Broken Time-reversal Symmetry in $\text{Sr}_2\text{RuO}_4$ and Other Novel Superconductors

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Peter Beyersdorf (San Jose State) - Sagnac design
Steve Kivelson (Stanford) - theoretical ideas, coffee partner!
Outline:

1. Time reversal breaking effects in unconventional superconductors
2. $\text{Sr}_2\text{RuO}_4$
3. Magneto-optical effects in solids
4. The Sagnac “Magnetometer”
5. The loopless version
6. Searching for Time-reversal symmetry breaking signals in $\text{Sr}_2\text{RuO}_4$
7. New measurements: High-Tc, Heavy Fermions, …
8. Conclusions
Unconventional superconductivity

In general $\Psi(\vec{k})$ Depends on $\vec{k}$

$$\langle \Psi(\vec{k}) \rangle_{\text{Fermi surface}} = \Psi_0$$

Conventional superconductors

$L = 0$ (isotropic)

Momentum average is harmless

For non magnetic impurities:

Anderson Theorem

$$\Psi_0 \neq 0$$

Unconventional superconductors

$L > 0$ (anisotropic)

$\Psi(\vec{k})$ depends on $\vec{k}$. Momentum average results in destructive interference.

$$\Psi_0 = 0$$

Suppression of superconductivity

A Hallmark of unconventional superconductors is their sensitivity to scattering, i.e. interference.
Search for Broken Time Reversal Symmetry in Unconventional Superconductors

For High Tc Superconductors:
- anyon superconductivity [Historically was first search]
  \[ d \chi \pm i d_{xy}, d \chi s_{xy}, \text{etc.}. \]
  D-density wave (staggered-flux state: Laughlin, Chakravarty, Lee, etc.)
  Loop-Current Order (does not break translation symmetry: C.Varma)

p-wave Superconductors:
- \( \text{Sr}_2\text{RuO}_4 \)
- \( \text{UPt}_3, \text{PrOs}_4\text{Sb}_{12}, \text{and other heavy fermions} \)
- \( \text{(TMTSF)}_2\text{ClO}_4 \text{ and other organic superconductors} \)

Ferromagnetic superconductors
- \( \text{ErRh}_4\text{B}_4, \text{UGe}_2, \text{…} \)

* A significant feature of the mixed symmetry states is that they may produce spontaneous currents and magnetic moments which can be measured using appropriate experimental techniques.
**Sr$_2$RuO$_4$**

Quasi 2-dimensional
Strongly correlated Fermi liquid
$T_C = 1.5$ K

Sr$_2$RuO$_4$ is a layered perovskite isostructural with La$_{2-x}$Ba$_x$CuO$_4$.


$T_C$ (as discovered) $\sim 0.93$ K
Sr$_2$RuO$_4$ is a strongly correlated Fermi liquid:

The superconductivity of Sr$_2$RuO$_4$ condenses from a **metallic state that is a strongly correlated two-dimensional Fermi liquid**. (Low temperature $T^2$ of resistivity, quantum oscillations)

Early measurements indicated that Sr$_2$RuO$_4$ shows evidence of strong **triplet($S=1$) correlations in the normal state**. (e.g. similarity to ferromagnetic SrRuO$_3$)

Fermi liquid parameters and $S=1$ bear strong quantitative similarity to those of $^3$He$^*$.  

* Rice and Sigrist, 1995
Early evidence for unconventional odd parity pairing

Destruction of superconductivity by nonmagnetic impurities


Knight-shift does not Change below $T_c$.

Phase sensitive measurements supporting odd parity:


Phase sensitive measurements:

Results consistent with odd parity!
What is the actual symmetry of the order parameter of \( \text{Sr}_2\text{RuO}_4 \)?

Given:

1. Spin triplet pairing
2. 4-fold symmetry with in-plane (2D) pairing
3. Weak coupling superconductor (low \( T_c \), tunneling)
4. Some Spin-orbit coupling

Which state is realized?

**Generalized order parameter**

\[
\Psi(\vec{k}) = \left\langle \psi \left| c_{\vec{k}s} c_{-\vec{k}'s'} \right| \psi \right\rangle = \Phi(\vec{k}) \cdot \chi(s,s')
\]

Recall that \(\Delta(k) \propto \Psi(k)\) (the superconducting gap function)

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**Even parity, spin singlet - order parameter is a scalar**

\[
\hat{\Delta}_{\vec{k}} = \begin{bmatrix}
0 & \Delta_{\uparrow\downarrow} \\
\Delta_{\downarrow\uparrow} & 0
\end{bmatrix} = \begin{bmatrix}
0 & \Delta(\vec{k}) \\
-\Delta(\vec{k}) & 0
\end{bmatrix} = i\sigma_y \Delta(\vec{k})
\]

\(\Delta(-\vec{k}) = \Delta(\vec{k})\)

---

**Odd parity, spin triplet - order parameter is a vector**

\[
\hat{\Delta}_{\vec{k}} = \begin{bmatrix}
\Delta_{\uparrow\uparrow} & \Delta_{\uparrow\downarrow} \\
\Delta_{\downarrow\uparrow} & \Delta_{\downarrow\downarrow}
\end{bmatrix} = \begin{bmatrix}
-d_x + id_y & d_z \\
d_z & d_x + id_y
\end{bmatrix} = i\sigma_y \hat{d}(\vec{k}) \cdot \hat{\sigma}
\]

\(\hat{d}(-\vec{k}) = -\hat{d}(\vec{k})\)
Classification of (unitary) states:

\[ \vec{J} = \vec{S} + \vec{L} \]

<table>
<thead>
<tr>
<th>( \Gamma )</th>
<th>( J )</th>
<th>( J_z )</th>
<th>( \vec{d}/\Delta_0 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \Gamma_1^- )</td>
<td>0</td>
<td>0</td>
<td>( \hat{x}k_x + \hat{y}k_y )</td>
</tr>
<tr>
<td>( \Gamma_2^- )</td>
<td>1</td>
<td>0</td>
<td>( \hat{x}k_y - \hat{y}k_x )</td>
</tr>
<tr>
<td>( \Gamma_3^- )</td>
<td>2</td>
<td>1±2</td>
<td>( \hat{x}k_x - \hat{y}k_y )</td>
</tr>
<tr>
<td>( \Gamma_4^- )</td>
<td>2</td>
<td>2±2</td>
<td>( \hat{x}k_y + \hat{y}k_x )</td>
</tr>
<tr>
<td>( \Gamma_5^- )</td>
<td>1</td>
<td>1±1</td>
<td>( \hat{z}(k_x \pm ik_y) )</td>
</tr>
</tbody>
</table>

Which state is realized?

Suggested preferred state:

\[ S_z = 0 \]

\[ L_z = \pm 1 \]

This is a chiral state with orbital magnetic moment and degeneracy = 2

Time Reversal Symmetry is Broken!
Is this an example of orbital magnetism?

Can we measure a spontaneous magnetization?

\[ \overrightarrow{M} = ? \]

NO! Because of Meissner Effect! \( \Rightarrow \overrightarrow{M} = 0 \)

In general no spontaneous magnetic moment due to compensating Meissner currents.

However:
sample will always contain surfaces and defects at which the Meissner screening of the TRS-breaking moment is not perfect, and a small magnetic signal is expected.
Muon spin rotation as local measurement:

Observation of a spontaneous extra relaxation of the spin-polarization function below the superconducting transition temperature.

Estimated local field: $\sim 0.5$ Oe

However:
1. The effect was isotropic
2. Signal could come from other sources

Bulk measurements are needed which do not depend on defects in the superconductor:

**Magneto-Optical-like Measurements!**

- **Magnetization ➔** Splitting of spin-states (however, no asymmetry between LCP and RCP)
- **Spin-orbit interaction ➔** Splitting of orbital states
- **Absorption of circular polarization ➔** Induction of circular motion of electrons
  ➔ different phase and amplitude for LCP and RCP
  \[ \tilde{n}_R \neq \tilde{n}_L \quad (\tilde{n} = n + i\kappa) \]
- Condition for large magneto-optical response:
- Presence of strong (allowed) transitions involving elements with large spin-orbit interaction

\[
\begin{align*}
\tilde{n}_R & \neq \tilde{n}_L \\
\end{align*}
\]
In the optical regime we cannot define a magnetic susceptibility.

We therefore set $\mu=1$ and describe the behavior of the electromagnetic waves in the matter by $\varepsilon(\omega)$ only, or equivalently by $\sigma(\omega) = i \omega \varepsilon(\omega)$.

The general form of the conductivity for a cubic lattice:

$$
\begin{pmatrix}
\sigma_{xx} & \sigma_{xy} & 0 \\
-\sigma_{xy} & \sigma_{xx} & 0 \\
0 & 0 & \sigma_{zz}
\end{pmatrix}
$$

$$
\sigma_{ij} = \sigma'_{ij} + i \sigma''_{ij}
$$
Because of the axial symmetry, the index of refraction for right and left circularly polarized light is related to the complex optical conductivity by

\[ \varepsilon_{R,L}(\omega) = (n_{R,L} + i\kappa_{R,L})^2 = 1 + i\frac{4\pi\sigma_{R,L}}{\omega} \]

Where:

\[ \sigma_{R,L} = \sigma_{xx} \pm i\sigma_{xy} \]

\[ J_{R,L} = J_x \pm iJ_y \]

For example, the imaginary part of the off-diagonal conductivity is:

\[ \sigma''_{xy}(\omega) = \frac{\pi e}{4h}\sum_n\sum_m \left( |\langle n | J_R | m \rangle|^2 - |\langle n | J_L | m \rangle|^2 \right) \times \left[ \delta(\omega_{mn} - \omega) + \delta(\omega_{mn} + \omega) \right] \langle n | \hat{\rho} | n \rangle \]

asymmetry due to magnetization  
Allowed transitions  
Ground state population
**Faraday Effect:**

Assume a plane wave with a wavelength in vacuum: \( \lambda_0 \)

The wavelength of its circular components in the medium will be: \( \lambda_0 / n_R \) and \( \lambda_0 / n_L \)

At \( z=0 \) the wave is linearly polarized along \( x \)

Then: 
\[
\vec{E} = \frac{1}{2} E_0 e^{i(\omega t - k z)} [\hat{x} \cos(\delta/2) + \hat{y} \sin(\delta/2)]
\]

\[
\theta_F = \left( \frac{\delta}{2} \right)_{z=L} = \frac{\pi \ell}{\lambda_0} (n_R - n_L)
\]

\[
\theta_F = -\frac{2\pi \ell}{c/n} \frac{\sigma'_{xy} + \sigma''_{xy}}{n^2 + \kappa^2} \approx -\frac{2\pi \ell}{cn} \sigma'_{xy}(\omega)
\]

\( \kappa \ll n \)
Kerr Rotation:

Consider a Polar Kerr Effect at normal incidence

\[
\frac{E_r}{E_0} \equiv r = |r| e^{i\phi} = -\frac{(n + i\kappa) - 1}{(n + i\kappa) + 1}
\]

\[
\frac{r_R}{r_L} = \left| \frac{r_R}{r_L} \right| e^{i(\phi_R - \phi_L)}
\]

After reflection the complex amplitudes are different. The polarization is now elliptical with the major axis rotated by:

\[
\theta_K = -\frac{1}{2} (\phi_R - \phi_L) \approx -\text{Im}\left( \frac{(n_R + i\kappa_R) - (n_L + i\kappa_L)}{(n_R + i\kappa_R)(n_L + i\kappa_L) - 1} \right)
\]

In the last equality we used a small phase difference and small difference of the \( n \)s.

For small \( \kappa \):

\[
\theta_K = \frac{4\pi}{n(n^2 - 1)\omega} \sigma_{xy}''(\omega)
\]
Example:

Kerr effect of thick film Ferromagnetic $\text{SrRuO}_3$:

Note size of effect
In the range of
~ 10 Millirad !!!

For some ferromagnets $\theta_K$ can be of order $\sim\text{rad}$!
We can distinguish between magneto optic signal (Kerr and Faraday) from depolarization effects if we measure the difference between a light beam with its time reversal counter part beam.
Considerations for the experiment:

1. Expected signal is very small (some estimates gave $\theta_K \sim 10^{-10}$ rad).
2. Linear birefringence and optical activity may be present with much larger signal!
3. Comparing two beam traveling opposite in time can reveal the TRSB effect.
4. An interferometric detection is preferred.

A simple cross polarization method will not be enough*!

* Note that for searching for TRSB no modulation is possible!
Solution:

The Sagnac Effect
A Sagnac Loop at rest is reciprocal!

\[ \Delta \phi = \frac{2\pi}{\lambda} \frac{4A}{c} \Omega \]
Fiber-optic implementation
Example: Earth Rotation

\[ \Delta \phi = \frac{2 \pi L D}{\lambda c} \Omega \]

\( D = 20 \text{ cm} \)
\( \lambda = 1.06 \mu \text{m} \)
\( \Omega = \frac{2 \pi}{24 \cdot 3600} \)
\( L = 1 \text{ km} = 10^5 \text{ cm} \)

(optional, as we did, partially point it to have exactly 100 \( \mu \text{rad} \))
The Sagnac magneto-optic device
Use Sagnac to measure magnetization

\[ \phi_{nr} = \tan^{-1} \left[ \frac{J_2(2\phi_m)I_\omega}{J_1(2\phi_m)I_{2\omega}} \right] \]

Detected intensity graphs
Search for anyons - round-1 (1990-1992)

$YBa_2Cu_3O_{7-\delta}$
Optimally doped $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ Thin Films in Transmission:

Results: No effect to within 1 $\mu$rad

No shot noise limit. Main problems: Drift, need for higher power ($\sim 1$ mW)

The loopless interferometer

If we fold the system along the dashed line, we measure in reflection. If we use the two axes of the fiber - we do not need a loop.
There is no Optic-viewport, allows lower temperatures (0.3 K)

It rejects depolarization effects such as linear birefringence.

Both DC and AC performance are shot-noise limited.

Jing Xia, Peter Beyersdorf, M. M. Fejer, and A. Kapitulnik, Appl. Phys. Lett. 89, 062508
Setup

Optics
Drift can be as small as 20 nrad over a day.

Performance:

- **Noise**
  - Shot-noise-limited above 10µW

- **Drift**
  - Integrate over 10s
  - Integrate over 10 min
  - Drift can be as small as 20 nrad over a day.

- **Temperature**
  - ±20 nanorad!

- **Check of conventional superconductor - Al**
  - Superconducting transition 1.2K
Kerr effect measurements of ferromagnetic
Transition in SrRuO$_3$

Polar Kerr effect from a 30 nm SrRuO$_3$ thin film. (a) Kerr rotation in zero magnetic field with temperature down to 0.5 K. (b) Kerr rotations of the same sample measured in different cool-downs in zero fields. (c) Kerr rotation in a saturation field of 200 Oe.

Back to $\text{Sr}_2\text{RuO}_4$

Beam size $= 20 \mu\text{m}$
Incident optical power $= 0.7 \div 2 \mu\text{W}$

Zero field cool

Beam size = 20 µm
Incident power = 0.7 - 2 µW

Dashed line is guide to the eye

Sign of zero-field-cool data is random

Maximum Kerr rotation of zero-field-cool ~ 65 nanorad
Some zero field cool change sign

Variation of sign with successive cooldown, and change of sign suggest that domain size is of order of beam size.
Train the chirality with magnetic field:

- Cool in $H=+97$ Oe
- Warm up in $H=0$

- Cool in $H=-47$ Oe
- Warm up in $H=0$

Last two points before field switched to zero.

Dashed lines are guide to the eye.
Dependence on incidence power

cool in H=+97 Oe, Warm up in zero field

Incident power = 0.7 µW
Incident power = 6 µW

No power dependence!
More on temperature dependence

![Graph showing temperature dependence with a dashed line and data points labeled with a notation for 100 nanorad and a symbol for Tc.]
Measurements are consistent with $\theta_K \propto \Delta_0^2$
Minimum training field

Fields below $\sim 5$ Oe do not affect the sign of the chirality.

A minimum field between 5 Oe and 10 Oe* is needed to train the sign of the chirality.

* Note that $H_{c1} \sim 7 \div 10$ Oe
Phase sensitive measurements: evidence for $p_x \pm ip_y$

**Dynamical Superconducting Order Parameter Domains in Sr$_2$RuO$_4$**

Francoise Kidwingira, J. D. Strand, D. J. Van Harlingen, Yoshiteru Maeno

- Interference patterns consistent with $p_x \pm ip_y$
- Switching effects consistent with surface domains of order $\sim 0.5 \, \mu m$

*Fig. 4.* (A) Graphical representation of an SRO crystal with parallel chiral domains showing the order parameter phase winding in opposite directions. The phase difference between domains, $\delta$, is zero in one tunneling direction and $\pi$ on the orthogonal face. (B and C) Computer simulations of the diffraction patterns for junctions on orthogonal crystal faces with 10 parallel domains of random size, compared with measurements on those junctions.
Some theory:

Start with the lagrangian:

\[
L = \left( \begin{array}{cc}
  i\partial_t + \nabla^2/2m + \mu & i(\nabla \cdot \Psi + \Psi \cdot \nabla)/2 \\
  i(\nabla \cdot \Psi^* + \Psi^* \cdot \nabla)/2 & i\partial_t - \nabla^2/2m - \mu
\end{array} \right)
\]

where:

\[
\Psi = \Delta_x \hat{x} + i \Delta_y \hat{y}
\]

Calculate the off-diagonal part of the conductivity:

The Kerr angle:

\[
\theta_K = \frac{4\pi}{n(n^2 - 1)\Omega d} \sigma''_{xy}(\Omega)
\]

\[
\theta_K = \frac{2\pi}{n(n^2 - 1)} \frac{e^2}{d} \frac{\Delta_0^2}{(\hbar\Omega)^3} \propto (T_c - T)
\]

~ 200 nanorad!

* May have a problem when a pure system is considered due to Meissmer effect.
However:

Among other consequences of $p^{\uparrow \downarrow}$ is the existence of edge currents and currents between domain walls.

Upper limit on spontaneous supercurrents in $\text{Sr}_2\text{RuO}_4$


In conclusion, scanning magnetic microscopy measurements place quite severe limits on the size of edge currents and/or on domain sizes in $\text{Sr}_2\text{RuO}_4$. The different experimental results taken as evidence for $p_x + ip_y$ pairing come to quite different conclusions about domain sizes. Since there are now detailed predictions for the field profile in the vicinity of domain walls in the bulk, muon spin resonance could now, in principle, provide detailed information about the validity of these predictions as well as quantitative information about the density of domains in the bulk.

No detected edge currents!
Summary of observations:

- Maximum signal is ~65 - 100 nanorad
- Signal onsets at $T_C$
- Temperature dependence of signal can be fitted with either linear or quadratic dependence on the gap.
- Chirality can be trained with a magnetic field. A minimum field is needed.
- Domain size is large, of order beam size
  - Zero-field cool show some fluctuations
- Signal cannot be explained by trapped flux
  - max. zero-field cool signal equals field cool
- There is no power dependence on the size of the signal.
- We need to understand why there are no edge currents!