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Electronic reconstruction forming a $C_2$-symmetric Dirac semimetal in Ca$_3$Ru$_2$O$_7$

M. Horio$^{1,2,3}$, Q. Wang$^2$, V. Granata$^{2,3}$, K. P. Kramer$^4$, Y. Sassa$^4$, S. Jöhr$^1$, D. Sutter$^1$, A. Bold$^1$, L. Das$^1$, Y. Xu$^1$, R. Frison$^5$, R. Fittipaldi$^{2,3}$, T. K. Kim$^6$, C. Cacho$^6$, J. E. Rault$^7$, F. Le Fèvre$^8$, F. Bertran$^8$, N. C. Plumb$^8$, M. Shi$^8$, A. Vecchione$^{10,3}$, M. H. Fischer$^1$ and J. Chang$^8$

Electronic band structures in solids stem from a periodic potential reflecting the structure of either the crystal lattice or electronic order. In the stoichiometric ruthenate Ca$_3$Ru$_2$O$_7$, numerous Fermi surface-sensitive probes indicate a low-temperature electronic reconstruction. Yet, the causality and the reconstructed band structure remain unsolved. Here, we show by angle-resolved photoemission spectroscopy, how in Ca$_3$Ru$_2$O$_7$ a $C_2$-symmetric massive Dirac semimetal is realized through a Brillouin-zone preserving electronic reconstruction. This Dirac semimetal emerges in a two-stage transition upon cooling. The Dirac point and band velocities are consistent with constraints set by quantum oscillation, thermodynamic, and transport experiments, suggesting that the complete Fermi surface is resolved. The reconstructed structure— incompatible with translational-symmetry-breaking density waves—serves as an important test for band structure calculations of correlated electron systems.

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INTRODUCTION

A Fermi-surface reconstruction refers to the sudden change of the electronic band structure as a function of a tuning parameter. As most electronic properties are governed by electrons in the vicinity of the Fermi level, a change of the Fermi-surface topology can have profound ramifications. Those reconstructions that are not obviously linked to a symmetry change of the crystal structure are of particular interest. Common triggers of Fermi-surface reconstructions are translational-symmetry breaking spin- or charge-density waves. Typically, this reduction of symmetry and the resulting folding of the Brillouin zone lead large Fermi-surface contours to split into smaller pockets. Once reconstructed, it is, however, often difficult to identify the Fermi surface structure. The high-temperature superconductor YBa$_2$Cu$_3$O$_7$ is a good example of this. In the underdoped regime, charge-density-wave order$^{1-3}$ clearly reconstructs the Fermi surface. Although quantum oscillations$^{4,5}$ and transport$^{6,7}$ experiments have revealed the existence of an electron pocket, the reconstructed Fermi-surface topology is, despite strong effort, still not clarified$^8$. Another prominent example is the orthorhombic bilayer ruthenate Ca$_3$Ru$_2$O$_7$. An initial A-type antiferromagnetic (AFM) order setting in at $T_N = 56$K$^9$ has no significant impact on the transport properties$^{10}$. A spin reorientation from the orthorhombic $a$- to the $b$-axis direction$^{11,12}$ occurs at $T_{s1} = 48$K. While this lattice-space-group preserving reorientation$^9$ naively might appear to be of minor consequence, the transition at $T_{s1} = 48$K marks a dramatic electronic transformation.

Across this transformation, the Seebeck coefficient undergoes a sharp sign change taking large negative values below the transition temperature$^{13,14}$. $T_{s1}$ is also the onset of in-plane anisotropic transport properties$^{14}$. Although transport and thermodynamic experiments provide information about Fermi surface area and electronic masses$^{15,16}$, the complete reconstructed Fermi surface has so far remained undetermined. Previously reported angle-resolved photoemission spectroscopy (ARPES) data have revealed the existence of a boomerang-shaped Fermi surface$^{17}$. Obviously, a single pocket is insufficient to explain the observed ambipolar electronic properties$^{18}$. Although a density-wave state has never been identified$^{12}$, a common assumption is that a translational-symmetry-breaking order reconstructs the Fermi surface into multiple sheets. However, as long as the complete Fermi surface and its orbital composition remain unidentified, so do the reconstructing mechanism.

Here, we provide by direct ARPES experiments the complete Fermi surface structure of Ca$_3$Ru$_2$O$_7$ across the electronic reconstruction. Above the reconstructing temperature, the low-energy electronic structure resembles that of a strongly correlated metal consistent with its orthorhombic crystal structure. The reconstructed Fermi surface by contrast consists of a small electron pocket formed by massive Dirac fermions along the short-axis orthorhombic zone boundary and a boomerang-like hole pocket in vicinity to the long-axis zone boundary. As the orthorhombic order parameter $|a - b|/(a + b)$ remains essentially unchanged across the reconstruction$^9$, the sudden emergence of such a dramatic Fermi surface anisotropy is unexpected. We furthermore demonstrate that the Fermi surface transformation appears in two steps. The anisotropic zone-boundary Fermi surfaces first appear below $T_{s1} = 48$K and eventually the Dirac fermions settle into the low-temperature structure below $T_{s2} = 30$K. Throughout this temperature evolution, no signature of Brillouin-zone folding is identified, excluding translational-symmetry breaking density-wave/orbital orders as the origin of the phase transition at $T_{s1} = 48$K. We argue that the reconstructed
Fermi surface should be understood from the $d_{xz}$ and $d_{yz}$ orbitals whereas the $d_{xy}$ sector is not crossing the Fermi level in the reconstructed phase. The revelation of the complete Fermi surface reconstruction provides an ideal test-bed for ab-initio band structure calculations beyond density-functional-theory concepts.

**RESULTS**

**High-temperature state**

The Fermi surface and low-energy electronic structure of the Ca$_3$Ru$_2$O$_7$ normal state—above the Néel temperature $T_N = 56$ K—are presented in Fig. 1a, d, and e. The orthorhombic zone boundary is indicated by black dashed lines in Fig. 1a. All quasi-particle dispersions are broad irrespective of whether linear $p$- or $s$-polarised light is used. Part of the Fermi surface consists of straight sectors running diagonally through the orthorhombic Brillouin zone. This quasi-one-dimensional structure remains essentially unchanged across the Néel transition at $T_N = 56$ K [see Fig. 1b, f, and g]. Furthermore, the orthorhombic zone boundary points $M_a = (±π/a, 0)$ and $M_b = (0, ±π/b)$ are virtually indistinguishable (See also Supplementary Fig. S1).

**Fermi surface anisotropy**

Across the structural transition at $T_s = 48$ K, however, the electronic structure undergoes a dramatic reconstruction. This is evidenced by the emergence of a fast dispersing band and a tiny Fermi surface around $M_a$—see Fig. 1c, h, and i. Remarkably, this small Fermi surface sheet is absent at $M_b$. Instead, as previously reported$^{17}$, boomerang-like Fermi surface sheets are found around the $M_b$ point. Therefore, in contrast to $T > T_s$, the low-temperature structure appears highly anisotropic featuring different Fermi surface topology around $M_a$ and $M_b$. This Fermi surface reconstruction appears without change of the crystal lattice space group and with minute (~1%) reduction of the orthorhombic order parameter$^9$.

To exclude the possibility that this $C_2$ symmetry is an artefact of photoionization-matrix-element effects, we follow a standard measurement protocol$^{18,19}$. That is to carry out Fermi surface mappings with azimuthal angles differing from each other by 90° [see Fig. 2a, b]. In situ azimuthal rotation implies that the Fermi surface maps in Fig. 2a, b are from the same surface. Here, $k_{||}$ ($k_{\perp}$) on the horizontal (vertical) axis represents the momentum parallel (perpendicular) to the electron-analyser slit. The electronic structure with a tiny Fermi pocket around the $M_a$ point and boomerang-like features near $M_b$ tracks the azimuthal rotation—see Fig. 2a–d. The $C_2$-symmetric electronic structure is also revealed by the band dispersions. Along the $M_a$–$Γ$ and $M_b$–$Γ$ directions, the band curvature around $M_a$ and $M_b$ are clearly different [Fig. 2e, g]. An electron pocket is formed around $M_a$ whereas two hole-like pockets are found on each side of $M_b$. In a similar fashion, dispersions along the $M_a$–$X$ and $M_b$–$X$ directions are inequivalent [Fig. 2f, h]. Electron-like band curvature is found around $M_a$ whereas no Fermi-level crossing is observed along $M_a$–$X$. These results exclude matrix-element effects as the source of the observed anisotropy.

**Two-stage Fermi surface reconstruction**

Tracking the temperature dependence of the band structure reveals two electronic temperature scales. The electronic band structure along $M_a$–$Γ$ and $M_b$–$Γ$ is shown for temperatures going from 16 to 50 K above $T_s = 48$ K [Fig. 3e, j and k], all bands appear with broad line-shapes. Once cooled below $T_s$, well-defined bands around the $M_a$ and $M_b$ points emerge [see Fig. 3d, i, and k]. The appearance of another electron-like band around the $Γ$ point is...
accompanied by a gap \( \Delta \) opening below \( T_1 \) [see Fig. 3g–i and Supplementary Fig. S4]. The band structures around \( M_a \) and \( M_b \) are inequivalent not only in terms of curvature but also in terms of temperature dependence. The \( M_a-i \) band dispersion is temperature-dependent whereas the corresponding structure around \( M_b \) is virtually insensitive to temperature. Examining the \( M_a-i \) direction, two inequivalent bands with different Fermi momenta are observed for \( 30 < T < 48 \) K whereas only a single set of bands is resolved for \( T > 30 \) K—see Fig. 3. The two bands around \( M_a \) display asymmetric matrix elements. Momentum distribution curves (MDCs) at \( E_F \) and \( 30 < T < 48 \) K are therefore not symmetric around \( M_a \). Upon cooling below \( 30 \) K, the single electron pocket around \( M_a \) displays symmetric Fermi momenta \( k_F \) despite the asymmetric matrix elements. To illustrate the transition between the two- and single-band situation, we define \( \Delta k_F \) as the reciprocal-space distance between the two MDC-intensity maxima. Across \( T_2 = 30 \) K, \( \Delta k_F \) drops by a factor of two. This observation is independent of incident-light polarisation (see Supplementary Figs. S5 and S6). On the other hand, the gap \( \Delta \) evolves smoothly across \( T_2 \) [see Fig. 3g–i and Supplementary Fig. S4], suggesting that the states around \( \Gamma \) are not involved in this transition. Although \( T_2 = 30 \) K remains to be identified as a thermodynamic temperature scale, it does coincide with the onset of a strong negative Nernst effect response\(^{14} \) [see Fig. 3i]. We also notice that the reduction of \( \Delta k_F \) below \( T_2 \) is consistent with an increasing Nernst effect \( \nu / T \propto \mu / \epsilon_F \) where \( \mu \) is the electron mobility and \( \epsilon_F \) the Fermi energy\(^{20} \), as lower \( \Delta k_F \) implies a smaller Fermi energy. The low-temperature Fermi surface thus emerges as a result of two reconstructions. First below \( T_1 = 48 \) K, a fast dispersing band appears around \( M_a \) and \( M_b \) with a gap opening for other bands. Next, the band dispersion along the \( M_a-i \) direction undergoes a second transformation across \( T_2 = 30 \) K.

**Low-temperature electronic structure**

With the exception of the features around the \( M_a \) and \( M_b \) points, all other bands are not crossing the Fermi level for \( T < T_2 \)—see Fig. 2e–h. Around the \( M_a \) point, two hole-like bands—forming an M-shaped structure—are found [Fig. 2g]. While the hole-like band touches \( E_F \) along the \( M_a-i \) direction [Fig. 2g], the band top sinks below \( E_F \) upon moving away from it by \( \sim 0.1 \) meV. (See Supplementary Fig. S7) consistent with a previous report\(^{17} \). The boomerang-like feature thus forms a closed hole-like Fermi surface. Around the \( M_b \) point, the electron-like Fermi surface pocket is revealed by a high-resolution map in Fig. 4(a). The electron pocket is elliptical with \( k_{\parallel}^2 = 0.04 \pi / a \) and \( k_{\perp}^2 = 0.07 \pi / b \) along the \( M_a-i \) and \( M_a-X \) directions, respectively. The Fermi surface area \( A_{FS} = \pi k_{\parallel} k_{\perp} \) corresponds to 0.23% of the orthorhombic Brillouin zone. Inspecting the band dispersion along the \( M_a-i \) direction reveals a Dirac-cone structure with the Dirac point placed about \( E_0 = 15 \) meV below \( E_F \) [Fig. 4b]. The two-peak MDC profile found at \( E_F \) merges into a single peak at \( E_D \) and then

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**Fig. 2** **Breaking of fourfold rotational symmetry.** a, b Fermi surface maps recorded at \( T = 16 \) K (\( h\nu = 75 \) eV) for two sample azimuthal angles that are 90° apart as indicated in the schematics. The azimuthal angle rotation was operated in situ and hence the sample surface is identical. \( k_{\parallel} \) \( (k_{\perp} \) represents momentum parallel (perpendicular) to the electron-analyser slit. The spectral intensity was integrated within \( E_F \pm 20 \) meV, washing-out spectral gaps within the integration window. c, d Fermi surfaces from the tight-binding model (see Supplementary note 1). The sheets not observed in the experiment are indicated in grey. a, b Energy distribution maps along \( M_a-i \) for \( s_1 \) and \( s_2 \). The geometric distribution curve (GDC) is inequivalent not only in terms of curvature but also in terms of temperature dependence. Around the \( M \) point, two inequivalent bands with different Fermi momenta are observed for \( 30 < T < 48 \) K whereas only a single set of bands is resolved for \( T > 30 \) K—see Fig. 3. The two bands around \( M_a \) display asymmetric matrix elements. Momentum distribution curves (MDCs) at \( E_F \) and \( 30 < T < 48 \) K are therefore not symmetric around \( M_a \). Upon cooling below \( 30 \) K, the single electron pocket around \( M_a \) displays symmetric Fermi momenta \( k_F \) despite the asymmetric matrix elements. To illustrate the transition between the two- and single-band situation, we define \( \Delta k_F \) as the reciprocal-space distance between the two MDC-intensity maxima. Across \( T_2 = 30 \) K, \( \Delta k_F \) drops by a factor of two. This observation is independent of incident-light polarisation (see Supplementary Figs. S5 and S6). On the other hand, the gap \( \Delta \) evolves smoothly across \( T_2 \) [see Fig. 3g–i and Supplementary Fig. S4], suggesting that the states around \( \Gamma \) are not involved in this transition. Although \( T_2 = 30 \) K remains to be identified as a thermodynamic temperature scale, it does coincide with the onset of a strong negative Nernst effect response\(^{14} \) [see Fig. 3i]. We also notice that the reduction of \( \Delta k_F \) below \( T_2 \) is consistent with an increasing Nernst effect \( \nu / T \propto \mu / \epsilon_F \) where \( \mu \) is the electron mobility and \( \epsilon_F \) the Fermi energy\(^{20} \), as lower \( \Delta k_F \) implies a smaller Fermi energy. The low-temperature Fermi surface thus emerges as a result of two reconstructions. First below \( T_1 = 48 \) K, a fast dispersing band appears around \( M_a \) and \( M_b \) with a gap opening for other bands. Next, the band dispersion along the \( M_a-i \) direction undergoes a second transformation across \( T_2 = 30 \) K.
The free electron mass. Assuming a parabolic band dispersion \( m^* \) on the electron pocket, Lifshitz–Kosevich analysis of the \( \sim 35 \) \( T \) quantum-oscillation frequency yield \( m^*_e \approx 0.65 m_e \), where \( m_e \) is the free electron mass. Assuming a parabolic band dispersion \( m^*_e = h^2 k^2 /2m_e \) where \( h_e = 15 \) meV is the Fermi energy, the effective mass \( m^*_e \) is significantly larger than that inferred from quantum-oscillation experiments. A linear band dispersion \( E_k = \nu_F k \) provides a much better agreement \( m^*_e = \nu_F /\nu_F \approx h^2 k^2 /E_F = 0.49 m_e \). This fact reinforces the interpretation of Dirac fermions around \( M_e \). The boomerang band along \( \Gamma-M_e \) has comparable Fermi velocity to that of the electron pocket along \( \Gamma-M_e \). Estimation of the Fermi energy from linear extrapolation of the M-shaped band dispersion yields \( \nu_F \approx 5 \) meV, which is about three times smaller than that obtained for the electron pocket. As the hole pocket area—according to quantum-oscillation measurements—is also about three times smaller than the electron sheet, we estimate the hole-like carriers to have a comparable effective mass of \( m^*_h = 0.49 m_e \). With two hole and one electron pocket per Brillouin zone, a Sommerfeld constant of \( \gamma = 2.1 \) mJ mol\(^{-1}\) K\(^{-2}\) is found, with mol referring to one formula unit. Here, we assumed two-dimensional band dispersions without bi-layer splitting and used the two-dimensional expression \( \gamma = n_e \nu_F^2 (ab/3)^{12} \). The Avogadro constant, \( h \) is the reduced Planck constant, and \( m_e \) is the effective mass. In addition, spin polarisation within the RuO\(_2\) plane was employed to treat the A-type AFM order. As our estimate is in reason-able agreement with the value \( \gamma = 2.8-3.4 \) mJ mol\(^{-1}\) K\(^{-2}\) obtained by specific heat experiments, we conclude that our experiments reveal the entire bulk Fermi surface.

The two-stage transformation of the electronic structure has a clear impact on all transport coefficients. A remarkable increase of in- and out-of-plane resistivity appears across \( T_{\text{ns}} \approx 48 \) K. Simultaneously, the Seebeck coefficient changes sign going from weak positive to large negative values across \( T_{\text{ns}} \). Although less sharp, the Hall coefficient also changes sign (from positive to negative) across \( T_{\text{ns}} \). While the Hall coefficient takes increasingly large negative values, the Seebeck coefficient displays a complicated temperature dependence that is different along the \( a \) and \( b \) directions. This temperature dependence is a typical signature of ambipolar transport behaviour, where both electron- and hole-like carriers are contributing. Furthermore, the low-temperature Hall coefficient \( R_H \) that (in different studies) ranges from \(-0.5 \times 10^{-7} \) m\(^2\) C\(^{-1}\) to \(-1.4 \times 10^{-7} \) m\(^2\) C\(^{-1}\) cannot be explained by the electron pocket that alone should generate \( R_H = -1/(n_e e) = -8.0 \times 10^{-7} \) m\(^2\) C\(^{-1}\). Using the combined ARPES and quantum-oscillation knowledge that \( n_e = 7.8 \times 10^{18} \) cm\(^{-3}\) and \( n_h \approx 2 \times n_e/3 \), a two band model yields \( R_H = (2a^2/3 - 1)/(n_e e) \).
temperature \((\text{the matrix-element effect of the electron pocket around M})\) 

- Whether they possess a finite mass as illustrated by green dispersions reproduced by the tight-binding model (see Supplementary Note 1).
- MDCs, extracted from b at binding energies as indicated.
- Fermi level, this gap is irrelevant for transport and thermodynamic measurements. While our tight-binding model based on the Ru \(t_{2g}\) orbitals is too simplistic to capture all the features and does not include the actual electronic instability, it reproduces the most salient features of both the high- and low-temperature dispersions. We thus conclude that the low-temperature low-energy band structure stems primarily from the \(d_{xy}\) and \(d_{yz}\) Ru orbitals.

A fundamental remaining question links to the triggering mechanism that induces the Dirac semimetal. Specific heat suggests that the phase transition at \(T_1\) involves a large entropy change and unlike other layered ruthenates, the ground state is a low density-of-state semimetal. It has been argued that the reorientation of the magnetic moments alone can not account for this large entropy change. Upon cooling, an energy gain of the system is manifested by an electronic reconstruction that opens a gap leaving only small Fermi surface pockets around the zone boundaries. Most likely, this Fermi surface reconstruction is triggered by an electronic mechanism. Density-wave orders breaking translation symmetry are, however, excluded since the reconstruction preserves the original Brillouin-zone boundaries. This leads us to speculate alternative scenarios, with electron correlations likely involved in some way. If so, the situation resembles that of the single-layer counterpart \(\text{Ca}_3\text{Ru}_2\text{O}_7\) where the instability toward a Mott-insulating state triggers a large \(c\)-axis lattice contraction. Indeed, a \(c\)-axis lattice contraction is found across the first (48 K) transition though this effect is much less pronounced in \(\text{Ca}_3\text{Ru}_2\text{O}_7\). Alternatively, it has been proposed that \(\text{Ca}_3\text{Ru}_2\text{O}_7\) hosts magnetic anapole order. This would connect \(\text{Ca}_3\text{Ru}_2\text{O}_7\) with hidden order problems in the sense that it is very difficult to demonstrate experimentally.

Note added after completion of this work: A recent complementary ARPES study conducted at \(T \geq 30\) K suggested using DFT calculations including Rashba-type spin-orbit coupling that the electronic reconstruction across \(T = 48\) K can be understood from the magnetic-moment reorientation alone without the need for additional hidden order.
METHODS
Sample characterisation

High-quality single crystals of Ca$_3$Ru$_2$O$_7$ were grown by floating zone technique. The electronic transition at $T_s$ = 48 K was checked by thermopower measurements (see Supplementary Fig. S10) and found in agreement with existing literature. Detwinning of orthorhombic domains was achieved with a thermo-mechanical device and monitored by polarised light microscopy. The resulting monodomain constitutes 99% (or more) of the sample volume according to X-ray diffraction measurements (see Supplementary Fig. S10).

ARPES experiments

ARPES experiments were carried out at the SIS$^{35}$, CASSIOPEEE (https://www.synchrotron-soleil.fr/en/beamlines/cassiopee), and I05 beamlines of the Swiss Light Source,SOLEIL synchrotron, and Diamond Light Source, respectively. Pristine surfaces were obtained by top-post cleaving at $T > T_s$ (80 K). Incident photons $h\nu = 31-115$ eV, providing high in-plane and modest out-of-plane momentum resolution, were used for this study. Consistent results were obtained on different crystals and upon cooling and heating through the critical temperature $T_s = 48$ K below which the electronic structure is reconstructed. ARPES data are presented using orthorhombic notation with lattice parameters $a = 5.37$ Å and $b = 5.54$ Å.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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AUTHOR CONTRIBUTIONS

V.G., R.F., and A.V. grew and prepared single crystals. L.D. and Y.X. performed thermopower measurements. S.I. and M.H. detwinned single crystals. M.H., Q.W., S.I., and R.F. carried out x-ray and Laue diffraction measurements. M.H., Q.W., K.P.K., Y.S., D.S., and A.B. performed and prepared carried out the ARPES experiment with the assistance of T.K.K., C.C., J.E.R., P.L.F., F.B., N.C.P., and M.S. M.H. analyzed the ARPES data. M.H., M. Horio, Q.W., K.P.K., Y.S., T.K.K., and I. V.G., R.Fi., and A.V. grew and prepared single crystals. L.D. and Y.X. performed ARPES experiments at the SIS, CASSIOPEEE, and I05 beamlines of the Swiss Light Source, SOLEIL synchrotron, and Diamond Light Source, respectively. We acknowledge Diamond Light Source for time at beamline I05 under proposal SI20259.

COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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