

Experimental report

27/11/2023

Proposal: CRG-2914

Council: 10/2022

Title: Temperature dependence of magnons and phonons in untwinned SrRuO₃

Research area: Physics

This proposal is a new proposal

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Samples: SrRuO₃

Instrument	Requested days	Allocated days	From	To
ORIENTEXPRESS	1	1		
IN22	6	6	17/05/2023	23/05/2023

Abstract:

SrRuO₃ is a fascinating material that exhibits close coupling between charge, lattice and magnetism. Previous inelastic neutron scattering experiments suffered from the twinning associated with the orthorhombic distortion. We propose to detwin large SrRuO₃ crystals by applying vertical magnetic fields along a pseudocubic [011] direction. We then wish to determine the intrinsic width of magnon as function of temperature. In addition we wish to deepen the analysis of the phonon dispersion towards the bond-bending and rotational modes which strongly suffer from the superposition of different modes in a twinned crystal.

Instrument	IN22
Proposal Number	CRG-2914
Proposal	Temperature dependence of magnons and phonons in untwinned SrRuO ₃
Experimentalist	Zahrasadat Ghazinezhad, Markus Braden
Local Contact	Wolfgang F SCHMIDT, Paul Steffens

This experiment aimed to study the magnetic excitations in the infinite-layer-ruthenate SrRuO₃ after detwinning by applying magnetic field. SrRuO₃ is the infinite layered ruthenate that exhibits ferromagnetic order at ambient pressure and zero field with the order moment of 1.6 μ B and T_c=165K [1, 2]. The relation between the anomalous Hall effect and Berry phase due to the Weyl point has been evidenced for SrRuO₃ and by now SrRuO₃ is known as a FM Weyl semimetal [3]. The observed anomalous temperature dependences of the magnon gap and magnon stiffness in SrRuO₃ both follow the AHE but not the magnetization [4, 5]. A better understanding of the magnon dispersion in SrRuO₃ is possible when detwinning the crystals by applying the vertical magnetic field. The IN22 experiment was performed with six single cylindrical crystals that were grown with the floating-zone technique in a mirror furnace at Cologne University and co-aligned together for the experiment. The samples were co-aligned and mounted in [100]/[011] pseudo cubic notation (Fig. 4). SrRuO₃ deviates from the cubic perovskite structure due to the rotation and tilting of RuO₆ Octahedra and reduces the symmetry to orthorhombic space group Pnma [1]. In this experiment, we always refer to a pseudo-cubic system (Pm-3m, a=3.9Å) since the Ru position still follows the cubic symmetry. Due to the two structural phase transitions, these crystals show a structural multidomain arrangement following the six different orthorhombic twins [6]. Therefore, we detwinned the SrRuO₃ crystals by applying vertical magnetic fields of 6T along a pseudocubic [011] direction, which can correspond to orthorhombic *c*, the easy axis of this material. PG(002) crystals were used as a monochromator and analyser. Scans were mostly performed with fixed final neutron momentum $K_f = 2.662 \text{ \AA}^{-1}$. To see whether the detwinning by applying the magnetic field was successful, we checked the superstructure peak at each temperature, which shows the reliable suppression of the peaks. In Fig. 1 the integrated intensity of the (1.5 0.5 0.5) superstructure peak versus temperature and magnetic field is shown. Starting from zero field at 220 K the intensity of this reflection could be almost completely suppressed documenting the nearly monodomain state we could induce.

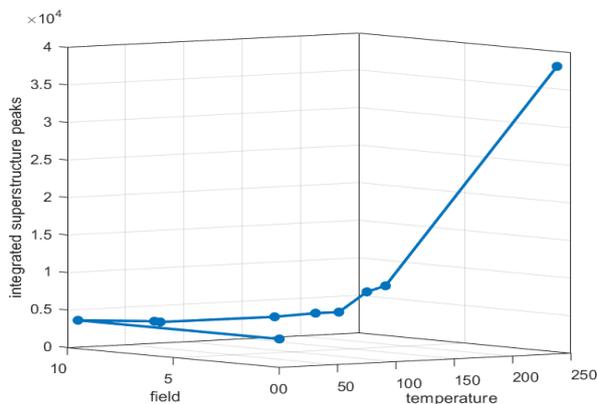


Fig. 1: Integrated superstructure peak intensities at different temperatures and magnetic fields. Suppression of the intensity shows that the detwinning process remains efficient during the measurement.

In overall, we can declare that this experiment was successful and productive in collecting data with high statistics in a reasonable time. We were able to detwin the crystals by applying the 6T magnetic field and kept this detwinning during the experiment (Fig. 1). The magnetic

detwinning let us quantitatively analyse the magnon gap and stiffness in different temperatures and improve our understanding of the acoustic phonon dispersion behaviour in this single crystal. Constant energy scans across the magnon dispersion at $Q=(1\ 0\ 0)$ in diagonal and longitudinal directions for different energies have been measured and the results in diagonal direction are depicted in Fig.2. The $(1\ 0\ 0)$ scattering vector is the most favourable for studying the low-energy (up to 12meV) magnons since there is no structural intensity at this Q value. The peaks arising from the magnon modes on both sides of the (100) Bragg points are noticeable and determine the magnon dispersion. The dispersion could be followed up to 12meV energies, but the drop of intensity and the perturbations by phonon scattering harden studies at higher energies. Only with noticeable efforts data above 12meV can be obtained on a TAS instrument. So, we limited our constant energy scans from 2meV to 12meV under the 6T magnetic field. Constant energy scans were done in 4, 6, and 10meV in different temperatures up to 200K and cover both sides of the magnetic zone center. The magnon signals can be easily followed for all the temperatures but the magnon gap closes above the ferromagnetic transition temperature. The characteristic two peak structure stays visible even above the Curie temperature, that proves the persistence of ferromagnetic correlations. We have already shown that the magnon gap and magnon stiffness soften upon cooling over a large temperature change which is nicely explained by the effect of Weyl points and perfectly reproduced by this new experiment [4].

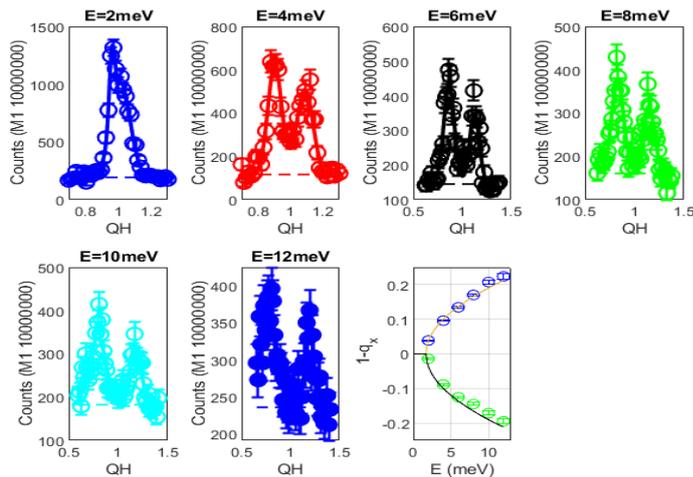


Fig. 2: Different constant energy scans across the magnon dispersion recorded at at $Q=(1, 0, 0)$ in longitudinal direction at 2K temperature. In the down part the magnon dispersion obtained from the scans is plotted.

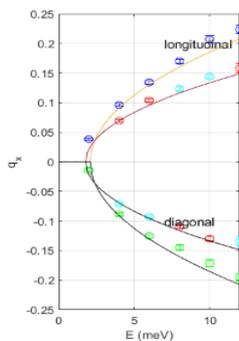


Fig. 3: Calculated magnon dispersion recorded at $Q=(1\ 0\ 0)$ in diagonal and longitudinal directions. The stiffness is smaller in diagonal direction.



Fig. 4: Six SrRuO_3 crystals oriented and co-aligned together [8].

Constant energy scans along high-symmetry directions around the ferromagnetic zone center $Q=(1\ 0\ 0)$ manifest the magnon dispersion since the position of the peaks varies with increasing energy transfer. In Fig.2 one can see the dispersion in longitudinal direction

recorder at $Q=(1\ 0\ 0)$. In the following Figure (Fig. 3) the comparison of magnon dispersion in two different directions is depicted. The magnon gaps and stiffness constants in diagonal and longitudinal directions amount to 1.8meV, 89.6 meVÅ², and 2.1meV, and 88.8 meVÅ², respectively at 2K. In orthorhombic SrRuO₃, there is thus no measurable anisotropy in the parabolic dispersion at low energies. The values nicely agree with our previous determination on a twinned crystal [4]. We could determine the intrinsic width of magnons as a function of temperature by recording the data at six different temperatures between 2 and 200K (Fig. 5) with different energies which also gives access to the intrinsic broadening of the magnon signals (i.e. without broadening due to the mixing of directions).

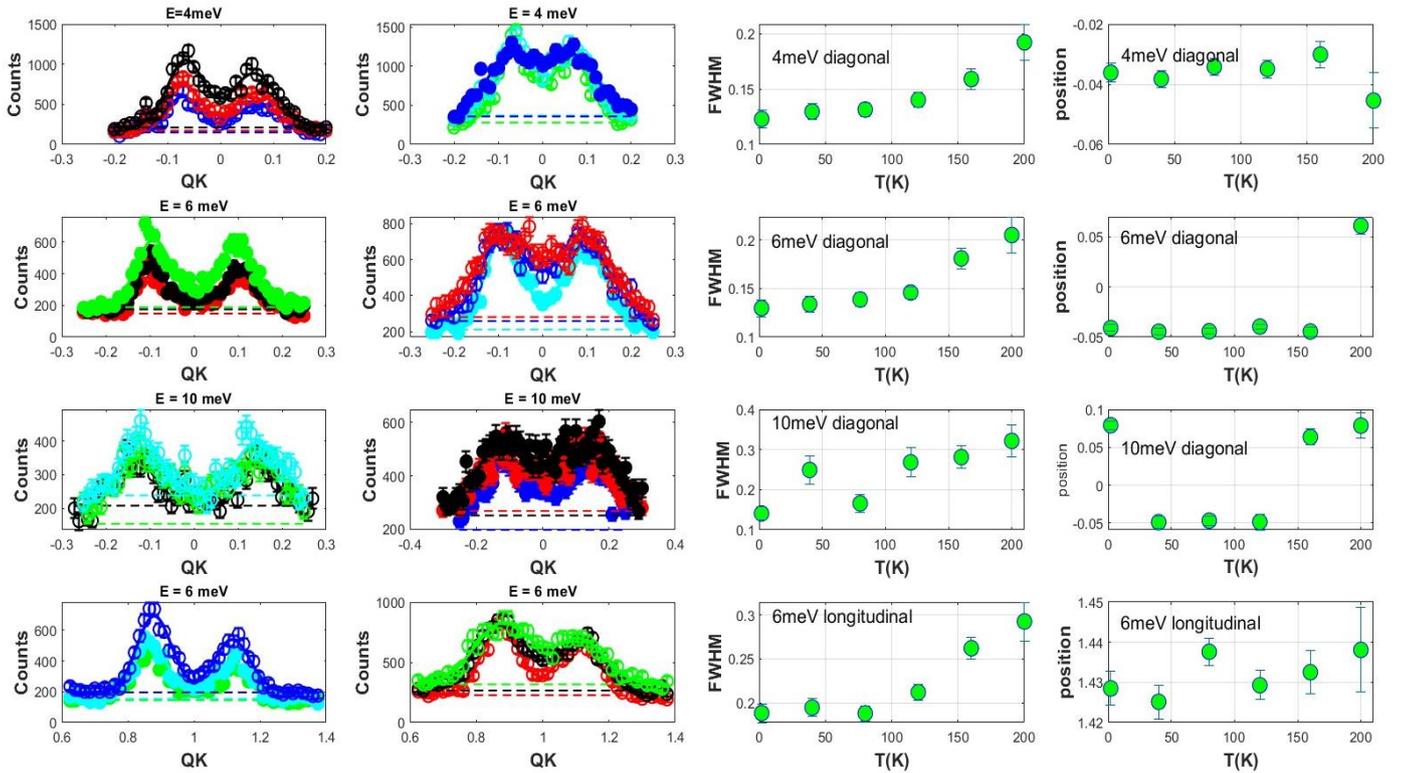


Fig. 5: Constant energy scans across the magnon dispersion measured at $Q=(1\ 0\ 0)$ in diagonal and longitudinal direction for various energies and in different temperatures and the average FWHM and Positions of the peaks

We also took advantage of the detwinning to get unambiguous results on the phonon dispersion. Due to the coupling between the rotational modes with structural distortions and orthorhombic directions, noticeable differences in rotation modes contributing to the same Q vector of the twinned crystal exist that render data on twinned samples difficult to interpret. Due to time limitations, we focussed on measurements on characteristic zone boundaries.

References:

- [1] S. Kunkemöller et al., *Crystal Research and Technology*, vol. 51, no. 4, pp. 299-305, 2016.
- [2] A. Kanbayasi et al., *Journal of the physics society of japan*, vol. 41, no. 6, pp. 1876-1878, 1976.
- [3] S. Itoh et al., *Nature Communications*, vol. 7, no. 1, p. 11788, 2016.
- [4] K. Jenni et al., *Physical Review letters*, vol. 123, no. 1, p. 017202, 2019
- [5] N. Nagaosa et al., *Review of modern physics.*, vol. 82, no. 2, p. 1539, 2010.
- [6] S. Kunkemöller et al., *Physical Review B*, vol. 96, no. 22, p. 220406, 2017.
- [7] M. Braden et al., *Physical Review B*, vol. 76, no. 1, p. 014505, 2007.
- [8] K. Jenni, Doctoral dissertation, Universität zu Köln, 2021.