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*

Deadlines for submission of Experimental Reports

Experimental reports must be submitted within the period of 3 months after the end of the experiment.

Experiment Report supporting a new proposal ("relevant report")

If you are submitting a proposal for a new project, or to continue a project for which you have previously been allocated beam time, you must submit a report on each of your previous measurement(s):

- even on those carried out close to the proposal submission deadline (it can be a *"preliminary report"),*
- even for experiments whose scientific area is different form the scientific area of the new proposal,
- carried out on CRG beamlines.

You must then register the report(s) as "relevant report(s)" in the new application form for beam time.

Deadlines for submitting a report supporting a new proposal

- 1st March Proposal Round - **5th March**
- 10th September Proposal Round - **13th September**

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.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report in English.
- include the experiment number 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.

	Experiment title: Magnetic Excitations across Topological Phase Transition in the Weyl Semimetal Mn3Sn	**Experiment number**: HC-4421
Beamline: ID32	**Date of experiment**: from: 20/4/2021 to: 26/4/2021	**Date of report**: 10/9/2021 *Received at ESRF:*
Shifts: 18	**Local contact(s)**: Davide Betto	

Names and affiliations of applicants (* indicates experimentalists):

Hakuto Suzuki*, Matteo Minola*, Zichen Yang*, Bernhard Keimer- Max Planck Institute for Solid State Research

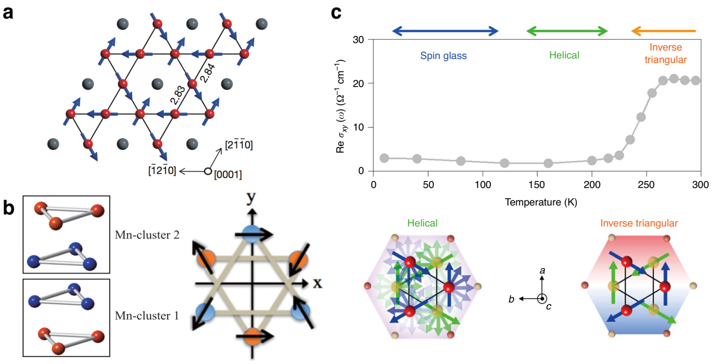

Figure 1 (a) Inverse triangular antiferromagnetic order of a Weyl semimetal Mn_3Sn. (b) Cluster octupole moments responsible for the Berry curvature. (c) Terahertz anomalous Hall effect in in the helical and inverse triangular spin structures.

Report:

Mn_3Sn is a magnetic Weyl semimetal exhibiting a large anomalous Hall effect without net magnetization at room temperature, opening the possibility for spintronic memory without stray fields. These fascinating properties emerge from the nonzero Berry curvature in the inverse triangular antiferromagnetic structure on the Kagome lattice of Mn atoms (Fig. 1a). The nonzero Berry curvature is attributed to the existence of local cluster multipole moments (Fig. 1b), a generalization of the uniform magnetization in ferromagnets. This notion has been supported by the disappearance of terahertz anomalous Hall effect upon entering the helical magnetic state (Fig. 1c). The goal of the present proposal was therefore to understand the evolution of collective spin excitations across the topological phase transition by utilizing high-resolution soft-x-ray RIXS at the Mn L_3 edge. Since no resonant x-ray magnteic diffraction study has been reported on this compound, we also tried to observe the out-of-plane magnetic Bragg peak from the helical magnetic state utilizing the photodiode installed in the chamber.

Single crystals of Mn_3Sn (1 x 1 x 0.1 mm^3 in size) were grown by the Bridgeman method and pre-aligned with Laue diffractometer. To maximize the momentum transfer from the x-rays at the Mn L_3 absorption edge (~640 eV), we fixed the scattering angle 2θ at 150 degrees. We first set the incident polarization to σ polarization and searched for Bragg peaks. Due to the small lattice parameters in Mn_3Sn and geometrical constraints only one lattice Bragg peak was accessible with hν < 2 keV in ID32. We first found a lattice Bragg peak (101) with $h\nu = 1950$ eV and searched for the magnetic Bragg peak of the helical order at 200 K with $h\nu = 640$ eV. A neutron scattering study in the early days [4] reported helical ordering vector of ~(1,0,0.09) and ~(1,0,0.07), but we did not detect the magnetic Bragg peak around (0,0,0.07) and (0,0,0.09). This is possibly because of small intensity of these peaks and/or because of different ordering vector in our Mn_3Sn sample, as the magnetism of Mn_3Sn is sensitive to the growth conditions and the amount of excess Mn.

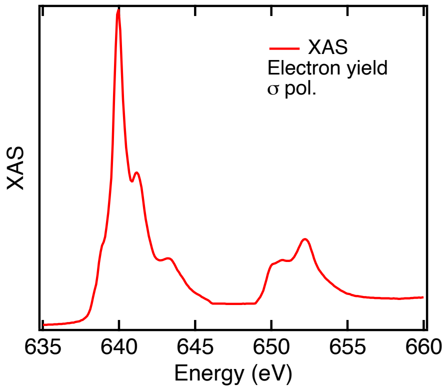

Figure 2 Electron-yield XAS of Mn_3Sn.

Therefore we moved on to RIXS measurement of the magnetic excitations in the inverse triangular state at room temperature, which has been well characterized by complementary neutron scattering studies. Figure 2 shows the electron-yield XAS spectrum of Mn_3Sn, which dominantly reflects the electronic states close to the surface. Mn_3Sn is a metallic alloy and therefore the shoulder structures seen below and above the main L_3 peak are most likely due to oxide particles unavoidable at the surface (consistent with complementary Raman scattering studies). Nevertheless, the RIXS spectra show broad features typical of metallic systems reflecting the bulk electronic states of Mn_3Sn (see below).

After the tuning of beamline conditions, the total energy resolution of 24 meV (in FWHM) was achieved, which enabled us to detect low-energy magnetic excitations. Within the allocated beamtime, we have collected RIXS data with σ polarization for $q = (H, 0)$ and (H, H) paths at 300 K, and with π polarization for $q = (H, 0)$ and (H, H) paths at 300 K and 200 K. We spent more time for the π polarization as it suppresses the charge elastic scattering and made the inelastic signal more visible. As a representative set of experimental data, we show in Figure 3 the low-energy RIXS spectra taken at 300 K with π polarization for the $q = (H, 0)$ path (upper curves correspond to the low H values). The observed quasi-elastic peaks have asymmetric lineshape with broad tails in the energy loss side, which can be ascribed to magnetic excitations. Furthermore, the tails show momentum dependence with minima close to the Γ point (middle curves in Fig.3), which is consistent with $\boldsymbol{q} = 0$ ordering vector of inverse triangular magnetic structure of Mn_3Sn. We aim to carefully decompose the quasielastic peaks into the elastic peaks and magnetic excitations, to reveal the dispersion relation of the magnetic excitations. Note here that visual inspection of the top and bottom curves (corresponding to (-0.45,0) and (0.45, 0), respectively) yields the small bandwidth of ~20 meV, highlighting the high resolving power of the ERIXS spectrometer.

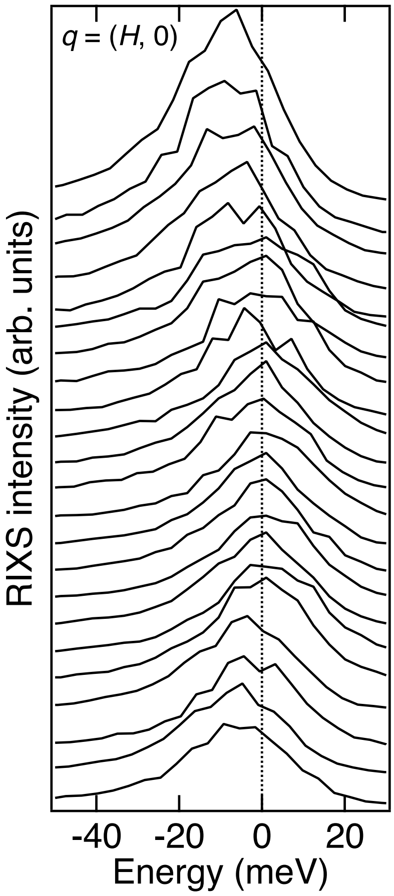

Figure 3 RIXS spectra of Mn_3Sn at room temperature along the $q = (H, 0)$ path.

To summarize, despite the small bandwidth of magnetic excitations and broad lineshape of the metallic Mn_3Sn, the experiment has been successfully completed thanks to the ultrahigh resolution of ERIXS around the Mn L_3 edge. Detailed data analysis for the extraction of magnon dispersion is under way. In particular, the differentiation of magnon dispersion taken with 300 and 200 K will give us insight into the two topologically distinct magnetic phases, which will be the main focus of the forthcoming publication. As mentioned in the proposal, we also aim to perform a further RIXS experiment under uniaxial stress, as the strain tuning of magnetism has proven to be effective [5]. The expertise of MPI-Stuttgart on uniaxial strain experiments accumulated through former works on cuprate superconductors [6] will be crucial in planning future measurements.

References

[1] S. Nakatsuji, N. Kiyohara, and T. Higo. “Large anomalous Hall effect in a non-collinear antiferromagnet at room temperature”, Nature 527, 212 – 215 (2015).

[2] M.-T. Suzuki, R. Arita et al., “Cluster multipole theory for anomalous Hall effect in antiferromagnets”, Phys. Rev. B. 95 094406 (2017)

[3] T. Matsuda, R. Matsunaga et al., “Room-temperature terahertz anomalous Hall effect in Weyl antiferromagnet Mn_3Sn thin films”, Nat. Commun. 11 909 (2020).

[4] J. Cable et al., “A neutron study of the magnetic structure of Mn_3Sn”, Solid State Commun. **88** 161 (1993)

[5] M. Ikhlas, S. Nakatsuji, C. Hicks et al., Appl. Phys. Lett. **117** 233502 (2020)

[6] H.-H. Kim, M. Le Tacon et al., Phys. Rev. Lett. **126** 037002 (2021)