

# Experimental report

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**Title:** Existence of a resonance mode in superconducting Sr<sub>2</sub>RuO<sub>4</sub>

**Research area:** Physics

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**Samples:** Sr<sub>2</sub>RuO<sub>4</sub>

Instrument	Requested days	Allocated days	From	To
THALES	7	7	16/09/2015	23/09/2015

## Abstract:

There is ongoing debate about the symmetry of the superconducting order parameter in Sr<sub>2</sub>RuO<sub>4</sub> which is one of the most promising materials for topological superconductivity. Also the question which electronic bands drive and which follow superconductivity remains subjects of controversy. We propose to search for a resonance mode or for a shift of spectral weight in the superconducting state in Sr<sub>2</sub>RuO<sub>4</sub> in order to verify or falsify clear predictions made by theory.

# Superconductivity-induced spin-gap in magnetic fluctuations of $\text{Sr}_2\text{RuO}_4$

$\text{Sr}_2\text{RuO}_4$  is one of the best studied unconventional superconductors [1–4] but its pairing symmetry and mechanism remain subject of very active debate. The strong nesting in the 1d bands results in strongly enhanced AFM fluctuations in pure  $\text{Sr}_2\text{RuO}_4$  [6–10] and minor chemical substitution even leads to static ordering of this AFM instability [11, 12]. There is no doubt that  $\text{Sr}_2\text{RuO}_4$  is closest to this nesting driven AFM instability, which, however, cannot be easily associated with the most likely chiral superconducting state [2].

As stated in reference 13, inelastic neutron scattering (INS) can yield valuable information concerning the role of the different bands in the pairing. If superconductivity directly arises from the one-dimensional bands as active bands there must be a clear impact on the associated incommensurate magnetic excitations. Several calculations predict the occurrence of a resonance mode in at least one of the spin excitation channels [13–15]. On the other hand, if superconductivity is mainly driven by the 2d band associated with ferromagnetic fluctuations, a lower gap in the 1d bands and a smaller impact on the magnetic fluctuations is expected. Here we report INS experiments across the superconducting transition in  $\text{Sr}_2\text{RuO}_4$ , which clearly show that nesting-induced magnetic fluctuations only sense a reduced gap indicating that the 1d bands are not the active ones in the superconducting pairing.

INS experiments were carried out at the recently upgraded THALES instrument. In the experiment we used an assembly of 12  $\text{Sr}_2\text{RuO}_4$  crystals with a total volume of  $2.2 \text{ cm}^3$ . The crystals were grown at the University of Kyoto using a floating-zone mirror furnace and similar crystals were studied in many different experiments [3]. We choose the  $[100]/[010]$  scattering geometry, because this yields the best INS signal due to the integration along the vertical direction along  $c$  where little modulation of magnetic response is expected. The crystal assembly was cooled with a dilution cryostat attaining minimum temperature of the order of  $\sim 50 \text{ mK}$ . There is some impact on the neutron absorption on the sample temperature of the order of  $10 \text{ mK}$ , which, however, is negligible compared to the transition temperature. In order to suppress the background we included a radial collimator and a Be filter in front of the analyzer and we used a Si (111) monochromator. We applied vertical focusing at the monochromator and horizontal at the analyzer. In addition, a velocity selector in front of the monochromator was inserted to suppress higher order contaminations. Most scans on THALES were performed with a fixed final momentum of  $k_f = 1.57 \text{ \AA}^{-1}$  where the Be filter effectively cuts all neutrons with only slightly larger final energy. Some scans were performed by scattering at the sample and at the analyzer in the same sense (U configuration), which reduces the background as the detector is positioned farther away from the direct beam but worsens the resolution.

Fig. 1 and 2 show constant energy scans at intermediate energies for temperatures above and below the superconducting transition. The data in Fig. 1 were taken with the PG monochromator on THALES (energy resolution at the elastic line  $\Delta E_0 = 0.22 \text{ meV}$ ) and those in Fig. 2 with a Si (111) monochromator and a radial collimator, which yields a lower background close to the elastic line and improves the resolution ( $\Delta E_0 = 0.12 \text{ meV}$ ). With the dilution cryostat used in these experiments, it is impossible to stabilize temperatures in the range  $1.2$  to  $1.6 \text{ K}$ , therefore we could not follow the signals close to  $T_c$ . The data shown in Fig. 1 and Fig. 2 unambiguously show that the nesting related fluctuations in the energy range  $0.6$  to  $1 \text{ meV}$  can be well studied by our INS experiment and that this signal is not affected by the superconducting transition concerning neither the intensity nor the width. We have studied the nesting signal at the two scattering vectors  $\mathbf{Q} = (0.3 \ 0.3 \ 0)$  and  $(0.7 \ 0.3 \ 0)$  which are not equivalent due to the centering of the body centered lattice in  $\text{Sr}_2\text{RuO}_4$  and due to the lower form factor at the latter reducing the magnetic signal. Because of the quasi-twodimensional nature of the magnetic correlations in  $\text{Sr}_2\text{RuO}_4$ , however, one does not expect an essential difference, and the signal at both scattering vectors is comparable and in particular there is no change at the superconducting transition for energies above  $0.6 \text{ meV}$  at both  $\mathbf{Q}$  values.

Experiments at lower energy transfer are more difficult as described above. Since the background depends on the length of the scattering vector (or the scattering angle), it is not constant in a straight transversal constant-energy scan like those shown in Fig. 1 and 2. Therefore, we performed inelastic rocking scans at lower energy transfer, which are shown in Fig. 3. These scans, which possess a perfectly constant background, clearly indicate that spectral weight is suppressed in the superconducting phase of  $\text{Sr}_2\text{RuO}_4$  due to the opening of the electronic gap. This reduction of spectral weight, however, only occurs at rather low energy transfer,  $E \sim 0.3$  and  $0.4 \text{ meV}$ . These results are summarized and corroborated by the constant  $\mathbf{Q}$  scans, see Fig. 4.

The upper panel of Fig. 4 shows the fitted peak heights of the scans taken in different configurations at the two scattering vectors. In order to allow comparison the data was normalized to the values at  $1 \text{ meV}$  and  $2 \text{ K}$ . The peak heights at larger energies clearly remain unchanged upon entering the superconducting state while a partial suppression is observed below  $0.4 \text{ meV}$ . The middle part of Fig. 4 shows constant  $\mathbf{Q}$  scans taken at the nesting scattering vector  $(0.3 \ 0.3 \ 0)$  above and below the superconducting transition as well as a scan taken at a scattering

vector of the same length but rotated sufficiently away from the nesting position to fully suppress this signal (U-configuration with  $\Delta E_0=0.16$  meV). Taking the latter results as a measure of the background at the nesting position we can deduce the magnetic signal at both temperatures. This clearly shows that the nesting scattering remains essentially unchanged for energies above 0.45 meV while there is a suppression of spectral weight due to the opening of the superconducting gap at lower energies.

The magnetic response of an itinerant system corresponds to a particle-hole excitation, which in a superconductor must be compared to twice the superconducting gap,  $2\Delta$ . There have been several reports on the superconducting gap in  $\text{Sr}_2\text{RuO}_4$  [5, 16–19]: The first tunneling experiments were interpreted as evidence for very large gap and  $\frac{2\Delta}{k_B T_c}$  values [16, 17] while more recent studies more conclusively suggest smaller values: Suderow et al.  $2\Delta=0.56$  meV [18], Kashiwara et al.  $2\Delta=0.93$  meV [19] and Fermo et al.  $2\Delta=0.7$  meV. None of these studies can safely identify the band carrying the largest gap, leaving the discussion about active and passive bands open. On the theoretical side three different studies arrive at nearly the same conclusion that opening the gap in the 1d sheets results in a full suppression of spectral weight below  $2\Delta_{1d}$  and even a resonance enhancement at or close to this value [13–15]. Such resonance enhancement of the magnetic response in the superconducting state has been reported in many unconventional superconductors [20]. In particular in superconductors, in which the pairing appears mediated by a well defined magnetic signal such as the cuprates or the FeAs-based compounds, strong resonance modes are found [20]. Such a behavior can be excluded for the nesting scattering in  $\text{Sr}_2\text{RuO}_4$  which exhibits only modest suppression of magnetic weight that, furthermore, only appears well below the maximum  $2\Delta$  reported in the tunneling experiments. We may, therefore, conclude that the 1d bands are not the active ones for the superconducting pairing.

Nodes of the gap function may lead to persisting magnetic scattering in the superconducting state for energies below the maximum values of  $2\Delta$ . But in the scenario of 1d bands being the active ones for the superconducting pairing mediated by nesting induced fluctuations, some effect of the gap opening must be observed. The fact that we see no change in the magnetic scattering (below 20% for  $E>0.5$  meV) well below the observed maximum values of  $2\Delta$  [5, 16–19] clearly excludes such a scenario.

In conclusion we have studied the low-energy magnetic fluctuations associated with the nesting of 1d bands in  $\text{Sr}_2\text{RuO}_4$ . The fact that we do not observe a significant change in this signal when passing the superconducting transition disagrees with a scenario of nesting related fluctuations driving superconductivity primordially in the 1d bands. Instead a prominent role of ferromagnetic fluctuations on charge carriers in the 2d bands seems more likely.

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- [1] Y. Maeno, H. Hashimoto, K. Yoshida, S. Nishizaki, T. Fujita, J. G. Bednorz and F. Lichtenberg, *Nature* **372**, 532 (1994).
  - [2] A. P. Mackenzie and Y. Maeno, *Rev. Mod. Phys.* **75**, 657 (2003).
  - [3] Y. Maeno, S. Kittaka, T. Nomura, S. Yonezawa and K. Ishida, *J. of Phys. Soc. Jpn.* **81**, 011009 (2012).
  - [4] C. Kallin, *Rep. Prog. Phys.* **75**, 042501 (2012).
  - [5] I. A. Fermo, S. Lederer, C. Lupien, A. P. Mackenzie, J. C. Davis and S. A. Kivelson, *Phys. Rev. B* **88**, 134521 (2013).
  - [6] Y. Sidis, M. Braden, P. Bourges, B. Hennion, S. Nishizaki, Y. Maeno, and Y. Mori, *Phys. Rev. Lett.* **83**, 3320 (1999).
  - [7] M. Braden, Y. Sidis, P. Bourges, P. Pfeuty, J. Kulda, Z. Mao, and Y. Maeno, *Phys. Rev. B* **66**, 064522 (2002).
  - [8] F. Servant, B. Fak, S. Raymond, J. P. Brison, P. Lejay, and J. Flouquet, *Phys. Rev. B* **65**, 184511 (2002).
  - [9] M. Braden, P. Steffens, Y. Sidis, J. Kulda, P. Bourges, S. Hayden, N. Kikugawa, and Y. Maeno, *Phys. Rev. Lett.* **92**, 097402 (2004).
  - [10] K. Iida, M. Kofu, N. Katayama, J. Lee, R. Kajimoto, Y. Inamura, M. Nakamura, M. Arai, Y. Yoshida, M. Fujita, K. Yamada, and S.-H. Lee *Phys. Rev. B* **84**, 060402(R) (2011).
  - [11] J.P. Carlo, T. Goko, I. M. Gat-Malureanu, P. L. Russo, A. T. Savici, A. A. Aczel, G. J. MacDougall, J. A. Rodriguez, T. J. Williams, G. M. Luke, C. R. Wiebe, Y. Yoshida, S. Nakatsuji, Y. Maeno, T. Taniguchi and Y. J. Uemura, *nature mat.* **11**, 323 (2012).
  - [12] S. Kunkemöller, A. A. Nugroho, Y. Sidis and M. Braden, *Phys. Rev. B* **89**, 045119 (2014)
  - [13] J.W. Huo, T.M. Rice and F.-C. Zhang, *Phys. Rev. Lett.* **110**, 167003 (2013).
  - [14] Hae-Houng Kee, *J. Phys.: Condens. Matter* **12**, 2279 (2002).
  - [15] D. K. Morr, P. F. Traumann, and M. Graf, *Phys. Rev. Lett.* **86**, 5978 (2001).
  - [16] F. Laube, G. Goll, H. v. Löhneysen, M. Fogelström, and F. Lichtenberg, *Phys. Rev. Lett.* **84**, 1595 (2000).
  - [17] M. D. Upward, L. P. Kouwenhoven, A. F. Morpurgo, N. Kikugawa, Z. Q. Mao, and Y. Maeno, *Phys. Rev. B* **65**, 220512(R) (2002).
  - [18] H. Suderow, V. Crespo, I. Guillamon, S. Vieira, F. Servant, P. Lejay, J. P. Brison, and J. Flouquet, *New Journal of Physics* **11**, 093004 (2009).
  - [19] S. Kashiwaya, H. Kashiwaya, H. Kambara, T. Furuta, H. Yaguchi, Y. Tanaka, and Y. Maeno, *Phys. Rev. Lett.* **107**, 077003 (2011).
  - [20] D. J. Scalapino, *Rev. Mod. Phys.* **84**, 1383 (2012).

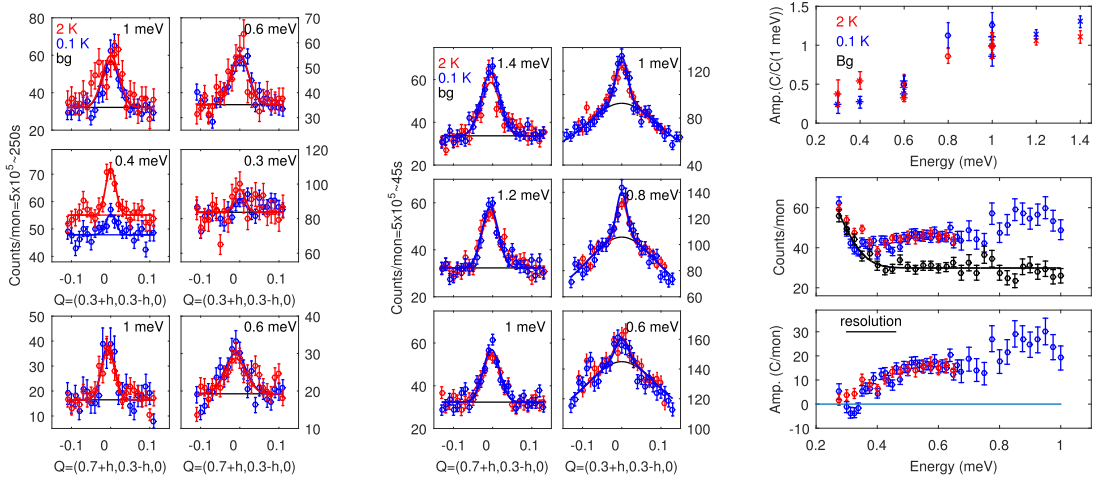


Figure 1: (left) Constant energy scans obtained on THALES with  $k_f=1.57 \text{ \AA}^{-1}$  using a Si monochromator and a PG analyzer with a radial collimation in front of the analyzer.

Figure 2: (middle) Constant energy scans obtained on THALES with  $k_f=1.57 \text{ \AA}^{-1}$  using PG monochromator and analyzer crystals. Note that even for the transversal scans through the  $(0.3 \ 0.3 \ 0)$  the length of the scattering vector varies implying larger background in the center of the scans. Lines denote fits with Gaussian profiles (colored) and curved or linear background (black).

Figure 3: (right) a) Fitted peak heights of the scans taken with the two configurations at the two scattering vectors. In order to allow comparison the data were normalized to the values at 1 meV and 2 K and applied a correction with the Bose factor. b) Constant-Q scans obtained on THALES with  $k_f=1.57 \text{ \AA}^{-1}$  using a Si monochromator and a PG analyzer with a radial collimation in front of the analyzer. Blue and red symbols denote the data taken at  $Q=(0.3 \ 0.3 \ 0)$  above and below the superconducting transition respectively, and the black symbols denote intensity observed at a Q vector of the same length but rotated by 16 degrees with respect to the Q position of the nesting response. c) Magnetic signal obtained by subtracting the background signal from that at the incommensurate nesting position for the two temperatures corrected with the Bose factor.

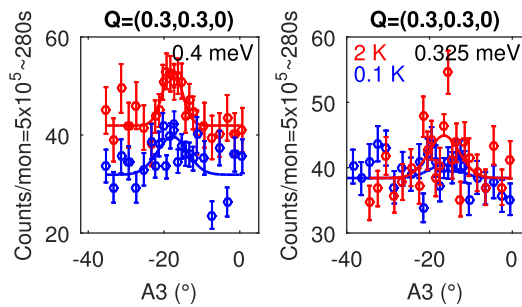


Figure 4: Inelastic rocking scans obtained on THALES with  $k_f=1.57 \text{ \AA}^{-1}$  using a Si monochromator and a PG analyzer with a radial collimation in front of the analyzer. The sample was rotated through the  $(0.3 \ 0.3 \ 0)$   $E=0.4$  and  $0.3 \text{ meV}$  positions at the center of the scans.