Momentum-resolved superconducting energy gaps of Sr$_2$RuO$_4$ from quasiparticle interference imaging

Rahul Sharma$^{a,b}$, Stephen D. Edkins$^c$, Zhenyu Wang$^d$, Andrey Kostin$^{a,b}$, Chanchal Sow$^{c,d}$, Yoshiteru Maeno$^e$, Andrew P. Mackenzie$^{a,b}$, J. C. Séamus Davis$^{a,g,i,j}$, and Vidya Madhavan$^{a,b}$

$^a$Laboratory of Atomic and Solid State Physics, Department of Physics, Cornell University, Ithaca, NY 14853; $^b$Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, NY 11973; $^c$Department of Applied Physics, Stanford University, Stanford, CA 94305; $^d$Department of Physics, University of Illinois, Urbana, IL 61801; $^e$Department of Physics, Kyoto University, Kyoto 606-8502, Kyoto, Japan; $^f$Department of Physics, Indian Institute of Technology-Kanpur, 208016 Uttar Pradesh, India; $^g$Physics of Quantum Materials Department, Max Planck Institute for Chemical Physics of Solids, D-01187 Dresden, Germany; $^h$School of Physics and Astronomy, University of St. Andrews, North Haugh, St. Andrews KY16 9SS, United Kingdom; $^i$Clarendon Laboratory, University of Oxford, Oxford OX1 3PU, United Kingdom; and $^j$Department of Physics, University College Cork, T12RSC Cork, Ireland

Sr$_2$RuO$_4$ has long been the focus of intense research interest because of conjectures that it is a correlated topological superconductor. It is the momentum space (k-space) structure of the superconducting energy gap $\Delta(k)$ on each band i that encodes its unknown superconducting order parameter. However, because the energy scales are so low, it has never been possible to directly measure the $\Delta(k)$ of Sr$_2$RuO$_4$. Here, we implement Bogoliubov quasiparticle interference (BQPI) imaging, a technique capable of high-precision measurement of multiband $\Delta(k)$. At $T = 90$ mK, we visualize a set of Bogoliubov scattering interference wavevectors $\tilde{\alpha} = 1 − 5$ consistent with eight gap nodes/minima that are all closely aligned to the $(\pm 1, \pm 1)$ crystal lattice directions on both the $\alpha$ and $\beta$ bands. Taking these observations in combination with other very recent advances in directional thermal conductivity [E. Hassinger et al., Phys. Rev. X 7, 011032 (2017)], temperature-dependent Knight shift [A. Pustogow et al., Nature 574, 72−79 (2019)], time-reversal symmetry conservation [S. Kashiyawa et al., Phys. Rev. B, 100, 094530 (2019)], and theory [A. T. Romer et al., Phys. Rev. Lett. 123, 247001 (2019); H. S. Roising, T. Scaffidi, F. Flicker, G. F. Langer, S. H. Simon, Phys. Rev. Res. 1, 033108 (2019); and O. Gingras, R. Nourafkan, A. S. Tremblay, M. Côté, Phys. Rev. Lett. 123, 217005 (2019)], the BQPI signature of Sr$_2$RuO$_4$ appears most consistent with $\Delta(k)$ having a $d_{x^2−y^2}$ ($B_2g$) symmetry.

Significance

Sr$_2$RuO$_4$ has been widely studied as a candidate correlated topological superconductor. However, the momentum space structure of the superconducting energy gaps, which encode both the pairing mechanism and its topological nature, has proven impossible to determine by conventional techniques. To address this challenge, we introduce Bogoliubov quasiparticle scattering interference visualization at millikelvin temperatures. We discover that the $\alpha$ and $\beta$ bands of Sr$_2$RuO$_4$ support thermodynamically prevalent superconducting energy gaps and that they each contain four gap nodes (or profound minima) that are contiguous to the $(0,0) \rightarrow (\pm 1, \pm 1)\pi$ lines in momentum space. In the context of other recent advances, these observations appear most consistent with a $d_{x^2−y^2}$ order parameter symmetry for Sr$_2$RuO$_4$.


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[^1]: To whom correspondence may be addressed. Email: jseamusdavis@gmail.com or vm1@illinois.edu.

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Sr$_2$RuO$_4$ as it has the proven capability of measuring extremely anisotropic $\Delta^\text{Δα}(k)$ having $d_{x^2-y^2}$ (even parity) or $p_{z\text{helical}}$ (odd parity) order parameters (18, 19). Obviously, this is because the maximum magnitude of any of these gaps (30, 31) is $|\Delta| \leq 350$ $\mu$eV so that temperature $T \leq 100$ mK and energy resolution with $\delta E \leq 100$ $\mu$eV are required to spectroscopically detect strongly anisotropic $\Delta^\text{min}(k)$ gap structures and/or their gap minima. Thus, techniques capable of band-resolved, high-resolution superconducting $\Delta(k)$ determination and specifically, of distinguishing the orientation of any gap minima on different bands are required. Bogoliubov quasiparticle interference (BQPI) imaging (32–38) has been proposed (39–41) to achieve these objectives for Sr$_2$RuO$_4$ as it has the proven capability of measuring extremely anisotropic (33–38), multiband (35, 36, 38) superconducting energy gaps with energy resolution (36, 38) $\delta E \leq 75$ $\mu$eV. Intuitively, this is possible because, when a highly anisotropic $\Delta^\text{Δα}$ opens on a given band, Bogoliubov quasiparticles $|k\epsilon(k)\rangle$ exist in the energy range $\Delta^\text{min}(k) < E < \Delta^\text{max}(k)$. Within this range, interference of impurity-scattered quasiparticles produces characteristic real-space ($r$-space) modulations in the density of electronic states (32, 39–41) $\delta N(r,E)$. The Bogoliubov quasiparticle dispersion $E_i(k)$ then exhibits closed constant energy contours surrounding FS $k$-points where minima in $\Delta^\text{Δα}$ occur. These $k$-space locations can be determined because $\delta N(r,E)$ modulations occur at the set of wavevectors $q_i(E)$ connecting them. These $q_i(E)$ are identified from maxima in $\delta N(q,E)$, the power spectral density Fourier transform of $\delta N(r,E)$.

For Sr$_2$RuO$_4$, BQPI signatures of different types of gap structures [for example, $\Delta^\text{Δα}(k)$; $\Delta^\text{Δβ}(k)$] may be anticipated by using a pedagogical Hamiltonian $\hat{H}(k) = \sum_\epsilon^\text{Δi}(k) \hat{H}(k) \psi(k)$, where

$$\hat{H}(k) = \begin{pmatrix}
\epsilon_\text{α}(k) & \Delta^\text{Δα}(k) & 0 & 0 \\
\Delta^\text{Δα}(k) & -\epsilon_\text{α}(k) & 0 & 0 \\
0 & 0 & \epsilon_\text{β}(k) & \Delta^\text{Δβ}(k) \\
0 & 0 & \Delta^\text{Δβ}(k) & -\epsilon_\text{β}(k)
\end{pmatrix},$$

in the basis $\psi^\text{±1}(k) = (c^\text{±1}_\alpha k \uparrow, c_{\alpha \rightarrow -k \uparrow}, c^\text{±1}_\beta k \downarrow, c_{\beta \rightarrow -k \downarrow})$ and $\epsilon_\text{α}(k)$, $\epsilon_\text{β}(k)$ are the band dispersion for the $\text{α,β}$ bands (39–41). The unperturbed Green’s function is $G^\text{0}(k,\epsilon) = (\epsilon + i\delta I - \hat{H}(k))^\text{−1}$, where $I$ is identity matrix and $\delta$ is the energy width broadening parameter. Both interband and intraband scattering could be considered using a $T$-matrix for all scattering processes as

$$T^{-1}(\omega) = (\langle V_{\text{interaction}} + V_{\text{internal}} \rangle \otimes \sigma_z) ^{-1} = \int \frac{dk}{(2\pi)^2} G^\text{0}(k,\omega).$$

However, interband scattering between the $\text{α,β}$ and $\gamma$ bands has not been the subject of any theoretical analysis for Sr$_2$RuO$_4$.
interference in Sr$_2$RuO$_4$ and, through comparison with the experimental challenge is to visualize Bogoliubov scattering from the SrO cleave surface (Fig. 1A). At the SrO termination layer occurs (31). The experimental challenge is to visualize Bogoliubov scattering interference in Sr$_2$RuO$_4$ and, through comparison with δN(q,E) predictions (39–41) to determine Δk(k).

To do so, we insert high-quality single crystals of Sr$_2$RuO$_4$ (Tc = 1.45 K) into a dilution refrigerator-based spectroscopic imaging scanning tunneling microscope (STM) and cleave them in cryogenic ultrahigh vacuum at T ≤ 1.8 K. This typically reveals an atomically flat SrO cleave surface (Fig. 1A), although sometimes, the RuO$_2$ termination layer occurs (31). At the SrO termination surfaces used throughout these studies (Fig. 1A), the tip sample differential tunneling conductance g(r,E) ≡ dI/dV(r,E = eV) is imaged to visualize scattering interference-induced modulations g(r,E) ∝ δN(q,E). In the normal state, g(r,E) measurements in the range −20 meV < E < 20 meV reveal g(q,E) ∝ δN(q,E) (Fig. 2A) with predominant scattering wavevectors q(E) shown as red and blue arrows in Fig. 2A. Quantitative comparison with the known FS k(E = 0) wavevectors (25) reveals that these arise from intraband scattering in both the β band and the α band (Fig. 1B) (SI Appendix, section II). As in previous quasiparticle interference studies of normal-state Sr$_2$RuO$_4$, the γ band is virtually undetectable, probably because the d$_{xy}$ character leads to small wavefunction overlap for tunneling into the STM tip (31). In any case, the αβ bands are directly identifiable from their normal-state scattering interference wavevectors throughout all of the BQPI studies reported below.

To measure Δk(k), we cool each sample to T = 90 mK (SI Appendix, section III) and typically measure g(r,E) ∝ δN(q,E) on a 128 × 128 grid in a 20-nm field of view. Typical junction formation parameters for these g(r,E) measurements are I$_S$ = 40 pA, V$_S$ = 1 mV, and |E| = 0, 100, 200, 300, 400 μeV spanning the maximum superconducting energy gap (SI Appendix, section III). The actual electron temperature is manifestly well below ~100 μeV/3.5 k$_B$ or ~300 mK because these BQPI images are distinct when the tip bias voltage is changed in energy steps of 100 μeV. A representative point spectrum from such a map is shown in Fig. 2C, showing the typical (30, 31) energy gap minimum Δ$_{max}$ ≈ 350 μeV. Fig. 2D shows a typical measured g(q, E = 100 μeV) deep within this superconducting gap. It is highly distinct from the g(q, E) measured near E$_F$ in the normal state (Fig. 2A) or at E > 350 μeV in the superconducting state (Fig. 4E), with many robust distinct q-space features. Differences in signal intensity between g(q, E = 400 μeV) measured in the normal and superconducting states occur due to
the greatly reduced bias modulation amplitude required for the latter. Most importantly, the distinct $g(q,E)_{\alpha}$ at $E = 0, 100, 200, 300 \mu$eV at $T = 90$ mK hold the key to understanding the superconducting magnitude on the FS for

\[ T = \frac{1}{2} \]

\[ E = \frac{1}{2} \]

$\alpha$ band with gap minima along $(\pm 1, \pm 1)$ on $\alpha \beta$ bands (Fig. 3A). In Fig. 3A, the hypothetical gap magnitudes $|\Delta_{\alpha}(k)|, |\Delta_{\beta}(k)|$ are indicated by the thickness of the curves overlaid on the $\alpha \beta$ FS. Fig. 3B identifies the consequent $k$-space regions where, because of minima in $\Delta_{\alpha}(k)$ and $\Delta_{\beta}(k)$, significant quasiparticle density of states is expected as $E \to 0$. The key BQPI wavevectors $q; j = 1, 2, 5$ (Fig. 3B) then connect these $k$-space

**Fig. 3.** Pedagogical Bogoliubov scattering interference model. (A) Gap magnitude on the FS for $\alpha \beta$ band with gap minima along $(\pm 1, \pm 1)$. (B) Regions of significant quasiparticle density $E \to 0$ for $\alpha \beta$ bands when gapped as shown in A. Major scattering vectors are labeled as $q_{i,j} = 1, 2, 3, 4, 5$. (C) Calculated $g(q,E)$ pattern from Eq. 3 for $\alpha \beta$ band from the model in A at $E = 100 \mu$eV. Key scattering wavevectors are indicated by $q_{i,j} = 1, 2, 3, 4, 5$.

**Fig. 4.** Imaging Bogoliubov scattering interference of Sr$_2$RuO$_4$. (A–D) Measured $g(q,E)$ images at $T = 90$ mK in superconducting state of Sr$_2$RuO$_4$ at $E = 1$ meV, 300 $\mu$eV, 200 $\mu$eV, and 100 $\mu$eV. Red crosses denote Bragg peaks. Typically, the features at lowest $|E| = 100$ eV, 300 $\mu$eV, and 100 $\mu$eV. Red crosses denote Bragg peaks. Typiﬁcally, the features at lowest $|E| = 100$ eV, 300 $\mu$eV, and 100 $\mu$eV. Red crosses denote Bragg peaks. The key BQPI wavevectors $q; j = 1, 2, 5$ (Fig. 3B) then connect these $k$-space

energy gap structure of Sr$_2$RuO$_4$ using Bogoliubov scattering interference (39–41). At the most elementary level, Fig. 2D reveals spectroscopically that, consistent with a wide variety of other techniques (S, 7–9), a strong Bogoliubov quasiparticle density of states exists deep within the superconducting gap of this material.

To aid with interpretation of these $g(q,E)$ data, we explore a pedagogical model for $\Delta(k)$ having gap zeros along $(\pm 1, \pm 1)$ on $\alpha \beta$ bands (Fig. 3A). In Fig. 3A, the hypothetical gap magnitudes $|\Delta_{\alpha}(k)|, |\Delta_{\beta}(k)|$ are indicated by the thickness of the curves overlaid on the $\alpha \beta$ FS. Fig. 3B identifies the consequent $k$-space regions where, because of minima in $\Delta_{\alpha}(k)$ and $\Delta_{\beta}(k)$, significant quasiparticle density of states is expected as $E \to 0$. The key BQPI wavevectors $q; j = 1, 2, 5$ (Fig. 3B) then connect these $k$-space

\[ q_{1,2} \]

\[ q_{2,5} \]
Fig. 5. Predominant $\Delta_{\alpha}(k)$, $\Delta_{\beta}(k)$ with gap min/nodes along (+1, ±1). (A) Predicted $\delta N(q,E)$ for $\Delta_{\alpha}(k)$, $\Delta_{\beta}(k)$ at $E = 100 \, \mu$eV with red (blue) circles denoting the features arising from scattering from $\alpha$- and $\beta$-bands. (B) Measured $g(q,E)$ pattern at $E = 100 \, \mu$eV with circles at similar locations as in A. (C) Measured $g(q,E)$ pattern at $E = 100 \, \mu$eV with circles at similar locations as in A. The angular width of maxima at $q_j = 3.4$ in this image indicates that minima in $\Delta_{\alpha}(k)$, $\Delta_{\beta}(k)$ occur at less than ∼0.05 rad from the (0,0) → (+1, ±1)π/a k-space lines. (D) Superconducting energy gap $\Delta_{\alpha}(k)$ structure of Sr$_2$RuO$_4$ consistent with the $g(q,E)$ data presented here and in Fig. 4.

locations as shown. Fig. 3C shows typical evaluations of $\delta N(q,E)$ from Eq. 3 for this model, with the key BOPI wavevectors overlaid. Here $K_{1,2,3,4}$ (Fig. 3B) occur due to the gap minima/nodes on the $\beta$ band, while $q_1, q_5$ (Fig. 3B) occur due to gap minima/nodes on the $\alpha$ band. Observation of BOPI intensity in $g(q,E)$ data at these specific wavevectors $q_j = 1.25$ would give direct evidence for a superconducting energy gap structure (Fig. 3A) with gap minima/nodes along the (+1, ±1) on the $\alpha, \beta$ bands of Sr$_2$RuO$_4$.

Fig. 4 contains the key experimental results of this study: the measured $g(q,E)$ at multiple energies within the superconducting gap of Sr$_2$RuO$_4$ at $T = 90$ mK. The $g(q,E = 1 \, \text{meV})$ in Fig. 4A is shown for comparison. Predictions from Eq. 3 for $\delta N(q,E)$ with the gap model in Fig. 3A are shown at corresponding energies to the measured $g(q,E)$ in Fig. 4E–H. The simultaneously measured $g(q,E = 1 \, \text{meV})$ exhibits direct signatures of $\alpha, \beta$-band scattering interference as identified from our normal-state studies (SI Appendix, section II). Since the electron tunneling manifestly occurs to the $\alpha, \beta$ bands and simultaneously exhibits a single-particle spectrum showing gap maximum $\Delta_{\text{max}} \approx 350 \, \mu$eV (Fig. 2C), we conclude that this superconducting gap is hosted by the $\alpha, \beta$ bands (31). Additionally, because $\Delta_{\text{max}} \approx 350 \, \mu$eV is a consistent gap maximum for the bulk superconducting critical temperature $T_c = 1.45 \, \text{K}$ (because $2 \Delta_{\text{max}}/k_B T_c \approx 4$), this indicates that $\Delta_{\alpha}(k)$, $\Delta_{\beta}(k)$ are principal energy gaps of Sr$_2$RuO$_4$.

Then, when Bogoliubov scattering interference is visualized at subgap energies $|E| < \Delta_{\text{max}}$, a distinctive $g(q,E)$ pattern emerges. It exhibits clear maxima at specific $q$ vectors (Fig. 4 B–D) that evolve but do not disappear as $E \to 0$. Theories of Sr$_2$RuO$_4$ BOPI demonstrate how these $q$ vectors encode the direction of the gap minima in $\Delta_{\alpha}(k)$, $\Delta_{\beta}(k)$, and also predict a very weak dispersion of the subgap $g(q,E)$ with energy (39–41). The observed pattern of $g(q,E = 100 \, \mu$eV) maxima in Fig. 4D is quite representative and conforms to predicted $\delta N(q,E = 100 \, \mu$eV) of the energy gap model in Fig. 3. Specifically, in Fig. 5A, the predicted BOPI wavevectors $q_1, q_2, q_3, q_4, q_5$ and $q_6$ from the $\alpha, \beta$-band model with nodes/minima along (+1, ±1) (circles in Fig. 5A) are compared with the locations of five distinct local maxima in $g(q,E = 100 \, \mu$eV) in Fig. 5B and found to be in good agreement. The immediate implication is that eight nodes/minima occur in $\Delta_{\alpha}(k)$, $\Delta_{\beta}(k)$ at the locations where the $\alpha, \beta$ bands cross the (0,0) → (+1, ±1)$\pi/a$ symmetry axes. Because the measured $g(q,E)$ are distinct for $E = 0,100,200,300 \, \mu$eV (Fig. 4), the energy resolution $\delta E$ is demonstrably $\delta E < 100 \, \mu$eV, while from the measurement parameters, we estimate that $\delta E \lesssim 75 \, \mu$eV. This means that, if minima (as opposed to nodes) occur in $\Delta_{\alpha}(k)$ and $\Delta_{\beta}(k)$, they exist below the energy scale $|E| = 75 \, \mu$eV. Moreover, analysis of the $g(q,E = 0)$ data shown in Fig. 5C indicates that all eight gap minima/nodes have an angular displacement about (0,0) in $k$ space within ±0.05 rad from the (0,0) → (+1, ±1)$\pi/a$ lines (SI Appendix, section IV). No features expected of $\Delta_{\alpha}(k)$ (SI Appendix, section II) are detected. As to the signature in $g(q,0)$ of the predicted minima on $\Delta_{\alpha}(k)$ in an odd-parity state (figure 2 of ref. 18, figure 5 of ref. 31), these are expected to appear as $g(q,0)$ maxima at wavevectors at least ±0.1 rad away from the (0,0) → (+1, ±1)$\pi/a$ lines (18, 19, 28, 31), or if the energy resolution is insufficient to resolve them, they should exhibit as a broad arc connecting these $g(q,0)$ maxima. As discussed in SI Appendix, section V, neither of these signatures has been detected within the available signal to noise ratio. Moreover, in the same models (18, 19, 28, 31), the minimum that occurs on $\Delta_{\alpha}(k)$ is typically shallow, whereas the measured minimum on $\Delta_{\alpha}(k)$ is deep reaching to within 75 μeV of zero (Fig. 5C). Therefore, a gap structure for both $\Delta_{\alpha}(k)$ and $\Delta_{\beta}(k)$ as shown in Fig. 5D seems most consistent with our present data.
In this project, we introduced momentum-resolved spectroscopic measurements of the superconducting gap structure in Sr$_2$RuO$_4$. They reveal eight nodes or deep minima in $\Delta_\alpha(k)$ and $\Delta_\beta(k)$, which occur in close proximity to where the $\alpha$: $\beta$ bands cross the $(0,0)\rightarrow(\pm 1, \pm 1)$/$a$ lines. In light of recent thermal conductivity (9), Knight shift (10), current-field reversal (13) experiments, and advanced theory (18–20, 28), several key implications emerge from this observation. If TRS was actually broken (14–17) by $\Delta(k)$ of Sr$_2$RuO$_4$, but the order parameter has even parity (10), then $s' + id_{xy}(\pm 2)$ (18) or $d_{x^2-y^2} + id_{xy}$ (42) states would be plausible. Based on our BQPI data along with thermodynamic/transport studies (6–9), $d_{x^2-y^2} + id_{xy}$ appears consistent because of its circumferential nodes in the $\kappa_h\kappa_h$ plane, but $s' + id_{xy}(\pm 2)$ might be consistent. However, for such order parameters, the transition temperature should split under a crystal symmetry-breaking field, but that effect is reportedly absent in multiple relevant studies (11, 12, 43–46). On the other hand, if TRS is preserved (13), the BQPI data (Figs. 3 and 4) are most consistent with a helical odd parity $\text{ph}e\text{lical}$ order parameter $\alpha$ (18, 19, 28) with $A_{1g}$ symmetry or an even parity $d_{x'y'}$ order parameter (18–20, 28) with $B_{1g}$ symmetry. In terms of the detailed $k$-space structure of $\Delta(k)$, these two cases are distinct. The former exhibits minima but not nodes on the $a\beta$ bands, their $k$-space locations are not constrained by crystal symmetry, and the magnetic fields on different bands are not necessarily co-aligned in $k$ space (18, 19). The latter exhibits true nodes on both the $a$ and $\beta$ bands, with $k$-space locations that are constrained precisely by crystal symmetry to lie along the $(\pm 1, \pm 1)$ directions. Our BQPI data (Fig. 4) imply that the four energy gap minima/nodes of both $\Delta_\alpha(k)$ and $\Delta_\beta(k)$ exist below the energy scale $|E| = 75$ $\mu$eV and that they occur within an angular distance from the $(0,0)\rightarrow(\pm 1, \pm 1)$/$a$ $k$-space lines of $\approx 0.05$ rad. Overall, therefore, these observations seem most consistent with a $d_{x'y'}$ order parameter symmetry for Sr$_2$RuO$_4$.

**Methods**

Additional information can be found in the **Methods** section of the manuscript.