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I Tupitsyn

P Stamp

Nikolai Prokof'ev

University of Massachusetts - Amherst, prokofev@physics.umass.edu

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Reply to the Comment on "Hole digging in ensembles of tunneling molecular magnets"

I. S. Tupitsyn^{1,2} P.C.E. Stamp² and N.V. Prokof'ev³

¹ *Russian Research Center "Kurchatov Institute",
Kurchatov Sq.1, Moscow 123182, Russia.*

² *Department of Physics and Astronomy,
and Pacific Institute for Theoretical Physics,
University of British Columbia, 6224 Agricultural Rd.,
Vancouver, B.C. V6T 1Z1, Canada*

³ *Department of Physics, University of Massachusetts,
Amherst, MA 01003, USA.*

Our work has argued for a particular scaling form governing the distribution $M(\xi, t)$ of magnetisation over bias ξ , for a system of dipolar-interacting molecular spins. This form, which was found in Monte Carlo (MC) simulations, leads inevitably to a short-time form $\sim t^{1/2}$ for the magnetisation relaxation in the system. The authors of the Comment argue that the magnetisation should decay rather as $\sim t^p$, with the exponent p depending on the lattice type- and they argue this form is valid up to infinite times. They also claim that our conclusion is based on an assumed exponential dependence of the function $M(\xi, t)$ on $\tau_{de}(\xi)$, the effective molecular relaxation time. In fact our results do not depend on any such dependence, which was used merely for illustrative purposes, but only on the scaling form we found. Repeating our MC simulations for different lattice types and different parameters, we always find a square root relaxation for short times. We find that the results of the comment are flawed because they try to fit their results over far too large a range of times (including the infinite time limit, where no simple theory applies).

The comment¹ misrepresents both our work and the physics of the problem. Our results are essentially that for both strongly polarised² and depolarised³ dipolar systems, the short-time relaxation (after an initial transient) has the form $\delta M \sim \sqrt{t}$, regardless of lattice type- with concomitant results for the hole width and the scaling function $M(\xi, t)$ introduced in³. It is argued in the comment¹ that (i) our results are based on the assumption of an exponential form for the function $M(\xi, t)$; and (ii) that the magnetisation relaxation in lattices of various symmetry may be described over huge time ranges, encompassing up to 3 orders of magnitude in magnetisation (from $M = 1$ to $M = 10^{-3}$) by a power law form $\delta M(t) \sim t^p$, with p dependent on lattice symmetry. We respond to these in turn:

(i) The effective relaxation rate $\tau_{de}^{-1}(\xi)$ was introduced in³, and extracted and studied using direct t -dependent MC simulations. It describes molecules in large bias fields ξ , and, we claimed, has a Lorentzian-tail dependence $\tau_{de}^{-1}(\xi) \sim \xi^{-2}$. As before we write the total normalised magnetisation of the system $M(t) = \int d\xi M(\xi, t)$, where $M(\xi, t)$ is the normalised distribution of magnetisation over the local bias fields ξ acting on each molecular spin; in terms of the probability $P_\sigma(\xi, t)$ for a spin to be in a bias field ξ at time t , one has $M(\xi, t) = [P_\uparrow(\xi, t) - P_\downarrow(\xi, t)]$. In commenting on our paper Alonso and Fernandez have introduced a function $f(\xi, t)$ which is nothing but $f(\xi, t) = -M(\xi, t)$. They argue that our conclusions are based on the assumption that our $M(\xi, t)$ depends on $\tau_{de}(\xi)$ via an exponential form, ie., that $M(\xi, t) \propto e^{-t/\tau_{de}(\xi)}$. In fact this is not correct- the crucial conclusion in our paper about $\tau_{de}(\xi)$ was that it scales as ξ^2 , so that we can write $M(\xi, t)$ as a function

of one scaled variable alone, satisfying the scaling law

$$M(\xi, t) \approx M(t/\xi^2) \equiv M(z), \quad (1)$$

where $z = t/\tau_{de} \sim t/\xi^2$. The actual form of $M(z)$ is not so crucial. In our paper we used an *exponential* decay form for $M(z)$ (this is just the standard τ -approximation result, and so the simplest form to use). This was used for *illustrative purposes only*, and none of our conclusions is based on the specific form of this function- contrary to the comment's claim! In fact it is very easy to see that the functional form of $M(z)$ is *irrelevant* for the final conclusion that the magnetization decay is $\sim t^{1/2}$, which simply and immediately follows from the use of the scaling form in $M(t) = \int d\xi M(\xi, t)$, ie., *from the scaling form alone*. This scaling form was found to be valid in our MC simulations for different lattice types, provided one assumes that $t > \tau_o$ (ie., assuming initial transients are over) and that $\delta M/M(t=0) \ll 1$ (ie., assuming that the long-time multispin correlations have not yet set in). We also assumed that $\xi_o \ll \xi \ll W_D$ (here W_D is the half-width of the dipole energy distribution, ξ_o is the nuclear spin bath parameter, and τ_o is the characteristic single-molecule relaxation time³- hereafter we measure time in units of τ_o and energies in units of ξ_o).

(ii) Alonso and Fernandez are basically arguing that the scaled variable z should be rather written as $t/\xi^{1/p}$ with the exponent p depending on the lattice type; correspondingly, $M(t) \sim t^p$ and for FCC lattices they found $p \simeq 0.73$. For SC lattices they found $p = 0.5$. In Fig. 1 we present MC solutions of kinetic equations for the magnetization decay in the FCC lattice with 60^3 spins (FCC, BCC and triclinic lattices were also analyzed in Ref.⁴).

After the usual initial transient behavior $\sim t$ (already discussed in Ref.² as well as³), we observe a long interval with the $\sim \sqrt{t}$ relaxation at least until the fractional change in $|M(t) - M(0)|/M(0) \sim 0.5$. At longer times relaxation deviates from the $\sim t^{1/2}$ behavior, but the duration of the $\sim t^{1/2}$ interval in a demagnetized sample is *longer* than in a polarized sample (compare figures in Refs.² and^{3,4}).

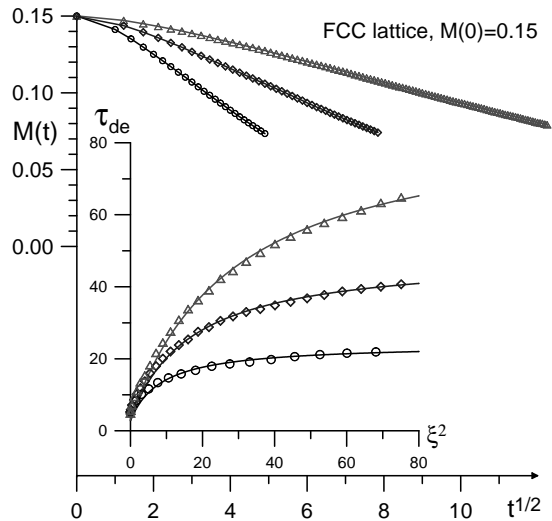


FIG. 1: $M(t)$ vs \sqrt{t} in the FCC lattice for initial magnetization $M(0) = 0.15$ and three values of E_D : $E_D = 2.5$ (circles), $E_D = 5$ (diamonds) and $E_D = 10$ (triangles). The inset shows the effective relaxation time $\tau_{de}(\xi)$ vs ξ^2 . Solid lines in the inset are the shifted Lorentzian curves described in the text with $\alpha(E_D) \approx 0.0415, 0.0205, 0.0114$ for $E_D = 2.5, 5, 10$. The time-step in the MC simulations was $\delta t/\tau_o = 10^{-2}$.

Simultaneously, we find that the functional form of the effective relaxation rate in the FCC lattice is well described by the shifted Lorentzian $\tau_{de}^{-1}(\xi) = (1/\pi)/(\xi^2 + 1) + \alpha(E_D)$ with small $\alpha(E_D) \sim 1/W_D$ (again, $\xi_o = 1$ in our units); at small values of initial polarization in the FCC lattice $W_D \approx 10E_D$, where E_D is the strength of the dipole-dipole interactions between nearest neighbor spins. We did not find any qualitative difference between the SC, triclinic, BCC and FCC cases.

It is important to notice that when the number of molecules in resonance is very small (as it is in the limit of very small values of ξ_o/W_D , particularly for FCC lattices), the usual *initial transient*² of $M(t)$ can be rather long, and its duration sensitive to the crystal structure because the dipole field spectrum is discrete in this limit. Then MC results on a small finite system are not relevant to experiments on macroscopic samples (where the number of molecules in resonance is also macroscopic).

In the comment, and in a previous paper⁶, the authors have argued that they can fit the whole time range, including both very short and even *infinitely* long times, using a single theory with a single power law exponent p (see, eg., Figs. 3,4 of the comment and Ref.⁶, paragraphs before Eqtn. 22 and after Eqtn. 24 of this paper). We find this implausible. Neither our MC simulations^{2,3,4,5} nor our analytic work² have ever claimed or attempted to explain more than the initial transient and the short-time relaxation that ensues after this transient, in the time interval before multi-spin correlations build up. It is in this restricted time range that we have argued for simple scaling and the associated square root relaxation.

¹ J.J. Alonso, J.F. Fernandez, preceding comment

² N.V. Prokof'ev and P.C.E. Stamp, Phys. Rev. Lett., **80**, 5794 (1998).

³ I.S. Tupitsyn, P.C.E. Stamp and N.V. Prokof'ev. Phys. Rev. B **69**, 132406, (2004).

⁴ I.S. Tupitsyn and P.C.E. Stamp, Phys. Rev. Lett. **92**, 119701 (2004).

⁵ N.V. Prokof'ev and P.C.E. Stamp, J Low Temp. Phys. **113**, 1147 (1998)

⁶ J.F. Fernandez and J.J. Alonso, Phys. Rev. B **69**, 024411 (2004). Analytical calculations of the exponent p were claimed in this paper, which assumed the form $\delta M \sim t^p$ to be valid both up to the limit $t \rightarrow \infty$, and also in the limit of vanishingly small values of ξ_o/W_D . No account was taken of either the discreteness of the dipole field spectrum in the small ξ_o limit, or of higher multi-spin correlations in the long time limit.