The surface layer of $\text{Sr}_2\text{RuO}_4$: A two-dimensional model system for magnetic-field-tuned quantum criticality

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Many of the exciting properties of strongly correlated materials are intricately linked to quantum critical points in their phase diagram. This includes phenomena such as high temperature superconductivity,$^{1,2}$ unconventional superconductivity in heavy fermion materials,$^{3}$ as well as exotic nematic states in $\text{Sr}_3\text{Ru}_2\text{O}_7$.$^4$ One of the experimentally most successful pathways to reaching a quantum critical point is tuning by magnetic field allowing studies under well-controlled conditions on ultra-clean samples. Yet, spectroscopic evidence of how the electronic states change across a field-tuned quantum phase transition, and what the importance of quantum fluctuations is, is not available so far. Here we show that the surface layer of $\text{Sr}_2\text{RuO}_4$ is an ideal two-dimensional model
system for a field-tuned quantum phase transition. We establish the existence of four van Hove singularities in close proximity to the Fermi energy, linked intricately to checkerboard charge order and nematicity of the electronic states. Through magnetic field, we can tune the energy of one of the van Hove singularities, with the Lifshitz transition extrapolated at ~32T. Our experiments open up the ability to directly study spectroscopically the role of quantum fluctuations at a field-tuned quantum phase transition in an effectively 2D strongly correlated electron material. Our results further have implications for what the leading instability in Sr$_2$RuO$_4$ is, and hence for understanding the enigmatic superconductivity in this material.

The occurrence of quantum criticality is often connected to van Hove singularities (vHs) in the electronic structure close to the Fermi energy ($E_F$). Tuning such a van Hove singularity to the Fermi energy leads to a Lifshitz transition that can explain many of the exotic properties of quantum materials$^{5-9}$. The Lifshitz transition is associated with a large change in the density of states (DOS) at the Fermi level together with a change in Fermi surface topology, leading to a quantum phase transition. For a magnetic-field tuned Lifshitz transition, this mechanism requires a van-Hove singularity within a few meV of $E_F$, so that a Zeeman splitting can drive one spin sub-band of the vHs to $E_F$ as illustrated in Figure 1a. Yet in many materials, it remains unclear whether the properties attributed to the quantum critical point are driven by quantum fluctuations, or simply a consequence of the Lifshitz transition in a single particle picture. Spectroscopic detection of the changes in the DOS in such a scenario would enable clarification of the role of quantum fluctuations in the Lifshitz transition, as well as provide the opportunity to test theories aiming at describing quantum criticality and the role of quantum fluctuations.
In this work, we show that the surface layer of Sr$_2$RuO$_4$ is an ideal model system to study a magnetic-field tuned Lifshitz transition. Whilst the bulk of Sr$_2$RuO$_4$ is a superconductor with a transition temperature $T_c \sim 1.5$ K, the clean surface exhibits a reconstruction suppressing superconductivity. We use ultra-low temperature scanning tunneling microscopy (STM) to demonstrate that the electronic structure of the surface layer enters a new emergent phase that cannot be explained simply by the structural change of the surface. We identify a checkerboard charge order concomitant with nematicity of the low energy electronic structure, giving rise to four vHs within 5 mV of the Fermi energy. In tunneling spectra in magnetic fields up to 13.4 T and at temperatures down to 50 mK, we observe a clear magnetic-field-induced splitting of the most prominent vHs, which can be tuned towards the Fermi level.

**Results**

The crystal structure of Sr$_2$RuO$_4$ consists of single layers of RuO$_2$ sandwiched between SrO layers, with a tetragonal unit cell. The bulk Fermi surface of Sr$_2$RuO$_4$, represented in Figure 1b, has been established by quantum oscillations and confirmed by angle-resolved photoemission spectroscopy (ARPES). It is mainly made up of the $4d$ $t_{2g}$ states of the Ru atoms, where the 2D $d_{xy}$ band (yellow line) has a vHs at the M-point of the Brillouin zone (BZ). The energy of the bulk vHs has been estimated at about 14 meV above $E_F$ and leads to a peak in the DOS (solid line in Figure 1c).

The surface layer of Sr$_2$RuO$_4$ exhibits a 6° rotation of the RuO$_6$ octahedra (Figure 1c, left inset). This rotation doubles the size of the unit cell and modifies the Fermi surface (Figure 1d) and band dispersion so that the vHs is shifted below $E_F$ (Figure 1c, dashed line).
Figure 1e shows a topographic image of Sr$_2$RuO$_4$, recorded at a temperature of 76mK, showing a SrO-terminated surface$^{18,19}$ with atomic resolution and demonstrating a low concentration of point defects (less than 0.1%). We find defects at the Ru site with two distinct orientations due to the octahedral rotation. The Fourier transform of the topography (upper inset in Figure 1e), shows the presence of quasiparticle interference (QPI), as well as Bragg peaks at (0,2π) due to the Sr lattice, and peaks at (π, π) from a modulation of the charge.
density (lower inset in Figure 1e) with the periodicity of the surface reconstruction. This modulation has been observed previously at the surface of Sr$_2$RuO$_4$,\textsuperscript{18} as well as at that of Sr$_3$Ru$_2$O$_7$.\textsuperscript{20} However, there is no obvious explanation for how the octahedral rotation leads to a charge modulation.

A typical differential conductance spectrum $g(V)$ in the range +/-80 mV is presented in Figure 1f. Here, kink- and gap-like features are observed at ±43 meV and ±5 meV respectively. The latter is associated with a reduction of the differential conductance by almost 30%. The spectra, as well as the absence of a superconducting gap, are in agreement with previous reports\textsuperscript{21–23}.

The additional modulation of the charge density leads to pronounced signatures in spectroscopic maps: while topographic images obtained at positive bias voltage show predominantly the Sr lattice (Figure 2a), differential conductance maps exhibit a clear checkerboard charge modulation superimposed to this lattice at bias voltages close to $E_F$ (Figure 2b, c). The checkerboard exhibits a contrast inversion across the Fermi energy. We plot in Figure 2d the energy dependence of the intensity of the checkerboard analyzed through a phase-referenced Fourier transformation (PR FFT, see S2C), which provides information about the amplitude and phase of the modulation. The amplitude of the checkerboard exhibits a pronounced maximum at -3.5mV with a width of only about 1mV and a weaker maximum at +3.5mV of similar width but with opposite phase. The phase change is consistent with what one would expect for a charge density wave.\textsuperscript{24}
Comparison of the checkerboard charge order centred at the Sr sites with the structure of the surface layer shows that the emergence of this charge order is equivalent to a reduction of C\(_4\) symmetry to C\(_2\) symmetry (Figure 3a) as a consequence of the octahedral rotation. The broken C\(_4\) symmetry means that the oxygen atoms become inequivalent in the [10] and [01] directions, as indicated in Figure 3a, resulting in nematicity of the low energy electronic states. This emergent nematicity is confirmed experimentally through unidirectional periodic modulations with atomic periodicity (Figure 3b, c) and anisotropy of the low-q quasi-particle interference (Figure 3d-i). The atomic scale symmetry breaking reveals that for changes in the
bias voltage of about 1mV the atomic-scale unidirectional periodicity changes direction between the high-symmetry directions [10] and [01]. This is also confirmed in the intensity of the atomic peaks (Fig. S4). The symmetry breaking of long-wavelength quasi-particle interference is seen in a real-space map around four defects (Figure 3d-i, defects marked by black crosses) and reveals a change from predominant quasiparticle scattering along [10] to scattering along [01] and back as a function of energy.

To understand the microscopic consequences nematicity and the observed charge modulation have on the electronic structure of this system, we have performed tight-binding simulations of the band structure of the surface of Sr$_2$RuO$_4$, accounting for the surface

Figure 3: Nematicity and the equivalence of C$_4$ symmetry breaking. a, Model of the surface atomic structure with the checkerboard charge order. The charge order on the Sr lattice combined with the octahedral rotations leads to a broken C$_4$ symmetry, which renders the oxygen atoms along the [10] and [01] directions (labelled O$_x$ and O$_y$) inequivalent. b, c, Nematicity in the atomic scale charge modulations (T=1.8 K, $V_{set}=7.8$ mV, $I_{set}=500$ pA, $V_L=370$ μV, scale bar: 1nm). d-i Real space images showing directional quasi-particle interference near defects (T=56 mK, $V_{set}=6.4$ mV, $I_{set}=225$ pA, $V_L=398$ μV, scale bar: 5nm).
reconstruction and with hopping parameters optimized against ARPES data\textsuperscript{15}. The surface reconstruction and the checkerboard charge order (Figure 2) are accounted for by including a weak intraband hybridization ($\Delta_{\text{hyb}}$). We account for the broken symmetry along the [10] and [01] lattice directions (Figure 3) through inclusion of a phenomenological nematic order parameter $\Delta_{\text{nem}} = \delta_{\text{nem}}(\cos(k_x) - \cos(k_y))$, which we apply only to the relevant $d_{xy}$ orbital. A similar order parameter has previously been introduced to describe nematicity in Sr\textsubscript{3}Ru\textsubscript{2}O\textsubscript{7}.\textsuperscript{25} The full Hamiltonian is then

$$H(k) = \begin{pmatrix} H_{\text{Ru}}(k) + \Delta_{\text{nem}} \hat{I}_{xy} & \Delta_{\text{hyb}} \hat{I} \\ \Delta_{\text{hyb}} \hat{I} & H_{\text{Ru}}(k + Q) + \Delta_{\text{nem}} \hat{I}_{xy} \end{pmatrix},$$

(1)

where $H_{\text{Ru}}(k)$ is the Hamiltonian for the ruthenium $4d$ bands in the unreconstructed Brillouin zone\textsuperscript{26}, and $Q = (\pi, \pi)$ accounts for the doubling of the unit cell (for details see S3). The band structure, calculated from Eq. 1, is presented for the most relevant part of the Brillouin zone around the M point in Figure 4a. We follow the notation by van Hove\textsuperscript{27} to label the resulting four van Hove singularities in the band structure: nematicity results in two saddle points, $S_1$ and $S_2$, on the high symmetry axis at the M point, whereas hybridization leads to a partial gap structure with a band maximum $\mathcal{M}$ and an additional saddle point $S_3$ around $E_F$. This can be seen from a 3D representation of the band structure around the M point (Figure 4b). The partial drop in the $g(V)$ spectrum around $E_F$, observed in Figure 1f, is naturally explained by the hybridization of the Ru $d_{xy}$-bands caused by the doubling of the unit cell. These four singularities lead to maxima in the density of states (see Figure 4c) in excellent agreement with the structure of the low energy $g(V)$ spectrum.
Having identified the presence and origin of the van Hove singularities at the surface of Sr₂RuO₄, we investigate the influence of an applied magnetic field, focusing on the behavior...
of the most prominent vHs, $\mathcal{M}$, at -3.4 mV. Figure 4d shows tunneling spectra recorded in magnetic fields from 0T to 13.4T, applied perpendicular to the surface. The spectra reveal a clear splitting of the vHs with increasing magnetic field. In Figure 4e, the field-dependence of the energy of the vHs reveals a linear behavior as expected for a Zeeman-like splitting. We find the slope to be +0.10 mV/T for the peak moving towards the Fermi energy and -0.07mV/T for the one moving away. The difference in slope can be attributed to an overall chemical potential shift of the $d_{xy}$ band of 17$\mu$V/T to ensure charge conservation. Taking this into account, we find a $g$-factor of $g^* \sim 3$ (with the splitting $\Delta E = g^* \mu_B B_z$). This value is consistent with the known Wilson ratio of bulk Sr$_2$RuO$_4$.\textsuperscript{28} From our experimental data, we can extrapolate that the vHs will cross the Fermi energy at a magnetic field of about 32T. At this field, the surface layer is expected to undergo a Lifshitz transition. As the energy of the van Hove singularity is associated with the checkerboard charge order, once it is split in a magnetic field, the charge order becomes spin-polarized. Our measurements demonstrate that the electronic structure of the reconstructed surface layer of Sr$_2$RuO$_4$ has all the ingredients for quantum critical behavior as described in the model in Figure 1a: (1) a vHs close to the Fermi energy, (2) the energy of the vHs can be tuned by magnetic field towards the Fermi energy, and (3) a Lifshitz transition of the electronic structure within reach of available magnetic fields.

**Discussion**

Our measurements show how the electronic structure of a two-dimensional model system evolves on approaching a magnetic-field induced Lifshitz transition, making it effectively a quantum simulator to study field-tuned quantum phase transitions. We can explain the surface electronic structure well by a phenomenological tight-binding model that incorporates the two orders we detect, nematicity and checkerboard charge order and reproduces all features we observe in our data. It naturally explains the pronounced gap-like
feature seen in tunneling spectra. What the model does not provide is an explanation for the origin of the checkerboard order and nematicity, and which of the two dominates, if either. Possible mechanisms include (1) nematicity driven by electron correlations, (2) charge order driven by nesting or a lattice distortion, (3) a cooperative effect of both, and (4) an antiferromagnetic order with formation of a spin-density wave, which stabilizes the checkerboard order and nematicity. For the first three scenarios, one would expect a significant structural distortion accompanying these, as is seen for charge density waves or nematicity in other systems. A structural distortion in the surface layer of Sr₂RuO₄ beyond the octahedral rotation has not been reported so far.²⁹ In a scenario where magnetic order or fluctuations drive nematicity and charge order, one would still expect a structural distortion, though much smaller.

A comparison with Sr₃Ru₂O₇ reveals intriguing parallels: Sr₃Ru₂O₇ exhibits a similar (albeit larger) octahedral rotation, and the energy of the vHs as detected by ARPES occurs at an energy below $E_F$³⁰, close to the energy at which we find the dominant vHs in the surface layer of Sr₂RuO₄. Due to stronger correlations and a $g$-factor of $g^* ≈ 14.6$ in Sr₃Ru₂O₇,³¹ the vHs can be tuned to the Fermi energy at 8T, whereas in the surface layer of Sr₂RuO₄, we find a $g$-factor that is almost 4 times smaller. Consequently, the vHs is expected to reach the Fermi energy only at 32T. It remains to be seen whether the parallels go any further when the surface layer in Sr₂RuO₄ undergoes the Lifshitz transition. There are also important differences though, including that the system we report here is strictly two-dimensional which puts the criticality of the Lifshitz transition into a different universality class than what is expected for the bulk of Sr₃Ru₂O₇.
Consistent with previous studies of the reconstructed surface,\textsuperscript{21,22} we do not observe any evidence for superconductivity, indicating that the emergent electronic order suppresses superconductivity in the surface layer. This may imply that the density of states of the $d_{xy}$ band around $E_F$, which becomes gapped out at the surface, and the fluctuations associated with the reconstruction play a crucial factor in superconducting pairing. This scenario naturally results in a competition of nematicity and the charge density modulations with superconductivity, reminiscent to what is found in other strongly correlated electron systems\textsuperscript{3,32,33}. Understanding the leading superconducting instability in this highly debated material therefore undoubtedly will have to account for this susceptibility towards density wave formation as observed in other unconventional superconductors.

Conclusion

Using the surface layer of Sr$_2$RuO$_4$ as a two-dimensional model system to study magnetic-field induced critical points and Lifshitz transitions provides a new well-controlled test bed to compare microscopic theories to experiment. Because the emergent surface phase is strictly two-dimensional, limited to the surface layer, all relevant information about the electronic states across the quantum phase transition are accessible spectroscopically. Magnetic-field tuned Lifshitz transitions have been proposed to be at the heart of the quantum critical behavior in a range of heavy fermion and strongly correlated electron materials, yet the ability to spectroscopically trace the electronic structure across a field-tuned quantum phase transition has remained elusive. Given the sensitivity of the energy of the van-Hove singularity in Sr$_2$RuO$_4$ to uniaxial strain,\textsuperscript{34} we expect that the magnetic field at which the surface undergoes the quantum phase transition can be reduced substantially by combining uniaxial strain with magnetic field. This creates the opportunity to spectroscopically verify the role of quantum fluctuations near a field-tuned quantum critical point, and provides important insights into the
leading instabilities of Sr$_2$RuO$_4$ with important implications for its enigmatic superconducting state.

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Supplementary Material for

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Figs. S1 to S7

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S1. MATERIALS AND METHODS

A. Single crystal growth.

The Sr$_2$RuO$_4$ crystals used in this work were grown by the floating-zone technique with Ru self-flux, using a commercial image furnace with double-elliptical mirrors and two 2.0kW halogen lamps (SI). Morphological and elemental characterization of the crystals was carried out using a Zeiss Leo EVO 50 scanning electron microscope (SEM) equipped with an Oxford INCA Energy 300 energy dispersive X-ray spectroscopy (EDS) system. The structure and crystalline qualities of the samples were assessed by a high-resolution X-ray diffractometer (Panalytical, X Pert MRD), with a Cu K-α source.

B. Characterization

1. *Transmission electron microscopy (TEM)*

To verify the crystal quality in the surface region following cleavage of the sample in the STM, we have performed Transmission Electron Microscopy on a sample on which STM measurements had been carried out. The sample for STEM analysis was prepared by conventional gallium focused ion beam (FIB) milling using an FEI Scios focused ion beam scanning electron microscope (FIBSEM) equipped with an EDAX Hikari Super electron back-scattered diffraction (EBSD) detector. The orientation of the sample was determined by EBSD prior to milling, to cut a lamella in the [010] plane. High angle annular dark field (HAADF) images were recorded using a probe corrected FEI Themis 200 scanning/transmission electron microscope operated at 200kV.

Figure S1(a) shows a TEM image of a cross section of the sample after cleaving. It shows a uniform phase along the c-axis on the scale of ≈ 80nm extending up to the surface. Figure S1B shows a zoom in with atomic resolution. It shows the expected stacking for Sr$_2$RuO$_4$, as evidenced by the inset, where both Strontium and Ruthenium atoms are visible, with the Ru atoms appearing with a slightly higher intensity. Oxygen atoms are not visible. The TEM image shows no evidence of inclusions of other members of the Ruddlesden-Popper series (i.e. Sr$_{n+1}$Ru$_n$O$_{3n+1}$ with $n > 1$).
FIG. S1: TEM images along the b-axis. (A) Image of the cross-section of a sample after STM measurements have been performed on its surface, demonstrating uniformity up to the surface. (B) High-resolution image showing atomic resolution. The inset shows a zoom in of the image, with the stacking expected for Sr$_2$RuO$_4$. Superimposed to the TEM image is the atomic structure, with purple spheres indicating strontium atoms, black ones ruthenium atoms and red ones oxygen atoms. The black dashed line delimits the unit cell of Sr$_2$RuO$_4$.

2. Resistance measurements

Electric transport measurements were performed using a four probe technique with a $^3$He refrigerator. The resistance as a function of temperature shows a superconducting transition at $T_C = 1.5$ K, in agreement with data reported in literature for good quality crystals($S2$).

We determine a residual-resistance ratio (RRR) $\lim_{T \to 0K} \frac{R(300K)}{R(T)} \approx 666$, extrapolated to 0K from fitting the resistance above $T_C$ to $R(T) = R_{\text{res}} + AT^2$ (Figure S2A). This value is comparable to that of high-purity crystals reported in the literature($S3$).

3. Scanning tunneling microscopy (STM)

Experiments were performed in a home-built ultra-low temperature STM operating in a dilution refrigerator($S4$) Samples were prepared by in-situ cleaving at low temperatures ($\sim 20$K) in cryogenic vacuum. We used STM tips cut from PtIr wire, and prepared them in-
FIG. S2: **Resistance as a function of temperature of Sr2RuO4 crystal measured applying a bias current along the a-b plane.** (A) Resistance up until room temperature. The inset shows the resistance as function of temperature in log-log scale (B) Low temperature resistance down to 0.5 K showing a superconducting transition at 1.5 K.

**situ** by field emission on a Au(111) single crystal. Bias voltages were applied to the sample, with the tip at virtual ground.

Spectroscopic measurements were performed using a lock-in amplifier to measure differential conductance $g(V)$. The bias voltage $V$ was modulated at a frequency of $\nu = 397$Hz, the RMS amplitude $V_L$ is given in figure captions where applicable. The bias and current setpoints, $V_{\text{set}}$ and $I_{\text{set}}$ respectively, are indicated for both topographies and spectroscopic maps.

After cleaving and inserting the samples into the STM head, we have always observed the reconstructed SrO termination as observed in previous reports(S5–S8) and as can be inferred from the appearance of the defects.

**S2. DIFFERENTIAL CONDUCTANCE MAPS**

In this section, we show the real space conductance maps underpinning the data shown in fig. 2D and how it has been processed.
A. Real space maps

The topographic image acquired simultaneously with the map taken at 2K is shown in Figure S3A and the corresponding real space differential conductance map, \( g(r, V) = dI/dV(r, V) \), for \( V = -3.4 \text{mV} \) in Figure S3B. The checkerboard charge order is clearly visible. In addition quasiparticle interference (QPI) effects are observed around the defect seen in the topography.

B. Processing of differential conductance maps

In Figure S3 we show images following each step of the data processing. The raw Fourier transformation of the \( g(r, V) \) map is displayed in Fig. S3C. It shows clear signatures of the atomic peaks at \((\pm 2\pi, 0)\) and \((0, \pm 2\pi)\) (in units of \(1/a\), where \(a\) is the lattice constant of the bulk tetragonal unit cell), as well as of the checkerboard charge order at \((\pm\pi, \pm\pi)\) and \((\pm\pi, \pm\pi)\). There are in addition weak higher order peaks. The Fourier transformation is first corrected for any linear drift by mapping the atomic peaks onto a perfect square (Fig. S3D). Next, the resulting images are mirror symmetrized along the horizontal or vertical direction (Fig. S3E). Finally, to suppress the high intensity at the centre of the image due to the distribution of defects, a Gaussian function with a width of 4 pixels is subtracted (Fig. S3F).

This procedure was implemented for all layers of the map. Fig. S4 shows the Fourier transformation after processing of the same \( g(r, V) \) map at different energies, between \(-8.2 \text{mV}\) and \(+7.8 \text{mV}\). The intensity of the \((\pm\pi, \pm\pi)\) peaks is strongly energy dependent, showing maximum intensity at \(-3.4 \text{mV}\) (highlighted by a red box). Additionally, we observed an anisotropy in the intensities of the \((2\pi, 0)\) and \((0, 2\pi)\) peaks, as is indicated in Fig. S4 by the solid red and dashed white circles. It can be seen that the high intensity switches from one direction to the other with energy. This reflects the atomic scale symmetry breaking shown in Fig. 3B and C in the main text.

C. Phase-referenced Fourier transformation

In fig. 2D of the main text, we show a phase-referenced Fourier transformation to deduce the characteristic energy scale of the checkerboard charge order and the relative phase at positive and negative bias voltages. The Fourier transformation \( \tilde{g}(q, V) \) of a differential
FIG. S3: **Processing of differential conductance maps.** (A) Topography acquired simultaneously with map, showing one defect in the top left corner. Scale bar: 5nm ($V_{set} = 7.8$mV, $I_{set} = 225$pA). (B) Real space $g(r,V)$ at $V = -3.4$ mV. (C) Absolute value of its Fourier transformation, $|\tilde{g}(q,V)|$. (D) after drift correction, (E) after symmetrizing along the horizontal (or vertical) direction, and (F) after subtraction of a gaussian function with a width of 4 pixels at the centre of the image ($V_L = 800\mu$V, $T = 2$K, $B_z = 0$T).

Conductance map $\tilde{g}(r,V)$ which shows the atomic lattice exhibits two pairs of Bragg peaks at $q_{lat} = (\pm 2\pi, 0)$ and $(0, \pm 2\pi)$ (in units of $1/a$, where $a$ is the lattice constant), as shown in Figure S5A by the black circle. The checkerboard order shown in Figure 2B, C of the main text has a wavelength that is $\sqrt{2}$ times larger, with the unit vector $45^\circ$ rotated. The periodicity due to the checkerboard charge order shows up as four peaks at $q_{CKB} = (\pm \pi, \pm \pi)$, one of which is indicated by the red circle in Fig. S5A. The amplitude of this checkerboard order is reflected in the intensity of Fourier peak, $|\tilde{g}(q_{CKB},V)|$, as shown in Fig. S4. We can determine the characteristic energy scale of the checkerboard modulation as a function of energy by plotting, $|\tilde{g}(q_{CKB},V)|$ as a function of applied bias $V$, however losing the phase information $\phi$ contained in the Fourier transformation contained in the
FIG. S4: Energy layers from the $\tilde{g}(q, V)$ map shown in Fig. 2 of the main text and Fig. S3. The intensity of the peaks at $q_{\text{CKB}} = (\pm \pi, \pm \pi)$ due to the checkerboard charge order is strongly energy dependent. The layer corresponding to the energy of the vHs is highlighted by a red box, where the peaks at $q_{\text{CKB}}$ exhibit the highest intensity. Additionally, the intensity of the Bragg peaks $q_{\text{at}} = (0, \pm 2\pi)$ and $(\pm 2\pi, 0)$ switches with energy due to the nematicity (cf. fig. 3B, C of the main text). For each layer, the higher intensity peak is highlighted by a red circle, and the lower intensity peak with a dashed white circle. The color bar for all images is scaled to the same range.
complex values of
\[ \tilde{g}(\mathbf{q}, V) = | \tilde{g}(\mathbf{q}, V) | e^{i\phi(\mathbf{q}, V)}. \]  
(S1)

While analyzing the phase $\phi$ is possible, it suffers from an overall global phase factor which is arbitrary. To remove this global phase factor, we use a phase-referenced Fourier transformation (PR-FFT) \((S9)\)

\[ \tilde{g}^R(\mathbf{q}, V) = \frac{\tilde{g}(\mathbf{q}, V)}{e^{i\phi(\mathbf{q}, V_0)}} = | \tilde{g}(\mathbf{q}, V) | e^{i(\phi(\mathbf{q}, V) - \phi(\mathbf{q}, V_0))}. \]  
(S2)

In this PR-FT, the phase at each $\mathbf{q}$-vector is referenced to the phase at a specific energy $V_0$, removing the global phase factor. This allows tracking of the change in phase as a function of energy relative to that layer by simply plotting the real part of the PR-FT

\[ \text{Re} \left[ \tilde{g}^R(\mathbf{q}, V) \right] = | \tilde{g}(\mathbf{q}, V) | \cos (\phi(\mathbf{q}, V) - \phi(\mathbf{q}, V_0)). \]  
(S3)

Here, we reference the phase to the map layer at $V_0 = -3.4 \text{mV}$. The real part of the PR-FFT images, Figure S5C, $\text{Re} \left[ \tilde{g}^R((\pi, \pi), V) \right]$ allows us to determine if there is a phase shift between the checkerboard order at different energies as well as the relative amplitude. A phase reversal means a change in sign. At the reference energy, $V_0$, $\text{Re} \left[ \tilde{g}^R((\pi, \pi), V) \right]$ will be positive by definition. Fig. S5C shows $\text{Re} \left[ \tilde{g}^R((\pi, \pi), V = +3 \text{mV}) \right]$, where the peaks at $\mathbf{q}_{\text{CKB}}$ appear with a negative sign, evidencing a phase shift with respect to the charge modulation at $V_0 = -3.4 \text{mV}$. Figure 2D of the main text shows that the checkerboard order appears predominantly at $V = -3.5 \text{mV}$ and $V = 3.5 \text{mV}$ with opposite phase. For better comparison between the maps at the two different temperatures, $\tilde{g}^R(\mathbf{q}_{\text{CKB}}, V)$ was normalized by its value, $\tilde{g}^R(\mathbf{q}_{\text{CKB}}, V_0)$, at the bias setpoint, $V_0$. This was $V_{\text{set}} = 7.8 \text{ mV}$ for the map measured at 2 K and $V_{\text{set}} = 7.0 \text{ mV}$ for the one at 59 mK.

S3. TIGHT-BINDING MODEL

The tight binding model for the surface of $\text{Sr}_2\text{RuO}_4$ may be defined, in an analogous manner to that of the bulk, via hopping associated with the three $t_{2g}$ Ru orbitals \((S10)\),

\[ H^r(\mathbf{k}) = \begin{pmatrix} E_{zz}(\mathbf{k}) & \gamma(\mathbf{k}) - \sigma i\eta & i\eta \\ \gamma(\mathbf{k}) + \sigma i\eta & E_{yz}(\mathbf{k}) & -\sigma \eta \\ -i\eta & -\sigma \eta & E_{xy}(\mathbf{k}) \end{pmatrix}. \]  
(S4)
FIG. S5: Phase-referenced Fourier transformation. (A) Amplitude $|\tilde{g}(q, V_0 = -3.4\text{mV})|$ for the Fourier transformation. (B) Real part $\text{Re}[\tilde{g}(q, V = +3.0\text{mV})]$; it can be seen that the peaks at $(\pi, \pi)$ and $(-\pi, \pi)$ have opposite sign. (C) Real part of the PR-FFT, $\text{Re}[\tilde{g}^R(q, V = +3.0\text{mV})]$, showing the reversed sign of the peaks at $(\pi, \pi)$ and $(-\pi, \pi)$ compared to panel A (map parameters as in fig. S3).

This Hamiltonian includes nearest neighbour hoppings between the $d_{xz}$ and $d_{yz}$ orbitals as well as up to third nearest neighbour hoppings between $d_{xy}$ orbitals,

$$E_{xz}(k) = -2t_1 \cos(k_x) - 2t_2 \cos(k_y) - \mu,$$
$$E_{yz}(k) = -2t_2 \cos(k_x) - 2t_1 \cos(k_y) - \mu,$$
$$E_{xy}(k) = -2t_3(\cos(k_x) + \cos(k_y)) - 4t_4 \cos(k_x) \cos(k_y) - 2t_5(\cos(2k_x) + \cos(2k_y)) - \mu_c$$

(S5)

The off diagonal term, $\gamma(k)$, describes inter-orbital hopping between the degenerate $d_{xz}$ and $d_{yz}$ states and is written as $\gamma(k) = -4t_{\text{inter}} \sin(k_x) \sin(k_y)$. The rest of the off-diagonal terms arise from the spin-orbit interaction, where $\eta$ is the spin-orbit coupling constant and $\sigma$ is defined as $+1$ for up spins ($\uparrow$) and $-1$ for down spins ($\downarrow$).

Using this basis, a tight binding model for the Ru electronic structure can be defined via

the Hamiltonian
$$H_{\text{Ru}}(k) = \begin{pmatrix} H^+(k) & 0 \\ 0 & H^+(k) \end{pmatrix}, \quad \text{(S6)}$$

representing now a $6 \times 6$ matrix. For the surface, we must account for the doubling of the unit cell due to the additional octahedra rotation. This surface tight binding model is then given by the $12 \times 12$ matrix

$$H_{\text{surf}}(k) = \begin{pmatrix} H_{\text{Ru}}(k) & 0 \\ 0 & H_{\text{Ru}}(k+Q) \end{pmatrix}, \quad \text{(S7)}$$

with $Q = (\pi, \pi)$.

For the bulk tight binding model, presented in Fig. 1B of the main text, we calculate the Eigenvalues using Eq. S6 with the hopping parameters from Ref. S10.

$$\begin{align*}
t_1 &= 0.15\text{eV} \\
t_2 &= 0.1t_1 \\
t_3 &= 0.8t_1 \\
t_4 &= 0.3t_1 \\
t_5 &= 0.1t_1 \\
t_{\text{inter}} &= 0.01t_1 \\
\mu &= 1.0t_1 \\
\mu_c &= 1.1t_1 \\
\eta &= 0.1t_1.
\end{align*}$$

Here $t_1$ is set to $150\text{meV}$ and all other parameters are defined relative to $t_1$.

For the surface tight binding description, which has been used in Fig. 4 of the main text, we use $t_5 = 0.095t_1$, $\mu = 0.75t_1$ and $\mu_c = 0.812t_1$. This places the vHs just above the Fermi level and slightly alters the Fermi wave vector $k_F$ of the $d_{xz}$ and $d_{yz}$ bands to best fit the ARPES data (S11). We introduce a hybridisation between the two Ru site, $\Delta_{\text{hyb}} = 3\text{meV}$, as off-diagonal elements in Eq. S7,

$$H_{\text{surf}}(k) = \begin{pmatrix} H_{\text{Ru}}(k) & \Delta_{\text{hyb}} \hat{I} \\ \Delta_{\text{hyb}}^* \hat{I} & H_{\text{Ru}}(k+Q) \end{pmatrix}, \quad \text{(S8)}$$

and include a phenomenological $C_4$ symmetry breaking term $\Delta_{\text{nem}}(k) = \delta_{\text{nem}}(\cos(k_x) - \cos(k_y))$ specifically to the $d_{xy}$ orbital in $H_{\text{Ru}}$, with $\delta_{\text{nem}} = 2 \text{ meV}$, to produce the full Hamiltonian defined in Eq. 1 of the main text. A similar nematic term has been discussed for Sr$_2$Ru$_2$O$_7$ previously. (S12)

$$H_{\text{surf}}(k) = \begin{pmatrix} H_{\text{Ru}}(k) + \Delta_{\text{nem}}(k) \hat{I}_{xy} & \Delta_{\text{hyb}} \hat{I} \\ \Delta_{\text{hyb}}^* \hat{I} & H_{\text{Ru}}(k+Q) + \Delta_{\text{nem}}(k+Q) \hat{I}_{xy} \end{pmatrix}, \quad \text{(S9)}$$
The density of states presented in Fig. 4B has been calculated via

$$N_0(\omega) = -\frac{1}{\pi} \text{Tr} \left[ \text{Im} \left[ \sum_k G(k, \omega) \right] \right].$$  \hspace{1cm} (S10)

Here, $G(k, \omega)$ is the Green's function defined as $G(k, \omega) = \frac{1}{\omega - H(k) + i\Gamma}$. where $\omega$ is the energy and $\Gamma$ a broadening parameter. We use a $k$-grid of 4096x4096 lattice points and $\Gamma = 0.1 \text{meV}$.

Fig. S6 shows the Fermi surface, band structure and DOS given by the tight-binding model described above for comparison. Fig. S6A-C show the case where only the doubling of the unit cell is taken into account (Eq. S7). The vHs was put above, but very close to, $E_F$ and it appears as a sharp peak with logarithmic divergence, cut off by the broadening $\Gamma$. Fig. S6D-F show the case were only the $C_4$-symmetry breaking term is included. The $d_{xy}$ band becomes $C_2$-symmetric, and the vHs splits into two peaks, one above $E_F$ and another below, although no gap opens around the Fermi energy. Fig. S6G-I show the case of eq. S9, where both the nematic term and the hybridization potential are included. A gap is opened between the $d_{xy}$ bands, creating four vHs. The calculated DOS reproduces the measured differential conductance spectrum, as shown in Fig. 4 of the main text.

S4. ANALYSIS OF MAGNETIC-FIELD DEPENDENT TUNNELING SPECTRA

Each $g(r, V)$ spectrum shown in Figure 4C of the main text is obtained from an average of 10 spectra. All spectra were acquired with the same setpoint conditions before turning off the feedback loop. To determine the energy of the van Hove singularity from the peak positions in the spectra, we first subtract a background from the spectra, and then fit the positions of the dominant peaks. To describe the background, we fit an arc tangent and a constant ($f(V) = a \cdot \arctan((V - V_0)/\Gamma) + c$) to the background in the data at 13.4T to describe the gap edge at negative energies and subtract the resulting function $f(V)$ as a base line. For the data at other fields, we fit the values of $a$ and $c$ to the background using the same arctangent function, but keep $V_0$ and $\Gamma$ fixed at the values of the fit at 13.4T, Figure S7A. To determine the energy of the magnetic field split van Hove singularity, we fit a Lorentzian, Figure S7B.
FIG. S6: Tight-binding model of the surface of Sr$_2$RuO$_4$. (A-C) Fermi surface, band dispersion along Γ-M-X and density of states between ±8 meV for a model without nematicity ($\Delta_{nem} = 0$ meV) and hybridization of bands due to the reconstruction $\Delta_{hyb} = 0$ meV. (D-F) Equivalent plots with nematicity included, $\Delta_{nem} = 2$ meV, but no hybridization between the bands due to the reconstruction $\Delta_{hyb} = 0$ meV. (G-I) Equivalent plots with nematicity $\Delta_{nem} = 2$ meV (as before) and a non-zero hybridization $\Delta_{hyb} = 3$ meV.

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FIG. S7: Determination of splitting of vHs in magnetic field. (A) $g(r,V)$ spectra in magnetic fields of $B = 0\ldots13.4\text{T}$, with the background fit shown as red lines. (B) $g(r,V)$ spectra after subtraction of the background. The red lines show the lorentzian fits to extract the peaks positions.

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