Phase-Sensitive Tests of the Pairing State Symmetry in Sr_2RuO_4

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Exotic superconducting properties of Sr_2RuO_4 have provided strong support for an unconventional pairing symmetry. However, the extensive efforts over the past decade have not yet unambiguously resolved the controversy about the pairing symmetry in this material. While recent phase-sensitive experiments using flux modulation in Josephson junctions consisting of Sr_2RuO_4 and a conventional superconductor have been interpreted as conclusive evidence for a chiral spin-triplet pairing, we propose here an alternative interpretation. We show that an overlooked chiral spin-singlet pairing is also compatible with the observed phase shifts in Josephson junctions and propose further experiments which would distinguish it from its spin-triplet counterpart.

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The unambiguous determination of the pairing state symmetry is one of the key steps towards understanding the pairing mechanism in a continuously growing class of unconventional superconductors [1]. Phase-sensitive experiments, capable of identifying the angular dependence of the superconducting order parameter, have provided crucial evidence for a dominant d-wave orbital symmetry in cuprate superconductors [2–4]. However, much less is known for other unconventional superconductors such as heavy fermions, charge transfer salts, and cobaltates. In particular, there is compelling evidence for an unconventional pairing in Sr_2RuO_4 [5,6], with the strong possibility of spin-triplet superconductivity which would be a solid-state analog of superfluid He^3 [7].

In superconductors with inversion symmetry an order parameter (gap matrix) can be expressed as $\hat{\Delta}(\mathbf{k}) = \Delta_0(\mathbf{k}) i\hat{\sigma}_y$ for spin-singlet and $\hat{\Delta}(\mathbf{k}) = \hat{\sigma}\cdot\mathbf{d}(\mathbf{k}) i\hat{\sigma}_y$ for spin-triplet pairing. Here $\hat{\sigma}$ are the Pauli spin matrices and scalar (vector) Δ_0 ($\mathbf{d}$) has even (odd) parity in the wave vector $\mathbf{k}$. Often the symmetry of both the orbital and the spin part of $\hat{\Delta}(\mathbf{k})$ remains to be identified and the lack of related understanding comes from the difficulty in performing phase-sensitive experiments.

While numerous previous experiments probed the pairing symmetry of Sr_2RuO_4 [6], in this context, recent phase-sensitive experiments [8] that provide angle-resolved information are particularly important. The measurements were performed in superconducting quantum interference device (SQUID) geometry, consisting of a pair of $Au_{0.5}In_{0.5}/Sr_2RuO_4$ Josephson junctions. Since $Au_{0.5}In_{0.5}$ is a conventional s-wave superconductor, the observed modulation of critical current in an applied magnetic field was interpreted as conclusive support for an odd-parity spin-triplet pairing in Sr_2RuO_4 [8,9].

A similar SQUID geometry was proposed [10] to study possible p-wave pairing in heavy fermions and later also used for identifying d-wave pairing in cuprates [2]. The critical current I_c is modulated in the applied magnetic field as [11] $I_c \propto \cos(\Phi/\Phi_0 + \delta_{12}/2)$, where Φ is the flux threading the SQUID, Φ_0 is the flux quantum, and δ_{12} is the intrinsic phase shift of the order parameter between the two tunneling directions. For a conventional s-wave SQUID $\delta_{12} = 0$ and I_c has a maximum at $\Phi = 0$. In contrast, a phase shift $\delta_{12} = \pi$, characteristic of unconventional pairing [12], yields a minimum I_c at $\Phi = 0$. The modulation of external flux together with the fabrication of junctions with varying tunneling directions in SQUID geometry therefore provide angle-resolved phase-sensitive information about the pairing symmetry.

The suggested chiral p-wave (CpW) state with the triplet order parameter [13],

$$\mathbf{d}(\mathbf{k}) \propto (k_x + ik_y)\hat{\mathbf{z}}, \tag{1}$$

in which the spins of the Cooper pairs lie in the RuO_2 plane ($\perp \mathbf{d}$), is indeed compatible with the experiment [8]. However, we show here that it is not the only candidate. There exists another pairing state, allowed by the tetragonal symmetry of Sr_2RuO_4, the singlet chiral d-wave (CdW) state ${}^1E_g(c)$ with $\Delta_0(\mathbf{k}) \propto (k_x + ik_y)k_z$, or, more accurately [14]

$$\Delta_0(\mathbf{k}) \propto (k_x + ik_y)\sin k_z c, \tag{2}$$

which is equally consistent [15] with the phase shifts observed in Ref. [8]. We use our findings to propose an experimental test which would discriminate between CpW and CdW pairing symmetries.

Could experimental and theoretical reasons be used to rule out CdW and favor only the CpW state? The two main arguments in favor of the CpW come from muon spin resonance and Knight shift experiments [6,16]. The former indicate a time-reversal symmetry breaking below the transition temperature T_c, incompatible with the $d_{x^2-y^2}$-wave state in cuprates, but fully compatible with either CpW or CdW symmetry. The Knight shift (K) was initially interpreted as firm evidence for a triplet state with in-plane spins (like CpW), since no change of the in-plane spin susceptibility below T_c was found.

Even in singlet superconductors (e.g., V, Hg, Sn), K could remain invariant below T_c. Such behavior is usually attributed to (a) spin-orbit induced spin-flip scattering, which suppresses the effect of the superconductivity on K or (b) an accidental cancellation of the spin, dipole, and orbital contributions of the Fermi-level electrons to K, which leaves only superconductivity-insensitive contributions such as the Van Vleck susceptibility. However, a quantitative analysis [17] shows that the spin-orbit coupling in Sr_2RuO_4 is too weak for scenario (a) while the accidental cancellation, required for scenario (b), does not occur [18]. Thus, neither of the two explanations of a constant K arising from singlet pairing is applicable.

This would have made the Knight shift argument for CpW very convincing, if not for the recent experiment showing the same result in a field perpendicular to the plane [19]. It was proposed [19] that the testing field of 0.02 T may be enough to induce a phase transition from the CpW in Eq. (1) to a state with $\mathbf{d}\|\hat{\mathbf{x}}$. However, this is highly unlikely: (i) the $\mathbf{d}\|\hat{\mathbf{x}}$ state would have an additional horizontal line node, as compared to the $\mathbf{d} \propto (k_x + ik_y)\hat{\mathbf{z}}$ state and, therefore, lose a large part of the pairing energy ($\sim\Delta$ per electron, $\Delta \gtrsim 1.4$ K $\gg 0.02$ T); (ii) although in the $\mathbf{d}\|\hat{\mathbf{x}}$ state the spins of the pairs lie in the yz plane, there is no crystallographic y-z symmetry and it is not *a priori* clear whether the magnetic susceptibility of the Cooper pair will be the same as for the normal electrons. Since $\mathbf{d}\|\hat{\mathbf{x}}$ is not allowed for a tetragonal symmetry, it may only appear as a result of a second phase transition below T_c, which has never been observed in Sr_2RuO_4; (iii) the spin-orbit part of the pairing interaction, which keeps the spins in the xy plane, despite z being the easy magnetization axis [20], would have to be weaker than $0.02\ \mathrm{T} = 1.1\ \mu\mathrm{eV} = 0.013$ K, an energy scale much too small for the spin-orbit coupling in Sr_2RuO_4. So, neither the old theories for the lack of a Knight shift reduction below T_c, nor the new explanation in terms of the magnetic-field rotated order parameter withstand quantitative scrutiny; the Knight shift in Sr_2RuO_4 remains a challenge for theorists. Until this puzzle is resolved, we cannot use the Knight shift argument.

We now turn to the experiments of Ref. [8] and compare Josephson tunneling between an s-wave superconductor and either (a) an even parity (spin-singlet) superconductor or (b) an odd-parity (triplet) superconductor. In the first case, the Josephson current between a conventional s-wave superconductor and an unconventional spin-singlet superconductor, represented by the order parameters $\Delta_{s\text{-wave}}$ and $\Delta_0(\mathbf{k})$, respectively, can be expressed as [21]

$$J \propto \langle T_{\mathbf{k}}\, \mathrm{Im}[\Delta^*_{s\text{-wave}}\Delta_0(\mathbf{k})]\rangle_{\mathrm{FS}}, \tag{3}$$

which depends on the relative phase between the superconducting order parameters. The averaging is over all states at the Fermi surface (FS) where the Fermi velocity, $\mathbf{v}_{\mathrm{F}}$, has a positive projection on the tunneling direction represented by the unit normal $\mathbf{n}$ (perpendicular to the interface plane, see Fig. 1) and $T_{\mathbf{k}}$ is the tunneling probability. For a thick rectangular barrier of width w and height U [22] we can obtain

$$T_{\mathbf{k}} = \frac{16m^2\kappa^2 v_L v_R \exp[-2\kappa w - k_\parallel^2 w/\kappa]}{(\kappa^2 + m^2 v_L^2)(\kappa^2 + m^2 v_R^2)}, \tag{4}$$

where $\kappa = \sqrt{2m(U-\mu)}$ such that $w\kappa \gg 1$ (in the thick-barrier limit), m is the free-electron mass, μ is the Fermi energy, and we set $\hbar = 1$. We use $v_{L,R}$ to denote normal components of the Fermi velocities in the two superconductors and $k_\parallel$ is the component parallel to the interface. From Eq. (4) we see that $T_{\mathbf{k}}$ is sharply peaked when $\mathbf{v}_{\mathrm{F}} \parallel \mathbf{n}$.

In the second case, the Josephson current between a singlet and a triplet superconductor becomes [21,23]

$$J \propto \langle \tilde{T}_{\mathbf{k}}\, \mathrm{Im}[\Delta_0^*(\mathbf{k})\mathbf{d}(\mathbf{k}) \cdot (\mathbf{n} \times \mathbf{k})]\rangle_{\mathrm{FS}}, \tag{5}$$

where we use $\tilde{T}_{\mathbf{k}}$ to denote that, unlike $T_{\mathbf{k}}$, it contains matrix elements corresponding to spin-flip tunneling, for example, due to magnetic interfaces or spin-orbit coupling. For nonmagnetic barriers and in the absence of spin orbit, there is no spin-flip scattering, therefore $\tilde{T}_{\mathbf{k}} = 0$ and the Josephson current vanishes identically [21,23,24].

From Eqs. (3) and (5) we can directly infer that for c-axis tunneling, $\mathbf{n} \parallel \mathbf{c}$, $J = 0$ for both CdW and CpW states [$\int dk_x dk_y (k_x + ik_y) = 0$]. For tunneling *precisely* in the ab plane, the current is also zero for CdW while for CpW it only vanishes at $\mathbf{n} \parallel \mathbf{k}$.

We consider a model of a quasi-two-dimensional (2D) layered superconductor which has a nearly cylindrical FS with a small c-axis dispersion originating from the weak interlayer hopping. In Fig. 1(a) we show the sample geometry used in Ref. [8] and in Fig. 1(b) represent a warping

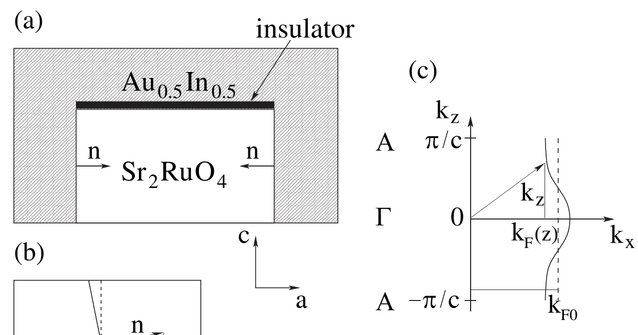

FIG. 1. Schematic sample and the Fermi surface geometry for phase-sensitive SQUID measurements from Ref. [8]. (a) $Au_{0.5}In_{0.5}/Sr_2RuO_4$ junction geometry with an interface normal $\mathbf{n}$. (b) Possible deviation of $\mathbf{n}$ from the ab crystallographic plane. (c) Warping of the Sr_2RuO_4 Fermi surface. The magnitude of the Fermi wave vector $\mathbf{k}_{\mathrm{F}}$ is generally different from the one corresponding to the cylindrical Fermi surface (k_{F0}).

of the Fermi surface. In the first approximation for Sr_2RuO_4 such a warping can be expressed as [25]

$$\mu = \frac{k_F^2(z)}{2m}[1 + \varepsilon \cos k_z c], \tag{6}$$

where $|\varepsilon| \ll 1$ is the warping parameter ($\varepsilon \approx -7 \times 10^{-4}$ [25]), c is the lattice constant along the crystallographic z direction, and $\mathbf{k}_F(z)$ is the z-dependent projection of the Fermi wave vector in the xy plane [see Fig. 1(c)]. It is convenient to resolve the Fermi wave vector in cylindrical coordinates $[k_F(z), \varphi, k_z]$ with $k_F(z) = k_{F0}/[1 + \varepsilon \cos k_z c]^{1/2}$, where $k_{F0} = (2m\mu)^{1/2}$ and for $\varphi = 0$ [see Fig. 1(c)], $k_F(z) \to k_{Fx}$.

A scheme of the SQUID geometry in Fig. 1(a) (adapted from Ref. [8]) is an oversimplification. While efforts were made to fabricate edges either precisely parallel or perpendicular to the c axis, in the actual samples the direction of interface planes or their corresponding normals changes gradually from the a to c direction. In several samples [8], an interface nearly parallel to the ab plane, at the $Au_{0.5}In_{0.5}/Sr_2RuO_4$ junction, was covered by an insulating oxide [see Fig. 1(a)]. It is then plausible to expect that the normal to such interface could deviate from the ab plane. In Fig. 1(b) we depict a generalized situation in which an interface normal, $\mathbf{n} = (n_\rho, n_\varphi, n_z)$ with $|n_z| \ll 1$, need not lie exactly in the crystallographic ab plane of Sr_2RuO_4. We show below that the analysis of phase-sensitive measurements in terms of the two small but *finite* parameters, ε and n_z, can provide a qualitatively different interpretation from those which *a priori* assume $\varepsilon = n_z \equiv 0$.

For a conventional superconductor with the FS larger than the one of Sr_2RuO_4, the Josephson tunneling across a thick rectangular barrier can be obtained from Eqs. (3) and (4) as

$$J \propto \int_{\mathbf{v}_F \cdot \mathbf{n} > 0} d\mathbf{k}\, \delta(\epsilon_{\mathbf{k}} - \mu) \mathbf{v}_F \cdot \mathbf{n} \exp(-k_\parallel^2 w/2\kappa) \times \mathrm{Im}(k_x + ik_y) \sin k_z c, \tag{7}$$

where $k_\parallel^2 = \mathbf{k}_F^2 - (\mathbf{k}_F \cdot \mathbf{n})^2$, $\kappa^2 \gg m v_{L,R}^2$, and the projection of the Fermi velocity in Sr_2RuO_4 along $\mathbf{n}$ is

$$\mathbf{v}_F \cdot \mathbf{n} = \frac{k_{F0}[1 + \varepsilon \cos k_z c]^{1/2}}{m} n_\rho - \frac{k_{F0}^2 \varepsilon c \sin k_z c}{2m[1 + \varepsilon \cos k_z c]} n_z. \tag{8}$$

For a thick barrier, the integration can be simplified by noting that the dominant contribution comes from $k_\parallel = 0$. The right-hand side of Eq. (7), in the leading order in ε and n_z, can be then reduced to $\sqrt{\pi\kappa/w} c k_{F0}^2 n_z (1 - \varepsilon)$, such that

$$J_\sqcap = A n_z (1 - \varepsilon), \tag{9}$$

where A characterizes the normal state barrier transparency. Thus, with a tilted interface ($n_z \neq 0$) there is a finite current even in the absence of any FS warping ($\varepsilon = 0$).

To verify that our findings of finite Josephson current in the CdW state are not limited to the specific assumption of a thick rectangular barrier, we also consider the rather different case of a strong δ-function barrier. The corresponding transmission probability is [22,26]

$$T_{\mathbf{k}} = \frac{4 v_L v_R}{(v_L + v_R)^2 + 4U^2}, \tag{10}$$

where $v_{L,R}$ are the normal components of the Fermi velocities in the two superconductors and U ($\gg v_{L,R}^2$) is the scattering strength. From Eqs. (3) and (10) we obtain

$$J \propto \int_{\mathbf{v}_F \cdot \mathbf{n} > 0} d\mathbf{k}\, \delta(\epsilon_{\mathbf{k}} - \mu) \mathbf{v}_F \cdot \mathbf{n}\, \mathrm{Im}(k_x + ik_y) \sin k_z c, \tag{11}$$

where, unlike in the case of a thick barrier, we perform φ and k_z integration. In the leading order, the right-hand side of Eq. (11) is $-\pi k_{F0}^2 \varepsilon n_z$, and yields

$$J_\delta = -A \varepsilon n_z, \tag{12}$$

where again A characterizes the normal state transparency. In contrast to the thick-barrier result, the current now vanishes in the absence of FS warping. From Eqs. (9) and (12) one can conjecture that for a general case $J \approx A(s - \varepsilon)$, where $0 \lesssim s \lesssim 1$.

The presence of small parameters ε and n_z in Eqs. (9) and (12) shows that the Josephson current in the CdW state would be reduced as compared to the conventional SQUID with s-wave electrodes. However, the alternative picture, based on the CpW state, also contains small parameters which should be kept in mind when interpreting the experiment of Ref. [8]. In addition to the small relative strength of the spin-orbit coupling (quantified by the admixture of $S_\downarrow$ into a nonrelativistic $S_\uparrow$ state, or the spin-orbit induced band shift relative to the band width [27]), there can also be another small factor—a ratio of the lattice constant and the superconducting coherence length [28], approximately 6×10^{-3} [6].

Results from Eqs. (9) and (12) confirm that the CdW state could be compatible with the phase shifts observed in Ref. [8]. Furthermore, the azimuthal dependence of an order parameter coincides for both the CdW and CpW states. While the proposed symmetry arguments [8,9] exclude most of the superconducting states allowed in the tetragonal symmetry [29], these arguments alone are not sufficient to unambiguously identify the odd pairing of the CpW state. Instead, to confirm that a CpW state has indeed been observed, one would need to accurately calculate the expected magnitudes of the Josephson current for both chiral states. In particular we propose a modification of the experimental configuration [8] such that the interface plane would be slanted at ≈45° with the c axis. If the corresponding ratio of the Josephson current to the normal state conductivity becomes substantially larger (n_z is no longer small) than in Ref. [8], it would be strong support for the chiral singlet state in Eq. (2).

Another important distinction between the two chiral states is the presence of nodes in the superconducting gap. In contrast to the CpW state, CdW requires by symmetry a horizontal line node [see Eqs. (1) and (2)]. The idea of a horizontal line node [6] has been entertained by experimentalists [30] and theorists [31] for a while, although recently it has fallen out of favor. Still, some researchers insist on the existence of a horizontal line node [32]. Moreover, in the Josephson experiments of Ref. [8] evidence was found for a substantial k_z dependence, albeit not necessarily for horizontal nodes, of the order parameter in Sr_2RuO_4 [33]. How could such a material with nearly 2D electronic structure develop a highly 3D superconducting state? To answer this question we point out the following facts: (a) practically no ferromagnetic spin fluctuations, favorable for a p-wave pairing, have been experimentally found in Sr_2RuO_4; (b) antiferromagnetic spin fluctuations at $\mathbf{q} = (2/3, 2/3, q_z)$ have negligible z dispersion; (c) the crystal structure of Sr_2RuO_4, as opposed to its electronic structure, is fairly 3D, so one can expect the electron-phonon coupling to be quite 3D as well; (d) there is a sizeable O isotope effect in Sr_2RuO_4, which strongly changes with the introduction of pair-breaking defects [34]. While electron-phonon coupling *per se* can only induce an s-wave pairing, such a pairing would be prevented by the strong antiferromagnetic spin fluctuations. However, for the proposed CdW state, any 2D interaction cancels out, including the magnetic interaction. Should the electron-phonon coupling have a maximum say, at $(1/2, 1/2, 1/2)$, as opposed to $(1/2, 1/2, 0)$, the CdW state would have been immediately stabilized providing a plausible scenario for spin-singlet superconductivity in Sr_2RuO_4 [35].

In conclusion, we have revealed that a completely overlooked chiral d-wave pairing state in Sr_2RuO_4 is equally compatible with the existing body of experimental data as the generally accepted chiral p-wave state. We have proposed phase-sensitive experiments in a SQUID geometry with a variable tilting angle capable of unambiguously distinguishing between the two chiral states.

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