Electronic theory for superconductivity in Sr$_2$RuO$_4$: triplet pairing due to spin-fluctuation exchange

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Using a two-dimensional Hubbard Hamiltonian for the three electronic bands crossing the Fermi level in Sr$_2$RuO$_4$ we calculate the band structure and spin susceptibility $\chi(q, \omega)$ in quantitative agreement with nuclear magnetic resonance (NMR) and inelastic neutron scattering (INS) experiments. The susceptibility has two peaks at $Q_1 = (2\pi/3, 2\pi/3)$ due to the nesting Fermi surface properties and at $q_0 = (0.6\pi, 0)$ due to the tendency towards ferromagnetism. Applying spin-fluctuation exchange theory as in layered cuprates we determine from $\chi(q, \omega)$, electronic dispersions, and Fermi surface topology that superconductivity in Sr$_2$RuO$_4$ consists of triplet pairing. Combining the Fermi surface topology and the results for $\chi(q, \omega)$ we can exclude s- and d-wave symmetry for the superconducting order parameter. Furthermore, within our analysis and approximations we find that f-wave symmetry is slightly favored over p-wave symmetry due to the nesting properties of the Fermi surface.

74.20.Mn, 74.25.-q, 74.25.Ha

The novel spin-triplet superconductivity with $T_c = 1.5$K observed recently in layered Sr$_2$RuO$_4$ seems to be a new example of unconventional superconductivity [1]. The presence of incommensurate antiferromagnetic and ferromagnetic spin fluctuations confirmed recently by inelastic neutron scattering (INS) [2] and NMR $^{17}$O Knight shift [3], respectively, suggests a pairing mechanism for Cooper-pairs due to spin fluctuations. This is further supported by the observed non s-wave symmetry of the order parameter. Likely Sr$_2$RuO$_4$ is another example of spin fluctuations induced superconductivity. This makes the theoretical investigation of ruthenates very interesting. NMR [4] and polarized neutron scattering [5] measurements indicate spin-triplet state Cooper-pairing. In analogy to $^3$He this led theorists to conclude that p-wave superconductivity is present [6]. However, by fitting the specific heat and the ultrasound attenuation Dahn et al. doubted p-wave superconductivity [7] and propose an f-wave symmetry of the superconducting order parameter. A similar conclusion was drawn in Ref. [8]. Recently it has been reported that also thermal conductivity measurements are most consistent with f-wave symmetry [9].

Clearly, it is important to analyze more definitely the origin of superconductivity, triplet pairing and also the symmetry of the order parameter on a basis of an electronic calculation. This is difficult, since there are three Ru$^{4+}$ t$_{2g}$ bands that cross the Fermi level with $\approx$2/3-filling of every band in Sr$_2$RuO$_4$. The hybridization between all three bands seems to cause a single $T_c$. All bands cross the Fermi level and hence, the cross-susceptibilities are not small and play an important role. In view of these facts the previous theoretical analysis of the gap symmetries and competition between p and d-wave superconductivity [10,11] must be re-examined and it is necessary to determine superconductivity within an electronic theory and to derive the symmetry of the order parameter from general arguments.

In this letter we present an electronic theory which takes into account the hybridization between all three bands. We calculate the Fermi surface (FS), energy dispersion and the spin susceptibility $\chi$ including all cross-susceptibilities. Then, we analyze the pairing interaction mediated by the spin fluctuations exchange in Sr$_2$RuO$_4$. Analyzing experimental results for the $^{17}$O Knight shift and INS data as well as the FS observed by Angle-Resolved-Photoemission-Spectroscopy (ARPES) [12] we obtain values for the hopping integrals and effective Coulomb repulsion $U$. Taking this as an input into the pairing interaction we analyze the p-, d- and f-wave superconducting gap symmetries. The delicate competition between weak ferromagnetic spin fluctuations and relatively strong incommensurate antiferromagnetic spin fluctuations due to nesting of the FS cause triplet Cooper-pairing. We get singulet d$_{x^2-y^2}$-wave symmetry is energetically less favorable.

We start from the two-dimensional three-band Hubbard Hamiltonian

\[ H = \sum_{k,\sigma} \sum_{\alpha} t_{k\alpha} a_{k,\alpha\sigma}^\dagger a_{k,\alpha\sigma} + \sum_{i,\alpha} U_{\alpha} n_{i\alpha \uparrow} n_{i\alpha \downarrow}, \]  

where $a_{k,\alpha\sigma}$ is the Fourier transform of the annihilation operator for the $d_{\alpha}$ orbital electrons ($\alpha = xy, yz, zx$) and $U_{\alpha}$ is an effective on-site Coulomb repulsion. $t_{k\alpha}$ denotes the energy dispersions of the tight-bindings bands calculated as follows: $t_{k\alpha} = -\epsilon_0 - 2t_x \cos k_x - 2t_y \cos k_y + 4t' \cos k_x \cos k_y$. In accordance with experimental measurements of the Fermi surface and energy dispersions we choose the values for the parameter set ($\epsilon_0, t_x, t_y, t'$) as (0.5, 0.42, 0.44, 0.14), (0.23, 0.31, 0.055, 0.01), and (0.24, 0.045, 0.31, 0.01) eV for $d_{xy}$-, $d_{xz}$-, and $d_{yz}$-orbitals [12]. The analysis of de Haas-van Alphen experiments [13] shows a substantial hybridization between $xz$- and $yz$-orbitals [14].
yz- orbitals about \( t_{\perp} = 0.1 \text{ eV} \), but not with the \( xy \)-orbital \[13\]. However, the observation of a single \( T_c \) implies the coupling between all three bands. Therefore, we choose a weak hybridization \( t_{hyb} = 0.01 \text{ eV} \) (hybridization between \( xy \)- and \( xz, yz \)-orbitals) \( \ll t_{\perp} \). Note, even such a weak hybridization transfers the nesting properties to the \( xy \)-orbital. In the inset of Fig. 1 we show the resultant energy dispersions of the obtained hole-like \( \alpha \)-band and electron-like \( \beta \)- and \( \gamma \)-bands after hybridization. Due to the small value of hybridization between \( xy \) and \( yz \), \( xz \) orbitals the dispersion curves and resulting FS (see also Fig. 4) look quite similar to the non-hybridized ones \[13\]. However, the importance of hybridization between these orbitals for the spin susceptibility, \( \chi(q, \omega) \), will be immediately seen from the analysis of the latter. The susceptibility is given by:

\[
\chi_{ij}^0(q, \omega) = \frac{1}{N} \sum_{k} f(e^i_{k, \gamma}) - f(e^j_{k+q, \gamma}) - e^i_{k, \gamma} + \omega + i0^+ ,
\]

where \( f(\epsilon) \) is the Fermi function and \( e^i_{k, \gamma} \) is the energy dispersion of the \( \alpha \), \( \beta \), and \( \gamma \) band \[13\].

In Fig. 1 we show the momentum dependence of the real part of \( \chi_{\gamma}^0 \). While hybridization between bands does not affect much the energy dispersion, it changes significantly the susceptibility of the \( \gamma \)-band. In particular, the nesting properties of \( xz \)- and \( yz \)-orbitals reflected by the peak at \( Q = (2\pi/3, 2\pi/3) \) in \( \chi_{\gamma}^0 \) are caused by the hybridization between \( xz, yz \) and \( xy \) bands. Note, without taking into account the hybridization one would not get the peak at \( Q \) in the \( \gamma \)-band, but only a broad hump as discussed earlier \[13\]. The small peak at \( q_i \approx (0.2\pi, 0) \) is due to the original tendency towards ferromagnetism of the \( xy \)-band and is not affected by the hybridization.

Our results for \( \chi(q, \omega) \) will have important consequences and were obtained in contrast to previous studies from observed band dispersions. The susceptibility matrix \( [\chi_{ij}] \) is calculated where \( i, j \) refers to the hybridized bands. As already mentioned the non-diagonalized susceptibilities are not small and thus cannot be neglected. After diagonalizing the matrix \( [\chi_{ij}] \) we get within RPA with an effective \( U(q) \) for \( \chi \):

\[
\chi(q, \omega) = \frac{\chi_0(q, \omega)}{1 - U(q)\chi_0(q, \omega)} ,
\]

where now \( \chi_0(q, \omega) = \sum_{i'} \chi_{0i'}^0(q, \omega) \). Here, \( \chi_{0i'}^0(q, \omega) \) \( (i' = \alpha' , \beta', \gamma') \) are the diagonal elements of the diagonalized matrix \( [\chi_{ij}^0] \). This obtained susceptibility characterizes the normal state magnetic properties of \( Sr_2RuO_4 \). Its spin fluctuations are given by \( \chi(q, \omega) \) with peaks at \( Q_s \) and \( q_i \). These wave vectors are important for determining the symmetry of the superconducting order parameter.

In Fig. 2 we present the results for \( \chi(q, \omega) \) obtained from Eq. \[3\] and the susceptibilities \( \chi_0^0(q, \omega) \) obtained after diagonalization of \( [\chi_{ij}^0(q, \omega)] \). Here, we approximate \( U(q) \) by \( U = 0.345 \text{ eV} \) which gives agreement with INS \[18\]. Remarkably, the peak in Re \( \chi \) at \( Q_s \) remains nearly the same as for \( \chi_{\gamma}^0 \). The peak at \( q_i \) is also present, but slightly shifted to a larger value. Clearly, \( \chi_{\gamma}^0 \) is much larger than \( \chi_{\beta'}^0 \) and \( \chi_{\alpha'}^0 \). Our calculations also have shown that cross-susceptibilities \( \chi_{ij}^0 \) \( (i \neq j) \) cannot be neglected.

In Fig. 3(a) we compare our calculation of the temperature dependence of the uniform spin susceptibility \( \chi(0, 0) \) which is measured by the \( ^{17}O \) Knight shift \[3\], and in Fig. 3(b) we compare \( \text{Im} \chi(Q_i, \omega) \) with INS data \[3\]. For the calculation of \( \chi(0, 0) \) we approximate \( U(q) \) by \( U(0) = 0.177 \text{ eV} \) \[13\] which gives agreement with Knight shift measurements and is also taken in previous calculations. These comparisons shed light on the validity of our results for \( \chi(q, \omega) \). Note, we also take into account that there are four electrons per three \( t_{2g} \)-bands that would give every \( \chi_0^0 \) an additional weight 4/3. Our results are in
uniform spin susceptibility with $U_0 = 0.177\, \text{eV}$ compared with the $^{17}$O Knight shift measurements. The peak is due to thermal activation involving $\gamma$ and $\alpha$, $\beta$ bands. (b) Calculated frequency dependence of $\text{Im} \chi(Q_i, \omega)$ compared to INS data using $U_{Q_i} = 0.345\, \text{eV}$.

fair agreement with experiment that shows a tendency towards ferromagnetism [20]. The maximum in $\chi_{RPA}(0,0)$ at about 25K results from thermally activated changes in the populations of the bands near $E_F$. In Fig. 3(b) we compare our results with INS data. In this case we must take $U_{Q_i} = 0.345\, \text{eV}$ in order to fit $\chi(q, \omega)$ to the peak position and height at $\omega = 6\, \text{meV}$ as observed in INS.

While an uncertainty in the INS data (shown in Fig. 3(b)) is present, our results for $\chi(q, \omega)$ should be a useful basis for further calculations. The antiferromagnetic spin excitations result in incommensurate antiferromagnetic Ru-spin alignment at distances larger than nearest neighbors. Hence, if Cooper-pairing involves nearest neighboring Ru spins also incommensurate antiferromagnetic fluctuations will cause triplet-pairing because neighboring Ru spins see also partly a ferromagnetic environment. Note, $\chi(q, \omega)$ controls the symmetry of the superconducting order parameter.

For the analysis of superconductivity in Sr$_2$RuO$_4$ we take into account that experiment observes non-s-wave symmetry of the order parameter which strongly suggests spin-fluctuation-mediated Cooper pairing. Assuming the spin-fluctuation-induced pairing it is possible to analyze the symmetry of the superconducting state from the gap equation and our calculated results for $\chi(q, \omega)$ with the pronounced wave vectors at $Q_i$ and $q_i$. We get for the gap equation:

$$\Delta_k = -\frac{1}{2} \sum_{k',\ell'} (V_{s\ell}^{\text{eff}}(k,k')) \Delta_{k',\ell'} \tanh \left( \frac{E_{k'}^{\ell'} - E_k^{\ell}}{2k_B T} \right),$$  \hspace{1cm} (4)

where $E_{k}^{\ell} = \sqrt{\epsilon_k^2 + \Delta_k^2}$ are the energy dispersions of the bands [21] and the pairing potential $V_{s\ell}^{\text{eff}}(k,k')$ is different for singulet ($\sigma = 0$) and triplet ($\sigma = 1$) Cooper pairing. The eigenvalue analysis of Eq. (4) will yield the symmetry with lowest energy. The $\gamma$-band plays the most important role.

For the determination of the pairing symmetry we follow the analysis by Anderson and Brinkmann for $^3\text{He}$ [23] and by Scalapino for the cuprates [24] and use the calculated FS and spin susceptibility for Sr$_2$RuO$_4$. For triplet pairing the effective pairing interaction is $(U \equiv U(Q_i))$

$$V_1^{\text{eff}}(k,k') = -\frac{U^2 \chi_0(k-k',0)}{1 - U^2 \chi_0^2(k-k',0)},$$

and for singlet pairing

$$V_0^{\text{eff}}(k,k') = \frac{U^2 \chi_0(k-k',0)}{1 - U^2 \chi_0^2(k-k',0)} + \frac{U^2 \chi_0^2(k-k',0)}{1 - U^2 \chi_0^2(k-k',0)}.$$

respectively [25]. It is important to note that for $E_F \gg \Delta_l$ the gap function can be expanded into spherical harmonics corresponding to the angular momentum $l = 1, 2, 3, \ldots$ and no mixture of $\Delta_l$ belonging to different symmetry representations can be present if a single $T_c$ is observed. Therefore, we can exclude the $(p + d)$-wave superconducting state. Using appropriate symmetry representations [8] we discuss the solutions of Eq. (4) for the $p$-, $d$-, and $f$-wave symmetries of the order parameter:

$$\Delta_p(k) = \Delta_0 \bar{\sigma} (\sin k_x + i \sin k_y),$$

$$\Delta_d(k) = \Delta_0 (\cos k_x - \cos k_y),$$

$$\Delta_f(k) = \Delta_0 \bar{\sigma} (\cos k_x - \cos k_y) (\sin k_x + i \sin k_y).$$

Note, the largest eigenvalue in Eq. (4) will yield the superconducting symmetry of $\Delta_l$ in Sr$_2$RuO$_4$. Solving Eq. (4) in the first BZ down to 5K, we find $f$-wave symmetry slightly favored. As expected $p$- and $f$-wave symmetry Cooper-pairing are close in energy ($\lambda_f = 0.76 > \lambda_p = 0.51$). A more complete analysis taking into account also the coupling between RuO$_2$ planes and interband $U$ might yield a definite answer [22]. Note, to obtain a combined energy gain from the antiferromagnetism and Cooper-pairing one expects an order parameter with nodes in the RuO$_2$ plane and possibly also with respect to the $c$-direction.

The solutions of Eq. (4) can be characterized by Fig. 4 where we present our results for the Fermi surface, wave
the contributions due to $Q$, where the sum is over all contributions due to $Q_i$. In a good approximation we linearize Eq. (1) in $\Delta_i$, i.e. $E^\prime_{k'} \to E^\prime_{k'}$, and safely put $\tanh(\Delta_i^2/2k_B T) = 1$. Therefore, the main contribution to the pairing comes from the Fermi level. Note, determining $\Delta_f$ for the RuO$_2$-planes it is sufficient to take into account only the $\gamma$-band, since only this band has a dispersion in the plane. The minus sign in Eq. (10) is cancelled for triplet pairing (see Eq. (9)). Furthermore, the summation over $k'$ in the first BZ is dominated by the contributions due to $Q_i$ and the one due to the background and $q_t$. Thus, we obtain approximately for the $\gamma$-band contribution ($l = f$ or $p$)

\[ \Delta_f(k) \approx \sum_i \frac{V_{rff}(Q_i)}{2k_{k+Q_i}} \Delta_f(k + Q_i) + \sum_i \frac{V_{rff}(q_t)}{2k_{k+q_t}} \Delta_f(k + q_t), \tag{10} \]

where the sum is over all contributions due to $Q_i$ and $q_t$. As can be seen from Fig. 4 in the case of $f$-wave symmetry the wave vector $q_t$ in Eq. (10) bridges the same number of portions of the FS with opposite and equal sign. Therefore, the second term in Eq. (10) is approximately zero for triplet pairing. We see from Fig. 4 that $Q_i$ bridges portions of the FS with equal signs of the superconducting order parameter. Thus, a solution of Eq. (10) for $\Delta_f$ is indeed possible.

In the case of $p$-wave pairing the real part of the order parameter has a node only along $k_x = 0$ in the $k_x, k_y$ plane. Then, using the corresponding Fig.4 the wave vectors $Q_i$ bridge portion of the FS where $\Re \Delta_p$ has the same or opposite sign. Regarding the $q_t$ contributions the situation is similar as in the case of the $f$-wave symmetry. Hence, we expect for the eigenvalues $\lambda_p \leq \lambda_f$ as in the result of the algebraic solution of Eq. (4). Note, for increasing nesting Fig. 4 also suggests that $f$-wave symmetry is favored more than $p$-wave. The eigenvalue analysis of the possible solutions $\Delta_f$ and $\Delta_p + i \Delta_f$ should increasingly rule out the latter for stronger nesting.

Also using similar arguments we can rule out singulet pairing on the basis of Eq. (9). In particular, assuming $d_{x^2-y^2}$-symmetry for Sr$_2$RuO$_4$ we get a change of sign of the order parameter upon crossing the diagonals of the BZ. According to Eq. (4) wave vectors around $Q_i$ connecting areas (+) and (-) contribute constructively to the pairing. Contributions due to $q_t$ and the background connecting the same sign areas subtract from the pairing (see Fig. 4 with nodes at the diagonals for illustration). Therefore, we get that the four contributions due to $Q_i$ and the background cancel the pair-building contribution due to $q_t$. As a consequence we obtain no $d_{x^2-y^2}$-wave symmetry. Note, this is in contrast to the cuprates where the cancelling contributions due to $q_t$ and the small background are negligible. For the $d_{xy}$-symmetry where the nodes are along $(\pi, 0)$ and $(0, \pi)$ directions we can argue similarly and thus exclude this symmetry.

Thus, as a result of the topology of the FS and the spin susceptibility we get for $p$- and $f$-wave the strongest pairing and can definitely exclude $d$-wave pairing. In our approximation we find that $f$-wave symmetry pairing is slightly favored over $p$-wave symmetry in Sr$_2$RuO$_4$.

In view of our Fig. 4 we also remark that while $\Re \Delta_f$ exhibits three line nodes, that can be seen by phase sensitive experiments, $|\Delta_f|^2$ shows nodes only along the diagonals, as recently found in measurements of ultrasound attenuation below $T_c$. However, note in view of the low eigenvalues for $p$ and $f$-wave symmetries and approximations used we cannot definitely conclude that $f$-wave is favored over $p$-wave.

In summary, we show that hybridization between all three bands is important and transfers the nesting properties of $xz$- and $yz$-orbitals to the $\gamma$ band in Sr$_2$RuO$_4$. Taking into account all cross-susceptibilities we successfully explain the $^{17}$O Knight shift and INS data. Most importantly, we calculate $\chi(q, \omega)$ and show on the basis of the Fermi surface topology and the calculated spin susceptibility $\chi(q, \omega)$ that triplet pairing is present in Sr$_2$RuO$_4$. Our analysis seems quite general and also predicts $d$-wave symmetry for electron- and hole-doped cuprates. To decide whether $p$- or $f$-wave symmetry pairing is present one needs to perform more complete calculations including coupling between RuO$_2$ planes, for example. If the interplane coupling involves also nesting, then corresponding nodes are expected.

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