実験報告書様式（一般利用課題・成果公開利用）

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Experimental Report	承認日 Date of Approval 2014/10/14 承認者 Approver Ryoichi Kajimoto 提出日 Date of Report 2014/10/10
課題番号 Project No. 2013B0245 実験課題名 Title of experiment Low dimensionality excitations in the Swedenborgite compound CaBaCo2Fe2O7 実験責任者名 Name of principal investigator Johannes Reim 所属 Affiliation JCNS,Forschungszentrum Juelich,52428 Juelich, Germany	装置責任者 Name of Instrument scientist Ryoichi Kajimoto 装置名 Name of Instrument/（BL No.） BL-01 実施日 Date of Experiment 27.2.2014 – 3.3.2014

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.

A single crystal of $CaBaCo_2Fe_2O_7$ of 5g.

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.

Geometrically frustrated spin system may exhibit a macroscopic manifold of competing ground states leading to emergence of new, sometimes exotic phenomena, like topological magnetic monopoles, and to a functional link for multiferroic behavior. The proposed system to study is $CaBaCo_2Fe_2O_7$, a compound with the swedenborgite structure. Similarly to the pyrochlores, magnetic ions form a highly frustrated network with tetrahedral coordination. In addition, inversion symmetry is broken and Dzyaloshinskii-Moriya interaction may contribute on all magnetic exchange paths.

We have now successfully grown large single crystals of $CaBaCo_2Fe_2O_7$ (~2cm^3), a swedenborgite compound which is so far unique in showing antiferromagnetic order below T_N≈160K and resembling features of the predicted complex ground state without changing the hexagonal structure. Magnetic ordering is characterized by a √3x√3-supercell, where geometric frustration is expected to promote a strong anisotropy in spin correlations with 1D spin chains perpendicular to more disordered kagome layers. Along c-direction previously measured data are not consistent with linear spin wave calculations based on the coplanar √3x√3-ground state and estimated exchange parameter. Including frustration effects leads to a decoupling of spin columns perpendicular to kagome layers through interlayer sites and favor chain-like excitations.

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

We proposed to use the thermal time-of-flight instrument 4Seasons making use of its simultaneous available wavelengths, because of intensity and adapted resolution for the energy ranges explored. The crystal has been cleaned and mounted at site. During the cooling procedure the crystal has been oriented using the sample rotation. For the experiment we have rotated the crystal around the c-axis covering a 60° section with 0.8° steps in three different sequences. First we measured the spin wave excitations at 4K with the incident energies: 10meV, 17meV, 33meV and 90meV. Subsequently a different set of incident energies was chosen (14meV, 24meV, 53meV and 199meV) in order to explore the excitations at even higher energies and intermediate energies. At last we measured the spin wave excitations at 80K using the first set of incident energies. Previous diffraction experiments show two different kinds of magnetic order, which are most prominent at the selected temperatures.
The investigated crystal had been preoriented along the c-axis during its growth process to a precision of about 2°, thus preliminary cuts taking with the Utsusemi software from the measured data show a nice ordered set of Bragg reflexes according to crystal symmetry (cf. Fig. 1a). Yet prior to an in depth analysis of the excitations the crystal has to be aligned more precisely within the software. Without this alignment, apart from slightly incorrect axes, cuts with a larger reciprocal volume to integrate over will only show blurred excitations. Such cuts are necessary to gather the available statistics especially for the data sets at higher incident energies. Therefore together with our local IT team we are working on a method to determine the crystal orientation from the gathered data.

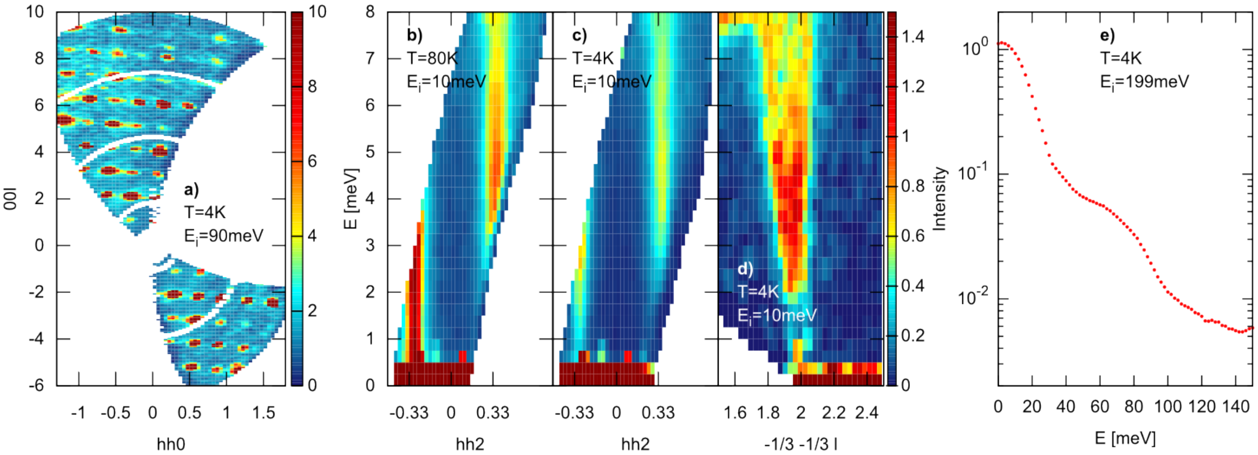

Fig. 1: Evaluation of selected measurement sets: a) shows a cut of the elastic scattering plane hhl containing nuclear and magnetic Bragg peaks; b) till c) are inelastic cuts along high symmetry directions of the magnetic Brillouin zone, b) and c) are cuts integrating over the whole excitation cone width, while d) is a thin cut through the cone center; e) displays the excitations spectra for an integration of the whole Q-space.

Nevertheless even in the current state the data reveals some very interesting and nice results. Using the Utsusemi software we have exported the data to a format, which can be evaluated using the software suite Horace developed at ISIS, as we are facing some troubles using a local installation of the Utsusemi software. The figures 1a) till 1d) are created from slim cuts in the S(Q,E)-space in order to minimize the effect of the misalignment.
Especially the measurements with an incident energy of 10meV show intriguing features, which have not been observed before due to the lack in resolution. In Fig. 1b) and c) a cut through the most prominent magnetic Bragg peaks -⅓ -⅓ 2 and ⅓ ⅓ 2 is displayed. Apparently an excitation gap arises between 80K and 4K of about 2meV. Furthermore a cut along the 001 direction through the magnetic Bragg peak -⅓ -⅓ 2 shows, that the excitations cones are not filled (cf. Fig 1d) meaning that the two magnon continuum for $CaBaCo_2Fe_2O_7$ is not as prominent as it has been observed e.g. on MgF_2. In order to determine the upper limit for the spin wave excitations the measurement with the highest incident energy is used. Here all Q-axes are integrated resulting in the plot in Fig. 1e). Basically two different structures can be observed: an area up to 40meV with high intensive excitations and a second one up to 90meV. Here due to the large Q-range also Phonon contributions will play a role. Yet with a corrected crystal orientation significantly better cuts can be created.

Finally we would like to thank the instrument responsibles for their great help and effort with preparing and conducting the experiment.