Experimental report

18/12/2023

Proposal: CRG-2913 **Council:** 10/2022

Title: Nature of magnetic correlations inCo-doped Sr2RuO4

Research area: Physics

This proposal is a new proposal

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Samples: Sr2Ru0.95Co0.05O4

Instrument	Requested days	Allocated days	From	To
IN12	5	5	09/06/2023	14/06/2023

Abstract:

There is a very active discussion about the symmetry and the pairing mechanism in superconducting Sr2RuO4. Small amounts of Co doping seem to induce a ferromagnetic glassy ordering while all other previously studied substitutions yielded antiferromagnetic spin-density wave ordering. Ferromagnetic ordering in only slightly doped Sr2RuO4 would be most relevant for the discussion about the superconducting pairing, but this ordering needs to be better characterized. We therefore propose to analyse the magnetic correlations in Co-doped Sr2RuO4 by polarized neutron scattering to determine the amplitude and correlation lengths as function of temperature.

Experimental Report

Instrument	IN12
Proposal Number	CRG-2913
Proposal	Nature of magnetic correlations in Co-doped Sr2RuO4
Experimentalist	Felix Wirth, Kevin Jenni, Markus Braden, Yvan Sidis
Local Contact	Karin Schmalzl

The superconducting (SC) pairing in Sr_2RuO_4 [1] remains the subject of very active debate [2-4]. Unconventional superconductivity and magnetic correlations are expected to be closely coupled in Sr2RuO4. The longtime advocated triplet p-wave pairing arises from coupling through quasi-ferromagnetic (FM) fluctuations. Sr_2RuO_4, however, seems to be closest to an antiferromagnetic (AFM) instability [5], and new studies of the NMR Knight shift and polarized neutron diffraction [4,6] reveal an unambiguous drop of the electronic susceptibility that is inconsistent with spin-triplet pairs parallel to the layers. The strong nesting in the one-dimensional bands results in strongly enhanced AFM fluctuations near Q_{IC}=(0.3 0.3 0) in pure Sr_2RuO_4 [5,7] and minor chemical substitution leads to the static spin density wave (SDW) ordering of this AFM instability. Only 2.5% of Ti or 3 % of Mn induces this AFM phase, and the same magnetic order was found to occur upon replacing Sr with isovalent Ca [8]. Other studies with Fe-doped samples suggest non-fermi surface nesting driven SDW ordering with a commensurate wave vector Q_C=(0.25 0.25 0), while the AFM fluctuations stay present at Q_{IC} [9]. Doping Sr_2RuO_4 with 1.5-10% of Co results in a consistent and sharp rise in remnant magnetization as temperature decreases. Concurrently, magnetic hysteresis and exponential magnetic relaxation are observed, indicating the formation of FM clusters by Co within the crystal. In our earlier investigations on IN22 (refer to report CRG-2575) involving a 5% Co-doped sample, we found no indications of static magnetic SDW ordering. Elastic scans conducted near Q_{IC} did not reveal any notable increase in intensity, while inelastic measurements at that position resemble the single relaxor behavior like in the pure compound.

In this experiment, we aimed to use polarization analysis to strengthen the evidence for FM ordering. The main goals were to confirm the magnetic origin of the previously seen low Q signal (report CRG-2575) and to determine the spatial anisotropy, which magnetometer studies suggested to be essentially c polarized. We further aspired to study the temperature dependence of the amplitude in more detail.

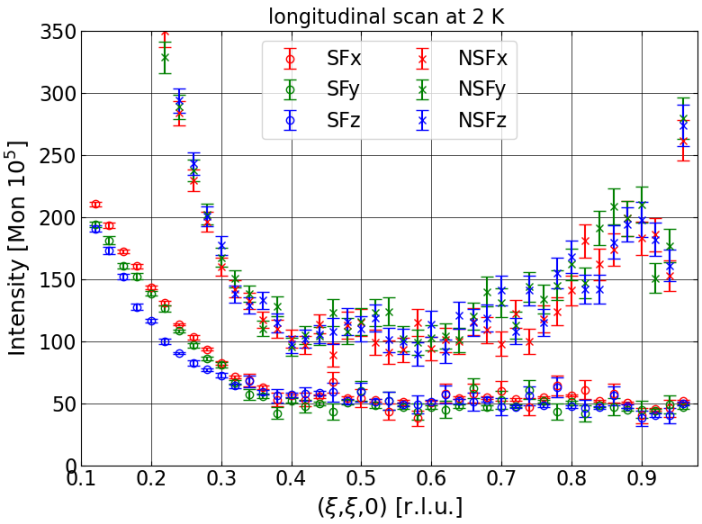

Fig. 1: longitudinal scan for all respective spin-flip and non-spin-flip channels at 2 K.

To detect the tiny magnetic moment with polarized neutrons we measured at IN12 in the following configuration. A focused Heusler analyzer and a monochromator were used to polarize the neutron beam and analyze the signals. To study the independent spin orientations with longitudinal *xyz* analysis the sample was placed inside a Helmholtz-coil setup. A 20x30 mm diaphragm was placed in front and a 30x40 mm one was placed behind the sample to effectively minimize the

background. A Be filter was installed to suppress the higher-order impurities. To maximize the flux of neutrons, we measured always with a final wave vector of 1.5 $Å^{-1}$.

After starting the experiment, we performed elastic longitudinal scans along the Brillouin zone and collected all spin-flip and non-spin-flip channels at 2 (see Fig. 1) and 30 K. One eye-catching feature is the difference between the y-spin-flip and the z-spin-flip channel around Q_1=(0.2,0.2,0), which already gives clear evidence for the magnetic moments being pointed along c. The none-spin-flip channels resemble our previous unpolarized experiment (see report CRG-2575). Further, we checked for potential magnetic response around the Q=(1,0,0), which shows broad FM fluctuations in the pure compound. Scans along (ξ 0 0) and (ξ ξ-1 0) at 2 and 30 K were performed. Fig. 2 shows the data after polarization analysis. Since all polarization gave essentially zero, no magnetic response could be detected.

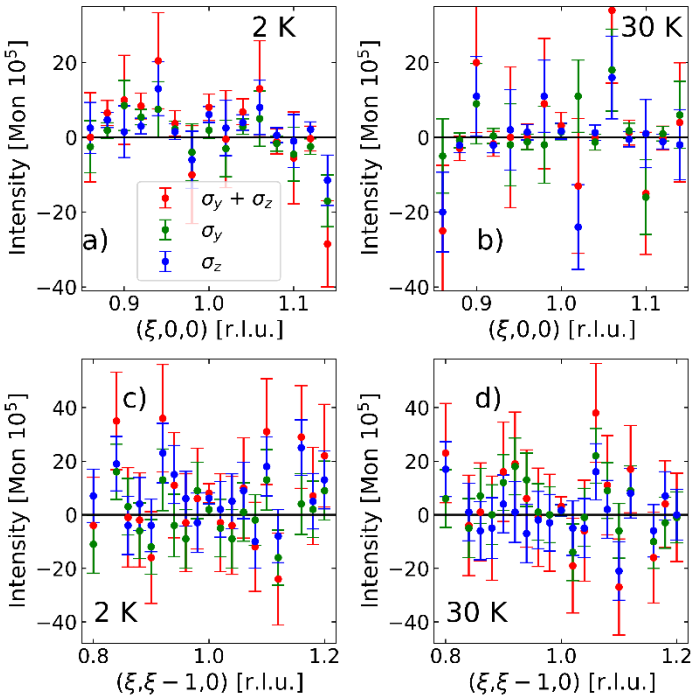

Fig. 2: Polarization analysis for different scans on Q=(1,0,0). a) (ξ 0 0) scan at 2 K, b) scan along (ξ 0 0) at 30 K, c) (ξ ξ-1 0) scan at 2 K, d) scan along (ξ ξ-1 0) at 30 K

After the detection of the magnetic signal at Q_1=(0.2,0.2,0) more beam time was spent to improve the statistics and to see, if the detected signal is also present around Q_2=(0.8,0.8,0), Q_3=(0.8,0.2,0) and Q_4=(0.2,0.8,0). Since the magnetic form factor declines rapidly with increasing |Q|, Q=(0.8,0.8,0) were hard to detect and needed a decent amount of counting time. The signals in the spin-flip channels around Q_1 and Q_2 are shown in Fig. 3 in more detail and also after polarization analysis. Fits for the polarization analysis were performed with two Gaussian functions plus a constant background. For the $\boldsymbol{\sigma_y}$ channel, a linear function fits the data best. At Q_1 the $\boldsymbol{\sigma_z}$ channel shows a pronounced peak, which lies almost perfectly on QH = 0.2, while the $\boldsymbol{\sigma_y}$+$\boldsymbol{\sigma_z}$ channel is shifted to smaller Q values, due to the increased background caused by the direct beam nearby. At Q_2 the signal is, as expected, smaller but $\boldsymbol{\sigma_y}$ and $\boldsymbol{\sigma_z}$ channels are separated. In addition, single point measurements on Q_3, Q_4, and a rocking scan over Q_1 draw the same picture and strengthen the evidence for Co causing magnetic ordering in Sr_2RuO_4. As in the pure and the Ti-, Mn-, and Ca-doped compounds the observed

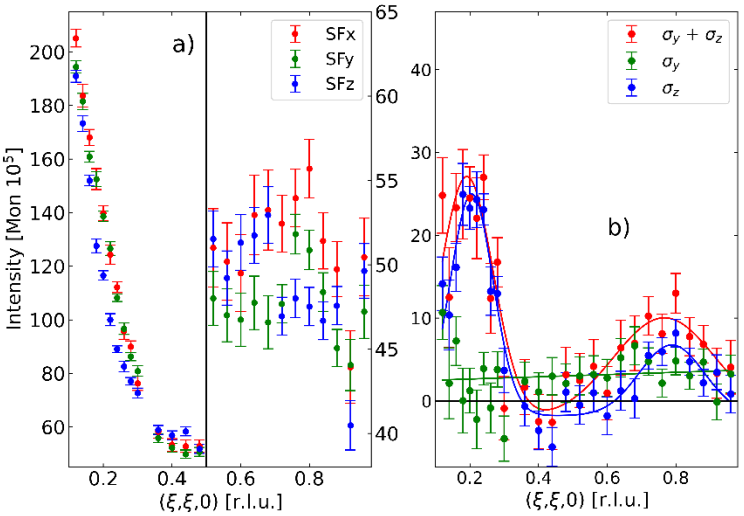

Fig. 3: a) longitudinal scan on spin-flip channels in more details, b) polarization analysis showing signals at Q_1 and Q_2

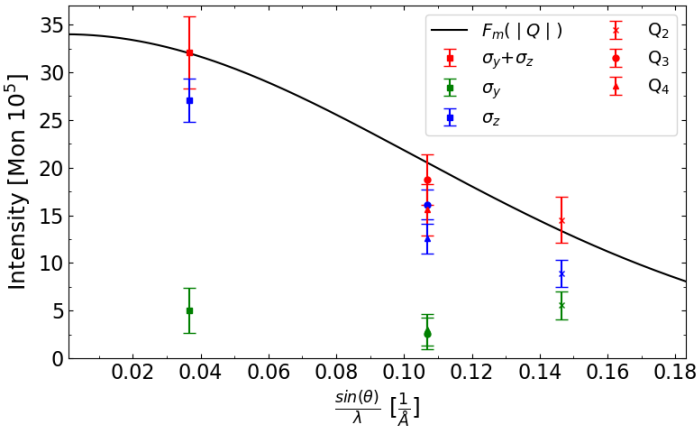

Fig. 4: polarization analysis on the different points of interest Q_1, Q_2, Q_3, and Q_4 translated to |Q| and magnetic moments point along the c-direction [8]. In Fig. 4 single point polarization analysis is shown for the four points of interest in the Brillouin zone compared to the magnetic form factor of Ru^{1+}. The intensity of all points follows the form factor almost exactly, which supports the idea of an intrinsic commensurate magnetic ordering, that is not fermi surface driven comparable to the previously reported Fe-doped samples [9].

Further, we studied the temperature dependence on the Q_1 signal. To have a better comparison to the previously measured data (see report CRG-2575) we performed a temperature scan on Q=(0.24,0.24,0). The temperature dependence is shown in Fig. 4. On the left-hand side the respective spin-flip channels are shown. The z-spin-flip channel is fitted with a constant line, while the x- and y-spin-flip channels are fitted with lines on the intervals above and below the transition. There the upturn at about 55 K indicates the transition. On the right-hand side, the data is shown after polarization analysis. There the transition temperature seems to be slightly reduced. Comparing the fits of the channels of $\sigma_y + \sigma_z$, σ_y, and σ_z, above and below the transition temperatures, the split between vertical and in-plane moments becomes unambiguous. The $\sigma_y + \sigma_z$ and σ_z channels are perfectly parallel. Hence the observed magnetic response is driven by the c-polarized moments plus a constant fluctuation in the plane.

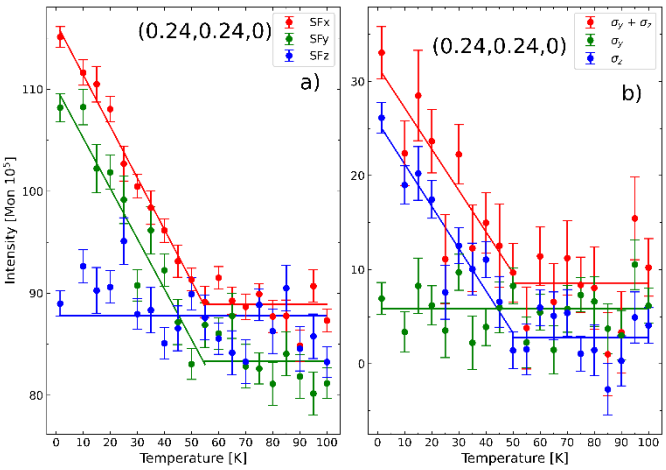

Fig. 5: Temperature dependence of the respective spin-flip channels and the Background

In summary, we have investigated the elastic magnetic response of Sr_2RuO_4 doped with 5 % of Co with polarization analysis. Here we observed magnetic ordering with a commensurate propagation vector of (0.2 0.2 0), while FM fluctuations at (1 0 0) could not been detected. The intensity of the respective point of Interest in the Brillouin zone agrees well with the magnetic form factor. We could confirm, that the magnetic moments are pointing along the crystallographic c-axis. The temperature dependence suggests a transition to the magnetic ordering at around 50 K.

References

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