Comment on "Magnetization Process of Single Molecule Magnets at Low Temperatures"

In a recent letter Fernandez et al. [1] present Monte Carlo (MC) simulations of relaxing interacting dipoles for various lattice structures and argue that the short-time behavior of the magnetization $M(t)$ in an initially depolarized sample ($M(0) = 0$) is lattice structure dependent. They claim that in a simple cubic (SC) lattice at short times, $\Delta M(t) = |M(t) - M(0)|$ follows the usual $\sqrt{t}$ law, but that in body centered (BCC) and face centered (FCC) cubic lattices, $\Delta M(t) \sim t^{0.7}$.

We disagree with this conclusion, and we believe it arises from finite size effects in the MC simulations in [1]. The $\sqrt{t}$ short-time relaxation is expected when the dipolar interaction dominates over the hyperfine coupling, i.e., when the spread of dipolar field bias $W_D \gg \xi_o$, where $\xi_o$ is the range of energies over which the nuclear bias fluctuates [2] (in a completely demagnetized sample $W_D$ is several times larger than the strength $E_D$ of nearest-neighbour dipolar interactions). It should persist at least until $t \sim (W_D/\xi_o)\tau_o$, where $\tau_o$ is a microscopic single molecule tunneling relaxation time. In the very short time limit the relaxation is linear in $t$. Since these predictions all depend on the long-range part of the dipolar interaction, they should not depend on lattice structure.

To show this, we perform MC simulations for BCC, FCC and triclinic lattices with and without periodic boundary conditions (P.B.C.). At $t = 0$ all spins orientations were random and uncorrelated, with $M(0) = 0$ and $\langle S_i \rangle = \pm \xi$ ($|\xi| = 1$). The dipolar interaction $V_D(r_{ij}) = E_D[S_i S_j - 3\langle S_i \rangle\langle S_j \rangle]/r_{ij}^3$, and dipole fields and spin configurations were updated at time intervals $\delta t$ by flipping individual spins with probability $1 - \exp( - \delta t \tau^{-1}_N(\xi) \exp(-\xi/T))$. Here $\tau^{-1}_N(\xi)$ is an in-coherent relaxation rate $\tau^{-1}_N(\xi) = \tau_o^{-1} e^{\xi/\xi_o}$ (see [2]); we put $\tau_o = 1$ and assume contact with a bath at temperature $T$. The total bias energy $\xi = \xi_{dip} - g_0\mu_BSH_z$, including the internal dipolar contribution $\xi_{dip}$.

In the case of BCC and FCC clusters with P.B.C., with dimensions $16 \times 16 \times 16$ for $E_D = 10\xi_o$ we, as in [1], found $M(t) \sim t^{-0.7}$. However, we also found that in the FCC cluster (for example) the number of spins in resonance ($|\xi| \approx \xi_o$) is very small, much smaller than for the corresponding SC cluster. This means that in small FCC clusters, the MC procedure may not accumulate proper statistics.

To solve this problem one can (i) increase the cluster size, or (ii) increase the fraction of spins in resonance, by reducing $E_D/\xi_o$. For a BCC lattice (2 molecules per unit cell) we applied the first scenario. For a FCC lattice (4 molecules per unit cell) we applied both, taking $E_D = 2.5\xi_o$. Since in a completely demagnetized FCC sample $W_D \sim 10E_D$, then $W_D/\xi_o \sim 25$, i.e., still $>>1$.

The results (Fig. 1) are clear: for larger samples one finds $\sqrt{t}$ relaxation when $W_D/\xi_o \gg 1$, after the usual initial linear behaviour. Only the duration of the initial transient and the subsequent $\sqrt{t}$ are sensitive to the crystal structure.

![FIG. 1: $M(t)$ vs $\sqrt{t}$ for $M(0) = 0$. (a) BCC lattice of size $32 \times 32 \times 32$, with P.B.C.; Diamonds - the MC result; solid line - the $\sqrt{t}$ slope of the curve; (b) FCC lattice, of size $32 \times 32 \times 32$, and P.B.C. (solid line), and of size $48 \times 48 \times 48$, no P.B.C. (diamonds); (c) Triclinic lattice of $60^3$ Fe $- 8$ molecules, with no P.B.C. (Diamonds - the MC result; solid line - the $\sqrt{t}$ slope of the curve). Each of the 8 spin $5/2$ Fe$^{+3}$ ions was oriented along the easy axis, with lattice positions obtained from the Cambridge Data Base (easy and hard axis orientations were taken from [3]). All curves were obtained after averaging over a few hundred random initial configurations of spins.]

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[1] J.F. Fernandez and J.J. Alonso, Phys. Rev. Lett. 91, 047202 (2003).

[2] N.V. Prokochev and P.C.E. Stamp, Phys. Rev. Lett., 80, 5794 (1998).

[3] A.L. Barra, D. Gatteschi and R. Sessoli, Chem. Eur. J. 6, 1608 (2000).