Experimental report

02/09/2022

Proposal: **5-51-545** **Council:** 10/2019

Title: Revisiting the spin susceptibilityof Sr2RuO4

Research area: Physics

This proposal is a new proposal

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Samples: Sr2RuO4

Instrument	Requested days	Allocated days	From	To
D33	4	4	27/05/2021	31/05/2021

Abstract:

The superconductivity of Sr2RuO4 HAD been believed to be not a spin singlet but a spin triplet with strong evidences that the NMR knight shift and the polarized neutron scattering measurements revealed a constant spin susceptibility across and well below Tc. In recent years, however, there appear several experimental evidences which support non p-wave symmetry. Moreover, in the beginning of this year, Pustogow et al. arXived that their 17O NMR night shift data clearly shows a drop below Tc, and then Prof. Ishida at Kyoto Univ. admitted that their previous NMR data was misleading due to the heat input by the RF pulses. But the previous polarized neutron results also supported the p-wave scenario since it also shows constant temperature dependence of susceptibility below Tc.
This makes us to propose another polarized neutron scattering experiment on Sr2RuO4.
In the present study, we want to measure the low-Q ferromagnetic susceptibility by using polarized neutron diffraction technique at D33 with a polarized option. Single crystals of Sr2RuO4 with total mass up to 25g are already available. We request 4 days of beamtime.

Experimental Report: Revisiting the pairing state in Sr_2RuO_4

Motivation

Our proposal was accepted in 2019, but delayed by Covid and then a leak in the cryomagnet sample can and was finally performed starting on 26/05/21. We planned to use two techniques to measure the conduction election magnetic susceptibility in Sr_2RuO_4.

(i) To investigate the small-*q* spin susceptibility from fluctuations in Sr_2RuO_4, in the normal and superconducting states, using D33 with a polarised incoming beam (but without polarization analysis).

(ii) To measure the flux lattice signal from the spin-polarized cores of the flux lines in Sr_2RuO_4.

The motivation was to establish whether superconducting pairing in this material is even or odd parity. For many years, it was believed to be a *p*-wave odd-parity material with parallel-spin pairing. However, NMR results [1] in recent years have shown that earlier experiments [2] were affected by NMR pulse heating, and that the spin susceptibility – at least for one magnetic field direction – falls on entering the superconducting state. This strongly suggests anti-parallel-spin even parity pairing. This result matches up with other experimental evidence that pointed away from *p*-wave symmetry, such as the first order transition in the high-field and low temperature corner of the superconducting phase [3], indicating a strong Pauli paramagnetic effect that contradicts the *p*-wave scenario [4]. It appears that the changes in the Knight shift are much stronger for magnetic field in the [100] direction than the [110] direction [5]. However, there remains evidence of time-reversal symmetry-breaking and two-component order parameters [14], so the exact pairing state is still a matter of debate [1, 5-9] and was in great need of further experiment.

Measurement (i) was in our proposal to measure the spin susceptibility at small *q* for two basal plane field directions. In the time-gap between submission and performance of the experiment, an alternative approach was developed. This relies on the fact that for fields close to the basal plane, the superconductivity appears to be Pauli-limited (as expected for anti-parallel spin pairing), in which case, there should be an extra contribution from the spin susceptibility in the flux line cores to the 'form factor'.

We had a fairly brief time to try two methods in a period allotted for one. Also, unlike other materials which show Pauli paramagnetic effects, this is not a heavy-fermion material, and our expectation was that the signals would be weak. Despite preparing large-mass samples of carefully co-aligned crystals, we were not able to obtain clear evidence of the effects we sought.

Experimental details

Our sample is shown in Fig. 1. There were two different mosaics: one was co-aligned to give a (hhl) horizontal scattering plane and the other gave a (h0l) horizontal scattering plane. The (001) faces were glued with hydrogen-free glue to aluminium plates. Reflections at grazing angles from these faces/plates were used to find the exact sample rotation that gave the crystal basal planes parallel to the neutron beam. The sample was mounted on a dilution refrigerator insert, which could be rotated relative to the ORTF horizontal-field cryomagnet, and allowed the required temperature range 50 mK to 2 K.

Fig. 1 ~16 g of co-aligned single crystals of Sr_2RuO_4.

(i) The small-*q* spin susceptibility

The aim was to use a horizontal magnetic field close to the [100] (and later the [110]) directions to induce spin polarization in the conduction electrons, and to compare the spin susceptibility in the superconducting mixed state with that in the normal state. For these measurements, we used a polarized incoming beam and a spin flipper to extract the magnetic component of the scattering from metallurgical scattering from the sample, and we had the field aligned approximately perpendicular to the beam. Measurements on D33 before the beamtime had established that the incoming neutron polarization is maintained even for "perpendicular" magnetic fields, by operating with the applied field at 85° to the neutron beam. This rotates the depolarizing field-zero away from the incoming beam path, so we used this arrangement. For fields parallel to the *ab* plane, $B_{c2}(0)$ is ~ 1.5 T. We established a field in the superconducting mixed state either by cooling in field or by lowering the field from 1.6 T at base temperature. We used horizontal magnetic fields in the range 0.0 to1.6 T, approximately perpendicular to the neutron beam, but exactly parallel to the sample *ab* planes. We searched for the magnetic component over several different *q*-ranges by looking for an interference term by flipping the neutron polarization. The 'moment orientation effect' indicates that such signals should not occur for $\boldsymbol{q} \parallel \boldsymbol{B}$. At the lowest *q* settings, we found an additional effect, which complicated the analysis. As shown on the LHS of Fig. 2. on the next page, there was a change in the signal very close to the beam (no beamstop at 12 Å) in the horizontal direction ($\boldsymbol{q} \parallel \boldsymbol{B}$) when the flipper was operated. The upper two traces in the RHS of Fig. 2 are horizontal scattering to left and right, and the lower two traces are vertical scattering. The sequence in the picture is flipper settings: + - - + + - - + +. There should be no magnetic effect in the horizontal *q*-direction, and yet this direction gives the largest effect. We suspect that this represents a slight focusing and defocusing of the beam, arising from the opposite Zeeman energy of the + & - spin polarizations in the stray field of the magnet. Since the field was horizontal and at 85 deg to the beam, the horizontal and vertical directions are not equivalent.

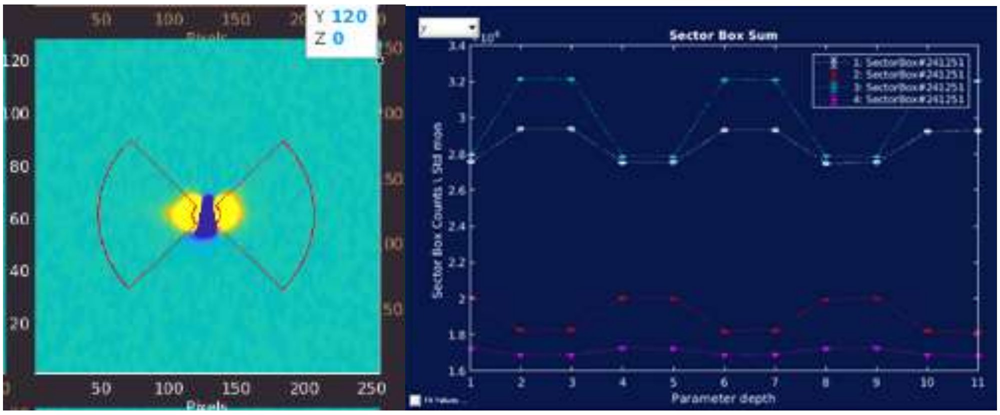

Fig. 2 (L) Detector picture showing the change in signal near the main beam between flipper on & off (R) Intensity of vertical and horizontal scattering of polarized beam as the flipper is turned on & off.

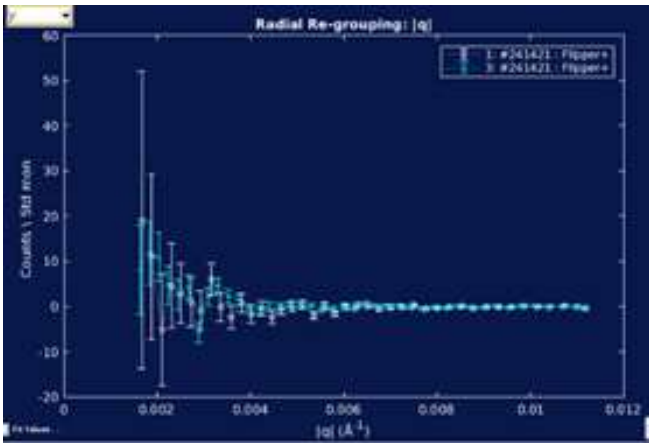

Fig. 3: Flipper effect vs *q* for upper (white) and lower quadrants (blue)

This effect meant that any changes at small q could not be relied upon, so we concentrated on setups at larger q, which give more intensity. However, no significant changes in the signal at larger q due to operating the flipper were observed. Typical results are shown in Fig. 3. We therefore changed to unpolarized beam with $\boldsymbol{B}$ // beam in order to look for Pauli paramagnetic effects, which would be a sign of even parity pairing.

(ii) Search for the flux lattice signal from the spin-polarized cores of the flux lines

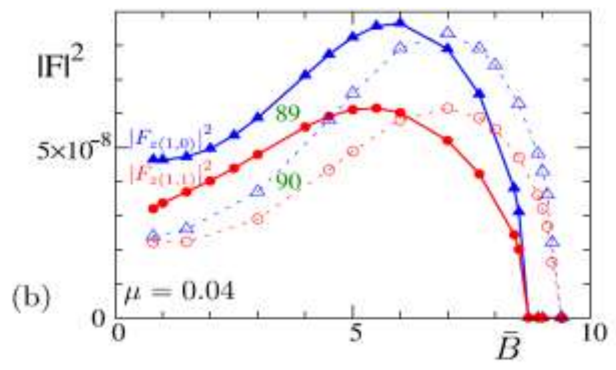

Fig. 4 Predicted field-dependence of PPE FLL form factor [9]

This experiment was proposed some years ago by the Machida group [9]. Against the prevailing *p*-wave opinion of the time, they calculated the Pauli paramagnetic contribution to the flux lattice signal, which is only present in a singlet superconductor. The huge anisotropy factor (~60) of this highly 2-dimensional material means that the conventional FLL signal becomes completely negligible for the field in the basal plane (but gives an easily-detected spin-flip FLL signal for fields just off the planes [10]). However, with B_{c2} ~ 1.5 T, Pauli paramagnetism may become important. Fig. 4 from [9] shows the predicted field-dependence of the FLL intensity for vertical and (nearly) horizontal PPE spots for the field parallel and 1 deg away from the planes. (The horizontal field scale is in theorist units, and B ~ 9 corresponds to 1.5 T.). Any PPE signal from the flux lattice would arise from the field-contrast between the spin polarization in the "normal" flux line cores and the reduced spin susceptibility of the superconducting regions between them. This effect has been clearly observed in $CeCoIn_5$ [11]. We estimated that detection of the horizontal spots would be preferable, because their much larger q would allow relaxed collimation and therefore larger beam intensity. Also, as these reflections had not been searched for before, we investigated them as well as the top and bottom spots. However, we found that the horizontal spots were not observable, due to the big metallurgical scattering/reflections from the basal planes in the samples, so we concentrated on the top/bottom spots. Our measurements at this point were somewhat disrupted by the up/down (phi) motion not always reaching its required value. We had to replace phi scans with a series of phi moves, with the instruction at each value repeated several times, in case the first movement did not reach its required value.

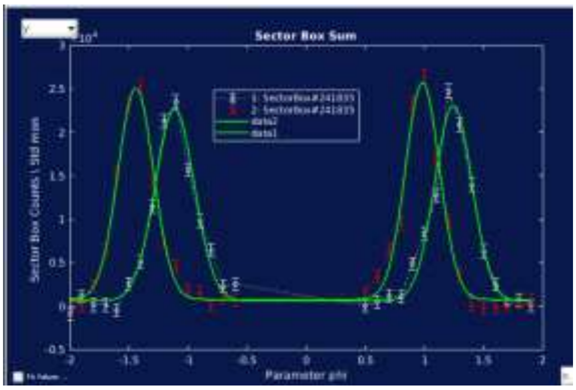

Fig. 5 Rocking curves of top and bottom spin-flip signals

Initially, we measured the FLL reflections arising from spin-flipped neutrons diffracted by transverse field components in the FLL, with a typical rocking curves at 0.4 T shown in Fig. 5. The backgrounds were taken at base temperature at a field of 1.6 T. We see that there are two peaks for the top spot (white points) and two for the bottom (red points). These arise from the energy change of the neutrons when they spin flip in the sample in the presence of a large magnetic field [12]. With an unpolarized input beam, the energy change can be either positive or negative, which shifts the rocking curve by ~+/- 1.2 deg in this case. For a signal arising from the PPE, there are no transverse fields, so there will be rocking curves centred at known positions in the middle of Fig. 5. We went to the calculated positions and took long counts of foreground and background. Again we saw no signal above the Poisson noise that could be attributed to PPE.

Both of the experimental techniques were repeated for both sample orientations. We conclude that either the spin susceptibility in this non-heavy-fermion sample is too small to measure by these means, or that our sample quality was not good enough. In view of other workers' data, we do not regard our null result as evidence against even parity superconductivity.

References

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