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► **To cite this version:**

H. Nojiri, Y. Tokunaga, M. Motokawa. MAGNETIC PHASE TRANSITION OF HELICAL CsCuCl₃ IN HIGH MAGNETIC FIELD. *Journal de Physique Colloques*, 1988, 49 (C8), pp.C8-1459-C8-1460. 10.1051/jphyscol:19888670 . jpa-00228902

HAL Id: jpa-00228902

<https://hal.science/jpa-00228902>

Submitted on 4 Feb 2008

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MAGNETIC PHASE TRANSITION OF HELICAL CsCuCl₃ IN HIGH MAGNETIC FIELD

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Abstract. – Magnetization measurements of CsCuCl₃ is performed using pulsed high magnetic fields at various temperatures below T_N . Magnetization shows a small jump at 12.5 T and saturation at 31 T at 1.1 K when $H_0 \parallel c$, while a small plateau at around 12 T when $H_0 \perp c$. The results are quite different from those reported so far in other triangular lattice substances and seem to be due to other type of incommensurate phase transition.

Hexagonal ABX₃ type compounds show complicated but interesting magnetic behaviors due to triangular lattice structure, where A = Rb, Cs, X = Cl, Br and B = Mn, Fe, Co, Ni, Cu. They have been investigated for more than two decades [1] but still remain some attractive problems. In this type of crystal structure, magnetic moments couple each other strongly along the c -chain and then these compounds can be regarded as quasi-one dimensional magnets. The inter-chain exchange interaction is usually weak compared to intrachain one. Because of the triangular network in the c -plane, the moments whose in-plane interaction is antiferromagnetic have frustration and then spin structure becomes complicated.

CsCuCl₃ is one of the materials of this category [2]. Strong intrachain ferromagnetic exchange interaction $J_1 = 16.7 \text{ cm}^{-1}$ makes almost ferromagnetic but low pitch helical spin structure along the c -axis below $T_N = 10.6 \text{ K}$. This is considered to be due to the antisymmetric Dzyaloshinsky-Moriya interaction. In the c -plane, the spin system shows a 120° structure caused by weak antiferromagnetic interchain interaction $J_2 = 2.7 \text{ cm}^{-1}$. The magnetic moment lies in the plane due to D-M interaction whose vector is parallel to the c -axis, although dipolar anisotropy apparently makes spins prefer to direct to the c -axis.

Magnetization measurements on CsCuCl₃ was first reported by Motokawa [3], and it has been found that this substance shows an abrupt magnetization change at $H_c = 12.5 \text{ T}$ at 1.1 K in a field parallel to the c -axis as is shown in figure 1. It seemed strange at that time because a smooth increase of magnetization was first expected up to saturation field ($H_s = 31 \text{ T}$) since the spins were in the c -plane with 120° structure at zero field and a commensurate cone structure was expected in a field. In a field perpendicular to the c -axis, a small plateau was observed at around 12 T instead of an abrupt transition. Recently Fedoseeva *et al.* reported magnetization measurements which were done in the vicinity of T_N in a field up to 8 T using superconducting magnet [4]. They also observed a small jump of magnetization and they ascribed it to a phase tran-

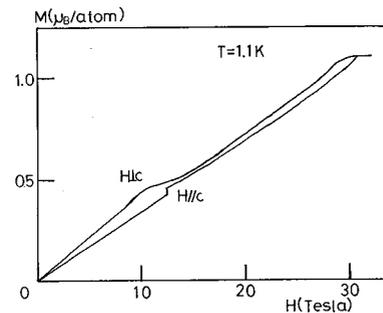


Fig. 1. – Magnetization curve of CsCuCl₃.

sition from the 120° structure to an incommensurate state. To investigate these phases in more detail and to make a phase diagram which covers high field region and intermediate temperature region, we reexamined magnetization measurements using pulsed high magnetic fields at various temperatures between 1.1 K and T_N .

When an external field H_0 is parallel to the c -axis, magnetization increases almost linearly with increasing field up to H_c as is shown in figure 1. Just above H_c , the increasing rate is slightly less than that below it and the extrapolation of magnetization curve above H_c to zero field intersects the ordinate at finite magnetization value, for example $0.06 \pm 0.01 \text{ m}\mu_B$ at 1.1 K. This means that the spin structure above H_c must be one which shows residual weak ferromagnetic moment if this structure is kept stable at zero field. This is different from usual spin flop phenomenon in which the extrapolation must intersect the origin. Above H_c , the magnetization increases waping upward and finally saturates at H_s . The temperature dependences of H_c and H_s are shown in figure 2 as a phase diagram. H_c decreases with increasing temperature and merges with paramagnetic-antiferromagnetic boundary just below T_N as is shown in figure 2. Our experimental result in the vicinity of T_N is consistent with that obtained by Fedoseeva *et al.* The magnitude of magnetization jump at H_c , ΔM , is temperature de-

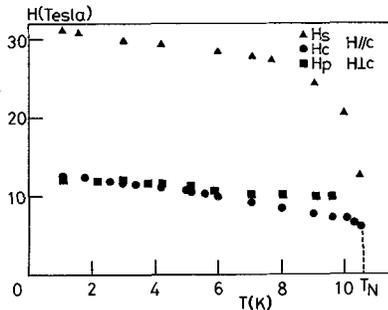


Fig. 2. — Temperature dependence of saturation field H_s , transition field H_c when $H_0 \parallel c$ and plateau field H_p when $H_0 \perp c$

pendent. It is almost constant below 3 K and shows a maximum at 6 K. The magnetization jump is sometimes separated by two steps in vicinity, main one and small satellite one, depending on sample and temperature. We consider there must be domains of two phases in a field between two steps.

It was first pointed out by Shiba [5] and Shiba and Suzuki [6] that incommensurate phase appears in such a triangular lattice system due to dipole-dipole interaction. They calculated mainly for RbFeCl_3 and it was a case of XY -type spin system. They also discussed magnetic field effect when the field is in-plane. In the case of CsCuCl_3 , however, it is Heisenberg type spin system and the field effect we are now discussing is the case of perpendicular field to the plane, i.e. parallel to the c -axis. Fedoseeva *et al.* calculated the phase boundaries from paramagnetic state to incommensurate phase or 120° structure phase according to Shiba's method. They used Landau expansion which is useful only when thermal average of spin is small

in the vicinity of the transition temperature. At low temperature, this method is no longer valid and no theoretical treatment to explain the transition at H_c has been done. Nagamiya discussed general case of helical spin system [7]. According to his theory, when an external field is applied in the XY -plane in the XY -spin system, the helical spin structure turns to fan structure at certain field which is $(\sqrt{2} - 1) H_s$ in the 120° spin structure case. In his theory, dipole-dipole interaction is not taken into account. Our experimental result shows H_c is equal to $0.4 H_s$. This is suggestive his theory is adaptable to our case, but applied field is perpendicular to the XY -plane in this case. Then the fact that $H_c = 0.4 H_s$ seems to be accidental.

When an applied field is perpendicular to the c -axis, magnetization increases with increasing field showing a small plateau at around 12 T and again increases up to the saturation field as is shown in figure 1. The temperature dependence of this plateau field is more gradual than that of H_c as is shown in figure 2. In this case, spin structure is considered to gradually change from 120° structure to fan structure.

The early stage of this study has been performed in Osaka University.

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