^{55}Mn NMR in Mn_{12} acetate: Quantum tunneling and magnetic relaxation of Mn_{12} cluster

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Abstract

^{55}Mn NMR in Mn^{4+} ions on Mn_{12} acetate oriented-powder crystal has been investigated in the external fields between T=1.4 K and 2.2 K to study the magnetic relaxation of Mn_{12} cluster associated with quantum tunneling. With increasing the longitudinal external field, the relaxation time decreased exhibiting significant dips at every 0.45 T, which is due to the effects of phonon-assisted quantum tunneling between the spin states at level crossings. The dependence of the relaxation time on the transverse field and the temperature dependence of the exponent in the stretched exponential function have been also studied.

Key words: Mn_{12} acetate; Quantum tunneling; Magnetic relaxation

1. Introduction

Recently, in quantum physics of mesoscopic system, nanoscale magnets Mn_{12}Ac has attracted much attention to quantum tunneling in magnetic relaxation at low temperatures [1]. Mn_{12}Ac is composed of Mn_{12} clusters (a large spin S=10), coupled each other through weak dipolar interactions, having the Ising anisotropy along the c axis. Magnetic relaxation in the external field is explained in terms of overcoming thermally the barrier intervening the two kinds of eigenstates $|\pm$m> and tunneling on level-crossing [1]. By means of observing a time change of ^{55}Mn spin-echo intensity of Mn^{4+} ions concerned with one side of frequency branches in frequency-field diagram, we have investigated relaxation process with quantum tunneling[2].

In the presence of an external field having components in both longitudinal H_z along the c-axis and transverse H_x, the spin Hamiltonian of a single Mn_{12} cluster can be described as

$$\mathcal{H}_S = -DS_z^2 - g\mu_B S_z H_z - g\mu_B S_x H_x \quad (1)$$

with the following constants [1], $D = 0.56 - 0.64$ K, and $g \approx 2$. After reversal of saturating H_z exerted on the oriented-powder Mn_{12}Ac crystal, we measured between T=1.4 K and 2.2 K the increase of spin-echo signal-amplitude h of ^{55}Mn NMR. The signal intensity reflects the fraction of Mn_{12} clusters relaxing their own magnetization along H_z. The recovery curve has been found to follow the stretched exponential formula:

$$h = h_0[1 - \exp\{-(t/\tau)^\beta\}] \quad (2)$$

from which we can obtain the relaxation time τ in case the exponent β is known.

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2. Experimental results and discussion

We obtained the dependence of the relaxation time on H_z at 1.88 K as shown in Fig. 1 in approximation of $\beta = 1/2$ in short-times[2]. The relaxation times indicated the thermally-assisted quantum tunneling at every resonance field $H_z = nH_1(H_1 \approx D/g\mu_B \approx$0.45T) of the level-crossing which is specified by the diagonal term of Eq.(1). The value H_1 is in good agreement with that obtained by magnetization measurements[3].

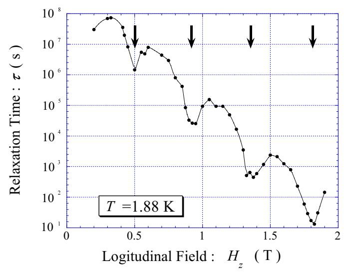

Fig. 1. The relaxation time τ vs the longitudinal field H_z for oriented-powder $Mn_{12}Ac$. The relaxation time is reduced from the Arrrhnius law at fields, shown by arrows, of resonant quantum tunneling.

In order to investigate quantitatively the effect of the transverse field H_x on the relaxation of clusters we measured at $T = 1.94$ K the relaxation time with applying H_x at constant H_z[4]. The result of relaxation times obtained by varying H_x between 0 and 0.6 T for $H_z = 1.25, 1.35$ and 1.50T has been shown in Fig. 2. For all curves in Fig. 2 we can see the gradual decrease of $\log\tau$ with increasing H_x. The trend of gradual decrease of the relaxation time τ is explained classically in terms of the barrier reduction due to an external field from the following equation:

$$\log\tau = \log[\tau_0 \exp\{(DS^2 - g\mu_B SH_z)/k_BT\}] - \{g\mu_B S \log e/k_BT\}H_x \quad . \qquad (3)$$

We showed, by the dotted line in Fig. 2, a typical example of $\log\tau = 4.45 - 2.92H_x$ calculated for $H_z = 1.25$ T from Eq. (3) using the following values: $S = 10, D = 0.64$ K, $g = 1.94$ and $\tau_0 = 8 \times 10^{-7}$ s.

Besides this classical effect, it is expected at the resonant position that the relaxation rate is more increased through tunneling by putting H_x than that at the off-resonant one, which can be clearly seen in Fig. 2 for τ measured at H_z=1.35T.

Next, we measured the temperature dependence of the exponent β of Eq. (2) at $H_z = 0.90$ and1.35 T without H_x, the result of which is shown in Fig. 3, where the result for β, obtained by Thomas et al. for $Mn_{12}Ac$ in H_z=0, [5], is depicted for reference.

In our case, the exponent β is more strongly temperature-dependent between 1.5 - 2.0 K than that

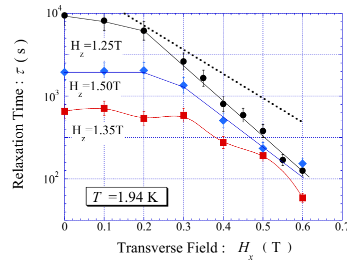

Fig. 2. The relaxation time τ vs the transverse field H_z for oriented-powder $Mn_{12}Ac$ under constant H_x. The dotted line shows the result calculated classically for $H_z = 1.25$ T, see the text.

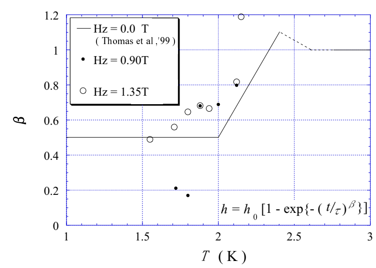

Fig. 3. The temperature dependence of the exponent β in the recovery curve, $h = h_0[1 - \exp\{-(t/\tau)\}^\beta]$, observed for oriented-powder $Mn_{12}Ac$.

of zero-field relaxation, which reflects the fact that the relaxation of clusters with quantum tunneling presently observed is thermally assisted. The further measurements are in progress.

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