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Using Muons to Uncover the Behavior of Superconducting Electron Pairs

Hidden Pitfalls Caused by Nearby Superconductors

Summary

Quantum materials, which have attracted growing attention in recent years, are materials whose macroscopic properties emerge from quantum-mechanical effects. Superconductors represent a prime example. Among them, so-called *unconventional superconductors*—which cannot be explained within the framework of standard theory—such as cuprate high-temperature superconductors, are a central focus of modern fundamental research. Superconductivity was discovered in the ruthenium oxide Sr_2RuO_4 about 30 years ago and is considered a prototypical example of an unconventional superconductor.

For many years, it had been believed that this material realizes an innovative state known as *spin-triplet superconductivity*, in which electron pairs retain magnet-like properties, namely a total spin of one, and can transport quantum information without electrical resistance. However, recent nuclear magnetic resonance (NMR) experiments have produced results that overturn this long-standing conclusion, making independent verification using other experimental techniques essential.

A research group led by Dr. Hisakazu Matsuki (Specially Appointed Assistant Professor at the Toyoda RIKEN–Kyoto University Collaboration Center, TRiKUC, Kyoto University; currently Assistant Professor at the Institute for Chemical Research, Kyoto University), Professor Yoshiteru Maeno (TRiKUC), Dr. Rustem Khasanov (Paul Scherrer Institute, Switzerland), and graduate student Kosuke Yuichi (Institute for Solid State Physics, The University of Tokyo) have now introduced a new approach to muon spin resonance experiments. Using this method, they have conclusively demonstrated that the superconductivity of Sr_2RuO_4 can be consistently explained by *spin-singlet superconductivity*.

In the course of this study, the researchers also uncovered an unexpected pitfall in many previous muon-based experiments on superconductors. When multiple superconducting single-crystal samples are arranged side by side to efficiently irradiate them with a muon beam, stray magnetic fields leaking from neighboring samples exhibiting the Meissner effect can generate significant spurious signals.

This work is expected to further advance the study of superconductors using muon-based magnetic resonance techniques, providing insights that are complementary to those obtained from nuclear magnetic resonance. These research results will be published online on February 9, 2026, in *Physical Review Letters*, a journal published by the American Physical Society.

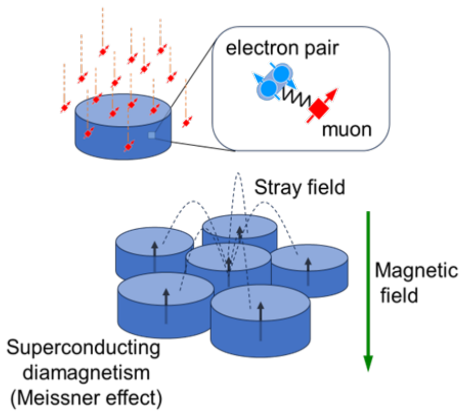

Fig. 1. Principle of probing the state of electron pairs using muons implanted into a superconductor. Arranging multiple samples side by side can cause problems due to stray magnetic fields.

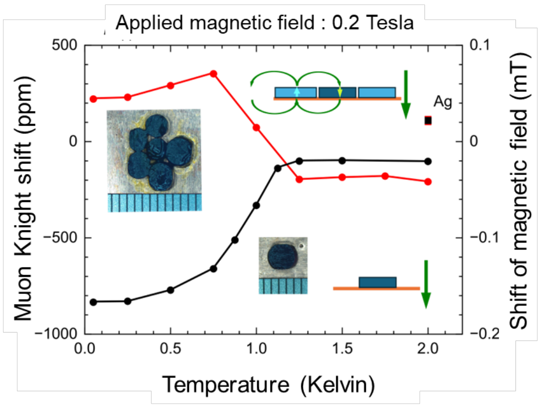

Fig. 2. A pitfall of a conventional experimental method. When multiple crystals are arranged side by side (red), a signal opposite to the intrinsic one is generated. Using one crystal (black) yields the correct signal.

1. Background

Superconductivity in the ruthenium oxide Sr_2RuO_4 was discovered in 1994 by Maeno and co-workers. This material shares the same crystal structure as the copper-oxide high-temperature superconductors and exhibits unconventional superconductivity that is characteristic of quantum materials. Because the properties of its normal (non-superconducting) state are well understood both experimentally and theoretically, and because extremely high-quality single crystals can be grown, Sr_2RuO_4 has long been regarded as an important model system for fundamental studies of superconductivity.

Despite this, the symmetry of its superconducting state remains unresolved and has been the subject of intense debate for many years. In 2019, nuclear magnetic resonance (NMR) measurements probing the spin state of superconducting electron pairs produced results that overturned conclusions deduced from early experimental measurements. Since then, a variety of studies based on new perspectives have been reported; nevertheless, the fundamental nature of the superconducting state—namely, the symmetry of the superconducting order parameter—has yet to be clarified [1, 2].

Current research efforts focus on several key issues including: (1) Whether the spin state can be definitively identified using methods complementary to NMR, in which sample heating due to electromagnetic pulses had posed challenges in earlier experiments. (2) Whether the spontaneous internal magnetic fields that emerge in the superconducting state, even in the absence of an external magnetic field, indicate time-reversal symmetry breaking (TRSB). (3) How to resolve the apparent inconsistencies between changes in sound velocity across the superconducting transition temperature T_c and changes in T_c under strain [3].

2. Research Methods and Results

In this study, we successfully determined the spin state of superconducting electron pairs using magnetic resonance based on muons (muon spin rotation/relaxation/resonance, μSR), in which elementary particles known as muons are implanted into a superconductor. High-quality single crystals of the ruthenium oxide Sr_2RuO_4 grown at Kyoto University were investigated using a newly developed μSR spectrometer that started its operation in 2022 at the Paul Scherrer Institute (PSI) in Switzerland. This state-of-the-art instrument enabled precise measurements of extremely small changes in the internal magnetic fields in the superconducting state when immersed in an external field.

These changes are known as the *muon Knight shift* and provide crucial information on how electrons form pairs and enter the superconducting state. As described below, by avoiding previously overlooked experimental pitfalls and by introducing a new measurement protocol that combines μSR with complementary measurements using a superconducting quantum interference device (SQUID), we clearly observed a reduction of the Knight shift upon entering the superconducting state. This finding provides compelling evidence that the superconductivity in Sr_2RuO_4 can be consistently explained in terms of *spin-singlet pairing*.

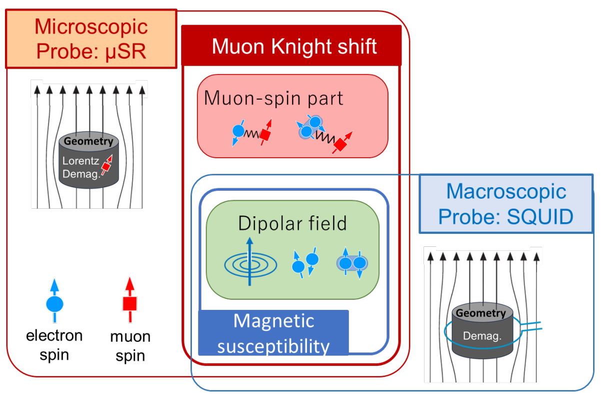

Fig. 3. The combination of muon spin rotation (μSR) experiments and superconducting quantum interference device (SQUID) measurements provides microscopic insights into the behavior of superconducting electrons.

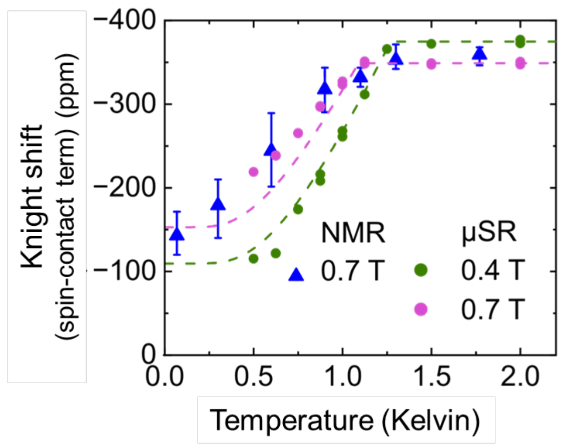

Fig. 4. Muon spin rotation (μSR) enabled us to verify the results with improved precision compared with widely used nuclear magnetic resonance (NMR) techniques.

The new μSR spectrometer allows independent measurements of signals from a silver reference sample, used for high-precision magnetic field calibration, and from the superconducting sample itself, leading to a dramatic improvement in the sensitivity to internal field changes. This advance revealed a serious pitfall in a commonly used experimental practice, in which multiple small crystals are arranged side by side to enhance signal intensity. Stray magnetic fields originating from the Meissner effect in neighboring superconducting crystals can generate spurious signals that appear in the μSR data despite not reflecting intrinsic properties of the material (Figs. 1 and 2).

In this study, we performed measurements using one single crystal and combined a microscopic technique—the muon Knight shift—with a conventional macroscopic measurement of dc magnetic susceptibility on the same sample. This unique approach allowed us to isolate the signal arising purely from the spin–spin interaction between muons and electrons (Fig. 3). As a result, we obtained highly precise results under various magnetic fields, which are in excellent agreement with recent NMR findings (Fig. 4).

3. Broader Impact and Future Perspectives

This study is expected to further advance research on superconductors using muon spin rotation/relaxation (μSR) as a complementary technique to nuclear magnetic resonance (NMR), which employs atomic nuclei as probes. The technical issues identified in this work—previously overlooked—will require careful consideration not only in superconductivity research but also in studies of magnetic materials, particularly as experimental precision continues to improve.

Fundamental research on unconventional superconductors is directly linked to the development of new experimental techniques and theoretical frameworks for quantum materials. In NMR experiments, heating effects can become problematic in superconductors with extremely low electrical resistance. In this context, μSR is expected to play an increasingly important complementary role in superconductivity research.

As a next step, for ruthenium-oxide superconductors, we plan to investigate superconducting states with broken time-reversal symmetry by conducting muon spin-relaxation experiments under no external magnetic field.

[1] Y. Maeno, A. Ikeda & G. Mattoni, "*Thirty years of puzzling superconductivity in Sr_2RuO_4*", Nature Physics **20**, 1712–1718 (2024). DOI: 10.1038/s41567-024-02656-0

[2] Y. Maeno, S. Yonezawa & A. Ramires "*Still Mystery after All These Years --Unconventional Superconductivity of Sr_2RuO_4--*", Journal of the Physical Society of Japan **93**, 062001-1-45 (2024). DOI: 10.7566/JPSJ.93.062001

[3] G. Mattoni, T. Johnson, A. Ikeda, S. Paul, J. Bobowski, M. Sigrist, Y. Maeno, "*Direct evidence for the absence of coupling between shear strain and superconductivity in Sr_2RuO_4*", Nature Communications. **17**, 700 (2026). DOI: 10.1038/s41467-025-67307-1

4. Research Project

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<Researcher's comment>

Fully understanding unconventional superconductivity is still an extremely challenging goal in modern physics. Our findings highlight that even well-established experimental approaches may involve unexpected technical pitfalls that have gone unnoticed, even by experts. Addressing such long-standing questions requires patience and sustained effort. With the key open issues in ruthenium-oxide superconductors now clearly identified, we are committed to resolving them step by step toward a final understanding. (Maeno)

<Paper title authors>

Title：Muon Knight shift as a precise probe of the superconducting symmetry of Sr_2RuO_4

Authors：Hisakazu Matsuki, Rustem Khasanov, Jonas A. Krieger, Thomas J. Hicken, Kosuke Yuchi, Jake S. Bobowski, Giordano Mattoni, Atsutoshi Ikeda, Ryutaro Okuma, Hubertus Luetkens, and Yoshiteru Maeno

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[Glossary]

Muon (μ)

A muon is a fundamental particle classified as a lepton, like the electron. It carries the same electric charge as the electron but has a mass about 207 times larger and an average lifetime of approximately 2.2 microseconds, after which it decays into an electron (or positron) and neutrinos.

In recent years, techniques known as *muon radiography (muography)*, which exploit the high penetrating power of muons, have been developed to non-destructively visualize the internal structures of large objects—such as pyramids, volcanoes, and nuclear reactors—in a manner similar to X-ray imaging.

Muon Spin Rotation/Relaxation/Resonance (μSR)

Muon spin rotation/relaxation/resonance (μSR) is an experimental technique that utilizes the rotation and relaxation of the spin of muons. In a magnetic field, the muon spin undergoes precession at a characteristic frequency. By analyzing the precession frequency and the time evolution of the spin polarization, μSR allows researchers to probe the local magnetic field experienced by implanted muons, including its magnitude, spatial inhomogeneity, and the time scale of magnetic fluctuations.

In this study, positively charged muons were implanted into a superconductor, and the distribution of positrons emitted during muon decay was analyzed to extract microscopic information on the spin state of superconducting electron pairs.

Superconducting Quantum Interference Device (SQUID)

A superconducting quantum interference device (SQUID) is an extremely sensitive magnetic sensor based on a superconducting ring incorporating Josephson junctions. SQUIDs are widely used in many fields, including magnetoencephalography and magnetocardiography for measuring weak magnetic signals from the brain and heart, research on magnetic materials, seismology, and the control of qubits in quantum computers.

SQUIDs operate by exploiting the quantum nature of superconductivity, in which the magnetic flux penetrating a superconducting ring is quantized in integer multiples. As a result, the electric current flowing through the ring varies periodically with magnetic flux, enabling the detection of extremely small changes in magnetic fields with high precision. In research on superconductors and magnetic materials, SQUID-based systems combined with superconducting coils are widely used in instruments that allow precise control of magnetic field and temperature.

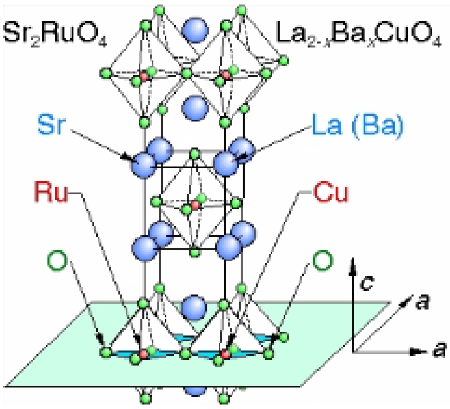
Unconventional Superconductivity

Superconductivity is a phenomenon in which electrons in a metal exhibit zero electrical resistance at low temperatures. It was discovered in 1911 by the Dutch physicist Heike Kamerlingh Onnes. The temperature below which superconductivity occurs is called the superconducting critical temperature (T_c). Below T_c, electrons form pairs, leading to a quantum condensed state analogous to Bose–Einstein condensation.

This mechanism is explained by the Bardeen–Cooper–Schrieffer (BCS) theory, which has long been regarded as the standard theory of superconductivity. In contrast, superconductivity in which the nature of electron pairing or interactions cannot be adequately explained within the BCS framework is referred to as *unconventional superconductivity*. Representative examples include high-temperature copper-oxide (cuprate) superconductors, heavy-fermion superconductors containing elements such as cerium or uranium, iron-based superconductors, and ruthenium-oxide superconductors.

Ruthenium-Oxide Superconductors

Ruthenium-oxide superconductors have the same crystal structure as cuprate high-temperature superconductors, with copper atoms replaced by ruthenium atoms (see figure). Although their superconducting critical temperature Tc is relatively low, ranging from approximately 1.5 to 3.5 kelvin, these materials exhibit strong electron–electron interactions and complex electronic structures involving multiple electron bands. Owing to these characteristics, they are classified as unconventional superconductors and have attracted particular attention as important platforms for fundamental research on functional quantum materials.



Spin-Singlet Electron Pairs

Electrons possess a quantum-mechanical property called spin, with a magnitude of 1/2. A *spin-singlet* electron pair is a pair of electrons whose spins cancel each other, resulting in a total (composite) spin of zero. In a classical picture, this corresponds to two spins aligned antiparallel to each other.

In contrast, superconductors composed of *spin-triplet* electron pairs, in which the two electron spins are aligned parallel, have been theoretically proposed and several experimental candidates have been reported; however, no definitive example has yet been conclusively established.

It is also important to note that even spin-singlet electron pairs can give rise to unconventional superconductivity when the paired electrons occupy special orbital states, such as configurations resembling a binary system in which the electrons orbit around each other.