

Experimental report

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Proposal: 4-02-617

Council: 10/2022

Title: π,π resonance mode in the superconducting state of Sr₂RuO₄

Research area: Physics

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Samples: Sr₂RuO₄

Instrument	Requested days	Allocated days	From	To
THALES	8	8	06/06/2023	14/06/2023

Abstract:

The most advocated scenario for the superconducting pairing in Sr₂RuO₄ was recently excluded by new NMR experiments resulting in a very active debate about the SC state. We propose to search for a spin resonance mode as a fingerprint of unconventional pairing symmetry. Our recent unpolarized experiment on THALES yields evidence for such a mode appearing in the SC phase at (0.5,0.5,0), as it is expected for a dx²-y² gap distribution. However, additional experimental support is required. We wish to apply longitudinal polarization analysis to further elucidate the magnetic response in the SC state of Sr₂RuO₄.

Experimental Report

Instrument	ThALES
Proposal Number	4-02-617
Proposal	π,π resonance mode in the superconducting state of Sr_2RuO_4
Experimentalist	Felix Wirth, Yvan Sidis, Markus Braden
Local Contact	Paul Steffens

The superconducting (SC) state in Sr_2RuO_4 remains mysterious after over 25 years of efforts. The longtime advocated triplet p-wave pairing arises from coupling through quasi-ferromagnetic (FM) fluctuations. However, the dominant magnetic excitations in Sr_2RuO_4 are incommensurate (IC) fluctuations originating from strong nesting in the Q1D bands [1]. These fluctuations do not seem to play the most active role in the SC pairing [2]. Polarized INS experiments have shown that there indeed exist quasi-FM fluctuations, which are significantly weaker than the IC ones but widely spread in Q-space [3], so their q-averaged impact is at least comparable. For the recently advocated $d_{x^2-y^2}$ gap distribution one expects no resonance modes or large gaps at the nesting vector, which is consistent with the previous observation [2]. However, this $d_{x^2-y^2}$ gap symmetry leads to a sign change at $\mathbf{q}=(0.5, 0.5, 0)$ (or π,π), which can lead to the occurrence of a resonance mode or a large gap. On a previous experiment on Thales, see report on 4-02-586, some evidence for such a gap was detected by unpolarized neutron scattering.

In this experiment, we aimed to use polarization analysis to further investigate the magnetic fluctuations in Sr_2RuO_4 in its superconducting state. The main goals were to confirm the suppression of the spectral weight at the nesting position at very low energies and to establish the magnetic nature of the unpolarized signal at the potential resonance mode or gap position at $(0.5, 0.5, 0)$.

To achieve the low background, the high energy resolution, and the high flux required for the described tasks, we measured at ThALES 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, the Cryopad device was used although we studied only longitudinal xyz analysis. In a superconductor, the Cryopad device is advantageous, because there are no guide fields at the sample that can become trapped or expelled. The Be filter was also installed to suppress the higher order impurities.

After starting the experiment, considerable time was spent on finding optimal measurement conditions. Since the elastic line had a broad tail at $K_f = 1.55 \text{ \AA}^{-1}$, smaller $K_f = 1.35$ and 1.2 \AA^{-1} were also tried. This improved the background although some tails persisted, but the induced intensities losses could not be accepted. Therefore, a K_f of 1.55 \AA^{-1} had to be kept and a radial collimator was installed to reduce the background.

We started with the low-energy response at the incommensurate positions $Q_{IC1} = (0.3, 0.3, 0)$, which is related to the nesting between the one-dimensional sheets of the Fermi surface. Fig.1 shows the easily detectable magnetic signal for $E = 2 \text{ meV}$ at this position and also the pronounced spin anisotropy

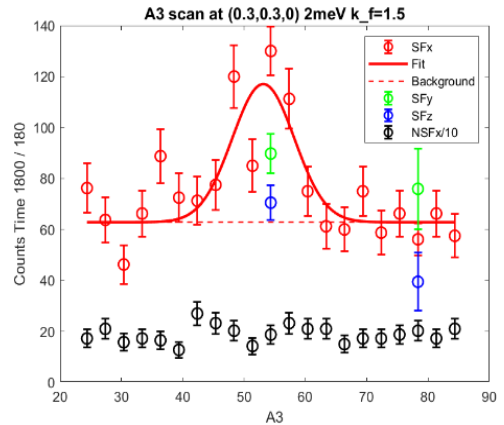


Fig. 1: A3 scan over the nesting signal at $(0.3, 0.3, 0)$ and 2 meV . The spin-flip x-channel and the non-spin-flip x-channel were collected. To determine the anisotropic character of the magnetic signal two points in y- and z-channels were also measured. Note that the NSFx data are scaled by $1/10$.

along the c-direction. Since the intensity is systematically low in polarized neutron experiments and in the given setup, long counting times were expected.

Fig. 2: Polarization analysis at the candidate resonance mode position at $Q=(0.5,0.5,0)$. (left) The different spin-flip channels are compared to the channel without spin-flip (NSF). (Right) Data after subtracting channels to determine magnetic components. Due to the strong background, the error bars remain large preventing a discrimination of weak magnetic signals of the expected strength.

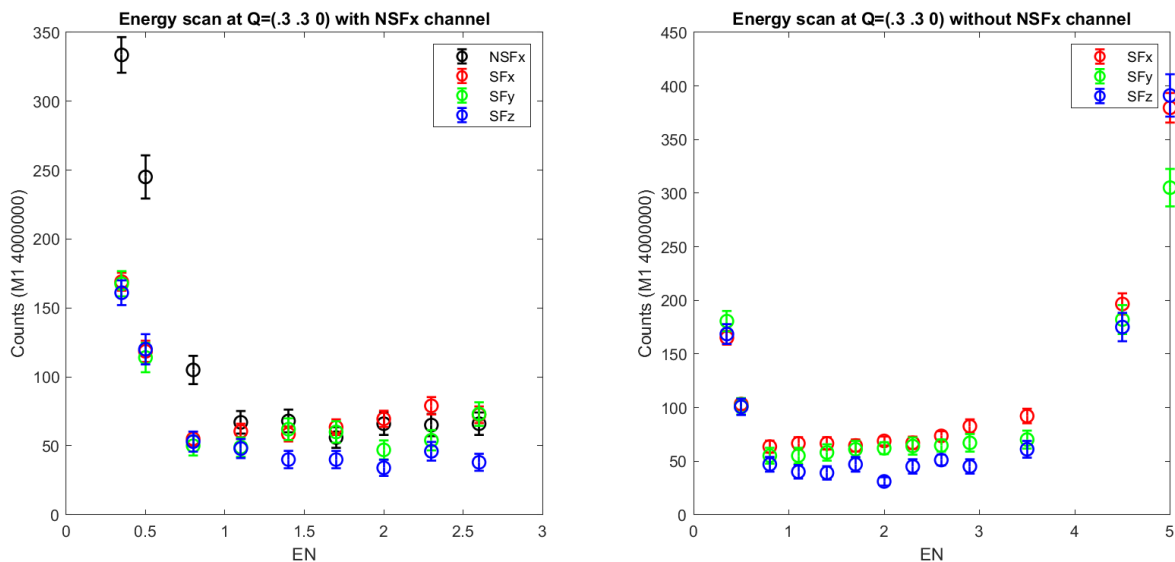
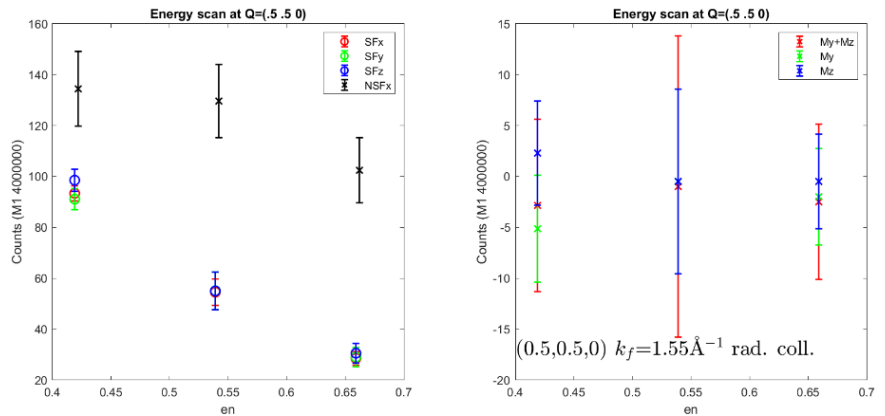


Fig. 3: Constant Q scans at the positions $(0.3,0.3,0)$. The background is represented by the NSFx channel. At low energies, the high background makes it difficult to separate the x, y, and z channels with spin flip. At higher energies, a clear magnetic signal polarized along the z-direction becomes visible. Longer counting times and thus better statistics were performed for energy values between 0.4 meV and 3.5 meV.

Most efforts were spent on the signal previously detected in the temperature dependence at $(0.5, 0.5, 0)$ and 0.42 meV. Fig. 2 shows the results at the three energies we focused on. The raw data presented at the left side indicate that in spite of extremely long counting times statistical errors remain too high due to the large background. Therefore, the subtraction of different channels to determine the magnetic components yields too large errors with respect to the expected small signal. To estimate this expected signal, one can compare the signal from the IC response at 0.6 meV and the signal at $(0.5, 0.5, 0)$ for an energy of 0.42 meV from the previous unpolarized experiment (see report 4-02-586). In that earlier experiment, the signal at $(0.5, 0.5, 0)$ was about 3 times smaller than at the IC position. Transferring this to the current experiment, one might expect about 3 counts per 4 million monitor counts for the resonant mode position (for the total magnetic scattering, i.e., the

sum of both components). To detect and document the magnetic character of such a signal, the statistics would have had to be improved with a long count time, much longer than the available time.

At the nesting position, an energy scan from 0.4 meV to 5 meV was performed to determine the energy dependence of the anisotropy. The left part of Fig. 3 shows the spin-flip channels and the non-spin-flip channel for x-polarization (NSFx). For energies below 1 meV, the intensity of the NSFx channel is greatly increased due to the tail of the elastic line. For higher energies, it is comparable to the other spin-flip channels. On the right side of Fig. 3, the full scan is shown for the spin-flip channels only. Due to the better signal-to-noise ratio, the error bars are small enough to separate the channels and confirm the magnetic nature of the signal. Fig. 4 shows the data with the full polarization analysis. In this energy scan at (0.3 0.3 0), the magnetic signal consists mainly of a c-polarized response that is roughly twice as strong as the in-plane response in the studied energy range. In an earlier polarized INS experiment the anisotropy was only determined at 8meV [4]. Note that the signal strength is roughly linear in energy in agreement with the mostly accepted single-relaxor behavior and a high characteristic energy.

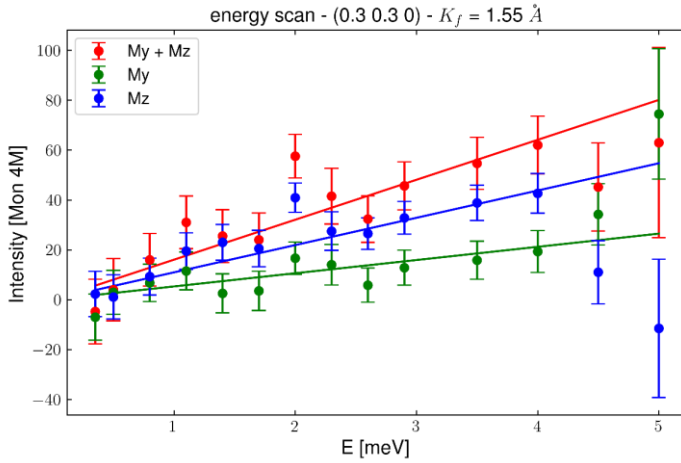


Fig. 4: Constant Q-scans at the nesting position (0.3 0.3 0). Spectral weight suppression at low energies is clearly visible. The polarization anisotropy of the magnetic excitations persists across the studied energy range, as it is suggested by the linear fits.

In summary, we have investigated the low-energy response of the magnetic fluctuation at the nesting point in the Brillouin zone below the SC transition with polarized neutrons. The data on the IC positions confirm and extend the anisotropy of the nesting signal with a dominance of the c-polarized component that corroborates the impact of spin-orbit coupling. Unfortunately, the data taken at $Q = (0.5 0.5 0)$ and at the energy of the weak signal observed in an unpolarized experiment had a high background that made it impossible to detect such a signal in the polarization channels. In general, polarization data acquisition, especially at very low energy, remains a challenge due to the high background leaking from the tail of the elastic line.

References

- [1] Y. Sidis et al., Phys. Rev. Lett. **83**, 3320 (1999); M. Braden et al., Phys. Rev. B **66**, 064522 (2002); F. Servant et al., Phys. Rev. B **65**, 184511 (2002); K. Iida et al., Phys. Rev. B **84**, 060402(R) (2011); K. Iida et al., J. of Phys. Soc. Jpn. **81**, 124710 (2012). [2] S. Kunkemöller et al., Phys. Rev. Lett., 147002 (2017). [3] P. Steffens et al., Phys. Rev. Lett. **120**, 047004 (2019). [4] M. Braden et al., Phys. Rev. Lett. **92**, 097402 (2004).