Triplet Pairing Superconductivity Induced by Short-Range Ferromagnetic Correlations in Sr$_2$RuO$_4$

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The microscopic origin of triplet superconductivity in Sr$_2$RuO$_4$ is discussed, paying attention to the role of Coulomb interaction, $U_{pp}$, at the O site. It is shown on the $d$-$p$ model that $U_{pp}$ induces a ferromagnetic exchange interaction between "d-electrons" (molecular orbital with $d_{xy}$-symmetry) at adjacent Ru sites, leading to short-range ferromagnetic correlations and promoting Cooper pairing with $(\sin p_x \pm i \sin p_y)$-symmetry on the $\gamma$-band. The reason why such ferromagnetic correlations work effectively may be traced back to the fact that the level of 4$d$-electrons at Ru sites is relatively low and located near that of 2$p$-electrons at O sites.

KEYWORDS: Sr$_2$RuO$_4$, superconductivity, ferromagnetic correlations, second-order perturbation

Since its discovery by Maeno et al.,

1 superconductivity in Sr$_2$RuO$_4$ has attracted much attention both experimentally and theoretically. Now it seems to have been established experimentally that the superconducting (SC) state is in the chiral spin triplet state.\textsuperscript{2,3} The existence of technically linelike nodes in the SC gap has also been suggested.\textsuperscript{4–6} Since these observations are in contrast with the expectation at the early stage of theoretical works,\textsuperscript{7,8} some phenomenological gap models have been proposed to explain those experiments.\textsuperscript{9–11} The results of ref. 9 were consistent with the observed gap with linelike nodes in [100] and [010] directions.\textsuperscript{12} On the other hand, microscopic calculations have been performed on various models and methods.\textsuperscript{13–16} The results on the multiband Hubbard model by Nomura and Yamada\textsuperscript{15} seem consistent with almost all the available measurements to date probing the gap structure. However, the Hubbard model with only on-site repulsion, $U_{dd}$, seems too stoical to discuss the superconductivity in Sr$_2$RuO$_4$ because the quasi-particles (QP) consist of 4$d$-electrons at Ru sites and 2$p$-ones at O sites. Then the intersite interaction between QP, arising through the Coulomb interaction at O sites, may not be neglected, while the direct Coulomb interaction $V$ between 4$d$-electrons at the nearest-neighbor Ru sites\textsuperscript{17} would be negligibly small. The interaction overlooked so far is

\[
\mathcal{H}_{\text{ex}} = -\frac{U_{pp}}{6} \sum_{m,i} \sum_{\alpha\beta\gamma\delta} p^\dagger_{mia} p^\dagger_{mic} p_{mid\alpha} p_{mid\beta} (\vec{\sigma}_{\alpha\beta} \cdot \vec{\sigma}_{\gamma\delta}),
\]
where $p_{m\sigma}$ is the operator for a $2p_m$-electron ($m = x, y$) of spin $\sigma$ at the $i$th site, and $\vec{\sigma}_{\alpha\beta}$'s are the Pauli matrices. Such a Coulomb interaction gives rise to an effective intersite correlation $J$ between the molecular orbital with $d_{xy}$-symmetry ("d-electron"). We expect that eq. (1) can be expressed phenomenologically as

$$\mathcal{H}_{ex} \simeq -\sum_{kk'} \sum_{q} J_q \tilde{d}_{k+q\sigma} \tilde{d}_{k'-q\sigma} \tilde{d}_{k'\delta} \tilde{d}_{k\gamma} (\vec{\sigma}_{\alpha\beta} \cdot \vec{\sigma}_{\gamma\delta}),$$

(2)

where $\tilde{d}$ denotes the operator for a "d-electron" and $J_q = 2J(\cos q_x + \cos q_y)$. Although exchange interaction (2) is a too simplified version, we derive a more realistic and complicated version later starting with the $d$-$p$ model and show that this kind of intersite interaction promotes short-range ferromagnetic correlations (SRFMCs), which have been measured quite recently by inelastic neutron scattering.\(^{18}\)

The purpose of this Letter is to demonstrate that SRFMC promotes triplet pairing with $(\sin p_x \pm i \sin p_y)$-symmetry, as proposed in ref. 9. We follow the weak-coupling calculations with the pairing interaction given by the second-order perturbation theory (SOPT) with respect to $U_{dd}$ and $U_{pp}$.

Let us consider the situation drawn in Fig. 1, where a $2p_x$- or $2p_y$-orbital is sandwiched between two $d$-orbitals with $d_{xy}$-symmetry. Hereafter, we restrict our discussion within the $\gamma$-band since it is expected to provide the main contribution.\(^{15}\) The noninteracting $d$-$p$ Hamiltonian, which can well describe the situation shown in Fig. 1, is given as follows:

$$\mathcal{H}_{dp}^{(0)} = \sum_{k,\sigma} \begin{pmatrix} d_{k\sigma}^\dagger & p_{xk\sigma}^\dagger & p_{yk\sigma}^\dagger \end{pmatrix} \begin{pmatrix} \varepsilon_d & V_{yk}^* & V_{zk}^* \\ V_{yk} & \varepsilon_p & W_k \\ V_{zk} & W_k & \varepsilon_p \end{pmatrix} \begin{pmatrix} d_{k\sigma} \\ p_{xk\sigma} \\ p_{yk\sigma} \end{pmatrix} \equiv \sum_{k,\sigma} \phi_{k\sigma}^\dagger \mathcal{H}_{kk'\sigma}^{(0)} \phi_{k'\sigma},$$

(3)

where $d_{k\sigma}$ and $p_{m\kappa\sigma}$ are the operators for $4d$- and $2p_m$-electrons of momentum $k$ and spin.

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Fig. 1. Schematic figure of RuO$_2$ plane. $t_{dp}$ and $t_{pp}$ are hopping integrals. Gray domains circled by dashed squares represent the molecular orbitals, which form a two-dimensional square lattice.
regarded as the using
Hereafter, we denote can perform the Fourier transformation of eq. (7), leading to
\[ J. \text{Phys. Soc. Jpn.} \]
\[ \sigma \]
\[ \vec{\phi} \]
\[ \text{Here, } \sigma \text{ is related to } \vec{\phi} \text{ by the relation } \sigma_{k\sigma} = \hat{U}_k^{-1} \vec{\phi}_{k\sigma}, \text{ where the matrix } \hat{U}_k \text{ is defined as } \hat{U}_k = (\bar{a}_{1k} \bar{a}_{2k} \bar{a}_{3k}). \]
Substituting the relations between \( \vec{\phi}_{k\sigma} \) and \( \vec{\psi}_{k\sigma} \) into eq. (1), we obtain, as shown below, the effective interaction between QP described by \( a_{1k\sigma} \). For this purpose, it is enough to confine our discussions within the case \( t_{pp} = 0 \); namely we need not consider the hopping \( t_{pp} \). Because electrons located on both sides of a certain O site can interact with each other effectively only through the O site, the term including \( t_{pp} \) corresponds to the next nearest-neighbor interaction (higher-order contribution). Then, hereafter, we can use simplified relations such as
\[ d_{k\sigma} = \frac{\lambda_{1k}}{L_{1k}} a_{1k\sigma} + \frac{\lambda_{2k}}{L_{2k}} a_{2k\sigma} + \frac{\lambda_{3k}}{L_{3k}} a_{3k\sigma}, \]
\[ p_{m\kappa\sigma} = \frac{V_{m\kappa}}{L_{1k}} a_{1k\sigma} + \frac{V_{m\kappa}}{L_{2k}} a_{2k\sigma} + \frac{V_{m\kappa}}{L_{3k}} a_{3k\sigma}, \]
where \( L_{1k} = (|V_{1k}|^2 + |V_{2k}|^2 + \lambda_{k}^2)^{1/2} \) \((i = 1, 2, 3)\). Moreover, it is sufficient to consider only the QP on the orbital \( i = 1 \), because the other bands are completely filled with electrons. Hereafter, we denote \( a_{1k\sigma} \) and \( L_{1k} \) simply as \( a_{k\sigma} \) and \( L_{k} \), respectively. The operator \( a_{k\sigma} \) is regarded as the \( \bar{d}_{k\sigma} \) in eq. (2).

Now we introduce the interaction terms. Using relations (5) and (6), we express such terms using \( a_{k\sigma} \)'s. Substituting relation (6), into the Fourier transformed form of eq. (1), we obtain
\[ H_{ex}^{(x)} = -\frac{U_{pp}}{6} \sum_{kk'q} \sum_{\alpha\beta\gamma\delta} \frac{V_{k+q-k'q} V_{k+k'} V_{k'}}{L_{k+q} L_{k'-q} L_{k'}} a_{k+q} a_{k'} a_{k'} a_{k'+q} a_{k+q} a_{k'} a_{k'+q} \sigma_{\alpha\beta} \sigma_{\gamma\delta}. \]
At first sight, it seems that this interaction Hamiltonian, eq. (7), has a complicated form in real space. However, if we neglect the \( k \)-dependence of \( L_{k} \)'s where \( L_k \equiv \Delta \sim O(\varepsilon_d - \varepsilon_p) \), we can perform the Fourier transformation of eq. (7), leading to
\[ H_{ex} = -\frac{U_{pp}^{(4)}}{3\Delta^4} \sum_{(i,j)} \sum_{\alpha\beta\gamma\delta} \frac{1}{(i,j)} (i, i, i) + (i, j, j)
+ (i, j, j) + (i, i, j) - (i, i, j) - (i, i, j) \]
- (j, i, i) - (i, j, i) \] \( \sigma_{\alpha\beta} \sigma_{\gamma\delta} \),
where we have added the term arising from the exchange process along the \( y \)-direction, the sum \( \sum_{(i,j)} \) is taken within the nearest-neighbor sites, and the abbreviation \((i, j, k, l)\) represents the term \( a_{1\alpha} a_{1\gamma} a_{k\delta} a_{l\beta} \). Eventually, it is revealed that eq. (7) contains all the types of interaction.
between two electrons at nearest-neighbor Ru sites. Performing the Fourier transformation again and executing straightforward calculations, we can rewrite eq. (8) into the following form:

$$H_{ex} = -\frac{1}{4} \sum_{kk'} \sum_{q} J_{k,k';q} a_{k}^\dagger \sigma_{\alpha\beta} q a_{k'}^\dagger \sigma_{\gamma\delta} q a_{k\beta} \delta_{\alpha\beta} \delta_{\gamma\delta},$$

(9)

$$J_{k,k';q} = \frac{2U_{pp}t_{dp}^4}{L_{k+q} L_{k-q} L_{k'} L_k} \sum_{m} \left\{ \frac{1}{2} + \cos q_m - \cos k_m - \cos k'_m + \frac{1}{2} \cos(k_m + k'_m) \right\}.$$  

(10)

In eq. (10), the $k$-dependence of $L_k$’s is recovered again. The second term in the curly brackets of eq. (10), $\cos q_m$, corresponds to the simplified version of eq. (2). There are many terms which are not included in the naive estimation, eq. (2). Therefore, the coupling constant $J$ cannot be written simply as $J_q$, but has a complicated momentum dependence $J_{k,k';q}$ as eq. (10).

The term $\delta_{\alpha\beta} \delta_{\gamma\delta}$ in eq. (9), which appears automatically through the procedure above, assures that, due to the Pauli principle, there is no interaction between electrons with the parallel spin direction originally on the same O site. For interaction (9) to play an important role, the ratio $t_{dp}/(\varepsilon_d - \varepsilon_p) \equiv t_{dp}/\Delta$ should not be too small (namely, $\Delta/t_{dp}$ should not be too large).

Such a condition is expected to be fulfilled in Ru-oxides because the level of 4$d$-electrons in Ru is deeper than that of 3$d$-electrons in Fe or of 5$d$-electrons in Os in general, and the level is located near that of 2$p$-electrons at the O site. This implies that there exists a strong hybridization between Ru and O. Indeed, as shown in Fig. 2, the partial density of states (DOS) for 4$d_{xy}$, 2$p_x$ and 2$p_y$ obtained by band structure calculations are reproduced by taking $\Delta/t_{dp} \approx 0.15$ with the use of eqs. (5) and (6); namely, $\rho_{\gamma} \approx 2.0 \rho_O$ ($\rho_{\gamma} \approx 0.5 \rho_{Ru}$) where $\rho_{\gamma}$, $\rho_{Ru}$ and $\rho_{O}$ denote the partial DOS per atom (at the Fermi level) of Ru (\gamma-band), Ru (total of \alpha-, \beta-, and \gamma-bands) and O (in-plane), respectively. The same procedure is applicable to the on-site Coulomb repulsion $U_{dd}$ between $d$-electrons, $U_{dd} \sum_i d_{i \uparrow}^\dagger d_{i \uparrow} d_{i \downarrow}^\dagger d_{i \downarrow}$. Along the way
parallel to obtaining exchange interaction (7) from the original interaction between p-electrons, eq. (1), the Hubbard interaction is expressed in terms of the \( a_{k\sigma} \) operators describing the QP band. Then, the total Hamiltonian is given by

\[
H = \sum_{k,\sigma} (\lambda_k - \mu) a_{k\sigma}^\dagger a_{k\sigma} + \sum_{kk'q} U_{k,k';q} a_{k+q\uparrow}^\dagger a_{k'\uparrow}^\dagger a_{k'\downarrow} a_{k\downarrow} + \mathcal{H}_{\text{ex}} ,
\]

(11)

where \( \mu \) denotes the chemical potential. It is noted that the repulsion \( U_{dd} \) in eq. (12) should be regarded as an effective interaction between QP, that is reduced by the electron correlation leaving a relatively heavy QP. We set \( t_{pp} = -0.4t_{dp} \) to reproduce the Fermi surface of the \( \gamma \)-band given by the band structure calculations. Then the bandwidth of the \( \gamma \)-band is \( W \sim 2.2t_{dp} \). The last two terms of eq. (11) can be rewritten into a more convenient form to perform the perturbation expansion as follows:

\[
\mathcal{H}_{\text{int}} = \sum_{kk'q} \tilde{J}_{k,k';q} a_{k+q\uparrow}^\dagger a_{k'\uparrow}^\dagger a_{k'\downarrow} a_{k\downarrow},
\]

(13)

\[
\tilde{J}_{k,k';q} = U_{k,k';q} + J_{k,k';q} + J_{k',k,k-k'+q},
\]

(14)

Hereafter, \( t_{dp} \) is adopted as a unit of energy.

The static spin susceptibility \( \chi_{\perp}(k,0) \) including vertex correction, up to the first-order perturbation, is given by

\[
\chi_{\perp}(q, i\omega) = -\frac{T}{\epsilon_F} \sum_{p, p'} \left\{ 1 - T \sum_{\epsilon, \epsilon'} \tilde{J}_{p+q, p'; -p-p'} G(p' + q, \epsilon' + i\omega) G(p', i\epsilon) \right\} G(p + q, i\epsilon + i\omega) G(p, i\epsilon),
\]

(15)

whose diagrammatic expression is shown in Fig. 3(b). It is enough to calculate \( \chi_{\perp} \) because the SU(2) symmetry is preserved. The results of numerical calculations are shown in Fig. 4.

\[
\chi_{\perp}(q, 0)
\]

(16)

Fig. 3. (a) Vertex \( \tilde{J}_{k,k';q} \). (b) Spin susceptibility \( \chi_{\perp}(q, i\omega) \).
which gives rise to a practical SRFMC. The effect of $U_{dd}$ is to enhance $\chi_{\perp}(\mathbf{q},0)$ in the entire $\mathbf{q}$-space as seen in Fig. 4. On the other hand, the peak around $\mathbf{q} = (0,0)$ is much more enhanced by $U_{pp}$ than by $U_{dd}$, while there is no extra increase by $U_{pp}$ at $(\pi, \pi)$. The qualitative behavior of $\chi_{\perp}(\mathbf{q},0)$ remains unchanged if we neglect the $\mathbf{k}$-dependence of $L_k$’s in eq. (10) or (12). Then, we can draw the picture that SRFMC is induced by the effect of on-site repulsion between $p$-electrons at the O site. The broad peak structure at $\mathbf{q} = (0,0)$ is consistent with

![Fig. 4. Momentum dependence of static spin susceptibility $\chi_{\perp}(\mathbf{q},0)$ for several values of $U_{dd}$ and $U_{pp}$. The dashed lines that have no parameter values correspond to, from the lower line, $(U_{dd}, U_{pp}) = (1,0), (2,0)$ and so on, respectively. Namely, we first applied $U_{dd}$, then applied $U_{pp}$.](image)

the SRFMC that has been measured quite recently by inelastic neutron scattering,\textsuperscript{18} and is responsible for the occurrence of triplet superconductivity in Sr$_2$RuO$_4$ as discussed below. As for the results of different electron fillings, the more electrons are doped, the more the peak structure around $\mathbf{q} = (0,0)$ is enhanced.

Diagrams for the irreducible pairing vertices up to SOPT in terms of $\tilde{J}$ are shown in Fig. 5. Its analytic expressions are given as follows:

![Fig. 5. Pairing interaction vertices $\Gamma^{\uparrow\uparrow}$ and $\Gamma^{\uparrow\downarrow}$ up to the second-order perturbation with respect to $\tilde{J}$. The solid line and the square denote the noninteracting Matsubara Green function $\mathcal{G}$ and the intersite coupling vertex $\tilde{J}$, respectively.](image)

$$\Gamma_{\mathbf{k},\mathbf{k}'}^{\uparrow\uparrow} = T \sum_{\epsilon, \mathbf{p}} \tilde{J}_{\mathbf{k},\mathbf{p}+\mathbf{k}-\mathbf{k}'} \tilde{J}_{\mathbf{p}+\mathbf{k}-\mathbf{k}'} \mathcal{G}(\mathbf{p}, i\epsilon) \mathcal{G}(\mathbf{p} + \mathbf{k} - \mathbf{k}', i\epsilon),$$

(16)
The pairing interaction in the triplet manifold, $V^t_{kk'}$, is given by eq. (16) directly, and that in the singlet manifold, $V^s_{kk'}$, is given by

$$V^s_{kk'} = \sum_{\xi, p} \tilde{J}_{k,p} \tilde{J}_{k',p} \tilde{G}(\xi, p) \tilde{G}(\xi + k, p + k').$$  \hspace{1cm} (18)

Note that SU(2) symmetry is preserved in the triplet manifold. Namely, the triplet component stemming from eq. (17) is the same as the one from eq. (16).

At $T = T_c$, we can use the linearized gap equation, in the weak-coupling formalism, given by

$$\Delta_k = - \sum_{k'} V^s_{kk'} \frac{k'}{\xi k'} \tanh \left( \frac{\xi k'}{2T_c} \right).$$  \hspace{1cm} (19)

A proper way of obtaining the gap structure with the highest $T_c$ is to solve eq. (19) as an eigenvalue problem without specifying the type of the gap $\Delta_k$. However, for simplicity, we seek the variational solution for eq. (19). Namely, we calculate $T_c$ by specifying the type of gap function, such as $\sqrt{2} \sin k_x (p_x$-pairing), $\sqrt{2} \sin (k_x + k_y$) ($p_{x+y}$-pairing) for triplet pairing, and $2 \sin k_x \sin k_y$ ($d_{xy}$-pairing), $\cos k_x - \cos k_y$ ($d_{x^2-y^2}$-pairing) for singlet pairing. However, it is rational to consider that the type of pairing giving the highest $T_c$ is dominant in the true gap function. The pairing interaction $V^t_{kk'}$, eq. (16), and $V^s_{kk'}$, eq. (18) is estimated at $T = 0.02$, because their values are not sensitive at temperatures $T < 0.02$. We adopt the first Brillouin zone divided into 45×45 k-meshes. The results for a series of parameters are shown in Fig. 6. We can see in Fig. 6 that $T_c$ is enhanced as $U_{pp}$ is applied. For a system with a fillings of $n = 1.33$, a small value of $\Delta$ (splitting between $p$- and $d$-levels), and a moderate value of $U_{pp}$, corresponding to Sr$_2$RuO$_4$, $p_x$-pairing would be realized as shown in Fig. 6(a). When the system is located away from the half-filling state, it is also enhanced as shown in Fig. 6(b). On the other hand, $p_{x+y}$-pairing has $T_c$ less than $10^{-3}$. In ref. 17, it is discussed that the next nearest-neighbor (diagonal) pairings, $d_{xy}$ or $p_{x+y}$-pairing, are promoted due to the nearest-neighbor Coulomb repulsion $V$. In our case, on the contrary, the nearest-neighbor pairing, $p_x$-pairing, is realized through SRFMC of $\mathcal{H}_{ex}$. As for the other symmetries, $d_{x^2-y^2}$ and $d_{xy}$, we cannot obtain a finite $T_c$ in the parameter region shown in Fig. 6. At first sight, it appears unrealistic that $U_{pp}$ is larger than $U_{dd} \sim W$, because $3d$-electrons are much more localized on the ion site than $2p$-electrons. However, it is crucial to note that the Coulomb interactions here are effective ones that are greatly renormalized. Indeed, the effect of $U_{pp}$ on QP cannot be screened by avoiding $U_{pp}$ because QP consisting of molecular orbitals at the nearest-neighbor sites cannot get rid of $U_{pp}$. On the other hand, the effective interaction $U_{dd}$ can be reduced by correlations and by making a heavy QP. Therefore the values adopted for
$U_{dd}$ and $U_{pp}$ are considered to be not unrealistic.

In conclusion, we have derived from the so-called $d$-$p$ model the effective intersite exchange interaction $H_{ex}$, whose origin is the on-site Coulomb interaction at an O site. SRFMC were induced by this exchange interaction. Then, we have shown that the triplet SC state of $\sin p_x \pm i \sin p_y)$-symmetry is promoted as applying $U_{pp}$ within SOPT. These results simulate quite well the properties observed in Sr$_2$RuO$_4$. While our results are derived on SOPT for the pairing interaction, they might make sense at least in describing the qualitative behavior. It is possible to take into account the effect of $\alpha$- and $\beta$-bands on the same footing as the $\gamma$-band since the exchange interaction arising from the same mechanism also works between QPs in these bands. According to preliminary calculations, the pairing with $[\sin(p_x + p_y) + i \sin(p_x - p_y)]$-symmetry could be promoted, which is consistent with a line-node-like gap along the diagonal direction $k_x = \pm k_y$.\textsuperscript{12,15}

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