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LETTER TO THE EDITOR

# $Sr_2RuO_4$: an electronic analogue of $^3He$?

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**Abstract.** $Sr_2RuO_4$ is a superconductor with a similar structure to a high-$T_c$ cuprate superconductor. Nevertheless, the superconducting state may have different symmetry than that of cuprate superconductors. Strong Hund's rule coupling favours triplet over singlet pairing, similar to $^3He$. A strong candidate is the odd-parity pairing state which is the two-dimensional analogue of the Balian–Werthamer state of $^3He$. Various experimental consequences and tests are analysed.

Recently Cava *et al* [1], while looking for a 4d analogue of the high-$T_c$ cuprates, explored the localized to itinerant transition in an alloy series, $Sr_2Ir_{1-x}Ru_xO_4$, with the same layered perovskite structure as the $La_{2-x}Sr_xCuO_4$ compounds. Shortly thereafter Maeno *et al* [2] discovered superconductivity in high-quality crystals of $Sr_2RuO_4$ but with a low value of $T_c = 0.93$ K and contrasted this material with the high-$T_c$ cuprates. At present, not much is known in detail about the superconducting state of $Sr_2RuO_4$ and any possible relation to the cuprates. The purpose of this letter is to raise an alternative possibility, namely a close analogy of the electronic superconductor $Sr_2RuO_4$ to the triplet superfluid phases of $^3He$.

A lot is known about the electronic bandstructure of $Sr_2RuO_4$. Quantum oscillations in high magnetic fields in the normal phase at low temperatures were observed in an elegant series of experiments by Mackenzie *et al* [3]. The Fermi surface consists of three approximately cylindrical pieces in agreement with bandstructure calculations [4]. The oxidation state $Ru^{4+}$ has four 4d electrons which partially fill the non-bonding $t_{2g}$ subband. Taking $z$ to be parallel to the $c$ axis, we can identify the relevant orbitals as an $xy$ orbital dispersing strongly along $k_x$ and $k_y$, and $\{xz, yz\}$ orbitals which are degenerate at the $\Gamma$ point but do not disperse along $k_x$ and $k_y$, respectively. All three bands are occupied at the $\Gamma$ point and the dispersion causes two bands to cross the Fermi surface along $k_x$ and along $k_y$. Along the $k_x = \pm k_y$ line all three bands disperse and cross the Fermi energy. The Fermi surface consists of a hole cylinder at the zone corner and two electron cylinders with almost square cross-sections centred at the $\Gamma$ point.

Maeno *et al* pointed out signs of strong correlations [2]. The resistivity shows a crossover at a temperature $T \sim 120$ K from 2D behaviour and incoherent transport along the $c$ axis to Fermi liquid behaviour and a $T^2$ law along all directions as the temperature is lowered. Mackenzie *et al* could measure the individual mass enhancements on different Fermi surface sheets and found values of $\approx$4 in each case [3]. If one assigns all of the paramagnetic susceptibility to a Pauli term $\chi_0$, then the Wilson ratio is 1.8 [2] giving a susceptibility enhancement $\approx$8. These values agree roughly with those of $^3He$ at atmospheric pressure. Note, however, Carter *et al* [5] argue for a substantial van Vleck term from an examination of the variation of the susceptibility in the $Sr_2Ir_{1-x}Ru_xO_4$ alloys, which could lower the enhancement.

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The alloy series does, however, show that dilute $Ru^{4+}$ acts as a local $S = 1$-moment impurity in $Sr_2IrO_4$ which can be interpreted as a sign of the importance of Hund's rule coupling of the two holes in $\{xz, yz\}$ orbitals. Further evidence favouring $S = 1$ or spin triplet correlation is the ferromagnetic metallic state observed in the perovskite, $SrRuO_3$. This metal is the three-dimensional analogue of the layered $Sr_2RuO_4$ material and below a Curie temperature of $T_c = 160$ K shows a saturated moment of 0.85 $\mu_B$ [6]—again an indicator for triplet correlation.

In summary as remarked by previous workers [1, 2, 3] there is considerable evidence for the presence of strong correlations in $Sr_2RuO_4$. However, since the closely related magnetic compound in this case is a ferromagnet and since there is an approximate correspondence to the many-body enhancements of the specific heat and susceptibility of $^3$He at atmospheric pressure, it is plausible to look for triplet or $S = 1$ pairing in the superconducting state here rather than the singlet pairing observed in the cuprates.

Because of the strongly two-dimensional electronic structure of $Sr_2RuO_4$ we wish to analyse triplet superconductivity in two dimensions on a square lattice. The low value of $T_c$ in comparison to the onset temperature of the well developed Fermi liquid state suggests weak coupling. Under these circumstances we should look for a two-dimensional triplet pairing state without any gap nodes, the analogue of the Balian–Werthamer (BW) state of $^3$He. We write the pairing matrix $\Delta_{ss'}(\boldsymbol{k})$ in the standard form (see Sigrist and Ueda [8]),

$$\begin{aligned} \hat{\Delta}(\boldsymbol{k}) &= \mathrm{i}\left(\sum_{\mu} d^{\mu}(\boldsymbol{k})\hat{\sigma}_{\mu}\right)\hat{\sigma}_y \\ &= \begin{bmatrix} -d_x(\boldsymbol{k}) + \mathrm{i}d_y(\boldsymbol{k}) & d_z(\boldsymbol{k}) \\ d_z(\boldsymbol{k}) & d_x(\boldsymbol{k}) + \mathrm{i}d_y(\boldsymbol{k}) \end{bmatrix} \end{aligned} \tag{1}$$

where $\hat{\sigma}^{\mu}$ denotes Pauli matrices ($\mu = x, y, z$) and $\boldsymbol{d}(\boldsymbol{k})$ is a vectorial function which is odd in $\boldsymbol{k}$. In table 1 we classify the triplet ('p wave') pairing states which, according to their symmetry, would be nodeless on the cylindrical Fermi surface sheets of $Sr_2RuO_4$. It is the absence of nodes that stabilizes them in a weak-coupling approach. In the absence of spin–orbit coupling all states would be degenerate, i.e. their transition temperature $T_c$ would be identical. Consequently, spin–orbit coupling although weak is essential to lift this degeneracy. The odd-parity representations of the crystal point group $C_{4v}$, $\Gamma^-_{1-4}$ (1D representations) and $\Gamma^-_5$ (2D representation), can be interpreted as a classification according to the total 'angular momentum'($\boldsymbol{J} = \boldsymbol{S} + \boldsymbol{L}$). We expect that the most symmetric state ($\boldsymbol{J} = \boldsymbol{0}$) would be favoured by spin–orbit coupling, i.e. $\boldsymbol{d}(\boldsymbol{k}) = \hat{\boldsymbol{x}}k_x + \hat{\boldsymbol{y}}k_y$ belonging to $\Gamma^-_1$. This state has the structure of the axial state arising as the limit of the three-dimensional BW state near to a planar wall as analysed by Anderson and Brinkman [7].

**Table 1.** A list of possible triplet pairing states including splitting due to spin–orbit coupling. The crystal point group symmetry is that of a 2D square lattice, $C_{4v}$. $\hat{\boldsymbol{x}}, \hat{\boldsymbol{y}}$ and $\hat{\boldsymbol{z}}$ denote the unit vectors along the $x$ , $y$ and $z$ directions, respectively. $J$ and $J_z$ denote the total anglular momentum of the pairing state and its $z$ component, respectively.

| $\Gamma$ | $J, J_z$ | $\boldsymbol{d}(\boldsymbol{k})$ |
|---|---|---|
| $\Gamma^-_1$ | 0, 0 | $\hat{\boldsymbol{x}}k_x + \hat{\boldsymbol{y}}k_y$ |
| $\Gamma^-_2$ | 1, 0 | $\hat{\boldsymbol{x}}k_y - \hat{\boldsymbol{y}}k_x$ |
| $\Gamma^-_3$ | 2, ±2 | $\hat{\boldsymbol{x}}k_x - \hat{\boldsymbol{y}}k_y$ |
| $\Gamma^-_4$ | 2, ±2 | $\hat{\boldsymbol{x}}k_y + \hat{\boldsymbol{y}}k_x$ |
| $\Gamma^-_5$ | 1, ±1 | $\hat{\boldsymbol{z}}(k_x \pm \mathrm{i}k_y)$ |

**Table 2.** The low-lying collective modes for the $\Gamma_1^-$ state classified according to the fluctuation channels.

| $\Gamma$ | $\delta d(k)$ | Type of mode | Possible probes |
|---|---|---|---|
| $\Gamma_2^-$ | $\hat{x}k_y - \hat{y}k_x$ | spin supercurrent | — |
| $\Gamma_3^-$ | $\hat{x}k_x - \hat{y}k_y$ | angular modulation of | ultrasound |
| $\Gamma_4^-$ | $\mathrm{i}(\hat{x}k_y + \hat{y}k_x)$ | the gap | ultrasound |
| $\Gamma_5^-$ | $\hat{z}(k_x, k_y)$ | spin rotation | microwave |

Firstly, however, we discuss the properties of all the states classified in table 1 without any preference for one of them. The pairing states belonging to the 1D representations, $\Gamma_{1-4}^-$, are so-called equal spin pairing (ESP) states, i.e. $\Delta_{\uparrow\uparrow}(k) = -\Delta_{\downarrow\downarrow}^*(k)$ and $\Delta_{\uparrow\downarrow}(k) = \Delta_{\downarrow\uparrow}(k) = 0$. On the other hand, the spin of the $\Gamma_5^-$ pairing state $d = \hat{z}(k_x \pm \mathrm{i}k_y)$ lies in the basal plane and is the analogue to the ABM state of $^3$He. This state breaks time-reversal symmetry and, thus, is twofold degenerate. Nevertheless, all states are unitary, i.e. $\hat{\Delta}^\dagger(k)\hat{\Delta}(k)$ is proportional to the unit matrix.

The key question is how to distinguish these states, firstly from singlet pairing states and then between each other. Clearly the existence of a finite energy gap leads to an exponential decay in the density of thermally excited quasiparticles at low temperature and the many properties dependent on the quasiparticle density of states will be indistinguishable from standard singlet s wave superconductivity. The spin susceptibility $\chi_s(T)$, however, offers the possibility of detecting the triplet state. As $T \to 0$, $\chi_s$ becomes highly anisotropic [7]. It takes the form

$$\begin{aligned} \frac{\chi_s(T)}{\chi_0} &= 1 \qquad && H \parallel \hat{z} \\ \frac{\chi_s(T)}{\chi_0} &= \frac{1}{2}\left[1 + Y\left(\frac{d}{k_B T}\right)\right] \qquad && H \perp \hat{z} \end{aligned} \tag{2}$$

for the 1D representations ($\Gamma_{1-4}^-$) and

$$\begin{aligned} \frac{\chi_s(T)}{\chi_0} &= Y\left(\frac{d}{k_B T}\right) \qquad && H \parallel \hat{z} \\ \frac{\chi_s(T)}{\chi_0} &= 1 \qquad && H \perp \hat{z} \end{aligned} \tag{3}$$

for the $\Gamma_5^-$ state, where $Y(x)$ is the Yosida function and $d$ is the gap magnitude. The spin susceptibility is not directly measurable in a superconductor but in type II superconductors it may be measured indirectly through the Knight shift. In this way we can also distinguish between the the 1D and 2D representation state. In the case of the $\Gamma_{1-4}^-$ states, the key prediction is that the Knight shift is unchanged upon cooling through $T_c$ in a magnetic field oriented along the $c$ axis but should show a substantial drop if the magnetic field lies in the *ab* plane. Essentially the opposite behaviour is true for the $\Gamma_5^-$ state. In the present case, however, the low value of the critical field leads to experimental difficulties. Thus, we present in the following several additional characteristic properties which may be used to identify the symmetry of the pairing state.

The analogy to $^3$He may be extended to the existence of collective modes characteristic of the pairing symmetry. The majority of modes for the classified pairing states are massive with energy gaps of the order of the quasiparticle excitation gap [9]. However, if we neglect spin–orbit coupling for the moment, there are three types of gapless modes originating from broken gauge, spin and orbit rotation symmetry. All modes can be understood as fluctuations of the pairing state into one of the other pairing channels listed in table 1. Let us first consider the mode connected with the broken gauge symmetry which occurs for the states belonging to the 1D representations. It is most easily understood if we write these ESP states in the following way,

$$\Delta_{\uparrow\uparrow}(\boldsymbol{k}) = \eta_{\uparrow}[-\mathrm{d}_x(\boldsymbol{k}) + \mathrm{i}\mathrm{d}_y(\boldsymbol{k})]$$

$$\Delta_{\downarrow\downarrow}(\boldsymbol{k}) = \eta_{\downarrow}[d_x(\boldsymbol{k}) + \mathrm{i}\mathrm{d}_y(\boldsymbol{k})] \tag{4}$$

$$\Delta_{\uparrow\downarrow} = \Delta_{\downarrow\uparrow} = 0$$

where $\eta_{\uparrow}$ and $\eta_{\downarrow}$ are complex amplitudes of the order parameter. In the absence of spin–orbit coupling the up and down spin pairing components can be considered independently [10]. From equation (4) we identify two modes corresponding to in-phase and out-of-phase oscillations of the phases of $\eta_{\uparrow}$ and $\eta_{\downarrow}$. The in-phase mode (related to the charged supercurrent), however, is raised to the plasma frequency due to the fact that we consider a 'superfluid' of charged particles. Therefore it is not of interest here. On the other hand, the out-of-phase mode (related to the spin supercurrent) does not involve charge transport and is indeed a gapless mode with a linear dispersion [9, 10]. In the case when the $\Gamma_1^-$ state is the ground state this mode corresponds to an oscillation to the $\Gamma_2^-$ channel.

The second type of mode has its origin in spin rotation (rotation of $\boldsymbol{d}$) and can occur for all representations through a fluctuation of any of the 1D representation channels into the $\Gamma_5^-$ state and vice versa. This mode is gapless and has also a linear dispersion.

Finally, the third mode is a fluctuation in the angular dependence of the gap,

$$\Delta(\theta) = \Delta_0 + \delta\cos(2\theta) \tag{5}$$

where $\theta$ is the angle of the momentum in the basal plane and $\delta$ is a small oscillatory amplitude. This mode exists for the 1D representations and occurs for the $\Gamma_1^-$ pairing state via fluctuations to the $\Gamma_3^-$- or $\Gamma_4^-$ channel. Also this mode is gapless and has a linear dispersion. All three types of mode are essentially equivalent in the sense that they represent oscillations as rotations in the spin–orbit space (table 2). However, their physical properties are different in a two-dimensional system.

When we turn on the spin–orbit coupling both types of mode become massive. As mentioned above, spin–orbit coupling lifts the degeneracy among the different representations in table 1 and the gap introduced by spin–orbit coupling is related to the splitting of the bare transition temperatures $T_c(\Gamma)$. Thus, considering the fluctuations of the $\Gamma_1^-$ state into the $\Gamma_2^-$ channel the lowest mode frequency or gap is given by $\omega^2 \propto \ln[(T_c(\Gamma_1^-)/T_c(\Gamma_2^-)]$ [11]. The proportionality factor depends on various details, in particular, Fermi liquid corrections which are not easy to estimate with our present understanding. Because, however, the spin–orbit coupling should be rather weak, the gap should be small and well below the quasiparticle gap. It is difficult to probe this mode directly, because it requires distinct coupling to the phases of the up and down equal spin pairs. A similar analysis concerning the gap applies to the other collective modes. The mode connected with spin rotation might be observable via microwave absorption. The last mode associated with axial deformation of the gap function on the Fermi surface couples to distortions of the cylindrical Fermi surface and can, consequently, lead to absorption of ultrasound with polarization in the basal plane.

Additional information could be obtained from the observation of the internal phase structure of the order parameter by integrating a single crystal of $Sr_2RuO_4$ into a SQUID device with an s wave superconductor [12, 13]. Although singlet s wave and triplet p wave superconductors would have no Josephson effect due to the different behaviour under the time reversal operation, in general, spin–orbit coupling leads to a finite Josephson tunnelling [8, 12]. Hence, the SQUID geometry described by Geshkenbein *et al* [12] could distinguish between even- and odd-parity pairing.

For the $\Gamma_5^-$ state an additional probe is possible. The twofold degeneracy of the $\Gamma_5^-$ representation may be lifted by applying uniaxial stress along one of the main axes of the basal plane. Thus, the superconducting phase transition would split into two consecutive ones with a high-temperature phase like $d(k) = \hat{z}k_x$ and a low-temperature phase, $d(k) = \hat{z}(k_x \pm i\varepsilon k_y)$ ($\varepsilon$ real) [14].

In this short letter we have presented arguements favouring unconventional superconductivity in $Sr_2RuO_4$, with odd-parity pairing. If this can be confirmed, and in our view an NMR measurement of the Knight shift would be the most direct test, then the analogy to the superfluid state of $^3$He will be quite close. Spin–orbit coupling should favour the most symmetric gapless pairing state belonging to the representation $\Gamma_1^-$. As we mentioned earlier, both $Sr_2$ $RuO_4$ and $^3$He are Landau Fermi liquids and the enhancement factors of the specific heat and spin susceptibility are comparable. Also the values of the $T_c$ in dimensionless units, i.e. $T_c/T_F$ (where $T_F$ is the Fermi temperature), are in the range $10^{-3}$–$10^{-4}$ in both systems. In $Sr_2RuO_4$ the lower crystalline symmetry will not allow texture formation and other high-symmetry structures of the superfluid, but still low-lying collective modes are predicted and some of them could lead to directly observable effects.

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*Note added in proof.* A further test for triplet pairing can be obtained by sandwiching a thin film of $Sr_2RuO_4$ between two singlet superconductors with a higher transition temperature. Above $T_c$ of $Sr_2RuO_4$ this system should behave like a standard SNS Josephson junction where the coupling is due to proximity-induced singlet pairs in the $Sr_2RuO_4$. Below $T_c$, however, the Josephson coupling should decrease because as triplet pairing appears in $Sr_2RuO_4$ the proximity-induced singlet order parameter will be suppressed [15]. The observation of such an anomalous temperature dependence of the Josephson effect would confirm the odd-parity symmetry of the order parameter.

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