Comment on “Hole digging in ensembles of tunneling molecular magnets”

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Tupitsyn et al. [Phys. Rev. B 69, 132406 (2004)] have recently reported results for the relaxation of crystalline systems of single-molecule magnets, such as Fe8, whose tunneling windows are much narrower than their dipolar field spread. They find that, independently of crystalline lattice structure, (i) the magnetization and hole widths of field distributions evolve with time $t$ as $\sqrt{t}$, and (ii) field-distribution line shapes are Lorentzian. We give a counter example to these conclusions, and show that the main assumption on which some of them rest is invalid.

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Tupitsyn, Stamp, and Prokof’ev (TSP) have recently reported results for the relaxation of crystalline systems of single-molecule magnets, such as Fe8, whose tunneling windows are much narrower than their dipolar field spread. They find that, independently of crystalline lattice structure, (i) the magnetization $m$ and hole widths of field distributions evolve with time $t$ as $\sqrt{t}$, and (ii) field-distribution line shapes are Lorentzian. The first conclusion is contrary to our prediction, of Ref. 2, that, in zero field, $m$ relaxes from weakly polarized states as $t^p$, where $p$ depends on crystal structure. We give below a counterexample to the conclusions TSP have reached, and show that the main assumption on which some of their conclusions rest is incorrect.

We mainly use the notation of Ref. 1, giving the bias field $\xi$ in terms of the tunnel window field $\xi_0$, but $\xi_0$ is given in terms of the nearest neighbor dipolar field $E_D$. We assume spins flip at rate $1/\tau_0$ if $|\xi|<1$, but not at all otherwise, and time $t$ is given in terms of $\tau_0$. The following numbers may be found useful: the rms value of the dipolar field $\langle \xi \rangle$ is 3.7$E_D$ and 8.3$E_D$ for simple cubic (SC) and face centered cubic (FCC), respectively.

Let $p_\uparrow(\xi,t)$ [$p_\downarrow(\xi,t)$] be the number density of up spins (down spins) with a field $\xi$ acting on them, and let $f(\xi,t)=p_\uparrow(\xi,t)-p_\downarrow(\xi,t)$. Note that $m(t)=-\int d\xi f(\xi,t)$. The main ingredient underlying Eq. (1) of Ref. 1 is the assumption that $f(\xi,t) \propto N(\xi)\exp[-t/\tau(\xi)]$, where $N(\xi)$ is of no interest to us here, and $\tau(\xi)$ is some time that depends only on $\xi$. The Monte Carlo (MC) results shown in Fig. 1 are contrary to the assumption of TSP, that $f(\xi,t)$ is exponential in $t$. [The probability density that a spin has field $\xi$ and has not yet flipped at time “$t$” behaves much as $f(\xi,t)$.]

From the assumption that $f(\xi,t) \propto \exp[-t/\tau(\xi)]$ and the further general statement TSP make, that $1/\tau(\xi)$ is a Lorentzian function of $\xi$, hole line widths that grow as $\sqrt{t}$ when $t \gg \tau_0$ follow in Ref. 1. Such $\sqrt{t}$ growth does take place in SC lattices, but not in general, as one can gather from the MC results exhibited in Figs. 2(a) and 2(b) for FCC lattices. Rather, from Figs. 2(c) and 2(d), one gathers that $\xi$ then scales as $t^p$, where $p=0.73$. Finally, in the region of interest, when $t \gg \tau_0$ but $t$ is still within the range where $m \approx \chi^p$, the line shape in Figs. 2(c) and 2(d) differ significantly from a Lorentzian function. For comparison, results from our own theory are also shown in Figs. 2(c) and 2(d).

For completeness sake, we examine further numerical evidence that supports our claim that $p=0.73$, not 1/2, for FCC lattices. To this end, note first that $\xi \sim t^p$ scaling implies, through the relation $m(t)=-\int d\xi f(\xi,t)$, that $m \sim t^p$. Thus, the value of $p$ can also be obtained from the time evolution of $m$.

The difference between the relaxation of the magnetization in SC and FCC lattices can be clearly appreciated in Fig. 3, as well as in Figs. 4(a) and 4(b). Note in Fig. 3 that, for FCC lattices, $m \propto t^p$ and $p=0.73$ for time spans that are increasingly larger for larger values of $1/\xi_0$. (This is as predicted in Ref. 2.) Note also that the $p=1/2$ slope that is claimed by TSP to hold universally for all lattices appears to ensue for FCC lattices only when the relaxation crosses over from the $m \propto \chi^0$ regime to saturation. This is in clear contrast with the data points for SC lattices in Fig. 3, for which $p \approx 0.5$. Linear $m/m_0$ vs time plots for SC and FCC lattices which further illustrate this point are also shown in Figs. 4(a) and 4(b). The small neighborhood of the inflection point

FIG. 1. (Color online) The field function $f(\xi,t)$ vs $t/\xi_0^2$ for SC lattices and the shown values of the dipole field $\xi$. The tunnel window’s half-width $\xi_0$ is 0.1. The initial value of the magnetization $m_0$ is 0.2 of saturation. All data points are for systems of 32768 spins, and follow from averages over 800 runs, over times much greater than $t \gg \tau_0$ but within the range where $m \propto \chi^p$. 

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cases, m = 65 536 spin system and spins. Data points for SC lattices are for 4096 spins. For all data shown values of H = 0.726. The dashed line is for the best fitting Lorentz curve. The full line is from our theory. (d) Same as in (c) but for ξ0 = 0.1. In all cases, m0 = 0.2. All data points are for systems of 65 536 spins, and follow from averages over 800 runs over times much greater than t ≳ τ0 but within the range where m ≳ m0. All the shown data points come from the 2ξ0 < ξ < 0.2δξ (nearly one and two decades for ξ0 = 0.1 and ξ0 = 0.01, respectively). In the shown range of time values, \(1 - m/m_0\) changes by approximately one and two decades (see Fig. 1), for ξ0 = 0.1 and ξ0 = 0.01, respectively.

FIG. 3. (Color online) \(1 - m/m_0\) vs time t. All data points follow from MC simulations. Triangles are for SC lattices. All other data points are for FCC lattices. All data points for the FCC lattice are for systems of 8192 spins, except for , which stand for 65 536 spins. Data points for SC lattices are for 4096 spins. For all data points, averages over 1200 runs were performed, except for the 65 536 spin system and t > 1200. For them, averages over 100 runs were performed.

FIG. 2. (Color online) (a) \(f(ξ, t)\) vs \(ξ/\rho\) for FCC lattices and the shown values of t, ξ0 = 0.01, and \(p = 0.5\). Lines are guides to the eye. (b) Same as in (a) but for ξ0 = 0.1. (c) Same as in (a) but for \(p = 0.726\). The dashed line is for the best fitting Lorentz curve. The full line is from our theory. (d) Same as in (c) but for ξ0 = 0.1. In all cases, m0 = 0.2. All data points are for systems of 65 536 spins, and follow from averages over 800 runs over times much greater than t ≳ τ0 but within the range where m ≳ m0. All the shown data points come from the 2ξ0 < ξ < 0.2δξ (nearly one and two decades for ξ0 = 0.1 and ξ0 = 0.01, respectively). In the shown range of time values, \(1 - m/m_0\) changes by approximately one and two decades (see Fig. 1), for ξ0 = 0.1 and ξ0 = 0.01, respectively.

\(m/m_0\) vs \(\sqrt{t}\). Data points are from averaging over at least 4000 MC runs for 4096 spins on SC lattices with ξ0 = 0.1. The straight line is a guide to the eye. (b) Same as in (a) but for 8192 spins on an FCC lattice. The straight line segment covers a neighborhood of the inflection point.

Fig. 4 (marked as a straight line segment) in Fig. 4(b) is, of course, nearly straight. This transient behavior might be misinterpreted as the onset of a \(m \propto \sqrt{t}\) regime if, as in Fig. 1 of Ref. 6, data points below \(m/m_0 = 0.5\) are not included in the plot.

Finally, comparison of the results shown in Figs. 2(a) and 2(b), Figs. 2(c) and 2(d), as well as among different curves shown in Fig. 3 for different values of ξ0 should allay any misgivings about spurious effects that might arise from the finite number of spins \(n_s\) in the tunnel window, since \(n_s \propto ξ_0\) if \(ξ_0 \ll δξ\). To the same end, data points for two FCC lattice sizes are shown in Fig. 3 for ξ0 = 0.01.

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\(^{3}\)This is because \(df(ξ, t)/dt = -f(h, t)/τ(ξ)\), since \(df(ξ, t)/dt\) depends, not only on f and ξ, but also on the rate of magnetization reversal which is going on within the tunneling window at time t, which in turn depends nontrivially on time.


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