Experimental Report

Proposal: **5-41-704** **Council:** 4/2012

Title: Re-examination of crystallographicand magnetic chirality in CsCuCl3

This proposal is resubmission of: 5-41-685

Researh Area: Physics

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Samples: CsCuCl3

Instrument	Req. Days	All. Days	From	To
D3	0	4	29/10/2012	02/11/2012

Abstract:

The relationship between crystallographic and helimagnetic chirality has been paid attention, because the sense of a screw spin structure depends on the right- or left-handed chiral crystal structure that allows a Dzyaloshinskii-Moriya (DM) interaction. CsCuCl3 is a very good candidate because it has a chiral crystal structure with a space group of P6122 and shows helimagnetism below 10.5 K. Some experimental groups have performed polarized neutron diffraction experiments. However, they got inconsistent results, because of difficulties in evaluating and controlling the crystallographic chirality. By our unique crystallization and X-ray diffraction technique, we recently succeeded in obtaining mm-ordered single crystals with well-controlled crystallographic chirality.

In order to solve the disagreements in the past neutron studies and to investigate the interplay between crystalline and magnetic chirality, it is strongly necessary to perform new polarized neutron diffraction experiments in CsCuCl3. For that reason, we are applying for 7 days of beam time in D23 instrument

Re-examination of crystallographic and helimagnetic chirality in $CsCuCl_3$

Chirality concept (right-handedness or left-handedness) is one of the most fundamental building blocks of nature, ranging from nano and biosciences to cosmic sciences. Therefore, it is very important to understand the chirality in molecules, crystals and magnetic structures both from theoretical and experimental viewpoints. Recently, the interplay between crystallographic and magnetic chirality have been paid attention because the properties of a helical magnetic structure strongly depends on the chiral crystal structure which allows an anti-symmetric Dzyaloshinskii-Moriya (DM) interaction [1, 2].

$CsCuCl_3$ have a chiral crystal structure with chiral space groups of right-handed $P6_122$ or left-handed $P6_522$ (Figure 1). Magnetization measurements show an antiferromagnetic response at T_N = 10.5 K. Cooling temperature below T_N, unpolarized neutron diffraction studies show helimagnetic ordering with the magnetic propagation vector $\boldsymbol{k}_{mag}$ = (1/3, 1/3, ±δ) [3]. To determine the domain ratio between right- and left-handed helimagnetic ordering in the right-handed crystal of $CsCuCl_3$, some experimental groups have performed polarized neutron diffraction experiments, and evaluated magnetic satellite intensities with the neutron polarization parallel and anti-parallel to the *c*-axis. However, their results are different probably due to the problems in evaluating the crystallographic chirality [4, 5].

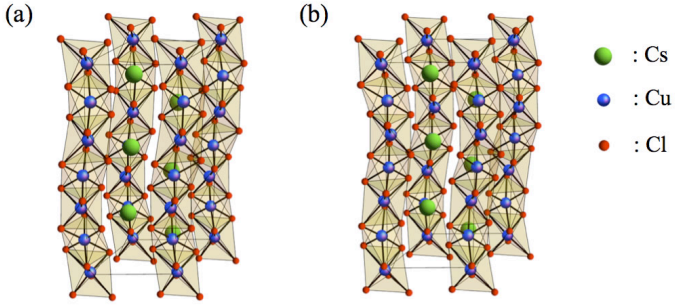

Fig. 1 (a) right-handed and (b) left-handed crystal structures in $CsCuCl_3$.

We have synthesized single crystals of $CsCuCl_3$ and evaluated the crystallographic chirality using circularly-polarized resonant X-ray diffraction. Our findings indicate that cm-ordered single crystalline samples are obtained by conventional spontaneous crystallization technique. However, all of such crystals form racemic twinned. To make the samples with the single crystallographic chirality, the sample size must be as small as sub-mm [6, 7]. However, by adapting a unique crystallization technique, we recently succeeded in controlling the crystallographic chirality and obtaining the mm-ordered single crystals of $CsCuCl_3$.

In order to solve the disagreement between reported polarized neutron diffraction results and to examine the interplay between crystalline and helimagnetic chirality in $CsCuCl_3$, we performed high-flux polarized hot neutron diffraction experiments at D3 in ILL using mm-ordered single crystals with well-controlled crystallographic chirality. The D3 diffractometer was operated with 2-axis mode and its wavelength λ = 0.84 Å, and the data was collected at 1.5 K. The single crystalline specimen with the size of 0.8 mm diameter and 2.5 mm length was used for the experiments. Single crystalline X-ray diffraction experiments indicated that the specimen formed only the right-handed ($P6_122$) crystallographic domain.

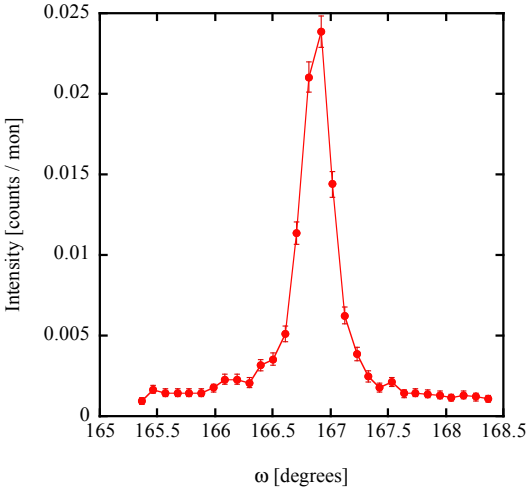

Fig. 2 ω-scan profile at the nuclear (0,0,6) reflection.

Figure 2 shows a ω-scan profile at the nuclear (0,0,6) reflection. It is obvious that a clear nuclear Bragg peak was observed. As the magnetic moment of the Cu site is 0.6 μ_B, the

expected magnetic satellite intensity is 1000 times smaller than the (0,0,6) reflection [3].

Figure 3 shows ω-scan profiles at the (1/3,1/3,6-δ) reflection. The magnetic satellite peak may be observed only when the incident neutron polarization was parallel to the scattering vector. To discuss the population of the right- and the left-handed helical structure, it is necessary to make quantitative discussions of the observed intensity. However, due to the large background and weak magnetic satellite signal, it is quite difficult to make the discussions. At the present, our results suggest that the right-handed $CsCuCl_3$ mainly forms right-handed helimagnetic structure.

To minimize the background intensity, we are planning to perform polarized cold neutron diffraction experiments with the triple-axis mode. To increase the magnetic satellite peaks, we will prepare larger enantiopure single crystals. And with takeing the 2 measures, we will make quantitative discussion in observed magnetic satellite intensity, and determine the sense of helimagnetic structure. To discuss the interplay of crystalline and helimagnetic chirality, we will take the data using the right- and the left-handed $CsCuCl_3$.

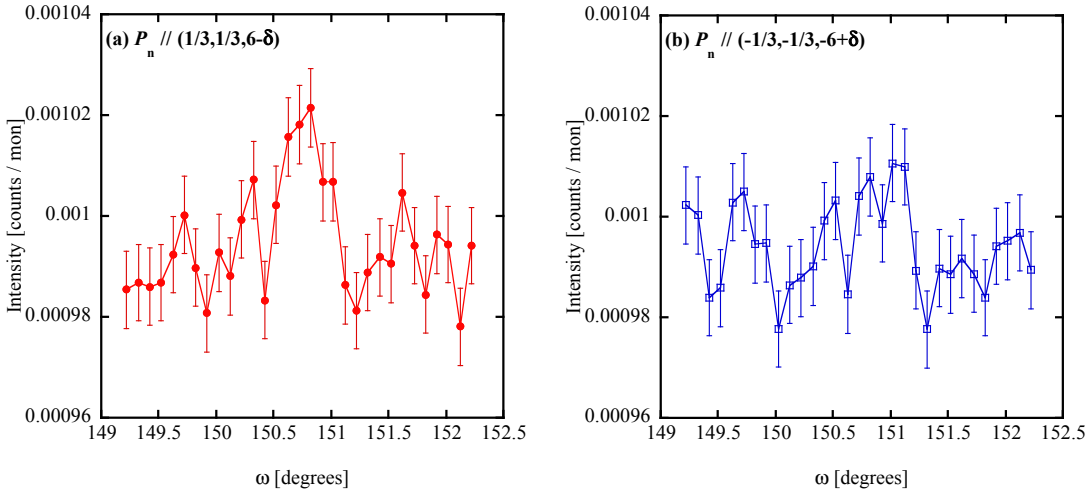

Fig. 2 ω-scan profiles at the magnetic (1/3,1/3,6-δ) reflection when the incident polarization is (a) parallel and (b) antiparallel to the scattering vector.

References

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