Model for Unconventional Superconductivity of Sr_2RuO_4: Effect of Impurity Scattering on Time-Reversal Breaking Triplet Pairing with a Tiny Gap

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A model for unconventional superconductivity of Sr_2RuO_4 is presented to resolve its puzzle. It is shown that the short-range ferromagnetic spin fluctuations give rise to the triplet pairing with p-like symmetry which is breaking the time-reversal symmetry and has a tiny gap due to the salient shape of the Fermi surface characteristic to Sr_2RuO_4. The effect of nonmagnetic-impurity scattering in the unitarity limit is shown to fill up easily the tiny gap giving rise to an appreciable residual density of states, which explains consistently the puzzling properties observed so far.

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The superconductivity of Sr_2RuO_4 has attracted much attention since its discovery [1], not only because it has the same structure as $La_{2-x}Sr_xCuO_4$ of high-T_c cuprates but also because the data are strongly suggestive of a triplet order parameter breaking the time-reversal symmetry [2,3]. A puzzle about its superconducting state is that the possible states allowed by the group theoretical argument have a finite gap over the Fermi surface [4], while the experimental data on the specific heat indicates that there remains the residual density of states (DOS) of about half of N_F, DOS in the normal state [5], and those on ^{101}Ru-NQR (and NMR) relaxation rate $1/T_1$ suggest the gap vanishes on the line of the Fermi surface [6]. A similar trend has also been observed in thermal conductivity [7].

In order to explain the former property, the nonunitary state in the triplet manifold has been proposed on the basis of group theoretical arguments [8,9]. However, it cannot explain the absence of any trace of the Hebel-Slichter peak near the transition temperature T_c [6]. Furthermore, it is not so evident why the nonunitary state is stabilized. It needs in general to take into account the explicit character of Sr_2RuO_4 which would have been overlooked by the group theoretical arguments. For example, it has been proposed that the superconducting gaps on the branch α and β of the Fermi surfaces are much smaller than that belonging to the main branch γ due to the band symmetry of quasiparticles [10,11]. However, it also encounters the difficulty of $1/T_1$ mentioned above [6].

A purpose of this Letter is to present a model to resolve the above mentioned puzzle on the phenomenology relying on the experimental observations. According to the analysis of NMR experiments [6,12], the short-range ferromagnetic spin fluctuations are developed in the RuO_2 plane [13]. Therefore, it is natural to assume the short-range pairing interaction works between quasiparticles with parallel spins located at the nearest neighbor sites. Namely, the interaction matrix element $V_{k,k'}$ is assumed to be in the form

$$V^{\alpha\beta,\gamma\delta}_{\mathbf{k},\mathbf{k}'} = -2V_0[\cos(k_x - k'_x) + \cos(k_y - k'_y)] \times (\vec{\sigma}_{\alpha\delta} \cdot \vec{\sigma}_{\beta\gamma}), \quad (1)$$

where we have assumed the 2D square lattice with unit lattice constant and $\vec{\sigma}$ is the vector formed by the Pauli matrices [14]. It is straightforward to take into account the effect of interaction between the second neighbors, the third neighbors, and so on [15]. Then, in the triplet manifold, the pairing interaction (1) is reduced to

$$V^{\mathrm{triplet}}_{\mathbf{k},\mathbf{k}'} = -2V_0(\sin k_x \sin k'_x + \sin k_y \sin k'_y). \quad (2)$$

It is difficult to determine, from purely theoretical considerations, the relative stability among the degenerate superconducting states in the triplet manifold. Namely, the relative importance of the spin-orbit coupling or the dipole interaction and the spin-fluctuation feedback effect, which lifts its degeneracy [16,17], is hard to estimate on the microscopic level. Then, we follow the experimental fact that the time-reversal breaking state seems to be realized [3]. Combining this fact and the type of pairing interaction (2), the $\mathbf{d}$ vector is identified as that of the Γ_5^- state classified in Refs. [4,17],

$$\mathbf{d}_{\mathbf{k}} = \hat{\mathbf{z}}\Delta_0[\sin k_x \pm i \sin k_y], \quad (3)$$

where we have used the fact that the xy plane is the easy plane of spin polarization [12]. This state satisfies the requirement of D_{4h} symmetry, which gives rise to a crucial difference from the state Γ_5^- discussed in Refs. [4,17] which is isotropic in the xy plane of k space. The amplitude of the gap, $|\mathbf{d}_{\mathbf{k}}|$, vanishes only if the following condition is satisfied:

$$\sin k_x = \sin k_y = 0, \quad (4)$$

which means $\mathbf{k} = (0,0)$, $(0, \pm\pi)$, $(\pm\pi, 0)$, and $(\pm\pi, \pm\pi)$. According to the band structure calculations [18], the Fermi surface of the main branch, γ branch, is close to circular and passes through very near those points, $(0, \pm\pi)$ and $(\pm\pi, 0)$. Therefore, the gap on the Fermi surface is extremely anisotropic leaving a tiny gap around those points. If we approximate the Fermi surface of the γ branch by $\mathbf{k}_{\mathrm{F}} = (\pi R \cos\theta_k, \pi R \sin\theta_k)$, where R parametrizes the diameter of the Fermi circle, and θ_k is the angle measured form k_x axis in the k space, the amplitude of the gap on the Fermi surface is given by

$$|\mathbf{d_k}| = \Delta_0[\sin^2(\pi R \cos\theta_k) + \sin^2(\pi R \sin\theta_k)]^{1/2}, \quad (5)$$

which exhibits rather anisotropic behavior for a realistic value of $R = 0.9$ [18] as shown by the solid line in Fig. 1. The dashed line in Fig. 1 is for

$$|\mathbf{d_k}| = \Delta_0[1 - r\cos(4\theta_k)] \quad (6)$$

with $r = 0.692$. The gap (6) is the simplest model gap with fourfold symmetry and has the minimum gap the same as (5) with $R = 0.9$.

Since the superconducting coherence length ξ_0 is estimated at about 1000 Å which is far larger than the mean distance of electrons, the weak-coupling approach may be valid so that the superconducting gap is formed by the quasiparticles located near the Fermi level. We further introduce a model pairing interaction reproducing the expected gap $|\mathbf{d_k}|$,

$$\mathcal{V}_{\mathbf{k},\mathbf{k}'} = -Vf(\theta_k)f(\theta_{k'}) \times \vartheta(\omega_c - |\xi_k|)\vartheta(\omega_c - |\xi_{k'}|), \quad (7)$$

where the usual notations are used, and the basis function $f(\theta_k)$ is defined as

$$f(\theta_k) \equiv \frac{[\sin^2(\pi R \cos\theta_k) + \sin^2(\pi R \sin\theta_k)]^{1/2}}{[1 - J_0(2\pi R)]^{1/2}} \times \mathrm{sgn}(\sin\theta_k), \quad (8)$$

where $J_0(x)$ is the Bessel function of zeroth order. The function $f(\theta_k)$ is normalized as $\int_0^{2\pi}(d\theta/2\pi)|f(\theta)|^2 = 1$.

The gap $\Delta_\mathbf{k}$, obtained from (7), takes the form

$$\Delta_\mathbf{k} = \Delta f(\theta_k), \quad (9)$$

where Δ is determined self-consistently by solving the gap equation as usual. Then, $N_s(E)$, the DOS in the superconducting state, is given by

$$\frac{N_s(E)}{N_F} = \mathrm{Re}\left\langle \frac{E}{\sqrt{E^2 - |\Delta_\mathbf{k}|^2}} \right\rangle_{FS}, \quad (10)$$

where N_F is the DOS at the Fermi level, which is assumed to be uniform over the Fermi surface, and $\langle\cdots\rangle_{FS}$ indicates that the average over the Fermi surface is taken. The result for $N_s(E)/N_F$ is shown by the solid line in Fig. 2. It is remarked that a tiny gap Δ_{min} ($\simeq 0.3 \times \Delta \ll \Delta_{max} \simeq 1.3 \times \Delta$) is opened and $N_s(E)/N_F \propto E/\Delta_{max}$ for $\Delta_{min} < E \ll \Delta$, which is similar to that of a polarlike state, while the DOS show a sharp peak at $E = \Delta_{max}$ which is similar to that of the axial-like state. The specific heat jump at T_c is given as [16]

$$\frac{\Delta C}{C_N} = \frac{1}{\kappa}\left(\frac{\Delta C}{C_N}\right)_{BCS}, \quad (11)$$

where $\kappa \equiv \int_0^{2\pi}(d\theta/2\pi)|f(\theta)|^4$ is calculated as

$$\kappa = \frac{5 + J_0(4\pi R) - 8J_0(2\pi R) + 2J_0(2\sqrt{2}\pi R)}{4[1 - J_0(2\pi R)]^2}. \quad (12)$$

For $R = 0.9$, $\kappa \simeq 1.336$ giving $\Delta C/C_N \simeq 1.07$. The above results contradict with the specific heat measurements and T dependence of $1/T_1$ at $T \ll T_c$, which show that there exist appreciable DOS at zero energy, $N_s(0) \sim 1/2 \cdot N_F$.

A key to solve this discrepancy may be taking into account the effect of impurity scattering, because ξ_0 is large of the order of 10^3 Å so that a tiny amount of impurities, even in the sample where the de Haas–van Alphen oscillations are detected [19], can give rise to the residual DOS if they are the scattering center of the

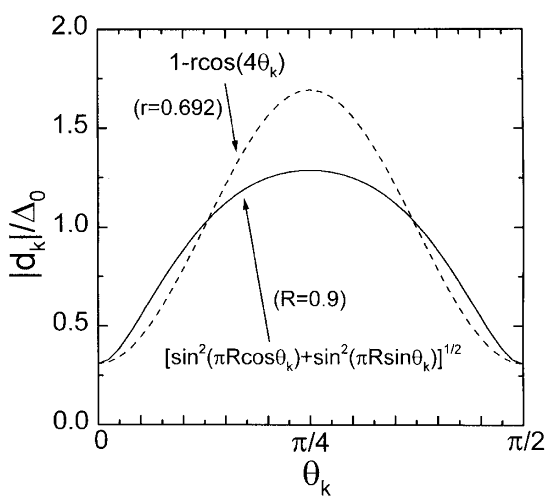

FIG. 1. Magnitude of superconducting gap $|\mathbf{d_k}|/\Delta_0$ as a function of $\theta_\mathbf{k}$, the angle on the Fermi surface. The solid line represents (5) for the parameter $R = 0.9$. The dashed line is for (6) with $r = 0.692$ which has the minimum gap the same as (5) with $R = 0.9$.

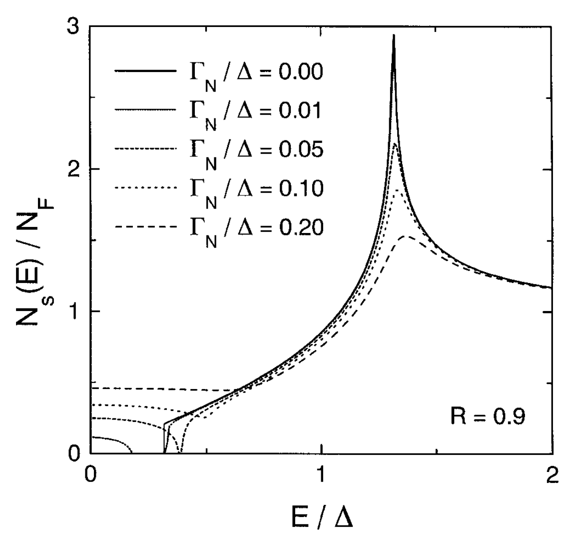

FIG. 2. Density of states, $N_s(E)/N_F$, for the pure system and that under the effect of impurity scattering in the unitarity limit. The degrees of impurity scattering are parametrized by Γ_N/Δ, where Γ_N is the scattering rate in the normal state and Δ is the gap parameter, defined by (9), at $0 \le T \le T_c$.

unitarity limit. The fact that the amount of the residual DOS is dependent on the sample quality and correlated with the variation of T_c is consistent with this conjecture [20,21]. Although this problem has been discussed in great detail in the other contexts [22–26], it is an open question whether the residual DOS appears easily even if the tiny gap exists in a pure system. We solve this problem following the above formalism [22,23].

The normal Green function $\bar{G}(\omega) \equiv \sum_{\mathbf{k}} G(\mathbf{k}, \omega)/N$, averaged over the k space, is given as

$$\bar{G}(\omega) = -i\pi N_{\mathrm{F}} \left\langle \frac{\tilde{\omega}}{\sqrt{\tilde{\omega}^2 - |\Delta_{\mathbf{k}}|^2}} \right\rangle_{\mathrm{FS}}, \tag{13}$$

where $\tilde{\omega}$ satisfies the self-consistent equation

$$\tilde{\omega} = \omega + \frac{i\Gamma_N}{\left\langle \frac{\tilde{\omega}}{\sqrt{\tilde{\omega}^2 - |\Delta_{\mathbf{k}}|^2}} \right\rangle_{\mathrm{FS}}}. \tag{14}$$

Here Γ_N is the scattering rate of quasiparticles in the normal state due to impurities in the unitarity limit. Then the DOS is given by the formula

$$N_{\mathrm{s}}(E) = -\frac{1}{\pi} \mathrm{Im}\bar{G}(E + i0^+). \tag{15}$$

Using the numerical solution of (14), the DOS (15) is calculated by means of (13). The results for a series of values of Γ_N are shown in Fig. 2. It is noted that the residual DOS appears even for the scattering rate which has little effect on the reduction of T_c. For the case $\Gamma_N/\Delta > 0.1$, the overall shape of $N_{\mathrm{s}}(E)$ is similar to that for the polar state with a small amount of impurities of the unitarity limit. Therefore, the T dependence of the physical quantities at low temperatures, $T \ll T_c$, is expected to look like that of the polar state with impurities.

The reduction of T_c due to impurity scattering is determined by the conventional Abrikosov-Gor'kov formula,

$$\ln\left(\frac{T_c}{T_{c0}}\right) = \psi\left(\frac{1}{2}\right) - \psi\left(\frac{1}{2} + \eta_c\right), \tag{16}$$

where T_{c0} is the transition temperature of the pure system, $\eta_c \equiv \Gamma_N/2\pi T_c$ is the pair breaking parameter, and $\psi(x)$ is the digamma function. By eliminating the explicit dependence of Γ_N from $N_{\mathrm{s}}(0)$, (15), and T_c, (16), we obtain a direct relation between T_c and $N_{\mathrm{s}}(0)$ which is shown in Fig. 3a.

The specific heat jump at T_c is given by

$$\frac{\Delta C}{C_N} = \frac{24[1 - \eta_c \psi^{(1)}(\eta_c + \frac{1}{2})]^2}{[\frac{1}{3}\eta_c \psi^{(3)}(\eta_c + \frac{1}{2}) - \kappa \psi^{(2)}(\eta_c + \frac{1}{2})]}, \tag{17}$$

where $\psi^{(n)}(x)$ is the nth derivative of the digamma function $\psi(x)$. The relation $\Delta C/C_N$ vs $N_{\mathrm{s}}(0)/N_{\mathrm{F}}$ is obtained after eliminating the explicit dependence of η_{c} and is shown in Fig. 3b.

For comparison with experiments, we also plot the experimental data points from Refs. [5] and [20] in Fig. 3.

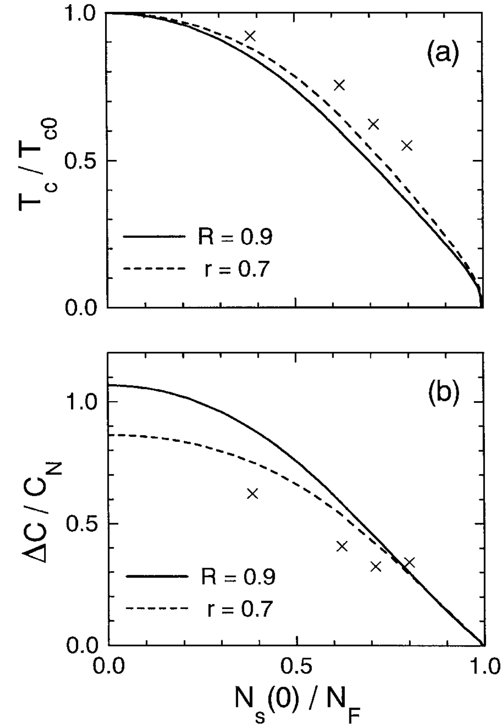

FIG. 3. (a) Transition temperature T_c and (b) specific heat jump $\Delta C/C_N$ vs residual density of states $N_{\mathrm{s}}(0)/N_{\mathrm{F}}$ under the effect of impurity scattering in the unitarity limit. Crosses are the experimental data points from Refs. [5] and [20]. Dashed lines are for the model pair function (18) with $r = 0.7$.

Although the quantitative agreement with the theory is not very good, the qualitative agreement is rather nice considering the crudeness of our model. The present model would offer at least a good starting point for understanding the heart of the superconducting state of Sr_2RuO_4. On the basis of the present model, we can revise the theory by taking into account a more realistic nature of Sr_2RuO_4, for instance, the effect of pairing interaction between second and third neighbor sites, the effect of other branches of band, nonuniformity of the DOS around the Fermi level, and so on.

For instance, the results shown by dashed lines in Fig. 3 are in better agreement with the experiments. These have been obtained by using the model pairing (7) with

$$f(\theta_k) \equiv \frac{[1 - r\cos(4\theta_k)]}{\sqrt{1 + r^2/2}}, \tag{18}$$

with $r = 0.7$, which gives approximately the same gap (6) with $r = 0.692$. The model gap (6) can be regarded as that taking into account the effect of second and third neighbor interaction [15]. It is because $\cos k_x = \cos k_y < 0$ at the Fermi surface corresponding to $\theta_k = \pi/4$, and Δ_1 and Δ_2 are in the sign opposite to Δ_0 (if both V_1 and V_2 are positive as expected) leading to a result that $[\Delta_0 + \Delta_1 \cos k_x + \Delta_2 \cos k_y]$ at $\theta_k = \pi/4$ is larger than Δ_0 and that at $\theta_k = 0$ (or $\pi/2$) is smaller than Δ_0.

The effect of branches other than the γ branch would certainly change a result quantitatively. In our model, the gap given by (3) is induced in general also on the $\{\alpha, \beta\}$ branches as long as the pair-hopping interaction remains no matter how small it is. The results for quantities concerning the transition point, such as T_c and ΔC, remain essentially the same as above if the magnitude of the induced gaps on $\{\alpha, \beta\}$ branches is small enough. Since C_N is increased by the contribution from the $\{\alpha, \beta\}$ branches, $\Delta C/C_N$ tends to decrease improving the discrepancy between the theory and experiments shown in Fig. 3b, if $N_s(0)$ would have remained unchanged. If the quasi-1D nature of the $\{\alpha, \beta\}$ branches is assumed [18], $\Delta^\alpha_{\max} \simeq \sqrt{3/2}\,\Delta_\alpha$ and $\Delta^\alpha_{\min} \simeq \sqrt{3}/2\Delta_\alpha$, and $\Delta^\beta_{\max} \simeq \sqrt{7}/2\Delta_\beta$ and $\Delta^\beta_{\min} \simeq \sqrt{3}/2\Delta_\beta$, giving rise to rather large anisotropy $\Delta^\alpha_{\max}/\Delta^\alpha_{\min} = \sqrt{2}$ and $\Delta^\beta_{\max}/\Delta^\beta_{\min} = \sqrt{7/3}$, respectively. This would cause some small structure in $N_s(E)$ at $E \sim \Delta_\alpha$ and Δ_β which are more easily smoothed, compared to the γ branch, by the impurity scattering giving rise to an additional contribution to the residual DOS $N_s(0)$. Such an effect tends to decrease the discrepancy between the theory and experiments shown in Fig. 3a, while the above mentioned improvement of Fig. 3b may be lessened to some extent. It is left for future study to estimate those values quantitatively on the basis of a model with a more microscopic base.

In conclusion, we have shown that the short-range ferromagnetic spin fluctuations induce a novel type of 2D triplet superconducting state in Sr_2RuO_4 with the help of the special nature of the Fermi surface characteristic to Sr_2RuO_4. The results obtained from this model are consistent with those of Sr_2RuO_4 observed so far resolving a puzzle about its superconductivity.

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