Quantum Hole Digging in Magnetic Molecular Clusters

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Abstract

Below 360 mK, Fe₈ magnetic molecular clusters are in the pure quantum relaxation regime. We showed recently that the predicted “square-root time” relaxation is obeyed, allowing us to develop a new method for watching the evolution of the distribution of molecular spin states in the sample. We measured the distribution \( P(H) \) of molecules which are in resonance at the applied field \( H \). Tunnelling initially causes rapid transitions of molecules, thereby “digging a hole” in \( P(H) \). For small initial magnetisation values, the hole width shows an intrinsic broadening which may be due to nuclear spins. We present here hole digging measurements in the thermal activated regime which may allow to study the effect of spin-phonon coupling.

Keywords: molecular clusters, quantum tunnelling of magnetisation, hyperfine coupling, spin-phonon coupling

Magnetic molecular clusters such as Mn₁₂ac and Fe₈ are ideal systems to study quantum tunnelling of the magnetisation (QTM) [1]. Crystals of these materials can be thought of as ensembles of identical, iso-oriented nanomagnets of net spin \( S = 10 \), and with a strong Ising-like anisotropy. Theoretical discussion of thermally-activated QTM assumes that thermal processes (principally phonons) promote the molecules up to high levels, not far below the top of the energy barrier, and the molecules then tunnel inelastically to the other side.

At temperatures below 360 mK, Fe₈ molecular clusters display a clear crossover from thermally activated relaxation to a temperature independent quantum regime [2]. In this regime only the two lowest levels of each molecule are occupied, and only “pure” quantum tunnelling through the anisotropy barrier can cause direct transitions between these two states.

Recently, we developed a method [3,4] for measuring the intrinsic line width broadening due to local fluctuating fields of the nuclear spins. It is based on the general idea that the short time relaxation rate is directly connected to the number of molecules which are in resonance at a given longitudinal applied field \( H \). In the low temperature regime, the Prokof'ev - Stamp theory [5] predicts that the magnetisation should relax at short times with a square-root time dependence, giving the rate function \( \Gamma_{\text{sqrt}}(H) \) which is proportional to the normalised distribution \( P(H) \) of molecules which are in resonance at the applied field \( H \).
Our measuring procedure is as follows. Starting from a well defined magnetisation state, we apply a magnetic field $H$ in order to measure the short-time relaxation behaviour, yielding the rate function $\Gamma_{\text{sqr}}(H)$ at the field $H$. Then, starting again from the same well defined magnetisation state, we measure $\Gamma_{\text{sqr}}(H)$ at another field $H$, yielding the field dependence of $\Gamma_{\text{sqr}}(H)$ which is proportional to the dipolar distribution $P(H)$.

This technique can be used for following the time evolution of molecular states in the sample during a tunnelling relaxation [4]. Starting from a well defined magnetisation state, and after applying a field $H_{\text{dig}}$, we let the sample relax for a time $t_{\text{dig}}$, called ‘digging field and digging time’, respectively. During the digging time, a small fraction of the molecular spins tunnel and reverse the direction of their magnetisation. Finally, we apply a field $H$ to measure the short time relaxation in order to get $\Gamma_{\text{sqr}}(H)$. The entire procedure is then repeated to probe the distribution at other fields $H$ yielding $\Gamma_{\text{sqr}}(H, H_{\text{dig}}, t_{\text{dig}})$ which is proportional to the number of spins which are still free for tunnelling.

We used this hole digging’ method, for studying Fe$_8$ and Mn$_{12}$ac molecular clusters [4] and found that tunnelling causes rapid transitions of molecules near $H_{\text{dig}}$, thereby "digging a hole" in $P(H, H_{\text{dig}}, t_{\text{dig}})$ around $H_{\text{dig}}$, and also pushing other molecules away from resonance. The hole widens and moves with time, in a way depending on sample shape; the width dramatically depends on thermal annealing of the magnetisation of the sample. For small initial magnetisation, the hole width shows an intrinsic broadening which may be due to nuclear spins [4]. The hole could be fitted to a Lorentzian function yielding the line width $\sigma$ which we studied as a function of temperature and digging time (fig. 1). We defined an intrinsic line width $\sigma_0$ by a linear extrapolation of the curves to $t_{\text{dig}} = 0$ (fig. 2). For temperatures between 0.04 and 0.4K, $\sigma_0 \approx 1.6 \text{ mT}$. For $T > 0.4 \text{ K}$, $\sigma_0$ increase rapidly.

The physical origin of the line width $\sigma_0$ at $T < 0.4 \text{ K}$ is assigned to the fluctuating hyperfine fields [5]. At higher temperature, spin-phonon coupling might be responsible for the observed line width.

References

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