

# Experimental report

13/09/2023

**Proposal:** 4-02-626

**Council:** 4/2023

**Title:** Magnetic excitations in layered ferromagnetic Sr<sub>4</sub>Ru<sub>3</sub>O<sub>10</sub>

**Research area:** Physics

**This proposal is a resubmission of 4-01-1764**

**Main proposer:** Zahrasadat GHAZINEZHAD

**Experimental team:** Zahrasadat GHAZINEZHAD

**Local contacts:** Paul STEFFENS

**Samples:** Sr<sub>4</sub>Ru<sub>3</sub>O<sub>10</sub>

Instrument	Requested days	Allocated days	From	To
THALES	4	4	07/09/2023	11/09/2023

## Abstract:

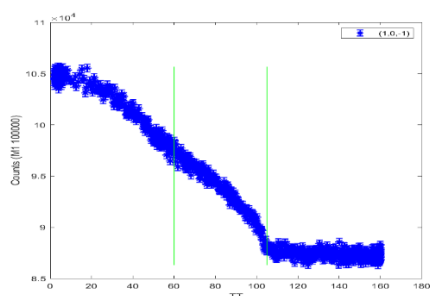
We propose to study the temperature dependence of the magnon gap in the triple-layer ruthenate Sr<sub>4</sub>Ru<sub>3</sub>O<sub>10</sub>. The material is closely related to ferromagnetic SrRuO<sub>3</sub> for which we established a characteristic impact of Weyl points on the spin dynamics. Similar effects are expected in Sr<sub>4</sub>Ru<sub>3</sub>O<sub>10</sub>. The better understanding of the electronic structure in Sr<sub>4</sub>Ru<sub>3</sub>O<sub>10</sub> and its smaller structural distortion will facilitate both the experimental and theoretical analyses. A large crystal could recently be grown that will allow time efficient measurements. In the last round beam-time on IN20 was accorded but the precise temperature dependence of the gap can only be determined on a cold TAS.

## Experimental Report

<b>Instrument</b>	THALES
<b>Proposal Number</b>	4-02-626
<b>Proposal</b>	Magnetic excitations in layered ferromagnetic Sr <sub>4</sub> Ru <sub>3</sub> O <sub>10</sub>
<b>Experimentalist</b>	Zahrasadat Ghazinezhad, Yvan Sidis, Markus Braden
<b>Local Contact</b>	Paul Steffens

The aim of the experiment was to study the temperature dependence of the magnon gap in the triple-layer-ruthenate Sr<sub>4</sub>Ru<sub>3</sub>O<sub>10</sub>. The material is closely related to ferromagnetic SrRuO<sub>3</sub>, for which we established a characteristic impact of Weyl points on the spin dynamics [1]. Similar effects can be expected in Sr<sub>4</sub>Ru<sub>3</sub>O<sub>10</sub>. The better understanding of the electronic structure in Sr<sub>4</sub>Ru<sub>3</sub>O<sub>10</sub>, its smaller structural distortion and in particular its layered structure should considerably facilitate both the experimental and the theoretical analyses.

Sr<sub>4</sub>Ru<sub>3</sub>O<sub>10</sub> is the triple-layer member of the Ruddlesden-Popper series of ruthenates [2], it exhibits ferromagnetic order at ambient pressure and zero field [2,3] below 105 K, but its ordered phase is split. Below T\*~60K another phase occurs that shows a double metamagnetic transition for applying in-plane magnetic fields [2-4]. The rough explanation of this behavior is given by a reorientation of the ordered moments, that rotate from an in-plane direction to perpendicular to the layers at low temperature [5]. The electronic structure [6,7] is essentially two-dimensional in nature and it bears strong resemblances with that of single-layered Sr<sub>2</sub>RuO<sub>4</sub>, in particular evidence for nesting. In a previous experiment on this material on the thermal TAS IN20 we studied the magnon dispersion as a function of temperature at higher energies, but the magnon gap could not be resolved with achievable energy resolution on this instrument, see report 4-01-1764.

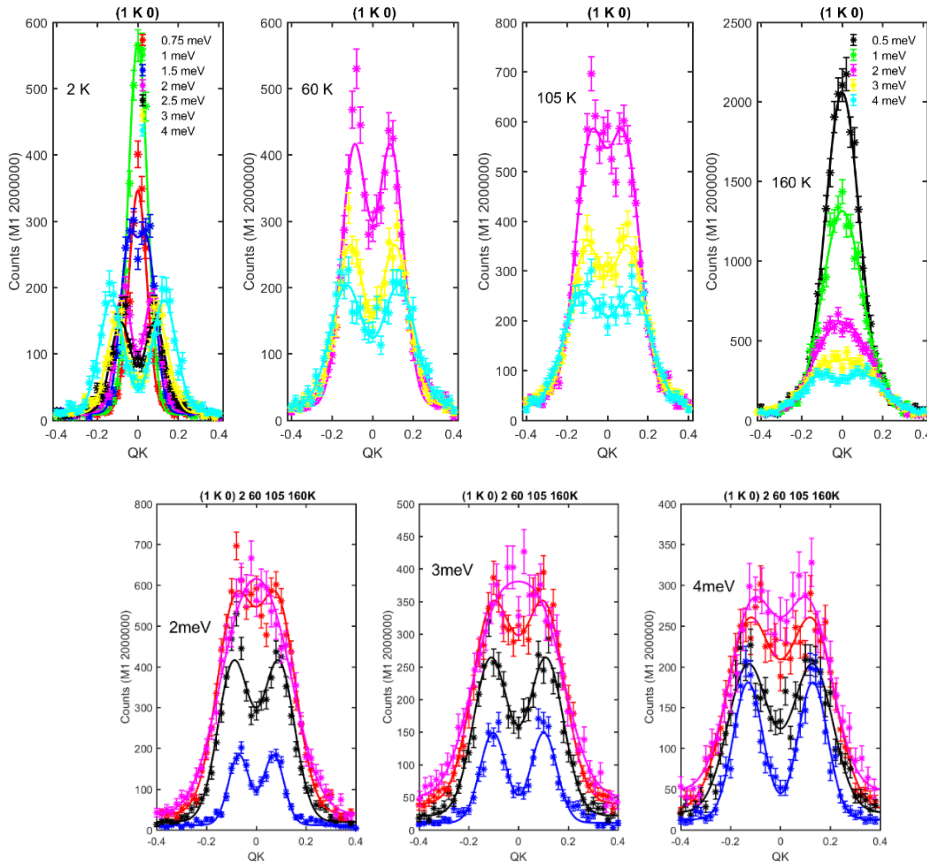


**Fig. 1** Temperature dependence of the Bragg intensity at (1 0 - 1) recorded upon cooling. This Bragg reflection exhibits nuclear and magnetic contributions; therefore, it does not vanish in the paramagnetic phase. Additional magnetic intensity appears at the onset of ferromagnetic ordering at 105 K, but also the reorientation transition is visible in these data.

The Thales experiment was performed with the same crystal of Sr<sub>4</sub>Ru<sub>3</sub>O<sub>10</sub> with a volume of about 850mm<sup>3</sup> that was studied on IN20. A single cylindrical piece was used in which the c direction is nearly parallel to the growth direction. The sample was mounted in [100]/[010] geometry. According to the literature [2], Sr<sub>4</sub>Ru<sub>3</sub>O<sub>10</sub> crystallizes in the orthorhombic space group *Pbam* which results from the ideal space group *I4/mmm* (*a*=3.90 and *c*=28.6 Å) due to the rotation of octahedrons around *c*. Here we always refer to the non-distorted lattice with the short *a* lattice constant. PG (002) crystals were used both as monochromator and as analyzer. In order to suppress higher-order contaminations we used a velocity selector in the incoming beam and a cooled Be filter in front of the analyzer. Scans were performed with fixed final neutron momentum, *k<sub>f</sub>*=1.5, 1.35 or 1.1 Å<sup>-1</sup> with the smaller values considerably enhancing the resolution (from 136 to 45μeV FWHM at the elastic line). Overall, we can state that this experiment was quite productive yielding high statistics within reasonable time, which was mandatory to apply the high-resolution conditions. The vertical focusing on this layered system is quite efficient.

With the small neutron momenta on a cold instrument only a few Bragg peaks can be reached in the used scattering geometry. Note that (1 0 0) actually is not a zone center as it is incompatible with body centering. Therefore, there is neither structural nor magnetic intensity arising from the 3-dimensional

magnetic order at this Q value. The nearest Bragg point is  $(1\ 0\ \pm 1)$  which could be reached in our instrument by tilting the cryostat, see Fig. 1. The recorded intensities show the two transition temperatures.



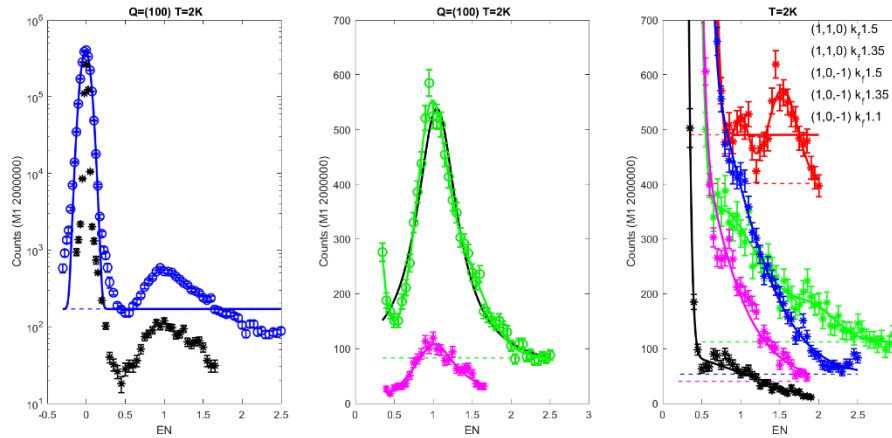
**Fig. 2:** Constant energy scans across the magnon dispersion recorded at  $\mathbf{Q}=(1\ 0\ 0)$  in transversal direction for various energies and four different temperatures. In the upper panels we compare data taken at fixed temperature, and the lower panels superpose data taken at different temperatures.

Fig. 2 displays constant energy scans recorded in the transversal direction at  $\mathbf{Q}=(1\ 0\ 0)$ . There is a strong low-energy magnon response near this Bragg point, because the inter-layer magnetic interaction is rather small. Studying  $\mathbf{Q}=(1\ 0\ 0)$  compared to  $(1\ 0\ 1)$  or  $(1\ 1\ 0)$  is very advantageous, because there is neither a strong tail from the nuclear Bragg peak nor a low-energy phonon contribution. And compared to  $(1\ 1\ 0)$  the form factor of the Ru yields a much stronger signal. These constant-energy data nicely fit to those taken at higher energies in the previous experiment on IN20. Again, an anomalous temperature dependence becomes visible, as the dispersion tends to become even stiffer at higher temperature, but the detailed temperature dependence seems to be even more subtle than in the similar experiments on  $\text{SrRuO}_3$  [1].

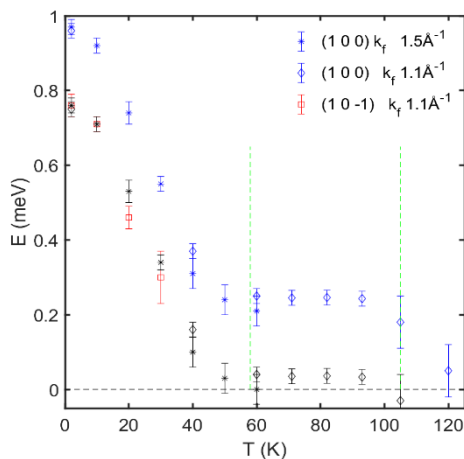
Also, the measurements of the magnetic gap performed at the  $(1\ 0\ 0)$  scattering vector yielded high statistics within little time, see Fig. 2. Note that  $(1\ 0\ 0)$  corresponds to the zone boundary and thus to the end of the  $c$ -axis dispersion of the magnon. Since there is only little interlayer interaction, the difference between measurements at  $(1\ 0\ 0)$  and a 3-dimensional Bragg peak is small, but as we could clearly establish, it is finite. The  $c$ -axis dispersion amount to  $\sim 0.2\text{meV}$ . The gap at  $(1\ 0\ 0)$  could be easily followed upon heating up to  $\sim 50\text{K}$  with  $k_f=1.5\ \text{\AA}^{-1}$ , but the measurements at the true Bragg points,  $(1\ 0\ -1)$  turned out to be more favorable. Note that the features observed at the very strong Bragg reflection  $(1\ 1\ 0)$  are most likely artifacts arising from the Bragg scattering.

By combining scans with different values of  $k_f$  at  $(1\ 0\ 0)$  and  $(1\ 0\ -1)$  we could establish the  $c$ -axis dispersion of the magnetic excitations and the temperature dependencies of the magnon gap at the

zone center and at the zone boundary. The black symbols denote the temperature dependence of the magnon gap when assuming a temperature independent  $c$ -axis dispersion. Comparing the temperature dependence of the magnon gap with that of the magnetization (see also Fig. 1) shows fundamentally different behavior contradicting the simple expectation. Our results can explain the character of the second magnetic transition. In the upper ordered magnetic phase, the anisotropy gap is tiny and thus irrelevant already in small magnetic fields. But below  $T^*$  the magnetic anisotropy increases reaching a low-temperature value comparable to that in  $\text{SrRuO}_3$ . Magnetic anisotropy seems thus to drive this transition.



**Fig. 3:** Constant  $Q$  scans across the magnon response recorded at  $Q=(1\ 0\ 0)$ ,  $(1\ 1\ 0)$  and  $(1\ 0\ -1)$  with different values of the final neutron momentum,  $k_f=1.5$ ,  $1.35$  or  $1.1\ \text{\AA}^{-1}$ . While the magnetic gap at  $(1\ 0\ 0)$  can be easily resolved with  $k_f=1.5\ \text{\AA}^{-1}$  at low temperature, the much stronger (at least two orders of magnitude) elastic signals at the 3-dimensional zone centers require an enhanced resolution. The measurements with the smaller  $k_f$  show always lower intensity. In the right panel red and blue correspond to data taken at  $(1\ 1\ 0)$ , and green, magenta and black to data at  $(1\ 0\ -1)$ , respectively.



**Fig. 4:** Temperature dependence of the magnon gaps at  $(1\ 0\ 0)$  and  $(1\ 0\ -1)$  extracted from scans with different  $k_f$  values. The black data correspond to the zone-boundary data reduced by the finite  $c$ -axis dispersion.

**References** [1] K. Jenni et al., Phys. Rev. Lett. **123**, 017202 (2019); *ibid.*, Phys. Rev. B Letter **105**, L180408 (2022); *ibid.* Phys. Rev. B **107**, 174429 (2023). K. Jenni dissertation Univ. Cologne 2021 <https://kups.ub.uni-koeln.de/55061/>. [2] M. K. Crawford et al., Phys. Rev. B **65**, 214412 (2002); G. Cao et al., Phys. Rev. B **68**, 174409 (2003). [3] F. Forte et al. B **100**, 104440 (2019); V. Granata et al. Phys. Rev. B **93**, 115128 (2016); *ibid.* J. Phys.: Condens. Matter **25** (2013) 056004. [4] W. Schottenhammel et al., Phys. Rev. B **94**, 155154 (2016). [5] M. Zhu et al., Scientific Reports **8**, 3914 (2018). [6] G. Gebreyesus et al., Phys. Rev. B **105**, 165119 (2022). [7] P. Ngabonziza et al., Scientific Reports **10**, 21062 (2020). [8] Jiajie Wan et al., Front. in Mater. **9**, 856000 (2022); Yan Liu et al., New J. of Phys. **18**, 053019 (2016).