実験報告書様式（一般利用課題・成果公開利用）

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CROSS TOKAI **Experimental Report** J-PARC	承認日 Date of Approval 2017/07/20 承認者Approver Takashi Ohhara 提出日 Date of Report 2017/06/15
課題番号 Project No. 2016B0262 実験課題名 Title of experiment Determination of possible antiferromagnetic Skyrmion-lattice like spin structure in CaBaCo2Fe2O7 実験責任者名 Name of principal investigator Johannes Reim 所属 Affiliation IMRAM, Tohoku University, Sendai	装置責任者 Name of responsible person Takashi Ohhara 装置名 Name of Instrument/（BL No.） Senju / 18 実施日 Date of Experiment 2017/03/22 – 2017/03/28

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.

Four small single crystal of CaBaCo2Fe2O7: 2*74mg and 2*35mg

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.

The layered kagome system in the hexagonal Swedenborgite structure displays similarly to the Pyrochlores a highly frustrated network of tetrahedral coordinated magnetic ions. However, its broken inversion symmetry raises further the complexity of ordering due to non-vanishing Dzyaloshinski-Moriya interactions. Recently investigated compounds of this family show various signs for unusual geometric frustration and disordered ground states despite of the typically strong antiferromagnetic exchange.
From X-ray powder and neutron single crystal scattering measurements the crystallographic structure of the compound $CaBaCo_2Fe_2O_7$ was successfully refined in $P6_3mc$ symmetry and determined to be structural invariant under temperature within the resolution limits. Further experiments using polarization analysis have evidenced a K-type antiferromagnetic order arising below $T_N \approx 160$K. High resolution neutron diffractometer revealed a coexistence of a commensurate and long periodic modulation within the antiferromagnetic phase. The latter one can be described with the 3-q star q^*_s of the propagation vector q_s = (0.342, 0.342, 0) noted in the crystallographic unit cell. This corresponds to a splitting of $\delta \approx 0.017Å^{-1}$ or rather a periodicity of 370Å. Based on the polarization analysis results the spin structure was deduced to be cycloidal. Taking these information into account a spin structure was established using a model of superimposed cycloidal waves along each arm of the star q^*_s, which is compatible with the crystal symmetry. The structure factor of the resulting spin structure corresponds qualitatively with the observations. Considering the information from polarization analysis the few peak intensities compare favorably with our model. This preliminary model of the spin structure itself is Skyrmion-lattice like with a winding number of w=-1, which indicates a topologically protected swirling.
Using the high resolution single diffractometer Senju we have addressed several open questions successfully.

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

After checking all three wavebands, short measurements with the 1st waveband frame providing the best Q coverage were used for the determination of the sample orientation while the 2nd waveband (4.6 ~ 8.8 Å) was ideal to resolve the peak splitting. The main target was to measure a large data set of peaks belonging to the long periodic modulation, which will allow us to refine the spin structure and compare it with our model calculations. Specifically, we were interested whether the refined spin structure exhibits a non-vanishing winding number. Selecting 12 sample orientations about 50 long periodic peak triplets were measured at 4K on a single sample. The determination of the integrated intensities for each single long periodic peak is currently under way.

Recent observations have revealed a very intriguing magnetic field dependence, specifically two different magnetic states have been identified, which are stable above T_N. This behavior is most likely closely related to the stabilization mechanism for the potential antiferromagnetic skyrmion lattice. Unveiling this mechanism is of utmost priority for the comprehension of this new topology but also for the search of other candidate materials. Thus, prior to the experiment the four single crystals cut from a larger piece of CaBaCo2Fe2O7 have been exposed to different magnetic field protocols (sample 1: applied field along a and c-axis then demagnetized at room temperature, sample 2: no field, sample 3: 5T along c-axis, sample 4: 5T along a-axis). From the scattering results we were able to confirm our hypothesis that is to say inducing a net magnetic moment using external magnetic field prevents the long periodic modulation from being stabilized exclusively (Fig. 3), while the net moment can be erased with a demagnetization protocol reestablishing the complete splitting (Fig. 2). Thanks to the outstanding support by the sample environment team we were able to study the magnetic field and temperature dependence in more detail using the 7T magnet. According to these results only close to room temperature the magnetic state can be influenced while at low temperatures (<=80K) no change in the magnetic order was observed (Fig. 4). However, at room temperature the initial state of the sample could be reversed successfully.

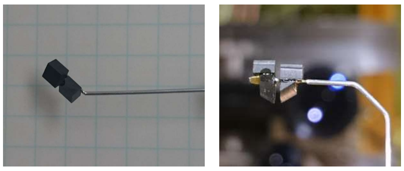

Figure 1: In order to to measure all samples in a short time, two or four samples were measured simultaneously. Since only the Bragg peaks of the commensurate and long periodic modulation are of interest, a slight misalignment allows to attribute the scattering contribution to the each sample (compare Fig. 3).

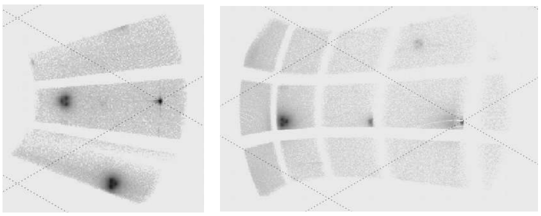

Figure 2: Reconstruction of the reciprocal space parallel to the (hk0) scattering plane for a measurement at 4K of the samples 1 and 2 with different orientation. Lines indicate the reciprocal lattice with nuclear peaks on intersections. The magnetic peaks (2/3 2/3 0) of sample 1 (left) and (1/3 1/3 2) of sample 2 (right) both show an absence of the commensurate modulation.

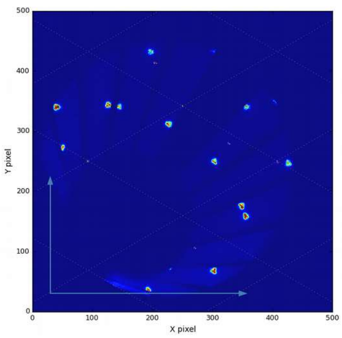

Figure 3: Reconstruction of the (hk0) scattering plane for a measurement of the samples 3 and 4 with different orientation at 4K. Lines indicate the reciprocal lattice for the sample 3 with nuclear peaks on intersections. Both samples exhibit non-vanishing scattering intensity at the commensurate magnetic Bragg peak position. This shows, that the magnetic state, still present in sample 2, was successfully changed using the external magnetic field. Furthermore, the mechanism appears to be independent from the field direction.

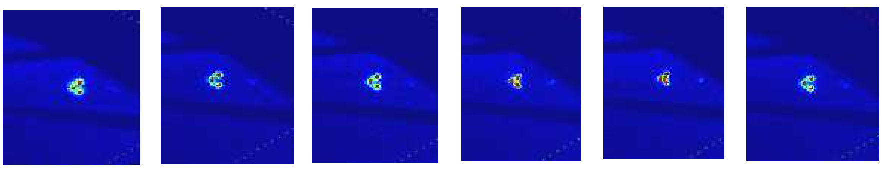

Figure 4: Reconstruction of the (hk0)-scattering plane close to the commensurate magnetic Bragg peak position (2/3 2/3 0). Figures are in order of measurement, from left to right: (a) 4K 6T, (b) 4K 0T, (c) 40K 6T, (d) 80K 0T, (e) 80K 6T, (f) 4K 0T. At 4K and without external magnetic field, the completely split scattering pattern is observed again despite the application of fields up to 6T.