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SPIN-CLUSTER ASSOCIATED OPTICAL TRANSITION AND MAGNETIZATION IN CsCoCl₃ UNDER HIGH MAGNETIC FIELDS

H. Hori, K. Amaya¹, J. Nakahara², I. Shiozaki, M. Ishizuka¹, Y. Ajiro³, T. Sakakibara⁴ and M. Date

Department of Physics, Faculty of Science, Osaka University, Toyonaka, Osaka 560, Japan

Abstract. - The magnetizations of Ising like 1D-AF magnet CsCoCl₃ in high magnetic fields show the metamagnetic behaviors of the chain. The spin-cluster associated transition model in the 1D-AF chain explains the observed optical spectral shifts.

1. Introduction

CsCoCl₃ gives a typical example of a quasi one dimensional Ising antiferromagnet (1D-AF) along the c-axis. The 1D-AF chains in CsCoCl₃ form a triangular lattice and the AF interchain exchange interaction induces partial disorder which is observed between the two transition temperatures $T_{N1} = 21.5$ K and $T_{N2} = 9.2$ K [1]. The AF chains order with a ferrimagnetic moment in the c-plane below $T_{N2}$. The metamagnetic transition in the magnetic field $H$ applied parallel to the Ising axis is shown in this paper.

It is well known that a typical magnetic excitation in the Ising spin chain is the spin-cluster excitations as was shown by Date and Motokawa in the ESR experiment [2]. Afterwards, a moving AF domain model or so-called Villain mode [3] was pointed out to explain the neutron diffraction data [4]. The side bands associated with the transition $^{4}T(P) - ^{2}T(P)$ around 18 500 cm⁻¹ have been considered so far to be magnon side bands. However, the spectral shift is too small for the usual magnon side bands and it is well explained by the spin-cluster side band model.

2. Experimental

At temperatures well below 300 K, i.e., the separation between the lowest doublets, the Co²⁺ spins in CsCoCl₃ are described by the following effective spin Hamiltonian with a fictitious spin 1/2 given by

$$\mathcal{H} = \sum_{i,j} [2J S_z^i S_z^j + 2J_{\perp} (S_x^i S_x^j + S_y^i S_y^j)] +$$

$$+ \sum_{i} [g\mu_B S_z H_z + g_{\perp} \mu_B (S_x H_x + S_y H_y)]. \quad (1)$$

The saturation field for $H \parallel c$ at absolute zero is estimated to exceed 40 T. Therefore, we started the magnetization measurements using the single turn coil system at ISSP in university of Tokyo and then the precise measurements were done by the double layer helical coil system in Osaka University. The magnetization $M$ and the field differential magnetization $dM / dH$ of the single crystal of CsCoCl₃ were measured using a conventional pick up coil method.

The optical spectra were observed in High Magnetic Field Laboratory, Osaka university where the standard method of the pulse field magneto-optics was used [5].

3. Experimental results and discussions

The figure 1 shows the $M$ vs. $H$ and $dM / dH$ vs. $H$ curves obtained at 4.2 K for $H \parallel c$. The linear increase of $M$ in the field range $0 < H < 30$ T is also found at 1.7 K and attributed to the Van Vleck temperature independent term. The metamagnetic transition in 32 T $< H < 44$ T and the fine structure shown by an arrow in the $dM / dH$ curve are found. We obtained the saturation moment of $3 \mu_B$ at $H = 44$ T as expected. The fine structure could not be observed in the decreasing magnetic field. The magnetization process is also studied for $H \perp c$. The result shows linear dependence of the magnetization with respect to the magnetic field up to 40 T. The examination of the $M$ vs. $H$ curve, however, shows the non linear increase above $H > 40$ T up to the measured field 57 T where the reached magnetization is 1.45 $\mu_B / Co^{2+}$.

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¹Faculty of Engineering Science, Osaka University, Japan.
²Faculty of Science, Hokkaido University, Japan.
³Faculty of Science, Kyoto University, Japan.
⁴ISSP, University of Tokyo, Japan.

Fig. 1. - $M$ and $dM / dH$ of CsCoCl₃ at $T = 4.2$ K, $H \parallel c$. 

[Insert graph]
The \( \frac{dM}{dH} \) signal obtained by a single turn coil system shows that the saturation field is about 60 T where the saturation moment reaches at about 1.5 \( \mu_B \)/Co

According to Yang and Yang [6], the observed metamagnetic behavior is clearly demonstrated by their theory. The tentative analysis of the experimental data using the theory gives \( J_1 = 83 \) K, \( J_\perp / J_1 = 0.1 \) and \( g_I = 5.8 \). The present value is close to 85 K reported in [7], but is slightly different from 75 K determined by the neutron diffraction [8]. The transverse magnetization gives \( g_A = 3.0 \) including the Van Vleck term, if it exists.

The magnetization in the partially disorder state is also studied [1]. The \( M \) vs. \( H \) curve in the case of \( H \parallel c \) shows that the small structure at 31 T disappears and the stepwise magnetization in higher fields is smoothed out up to measured 48.5 T.

The experimental results of the optical study are shown in figure 2. (A) shows the whole profile of the absorption spectra at \( H = 0 \) and the Zeeman effect is shown in (B). A in (A) is a hot band and others belong to the cold band. B in (A) is the zero-line of these lines. The D-line and its Zeeman effect can be well explained by the usual magnon side band model with the exchange parameter given by the present magnetization data. However, A and C lines are difficult to explain by this model, because the energy difference from B is about half of the corresponding exchange energy. A and C can be explained by introducing a new concept of the spin-cluster associated optical transition. Consider four spins \( S_1, S_2, S_3 \) and \( S_4 \) in a chain as is shown in figure 2C. We assume that \( S_1 \)-site ion absorbs a photon and the excited spin is given by a dotted arrow in B where the exchange energy for four spins is given by \( G + J' - J / 2 \). \( G \) is the optical excitation energy and \( J \) and \( J' \) gives the ground-ground and ground-excited exchange coupling constants, respectively. The \( C \) state is the \( S_3 \)-flipped state associated with the optical transition and the corresponding energy is given \( G + J / 2 \) where \( J' \) term is canceled as is easily seen in figure 2C. Therefore, the energy difference B-C is \( J - J' \) which is about half of the usual magnon side band energy of 2 \( J \) if \( J' \ll J \). This is the reason why the C-line is between B and D. The A-line is understood similarly as the hot band of the spin-cluster associated line and the energy difference between A- and B-line is given by \(- (J + J')\). Observed energy differences of A-B and C-B are 51 and 43 cm

obtained. Mogi et al. [9] explained the hot band by one-spin flip in the Villain mode. The transition, however, is not applicable to C and D because the Villain mode is not excited optically. Therefore, it may be natural to understand the whole spectra by the simple Ising-spin-flip model.

The splitting factors of C, D and H are \( g_C = g_D = 5.3 \) and \( g_H = 0.95 \), respectively. The corresponding excited-state \( g \)-values are obtained as \( g_C = g_D = 1.8 \) and \( g_H = 4.4 \), and these values are well explained by the spin-cluster associated transition model. The detailed analysis is to be shown in our successive paper.

\[ \text{[3] Villain, J., \textit{Physica B} \textbf{79} (1975) 1.} \]