Triplet superconductivity in Sr_2RuO_4 in terms of the $t-J-I$-model

E. V. Kuz'min

Krasnoyarsk State University, 660062 Krasnoyarsk, Russia

S. G. Ovchinnikov*)

L. V. Kirenskii Physics Institute, Siberian Branch, Russian Academy of Sciences, 660036 Krasnoyarsk, Russia

(Submitted March 11, 1999)
Fiz. Tverd. Tela (St. Petersburg) **41**, 1936–1938 (November 1999)

A generalized $t-J-I$-model is proposed for Sr_2RuO_4 that takes the strong intra-atomic correlations of the d electrons and the features of the electronic structure of Sr_2RuO_4 into account. It is shown that, in the limit of strong correlations, there are no singlet s-type solutions for the superconducting state, but triplet solutions exist because of ferromagnetic spin correlations. For typical values of the model parameters, $T_c \sim 1$ K, consistent with the value of T_c for Sr_2RuO_4. © *1999 American Institute of Physics.* [S1063-7834(99)00511-0]

Despite its low $T_c \sim 1$ K,[1] superconductivity in Sr_2RuO_4 is of great interest for two reasons. First, this is the only superconducting oxide that is isostructural with La_2CuO_4 but does not contain copper, so that a comparison with the HTSC oxides may clarify the role of the copper. Second, the superconducting state in Sr_2RuO_4 is analogous to superfluid ^{3}He and has p-type pairing.[2,3] The triplet superconductivity in Sr_2RuO_4 has been discussed in terms of a band theory as a consequence of pairing induced by ferromagnetic spin fluctuations. It was not clear beforehand whether the triplet state would be maintained in the presence of the strong electronic correlations owing to the high effective mass of the electrons in the γ band ($12m_e$ from data on quantum mechanical oscillations[5]) and the closeness of Sr_2RuO_4 to the boundary for a Mott–Hubbard transition (the isostructural, isoelectronic crystal Sr_2FeO_4 is a Mott dielectric[6]). In this paper we propose a generalization of the $t-J$ model for strongly correlated electrons in the form of an additional ferromagnetic exchange (I) term caused by the specific features of the electronic structure and show that, with strong correlations, triplet solutions exist for the superconducting ordering parameter with $T_c \sim 1$ K for typical values of the model parameters. There are no singlet s-type solutions since they do not satisfy the sum rule.

A $(d_{x^2-y^2}-p)-\sigma$ coupling is characteristic of the copper oxides; here the interaction of neighboring copper spins only occurs through a superexchange (indirect) interaction ($t-J$-model, $J \sim t^2/U$). Sr_2RuO_4 is distinguished by the fact that bands formed by a π bond of the type $(d_{xy}-p)$ exist in the neighborhood of the Fermi level.[7,8] A simple quantum mechanical analysis shows that in this case the d_{xy} orbitals of neighboring cations overlap and this leads to a Heisenberg exchange interaction, in addition to the superexchange interaction through oxygen. For describing strongly correlated electronic states with developed ferromagnetic fluctuations, we propose a generalized $t-J-I$-model with the Hamiltonian

$$H=\sum_{\mathbf{f},\sigma}(\varepsilon-\mu)X_{\mathbf{f}}^{\sigma,\sigma}-t\sum_{\mathbf{f},\boldsymbol{\delta},\sigma}X_{\mathbf{f}}^{\sigma,0}X_{\mathbf{f}+\boldsymbol{\delta}}^{0,\sigma}$$

$$+J\sum_{\mathbf{f},\boldsymbol{\delta}}K_{\mathbf{f},\mathbf{f}+\boldsymbol{\delta}}^{(-)}-I\sum_{\mathbf{f},\boldsymbol{\delta}}K_{\mathbf{f},\mathbf{f}+\boldsymbol{\delta}}^{(+)},$$

$$K_{\mathbf{f},g}^{\pm}=\hat{\mathbf{S}}_{\mathbf{f}}\hat{\mathbf{S}}_{\mathbf{g}}\pm\frac{1}{4}\hat{n}_{\mathbf{f}}\hat{n}_{\mathbf{g}}, \tag{1}$$

where we have introduced the Hubbard operators $X_{\mathbf{f}}^{pq}=|p><q|$, which act on the intra-atomic states $|0>$ (no holes) and $|\sigma>$ (a hole with spin $\sigma=\pm 1/2$) and automatically exclude two-particle states, and $\hat{\mathbf{S}}_{\mathbf{f}}$ and $\hat{n}_{\mathbf{f}}$ are the spin- and particle-number operators at site $\mathbf{f}$. The signs in Eq. (1) have been chosen so that all the parameters (t, J, I) are positive and, for simplicity, only the sum over the nearest neighbors is retained (the vector $\boldsymbol{\delta}$). For infinitely large correlations, i.e., $U\to\infty$, the antiferromagnetic exchange $J\to 0$. We restrict ourselves to the case $I>J$. The electron concentration $n_e=N_e/N$ varies over the range $0\leqslant n_e\leqslant 1$. As $n_e\to 1$ the kinetic energy goes to zero and a dielectric state exists with a long-range ferromagnetic ordering. As the hole concentration increases, the ferromagnetism breaks up and a superconducting state can appear.

The Hamiltonian (1) describes quasiparticles in the lower Hubbard band. For comparison with Sr_2RuO_4 it is necessary to explain that the hole-α and electron-β bands in this model are a reservoir of particles and determine the chemical potential. The electron-α band, which is also responsible for the superconductivity,[9] splits into upper and lower Hubbard bands when strong electron correlations are taken into account. Calculations[8] show that the β band is half filled, the α band contains $n_\alpha=0.28$ holes, and the γ band is filled somewhat beyond half with electrons to $n_{e\gamma}=1.28$. This means that in the electron representation, the lower Hubbard γ band is completely filled, while the upper band contains 0.28 electrons. We shall work in the hole rep-

resentation, where the number of holes in the lower Hubbard band is $n_h=1-n_\alpha$, $0\leqslant n_h\leqslant 1$.

The exact equation of motion for the operator $X_{\mathbf{k}\sigma}$, the Fourier transform of the operator $X_{\mathbf{f}}^{0\sigma}$, is ($\hbar=1$)

$$i\dot{X}_{\mathbf{k}\sigma}=\xi_{\mathbf{k}}X_{\mathbf{k}\sigma}+L_{\mathbf{k}\sigma},$$

$$L_{\mathbf{k}\sigma}=-\frac{1}{\sqrt{N}}\sum_{\mathbf{p}}\gamma_{\mathbf{p}}(X^{-\sigma,\sigma}_{\mathbf{k}-\mathbf{p}}X_{\mathbf{p},-\sigma}-X^{-\sigma,-\sigma}_{\mathbf{k}-\mathbf{p}}X_{\mathbf{p},\sigma})$$

$$+\frac{1}{\sqrt{N}}\sum_{\mathbf{p}}\gamma_{\mathbf{k}-\mathbf{p}}[(g-\lambda)X^{-\sigma,\sigma}_{\mathbf{k}-\mathbf{p}}X_{\mathbf{p},-\sigma}$$

$$-gX^{-\sigma,-\sigma}_{\mathbf{k}-\mathbf{p}}X_{\mathbf{p},\sigma}-\lambda X^{\sigma,\sigma}_{\mathbf{k}-\mathbf{p}}X_{\mathbf{p},\sigma}], \tag{2}$$

where $\xi_{\mathbf{k}}=-\gamma_{\mathbf{k}}-\overline{\mu}$, $\gamma_{\mathbf{k}}=z^{-1}\Sigma_{\boldsymbol{\delta}}\exp(i\mathbf{k}\boldsymbol{\delta})$, $g=J/t$, and $\lambda=I/t$. Here all energies are expressed in units of zt (the band half-width), $\overline{\mu}=(\mu-\varepsilon)/zt$, and z is the number of nearest neighbors.

A convenient procedure for linearizing the nonlinear operator L with separation of the normal and anomalous averages is the irreducible operator method,[10]

$$L_{\mathbf{k}\sigma}=\overline{L}_{\mathbf{k}\sigma}-\frac{\langle\{L_{\mathbf{k}\sigma},X^{+}_{\mathbf{k}\sigma}\}_{+}\rangle}{\langle\{X_{\mathbf{k}\sigma},X^{+}_{\mathbf{k}\sigma}\}_{+}\rangle}X_{\mathbf{k}\sigma}$$

$$-\frac{\langle\{L_{\mathbf{k}\sigma},X^{+}_{-\mathbf{k},-\sigma}\}_{+}\rangle}{\langle\{X^{+}_{-\mathbf{k},-\sigma},X_{-\mathbf{k},-\sigma}\}_{+}\rangle}X^{+}_{-\mathbf{k},-\sigma}. \tag{3}$$

Neglecting the irreducible part $\overline{L}_{\mathbf{k}\sigma}$, we write down the equation of motion in the average field approximation

$$i\dot{X}_{\mathbf{k},\uparrow}=\tilde{\xi}_{\mathbf{k}}X_{\mathbf{k},\uparrow}-\frac{\Delta_{\mathbf{k}}}{c(n)}X^{+}_{-\mathbf{k},\downarrow},$$

$$i\dot{X}^{+}_{-\mathbf{k},\downarrow}=-\tilde{\xi}_{\mathbf{k}}X^{+}_{-\mathbf{k},\downarrow}-\frac{\Delta^{*}_{\mathbf{k}}}{c(n)}X_{\mathbf{k},\uparrow}. \tag{4}$$

Here the renormalization of the dispersion relation is taken into account in the simplest ''Hubbard 1'' form for a nonmagnetic state, $n_\uparrow=n_\downarrow=n_h/2$, $c(n)=1-n_h/2$,

$$\tilde{\xi}_{\mathbf{k}}=c(n)(-\gamma_{\mathbf{k}}-m),\quad m=[(g+\lambda)n/2+\overline{\mu}]/c(n), \tag{5}$$

and m is the effective chemical potential. The gap is given by

$$\Delta_{\mathbf{k}}=\Delta_{-\mathbf{k},\downarrow}=-\Delta_{\mathbf{k},\uparrow},$$

$$\Delta_{\mathbf{k}}=\frac{2}{N}\sum_{\mathbf{p}}\left(-\gamma_{\mathbf{p}}+\frac{1}{2}(\gamma_{\mathbf{k}+\mathbf{p}}+\gamma_{\mathbf{k}-\mathbf{p}})\right)B_{\mathbf{p}}$$

$$-\frac{\lambda}{N}\sum_{\mathbf{p}}\gamma_{\mathbf{k}+\mathbf{p}}B_{\mathbf{p}}, \tag{6}$$

where we have introduced the anomalous average $B_{\mathbf{p}}=\langle X_{-\mathbf{p}\downarrow}X_{\mathbf{p}\uparrow}\rangle$. Writing out the solution of the system of Gorkov equations for the normal and anomalous Green functions in the standard way, we obtain the following expressions for the averages in the superconducting phase:

$$n_{\mathbf{k}}=\langle X^{+}_{\mathbf{k},\uparrow}X_{\mathbf{k},\uparrow}\rangle=c(n)\frac{1}{2}\left(1-\frac{\tilde{\xi}_{\mathbf{k}}}{E_{\mathbf{k}}}\tanh\frac{E_{\mathbf{k}}}{2\tau}\right)\equiv c(n)f_{\mathbf{k}}, \tag{7}$$

$$kB^{*}_{\mathbf{k}}=\langle X^{+}_{\mathbf{k},\uparrow}X^{+}_{-\mathbf{k},\downarrow}\rangle=\frac{\Delta^{*}_{\mathbf{k}}}{2E_{\mathbf{k}}}\tanh\frac{E_{\mathbf{k}}}{2\tau}, \tag{8}$$

where $E^2_{\mathbf{k}}=\tilde{\xi}^2_{\mathbf{k}}+|\Delta_{\mathbf{k}}|^2/c^2(n)$ and $\tau=k_BT/zt$ is the dimensionless temperature. Besides the ordinary self consistent equations for the chemical potential and gap $\Delta_{\mathbf{k}}$ in the theory of strongly correlated systems, it is necessary to include the sum rules for the anomalous averages (a consequence of the algebra of X-operators that excludes two-particle states),

$$\frac{1}{N}\sum_{\mathbf{k}}B_{\mathbf{k}}=0. \tag{9}$$

Breaking up the anomalous averages into a sum of symmetric and antisymmetric components,

$$B_{\mathbf{p}}=B^{(S)}_{\mathbf{p}}+B^{(a)}_{\mathbf{p}},\quad B^{(S)}_{\mathbf{p}}=B^{(S)}_{-\mathbf{p}},\quad B^{(a)}_{-\mathbf{p}}=-B^{(a)}_{\mathbf{p}},$$

it is easy to write down an expression for the gap of the form

$$\Delta_{\mathbf{k}}=\alpha_{\mathbf{k}}\Delta_S+\lambda_{\mathbf{k}}\Delta_a,$$

$$\alpha_{\mathbf{k}}=2-(2g-\lambda)\gamma_{\mathbf{k}},\quad \lambda_{\mathbf{k}}=\lambda S_{\mathbf{k}},$$

$$S_{\mathbf{k}}=(\sin k_x+\sin k_y)/2. \tag{10}$$

The symmetric solutions $\Delta^{(S)}_{\mathbf{k}}=\alpha_{\mathbf{k}}\Delta_S$ correspond to singlet pairing and the antisymmetric solutions $\Delta^{(a)}_{\mathbf{k}}=\lambda_{\mathbf{k}}\Delta_a$, to triplet pairing. The first term in $\alpha_{\mathbf{k}}$, which equals 2, arises from a kinematic mechanism for pairing.[11] It is easy to see that the symmetric solution does not satisfy the sum rule.[12] At the same time, this rule is automatically satisfied for the antisymmetric solution. The ordering parameter Δ_α can be written in the form

$$\Delta_\alpha=\frac{i}{\sqrt{2}z}\sum_{\boldsymbol{\delta}}\frac{1}{N}\sum_{\mathbf{f}}\left\langle\frac{1}{\sqrt{2}}(X^{0,\downarrow}_{\mathbf{f}}X^{0,\uparrow}_{\mathbf{f}+\boldsymbol{\delta}}-X^{0,\downarrow}_{\mathbf{f}+\boldsymbol{\delta}}X^{0,\uparrow}_{\mathbf{f}})\right\rangle, \tag{11}$$

containing the average of the annihilation operator for a triplet pair with $S^z=0$ at neighboring lattice sites. For this solution, at $T=0$ we have the following equations for the gap

$$\frac{2-n_h}{\lambda}=\frac{1}{N}\sum_{\mathbf{p}}\frac{S^2_{\mathbf{p}}}{\sqrt{(\gamma_{\mathbf{p}}+m)^2+S^2_{\mathbf{p}}D^2_a}} \tag{12}$$

and for the chemical potential

$$\frac{n_h}{2-n_h}=\frac{1}{2N}\sum_{\mathbf{p}}\left(1+\frac{\gamma_{\mathbf{p}}+m}{\sqrt{(\gamma_{\mathbf{p}}+m)^2+S^2_{\mathbf{p}}D^2_a}}\right), \tag{13}$$

where $D^2_a=\lambda^2|\Delta_a|^2/c^4(n)$, which have solutions only for $\lambda>0$. The equations for T_c have the form

$$\frac{2-n_h}{\lambda}=\frac{1}{N}\sum_{\mathbf{p}}\frac{S^2_{\mathbf{p}}}{|\gamma_{\mathbf{p}}+m|}\tanh\left\{\frac{c(n)|\gamma_{\mathbf{p}}+m|}{2\tau_c}\right\}. \tag{14}$$

Numerical solutions of Eqs. (12)–(14) show that the gap and T_c depend nonmonotonically on the hole concentration and are nonzero within a finite interval of concentrations (n_{h1},n_{h2}) whose boundaries depend on the interaction constant λ. These dependences are characterized by a smooth maximum near populations $n_h\approx 0.7-0.8$. For Sr_2RuO_4 with $n_h=1-n_\alpha$ and $n_\alpha\approx 0.28$, this means close to optimum dop-

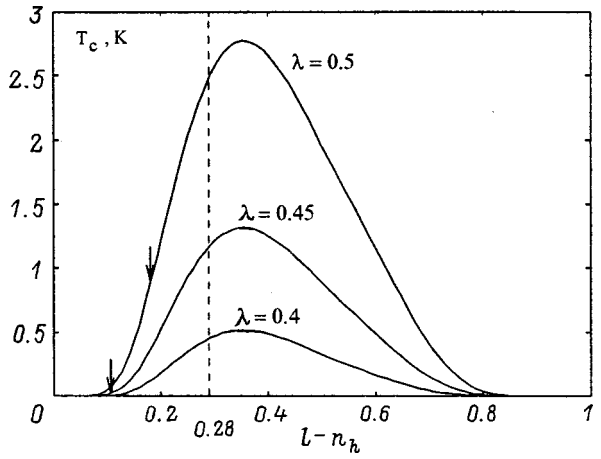

FIG. 1. T_c as a function of the hole concentration in Sr_2RuO_4 for three values of the dimensionless interaction parameter λ; the dashed curve is the hole concentration in the γ band, whose half width is 0.4 eV.

ing. $T_c \sim 1$ K for the typical model parameters. Thus, for $t=0.1$ eV, $n_\alpha = 0.28$, and $\lambda = 0.5$, we have $T_c \approx 2$ K.

As opposed to the BCS theory, the entire band is significant in Eqs. (12)–(14) and not just a narrow layer near the chemical potential. An approximate analytic solution for T_c can be written down if, when taking the integral with respect to the energy, a narrow layer of width 2δ ($\delta \approx 10^{-2}$) is selected near the chemical potential where the ordinary logarithm is obtained and, outside this, $\tanh x \approx 1$. This yields the following expression for T_c:

$$T_c = 1.14 z t c(n) \delta \exp\left\{ \frac{-c(n)}{(\lambda+\lambda_0)S^2(m)} \right\} \tag{15}$$

with values that differ from the numerical solution by a few percent. In Eq. (15) the parameter λ_0 describes the contribution from the outer part of the band,

$$\lambda_0 = \lambda^2 \frac{A(m,\delta)}{2-n_h-\lambda A(m,\delta)},$$

$$A(m,\delta) = \int_{-1}^{m-\delta} \frac{S^2(\omega)d\omega}{|\omega-m|} + \int_{m+\delta}^{1} \frac{S^2(\omega)d\omega}{|\omega-m|}, \tag{16}$$

where $S^2(\omega)$ is the average value of $S_{\mathbf{k}}^2$ on the isoenergetic surface $\omega = \gamma_{\mathbf{k}}$. T_c is plotted as a function of the hole concentration for different values of the parameter λ in Fig. 1. The arrows indicate the point where the transition to the ferromagnetic phase takes place, as defined by equality between the energies of the ferromagnetic and superconducting phases.

In conclusion, we note that generalizing the $t-J$-model by adding a ferromagnetic interaction of neighboring spins yields the $t-J-I$-model, which is a realistic model for describing the strongly correlated electrons in Sr_2RuO_4. The triplet superconductivity obtained previously[4] in terms of a band picture of Sr_2RuO_4 is also retained with strong electron correlations. An earlier attempt was made to describe a phononless mechanism for superconductivity in Sr_2RuO_4 in a strong electron correlation regime,[13] but the questions of the symmetry of the order parameter and the role of ferromagnetic fluctuations were not examined there.

This work was supported by the federal program ''State Support for the Integration of Fundamental Science and Higher Education'' (Grant No. 69) and the Krasnoyarsk Regional Science Foundation (grant 8F32).

*)E-mail: sgo@post.krascience.rssi.ru

[1] Y. Maeno, H. Hasimoto, K. Yoshida, S. Nichizaki, T. Fujita, J. G. Bednorz, and F. Lichtenberg, Nature (London) **372**, 532 (1994).
[2] T. M. Rice and M. Sigrist, J. Phys.: Condens. Matter **7**, L643 (1995).
[3] T. M. Rice, Nature (London) **396**, 627 (1998).
[4] I. I. Mazin and D. J. Singh, Phys. Rev. Lett. **79**, 733 (1997).
[5] A. P. Mackenzie, S. R. Julian, A. J. Diver, G. J. McMullan, M. P. Ray, G. G. Lonzarich, Y. Maeno, S. Nishitaki, and T. Fujita, Phys. Rev. Lett. **76**, 3786 (1996).
[6] P. Adler, A. F. Goncharov, K. Syassen, and E. Schonher, Phys. Rev. B **50**, 11396 (1994).
[7] T. Oguchi, Phys. Rev. B **51**, 1385 (1995).
[8] D. J. Singh, Phys. Rev. B **52**, 1358 (1995).
[9] T. M. Riseman, P. G. Kealey, E. M. Forgan, A. P. Mackenzie, L. M. Galvin, A. W. Tyler, S. L. Lee, C. Ager, D. Mck. Paul, C. M. Aegerter, R. Cubitt, Z. O. Mao, T. Akima, and Y. Maeno, Nature (London) **396**, 242 (1998).
[10] S. V. Tyablikov, *Methods in the Quantum Theory of Magnetism* (Nauka, Moscow, 1975).
[11] R. O. Zaĭtsev and V. A. Ivanov, Fiz. Tverd. Tela (Leningrad) **29**, 2554 (1987) [Sov. Phys. Solid State **29**, 1475 (1987)].
[12] N. M. Placida, V. Yu. Yushanhai, and I. V. Stasyk, Physica C **160**, 80 (1989).
[13] R. O. Zaitsev and Ju. V. Mihailova, Physica C **282–287**, 1677 (1997).

Translated by D. H. McNeill