（ ※本報告書は英語で記述してください。ただし、産業利用課題として採択されている方は日本語で記述していただいても結構です。 ）

TOKAI CROSS Experimental Report J-PARC	提出日 Date of Report 2013.2.7
課題番号 Project No. 2012A0064 実験課題名 Title of experiment Real-time observation of magnetic structural change in frustrated magnets 実験責任者名 Name of principal investigator Kiyoichiro Motoya 所属 Affiliation Tokyo University of Science	装置責任者 Name of responsible person Takashi Ohara 装置名 Name of Instrument/（BL No.） BL18 実施日時 Date and time of Experiment 2012. 10/31-11/2

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。（適宜、図表添付のこと）

Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.

$Ca_3Co_2O_6$, single crystal, 12.8mg

$Ca_3Co_2O_6$ crystallizes in a hexagonal structure. It consists of Co chains along the c-axis and these chains are arranged in a triangular lattice in the c-plane. This compound shows two magnetic transitions at T_{c1}=25 K and T_{c2}=13 K and 5-step metamagnetic transitions below 7 T at 1.9 K.

Previous neutron scattering measurement showed that the magnetic Bragg peak observed at (1, 0, 0.0085) immediately after cooled to 10 K moved towards (1, 0, 0) with time. On the basis of the neutron scattering intensity after sufficient waiting time at T =10.5 K, we determined that the magnetic moments of Co atoms form a series +++ · · · +++/− − − · · · − − −/+++ · · · +++ along a Co chain where + and − mean the moment direction parallel and antiparallel to the c direction, respectively. Therefore, the shift of the peak position with time corresponds to an increase of the ferromagnetically coupled length of the Co chain.

2. 実験方法及び結果（実験がうまくいかなかった場合、その理由を記述してください。）

Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.

Neutron scattering experiment on single crystal of $Ca_3Co_2O_6$ was made using the BL-18 time of flight instrument at the J-PARC. To get information for the magnetic transition at T_{c2}, temperature variation of intensity of magnetic reflection was measured between 2.8 and 26 K.

Figure 1 shows the neutron intensity pattern in the ($h0l$) reciprocal lattice plane at T=2.8 K. This pattern was measured using first frame (214 kW, 1hour). Some nuclear and magnetic Bragg reflections were observed.

Figure 2 shows the pattern of the (-2 0 +δ) and (-2 0 -δ) magnetic reflections in the ($h0l$) reciprocal lattice plane at 2.8 K. This pattern was measured using second frame (214 kW, 2hour).

In these measurements, diffuse scattering were observed around magnetic reflections along the a^* axis.

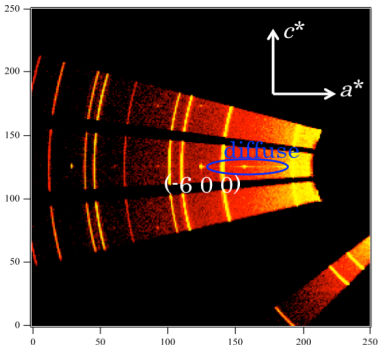

Figure 1

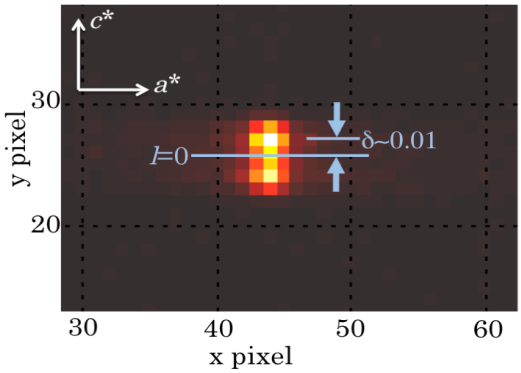

Figure 2

2. 実験方法及び結果（つづき） Experimental method and results (continued)

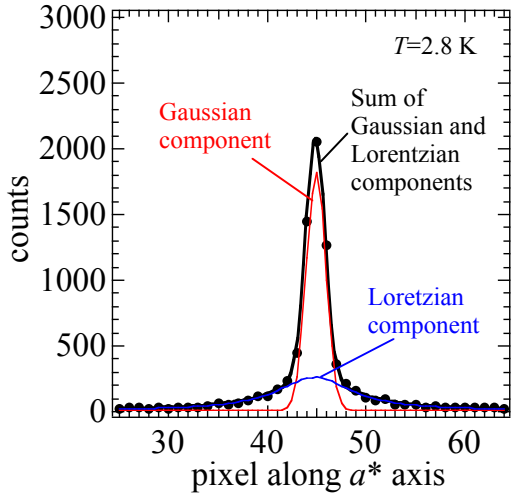

Figure 3

Temperature variation of magnetic reflections was measured in the (h0l) plane using the second frame. Each measurement was made for about 2 hours. Figure 3 shows profiles along the a^*-axis of a sum of (2 0 +δ) and of (2 0 -δ) magnetic reflections observed at 2.8 K. We analyzed the observed patterns assuming that the scattering function consists of a sum of a Gaussian and a Lorentzian functions. Curves in Fig. 3 show the results of the fittings.

Figure 4(a) shows the temperature variation of intensities of the (300) nuclear reflection and the sum of (2 0 +δ) and (2 0 -δ) reflections. The intensity of the (2 0 +δ) and (2 0 -δ) reflections shows a broad peak at approximately T=13 K (=T_{c2}).

Figure 4(b) shows intensities of the Gaussian and the Lorentzian components. Figures 4(c) and 4(d) show the width of the Gaussian and Lorentzian components, respectively. The intensity of both components show anomalous behavior at ~13K in the heating process. Temperature variation of the width of both components also shows a characteristic behavior at ~13K.

Below 13K, the intensity of the sum of the (2 0 +δ) and (2 0 -δ) magnetic reflections decreases with decreasing temperature. We set to work on construct a scenario to explain these temperature variations of magnetic scattering intensity and width.

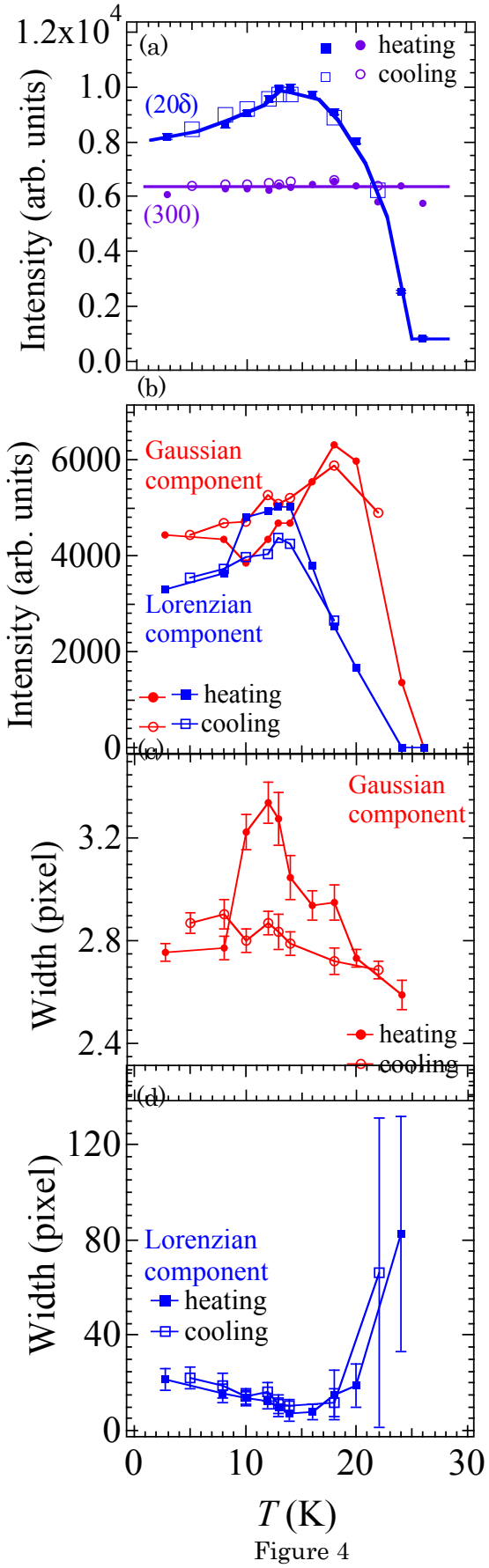

Figure 4