ESR Study on Metamagnetic Transition in $CsFeCl_3$ up to 40 T

Meiro Chiba [a,1], Tomoyuki Higuchi [b], Kiyofumi Kitai [b], Seitaro Mitsudo [b], Toshitaka Idehara [b], Mitsuru Toda [c], Shizumasa Ueda [d]

[a] *Department of Applied Physics, Fukui University, Fukui 910-8507, Japan*
[b] *Research Center for Development of Far Infrared Region, Fukui University, Fukui 910-8507, Japan*
[c] *Institute of Materials Structure Science, KEK, Tsukuba 305-0801, Japan*
[d] *Institute of Advanced Energy, Kyoto University, Uji, Kyoto 611-0011, Japan*

Abstract

In the $S = 1$ fictitious spin system $CsFeCl_3$ an anomalous magnetization has been observed under the magnetic field around 33 T, being much higher than the saturation field. It can not be explained in terms of the fictitious spin $S = 1$. In order to clarify the nature of the metamagnetic transition, an ESR experiment was performed under magnetic fields up to 40 T. The mechanism of the metamagnetic transition is discussed under the basis of the ESR frequency-field diagram.

Key words: ESR, high field, fictitious spin, metamagnetic transition

1. Introduction

The magnetic compound $CsFeCl_3$ is an interesting material as one of the family of ABX_3 type hexagonal crystal relating to the magnetic frustration caused by the triangular-lattice antiferromagnetism. Usually the Fe^{2+} spin system in this material is treated within the framework of the fictitious spin $S = 1$ [1]. At zero magnetic field the spin states are composed of the singlet ground- and doublet excited state separated by D due to the crystallographic anisotropy. It is also interesting that the material does not have any long range order at zero magnetic field. On the other hand under a magnetic field $\boldsymbol{B}$ applied parallel to the crystal c-axis, ($\boldsymbol{B}$ // c-axis), one of the states of the doublet excited state comes down to cross the ground state at 7.5 T. It is reported that the long range magnetic order occurs around the level-cross field below 2.5 K [2,3].

A high field magnetization has been observed at 1.3 K with $\boldsymbol{B}$ // c-axis by one of the authors [4]. The magnetization seems to saturate at 11 T, which is consistent with the framework of fictitious spin $S = 1$. However, an anomalous magnetization has been observed under higher magnetic field B_c around 33 T [4]. The presence of the anomalous magnetization suggests the appearance of a new type of magnetic structure, which can not be explained within the framework of the fictitious spin $S = 1$.

In order to clarify the nature of the above-mentioned anomalous magnetization, a submillimeter ESR experiment was performed. A preliminary result has already been reported [5].

2. High field magnetization

Here we briefly survey the high field magnetization observed at 1.3 K with $\boldsymbol{B}$ // c-axis. As is shown in Fig. 1, under the magnetic field from zero to 4 T the magnetization is weak with a slight linear increasing due to the Van Vleck paramagnetism. The magnetization increases rapidly with increasing magnetic field from 4 to 11 T. The linear increase of the magnetization suggests the appearance of the magnetic order around the

[1] Corresponding author. E-mail: chiba@quantum.apphy.fukui-u.ac.jp

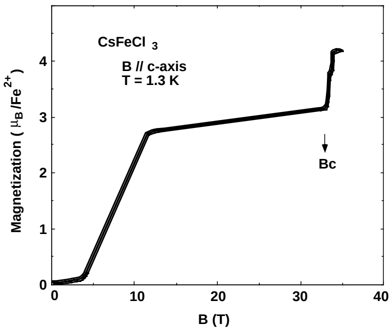

Fig. 1. High field magnetization in $CsFeCl_3$. B // c-axis. After ref.[4].

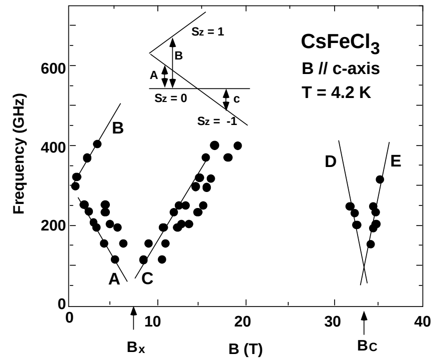

Fig. 2. Frequency-field diagram of high field ESR in $CsFeCl_3$. B // c-axis.

field of the ground state crossover, which is consistent with the theory by Tsuneto and Murao [6]. The magnetization seems to saturate at 11 T. Afterwards, up to about 32 T the magnetization increases gradually, which reflects the Van Vleck paramagnetism.

Thus below 32 T the framework of the fictitious spin $S = 1$ has been found to work well for the explanation of the magnetization. However, the metamagnetic transition around $B_c = 33$ T can not be explained in the framework of the fictitious spin $S = 1$. As is indicated in Fig. 3 in ref.[4] the calculated energy level scheme shows no other ground state crossover under the available magnetic field up to 40 T.

3. ESR experiments and discussion

An ESR experiment was performed at 4.2 K under pulsed magnetic fields up to 40 T with operating frequencies in the submillimeter region. The applied field was parallel to the c-axis of a single crystal $CsFeCl_3$.

The ESR absorption spectrum has several branches as is denoted by A, B, C, D and E in Fig. 2. The branches A and B are consistent with the experiment carried out by another group [7]. Other branches C, D and E are the spectrum observed for the first time.

Now we consider the branches A, B and C in the framework of fictitious spin $S = 1$. The branches A and B correspond to the transitions from the state $S_z = 0$ to $S_z = -1$, and from the state $S_z = 0$ to $S_z = 1$, respectively. Under the magnetic field exceeding the first level cross field $B_x = 7.5$ T appears the branch C which corresponds to the transition from the state $S_z = 1$ to $S_z = 0$. The slope of the straight lines in Fig. 2 corresponds to $g = 2.6$, which is consistent with that determined from the saturation magnetization [4]. The origin of the groups of the resonance lines, which belongs to neither A nor C, are not interpreted so far. One of the possible mechanisms is the contribution from some collective spin mode.

According to the results of high field magnetization [4] the value of the anomalous magnetization is a little larger than 4 μ_B/Fe^{2+}. By assuming that g is about 2, the possible ground state at the field above 33 T is $S_z = -2$ in $J = 2$ spin multiplet. However, it is not well understood so far why the excited state of $S_z = -2$ comes down at such a low magnetic field. The branches D and E, composed of strong absorption spectral lines, were observed around 33 T. This field coincides the field where the anomalous magnetization jump appears, namely, the second level cross field B_c. The straight lines in branches D and E are drawn to fit the experimental data. If we formally determine the g-factor from their slopes, we obtain $g = 3.7$ for both branches of D and E. The value is too large considered from the calculated energy level diagram [4]. This fact means the rapid decreasing of the energy level $S_z = -2$ in $J = 2$ spin multiplet enhanced by the strong ferromagnetic intrachain coupling as has been proposed by Hori et al. [8]. The detailed qualitative discussion is a future problem.

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