Experimental report

14/12/2021

Proposal: 4-01-1663 **Council:** 10/2019

Title: Longitudinal spin fluctuations in the metallic ferromagnet $SrRuO_3$

Research area: Physics

This proposal is a new proposal

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Samples: $SrRuO_3$

Instrument	Requested days	Allocated days	From	To
IN20	7	6	22/03/2021	30/03/2021

Abstract:

Our previous unpolarized INS study of ferromagnetic $SrRuO_3$ revealed a remarkable impact of Weyl points on the spin dynamics. Both, the magnon gap and the stiffness soften upon cooling in the ferromagnetic phase, which can be attributed to the same renormalization factor that describes the anomalous Hall effect. We would like to extend our study to the longitudinal spin fluctuations and to the temperature range close and above the Curie temperature, which will further elucidate the ferromagnetic state and its emergence in $SrRuO_3$.

Experimental Report

Instrument	IN20
Proposal Number	4-01-1663
Proposal	Longitudinal spin fluctuations in the metallic ferromagnet $SrRuO_3$
Experimentalist	Kevin Jenni, Akshay Tewari, Markus Braden University of Cologne
Local Contact	Paul Steffens

$SrRuO_3$ is of special interest due to its application as a conducting oxide layer in thin-film applications; it is one of the very few known metallic ferromagnetic (FM) oxides, moment of 1.6 μ_B, and T_c=165 K [2-7] and it is related to the unconventional superconductivity in Sr_2RuO_4 [1]. Many observations are highly anomalous [2-5]: Thermal expansion shows an invar effect [8], magnetization interferes with the DC transport [9], and there is evidence for non-Fermi-liquid behavior [10,11]. The high magnetic anisotropy indicates strong spin-orbit coupling. Our previous comprehensive unpolarized INS experiments on $SrRuO_3$ determine the main characteristics of the magnon dispersion [14,15]. At low temperature the dispersion is isotropic with stiffness D=87(2) meVÅ^2 and a well-defined magnon gap Δ=0.94(3)meV [14]. The temperature dependence of both spin-dynamics parameters, namely gap energy and magnon stiffness, can be explained as a fingerprint of Weyl points and is coupled to the anomalous Hall effect [14]. Strong SOC in $SrRuO_3$ intertwines charge and spin degrees of freedom resulting in Weyl points where the charge dynamics couples to the spin dynamics. $SrRuO_3$ is thus a remarkable example for the impact of Weyl physics on the magnon dynamics [14].

The main aim of this polarized experiment was investigating longitudinal modes. Here longitudinal designates a fluctuation along the magnetization that in a system with local moments would be suppressed. The analysis of the magnetization density in $SrRuO_3$ clearly shows that oxygen carries a large magnetic moment [19] which can enhance the tendency to form low-energy longitudinal modes. In addition we wanted to study the magnetic excitations in general near and above the Curie temperature, where phonon scattering becomes too important and polarization analysis is needed to isolate the magnetic signal.

For the experiment we used 6 large coaligned single crystals grown at Cologne University. This assembly was already studied on previous INS experiments [14,15]. A large horizontal magnetic field of up to 3.8T (cryomagnet 134OXHV38) was applied in order to avoid the neutron depolarization. However, the field was found to heavily reduce the operation of the spin-flippers which needed to be calibrated as function of the angle between the field and the beam (i.e. A3-A4 for the second flipper). For most of the experiment we only flipped on k_f. Figure 1 shows the flipping ratios at various temperature and magnetic fields of 3.8 and 1T. One clearly sees that the quality of the polarization gets lost when the FM order sets in and that the smaller field can only yield sufficient polarization for small values of the FM magnetization. The sample was mounted with (110) and (001) directions spanning the scattering plane (in pseudocubic notation) and the field was set along [110]. With this geometry one can study the following Bragg points: (110), (001) and (002) in SF and nSF channels. However, it is not possible to vary the neutron polarization direction at a given scattering vector as this is given by the horizontal field. Therefore, we only see the *x* direction at (110) and the *y* direction at (001) and (002) with *x,y,z* being the standard coordinate system in polarized INS experiments. This yields the following selection rules:

- Q=(hh0) transversal magn. excitations in SF / nuclear excitations in nSF

- Q=(00L) nuclear and long. magn. excitations in nSF / transversal magn. excitations in SF

The transversal magnetic excitations in SF at (hh0) must be two times stronger than the signal in SF at (00L) unless there is an anisotropy. Further restrictions of the experiment arise from the constraints enforced by the geometry of the magnet yielding the condition 20<A3<160 and (A3-A4<-20 or A3-A4>20). While this yields little impact at Q=(110) it prohibits longer energy scans at (00L) without changing the L value. Before the experiment we simulated the possibilities with the other available horizontal magnet (258OXHV49), which however does not allow for any reasonable scanning.

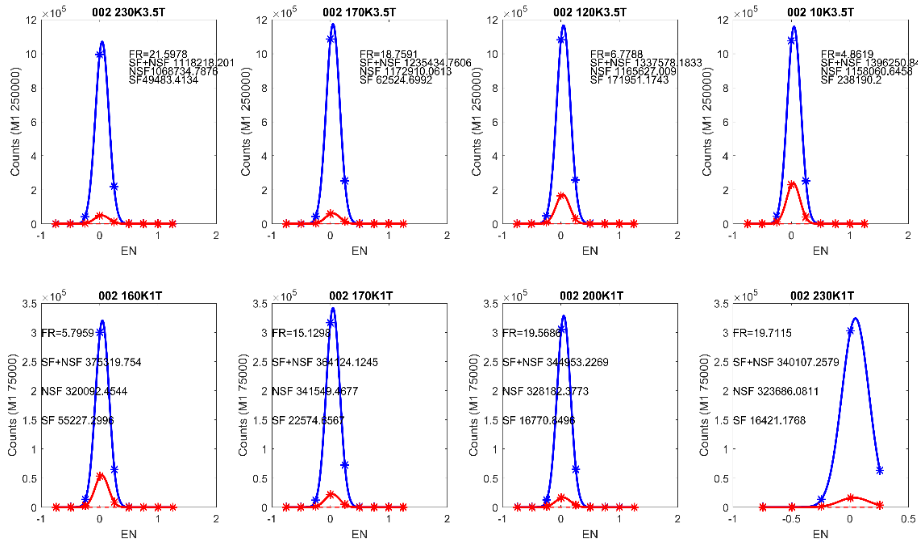

Figure 1: Energy scans across the (002) peak that illustrate the loss of polarization when the magnetization of $SrRuO_3$ increases. Note that at (002) with magnetic field along (110) all scattering should be found in the nSF channel. Clearly a high field of 3.8T is needed in the FM phase to maintain polarization.

The main inelastic results are shown in Figure 2 for the raw data. Concerning the longitudinal excitations at low temperatures, we do not find a clear hint in the energy range accessible, but the phonon contribution cannot be precisely estimated, because the separated measurement is only possible at (110), i.e. a different Bragg point. The signal at 15 to 20meV at (001) at low temperature nevertheless seems to be fully phononic. Only close to the magnetic transition, which however is smeared out in the finite field, there is some evidence for longitudinal excitations at low energies visible in the nSF spectra taken at (002) for different temperature and field values. But this signal is superposed on a large phonon contribution that requires further quantitative analysis in order to identify the longitudinal magnetic contributions. The origin of such modes does however not necessarily stem from excitations across the exchange gap.

The most prominent temperature driven change in the spectra is found at (110) at energies around 20meV. Here the phonon signal should exclusively contribute to nSF processes while the SF signal must be magnetic. However, the strong increase in the SF signal appears suspicious as it is not consistent with the data at SF (001) that senses half of transversal magnetic excitations. We carefully checked the flipping ratio at the value of the corresponding incoming energy but did not find an indication for a loss of polarization and several scans at varying scattering vectors were taken.

The SF data at (001) and (110), however, consistently indicate a general uptake of magnetic scattering when approaching the paramagnetic phase. For sharp magnons one does not expect magnetic scattering right at the magnetic zone centre and well above the magnon gap. This increase of scattering is consistent with previous unpolarised INS experiments [14,15] that indicate serious broadening of magnons upon heating. The polarized data underline an essential change in the magnon response from a simple magnon dispersion with well-defined modes at low temperature to a more continuum-like scattering distribution resembling a paramagnon response.

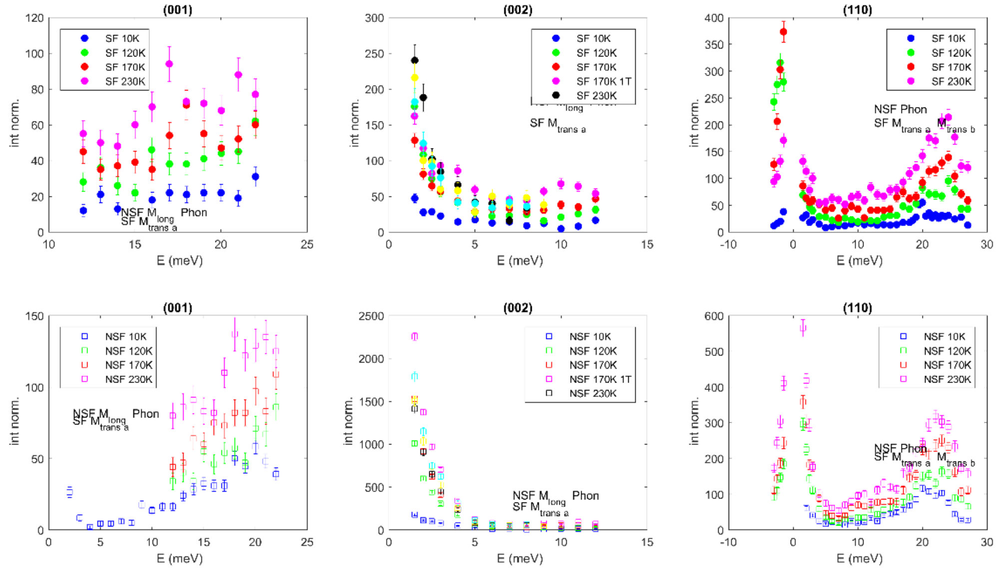

Figure 2: Comparison of the polarized energy scans at the three Q vectors studied most. SF and nSF data are shown in the upper and lower panels, respectively and the color coding is identical in all panels. At 10 K the data are fully consistent with well-defined magnon scattering, and there is no evidence for longitudinal excitations. However, at higher temperature there are additional scattering contributions.

The SF data taken at (110) reveal a strong asymmetry of negative and positive energies. Following the commutation rules of the spin operators, the FM magnon must be right-handed unless very strong spin-orbit coupling implies a change. The chirality of the magnon in $SrRuO_3$ was intensively studied on IN12 [15] finding exclusively right-handed magnons well in the FM phase. This is further supported by the SF data at (110) obtained on IN20 which exhibit the magnon gap signal only at negative energy, as it is expected for a right-handed excitation.

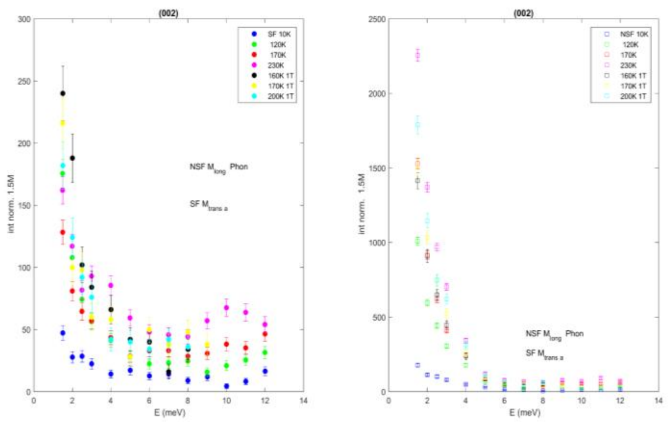

Figure 3: Comparison of the polarized energy scans at the Q=(002) for different temperature and magnetic field values. At high temperature nSF and SF data are dominated by the strongly enhanced phonon scattering, and possible evidence for a longitudinal low-energy contribution around the onset of FM magnetization requires a carefull analysis of the temperature and energy dependence.

[1] Y. Maeno et al., Nature **372**, 532 (1994). **[2]** R.S. Perry et al., PRL **86**, 2661 (2001). **[3]** Nakatsuji et al., PRL **90**, 137202 (2003) **[4]** Nakatsuji et al., Phys. B **259**, 949 (1999). **[5]** M. Braden et al., PRL **88**, 197002 (2002). **[6]** G. Koster et al., Rev. Mod. Phys. **84**, 253 (2012). **[7]** A. Kanbayasi, J. Phys. Soc. of Japan **41**, 1876 (1976). **[8]** T. Kiyama et al., PRB **54**, R756 (1996). **[9]** L. Klein et al. PRL **77**, 2774 (1996). **[10]** P. Kostic et al., PRL **81**, 2498 (1998). **[11]** M.S. Laad PRL **87**, 246402 (2001). **[12]** S. Kunkemoeller et al. Chrys. Res Tec. **51**, 299 (2016) ; PRB **96**, 220406(R) (2017). **[13]** S. Itoh et al. Nat Com. **7**, 11788 (2016). **[14]** K. Jenni et al., PRL **123,** 017202 (2019). **[15]** K. Jenni, Dissertation, University of Cologne 2021.