Interband Proximity Effect and Nodes of Superconducting Gap in Sr_2RuO_4

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The power-law temperature dependences of the specific heat, the nuclear relaxation rate, and the thermal conductivity suggest the presence of line nodes in the superconducting gap of Sr_2RuO_4. These recent experimental observations contradict the scenario of a nodeless $(k_x + ik_y)$-type superconducting order parameter. We propose that interaction of superconducting order parameters on different sheets of the Fermi surface is a key to understanding the above discrepancy. A full gap exists in the active band, which drives the superconducting instability, while line nodes develop in passive bands by the interband proximity effect.

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The layered perovskite Sr_2RuO_4 with $T_c \simeq 1.5$ K [1] is an example of an unconventional superconductor with non-s-wave Cooper pairing [2]. The theoretical proposal [3–5] of a spin-triplet p-wave order parameter $\Delta_{\alpha\beta}(\mathbf{k}) = (i\sigma^y\sigma^z)_{\alpha\beta}d(\mathbf{k})$, $d(\mathbf{k}) \propto (k_x + ik_y)$ is supported by experimental observations of a temperature independent Knight shift for $H \perp c$ [6] and an increased muon spin relaxation below T_c [7]. Such an axial gap function has a nonvanishing amplitude on the cylindrical quasi-two-dimensional Fermi surface of Sr_2RuO_4 [8–10]. This property favors the axial state as a natural choice in a weak-coupling theory, which generally supports nodeless solutions [3]. Recent experimental data collected on high quality samples, however, seem to invalidate the above conclusion. The power-law temperature dependences as $T \to 0$ found for the specific heat, $C(T) \propto T^2$ [11,12], the nuclear quadrupole-resonance relaxation rate, $T_1^{-1} \propto T^3$ [13], the thermal conductivity, $\kappa(T) \propto T^2$ [14,15], the penetration depth [16], and the ultrasonic attenuation [17] point to lines of zeros in the superconducting gap and, thus, question the consistency of the whole picture.

There have been several theoretical attempts to resolve this controversy [18–21]. Most suggest replacing the axial p-wave order parameter $d_p(\mathbf{k}) \propto (k_x + ik_y)$ by a suitable f-wave gap: $d_f(\mathbf{k}) \propto (k_x + ik_y)g(\mathbf{k})$, where the even parity function $g(\mathbf{k})$ is chosen to have zeros, e.g., k_xk_y or $(k_x^2 - k_y^2)$ [19–21]. There is no clear microscopic mechanism for such an f-wave instability. More importantly, nodes in an f-wave gap are only *marginally* stable; i.e., they disappear if all symmetry allowed harmonics are included in the expansion of the gap function. For example, for $g(\mathbf{k}) = k_xk_y$ one finds

$$d(\mathbf{k}) = \eta_1(k_x + ik_y)k_xk_y + i\eta_2(k_x - ik_y), \quad (1)$$

where η_1 and η_2 are real. Both terms in Eq. (1) transform in the same way under operations of the symmetry group of the superconducting state. In particular, both harmonics are symmetric with respect to a fourfold rotation $e^{i\pi/2}C_4$ and time reversal in combination with a reflection in the (100) plane, $e^{i\pi}\hat{\mathcal{T}}\hat{\sigma}_x$. Therefore, the f- and the p-wave harmonics are symmetry indistinguishable in tetragonal crystals and mix with each other producing a finite gap $|\Delta|_{\min} \sim \eta_2$. Very recently, two groups [14,15] have measured the basal plane anisotropy of the thermal conductivity $\kappa(\theta)$ in Sr_2RuO_4 at finite magnetic fields. Their results also discard an f-wave gap together with a so-called "anisotropic" p-wave state [18] as possible candidates to explain line of nodes in Sr_2RuO_4: All these superconducting states have a substantial anisotropy of $\kappa(\theta)$ in the magnetic field determined by in-plane node structure, whereas experimentally the basal plane anisotropy is much smaller. Instead, these experiments suggest horizontal nodes in the superconducting gap [14,15].

An opposite conclusion has been reached by Lupien *et al.* [17] from the anisotropy of the ultrasonic absorption. However, the measured anisotropy appears mainly in the absolute magnitude of the attenuation and not in the exponent of the temperature power law. For this reason it is not clear that these results are in conflict with horizontal lines of zeros and, as Lupien *et al.* stress, detailed calculations based on the actual electronic structure are necessary for a definitive interpretation of their results.

Here, we propose a mechanism for the formation of horizontal line nodes in the superconducting gap of Sr_2RuO_4. The Fermi energy crosses three bands, determined by the d_{xy} (γ sheet of the Fermi surface) and the hybridized d_{xz} and d_{yz} (α and β sheets) orbitals of Ru [8–10]. Magnetic fluctuations, responsible for anisotropic Cooper pairing [3,22], have significant orbital dependence [23]. Therefore, the intrinsic temperature of the superconducting instability should vary from band to band with one sheet being the active source for superconducting instability and the others being the passive sheets. In reality, interband scattering of Cooper pairs, or proximity effect in the momentum space, will induce the superconducting gap simultaneously on all parts of the Fermi surface. Such interband scattering is generally a strong effect, which allows one to treat numerous multiband superconductors by an effective single band Fermi surface. Agterberg and co-workers [4] have argued that Sr_2RuO_4 is different: A direct in-plane

scattering of the p-wave Cooper pairs between bands is significantly suppressed by the orbital symmetry. Therefore, they conclude, one or two bands develop only tiny superconducting gaps, which show up at intermediate temperatures as a residual density of states with a subsequent crossover as $T \to 0$ to a full gap behavior. In this Letter, we study additional interlayer contributions to interband scattering of Cooper pairs, which become important when direct in-plane scattering is suppressed. We find that a nodeless axial order parameter $d_p(\mathbf{k}) \propto (k_x + ik_y)$ in the active band can induce superconducting gaps with zeros in the passive bands: $d'_p(\mathbf{k}) \propto (k_x + ik_y)\cos(k_z/2)$. Thus, circular nodes of the superconducting gap develop about the c axis on one or two of the three Fermi surface sheets. This model of weakly coupled superconducting order parameters in different bands fits well $C(T)$ in zero field [11] and helps to explain the observed field behavior of the specific heat [12].

We start with a general two-particle interaction:

$$\hat{V} = \int d\mathbf{r}\, d\mathbf{r}'\, U(\mathbf{r},\mathbf{r}')\psi^\dagger(\mathbf{r})\psi^\dagger(\mathbf{r}')\psi(\mathbf{r}')\psi(\mathbf{r}), \qquad (2)$$

where for simplicity we omit all spin indices assuming a fixed spin structure of the triplet order parameter. The band representation for the effective interaction $\hat{V}$ is obtained by (i) expanding the field operators $\psi(\mathbf{r})$ in terms of the band operators: $\psi(\mathbf{r}) = \sum_{l,\mathbf{k}} \varphi_{l\mathbf{k}}(\mathbf{r}) c_{l\mathbf{k}}$ (l is a band index); and (ii) representing the Bloch function $\varphi_{l\mathbf{k}}(\mathbf{r})$ in a given band as a lattice sum over the Wannier function of the Ru orbitals: $\varphi_{l\mathbf{k}}(\mathbf{r}) = \sum_n e^{i\mathbf{k}\mathbf{R}_n}\phi_l(\mathbf{r} - \mathbf{R}_n)$. The interaction in the Cooper channel is

$$\hat{V} = \frac{1}{2}\sum_{ll',\mathbf{k}\mathbf{k}'} V_{ll'}(\mathbf{k},\mathbf{k}') c^\dagger_{l\mathbf{k}} c^\dagger_{l-\mathbf{k}} c'_{l'-\mathbf{k}} c'_{l'\mathbf{k}}, \qquad (3)$$

where the scattering vertex is given by

$$V_{ll'}(\mathbf{k},\mathbf{k}') = \int d\mathbf{r}\, d\mathbf{r}'\, U(\mathbf{r},\mathbf{r}') \sum_{nn'mm'} e^{-i\mathbf{k}(\mathbf{R}_n - \mathbf{R}_{n'})} e^{i\mathbf{k}'(\mathbf{R}_m - \mathbf{R}_{m'})} \phi^*_l(\mathbf{r} - \mathbf{R}_n)\phi^*_l(\mathbf{r}' - \mathbf{R}_{n'})\phi_{l'}(\mathbf{r}' - \mathbf{R}_{m'})\phi_{l'}(\mathbf{r} - \mathbf{R}_m). \qquad (4)$$

Following Ref. [4] we now consider interband scattering processes ($l \neq l'$) in the tight-binding approximation, i.e., assume that the Wannier functions are well localized and, therefore, the main contribution to $V_{ll'}(\mathbf{k},\mathbf{k}')$ comes from a few neighboring sites. The largest on-site contribution ($\mathbf{R}_n = \mathbf{R}_{n'} = \mathbf{R}_m = \mathbf{R}_{m'}$) is independent of $\mathbf{k}$ and $\mathbf{k}'$. It causes coupling only between conventional s-wave order parameters in two bands. The coupling of the p-wave order parameters appears first in the sum (4) for $\mathbf{R}_m = \mathbf{R}_n$, $\mathbf{R}_{m'} = \mathbf{R}_{n'}$, and $(\mathbf{R}_{n'} - \mathbf{R}_n) = \boldsymbol{\delta}_i = \pm a\hat{\mathbf{x}}(\hat{\mathbf{y}})$ (a is lattice constant):

$$V^{pp}_{ll'}(\mathbf{k},\mathbf{k}') = \sum_{i=x,y} \sin k_i a \sin k'_i a \times \int d\mathbf{r}\, d\mathbf{r}'\, U(\mathbf{r},\mathbf{r}') \times \phi^*_l(\mathbf{r})\phi_{l'}(\mathbf{r})\phi^*_l(\mathbf{r}' - \boldsymbol{\delta}_i)\phi_{l'}(\mathbf{r}' - \boldsymbol{\delta}_i).$$

This direct in-plane scattering of the Cooper pairs induces the same nodeless superconducting gap,

$$d_1(\mathbf{k}) \propto (\sin k_x a + i \sin k_y a), \qquad (6)$$

on all sheets of the Fermi surface. If we now approximate in a tight-binding spirit $U(\mathbf{r},\mathbf{r}') \approx U(\boldsymbol{\delta}_i)$, then the double integral in Eq. (5) factorizes in a product of two spatial integrals. Each of these integrals vanishes separately for $l = \gamma$ and $l' = \alpha, \beta$ because the orbitals from these bands have different parity with respect to $\hat{\sigma}_z$. Thus, it is essential to keep spatial dependence of $U(\mathbf{r},\mathbf{r}')$. For a Coulomb-type interaction, the off-diagonal matrix element ($l \neq l'$) in Eq. (5) has a dipolar reduction $(b/a)^2 \simeq 0.02$ [4] compared to the diagonal matrix elements ($l = l'$), where b is a characteristic spatial extension of the Wannier functions. In reality, the matrix element will be reduced even further because d_{xz} and d_{yz} orbitals mix in α and β bands only in a close vicinity of the Brillouin zone diagonals. Away from these directions, there is an extra approximate symmetry $\hat{\sigma}_{x(y)}$, which introduces an effective quadrupolar reduction of the matrix element in Eq. (5). Thus, the direct interaction of the p-wave order parameters between γ and α or β bands is significantly reduced, and the amplitude of the type-I gap (6) in passive bands should be much smaller than in the active band [24].

The next contribution to the interband scattering of the p-wave pairs in Eq. (4) comes from interlayer terms with $\mathbf{R}_m = \mathbf{R}_n$ and $\mathbf{R}_{n'} = \mathbf{R}_n \pm a\hat{\mathbf{x}}(\hat{\mathbf{y}})$, $\mathbf{R}_{m'} = \mathbf{R}_n \pm \frac{a}{2}\hat{\mathbf{x}} \pm \frac{a}{2}\hat{\mathbf{y}} \pm \frac{c}{2}\hat{\mathbf{z}}$ on a body-centered tetragonal lattice of Ru atoms. Summing over all contributing sites, the p-wave gap of the type (6) in the γ band induces

$$d_2(\mathbf{k}) \propto \left(\sin\frac{k_x a}{2}\cos\frac{k_y a}{2} + i\sin\frac{k_y a}{2}\cos\frac{k_x a}{2}\right)\cos\frac{k_z c}{2}, \qquad (7)$$

in α and β bands and vice versa. Existence of the type-II p-wave gap, but in all bands simultaneously, has been conjectured by Hasegawa *et al.* [19]. They, however, based their suggestion on an (unjustified) assumption of a repulsive interaction between electrons in a single Ru-O plane and an attraction only for electrons in adjacent layers. The type-II superconducting gap $d_2(\mathbf{k})$ has circular line nodes at $k_z = \pm\pi/c$. Importantly, they are stable with respect to an admixture of a small amount of the type-I gap, which shifts only the position of zeros along k_z. The two gaps are mixed with a real phase, as required by the time-reversal symmetry ($\hat{\mathcal{T}}\,\hat{\sigma}_x$), and nodes of $d_1 + d_2$ disappear only for $|d_1|_{\max} > |d_2|_{\max}$.

A reliable estimate for the strength of the scattering vertices corresponding to the two types of induced gaps (6) and (7) is possible only in the framework of complete microscopic theory of the Fermi liquid state in Sr_2RuO_4.

Note that scattering processes contributing to Eq. (7) have one symmetry cancellation factor less than Eq. (6), but instead they are reduced by a small overlap of the orbitals in adjacent layers. Information on the interlayer overlap can be obtained by analyzing the results of the high-precision de Haas–van Alphen measurements [10], which determined corrugation of the Fermi surface cylinders along k_z. Bergemann *et al.* [10] found the strongest corrugation in the β sheet of the Fermi surface: $\Delta k_{F,\beta} \sim \cos(k_z c/2)$ and a much weaker corrugation of the γ sheet: $\Delta k_{F,\gamma} \sim \cos(k_z c)$. This different periodicity naturally appears in a tight-binding model: A direct interlayer overlap $t_\perp'$ of d_{xz} (d_{yz}) orbitals yields a diagonal first-order contribution to the band energy: $\Delta\varepsilon_{\alpha,\beta}(\mathbf{k}, k_z) \simeq 8t_\perp' \cos(k_x a/2) \times \cos(k_y a/2)\cos(k_z c/2)$. On the other hand, planar d_{xy} orbitals do not hybridize across the layers. However, they can hybridize with d_{xz} (d_{yz}) orbitals in adjacent planes with the hopping amplitude $t_\perp''$, which produces the interband matrix element $8t_\perp'' \cos(k_{x(y)}a/2)\sin(k_{y(x)}a/2)\sin(k_z c/2)$. As a result, the corrugation of the γ sheet is a relatively weaker second-order effect $\Delta\varepsilon_\gamma(\mathbf{k}, k_z) \simeq [t_\perp'' \cos(k_z c/2)]^2/[\varepsilon_\gamma(\mathbf{k}) - \varepsilon_{\alpha,\beta}(\mathbf{k})]$, where smallness of the interband-interlayer hopping is partially compensated by closeness of the Fermi surfaces. Comparing to the experimental results, we find $t_\perp' \simeq -1$ meV and a somewhat larger magnitude for $t_\perp'' \simeq 3$ meV. The degree of mixing of different orbitals is controlled by the parameter $t_\perp''/[\varepsilon_\gamma(\mathbf{k}) - \varepsilon_{\alpha,\beta}(\mathbf{k})] \simeq 0.1$, which is comparable to or even exceeds the reduction of the matrix elements in Eq. (5).

We consider now the effect of line nodes in passive bands on thermodynamic properties of the superconducting state. In particular, we calculate the specific heat $C(T)$. Since the Cooper pair scattering between the α and the β sheets is not small, we adopt an effective two-band model for Sr_2RuO_4 and split the total density of states at the Fermi level according to $N_{01} : N_{02} = N_{0\gamma} : (N_{0\alpha} + N_{0\beta}) = 0.57 : 0.43$ based on the de Haas–van Alphen measurements [8]. We also assume that the active sheet for the superconducting instability is the γ sheet. Our main motivation for this assumption comes from comparison with the experimental data below. We adopt a weak-coupling approach and parametrize the pairing potential in the two bands by three parameters: $V_{11}(\mathbf{k}, \mathbf{k}') = -g_1 f(\mathbf{k}) f(\mathbf{k}')$, $V_{22}(\mathbf{k}, \mathbf{k}') = -g_2 \tilde{f}(\mathbf{k})\tilde{f}(\mathbf{k}')$, and $V_{12}(\mathbf{k}, \mathbf{k}') = -g_3 f(\mathbf{k}) \times \tilde{f}(\mathbf{k}')$, where we choose for simplicity $f(\mathbf{k}) = \mathbf{k}/k_F$ and $\tilde{f}(\mathbf{k}) = \sqrt{2}\,(\mathbf{k}/k_F)\cos(k_z/2)$, i.e., we presume that only interlayer processes contribute to the interband scattering of the Cooper pairs [25]. The pairing interaction in the active band is attractive ($g_1 > 0$), while interaction constants in the passive band (g_2) and between the bands (g_3) can have arbitrary sign. Solving the system of the two gap equations numerically, we determine the specific heat from

$$C(T) = 2\sum_{l,\mathbf{k}} E_{l\mathbf{k}} \frac{df(E_{l\mathbf{k}})}{dT}, \qquad (8)$$

where $E_{l\mathbf{k}}$ is the quasiparticle excitation energy $\sqrt{\epsilon_{l\mathbf{k}}^2 + \Delta_l^2(\mathbf{k})}$ (we consider only unitary triplet states), and $f(E_{l\mathbf{k}})$ is the corresponding Fermi distribution.

We present in the upper panel of Fig. 1 the specific heat for different choices of the coupling constants in the two-band model. The calculations have been done for a "typical" weak-coupling magnitude of $g_1 = 0.4$ (in units of the inverse total density of states) varying the two other parameters. The first three curves correspond to weak $g_3 = 0.01 g_1$ (No. 1), intermediate $g_3 = 0.07 g_1$ (No. 2), and strong $g_3 = 0.2 g_1$ (No. 3) interband scattering of the Cooper pairs, keeping in all cases $g_2 = 0.85 g_1$. Such a moderate change in the coupling constants between the two bands $\lambda_2/\lambda_1 = g_2 N_{02}/g_1 N_{01} = 0.64$ results in an order of magnitude difference in their bare transition temperatures: $T_{c2}/T_{c1} = 0.086$ in the weak-coupling theory. For the weaker interband coupling, the heat capacity develops a second peak, which reflects a nonzero bare transition temperature in the passive band. For the stronger interband coupling, the two gaps are tightly bound to each other and we return to an effective one-band behavior. The

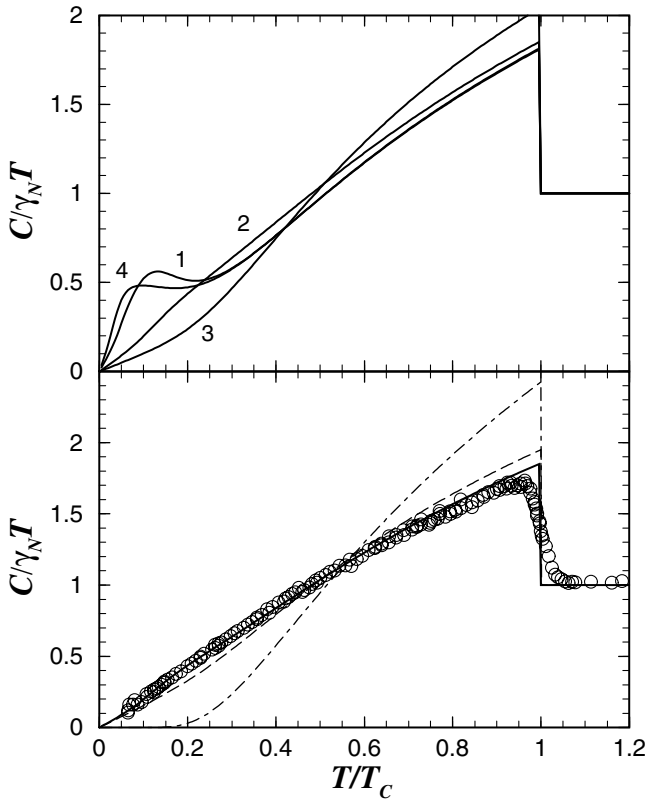

FIG. 1. Temperature dependence of the normalized specific heat. Upper panel: The two-band model results for various choices of the interaction parameters. Curves No. 1–3 correspond to $g_2/g_1 = 0.85$ and $g_3/g_1 = 0.01, 0.07, 0.2$, respectively. The curve No. 4 is for $g_2/g_1 = 0.1$ and $g_3/g_1 = 0.07$. Lower panel: Circles are experimental data for Sr_2RuO_4 [11]. One-band results are shown for an anisotropic gap with line nodes (dashed line) and for an isotropic gap (dot-dashed line). The solid line is the two-band model fit with $g_2 = 0.85 g_1$ and $g_3 = 0.07 g_1$.

curve No. 4 is an example of a shoulder in the temperature dependence of C/T, which arises for a reduced pairing interaction in the passive band $g_2 = 0.1g_1$ if we keep the same $g_3 = 0.07g_1$ as for the curve No. 2.

In the lower panel of Fig. 1 we present our fit to the experimental data of NishiZaki *et al.* [12] (same as the curve No. 2). The strength of the interband scattering vertex, g_3, is an order of magnitude smaller than the intraband vertices, g_1, g_2. The relative value is rather more than the simplest estimates which give the square of the interlayer mixing of orbitals in the bands at the Fermi energy. *A priori* estimates of g_1, g_2, and g_3 are difficult. We note, however, that direct Coulomb processes can contribute to g_3, and this may account for the increase in its value over the simplest estimate. One point which shows remarkably good agreement is the size of the specific heat jump at $T = T_c$ ($\Delta C/C_n \simeq 0.82$). This value coincides with the BCS value for the γ band alone ($1.43N_\gamma/N_0$) and is a direct confirmation of our choice of γ as the active band. Though it is impossible to fix all three parameters of the two-band model uniquely, this model can naturally explain a clear convex shape of the experimental data for C/T at low temperatures by choosing an intermediate strength of the interband scattering matrix element. In contrast, a one-band model with an anisotropic gap or the two-band model with a strong interband scattering predict a concave shape for C/T. From our fit we cannot also exclude a possibility of a small but finite $|\Delta_{\min}|$ in the passive bands, which appears if in-plane scattering amplitude slightly exceeds the interplane contribution (7).

The field dependence of the residual density of states at low temperatures suggests another argument in favor of the multiband scenario. Small magnetic fields ($\ll H_{c2}$) quickly restore about 40% of the total density of states [12]. In our model, this corresponds to the behavior exhibited by the curve No. 4, the upper panel in Fig. 1, in the case of temperature effects. Such a new feature in the external field arises because of an additional suppression of superconductivity in the passive α and β bands for $H \perp c$. Stronger c-axis dispersion in these bands leads to a larger coherence length ξ_c and an extra reduction of the bare H_{c2}^{ab}. This effect should disappear for $H \parallel c$ because of similar values of the in-plane Fermi velocities, which also agrees with the experiment [12]. It would be also interesting to reinvestigate the impurity effect on the residual density of states. Such an analysis has been done previously for the two-band model with a constant gap amplitude in the passive band [26]. Line nodes can modify the expected behavior and produce a gapless superconducting state in the passive bands.

In summary, we have shown that circular horizontal line nodes in the superconducting gap of Sr_2RuO_4 appear due to a weak and anisotropic interband proximity effect. This effect is a consequence of (i) non-s-wave symmetry of the Cooper pairs [conventional superconductors have generally a strong isotropic interband coupling dominated by the on-site term in Eq. (4)] and (ii) specific symmetry of the Ru orbitals, which give extra suppression of the matrix element in Eq. (5). Further experimental tests of our scenario should include studying effects of pressure, which can modify the strength of interlayer scattering amplitude for the Cooper pairs.

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