Superconducting double transition and substantial Knight shift in \( \text{Sr}_2\text{RuO}_4 \)

R. Gupta\(^1\), T. Saunderson\(^1\), S. Shallcross\(^2\), M. Gradhand\(^1\), J. Quintanilla\(^3\), and J. Annett\(^4\)

\(^1\) H. H. Wills Physics Laboratory, University of Bristol, Tyndall Ave, BS8 1TL, UK
\(^2\) Max-Born-Institute for non-linear optics, Max-Born Strasse 2A, 12489 Berlin, Germany
\(^3\) Physics of Quantum Materials, School of Physical Sciences, University of Kent, Canterbury CT2 7NH, United Kingdom

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Recent nuclear magnetic resonance experiments measuring the Knight shift in \( \text{Sr}_2\text{RuO}_4 \) have challenged the widely accepted picture of chiral pairing in this superconductor. Here we study the implications of helical pairing on the superconducting state while comparing our results with the available experimental data on the upper critical field and Knight shift. We solve the Bogoliubov-de-Gennes equation employing a realistic three-dimensional tight-binding model that captures the experimental Fermi surface very well. In agreement with experiments we find a Pauli limiting to the upper critical field and, for a finite in-plane magnetic field, a double superconducting transition below a temperature \( T^* \). These transitions are first-order in nature and merge into a single second-order transition at a critical point \((T^*, H^*)\), for which we find \((0.8 \text{ K}, 2.35 \text{ T})\) with experiment reporting \((0.8 \text{ K}, 1.2 \text{ T})\) \([J. \text{ Phys. Soc. Jpn.} \textbf{71}, 2839 (2002)]\). Furthermore, we find a substantial drop in the Knight shift in quantitative agreement with recent experiments.

I. INTRODUCTION

More than two decades after the discovery of superconductivity in \( \text{Sr}_2\text{RuO}_4 \)\(^3\) the nature of the pairing symmetry in this material remains unsettled. It has been speculated\(^2\) to be a long sought metallic analogue of superfluid helium-3 \((^3\text{He})\), and the possibility of triplet superconductivity has been explored by various groups (see Ref. \[^{2,4}\] and references therein). Theoretically, it was found that the free energy differences between different possible pairing symmetries were so small as to be nearly degenerate, rendering it a far from trivial problem to predict the pairing symmetry\(^5\), a situation exacerbated by the large number of symmetry-distinct superconducting order parameters\(^5\) compatible with the body centred tetragonal structure. Distinguishing between different order parameters therefore requires experiments to be performed under very stringent conditions. An indirect approach, where one determines specific experimental signatures of each pairing symmetry, thus provides an attractive alternative route to understanding this material\(^5\).

Early experiments pointed to \( \text{Sr}_2\text{RuO}_4 \) being an odd-parity chiral superconductor. Specifically, measurements of the Knight shift at both \( \text{O} \)\(^{16}\) and \( \text{Ru} \)\(^{11}\) sites showed almost no drop in value under a magnetic field applied in the \( x-y \) plane, exactly as expected for the chiral \( p \)-wave state. Confirmation of this result was found in direct measurements of the field dependent magnetic moment by neutron scattering\(^12\), although the large experimental error bars implied that a small Knight shift could not be ruled out. The chiral \( p \)-wave pairing state was further supported by phase sensitive measurement\(^13\)-\(^14\) which, under inversion, reported a phase change of \( \pi \) in the superconducting order parameter. The \( p \)-wave chiral pairing state picture was also consistent with experiments such as muon spin rotation \((\mu\text{SR})\)\(^15\) and polar Kerr rotation\(^16\) which revealed the time reversal symmetry breaking (TRSB) when \( \text{Sr}_2\text{RuO}_4 \) enters the superconducting phase. In contrast, the surface magnetic fields or associated edge supercurrents expected in the chiral state were never observed, despite many experimental efforts\(^16\). Furthermore, recent experiments on \( x-y \) plane uniaxial strain dependence of \( T_c \) did not show the expected linear change in \( T_c \) for small strains, as required theoretically for a \( p_x + ip_y \) chiral state\(^16\), raising further doubts as to the existence of chiral \( p \)-wave pairing in this material.

Additionally, studies of the upper critical field\(^12\)-\(^24\) revealed another serious discrepancy. At low temperatures, a first-order superconducting to normal transition in the magneto-caloric effect\(^25\) the specific heat\(^22\) and magnetization\(^21\) was observed under a magnetic field applied in the \( x-y \) plane, characteristic of Pauli-limiting\(^23\) and inconsistent with the Knight shift measurements. For about 20 years there have been a number of attempts to understand this puzzling behaviour with little or no success. Recently, new Knight shift experiment\(^15\) contradicting the original experiments, observed a large drop in its value below \( T_c \) for \( x-y \) plane fields, with the previously observed temperature independent Knight shift attributed to sample heating during measurements\(^26\). These new measurements decisively rule out the chiral \( p \)-wave pairing state and instead are consistent with the helical- or singlet-pairing in the superconducting state\(^25\). Furthermore, the recent observation of half-quantized fluxoids\(^27\)-\(^29\) which requires multiple order parameters for the pairing function with both the spin and orbital degrees of freedom active, implies the possibility of spin-triplet pairing.

Here we investigate a time reversal symmetry preserving helical pairing\(^25\)-\(^30\)\(^33\) state under an in-plane magnetic field using a realistic three-dimensional (3D) tight-binding (TB) model. We focus on results from two experimental studies\(^15\)-\(^25\) to probe the internal symmetry of the Cooper pairs, and report two key findings. Firstly, as
in Ref. \cite{19}, we find a double superconducting transition below a temperature $T^*$, as a spin-only magnetic field is applied. These transitions are first-order in nature and merge into a single, second-order transition at a critical point ($T^*, H^*$), for which we find (0.8 K, 2.35 T) with experiment reporting (0.8 K, 1.2 T)\cite{19}. Secondly, our Knight shift results are in good quantitative agreement with Ref. \cite{27}. We find a 44% drop in its $T = 0$ K value from the normal-state value at a field of 0.7 Tesla. Our results therefore suggest that time reversal symmetry preserving helical pairing could be the appropriate pairing symmetry to explain many of the experimental features of $Sr_2RuO_4$. Evidently, this would then require separate explanation for other phenomena that have been interpreted as evidence of TRSB, including the increased zero-field muon spin relaxation rate in the superconducting state and the Kerr effect. A discussion of this is offered towards the end of the paper.

The remainder of this article is structured as follows. In Sec. \[II\] we describe the theoretical model employed in this work. The next section (Sec. \[III\]) detailing our results is divided into four subsections where we discuss the gap-function, specific heat, spin susceptibility and Knight shift, and variation of polar angle. All the calculations are performed both at fixed temperature (varying the magnetic field) and vice-versa. Thereafter, we conclude our results with a discussion of possible future research in Sec. \[IV\].

**II. THREE DIMENSIONAL TIGHT-BINDING MODEL**

We employ a 3D TB Hamiltonian consisting of $d_{xy}$, $d_{xz}$, and $d_{yz}$ orbitals following the approach of Ref. \cite{34}, which was previously applied to the study of chiral pairing in the superconducting state. The model is built upon the full 3-dimensional Fermi surface consisting of three sheets, as determined experimentally\cite{25}. Superconductivity is introduced into the model by adding a minimal set of site and orbital dependent negative $U$ pairing interactions. By introducing horizontal nodal lines into two of the sheets of the Fermi surface, it was shown that for the chiral superconducting state the model described the experimental specific heat very well. These line nodes have recently been revealed by spin resonance in inelastic neutron scattering experiment\cite{25}. The key difference from Ref. \cite{34} that we introduce here is to consider a pairing interaction that leads to helical pairing (between the same spin-types) instead of chiral pairing (between the opposite spin types). This choice of helical pairing is motivated, as explained in the introduction, by new experiments\cite{27,37} in which a substantial drop in the Knight shift and magnetic susceptibility has been observed under a magnetic field applied parallel to the $RuO_2$ plane.

Our effective pairing Hamiltonian is a multi-band attractive $U$ Hubbard model with an “off-site” pairing\cite{35}

$$\hat{H} = \sum_{ijmm'} ((\varepsilon_m - \mu)\delta_{ij}\delta_{mm'} - t_{mm'}(ij))c_{ijm}\sigma c_{jm'}\sigma - 1/2 \sum_{ijmm',\sigma,\sigma'} U_{mm'}(ij)\hat{n}_{ijm}\sigma \hat{n}_{jm'}\sigma', \hspace{1cm} (1)$$

where $m$ and $m'$ stand for the three Ruthenium $t_{2g}$ orbitals $a = d_{xy}, b = d_{xz}, c = d_{yz}$ and $i, j$ refer to the sites of a body centered tetragonal lattice. The hopping integrals $t_{mm}(ij)$ and on-site energies $\varepsilon_m$ have been reported in Ref. \cite{34}, which were fitted to reproduce the experimentally determined Fermi surface. The off-site pairing interaction involves two interaction constants, $U_{||}$ for nearest neighbours in the plane and $U_{\perp}$ for nearest neighbours in adjacent planes. Also, the in-plane interaction is taken finite only for the $a - a$ pairing and the out-of-plane interaction is assumed finite for the $b - b$, $c - c$, $b - c$ types of pairings written in terms of a $3 \times 3$ matrix

$$\hat{U}_{m, m'} = \begin{pmatrix} U_{||} & 0 & 0 \\ 0 & U_{\perp} & U_{\perp} \\ 0 & U_{\perp} & U_{\perp} \end{pmatrix}, \hspace{1cm} (2)$$

with the matrix indices ordered as $a$, $b$ and $c$ orbitals. This choice was motivated by the spatial symmetries of different orbitals: the “a” orbitals are confined to the $x-y$ plane and hence give rise to dominant in-plane interactions whereas the “b” and “c” orbitals having only one component lying in the plane and so contribute dominantly to the out-of-plane interaction. Note that in this paper, for simplicity, we do not include any spin-orbit coupling terms in the TB model Hamiltonian.

The pairing basis functions for triplet superconductivity are the odd-parity functions in $k$-space given by (where for simplicity we have chosen units of length such that the in-plane lattice constant $a = 1$)

$$\sin k_x, \sin k_y \hspace{1cm} (3)$$

and

$$\sin \frac{k_x}{2} \cos \frac{k_y}{2} \cos \frac{k_z c}{2}, \cos \frac{k_x}{2} \sin \frac{k_y}{2} \cos \frac{k_z c}{2}, \hspace{1cm} (4)$$

for in-plane and out-of-plane interactions respectively. The general form of gap-function for an odd-parity triplet state can be represented by a $2 \times 2$ matrix in spin-space as

$$\Delta(k) = \begin{pmatrix} \Delta_{\uparrow\uparrow}(k) & \Delta_{\uparrow\downarrow}(k) \\ \Delta_{\downarrow\uparrow}(k) & \Delta_{\downarrow\downarrow}(k) \end{pmatrix} \hspace{1cm} (5)$$

which can be conveniently written in the form

$$\begin{pmatrix} -d_z(k) + id_y(k) \\ d_z(k) \end{pmatrix} = i[d(k), \sigma] \sigma \hspace{1cm} (6)$$
where the vector \( \mathbf{d}(\mathbf{k}) \) is given by \( \mathbf{d}(\mathbf{k}) = (d_x(\mathbf{k}), d_y(\mathbf{k}), d_z(\mathbf{k})) \) and \( \mathbf{\sigma} = (\sigma_x, \sigma_y, \sigma_z) \) is the vector of Pauli spin matrices.

The Bogoliubov de Gennes (BdG) equation

\[
(\hat{H}_k(r) - \Delta_k(r)) \begin{pmatrix} u_n(k) \\ \nu_n(k) \end{pmatrix} = E_{nk} \begin{pmatrix} u_n(k) \\ \nu_n(k) \end{pmatrix},
\]

(7)
is solved self-consistently at every \( k \)-point. In our TB model, a spin-only magnetic field \( \mathbf{H} = (H_x, H_y, H_z) \) can be added to Eq. (7) by replacing \( \hat{H}_k(r) \) with

\[
\hat{H}_k(r) = H_k(r)\mathbf{\sigma} + \mu_B \mu_0 \mathbf{H},
\]

(8)

\( \mu_B \) being the Bohr magneton and \( \mu_0 \) being the vacuum permeability (in what follows we set \( \mu_0 = 1 \) for convenience).

### A. Pairing vector

As \( \text{Sr}_2\text{RuO}_4 \) has a body-centered tetragonal crystal structure there exist several choices for the \( d \)-vector corresponding to different irreducible representations of the point group symmetry. In this work we consider the form \( \mathbf{d} = (X, Y, 0) \), which corresponds to the \( A_{1u} \) representation and is known as “helical” \( d \)-vector. \( X \) and \( Y \) are the basis functions as described in Eqs. (3) and (4). Following the approach of Ref. [39] and using Eqs. (3), (4) and (5), we can write expressions for the components of matrix in Eq. (5) as follows

\[
\Delta_{aa}^{\sigma \sigma}(\mathbf{k}) = \left( \eta \Delta_{aa}^{\sigma \sigma}(\mathbf{k}) \sin k_x + i \Delta_{aa}^{\sigma \sigma}(\mathbf{k}) \sin k_y \right)
\]

(9)

for in-plane components and

\[
\Delta_{ij}^{\sigma \sigma}(\mathbf{k}) = \left( \eta \Delta_{ij}^{\sigma \sigma}(\mathbf{k}) \sin \frac{k_x}{2} \cos \frac{k_y}{2} + i \Delta_{ij}^{\sigma \sigma}(\mathbf{k}) \cos \frac{k_x}{2} \sin \frac{k_y}{2} \right) \sin \frac{k_z}{2}
\]

(10)

for out-of-plane components where \( ij = bb, cc \) and \( bc \), and \( \eta = +1 \) for \( \sigma = \downarrow \) and \( \eta = -1 \) for \( \sigma = \uparrow \). As previously mentioned, \( a = d_{xy}, b = d_{xz}, \) and \( c = d_{yz} \) represent different orbitals. The coefficients involved are given by

\[
\Delta_{aa}^{\sigma \sigma} = U_{||} \sum_n \int d^3(k) |u_{a,n}^\sigma(k) v_{a,n}^{\sigma*}(k)| \sin k_x f(T, E_n),
\]

\[
\Delta_{ij}^{\sigma \sigma} = 4 U_{\perp} \sum_n \int d^3(k) |u_{a,n}^\sigma(k) v_{b,n}^{\sigma*}(k) + v_{b,n}^{\sigma*}(k) u_{a,n}^{\sigma}(k)| \sin \frac{k_x}{2} \cos \frac{k_y}{2} \cos \frac{k_z}{2} f(T, E_n),
\]

(11)

where \( f(T, E_n) \) is the Fermi function at a temperature \( T \) and eigenvalue \( E_n \) corresponding to the \( n^{th} \) band. Similar relations hold for the \( y \)-components \( \Delta_{aa}^{\sigma \sigma} \) and \( \Delta_{ij}^{\sigma \sigma} \).

Using the above equations, along with the symmetry induced relations

\[
\Delta_{aa}^{\sigma \sigma} = \Delta_{bb}^{\sigma \sigma},
\]

\[
\Delta_{bb}^{\sigma \sigma} = \Delta_{cc}^{\sigma \sigma}/z,
\]

we self-consistently solve Eq. (7). The only unknown constants are the in-plane ad out-of-plane interaction parameters \( U_{||} \) and \( U_{\perp} \). These are chosen such that both the in-plane and out-of-plane components of the zero-field gap-function have a common superconducting critical temperature of 1.5 K. Under this requirement we find

\[
U_{||} = 0.461 t
\]

(12)

\[
U_{\perp} = 0.624 t
\]

(13)

where \( t = 0.08162 \) eV. Fig. [1] illustrates the Fermi surface of \( \text{Sr}_2\text{RuO}_4 \) obtained from our model along with the variation of superconducting gap, obtained by solving the BdG equation self-consistently. The line nodes incorporated into the model are visible on the \( \alpha \) and \( \beta \) sheets where the gap vanishes at \( k_z = \pm\pi/c, c = 12.722 \text{ Å} \) being

![Figure 1](image-url)
Figure 2: (Colour online.) Field dependence of the gap-function at temperatures (a) 0.2 K, (b) 0.6 K, (c) 0.8 K and temperature dependence of the gap-function at fields (d) 0 T, (e) 1.49 T, (f) 2.67 T. Different plots within each panel correspond to the different components of the gap-function as labeled in the legend, where the subscripts of the gap-function denote orbitals as $a = d_{xy}$, $b = d_{xz}$, and $c = d_{yz}$. The superscript refers to the component of the gap-function; we show only the $x$ component and the similar physics holds for the $y$ component. Two clear first-order transitions can be seen in panels (a) and (b) at $H_{p1}$ and $H_{p2}$ that merge into a single superconducting transition in (c). The superconducting transition in (d)-(f) is of second or first order depending upon whether the field $H < H_{p1}$ or $H_{p1} < H < H_{p2}$ respectively.

the lattice constant along $z$-axis. These nodes are a direct consequence of the assumed interlayer pairing interaction acting among the $d_{xz}$ and $d_{yz}$ orbitals which are primarily oriented perpendicular to the plane. In contrast, the $\gamma$ sheet of the Fermi surface predominantly corresponds to the $d_{xy}$ orbital lying in the $x$-$y$ plane. The quasiparticle gap on this sheet has no nodes, but does have deep minima for $k$ in the $(1,0,0)$ and $(0,1,0)$ directions, as shown in Fig. [1]

III. RESULTS AND DISCUSSION

Using the model described in previous section, we numerically solve the BdG equation (Eq. [7]). In the following we divide our presentation of results into three subsections. In Sec. [III A] we study the gap-function as a function of applied magnetic field for a fixed temperature, and as a function of temperature for fixed magnetic field. In this way we build up a magnetic field versus temperature phase diagram for the superconductor. In Sec. [III B] we show the results for specific heat as a function of temperature with fixed magnetic field and vice-versa. Finally Sec. [III C] is dedicated to the study of Knight shift and Sec. [III D] to the variation of polar angle. In each case we compare our results with experiment.

A. Gap-function and phase diagram

One of the key findings of the experiment of Ref. [19] was the emergence of a double superconducting transition below $T = 0.8$ K upon variation of magnetic field. Motivated by this, we study the gap-function as a function of magnetic field (aligned along the [100] direction) in Fig. [2] panels (a)-(c), and as a function of temperature in panels (d)-(f). Different plots within each panel represent the different components of the gap-function as labeled in the legend.

Field sweep at fixed temperature: In panel (a) we see two first order transitions at the lower critical field $H_{p1} = 2.35$ T and the upper critical field $H_{p2} = 2.77$ T, with the temperature fixed at 0.2 K. This feature of a double superconducting transition, in our model, results from different critical fields for the gap-functions on the $d_{xy}$ ($\Delta_{x}^{aa}$) and $d_{xz}/d_{yz}$ ($\Delta_{x}^{bb}/\Delta_{x}^{cc}$) orbitals respectively, represented by $H_{p1}$ for the former and by $H_{p2}$ for the latter. The larger value of $H_{p2}$ implies that whereas the gap-function on $d_{xy}$ orbitals becomes zero at a lower value of the field, it remains finite on the $d_{xz}$ and $d_{yz}$ orbitals until a higher field of $H_{p2}$. When the temperature is increased to a value of 0.6 K in panel (b), the difference between $H_{p2}$ and $H_{p1}$ reduces and the two transitions move closer to each other. When temperature is further raised to $T = 0.8$ K, panel (c), this difference falls to zero which corresponds to a single critical field of
the value $H_p = 2.4$ T. Above $T = 0.8$ K, the superconducting transition is of second order, which will become clearer from the specific heat results in the next section. This temperature of 0.8 K, which we denote $T^*$, matches the temperature reported in Ref. [19] below which a first or second order transition. The bicritical point ($T^*$, $H^*$), the point on the phase diagram where the two critical fields merge into one, is (0.8 K, 2.35 T). Seemingly, the spin-only field controls only the upper critical field as a function of temperature whereas experimental results suggest both $H_{p1}$ and $H_{p2}$ vary significantly with temperature.

To explore this further in Fig. 4 we display the variation of superconducting quasiparticle energy gap on three different bands of the Fermi surface under a magnetic field of $H_z = 2.67$ T. Comparison with Fig. 1 reveals that the gap on the parts of the Fermi surface corresponding to the $d_{xz}$ and $d_{yz}$ orbitals is significantly reduced. On the $\gamma$ sheet, which almost purely consists of the $d_{xz}$ orbitals, it reduces to approximately half of the average value of the original gap. On parts of the $\alpha$ and $\beta$ sheets which are mainly $d_{xz}$ orbital in character, it reduces to a very small value. Interestingly the nodal structure of the field dependent quasiparticle gap shown in Fig. 4 is significantly different from the zero field case seen in Fig. 1 especially on the $\beta$ sheet.

B. Specific heat

Contradicting the expectation of a $C_\text{e}/T$ versus $T$ curve deviating downward near $T_c$ from the linear extrapolation of the data at lower temperatures, an unusual upward-deviation was observed at a field below 1.2 T[20]. While Ref. [19] reported a double peak structure in $C_\text{e}/T$ plots at field values of 1.40 T and 1.42 T. Ref. [20] also studies $C_\text{e}/T$ versus $H$ at fixed temperature, with again a downward deviation of the $C_\text{e}/T$ versus $H$ curve near $H_{p2}$ observed at 0.5 K and 0.7 K and for $H \parallel [100]$, a double-peak structure was reported below $T = 0.8$ K[19]. In Fig. 3 we present our results for the calculations of $C_\text{e}/T$ against $H$ for a range of temperatures. In accordance with the results for the gap-function (Fig. 2),
we find a single phase transition above $T^* = 0.8$ K, and a double peak structure below $T^*$. Our results below $T^*$ are in qualitative agreement with the experimental result\cite{19,20} where we see a upward slope for $C_e/T$ versus $H$ graph near $H_{p1}$ and $H_{p2}$ at low temperatures. An important difference is that we have a larger difference between the upper and lower critical fields as compared to the experiments. As previously mentioned, this is a consequence of the fact that our spin-only field controls only the $H_{p2}$ as a function of temperature leaving $H_{p1}$ almost unchanged.

Turning to variation of the heat capacity with temperature we first consider the zero field case, finding a very good agreement with the experimentally measured specific heat\cite{20} as shown Fig. 6. The feature that at low temperature, specific heat scales linearly with $T$ is a consequence of horizontal line nodes built into our model\cite{20}.

The results for the specific heat calculations at fixed magnetic field are shown in Fig. 7. As the field is increased, $T_c$ decreases with little change in the height of the jump until above the field $H^* = 2.35$ T where the slope of the $C_e/T$ versus $T$ curve increases near $T_c$ and a peak begins to appear. This result is again in accordance with our results of the gap-function and the height of this peak increases with the increase in field. This peak is related to the Pauli paramagnetic effect\cite{19} which results in a first-order transition and can be mathematically understood as arising from the energy derivative term, when the temperature derivative of the energy eigenvalues diverges\cite{20}.

\[
C_v = \sum_{n,k} \left( \frac{k_B}{2} \left( E_{n,k} + \beta \frac{dE_k}{d\beta} \right) E_k sech^2 \frac{\beta E_k}{2} \right). \tag{14}
\]
Figure 7: (Colour online.) Temperature dependence of $C_e/T$ at various values of the applied field $H \parallel [100]$. As the field is increased, a peak begins to develop at $H^* = 2.35$ T, characteristic of first-order transition.

C. Spin susceptibility

The measurement of spin susceptibility has proved to be a useful technique for determination of the internal pairing state of Cooper pairs in superconductors. Contrary to early results\textsuperscript{10,11}, recent results report a very large drop in Knight shift\textsuperscript{27} and in magnetic susceptibility\textsuperscript{37} in the superconducting state as compared to the normal state. This throws into doubt the widely accepted picture of chiral pairing in Sr$_2$RuO$_4$\textsuperscript{47} and leads to the possibility of helical pairing. As in our work we consider a magnetic field which couples only to the spin degree of freedom, we calculate a similar quantity, the spin susceptibility and compare our results with the available experimental data. We plot the ratio of spin moments in the superconducting state to the normal state in Fig. 8. We choose the values of field to be 0.7 T from nuclear magnetic resonance (NMR)\textsuperscript{27} and 0.5 T\textsuperscript{12}, 1 T\textsuperscript{12} from neutron scattering experiments performed on Sr$_2$RuO$_4$. Also, the data from experiments is presented for comparison.

Our results can be closely compared to the NMR experiments as long as our choice of magnetic field lies in the linear-response regime so that

$$K(T) = \frac{\partial M(T)}{\partial H} = \frac{M(T)}{H}$$  \hspace{1cm} (15)$$

holds, where $K(T)$ is the Knight shift measured at temperature $T$ and $M(T)$ is the corresponding spin magnetic moment. As shown in the inset of Fig. 8, the linear-response holds up to a large value of the field of $\approx 1.4$ T. Our results in Fig. 8, where we see a 44% and 46% drops in the $T = 0$ K moment compared to the normal state value at 0.7 T and at 0.5 T respectively are in fair agreement with the experiments. Clearly, our results for 1 T do not agree with the early neutron scattering experiment\textsuperscript{12} where almost no drop in the magnetic moment was measured in the superconducting phase. This suggests that our results can be compared with the neutron scattering data only for low values of the field when Meissner screening effect is weak. Also, as mentioned in Ref. 27, the experimental drop of a few extra percent below 50%, a number limited by the expression for the susceptibility tensor for helical pairing\textsuperscript{44}

\begin{equation}
\hat{\chi}_s(T) = \frac{\chi_n}{2} \text{diag}(1 + Y(T), 1 + Y(T), 2) \hspace{1cm} (16)
\end{equation}

can possibly be captured by Fermi-liquid correction, where $\chi_s, \chi_n$ represent spin susceptibilities in the superconducting and normal state respectively and $Y(T)$ is the Yosida function\textsuperscript{42}.

Further, Ref. 27 presented the Knight shift ratio in the superconducting and normal state as a function of field, at a fixed temperature of 66 mK and the similar ratio from neutron scattering experiment was shown\textsuperscript{37}. In Fig. 9 we present the ratio of spin susceptibilities in the superconducting and normal states ($\chi_s/\chi_n$) with varying field for a range of temperatures, including $T = 66$ mK. As long as Eq. (15) holds, i.e. we are in linear response regime, our results can be compared with the Knight shift ratio from experiment and we see a reasonable agreement. At higher fields, where the linear approximation does not hold, the expected jumps at critical fields $H_{p1}$ and $H_{p2}$ are observed.
D. Varying the polar angle

Ref. [19] also studies the critical field by varying the polar angle between the normal to the \( \text{RuO}_2 \) plane and the direction of the applied magnetic field, reporting a very strong dependence on angle with \( H_{c2} \) reducing sharply with the angle. This effect cannot be explained by helical pairing as it is well known that a field perpendicular to the \( x-y \) plane for a helical \( d \)-vector would leave the gap-function almost unchanged. In Fig. 10 we present the gap-function for \( d_{yz} \) orbitals with a field inclined at angle \( \theta \) with respect to the normal. At \( \theta = 0 \), when the magnetic field is out of plane, the critical field tends to infinity. As \( \theta \) increases, the component of the field in the plane increases as a result of which \( H_c \) decreases and becomes minimal at \( \theta = 90^\circ \). A similar effect is seen for the other components of the gap-function. Correspondingly, the Knight shift will remain unaffected for a choice of \( \theta = 0^\circ \) (See Eq. 16).

IV. DISCUSSION

A thorough study of helical pairing in \( \text{Sr}_2\text{RuO}_4 \) has been made using a realistic 3D tight-binding model, with results compared to experiments where available. Our model based upon helical pairing agrees with many of the experimental observations such as the double superconducting transition, first-order transition to the normal state, and the substantial drop of Knight shifts and magnetic moments in the superconducting phase. Although the temperature \( T^* = 0.8 \) K of the bicritical point on the \( H - T \) phase diagram agrees with experiment, the corresponding experimental values of \( H^* = 1.2 \) T and the upper critical field \( H_{c2} = 1.5 \) T does not agree, with our results for these fields being 2.35 T and 2.67 T respectively. Furthermore, the temperature dependence of \( H_{c1} \) also differs from experiments with our results showing a much weaker dependence.

These differences can likely be attributed to the orbital contribution to the critical field, which we do not include in our model. The orbital limit of the upper critical field can be estimated using the Wethamer-Helfand-Hohenberg (WHH) formula as \( H_{c2}^{orb}(0) = -0.75|dH_{c2}/dT|T_c/T_c \). This formula, applied to \( \text{Sr}_2\text{RuO}_4 \), gives a value of 3.3 T which would correspond to a value of \( H_{c2} \) if the superconductivity was orbitally limited, significantly larger than the experimental value of 1.5 T. This strongly indicates that the superconductivity in \( \text{Sr}_2\text{RuO}_4 \) is Pauli limited. Nevertheless, vortex lattice contribution to critical fields can not be ignored. Furthermore, it needs to be stressed that in our calculation we assumed that the Cooper pairs have a net zero momentum thereby excluding the possibility of FFLO phase at high field, as found, for example, in \( \text{CeCoIn}_5 \), a Pauli-limited heavy-fermion superconductor. Inclusion of orbital effects may also ameliorate the disagreement with experiment for the dependence of upper critical field on polar angle.

We should also stress that our model does not support experiments which show that TRS is broken in the superconducting phase. The in-plane anisotropy of \( H_{c1} \) and \( H_{c2} \) measured via ac susceptibility studies is obviously missing from our model since we consider a spin-only magnetic field. In general helical pairing states preserve TRS, unlike the chiral state. This is a direct consequence of spin-orbit coupling, which implies that the four states of helical type are non-degenerate.
The possible realisation of any specific pairing TRS state such as this will depend on details of the pairing interaction as well as the strength of spin-orbit coupling, which requires further study. It also seems plausible that TRS breaking states of the form \( d = (X, iY, 0) \), each corresponding to one of the 1d irreducible representations \( A_1u, A_2u, B_1u, B_2u \) of the \( D_{4h} \) point group. However in the absence of spin-orbit coupling they all derive from the \( E_u \) irreducible representation of the tetragonal point group and among the distinct pairing states allowed are TRS breaking states of the form [2]

\[
d = (X, Y, 0)
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(17)

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(18)

search for the internal pairing symmetry of the Cooper pairs. Improvements to the model could include coupling between different orbitals due to the addition of spin-orbit coupling which may affect the \( H - T \) phase diagram and which will be the subject of our next work. However the possibility of other types of singlet pairings such as d-wave or extended s-wave can not be ruled out [3], in particular since the sharp variation of \( H_{p2} \) with polar angle cannot be explained with helical pairing. Further experiments on the NMR measurements with a field applied along \( z \)-axis can help resolve the issue to some extent since no drop in Knight shift is expected for a helical pairing and such an observation would rule out any possibilities of singlet \( s \) or \( d \)-wave pairing.

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