Experimental report

Proposal:	4-01-1764
Title:	Magnetic excitations in layered ferromagnetic Sr4Ru3O10
Research area:	Physics

This proposal is a new proposal

Council:	10/2022

Main proposer:	**Zahrasadat GHAZINEZHAD**
Experimental team:	Markus BRADEN
	Zahrasadat GHAZINEZHAD
	Yvan SIDIS
Local contacts:	Paul STEFFENS
	Mechthild ENDERLE
	Ursula Bengaard HANSEN

Samples: Sr4Ru3O10

Instrument	Requested days	Allocated days	From	To
IN8	8	0		
THALES	5	0		
IN20	8	4	13/04/2023	19/04/2023

Abstract:

We propose to study magnetic excitations in the triple-layer compound Sr4Ru3O10. The material is closely related to ferromagnetic SrRuO3 for which an impact of Weyl points on the spin dynamics could be established. Similar effects are expected in Sr4Ru3O10. In addition the understanding of the electronic structure is deeper in Sr4Ru3O10 and the presence of nested bands should give rise to magnetic excitations. Also for analysing the magnon width and in view of a complete picture this compound is highly advantageous. A large crystal could recently be grown that will allow time efficient measurements.

Instrument	IN20
Proposal Number	4-01-1764
Proposal	Magnetic excitations in layered ferromagnetic $Sr_4Ru_3O_{10}$
Experimentalist	Zahrasadat Ghazinezhad, Markus Braden
Local Contact	Ursula Bengaard Hansen, Paul Steffens

The aim of the experiment was to study the magnetic excitations in the triple-layer-ruthenate $Sr_4Ru_3O_{10}$. The material is bridging the unconventional superconductor Sr_2RuO_4 with ferromagnetic $SrRuO_3$, it is the simplest layered ruthenate that exhibits ferromagnetic order at ambient pressure and zero field. In the single-layered superconductor both incommensurate AFM and quasi-FM fluctuations exist and compete, while the perovskite only shows ferromagnetic correlations, for which we established a characteristic impact of Weyl points [1]. Both the magnon gap and the magnon stiffness exhibit an anomalous temperature dependence that clearly does not follow that of the magnetization. Similar effects can be expected in $Sr_4Ru_3O_{10}$. The better understanding of the electronic structure in $Sr_4Ru_3O_{10}$, its smaller structural distortion and in particular its layered structure should considerably facilitate both the experimental and the theoretical analyses.

$Sr_4Ru_3O_{10}$ 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 $T^* \sim 60K$ 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_2RuO_4, in particular evidence for nesting. Furthermore, there is a strong anomalous Hall effect [8].
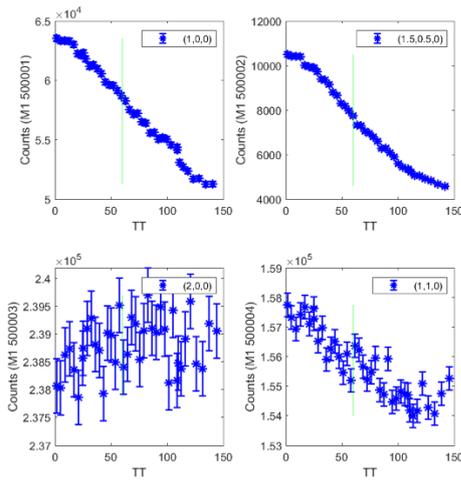

Fig. 1 Temperature dependence of the Bragg intensity at (1 0 0), (1.5 0.5 0), (2 0 0) and (1 1 0) recorded upon cooling. (1 0 0) breaks the body centering and does not sense the FM ordering but only some parts via stacking faults. (1.5 0.5 0) senses the structural distortion, i.e., the rotation of the octahedra. One recognizes anomalies at both magnetic transitions. At the true 3-dimensional Bragg point (1 1 0) the inset of the magnetic order and the reorientation are hardly visible due to the strong nuclear contribution. At (2 0 0) no magnetic intensity can be identified.

The experiment was performed with a large crystal of $Sr_4Ru_3O_{10}$ (volume of about 850mm^3) that was grown with the floating-zone technique in a mirror furnace at Cologne university. 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], $Sr_4Ru_3O_{10}$ 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 PG filter in front of the analyzer. Scans were mostly performed with fixed final neutron momentum k_f=2.662Å^{-1}. Overall, we can state that this experiment was quite productive yielding high

statistics within reasonable time, which was important to study temperature effects in quite limited time. The vertical focusing operates most efficiently in this layered system.

Temperature dependencies of selected Bragg intensities are shown in Fig. 1. There is a clear signature of the FM ordering at (1 0 0) although this Q values corresponds to a zone boundary in the body-centered lattice. However, the intensity uptake at the nuclear Bragg points (1 1 0) and (2 0 0) is limited. In contrast the superstructure reflections (1.5 0.5 0) that does not sense the FM order clearly increases in the FM phase indicating an enhancement of the rotation angle.
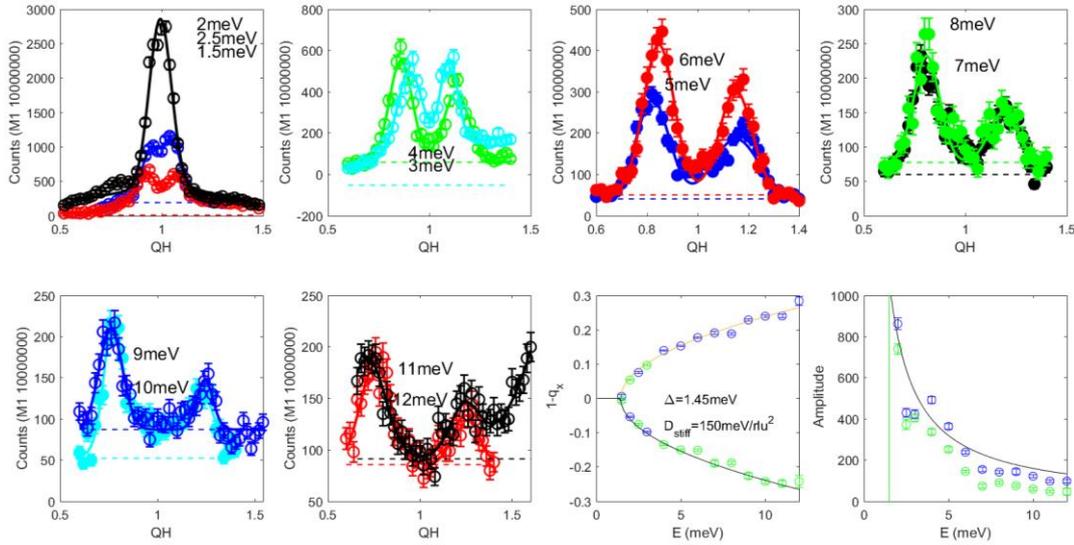

Fig. 2: Constant-E scans across the magnon dispersion recorded at **Q**=(1 0 0) in longitudinal direction for various energies. Data were fitted by two gaussian distributions yielding a quadratic dispersion and a rough estimate of the gap. The amplitudes of the signal rapidly drop with increasing energy.

Fig. 2 displays constant energy scans recorded in the longitudinal direction at **Q**=(1 0 0). The magnon dispersion could be easily followed up to 12 meV. However, the drop of intensity with increasing energy and the perturbation by phonon scattering render studies at higher energies very challenging. Only with considerable efforts data above 12 meV can be obtained on a TAS instrument. As we needed to study also the detailed temperature dependency we had to refrain from such studies, which are better place on a TOF instrument.

The (1 0 0) scattering vector is most favorable for studying the low-energy (up to 12meV) magnons, because it does not exhibit scattering from acoustic phonons. However, one has to keep in mind that the *c*-axis dispersion is fully neglected. Due to the layered character of the nuclear and electronic structure approach is well justified. The magnon gap at (1 0 0) could not be determined on IN20 at it is of the order of 1 meV.

We also took data in the transversal direction at (1 0 0) and along [110] direction. The latter data were analyzed analogously and are shown in Fig. 3. The temperature dependence of the magnon dispersion was studied by recording sets of scans at six temperatures between 1.6 and 160 K. Part of these data are shown in Fig. 4. Similar to the observation in $SrRuO_3$ [1] the temperature dependence is clearly anomalous, as at considerably higher temperature the dispersion even gets stiffer, while one would expect softening in a *normal* system.

The ARPES and DFT analyzes of the electronic structure of $Sr_4Ru_3O_{10}$ indicate strong resemblance with that of the single-layered superconductor [6,7]. In particular one can find similar evidence for electronic nesting. Therefore, we searched for other magnetic correlations besides the ferromagnetic magnon response. In Fig. 5 we show a map of the scattered intensity with an energy transfer of 5 meV.

There is no indication for any additional signal besides the ring of magnon scattering around (100). Any such incommensurate contribution must be considerably smaller or exhibit a different energy scale, which, however, would be surprising. However, the ring of the magnon scattering seems not to be fully isotropic but exhibits enhanced signal strength along the pseudo tetragonal directions. This may suggest a more complex impact of the electronic structure.
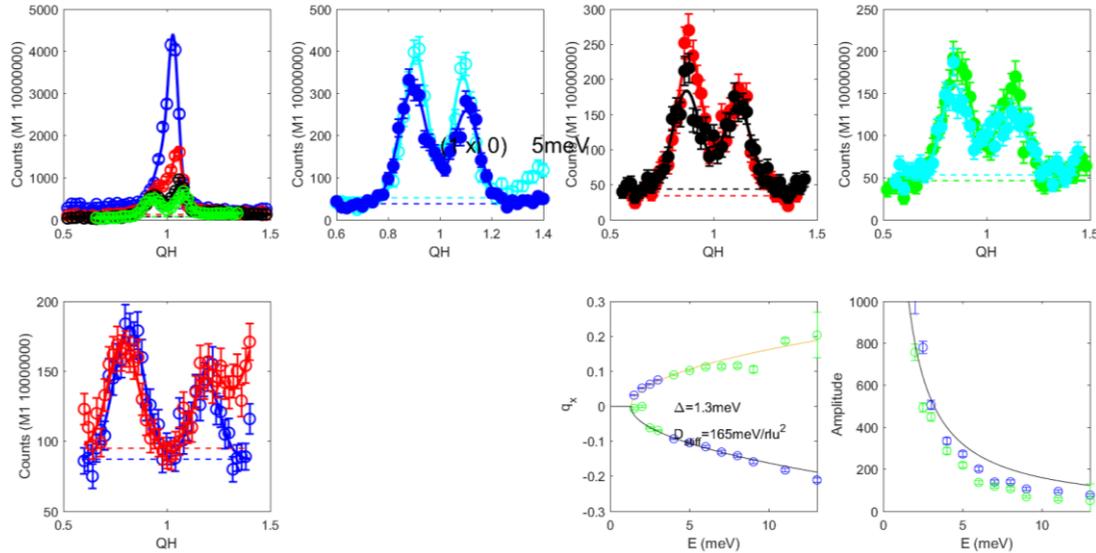

Fig. 3: Constant energy scans across the magnon dispersion recorded at **Q**=(1 0 0) in [110] direction for various energies. The data were fitted by two gaussian distributions yielding a quadratic dispersion and a rough estimate of the gap similar to the analysis in Fig. 2.
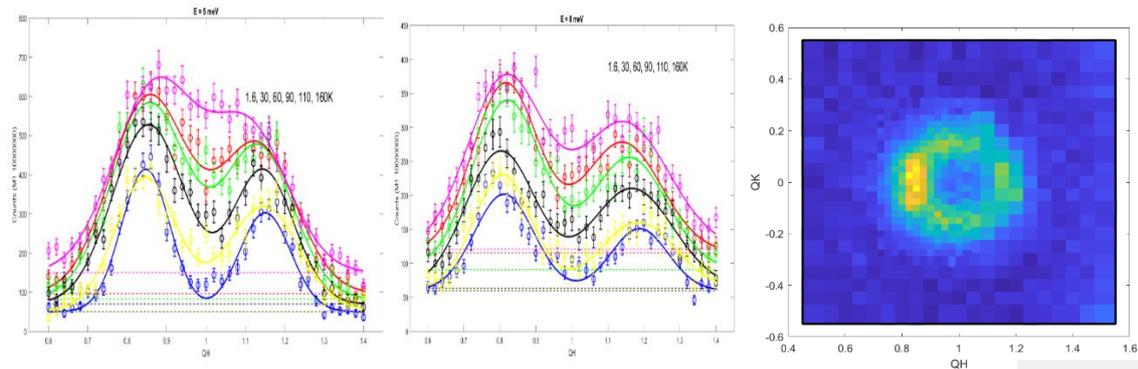

Fig. 4: Temperature dependence of the magnon scattering at 5 and 8 meV. One clearly recognizes an anomalous stiffening at larger temperature, while a simple ferromagnet should exhibit softening. Furthermore, the 8 meV scan at 160 K, i.e. well above the ferromagnetic transition, clearly exhibits a double peak structure indicating that ferromagnetic correlations persist in this itinerant system. **Right panel:** Map of scattered intensity for an energy transfer of 5 meV. Besides the ring of magnon signals no significant feature can be found. However, the magnon scattering seems to be slightly anisotropic.

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).