Superconducting Pairing Mechanism of Sr$_2$RuO$_4$

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Recent ultrasound experiments unveiled two-component nature of the superconducting order parameter in Sr$_2$RuO$_4$ and proposed $d_{x^2-y^2} + i d_{xy}$ and the accidentally degenerate $d_{x^2-y^2} + i g$ as possible candidates. To clarify the true pairing symmetry, we construct a phenomenological model containing all symmetry-allowed multipole fluctuations as potential pairing glues and make a systematic survey of major pairing states within the Eliashberg framework. This allows us to exclude with certainty $d_{x^2-y^2} + i d_{xy}$ based on the quasi-two-dimensionality of the Fermi surface topology and establish $d_{x^2-y^2} + i g$ as the most probable pairing state. The latter is found to arise from the interplay of antiferromagnetic, ferromagnetic, and electric multipole fluctuations and agree well with most current experiments. Our work provides a promising basis for understanding the pairing mechanism in superconducting Sr$_2$RuO$_4$.

I. INTRODUCTION

For over two decades, superconductivity in Sr$_2$RuO$_4$ had been proposed to be of spin-triplet pairing both in theory and in experiment [1–5]. This belief has lately been overturned when refined nuclear magnetic resonance (NMR) [6, 7] and polarized neutron scattering (PNS) [8] experiments detected a drop in the spin susceptibility below $T_c$. Muon spin relaxation ($\mu$SR) and polar Kerr effect revealed time-reversal symmetry breaking (TRSB) of the superconducting order parameter [10, 11]. A line-node gap was then supported by specific heat [12, 13], penetration depth [15], thermal conductivity [16, 17], spin-lattice relaxation rate [18], and quasiparticle interference imaging [19]. Very recently, ultrasound experiment reported a thermodynamic discontinuity in the shear elastic modulus and put further constraint on the pairing symmetry [20, 21]. Candidate proposals of two-component TRSB order parameters include $d_{x^2-y^2} + ig$ [22, 23], $s + id_{x^2-y^2}$ [24], $s + id_{xy}$ [25], chiral or helical or mixed p-wave [26–34], $d_{xz} + id_{yz}$ [35], and exotic interorbital pairings [36–41]. Among them, $d_{xz} + id_{yz}$ and $d_{x^2-y^2} + ig$ can satisfy the ultrasound requirement. While $d_{xz} + id_{yz}$ also seems supported by $\mu$SR under pressure [42, 43], its nodal structure is inconsistent with spectroscopic measurements by scanning tunneling microscope (STM) [19]. The accidentally degenerate $d_{x^2-y^2} + ig$ state can fit most experiments including the STM, but how the $g$-wave can arise and become degenerate with $d_{x^2-y^2}$ remains unclear. The exact pairing symmetry of Sr$_2$RuO$_4$ has not been decided.

To resolve this issue, we construct here a general model Hamiltonian combining realistic band structures from angle-resolved photoemission spectroscopy (ARPES) and multipole pairing interactions allowed by symmetry for the spin-orbit coupled Ru-4$d$ electrons. The superconducting gap structures are then evaluated systematically by solving the linearized Eliashberg equations with anti-ferromagnetic (AFM), ferromagnetic (FM), electric multipole fluctuations and their mixtures. We find that the $d_{x^2-y^2} + ig$ (pseudospin) singlet pairing is the most probable candidate and can be realized by the interplay of all three types of multipole fluctuations, while $d_{xz} + id_{yz}$ is theoretically not favored within proper parameter range from experiments due to the quasi-two-dimensional Fermi surface topology. A candidate $s + id_{x^2-y^2}$ state can also be obtained to have the desired gap structure for STM [19], but fails to conform with the ultrasound experiment. Our work may help to clarify the nature of superconductivity in Sr$_2$RuO$_4$.

II. MODEL

Spin-orbit coupling (SOC) is considered important in Sr$_2$RuO$_4$ [44]. To capture its superconducting symmetry, we first construct a general model Hamiltonian based on multipole representations of the pairing interactions. By Stevens operator-equivalent technique, multipole operators $Q^{jq}$ ($k = 0, 1, \ldots, 2j; q = -k, -k + 1, \ldots, k$) for a given angular momentum $j$ can be obtained from the $(2j + 1) \times (2j + 1)$ tensor operator $\hat{J}_{kq}$ satisfying [15, 46]:

$$
\hat{J}_{kk} = (-1)^j \sqrt{\frac{(2k-1)!}{(2k)!!}} (\hat{J}_+)^k, \\
[\hat{J}_\pm, \hat{J}_{kq}] = \sqrt{(k+q)(k+q+1)} \hat{J}_{k,q \pm 1} \quad (q < k),
$$

where $\hat{J}_\pm$ is the raising/lowering operator within the corresponding $j$-subspace. These multipole operators are further projected into the irreducible representation (IR) $\Gamma$ of the $D_{4h}$ point group of Sr$_2$RuO$_4$ and denoted as...
More details are explained in Appendix A.

| IR (Γ) | Multipole operator $\hat{Q}^{\Gamma\alpha}$ |
|--------|---------------------------------|
| $A^+_1$ | $\hat{1}$, $\tilde{O}_{20}$, $\hat{H}_0$, $\hat{H}_4$ |
| $A^+_2$ | $\hat{H}_{zz}$ |
| Electric | | |
| $B^+_1$ | $\tilde{O}_{22}$, $\hat{H}_2$ |
| $B^+_2$ | $\hat{O}_{xy}$, $\hat{H}_{xy}$ |
| $E^+_g$ | $(\tilde{O}_{xz}, \tilde{O}_{yz}), (\hat{H}_{yz}, \hat{H}_{xy}), (\hat{H}_{z\hat{y}b}, \hat{H}_{z\hat{y}b})$ |
| Magnetic | | |
| $A^+_1$ | $\tilde{D}_4$ |
| $A^+_{2g}$ | $\tilde{T}_{za}, \tilde{D}_{z\alpha 1}, \tilde{D}_{z\alpha 2}$ |
| $B^+_{1g}$ | $\tilde{T}_{xyz}, \tilde{D}_2$ |
| $B^+_{2g}$ | $\tilde{T}_{zb}, \tilde{D}_{zb}$ |
| $E^-_g$ | $(\tilde{D}_{z\alpha 1}, \tilde{D}_{z\alpha 1}), (\tilde{D}_{z\alpha 2}, \tilde{D}_{z\alpha 2}), (\tilde{D}_{zb}, \tilde{D}_{zb})$ |

$\hat{Q}^{\Gamma\alpha}$ for the $\alpha$-th component in $\Gamma$ \[47, 48\]. Table II gives all multipole operators for the $j = 3/2$ and $5/2$ manifolds of Ru-4$d$ electrons. The electric multipoles are of even-rank and time-reversal symmetric and listed on the top of the table, while on the bottom are the magnetic multipoles (odd-rank and time-reversal antisymmetric) \[48, 49\]. More details on the definition of these multipole operators can be found in Appendix A.

We then write down a general interaction containing all symmetry-allowed multipole fluctuations as potential superconducting pairing glues:

$$H_{\text{int}} = -\sum_{j,\Gamma} \sum_{\alpha \beta} \sum_{m' m} g_{\alpha \beta}^{\Gamma} V^j_{\Gamma}(q) \hat{Q}^{\Gamma\alpha \dagger}(q) \hat{Q}^{\Gamma\beta}(q)$$

$$= -\sum_{j,\Gamma} \sum_{\alpha \beta} \sum_{m m'} \sum_{k, k'} g_{\alpha \beta}^{\Gamma} V^j_{\Gamma}(q) Q_{m l m' n l m}^{\Gamma\alpha} Q_{m l m' n l m}^{\Gamma\beta}$$

$$\times c_{l, k, -q m, k}^{\dagger} c_{l', q m'}^{\dagger} c_{l', q m'} c_{l, -q m},$$

where $Q_{m l m}^{\Gamma\alpha}(q) = \sum_{k, l m} Q_{l m}^{\Gamma\alpha} c_{l, k, q m, k}^{\dagger} c_{l, -q m, k}$ and $c_{m, k} (c_{m, k}^{\dagger})$ is the electron annihilation (creation) operator with $k$ being the momentum and $m$ the $z$-projection of the total angular momentum $j$. The matrix elements $Q_{l m}^{\Gamma\alpha}$ are normalized with $Q_{l m}^{\Gamma\alpha} \rightarrow Q_{l m}^{\Gamma\alpha} / \sqrt{\sum_{l' m'} |Q_{l m}^{\Gamma\alpha}|^2}$ for comparison of different multipole fluctuations, $V^j_{\Gamma}(q)$ is the momentum-dependent interaction vertex, and $g_{\alpha \beta}^{\Gamma}$ controls the fluctuation strength between the multipole components $\alpha$ and $\beta$, as illustrated in Fig. 1(a). The values of $g_{\alpha \beta}^{\Gamma}$ are highly restricted as the multipole product should be projected to the identity representation to keep the overall symmetry of the Hamiltonian. Thus only multipoles belonging to the same IR can be coupled. For the two-dimensional IR $E^+_g$, such projection yields $(Q_{E^+_g}^{\Gamma\alpha} \hat{Q}^{\Gamma\beta} + Q_{E^+_g}^{\Gamma\beta} \hat{Q}^{\Gamma\alpha})/2$, which will be denoted as $Q_{E^+_g}^{\Gamma\alpha} \hat{Q}^{\Gamma\beta}$ for simplicity. There are a total number of 6 electric multipole fluctuation channels and 11 magnetic multipole fluctuation channels in the $j = 3/2$ manifold, and 23 electric components and 38 magnetic components in the $j = 5/2$ manifold, that are allowed by symmetry in Sr$_2$RuO$_4$. For clarity, we arrange them according to their IR and rank. For example, the 6 electric multipole channels for $j = 3/2$ are listed as $\sum_{l m}$ \[1\], \[1\] $\tilde{O}_{20}$, \[2\] $\tilde{O}_{22}$, \[2\] $\tilde{O}_{xy}$, \[2\] $\tilde{O}_{z\hat{z}}$, \[2\] $\tilde{O}_{z\hat{z}}$, \[2\] $\tilde{O}_{z\hat{z}}$.

The above procedures lay out a general phenomenological framework for studying electron pairing induced by multipole fluctuations. To apply it to Sr$_2$RuO$_4$, we consider the following three dimensional (3D) tight-binding (TB) model, $H_K = H_0 + H_z$, where $H_0 = \sum_k \psi_{s}(k) h_0(k, s) \psi_{s}(k)$ describes the $k_z$-independent band structure from ARPES measurements \[51\]. $\psi_{s}(k) = [c_{xz, x}(k), c_{yz, y}(k), c_{zy, -y}(k)]^T$ is the basis of the low-lying
Ru-4d \( t_{2g} \) orbitals \( (d_{xz}, d_{yz}, d_{xy}) \). We have

\[
h_0(k, s) = \begin{pmatrix}
    \epsilon_k^{xx} - \mu_0 & \epsilon_k^{xy} - i\lambda_{SOC} \\
    \epsilon_k^{xy} + i\lambda_{SOC} & -\mu_0
\end{pmatrix},
\]

with \( s = \pm \) for the spin and

\[
\epsilon_k^{xx} = -2t_1 \cos(k_x) - 2t_2 \cos(k_y),
\]

\[
\epsilon_k^{xy} = -2t_2 \cos(k_x) - 2t_1 \cos(k_y),
\]

\[
\epsilon_k^{xz} = -2t_3(\cos(k_z) + \cos(k_y)) - 4t_4 \cos(k_z) \cos(k_y) - 2t_5(\cos(k_x) + \cos(k_y)),
\]

\[
\epsilon_k^{off} = -4t_6 \sin(k_z) \sin(k_y).
\]

The \( H_z \) term describes the hopping along \( z \)-direction and is introduced to deal with out-of-plane pairing such as \( d_{xz} \) and \( d_{yz} \). Under the same basis \( \psi_s(k) \), it takes the form,

\[
H_z(k) = -8t_0 \cos(k_x/2) \cos(k_y/2) \cos(k_z/2).
\]

The best ARPES fit yields \( |t_1, t_2, t_3, t_4, t_6, \mu_0, \lambda_{SOC}| = [0.145, 0.015, 0.081, 0.039, 0.05, 0.122, 0.032] \) eV \([51]\).

Candidate pairing symmetries of the superconductivity can be analyzed using the linearized Eliashberg equations \([58, 59]\). The kernel functions \( K^N_{\mu\mu'}(k, i\omega_n; k', i\omega_{n'}) \) and \( K^\Lambda_{\mu\mu'}(k, i\omega_n; k', i\omega_{n'}) \) are given by

\[
K^N_{\mu\mu'}(k, i\omega_n; k', i\omega_{n'}) = \sum_{lml'm'\eta'\eta} \sum_{\alpha\beta} \sum_{j^T} g_{\alpha\beta}^{lT} V^{1T}(k - k', i\omega_n - i\omega_{n'}) Q_{ml}^{1T} Q_{ml'}^{1T} \psi_{j^T}(k, i\omega_n) \epsilon_{j^T}(k, i\omega_n) Q_{ml'}^{1T} \psi_{j^T}(k', i\omega_{n'}),
\]

\[
K^\Lambda_{\mu\mu'}(k, i\omega_n; k', i\omega_{n'}) = \sum_{lml'm'\eta'\eta} \sum_{\alpha\beta} \sum_{j^T} g_{\alpha\beta}^{lT} V^{1T}(k - k', i\omega_n - i\omega_{n'}) Q_{ml}^{1T} Q_{ml'}^{1T} \psi_{j^T}(k, i\omega_n) \epsilon_{j^T}(k, i\omega_n) Q_{ml'}^{1T} \psi_{j^T}(k', i\omega_{n'}),
\]

where \( \hat{\epsilon}^{k} \) is the matrix diagonalizing the 3D (2D) TB Hamiltonian \( H_K \) (\( H_0 \)), projected in the \( j \) representa-
For AFM fluctuations, inelastic neutron scattering (INS) experiments estimate $\xi_{xy}^\text{AFM} = 9.7$ Å and $\omega_q^\text{AFM} = 11.1$ meV at the AFM wave vector $Q_{\text{AFM}} = (0.3, 0.3, 0)$ \cite{53, 55}. The longitudinal correlation length is set to $\xi_{xy} = 0.1\xi_{xy}^\text{AFM}$ to reflect the absence of $z$-axis signal \cite{54}. Among all 11 AFM multipole fluctuation channels, most of them support $d_{x^2-y^2}$ or $s$. Two leading fluctuation channels from RPA analysis, $\hat{J}_{2z}$ and $\hat{T}_{za}\hat{T}_{ra}$, gives predominant $d_{x^2-y^2}$-wave pairing. The subordinate channels, $\hat{J}_{s,T_{za}}\hat{T}_{xy}, \hat{T}_{zy}, \hat{T}_{xy}\hat{T}_{rb}$, also support $d_{x^2-y^2}$, while the subordinate $\hat{T}_{zy}\hat{T}_{rb}, \hat{J}_{r,r}, \hat{J}_{r,T_{ra}}, \hat{J}_{r,T_{rb}}$ favor $s$-wave and $\hat{T}_{za}\hat{T}_{za}$ favors $(p_x, p_y)$ or $p_x + ip_y$. All these pairing states are $m_z$-symmetric, i.e., symmetric about the $k_z = 0$ plane. The $m_z$-antisymmetric $d_{x^2+y^2}$ or $(d_{xz}, d_{yz})$ pairing state can also be obtained but is not favored for the TB Hamiltonian which is quasi-2D and weakly dispersive along $k_z$ direction. This conclusion is robust against reasonable tuning of $\xi_0$ and $\xi_z$. The results for $\xi_0 = \xi_{xy}$ are summarized in Appendix C.

FM pairing interactions have previously been considered because Sr$_2$RuO$_4$ has similar electronic structures as the itinerant ferromagnets SrRuO$_3$ and Sr$_3$Ru$_2$O$_7$ and the metamagnet Sr$_2$Ru$_2$O$_7$ \cite{53, 61}. PNS experiment has reported a broad FM response \cite{55}, giving $Q_{\text{FM}} = (0, 0, 0)$, $\xi_{xy}^\text{FM} = 2.5$ Å, and a characteristic energy $\omega_0^\text{FM} = 15.5$ meV. Since there is no experimental data for the spin wave dispersion, we use $\omega_q = \omega_0^\text{FM}|_q$ and choose $v_0$ such that $\omega_q$ reaches the order of $\omega_0^\text{FM}$ at the zone boundary. A slight variation of $v_0$ makes no qualitative change on our main conclusions. Figure 2(b) shows the typical results of five major pairing states induced by FM pairing interactions. Similarly, we find predominant $d_{x^2-y^2}$-wave pairing from leading dipole fluctuations $\hat{J}_{2z}, \hat{T}_{xy}$ and $p$-wave from leading octupole $\hat{T}_{za}\hat{T}_{ra}$ and $\hat{T}_{zy}\hat{T}_{rb}$, while the $s$-wave is supported by some subordinate multipole channels.

Electric fluctuations may arise from multi-orbital nature of Sr$_2$RuO$_4$ \cite{62, 66} and have similar interaction vertex as AFM ones, but with $Q_{\text{E}} = (0.2, 0.2, 0)$, $\xi_{xy}^\text{E} = \xi_{xy}^\text{AFM}$, and $\omega_0^\text{E} = \omega_0^\text{AFM}$. As shown in Fig. 2(c), all six multipole channels support $s$-wave pairing, which is robust under the tuning of $\xi_{xy}^\text{E}$ and $\omega_0^\text{E}$. Figure 2(d) plots the eigenvalues of five major pairing states as a function of $Q_{\text{E}}$ along the (110) direction. We see that the $s$-wave pairing always has a much larger eigenvalue than others. This is expected since superconductivity induced by charge fluctuation is typically $s$-wave. But quite interestingly, the eigenvalue of $s$ reaches a maximum around $Q_{\text{E}} = (0.2, 0.2, 0)$, exactly the wave vector proposed by the RPA charge susceptibility \cite{67}, implying a potential role of electric multipole fluctuations in superconducting Sr$_2$RuO$_4$.
wave can be induced by a moderate FM pairing interaction \((r_2/r_1 > 0.5)\), while for a strong electric interaction \((r_2/r_1 > 1)\), we find a predominant nodeless \(s\)-wave with gap minima on \(\gamma\) band. For distinction, we will use \(s'\) to denote nodal \(s\)-wave in the following. As discussed earlier, \(d_{x^2-y^2}\) and \(s\) (or \(s'\)) are major pairing states for pure AFM, FM or electric multipole fluctuations. But quite surprisingly, Fig. 4 shows that \(g\)-wave pairing can become dominant over a large portion of the phase diagram where both FM and electric multipole fluctuations are equally important as the AFM ones.

Under this premise, accidentally degenerate \(d_{x^2-y^2}+ig\) pairing may appear at the phase boundary with a somewhat weaker FM pairing interaction than the AFM one, namely \(r_2/r_1 \approx 0.5\). However, a moderate electric pairing interaction \((0.5 < r_2/r_1 < 2)\) is also required. If electric fluctuations are too weak, a two-component \(s' + id_{x^2-y^2}\) state might appear as an alternative candidate. In any case, electric multipole fluctuations, such as the 0-rank charge fluctuations, seem to play a crucial role for \(d_{x^2-y^2} + ig\) to actually appear in Sr\(_2\)RuO\(_4\), and should be better examined by future X-ray diffraction or Raman experiments [63–71].

The emergence of \(g\)-wave is robust for such mixed AFM, FM, and electric pairing interactions. Appendix E shows the phase diagrams of two other examples of averaged pairing interactions. In the first case, we include \(j = 5/2\) and take an averaged pairing interaction over all multipole fluctuations; and in the second case, we keep \(j = 3/2\) but consider only leading multipole components in the averaged AFM and FM interactions. In both cases, the phase diagrams are similar as in Fig. 3b) and the \(g\)-wave state covers a large portion of the phase diagram for mixed AFM, FM, and electric pairing interactions, where \(d_{x^2-y^2}\) and \(s\) (or \(s'\)) solutions supported by leading multipole fluctuations are suppressed. If these leading fluctuations dominate the pairing interaction, the phase boundaries will shift slightly towards a larger \(r_2/r_1\). We conclude that the competition and interplay of AFM, FM, and electric multipole fluctuations may give rise to a mechanism for this unusual \(g\)-wave pairing in Sr\(_2\)RuO\(_4\). Whether or not this reflects the true situation in real materials requires future experimental scrutiny on their relative weights.

To have an idea of the gap structures for these pairing states, Fig. 3 presents their projection on the 2D Fermi surfaces and evolution with the azimuth \(\phi\). Both \(d_{x^2-y^2}\) and \(g\) show clear nodes on three bands along the zone diagonal (\(\phi = \pm \pi/4\) and \(\pm 3\pi/4\)). The resulting \(d_{x^2-y^2} + ig\) gap has nodes along the zone diagonal, which is protected by symmetry and fits well the STM data [19]. Quite unexpectedly, we also find that the \(s'\)-wave can change sign or have gap minima near the zone diagonal. This interesting feature arises from the particular orbital character of the three bands. Along the diagonal direction, \(\alpha\) and \(\beta\) bands contain no contribution from \(|j = 3/2, j_z = \pm 1/2\rangle\), and the \(\gamma\) band contains no contribution from \(|j = 3/2, j_z = \pm 3/2\rangle\). Hence, an \(s'\)
wave paired supported by multipole fluctuations with only \( j_z = \pm 1/2 \) or \( \pm 3/2 \) components must have nodes along the zone diagonal on \( \alpha/\beta \) or \( \gamma \) bands, respectively. This gives rise to the gap minima after other \( j_z \) contributions are included. As a result, the two-component \( s'+id_{x^2-y^2} \) also exhibits gap minima near the zone diagonal. For the above averaged pairing interaction, we find a relative gap ratio \(|\Delta_{\min}/\Delta_{\max}| \approx 0.11, 0.01, 0.16\) for \( \alpha, \beta, \gamma \) bands, respectively. Note that the previous STM experiment has an energy resolution of about 75 \( \mu eV \), which is roughly 21% of the measured gap of 350 \( \mu eV \) [19]. Hence, within the resolution of the STM experiment, it might not be possible to distinguish the predicted \( s'+id_{x^2-y^2} \) and \( d_{x^2-y^2}+ig \) pairing symmetry.

VI. DISCUSSION AND CONCLUSION

Under the assumption of a two-component order parameter that breaks the time reversal symmetry, we have examined candidate pairing states \( px+ipy, dx+idy, s'+id_{x^2-y^2} \), and \( d_{x^2-y^2}+ig \) for \( Sr_2RuO_4 \) by constructing a general model Hamiltonian with all symmetry-allowed multipole fluctuations as the pairing interaction.

The spin-triplet \( px+ipy \) pairing has been excluded by a series of NMR [4, 8] and PNS [9] experiments; its nodeless gap is also inconsistent with STM experiment. In our theory, it is only supported by the relatively weaker ferromagnetic octupole fluctuations. A mixed state of spin singlet-triplet pairing with line nodes might be possible, but is too complicated to realize.

The \( d_{xz}+idy \) pairing can also be excluded with certainty due to the quasi-two-dimensional Fermi surface topology of \( Sr_2RuO_4 \). It is featured with horizontal line nodes on the \( k_z = 0 \) plane and is supposed to cause \( L \) modulated intensity of the spin resonance [72]. But this expected resonance peak at \( L = \pm 0.5 \) was not observed in latest neutron scattering experiment [73]. The \( d_{xz}+idy \) pairing was mostly supported by \( \mu\text{SR} \) measurements that reported the splitting of superconductivity and TRSB under uniaxial pressure opposed to hydrostatic pressure [42, 43]. However, the splitting was questioned by specific heat measurements, which found no sign of bulk phase transition induced by uniaxial pressure [74]. More accurate experiments will clarify how exactly superconductivity evolves under pressure. Alternatively, a spin-triplet odd-orbital \( d_{xz}+idy \) pairing has been proposed based on momentum-dependent SOC [23, 36, 37], but it is inconsistent with NMR [8] and PNS [9] experiments.

The \( d_{x^2-y^2} \) wave has the desired vertical line nodes revealed by thermal conductivity [17] and nodes or gap minima on \( \alpha \) and \( \beta \) bands in STM measurements [10]. From our calculations, it is indeed supported by AFM fluctuations and can form a two-component order parameter with accidentally degenerate \( s' \) or \( g \) in the presence of moderate FM and electric fluctuations. An \( s'+id_{x^2-y^2} \) has been proposed in previous theory but was nodeless along the zone diagonal [24]. By contrast, our derived \( s'+id_{x^2-y^2} \) has nodes or gap minima near the 2D zone diagonal and agrees with STM experiments. However, \( s'+id_{x^2-y^2} \) seems inconsistent with ultrasound experiment, where the observed thermodynamic jump of the shear elastic modulus \( \delta c_{66} \propto \alpha_2^4 \) reflects the coupling term \( \alpha_4 u_{xy}(\Delta_s, \Delta_{d_{x^2-y^2}}, \Delta_{d_{x^2-y^2}}, \Delta_s) \) between the strain \( u_{xy} \) and two superconducting components in the Landau free energy [20, 21]. But such coupling is prohibited by symmetry because \( B_{2g}(u_{xy}) \cdot A_{1g}(\Delta_s) \cdot B_{1g}(\Delta_{d_{x^2-y^2}}) = A_{2g} \neq A_{1g} \). To overcome this [23, 21], an \( s'+id_{xy} \) pairing has been introduced by considering nearest-neighbor...
mechanism of Sr provides a most plausible basis for understanding the pairing and fluctuations. O
harmonics as the basis [47, 50]. For 1D or 2D IR of 151, the China Postdoctoral Science Program of the Chinese Academy of Sciences (Grant No. 2017YFA0303103), the Strategic Priority Research Program of MOST of China (Grant No. 2017YFA0303103), the China Postdoctoral Science Foundation (Grant No. 2020M670422), and the Youth Innovation Promotion Association of CAS.

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Appendix A: Multipole operators

Definitions of the multipole operators under \( D_{5g} \) point group are listed in Table I and formed by the Hermitian tensor operators (for \( q \neq 0 \))

\[
\hat{J}_{kpc} = \frac{1}{\sqrt{2}} \left[ (-1)^q \hat{J}_{kq} + \hat{J}_{k,-q} \right],
\]

\[
\hat{J}_{kqs} = \frac{1}{\sqrt{2i}} \left[ (-1)^q \hat{J}_{kq} - \hat{J}_{k,-q} \right].
\]

(A1)

For \( q = 0 \), \( \hat{J}_{k0} \) is itself Hermitian. Different notations have been used for multipole operators in the literature [47, 49, 80, 81]. Here we follow the convention in Ref. 49 and use the tesseral harmonics in \( O_{5g} \) point group or cubic harmonics as the basis [47, 50]. For 1D or 2D IR of \( O_h \), the subscript denotes the tesseral harmonics \( Z_{kq}(\hat{f}) \). For 3D IR of \( O_h \), the subscripts in \( \hat{O}_{xz}, \hat{O}_{yz}, \hat{O}_{xy} \) represent the basis function \( (xz, yz, xy) \), while other multipoles are marked by the subscript \( (x, y, z) \) with additional \( a (b) \) denoting the \( T_{1g}/u \) (\( T_{2g}/u \)) IR, 1 (2) for different equal rank basis in the same IR, and \( g/u \) for inversion symmetric/antisymmetric. For instance, \( (\hat{O}_{zz}, \hat{O}_{zz}) \) in the \( E_g \) IR correspond to tesseral harmonics \( r^2 \hat{Z}_{zz}(\hat{f}) \propto x^2 - y^2 \) and \( r^2 \hat{Z}_{zz}(\hat{f}) \propto 3z^2 - r^2 \), respectively; \( \{\hat{D}_{xx1}, \hat{D}_{xy1}, \hat{D}_{yy1}\} \) correspond to the first basis in \( T_{1u} \) IR [47, 49, 80, 81]. For simplicity, we have dropped the label \( j \) of the total angular momentum. Multipole operators belonging to different \( j \)-spaces may have same notations but different representation matrices. As examples, the matrices for the dipole \( J_2 \) and octupoles \( T_{xa}, T_{xyz} \) are

\[
\hat{J}_z = \frac{1}{2\sqrt{5}} \begin{pmatrix} -3 & -1 & 1 \\ 1 & 1 & 3 \end{pmatrix},
\]

\[
\hat{T}_{xa} = \frac{1}{4\sqrt{5}} \begin{pmatrix} -\sqrt{3} & 3 & 5 \\ 3 & 3 & -\sqrt{3} \end{pmatrix},
\]

\[
\hat{T}_{xyz} = \frac{i}{2} \begin{pmatrix} -1 & 1 & 1 \\ 1 & 1 & -1 \end{pmatrix},
\]

in the \( j = 3/2 \) subspace and

\[
\hat{J}_z = \frac{1}{\sqrt{70}} \begin{pmatrix} -5 & -3 & 1 \\ -3 & 1 & 3 \\ 1 & 3 & 5 \end{pmatrix},
\]

\[
\hat{T}_{xa} = \frac{1}{12} \begin{pmatrix} -3 & -3 & \frac{3}{\sqrt{10}} & \frac{5}{\sqrt{2}} & 2\sqrt{5} \\ \frac{3}{\sqrt{2}} & \frac{6}{\sqrt{5}} & \frac{3}{\sqrt{10}} & \frac{5}{\sqrt{2}} & -3 \\ \frac{5}{\sqrt{2}} & \frac{3}{\sqrt{10}} & \frac{3}{\sqrt{10}} & -3 \end{pmatrix}
\]

\[
\hat{T}_{xyz} = \frac{i}{2\sqrt{6}} \begin{pmatrix} -\sqrt{5} & -1 & -1 \\ 1 & \frac{1}{\sqrt{5}} & 1 \\ \frac{1}{\sqrt{5}} & -1 & \frac{1}{\sqrt{5}} \end{pmatrix}
\]

in the \( j = 5/2 \) subspace. All multipole matrices are normalized with \( Q_{lm}^{T\alpha} \rightarrow Q_{lm}^{T\alpha}/\sqrt{\sum_{l'm'}|Q_{lm}^{T\alpha}|^2} \). For two-dimensional IR \( E_g^\pm \), we fix the sign in the definition of
Table II: Definition of the multipole operators in Table I under $D_{4h}$ point group.

| $\Gamma$ | $\alpha$ | $\hat{Q}^{I\alpha}$ | Basis |
|----------|----------|----------------------|--------|
| $A^+_{1g}$ | 1 | $\hat{1}$ | $J_{00}$ |
|          | 2 | $\hat{O}_{20}$ | $\hat{J}_{20}$ |
|          | 3 | $\hat{H}_{0}$ | $(\sqrt{7}\hat{J}_{40} + \sqrt{5}\hat{J}_{44})/\sqrt{12}$ |
|          | 4 | $\hat{H}_{4}$ | $(\sqrt{7}\hat{J}_{40} - \sqrt{7}\hat{J}_{44})/\sqrt{12}$ |
| $A^+_{2g}$ | 1 | $\hat{H}_{1a}$ | $J_{44}$ |
| Electric | $B^+_{1g}$ | 1 | $\hat{O}_{22}$ | $\hat{J}_{22}$ |
|          |          | 2 | $\hat{H}_{2}$ | $-\hat{J}_{42}$ |
|          | $B^+_{2g}$ | 1 | $\hat{O}_{xy}$ | $\hat{J}_{2s}$ |
|          |          | 2 | $\hat{H}_{sb}$ | $\hat{J}_{42}$ |
| $E^+_{g}$ | 1 | $\hat{O}_{xx}, \hat{O}_{ya}$ | $J_{31c}, J_{12}$ |
|          | 2 | $\hat{H}_{xa}, \hat{H}_{ya}$ | $-\hat{J}_{43c} + \sqrt{7}\hat{J}_{41c})/\sqrt{8}, (\hat{J}_{43s} + \sqrt{7}\hat{J}_{41s})/\sqrt{8}$ |
|          | 3 | $\hat{H}_{yb}, \hat{H}_{yb}$ | $-\sqrt{7}\hat{J}_{43c} - J_{41c})/\sqrt{8}, (\sqrt{7}\hat{J}_{43s} - J_{41s})/\sqrt{8}$ |
| $A^-_{1g}$ | 1 | $\hat{D}_{1a}$ | $J_{54}$ |
|          | 2 | $\hat{J}_{10}$ |
| $A^-_{2g}$ | 3 | $\hat{D}_{sa1}$ | $\hat{J}_{50}$ |
|          | 4 | $\hat{D}_{sa2}$ | $\hat{J}_{54}$ |
| Magnetic | $B^-_{1g}$ | 1 | $\hat{T}_{xy}z$ | $\hat{J}_{32}$ |
|          | 2 | $\hat{D}_{2}$ | $-\hat{J}_{52}$ |
| $B^-_{2g}$ | 1 | $\hat{T}_{z}b$ | $\hat{J}_{32}$ |
|          | 2 | $\hat{D}_{1b}$ | $\hat{J}_{52}$ |

Its two components so that $\hat{Q}^{I\alpha}_{r} \hat{Q}^{I\beta}_{r} \equiv (\hat{Q}^{I\alpha}_{r} \hat{Q}^{I\beta}_{r} + \hat{Q}^{I\alpha}_{r} \hat{Q}^{I\beta}_{r})/2$ belongs to the identity representation.

**Appendix B: Leading multipole fluctuations**

We evaluate the dynamical susceptibility $\chi^{RPA}$ from random phase approximation (RPA), project it into the $j$-space, and define an effective strength for each multipole channel using the correlation function $\Gamma$:

$$ \langle \hat{Q}^{I\alpha}_{l} \hat{Q}^{I\beta}_{m} \rangle = \sum_{l''m''} Q^{I\alpha}_{l'm'} \chi^{RPA}_{l''m''}(Q, \omega \rightarrow 0) Q^{I\beta}_{l'm'} .$$  

(B1)

The RPA susceptibility $\chi^{RPA}$ is given by

$$ \chi^{RPA}(q) = \left(1 - \Gamma_{0}\chi_{0}(q)\right)^{-1} \chi_{0}(q). $$  

(B2)

where $q$ denotes both momentum and bosonic Matsubara frequency, and $\chi_{0}(q)$ is the Lindhard susceptibility,

$$ [\chi_{0}]^{ij}_{l''l_1l_2}(q) = -T \sum_{k} G^{0}_{l_1l_2}(k) G^{0}_{l''l_1l_2}(k - q). $$  

(B3)

They are calculated based on the 2D TB Hamiltonian $H_{0}$ with an additional local Coulomb term,

$$ H_{U} = \sum_{i} \sum_{l_1l_2l_3l_4} [\Gamma^{0}]^{ij}_{l_1l_2l_3l_4} c_{i}^{\dagger} c_{l_2} c_{l_3} c_{l_4}, $$  

(B4)
where \( l \) represents both orbital and spin quantum numbers. The interaction matrix \( \Gamma_0 \) is given by

\[
\begin{align*}
\Gamma_0^{ll'} &= \Gamma_0^{ll'} = U; \\
\Gamma_0^{llm} &= \Gamma_0^{ll'm} = J; \\
\Gamma_0^{ll'm'} &= \Gamma_0^{ll'm'} = -J; \\
\Gamma_0^{ll'm'} &= \Gamma_0^{ll'm'} = -J'; \\
\Gamma_0^{llm} &= \Gamma_0^{ll'm} = U'; \\
\Gamma_0^{llm'} &= \Gamma_0^{ll'm'} = U - J; \\
\Gamma_0^{llm'} &= \Gamma_0^{ll'm'} = J - U',
\end{align*}
\]

where spin up or down is distinguished by the indices with or without prime. The parameters are fixed as \( J = 0.17U, J' = J \) and \( U' = U - 2J \) according to previous LDA+DMFT study \[63\]. The obtained RPA spin susceptibility peaks at \( Q_{\text{RPA}} = (0.37, 0.37) \), close to the experimental \( Q_{\text{AFM}} \).

Figure 5 compares the symmetry-allowed AFM, FM, and electric multipole correlations as functions of the Stoner factor \( \alpha_S \) at their respective wave vectors. The Stoner factors are defined as the largest eigenvalue of the matrix \( \hat{\Gamma}_0 \chi_0(q) \) for given \( Q \) \[84\]. For \( j = 3/2 \), we find, among all 11 magnetic multipoles, two leading components, \( \langle \hat{J}_s \hat{J}_z \rangle \) and \( \langle \hat{T}_{raT_{ra}} \rangle \), in the AFM channel, and three leading diagonal components, \( \langle \hat{J}_z \hat{J}_z \rangle, \langle \hat{T}_{raT_{ra}} \rangle, \langle \hat{T}_{rbT_{rb}} \rangle \), in the FM channel. For \( j = 5/2 \), among all 15 diagonal magnetic multipoles, we find two leading diagonal components, \( \langle \hat{J}_z \hat{J}_z \rangle, \hat{D}_{2a22} \hat{D}_{2a22}, \) for AFM fluctuations, and three leading diagonal components, \( \langle \hat{J}_z \hat{J}_z \rangle, \hat{D}_{ra1} \hat{D}_{ra1}, \hat{D}_{rb1} \hat{D}_{rb1}, \) for FM fluctuations. The off-diagonal multipole fluctuations, \( Q^{ij}_F Q^{jF} \beta \) with \( \alpha \neq \beta \), are also included in our analysis of individual or mixed pairing interactions. They may induce a negative renormalization function and were usually ignored in the literature for determining the pairing symmetry \[40, 82\]. The electric multipole fluctuations show no significant variation for \( \alpha_S^F < 1 \) in both \( j \)-spaces and all their components are considered equally.

**Appendix C: Pairing states with \( \xi_z = \xi_{xy} \) for \( j = 3/2 \)**

Our conclusion that \( d_{xz} + id_{yz} \) is not favored is robust against parameter tuning. As an example, Fig. 6 shows five major pairing states with an artificially enlarged \( \xi_z = \xi_{xy} \) for all three channels (AFM, FM, electric) in the \( j = 3/2 \) manifold. All other parameters are the same as in Fig. 2. A larger value of \( \xi_z \) does enhance \( d_{xz} + id_{yz} \) and other \( m_z \)-antisymmetric pairing states, but it is still not enough to make them predominant. This excludes with certainty \( d_{xz} + id_{yz} \) as the candidate pairing symmetry in the quasi-2D superconductor Sr2RuO4.
Appendix D: Pairing states for $j = 5/2$

The $d_{x^2} + id_{y^2}$ pairing is also not favored for $j = 5/2$. Figure 7 shows five major pairing states induced by 38 AFM or FM multipole fluctuation channels and 23 electric channels. For the AFM channel, the leading dipole component $\hat{J}_z \hat{J}_z$ supports $d_{x^2 - y^2}$, while the leading dotriacontapole $\hat{D}_{2a2} \hat{D}_{2a2}$ supports $s$-wave pairing. For the FM channel, the leading dipole $\hat{J}_z \hat{J}_z$ favors $s$-wave, while the leading dotriacontapole $\hat{D}_{ra1} \hat{D}_{ra1}$ and $\hat{D}_{rb} \hat{D}_{rb}$ support $p$-wave. Thus, compared to $j = 3/2$, the $j = 5/2$ multipole fluctuations on average tend to enhance more $s$-wave pairing than $d_{x^2 - y^2}$. As a result, this may reduce the phase boundary of $d_{x^2 - y^2}$ on the theoretical phase diagram of superconducting Sr$_2$RuO$_4$.

Appendix E: Robustness of $g$-wave

To show the robustness of $g$-wave for mixed AFM, FM, and electric multipole fluctuations, we have also calculated the phase diagram for the pairing interaction averaged over all multipole components for both $j = 3/2$ and $j = 5/2$ manifolds. As shown in Fig. 8(a), there is no sig-

![FIG. 7: Comparison of five major pairing states obtained by diagonalizing the linearized Eliashberg equations on the 3D Fermi surfaces of Sr$_2$RuO$_4$ from (a) 38 AFM multipole fluctuation channels; (b) 38 FM channels; and (c) 23 electric channels for $j = 5/2$. All parameters are the same as in Fig. 4. The table on the bottom lists all multipole fluctuation channels for $j = 5/2$ sorted according to their IR and rank.](image1)

![FIG. 8: Theoretical phase diagram of the superconductivity in Sr$_2$RuO$_4$ for the mixed pairing interaction averaged over (a) all multipole channels in both $j = 3/2$ and $j = 5/2$; (b) only leading multipole channels for AFM and FM and in $j = 3/2$. The insets show typical gap structures in each region. All parameters are the same as in Fig. 4.](image2)
ificant change in the phase diagram. Only the $d_{x^2-y^2}/s'$ phase boundary is shifted slightly towards a smaller $r_2/r_1$ as discussed in Appendix D. One still obtains a dominant $g$-wave in the broad intermediate region and possible two-component $d_{x^2-y^2} + ig$ pairing with strong AFM pairing interaction and moderate FM and electric multipole fluctuations. On the other hand, if only the few leading multipole components are considered in the average for AFM and FM pairing interactions, $d_{x^2-y^2}$ and $s'$ will be enhanced, causing a slight shift of the phase boundary towards a larger $r_3/r_1$, as shown in Fig. 8(b). The situation in real materials is expected to lie in between, but presently the relative strengths of different multipole fluctuations are not known for Sr$_2$RuO$_4$. We leave this for future experimental examination.

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