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J. Phys.: Condens. Matter 25 (2013) 106006 (6pp)

Enhancement of the magnetic anisotropy barrier in critical long range spin systems

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Received 11 December 2012, in final form 20 January 2013 Published 12 February 2013 Online at stacks.iop.org/JPhysCM/25/106006

Abstract

Magnetic materials are usually characterized by anisotropy energy barriers which dictate the timescale of the magnetization decay and consequently the magnetic stability of the sample. Here we consider magnetization decay for spin systems in a d = 3 cubic lattice with an isotropic Heisenberg interaction decaying as a power law with a critical exponent $\alpha = d$ and on-site anisotropy. We show that the anisotropy energy barrier can be determined from the ergodicity breaking energy of the corresponding isolated system and that, unlike in the case of nearest neighbour interaction, the anisotropy energy barrier grows as the particle volume, V, and not as the cross-sectional area.

(Some figures may appear in colour only in the online journal)

1. Introduction

From a theoretical point of view the problem of magnetization decay in nanosystems is difficult to treat: nanoscopic systems are too big to be solved by brute force calculation and too small to be tackled by the tools of statistical mechanics at equilibrium. Indeed, the problem of magnetization decay is a typical example of an out of equilibrium phenomenon, which is the decay out of a metastable state.

On the other hand, nanomagnetism has important consequences in the technology of memory and information processing devices due to the quest for improving magnetostorage density and for the realization of smaller and smaller magnetic units. Significant improvements in experimental techniques allowed investigations of magnetic properties in nanoparticles and nanowires [1]. The analysis of magnetic decay is crucial for understanding ferromagnetic behaviour; indeed one-dimensional nanoscopic systems can also show ferromagnetic behaviour at low but finite temperature, even if a ferromagnetic phase transition is theoretically forbidden [2], due to large magnetic decay times [3–7].

The modelling of the magnetic decay through an on-site anisotropic barrier is typical in the literature where mainly

short range interactions have been considered. Nevertheless, in many realistic situations, one needs to go beyond nearest neighbour coupling, taking into account the long range nature of the interaction. The latter is usually introduced as a two-body interaction coupling decaying at large distance with a power law exponent α not larger than the embedding spatial dimension d [8]. It is the case, for instance, for the dipolar interaction in 3D systems, or of the so-called RKKY (Ruderman-Kittel-Kasuya-Yosida) interaction, which decays as r^{-d} , where r is the distance between spins, and d is the dimension of the lattice system. In particular, long range interactions might be responsible for the ferromagnetic behaviour of diluted magnetic semiconductors (DMS) [9] and diluted magnetic oxides (DMO) [10], promising materials for the realization of spintronics devices. Note that in both cases the interaction decays with a power, α , equal to the lattice dimension, d, which is the case that we will analyse in this work and that it is termed 'critical' in the literature [8].

One of the first attempts to understand magnetic decay times in nanoparticles is due to Neél [11] and Brown [12], who considered that all the spins in a magnetic particle move coherently as a single spin, so magnetization decay can be described as due to thermal activation over a single energy barrier. In Brown's theory the magnetic decay time, τ , is shown to follow an Arrhenius law:

$$\tau \propto e^{\beta \Delta E} \tag{1}$$

where $\beta = 1/k_{\rm B}T$ is the inverse temperature and $\Delta E \propto V$ is the anisotropic energy barrier proportional to the particle volume, V. The question of whether a microscopic model would support this conclusion has been analysed by many authors, mainly assuming nearest neighbour interactions. For instance in Braun's theoretical approach [13] a sufficiently elongated system of nearest neighbour interacting spins with an on-site anisotropy barrier have been shown to reverse the magnetization (thus producing an average magnetic decay) through a process called nucleation, energetically convenient with respect to coherent rotation. In this mechanism, accomplished by the formation of a soliton-antisoliton domain wall, the magnetic anisotropic energy to be overcome turns out to be proportional to the cross-sectional area of the particle, $\Delta E \propto A$, and not to its volume, V. Studies of different mechanisms of magnetic decay have been the objective of intensive investigation [14] until recently, where also 3D spherical samples with short range interactions and on-site anisotropy are shown to produce nucleation for sufficiently large radius [15]. Thus, for short range interaction, Brown's theory and a consequent Arrhenius law with an exponent proportional to the volume V of the particle are valid only for very small particles, while in general, for large or elongated particles, the exponent is given by the cross-sectional area of the particle. A smaller exponent means smaller decay times for the same temperature. The size and shape dependences of the magnetic anisotropy barrier, and consequently of the decay times, have also been confirmed experimentally in [16].

Magnetic decay, in a macroscopic model with long range interaction, has been much less investigated. Long range interaction can affect the decay out of a metastable state in a significant way; in the seminal paper [17] it was shown that the time of decay out of a metastable state in a toy model with infinite range interaction is given by an exponential law with an exponent proportional to the squared volume of the particle.

The main goal of this paper is to analyse magnetic decay beyond nearest neighbour interaction, focusing on a critical three-dimensional system.

In order to estimate the anisotropic energy barrier, we propose a different point of view which does not assume the specific motion of the spins upon the magnetic reversal; rather we estimate the minimal energy barrier that must be encountered during the magnetic reversal process. We related the energy barrier to the recently found topological non-connectivity threshold (TNT) in anisotropic spin systems [18–21].

2. The topological non-connectivity threshold (TNT)

Following [18], we briefly review the topological nonconnectivity threshold. Let us consider a generic anisotropic spin system, with an easy axis of magnetization (the direction \hat{n}_{easy} of the magnetization in the ground state), with a microcanonical energy

$$H(\vec{S}_1,\ldots,\vec{S}_N)=E$$

Let us also set

$$m = \frac{1}{N} \sum_{k} \vec{S}_{k} \cdot \hat{n}_{\text{easy}}, \qquad (2)$$

as the magnetization along the easy axis. Note that in our paper it will be $\hat{n}_{easy} = \hat{z}$.

It was proven [18] that below a suitable threshold, E_{tnt} , given by the minimal energy attainable under the constraint of zero magnetization, *m*, along the easy axis:

$$E_{\text{tnt}} = \operatorname{Min}(H(\dots \hat{S}_i \dots) | m = 0), \qquad (3)$$

the constant energy surface is disconnected into two portions, characterized by a different sign of the magnetization. From the dynamical point of view one has a case of ergodicity breaking: a trajectory at fixed energy cannot change the sign of magnetization since it is confined forever in one region of the phase space.

It was also demonstrated that in the case of long range interaction among the spins [20], the disconnected energy portion determined by the TNT remains finite when the number of particles becomes infinite.

While for isolated systems the magnetization cannot reverse its sign if the microcanonical energy *E* is below the energy threshold E_{tnt} , when the system is put into contact with a heat bath this may happen for any (even extremely low) temperature. In the following we will address the question of whether the TNT influences in some way the magnetization decay.

3. The α -ranged model with on-site anisotropy

The model that we consider is a spin system characterized by an isotropic α -ranged exchange interaction and on-site anisotropy and it is described by the following Hamiltonian:

$$H = -J \sum_{i>j} \frac{\bar{S}_i \bar{S}_j}{r_{i,j}^{\alpha}} - D \sum_i (S_i^z)^2,$$
(4)

where the \vec{S}_i are the spin vectors with unit length, α determines the range of the interaction among the spins, J > 0 is the exchange coupling and D > 0 is the on-site energy anisotropy. As one can see in equation (4), the first term is not the usual Heisenberg (or exchange) term, but it contains a power law decaying factor $r_{i,j}^{-\alpha} = |\vec{x}_i - \vec{x}_j|^{-\alpha}$, where \vec{x}_i indicates the position of the *i*th spin in the lattice. α is called the range of interaction and, as one can observe, for $\alpha = 0$ one has the so-called 'infinite-ranged' model where all spins interact in the same way with all others. On the other hand, for $\alpha = \infty$, one gets the usual nearest neighbour (or Heisenberg) interaction. This model has been considered for instance in [19, 22].

The minimal energy for this class of spin systems is attained when all the spins are aligned along the \hat{z} direction,

which thus defines the easy axis of magnetization. In the following we will consider the 'critical' case $\alpha = d$.

For this class of systems, the energy, E_{tnt} (see equation (3)), can be computed numerically using for instance a minimizing constrained algorithm, but it can also be estimated analytically. To this end let us consider two configurations with m = 0:

- The first one has all spins aligned perpendicular to the easy axis (the z axis in our case). The energy difference of this configuration from the one having minimal energy is DN, which is the energy barrier due to the coherent rotation of all spins.
- The second is a configuration, labelled $\uparrow\downarrow$, consisting of two neighbouring identical blocks with opposite magnetization along the easy axis. This configuration roughly corresponds to what is called nucleation. The energy difference of this configuration from the minimal energy, for large N values, is given by [20]

$$\Delta E_{\uparrow\downarrow} \simeq JCN \tag{5}$$

where C is a suitable, shape dependent, constant.

For the sake of clarity we give here below the same general argument as in [20], leading to equation (5). In the $\uparrow\downarrow$ configuration the spins in each of the two blocks are aligned along the easy axis, so the energy of each block is just the minimal energy of N/2 spins, $E_{\min}(N/2)$. Thus, the energy of the system in the $\uparrow\downarrow$ configuration is the sum of the energies of each of the blocks plus the energy of interaction, V, between the two blocks:

$$E_{\uparrow\downarrow} = 2E_{\min}(N/2) + V. \tag{6}$$

On the other hand, we can write in a similar way the minimal energy for a block of N spins: $E_{\min}(N) = 2E_{\min}(N/2) - V$, and compute the disconnected portion of the spectrum as $\Delta E_{\uparrow\downarrow} =$ $E_{\uparrow\downarrow} - E_{\min} = 2V$. This implies that we can compute $\Delta E_{\uparrow\downarrow}$ only from the knowledge of the minimal energy, since we have $V = 2E_{\min}(N/2) - E_{\min}(N)$, from which equation (5) follows since $E_{\min}(N) \propto N \ln N$ for $\alpha = d = 3$; see [20] and references therein. Note that the same argument can be applied for any α ; in particular for $\alpha \leq d$, we have $\Delta E_{\uparrow\downarrow} \propto N^{2-\alpha/d}$.

Moreover it can be shown (see [20]) that the energy of these two configurations is a good approximation of E_{tnt} , so we can write

$$\Delta E_{\text{tnt}} \equiv E_{\text{tnt}} - E_{\text{min}} \approx \text{Min}(DN, \Delta E_{\uparrow\downarrow}). \tag{7}$$

Equation (7) is valid whenever DN is not too close to $\Delta E_{\uparrow\downarrow}$. The magnetic decay times have been analysed in the canonical ensemble, using a modified Monte Carlo simulation [23, 5]. In this modified Monte Carlo approach, at each step the spin is allowed to move inside a cone with a temperature dependent size. This method allows us to connect the unrealistic Monte Carlo time to the physical time given by the Langevin equations in the high damping limit [5]. As the initial condition we chose all spins aligned along the easy axis, and from the exponential decay in time of the average magnetization, $\langle m(t) \rangle \propto e^{-t/\tau}$, we computed the magnetic decay time, τ .



3

β

Figure 1. Decay time τ versus the inverse temperature β for a parallelepiped $L \times L \times \epsilon L$ with $\alpha = 3, J = 1/20, D = 0.5, \epsilon = 10$. Symbols refer to numerical data, while full lines are the analytical prediction from equations (7) and (8); see the text. Black squares are for L = 3, green circles are for L = 4 and red asterisks are for L = 5. Arrows indicate the corresponding critical temperatures β_{stat} defined in equation (9).

4. Results

In order to prove that the energy barrier depends on the volume and not on the cross-section, in the following we will consider the 'worst' possible case, namely a thin parallelepiped, $L \times L \times \epsilon L$, in the presence of a 'critical' interaction $\alpha = d = 3$. We chose the aspect ratio $\epsilon = 10$.

Magnetic decay times as a function of the inverse temperature, β , for different sizes, L, are shown in figure 1. As one can see, two different regimes are clearly identified: one for high temperature (small β) where all curves are superimposed, and another one, for sufficiently low temperature, where times grow exponentially fast. In the same figure we also show, as full lines, the magnetic decay times given by the following formula:

$$\tau = \tau_0 \, \mathrm{e}^{\beta \Delta E_{\mathrm{tnt}}},\tag{8}$$

where τ_0 is a factor that may depend on temperature too, $\beta = 1/k_{\rm B}T$, and $\Delta E_{\rm tnt}$ has been obtained from equation (7). Note that for the parameters used in figure 1 we have $\Delta E_{\text{tnt}} \simeq$ $\Delta E_{\uparrow\downarrow}$, so we are in the nucleation regime.

The good agreement with numerical simulations shows that the anisotropic energy barrier, ΔE , is very well approximated by ΔE_{tnt} . Note that in this case (nucleation) we have $\tau_0 \propto e^{D\beta}$ [4]. Indeed a single spin can continuously change from S = 1 to -1 only passing through the state S = 0. While the two extreme states have the same magnetic anisotropy (since it is proportional to DS^2), the intermediate state S = 0 has a barrier higher by a factor D. This additional spin flip does not occur, for instance, in discrete models, such as the Ising model, since there are no intermediate states between S = 1 and -1.

We also checked that equation (8) is valid in the coherent rotation regime, where the anisotropic energy barrier is given by $\Delta E_{\text{tnt}} = DN$.

4

We may expect that the exponential law given by equation (8) holds when $k_{\rm B}T \ll \Delta E$, see [12]. Clearly this gives an upper bound for the temperature for which equation (8) is valid. Indeed, it should also be the case that $T \ll T_{\rm stat}$, where the latter is the temperature at which the barrier at m = 0 in the free energy vanishes (which coincides, in the thermodynamic limit, with the critical temperature, $T_{\rm cr}$, at which a phase transition occurs). Clearly in the absence of a free energy barrier, i.e. when $T \ge T_{\rm stat}$, one cannot expect the magnetic decay to be described by the Arrhenius law.

 T_{stat} can be computed numerically from the probability distribution $P_T(m)$ of the magnetization in the canonical ensemble at the temperature *T*. An example of these probability distributions is shown in figure 2 where it is clear that on increasing the temperature, and thus decreasing β , the distribution changes from bi-modal to singly peaked. It is thus natural to define T_{stat} as the temperature at which the second derivative of such a probability distribution at m = 0 changes sign:

$$\frac{\mathrm{d}^2 P_{T_{\text{stat}}}}{\mathrm{d}m^2}(0) = 0. \tag{9}$$

The related values of $\beta_{\text{stat}} = 1/k_{\text{B}}T_{\text{stat}}$ for different *L* values are indicated as arrows in figure 1 and agree very well with the temperatures at which the exponential law starts to be valid. Work is in progress in an effort to understand whether the onset of the exponential law at the temperature T_{stat} occurs also for a generic interaction range $\alpha \neq d$. One should note that on increasing the system size, T_{stat} increases; see figure 1. This is typical of long range interacting systems when the coupling has not been rescaled in order to get an extensive energy.

For $\alpha < d$ and D = 0 a simple mean field approach allows us to compute the critical temperature of the thermodynamic phase transition:

$$T_{\rm cr} = JN^*/3, N^* = \propto \begin{cases} \frac{2N^{1-\alpha/d} - 1}{(2-\alpha/d)(1-\alpha/d)} & \text{for } \alpha < d \\ 2\ln(N) & \text{for } \alpha = d. \end{cases}$$
(10)

Let us note that for the parameters chosen in figure 1, T_{stat} behaves as T_{cr} for $\alpha = d$, namely it grows with the number of spins as $\ln N$, even if $D \neq 0$. We do not know whether such scaling is valid for any values of D.

It is possible to give a heuristic justification of equation (8). Magnetic decay occurs through fluctuations of the magnetization around its equilibrium value. The probability of a magnetization fluctuation is determined by the free energy barrier, $\Delta F = \Delta E - k_{\rm B}T\Delta S$, through the Arrhenius factor, $e^{-\beta\Delta F}$. Whenever the entropic barrier, ΔS , is negligible at low temperature, the accessible spin configurations can be determined by minimizing the energy only. In order to reverse its sign, the value of the magnetization, *m*, has to go, say, from m = 1 to 0. Since for m = 1 the system has minimal energy, it is clear that $\Delta E_{\rm tnt}$ represents the minimal energy barrier encountered by the system while reversing its magnetization.





Figure 2. Magnetization probability distribution for a cubic lattice, $4 \times 4 \times 40$, at different inverse temperatures β as indicated in the legend. The critical curve corresponding to β_{stat} , where the curve changes concavity (see equation (9)), has been shown as a dashed line.



Figure 3. Anisotropic energy barrier ΔE as a function of the side *L* of the parallelepiped, $L \times L \times \epsilon L$, obtained by an exponential fitting of the decay time versus the inverse temperature β (namely fitting the points after the arrows in figure 1). The (full) line L^3 and the (dashed) line L^2 have also been added for the sake of comparison. Here, $\epsilon = 10$, $\alpha = 3$, J = 1/20 and D = 0.5.

From the data shown in figure 1 we have that $\Delta E \approx \Delta E_{\text{tnt}}$. Moreover, from equations (5) and (7), we have that, independently of the size and the shape of the magnetic particle, $\Delta E_{\text{tnt}} \propto N$. It follows that the anisotropic energy barrier is proportional to the particle volume since $V \propto N$. In particular, this means that even in the case of 3D elongated particles, in contrast to what happens for nearest neighbour interactions, the energy barrier depends not on the cross-sectional area but on the whole particle volume.

In order to further confirm such theoretical prediction we discuss the dependence of the anisotropic energy barrier, ΔE , as a function of the side, L, of the parallelepiped. Results are shown in figure 3. For the sake of comparison, the lines proportional to the cross-section (L^2) and to the volume (L^3) have been drawn. As one can see, the barrier grows as the particle volume even in this elongated quasi-1D geometry.

Last but not the least, we would like to mention that the term 'nucleation' that we used above is meaningful even for α -ranged interactions and that the proportionality between the energy barrier, ΔE , and the number of particles, N, is not due to coherent rotation.

To prove that, we first consider in figure 4 the time evolution of a single system taken from figure 1. In particular we plot for one single Monte Carlo trajectory the values of

$$M = \left| \frac{1}{N} \sum_{i=1}^{N} \vec{S}_i \right|,\tag{11}$$

and

$$m = \frac{1}{N} \sum_{i=1}^{N} S_i^{z},$$
 (12)

as a function of the Metropolis time. As one can see, every time m changes its sign, the modulus of the magnetization, M, goes to zero, which is consistent with a coherent movement of all spins as in the soliton–antisoliton mechanism.

Let us note that in this work we mainly considered the case $D \gg J$ for which, during magnetic reversal, we have the formation of a sharp domain wall, i.e. with a width much smaller than the particle size. Nevertheless we find that the energy barrier scales with the volume, even if the magnetic reversal mechanism is given by nucleation, as shown in figure 4. This is at variance with the short range interacting case, found in the literature, where nucleation always implies a scaling with the cross-sectional area and not with the volume. This crucial difference from results found in the literature for nearest neighbour interactions is one of the main points of our paper and a generic hallmark of long range interaction.

5. Conclusions

In conclusion, we have analysed a 3D spin system with long range interaction characterized by an exponent $\alpha = d =$ 3. We have predicted and numerically confirmed that the magnetic decay time depends exponentially on the volume of the particle, independently of shape and size, and not on its cross-sectional area, as happens for nearest neighbour interactions in the large size limit.

The fast growth of the anisotropic energy barrier with the volume gives rise to the possibility of observing long-lived metastable ferromagnetism in nanosystems at high temperature, which is difficult to achieve for short range interaction.

Another important result is that we were able to compute the anisotropic energy barrier at finite temperature from the corresponding topological non-connectivity threshold of the isolated system.

One might ask whether the TNT gives the anisotropic energy barrier for any range of interaction. Our preliminary results indicate that this is the case for short range interaction; in particular for nearest neighbour interacting chains, the TNT agrees with the anisotropic energy barrier as computed in the literature (see [4]). For the long range case, $\alpha < d$, the relation between the TNT and the anisotropic energy barrier needs further investigations to be clarified. Nevertheless our



Figure 4. Magnetization as a function of the Metropolis time for a $4 \times 4 \times 40$ system with $\beta = 3.5$, J = 1/20, D = 0.5 and $\alpha = 3$. The light (red) curve refers to *M* (see equation (11)), while the dark (black) one refers to *m* (see equation (12)). As one can see, whenever *m* changes sign, *M* goes to zero, so a process close to nucleation occurs. It is important to stress that in the case of coherent rotation one would have $M \neq 0$ when *m* changes sign.

preliminary data indicate that the anisotropic energy barrier can grow even faster than the volume $\Delta E \propto V^{2-\alpha/d}$. Such an effect would be a distinguishing feature of long range interactions and cannot occur for short range ones.

Acknowledgments

We acknowledge useful discussion with B Goncalves, D Mukamel, S Ruffo, R Trasarti-Battistoni and A Vindigni. This work was supported by Regione Lombardia and CILEA Consortium through a Laboratory for Interdisciplinary Advanced Simulation (LISA) Initiative (2010) grant [link:http://lisa.cilea.it]. Support by the grant D.2.2 2010 (Calcolo ad alte prestazioni) from Universitá Cattolica is also acknowledged.

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