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 symmetries, of Sr_2RuO_4. 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 quasiparticle spectra has nodes on the α and β sheets of the Fermi surface, but which is complex with no nodes on the γ sheet. We show that this state is consistent with the available experimental data. In particular, we present the results of our calculations of the specific heat and penetration depth as functions of temperature.

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Ever since triplet pairing was discovered in superfluid ^{3}He, there has been a constant search for the superconducting analogue. Although, as yet, there is no metal for which triplet pairing has been definitively demonstrated, there are a number of good candidates. The evidence that Sr_2RuO_4 is a triplet superconductor is particularly strong.[1,2] Nevertheless, even in this case the full symmetry of the equilibrium state below T_c remains open to debate.[1–11]

One of the puzzles currently at the center of attention is the apparent incompatibility between experimental evidence for broken time-reversal symmetry in the superconducting state[9,12] and equally convincing measurements indicating that the order parameter $\mathbf{d}(\mathbf{k})$ has a line of nodes on the Fermi surface.[13,14] This presents a dilemma because 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.[15] Consequently, due to their lower condensation energy, line nodes are unlikely. 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 paper we propose and investigate a family of such physically motivated models.

Our models are prompted by the observation, of Hasegawa *et al.*,[10] that coupling between the ruthenium layers in Sr_2RuO_4 leads to convenient, horizontal, $k_z=\pm\pi/c$, lines of zeros on the Fermi surface. A particularly simple example of such models features two, intraplane and interplane, interaction constants, $U_\parallel$ and $U_\perp$ respectively. They describe the attraction between electrons each occupying one of three t_{2g} orbitals on Ru atoms which are nearest neighbors either in plane or out of plane. As we shall show, this simple physical picture yields nodes on the α and β sheets of the Fermi surface, while the γ sheet is fully gapped with $\mathbf{d}(\mathbf{k}) \sim (\sin k_x + i \sin k_y)\hat{\mathbf{e}}_z$, in quantitative as well as qualitative agreement with a number of experiments.

Hasegawa *et al.*[10] treated the case of a single band only and, unlike us, they made no quantitative contact with experiments. More recently Zhitomirsky and Rice[11] proposed a two band model and showed that its parameters can be chosen so that it reproduces the experimental specific heat data.[13] They coupled electrons on different layers and found line nodes on one sheet but a gap on the other.

We take the matter further by describing the experimentally observed three sheets of the Fermi surface realistically and calculate more experimental observables. However, our strategy is very different. In a multiband 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 gapped, successively, on lowering the temperature.[16] Since experimentally there is only one jump in the specific heat for Sr_2RuO_4, at $T_c=1.5$ K, in constructing a viable model one must tailor the form of the attractive interaction to eliminate such multiple transitions. Zhitomirsky and Rice[11] 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 interband proximity effect. We, on the other hand, build our models in terms of real space bonds between electrons, with spins σ and σ', occupying specific orbitals labeled by m and m', centered on specific sites i and j. These are described by the interaction constants $U^{\sigma,\sigma'}_{m,m'}(ij)$. We have investigated a number of such models and found that two non zero coupling constants: $U_\parallel$ and $U_\perp$ form a useful minimal set. Then we adjusted these values so that the transitions on the α and β sheets would occur at more or less the same temperature, $T_c \sim 1.5$ K, as on γ. 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.

To describe the superconducting state we employ a simple multiband attractive Hubbard model,

$$\hat{H}=\sum_{ijmm',\sigma}[(\varepsilon_m-\mu)\delta_{ij}\delta_{mm'}-t_{mm'}(ij)]\hat{c}^+_{im\sigma}\hat{c}_{jm'\sigma} - \frac{1}{2}\sum_{ijmm'\sigma\sigma'}U^{\sigma\sigma'}_{mm'}(ij)\hat{n}_{im\sigma}\hat{n}_{jm'\sigma'}, \quad (1)$$

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 ε_m were fitted to reproduce the experimentally determined three-dimensional Fermi surface.[17] For this Hamiltonian we solve, self-consistently, the Bogoliubov–de Gennes equation

$$\sum_{jm'\sigma'} \begin{pmatrix} E^\nu \delta_{ij} - H_{mm'}(ij) & \Delta^{\sigma\sigma'}_{m,m'}(ij) \\ \Delta^{*\sigma\sigma'}_{mm'}(ij) & E^\nu \delta_{ij} + H_{mm'}(ij) \end{pmatrix} \begin{pmatrix} u^\nu_{jm'\sigma'} \\ v^\nu_{jm'\sigma'} \end{pmatrix} = 0, \tag{2}$$

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

$$\Delta^{\sigma\sigma'}_{mm'}(ij) = U^{\sigma\sigma'}_{mm'}(ij)\chi^{\sigma\sigma'}_{mm'}(ij). \tag{3}$$

Furthermore, as usual,

$$\chi^{\sigma\sigma'}_{mm'}(ij) = \sum_\nu u^\nu_{im\sigma} v^{\nu *}_{jm'\sigma'}[1-2f(E^\nu)], \tag{4}$$

where ν enumerates the solutions of Eq. (2).

In short, the hopping integrals and site energies for all three bands, which constitute $H_{mm'}(i,j)$, are fixed by experiments in the normal state and we adjust the interaction constants $U^{\sigma\sigma'}_{mm'}(ij)$ to reproduce measurements of superconducting properties. We have investigated many sets of U's, but here we report only on a particularly interesting, minimal nonzero, set. It consists of the ''out of plane'' nearest neigbour interaction $U^{\sigma\sigma'}_{aa}(ij) = U^{\sigma\sigma'}_{bb}(ij) = U_\perp$ and the in plane nearest neighbor interaction $U^{\sigma\sigma'}_{cc}(ij) = U_\parallel$. The need to avoid two transitions and to reproduce the measured T_c determines both $U_\perp$ and $U_\parallel$ (with values of 48 and 40 meV respectively). All further consequences of our calculations are quantitative predictions of the model. The remarkable agreement of such predictions with a number of experiments is the principal result of this paper.

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

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

$$\Delta_{cc}(\mathbf{k}) = \Delta^x_{cc} \sin k_x + \Delta^y_{cc} \sin k_y \tag{5}$$

for c orbitals, and

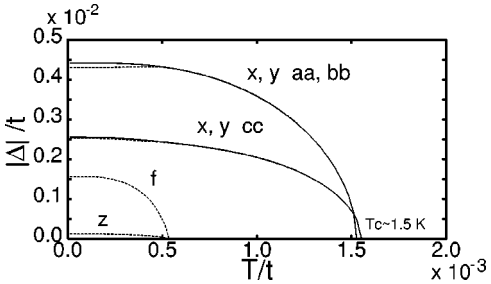

FIG. 1. Order parameters $|\Delta^x_{aa}|$, $|\Delta^x_{cc}|$, $|\Delta^z_{aa}|$, and $|\Delta^f_{aa}|$ as functions of temperature (dashed lines); and excluding z and f (full lines); $t \approx 0.08$ eV.

$$\Delta_{mm'}(\mathbf{k}) = \Delta^z_{mm'} \sin\frac{k_z c}{2}\cos\frac{k_x}{2}\cos\frac{k_y}{2} + \Delta^f_{mm'} \sin\frac{k_x}{2}\sin\frac{k_y}{2}\sin\frac{k_z c}{2} + \left(\Delta^x_{mm'} \sin\frac{k_x}{2}\cos\frac{k_y}{2} + \Delta^y_{mm'} \sin\frac{k_y}{2}\cos\frac{k_y}{2} \right) \cos\frac{k_z c}{2} \tag{6}$$

for $m,m' = a,b$. Note that beyond the usual p-wave symmetry of the $\sin k_i$ type for the c orbitals, we include all three additional p-wave symmetries of the $\sin(k_i/2)$ 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, $\Delta^f_{mm'}$, transforming as B_{1u}.[15] 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.[6–8] The p_z order parameters $\Delta^z_{mm'}$ are of A_{2u} symmetry. In contrast the pairs $\Delta^x_{mm'}, \Delta^y_{mm'}$ for $m,m' = a,b$ are of the same E_u symmetry as $\Delta^x_{cc}, \Delta^y_{cc}$. In general the order parameters transforming according to distinct irreducible representations will have different transition temperatures. Surprisingly, even though they have the same symmetry, the fact that Δ^x_{cc}, Δ^y_{cc} and $\Delta^x_{m,m'}$, $\Delta^y_{m,m'}$ go to zero at $T_c \sim 1.5$ K determines both $U_\perp$ and $U_\parallel$. This is because in the gap equation they are not coupled by any two-point Hubbard interaction of the form included in Eq. (1). Coupling only arises via the ''interband proximity effect,''[11] as discussed in detail below.

Figure 1 shows the calculated order parameters as a function of temperature. Order parameters transforming according to distinct irreducible representations have different transition temperatures, and so the f-wave and p_z components have transition temperatures at T^f_c and T^z_c below T_c. Above T^f_c but below T_c the order parameters have the symmetries $\Delta^y_{cc} = i\Delta^x_{cc}$, $\Delta^y_{bb} = i\Delta^x_{aa}$ as for[3] $(k_x + ik_y)\hat{\mathbf{e}}_z$ corresponding to the time reversal broken pairing state of ^3He-A. The off-diagonal components, such as Δ^x_{ab} are small but nonzero, as are Δ^x_{bb} and Δ^y_{aa}. It should be stressed that the k-space pairing potentials $\Delta_{mm'}(\mathbf{k})$ do not directly correspond to the energy gaps on the Fermi surface sheets, shown in Fig. 2, because, in general, the tight-binding Hamiltonian is nondiagonal in the orbital indices.

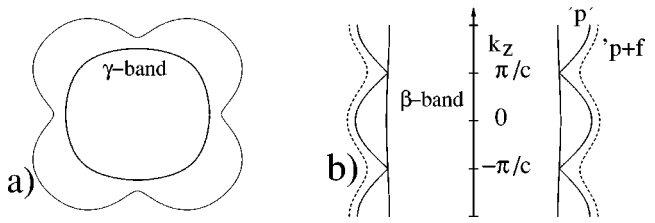

FIG. 2. Lowest energy quasiparticle eigenvalues $E^{\nu}(\mathbf{k})$ on the Fermi surface, (a) Polar plot of the γ sheet in the plane $k_z=0$; $E_{\gamma,max}(\mathbf{k}_F)=0.22$ meV and $E_{\gamma,min}(\mathbf{k}_F)=0.056$ meV. (b) Vertical cross-section of the cylindrical β sheet in the plane $k_x=k_y$; $E_{\beta,max}(\mathbf{k}_F)=0.32$ meV. A nonzero *f*-wave order parameter (dotted line) lifts the *p*-wave line nodes (solid lines).

Using the above quasiparticle 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 27 mJ/mol, in essentially exact agreement with the experiment.[13] Given that this value does not correspond to the BCS result $\Delta C/C=1.41$ this fact strongly supports the two parameter, $U_\perp$ and $U_\parallel$, model we are investigating. At low temperatures the full solution corresponds to the dotted curve. The peak at $0.3T_c$ is the reflection of the *f* component of Δ becoming nonzero and is clearly inconsistent with the experiments. Evidently, this unwanted second transition could be eliminated by introducing further interaction constants. Instead of doing that we remove this peak by setting the *f* and *z* components of Δ to zero in each step of the iterative procedures. This can be readily justified by invoking a small concentration of impurities described by the scattering rate $1/\tau$. We have checked by explicit calculations, based Abrikosov-Gorkov theory,[16] that such weak disorder will suppress $\Delta^f_{m,m'}$ without much influence on the other components. The details of these calculations will be published elsewhere.[18] We have also found that the predictions of the above ''two *U* model'' are remarkably stable to introduction of further interaction constants such as $U_\parallel$ between the *a* and *b* orbitals. Such terms do not eliminate the line node but shift its position in k_z.

As can be seen in Fig. 1 the *x*,*y* *a*, and *c* order parameters are only slightly affected by the presence of the *f*-wave gap. Moreover, as shown by the full curve in Fig. 3, the calculation without the *f* and *z* components reproduces the experimental result very accurately. Note that the low temperature limit of the specific heat is power law because, without *f*, our energy gap has line nodes. These are a horizontal circle around the cylindrical β Fermi surface sheet, as illustrated in Fig. 2, and eight point nodes on the α Fermi surface.[18] The γ sheet is nodeless. 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 nodes are only present on α and β, as in our model.

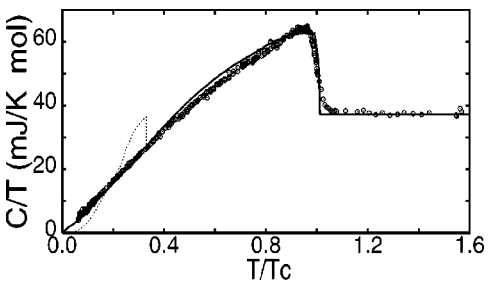

FIG. 3. Calculated specific heat *C* as a function of temperature *T*, compared to the experimental data of NishiZaki *et al.* (Ref. 13). The dotted line includes the *f*-wave order parameter, and the solid line is calculated after setting the *f*-wave gap parameter to zero.

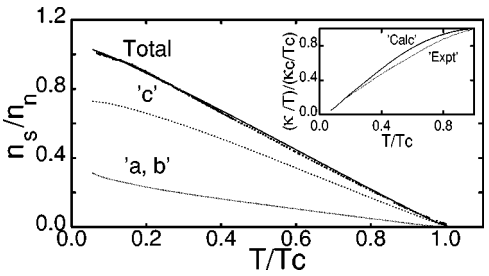

FIG. 4. Superfluid density as a function of T (solid line), and experimental points from Sample 1 of Bonalde *et al.* (Ref. 14). The relative contributions of *c* and *a* and *b* order parameters are indicated (dashed lines). The inset shows thermal conductivity compared to the experimental data of Izawa *et al.* (Ref. 22).

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 $\Delta^{x,y}_{mm'}$ to zero for $m=m'=c$ or $m,m'=a,b$ and repeating the calculation. The *c* orbital only contribution gives about 70% of the zero temperature superfluid density, with the remaining 30% derived from the *a* and *b* orbital order parameters. One can see that the finite slope at zero temperature derives only from *a*,*b* contributions, consistent with the line nodes on the α and β Fermi surfaces. The calculations agree well with the experiments[14] at all temperatures, even though there is no T^2 behavior in either the *a*, *b*, or *c* superfluid contributions. The only disagreement is that the absolute magnitude of our calculated zero temperature *x*-*y* plane penetration depth is only 450 Å compared to the 1900 Å determined experimentally.[19,20] This discrepancy may be due to non-local electrodynamic effects associated with the line nodes.[21] We have also calculated the temperature dependent zero-field thermal conductivity, which, as shown in the inset of Fig. 4, is also in good quantitative as well as qualitative agreement with the experiments of Izawa *et al.*[22]

In summary we would like to emphasize two points. First, we have proposed an alternative to the ''intraband proximity effect'' model of Zhitomirsky and Rice[11] for describing horizontal line nodes on the β sheet of the Fermi surface in superconducting Sr_2RuO_4. Our bond model differs from theirs in the way the interlayer coupling is implemented. Our description is a real space, two point tight-binding interaction such as naturally arises in any multiband, extended, negative-*U* Hubbard model [Eq. (1)]. To be quite clear about this matter we recall that a generic pairwise interaction like $U(\mathbf{r},\mathbf{r}')$, 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 $U_i^{(1)}=U_{ii,ii}$ while the extended Hubbard model is based on two point parameters $U_{i,j}^{(2)}=U_{ij,ij}$. Evidently our ''bond'' model is a negative-U version of the latter.[23] On the other hand, the ''proximity effect model''[11] corresponds to using $U_{i,j,l}^{(3)}=U_{ij,lj}$. The physics of this is often referred to as assisted hopping.[24] 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 a stronger coupling than assisted hopping and should be the preferred coupling mechanism. However, for the tetragonal arrangement of Ru atoms in Sr_2RuO_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 further experiments. Second, 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 $U_\parallel$ and $U_\perp$. Thus the calculated specific heat (Fig. 3), the superfluid density, and the thermal conductivity (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 the superfluid density, on the α and β sheets of the Fermi surface are comparable to that on the γ sheet. This is unlike the case suggested by the ''interband proximity effect'' picture[11] where γ is the active band and α and β play a passive role. A more detailed comparison between the present model and that of Zhitomirsky and Rice is presented elsewhere.[25] Finally, we note that since in our two parameter model the possibility of two transitions has been eliminated by fixing the relative sizes of $U_\perp$ and $U_\parallel$ we are forced to predict that uniaxial pressure, which presumably affects $U_\perp$ differently from $U_\parallel$, could split the transition.

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