Spin-Orbit Coupling and Symmetry of the Order Parameter in Strontium Ruthenate

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(Dated: November 21, 2018)

Determination of the orbital symmetry of a state in spin triplet Sr$_2$RuO$_4$ superconductor is a challenge of considerable importance. Most of the experiments show that the chiral state of the $\pm (k_x \pm i k_y)$ type is realized and remains stable on lowering the temperature. Here we have studied the stability of various superconducting states of Sr$_2$RuO$_4$ in the presence of spin-orbit coupling. Numerically we found that the chiral state is never the minimum energy. Alone among the five states studied it has $\langle \mathbf{L}\mathbf{S} \rangle = 0$ and is therefore not affected to linear order in the coupling parameter $\lambda$. We found that stability of the chiral state requires spin dependent pairing interactions. This imposes strong constraint on the pairing mechanism.

Despite 10 years of intensive research [1], the mechanism of superconductivity in strontium ruthenate Sr$_2$RuO$_4$ is still not clarified [2, 3]. A necessary first step in identifying the effective interactions responsible for the superconductivity is to fully identify the symmetry of the pairing state and to explain the various relevant experimental properties below $T_c$. The current consensus is that most experiments point towards a p-wave spin triplet, nodeless chiral state of the type $d(k) = \hat{z}(\sin k_x + i \sin k_y)$, but with additional lines of gap nodes somewhere on the Fermi surface [3, 4]. In the Zhitomirsky and Rice model [3] the lines of gap nodes arise because of an inter-band proximity effect which couples of the Cooper pairs between the “active” and “passive” sheets of the Fermi surface. In our alternative proposal [2, 4] we introduced an effective interlayer coupling model, which also leads to a chiral symmetry state with lines of gap nodes. This model has a fully gaped chiral state on the $\gamma$ Fermi surface sheet and horizontal lines of nodes at $k_z = \pm \pi / c$ on the $\beta$ sheet of the Fermi surface. Moreover, we have shown that it is in good agreement with the experimental measurements of specific heat, penetration depth and thermal conductivity below $T_c$.

In this letter we address the crucial question of the stability of the chiral symmetry state in the presence of spin-orbit coupling. For p-wave spin-triplet superfluids a large number of different pairing states are possible, and in general many different states are degenerate in the weak coupling limit. In the well known case of superfluid $^3$He, weak coupling theory favors the fully gaped Balian-Werthammer (B) phase relative to the Anderson-Brinkman-Morrel (A) phase which has point gap nodes [3]. In this case the A phase is stabilized (in zero magnetic field) only by a spin-fluctuation feedback effect in which the strength of the effective pairing interaction itself is modified by the pairing symmetry. The broken symmetry ground state effectively leads to a magnetic anisotropy in the spin fluctuation spectrum, which in turn favors the chiral solution. The analogous question in Sr$_2$RuO$_4$ is to ask whether the experimentally determined chiral state is stable simply because of magnetic anisotropy arising from spin-orbit interactions in the normal state, or whether it is also necessary to invoke a directly spin-dependent pairing interaction. Below we argue that this latter case applies, and therefore the very existence of the chiral pairing state provides evidence for a magnetic contribution to the pairing mechanism.

We follow earlier calculations [5, 6, 10, 11] where the role of spin-orbit coupling in the superconducting and normal states, respectively have been investigated. Our results are qualitatively consistent with the superconducting state calculations of Ng and Sigrist [9, 10, 11], but, importantly, differ in conclusion. We show below that single-particle spin-orbit coupling alone is unlikely to stabilize the chiral state. The chiral state is stabilized only after allowing for a spin-dependent effective interaction. Such an effective interaction is consistent with the implication of spin-orbit interaction for transverse and longitudinal spin fluctuations, in the work of Eremin, Manske and Bennemann [12] and Kuwabara and Ogata [13]. Therefore we suggest that the experimental observation of a chiral pairing state is conclusive proof of a spin-dependent pairing interaction. Whilst this conclusion strongly supports the spin fluctuation mechanism of pairing, it could also be consistent with other spin-dependent interaction models, such as Hund’s rule coupling [14].

The effective pairing Hamiltonian we consider is a simple three-band, attractive $U$, Hubbard model:

$$
\hat{H} = \sum_{ijmm',\sigma} \left( (\varepsilon_m - \mu) \delta_{ij} \delta_{mm'} - t_{mm'}(ij) \right) \hat{c}_{i\alpha\sigma}^{\dagger} \hat{c}_{j\beta\sigma} + \frac{1}{2} \sum_{ijmm'\sigma\sigma'} U^{\alpha\beta,\gamma\delta}_{mm'}(ij) \hat{c}_{i\alpha\sigma}^{\dagger} \hat{c}_{j\beta\sigma'}^{\dagger} \hat{c}_{j\beta\sigma'} \hat{c}_{i\alpha\sigma},
$$

(1)

where $m$ and $m'$ refer to the three Ruthenium $t_{2g}$ orbitals $a = xx$, $b = yz$ 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 [12, 16]. In our previous papers [13, 17] we chose the simplest set of attractive pairing interactions $U_{mm'}(ij)$ which lead to results consistent with experiment. However, more generally, for a spin-dependent pairing interaction the effective Hubbard parameters $U^{\alpha,\beta,\gamma,\delta}_{mm'}(ij)$ are spin as well as orbital dependent.

In the presence of spin-orbit coupling the original Hamiltonian (Eq. 1) should be supplemented by the term $\lambda \mathbf{L} \cdot \mathbf{S}$ for each Ru atomic site. This can be expressed in the form

$$H^{SO} = \frac{1}{2} \sum_{i,\sigma} \sum_{m,m'} \varepsilon^{\kappa mm'} \sigma^{\kappa}_{i\sigma} c^{\dagger}_{im\sigma} c_{im'i\sigma'},$$

where $\sigma^{\kappa}_{i\sigma}$, $\kappa = x, y, z$, are the Pauli matrices, $\varepsilon^{\kappa mm'}$ denotes the completely antisymmetric tensor, and the sign convention implies that here the Ru orbital indices must be ordered as $m = (y, z, x, xy)$ or $(b, a, c)$ in our notation [6].

In the Hartree-Fock-Gorkov approximation the gap equation takes the simplified form

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

where the pairing amplitude, or order parameter, $\chi^{\sigma\sigma'}_{mm'}(ij)$, is defined by the usual relations [12]

$$\chi^{\sigma\sigma'}_{mm'}(ij) = \sum_{\nu} \alpha^{\nu}_{im\sigma} v^{\nu*}_{jm'\sigma'} (1 - 2 f(E^{\nu})) \quad \text{for} \quad \sigma = \sigma'$$

$$\chi^{\sigma\sigma'}_{mm'}(ij) = \frac{1}{2} \sum_{\nu} (\alpha^{\nu}_{im\sigma} v^{\nu*}_{jm'\sigma'} + v^{\nu*}_{im\sigma'} \alpha^{\nu*}_{jm'\sigma}) (1 - 2 f(E^{\nu})) \quad \text{for} \quad \sigma \neq \sigma'.$$

Note that the full spin and orbital dependent effective interaction $U^{\alpha,\beta,\gamma,\delta}_{mm'}(ij)$ from Eq. [10] is reduced to the simpler set of parameters $U^{\sigma\sigma'}_{mm'}(ij)$ for which there are only two distinct cases: $U^{\uparrow\uparrow}_{mm'}(ij) = U^{\downarrow\downarrow}_{mm'}(ij)$ and

$U^{\uparrow\downarrow}_{mm'}(ij) = U^{\downarrow\uparrow}_{mm'}(ij)$. Other more general spin dependent terms, such as spin-flipping terms, might occur, but these would not have any effect on the states we consider.

The group theory of pairing symmetry of triplet (odd-parity) superconductivity in tetragonal crystals has been discussed by Volovik and Gorkov [12] and reviewed in Annett [14] and Mineev and Samikhin [20]. In this case there are five distinct symmetry gap functions which are relevant. Transforming to k-space and using the standard representation $\Delta^{\sigma\sigma'} = i \sigma_{\sigma} \mathbf{d}_{k}$, where $\Delta$ and $\mathbf{d}$ are matrices in orbital indices, they have the characteristic order parameters

$$(a) \quad \mathbf{d}_{k} \sim (0, 0, X + iY)$$
$$(b) \quad \mathbf{d}_{k} \sim (X, Y, 0)$$
$$(c) \quad \mathbf{d}_{k} \sim (Y, -X, 0)$$
$$(d) \quad \mathbf{d}_{k} \sim (X, -Y, 0)$$
$$(e) \quad \mathbf{d}_{k} \sim (Y, Y, 0)$$

corresponding to the $E_u$, $A_{1u}$, $A_{2u}$, $B_{1u}$ and $B_{2u}$ pairing symmetry states [19]. Here $X(k)$ and $Y(k)$ are pairs of basis functions transforming as $k_x$ and $k_y$, respectively, under crystal point group symmetries. For in-plane interactions $X = \sin k_x$ and for inter-plane interactions $X = \sin (k_2/2) \cos (k_4/2) \cos (k_2/2)$ in the body centered tetragonal crystal Sr₂RuO₄. For brevity we shall refer to these five pairing states as (a)–(e) below.

If the pairing interaction were spin-independent, then the interaction parameters would obey

$$U^{\uparrow\uparrow}_{mm'}(ij) = U^{\downarrow\downarrow}_{mm'}(ij).$$

With this assumption we solved the full self-consistent

FIG. 1: Critical temperature for solutions of various symmetries denoted by symbols (a)–(e) defined in Eq. 5.

FIG. 2: Condensation free energy for solutions of various symmetries defined in Eq. 6. For $\lambda > 0$ the most stable state is (b) $\mathbf{d}(k) = (X, Y, 0)$, while for $\lambda < 0$ the state (e) $\mathbf{d}(k) = (X, Y, 0)$. The chiral state (a) is never stable for nonzero spin-orbit coupling. The inset shows the condensation energy for small $\lambda$ calculated for the states (b)–(e) with respect to the state (a).
gap equations (Eqs. 3, 4) in presence of a weak spin-orbit coupling parameter $\lambda$. The band structure and the interaction parameters were chosen as in our previous works [6, 11, 21]. The results for the critical temperature $T_c$ are shown in Fig. 1 and the condensation free energies ($F_s - F_n$) at $T = 0$ are shown in Fig. 2. Both figures show clearly that all five pairing states become exactly degenerate at $\lambda = 0$, as expected. For positive values of $\lambda$ the states split in such a way that the (b) and (c) states have the highest $T_c$ and the greatest (negative) condensation energy. Conversely, for negative values of $\lambda$ states (d) and (e) have the highest $T_c$ and the greatest condensation energy. Surprisingly, the chiral state, (a), is never stable compared to the others.

This counter-intuitive result deserves some comment. Firstly, one should note that experiments point unequivocally to a chiral solution, since only this one would have local to a chiral solution, since only this one would have

Although we draw different conclusions, on closer inspection, our results are indeed very similar to theirs. They used a three-band model characterized by two main pairing parameters $V_1 = V_2$ and $V_3$. They found that the chiral state is favored for positive $\lambda$ only in the region $V_3/V_2 < 1$. For $V_3/V_2 > 1$ they found that states (b) and (c) (III and II in their notation) were favored, and that these two states were nearly degenerate being separated only by an additional inter-band proximity effect coupling term. Our results are qualitatively consistent with this finding, if we interpret our effective Hubbard interaction terms $U_{\parallel L}$ and $U_{\perp L}$ as giving a corresponding value of $V_3/V_2 \approx 2U_{\parallel L}/U_{\perp L} = 2.4$. In our model we have no inter-band proximity effect coupling, and hence states (b) and (c) are essentially degenerate.

The physical values of the spin-orbit coupling parameter are positive, and believed to be of order $\lambda = 40 - 80$ meV, corresponding to $\lambda/t \approx 0.5 - 1$ in our units. We have also carried out fully self-consistent calculations at $\lambda = 0.5$, and found the same ordering of the ground states as shown in Fig. 2. But for such large values of $\lambda$ we observed that the Fermi surface shape begins to change, and so it was necessary to refit the Hubbard model hopping parameters to the experimental Fermi surface [16]. However this refitting of the Fermi surface did not change the results qualitatively from those expected from Fig. 2 and so we conclude that the precise Fermi surface shape is not essential in determining the relative stability of the different pairing states.

In order to understand more fully the physical origin of the spin-orbit splitting shown in Figs. 1-2, we consider small perturbations about the point $\lambda = 0$. Here the change in condensation energy to the linear order is

$$\delta F = \frac{\lambda}{2} (\langle \hat{L}_z \rangle) = \frac{\lambda}{2} \sum_{m,m'} \langle m | \hat{L}_z | m' \rangle n_{m,m'}^{\uparrow \uparrow},$$

$$+ \frac{\lambda}{2} \sum_{m,m'} \langle m | \hat{L}_x - i \hat{L}_y | m' \rangle n_{m,m'}^{\uparrow \downarrow},$$

$$+ \frac{\lambda}{2} \sum_{m,m'} \langle m | \hat{L}_x + i \hat{L}_y | m' \rangle n_{m,m'}^{\downarrow \uparrow},$$

$$- \frac{\lambda}{2} \sum_{m,m'} \langle m | \hat{L}_z | m' \rangle n_{m,m'}^{\downarrow \downarrow},$$

where $n_{m,m'}^{\sigma \sigma'} = \langle c_{i,m}^\sigma c_{i,m'}^{\sigma'} \rangle$ are on-site density matrices for the Ru $d$ orbitals, in the absence of spin-orbit coupling. In fact, in our model, the $\gamma$ band associated with the $d_{xy}$ orbital is strongly decoupled from the $d_{xz}$ and $d_{yz}$ orbitals, and so the only significant off-diagonal density matrix elements are $n_{ab}^{\sigma \sigma'}$ and $n_{ba}^{\sigma \sigma'}$. Thus we find the spin-orbit correction

$$\delta F = \frac{\lambda}{2} \left( n_{ab}^{\uparrow \uparrow} - n_{ab}^{\uparrow \downarrow} - n_{ba}^{\uparrow \downarrow} + n_{ba}^{\uparrow \uparrow} \right),$$

For the chiral (a) state $n_{ab}^{\uparrow \uparrow} = n_{ab}^{\downarrow \downarrow}$ and so the spin-orbit coupling is zero to linear order in $\lambda$. On the other hand for (b)-(e) $n_{ab}^{\uparrow \downarrow} = -n_{ab}^{\downarrow \uparrow}$ and so the spin-orbit energy is of order $\lambda$. This explains the small $\lambda$ behavior seen in our numerical calculations, and shown in the inset to Fig. 2. It is interesting to note that for all the states (a)-(e) the expectation value of the spin $\langle \hat{S} \rangle = 0$ for each orbital $m$. On the other hand the $z$ component of the orbital angular momentum is

$$\langle \hat{L}_z \rangle = n_{ab}^{\uparrow \uparrow} + n_{ab}^{\downarrow \downarrow} - \left( n_{ba}^{\uparrow \uparrow} + n_{ba}^{\downarrow \downarrow} \right),$$

which is non-zero only for the chiral state. Therefore $\langle \hat{L}_z \rangle$ is non-zero for four states (b)-(e) which have $\langle \hat{S} \rangle = 0$ while the (a) state has $\langle \hat{S} \rangle = 0$, $\langle \hat{L}_z \rangle \neq 0$.

Given this surprising result, how we can understand the stability of the chiral state, which is required for understanding the susceptibility experiments? Clearly, if the effective pairing interaction is caused, even in part, by spin fluctuations, then in general we would expect that $U_{mn}^{\uparrow \uparrow}(ij) \neq U_{mn}^{\downarrow \downarrow}(ij)$, corresponding to the two Fermi liquid parameters $F_{\uparrow \uparrow}, F_{\downarrow \downarrow}$. In the spin-fluctuation theory of Kuwabara and Ogata [13] one can see that the parallel spin and opposite spin effective interactions are derived from transverse or longitudinal spin fluctuations, respectively, and hence they become different when spin-orbit interactions are present. The role of spin-orbit coupling in the transverse, $\chi^\perp(q)$, and longitudinal, $\chi^\parallel(q)$, spin susceptibilities has been explored more fully by Eremin, Manske and Bennemann [12], and their theory agrees well with the results of NMR [22] and neutron scattering experiments [23]. Therefore spin-orbit coupling

has two quite distinct effects on a spin triplet superconductor. Firstly, the spin-orbit term, Eq. 2, directly enters the gap equation leading to small changes in the single-particle band structure and density of states, and secondly, it leads to spin anisotropy in the effective pairing interaction. Both of these effects must be included in order to establish the relative stability of the different possible pairing symmetry states [25].

We have found that the chiral state becomes stable when we increased the interaction $U^{\uparrow\downarrow} = U'$ by 1% with respect to $U^{\uparrow\uparrow} = U$. These results in Fig. 3. The phase diagram in the $U'/U$ vs. $\lambda/t$ space is presented in Fig. 4. It shows that to stabilize the chiral state in the presence of sizeable spin-orbit coupling requires increase of $U'$ by less than 1%.

It follows from these results, that the pairing interaction itself must directly favor the chiral symmetry state and would be an analogue of the well known spin-fluctuation feedback mechanism which stabilizes the A-phase in superfluid $^3$He [8]. Eremin, Manske and Bememann [12] calculated the spin susceptibility in the normal state of Sr$_2$RuO$_4$. They found that spin-orbit interaction led to a magnetic anisotropy between $\chi^z(q, \omega)$ and $\chi^\pm(q, \omega)$. The well known argument for $^3$He would then suggest that these susceptibilities lead to different effective pairing interactions between spin-triplet, $S = 1$, quasiparticle pairs in the $m = 0$ or $m = \pm 1$ channels. As it turns out a small increase of the interaction, $U'/U = 1.01$, stabilizes the (a) state.

This work has been partially supported by the KBN grant No. 2P03B06225 and the NATO Collaborative Linkage Grant No. 979446 and the INTAS grant No. 01-654. GL would like to thank Max Planck Institute for the Physics of Complex Systems in Dresden for hospitality. The authors are grateful D. Manske and I. Eremin for discussions.

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