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Experimental observation of the selection rule in Josephson coupling between In and Sr_2RuO_4

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Abstract. – We have carried out experimental studies on the Josephson coupling between a conventional *s*-wave superconductor (In) and Sr_2RuO_4 and found that the coupling is allowed in the in-plane direction, but not along the *c*-axis. This selection rule indicates that the symmetry of superconducting order parameter of Sr_2RuO_4 is either *p*- or, alternatively, purely *d*-wave. If Sr_2RuO_4 is a *p*-wave superconductor, as strongly favored by other experimental evidence, our result suggests that the pairing state of Sr_2RuO_4 is Γ_5^-, with $\boldsymbol{d}(\boldsymbol{k}) = \boldsymbol{z}(k_x \pm ik_y)$, a nodeless state in which the spins of the superconducting electrons lie in the RuO_2 planes.

Recently, the first known Cu-free layered perovskite superconductor, Sr_2RuO_4 [1], has emerged as a new focus of superconducting materials research. The main issue is whether the pairing symmetry of Sr_2RuO_4 is spin triplet with odd parity (*p*-wave) as predicted theoretically [2]. A growing body of experimental evidence, including results obtained from muon spin relaxation [3], NMR $1/T_1$ and Knight shift [4], neutron scattering [5], impurity effect [6], proximity Josephson coupling effect [7], and specific heat [8] measurements, has shown that the pairing symmetry is unconventional, most likely *p*-wave. In particular, the flat Knight shift observed across the T_c of Sr_2RuO_4 may be considered as direct evidence for the *p*-wave pairing state in Sr_2RuO_4 [4] (see below). In addition to the only known *p*-wave superconductor, ^{3}He, certain heavy-fermion compounds are also possible candidates for *p*-wave superconductors. However, Sr_2RuO_4 has an advantage that its electronic band structure is considerably simpler than those of heavy-fermion compounds, making it perhaps a more tractable material for demonstrating a *p*-wave pairing state.

Assuming a *weak* spin-orbit coupling, five possible *p*-wave states are allowed by the crystal symmetry of Sr_2RuO_4 [2]. Among them, the Γ_5^- state, with the $\boldsymbol{d}$-vector given by $\boldsymbol{d}(\boldsymbol{k}) = \boldsymbol{z}(k_x \pm ik_y)$ ($\boldsymbol{z}$ denotes the unit vector along the *c*-axis), is favored by muon spin

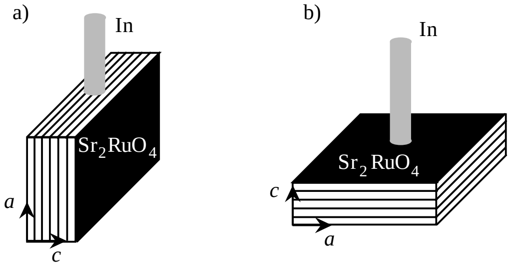

Fig. 1

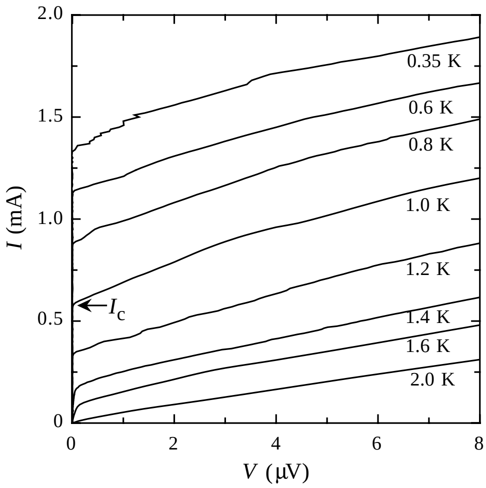

Fig. 2

Fig. 1 – Schematics of (a) in-plane and (b) c-axis In/Sr_2RuO_4 junctions.

Fig. 2 – I-V curves at various temperatures for an in-plane In/Sr_2RuO_4 junction (sample #11). Finite critical current I_c is indicated.

relaxation [3] and NMR Knight shift [4] results. The physical meaning of $\boldsymbol{d}$-vector is as follows. The magnitude of the $\boldsymbol{d}$-vector is the amplitude of the superconducting order parameter. When projected to the direction of the $\boldsymbol{d}$-vector, the component of total superconducting electron spins is zero. The Γ_5^- state for Sr_2RuO_4 is nodeless with all spins of the superconducting electrons lying in the RuO_2 planes. Our experiment on Josephson coupling between an s-wave superconductor, In, and Sr_2RuO_4 along different crystalline orientations, is shown schematically in fig. 1. This experiment can be used to determine which pairing state within the p-wave scenario is adopted by Sr_2RuO_4. In this letter, we present our experimental finding of a selection rule in the Josephson coupling between In and Sr_2RuO_4. It was found that this coupling is allowed in the in-plane direction (fig. 1a) but not along the c-axis (fig. 1b). If Sr_2RuO_4 is a spin-triplet superconductor, as strongly favored by other experimental results, our observation provides *direct* experimental evidence that the pairing state of Sr_2RuO_4 is indeed Γ_5^-. In the context of the spin-singlet d-wave scenario [9], which is not inconsistent with our selection rule but contradicts the NMR Knight shift result [4], the present work suggests that the pairing state in Sr_2RuO_4 is purely d-wave.

Single crystals of Sr_2RuO_4 were grown by a floating-zone method using an image furnace [1]. Results from a.c. magnetic susceptibility measurements showed a superconducting transition at $T = T_c = 1.45\,\mathrm{K}$ (onset) and a transition width around 0.05 K for crystals prepared in two separate growth runs. Its superconducting coherence lengths at zero temperature are $\xi_{ab} = 660\,\text{Å}$ and $\xi_c = 33\,\text{Å}$ for the in-plane and c-axis directions, respectively [10]. To prepare c-axis In/Sr_2RuO_4 junctions, a Sr_2RuO_4 single crystal was cleaved along the ab-plane. Atomic force microscope (AFM) studies of the cleaved surface show an atomically flat surface over an area of up to $(10\,\mu\mathrm{m})^2$. A freshly cut In wire of 0.25 mm in diameter was pressed on the crystal immediately after it was cleaved. The in-plane junctions were prepared on Sr_2RuO_4 single crystals with a finely polished a.c. face. AFM imaging showed that the polished face is fairly rough with micron-size mechanical damage. To our knowledge, no chemical solution can

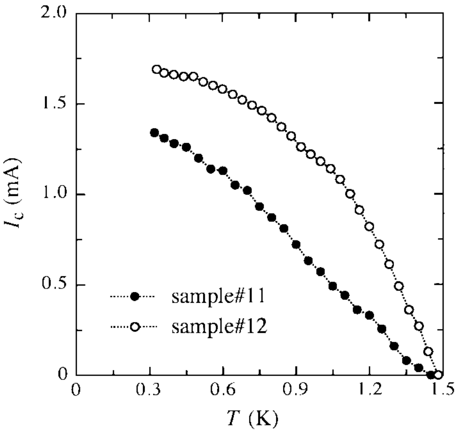

Fig. 3 – Temperature dependence of the critical current, $I_c(T)$, for two in-plane In/Sr_2RuO_4 junctions (samples #11 and #12).

etch Sr_2RuO_4 to obtain a smooth surface. Thus, the freshly cut In wire was directly pressed on clean, but as-polished a.c. face of Sr_2RuO_4 to form an in-plane junction. For both types of junctions, the maximum junction area is $\sim 0.05\,\mathrm{mm}^2$ with junction resistances ranging from 0.1 to 100 Ω. Electrical measurements were carried out in d.c. in a ^{3}He cryostat with a base temperature of 0.3 K. All electrical leads entering the sample enclosure were filtered by RF filters with the insertion loss of 10 dB at 10 MHz, 30 dB at 100 MHz and 50 dB at 300 MHz. A μ-metal box shielded the samples from residual magnetic field.

In fig. 2, the I-V curves of an in-plane In/Sr_2RuO_4 junction (sample #11) are shown. In this case, the current (I) flows along the in-plane direction. A non-zero supercurrent, followed by a linear I-V characteristic, was evident. Qualitatively the same behaviors have been found in other in-plane junctions. (Among 11 in-plane In/Sr_2RuO_4 junctions we have prepared, five showed non-zero supercurrent.) The temperature dependence of I_c, shown in fig. 3 for samples #11 and #12, has the general shape of that for a superconductor-normal metal-superconductor (SNS) junction [11]. The magnetic-field dependence of I_c was measured for one in-plane junction (without μ-metal shield). While I_c was found to decrease with increasing field, no Fraunhofer pattern was observed, suggesting that the junction is not very uniform. Because of the stress, structural defects, oxygen deficiency, and the possible formation of an insulating indium oxide layer, we believe that a potential barrier with spatially varying height is present at the In/Sr_2RuO_4 interface. The observed Josephson coupling is through regions where the potential is low.

For two dissimilar s-wave superconductors, the Ambegaokar-Baratoff (A-B) limit for I_cR_N is given by $I_cR_N \le (\Delta_1/e)K\{[1-(\Delta_1/\Delta_2)^2]^{1/2}\}$, where R_N is junction resistance in the normal state, Δ_1 and Δ_2 are zero-temperature energy gaps for two superconductors, and the function K is the elliptic integral of the first kind [12]. Unfortunately, the gap for Sr_2RuO_4 is yet to be determined experimentally [13]. However, if one estimates the gap using the BCS result, $\Delta = 1.764k_BT_c \approx 0.22\,\mathrm{meV}$, this leads to an A-B limit of 0.6 mV. At $T = 0.3\,\mathrm{K}$, values of I_cR_N are 0.16 and 0.18 mV for junctions #11 and #12, and 15, 16, and 45 μV for three other (in-plane) samples, respectively. The numbers for junctions #11 and #12 are a substantial fraction of the A-B limit, suggesting that, at least for these two junctions, the observed I_c is due to a finite Josephson coupling between In and Sr_2RuO_4 in the in-plane direction.

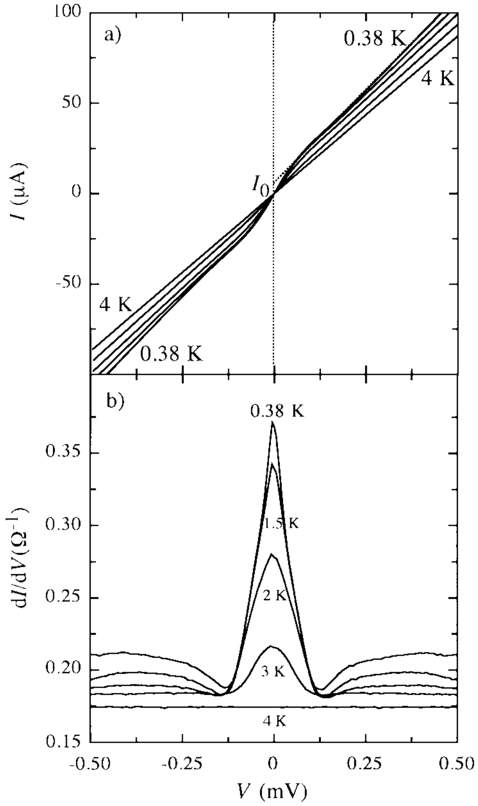

Fig. 4 – a) I-V curves for a c-axis In/Sr_2RuO_4 junction (sample #17). b) Dynamic conductance of the same junction.

In 12 c-axis In/Sr_2RuO_4 junctions, no finite supercurrent was found. While it is not clear why the potential barrier is higher than the c-axis junctions, experimentally most c-axis junctions were found to exhibit tunneling behavior. In principle, it is possible that the short c-axis superconducting coherence length of Sr_2RuO_4 together with the presence of a tunnel barrier suppresses the amplitude of the superconducting order parameter [14], resulting in a vanishing supercurrent, independent of the pairing symmetry of Sr_2RuO_4. In two c-axis In/Sr_2RuO_4 junctions, however, instead of tunneling features, an excess current (I_0) or zero-bias conductance peak (fig. 4) was seen. The excess current or zero-bias conductance peak is a signature of the Andreev reflection process at a normal metal-superconductor (N-S) interface, where an incoming normal electron with energy below the gap of the superconductor combines with another electron to form a Cooper pair which enters the superconductor. As a result, a hole is reflected, giving rise to "extra" charge passing through the interface. Since the Andreev reflection occurs only when the potential barrier at the interface is low [15], its presence above the T_c of Sr_2RuO_4 indicates that the interface between In and Sr_2RuO_4 is essentially metallic. This excess current was seen to persist below the T_c of Sr_2RuO_4, which may be explained by the existence of a normal layer at the ab face of the Sr_2RuO_4 crystal. This is further supported by the observation that no gap features associated with Sr_2RuO_4 were present below the T_c of Sr_2RuO_4 in all c-axis tunnel junctions we have studied [13]. No supercurrent was observed down to 0.38 and 0.65 K, respectively, in either of these two c-axis junctions showing Andreev reflection, suggesting that no Josephson coupling was established in these c-axis junctions.

An important question is whether the lack of finite Josephson coupling between In and Sr_2RuO_4 along the c-axis is of an intrinsic or extrinsic origin. In addition to a metallic contact between In and the normal top layer of Sr_2RuO_4, the interface between the normal layer and the bulk superconducting Sr_2RuO_4 should be metallic as well since it is naturally formed with no oxygen deficiency or mechanical stress expected. As a result, the two c-axis In/Sr_2RuO_4 junctions showing Andreev reflection should be SNS junctions. If Sr_2RuO_4 is an s-wave superconductor, Josephson coupling should be possible in these c-axis junctions as long as the thickness of the N-layer is within a few times the normal coherence length ξ_N [11]. In the clean limit, which we believe is appropriate for Sr_2RuO_4, $\xi_N = \hbar v_N/(2\pi k_B T)$, where v_N is the Fermi velocity. Using $v_N = 1.4 \times 10^6$ cm/s along the c-axis [16], we have $\xi_N = 774\,\text{Å}$ for Sr_2RuO_4 at $T = 0.38\,\text{K}$, the lowest temperature measured for sample #17. Compared with the distance between two adjacent RuO_2 layers, 6.4 Å, a length a few times ξ_N would be of a few hundreds the inter-layer distance. It is very unlikely that the normal layer formed at a freshly cleaved Sr_2RuO_4 single crystal can be so thick. Therefore the absence of supercurrent in c-axis In/Sr_2RuO_4 junctions cannot be due to an overly thick N-layer.

Is it possible that the supercurrent for the c-axis junctions is smeared out by thermal fluctuations? The characteristic current I_{th} due to thermal fluctuation effects is given by $I_{th}(\mu\text{A}) = 0.042T(\text{K})$ [17], which is $0.016\,\mu\text{A}$ at $T = 0.38\,\text{K}$. Supercurrent above this value should not be thermally smeared. If the A-B limit for $I_c R_N$ (0.6 mV) is a good guide, we expect a critical current on the order of $100\,\mu\text{A}$ for sample #17 with $R_N = 5.7\ \Omega$. Even using the experimental values of $I_c R_N$ for two in-plane In/Sr_2RuO_4 junctions shown in fig. 3 (0.16 and 0.18 mV, respectively, at 0.3 K), we still expect $I_c > 28\,\mu\text{A}$, well above I_{th} and our measurement limit. Therefore, the absence of Josephson coupling between In and Sr_2RuO_4 along the c-axis must be due to intrinsic reasons. It should be noted that, unlike experiments on c-axis tunneling between Pb and high-T_c superconductors [18, 19], we are attempting to demonstrate the absence, not the presence, of a c-axis Josephson coupling between the two superconductors. Hence, whether or not the in-plane coupling due to the presence of steps on the cleaved ab face may be present in our c-axis junctions is not an issue.

Josephson coupling between a spin-singlet (even-parity) and a spin-triplet (odd-parity) superconductor was first thought to be impossible [20]. However, it was subsequently pointed out that the first-order Josephson coupling between two superconductors with different parities could arise from spin-orbit coupling [21]. In the presence of the spin-orbit coupling, the Cooper pairs of different parities will be mixed at the interface between the s- and the p-wave superconductor, resulting in a direct Josephson coupling between them. In fact, it has been shown that, in the presence of a finite potential barrier, I_c is proportional to [22–24]

$$I_c \sim \langle \boldsymbol{d}(\boldsymbol{k}, \boldsymbol{x}) \cdot (\boldsymbol{k} \times \boldsymbol{n}) \rangle_{FS}\,, \tag{1}$$

where $\boldsymbol{n}$ is the unit vector normal to the interface, $\boldsymbol{x}$ and $\boldsymbol{k}$ are real- and momentum-space coordinates, and $\langle \ldots \rangle_{FS}$ denotes average over the Fermi surface. Since $\boldsymbol{d}$ and $\boldsymbol{k} \times \boldsymbol{n}$ represent essentially the spin and the orbital angular momentum of the superconducting condensate at the interface, respectively, $[\boldsymbol{d}(\boldsymbol{k}, \boldsymbol{x}) \cdot (\boldsymbol{k} \times \boldsymbol{n})]$ merely reflects the spin-orbit coupling strength of the p-wave superconductor, which gives rise to the (orientation-dependent) Josephson coupling between the p- and the s-wave superconductor as mentioned above. If the pairing symmetry in Sr_2RuO_4 is indeed p-wave, eq. (1) implies that the Josephson coupling between In and Sr_2RuO_4 is orientation-dependent. In particular, among five possible representations (Γ^-_{1-5}) [2], eq. (1) states that, for in-plane junctions, $I_c \neq 0$ only if the pairing state of Sr_2RuO_4 is Γ^-_5, with $\boldsymbol{d}(\boldsymbol{k}) = \boldsymbol{z}(k_x \pm ik_y)$.

The above result is consistent with *all other* experimental findings obtained thus far. In particular, in the NMR experiment [4], the electron spin susceptibility of Sr_2RuO_4 was found to be a constant within experimental error as temperature was brought from above T_c to 15 mK, as expected for the Γ_5^- state. While the standard theory predicts exponentially small electron spin susceptibility in the zero-temperature limit for *s*-wave superconductors [25], it was found that Hg [26] and Sn [27] showed finite electron spin susceptibility well below T_c. This was explained [28] by the presence of spin-orbit coupling within an *s*-wave picture. Nevertheless, a finite drop in electron spin susceptibility was still observed across T_c for both Hg [26] and Sn [27]. A constant electron spin susceptibility (or Knight shift) in Sr_2RuO_4 containing relatively light elements (corresponding to weak spin-orbit coupling) is difficult to be explained within a spin-singlet picture.

It should be pointed out that a *d*-wave scenario has recently been proposed for Sr_2RuO_4 [9]. This scenario contradicts the NMR Knight shift result. However, if it turns out to be true, the result of the present work suggests that the superconducting order parameter in Sr_2RuO_4 is purely *d*-wave. Given that the issue of whether the pairing symmetry in high-T_c cuprates is purely *d*-wave is still being debated, our selection rule result would be of significance in that context as well.

In conclusion, we have found a selection rule in the Josephson coupling between In and Sr_2RuO_4. While a phase-sensitive experiment may ultimately be required to settle this issue, the totality of the available results on Sr_2RuO_4, and the remarkable consistency among them, makes a compelling case for a *p*-wave pairing state in Sr_2RuO_4. In this context, the present work shows that the pairing state of Sr_2RuO_4 is Γ_5^-.

* * *

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