Orbital magnetic moment of a chiral p-wave superconductor

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Abstract. The existence and magnitude of a bulk orbital angular momentum of the chiral condensate in the A phase of superfluid helium-3 is a longstanding matter of controversy. The analogous problem in a chiral p-wave superconducting material is the existence of a finite orbital magnetic moment in the bulk. In Sr$_2$RuO$_4$, the existence of such an orbital moment is strongly suggested by experimental evidence for spontaneous time-reversal symmetry breaking (TRSB) in the superconducting state, but the theories disagree on the expected magnitude of this moment. We show that a nonzero orbital magnetization density arises naturally in a realistic band model for Sr$_2$RuO$_4$, and its temperature dependence is qualitatively similar to those of the muon spin rotation and Kerr effect experimental results. The simplest model that leads to the orbital moment requires at least two degenerate atomic orbitals per Ru, which correspond to the Ru d$_{xz}$ and d$_{yz}$ states. This is in contrast to the theories of orbital angular momentum in the isotropic superfluid $^3$He, or models of orbital moment in Sr$_2$RuO$_4$ which assume only a single band at the Fermi level. The implications of this surprising result are explored.

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1. Introduction

The superconductor Sr$_2$RuO$_4$ is probably the strongest candidate available currently for a spin-triplet p-wave paired superconductor [1, 2]. The material is superconducting below $T_c \approx 1.5$ K and has a tetragonal crystal structure, which is an analogue of the high-$T_c$ parent compound La$_2$CuO$_4$, where the Cu sites are occupied by Ru ions. These Ru$^{2+}$ ions lead to a three-sheet Fermi surface (with $\alpha$, $\beta$ and $\gamma$ sheets) which has been well mapped out by de Haas experiments [3]. These three sheets arise from hybridization between oxygen and the Ru d orbitals: the $d_{xz}$ and $d_{yz}$ orbitals giving the $\alpha$ and $\beta$ sheets and the $d_{xy}$ orbital giving the $\gamma$ sheet.

In particular, it is believed that the pairing state in this material is a chiral state, with a gap parameter of the form

$$d(k) \propto (\sin k_x + i \sin k_y)\hat{e}_z, \quad (1)$$

where $\mathbf{k} = (k_x, k_y, k_z)$ is the wave vector in the body-centred tetragonal Brillouin zone and $\hat{e}_z$ is a unit vector along the tetragonal crystal $c$-axis. On the three cylindrical Fermi surface sheets of Sr$_2$RuO$_4$ such a symmetry state is a two-dimensional analogue of the ABM state of superfluid $^3$He-A [4, 5].

The purpose of this paper is to further explore the analogy between this chiral pairing state and the ABM state in superfluid $^3$He-A. Specifically we address questions of the magnitude and temperature dependence of the orbital magnetic moment expected in the bulk of this material. The existence of such a moment may be expected because of the presence of the $i$ in equation (1), implying that time-reversal symmetry is spontaneously broken in the superconducting state. Direct experimental evidence for this fact comes both from muon spin rotation ($\mu$-SR) experiments [6] and from the optical Kerr effect [7].

The central issue, however, is that the theory of the orbital magnetic moment in a spin triplet superconductor is analogous to the question of the orbital angular momentum in the chiral ABM phase of superfluid $^3$He-A. The correct calculation of this total orbital angular momentum in the superfluid has been debated over many years, in fact since 1961 [8], and is still not fully resolved. Predictions of the ‘ideal’ (ignoring effects of vortices or surface textures) total orbital angular momentum of the condensate in a chiral superfluid range over six orders of magnitude between numbers of order $\hbar$ per Cooper pair to $(\Delta/s_F)^2\hbar$ where $\Delta$ is the gap and $s_F$ is the Fermi energy [5].

Naively taking these superfluid $^3$He-A results over to the case of chiral superconductivity in Sr$_2$RuO$_4$ we might expect a condensate induced spontaneous orbital magnetic moment of...
magnitude either \( 1 \times \mu_B \) or \( 10^{-6} \times \mu_B \) per unit cell, respectively, where \( \mu_B \) is the Bohr magneton \( \mu_B = \frac{e\hbar}{2m_e} \) in SI units. The former estimate would make the superconducting condensate of Sr\(_2\)RuO\(_4\) almost as magnetic as iron! On the other hand, the lower value might perhaps be more consistent with the rough order of magnitude which might be expected from the extremely small measured optical Kerr angle of the order of \( 10^{-8} \) radians [7].

Below, we present a calculation of the orbital magnetic moment in a realistic three-band model of Sr\(_2\)RuO\(_4\). Within our model we find that the zero temperature moment is roughly consistent with the estimate \( 10^{-6} \times \mu_B \) per Ru, rather than any much larger value. We also find that the temperature dependence of the moment is consistent with an expression of the form \( (\Delta(T)/\Delta(0))^2 \), similar to the temperature dependences observed in both \( \mu \)-SR [6] and Kerr effect [7] experiments.

A surprising theoretical feature of our calculation is that we use a simple atomic-like expression for the orbital moment per Ru ion, which differs fundamentally from the earlier work in superfluid \(^3\)He-A. A key feature of our atomic estimate is that the orbital moment arises from coupling of \( d_{xz} \) and \( d_{yz} \) symmetry Wannier orbitals associated with the \( \alpha \) and \( \beta \) Fermi surface sheets. In this formalism, the \( \gamma \) band arising from the \( d_{xy} \) symmetry Wannier states makes no on-site contribution to the orbital moment. We discuss this very surprising prediction in the light of a new and very general theory for orbital magnetism in metals, insulators and Chern insulators, which has been introduced by Thornhausser et al [9] and Ceresoli et al [10].

2. The angular momentum paradox in superfluid \(^3\)He-A

A longstanding and surprisingly difficult problem in the theory of superfluid helium-3, is the existence and magnitude of a finite orbital angular momentum of the condensate in the chiral A phase [5, 11]. The p-wave superfluid gap function in this phase can be written as

\[
d(k) = \frac{\Delta}{k_F}(k_x + ik_y)\hat{e}_z,
\]

where \( \Delta \) is the maximum value of the gap on the spherical Fermi surface, and \( k_F \) is the Fermi momentum. In this state, each Cooper pair has an internal angular momentum \( l_z = \hbar \) relative to its centre of mass. Therefore at zero temperature, it may be expected that the total \( z \)-component of the angular momentum of a macroscopic sample of \( N \) particles state should be \( \langle L_z \rangle = N\hbar/2 \) relative to the common centre of mass [12, 13]. This would correspond to the expectation for a Bose–Einstein condensate (BEC) of \( N/2 \) ‘molecules’ each with \( l_z = \hbar \). At higher temperatures, it is expected that this spontaneous angular momentum would decrease, in the same way as the superfluid density, to become zero at the critical temperature \( T_c \) [14, 15].

On the other hand, there are reasons to believe that the total angular momentum of such a chiral condensate will be considerably reduced from this ideal value, possibly by many orders of magnitude. The orbital angular momentum was originally predicted by Anderson and Morel [8] to be a factor of \( \Delta/\epsilon_F \) smaller than the ideal value of \( N\hbar/2 \), where \( \epsilon_F \) is the Fermi energy. In superfluid \(^3\)He this factor is of the order of \( 10^{-3} \). This result was obtained by considering the pair correlations in the superfluid state. Each particle attracts fluctuating Cooper pair partners to its vicinity, within a typical range of the coherence length \( \xi_0 \), and carrying one unit of orbital angular momentum. However, at any point in the bulk of the superfluid, the orbital currents from nearby Cooper pairs should cancel, and the only remaining contributions would be from the surface region of the order of the coherence length in thickness, leading to the substantially
reduced total orbital angular momentum expected to be of the order of $\xi_0 k_F \sim \Delta / \epsilon_F$ smaller than $N \hbar / 2$.

Other calculations predicted an even smaller angular momentum density, of the order of $(\Delta / \epsilon_F)^2$ times $N \hbar / 2$ per particle, corresponding to $10^{-6} \hbar / 2$ per particle in the A-phase of superfluid $^3$He. Cross [16] first obtained this result by considering the effects of particle–hole symmetry, which lead to cancellation of hole and particle contributions to the orbital current. The only residual current contributions which remain are of the order of $(\Delta^2 / \epsilon_F)^2$ arising from the small particle–hole asymmetry in the quasiparticle states within the gap energy $\pm \Delta$ at the Fermi energy $\epsilon_F$. Similar results were obtained by Volovik and Mineev [17] and by Balatskii and Mineev [18]. Gaitan [19] showed that the same result could be obtained as a consequence of a near exact cancellation between the bare quasiparticle contribution, of the order of $\hbar / 2$ per particle, and a collective mode backflow current.

More recent results show that the surface contribution is much larger than that proposed by Anderson and Morel [8]. Essentially there is a topological edge current, which is capable of carrying the full orbital angular momentum of $N \hbar / 2$ of the fluid [20, 21]. This theoretical expectation has been demonstrated by explicit numerical solutions of the Bogoliubov–de Gennes equations for a chiral condensate of Fermi particles in a circular harmonic trap [11]. These calculations not only confirmed the prediction $\langle \hat{L}_z \rangle = N \hbar / 2$ at zero temperature, but clearly show that the total angular momentum density is a sum of both bulk and surface contributions. The relative sizes of these two contributions is different in the case of the harmonic trap compared to the hard wall boundary conditions of Kita [13], but in both cases lead to the same total orbital moment of $\langle \hat{L}_z \rangle = N \hbar / 2$.

### 3. Orbital magnetism in crystals

When we consider the corresponding orbital magnetic moment of a chiral superconductor there is a further complication to add to this debate, such as the presence of the underlying crystal lattice. Therefore there is no continuous rotational symmetry in the Fermi surface, and the $N$-body wave function, $\Psi^N(\mathbf{r}_1 \ldots \mathbf{r}_N)$, will not be an eigenstate of the total $N$-particle angular momentum operator $\hat{L}_z^N = \sum_i \hat{L}_z^i = \sum_i [\mathbf{r}_i \times \hat{p}_i]$. To the extent that the electron band structure near to the Fermi surface is near to jellium-like, leading to an isotropic spherical or cylindrical Fermi surface, there may be an effective conservation of angular momentum of electron states near to the Fermi surface, but this will not be an exact symmetry and it is therefore not at all obvious how to define an orbital angular momentum or orbital magnetic moment. This problem applies equally to discussions of the orbital magnetic moment in normal metals and insulators, as well as in superconductors.

On the other hand, the orbital magnetism of metals and insulators is a well-known phenomenon. In relativistic band structure theory, the periodic Bloch wave solutions are found for the Dirac equation [22]. Spin–orbit coupling implies that any magnetic solutions of the relativistic Kohn–Sham equations have both spin and orbital contributions to the total magnetic moment. These separate contributions can be measured experimentally by a number of techniques, including for example, x-ray circular dichroism [23, 24] and x-ray resonant Raman scattering [25].

In the relativistic band theory, the orbital magnetic moment is usually simply defined in terms of a simple atomic-like angular momentum operator applied to each ion site in the crystal. For example, Bruno [26] derived expressions for orbital moment and magneto-crystalline
anisotropy using perturbation theory in the atomic spin–orbit coupling interaction \( \lambda \mathbf{L} \cdot \mathbf{S} \). Matrix elements of this operator are easily found within the tight-binding approximation for the Wannier functions of the electron bands.

However, this seemingly straightforward definition of orbital moment hides an important problem. At the atomic sites located at \( \mathbf{R}_s \) in the crystal, we can define a set of Wannier orbitals \( |w_{ni}\rangle \equiv w_n(\mathbf{r} - \mathbf{R}_i) \), so that the (nonrelativistic) Bloch states are given by

\[
\psi_{nk}(\mathbf{r}) = \frac{1}{N_s^{1/2}} \sum_{j} e^{i\mathbf{k} \cdot \mathbf{R}_j} w_n(\mathbf{r} - \mathbf{R}_i),
\]

where \( N_s \) is the number of atomic sites, \( \mathbf{R}_s \), in the solid. For simplicity we implicitly include the spin label \( \sigma \) of the Bloch states into the band index, \( n \). The total orbital angular momentum relative to a specific site \( \mathbf{R}_0 \) could be defined as

\[
\langle \hat{L}_z \rangle_0 = \langle \Psi^N | \sum_{i=1, N} \left( [(\mathbf{r}_i - \mathbf{R}_0) \times \hat{\mathbf{p}}]_z \right) |\Psi^N \rangle
\]

\[
= \sum_{nk} \langle \psi_{nk} | [(\mathbf{r} - \mathbf{R}_0) \times \hat{\mathbf{p}}]_z |\psi_{nk} \rangle
\]

\[
= \sum_{nk} \frac{1}{N_s} \sum_{ij} e^{i\mathbf{k} \cdot (\mathbf{R}_j - \mathbf{R}_i)} \langle w_{ni} | [(\mathbf{r} - \mathbf{R}_0) \times \hat{\mathbf{p}}]_z |w_{nj}\rangle,
\]

where the sum over bands and spin, \( n \), and Bloch states, \( \mathbf{k} \), is carried out over the occupied Fermi states comprising the filled Fermi sea, \( \epsilon_{nk} < \epsilon_F \) where \( \epsilon_F \) is the Fermi level.

We now use the trivial identity \( \langle \hat{p}_z | w_{nj} \rangle = \sum_{i} [\langle \mathbf{r} - \mathbf{R}_0 | \hat{\mathbf{p}}]_z |w_{nj}\rangle \] to shift the origin of the coordinates to \( \mathbf{R}_j \). This leads to

\[
\langle \hat{L}_z \rangle_0 = \sum_{nk} \left\{ \sum_{i} e^{i\mathbf{k} \cdot (\mathbf{R}_j - \mathbf{R}_i)} \langle w_{ni} | [(\mathbf{r} - \mathbf{R}_j) \times \hat{\mathbf{p}}]_z |w_{nj}\rangle + \frac{1}{N_s} \sum_{ij} [(\mathbf{R}_j - \mathbf{R}_0) \times \langle w_{ni} | \hat{\mathbf{p}}]_z |w_{nj}\rangle \right\}
\]

after using translational invariance to sum over the \( N_s \) equivalent sites \( j \).

The first term in equation (5) is now independent of the centre of rotation \( \mathbf{R}_0 \), and is clearly an extensive number proportional to the total number of atomic sites in the crystal. Therefore this term is one possible definition of an orbital moment per unit cell

\[
\langle \hat{L}_z \rangle = \sum_{n} \int \frac{d^3k}{(2\pi)^3} \sum_{i} e^{i\mathbf{k} \cdot (\mathbf{R}_j - \mathbf{R}_i)} \langle w_{ni} | [(\mathbf{r} - \mathbf{R}_j) \times \hat{\mathbf{p}}]_z |w_{nj}\rangle
\]

in the bulk. The usual procedure [26] for evaluating the orbital moment is then seen to be equivalent to taking only the site diagonal matrix elements, \( i = j \), of the Wannier functions in the first term in equation (5), giving

\[
\langle \hat{L}_z \rangle \approx \sum_{n} \int \frac{d^3k}{(2\pi)^3} \langle w_{nj} | [(\mathbf{r} - \mathbf{R}_j) \times \hat{\mathbf{p}}]_z |w_{nj}\rangle.
\]

For a choice of Wannier functions which resemble atomic orbitals, this will then define an orbital moment for each atomic orbital in the cell, and hence a corresponding total atomic orbital moment. Clearly in the limit of zero bandwidth (large lattice spacing) the Wannier functions \( |w_{nj}\rangle \) reduce to molecular spin–orbitals, and this expression yields the standard usual free atom orbital angular momentum and the corresponding magnetic moment per atom.

New Journal of Physics 11 (2009) 055063 (http://www.njp.org/)
Figure 1. The projection of the full three-dimensional Fermi surface onto the $k_z = 0$ plane. The width of the lines describes the corrugation of the Fermi surface along the $k_z$ direction. The spin–orbit coupling is chosen as $\lambda = -0.08t$.

The above orbital density should be a good approximation provided the Wannier functions are well localized on each site. However, we have neglected the term in equation (5) which depended on the centre of coordinates, $R_0$, about which the angular momentum is determined. This term might be expected to be small, but in general the matrix elements

$$\langle w_{ni}|\hat{p}|w_{nj}\rangle$$

are nonzero when there are macroscopic circulating currents. Thus for bulk metals, there is no guarantee even that these matrix elements will be small. Indeed, the effective moment grows with the distance from the centre of rotation, $|R_j - R_0|$, and so these terms can become arbitrarily large in a macroscopic sized sample. In particular, in light of the discussion above of the surface edge current effect in the chiral state of superfluid $^3$He, it is clear that the total orbital magnetic moment of a finite size sample will be the sum of both the bulk contributions, equation (6), and surface contributions which might be much larger than the bulk terms.

4. Model of the chiral state in Sr$_2$RuO$_4$

Before presenting the calculation of the orbital moment in Sr$_2$RuO$_4$ we shall first briefly summarize our phenomenological model of the chiral pairing state in this material. The model is based upon a realistic three-dimensional fit to the three Fermi surface sheets $\alpha$, $\beta$ and $\gamma$. We now discuss this fit to the Fermi surface, before briefly summarizing our model. Full details of the model and comparisons of its predictions with experimental results have been presented elsewhere [27]–[30].

The measured Fermi surface [3] consists of three slightly warped cylinders parallel to the tetragonal crystal $c$-axis. Our tight-binding fit is shown in figure 1. The largest electron-like sheet, $\gamma$ derives from the Ru d$_{xy}$ orbitals and is well represented by a two-dimensional tight-binding band involving nearest neighbour hopping, $t$, and next nearest neighbour hopping, $t'$ integrals. The states in this band are even in the mirror symmetry of the RuO$_2$ plane, and
so it is largely decoupled from the odd symmetry bands derived from the Ru \( d_{xz} \) and \( d_{yz} E_g \) doublet. Hopping between nearest neighbour Ru sites leads to two distinct hopping parameters between neighbouring \( d_e \) orbitals, either for hopping in the \( x \)-direction, \( t_{xx} \), or for hopping in the \( y \)-direction, \( t_{yx} \). The first allowed hopping between \( d_{xz} \) and \( d_{yz} \) orbitals occurs across the diagonal of the unit cell, at second neighbour, \( t_{ab} \). The resulting tight-binding band structure leads to two quasi-one-dimensional sheets which nearly touch, giving a slightly square shape to the \( \alpha \) and \( \beta \) Fermi surface sheets, as shown in [28]. A small out of plane hopping leads to a slight warping of these three Fermi surface sheets [28], which is comparable to the warping observed experimentally [3].

When spin–orbit interactions are included on the Ru sites, the tight-binding band structure must be supplemented by an additional term [31]

\[
H^\sigma = \frac{i\lambda}{2} \sum_{i,\sigma \sigma', mm'} \varepsilon^{\kappa mm'} \sigma_{\kappa \sigma} \sigma_{\kappa \sigma'} c^{\dagger}_{i mm'} c_{j mm'},
\]

where \( m \) and \( m' \) denote the Ru \( d \) orbitals, \( \sigma_{\kappa \sigma}, \kappa = x, y, z, \) 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 \( m = (d_{yz}, d_{xz}, d_{xy}) \) [27]. It is worth noting that the shape of the Fermi surface and relative positions of it various sheets depend on the spin–orbit coupling \( \lambda \). Figure 1 shows the projection of full Fermi surface calculated for \( \lambda = -0.08t \). Because of the small size of \( \lambda \) the overall shape is visually the same as that obtained for a smaller spin–orbit coupling of \( \lambda = -0.02t \) [30]. In order to avoid completely refitting the experimental Fermi surface for each different value of the spin–orbit coupling parameter \( \lambda \), our calculations make use of relatively small values in the range \( \lambda = -0.02t \) to \(-0.08t \).

To model the superconducting state of \( \text{Sr}_2\text{RuO}_4 \) we consider a simple multi-band attractive \( U \) Hubbard model

\[
H = \sum_{ij mm', \sigma} \left( (\varepsilon_m - \mu) \delta_{ij} \delta_{mm'} - t_{mm'}(ij) \right) \hat{c}^{\dagger}_{i mm' \sigma} \hat{c}_{j mm' \sigma} - \frac{1}{2} \sum_{ij mm' \sigma \sigma'} U_{mm'}^{\sigma \sigma'}(ij) \hat{n}_{i \sigma} \hat{n}_{j \sigma'},
\]

where \( i \) and \( j \) label the sites of a body-centered tetragonal lattice of Ru sites and the hopping integrals \( t_{mm'}(ij) \) and site energies \( \varepsilon_m \) correspond to the tight-binding band fit described above. 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 \( m' \), and are chosen [27]–[29] in such a way as to reproduce the experimental superconducting transition temperature of \( T_c = 1.5 \) K.

With this model Hamiltonian we make the Hartree–Fock–Gorkov approximation and numerically solve the coupled set of Bogoliubov–de Gennes equations [32]

\[
\sum_{jm' \sigma'} \begin{pmatrix} E^N - H^{\sigma \sigma'}_{mm'}(ij) & \Delta^{\sigma \sigma'}_{m, m'}(ij) \\ \Delta^{\sigma \sigma'}_{m, m'}(ij) & E^N + H^{\sigma \sigma'}_{mm'}(ij) \end{pmatrix} \begin{pmatrix} u_{jm' \sigma}^N \\ v_{jm' \sigma}^N \end{pmatrix} = 0.
\]

Using translational symmetry we Fourier transform (10) and get the gap equations in \( k \) space for energies \( E^N(k) \) and quasiparticle amplitudes \( (u^N(k), v^N(k)) \), where the index \( N \) denotes both band and spin indices.

In our three-dimensional model [28] the chiral state has the following explicit form of the pairing potential matrix [32]:

\[
\hat{\Delta}_{mm}(k) = \Delta_{mm}(T) \begin{pmatrix} 0 & \sin k_x + i \sin k_y \\ \sin k_x + i \sin k_y & 0 \end{pmatrix}
\]

[New Journal of Physics 11 (2009) 055063 (http://www.njp.org/) ]
for the Ru d$_{xy}$ orbital (in-plane state) and, 
\[ \hat{\Delta}_{mm'}(\mathbf{k}) = \Delta_{mm'}(T) \begin{pmatrix} 0 & X + iY \\ X + iY & 0 \end{pmatrix} \] (12)

for $m, m'$ equal to the Ru d$_{xz}$ and d$_{yz}$ orbitals (out-of-plane state), where here $X = \sin \frac{k_x}{2} \cos \frac{k_y}{2} \cos \frac{k_z}{2}$ and $Y = \cos \frac{k_x}{2} \sin \frac{k_y}{2} \cos \frac{k_z}{2}$ are the basis functions of the Eu symmetry representation corresponding to the assumed interlayer interaction between Ru d$_{xz}$ and d$_{yz}$ orbitals [27]. It is the cos $\frac{k_z}{2}$ term in both $X$ and $Y$ which is responsible for line nodes at the $\alpha$ and $\beta$ Fermi surfaces in this model. It is worth noting that in this model all bands become superconducting simultaneously at $T_c$ [27, 28]. Therefore there is no assumption of ‘interband proximity effect’ [33] in our picture.

5. Ruthenium orbital moment in Sr$_2$RuO$_4$

We now apply this model of the pairing state and the general procedure outlined above to calculate the orbital moment per Ru ion in the bulk of a homogeneous sample of superconducting Sr$_2$RuO$_4$. We concentrate below on the orbital moment density in the bulk of a chiral superconductor, based on equation (6), rather than the alternative question of calculating the total magnetic moment of a finite superconducting sample. Clearly, as in the case of superfluid $^3$He-A these quantities are possibly quite different and are obtained by very different theoretical approaches. We therefore neglect for the moment any issues of edge currents [34] or their contribution to the total moment of a finite sample of Sr$_2$RuO$_4$.

In terms of orbital $m$ and spin $\sigma$ quantum numbers, we write the orbital momentum operator [29]
\[ \hat{L}_z = \sum_{m\sigma} \sum_{m'\sigma'} L_{mm'}^{\sigma\sigma'} c_{m\sigma}^{\dagger} c_{m'\sigma'} , \] (13)
where
\[ L_{mm'}^{\sigma\sigma'} = \langle m\sigma | -i\hbar \frac{\partial}{\partial \varphi} | m'\sigma' \rangle \] (14)
and calculate its thermodynamic average $\langle \hat{L}_z \rangle$ using the known properties of spherical harmonics. The result is
\[ \langle \hat{L}_z \rangle = -i\hbar \left\{ n_{ab}^{\uparrow\uparrow} + n_{ba}^{\downarrow\downarrow} - (n_{ab}^{\uparrow\downarrow} + n_{ba}^{\down\uparrow}) \right\} \] (15)
with $a = d_{yz}$, $b = d_{zx}$ and the single-particle density matrix at each Ru site is
\[ n_{mm'}^{\sigma\sigma} = \frac{1}{N_s} \sum_k \langle c_{k\sigma\sigma}^{\dagger} c_{k\sigma'\sigma} \rangle . \] (16)
This single-particle density matrix must be evaluated with respect to the Hartree–Fock–Gorkov many-particle state of the superconductor. Using the fact that it is a single-particle operator, this can be expressed in terms of solutions of Bogolubov–de Gennes equation as
\[ n_{mm'}^{\sigma\sigma} = \frac{1}{N_s} \sum_k \sum_N \left[ u_N^{m\sigma} (\mathbf{k}) u_N^{m'\sigma} (\mathbf{k}) f (E_N (\mathbf{k})) + v_N^{m\sigma} (\mathbf{k}) v_N^{m'\sigma} (\mathbf{k}) (1 - f (E_N (\mathbf{k}))) \right] . \] (17)

Figure 2 shows the temperature-dependent orbital angular momentum obtained after evaluating this density matrix numerically using the pairing state defined by equations (11)
Figure 2. Temperature dependence of the normalized orbital moment \( \text{Sr}_2\text{RuO}_4 \) in chiral state of \( \text{Sr}_2\text{RuO}_4 \) calculated according to equation (15). Also shown are \( \Delta(T)/\Delta(0) \) and \( (\Delta(T)/\Delta(0))^2 \).

and (12). We find that the calculated angular momentum per Ru ion is of the order of \( 2.5 \times 10^{-6} \hbar \) at \( T = 0 \) K. The orbital angular momentum vanishes at \( T = T_c \), as shown in figure 2, where the temperature dependence of \( \langle \hat{L}_z(T) \rangle \) (normalized to its zero temperature value \( \langle \hat{L}_z(0) \rangle \)) is shown together with the normalized values of \( \Delta(T)/\Delta(0) \) and \( (\Delta(T)/\Delta(0))^2 \). Clearly as can be seen from the figure, the orbital angular momentum varies approximately as \( (\Delta(T)/\Delta(0))^2 \).

An interesting feature of the expression, equation (15) is that the Ru orbital moment arises only from the \( a \) and \( b \), atomic orbitals, i.e. \( d_{xz} \) and \( d_{yz} \). These orbitals are the ones which give rise to the \( \alpha \) and \( \beta \) Fermi surface sheets. Therefore within our formalism there is no local on-site orbital moment arising from the \( d_{xy} \) orbitals or the \( \gamma \) band. As noted above, our model has a similar magnitude of the gap \( \Delta \) on all three Fermi surface sheets, and all components vanish together at \( T_c \) [27, 29]. On the other hand, the ‘interband proximity effect model’ would predict a different temperature dependence on the \( \alpha–\beta \) sheets compared to the dominant \( \gamma \) sheet [33]. The experimental temperature dependencies of both the \( \mu \)-SR signal [6] or the Kerr effect [7] appear also to follow the functional form \( (\Delta(T)/\Delta(0))^2 \), consistent with the temperature dependence in figure 2. It is also worth noting that in the normal state of \( \text{Sr}_2\text{RuO}_4 \) the off-diagonal terms of the density matrix, \( n_{\sigma\sigma}^{ab} \), are zero by symmetry. They are also zero when evaluated relative to non-chiral p-wave pairing states, such as the two-dimensional analogues of the \(^3\text{He-B} \) phase \( d(k) \sim (k_x, k_y, 0) \) [29]. Therefore the appearance of these off-diagonal components in the on-site density matrix, and hence the orbital moment, is a unique signature of the chiral pairing state.

The spin–orbit coupling term, equation (8), couples the \( d_{xz} \) and \( d_{yz} \) orbitals most strongly to \( \hat{L}_z \), and so it is interesting to ask how the predicted Ru moment depends on the strength of the spin–orbit coupling parameter \( \lambda \). We find that the zero temperature value of the moment \( \langle \hat{L}_z(0) \rangle \) does depend weakly on the spin–orbit parameter \( \lambda \), as is presented in table 1. However, it is clear from the table that the moment remains finite even in the absence of spin–orbit coupling.
Table 1. The dependence of the zero temperature Ru orbital moment on the spin–orbit coupling $\lambda$.

| $\lambda/t$ | $\langle \hat{L}_z \rangle [10^{-6}\hbar]$ |
|------------|------------------------------------------|
| -0.08      | 2.452                                    |
| -0.04      | 2.676                                    |
| -0.02      | 2.638                                    |
| 0.0        | 2.559                                    |
| 0.02       | 2.639                                    |
| 0.04       | 2.672                                    |
| 0.08       | 2.458                                    |

when $\lambda = 0$. This is different from the usual case of orbital magnetism in metals, in which the orbital moment arises from a combination of a nonzero spin-moment $\langle \hat{S} \rangle$ and spin–orbit coupling $\lambda \hat{L} \cdot \hat{S}$ [26]. Here the spin moment is zero and the orbital moment arises solely from the chiral state of the Cooper pairs.

6. Discussion and future directions

We have calculated the magnitude and temperature dependence of the orbital moment on a Ru site in the chiral superconducting state of Sr$_2$RuO$_4$. These results resemble qualitatively the behaviour seen in $\mu$-SR [6] and optical Kerr effect [7] measurements, namely that the orbital moment is small (many orders of magnitude smaller than $\sim 1 \times \mu_B$ per Ru site) and changes with temperature approximately as $(\Delta(T)/\Delta(0))^2$. A surprising feature of the calculations is that in the theoretical calculations of the Ru ion orbital angular momentum operator, $\langle \hat{L}_z \rangle$, it is only the Ru d$_{xz}$ and d$_{yz}$ orbitals (corresponding to the $\alpha$ and $\beta$ bands) which contribute. Any contributions to the orbital moment from the d$_{xy}$ orbitals (corresponding to the $\gamma$ band) have zero matrix elements on the Ru site, even though it is generally expected that this band is the dominant one for the superconductivity [33]. Therefore the bulk orbital moment that we have obtained is arising only from circulating currents within the unit cell associated with the Ru d$_{xz}$ and d$_{yz}$ orbitals. This is surprising, but is not necessarily inconsistent with the expectation from the theory of the orbital angular momentum in superfluid $^3$He-A. Regarding the $\gamma$ band as essentially jellium-like, we can expect its total orbital angular momentum to reside essentially entirely on the surface, as is found to be the case in superfluid $^3$He-A [11, 13]. This is consistent with the predicted existence of a chiral edge current in Sr$_2$RuO$_4$ [34]. Our work evaluates only the bulk contributions to the orbital moment and we have neglected any surface contributions arising from the edge currents.

In our calculations, we have made use of a particular form of the angular momentum operator for a Ru site, equation (7). This is consistent with previous definitions of orbital magnetism in crystals [26] but, as noted above, this definition of the local orbital moment operator is not unique. The difficulties of the proper definition of the magnetization in insulators have recently been overcome, at least in the case of insulators for which it is possible to define exponentially localized Wannier functions. The key idea is to replace the ill-defined matrix elements of the $r$ operator by well-defined quantities defined in reciprocal space. In particular, if $u_{nk} = e^{-ik\cdot r}\psi_{nk}$ is the periodic part of the Bloch function, then the operator $-i\nabla_k$ acting on $u_{nk}$ plays a role similar to $r$ acting on the Wannier function. The proposal by Thornhauser et al [9] and Ceresoli et al [10] is that the following expression may be used for the orbital moment of a metal, insulator or Chern insulator

$$M = \frac{1}{2\pi} \Im \sum_n \int_{\epsilon_{nk} \leq \mu} d^3k (\nabla_k u_{nk} | \times (\hat{H}_k + \epsilon_{nk} - 2\mu) | \nabla_k u_{nk}),$$

(18)
where $u_{nk}$ is the periodic part of the Bloch state $\psi_{nk}$, the band energy is $\epsilon_{nk}$ and $\hat{H}_k = e^{-ik\cdot r} \hat{H} e^{ik\cdot r}$ where $\hat{H}$ is the single-particle crystal Hamiltonian. This formula has been derived rigorously for insulators [9] and is conjectured to be correct for the orbital moment in a metal [10]. The justification of equation (18) is provided by the semiclassical arguments of Xiao et al [35]. The first fully quantum mechanical derivation of the orbital magnetization in metals was given by Shi et al [36]. These authors used thermodynamic arguments to obtain the formula (18) by calculating the lowest order change of the thermodynamic potential due to an external magnetic field.

In the case of a chiral p-wave superconductor, the analogous question concerns both the orbital angular momentum of the Cooper pairs, and the corresponding orbital magnetic moment which will arise from the circulating flow of charged quasiparticles. In other words, when the Bogoliubov–de Gennes equations are solved self-consistently in the presence of a magnetic vector potential $A$ there will be contributions to the response arising both from the internal angular momentum within each Cooper pair, as well as contributions from the overall diamagnetic response of the condensate arising from macroscopic circulating supercurrents [37]. It would be very interesting to generalize the single-particle expression for the orbital moment, equation (18), to obtain the corresponding orbital magnetization arising from the quasiparticles in a chiral superconductor. In principle, this should be done within the framework of the fully relativistic theory of superconductivity [38, 39]. We leave this important problem for future work.

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