Comment on ’Pulsed field studies of the magnetization reversal in molecular nanomagnets’

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Abstract

In a recent paper, \url{http://xxx.lanl.gov/abs/cond-mat/0404041} J. Vanacken et al. reported experimental studies of crystals of Mn\textsubscript{12}-ac molecular nanomagnets in pulsed magnetic fields with sweep rates up to 4000 T/s. Steps in the magnetization curve were observed. The data were explained by collective dipolar relaxation. We give here an alternative explanation that is based on thermal avalanches triggered by \textit{defect} molecules (faster relaxing species). These species are always present in Mn\textsubscript{12}-ac molecular nanomagnets. We propose a simple method to test this interpretation. Note also that we do not question the possibility of collective effects that are based on spin–spin interactions.

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In a recent paper, J. Vanacken et al. reported low temperature magnetization studies of Mn$_{12}$-ac single crystals at a field sweep rate up to 4000 T/s. Their main finding was that at such high sweep rates the position of the steps in the magnetic relaxation shifts by $\Delta H$ that increases as the sweep rate goes up. The authors scaled the relaxation curves obtained at different sweep rates onto one curve. The scaling was explained within a model of collective magnetic relaxation of the crystal.

Instead of commenting on the interpretations by J. Vanacken et al., we point out here important facts about Mn$_{12}$-ac that may have disrupted their experiments and propose an alternative explanation that is based on thermal avalanches triggered by defect molecules (faster relaxing species). These species are always present in Mn$_{12}$-ac molecular nanomagnets. We then propose a simple method to test this interpretation. Note also that we do not question the possibility of collective effects that are based on spin–spin interactions. Such effects can lead to ‘off resonance’ tunneling in the case of sufficient energy level mixing (activated levels, high fields, etc.).

Fig. 1 shows typical hysteresis loops for a single crystal of Mn$_{12}$-ac. The cryostat temperature was about 60 mK. When the applied field is near an avoided level crossing, the magnetization relaxes faster, yielding steps separated by plateaus. The loops show two series of steps: in the low field region (0 to 2 T) and in the high field region (2.5 to 5 T). It is now well known that the steps in the low field region are due to faster relaxing species whereas the others are due to the normal species of Mn$_{12}$-ac. The step heights decrease for faster field sweep rates. However, concerning the four fastest field sweep rates, a thermal avalanche is observed at about 4 T that reverses the entire magnetization in a time scale faster than a millisecond (the time resolution of the looking amplifier). The avalanche field decreases for faster sweep rates but the avalanche is always observed close to a resonance field.

When comparing the measurements in Fig. 1 with the pulsed field ones of J. Vanacken et al. (Fig. 2 in [1]), we note that their field sweep rates are more than 1000 times larger than our rates. Furthermore, our crystal size is much smaller than in [1]. Both points should enhance the thermal avalanche effect in the pulsed field experiment. We also note in the experiment by Vanacken et al. that the entire magnetization reverses in a time scale of about 0.2 ms and in the low field region (below 2 T) (see Fig. 2 in [1]). We propose therefore that this fast magnetization reversal could have been triggered by the heat emission of the
FIG. 1: (color) (a) Hysteresis loop measurements for a single crystal of Mn$_{12}$-ac at several field sweep rates. The field was applied at an angle of about 6° in respect to the easy axis of magnetization. (b) Enlargement for the low field region showing the relaxation of the fast species.
fast relaxing species reversing below 2 T \cite{21}. The field sweep rate dependence of the step position could reflect the time needed to trigger the thermal avalanche.

This interpretation can be checked with the following method. First, a high negative field should be applied to saturate the magnetization. Then, the field should be swept slowly up to 2 T reversing only the fast relaxing species. Finally, the field should be ramped to zero field and a positive field pulse applied. The magnetization reversal should then occur above 3 T because the fast relaxing species cannot trigger any more a thermal avalanche. Note that all relevant tunnel splittings of the normal species are too small to induce a significant reversal below 3 T and below 0.6 K. We do not expect therefore an thermal avalanche below about 3 T.

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et al., all molecules reverse in a time window of 0.2 ms, at about 1.5 T. This leads to an energy release of $2 \times g \mu_B/k_B S \times 1.5 \ T = 40 \ K$ per molecule. Knowing that there are about 5% of faster relaxing species, the energy released by it is quite important and might trigger the avalanche.