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:** RIXS study of the magnetically driven metal-insulator transition in $NaOsO_3$	**Experiment number**: **HC- 1527**
Beamline:	**Date of experiment**: from: 21st January 2015 to: 27th January 2015	**Date of report**: 23/09/2015
Shifts:	**Local contact(s)**: Marco Moretti	*Received at ESRF:*

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Report:

The metal insulator transition (MIT) is of enduring interest, underlying key principles of condensed matter physics. Many materials exhibit a Mott MIT, i.e. one driven by Coulomb interactions (U). Such a transition is purely electronic and is independent of magnetic correlations. In contrast, a “Slater transition” is one driven purely by magnetic correlations: the development of antiferromagnetic (AFM) order. The onset of Néel order induces a periodic potential on each neighbouring ion and opens up a continuous charge gap below the MIT. This MIT is also distinguished by an absence of any structural distortion through the MIT.

Osmates constitute the only realization of the Slater transition in three dimensions to date. Bulk measurements [1] revealed that the perovskite osmate $NaOsO_3$ undergoes an MIT concomitantly with antiferromagnetic order ($T_N = T_{MIT} = 410$ K). Neutron powder diffraction [2] revealed that this magnetic order was G-type, and optical conductivity measurements [3] demonstrated a zero temperature charge gap of 100 meV, which closes continuously towards the MIT. All of these observations are consistent with a Slater picture. However at the time of the experiment there had not been any determination of the magnetic or orbital excitations for $NaOsO_3$. Since T_N is coincident with the MIT, then it is likely that the electronic and magnetic correlations are intimately linked. The motivation was thus to determine the behaviour of the magnetic excitations.

The RIXS experiment was performed at the Os L_3 edge (10.871 keV) on a single crystal of $NaOsO_3$. This crystal had dimensions 0.3 x 0.3 x 0.3 mm^3 and was oriented such that the (1 0 1) direction was normal to the crystal face. Low resolution measurements were carried out using a Si (311) channel-cut secondary monochromator and Si (664) diced analyser, providing a total energy resolution of 275 meV. For high resolution measurements, a Si (664) backscattering channel cut secondary monochromator was used along with the aforementioned analyser, and total energy resolution of 46 meV.

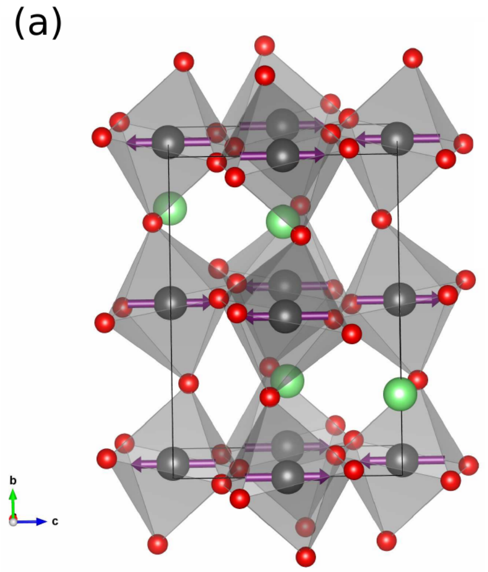

Figure 1: Crystal structure of $NaOsO_3$. The magnetic moments are oriented in the c-direction and coupled antiferromagnetically.

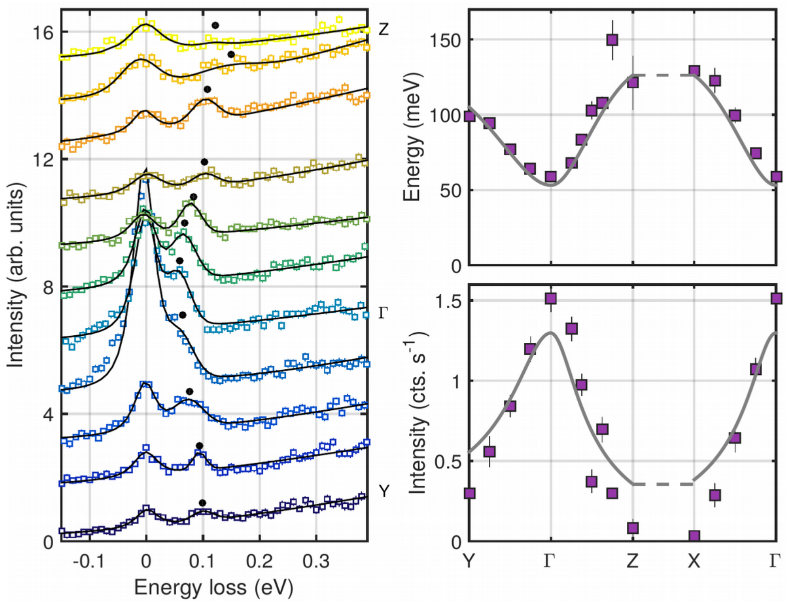

Figure 2: Left panel: Low energy RIXS spectra collected at 30 K in the (5 3 4) Brillouin zone. Right panel: Spin wave energy (top) and intensity (bottom) as a function of momentum transfer. Solid line is the result of a fit to a minimal J_1-J_2 Hamiltonian with single-ion anisotropy ($J_1 = J_2 = 14$ meV, $D = 4$ meV)

Low resolution measurements determined the choice of incident energy to use for the remainder of the experiment. These measurements revealed orbital excitations consistent with those collected previously at the APS, and hence the incident energy was selected as 10.880 keV, since this maximised the intensity of the intra-t_{2g} excitations.

The low energy RIXS spectra collected in the (5 3 4) Brillouin zone are displayed in Figure 2. Each spectrum was collected for four hours using the high-resolution setup. There is a clearly dispersing feature at approximately 100 meV, which appears to be gapped. Fitting the intensity and energy of this feature to a J_1-J_2 Hamiltonian plus some degree of single-ion anisotropy gave exchange parameters consistent with quantum chemistry calculations (unpublished), however the gap is somewhat larger than that predicted.

Unfortunately it was not possible to gain a full temperature dependence of the observed feature for two main the reasons. The first was that there were approximately three shifts lost due to accelerator problems. The second is an intrinsic weak signal which is distinct from the aforementioned accelerator issue. The poor signal-to-noise ratio (despite counting for four hours per spectrum) meant that data collection was limited. A similarly weak signal for the following experiment (HC-1534) may imply a transient beamline related problem. Preliminary measurements suggest that the spin gap may close continuously towards the MIT, however this is somewhat unclear.

Nevertheless these results show a distinct difference in the low energy excitations compared to the pyrochlore osmate $Cd_2Os_2O_7$, despite a similar local crystalline environment [4]. The well-defined spin wave excitations seen at low temperature for $NaOsO_3$ are a clear indication that a localized spin picture is appropriate for this material. Further measurements are proposed to look at the temperature dependence in more detail.

[1] Shi et al., Phys. Rev. B 80, 161104(R) (2009)
[2] Calder et al., Phys. Rev. Lett. 108, 257209 (2012)
[3] Lo Vecchio et al., Scientific Reports 3, 02990 (2013)
[4] Calder et al., arXiv:1508.01848 (2015)