Enhancement of magnetic anisotropy barrier in long range interacting spin systems

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Magnetic materials are usually characterized by anisotropy energy barriers which dictate the time scale of the magnetization decay and consequently the magnetic stability of the sample. Here we present a unified description, which includes coherent rotation and nucleation, for the magnetization decay in generic anisotropic spin systems. In particular, we show that, in presence of long range exchange interaction, the anisotropy energy barrier grows as the volume of the particle for on site anisotropy, while it grows even faster than the volume for exchange anisotropy, with an anisotropy energy barrier proportional to $V^{2-\alpha/d}$, where V is the particle volume, $\alpha \leq d$ is the range of interaction and d is the embedding dimension. These results shows a relevant enhancement of the anisotropy energy barrier w.r.t. the short range case, where the anisotropy energy barrier grows as the particle cross sectional area for large particle size or large particle aspect ratio.

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A truly comprehensive understanding of magnetism at the nanoscale is still lacking. 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 statistical mechanics at the equilibrium. Indeed, the problem of magnetization decay is a typical example of out of equilibrium phenomenon, which is the decay out of a metastable state.

On the other hand, magnetism at the nanoscale has also important consequences in the technology of memory and information processing devices. The quest for improving magneto-storage density calls for the realization of smaller and smaller magnetic units. Significant improvements in experimental techniques allowed investigations of magnetic properties in nanoparticles and nanowires[1]. In particular, recently, there has been great interest in Single Chain Magnets (SCM) [2–4], which are possible candidates for nanoscopic memory units. In the experiment reported in [5] a 1–d chain of Co atoms, on average 80 atoms long, shows ferromagnetic behavior at low but finite temperature, even if a ferromagnetic phase transition is theoretically forbidden [6]. The theoretical microscopic models suggested [2, 3, 7] focused on short range interaction with on–site anisotropy. Nevertheless in many realistic situations one needs to go beyond nearest neighbor coupling, taking into account the long range nature of the interaction defined by a two–body spin interaction constant 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, of the dipolar interaction in 3–d systems, or of the so–called RKKY (Ruderman-Kittel-Kasuya-Yosida) interaction, which decays as the inverse of the distance between spins. In particular, the latter might be responsible for the ferromagnetic behavior of Diluted Magnetic Semiconductors (DMS) [9] and Diluted Magnetic Oxides (DMO) [10], promising materials for the realization of spintronics devices.

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, so that they can be considered as a single spin and described magnetization decay as due to thermal activation over a single energy barrier. In Brown's theory this time, τ, is shown to follow an Arrhenius Law (AL) :

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

where $\beta = 1/k_B T$ is the inverse temperature and ΔE is the anisotropic energy barrier proportional to the particle volume. A step forward Brown's theory has been realized by Braun [13]. In his theoretical approach a sufficiently elongated system of short range interacting spins have been shown to reverse their magnetic moment (thus producing an average magnetization 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 and not to its volume. Studies of different mechanisms of magnetic decay have been the objective of intense investigation[14] until recently, where also 3–d spherical samples with short range interactions are shown to produce nucleation for sufficiently large radius[15]. Thus, for short range interaction, Brown's theory, and a consequent AL with a volume dependent exponent is 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 dependence of the magnetic anisotropy barrier, and consequently of the decay times, have been also confirmed experimentally[16].

The main goal of this paper is to extend the theory of magnetic decay beyond nearest-neighbor interaction, focusing on realistic long range interacting systems.

In order to estimate the dependence of magnetic decay times on temperature, we propose a different point of view, which turns out to be independent of the decay mechanism and related to the recently found Topological Non-connectivity Threshold (TNT) in anisotropic spin systems [17]. Given a generic anisotropic spin system, with an easy axis of magnetization, (the direction $\hat{n}_{easy}$ of the magnetization in the ground state), with energy $H(\vec{S}_1, \ldots, \vec{S}_N) = E$, it was proven that below a suitable threshold, E_{tnt}, given by the minimal energy attainable under the constraint of zero magnetization along the easy axis:

$$E_{tnt} = \mathrm{Min}(H(...\vec{S}_i...) \mid m = (1/N)\sum_k \vec{S}_k \cdot \hat{n}_{easy} = 0), \tag{2}$$

the constant energy surface is disconnected in two portions, characterized by a different sign of the magnetization. From hereafter with magnetization we mean the magnetization along the easy axis. It was also demonstrated that in case of long range interaction among the spins[18], 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 below E_{tnt}, when the system is put in contact with a heat bath, we have found that $\Delta E_{tnt} = E_{tnt} - E_{min}$, represents an effective energy barrier for magnetic decay and that the decay time depends exponentially on such energy barrier:

$$\tau = \tau_0 \; e^{\beta(E_{tnt} - E_{min})} = \tau_0 \; e^{\beta \Delta E_{tnt}}, \tag{3}$$

where, τ_0 is a factor, dependent on temperature too, and E_{min} is the ground state energy. Note that with magnetic decay we mean the decay in time of the magnetization averaged over an ansemble of identical systems. While Eq.(3) was confirmed in simple toy models with all-to-all interaction among the spins [19], here our aim is to generalize such result to realistic spin systems.

It is possible to give an heuristic justification of Eq. (3). The decay of the ensemble average magnetization is determined by the magnetic reversal (magnetization reversing its sign) of each system. Note that magnetic decay times and magnetic reversal times are proportional to each other. Magnetic reversal occurs through fluctuations of the magnetization around its equilibrium value. The probability of a fluctuation of the magnetization along the easy axis is determined by the free energy barrier through the Arrhenius factor: $e^{-\beta \Delta F}$, with $\Delta F = \Delta E - k_B T \Delta S$, and where the second term represents the entropic barrier. Since the entropic barrier is usually negligible at low temperature, the accessible spin configurations can be determined minimizing the energy only. In order to reverse its sign, the value of the magnetization has to go, say, from $m = 1$ to $m = 0$. Since for $m = 1$ the system is in its minimal energy, it is clear that ΔE_{tnt} represents the minimal energy barrier found by the system while reversing its magnetization. The magnetic reversal time can be also obtained from the knowledge of the probability distribution of the magnetization at a given temperature, $P_T(m)$. Indeed we have $e^{\beta \Delta F} = P_T(m_{max})/P_T(m_{min})$, where $P_T(m_{max,min})$ stands for the maximum/minimum of the probability distribution. In order to obtain an approximate expression for $P_T(m)$ at low temperature we consider the following : at low temperature it is usually inconvenient for the magnetization to visit high entropy regions, so that magnetic reversal occurs though a coherent motion of the spins, such as coherent rotation or nucleation. In this case it is possible to neglect the entropic term and to approximate the energy of the system as a function of the magnetization only: $E = E(m)$. We can thus write $P_T(m) \propto e^{-\beta E(m)}$, and from this we regain Eq.(3):

$$\tau = \frac{P_T(m_{max})}{P_T(m_{min})} \simeq \frac{e^{-\beta E(m_{max})}}{e^{-\beta E(m_{min})}} \simeq e^{\beta \Delta E_{tnt}} \tag{4}$$

Let us now focus on realistic spin systems with isotropic long–range exchange interaction and on–site anisotropy, described by the following Hamiltonian:

$$H = -J \sum_{i>j} \frac{\vec{S}_i \vec{S}_j}{r_{i,j}^{\alpha}} - D \sum_i (S_i^z)^2 \tag{5}$$

where, $\vec{S}_i$ are the spin vectors with unit length, α determines the range of the interaction among the spins, J is the exchange coupling and D is the on–site energy anisotropy. Even if in the Hamiltonian (5) the exchange interaction is isotropic and the anisotropy is given by the on site energy term, we will also comment later, on the case of exchange anisotropy.

Here, we focus on the case $\alpha = 1$ because it is related to the RKKY interaction which is an effective interaction among magnetic impurities mediated by mobile carriers. In the RKKY interaction the coupling between spins is given by: $J_{RKKY} = (2k_F R \cos(2k_F R) - \sin(2k_F R))/R^4$, where k_F is the Fermi wavevector and R the distance among spins. In the limit of small density of carriers, $k_F R \to 0$ and the RKKY interaction is always ferromagnetic, decaying as $1/R$. At large distance the RKKY coupling oscillates in sign but sometimes a cutoff, related to the carriers localization length has been introduced [21, 22] so that such oscillatory behavior can be neglected.

The energy E_{tnt} can be computed numerically using a minimizing constrained algorithm. We can also estimate analytically E_{tnt} for a generic range of the interaction α. To this purpose let us consider two configurations with $m = 0$: *i*) the first one with all the spins aligned perpendicular to the easy axis. The energy difference of this configuration from the minimal energy is DN, which is the energy barrier due to coherent rotation of all spins; *ii*) a configuration, labeled $\uparrow\downarrow$, consisting ot two neighbors identical blocks with opposite magnetization along the easy axis. This configuration roughly corresponds to what is called nucleation configuration in literature. The

energy difference of this configuration from the minimal energy in the case $\alpha \leq d$, is given by[18]:

$$\Delta E_{\uparrow\downarrow} = JC_{\alpha,d}N^{2-\alpha/d} \tag{6}$$

where $C_{\alpha,d}$ is a constant, and in the case considered here $C_{1,1} \simeq 4\ln 2$.

Following similar reasoning as in [18], it can be shown that the energy of these two configurations is a good approximation of E_{tnt}, so that we can write

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

Eq. (7), valid whenever DN is not close to $\Delta E_{\uparrow\downarrow}$, gives an estimation of the anisotropic energy barrier, which can be used in Eq. (3) to get magnetic decay times. Note that in case of nucleation ($DN > \Delta E_{\uparrow\downarrow}$) , a single spin-flip should be added, so that, $\tau_0 \propto e^{D\beta}$ [3].

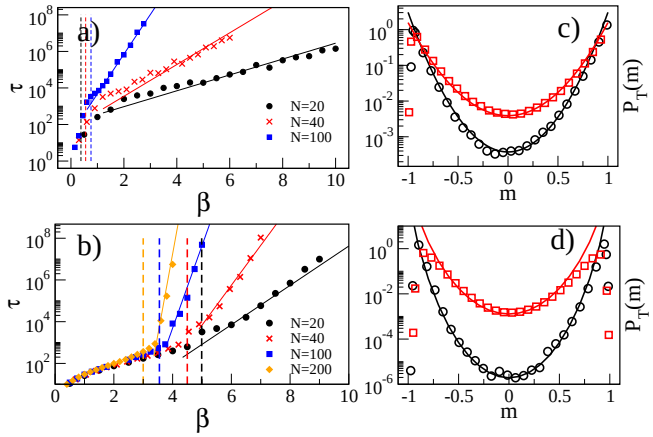

FIG. 1: In panels (a,b), magnetic decay times, τ *vs* the inverse temperature, β, are shown. In panels (c,d), the probability distribution at fixed temperature of the magnetization, $P_T(m)$, is plotted *vs* m. In the panel (a) characterized by coherent rotation we have $J = 1$ and $D = 0.05$, while in the lower (b) (nucleation) $J = 1/16$ and $D = 0.5$. Different symbols refer to numerical results for different number of spins N, as indicated in the legend. Full lines in (a-b) are the theoretical predictions $\exp(\beta\Delta E_{tnt})$. Specifically for coherent rotation, (a), we have $\Delta E_{tnt} = DN$, while for nucleation, (b), we have $\Delta E_{tnt} = \Delta E_{\uparrow\downarrow} + D$ (see text for explanations). Vertical dashed lines in (a-b) refer to the inverse statistical temperature $\beta_{stat} = 1/k_B T_{stat}$. In (c) the probability distribution of the magnetization $P_T(m)$ is shown for $\beta = 6$ (squares), $\beta = 9$ (circles) and $N = 20$. In (d), $P_T(m)$ is shown for the same parameters of (b), $\beta = 8$ (squares), $\beta = 11$ (circles) and $N = 20$. In (c-d) symbols represent numerical data, while curves analytical results.

We analyzed the magnetic decay time in the canonical ensemble, using a modified Monte Carlo simulation [14, 20]. As initial condition we choose all spins aligned along the easy axis, and from the exponential decay of the ensemble average magnetization, $\langle m(t)\rangle \propto e^{-t/\tau}$, we computed the magnetic decay time, τ. Let us consider first the 1–d case: results for magnetic decay times are shown in Fig. 1a) for coherent rotation ($D \ll 4J\ln 2$) and in Fig. 1b) for nucleation ($D \gg 4J\ln 2$) where the latter inequivalences have been obtained comparing Eq. (6) and (7). The good agreement with the theoretical prediction (shown by the straight lines in Fig. 1 a) clearly indicates that Brown's theory of coherent rotation with an anisotropy barrier proportional to the particle volume (DN here) is still at work for long range interacting systems. In Fig. 1b), the case of nucleation is shown. Samples with different number of particles experience different anisotropy energy barriers. As one can see the numerical results indicated by symbols well agree with the theoretical prediction given by Eq. (7), shown as lines. Note that for high temperature (small β values) symbols with different number of spins lye upon the same curve, that turns out to be independent from N.

We can now estimate $P_T(m)$ at low temperature, following the considerations given above. Let us compute $E(m)$ for coherent rotation and nucleation. When the reversal of the magnetization occurs through coherent rotation we have $\vec{S}_i\vec{S}_j = 1$, and $S_i^z = \langle S^z\rangle = m$ so that $E(m) = -J/2\sum 1/r_{i,j}^{\alpha} - DNm^2$. Since the first term is independent of m, for coherent rotation we have: $P_T(m) \propto e^{\beta DNm^2}$. In case of nucleation the possible configurations which the system can visit can be obtained assuming that magnetic reversal occurs by first reversing one of the edge spins, then its nearest neighbor, and then the spin immediately after until all spins are reversed. If we have k spins on the left pointing upwards and $N-k$ spin pointing downwards, we can write the energy of this configuration as $E(k) = E_{min}(k) + E_{min}(N-k) + V$, where V is the interaction energy between the two blocks and $E_{min}(k) = -J/2\sum_{i=1}^{k}\sum_{j\neq i}^{k} 1/r_{i,j}^{\alpha} - Dk$ is the minimal energy for a system of k spins. Collecting all together, we have $E_{min}(N) = E_{min}(k) + E_{min}(N-k) - V$, so that $V = E_{min}(k) + E_{min}(N-k) - E_{min}(N)$, and $E(m) = 2E_{min}(N(1-m)/2) + 2E_{min}(N(1+m)/2) - E_{min}(N)$. From the knowledge of $E(m)$ we obtain $P_T(m) \propto e^{-\beta E(m)}$ which remarkably agrees very well (apart close to $m = \pm 1$) with numerical results in the coherent rotation regime, Fig. 1c) , and in the nucleation regime as well, Fig. 1 d).

Another important point here is in which temperature range the AL with the exponent given by Eq. (7) holds. In the cases of coherent rotation and nucleation, see also Ref.[12], we might expect that the AL is valid only when $k_B T \ll \Delta E_{tnt}$. Clearly this gives an upper bound for the temperature for which Eq. (3) is valid. Moreover it should be $T \ll T_{stat}$, where the latter is the temperature at which the barrier at $m = 0$ in the free energy vanishes (and that coincides with the temperature at which a phase transition occurs in the thermodynamic limit). It is very interesting that, computing T_{stat} by means of a standard mean field approach, one gets a very nice estimate of the validity range of the AL (see dashed vertical

lines in Figs. 1a,b) and 2a,b)).

The exponential dependence of the magnetic decay time on the number of spins, shows the possibility of stable ferromagnetic behavior for nanoscopic single chain magnets with RKKY interactions. Indeed a chain with only 10^5 spins, with an exchange coupling of $J \approx 60$ K and an on-site anisotropy of $D = 3$ K is enough to have a ferromagnetic behavior below 500 K !

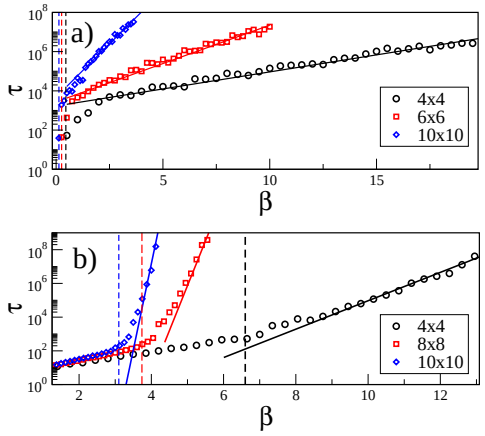

FIG. 2: Decay time τ *vs* the inverse temperature β for a 2–d square lattice with $\alpha = 1$. for $J = 1$ and $D = 0.025$ (a) (coherent rotation); (b) for $J = 1/40$ and $D = 1/2$ (nucleation). Symbols refer to numerical data, while full lines are the analytical prediction Eq. (7). Vertical dashed lines represent the inverse statistical temperature $\beta_{stat} = 1/T_{stat}$. Note that in case of coherent rotation (a), the anisotropy energy is proportional to DN, while in the case of nucleation (b), the anisotropy energy is proportional to $N^{3/2}$. Lattice dimensions have been indicated in the legend

Our results can be extended to higher dimensions. Since $\Delta E_{\uparrow\downarrow} \propto N^{2-\alpha/d}$, from Eq. (7) we have $DN < \Delta E_{\uparrow\downarrow}$, for large N and $\alpha < d$. This implies that the anisotropic energy barrier grows as the volume of the particle, V (with $V \propto N$), in all dimensions, as the particle volume becomes large enough. In the critical case $\alpha = d$, both terms grows like N, and again we have a volume dependent energy barrier. This is confirmed by our simulations for a 2–d system, see Fig. 2a). Note that Eq. (7) also implies that for small particles we can have an anisotropy energy barrier which grows faster than the volume of the particle, see Fig. 2b). Indeed in that case we have an anisotropy energy barrier growing as $N^{3/2} \propto V^{3/2}$. Another interesting consequence of Eq. (7) is that in the case of a long range exchange anisotropy without on–site anisotropy ($D = 0$) we have $\Delta E_{tnt} \propto V^{2-\alpha/d}$, and thus faster than the volume for *any particle size*. For instance for the relevant case $\alpha = 1$, we have that the anisotropy energy barrier grows as $V^{3/2}$ for 2–d systems and as $V^{5/2}$ for 3–d systems.

Finally we stress that the validity of Eq. (3) and of our method to approximate $P_T(m)$ at low temperature, have been tested for a wide range of values of α (even for short range interactions) and it will be reported elsewhere [23].

In conclusion we propose a general method to determine anisotropic energy barrier in spin systems. The barrier, which determines magnetic decay times, can be computed from the disconnected energy portion in the corresponding isolated systems. Our analysis shows that adding a small on site anisotropy to an isotropic long range exchange interaction induces a magnetic decay time which depends exponentially on the volume of the particle, for large enough particle size. We also pointed out that for long range interaction and in presence of exchange anisotropy, the anisotropic energy barrier grows faster than the volume of the particle. This remarkably contrast with the behavior of short range interacting systems, where the energy barrier is proportional to the cross-sectional particle area, rather than to its volume, for large enough particle size or aspect ratio. Finally we pointed out that the predicted enhancement of the anisotropic magnetic barrier can induce stable ferromagnetic behavior in finite size systems, in particular the possibility to have stable ferromagnetism at room temperature in nanomagnets with RKKY interaction has been discussed.

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