Search for exotic magnetic phases with µSR

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Superconductors that break time-reversal symmetry (BTRS)



Two-component order parameter $|\Delta_{1(2)}| e^{i\varphi_1(2)}$ with double degenerate states: $\{|\Delta_1|, |\Delta_2|, \varphi_1 - \varphi_2 = \theta_{12}\} \rightarrow \{|\Delta_1|, |\Delta_2|, \varphi_1 - \varphi_2 = -\theta_{12}\}$

Degenerate order parameter Intrinsically $|\Delta_1| = |\Delta_2|$, $\theta_{12} = \pm \pi/2 \longrightarrow T_c = T_{BTRS}$ Examples: $p_x \pm i p_{yy}$, $d_{xz} \pm i d_{yz}$, ...



Non-degenerate or accidentally degenerate order parameter

 $|\Delta_1| \neq |\Delta_2|, \theta_{12} \neq 0, \pi$ fine tuning for $T_c = T_{BTRS}$ Examples: $s \pm is', s \pm id, d \pm ig, ...$



The degeneracy is lifted by any perturbation.

V. Grinenko et al., Nat. Phys. 17, 1254–1259 (2021).

V. Grinenko, S. Ghosh et al. Nat. Phys. 17, 748–754 (2021).

lift the degeneracy.

µSR investigations of Sr₂RuO₄



µSR is local probe and it is sensitive to all types of BTRS states!

- > Split superconducting and BTRS transition under uniaxial strain in Sr₂RuO₄;
- > Strain induced spin density wave (SDW) with magnetic moment ~ $0.1\mu_{\rm B}$;

V. Grinenko, S. Ghosh et al., Nat. Phys. 17, 748–754 (2021). V. Grinenko et al., Phys. Rev. B 107, 024508 (2023).

BTRS superconductivity under hydrostatic pressure.

V. Grinenko et al., Nat Commun 12, 3920 (2021).



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en





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Sr₂RuO₄ is multiband superconductor



Crystal structure

C. Hicks, MPI Dresden



S. Raghu et al., Journal of Physics: Conference Series 449, 012031 (2013).

Sr₂RuO₄ has a complex electronic structure, makes it similar to Fe-based superconductors.

Electronic orbitals relevant for conductivity

Some experimental evidences for multicomponent order parameter in Sr₂RuO₄





Luke, G. M. et al. Nature 394, 558-561 (1998)

Superconducting diode effect



M.S Anwar et al., Commun Phys 6, 290 (2023).

S.Ghosh et al., Nat. Physics (2020), S. Benhabib et al., Nat. Physics (2020)

Evidences against chiral superconductivity in Sr₂RuO₄



Scanning SQUID microscopy



Problems with interpretation Josephson effect:

S. Kashiwaya et al., PRB 100, 094530, S.-I. Suzuki et al., PRB 101, 054505 (2020),

The negative result can be explained by the suppression of superconductivity through surface reconstruction and surface magnetism.

C. A. Margues et al., arXiv:2005.00071 A. Kreisel et al., arXiv:2103.06188 R. Fittipaldi et al., Nat Commun. 12, 5792 (2021).

Are T_c and T_{TRSB} transitions split under uniaxial strain?

Missing cusp in T_c under strain





C. W. Hicks et al., Science 344, 283-285 (2014). A. Steppke, et al., Science 355, eaaf9398 (2017).



The reduction of the NMR Knight shift below T_{c}

A. Pustogow et al., Nature 574, 72 (2019).

Non-chiral p-wave!?

Sr₂RuO₄ is multiband superconductor



Variety of possible superconducting order parameters in Sr₂RuO₄

Multi-band nature of the Fermi surface results in a possibility of mixed pairing states with the pairing mediated by spin fluctuations, candidates: *s+id*, *d+ig*, *s+ip*

A. T. Rømer, et al., PRL 123, 247001 (2019); PRB S. A. Kivelson, *et al.*, npj Quantum Mat. 5, 43 (20

Alternatively, the orbital degree pairing originated from local inte

A. Ramires, and M. Sigrist, *Physical Review B* (20 H. G. Suh *et al.*, Phys. Rev. Research **2**, 032023(F A. Ramires, arXiv:2110.10621.

Mixed pairing or chiral



dx,+idv,



A chiral p-wave is excluded according to the NMR Knight shift measurements:

A. Pustogow *et al., Nature* 574, 72 (2019).
K. Ishida, *et al.*, J. Phys. Soc. Jpn. 89, 034712 (2020).
A. Chronister *et al.*, PNAS 118 (25) e2025313118 (2021).

How can we distinguish between chiral and non-chiral superconductivity using μ SR?



- $\tilde{\Gamma}_7 = E_g$: chiral *d*-wave ($d_{xz} \pm i d_{yz}$)
- > $T_{\rm c}$ and $T_{\rm BTRS}$ transitions must split under symmetry breaking uniaxial \vdash strain.
- Do not split with the hydrostatic pressure and isotropic disorder.



Accidentally degenerated sates:

$$\begin{split} \tilde{\Gamma}_1 &= A_{1g} \oplus A_{2g} : \quad s + ig \text{-wave} \\ \tilde{\Gamma}_2 &= A_{1g} \oplus B_{1g} : \quad s + id \text{-wave} \\ \tilde{\Gamma}_3 &= A_{1g} \oplus B_{2g} : \quad s + id' \text{-wave} \\ \tilde{\Gamma}_4 &= B_{1g} \oplus A_{2g} : \quad d + ig \text{-wave} \\ \tilde{\Gamma}_5 &= B_{2g} \oplus A_{2g} : \quad d' + ig \text{-wave} \\ \tilde{\Gamma}_6 &= B_{1g} \oplus B_{2g} : \quad d + id' \text{-wave} \end{split}$$



- T_c and T_{BTRS} transitions must split under uniaxial strain.
- Do split with the hydrostatic pressure and disorder.

V. Grinenko et al., Nat Commun 12, 3920 (2021).

- > Splitting of T_c and T_{BTRS} is expected for both: chiral and accidentally degenerated orders.
- > Important demonstration that T_c and T_{BTRS} are related to a different phase transitions.



Sr₂RuO₄ under the (100) strain





A. Steppke, et al., Science **355**, eaaf9398 (2017).

- > What is the effect of van Hove on BTRS state?
- Can van Hove induce magnetism?

Experimental setup for the µSR:



S. Ghosh *et al.*, Review of Scientific Instruments **91**, 103902 (2020).
V. Grinenko, S. Ghosh *et al. Nat. Phys.* **17**, 748–754 (2021).

*T*_c is measured by AC susceptibility.
 *T*_{BTRS} by zero-field μSR.

BTRS superconductivity of Sr₂RuO₄ under the (100) strain



-0.68 GPa

-0.75 GPa

-0.82 GPa

5

6



0.03

0

- \checkmark Significant splitting of $T_{\rm BTRS}$ and $T_{\rm c}!$
- BTRS state exists above van Hove \checkmark singularity!

V. Grinenko et al., Nat. Phys. 17, 748–754 (2021)

Temperature (K)

6

Strain induced SDW phase in Sr₂RuO₄





- > An incommensurate SDW phase well above the van Hove singularity with $T_N \sim 7$ K;
- > Small internal magnetic fields correspond roughly to **0.1** $\mu_{\rm B}$ (similar to Sr₃Ru₂O₇);
- > The SDW internal field is 100 times stronger than spontaneous fields due to BTRS superconductivity.

µSR measurements of Sr₂RuO₄ under uniaxial strain





Superconducting T_c and T_{BTRS} split by uniaxial strain \rightarrow two distinct phases!



Uniaxial strain induces spin density wave order (SDW) above the Lifshitz transition in odd with early theoretical predictions.



YS. Li et al., Nature 607, 276–280 (2022).

-0.2

-0.1

SDW phase detected by µSR is confirmed by elastocaloric effect measurements.

Spontaneous superconducting diode effect



M. S. Anwar et al., arXiv:2211.14626v1

✓ Split T_c and T_{BTRS} is confirmed by spontaneous superconducting diode effect measurements.

What is the evidence from other probes in favor of the µSR phase diagram?

Sr₂RuO₄ under the (110) strain, Sample A





Very weak suppression of T_c by the (110) strain consistent with previous studies.

Splitting of $T_{\rm BTRS}$ and $T_{\rm c}$ under the strain!



V. Grinenko et al., Phys. Rev. B 107, 024508 (2023).

Summary: splitting between T_c and T_{TRSB} under uniaxial strain





Electronic structure is very sensitive to <100> strain \rightarrow difficult to perform a quantitative analysis of the strain dependencies of $T_{\rm c}$ and $T_{\rm BTRS}$.

- ➤ The electronic structure is less affected by <110> strain → the splitting is due to lifted degeneracy between the components.
- > Fast suppression of T_{TRSB} under <110> stress indicates that the condensation energy associated with the TRSB phase can be small:

$$\frac{\Delta C_1}{T_{\rm c}} \frac{dT_{\rm c}}{d|\sigma|} = -\frac{\Delta C_2}{T_{\rm BTRS}} \frac{dT_{\rm BTRS}}{d|\sigma|}$$

V. Grinenko et al., Phys. Rev. B 107, 024508 (2023).

How can we distinguish between chiral and non-chiral superconductivity using μ SR?



- $\tilde{\Gamma}_7 = E_g$: chiral *d*-wave ($d_{xz} \pm i d_{yz}$)
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- Do not split with the hydrostatic pressure and isotropic disorder.





- At small concentrations the dominant effect of La on T_c is related to a pair breaking effect.
- La-doping results in isotropic in-plane disorder without anisotropic internal strain.

Accidentally degenerated sates:

$$\begin{split} \tilde{\Gamma}_1 &= A_{1g} \oplus A_{2g} : \quad s + ig\text{-wave} \\ \tilde{\Gamma}_2 &= A_{1g} \oplus B_{1g} : \quad s + id\text{-wave} \\ \tilde{\Gamma}_3 &= A_{1g} \oplus B_{2g} : \quad s + id'\text{-wave} \\ \tilde{\Gamma}_4 &= B_{1g} \oplus A_{2g} : \quad d + ig\text{-wave} \\ \tilde{\Gamma}_5 &= B_{2g} \oplus A_{2g} : \quad d' + ig\text{-wave} \\ \tilde{\Gamma}_6 &= B_{1g} \oplus B_{2g} : \quad d + id'\text{-wave} \end{split}$$



- > $T_{\rm c}$ and $T_{\rm BTRS}$ transitions must split under uniaxial strain.
- > Do split with the hydrostatic pressure and disorder.

V. Grinenko et al., Nat Commun 12, 3920 (2021).

La-doping is an ideal tool to study the effect of disorder on T_c and T_{BTRS}

N. Kikugawa et al., Phys. Rev. B **70**, 060508(R) (2004)

Time reversal symmetry breaking in Sr_{2-y}La_yRuO₄





1% of La doping suppresses T_c by 50% as compared

- → The normal state electronic specific heat is nearly the same for all samples, $\gamma_n \approx 40$ mJ/mol-K² -> the DOS are unchanged.
- Sharp superconducting transition -> homogeneous suppression of superconductivity by a pair braking effect.



 $T_{BTRS} = T_{c}$ in the sample with **a half of the disorder free** T_{c0} ;

Together with the splitting between T_{BTRS} and T_{c} under uniaxial strain

\rightarrow evidence in favor of a chiral superconductivity in Sr₂RuO₄

V. Grinenko et al., Nat Commun 12, 3920 (2021).

Theory: Splitting of the T_c and T_{TRSB} under disorder for accidentally degenerated orders



Accidentally degenerated sates:

- $\tilde{\Gamma}_1 = A_{1g} \oplus A_{2g}: \quad s + ig\text{-wave}$ $\tilde{\Gamma}_2 = A_{1g} \oplus B_{1g}: \quad s + id\text{-wave}$ $\tilde{\Gamma}_3 = A_{1g} \oplus B_{2g}: \quad s + id'\text{-wave}$ $\tilde{\Gamma}_4 = B_{1g} \oplus A_{2g}: \quad d + ig\text{-wave}$ $\tilde{\Gamma}_5 = B_{2g} \oplus A_{2g}: \quad d' + ig\text{-wave}$ $\tilde{\Gamma}_6 = B_{1g} \oplus B_{2g}: \quad d + id'\text{-wave}$
- > $T_{\rm c}$ and $T_{\rm BTRS}$ transitions must split under uniaxial strain.
- Do split with the hydrostatic pressure and disorder.

V. Grinenko et al., Nat Commun 12, 3920 (2021).







Fine-tuning is possible that transitions do not split with disorder.

Therefore, additional experiments are needed!

How can we distinguish between chiral and non-chiral superconductivity using μ SR?



- $\tilde{\Gamma}_7 = E_g$: chiral *d*-wave ($d_{xz} \pm i d_{yz}$)
- > $T_{\rm c}$ and $T_{\rm BTRS}$ transitions must split under symmetry breaking uniaxial \vdash strain.
- Do not split with the hydrostatic pressure and isotropic disorder.



Hydrostatic pressure preserves tetragonal symmetry *a* = *b*



D. Forsythe *et al.,* Phys. Rev. Lett. 89, 166402 (2002) N. Shirakawa *et al.,* Phys. Rev. B 56, 7890 (1997)

Accidentally degenerated sates:

$$\begin{split} \tilde{\Gamma}_1 &= A_{1g} \oplus A_{2g} : \quad s + ig\text{-wave} \\ \tilde{\Gamma}_2 &= A_{1g} \oplus B_{1g} : \quad s + id\text{-wave} \\ \tilde{\Gamma}_3 &= A_{1g} \oplus B_{2g} : \quad s + id'\text{-wave} \\ \tilde{\Gamma}_4 &= B_{1g} \oplus A_{2g} : \quad d + ig\text{-wave} \\ \tilde{\Gamma}_5 &= B_{2g} \oplus A_{2g} : \quad d' + ig\text{-wave} \\ \tilde{\Gamma}_6 &= B_{1g} \oplus B_{2g} : \quad d + id'\text{-wave} \end{split}$$

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- T_c and T_{BTRS} transitions must split under uniaxial strain.
- Do split with the hydrostatic pressure and disorder.

V. Grinenko et al., Nat Commun 12, 3920 (2021).

"Fine tuning" is impossible for both disorder and hydrostatic pressure!

The first µSR measurements of BTRS superconductivity under pressure







Suppression of superconductivity and time-reversal symmetry breaking under pressure.



 $T_{BTRS} \approx T_{c}$ due disorder and under hydrostatic pressure

Evidence for two component chiral superconductivity in Sr₂RuO₄.

V. Grinenko et al., *Nat Commun* **12**, 3920 (2021)

Magnetic field effect on the BTRS state





V. Grinenko, S. Ghosh et al., Nat. Phys. 17, 748–754 (2021).

- The BTRS transition is not seen in the applied magnetic field above H_{c2} in both Knight shift and muon depolarization rate.
- The new measurements Y. Maeno *et al.*, shows that in magnetic fields $T_c \sim T_{BTRS}$ up to H_{c2} .

Evidence against BTRS superconductivity in Sr₂RuO₄



Missing anomaly at T_{TRSB} in the specific heat under uniaxial strain



Y.-S. Li et al., PNAS 118 (10) e2020492118 (2021)

Elastocaloric effect



YS. Li et al., Nature 607, 276–280 (2022).

No evidence for the BTRS transition in the specific heat and electrocaloric effect measurements under uniaxial strain.

Is the enhanced relaxation seen in µSR related to superconductivity at all?

Can µSR provide a thermodynamic evidence for BTRS ?





Is the BTRS transition affects the vortex distribution in the sample when T_c and T_{BTRS} are split?

> The hypothesis is that divergent TRSB coherence length at T_{TRSB} results in long-range intervortex attraction, while a short-range interaction, mediated by superconducting coherence length, is always repulsive. This should lead to vortex clustering, which can be observed in the field distribution probed by muons for sufficiently weak pinning and low vortex density (more noticeable in smaller fields).

What do we expect from a regular vortex lattice?



J. Phys.: Condens. Matter 21 (2009) 075701





Figure 1. Example of a spatial distribution of the magnetic field $B(\mathbf{r})$ and the corresponding local magnetic field distribution $P_{id}(B)$ for an ideal hexagonal FLL determined by the NGL method. The parameters used for the calculations are $\lambda = 50$ nm, $\xi = 20$ nm, and $\langle B \rangle = 0.3B_{c2} \simeq 246.8$ mT, and intervortex distance a = 69.5 nm.

Figure 2. Local magnetic field distribution $P_{id}(B)$ for an ideal hexagonal FLL obtained using the NGL model for different values of λ , at fixed ξ and applied field $B_{app} \simeq \langle B \rangle$. The curves are normalized so that $\int P_{id}(B) dB = 1$. Note that the shape of $P_{id}(B)$ strongly depends on λ .

A Maisuradze et al

Sample C (spring cell nominally 300N)



At the beginning we had troubles to apply strain.

The spectra in TF looks as expected for the samples with $T_c \sim T_{BTRS}!$

What do we expect from in the case $T_{BTRS} < T_c$?





Sample with naturally split transitions at zero strain, zf and B||ab







> The significant splitting about $\Delta T \sim 0.4$ K at zero stain;

- > The measurements without pressure cell -> bg \sim 0;
- > For the field applied in the *ab*-plane the spectra don't show apparent anomalies around T_{TRSB} .

Sample with naturally split transitions at zero strain, zf and B||c





Sample with split transitions at zero strain, B||c





- We observe an increase of the field of the high-field component and the fraction with zero field close to the expected T_{TRSB} in applied field.
- In addition double structure of the spectra appears close to T_{TRSB} with noticeable high-field shoulder.

Zero-field measurements of Sr₂RuO₄ under uniaxial strain





- The applied uniaxial pressure ~ 0.5GPa with $T_c^{50\%}$ ~ 2.7K;
- TRSB transition temperature, $T_{\text{TRSB}} \sim 1.4$ K.

Can we verify under uniaxial strain the observation obtained on the sample with the split transitions?

Analysis of the sample volume fraction using TF measuremnts



Standard experiment: The field is applied above T_c , the sample is cooled to the base temperature, and the measurements are performed in steps while heating.



Pinning experiments:

changing to 100 G and measuring.

ZF cooling to T = 0.8K application B||c = 120 G,

The central peak close to the applied field is not related to the background since it's position shifts systematically with the temperature.

The background contribution estimated from the pinning experiments is about of 15% of the signal amplitude.

30



✓ The high-field component moved to the highest field near $T_{\text{TRSB}} \approx 1.4$ K.

 \checkmark Around this temperature, there is a jump at zero field fraction.

3

2

Temperature (K)

31

0.00





High-field fraction is not an artifact of Fourier transform





- The high-field component is not an artifact of fit or Fourier Transform;
- The well defined high-field component is absent at lower temperatures.

TF-µSR data in 100 and 200 G applied fields ||c







- The background contribution cannot account the two-peak structure of the signal;
- Both peaks coming from the sample indicating two well-defined regions with different intervortex distances.

TF-µSR data in 100 G applied fields ||c





 \geq 2 peak structure develops around BTRS transition.



Summary



