Curie law above the temperature  $T_m = 70$  K. The difference between the FC and ZFC magnetizations diminishes above  $T_m$ . Such behaviors are key features of nanomagnets, showing up in super-spin glass systems [13] and interacting nanoparticles [22,23]. When  $J$  is larger, the magnetization in the FC and ZFC curves becomes substantially smaller, as shown in Fig. 1. When  $J$  becomes further larger, a minimal magnetization can be seen below  $T_B$ . It is at 30 K for 2.0 meV. When  $J$  is less than 0, the magnetization in such curves becomes larger but the curve shape remains nearly the same.

### 3.2. Simulated ZFC and FC memory effects over time

Cool the system under zero field to a low temperature, 30 K, and then apply a field 500 Oe and let the system relax from  $t = 0$ . At  $t_1 = 814$  s, we change  $T$  to 10 K and reverse the field, and at  $t_2 = 4124$  s, we recover the original temperature and field. We calculate the average magnetization when the system relaxes with time from  $t = 0$  to  $t = 15000$  s, and present the results in Fig. 2a. The magnetization at  $t_2$  is equivalent to that at  $t_1$ , that is, the system keeps the memory at  $t_1$  although it undergoes relaxation from  $t_1$  to  $t_2$  under the different temperature and field. This is a ZFC memory effect over time. In addition, we cool the system under a field 500 Oe to 30 K, and then remove the field and let the system relax from  $t = 0$ , but we change the temperature to 10 K and the field to  $-500$  Oe between  $t'_1 = 3416$  s and  $t'_2 = 6730$  s. Meanwhile we calculate the magnetization from  $t = 0$  to  $t = 20000$  s. The result, shown in Fig. 2b, shows an FC memory effect over time,  $m_{t_2} = m_{t'_1}$ . Similar effect was observed in super-spin glass systems and interacting nanoparticles [14,22,23]. Our further calculations

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