Orbital Fluctuation-Induced Triplet Superconductivity: 
Mechanism of Superconductivity in Sr$_2$RuO$_4$

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The mechanism of superconductivity in Sr$_2$RuO$_4$ is studied using a degenerate Hubbard model within the weak coupling theory. When the system approaches the orbital instability which is realized due to increasing the on-site Coulomb interaction between the electrons in the different orbitals, it is shown that the triplet superconductivity appears. This superconducting mechanism is only available in orbitally degenerate systems with multiple Fermi surfaces.

Since the discovery of the superconductivity in Sr$_2$RuO$_4$ below $T_c=1.5$K experimental and theoretical investigations on this compound have been carried out intensively. The main features of the results of investigations may be summarized as follows; (1) this compound has the same crystal structure as the high $T_c$ cuprate La$_{2-x}$Sr$_x$CuO$_4$, (2) the conduction electrons exhibit the 2D Fermi liquid property, (3) there are three cylindrical $t_{2g}$ Fermi surfaces, (4) $T_c$ is strongly suppressed to less than 1.5K by the non-magnetic impurities, (5) no Hebel-Slichter peak was seen in 1/K experiments. (6) The NMR Knight shift and $\mu$-SR experiments indicate that the spin triplet state breaking the time reversal symmetry is realized.

In an early stage of investigations, it was considered that the symmetry of the superconducting order parameter in Sr$_2$RuO$_4$ is of the $p$-wave similar to that in $^3$He and the pairing is caused by ferromagnetic spin fluctuation enhanced by the Hund’s coupling. From these facts, it was considered that the most consistent superconducting order parameter is represented by $d(k) = \tilde{\varepsilon}(k_x + ik_y)$. Recently, some experimental results suggest that the existence of line nodes in the superconducting gap. This result is incompatible with the above assumed form of $d(k)$. Also, the inelastic neutron scattering data does exhibit the incommensurate magnetic response consistent with the nesting vector predicted from the band-structure calculation, but no discernible response around $q = 0$. In view of these experimental results throwing some doubts about the $p$-wave scenario, it is desirable to develop a more detailed theory from a microscopic point of view.

In this letter, we investigate the mechanism of the superconductivity in Sr$_2$RuO$_4$ based on a degenerate Hubbard model having four on-site interaction parameters; the Coulomb integral between the same orbital electrons $U$, the direct Coulomb integral between the different orbital electrons $U'$, the Hund’s coupling constant $J$ and the pair-hopping constant $J'$. The model Hamiltonian mentioned above is described as follows;

$$ H = H_0 + H_1, $$

$$ H_0 = \sum_{k,\sigma} \epsilon_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma} $$

$$ H_1 = \sum_i \left[ U \sum_{\alpha} n_{i\alpha^+} n_{i\alpha^-} + J \sum_{\alpha \neq \alpha'} S_{i\alpha} \cdot S_{i\alpha'} ight] + U' \sum_{\alpha \neq \alpha'} n_{i\alpha^+} n_{i\alpha^-} + J' \sum_{\alpha \neq \alpha'} \bar{a}_{i\alpha^+} \bar{a}_{i\alpha^+} a_{i\alpha^-} a_{i\alpha^-} $$

where $a_{k\sigma\alpha}$ is the Fourier transform of the annihilation operator for the $d_{\alpha}$-orbital electrons ($\alpha=xy, yz, zx$). $\epsilon_{k\sigma}$ describes the energy dispersions of the tight binding bands measured from the Fermi level as follows; $\epsilon_{k\sigma} = -\epsilon_0 - 2t_x \cos k_x - 2t_y \cos k_y + 4t' \cos k_x \cos k_y$. In what follows we choose the same values for the parameter sets ($\epsilon_0, t_x, t_y, t'$) as in Ref.[13], (0.50, 0.44, 0.44, -0.14), (0.24, 0.045, 0.31, 0.01), (0.24, 0.31, 0.045, 0.01) eV for $d_{xy}, d_{yz}, d_{x^2-y^2}$-orbital, respectively. These bands are occupied by four electrons per site, corresponding to a tetra-valent Ru atom. We show the results of calculation for the Fermi surfaces and the $q$-dependence of the static susceptibilities $\chi^{(0)}_{\alpha\alpha}(q)$ in Fig. 1(a) and 1(b), respectively. Each peak of $\chi^{(0)}_{\alpha\alpha}(q)$ corresponds to the nesting vector depicted in Fig. 1(a).

Following the standard prescription [13,14], we can derive the following effective interaction Hamiltonian which is rotationally invariant in the spin space;

$$ H_1^{\text{eff}} = \frac{1}{4} \sum_{\alpha, \beta} \sum_{\mu \nu \zeta \eta} V_{\alpha\beta}(k-k') a_{\alpha \mu \eta} a_{\beta \nu \zeta} a_{\beta \nu \zeta} a_{\alpha \mu \eta} $$

$$ + \frac{1}{4} \sum_{\alpha \neq \beta} \sum_{\mu \nu \zeta \eta} V_{\alpha\beta}(k-k') a_{\alpha \mu \eta} a_{\beta \nu \zeta} k' a_{\alpha \mu \eta} a_{\beta \nu \zeta} $$

$$ + \sum_{\alpha \beta} \bar{V}_{\alpha\beta}(q) S_{\alpha\beta}(q) \sigma_{\mu \eta} \cdot \sigma_{\nu \zeta} - C_{\alpha\beta}(q) \delta_{\mu \eta} \delta_{\nu \zeta} $$

$$ + \sum_{\alpha \beta} \bar{V}_{\alpha\beta}(q) S_{\alpha\beta}(q) \sigma_{\mu \eta} \cdot \sigma_{\nu \zeta} - C_{\alpha\beta}(q) \delta_{\mu \eta} \delta_{\nu \zeta} $$

where $\sigma$ is the Pauli matrix. $S_{\alpha\beta}(q)$ and $C_{\alpha\beta}(q)$ are the scattering matrices in which the particle-hole pair is scattered from the $d_{\alpha}$-orbital into the $d_{\beta}$-orbital, while $\bar{S}_{\alpha\beta}(q)$ and $\bar{C}_{\alpha\beta}(q)$ describe the scattering matrices in which $d_{\alpha}$-particle and $d_{\beta}$-hole is scattered into $d_{\beta}$-particle and $d_{\alpha}$-hole, respectively. These four scattering matrices are given as follows by treating the degenerate Hubbard model within RPA:
\[ S_{\alpha\beta}(q) = -U_{\alpha\beta}^* + \sum_\gamma (\hat{U}^\gamma \hat{\chi}_0(q))_{\alpha\gamma} S_{\gamma\beta}(q), \]
\[ C_{\alpha\beta}(q) = -U_{\alpha\beta}^* - \sum_\gamma (\hat{U}^c \hat{\chi}_0(q))_{\alpha\gamma} C_{\gamma\beta}(q), \]
\[ \tilde{S}_{\alpha\beta}(q) = -\frac{J'}{1 - (U' - J/2)\chi^{(0)}_{\alpha\beta}(q)^2 - [J'\chi^{(0)}_{\alpha\beta}(q)]^2}, \]
\[ \tilde{C}_{\alpha\beta}(q) = -\frac{J'}{1 - (U' + 3J/2)\chi^{(0)}_{\alpha\beta}(q)^2 - [J'\chi^{(0)}_{\alpha\beta}(q)]^2}. \] (3)

where
\[
\hat{U}^s = \begin{pmatrix} U & -J & -J \\ -J & U & -J \\ -J & -J & U \end{pmatrix}, \quad \hat{U}^c = \begin{pmatrix} U & 2U' & 2U' \\ 2U' & U & 2U' \\ 2U' & 2U' & U \end{pmatrix}, \\
\hat{\chi}_0(q) = \begin{pmatrix} \chi_{xy}(q) & 0 & 0 \\ 0 & \chi_{yz}(q) & 0 \\ 0 & 0 & \chi_{zx}(q) \end{pmatrix}, \\
\chi^{(0)}_{\alpha\beta}(q) = \sum_k \frac{f(\epsilon_{\alpha\beta} + \epsilon_{k+q\beta} - \epsilon_{k\alpha})}{\epsilon_{k+q\beta} - \epsilon_{k\alpha}} \chi^{(0)}_{\alpha\beta}(q), \quad \chi^{(0)}_{\alpha\beta}(q) \equiv \chi^{(0)}_{\alpha\beta}(q) \] (4)

where \( f(\epsilon) \) is the Fermi distribution function. \( S_{\alpha\alpha}(q) \) and \( C_{\alpha\alpha}(q) \) are connected with the spin and charge fluctuations for \( d_{\alpha-\text{orbital}} \), respectively.

The magnetic (charge or orbital) instability appears when the following relation is satisfied:
\[ \det(1 + \hat{U}^s(\epsilon) \hat{\chi}_0(q)) = 0 \quad \text{for} \quad S_{\alpha\beta}(q)/C_{\alpha\beta}(q). \] (5)

This equation of the instability for \( S_{\alpha\beta}(q) \) \((C_{\alpha\beta}(q))\) is reduced to the following cubic equation with respect to \( J (2U') \):
\[ x^3 + a^s x^2 + c^s = 0, \quad x = J_q (2U'_q), \]
\[ a^s = -\frac{1}{2} \sum_{\alpha} \frac{1}{\chi^{(c)}(q)}, \quad c^s = \frac{1}{2} \prod_{\alpha} \chi^{(c)}(q), \]
\[ \frac{1}{\chi^{(c)}(q)} = \frac{1}{\chi^{(0)}(q)} + U \] (6)

where \( J_q \) \((U'_q)\) is the solution of the above cubic equation for each \( q \). When \( \chi^{(c)}(q) > 0 \) as expected, it can be proved that the above cubic equation gives always three real solutions (one negative and two positive). Since we have \( J < 0 \) and \( U' > 0 \), we should assign the negative solution of the upper one of eq. (5) to \( J_q \) and the smaller one of the positive solutions of the lower one of eq. (5) to \( 2U'_q \). From this cubic equation, it is always expected that the absolute value of the solution for \( J_q \) decreases with the increase of \( U \), while the value of the solution for \( 2U'_q \) increases with \( U \). Thus, \( S_{\alpha\beta}(q) \) is enhanced with the increase of \( U \) or \(|J|\), while \( C_{\alpha\beta}(q) \) develops due to the increase of \( U' \). Also, it is expected that both instabilities of \( S_{\alpha\beta}(q) \) and \( C_{\alpha\beta}(q) \) should occur only within small regions in the \( q \)-space where \( \chi^{(0)}_{\alpha\beta}(q) \) has the peak structures. Instabilities for \( S_{\alpha\beta}(q) \) and \( C_{\alpha\beta}(q) \) are not found for realistic parameter values in the present case because of the small value of \( \chi^{(0)}_{\alpha\beta}(q) \) \((\alpha \neq \beta)\).

FIG. 1. Calculated results of (a) Fermi surfaces and (b) \( q \)-dependences of \( \chi^{(0)}_{\alpha\beta}(q) \) using the parameter set described in the text. \( q_A \) and \( q_B \) are nesting vectors.

Carrying out the mean field prescription with respect to the obtained effective Hamiltonian, we derive the gap equation for the superconducting transition with the assumption that the effect of \( \langle a_{k\alpha \sigma}^\dagger a_{k'\beta \sigma} \rangle \) for \( \alpha \neq \beta \) is negligibly small. This assumption seems to be reasonable, since the non-zero value of \( \langle a_{k\alpha \sigma}^\dagger a_{k'\beta \sigma} \rangle \) for \( \alpha \neq \beta \) can be important only at the wave vector \( k \) where two Fermi surfaces cross with each other. We thus obtain the following gap equation for the singlet \((\eta = s)\) and triplet \((\eta = t)\) spin state:
\[ \Delta^s_{\alpha\beta}(k) = \sum_{\beta' \neq \alpha} \sum_{k'} V_{\alpha\beta}^s(k - k') \frac{\tanh(\epsilon_{k'\beta}/2k_B T)}{2\epsilon_{k'\beta}} \Delta^s_{\beta\beta'}(k') \]
\[ V_{\alpha\alpha}^s(q) = c_0 S_{\alpha\alpha}(q) + \frac{1}{2} C_{\alpha\alpha}(q) \]
\[ V_{\alpha\beta}^s(q) = c_\eta S_{\alpha\beta}(q) + \frac{1}{2} C_{\alpha\beta}(q) \] \((\alpha \neq \beta)\)
\[ c_s = \frac{3}{2}, \quad c_t = -\frac{1}{2} \] (7)
where \( k_B \) is the Boltzmann constant. We note that the superconducting transition occurs simultaneously at a
temperature for all orbitals as far as we keep $J' \neq 0$ or non-vanishing values of $S_{\alpha\beta}(q)$ and $C_{\alpha\beta}(q)$, as pointed out earlier by Agterberg et al. \[7\]

![Graph](image)

**FIG. 2.** Calculated phase diagrams on $U$-$|J|$ plane (a) for $U' = U$ and (b) for $U' = 1.14U$. $|J| = J'$ is assumed. Inset: Calculated phase diagrams on $U$-$U'$ plane.

Hereafter, we restrict ourselves to the small $|J|$ region with $J' = |J|$, the relation satisfied practically in any transition metals. \[4\] We carried out the calculation at a fixed temperature $T = 0.005$eV. Although this temperature looks too high compared with the observed value in the ruthenate, it seems to be sufficient insofar as we look for the mechanism of the superconductivity within the weak coupling theory without energy cut-off which over-estimates the value of $T_c$. When a solution is found at this temperature, lower values of $T_c$ may be obtained easily by changing parameter values. In the parameter space around both instabilities for $S_{\alpha\beta}(q)$ and $C_{\alpha\beta}(q)$, we solve the gap equation for both the singlet and triplet spin states numerically, using a finite lattice with $128 \times 128$ meshes.

![Graph](image)

**FIG. 3.** Calculated $q$-dependences of (a) $S_{\alpha\alpha}(q)$ and (b) $C_{\alpha\alpha}(q)$ at a crossed point in Fig. 2(b). Orbital and charge component of $C_{\alpha\alpha}(q)$ ($\alpha = yz, zx$) are shown in the inset of Fig. 3(b).

In order to decide the phase diagram at fixed temperature, we have to find out, first of all, the instabilities of $S_{\alpha\beta}(q)$ and $C_{\alpha\beta}(q)$. The instability of $S_{\alpha\beta}(q)$ ($C_{\alpha\beta}(q)$) is given by $|J_{\text{cri}}| = \min_{|q|} |U'_{\text{cri}}| = \min_{U'} |q|$. We show the phase diagrams of this model for $U' = U$ and $U' = 1.14U$ in Fig. 2(a) and 2(b), respectively. The mode of the magnetic state in Fig. 2 is $q_A$ shown in Fig. 1. Thus, it is easily understood that the single-glet superconductivity in Fig. 2(a) has the symmetry of $d_{xy}$ due to the enhanced spin fluctuations around the antiferromagnetic mode \[12\]. On the other hand, it is seen in Fig. 2(b) that the instability line of $C_{\alpha\beta}(q)$ at $q = q_B$ gets into the paramagnetic parameter space in the small $|J|$ region. We find that both $S_{\alpha\alpha}(q)$ and $C_{\alpha\alpha}(q)$ have the peak structures at the same $q$-points as shown in Fig. 3. Thus, from the gap equation we see that the triplet pairing interaction is enhanced due to the strong and relatively wide peak structure of $C_{\alpha\alpha}(q)$ ($\alpha = yz, zx$) around $q = q_B$ while the singlet one is considerably reduced with the increase of $U'$. Thus, increasing the value of $U'$ favors the triplet superconducting state rather than the singlet one. In order to clarify what contributes to $C_{\alpha\alpha}(q)$, we define the orbital (charge) operators from $d_{yz}$ and $d_{zx}$-orbitals as $n_{iyz} \mp n_{izz}$, and
show in the inset of Fig. 3(b) the q-dependences of $C_{\alpha\alpha}(q) + C_{\beta\beta}(q) + 2C_{\alpha\beta}(q)$ ($\alpha = yz, \beta = zx$) where ‘minus’ and ‘plus’ are connected with the orbital and charge fluctuations, respectively. It is evident that the orbital fluctuation between $d_{yz}$- and $d_{zx}$-orbitals mostly contributes to the q-dependences of $C_{\alpha\alpha}(q)$ ($\alpha = yz, zx$). Also, we show in Fig. 4 the k-dependence of $\Delta^{t(y)}(k)$ where $\Delta^{t(y)}(k)$ is odd with respect to $k_\xi$. Under this condition the magnitude of $\Delta^{t(y)}(k)$ is much larger than $\Delta^{t(y)}_{xz}(k)$ and $\Delta^{t(y)}_{xy}(k)$. From this figure, it is seen that the triplet superconducting state in Fig. 2(b) belongs to the $A_{1g} \times E_u$ irreducible representation under tetragonal symmetry $D_{4h}$, because $\Delta^{t(y)}(k)$ at $k_x = 0$ has the non-zero value and the direction of the node deviates from the diagonal. We may think of the form of the gap function $\Delta^{t(y)}(k)$ proportional to $\sin k_y(A + B(\cos k_x + \cos k_y))$. We show in Fig. 4 a rough fitting result due to this function. Non-zero value of $A$ seems to be essential for better fitting. For comparison, we also show a fitting by using the function $\sin k_y(\cos k_x - \cos k_y)$ (belonging to $B_{1g} \times E_u$). Thus, summarizing these results, it is understood that the superconducting phase obtained in Fig. 2(b) is the phase of the orbital dependent triplet superconducting state which is induced by the strong orbital fluctuations between $d_{yz}$- and $d_{zx}$-orbital with the increase of $U'$. However, it remains to discuss if the value of the parameter $U'$ used in Fig. 2(b) is realistic.

It seems difficult to explain the larger value of $U'$ compared with $U$ insofar as we use the bare values for them. In actuality we should use renormalized values for these parameters in the present RPA scheme. In order to discuss the values of the renormalized interactions, we extend the multiple scattering problem [14] between two electrons to the present case. For simplicity, we assume $|J| = J' = 0$, since we are interested only in the small $|J|$ region. As usual, the renormalized interaction strengthens $U_{\alpha\alpha}, U'_{\alpha\beta}$ with the momentum $q'$ for the center of gravity of two particles are given as follows:

$$U_{\alpha}(q') = \frac{U_{\text{bare}}}{1 + U_{\text{bare}}\phi_{\alpha\alpha}(q')},$$

$$U'_{\alpha\beta}(q') = \frac{U'_{\text{bare}}}{1 + U'_{\text{bare}}\phi_{\alpha\beta}(q')} \quad (\alpha \neq \beta),$$

$$\phi_{\alpha\beta}(q') = \sum_k \frac{1 - f(\epsilon_{-k\alpha}) - f(\epsilon_{k+q'\beta})}{\epsilon_{-k\alpha} + \epsilon_{k+q'\beta}}.$$  

where $U_{\text{bare}}$ and $U'_{\text{bare}}$ are the bare interaction constants which satisfy the usually expected relation $U'_{\text{bare}} = U_{\text{bare}}$. Now, we consider the case in which a particle at $k + q_B$ and a hole at $k$ are scattered to a particle at $k' + q_B$ and a hole at $k'$ by $U_{\alpha}$ or $U'_{\alpha\beta}$. Because the value of $\phi_{\alpha\beta}(q_B)$ is principally attributed to the contribution around $k = k_B$ shown in Fig. 1(a), we have $k \approx k' \approx k_B$, that is, $q' = k + k' + q_B \approx (2\pi, 2\pi)$. It is expected that $\phi_{\alpha\beta}(q')$ is much larger than $\phi_{\alpha\beta}(q')$ ($\alpha \neq \beta$) around $q' = 0$ because of the different geometry between these two Fermi surfaces. As a result, $U_{\alpha}(q')$ is more suppressed than $U'_{\alpha\beta}(q')$ around $q' = 0$. Thus, if $U$ and $U'$ are regarded as the renormalized interactions, $U' > U$ seems to be satisfied in our case. This result seems to favor the present mechanism for the orbital instability as shown in Fig. 2(b). For more quantitative estimation, we need more detailed studies in future.

It is interesting to note that even though the calculated intensity of the spin fluctuations as shown in Fig. 3(a) has the peaks around the antiferromagnetic mode in accordance with recent inelastic neutron experimental results [15], we obtain the triplet superconducting phase rather than singlet one in a fairly plausible parameter regime. It is evident that the strong orbital fluctuations enhanced by the increase of $U'$ as compared with $U$ play the most important role in this mechanism. We expect the present mechanism to apply to other strongly correlated electron systems with degenerate orbitals and multiple Fermi surfaces. In particular, it is tempting to consider a possible application of this mechanism to U$^+_{T_3}$ which is another triplet superconductor with both the multiple Fermi surfaces and the strong antiferromagnetic correlation.

In conclusion, we have investigated the mechanism of superconductivity in Sr$_2$RuO$_4$ using the Hubbard model with degenerate orbitals within the weak coupling theory. It is shown that the triplet superconductivity appears, when the strongly correlated system approaches the orbital instability achieved by the increase of the on-site Coulomb interaction $U'$ between the electrons in different orbitals as compared with $U$ between the electrons in the same orbital. It should be stressed that this superconducting mechanism is only available in the orbitally degenerate system with multiple Fermi surfaces.

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