Topological competition of superconductivity in Pb/Ru/Sr$_2$RuO$_4$ junctions

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(Dated: July 1, 2011)

We devise a new proximity junction configuration where an $s$-wave superconductivity and the superconductivity of Sr$_2$RuO$_4$ interfere with each other. We reproducibly observe in such a Pb/Ru/Sr$_2$RuO$_4$ junction with a single Pb electrode that the critical current $I_c$ drops sharply just below the bulk $T_c$ of Sr$_2$RuO$_4$ and furthermore increases again below a certain temperature below $T_c$. In order to explain this extraordinary temperature dependence of $I_c$, we propose a competition effect involving topologically distinct superconducting phases around Ru inclusions. Thus, such a device may be called a “topological superconducting junction”.

PACS numbers: 74.70.Pq, 74.25.Sv, 74.45.+c, 74.81.-g

Despite extensive studies during the past several decades the realization of spin-triplet superconductivity has not been thoroughly established. Among a number of the candidates, the layered superconductor Sr$_2$RuO$_4$ is a most promising case for spin-triplet pairing.\textsuperscript{2} The invariant spin susceptibility across the superconducting transition, observed by both NMR and polarized neutron scattering, provides indeed strong evidence for equal-spin pairing.\textsuperscript{3,4} The odd parity nature of the orbital wave function, required for triplet states, is compatible with observation using the $\pi$-junction SQUID experiment.\textsuperscript{5} There has been accumulated evidence to support the odd parity superconductivity of Sr$_2$RuO$_4$, however, it is highly desirable to conduct alternative direct experiments for the parity determination.

An important experiment aiming to detect the parity of Sr$_2$RuO$_4$ was introduced in 1999.\textsuperscript{9} It was revealed that the critical current $I_c$ of Pb/Sr$_2$RuO$_4$/Pb junctions with an $s$-wave superconductor Pb is sharply suppressed just below the superconducting transition temperature of Sr$_2$RuO$_4$, $T_{c,SRO} = 1.5$ K, and increases again at lower temperatures. The anomalous temperature dependence of $I_c$ was interpreted as due to the change of the phase difference between the two Pb electrodes from $0$ to $\pi$, driven by the odd parity superconductivity of Sr$_2$RuO$_4$.\textsuperscript{10,11} In this study, we report similar behavior but using Pb/Ru/Sr$_2$RuO$_4$ junctions with essentially different configuration containing only a single Pb electrode. Thus we newly interpret the anomalous $I_c$ as due to the change of the winding number between Pb and Sr$_2$RuO$_4$. Such behavior has never been reported in other superconductor/normal-metal/superconductor (SNS) junctions with an even-parity spin-singlet superconductor or those with odd-parity candidates UPt$_3$ or UBe$_{13,12,13}$ The present finding provides evidence for a most intriguing interplay between the superconductors Pb and Sr$_2$RuO$_4$ connected via Ru metal inclusions, reflecting the distinct pairing symmetry of the two systems.

An SNS junction provides an opportunity to examine the quantum interference involving Sr$_2$RuO$_4$, since such interference sensitively affects its voltage-current ($V$-$I$) characteristics. Technically, poor electrical contact to the surface parallel to the basal $ab$-plane of a Sr$_2$RuO$_4$ crystal hampers the fabrication of a high-quality normal-metal/Sr$_2$RuO$_4$ junction. A Ru metal inclusion in a eutectic Sr$_2$RuO$_4$-Ru crystal\textsuperscript{14} serves as an ideal normal-metal electrode, because of its atomically regular interface.\textsuperscript{15} This eutectic system exhibits higher onset $T_c$ of up to 3.5 K than $T_{c,SRO} (= 1.5$ K) and $T_c$ of Ru ($\sim 0.5$ K), thus called the 3-K phase.\textsuperscript{16} A number of experiments have revealed that the enhanced superconductivity occurs in the Sr$_2$RuO$_4$ region around Ru lamellae and possesses the same parity as that of Sr$_2$RuO$_4$.\textsuperscript{17,18}

FIG. 1. (color online) (a) Differential resistance vs. current, and (b) voltage-current characteristics of a Pb/Ru/Sr$_2$RuO$_4$ junction. The inset to (b) is a schematic of a junction. The critical current $I_{c,+}$ is defined at the peak with positive bias current; $-I_{c,-}$ with negative bias current.
The eutectic crystals were grown by a floating-zone method. The ab-surface of the crystal cut into the dimension of $1.9 \times 2.5 \times 0.2$ mm$^3$ was polished with diamond slurry. The area of Ru inclusions at the surface is typically less than 1% of the total sample area. A 1-$\mu$m thick film was deposited onto the polished surface using 6N-pure lead. In order to obtain the $V$-$I$ characteristics between Pb and Sr$_2$RuO$_4$ using a four-probe method, two terminals each were put on Pb and Sr$_2$RuO$_4$ as illustrated in the inset to Fig. 1(b).

The differential resistance $dV/dI$ of Pb/Ru/Sr$_2$RuO$_4$ junctions was measured by applying an AC current at a frequency of 887 Hz using a lock-in amplifier. Fig. 1(a) is a representative curve of its dependence on the DC bias current at 1.61 K. Fig. 1(b) represents $V$-$I$ characteristics at various temperatures obtained by integrating $dV/dI$, indicating the behavior of a typical superconducting junction. At each temperature, the data were taken after the junction was heated to 1.7 K and cooled to the measurement temperature with a negative DC current exceeding $-I_c$. At the target temperatures, the bias current was swept to a positive value (an up sweep) followed by a down sweep. All the curves in Fig. 1 represent the data taken on down sweeps. Fig. 2 represents the behavior in the up sweeps. As in Fig. 1(a), finite junction voltage emerges with a sharp peak in the differential resistance. Considering the amplitude of the AC current of 20 $\mu$A-rms, the critical current $I_c$ is defined more precisely and accurately at the peak of $dV/dI$, rather than at the onset of non-zero $dV/dI$.

As shown in the top of Fig. 2, $dV/dI$-$I$ curves are almost flat above 2.1 K. Below 2.1 K, the curves exhibit a dip-and-peak structure centered at the zero-bias current and the dip reaches zero at about 1.9 K, which is clearly higher than $T_{c,SRO}$. $I_c$ increases upon cooling below 2.1 K and the $I_c$-$T$ curve in Fig. 3 exhibits a super-linear temperature dependence down to $T_{c,SRO}$: $I_c \propto (T_c-T)^n$ with $n = 2.6$ for $T_c = 2.5$ K, or $n = 3.7$ for $T_c = 3.0$ K. For a tunneling junction with an insulator between the two superconductors, the Ambegaokar-Baratoff (A-B) theory gives $I_c = (eR)^{-1}\Delta_1 K((1 - \Delta_1^2/\Delta_2^2)^{1/2})$ where $R$ is the junction resistance in the normal state, $\Delta_{1,2}$ ($\Delta_1 <\Delta_2$) is the gap function of each superconductor, and $K(x)$ is a complete elliptic integral of the first kind. For $\Delta_1$ substantially smaller than $\Delta_2$, as in the junctions studied here, the temperature dependence of $I_c$ should actually be sub-linear, contrary to the behavior shown in Fig. 3. Furthermore, the $I_c$-$R$ values of our junctions, for example 0.01 $\mu$V at 1.4 K, are much smaller compared to an $I_c$-$R$ curve in Fig. 3 exhibits a super-linear temperature dependence down to $T_{c,SRO}$: $I_c \propto (T_c-T)^n$ with $n = 2.6$ for $T_c = 2.5$ K, or $n = 3.7$ for $T_c = 3.0$ K. For a tunneling junction with an insulator between the two superconductors, the Ambegaokar-Baratoff (A-B) theory gives $I_c = (eR)^{-1}\Delta_1 K((1 - \Delta_1^2/\Delta_2^2)^{1/2})$ where $R$ is the junction resistance in the normal state, $\Delta_{1,2}$ ($\Delta_1 <\Delta_2$) is the gap function of each superconductor, and $K(x)$ is a complete elliptic integral of the first kind. For $\Delta_1$ substantially smaller than $\Delta_2$, as in the junctions studied here, the temperature dependence of $I_c$ should actually be sub-linear, contrary to the behavior shown in Fig. 3. Furthermore, the $I_c$-$R$ values of our junctions, for example 0.01 $\mu$V at 1.4 K, are much smaller compared to an estimate of 400 $\mu$V using the A-B theory with $T_{c1} = 3$ K and $T_{c2} = 7$ K. These facts suggest that the Pb/Ru/3-K-phase configurations should be interpreted in terms of an SNS device with clean SN interfaces where the proximity effect induced coupling plays a crucial role.

Just below $T_{c,SRO} = 1.4$ K, used in the device represented here, the $I_c$ suddenly drops to nearly zero. This
is a surprising result because below this temperature the
interfacial superconductivity of the 3-K phase develops
into the bulk superconductivity of Sr$_2$RuO$_4$ and under
usual circumstances $I_c$ is expected to increase rapidly.
This distinct behavior marks clear evidence for uncon-
tentional interference effects. In addition, it indicates a
qualitative change of the interfacial state of Sr$_2$RuO$_4$ at
$T_{c\text{SRO}}$. Below $T_{c\text{SRO}}$, $I_c$ remains suppressed in a narrow
temperature range and exhibits complicated temperature
dependence. As additional peculiar behavior, $I_c$ starts to
increase sharply below a certain temperature at about 1 K,
which we designate as $T^*$. This anomalous overall
temperature dependence is well reproducible among sev-
eral samples. Another unusual feature of this junction is
that the differential resistance as well as the associated
dependence of the superconducting phase $\phi$ at each spatial
position; the arrows denote the momentum direction for which
$\phi = 0$. The angle $\theta$ is defined as normal to the interface (see
(a)). The lower panels represent the superconducting phases
$\phi$($\theta$) at the Sr$_2$RuO$_4$/Ru interface under no external current.
The solid lines represent the phase of s-wave superconduct-
licity in Ru and the broken lines that of p-wave supercon-
ductivity in Sr$_2$RuO$_4$. (a) $T_{c\text{SRO}} < T < T_c$: the $p_\parallel \pm ip_\perp$ state with the winding number $N = 0$ is realized (A'-phase),
matching with the s-wave order parameter induced in Ru.
(b) $T \lesssim T_{c\text{SRO}}$: replacement by $p_\parallel \pm ip_\parallel$, the bulk state of
Sr$_2$RuO$_4$, with $N = \pm 1$ (B-phase). (c) $T \ll T_{c\text{SRO}}$: increasing
interfacial energy enlarges the phase deformation in the
s-wave, strengthening the Josephson coupling.

Previous experiments and theories suggest that the 3-
K phase originates most likely from the nucleation of su-
perconductivity at the interface of Sr$_2$RuO$_4$ and Ru. It
coincides with a single p-wave component existing in a
narrow spatial range on the Sr$_2$RuO$_4$ side with its mo-
mentum parallel to the interface, denoted as $p_\parallel$-wave. This
time-reversal symmetry conserving state, called "A-
phase", corresponds topologically to a superconducting
state without phase winding ($N = 0$) from the viewpoint
of the Ru-inclusion. Below $T_c = 2.4 - 2.6$ K the time-
reversal symmetry breaking appears by adding an addi-
tional order parameter component, $p_\parallel + i\varepsilon_p\perp$, which
may correspond to the "A'-phase" with $N = 0$ or the
"B-phase" with $N = 1$ within the theory introduced by
Kaneyasu et al. The latter is topologically compatible
with the chiral p-wave bulk phase.

For the present junction geometry we assume that Pb
induces superconductivity of s-wave symmetry in the Ru
inclusions by proximity effect, which through spin-orbit
interaction yields a direct coupling to the $p_\parallel$-wave or-
der parameter in Sr$_2$RuO$_4$. The topological match-
ing with $N = 0$ of Ru naturally favors the A'-phase
over the B-phase as depicted in Fig. 5(a), because the phase
difference $\Delta \phi(\theta)$ at the interface can be set to zero for all angles $\theta$ around the circumference (junction ground state). While the A-phase consisting of only the
$p_\parallel$-component is strongly localized at the interface, the
A'-phase with the additional $p_\perp$-component is more extended
(large normal-metal coherence length $\xi_n$ perpen-
dicular to the interface). This extension can explain why
$I_c(T)$ starts to increase slowly with lowering $T$ below
$T_c$; the gradually growing order parameter $p_\perp$ strengthens the superconducting connections between different Ru-inclusions and to the external contacts. Thus, $I_c$ is expected to grow approximately as $\exp[-d/\xi_n(T)]$ with $d$ the average connecting distance among Ru-inclusions and contacts and $\xi_n(T) \propto (T - T_{c\text{SRO}})^{-1/2}$. This yields behavior qualitatively compatible with the experimental
results.

With the onset of bulk superconductivity at $T_{c\text{SRO}}$ the order parameter at the interface changes its topology to that of the B-phase (Fig. 5(b)), which due to its winding number $N = \pm 1$ is frustrated with the phase of the s-wave order parameter in Ru. For the over-
all Josephson current $I = \int_0^{2\pi} d\theta J_c(\theta, T) \sin \Delta \phi(\theta) \approx J_c(T) \int_0^{2\pi} d\theta \sin \Delta \phi(\theta)$, this topological mismatch leads to an almost complete cancellation. At the same time, this frustration induces mild phase deformation just below $T_{c,\text{SRO}}$. With decreasing temperature, the growing interfacial superconductivity in Sr$_2$RuO$_4$ requires less phase mismatch to minimize the total junction energy. As a result the region of significant $\Delta \phi$ is confined into a shrinking range of $\theta$ (Fig. 5(c)).$^{28}$ With the application of external current, the resulting phase deformation is such that the accompanying Josephson current density $J_c(\theta, T) \sin \Delta \phi(\theta)$ is constructively added and grows at lower temperatures.$^{29}$

The extraordinary temperature dependence of $I_c$ is explained in terms of a junction consisting of a topological superconductor encapsulating a conventional superconductor. For this reason, such a device may be called “topological superconducting junction”, in which non-trivial character of a topological superconductor becomes observable by appropriate design of the geometry.

The low-temperature phase ($T < T_{c,\text{SRO}}$) introduces new features. One is the variation of the $dV/dI$-vs-$I$ curves on the sweep direction of the supercurrent (see Fig. 4). This may involve complex phase frustration effects introduced below $T_{c,\text{SRO}}$ through the phase winding of the B-phase, which can lead to various metastable states. Similar variations have been reported in different junctions of Sr$_2$RuO$_4$.$^{22,24}$ Since our present junctions contain a number of Ru inclusions acting as parallel contacts, future experiments using junctions consisting of a single Ru inclusion may resolve this issue.

The coupling of a Pb to Sr$_2$RuO$_4$ via a Ru-inclusion provides a unique opportunity to investigate quantum interference effects between topologically incompatible superconducting phases. The unusual sharp drop of the critical current $I_c$ below $T_{c,\text{SRO}}$ and its curious recovery below $T^*$ can be explained by the change of the topology of the superconducting order parameter in Sr$_2$RuO$_4$ surrounding the Ru-inclusion. Thus, the device investigated here can be classified as a new class of superconducting junctions: a “topological superconducting junction”.

We thank Y. A. Ying, S. Kittaka, K. Ishida, T. Sumi and Y. Yamaoka for their support, and H. Kaneyasu and D. F. Agterberg for useful discussions. This work is supported by Grants-in-Aid for Global COE program “the Next Generation of Physics Spun from Universality and Emergence”, for Scientific Research on Priority Areas “Physics of New Quantum Phases in Superclean Materials” and for Scientific Research in Innovative Areas “Topological Quantum Phenomena” from MEXT of Japan. T. N. is supported by the JSPS.

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