EXCITON AND MAGNON SIDEBAND IN THE ONE-DIMENSIONAL ANTIFERROMAGNET $\text{CsCoCl}_3 \cdot 2\text{H}_2\text{O}$ UNDER HIGH MAGNETIC FIELD

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ABSTRACT

The absorption spectra corresponding to the transition ${}^4T_1({}^4F) \rightarrow {}^2T_1({}^2P)$ of Co^{2+} in $\text{CsCoCl}_3 \cdot 2\text{H}_2\text{O}$ were investigated under high magnetic fields up to 230kOe with a hybrid magnet. The anomalous line shapes of the magnon sideband with a double peak were observed under the magnetic fields parallel to the a- and c-axes. The behavior of the magnon sideband under the external magnetic fields is related to the exciton transfer. The intrachain exciton transfer becomes large under the magnetic field that induces the ferromagnetic components of the spins. This change of the exciton dispersion is reflected in the line shape of the magnon sideband. The anomalous behavior of the magnon sideband under the external fields was elucidated by the calculation of the line shape taking into account the field dependence of the exciton transfer.

1. INTRODUCTION

$\text{CsCoCl}_3 \cdot 2\text{H}_2\text{O}$ (CCC) is a one-dimensional Ising-like antiferromagnet. Its magnetic properties have been intensively studied by means of NMR [1,2], heat capacity[3], magnetic susceptibility[4] and magnetization[5]. On the other hand, the optical properties are particularly intriguing as well as the magnetic ones. As reported in a previous paper for the absorption spectra of CCC[6], exciton lines with the Davydov splitting and their magnon sideband were observed at the transition ${}^4T_1({}^4F) \rightarrow {}^2T_1({}^2P)$ of Co^{2+} . The magnetic one-dimensionality was found to be reflected in the temperature dependence of the peak energy and the integrated intensity of the magnon sideband. In contrast with the one-dimensionality of the exchange interaction for the ground state, the exciton, called as A-exciton, shows the two-dimensional migration, which was elucidated from the analysis of the Davydov splitting[7]. The two-dimensionality of the A-exciton is attributed to the spin structure. This exciton dispersion must be reflected in the line shape of the magnon sideband. The purpose of this paper is to investigate the behavior of the magnon sideband of CCC under a high magnetic field, where the change of the exciton dispersion is expected to be induced by the change of the spin structure.

The crystal of CCC belongs to the space group of orthorhombic $Pcca(D_{2h}^8)$ and the chemical unit cell contains four translationally inequivalent Co^{2+} ions[8]. Linear chains of Co^{2+} ions extend in the a direction. The magnetic structure below $T_N (=3.38\text{K})$ is shown in Fig.1. All spins lie in the ac-plane canting by 10° from the c-axis, so that each chain has a net moment along the a-axis.[2]

Fig.1. Magnetic structure of $\text{CsCoCl}_3 \cdot 2\text{H}_2\text{O}$. Eight spins in the magnetic unit cell are labeled with the numbers 1 to 8.

2. EXPERIMENT

Single crystals of CCC were grown at room temperature by slow evaporation from the aqueous solution of $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ and CsCl in the molar ratio of 5:1. Thickness of the sample used in the present experiments was 1.0-1.5mm.

The absorption spectra were measured with a Jasco model CT-50 spectrometer. Static magnetic fields were generated by a hybrid magnet in Tohoku University.

3. ABSORPTION SPECTRA IN $H_0//a$

Figure 2 shows the behavior of the magnon sideband B1 with the transition ${}^4T_1({}^4F) \rightarrow {}^2T_1({}^2P)$ of Co^{2+} in CCC under the magnetic field H_0 parallel to the a-axis at 4.2K. A1 is the one of the Davydov-split components of the A-exciton. Under the field higher than 120kOe the shape of the magnon sideband changes into the anomalous one with a double peak B1 and B1'. As reported earlier [9], this double peak is neither the Zeeman splitting nor a sublattice splitting. The change of the line shape is ascribed to the exciton transfer.

The A-exciton migrates among four different sublattices in the ac-plane. In the magnetic field parallel to the a-axis, the tilt angle θ of the spins away from the c-axis increases with the field, so that the exciton transfers for 1-4 and 1-3 Co^{2+} pairs become large proportionally to $\sin^2 \theta$ [10]. This change of the exciton dispersion is reflected in the shape of the magnon sideband. The calculation of the shape of the magnon sideband for CCC considering the field dependence of the exciton dispersion has been reported in ref.9. The calculated results taking the parameters as $V_{12}^0 = 1.3\text{cm}^{-1}$, $V_{14}^0 = 1.8\text{cm}^{-1}$ and $V_{13}^0 = 0.13\text{cm}^{-1}$ are shown in Fig.3, where V_{ij}^0 represents the matrix element of the resonance exciton transfer between i-th and j-th Co^{2+} sites at zero field [7]. For $H_0 = 0$, the line shape is asymmetric with a cut-off on the higher energy side (Fig.3 (a)). As V_{14} and V_{13} increase with $H_0//a$, the shoulder on the lower energy side grows as a peak

Fig.2. Behavior of the magnon sideband of $\text{CsCoCl}_3 \cdot 2\text{H}_2\text{O}$ in $H_0//a$ at 4.2K. E denotes the electric vector of the incident light.

Fig.3. Calculated line shapes of the magnon sideband with the parameters as $V_{12}^0 = 1.3\text{cm}^{-1}$, $V_{14}^0 = 1.8\text{cm}^{-1}$ and $V_{13}^0 = 0.13\text{cm}^{-1}$. The origin of the abscissa corresponds to the center of gravity of the Davydov-split A-exciton lines.

Fig.4 Behavior of the magnon sideband of $\text{CsCoCl}_3 \cdot 2\text{H}_2\text{O}$ in $H_0//c$ at 1.3K.

Fig.5. Dependence of the peak energies of the magnon sideband on the magnetic field parallel to the c-axis.

as shown in Fig.3(b)-(d). Consequently, the observed double peak of B1 and B1' is caused mainly by the increase of the intrachain exciton transfer V_{14} in $H_0//a$. Since short range magnetic order persists above T_N in a one-dimensional magnet, the exciton transfers resonantly even at 4.2K. A more detailed analysis considering the exciton-magnon interaction is necessary for the complete agreement between the observed and calculated line shapes of the magnon sideband.

4. ABSORPTION SPECTRA IN $H_0//c$

The behavior of the magnon sideband in $H_0//c$ at 1.3K is shown in Fig.4 and the peak energies are plotted as a function of H_0 in Fig.5. When an external field is applied along the c-axis, B1 shows the sublattice splitting up to 90kOe. Its higher energy component is remarkably weaker than the lower and disappears above 100kOe. The lower component of B1 shifts to lower energies with the field up to 130kOe and then shifts to higher energies above 160kOe. In the field range from 130 to 160kOe, B1 shows the anomalous shape with a sharp cut-off on the lower energy side, in contrast with the shape at zero field. On the higher energy side of B1, a new peak B1'' appears above 110kOe and shifts to higher energies with the field. Above 200kOe one more new peak appears between B1 and B1''. As the field is increased up to 230kOe, the intensities of B1 and B1'' decrease gradually.

The high field magnetization of CCC at 1.3K has been reported in a previous paper[5]. The magnetization along the c-axis does not show a clear metamagnetic transition but a broad magnetic phase transition, which is due to spin canting. The proposed model of this magnetization process is schematically illustrated in Fig.6, where S_1 and S_2 denote the Co^{2+} spins and, for simplicity, the two-sublattice model is employed. In the absence of an external field, there is no net moment along the c-axis. When an external field is applied along the c-axis, the antiferromagnetic phase transforms to the configuration in which the spins of the two sublattices are oriented at different angles with respect to the c-axis as shown in Fig.6(b). Under the field higher than about 180kOe, the spins align as shown in Fig.6(c).

Now we consider the relation between the spin configuration and the field dependence of the magnon

Fig.6. Schematic view of the spin arrangement in $H_0//c$. S_1 and S_2 denote the Co^{2+} spins.

sideband. In the spin configuration of Fig.6(b), the sublattice-split lower component of B1 shifts to lower energies with the field, but the shift is not linear against the field as shown in Fig.5. This is because the spin S_2 tends to tilt away from the c-axis as the field is increased. In the configuration of Fig.6(c), the magnon sideband shifts to higher energies with the field. Since the ferromagnetic components of the spins are dominant in this configuration, the intensity of the cold magnon sideband decreases, on the other hand, the intrachain exciton transfer V_{14} becomes large. Consequently, the change of the line shape of the magnon sideband in H_0/c is probably ascribed to the increase of the intrachain exciton transfer.

When the spin configuration of Fig.6(b) transforms to that of Fig.6(c) in the field range from 130 to 160kOe, the anomalous shape of B1 with a sharp cut-off on the lower energy side is observed. This change of the line shape cannot be explained only by the field dependence of the exciton transfer. It is necessary to consider the exciton-magnon interaction.

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