EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office via the User Portal:

https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do

Reports supporting requests for additional beam time

Reports can be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

Reports on experiments relating to long term projects

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF	**Experiment title:** Magnon-Phonon Coupling in Triangular Antiferromagnet alpha-CaCr2O4	**Experiment number**: HS-4532
Beamline: ID28	**Date of experiment**: from: 13.07.2011 to: 19.07.2011	**Date of report**: 28.02.2012 *Received at ESRF:*
Shifts: 18	**Local contact(s)**: Dr. Andrew Walters	

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Dr. Bella Lake

Dr. Andrew Colin Walters

Sándor Tóth

Report:

α-$CaCr_2O_4$ is a Heisenberg triangular lattice antiferromagnet with spin 3/2 magnetic Cr^{3+} ions located on stacked triangular layers. The spin moments order in a 120° magnetic structure at T_N=42K [1-2]. Inelastic neutron scattering below the magnetic ordering temperature showed large deviation from the classical spin-wave spectra of the spatially anisotropic stacked triangular lattice. Fitting the data showed that the deviations are due to the orthorhombic crystal symmetry, where there are four different nearest neighbour distances in the triangular planes and thus four inequivalent Heisenberg exchange interactions have to be taken into account. Moreover to explain the softening of spin wave modes at specific ***Q*** points: (1,1,1/2), (3,1,1/2), etc. further mechanisms have to be considered. Besides next-nearest neighbour inplane interactions, exchange striction would also cause the observed softening [3]. We measured inelastic X-ray scattering on ID28 to find evidence for the possible exchange-striction by observing the change in the phonon excitation spectra below and above the magnetic ordering. This technique enable us to distinguish the phononic excitations from the overlapping magnetic scattering by being insensitive to the latter.

We measured an α-$CaCr_2O_4$ single crystal sample oriented in the (H K 0) plane. The sample shape was a flat plate with thickness of 200 μm, the surface is perpendicular to the ***a***-axis. The area was 5x5 mm^2. An X-ray wavelength of 0.5226 Å selected by the Si(12,12,12) reflection of the monochromator was used. This wavelength is optimal for best resolution, which is 1.2 – 1.4 meV, according to calibration measurements on plexi glass. ***Q*** resolution perpendicular to the momentum transfer vector was $8\cdot 10^{-3}$ $Å^{-1}$ and $2\cdot 10^{-2}$ $Å^{-1}$ horizontally and vertically, parallel to the momentum transfer vector the resolution was much better. The sample temperature was controlled by a closed cycle refrigerator, which physically limited the available 2Θ angle.

The sample contains 3 twins, rotated by 60° and 120° along the ***a***-axis. The twin ratios were approximately determined by measuring a Laue pattern on ID28, using an area detector after the sample. On the selected beam spot, the twin ratio was 1:1:2. The sample was then accurately oriented on ID28 using nuclear Bragg reflections. During measurement the sample had to be repositioned after changing ***Q*** or temperature. The sample was measured at temperatures of 15 K, 50 K, 100 K, 200 K and 295 K.

At base temperature a few scans were performed along the (12.25,K,0) direction for direct comparison with neutron inelastic measurements along the equivalent direction of (4.25,K,0), see Fig. 1. Although the selection rules are the same for phonons and neutrons the nonmagnetic signal on the neutron scan around (4.25,0,0) differs from the X-ray scattering result, this may be because of the different relative scattering powers of Ca, Cr and O with X-rays and neutrons and the difference in ***Q*** value.

The temperature dependence of spectra were measured at several ***Q*** points as well. Two representative points are (4.25,0,0) and (4,4,-0.2), see Fig. 2. On all scans the strong incoherent signal was subtracted and the different temperature scans were corrected with the temperature dependent Bose factor. For the incoherent line subtraction at zero energy transfer the calibrated instrumental line shape was used. But due to the very strong signal, artifacts are visible on the corrected data around zero energy transfer.

The scan at (4.25,0,0) can be compared to the base temperature scan at ***Q***=(12.25,0,0) and one can see that the two ***Q*** points are equivalent. In Fig. 2.(a) the two room temperature scan was measured on two different points of the sample. The difference between the two scans is probably due to the different twin ratios in the two samples. Fig. 2(b) shows the (4,4,-0.2) scans, where the systematic temperature dependence is clearly visible. A large change of ~3meV is observed between 15K and 100K. This change can not attributed to the change of the stiffness of the crystal, because the typical phonon frequency variation due to thermal expansion are below 1 meV. To explain this change other effects have to be taken into account. One possibility is exchange striction which according to theory might also explain the softening of the magnons observed at low temperatures.Band structure calculations are currently taking place to model the phonon spectrum. To conclude, α-$CaCr_2O_4$ shows evidence of anomolous shifts in the phonon energies with temperature. However the data obtained from ID28 showed some inconsistencies because of the large sample, small beam size and variable twinning ratio across the sample surface. A measurement where the beam size was comparable or larger than the sample size would give more reliable results.

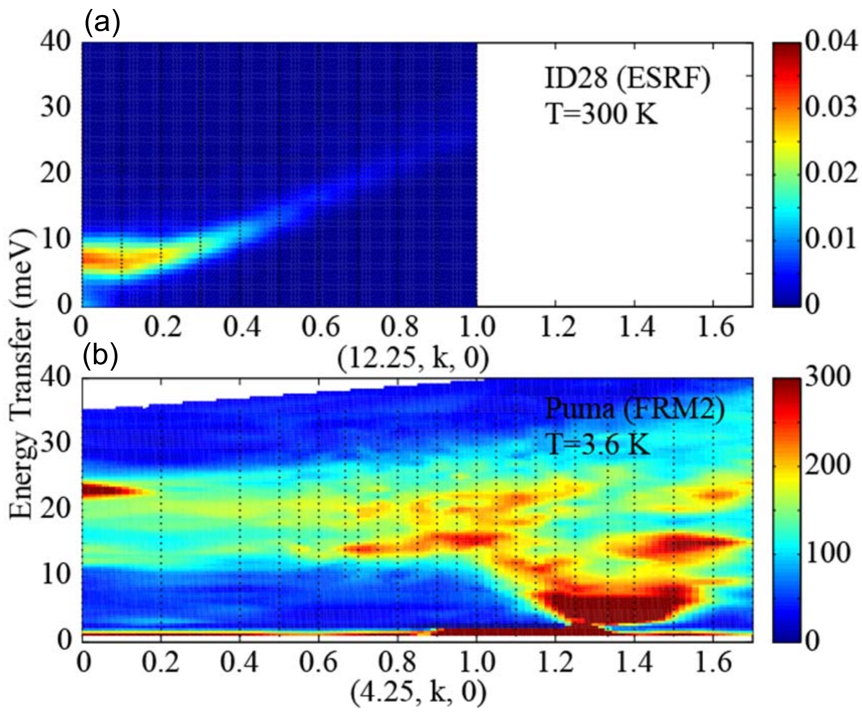

Figure 1: Comparison of inelastic X-ray measured on ID28 and previous neutron scattering result along equivalent directions.

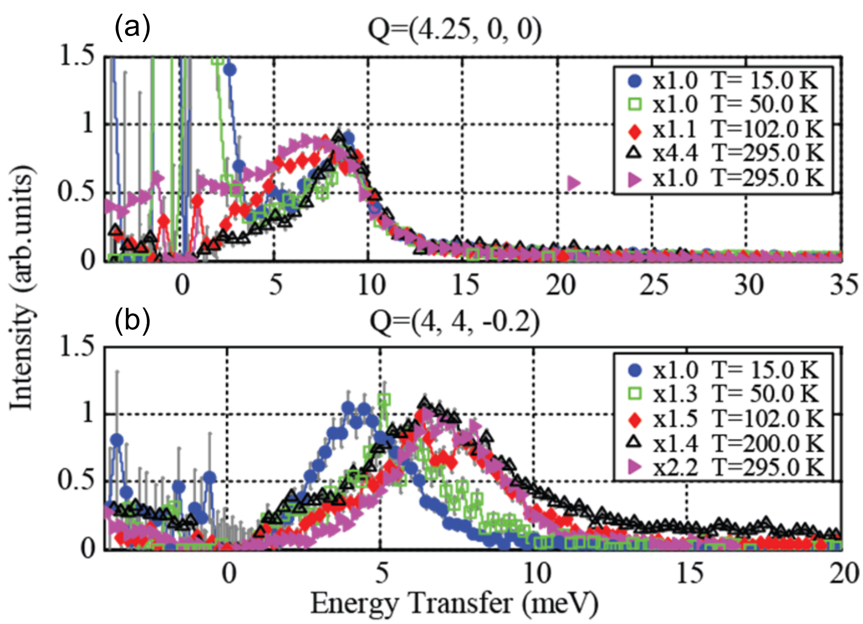

Figure 2: Phonon excitation intensities as a function of energy measured at different temperatures using ID28.

References

[1] L. C. Chapon, P. Manuel, F. Damay, P. Toledano, V. Hardy *et al.*, *Phys. Rev. B* **83**, 024409 (2011).
[2] S. Toth, B. Lake, S. A. J. Kimber, O. Pieper, M. Reehuis *et al.*, *Phys. Rev. B* **84**, 054452 (2011).
[3] Jung Hoon, K., & Jung Hoon, H., *Phys. Rev. B, 83*, 024409 (2011).