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## High field ESR measurements of CsFeCl<sub>3</sub>

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### Abstract

CsFeCl<sub>3</sub> is known to have a singlet ground state system with strong ferromagnetic interaction with the crystallographic c-axis and weak antiferromagnetic interaction in the plane perpendicular to c-axis. Under a magnetic field B applied parallel to the crystal c-axis, one of the states of the doublet excited state comes down to cross the ground state at 7.5 T.

The temperature dependences of the ESR were measured with the frequency 120 GHz and 301 GHz. At the frequency 120 GHz, three absorptions were observed at 4.2 K. One of them was observed below the magnetic field 7.5 T. The other two absorptions were observed above the magnetic field 7.5 T. Above 5 K, however only one absorption was observed above the magnetic field 7.5 T. One absorption was observed below the magnetic field 7.5 T. At the frequency 301 GHz, one absorption was observed below the magnetic field 7.5 T at 4.2 K. Above 5K, no absorption was observed below the magnetic field 7.5 T. On the other hand, above 4.2 K, one absorption was observed above the magnetic field 7.5 T.

## 1. Introduction

The magnetic compound CsFeCl<sub>3</sub> is an interesting material as one of the family of ABX<sub>3</sub> type hexagonal crystal relating to the magnetic frustration caused by the triangular-lattice antiferromagnetism.

Usually the Fe<sup>2+</sup> spin system in this material is treated within the framework of the fictitious spin S=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. CsFeCl3 is a singlet ground state system with strong ferromagnetic interaction with the crystallographic c-axis and weak antiferromagnetic interaction in the plane perpendicular to c-axis. The figure 208

of a crystal structure is shown in Fig. 1. There is a super-exchange interaction which B ion and X ion are considered as the cause of an interaction. Although it has a super-exchange interaction through one chlorine ion at intra-chain, in order to have to mediate two chlorine ions, a super-exchange interaction occurs because it becomes weak in the inter-chain. The ratio of exchange interaction of the intra-chains is about 20 times of the inter-chains exchange interaction. It has the character of 1-D antiferromagnetic chain along c-axis. 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 B applied parallel to the crystal c-axis, one of the states of the doublet excited state comes down to cross the ground state at 7.5T. The energy level diagram and fictitious spin of Fe<sup>2+</sup> are explained. The magnetic ion in CsFeCl<sub>3</sub> is Fe<sup>2+</sup> ion. If this free ion will be in a double clause state and the triple clause state, it is known. And it is divided in three energy level by receiving the effect of spin orbital interaction  $\lambda$ . It will be in a singlet ground state and an excitation doublet state. So, considering the fictitious spin S=1, explanation makes it strain oneself by this system. As for this substance, the crossover of singlet ground state and 1-doublet excite state takes place near 7.5 T. Singlet ground state interchanges by means of this. It is because having observed the changes here in the low magnetic field from magnetic field 7.5 T moved to observation of the changes here bordering on.

Two or more absorptions had been observed around the magnetic field at which a crossover occurs at the recent experiment in which we examined (fig.2). It seemed that it could be ascribed to the crystal slightly unparalleled to c-axis. The phenomenon can be called twin. The experiment was conducted to examine this case.

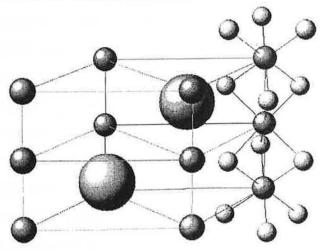


Fig.1 The crystal structure of CsFeCl<sub>3</sub>

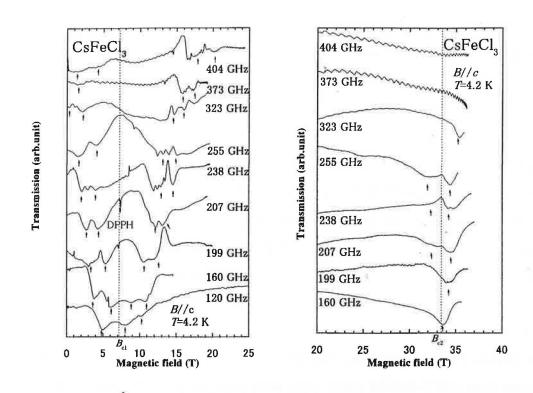


Fig.2 The observed ESR spectra of CsFeCl3 at different frequencies

CsFeCl<sub>3</sub> belongs to an ABX<sub>3</sub> type hexagonal lattice crystal group. This is the substance which is interesting to the singlet ground state system.

This substance was interested in the singlet ground state system. This figure showed pulsed magnetic field and stationary magnetic field for the magnetization curve. As was shown in fig.3[1], under the magnetic field from zero to 4 T the magnetization was weak with a slight linear increasing due to the Van Vleck paramagnetism. The magnetization increased rapidly with increasing magnetic field from 4 T to 12 T. The linear increase of magnetization suggested the appearance of the magnetic order around the field of the ground state crossover, which was consistent with the theory by Tsuneto and Murao[2]. The magnetization seemed to saturate at 12 T. And an adiabatic magnetization process was observed in the pulsed magnetic field. Similarly the magnetization increased linearly 33 T. Up to about 32 T magnetization increases gradually, which reflected the Van Vleck paramagnetism. Although the increase in the magnetization also from 33 T was observed, this could be considered from a crossover. It was possible that meta-magnetism transition took place as this reason. A magnetization curve became blunt unlike the magnetization curve in this pulsed magnetic field, and could consider the process of an adiabatic magnetization in a pulse magnetic field as the cause.

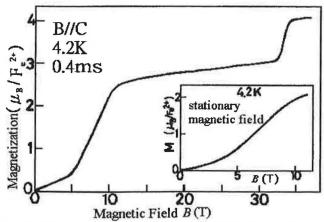


Fig.3 pulsed magnetic field and stationary magnetic field for the magnetization curve

## 2. The experiment method of high field ESR

The experiment method of high field ESR is measuring to detect signal by use of pulsed magnetic field. We make use of Gyrotron FU IV(301 GHz) and Gunn oscillator(120 GHz) as light source. The figure of experiment equipment is shown in Fig. 4.The InSb detector is used as a detector. In order to detect the signal in a pulse magnetic field, even if correspondence wavelength is large like a heat type, the detection machine with a slow response speed cannot be used.

Therefore, since it was necessary to use a quantum type detection machine with a response speed quick as a detector, the InSb detector was used.

Gyrotron FU IV of light source has high frequency in FU Series. The gyrotron consists of 12 T superconducting magnet and shield-off tube. We are using a triode magnetron injection gun. It operates in the additional coils which generate a relatively low magnetic field independently of the main magnetic field generated by a 12 T superconducting magnet.

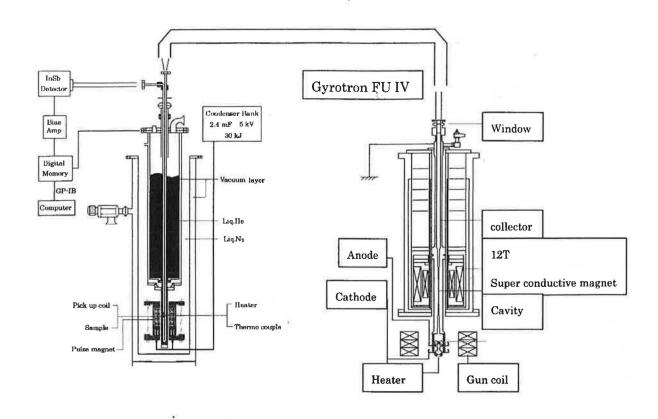
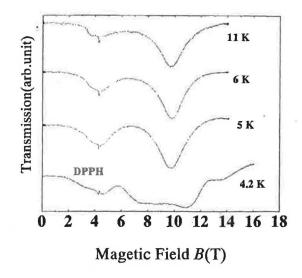


fig.4 The schematic drawing of ESR spectrometer using Gyrotron FU IV

## 3. Experiment result

An ESR experiment result was performed at 4.2 K-11 K under pulsed magnetic field up to 20 T with operating frequencies in the submillimeter region. The applied field was parallel to the c-axis of a single crystal CsFeCl<sub>3</sub>. Fig 5 is temperature dependences of ESR spectra were observed at 120 GHz(4.2 K-11 K). When temperature dependences was roughly divided into the low magnetic field and high magnetic field side bordering on 7.5 T, two ESR absorptions was seen. Or more at 5 K, one ESR absorption is observed at a low magnetic field of 7.5 T, and high magnetic field side, respectively. At 4.2 K, two or more broadcloth absorption is observed to it. Fig 6 is temperature dependences of ESR spectra were observed at 301 GHz(4.2 K-8 K). Although an ESR absorption was observed at 4.2K at the low magnetic field side, an ESR absorption was not checked or above 5 K. At the high magnetic field side, an ESR absorption was checked at any temperature.



6K 5K 42K 0 2 4 6 8 10 12 14 16 18 20 Magnetic Field B(T)

fig.5 Temperature dependences of ESR ESR spectra observed at 120 GHz

fig.6 Temperature dependences

ESR spectra observed at 301 GHz

## 4. Summary

We experimented the high field ESR of CsFeCl<sub>3</sub>. Temperature dependences of ESR spectra were observed at 120 GHz(4.2 K-11 K) and 301 GHz(4.2 K-8 K). At the frequency 120 GHz, three absorptions were observed at 4.2 K. One of them was observed below the magnetic field 7.5 T. The other two absorptions were observed above the magnetic field 7.5 T. Above 5 K, however only one absorption was observed above the magnetic field 7.5 T. One absorption was observed below the magnetic field 7.5 T. It can say that the sample does not have twins. At the frequency 301 GHz, one absorption was observed below the magnetic field 7.5 T at 4.2 K. Above 5 K, no absorption was observed below the magnetic field 7.5 T. On the other hand, above 4.2 K, one absorption was observed above the magnetic field 7.5 T.

### References

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- 2. T.Tsuneto, T.Murao, Physica 51(1971)186.