Interlayer Coupling and p-wave Pairing in Strontium Ruthenate

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On the basis of a three orbital model and an effective attractive interaction between electrons we investigate the possible superconducting states, with p and f-wave internal symmetry, of Sr2RuO4. For an orbital dependent interaction which acts between in plane and out of plane nearest neighbor Ruthenium atoms we find a state for which the gap in the quasi-particle spectra has a line node on the α and β sheets of the Fermi Surface, but it is complex with no nodes on the γ-sheet. We show that this state is consistent with all the available experimental data. In particular, we present the results of our calculations of the specific heat and penetration depth as functions of the temperature.

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Ever since triplet pairing was discovered in superfluid 3He [1], during the early seventies, there has been a constant search for the superconducting analogue of this intriguing macroscopic quantum phenomenon. Although, as yet, there is no metal for which triplet paring has been definitively demonstrated there are a number of good candidates. The evidence that Sr2RuO4 is a triplet superconductor is particularly strong [2,3]. Nevertheless, even in this case the full symmetry of the equilibrium state below Tc remains open to debate [2,12].

One of the puzzles currently at the centre of attention is the apparent incompatibility between experimental evidence for broken time-reversal symmetry in the superconducting state [13,14] and equally convincing measurements indicating that the order parameter d(k) has a line of nodes on the Fermi Surface [15]. The reason why this state of affairs represents a dilemma is that for all odd parity spin triplet pairing states in tetragonal crystals, group theory does not require the simultaneous presence of both broken time-reversal symmetry and line nodes [14]. Consequently, due to their lower condensation energy, line nodes are unlikely. For instance, the pairing state d(k) ~ (kx + iky)ez, proposed by Agterberg et al. [4] on the grounds that it minimizes the free energy, obviously breaks time reversal invariance and has no line nodes. Of course, such states as d(k) ~ (kx + iky)(kxky)ez discussed by Graf and Balatsky [5] and other f-wave states [3,6,11] are allowed by symmetry considerations but, the point is that, the nodes are not required by symmetry and hence it is not very attractive ansatz to build a theory on. Under these circumstances it is more advantageous to study physically motivated microscopic models even if the question of the actual mechanism of pairing is to be avoided. In this letter we propose and investigate one such physically motivated model.

Our model is prompted by the observation of Hasegawa et al. [1], that coupling between the Ruthenium layers in Sr2RuO4 leads to convenient, horizontal, kz = ±π/4 lines of zeros on the Fermi Surface. It features two, intra and inter plane, interaction constants U∥ and U⊥ respectively, which describe the attraction between electrons each occupying one of three, t2g, orbitals on Ru atoms which are nearest neighbours either in plane and out of plane. As we shall show, this simple physical picture yields horizontal nodes on the α, β sheets of the Fermi Surface whilst the γ-sheet is fully gapped with d(k) ~ (sin kx + i sin ky)ez, in quantitative as well as qualitative agreement with a number of experiments.

Hasegawa et al. [1], treated the case of a single band only and, unlike us, they made no quantitative contact with experiments. The more recent work of Zhitomirsky and Rice [12] is closer to ours, although dealing with a two band model only. They also couple electrons on different layers and find line nodes on the α, β sheets but a gap on γ. However, the physics behind their model, as well as its consequences, are quite different from ours. Largely, this is due to a difference in strategy. In a multi-band BCS like model, with different coupling constants for each band, one generically finds multiple phase transitions as the different sheets of the Fermi Surface are gaped on lowering the temperature. Since experimentally there is only one jump in the specific heat, at Tc = 1.5K, in constructing a sensible model one must tailor it to eliminate such double transitions. Zhitomirsky and Rice [12] chose to couple the order parameters on different sheets of the Fermi Surface. This hybridized the different order parameters and led to a single transition. They referred to such hybridization as an inter-band proximity effect. We, on the other hand, adjusted the two coupling constants, U∥ and U⊥, so that the transition on the α, β sheets would occur at more or less the same
temperature as as on $\gamma$. Somewhat surprisingly these two approaches imply different physics. The proximity model requires a three point interaction to mix the in-plane and out of plane Cooper pairs whilst our “local bond” model is a strictly a two point effect. Moreover, the “local bond” model has fewer free parameters by construction and therefore, as will be shown below, is more readily compared with experiments.

To describe the superconducting state we employ a simple multi-band attractive Hubbard model:

$$\hat{H} = \sum_{ijm'm',\sigma} ((\varepsilon_m - \mu) \delta_{ij} \delta_{mm'} - t_{mm'}(ij)) \hat{c}_{i\sigma}^+ \hat{c}_{j\sigma'}$$

$$-\frac{1}{2} \sum_{ijmm'\sigma\sigma'} U_{mm'}^\sigma(\sigma')(ij) \hat{n}_{i\sigma} \hat{n}_{j\sigma'}$$

(1)

where $m$ and $m'$ refer to the three Ruthenium $t_{2g}$ orbitals $a = xz, b = yz$ and $c = xy$ and $i$ and $j$ label the sites of a body centered tetragonal lattice. The hopping integrals $t_{mm'}(ij)$ and site energies $\varepsilon_m$ were fitted to reproduce the experimentally determined Fermi Surface. The set of interaction constants $U_{mm'}^\sigma(\sigma')(ij)$ describe attraction between electrons on nearest neighbour sites with spins $\sigma$ and $\sigma'$ and in orbitals $m$ and $m'$. Thus our actual calculations consist of solving, self-consistently, the following Bogoliubov-de Gennes equation:

$$\sum_{jm'\sigma'} \left( E^\nu - H_{mm'}(ij) \right) \Delta_{mm'}^\sigma(\sigma')(ij) \left( u_{jm'\sigma'}^\nu \right) = 0 ,$$

(2)

where $H_{mm'}(ij)$ is the normal spin independent part of the Hamiltonian, and the $\Delta_{mm'}^\sigma(\sigma')(ij)$ is self consistently given in terms of the pairing amplitude, or order parameter, $\chi_{mm'}^\sigma(\sigma')(ij)$,

$$\Delta_{mm'}^\sigma(\sigma')(ij) = U_{mm'}^\sigma(\sigma')(ij) \chi_{mm'}^\sigma(\sigma')(ij) .$$

(3)

defined by the usual relation

$$\chi_{mm'}^\sigma(\sigma')(ij) = \sum_{\nu} u_{im\sigma}^\nu v_{jm'\sigma'}^\nu (1 - 2f(E^\nu)),$$

(4)

where $\nu$ enumerates the solutions of Eq. 3.

We solved the above system of Bogoliubov de Gennes equations including all three bands and the experimental three dimensional Fermi surface. We assumed that the pairing interaction $U_{mm'}^\sigma(\sigma')(ij)$ for nearest neighbours in plane is only acting for the $c$ ($d_{xy}$) Ru orbitals. We further assumed that the nearest neighbour inter plane interaction acts only in $a$ and $b$ orbitals ($d_{xz}$, $d_{yz}$). The motivation for this is that the dominant hopping integrals in plane are between $c$ orbitals, and the largest out of plane hopping integrals are for $a$ and $b$. Therefore we have only two coupling constants $U_{aa}$ and $U_{bb}$ describing these two physically different interactions. Our strategy is to adjust these phenomenological parameters in order to obtain one transition at the experimentally determined $T_c$. Thus, beyond fitting $T_c$, there are no further adjustable parameters, and one can compare directly the calculated physical properties of the superconducting states to those experimentally observed. Consequently, if one obtains a good overall agreement one can say that one has empirically determined the form of the pairing interaction in a physically transparent manner.

![FIG. 1. Order parameters, $|\Delta_{x}^a|$, $|\Delta_{y}^a|$, $|\Delta_{x}^b|$, $|\Delta_{y}^b|$ as functions of temperature, (dashed lines), and excluding $z$ and $f$ (full lines).](image)

Because the pairing interactions $U_{mm'}^\sigma(\sigma')(ij)$ were assumed to act only for nearest neighbour sites in or out of plane, the pairing potential $\Delta_{mm'}^\sigma(\sigma')(ij)$ is also restricted to nearest neighbours. We further focus on only odd parity (spin triplet) pairing states for which the vector $d \sim (0,0,d^2)$, i.e. $\Delta_{mm'}^{\uparrow\downarrow}(ij) = \Delta_{mm'}^{\downarrow\uparrow}(ij)$, and $\Delta_{mm'}^{\uparrow\downarrow}(ij) = \Delta_{mm'}^{\downarrow\uparrow}(ij) = 0$. Therefore in general we have the following non-zero order parameters (i) for in plane bonds: $\Delta_{cc}(\hat{e}_x), \Delta_{cc}(\hat{e}_y)$, and (ii) for inter-plane bonds: $\Delta_{ab}(\hat{R}_{ij}), \Delta_{ab}(\hat{R}_{ij})$, $\Delta_{ab}(\hat{R}_{ij})$ for $\hat{R}_{ij} = (\pm a/2, \pm a/2, 0)$.

Taking the lattice Fourier transform of Eq. 3 the corresponding pairing potentials in k-space have the general form (suppressing the spin indices for clarity):

$$\Delta_{cc}(\mathbf{k}) = \Delta_{xx}^c \sin k_x + \Delta_{yy}^c \sin k_y$$

(5)

for c orbitals and,

$$\Delta_{mm'}(\mathbf{k}) = \Delta_{mm'}^z \sin \frac{k_z}{2} \cos \frac{k_x}{2} \cos \frac{k_y}{2}$$

$$+ \Delta_{mm'}^f \sin \frac{k_x}{2} \sin \frac{k_y}{2} \sin \frac{k_c}{2}$$

$$+ \left( \Delta_{mm'}^x \sin \frac{k_x}{2} \cos \frac{k_y}{2} + \Delta_{mm'}^y \sin \frac{k_y}{2} \cos \frac{k_x}{2} \right) \cos \frac{k_z}{2}$$

(6)

for $m, m' = a, b$. Note that beyond the usual p-wave symmetry of the $\sin k_x$ and $\sin k_y$ type for the c orbitals, we include all three additional p-wave symmetries of the $\sin k/2$ type which are induced by the effective attractive
interactions between carriers on the neighboring out-of-plane Ru orbitals. These interactions are also responsible for the f-wave symmetry order parameters, $\Delta_{mm'}$, transforming as $B_{1u}$ in the notation of [16]. This latter is symmetry distinct from all p-wave order parameters in a tetragonal crystal, unlike some other f-wave states which have been proposed [3,4]. The $p_z$ order parameters $\Delta_{\nu_{\nu'}}$ are of $A_{2u}$ symmetry. In contrast the pairs $\Delta_{\nu_{\nu'}}$, $\Delta_{\nu_{\nu'}}'$ for $m, m' = a, b$ are of the same $E_u$ symmetry as $\Delta_{\nu_{\nu'}}$, $\Delta_{\nu_{\nu'}}$. In general the order parameters in each distinct irreducible representations will have different transition temperatures.

Using the above quasi-particle spectra we have calculated the specific heat, shown in Fig. 3. Remarkably, although we fitted to $T_c$ only, the specific heat jump at $T_c$ is $27mJ/mol$, in essentially exact agreement with the experiment [14]. Thus, contrary to the argument given by Zhitomirsky and Rice [13], it is not necessary to assume a gap opening up on the active $\gamma$ sheet only in order to obtain the correct discontinuity at $T_c$. Furthermore the calculated specific heat also follows the experiment closely at lower temperatures, except for the second transition at about $0.3T_c$. Clearly from Fig. 3 this is due to the f-wave and $p_z$ pairing, which are symmetry decoupled from the $p_x, p_y$ states, and is not consistent with the experiments [14]. However, if we suppress the f-wave and $p_z$ pairing then the specific heat agrees perfectly down to the lowest temperatures. We believe that this is physically reasonable, because the f-wave states will be more strongly influenced by impurities than the p-wave states, due to a $(2l+1)$ prefactor in the impurity pair breaking parameter [19]. As can be seen in Fig. 3, the $x, y$ $aa$ and $cc$ order parameters are only slightly affected by the presence of the f-wave gap.

The low temperature limit of the specific heat is power law, because our gap parameters have line nodes. These are horizontal circles around the cylindrical $\alpha$ and $\beta$ Fermi surface sheets, as illustrated in Fig. 2, while the $\gamma$ sheet is node-less. As can be seen, when the f-wave order parameter also becomes finite, the line nodes disappear. The fact that the slope of specific heat at low temperatures (without f) agrees quantitatively with the experiments suggests that these horizontal nodes are only present on $\alpha$, $\beta$, as in our model.

A further independent test of our model is the calculation of the superfluid density [3] shown in Fig. 4. Again there is excellent agreement over the whole temperature range between $T_c$ and zero. Some physical insight into the different contributions to the superfluid density can be found by setting $\Delta_{\nu_{\nu'}}$ to zero for $m = m' = c$ or $mm' = a, b$ and repeating the calculation. The $c$ orbital only contribution gives about 70% of the zero temperature superfluid density, with the remaining 30% derives from the $a, b$ orbital order parameters. One can see that the finite slope at zero temperature derives only from $a, b$ contributions, consistent with the the line nodes on the $\alpha, \beta$ Fermi surfaces. The only disagreement is that the absolute magnitude of our calculated zero temperature x-y plane penetration depth is only 450Å compared to 1900Å determined experimentally [24,25]. This discrepancy may be due to impurities (the experimental samples had $T_c \sim 1.1-1.3K$) or to non-local electrodynamic effects associated with the line nodes [26]. We have also calculated the temperature dependent zero-field thermal conductivity, which is also in good qualitative agreement.
with the experiments of Izawa et al. [23].

In summary we would like to emphasize two points. Firstly, we have proposed an alternative to the the ‘intra-band proximity effect’ model of Zhitomirsky and Rice [12] for describing horizontal line nodes on the α, β sheets of the Fermi Surface in superconducting Sr$_2$RuO$_4$. Our bond model differs from theirs in the way the inter-layer coupling is implemented. Our description is a real space, two point tight-binding interaction such as naturally arises in any multi-band, extended, negative $U$ Hubbard model, Eq. (1). To be quite clear about this matter we recall that a generic pair-wise interaction like $U(r,r')$, when expressed in the language of a tight-binding model Hamiltonian will, in general, give rise to four point interaction parameters $U_{ij,kl}$. The original Hubbard Hamiltonian makes use of the one point parameters $U^{(1)}_{ij} = U_{ii,ij}$ whilst the extended Hubbard model is based on two point parameters $U^{(2)}_{ij} = U_{ij,ij}$. Evidently our ‘bond’ model is a negative U-version of the latter [23]. On the other hand the ‘proximity effect model’ [12] corresponds to using $U^{(3)}_{ij}$. The physics of this is often referred to as assisted hopping [23]. If one assumes, as is normally the case in an isotropic substance, that $U^{(1)} > U^{(2)} > U^{(3)} > U^{(4)}$ then the ‘bonds’ represent stronger coupling than assisted hopping and should be the preferred coupling mechanism. However, for the tetragonal arrangement of Ru atoms in Sr$_2$RuO$_4$ this is no more than a suggestion at present. Thus the relative merits of the two models will eventually be settled by appeal to experiments.

Secondly, we wish to stress that in our ‘bond’ approach to the problem the parameters which describe the normal state are determined by fitting to the very accurately known Fermi Surface and the measured $T_c$ determines both coupling constants $U_{||} =$ 40meV, $U_{\perp} =$ 48meV. Thus, the calculated specific heat, Fig. 3, and superfluid density, Fig. 4, are parameter free quantitative predictions of the theory. Consequently, their good agreement with experiments can be construed as strong support for the physical picture represented by our model. Interestingly a significant feature of this is that the amplitude of the gap function, and correspondingly the superfluid density, on the α, β sheets of the Fermi Surface are comparable to that on the γ sheet. This is unlike the case suggested by the ‘inter-band proximity effect’ picture where γ is the active band and α, β play a passive role. Hopefully, experiments will soon clarify which of the two situations prevails. As for the physical mechanism of pairing, the fact that $U_{\perp} \approx U_{||}$ implies that the pairing interaction is fairly isotropic in spite of the layered structure.

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\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure4.png}
\caption{Superfluid density as a function of $T$ (solid line), and experimental points from Sample 1 of Bonalde et al.\cite{15}. The relative contributions of $c$ and $a,b$ order parameters are indicated (dashed lines).}
\end{figure}

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