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Nonexponential magnetization relaxation in a manganese single-molecule magnet

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Abstract

The magnetization relaxation in a tetranuclear manganese cluster Mn_4(pdmH) has been investigated with an extraction and a Faraday magnetometer at 0.14–0.75 K. The relaxation curves in the tunneling regime show the "square-root time" dependence for a short time after reversing the magnetization. This fact suggests the hyperfine and inter-cluster dipolar interactions play important roles for the relaxation.

Keywords: Single molecule magnet; Cluster; Quantum tunneling; Magnetization relaxation

Single molecule magnets (SMMs) that serve as a nano-scale magnet are attracting an increasing interest since they show a quantum mechanical behavior, such as a quantum tunneling and coherence, providing a possibility of the application to a quantum computer [1]. In this point of view it is important to understand the mechanism of an incoherent tunneling that comes from the interaction with their environments. Recent extensive study on the Mn_{12}-ac, Mn_4 or Fe_8 have revealed the importance of a hyperfine, a dipolar coupling and an intercluster exchange interaction as possible origins of the decoherence [2–4]. In this report, we present the relaxation curves for one of SMMs, a tetranuclear manganese cluster Mn_4(pdmH). The magnetization relaxation as a function of time gives information on the mechanism of the incoherent quantum tunneling.

The sample was prepared by the method previously described [5]. Mn_4(pdmH) consists of a planar Mn_4 rhombus that is a mixed valent, Mn_2^{2+}–Mn_2^{3+}. It was already confirmed by ac susceptibility and DC magnetization measurements that it behaved as a SMM with a high spin ground state $S = 8$ and anisotropic constant $D/k_B = 0.35$ K [5–7]. Magnetization measurements were initially carried out with a home-made extraction magnetometer on a ^{3}He refrigerator above 0.5 K and followed by a home-made Faraday force Magnetometer installed on a dilution refrigerator [8]. The sample, 21.7 mg, is aligned in eicosane under high magnetic fields. The relaxation curves were collected as follows; At first the sample was magnetized in one direction by applying the field, then it was reversed to the measuring field.

Fig. 1(a) gives the relaxation curves at zero field at various temperatures. The curves below 0.3 K are temperature independent, indicating in the pure quantum regime. The relaxation curve does not follow a simple exponential function. A tentative fit to a

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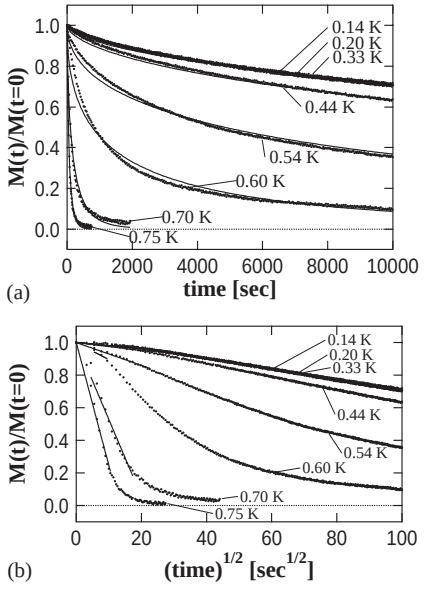

Fig. 1. (a) The magnetization relaxation curves at zero field for various temperatures. Solid lines are the fitted ones to the stretched exponential law; (b) The same as a function of $t^{1/2}$. The data is normalized by the value at $t = 0$ (starting time).

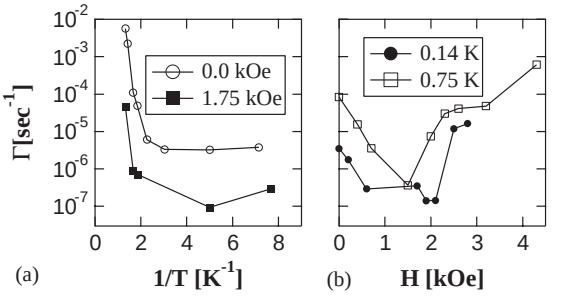

Fig. 2. The square-root relaxation rate (a) as a function of temperature; (b) as a function of the applied magnetic fields.

stretched exponential law, $M(t) = M_0(\exp[-(t/\tau)^{\beta}])$ gives a better agreement as shown in Fig. 1(a). The characteristic time τ follows Arrehenius law above 0.4 K and almost constant below it. The temperature dependent exponent β varies from 0.8 at 0.75 K to about 0.6 at the lower temperatures. A deviation from an exponential law suggests the effect of dipolar interaction as previously discussed [2]. For simplicity we assume the ellipsoidal shape of the crystals. At the beginning of measurements the aligned magnetic moment of the multicluster system produce an almost uniform dipolar field, whereas the reverse of some magnetic moments due to the quantum tunneling or thermal process, cause a time-dependent redistribution. Therefore the relaxation in the quantum tunneling, which can be tuned or de-tuned by the dipolar field, does not follow a simple exponential law.

In pure quantum regime a square-root law, $M(t) = M_0(1 - \sqrt{\Gamma t})$, was proposed for an array of SMMs with dipolar and hyperfine couplings [9]. As shown in Fig. 1(b) where the normalized magnetization is plotted as a function of square-root time, such a behavior can be observed for the data below 0.4 K for relatively long time region (20–100 $s^{1/2}$). At higher temperatures, the curve follows the square-root law in much shorter time region and the fit gives uncertainty on the Γ, depending on the fitting region. Fig. 2(a) gives the temperature dependence of the square-root relaxation rate Γ. The value is almost constant, $\Gamma_{qtm} = 4 \times 10^{-5}\ s^{-1}$, in the quantum regime. The $M(t)$ at 0.75 and 0.14 K were measured in the same way under various magnetic fields that are parallel to the easy axis. Each relaxation curve gives the square-root law for short times, although the region depends on both temperature and magnetic field. The Γ obtained from the fitting the data in 10–40 $s^{1/2}$ is given in Fig. 2(b). The enhancement at 0 and 2.5 kOe is an evidence of the resonant quantum tunneling, which is consistent with the hysteresis measurements [7]. As shown in Fig. 2(a), the temperature dependence of Γ at 1.75 kOe, where the energy levels are considered at "off-resonance", gives a small value of $\Gamma_{qtm} = 2 \times 10^{-7}\ s^{-1}$, indicating the contribution of the quantum tunneling still exist in such condition.

According to the theory the Γ_{qtm} is proportional to a square of tunneling splitting Δ in ground states and also depends on the hyperfine coupling [9]. The Γ_{qtm} observed here at zero magnetic field is much larger than the value reported for the Mn_{12}-ac, $6 \times 10^{-10}\ s^{-1}$ [2]. The large difference in the Γ_{qtm} might be attributed to difference of the tunneling splitting parameters rather than the hyperfine coupling.

In summary the behavior described above is quite similar to that observed for the Mn_{12}-ac, indicating that a common mechanism such as a dipolar and hyperfine coupling controls the quantum tunneling of both SMMs.

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