Unusual magnetization reversal of Mn_{12} cluster in ultra-fast sweeping fields

Tomohiro Taniguchi [a], Hiroyuki Nojiri [a,1], Keiji Takeda [b], Kunio Awaga [c]

[a] *Department of Physics, Okayama University, Tsushimanaka 3-1-1, Okayama 700-8530, Japan*
[b] *Graduate School of Science, Hokkaido University, Sapporo 060-0810, Japan*
[c] *Graduate School of Science, Nagoya University, Nagoya 464-8602, Japan*

Abstract

Magnetization reversal of Mn_{12} cluster has been studied in ultra-fast sweeping magnetic fields. Below 3 K, a jump of magnetization is found in magnetization process in pulsed fields. A temperature dependence of magnetization saturates below the blocking temperature of 1.3 K. Time and critical field of the reversal show strong sweep velocity dependences.

Key words: quantum tunneling; high magnetic field; molecular magnet; magnetization

1. Introduction

A problem of magnetization reversal in a single-molecular magnet has attracted a great deal of attention in this decade. In a single molecular magnet such as Mn_{12} cluster, a reversal of magnetization occurs only at crossing points of the discrete energy levels. A concept of "quantum tunneling" of magnetization should be adopted for this case.[1] A most simplified model for this phenomenon is so called "Landau-Zener-Stückelberg-mechanism"(LZS-mechanism) and many theoretical and experimental investigations have been made on this model.[2] One of the most important characteristics of the LZS-mechanism is a non-adiabatic transition. For example, the tunneling probability depends on the sweeping speed of a magnetic field at crossing point and thus the change of magnetization also depends on the sweep speed. An investigation of magnetization process under a fast sweeping magnetic field is thus very important for the study of a non-adiabatic transition and a quantum tunneling phenomenon.

2. Results and discussion

Single crystals of $[Mn_{12}O_{12}(O_2CC_6H_5)_{16}(H_2O)_4]$ $\cdot 2C_6H_5CO_2H$ molecule cluster have been used. This cluster contains two inequivalent Mn_{12}-molecules sites. The directions of the easy axes are nearly orthogonal each other. The present measurements are made for $H \| b$. In this direction, the angle between the molecular axis and the external field is identical for two sites. The angle is about 40 degree and thus a strong transverse field is applied to the molecules.To avoid any extrinsic phenomena due to the sample misalignment, single piece of crystal is used for each measurement. Fast sweeping magnetic fields have been generated by using a pulsed field generator consists of a capacitor bank and a inductive coil. To change the sweep velocity in wide range, several different combinations of capacitance and inductance have been employed. Since the field pulse is sinusoidal in this case, the sweep velocity increases with increasing field maximum.

Figure 1 shows the magnetization curves measured at different maximum fields and at different sweep velocities. Since the blocking temperature is 1.3 K, the temperature dependence of the magnetization curve is negligibly small below 1.5 K. A magnetization jump

[1] Corresponding author. E-mail: nojiri@science.okayama-u.ac.jp

occurs only when the maximum field exceeds a threshold field$H_{\rm th}$.

In a conventional tunneling of magnetization, the reversal occurs only at the level crossing points and this typical behavior is not found in the present case. It indicates that the jump is not a tunneling of magnetization. A similar phenomenon was found in Mn_{12}ac and was named as "avalanche effect".[1] It is interpreted that a fractional reversal of magnetization at a crossing point emits heat and thus triggers the avalanche reversal of magnetization. Because of the thermal process, a large distribution of the critical field $H_{\rm c}$ was found, i.e. the critical field changes sweep by sweep. The reversal time was found to be about 1 msec and it is consistent with the thermal mechanism. In the present case, it is noticed that $H_{\rm th}$ is close to the level crossing point between $S_{\rm z}$=-10 and $S_{\rm z}$=+9 levels. This coincidence shows that the present reversal is also triggered by the tunneling. The distribution of $H_{\rm c}$ is not found, however, in the present case. More over, the time of the magnetization reversal is much faster than that of the previous case.

To investigate a possible mechanism of the magnetization reversal, the time of reversal is examined at different sweep speed. It is found that the reversal time decreases with increasing sweep speed. In the thermal avalanche effect, it is expected that the reversal time depends on the initial heat emitted by the tunneling of magnetization. Since the tunneling probability decreases for the fast field sweep, the longer reversal time should be needed in fast sweep. It is opposite to the experimental finding. For these discrepancies, the "thermal avalanche" is not adoptable for the present case. We propose an alternative mechanism as follows.

In the case of Mn_{12}ac, all the spins are parallel and thus the dipolar field by the nearest neighbor molecules is parallel to the spins. It is considered that the longitudinal static dipolar field does not contribute to the tunneling. In the present case, the situation is different. Because of the nearly orthogonal arrangement of nearest neighbor molecules, a transverse dipolar field presents. When a tunneling occurs, this transverse field changes and may induce the reversal at nearest neighbor sites. It should be also noticed that a strong external transverse field is applied in the present case. A pure LZS-mechanism cannot be adoptable for such case. A reversal may occurs in wide range of fields apart from the crossing points because of the strong mixing due to the transvers field. A rather fast reversal of magnetization such as 10 μs may be interpreted by this "dipolar field avalanche". A theoretical investigation of such mechanism as well as the treatment of non-LZS transition in strong transverse field are challenging subjects for future theoretical investigations.

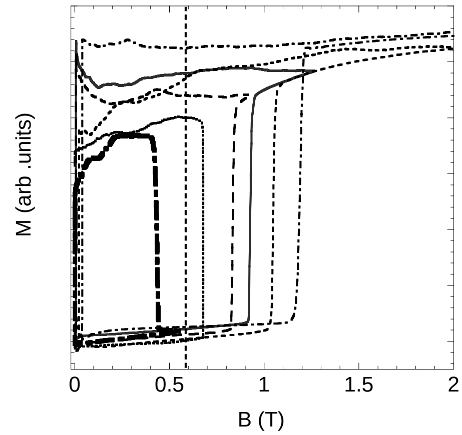

Fig. 1. Sweep field dependence of magnetization process for $H \| $b at 1.5 K. The vertical dashed line shows the $H_{\rm th}$.

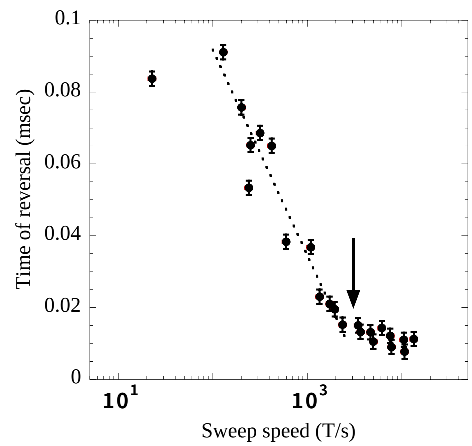

Fig. 2. Sweep speed dependence of the time of magnetization reversal for $H \| $b at 1.5 K. Sweep speed dependence saturates around the point indicated by an arrow.

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