^{133}Cs nuclear spin-lattice relaxation on field induced two-step phase transition in singlet-ground-state antiferromagnet $CsFeBr_3$

Tsuyoshi Nakamura [a,1], Takayuki Ishida [a], Yutaka Fujii [a], Hikomitsu Kikuchi [a], Meiro Chiba [a], Takeji Kubo [b], Yoshiyuki Yamamoto [c], Hidenobu Hori [c]

[a] *Department of Applied Physics, Fukui University, Fukui 910-8507, Japan*
[b] *Physics Department, Faculty of Education, Nara University of Education, Nara 630-8528, Japan*
[c] *School of Materials Science, JAIST, Ishikawa 923-1292, Japan*

Abstract

In order to study the spin dynamics associated with the field induced two-step phase transition, we have performed an NMR experiment on ^{133}Cs in $CsFeBr_3$ under magnetic fields applied along the *c*-axis. The phase transition temperatures T_{N1} and T_{N2} were determined from the anomalies in the temperature dependence of the nuclear spin-lattice relaxation rate T_1^{-1}. The transition points T_{N1} and T_{N2} thus obtained are consistent with those determined by the specific heat measurement.

Key words: $CsFeBr_3$; singlet-ground-state; phase transition; NMR; nuclear spin-lattice relaxation

1. Introduction

A hexagonal magnetic substance $CsFeBr_3$ is in the family of ABX_3–type crystal which is well known as the frustrating spin system due to triangular lattice antiferromagnet [1] [2]. Usually, the Fe^{2+} spin in $CsFeBr_3$ is treated as the fictitious spin $S = 1$. The spin state is composed of the singlet ground and doublet excited states due to a large easy plane type single ion anisotropy D along the *c*–axis. At zero magnetic field, this material does not have any long–range order down to 0 K. On the other hand, under the magnetic field applied along the *c*–axis, one of the levels in the doublet excited state comes down to cross the ground state. Around the level cross field, a two–step successive phase transition has been reported through the specific heat measurement [3]. In general, the Ising–type frustrated spin system has been known to exhibit the successive phase transition, while that of the easy plane type is expected to bring only a single step phase transition. Indeed, in the case of the isomorphic crystal $CsFeCl_3$ we have observed a single step transition [4]. It is peculiar that the two-step phase transition has been observed in $CsFeBr_3$. In order to study the spin dynamics associated with these phase transitions, we have performed an NMR experiment in $CsFeBr_3$. The nuclear spin–lattice relaxation time T_1 of the ^{133}Cs has been measured under the magnetic field up to 15 T applied along the *c*–axis in the temperature range T from 1.5 to 4.2 K. A preliminary result has been already reported [5].

2. Experimental results of ^{133}Cs nuclear spin–lattice relaxation rate

A typical result of the spin lattice relaxation rate T_1^{-1} is shown in Fig 1. The phase transition temperatures T_{N1} and T_{N2} are determined from the anomalies in the temperature dependence of the relaxation rate. In the paramagnetic phase ($T > T_{N1}$) the relaxation rate was almost independent of the temperature. The

[1] E-mail: nakamura@curie.apphy.fukui-u.ac.jp

slope of the temperature dependence of the relaxation rate became steeper for $T_{N2} < T < T_{N1}$. At T_{N2}, a small kink appeared in the slope of the relaxation rate. The transition temperature T_{N1} and T_{N2} thus determined are consistent with those determined by the experiment of the specific heat and the magnetization.

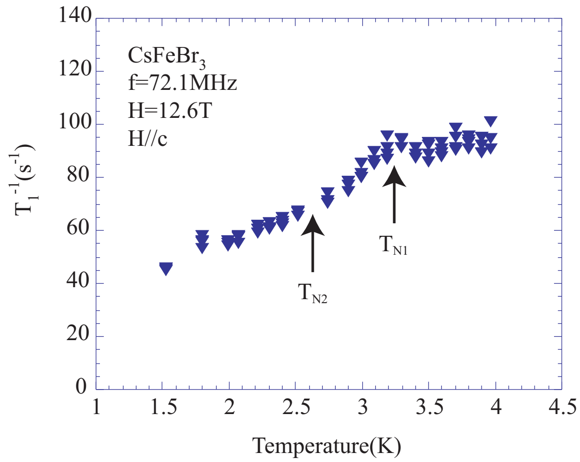

Fig. 1. Typical temperature dependence of nuclear spin-lattice relaxation rate T_1^{-1} of ^{133}Cs in $CsFeBr_3$. Arrows in the figure show anomalies in the relaxation rate from which the phase transition temperatures T_{N1} and T_{N2} are determined.

In the usual magnetic materials, the spin-lattice relaxation rate T_1^{-1} diverges near the phase transition temperature owing to the critical slowing down in the spin fluctuation. In the present case, however, T_1^{-1} did not diverge even at the transition temperatures, T_{N1} and T_{N2}. The behavior of T_1^{-1} in $CsFeBr_3$ is strange, by considering the fact that in the isomorphous crystal $CsFeCl_3$ the diverging behavior in T_1^{-1} has been reported at the temperature of the field induced phase transition [6]. The behavior of the nuclear relaxation rate shows that the spin fluctuation is large even in the order phase.

3. Results and discussion

The phase transition temperatures T_{N1} and T_{N2} in $CsFeBr_3$ are determined from the anomaly in the temperature dependence of the nuclear spin lattice relaxation rate of ^{133}Cs. We have already reported a preliminary experimental result under the magnetic field up to 15 T [5]. Unfortunately, the transition temperatures at the magnetic fields exceeding 10 T were suspicious because the experimental crystal orientation was ambiguous. This time the experiments were carried out with careful crystal orientation. The transition points for the field larger than 10 T did not reproduce those already reported. Thus we believe that the newly obtained T_{N1} and T_{N2} should be correct.

The temperature-field phase diagram is summarized in Fig 2. The transition temperatures T_{N1}, determined by the experiments of NMR, the specific heat and the magnetization, are all consistent each other. As for the second transition temperature T_{N2}, the results from the experiments of NMR and the specific heat are consistent, but the second transition is not observed in the magnetization experiment. The mechanism of the two-step phase transition is still an open question.

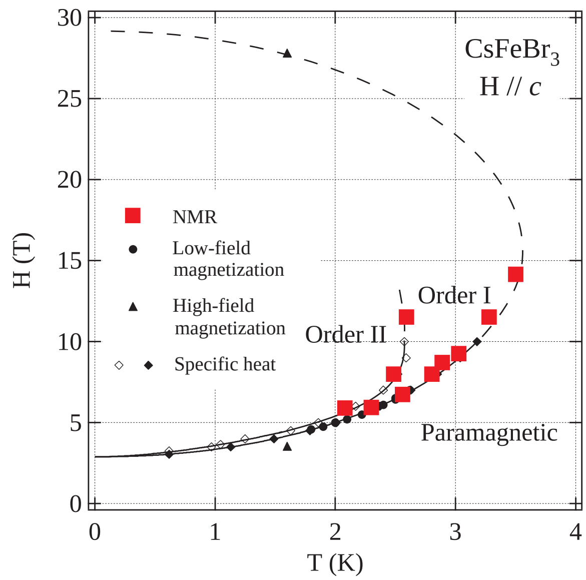

Fig. 2. Temperature-field phase diagram of $CsFeBr_3$ under $H//c$-axis. Solid quadrangles are the transition point determined by T_1^{-1}. Other symbols are the phase boundaries determined by the specific heat [3] and the magnetization [3] measurements. Broken lines are the guides to the eye.

References

[1] Y. Takeda, M. Shimada, F. Kanamaru and M. Koizumi, J. Phys. Soc. Jpn. **37** (1974) 276.

[2] For a recent review, see M. F. Collins and O. A. Petrenko, Can. J. Phys. **75** (1997) 605.

[3] Y. Tanaka, T. Tanaka, T. Ono, A. Oosawa, K. Morishita, K. Iio, T. Kato and H. A. Katori, J. Phys. Soc. Jpn. **70** (2001) 3068.

[4] M. Chiba, S. Ueda, T. Yanagimoto, M. Toda and T. Goto, Physica B **284–288** (2000) 1529–1530.

[5] T. Ishida, T. Nakamura, Y. Fujii, H. Kikuchi, M. Chiba, T. Kubo, Y. Yamamoto and H. Hori, Prog. Theor. Phys. in press.

[6] M. Toda, T. Goto, M. Chiba, K. Adachi and N. Suzuki, J. Phys. Soc. Jpn. **68** (1999) 2210.