

Electronic theory for superconductivity in Sr_2RuO_4 : Triplet pairing due to spin-fluctuation exchange

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Abstract. – Using a Hubbard Hamiltonian for the three electronic bands crossing the Fermi level in Sr_2RuO_4 , we calculate the band structure and spin susceptibility $\chi(\mathbf{q}, \omega)$ in quantitative agreement with nuclear magnetic resonance (NMR) and inelastic neutron scattering (INS) experiments. The susceptibility has two peaks at $\mathbf{Q}_i = (2\pi/3a, 2\pi/3a, 0)$ due to the nesting Fermi surface properties and at $\mathbf{q}_i = (0.2\pi/a, 0, 0)$ due to the tendency towards ferromagnetism. Applying spin-fluctuation exchange theory as in layered cuprates we determine from $\chi(\mathbf{q}, \omega)$, electronic dispersions, and Fermi surface topology that superconductivity in Sr_2RuO_4 consists of triplet pairing. Using $\chi(\mathbf{q}, \omega)$ we can exclude *s*- and *d*-wave symmetry for the superconducting order parameter. Furthermore, within our analysis and approximations we find that the order parameter will have a node between neighboring RuO_2 -planes and that in the RuO_2 -plane $f_{x^2-y^2}$ -wave and *p*-wave symmetry are close in energy.

The novel spin-triplet superconductivity with $T_c = 1.5\text{K}$ observed recently in layered Sr_2RuO_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_2RuO_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. Regarding the order-parameter symmetry some studies concluded that in analogy to ^3He *p*-wave superconductivity is present [6, 7]. However, by fitting the specific heat and the ultrasound attenuation, Dahm *et al.* [8] propose an *f*-wave symmetry of the superconducting order parameter with node in the RuO_2 -planes while in refs. [9, 10] nodes were predicted to lie half-way between the RuO_2 -planes. Also thermal-conductivity measurements are most consistent with *f*-wave symmetry with nodes between the RuO_2 -planes [11]. Note, however, that other measurements seem more consistent with a node in the RuO_2 -plane [12].

Clearly, it is important to analyze more definitely the origin of superconductivity, triplet pairing and also the symmetry of the order parameter on the basis of an electronic calculation. This is difficult, since there are three Ru^{4+} t_{2g} -bands which cross the Fermi level with $\approx 2/3$ -filling of every band in Sr_2RuO_4 . The hybridization between all three bands seems to cause a single T_c . The cross-susceptibilities between bands are not small and play an important role. In view of these facts the previous theoretical analysis of the gap symmetry and competition between p - and d -wave superconductivity [13–15] must be re-examined. It is necessary to determine superconductivity within an electronic theory and to derive the symmetry of the order parameter from electronic calculations as well as from general arguments.

In this letter we present an electronic theory which takes into account the hybridization between bands. We calculate the Fermi surface (FS), energy dispersion and the spin susceptibility χ including cross-susceptibilities. Then, we analyze the pairing interaction mediated by the spin fluctuations exchange in Sr_2RuO_4 by analyzing experimental results for the ^{17}O Knight shift and INS data as well as the FS observed by Angle-Resolved Photoemission Spectroscopy (ARPES) [16]. 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 that singlet $d_{x^2-y^2}$ -wave symmetry is energetically less favorable.

We start from the three-band Hubbard Hamiltonian

$$H = \sum_{\mathbf{k},\sigma} C_{\mathbf{k},\sigma}^\dagger \begin{pmatrix} t_{\mathbf{k}}^{xy} & 0 & 0 \\ 0 & t_{\mathbf{k}}^{yz} & t_{\perp} \cos k_z c \\ 0 & t_{\perp} \cos k_z c & t_{\mathbf{k}}^{xz} \end{pmatrix} C_{\mathbf{k},\sigma} + \sum_{i,\alpha} U_{\alpha} n_{i\alpha\uparrow} n_{i\alpha\downarrow}, \quad (1)$$

where $C_{\mathbf{k},\sigma} = (c_{\mathbf{k},\sigma,xy}^\dagger, c_{\mathbf{k},\sigma,yz}^\dagger, c_{\mathbf{k},\sigma,xz}^\dagger)$ is the Fourier transform of the annihilation operator for the d_{α} -orbital electrons ($\alpha = xy, yz, zx$) and U_{α} their effective on-site Coulomb repulsion. $t_{\mathbf{k}\alpha}$ denotes the energy dispersions of the tight-binding bands calculated as follows: $t_{\mathbf{k}\alpha} = -\epsilon_0 - 2t_x \cos k_x a - 2t_y \cos k_y a + 4t' \cos k_x a \cos k_y a$. Note that a and c are the lattice constants of Sr_2RuO_4 . 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_{zx} -, and d_{yz} -orbitals [16]. The analysis of de Haas-van Alphen experiments [17] shows a substantial hybridization between xz - and yz -orbitals about $t_{\perp} = 0.1$ eV, but not with the xy -orbital [18]. We propose here that this dispersion arises due to the hopping between neighboring RuO_2 -planes. In the inset of fig. 1 we show the resultant energy dispersions of the obtained hole-like α -band and electron-like β - and γ -bands after hybridization. Due to the introduced hybridization between yz - and xz -orbitals their dispersion curves and resulting FS change as one goes along the z -direction (see also fig. 3 below). This will be seen in the analysis of the spin susceptibility.

The susceptibility for the non-interacting electrons is given by

$$\chi_0^{ij}(\mathbf{q}, \omega) = \frac{1}{N} \sum_{\mathbf{k}} |M_{\mathbf{k}+\mathbf{q}}^{ij}|^2 \frac{f(\epsilon_{\mathbf{k}}^i) - f(\epsilon_{\mathbf{k}+\mathbf{q}}^j)}{\epsilon_{\mathbf{k}+\mathbf{q}}^i - \epsilon_{\mathbf{k}}^j + \omega + i0^+}, \quad (2)$$

where $f(\epsilon)$ is the Fermi function and $\epsilon_{\mathbf{k}}^i$ is the energy dispersion of the α -, β -, and γ -band. The matrix element $M_{\mathbf{k}}^{ij}$ was calculated previously [18]. In particular it was found out that

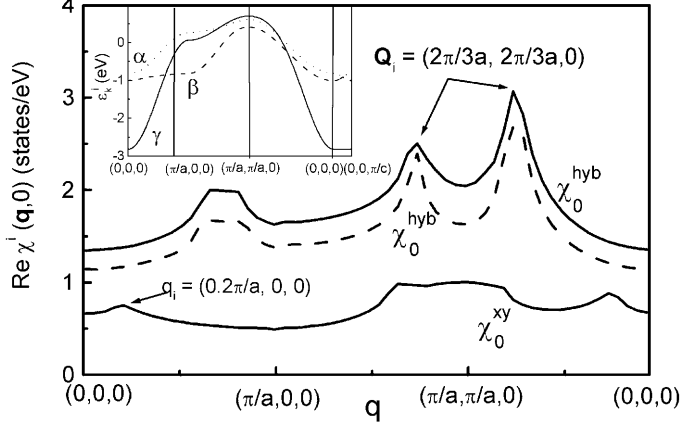


Fig. 1 – (a) Calculated susceptibility $\text{Re } \chi_0^{\text{hyb}}(\mathbf{q}, \omega = 0)$ and $\chi_0^{\gamma\gamma}(\mathbf{q}, \omega = 0)$ (solid curves) obtained from electronic calculations using the hybridized bands in the RuO_2 -plane ($k_z = 0$) and $\text{Re } \chi_0^{\text{hyb}}(\mathbf{q}, \omega = 0)$ (dashed curve) between the neighboring RuO_2 -planes ($k_z = \pi/2c$). The wave vectors $\mathbf{Q}_i = (\frac{2\pi}{3a}, \frac{2\pi}{3a}, 0)$ and $\mathbf{q}_i = (0.2\pi/a, 0, 0)$ reflect nesting of the α - and β -bands and the original tendency of the γ -band towards ferromagnetism, respectively. These wave vectors play a role for Cooper pairing. The inset shows results for the energies ϵ_k^i of the hybridized bands ($i = \alpha, \beta, \gamma$).

due to introduced hybridization the cross-susceptibility, $\chi^{\alpha\beta}(\mathbf{q}, \omega)$, becomes non-zero and enhances substantially the nesting properties at $\mathbf{Q}_i \approx (2\pi/3a, 2\pi/3a, 0)$ of the α - and β -band in the RuO_2 -plane.

In fig. 1 we show the momentum dependence of the real part of $\chi_0^{\text{hyb}} = \chi_0^{\alpha\alpha} + \chi_0^{\beta\beta} + 2\chi_0^{\alpha\beta}$ and $\chi_0^{\gamma\gamma}$ in the RuO_2 -plane. Due to the transitions between α - and β -bands the nesting properties of χ_0^{hyb} at $\mathbf{Q}_i = (\frac{2\pi}{3a}, \frac{2\pi}{3a}, 0)$ are larger than the nesting properties of xz - and yz -orbitals alone. $\chi_0^{\gamma\gamma}$ shows only slight structures at $\mathbf{q}_i \approx (0.2\pi/a, 0, 0)$ due to the original tendency of the xy -orbital towards ferromagnetism and broad hump around $(\pi/a, \pi/a, 0)$.

The situation is different if one goes between the RuO_2 -planes ($k_z = \pi/2c$). In this case the hybridization between the bands becomes zero and thus the inter-band nesting is reduced. Therefore the spin susceptibility χ^{hyb} has a smaller peaks at $\mathbf{Q}'_i = (\frac{2\pi}{3}, \frac{2\pi}{3}, \frac{\pi}{2c})$, while $\chi_0^{\gamma\gamma}$ is unchanged. Note, the wave vectors \mathbf{Q}_i and \mathbf{q}_i play an important role for the Cooper pairing. Moreover, as we will see later, the reduction of the spectral weight of χ_0^{hyb} between the RuO_2 -planes will have important consequences and can lead to a node of the superconducting gap between RuO_2 -planes in good agreement with experiment.

Also for comparison with experiment we use the RPA approximation for χ . Thus, we take into account the correlation effects in Sr_2RuO_4 . We get $\chi^{ij} = \chi_0^{ij} + \chi_0^{il} U_l \chi'^{lj}$. Assuming $U_{ij} = \delta_{ij} U$ one gets

$$\chi^{ij}(\mathbf{q}, \omega) \approx \frac{\chi_0^{ii}(\mathbf{q}, \omega)}{1 - U \chi_0^{ii}(\mathbf{q}, \omega)}. \quad (3)$$

The total spin susceptibility is defined as $\chi^{\text{tot}}(\mathbf{q}, \omega) = \chi^{\text{hyb}}(\mathbf{q}, \omega) + \chi^{\gamma\gamma}(\mathbf{q}, \omega)$. Note that the inclusion of the correlation effect enhances slightly the incommensurate antiferromagnetic fluctuations.

In fig. 2(a) we compare our calculation of the temperature dependence of the uniform spin susceptibility $\chi^{\text{tot}}(0, 0)$ which is measured by the ^{17}O Knight shift [3]. In fig. 2(b) we compare $\text{Im } \chi(\mathbf{Q}_i, \omega)$ with INS data [2]. For the calculation of $\chi(0, 0)$ we approximate $U = 0.177 \text{ eV}$ [19] 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 $\chi(\mathbf{q}, \omega)$. Note that we also

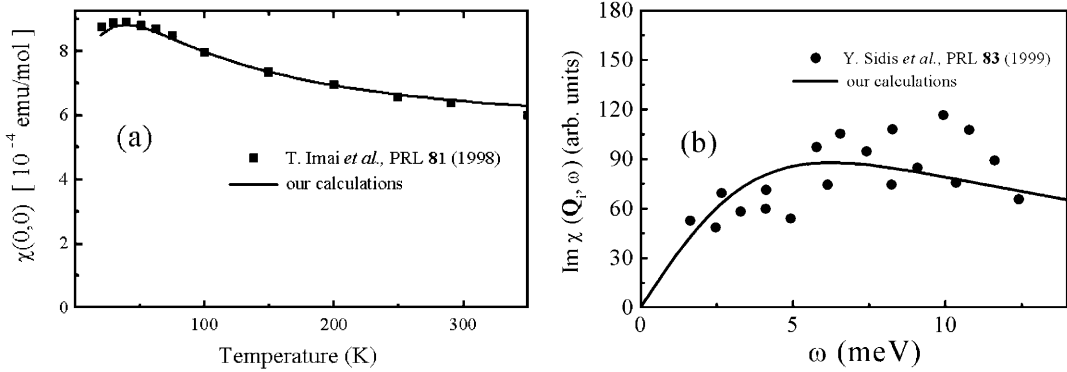


Fig. 2 – (a) The calculated temperature dependence of the uniform spin susceptibility is compared with the ^{17}O Knight shift measurements in the RuO_2 -plane. The peak is due to thermal activation involving γ - and α -, β -bands. (b) Calculated frequency dependence of $\text{Im } \chi(Q_i, \omega)$ compared to INS data.

take into account that there are four electrons per three t_{2g} -bands that would give every χ_0^{tot} an additional weight $4/3$. Our results are in fair agreement with experiments that show a tendency towards ferromagnetism [20]. The maximum in $\chi^{\text{tot}}(0,0)$ at about 25 K results from thermally activated changes in the populations of the bands near E_F . Despite an uncertainty in the INS data fig. 2(b) indicates that our results for $\chi^{\text{tot}}(\mathbf{q}, \omega)$ seem to be a useful basis for further calculations. Note that $\chi^{\text{tot}}(\mathbf{q}, \omega)$ controls the symmetry of the superconducting order parameter. 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 since neighboring Ru spins see also partly a *ferromagnetic* environment.

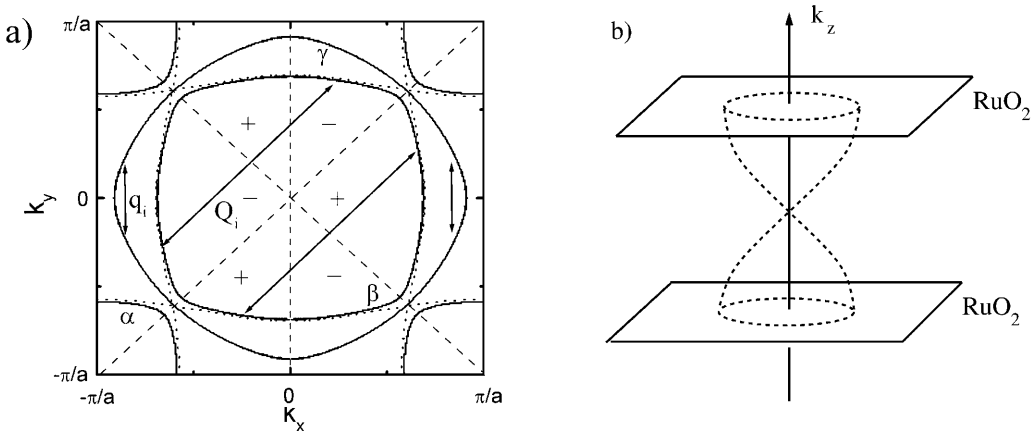


Fig. 3 – (a) Symmetry analysis of the order parameter for the triplet pairing in the first BZ for $k_z = 0$ (solid curves) and $k_z = \pi/2c$ (dotted curves). α , β , and γ denote the FS of the corresponding hybridized bands. The wave vectors Q_i and q_i are the pronounced wave vectors resulting from the susceptibility shown in fig. 1. For $f_{x^2-y^2}$ -wave symmetry the nodes of the real part of the order parameter are shown (dashed lines) and also the regions + (–) where the $f_{x^2-y^2}$ -wave superconducting gap is positive (negative). Note that for the real part of the p -wave order parameter the node occurs along $k_x = 0$. (b) Schematic representation of the k_z -dependence of the f_z -wave order parameter as given by eq. (10). Here, the amplitude of the order parameter in k_z has been drawn in cylindrical coordinates between RuO_2 -planes.

For the analysis of superconductivity in Sr₂RuO₄ we take into account that experiment observes non-*s*-wave symmetry of the order parameter which strongly suggests spin-fluctuation-mediated Cooper pairing. Then by assuming spin-fluctuation-induced pairing it is possible to analyze the symmetry of the superconducting state from the gap equation and our calculated results for $\chi(\mathbf{q}, \omega)$ with the pronounced wave vectors at \mathbf{Q}_i and \mathbf{q}_i . We get, for the gap equation,

$$\Delta_{\mathbf{k}}^i = - \sum_{\mathbf{k}', j} [V_{\sigma}^{\text{eff}}(\mathbf{k}, \mathbf{k}')]^{ij} \frac{\Delta_{\mathbf{k}'}^j}{2E_{\mathbf{k}'}^j} \tanh\left(\frac{E_{\mathbf{k}'}^j}{2k_{\text{B}}T}\right), \quad (4)$$

where $E_{\mathbf{k}}^i = \sqrt{\epsilon_{\mathbf{k}}^{i2} + [\Delta_{\mathbf{k}}^i]^2}$ are the energy dispersions of the bands and the pairing potential $V_{\sigma}^{\text{eff}}(\mathbf{k}, \mathbf{k}')$ is different for singlet ($\sigma = 0$) and triplet ($\sigma = 1$) Cooper pairing. The eigenvalue analysis of eq. (4) will yield the symmetry with lowest energy. We expect a node half-way between the RuO₂-planes, since their superfluid density $n_s \rightarrow 0$, while in the RuO₂-planes nesting will control the formation of the nodes. Hence, we will solve the gap equations in the RuO₂-plane ($k_z = 0$) and also between two neighboring RuO₂-planes ($k_z = \pi/2c$).

For the determination of the pairing potential we follow the analysis by Anderson and Brinkmann for ³He [21] and by Scalapino for the cuprates [22] and use the calculated FS and spin susceptibility for Sr₂RuO₄. For triplet pairing the effective pairing interaction is

$$\begin{aligned} [V_1^{\text{eff}}(\mathbf{k}, \mathbf{k}')]^{ij} &= \\ &= -\frac{U^2}{2} \left(\frac{\chi_0^{ii}(\mathbf{k} - \mathbf{k}', 0)}{1 - U\chi_0^{ii}(\mathbf{k} - \mathbf{k}', 0)} + \frac{\chi_0^{ii}(\mathbf{k} - \mathbf{k}', 0)}{1 + U\chi_0^{ii}(\mathbf{k} - \mathbf{k}', 0)} \right) - \\ &\quad - 2U^2\chi_0^{ij}(\mathbf{k} - \mathbf{k}', 0), \end{aligned} \quad (5)$$

where the last term occurs only for α - and β -bands. For singlet pairing the effective pairing interaction has the form [23]

$$\begin{aligned} [V_0^{\text{eff}}(\mathbf{k}, \mathbf{k}')]^{ij} &= \\ &= \frac{U^2\chi_0^{ii}(\mathbf{k} - \mathbf{k}', 0)}{1 - U\chi_0^{ii}(\mathbf{k} - \mathbf{k}', 0)} + \frac{U^3[\chi_0^{ii}(\mathbf{k} - \mathbf{k}', 0)]^2}{1 - U^2[\chi_0^{ii}(\mathbf{k} - \mathbf{k}', 0)]^2} - \\ &\quad - U^2\chi^{ii}(\mathbf{k} - \mathbf{k}', 0). \end{aligned} \quad (6)$$

Here, the last term corresponds to double-counting exclusion. Note that the cross-terms are small due to the Pauli principle and thus we neglect them for singlet pairing. Using appropriate symmetry representations [9] we discuss the solutions of eq. (4) for the *p*-, *d*-, and two *f*-wave symmetries of the order parameter:

$$\Delta_p(\mathbf{k}) = \Delta_0 \hat{z} (\sin k_x a + i \sin k_y a), \quad (7)$$

$$\Delta_d(\mathbf{k}) = \Delta_0 (\cos k_x a - \cos k_y a), \quad (8)$$

$$\Delta_{f_{x^2-y^2}}(\mathbf{k}) = \Delta_0 \hat{z} (\cos k_x a - \cos k_y a) (\sin k_x a + i \sin k_y a), \quad (9)$$

$$\Delta_{f_z}(\mathbf{k}) = \Delta_0 \hat{z} (\cos k_z c + \text{const}) (\sin k_x a + i \sin k_y a). \quad (10)$$

These symmetries must be substituted into eq. (4). The first three formulae here are projecting onto the RuO₂-planes. Note that the largest eigenvalue in eq. (4) will yield the superconducting symmetry of Δ_l in Sr₂RuO₄. Solving eq. (4) in the first BZ down to 5 K we have found

that p -wave and f_z -wave symmetries are the most stable solutions for the γ -band, while for the α - and β -bands the situation is dependent on the position along the z -direction.

Let us first consider the situation in the RuO₂-plane. One sees that the gap equations for the γ - and α -, β -bands can be separated. Moreover, for the γ -band the p -wave and f_z -wave are the most stable solutions, while for the α - and β -bands the $f_{x^2-y^2}$ -wave symmetry has a largest eigenvalue due to the strong nesting of the α - and β -bands.

Figure 3 characterizes the solutions of eq. (4). In a good approximation we linearize eq. (4) in Δ_i^i , *i.e.* $E_{\mathbf{k}'}^i \rightarrow \epsilon_{\mathbf{k}'}^i$, and safely put $\tanh(\epsilon_{\mathbf{k}'}^i/2k_B T) = 1$. Therefore, the main contribution to the pairing comes from the Fermi levels. We present our results for the Fermi surface in the RuO₂-plane ($k_z = 0$) and between the neighboring RuO₂-planes ($k_z = \pi/2c$). The wave vectors \mathbf{Q}_i and \mathbf{q}_i refer to the peaks in $\chi(\mathbf{q}, \omega)$. The areas with $\Delta_{f_{x^2-y^2}} > 0$ and $\Delta_{f_{x^2-y^2}} < 0$ are denoted by (+) and (-), respectively. Note that the minus sign in eq. (4) is cancelled for triplet pairing. The summation over \mathbf{k}' in the first BZ is dominated by the contributions due to \mathbf{Q}_i for the α - and β -bands and the one due to \mathbf{q}_i for the γ -band. As can be seen from fig. 3(a) in the case of $f_{x^2-y^2}$ -wave symmetry for the γ -band the wave vector \mathbf{q}_i bridges the same number of portions of the FS with opposite and equal sign. Therefore, $f_{x^2-y^2}$ has no solution in the γ -band. On the other hand, we also see from fig. 3(a) that \mathbf{Q}_i bridges portions of the FS with *equal* signs of the $f_{x^2-y^2}$ -superconducting order parameter for the α - and β -band. Moreover, the eigenvalue of this order parameter is also enhanced due the inter-band nesting effect between α - and β -bands. Thus, superconductivity in the RuO₂-plane is indeed possible yielding an $f_{x^2-y^2}$ -wave order parameter in the α - and β -band. At the same time, the γ -band in the RuO₂-plane has a stable solution for p -wave pairing or f_z -pairing which are the same in the RuO₂-plane meaning no nodes in the RuO₂-planes. However, since we did not reach $\lambda = 1$ in the eigenvalue analysis, we cannot conclude presently which of the order parameter gives a lower energy in the RuO₂-plane.

Let us also consider the solution of the gap equation (4) between neighboring RuO₂-planes ($k_z = \pi/2c$). In this case the hybridization between α - and β -bands is almost zero and the corresponding Fermi surface is changing towards the LDA dispersion [19] as shown in fig. 3(a). Most importantly, triplet pairing involving different bands does not contribute. The cross-susceptibility between α - and β -bands is zero. Therefore, the eigenvalue for α - and β -bands lowers due to the decreased nesting effects. Moreover, due to the superfluid density $n_s \rightarrow 0$, one expects that $\Delta_0 \rightarrow 0$ also, as illustrated in fig. 3(b). This also seems in agreement with experimental observation [11].

Also with similar arguments we can rule out singlet pairing using eq. (6). In particular, assuming $d_{x^2-y^2}$ -symmetry for Sr₂RuO₄ eq. (4) yields no $d_{x^2-y^2}$ -symmetry. Its eigenvalue is lower than in the case of triplet pairing. This is plausible as can be seen as follows using fig. 3(a). Note that we get a change of sign of the order parameter upon crossing the diagonals of the BZ. According to eq. (4), wave vectors around \mathbf{Q}_i connecting areas (+) and (-) contribute constructively to the pairing. Contributions due to \mathbf{q}_i and the background connecting the same sign areas subtract from the pairing (see fig. 3(a) with nodes at the diagonals for illustration). Therefore, we get that the four contributions due to \mathbf{q}_i in the γ -band do not allow to have $d_{x^2-y^2}$ -wave symmetry in the γ -band. Moreover, despite the pair-building contribution due to \mathbf{Q}_i , one gets that the eigenvalue of the $d_{x^2-y^2}$ -wave symmetry in the γ -band is smaller than for the $f_{x^2-y^2}$ -wave symmetry. This is due to the large contribution from \mathbf{Q}_i to the cross-terms for the triplet pairing which are absent for the singlet pairing. 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 summary, taking into account cross-susceptibility between α - and β -bands we successfully explain the ^{17}O Knight shift and INS data. Most importantly, we calculate $\chi(\mathbf{q}, \omega)$ and show on the basis of the Fermi surface topology and the calculated spin susceptibility $\chi(\mathbf{q}, \omega)$ that triplet pairing is present in Sr_2RuO_4 . To decide whether p - or f -wave symmetry pairing is present one needs to perform more complete calculations including spin-orbit coupling effects for example. In contrast to previous study, we find from fig. 1 that all bands (α, β, γ) are important and contribute to superconductivity. Triplet pairing should give a rich phase diagram in the presence of a magnetic field and spin-orbit coupling.

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