Vortex Lattice Structures of Sr₂RuO₄

D.F. Agterberg

Theoretische Physik, Eidgenossiche Technische Hochschule-Hönggerberg, 8093 Zürich, Switzerland (Received 4 February 1998)

The vortex lattice structures of Sr_2RuO_4 for the odd-parity representations of the superconducting state are examined. Particular emphasis is placed upon the two dimensional representation which is believed to be relevant to this material. It is shown that when the zero-field state breaks time reversal symmetry, there must exist *two* superconducting transitions when there is a finite field along a high symmetry direction in the basal plane. Also it is shown that a *square* vortex lattice is expected when the field is along the *c* axis. The orientation of the square lattice with respect to the underlying ionic lattice yields information as to which Ru 4*d* orbitals are relevant to the superconducting state. [S0031-9007(98)06315-7]

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The oxide superconductor Sr₂RuO₄ is structurally similar to the high T_c materials but differs markedly from the latter in its electronic structure [1]. In particular, the normal state near the superconducting transition of Sr₂RuO₄ is well described by a quasi-2D Landau Fermi liquid [2]. There now exists considerable evidence that the superconducting state of Sr_2RuO_4 [1] is not a conventional *s*-wave state. Nuclear quadrupole resonance measurements show no indication of a Hebel-Slichter peak in $1/T_1T$ [3], T_c is strongly suppressed by nonmagnetic impurities [4], and tunneling experiments are inconsistent with s-wave pairing [5]. While these measurements demonstrate that the superconducting state is non-s-wave, they do not determine what pairing symmetry actually occurs in this material. The determination of the pairing symmetry in unconventional superconductors is a notoriously difficult problem and theoretical insight provides a useful guide. The observations that the Fermi liquid corrections due to electron correlations are similar in magnitude to those found in superfluid ³He and that closely related ruthenates are itinerant ferromagnets have led to the proposal that the superconducting state in Sr₂RuO₄ is of odd parity [6]. Even with this insight there still remain five odd-parity states that have different symmetry-all of which have a nodeless gap and therefore similar thermodynamic properties [6]. Recently, muon spin rotation (μ SR) measurements indicate that a spontaneous internal magnetization begins to develop at T_c [7]. The most natural interpretation of this moment is that the superconducting state breaks time reversal symmetry (\mathcal{T}) . This places a strong constraint on the pairing symmetry in Sr₂RuO₄ since it implies that the superconducting order parameter must have more than one component [8]. Of the possible representations (REPS) of the D_{4h} point group only the two dimensional (2D) Γ_{5u} and Γ_{5g} REPS exhibit this property. Of these two the Γ_{5u} REP is the most likely to occur in Sr₂RuO₄ due to arguments of Ref. [6] and the quasi-2D nature of the electronic properties. The order parameter in this case has two components (η_1, η_2) that share the same rotation-inversion

symmetry properties as (k_x, k_y) [8]. The broken \mathcal{T} state would then correspond to $(\eta_1, \eta_2) \propto (1, i)$. The *z* component of the local magnetic field h_z couples linearly to this state via the symmetry allowed coupling $ih_z(\eta_1^*\eta_2 - \eta_2^*\eta_1)$. Magnetic properties of such a broken time reversal state are discussed in Ref. [8]. The identification of this state as the superconducting ground state would prove a significant advance since the only other non-*s*-wave state for which the symmetry has been unambiguously identified is the $d_{x^2-y^2}$ state in high- T_c cuprates (even after more than a decade of research into this problem for the heavy fermion compounds UPt₃ and UBe₁₃). Below some observable consequences of a $(\eta_1, \eta_2) \propto (1, i)$ state are revealed which, if seen, would identify this as the relevant pairing state for Sr₂RuO₄.

I investigate within Ginzburg Landau (GL) theory the vortex lattice structures expected for the odd-parity REPS of the superconducting state, focusing mainly on the Γ_{5u} REP. It is initially shown that a general consequence of the broken \mathcal{T} state described above is that in a finite magnetic field oriented along a high symmetry direction in the basal plane there will exist a second superconducting transition in the mixed phase as temperature is reduced. The high field state is a vortex lattice for a single component order parameter with line nodes. These nodes vanish when the second transition occurs. It is then shown that a square vortex lattice is expected to appear for all the odd-parity REPS when the field is along the c axis. Observable differences are shown to exist between the 1D and the 2D REPS for this field orientation. Finally, within the recently proposed model of orbital dependent superconductivity of Sr_2RuO_4 [9] it is also shown that the orientation of square vortex lattice with respect to the underlying crystal lattice dictates which of the Ru 4d orbitals give rise to the superconducting state.

To demonstrate the presence of the two superconducting transitions described above consider the magnetic field along the \hat{x} direction (x is chosen to be along the basal plane main crystal axis) and a homogeneous zero-field state $(\eta_1, \eta_2) \propto (1, i)$. In general the presence of a magnetic field along the \hat{x} direction breaks the degeneracy of the (η_1, η_2) components, so that only one of these two components will order at the upper critical field [e.g., $(\eta_1, \eta_2) \propto (0, 1)$]. As has been shown for type II superconductors with a single component the order parameter solution is independent of x [10] so that σ_x (a reflection about the \hat{x} direction) is a symmetry operation of the $(\eta_1, \eta_2) \propto (0, 1)$ vortex phase. Now consider the zero-field phase $(\eta_1, \eta_2) \propto (1, i), \sigma_x$ transforms (1, i) to

 $(-1, i) \neq e^{i\phi}(1, i)$, where ϕ is a phase factor. This implies that σ_x is *not* a symmetry operator of the zero-field phase. It follows that there must exist a second transition in the finite field phase at which η_1 becomes nonzero. Similar arguments hold for the field along any of the other three crystallographic directions in the basal plane. The existence of two transitions for all four crystallographic axes in the basal plane is a consequence of the zero-field broken \mathcal{T} state.

For a more detailed analysis consider the following dimensionless GL free energy density for the Γ_{5u} REP

$$f = -|\boldsymbol{\eta}|^{2} + |\boldsymbol{\eta}|^{4}/2 + \beta_{2}(\eta_{1}\eta_{2}^{*} - \eta_{2}\eta_{1}^{*})^{2}/2 + \beta_{3}|\eta_{1}|^{2}|\eta_{2}|^{2} + |D_{x}\eta_{1}|^{2} + |D_{y}\eta_{2}|^{2} + \kappa_{2}(|D_{y}\eta_{1}|^{2} + |D_{x}\eta_{2}|^{2}) + \kappa_{5}(|D_{z}\eta_{1}|^{2} + |D_{z}\eta_{2}|^{2}) + \kappa_{3}[(D_{x}\eta_{1})(D_{y}\eta_{2})^{*} + \text{H.c.}] + \kappa_{4}[(D_{y}\eta_{1})(D_{x}\eta_{2})^{*} + \text{H.c.}] + h^{2},$$
(1)

 $\mathbf{h} = \nabla \times \mathbf{A}, \quad D_{\nu} = \nabla_{\nu}/\kappa - iA_{\nu},$ where f is in units $B_c^2/4\pi$, lengths are in units $\lambda =$ $[\hbar^2 c^2 \beta_1 / (16e^2 \kappa_1 \alpha \pi)]^{1/2}$, h is in units $\sqrt{2} B_c =$ $\Phi_0/(4\pi\lambda\xi), \quad \alpha = \alpha_0(T - T_c), \quad \xi = (\kappa_1/\alpha)^{1/2}, \text{ and}$ $\kappa = \lambda/\xi$. Note that λ , ξ , B_c , and κ are simply convenient choices and do not correspond to measured values of these parameters. The coupling $ih_z(\eta_1^*\eta_2 - \eta_2^*\eta_1)$ mentioned in the introduction is equivalent up to a surface term to the difference of the κ_3 and κ_4 terms in Eq. (1). For the application of Eq. (1) to Sr_2RuO_4 it is reasonable to determine the phenomenological coefficients in the weak-coupling limit since $T_c/T_F \approx 10^{-4}$. Furthermore, the measurements of Mackenzie et al. of T_c as a function of impurity concentration show that the ratio of the mean free path to the zero-temperature coherence length is >8 for $T_c > 1.3$ K [4], indicating that the clean limit should also be a reasonable approximation for Sr_2RuO_4 . Taking for the Γ_{5u} REP the gap function described by the pseudo-spin-pairing gap matrix: $\hat{\Delta} = i [\eta_1 v_x / \sqrt{\langle v_x^2 \rangle} + \eta_2 v_y / \sqrt{\langle v_x^2 \rangle}] \sigma_z \sigma_y$, where the brackets $\langle \rangle$ denote an average over the Fermi surface and σ_i are the Pauli matrices, it is found that $\beta_2 = \kappa_2 = \kappa_3 = \kappa_4 = \gamma$ and $\beta_3 = 3\gamma - 1$, where $\gamma = \langle v_x^2 v_y^2 \rangle / \langle v_x^4 \rangle$. Note that $0 \le \gamma \le 1$ and that $\gamma = 1/3$, respectively, for a cylindrical or spherical Fermi surface. These parameters agree with the cylindrical Fermi surface results of Ref. [11]. It is easy to verify that in zero field $(\eta_1, \eta_2) \propto (1, i)$ is the stable ground state for all allowed γ .

It is informative to determine the values of γ that are relevant to Sr₂RuO₄. Local density approximation band structure calculations [12,13] reveal that the density of states near the Fermi surface are due mainly to the four Ru 4*d* electrons in the t_{2g} orbitals. There is a strong hybridization of these orbitals with the O 2*p* orbitals giving rise to antibonding π^* bands. The resulting bands have three quasi-2D Fermi surface sheets labeled α , β , and $\tilde{\gamma}$ (see Ref. [2]). The α and β sheets consist of {*xz*, *yz*} Wannier functions and the $\tilde{\gamma}$ sheet of *xy* Wannier functions. In general γ is not given by a simple

average over all the sheets of the Fermi surface. A knowledge of the pair scattering amplitude on each sheet and between the sheets is required to determine γ [9,14]. Recently, to account for the large residual density of states (DOS) observed in the superconducting state, it has been proposed that either the xy or the $\{xz, yz\}$ Wannier functions exhibit superconducting order [9]. This model implies that there are two possible values of γ , one for the $\tilde{\gamma}$ sheet (γ_{xy}) and one for an average over the $\{\alpha, \beta\}$ sheets $(\gamma_{xz,yz})$. A tight binding model indicates $\gamma_{xy} = 0.67$ and $\gamma_{xz,yz} = 0.11$ [15]. These values are sensitive to changes in the parameters of the tight binding model; however, the qualitative result that $\gamma_{xy} > 1/3$ and $\gamma_{xz,yz} < 1/3$ is robust. Physically $\gamma_{xy} > 1/3$ because of the proximity of the $\tilde{\gamma}$ Fermi surface sheet to a Van Hove singularity and $\gamma_{xz,yz} < 1/3$ due to the quasi-1D nature of the $\{\alpha, \beta\}$ surfaces [12,13].

Following Burlachkov [16] for the solution of upper critical field $H_{c_2}^{ab}$ for the field in the basal plane, the vector potential is taken to be $\mathbf{A} = H_z(\sin\theta, -\cos\theta, 0)$ (θ is the angle the applied magnetic field makes with the \hat{x} direction). After setting the component of **D** along the field to be zero it is found that $H_{c_2}^{ab}(\theta) = \kappa / [\kappa_5 \lambda(\theta)/2]^{1/2}$, where $\lambda(\theta) =$ $1 + \gamma - [(1 - \gamma)^2 - (1 + \gamma)(1 - 3\gamma)\sin^2 2\theta]^{1/2}$.

A measurement of the temperature independent fourfold anisotropy in $H_{c_2}^{ab}$ thus determines γ . To determine the field at which the second transition discussed above occurs consider the magnetic field along the \hat{x} direction. The free energy of Eq. (1) is then similar to that studied in UPt₃ [10,17,18] and since Sr₂RuO₄ is a strong type II superconductor with a GL parameter of 31 for the field in the basal plane [20] the procedure of Garg and Chen [17] to study the second transition can be applied here. At $H_{c_2}^{ab} \eta_1$ orders and the vortex lattice solution is given by [10,17,18]

$$\eta_1 = \sum_n c_n e^{inqz} e^{-(\kappa_5/\gamma)^{1/2} \kappa H [y-qn/(\kappa H)]^2/2},$$
(2)

5185

where $c_n = e^{in^2\pi/2}$ and q has the two possible values $q_1^2 = \sqrt{3} H \kappa \pi (\gamma/\kappa_5)^{1/2}$ or $q_2^2 = H \kappa \pi (\gamma/\kappa_5)^{1/2}/\sqrt{3}$ (these two solutions are degenerate). At the second transition the η_2 component becomes nonzero. As is discussed in Refs. [17,18] the solution for η_2 corresponds to a lattice that is displaced relative to that of η_1 by $\mathbf{d} = (\bar{y}, \bar{z})$. Accordingly, the field at which the second transition occurs is found by substituting

$$\eta_{2} = ir \sum_{n} c_{n} e^{i(nq + \kappa H\bar{y})(z-\bar{z})} e^{-\sqrt{\kappa_{5}} \kappa H[y-\bar{y}-qn/(\kappa H)^{2}]/2}$$
(3)

and Eq. (2) into the free energy, minimizing with respect to the displacement vector **d**, and determining when the coefficient of r^2 becomes zero. The numerical solution for the ratio of the second transition field H_2 to the upper critical field H_{c_2} and the specific heat jump of the second transition ΔC_2 to that of the first transition ΔC_1 is shown in Fig. 1. Three vortex lattice configurations are found to be stable as a function of γ (depicted in Fig. 1). For $0 < \gamma < 0.187$ $q = q_2$ and $\mathbf{d} = (T_y, T_z)/4$ $(T_y$ and T_z are the translation vectors of the centered rectangular cell for the η_1 lattice), for $0.187 < \gamma < 0.433$ $q = q_1$ and **d** = $(T_{y}, T_{z})/4$, and for $0.433 < \gamma < 1$ $q = q_{2}$ and $\mathbf{d} = 0$. For the field along $\hat{x} \pm \hat{y}$ the ratio H_2/H_{c_2} is given by Fig. 1 with γ replaced by $(1 - \gamma)/(1 + 3\gamma)$. The arguments in Ref. [19] imply that the shape of the vortex lattice unit cell for $H < H_2$ will be strongly field dependent.

The second phase transition will reveal itself through a discontinuity in both the specific heat and the dc magnetic susceptibility. It is of interest to note that evidence for this transition may already exist in the ac magnetic susceptibility measurements of Yoshida *et al.* [20]. They observed a second peak in the imaginary part of the magnetic susceptibility only when the flux lines were parallel to the basal plane. Another intriguing feature of the above described phase diagram is that the high-field phase corresponds to a vortex lattice for a gap with *line* nodes. In



FIG. 1. The ratio of the two transition lines and the two specific heat jumps for the field along \hat{x} as a function of γ . The open (closed) circles correspond to the zeros of the η_2 (η_1) lattice. The vertical lines separate regions where the depicted vortex lattice structures are favored. For all three lattice structures the *y* and *z* axes have the same orientation and the dimensions of the rectangular cell are the same.

particular for the field along the \hat{x} direction the gap matrix is given by $\hat{\Delta} = i[\eta_1(\mathbf{r})v_x(\mathbf{k})/\sqrt{\langle v_x^2 \rangle}]\sigma_z \sigma_y$ which is zero for $k_x = 0$ (note that symmetry allows a further contribution to the gap proportional to $iv_z \sigma_x \sigma_y$ which will reduce the line nodes to point nodes; however, it is expected that this contribution is small). If $H_2/H_{c_2} \ll 1$ then the arguments of Volovik [21] imply that for $H_2 < H \ll H_{c_2}$ the DOS varies as $\sqrt{H/H_{c_2}}$ (provided the residual DOS observed in the zero-field superconducting state is subtracted). In the low-field phase the line nodes vanish and as a consequence transport measurements should also be sensitive to the second transition.

Now consider the magnetic field oriented along the *c* axis. Setting $D_z = 0$ writing $\Pi_+ = \kappa (iD_x + D_y)/2H$, $\Pi_- = \kappa (iD_x - D_y)/2H$, $\eta_+ = (\eta_x + i\eta_y)/\sqrt{2}$, and $\eta_- = (\eta_x - i\eta_y)/\sqrt{2}$, minimizing the quadratic portion of Eq. (1) with respect to η_+ and η_- yields

$$2\kappa \begin{pmatrix} \eta_+\\ \eta_- \end{pmatrix} = H \begin{pmatrix} (1+\gamma)(1+2N) & (1+\gamma)\Pi_+^2 + (1-3\gamma)\Pi_-^2\\ (1+\gamma)\Pi_-^2 + (1-3\gamma)\Pi_+^2 & (1+\gamma)(1+2N) \end{pmatrix} \begin{pmatrix} \eta_+\\ \eta_- \end{pmatrix},$$
(4)

where $N = \Pi_{+}\Pi_{-}$. The maximum value of H that allows a nonzero solution for (η_{+}, η_{-}) yields the upper critical field $H_{c_2}^c$. For $\gamma \neq 1/3$ $H_{c_2}^c$ must be found numerically (note that for $\gamma = 1/3$ the solution can be found analytically [8, 22]). Expanding (η_{+}, η_{-}) in terms of the eigenstates of N (Landau levels) and diagonalizing the resulting matrix yields $H_{c_2}^c(\gamma)$. The form of the eigenstate at $H_{c_2}^c$ is found to be $\eta_{+}(\mathbf{r}) =$ $\sum_{n\geq 0} a_{4n+2}\phi_{4n+2}(\mathbf{r})$ and $\eta_{-}(\mathbf{r}) = \sum_{n\geq 0} a_{4n}\phi_{4n}(\mathbf{r})$, where $\phi_n(\mathbf{r}) = \sum_m c_m e^{iqm\bar{y}}2^{-n/2}H_n[\tilde{x} - qm/(\kappa H)] \times e^{-\kappa H[\tilde{x}-qm/(\kappa H)]^2/2}/(n!)^{1/2}$, the coefficients a_n are real, (\tilde{x}, \tilde{y}) is the vector (x, y) rotated by an angle $\tilde{\theta}$ about the z axis, and $H_n(x)$ represent Hermite polynomials. The solution for the form of the vortex lattice represents a complex problem due to the presence of many Landau levels in the solution of (η_+, η_-) and the weak type II nature of Sr₂RuO₄ for the field along the *c* axis (Ref. [20] indicates $\kappa \approx 1.2$). Here I present results that are strictly valid in the large κ limit and leave the treatment for general κ to a later publication [a perturbative expansion in $(1 - 3\gamma)/(1 + \gamma)$ indicates that the qualitative results are unchanged for $\kappa = 1.2$ [15]]. In the large κ limit the form of the vortex lattice is found by minimizing $\beta = \overline{f_4}/(|\eta|^2)^2$ [f_4 is the quartic part of Eq. (1)] with respect to the coefficients c_n , q, and $\tilde{\theta}$. It is assumed that $c_n = c_{n+2}$. This restricts the vortex lattice structures

to be centered rectangular with a short axis $L_y = 2\pi/q$ and a long axis $L_x = 2q/(\kappa H)$. The ratio $t = L_x/L_v$ is $\sqrt{3}$ for a hexagonal vortex lattice and is 1 for a square vortex lattice. I further restrict the analysis to the two orientations $\tilde{\theta} = \{0, \pi/4\}$ since these correspond to aligning one of the vortex lattice axes with one of the high symmetry directions in the basal plane. Remarkably, the treatment of the many Landau levels in the solution of η_+ and η_- becomes numerically straightforward when β is expressed as a sum over the reciprocal lattice given by $\mathbf{l} = \hat{x} l_1 2\pi / L_x + \hat{y} l_2 2\pi / L_y$ [15] (see also Ref. [23]). It is found that β is minimized for $c_n = e^{in^2\pi/2}$ and that the values of t and $\tilde{\theta}$ depend upon γ . For $\gamma \leq 1/3$ $(\gamma \ge 1/3) \ \tilde{\theta} = 0 \ (\tilde{\theta} = \pi/4)$ and t varies continuously from $\sqrt{3}$ to 1 as γ decreases (increases) from 1/3 to 1/3 - 0.0050 (1/3 + 0.0050). For $\gamma < 1/3 - 0.0050$ and $\gamma > 1/3 + 0.0050$ the minimum β corresponds to t = 1. This implies that for $\gamma_{xz,yz}$ a square vortex lattice rotated $\pi/4$ about the c axis from the crystal lattice is expected and for γ_{xy} a square vortex lattice that is aligned with the underlying crystal lattice is expected near $H_{c_2}^c$. Note the appearance of the square vortex lattice correlates with an anisotropy in $H_{c_2}^{ab}$ of $|1 - H_{c_2}^{ab}(\theta = 0)/H_{c_2}^{ab}(\theta = \pi/4)| > 0.01.$

The recent observation of a square vortex lattice in Sr_2RuO_4 [24] makes it of interest to compare the above behavior to that expected for the 1D REPS of D_{4h} . It is known that for single component order parameters nonlocal corrections to the standard GL theory stabilize a square vortex lattice [25–27]. In particular the following nonlocal term will stabilize the square lattice

$$\epsilon[|(D_x^2 - D_y^2)\psi|^2 - |(D_x D_y + D_y D_x)\psi|^2].$$
(5)

Treating this term as a perturbation to the GL free energy leads to $\psi = \phi_0 - \tilde{\epsilon} \phi_4$, where $\tilde{\epsilon} = \sqrt{6} \epsilon H/\kappa$ (κ is the GL parameter) near $H_{c_2}^c$. As $\tilde{\epsilon}$ increases (note $\tilde{\epsilon} = 0$ at T_{c}) the vortex lattice continuously distorts from hexagonal to square [25-27] until the square vortex lattice is stable for $|\tilde{\boldsymbol{\epsilon}}| > 0.024$. The sign of $\boldsymbol{\epsilon}$ determines the orientation of the vortex lattice; for $\epsilon > 0$ the vortex lattice is aligned with the underlying lattice while for $\epsilon < 0$ the lattice is rotated $\pi/4$ with respect to the underlying crystal lattice [25,27]. The sign of ϵ has been determined within a weak coupling clean limit approximation for the 1D odd-parity REPS. For the A_{1u} REP using $\hat{\Delta} = \psi(\hat{x}v_x/\sqrt{\langle v_x^2 \rangle} +$ $\hat{y}v_y/\sqrt{\langle v_x^2 \rangle}$ $\dot{\sigma}i\sigma_y$ the sign of ϵ is determined by the sign of $3\langle (v_x^2 + v_y^2)v_x^2v_y^2 \rangle / \langle (v_x^2 + v_y^2)v_x^4 \rangle - 1$. Using a form for $\hat{\Delta}$ that is analogous to that used for the $A_{1\mu}$ REP, the same result is found for all the 1D odd-parity REPS. This implies that the final orientation of the square vortex lattice for the 1D REPS is the same as that found for the 2D REP for superconducting order in the xy or the $\{xz, yz\}$ orbitals. The behavior of the vortex lattice for the 1D REPS as a function of $\tilde{\epsilon}$ is very similar to that for the 2D REP as a function of γ . An observable difference

between the 2D and the 1D REPS is that for the 2D REP the vortex lattice remains square up to T_c while for the 1D REPS the vortex lattice is hexagonal at T_c . Also, the GL theories for the 1D and the 2D REPS predict a fourfold anisotropy in $H_{c_2}^{ab}$ but this anisotropy vanishes at T_c for the 1D REPS and does not vanish at T_c for the 2D REP.

In conclusion I have examined GL models for the oddparity REPS of the superconducting state for Sr_2RuO_4 . It was found that if the zero-field ground state breaks \mathcal{T} symmetry (the 2D REP) then there should exist a second transition in the mixed state when the magnetic field is applied along a high symmetry direction in the basal plane. It was also shown that when the field is along the *c* axis there will be a square vortex lattice for all the possible odd-parity superconducting states.

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