Time relaxation of interacting single–molecule magnets

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Abstract

We study the relaxation of interacting single–molecule magnets (SMMs) in both spatially ordered and disordered systems. The tunneling window is assumed to be, as in Fe₈, much narrower than the dipolar field spread. We show that relaxation in disordered systems differs qualitatively from relaxation in fully occupied cubic and Fe₈ lattices. We also study how line shapes that develop in “hole–digging” experiments evolve with time t in these fully occupied lattices. We show (1) that the dipolar field h scales as tᵖ in these hole line shapes and show (2) how p varies with lattice structure. Line shapes are not, in general, Lorentzian. More specifically, in the lower portion of the hole, they behave as (|h|/tᵖ)^(1/p)−1 if h is outside the tunnel window. This is in agreement with experiment and with our own Monte Carlo results.

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I. INTRODUCTION

Magnetic relaxation in crystals of single–molecule magnets (SMM’s), such as Fe$_8$, has become a subject of great interest. At low temperature, $T$, each SMM behaves approximately as a single spin $S$. Magnetic relaxation at $k_BT \lesssim 0.1U/S$, where $U$ is a magnetocrystalline anisotropy barrier, is temperature independent, and is duly attributed to quantum tunneling under the barrier. Hyperfine interactions with nuclear spins as well as dipole–dipole interactions among all SMM electronic spins give rise to a variety of phenomena that are not yet fully understood. Hyperfine interactions enable spins to tunnel even when the ensuing Zeeman energy change $2\varepsilon_h$ is much larger than the tunnel splitting energy $\Delta$, provided $|\varepsilon_h|$ is smaller than some $\varepsilon_w$. For Fe$_8$, for instance, $\Delta \sim 10^{-3}$ mK, $\varepsilon_w \approx 10$ mK, and the rms value of the Zeeman energy $\delta\varepsilon_h$ is approximately 400 mK. We shall restrict ourselves to systems with $\varepsilon_w \ll \delta\varepsilon_h$. In these systems, the relaxation of the magnetization goes on long after time $\Gamma^{-1}$, where $\Gamma$ is the spin tunneling rate for spins within the tunneling energy window (that is, spins for which $|\varepsilon_h| \approx \varepsilon_w$). Tunneling spins give rise to changing dipolar fields, which in turn bring new spins into the tunneling energy window, thus keeping a magnetic relaxation process from extinction. In this paper, we focus our attention on effects that stem from this process.

We consider here experiments of the sort that were reported by Wernsdorfer et al. in Ref. [6]. In them, a crystalline sample of SMM’s is first quenched from $k_BT \sim U/S$ to $k_BT \lesssim 0.1U/S$ in either (a) a weak applied magnetic field of a few mT, and later observed after the field is switched off at time $t = 0$ or (b) a zero field, and later observed after a weak field is applied at $t = 0$. We shall refer to the former as a FC (for field cooled) experiment and to the latter as a ZFC (for zero field cooled) experiment. Both types of experiments can be lumped into one defining $\mbox{\~m} \equiv 1 - m/m_0$, where $m_0$ is the initial magnetization in a FC experiment, and $\mbox{\~m} \equiv m/m_s$, where $m_s$ is the final steady state magnetization in a ZFC experiment. Wernsdorfer et al. observed the $\mbox{\~m} \propto t^{1/2}$ over roughly two time decades. The time evolution of “holes” that, under suitable conditions, develop in a magnetization density function, have also been reported. The existing theory at the time predicted a universal $\sqrt{t}$ short time relaxation from a fully polarized system, but said little about relaxation from or into weakly polarized states. Other theories take hyperfine interactions into account but disregard dipole–dipole interactions. They therefore apply if $\delta\varepsilon_h \lesssim \varepsilon_w$ which is not
within the scope of this paper.

We have developed a theory\cite{12,14} that gives the time evolution both of $\tilde{m}$ and of the holes' line shapes in weakly polarized systems of interacting SMMs, such as Fe$_8$, in which $\varepsilon_w \ll \delta \varepsilon_h$. There are three clearly discernible time regimes. For $\Gamma t \lesssim 1$, $\tilde{m} \propto \Gamma t$. In the second time stage, when $1 \lesssim \Gamma t$, up to some time before $\tilde{m} \sim 1$, $\tilde{m} \propto t^p$, at least for all fully occupied cubic lattices. Moreover, the theory gives a simple relation that specifies how $p$ varies with lattice structure. In FC experiments, $\tilde{m} \sim 1$ in the third time stage, that is, $m \approx 0$. The third time stage is more interesting in ZFC experiments. Then $m(t)$ settles down temporarily to a quasi stationary value $m_s$, which the theory predicts, if either $k_B T \gg \varepsilon_w$ or if the heat exchange rate with the lattice is much smaller than $\Gamma$; on the other hand, if $k_B T \gg \varepsilon_w$ is not fulfilled and heat exchange rate with the lattice is not much smaller than $\Gamma$, then the relaxation of the magnetization shifts into a thermally driven approach to equilibrium, skipping the quasi stationary state.

A quite different treatment of relaxation from weakly polarized states that gives $1 - m/m_0 \propto \sqrt{t}$, independently of the spins' spatial distribution, is given by Tupitsyn, Stamp, and Prokof'ev (TSP) in Ref. \cite{15}, criticized in Ref. \cite{13}, and defended in Ref. \cite{16}. TSP’s treatment of relaxation from weakly polarized states is rather unrelated to the earlier theory\cite{5} of Prokof’e’v and Stamp for a $\sqrt{t}$ relaxation from fully polarized states, about which we have nothing to say here. According to TSP, $\sqrt{t}$ relaxation in weakly polarized systems holds as long as $1 \lesssim \Gamma t$ and $\tilde{m} \ll 1$, that is, roughly, over the second time stage. This is also the time domain where our theory gives $\tilde{m} \simeq (\varepsilon_w/\delta \varepsilon_h)(\Gamma t)^p$ for fully occupied lattices. This time stage, sometimes referred to as “short times”, can in fact be arbitrarily large, for arbitrarily small $\varepsilon_w$, since $\tilde{m} \ll 1$ only implies $\Gamma t \ll (\delta \varepsilon_h/\varepsilon_w)^{1/p}$. Note also that $\sqrt{t}$ relaxation from fully polarized systems ends sooner, when $t \sim \delta \varepsilon_h/\varepsilon_w$. This may explain why Wernsdorfer\cite{6} et al. were able to observe $\tilde{m} \sim \sqrt{t}$ in Fe$_8$ ($p \approx 0.5$ for Fe$_8$) under weak applied magnetic fields for up to some $10^3$ s while Ohm\cite{17} et al. observed $\sqrt{t}$ relaxation from fully polarized Fe$_8$ for only up to $10^2$ s.

The two approaches, ours and TSP’s, are quite different. The underlying assumptions are as follows. In Ref. \cite{15}, the main result follows from a claim that is made on the function $f(h, t) = p_\downarrow(h, t) - p_\uparrow(h, t)$, where $p_\uparrow(h, t)$ and $p_\downarrow(h, t)$ are the number densities of up- and down–spins, respectively, with a dipolar field $h$ acting on them at time $t$. In its latest form\cite{16} the claim is that the scaling form $f(h, t) = f(h/t^2)$ “was found to be valid in our MC simulations for different lattice types...” when $1 \lesssim \Gamma t$ and $\tilde{m} \ll 1$. Since, the magnetization $m$
and \( f(h, t) \) are clearly related by \( m(t) = -\int dh f(h, t) \), a \( \sqrt{t} \) relaxation follows immediately. As we have shown recently, the above scaling form holds approximately for SC and Fe\(_8\) lattices (as defined in Ref. [18]), but not in general. It fails, for example, for FCC, BCC, and diamond lattices. Our theory also gives the time evolution of \( f(h, t) \), but follows from a more fundamental assumption: that the dipolar field on any one given site changes by some random amount \( \Delta h \), whenever a spin flips somewhere else for the first time, and that \( \Delta h \) follows a Lorentzian distribution (for more details see Sects. II A and III A, and Ref. [14]). The integro-differential equations for the evolution of \( f(h, t) \) and of the magnetization that obtain in our theory, follow from this assumption.

Unfortunately, as far as we know, only experiments on a crystalline Fe\(_8\) structure have thus far been performed. However, MC simulations have been performed for various fully occupied cubic lattices, which have given values of \( p \) that agree with our predictions.13,14 This paper’s first aim is to extract from our theory how the magnetization is supposed to relax in Fe\(_8\), compare this with experiment over the time span where published experimental data exists and make predictions for later times. It is also our purpose to predict, and check with MC simulations, how \( m \) relaxes with \( t \) for other spatial distributions, namely, under full spatial disorder, thus providing another test for our theory.

The holes observed in the experiments described above are also of interest. They correspond to “wells” that develop in the function \( f(h, t) = p_\downarrow(h, t) - p_\uparrow(h, t) \) (defined above). From the relation \( m(t) = -\int dh f(h, t) \), the time evolution of \( m \) follows, but \( f(h, t) \) provides additional information about the magnetic evolution of the system that \( m(t) \) does not. For short times, that is, for \( \Gamma t \lesssim 1 \), the hole’s width is equal to \( \varepsilon_w \). This was first surmised by Wernsdorfer et al.\textsuperscript{6} to propose a number, approximately 10 mK, for the tunneling energy window, \( \varepsilon_w \). However, we know of no published data for the holes’ line shape evolution well into the intermediate time range, that is, for \( 1 \ll \Gamma t \). Our second aim is to fill this gap. To this end, we work out from our theory the time evolution of holes’ line shapes in this time stage in fully occupied cubic systems and Fe\(_8\) crystals, and check the results obtained against our MC results. We also obtain new results of a more general nature for the holes’ line shape. Before we state our results, we specify the model.
A. The model

All spins are on a lattice, they point along the easy anisotropy axis, and interact as magnetic dipoles. We consider both fully and partially occupied lattices. Let the magnetic field at site \( i \) produced by spin \( S_j \) at site \( j \) be given, in the usual notation, by

\[
h_{ij} = h_d \left( \frac{a}{r_{ij}} \right)^3 \left[ 1 - 3 \left( \frac{z_{ij}}{r_{ij}} \right)^2 \right]
\]

where \( r_{ij} \) is the distance between the \( i \) and \( j \) sites, \( a \) is the distance between nearest neighbor sites, \( h_d = \left( \frac{\mu_0}{4\pi} \right) g \mu_B S / a^3 \), and \( S_i = \pm S \) for all \( i \). Furthermore, let the magnetic field \( h_i \) at site \( i \) be given by \( h_i = \sum_j h_{ij} \), where \( \sum_j \) is over all occupied lattice sites. The tunnel window size and tunneling rate \( \Gamma \) are defined next. At very low temperature, that is, if \( k_B T \lesssim \frac{0.1 U}{S} \), a spin can flip only if the field \( h \) acting on it satisfies \( |h| < h_w \). The flipping rate is \( \Gamma \) if upon tunneling the energy decreases, but if the energy increases by \( \Delta E \), then the rate is \( \Gamma \exp(-\Delta E/k_B T) \), following detailed balance. (Even though \( |\Delta E| < \varepsilon_w \), and usually \( k_B T \gg \varepsilon_w \), \( \exp(-\Delta E/k_B T) \) is not quite equal to 1, and this makes a difference after a sufficiently long time, as is shown below.) We also simulate relaxation processes in which the energy \( E \) is assumed to remain constant (such as if no spin-lattice relaxation takes place). Then, we assume a value of \( T \) is such that \( E \) remains approximately constant. No tunneling window restriction applies for spin flips if \( k_B T \gtrsim \frac{U}{S} \).

We let \( p(h, t) \) be the probability density function (PDF) that any one given spin have field \( h \) at time \( t \), let \( p_0(h) \) be the same distribution for a completely random spin configuration, and let

\[
\sigma = \left[ \sqrt{2\pi} p_0(0) \right]^{-1}.
\]  

(1)

For a Gaussian field distribution, \( \sigma \) is equal to the dipole field rms value \( \delta h \) for a random spin configuration (see Table I), but this is not so in general. Values of \( \sigma \) that follow from MC simulations for cubic and Fe\(_8\) lattices with randomly oriented spins are given in Table I. Finally, let \( h_0 = (8\pi^2/3^{5/2}) h_d \tilde{n} \), where \( \tilde{n} \) is the number of dipoles per unit cubic cell.\(^{25}\) Randomly oriented spins on a cubic lattice give a Lorentzian field distribution of \( h_0 \) half width at half maximum if \( \tilde{n} \ll 1 \).\(^{26}\) Values we will be using for \( h_0 \) are given in Table I. From here on, unless otherwise stated, all magnetic fields and energies are given in terms \( h_d \) and \( g\mu_B h_d S \), respectively.
TABLE I: Quantities $h_0$, $\sigma$, and $\sqrt{\langle h^2 \rangle_0}$ are given for randomly oriented spins on the lattices specified. $h_0 = \tilde{n} 8\pi^2 / 3^{5/2}$, $\sigma \equiv [\sqrt{2\pi p(0)}]^{-1}$, where $p(0)$ stands for the PDF at $h = 0$, and $\sqrt{\langle h^2 \rangle_0}$ is the rms spatial average of the dipolar field. For all lattices except for Fe$_8$, $h_0$, $\sigma$, and $\sqrt{\langle h^2 \rangle_0}$ are given in terms of $h_d$. All lattices are fully occupied, except for the “dilute SC”, which stands for a SC lattice with $\tilde{n} \ll 1$ sites occupied. Finally, $\alpha = 8\pi^2 / 3^{5/2}$.

| LATTICE   | $h_0$  | $\sqrt{\langle h^2 \rangle_0}$ | $\sigma$ |
|-----------|--------|-------------------------------|---------|
| SC        | $\alpha$ | 3.655 | 3.83(2) |
| BCC       | $2\alpha$ | 3.864 | 4.03(2) |
| FCC       | $4\alpha$ | 8.303 | 8.44(2) |
| Fe$_8$    | $47(1)$ mT | $46(1)$ mT | 31(1) mT |
| dilute SC | $\alpha \tilde{n}$ | $\sqrt{\pi / 2h_0}$ |         |

$^b$ We assume an easy anisotropy axis as given in Refs. [23,24,28].

B. Plan and main results

Section II is devoted to the relaxation of the magnetization. The equations from our theory which we use to calculate the time evolution of the magnetization are restated in Sect. II A. The results that follow from them for Fe$_8$ crystals are shown to agree rather well with experiment and with our own MC results in Sect. II B. The evolution we predict for $m(t)$ as well as our MC results cover a time span that is 2 orders of magnitude longer than the experimental one $\tau_E$ ($\tau_E \approx 20$ minutes). Let $\tau_w$ be the end time for the regime where $m \propto t^{p}$. We showed in in Ref. [14] that $\tau_w \approx \Gamma^{-1}(\sigma / h_w)^{1/p}$, from which we obtain $\tau_w \sim 10\tau_E$. For $\tau_w \lesssim t$, the evolution of $m(t)$ is shown to depend sensitively on $T$ if good thermal contact with a heat reservoir is assumed. If on the other hand we assume constant energy processes (i.e., no spin-lattice relaxation), then $m$ levels off, if only temporarily, to a stationary value $m_s$ when $\tau_w \lesssim t$. The value of $m_s$ we obtain from theory is unrelated to the equilibrium value of $m$, which only obtains much later. This stage, when $m \rightarrow m_s$, sets in after most spins in the system have tunneled at least once after the magnetic field is applied. Simulations bear this out. We also obtain, from theory as well as from MC simulations, $m(t)$ for spatially disordered systems. More specifically, we make a random selection of a fraction $\tilde{n}$ of $L \times L \times L$ SC lattice sites and place spins on them. For $\tilde{n} \lesssim 0.1$, we assume full disorder.
Then, theory predicts a magnetic relaxation that bears no resemblance to a $\sqrt{t}$ rule, not even to the $tp$ rule that we obtain for fully occupied SC lattices. Instead, for $\tilde{n} \lesssim 0.1$,

$$\tilde{m} \simeq \frac{80h_w}{\sigma} e^{-(t/\tau_m)p},$$  

approximately, where $\tau_m \simeq 10^6 \Gamma^{-1}$ and $q \simeq -0.105$. Results from our MC simulations are in fair agreement with this. In Sect. III we report results for holes’ line shapes in fully occupied crystal lattices. The main results for line shapes, which are derived in Sect. IIIA follow. When $1 \ll \Gamma t \ll (\sigma/h_w)^{1/p}$ and $h_w \ll |h + H| \ll \sigma$,

$$f(h, t)/f(h, 0) \propto |\eta|^{\frac{1}{p} - 1}$$  

if $|\eta| \ll 1$, where

$$\eta \equiv \frac{h + H}{h_w(\Gamma t)^p}. \tag{4}$$

$H$ is an applied field, and $p$ is given by

$$p^{-1} \sin \pi p = \sqrt{2\pi \sigma / h_0}. \tag{5}$$

In Sect. IIIA we also derive relation for holes line shapes that hold over longer time spans: $1 \lesssim \Gamma t \lesssim (\sigma/\varepsilon_w)^{1/p}$. In Sect. IIIB we apply these results to published experimental data for Fe$_8$ and to MC data for Fe$_8$ as well as to fully occupied FCC lattices. Finally, concluding remarks appear in Sect. IV.

II. RELAXATION OF THE MAGNETIZATION

A. Theory

We first describe a stochastic model which helps to understand the physics of the problem as well as the statistical assumption we have made in order to solve it. Consider two tracks, both filled with particles. Let there be one particle on the “up-track” for each up spin on a lattice, and one particle in the “down-track” for each down spin. Let all particles on each track be ordered according to the value of the magnetic field $H + h$ acting on each spin. To mimic tunneling, random select a particle within the tunnel window, that is, a particle fulfilling $-h_w < H + h < h_w$, whether on the up- or down-track, and move it to the “point” $H + h$ on the opposite track. In order to mimic the effect such a spin flip has on the
dipolar fields on other spins, draw a random value of $\Delta h$ for each particle from a Lorentzian distribution of half width $h_0/N$, where $N$ is the total number of spins, and let $h \rightarrow h + \Delta h$ if the particle that just shifted track has done so for the first time. This latter proviso is related to the fact that no effect on the dipolar field follows when the same spin flips twice (for a more detailed explanation, see Ref. [14]). Repeat this whole process at every tic of a clock. Clearly, the whole process stops when all particles have jumped track at least once.

We can get a feeling for the relevance of lattice structure or spatial spin distribution from the following simple consideration. $Np(0)2h_w$ is the number of particles in the tunnel window, which is the number of times the clock tics in time $\Gamma^{-1}$, which, in turn, would be the relaxation time for the equilibration of the number of up and down particles in the tunneling window if there were no “field shifting”. Now, the median field shift, or diffusion length, in time $\Gamma^{-1}$ is $2p(0)h_0h_w$, that is, a fraction $p(0)h_0$ of the tunnel window’s width. Thus, $p(0)h_0$ is a measure of the amount by which the unbalance between up and down particles is restored in the tunnel window in time $\Gamma^{-1}$. Therefore, the relaxation rate clearly depends on $p(0)h_0$, which in turn depends on lattice structure. This shows why the latter is relevant to the relaxation of the magnetization.

The simple statistical assumption above has enabled us to derive the equations we need for the calculation of the time evolution of the magnetization. These equations, which we reproduce in a compact form immediately below, give the magnetization $m(t)$ at time $t$, and $n(t)$, the fractional number of spins that flip at least once in time $t$. We first recall an important ingredient of the theory for ZFC experiments, the energy per spin at the time when the system is quenched, which we refer to as the “annealing energy”, is $-\varepsilon_a$. Let $x_1 = mg\mu_B S\sigma(\langle h^2 \rangle_0/(\varepsilon_a h_w H)$ and $x_1 = 2(\sigma/h_w)(1-m/m_0)$, for ZFC and FC experiments (defined in the Introduction), respectively, and $x_2 = n\sigma/h_w$ for both FC and ZFC experiments. The desired equation follows,

$$
\frac{dx_j}{dt} \simeq a_j \sqrt{\frac{2}{\pi}} - b_j \int_0^t d\tau \frac{dx_j(\tau)}{\omega(t-\tau) + 1},
$$

(6)

where

$$
\omega(t') \simeq \min \left[ \frac{\pi h_0}{2\sigma} x_2(t'), \sqrt{\frac{\pi\sigma}{2h_w}} x_2(t') \right],
$$

(7)

$a_1 = 4$, $b_1 = 2$. In order to obtain $x_1$, Eq. (6) must first be solved for $j = 2$, letting $a_2 = 1$ and $b_2 = 1$, in order to then use $x_2(t)$ in Eq. (7), and thus enable substitution of $\omega$ into Eq. (6) for $j = 1$. 

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The theory applies if $h_w \ll \sigma$ and the energy is constant, that is, if no energy transfer to the phonon bath takes place. This is also approximately so if $kT \gg \varepsilon_w$. If the constant energy condition is not met in a ZFC experiment, a linear in time magnetization relaxation that is thermally driven takes over before $m(t) \rightarrow m_s$. The theory also gives

$$m_s \simeq 2H\varepsilon_a/(gS\mu_B\langle h^2 \rangle_0).$$

(8)

Note that the definition of $x_1(t)$, together with Eqs. (6), (7), and (8) imply that the time variation of $m(t)/m_s$ in a ZFC experiment is the same as $1 - m/m_0$ in a FC experiment.

**FIG. 1:** $\tilde{m}$ and $n$ (the fractional number of spins that have flipped in time $t$ at least once) versus time. Full lines are for theory and symbols stand for MC results for 65536 spins that are initially up or down at random, with probabilities 0.6 and 0.4, respectively, on a FCC lattice. $\blacklozenge$ ($\circ$) stand for $\tilde{m}$ for $h_w = 0.1$ (0.01); $\square$ stand for $n$ for $h_w = 0.01$. The dash–dotted lines stand for slopes $p = 0.73$, and $p = 0.5$, which follow for FCC lattices from Eq. (5) and from Ref. [15], respectively. Data points taken from Ref. [16] are shown as $\blacklozenge$. Our data points for $\Gamma t < 1024$ come from averages over 1400 MC runs, and for 100 runs for $t > 1200$.

A few remarks about Eqs. (6) and (7) are in order. Clearly, $x_1(t)$ only depends on two parameters: $h_0/\sigma$ and $\sigma/h_w$. The latter only comes into play at the later portion of the time evolution, when $n(t) > 2\sigma^2/(\pi h_0^2)$, which is when $m(t)$ starts leveling off. Now, $0.4 \lesssim \sigma/h_0 \leq \sqrt{\pi/2}$ for all cubic lattices, whether fully occupied or not. It follows that leveling off of $m(t)$ is triggered some time (see below) while $n(t) \leq 1$. This has to do with the fact that a spin flip contributes to hole widening in $f(h, t)$ the first time it takes place after the field is switched on. When a spin flips a second time, it only returns to its initial state, thus it cancelling the effect of the first flip. Since $m$ is proportional to the width
of the hole in \( f(h,t) \) when \( \Gamma t \gtrsim 1 \) it follows that \( m(t) \) becomes constant when \( n \sim 1 \).

The time when \( m(t) \) levels off is illustrated in Fig. 1 for fully occupied FCC lattices. For instance, for \( h_w = 0.01 \), theory predicts \( m(t) \) to start leveling off when \( n(t) \simeq 0.11 \) since \( 2\sigma^2/(\pi h_0^2) \simeq 0.11 \) then. Thus, a significant portion of the evolution, up to \( n \sim 1 \), only depends on \( h_0/\sigma \). Information about the lattice or degree of spatial disorder only comes into the equations through this number. Other numbers, such as \( h_w, \varepsilon_a, \) and \( H \) only come into the proportionality factor between \( x_1 \) and either \( m \) or \( 1 - m/m_0 \). The temperature does not enter anywhere into the equations.

We have shown analytically in Ref. [14] that \( m \propto t^p \) and \( n \propto t^p \) fulfill Eq. (6) for \( j = 1 \) and \( j = 2 \) in the \( 1 \ll \Gamma t \ll (\sigma/h_w)^{1/p} \) time range if \( p \) is given by Eq. (5). Numerical solutions of Eq. (6) shows that \( m \propto t^p \) ensues in the wider \( 1 \lesssim \Gamma t \lesssim (\sigma/h_w)^{1/p} \) time span for all fully occupied cubic lattices. We show below that this is also so for a fully occupied Fe\(_8\) lattice.

Spatially disordered systems behave differently. Consider a very small fraction (\( \tilde{n} \ll 1 \)) of sites of a SC lattice to be occupied by randomly oriented spins. Then, a Lorentzian dipolar magnetic field distribution of half–width \( h_0 \) ensues. It follows then, from the definition of \( \sigma \) and of \( h_0 \), that \( \sigma/h_0 = \sqrt{\pi/2} \). The number \( p = 0 \) follows then from Eq. (5). Recall, however that theory only implies that this ensues only when \( \Gamma t \gg 1 \). For earlier times, more specifically, for all \( \Gamma t \gtrsim 1 \), we find that the numerical solution from Eq. (6) is well fitted by Eq. (2) if \( \tilde{n} \lesssim 0.1 \). Numerical solutions of Eq. (6) are plotted in Fig. 2a for \( \tilde{n} = 1 \) and \( \tilde{n} = 0.6 \), both for \( h_w = 0.02 \) and in Fig. 2b for (1) \( \tilde{n} = 0.1 \) and \( h_w = 0.02 \), and for (2) \( \tilde{n} = 0.03 \) and \( h_w = 0.006 \). Note that the same solution obtains for the latter two cases. We come back to these figures in Sect. II B.

B. Comparison with experiments and simulations

We first make use of Eqs. (6) and (7) to obtain \( m(t) \) for Fe\(_8\). Some numbers must first be fed into Eqs. (6) and (7). For \( h_w \), we use \( 0.8 \) mT, as given in Ref. [6]. We use \( \Gamma = 0.04 \) s\(^{-1} \), (see Refs. [14] and [21]). With the numbers given in Table I for \( \sigma \) and \( h_0 \), we obtain \( x_1(t) \) and \( x_2(t) \) numerically from Eqs. (6) and (7). Finally, the value of \( -\varepsilon_a \), the annealing energy, is needed in order to obtain \( m \) from \( x_1 \). Not knowing \( \varepsilon_a \), we treat it as a fitting parameter. We find \( \varepsilon_a \simeq 36 \) mK fits best the experimental data points from Ref. [6], which
FIG. 2: (a) Magnetization versus $\Gamma t$. Lines are for theory and symbols are for data from MC simulations of $L \times L \times L$ SC lattices with a fraction $\tilde{n}$ of their sites occupied. For $\tilde{n} = 0.6$ the dashed line is for theory, ♦, and ♠ are for MC data for $L = 32$ and $L = 16$, respectively. For $\tilde{n} = 1$ the full line is for theory, △, and ▲ are for MC data for $L = 32$ and $L = 16$, respectively. All symbols are from averages of 800 MC runs while $\Gamma t < 10^3$ and 40 MC runs when $\Gamma t > 10^3$. (b) Same as in (a) but for $\tilde{n} = 0.1$ and $h_w = 0.02$, and for $\tilde{n} = 0.03$ and $h_w = 0.006$. The full line is for theory for both values of $\tilde{n}$. For $\tilde{n} = 0.03$, ○, and ● stand for MC data for $L = 64$ and $L = 32$, respectively. For $\tilde{n} = 0.1$, ▼, and ◄ stand for MC data for $L = 32$ and $L = 16$, respectively. As in (a), all symbols are from averages of 800 MC runs while $\Gamma t < 10^3$ and 40 MC runs when $\Gamma t > 10^3$.

are shown in Fig. 3 for a few applied fields. The energy $-36$ mK may be compared to the approximate value, $-500$ mK, of the ground state energy.28

The MC data points shown in Fig. 3 follow from simulations in which the system first evolves at some high temperature (a few Kelvin) for a short time (less than 1 MC sweep) until the energy equals $-36$ mK. At such temperatures, Fe$_8$ cluster spins are not forced to tunnel through the ground state doublet. Accordingly, all spins are allowed to flip, regardless of the dipolar field acting on them. We explore different scenarios after quenching. In our theory, we assume no energy exchange takes place between the spin system and a heat reservoir. We have also performed MC simulations under this assumption. This is approximately realized for the time range exhibited in Fig. 3 by flipping only spins within the tunnel window,
and then with equal probabilities for upward or downward flips. If, on the other hand, heat exchange does take place readily, as one gathers from Ref. 29, where heat exchange rates that are comparable to $\Gamma$ are found, then detailed balance should be enforced in MC simulations. The results of doing this lead to the plots shown in Fig. 3 for $T = 40$ and 300 mK.

![Graph showing magnetization versus time](image)

**FIG. 3:** Magnetization versus time, in hours. Units for $m$ are such that $m = 1$ for full polarization. ♦, □ and ○ are for experimental data taken from Ref. [6] for $H = 3.92, 2.24, \text{ and } 1.12\text{ mT, respectively. Full lines are from our MC simulations for } kT \gg \varepsilon_w, \text{ and dashed lines are for theoretical predictions, that is, from Eqs. (6) and (7). Data points that follow from MC simulations for } H = 3.92\text{ mT are also shown for } T = 40 (\diamond) \text{ and } T = 300 \text{ mK (△). We assumed } h_w = 0.8 \text{ mT and used the values of } \sigma \text{ and } h_0 \text{ that are given in Table I. In the simulations, the initial state was prepared at } T = 2 \text{ K. At this temperature we allowed the simulation to proceed in time up to the point when the energy of the system reached } -36\text{ mK, which is } 0.07 \text{ of the ground state energy. This is the value of } \varepsilon_a \text{ we used in order to relate } x_1 \text{ and } m, \text{ just above Eq. (6). We treat one MC sweep as } \Gamma t = 1 \text{ and assumed } \Gamma = 0.04 \text{ s}^{-1} \text{ in order to convert MC sweeps to hours. Note that } \tau_w \simeq 3 \text{ hours, since } \sigma \simeq 31 \text{ mT and } h_w \simeq 0.8 \text{ mT.}

Results obtained from theory, in Sect. IIIA for very disordered systems are shown in Fig. 2 for $\tilde{n} = 0.1 \text{ and } h_w = 0.02 \text{ and for } \tilde{n} = 0.03 \text{ and } h_w = 0.006$. Monte Carlo data points are also shown for the same values of $\tilde{n}$ and of $h_w$. Data points for $\tilde{n} = 0.1 \text{ and } h_w = 0.02 \text{ fall on top of data points for } \tilde{n} = 0.03 \text{ and } h_w = 0.006$. This is as expected, since $\sigma \propto \tilde{n}$, for for full spatial disorder, implies that $\sigma/h_w$ has the same value in both cases and theory predicts independence from any other parameter. The fitting function from Eq. (2) falls
right on top of the curve for theory in Fig. 2b, and cannot therefore be shown separately.

Finally, we consider size effects. For a $16 \times 16 \times 16$ lattice and $\tilde{n} = 0.1$, for instance, approximately 410 spins make up the system. Of these, only approximately a fraction $2p(0)h_w$ are within the tunnel window. That is, approximately $820h_w/\pi h_0$ spins, which only amounts to some 12 spins, are within the tunnel window. For this reason, we also simulated $32 \times 32 \times 32$ and $64 \times 64 \times 64$ lattices for $\tilde{n} = 0.1$ and $\tilde{n} = 0.03$ respectively. Monte Carlo data points are shown in Fig. 2. Clearly, no significant size effects are observed.

III. TIME EVOLUTION OF THE LINE SHAPE

In this section we first derive some results for $f(h, t)$ that are valid whenever $m \propto t^p$ holds. We know from Ref. [14] and from the previous section that $m \propto t^p$ holds in the time span $1 \lesssim \Gamma t \lesssim (\sigma/h_w)^{1/p}$ for fully occupied SC, FCC, BCC, and Fe$_8$ lattices, but we now know it is not so for spatially random systems. The results we derive below are applied to fully occupied Fe$_8$ and FCC lattices and are compared to results from experiment (for Fe$_8$) and from MC simulations.

A. Theory

The starting point for the derivation are the following two equations, from Ref. [14],

$$g(h, t) \simeq \int_0^t d\tau \frac{dm(\tau)}{d\tau} G(h + H, t - \tau) \tag{9}$$

and

$$G(h, H, t - \tau) \simeq \frac{u(t - \tau)}{\pi[(h + H)^2 + u(t - \tau)^2]}, \tag{10}$$

where $g(h, t) \equiv f(h, 0) - f(h, t)$, $u(t - \tau) \equiv h_0 n(t - \tau)$, and $n(t - \tau)$ is the fractional number of spins that flip at least once in time $t - \tau$. The rationale for these two equations is given next, but (if $|h + H| \ll \sigma$ and $\Gamma t \ll (\sigma/h_w)^{1/p}$) Eqs. (9) and (10) also follow from Eqs. (13) and (14) of Ref. [14], respectively. Assume that, between times $t$ and $\tau$, a fraction $n(t - \tau)$ of all spins flip at least once and that $n(t - \tau) \ll 1$. This can later be checked to be fulfilled if $t \ll \tau_w$, where $\tau_w \equiv (\sigma/h_w)^{1/p}$. Then, Eq. (10) gives the probability density $G(h, H, t - \tau)$ that, at time $t$, the field is $h + H$ at a site where the field at time $\tau$ was $0.26$. To understand Eq. (9), note first that the definition of $g(h, t)$ implies that $g(h, t)$...
FIG. 4: (a) $f$ versus $\eta$, defined in Eq. (4), for the times shown. All symbols stand for Wernsdorfer’s experimental data for Fe$_8$ for $|h| > 2h_w$. Thin lines are for data points for $|h| < h_w$, to which Eq. (8) does not apply. The full line stands for Eq. (12), using $h_w = 0.8$ mT and $p = 0.58$, from Eq. (5), and $\sigma/h_0 = 0.66$, from Table I. (b) Same as in (a) but for MC simulations of Fe$_8$. Points from within the tunnel window have been excluded. Times are given in seconds, after converting each MC sweep into 25 seconds, which is the value we assign to $\Gamma^{-1}$. All symbols stand for averages over $1.6 \times 10^5$ MC runs of systems of $16 \times 16 \times 16$ spins. The full line stands for Eq. (12).

must fulfill $\int dh g(h,t) = m(t) - m(0)$. Equation (9) does give $m(t) - m(0) = \int d\tau dm/d\tau$, since $\int dh G(h,H,t-\tau) = 1$. Similarly, a variation in the magnetization $(dm/d\tau)d\tau$ coming from some spin flipping between times $\tau$ and $\tau + d\tau$, when the field $h + H$ acting on them was within the tunnel window, contributes to $g(h,t)$ with a Lorentzian curve whose width is $h_0 n(t-\tau)$, where $n(t-\tau)/2$ is approximately the fraction of the total number of sites where spins at time $t$ point opposite to the way they did at time $\tau$. $\tilde{m}(\tau)$, it is

Furthermore, the area under the Lorentzian must be given by $(dm/d\tau)d\tau$. That explains Eq. (9).

We make use of $n(t-\tau) \simeq \tilde{m}(t-\tau)$ and of

$$\tilde{m}(\tau) \simeq 1.1 \frac{h_w}{\sigma}(\Gamma\tau)^p$$

which has been shown to hold for all fully occupied cubic and Fe$_8$ lattices in Ref. [14] and
in Sect. II B, respectively, while $1 \lesssim (\Gamma \tau)^p \lesssim \sigma/h_w$. We then divide by $f(h, 0)$, which, for $h \ll \sigma$ is approximately given by $m_0/\sqrt{2\pi\sigma}$. Finally, the change of variable $x \equiv \tau/t$ brings, if $1 \ll \Gamma t \ll (\sigma/h_w)^{1/p}$ and $h_w \ll |h + H| \ll \sigma$, Eq. (9) into

$$\frac{f(h, t)}{f(h, 0)} \simeq 1 - \frac{\sin \pi p}{\pi} \int_0^1 dx x^{p-1} \frac{(1 - x)^p}{\alpha \eta^2 + (1 - x)^{2p}},$$

(12)

where $\alpha \simeq 0.8(\sigma/h_0)^2$. Both Eqs. (3) and (12) show that the field $h$ scales as $t^p$ in holes’ line shapes.

Finally, to obtain Eq. (3) from Eq. (12), note first that $f(0, t)/f(0, 0) \to 0$ as $t \to \infty$ in Eq. (12), since the integral therein equals $\pi/\sin \pi p$ if $\eta = 0$. Then, breaking up the integration interval into two pieces, (1) from 0 to $(\alpha \eta^2)^{1/2p}$, and (2) from $(\alpha \eta^2)^{1/2p}$ to 1, and expanding $x^p/(\alpha \eta^2 + x^{2p})$ in powers of $\epsilon$ and $1/\epsilon$ in the first and second integration intervals, respectively, where $\epsilon \equiv x^{2p}/\alpha \eta^2$, gives $|\eta|^{1/p-1}$ for the leading term, which is desired result, that is, Eq. (3).

B. Comparison with experiments and simulations

In this section we test our results, that is, the validity of Eqs. (3) and (12), against experiments and against our MC simulations.

We first apply Eqs. (3) and (12) to Fe$_8$. From Table I, $\sigma/h_0 = 0.66$ follows. Substitution of this number into Eq. (3) gives $p = 0.58$. Knowing the value of $p$ enables us to plot the data points for Fe$_8$ shown in Fig. 4a. Unfortunately, data for holes in Fe$_8$ have only been published for $t \leq 40$ s, that is, for $\Gamma t \lesssim 1.6$, a time which falls short of the validity range for Eqs. (3) and (12). Still, one can appreciate in Fig. 4a how the data points seem to approach the theory curve for $f(h, t)/f(h, 0)$ as $t$ increases up to $\Gamma t \lesssim 1.6$.

In order to see how this would go for longer times, we have used our model to simulate an experiment on Fe$_8$. The results are shown in Fig. 4b. We have let one MC sweep equal $\Gamma t = 1$, which, by the argument given above, implies $t \simeq 25$ s for Fe$_8$. The agreement with theory is remarkable. This is better appreciated in the log–log plot shown, with the same data, in Figs. 4a and b. On the other hand, rescaling these plots, using $p = 1/2$ gives rise to some data point scatter, but not sufficiently large to convincingly rule out $p = 1/2$. This is not too surprising, given the small difference between $p = 1/2$ and the value $p = 0.58$ that is given by Eq. (3). Still, one might have hoped that these data would have been sufficient.
FIG. 5: (a) Everything is as in Fig. 4a, except that a Log–log scale is used here. (b) Same as in (a) but for MC simulations, instead of experiments. Everything else is as in Fig. 4b. The dashed line stands for $\eta^{1/p-1}$, as predicted by Eq. (3) for $p = 0.58$, for Fe$_8$.

to discriminate between Eq. (3), where $\eta$ is raised to the $1/p - 1$ power and the Lorentzian curve of Ref. [15]. Again, data for smaller values of $\eta$ would be required for this. We know of no other experimental results for hole digging we can make use of. As far as we know, all other reported experiments for SMM systems start from strongly polarized initial states.

Consequently, we decided to do simulations of SMM’s in FCC lattices, because Eq. (5) gives then a value, 0.73, for $p$, which differs significantly from 1/2. We are now at liberty to choose the value of $h_w$. In order to be able to obtain holes’ line shapes down to rather small values of $\eta$, and still meet the validity criterion for Eqs. (3) and (12), we let $h_w$ take values down to 0.01.

We show how $m$ varies with $t$ in Fig. 6 in a simulated experiment in which all spins are up and down with probabilities 0.6 and 0.4 in the initial state, and evolve thereafter with no applied field. For $\Gamma t \lesssim 1$ (not shown), $\tilde{m} \propto \Gamma t$. Note that $\tilde{m} \propto (\Gamma t)^p$ up to $\Gamma t \sim (\sigma/h_w)^{1/p}$, which for $h_w = 0.01$, for instance, $\Gamma t \sim 9 \times 10^3$, as predicted. Note also the good agreement with the value, 0.73, which Eq. (3) gives for $p$. 
FIG. 6: (a) $f(h,t)/f(h,0)$ versus $\eta$, defined in Eq. (4), for the shown times. Times are in MC sweeps. $h_w = 0.01$. Symbols stand for averages over 1400 MC runs for 65536 spins in a FCC lattice. The full line is from Eq. (12). As in Fig. 4, initially, all 65536 spins are randomly up or down with probabilities 0.6 and 0.4, respectively. (b) Same as in (a) but in a Log–Log scale. The solid line stands for Eq. (12) and the dashed line is for the best fitting Lorentzian curve. The dash–dot line stands for $\eta^{1/p-1}$, as predicted by Eq. (3), for $p = 0.73$, given by Eq. (5) for FCC lattices.

Holes’ line shapes obtained from MC simulations are shown in Figs. 6a and b. The nice agreement with theory is reassuring. Similar plots but using $p = 0.5$ give unsatisfactorily wide data point scatter. The data clearly follow Eq. (3) for $\eta \ll 1$ and deviate sharply from a Lorentzian line shape. We do not exhibit results for SC or BCC lattices, but we have found them to follow our predictions equally well.

IV. CONCLUSIONS

Results that follow from our theory for the relaxation of the magnetization of interacting SMMs are reported. They are in fair agreement with the experimental relaxation of the
magnetization observed in Fe₈ as well as with our own MC results for Fe₈ and for other lattices. Furthermore, we make some predictions for Fe₈ that can be checked experimentally. Experiments following the lead of Ref. [6] would be made feasible by the application of a transverse field \( H_\perp \) of approximately 0.3 T, since \( \Gamma \), which increases as \( \Delta^2 \), would then increase by a factor of roughly 50 if \( H_\perp \) is applied along the easier magnetization direction on the \( xy \) plane (see Figs. 2 and 3 of Ref. [4]). This would in effect approximately reduce the time scale in Fig. 3 from hours to minutes. Comparison of experimental results with MC data shown in Fig. 3 would be interesting. It would, for instance, show whether heat exchange takes place readily, as inferred in Ref. [29] from nuclear magnetic resonance experiments, or not.

We have also shown, from theory and MC simulations, that the magnetization of spatially disordered SMMs relaxes, not as any power of time, but approximately as given by Eq. (2). A counterintuitive prediction that follows from our theory and from MC simulations can be gathered from Figs. 2a and 2b. One might have thought that dilution would lead to weaker dipole interactions and, consequently, to unhindered, faster relaxation. Instead, the opposite effect takes place for \( 0.1 \lesssim \tilde{n} \leq 1 \) after some time.

Line shapes that develop in crystals of Fe₈ clusters have been obtained from our theory. We have shown that \( f(h, t) \) is only a function of \( h/t^p \) for all \( |h| > h_w \), that is, for all \( h \) outside the tunnel window. This is the main content of Eq. (12). Furthermore, we have shown that data points from experiments on Fe₈, taken from Ref. [6], as well as results from MC simulations we have performed for the same system, follow this rule. Scaling also ensues for the data from our MC simulations of SMM’s on FCC lattices for \( p = 0.73 \), as given by Eq. (13), but not for the otherwise predicted \( p = 1/2 \) value that is supposed to hold universally.

We have also shown that \( f(h, t) \sim |h/t^p|^{\frac{1}{(1/p)-1}} \) if \( h_w/\sigma \ll \eta \ll 1 \) and \( h \) is outside the tunnel window. Again, this is in agreement with experimental and MC results for Fe₈ (see Figs. 5a and b), FCC (see Figs. 6a and b), and (not shown) SC and BCC lattices. A rough argument that explains why holes’ line shapes are not Lorentzian follows. Note first that while field distributions from dilute systems of dipoles are indeed Lorentzian, only spin flips that take place after time \( t \) contribute to the diffusion of a hole that was “dug” at time \( t \). Since a full hole is only dug gradually in the course of time, a sum of Lorentzian functions of \( h \) [see Eq. (12)] of various widths is expected. Not surprisingly, Lorentzian function does
not ensue for \( f(h, t) \) [see, Eq. (3)]. Here, experimental data for holes in the hundreds of seconds time range, over which the magnetization has already been observed experimentally, would be helpful.

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