Is the anisotropy of the upper critical field of Sr$_2$RuO$_4$ consistent with a helical $p$-wave state?

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Abstract
We calculate the angular and temperature $T$ dependencies of the upper critical field $H_{c2}(\theta, \phi, T)$ for the $C_{4v}$ point group helical $p$-wave states, assuming a single uniaxial ellipsoidal Fermi surface, Pauli limiting and strong spin–orbit coupling that locks the spin-triplet $d$-vectors onto the layers. Good fits to the Sr$_2$RuO$_4$ $H_{c2}(\theta, T)$ data of Kittaka et al (2009 Phys. Rev. B 80 174514) are obtained. Helical states with $d(k) = k_x \hat{x} - k_y \hat{y}$ and $k_x \hat{x} + k_y \hat{y}$ (or $k_x \hat{x} + k_y \hat{y}$ and $k_x \hat{x} - k_y \hat{y}$) produce $H_{c2}(90^\circ, \phi, T)$ that greatly exceed (or do not exhibit) the fourfold azimuthal anisotropy magnitudes observed in Sr$_2$RuO$_4$ by Kittaka et al and by Mao et al (2000 Phys. Rev. Lett. 84 991), respectively.

Keywords: superconductivity, triplet pairing, helical $p$-wave states, upper critical field, strontium ruthenate

(Some figures may appear in colour only in the online journal)
tunneling microscopy (STM) experiments was consistent with a single nodeless gap on all three Fermi surfaces of Sr$_2$RuO$_4$ [25], but in another STM experiment, the tip was placed in a spot with substantial normal regions for $T < T_c$ [26], completely disguising any possible superconducting order parameter form. To gain a possibly consistent interpretation of all pieces of experimental evidence, it appears indispensable to introduce a new mechanism to describe the nontrivial interaction between spin-triplet superconductivity and $H$. Beforehand, one could nevertheless assume that the Pauli limit was essential to determine the in-plane $H_{c2,ab}$. In addition, since many examples of anomalous Knight shift results in singlet-spin layered and heavy fermion superconductors have been obtained, a new theory of the Knight shift is sorely needed [27, 28].

The possible spin-triplet $p$-wave states for Sr$_2$RuO$_4$ are limited by the tetragonal crystal structure with two-dimensional square lattice point group symmetry $C_{4v}$, to the six degenerate states with the $d$-vectors $\hat{d} \parallel \hat{e}$ and with $\Delta(\hat{k} \pm i\hat{k}_F)\xi$ [29, 30]. The two chiral states $d = \hat{d} \parallel \hat{e}$ and $\Delta(\hat{k} \pm i\hat{k}_F)\xi$ with $d \parallel \hat{e}$ are believed to be stabilized by setting $\hat{k}_F \rightarrow \sin(k_a\alpha)$ and $\hat{k}_F \rightarrow \sin(k_a\beta)$ [26], could fit the in-plane $H_{c2,ab}$ measurements; for comparison, even the conventional $s$-wave state without Pauli limiting has $H_{c2}(T)$ well above the experimental data of Kittaka et al for $H \parallel \hat{a}$ (figure 2) [13]. Instead, for the helical states there is a chance that Pauli limiting can play the crucial role in suppressing $H_{c2,ab}(T)$, as long as $H$ cannot cause the $d$-vectors to rotate.

Hence we model Sr$_2$RuO$_4$ as a clean homogeneous weak-coupling type-II superconductor. Since close to $H_{c2}$, $\Delta(R) = \sum_{n=0}^{\infty} a_n \mu B^n(R)$ for the vortex lattice in the mixed state, is constructed from the harmonic oscillator states $|n\rangle$, and is vanishingly small, the Gor’kov equations for $p$-wave superconductors with a single ellipsoidal Fermi surface can be linearized and transformed to yield [40, 41, 44]

$$\Delta(R) = 2\pi TN(0) V_0 \sum_{q_{\perp}} \int \frac{d\Omega}{4\pi} \int_0^\infty d\xi \exp[-2\mu B(R)\xi] \times \exp[-i\mu B(R)|\nu \cdot \xi| + 2\xi] \cdot \Delta(R) \times \left( |d(\hat{k}^\prime)|^2 + \left| \frac{\cos(a_{\parallel} B^\perp)}{\cos(\alpha B^\perp)} - 1 \right| \cdot d(\hat{k}^\prime)|^2 \right). \quad (1)$$

Figure 1. Illustration of the $d$-vectors for the helical states. In terms of $H_{c2,ab}(<\theta, \phi, T)$, the helical states shown in (a) and (d) are isotropic, while those in (b) and (c) exhibit fourfold in-plane anisotropies due to the Pauli paramagnetic effect and strong spin–orbit coupling.

2. Model

The Fermi surface of Sr$_2$RuO$_4$ consists of three sheets: a quasi-two-dimensional $\gamma$ band, and a pair of quasi-one-dimensional ($\alpha$, $\beta$) bands [32]. Although still under debate [33–35], the cylindrical $\gamma$ band is widely considered to be the primary source of $p$-wave pairing [3, 36]. The small $c$-axis dispersion in this nearly cylindrical $\gamma$ Fermi surface can be incorporated by treating it as an elongated uniaxial ellipsoid, characterized by the effective mass anisotropy of the quasi-particles $m_c \rightarrow m_a = m_b$. The primary pair-breaking effects established in superconductivity fall into two categories: 1. the orbital effect arising from the competition between the coherence of two quasi-particles in a Cooper pair and their individual orbital motions in a magnetic field, i.e., the Landau levels governed by the effective vector potential $A$ [37]; 2. the paramagnetic effect due to the Zeeman energy gained from the interactions between their spins and the field [15]. Highly anisotropic Zeeman interactions are expected in the layered compound Sr$_2$RuO$_4$, described here by an effective diagonal $g$-tensor $g = g_\alpha g_b g_b$ and $g_\alpha = g_b$ as $-\mu BS \cdot \hat{m}_{12}$. The possible spin-triplet $p$-wave states without Pauli limiting has $H_{c2}(T)$ well above the experimental data of Kittaka et al for $H \parallel \hat{a}$ (figure 2) [13]. Instead, for the helical states there is a chance that Pauli limiting can play the crucial role in suppressing $H_{c2,ab}(T)$, as long as $H$ cannot cause the $d$-vectors to rotate.
where \( N(0) \) is the density of states per spin at the Fermi level, \( V_0 \) is the pairing amplitude, \( \omega_n \) are the fermion Matsubara frequencies, \( v_F = k_B/m \) is the effective Fermi velocity, \( \alpha = (m_0 \sin^2 \theta + m_0 \cos^2 \theta)^{1/2} \) characterizes the geometric anisotropy of the Fermi surface, \( g_{eff} = [g_0^2 \cos^2 \theta + g_0^2 \sin^2 \theta]^{1/2} \) is the effective \( g \)-factor experienced by the spins with \( \cos \theta = \sqrt{\alpha} \), \( \epsilon \) is the electronic charge and the convention \( h = e = k_B = 1 \) is adopted. We note that the Klemm–Clem (KC) transformations have been performed so that the \( \xi' \) direction in (1) is always along \( B' \) [42–44].

All of the helical states in figure 1 are degenerate in terms of the KC transformed \( \text{ld} (\hat{k}')^2 = (\hat{k}^x \cdot \hat{\epsilon})^2 \geq 0 \), which contribute to the Zeeman energy, are distinct for each of the four helical states. For the helical state \( d = \hat{k} \hat{x} - \hat{k} \hat{y} \) in figure 1(b), the KC transformed

\[
\text{ld} (\hat{k}')^2 = (g_{ab} g_{ab}^2)^2 \sin^2 \theta', \\
\times \left[ (\hat{k}^x \cdot \cos \theta + \hat{k}^y \cdot \sin \theta) \cos 2\phi - \hat{k}^y \sin 2\phi \right]^2, \tag{2}
\]

is anisotropic in the basal plane [42, 43], where \( \phi' = \phi \) for \( m_a = m_0 \) and for consistency we set \( \hat{k}^x \rightarrow \hat{k}^x \) and \( \hat{k}^y \rightarrow -\hat{k}^y \).

ld (\hat{k}')^2 in state (c) is obtained from that of helical state (b) in (2) by letting \( \phi = \pi/4 \), while \( \text{ld} (\hat{k}')^2 \) for the helical states (a) and (d) are respectively obtained by setting \( \phi = 0 \) and \( \phi = \pi/4 \) in (2). These latter two helical states are therefore isotropic in the basal plane. Accordingly, the helical state (b) with \( d = \hat{k} \hat{x} - \hat{k} \hat{y} \) can be used to present the formulation.

We introduce the dimensionless quantities \( t = T/T_c(0) \), \( b_{22} = B_{22}/B_0 \) and for the \( g \)-tensor (via its elements) \( g = g/l_g \), where \( T_c(0) = (2e^2 \omega_c/\pi e)^{-1/2} \) is the superconducting transition temperature in zero field, \( C = 0.577 \) is the Euler constant, \( \omega_c \) is the energy cutoff from the BCS theory,
3. Results

Figure 3 shows our fits to the angular dependent $H_{c2}(\theta, T)$ measurements of Kittaka et al on a sample of Sr$_2$RuO$_4$ ($T_c(0) = 1.503$ K) [13] using helical state (b) with $d = \hat{k}_x \hat{x} - \hat{k}_y \hat{y}$. The appropriateness of an elongated uniaxial ellipsoidal Fermi surface for the $\gamma$ band is verified by the huge effective mass anisotropy $m/m_{ab} = 1067$ estimated from the slopes of $H_{c2}(T)$ at $T_c(0)$ in the [100] and [001] crystal directions where Pauli limiting effects are negligible. Down to low $t$, a suitable choice of the effective $g$-factor will further suppress the $H_c2$ curves, especially for those with $\theta < 5^\circ$ (see figure 2). Although the $H_{c2}(T, \theta > 5^\circ)$ data appear to follow the anisotropic effective mass model [13, 24, 44], one should nevertheless take into consideration the intrinsic anisotropy of $H_{c2}(\theta)$ arising from the point nodal structures of the helical states ($(H_{c2,ab}/H_{c2,abc})_{\theta=0} = \sqrt{2}$ for an isotropic Fermi surface) [24]. For an overall best fit, the effective $g$-tensor was evaluated to have the diagonal elements $g_x = 0.2$ and $g_{ab} = 1.9$. Obviously, the small-valued $g_x$ doesn't contribute to $H_{c2,ab}$ since $d \perp \hat{e}$ for the helical states, but it plays a role in determining $H_{c2}(\theta)$ for $0^\circ < \theta < 90^\circ$.

We remark that all the helical states listed in figure 1 could equally well fit the data shown in figure 3, as the differences in their $H_{c2}$ values appear only in their in-plane ($\phi$) anisotropies. As seen from (1), in the absence of Pauli limiting, $H_{c2,ab}(\phi)$ for the helical states are isotropic in the basal plane. However, with the fitting parameter $g_{ab} = 1.9$, the $H_{c2,ab}(\phi)$ at 0.13 K for the helical states (b) and (c) in figure 1 exhibit fourfold in-plane azimuthal anisotropies with a relative amplitude as large as 30% (figure 4(a)) and a phase shift of $\pi/4$ between them, while those for states (a) and (d) remain isotropic in the $ab$ plane. The observed in-plane anisotropy of $H_{c2,ab}(\phi)$ is at most 3% and disappears either above 0.8 K or with a field misalignment of less than 1° [13, 19]. The calculated anisotropy for helical state (b) with $d = \hat{k}_x \hat{x} - \hat{k}_y \hat{y}$ persists for $T > T_c/2$ and for field misalignments greater than $2^\circ$ (figure 4(b)). Thus, this parallel-spin $p$-wave state can explain the strong Pauli limiting for $B \perp \hat{e}$, but the details are not in precise agreement with the experimental observations [13, 19].

4. Discussion

A multi-component order parameter proposed to interpret the in-plane $H_{c2,ab}(\phi)$ anisotropy in reference [19] turns out to have a similar problem of a large magnitude of the in-plane anisotropy [21]. There could also be two slightly misaligned crystals in the same sample [13], and the smaller region of the hysteretic magnetization data below 0.8 K in the more recent data of Yonezawa et al than in the older Mao et al and Deguchi et al data are consistent with this scenario [12–14], [19]. Others think that this first-order transition below 0.8 K is more intrinsically due to a Fulde–Farrell–Larkin–Ovchinnikov state, entered below 0.55$T_c$ (close to 0.8 K in Sr$_2$RuO$_4$) [45]. Based on the present calculations, if the Pauli pair-breaking effect is demanded as the source for the suppression on $H_{c2,ab}$, helical state (b) with $d = \hat{k}_x \hat{x} - \hat{k}_y \hat{y}$ has the same fourfold anisotropy with the same phase as in the experiments. Helical state (c) with $d = \hat{k}_x \hat{x} + \hat{k}_y \hat{y}$ has the fourfold anisotropy differing in phase by $\pi/4$. But, both of these azimuthal anisotropies are much stronger than that observed in experiment. However, the other helical (a) and (d) $p$-wave states with $d = \hat{k}_x \hat{x} + \hat{k}_y \hat{y}$ and $\hat{k}_x \hat{x} - \hat{k}_y \hat{y}$ are predicted to have no azimuthal anisotropies at all. Including $ab$-planar anisotropy on the $\gamma$ Fermi surface could lead to a small azimuthal anisotropy of $H_{c2}(90^\circ, \phi, T)$, but normally Fermi surface anisotropy is largest near to $T_c$. Thus, a single purported triplet-spin order parameter for Sr$_2$RuO$_4$ is still elusive. We note, however, that there are many examples in which the Knight shift observations have been
misleading and/or are also in apparent conflict with the upper critical field results [28, 46], strongly suggesting that a new theory of the Knight shift might lead to a possible resolution of the symmetry of the order parameter in Sr2RuO4 [27, 28].

In summary, we studied the four helical p-wave states potentially realized in Sr2RuO4 at Hc2 by fitting the angular dependent \( H_{c2}(\theta, \phi, T) \) measurements, taking the Pauli paramagnetic effects into account by imposing strong spin–orbit coupling effects as the origin of the \( H_{c2,ab} \) suppression. In the ranges of the fitting parameters, one of the four helical states was predicted to have in-plane \( H_{c2}(90°, \phi, T) \) fourfold azimuthal anisotropy with the same phase as observed, but both that azimuthal anisotropy and that from the (c) helical state were predicted to be much stronger than that observed in Sr2RuO4. The \( H_{c2}(90°, \phi, T) \) behaviors of the two other helical p-wave states were predicted to be completely independent of \( \phi \), as long as in-plane Fermi surface anisotropy could be safely ignored. Other attempts to fit an order parameter such as \( \Delta_0 \sin(k_a \alpha) + i \sin(k_a \alpha) \) with the low-T specific heat \( C_v \sim T^2 \) dependence failed to confront the very strong Pauli limiting of \( H_{c2}(90°, \phi, T) \) [26]. Thus, the thermodynamic zero-field specific heat measurements, which are sensitive to the normal Ru inclusions, appear to be in direct conflict with the field-dependent thermodynamic specific heat and magnetization measurements of the upper critical field, which are not [12–14], [19]. Further calculations to try to fit the excellent STM results of Suderow et al with a p-wave order parameter are also needed [25]. A point node in a helical p-wave order parameter might smear the sharp density of state walls they observed, but an accurate calculation is needed to quantify this possible disagreement.

Acknowledgments

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