Observation of avalanche effect of the spin reversal in Mn_{12} Ac by ^{55}Mn NMR

Takao Goto [a,1], Takeshi Koshiba [a], Akira Oyamada [a], Takeji Kubo [b], Yoshihito Suzuki [c], Kunio Awaga [c], Bernard Barbara [d], Jean-Paul Boucher [e]

[a] *Graduate School of Human and Environmental studies, Kyoto University, Kyoto 606-8501, Japan*

[b] *Department of Physics, Faculty of Education, Nara University of Education, Nara, 630-8528, Japan*

[c] *Department of chemistry, Graduate School of Science, Nagoya University*

[d] *Laboratoire de Magn'etisme Louis N'eel, CNRS, BP166, 38042 Grenoble, Cedex9, France*

[e] *Laboratoire de Spectrometrie Physique Universite J.Fourier-Grenoble I, 38402 France*

Abstract

By using the pulsed ^{55}Mn NMR in Mn_{12}Ac, the avalanche effcet for the magnetization recovery was observed in assembly of small pieces of single crystals between 0.1K and 1.6K in the field range from 0.7T to 1.5T. These results are consistent with the result of the previous measurement of hysteresis curve of the magnetization by SQUID.

Key words: Mn_{12}Ac; ^{55}Mn NMR, avalanche effect

1. Introduction

Recently, there has been a great interest in nano-scale molecular cluster magnets Mn_{12}Ac and Fe_8 on viewpoint of the quantum natures such as macroscopic quantum tunneling. In order to study the magnetic properties of Mn_{12}Ac from the microscopic standpoint, we have been studying ^{55}Mn NMR in Mn_{12}Ac. One of important information obtained from the ^{55}Mn NMR is concerned with the time-dependent magnetization recovery through the change in the signal intensity of ^{55}Mn NMR associated with the manganese ions belonging to the relevant cluster[1].

In the present work, we have found, by monitoring ^{55}Mn NMR in Mn^{+4} ion, an experimental evidence for the avalanche effect in the recovery of the magnetization at very low temperatures. The magnetic avalanche was observed first by Uehara et al. in various system[2],[3], and in particular in Mn12 by Paulsen et al. in SQUID measurement for the recovery of cluster magnetization [4]. Our result is consistent with the result given in ref.[4].

2. Experimental results and Discussions

The present experimental procedure is as follows. Some small pieces of the single crystal were attached on the stycast plate mounted in the sample holder with the *c*-axis along the same direction. The holder has the thermal contact, via the bundle of the meshed copper, to the mixing chamber in the dilution refrigerator. The sample was set with the *c*-axis parallel to the external field. The temperature was monitored in the thermometer in the mixing chamber. In zero field, the ^{55}Mn NMR for Mn^{+4} ion appears at the resonance frequency $\omega_N/2\pi = 230$MHz, which corresponds to internal field of H_{int} =21.3T. In the presence of an external field H_0 applied along the *c*-axis, the resonance line splits into two branches with the frequencies of $\omega_N = \gamma_N$ $(H_{int} \pm H_0)$, corresponding to the cluster whose magnetization is parallel or anti-parallel to H_0. These are

[1] E-mail:goto@nmr.jinkan.kyoto-u.ac.jp

referred to as upper (+) or lower (-) branches, respectively.

In the measurement, the resonance frequency was set at 237.4 MHz, which corresponds to the external field $H_0 \pm 0.7$ T for the (+) and (-) branches, respectively. The pulse sequence for the observation of the NMR signal was repeated with time duration longer than about 5 second to avoid increase in the temperature of the sample caused by rf-pulses. First we cooled the sample from 4K down to 1.5K with H_0 of about 4T, which is enough large for the saturation of the magnetization. We confirmed that the NMR signal corresponding to the (+) branch appears around +0.7T with the half-line width of about 0.4T. Then we decreased the temperature down to 0.15K. Next, after passing zero-field, H_0 is increased in the opposite direction. Unless any fraction of the reversed clusters exist, no NMR signal is expected to appear around H_0=- 0.7T.

We found that the almost full amplitude of the NMR signal for the (-) branch emerges abruptly at a certain value of H_0 within the NMR line-width. This means that the recovery of the full magnetization occurs instantaneously. This is an evidence for the avalanche. Such a sudden appearance of the NMR signal was obsedved in each run of the above in either direction of the field and at higher temperatures up to around 1.6K. Figure 1 shows the typical example of the recorder chart, which exhibits the abrupt recovery of the signal intensity from the zero level. No effect was observed by changing the sweeping speed. In Fig.2 are plotted the values of the external field at which the avalanche effect occurred as a function of the temperatures

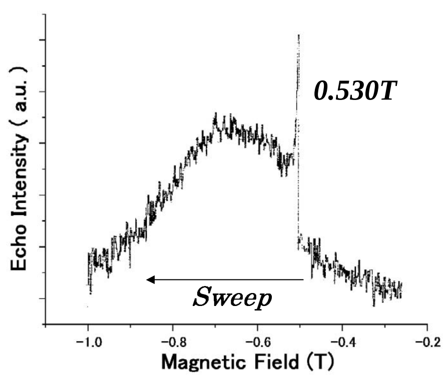

Fig. 1. Example of the recoder chart for obserbing the avalanche at 0.55K.

The avalanche effect may happen through the local increase of the temperature in the sample. In the present case the sample has thermal contact with the mixing chamber via the copper mesh wire, instead of imersing directly in the mixture. In vew of such an experimental condition, our present results may be in-

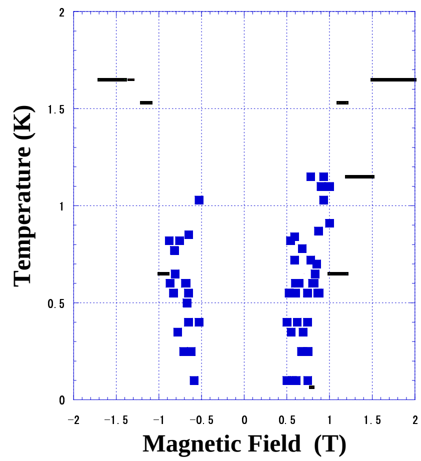

Fig. 2. The relation between the temperature and the field for the occurence of the avalanche

terpreted as follows. When the sweeping field reaches the crossing values some cluster spins flip by tunneling, giving some heat, but the signal should be negligible because there are little molecules in coincidence. But this heat adds, and cannot be evacuated immediately to the cryostat bath. The temperature becomes high enough to cause the jump of a large number of clusters which occurs for duration, for example, within 1ms. If, after the jump finishes, the system cools down, say, within 100 ms, we can observe the NMR signal corresponding to full reversed magnetization. This effect was observed even at 1.6K in the case of sample holder for the dilution refrigerator. However, when we put the oriented powder sample directly into liq.He, such an effect did not occur at the lowest temperature around 1.4K, because of bettter thermal coupling with the cryostat bath. It might be probable that the fluctuation of manganese nuclear spin associated with the ^{55}Mn NMR plays some important role in addition to the thermal effect. Further measurement will be necessary to clarify the mechanism of the avalanches in Mn_{12}.

References

[1] T. Kubo, T.Goto, T.Koshiba, K.Takeda, and K.Awaga, Phys. Rev. B**65** (2002) 22425

[2] M.Uehara, B.Barbara, B.Dieny, P.Stamp, Phys.Lett. **114A**(1986), 1, 23,

[3] B.Barbara,et.al., Proc.4th Int.Symp.Foundations of Quantum mechanics, Tokyo,JJAP Series 9 (0993) 237

[4] C.Paulsen, J.-G.Park, B.Barbara, R.Sssolo, and A.Chaneschi, J.Mag .and Mag.Mater. 140-144(1995)1819