Experimental report

12/08/2021

Proposal: **5-51-571** **Council:** 4/2020

Title: The low-field susceptibility of the superconducting state of Sr2RuO4

Research area: Physics

This proposal is a new proposal

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Samples: Sr2RuO4

Instrument	Requested days	Allocated days	From	To
IN20	14	9	02/03/2021	15/03/2021

Abstract:

Sr2RuO4 has long been considered as a textbook example of a system where superconductivity develops from a strongly correlated Fermi liquid. It has generated considerable interest as a test of theory for many reasons including: (i) it has a relatively simple layered structure; (ii) very clean samples can be prepared and, (iii) its magnetic excitations are well-characterised. New NMR Knight shift and polarized neutron scattering (PNS) measurements of the susceptibility in the superconducting state have brought into question the accepted pairing wavefunction. Thus, in the last year numerous candidate superconducting states based on magnetically mediated pairing and ab-initio calculations have been proposed. Here we propose to make further PNS measurements to constrain the allowed superconducting states.

EXPERIMENT DIR-183 / 5-51-571 ON IN20 IN MARCH 2021
THE LOW-FIELD SUSCEPTIBILITY OF THE SUPERCONDUCTING STATE OF SR_2RUO_4

A. Abstract

Sr_2RuO_4 is a correlated transition metal oxide which forms in its normal state a Fermi liquid, shows strong spin-orbit coupling (SOC) which significantly modifies its Fermi surface [1] and undergoes a transition into an unconventional superconducting state at $T_c \approx 1.5\,\mathrm{K}$ and $H_{c2} \approx 1.47\,\mathrm{T}$. Whereas the previous picture of an odd-parity chiral-triplet state with $\mathbf{d} \parallel \hat{\mathbf{z}}$ is now rejected [2] the parity and the paring symmetry of the superconducting state is still unclear. In a previous polarised neutron scattering (PNS) experiment [3, 4] we observed analogue to ^{17}O-nuclear magnetic resonance (NMR) Knight shift K measurements [2, 5, 6] a reduction of the spin susceptibility χ_{spin} in the superconducting state Sr_2RuO_4 for magnetic fields H applied along the crystallographic a-axis. However our data suggested a rather large zero-field and zero-temperature residual $\chi_{\mathrm{spin}}(0)$ which appeared less prominent in the NMR K-measurements. Furthermore, we established that an earlier PNS experiment [7] suffered from bad statistics and thus failed to observe the change in χ_{spin}.
Throughout various techniques signatures are observed that the superconducting state in Sr_2RuO_4 undergoes a first-order transition for H near H_{c2} [8, 9]. This was recently also observed in the electronic specific heat [10] and in NMR [5] and hence it is also expected to be observable in PNS.
The technique we applied to study the χ_{spin} and to avoid screening by supercurrents is PNS which was pioneered by Shull and Wedgwood [11]. The aim of this experiment [12] was to further study the H-dependence of the χ_{spin} in the superconducting as well as normal state at low temperatures. The experiment hereby specifically focused on the low field regime to examine any residual χ_{spin} and on H H_{c2} to study a possible first-order transition. We observed a large first order transition but also find a residual $\chi(0)$ of almost 50% of the normal state susceptibility $\chi(n)$.

B. Experimental details

PNS [12] measurements were carried out on IN20 with the sample mounted in a dilution refrigerator with a 2.5 T magnet where magnetic fields were applied vertically and parallel to [100]. Any measurements were performed on the (011) Bragg reflection which mainly includes magnetic scattering from the Ruthenium side. The alignment, the calibration of the flippers and the polarisation of the beam were performed with the complete instrument whereas measurements of the flipping ratio on the sample to study χ were performed without the analyser to increase the flux. To improve the statistics, the data normalisation and corrections for fluctuations in the neutron beam flux in comparison to previous experiment [3, 4] the detector and the monitor were replaced by a diffraction detector and monitor, respectively. The data in the superconducting state were collected at 60 mK ($T \ll T_c$) and fields between 0.2 T to 1.2 T with $\mathbf{H} \parallel [100]$. Normal state data was collected at 60 mK and 1.5-2.5 T and for 2.5 T at various temperatures.

C. Results

The final results are mainly presented in Fig. 1 and Fig. 2.
At first the normal state was characterised. Values in good agreement with results from previous experiment [3, 4] were measured. A series of temperatures at 2.5 T were probed to search for contribution to the flipping ratio R by polarisation of nuclear isotopes of Ruthenium, Strontium and oxygen with non-zero nuclear spin [13–15]. Due to the small moments of the nuclei the polarisation is expected to be linear in H/T (small-H/high-T limit of the Brillouin function) a the applied H and T but no such dependency was observed.
The measurements in the superconducting state only show a small reduction with decreasing fields for $H < 0.8\,H_{c2}$ but show a large increase near H_{c2} which is consistent with a first-order transition due to Pauli limiting. Maki suggested that strong SOC might favour Pauli limiting [16] over orbital limiting which might be the case here. Our results point towards a residual susceptibility $\chi(0)$ of about 50% of the normal state susceptibility $\chi(n)$. The susceptibility here includes an orbital part estimated to up to 22% of $\chi(n)$ but χ_{spin} is subject to enhancement which counter-intuitive leads to a smaller $\chi_{\mathrm{spin}}(0)$. These two effects might partially or even almost completely cancel and hence $\chi(0)/\chi(n)$ might be a good estimate for $\chi_{\mathrm{spin}}(0)/\chi_{\mathrm{spin}}(n)$.
Sr_2RuO_4 exhibits strong SOC which couples the orbital and spin moments and create a pseudo-spin space [17]. This implies that PNS and NMR K-shift measurements in Sr_2RuO_4 probe the parity of the superconducting state rather than the paring symmetry as in a superconductor without SOC. It further implies that in spin-space the paring states are non-diagonal which results even for even-parity states in non-zero $\chi_{\mathrm{spin}}(0)$ induced by triplet components in spin-space. Our data generally favour an even-parity state with very strong SOC and a nodal gap structure but are also consistent with an odd-parity state.
Altogether the large residuum is consistent with three proposals which are depicted in Figure 1) where a large spin part is suggested either due to an odd-parity state with in-plane $\mathbf{d}$-vector, due to triplet contributions induced by the

spin orbit coupling in an even-parity state or due to scattering on impurities. Qualitatively we come to similar results as the electronic specific heat γ_e measurements and the NMR Knight shift K measurements but observe a smaller first order transition and a larger residual χ which is partially due to orbital contributions.

D. Conclusions and publication of this work

We have collected high quality polarised neutron scattering data with fields down to 0.2 T and flipping ratios down to $R-1 \approx 2 \times 10^{-}4$. The field dependence of the susceptibility in and outside of the superconducting phase were mapped out.
A reduced spin susceptibility with a large first-order transition and a large zero-field and zero-temperature residual term was observed alongside a field dependence which suggested a (near-)nodal gap structure. The results are consistent an odd-parity state or an even-parity with a large residual term due to spin-orbit coupling. The gap structure might be consistent with a nodal-s'-wave [17], a $d_{x^2-y^2}$-wave [17], a d_{xy}-wave [18] or also a $g_{xy(x^2-y^2)}$-wave.

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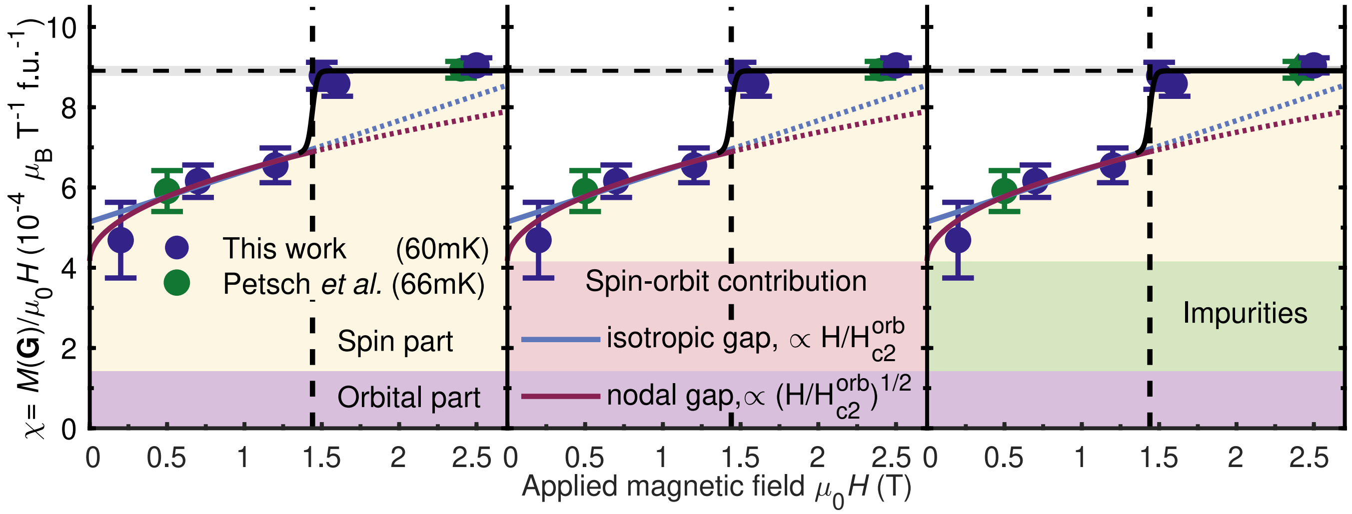

FIG. 1: Field dependence of the measured bulk susceptibility χ (blue) together with results from previous experiment (green) [3, 4]. The three panels (i-iii) describe different proposals which could explain the large non-zero residuum. Here, panel (i) suggests an odd-parity state, panel (ii) suggest an even-parity state with strong spin-orbit coupling and panel (iii) suggest residual density of states due to scattering on impurities.

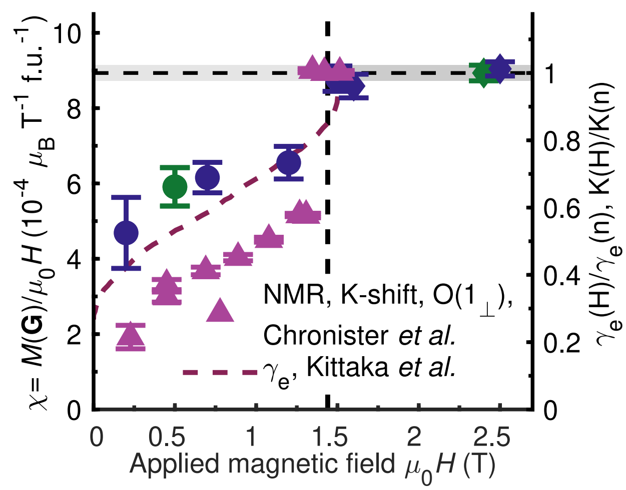

FIG. 2: The results are compared with the field dependence of the ^{17}O-NMR Knight shift K [5] and the linear coefficient of the electronic specific heat γ_e [10]. Both show qualitatively the same features but yield smaller low field values and smaller residual values. However, the K-shift shows a significantly larger first order transition which makes up for most of the difference.