Journal of Magnetism and Magnetic Materials 104–107 (1992) 815–816
North-Holland

journal of magnetism and magnetic materials

Phase transition in $CsCoCl_3$ and $CsCo_{1-x}Mg_xCl_3$ in magnetic fields

H. Hori [a], H. Mikami [a], M. Date [a], Y. Ajiro [b] and N. Mori [b]

[a] *Faculty of Science, Osaka University, Toyonaka, Osaka, Japan*
[b] *Faculty of Science, Kyoto University, Sakyo-ku, Kyoto, Japan*

Two peaks of the differential magnetization are observed in $CsCoCl_3$ at 32.9 and 43.5 T and two additional peaks at 16 and 24 T in Mg-doped crystal, $CsCo_{1-x}Mg_xCl_3$. The two peaks in pure $CsCoCl_3$ are attributed to the antiferromagnetic (AF)–ferrimagnetic (FRI) and FRI–ferromagnetic (FRO) boundaries and the two additional peaks in Mg-doped crystal are understood by the local spin-flip of Co^{2+} ions around the Mg^{2+} impurities. The intrachain and the first nearest interchain exchange parameters are consistently obtained as $J_0 = -54$ cm^{-1} and $J_1 = -5.2$ cm^{-1}, respectively.

$CsCoCl_3$ is a one-dimensional Ising-like antiferromagnet which is characterized by a main intrachain exchange interaction J_0 of 52 cm^{-1} (74.8 K) [1]. The nearest and second nearest weak interchain exchange interactions J_1 and J_2 on the triangular-plane induce the spin frustration effect and two transition temperatures $T_{N1} = 21.3$ K and $T_{N2} = 9.2$ K [1] have been found. The antiferromagnetic structure is stable below T_{N2}. An ideal one-dimensional model with very weak interchain interactions is believed to describe in this compound, since a theoretical estimation of $J_1 = 0.52$ cm^{-1} (0.75 K) and $J_2 = 0.0052$ cm^{-1} (0.0075 K) was published associated with the existence of the two transition temperatures [2]. Recently, however, two sharp peaks in the differential magnetization dM/dH_0 were observed at H_{c1} of 32.9 T and H_{c2} of 43.5 T. A model based on the one-dimensional anisotropy spin theory by Yang and Yang [3] has been proposed. If the ideal one-dimensional model is assumed, the observed two-peaks could be explained by the broadening of the antiferro–ferromagnetic transition in the presence of the transverse component of the intrachain exchange interactions [4]. However, the observed two-peak profile is not in agreement with the theory and the high field data induce doubt about the simple one-dimensional model. On the other hand, the two-peaks can simply be explained by successive magnetic phase transition model of AF–FRI and FRI–FRO, if J_1 and J_2 are considerably larger than expected [5]. An extensive study has been done in $CsCo_{1-x}Mg_xCl_3$ to investigate the magnetization mechanism in $CsCoCl_3$.

The magnetization measurements were done in Research Center for Extreme Materials, Osaka University. A description of the measurement system and the methods has been given elsewhere [6,7], so that only a brief outline of the system will be presented here. The magnet with the inner diameter of 20 mm is used and it can produce the field up to 60 T. A transient recorder (IWATSU Model DM-2350) is used for the detection of differential magnetization signal together with the monitored signal of the magnetic field. Rod-like samples with the length of about 5 mm and 2.5 mm in diameter were used for the magnetization measurement.

Differential magnetization curves of both $CsCoCl_3$ and $CsCo_{1-x}Mg_xCl_3$ are shown in fig. 1. The two weak peaks at 16 T (H_B) and 24 T (H_A) in addition to the two strong peaks at 34 T (H_{c1}) and 43.5 T (H_{c2}) are observed in $CsCo_{1-x}Mg_xCl_3$. H_{c1} and H_{c2} are explained by the model of AF–FRI and FRI–FRO transitions in an external field [5] which are shown in fig. 2. The spin structures of $CsCoCl_3$ below T_{N2} [1] and the exchange parameters are also shown in fig. 2. Using the exchange parameters J_0, J_1, J_2 and J_1', the critical fields H_{c1} and H_{c2} are written by

$$H_{c1} = -2J_0/g_{\parallel}\mu_B, \tag{1}$$

$$H_{c2} = -[2J_0 + 6(J_1 + J_1')]/g_{\parallel}\mu_B, \tag{2}$$

where $g_{\parallel}$ and μ_B are the g-value along the c-axis and Bohr magneton, respectively. It is noticed that H_{c1} and H_{c2} do not depend on J_2 so that the parameter cannot

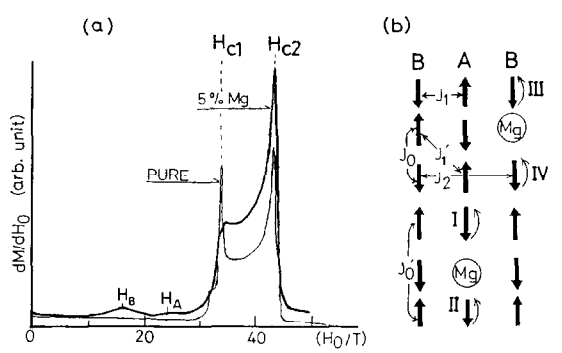

Fig. 1. (a) Experimental differential magnetization curves of both $CsCoCl_3$ and $CsCo_{1-x}Mg_xCl_3$. (b) Schematic local spin-flip modes around Mg^{2+}.

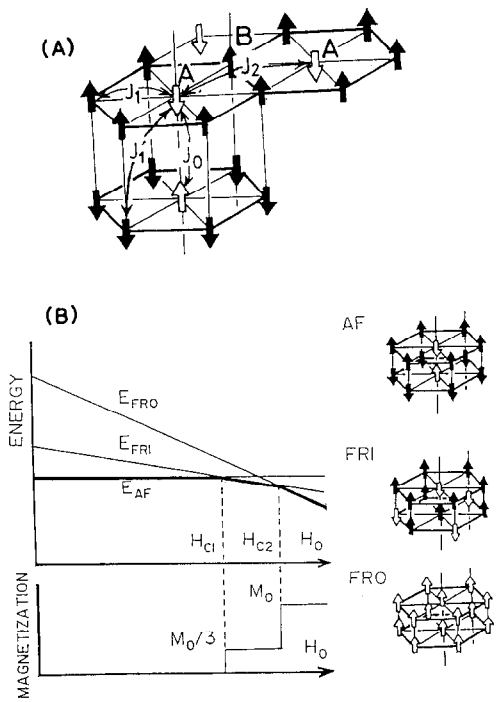

Fig. 2. (A) The schematic spin structure of $CsCoCl_3$ below T_{N2}. (B) Field dependence of the energies in the phases AF, FRI and FRO and the theoretical magnetization process.

be determined from the critical field data. As was discussed in ref. [5], the positive sign of J_2 is necessary to stabilize the AF phase in fig. 2. However, J_2 may be very small.

The additional weak peaks at 16 and 24 T arise from the effect of the Mg^{2+} impurity. They are analyzed by use of the local spin-flip model [8]. When the applied field exceeds the local exchange field on a spin, the spin-flip occurs, if the spin is highly Ising-like. As seen in fig. 1, the spins I and II in the chain A and spins III and IV in chain B see the weak exchange fields, because Mg^{2+} ion has no magnetic moment. The spin-flips are expected at about half of H_{c1} and H_{c2}, because the main exchange fields come from the intrachain interactions. The corresponding two critical fields of the local spin flips on A- and B-chains are given by

$$H_A = (-J_0 - 6J_1 + 6J_2 + 12J_1')/g_{\parallel}\mu_B, \quad (3)$$

$$H_B = (-J_0 + 6J_2)/g_{\parallel}\mu_B. \quad (4)$$

Neglecting J_2 and using the experimental values of H_{c1}, H_{c2}, H_A and H_B, the exchange parameters J_0, J_1 and J_1' are determined from eqs. (1)–(4) as -54, -5.2 and -0.6 cm^{-1}, respectively.

The estimated value of J_0 is in agreement with the value in ref. [1] but a large difference is found by comparing J_1 of ref. [2]. It is noted that the model in ref. [2] can explain the additional peak at H_B but H_A-peak is impossible to describe. On the other hand, our model can explain both H_A and H_B with the exchange parameters required from the H_{c1} and H_{c2} data. The model in this work is also consistent with the theory by Kaburagi et al. [9]. A detailed discussion will be published elsewhere.

The authors would like to express their thanks to Professor Yamagishi for his kind agreement of the use of the high magnetic field facilities. This work was supported in part by the Grant in Aid Scientific Research from the ministry of Education, Science and Culture.

References

[1] M. Mekata, J. Phys. Soc. Jpn. 42 (1977) 76.
[2] H. Shiba, Prog. Theor. Phys. 64 (1980) 466.
[3] C.N. Yang and C.P. Yang, Phys. Rev. 151 (1966) 258.
[4] K. Amaya, H. Hori, I. Shiozaki, M. Date, M. Ishizuka, T. Sakakibara, T. Goto, N. Miura, H. Kikuchi and Y. Ajiro, J. Phys. Soc. Jpn. 59 (1990) 1810.
[5] H. Hori, K. Amaya, H. Mikami and M. Date, J. Magn. Magn. Mater. 90 & 91 (1990) 269.
[6] M. Date; IEEE Trans. Magn. MAG-12 (1976) 1024.
[7] H. Mollymoto, M. Motokawa and M. Date, J. Phys. Soc. Jpn. 49 (1980) 108.
[8] For example, A.R. King, V. Jaccarino, T. Sakakibara, M. Motokawa and M. Date, High Field Magnetism (North-Holland, Amsterdam, 1983) p. 11.
[9] M. Kaburagi, T. Tonegawa and J. Kanamori, J. Phys. Soc. Jpn. 53 (1984) 1971.