Discovery of nanoscale phase coexistence of heavy Fermi-liquid and metallic spin-liquid in geometrically frustrated Pr$_2$Ir$_2$O$_7$

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The interplay of spin-orbit interaction and geometric frustration of local Ir spins drives the long-range magnetic order in pyrochlore iridate family, R$_2$Ir$_2$O$_7$ (R = lanthanide), into a paramagnetic state at near-zero temperatures$^1$. Pr$_2$Ir$_2$O$_7$ lies at a tuning-free quantum critical point and exhibits a diverse array of complex phenomena including an underscreened Kondo effect, biquadratic band structure, metallic spin-liquid (MSL), and anomalous Hall effect$^{2-5}$. Using spectroscopic imaging with the scanning tunneling microscope, complemented with machine learning K-means clustering analysis, density functional theory, and theoretical modeling, we probe the local electronic states in single crystal of Pr$_2$Ir$_2$O$_7$ and discover an unprecedented nanoscale electronic phase separation. Regions of heavy Fermi-liquid (HFL) with a well-defined Kondo resonance are interwoven with a non-magnetic metallic phase with Kondo-destruction that is suggestive of the candidate MSL. Remarkably, the spatial nanoscale patterns display a correlation-driven fractal geometry with power-law behavior extended over two and a half decades, consistent with being in proximity to a critical point. Our discovery reveals a new nanoscale tuning route to realize a MSL, viz. using a spatial variation of the electronic potential as a means of adjusting the balance between Kondo entanglement and geometric frustration.
A Kondo lattice, a matrix of localized spins embedded in a sea of itinerant electrons, is traditionally understood as giving rise to maximally-entangled singlets between the two types of spins, leading to a HFL. When it is exposed to strong geometric frustration, the enhanced quantum fluctuations among the local moments are expected to break down the Kondo effect and may lead to the emergence of a novel and rare phase of quantum matter - a MSL. Analogous to the elusive quantum spin-liquid anticipated in frustrated magnetic insulators, this exotic phase of quantum matter may host unusual fractionalized excitations and bare the essence of unconventional superconductivity.

Over the past decade, theoretical efforts focused on the description of a unified global magnetic phase diagram that merges geometric frustration (G) and Kondo hybridization (Jₖ) (see Fig.1a). In the limit of small G and Jₖ, local moments form a long-range magnetic order governed by the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction. However, the magnetic order becomes unstable as either G or Jₖ is enhanced. The regime of large Jₖ has been sufficiently investigated in the past, both theoretically and experimentally. Simply put, the Kondo entanglement delocalizes the f-electrons and the resulting HFL has a large Fermi surface that incorporates both the itinerant- and f-electrons.

The regime of strong geometric frustration (large G) is not yet well established. In this regime, long-range antiferromagnetic order is no longer sustained due to enhanced quantum fluctuations, giving way to a MSL ground state that preserves the long-range entanglement of spin singlets. In this phase, the spins are localized and the Fermi surface is determined by the itinerant conduction electrons only, characterized by the small Fermi surface. As the HFL and MSL involve the same local magnetic moments and therefore locally compete, the mutual territory of strong geometric frustration and Kondo hybridization (large G and large Jₖ) may give rise to unconventional forms of quantum criticality. Experimentally, this territory is largely unexplored.

From a materials perspective, the pyrochlore iridate Pr₂Ir₂O₇ is one of the very few existing systems that may enable us to investigate this regime. The 4f-electron moments of the Pr are located on the edges of the corner-sharing tetrahedral of the pyrochlore structure, forming a Kagome lattice of Ising spins pointing along the [111] direction. Susceptibility measurements indicate a negative Curie-Weiss temperature Θₑw ≈ -20 K due to enhanced magnetic fluctuations induced by this geometric frustration. Pr₂Ir₂O₇ is the only member of the pyrochlore iridate family that remains magnetically disordered and metallic down to the lowest measured temperature and is argued to be located in proximity to a quantum critical point. The 5d electrons of the Ir tetrahedral states, which are in the Jₐeff = ½ state, are semi-metallic and form a quadratic band-touching at the zone center in Pr₂Ir₂O₇ as confirmed by angle resolved photoemission spectroscopy (ARPES). This is in contrast to most other members of the pyrochlore family, where they lead to an insulating AFM. These metallic states, therefore, mediate Kondo hybridization of the Pr 4f-moments. The non-Kramers doublet ground state of the J = 4 Pr orbitals indicates a possible two-channel Kondo effect with the excited Kramers doublet. Indeed, resistivity measurements show a minimum and a weak upturn below 60 K providing evidence of Kondo behavior. On the other hand, the magnetic susceptibility diverges starting below about 40 K suggesting that 4f moments, at least partially, are not Kondo-quenched, rather fluctuating down to the lowest temperature as a result of the magnetic frustration. Spin freezing is observed only below Tₘ = 300 mK, two orders of magnitude.
magnitude smaller than both \( T_K \) and \( \Theta_W \). This makes Pr\(_2\)Ir\(_2\)O\(_7\) a distinct example of a magnetically frustrated Kondo-lattice that may display MSL behavior.

Here we use spectroscopic imaging with the scanning tunneling microscope to spatially visualize and probe the local electronic states in single crystal of Pr\(_2\)Ir\(_2\)O\(_7\). To our knowledge, this is the only STM experiment on any pyrochlore iridate system to date. Figure 1b shows a schematic of the crystal structure revealing the Kagome lattice of Pr- and Ir-rich layers alternating along the [111]-direction\(^\text{27}\). The lack of a natural cleaving plane in this cubic material system, as examined in our density functional theory (DFT) calculations, makes STM experiments particularly challenging. Over 20 different attempts of single crystal cleaves have been carried out to achieve the desired atomically resolved surface topographs. Fig.1c shows a topographic STM image (65 nm) of the cleaved (111) surface revealing the atomically resolved Kagome lattice with an in-plane spacing of 7.6 Å corresponding to the bulk crystal structure of Pr\(_2\)Ir\(_2\)O\(_7\). We also observe topographs with atomically resolved terraces separated by ~ 6 Å, corresponding to the unit-cell along the [111]-direction, within which small islands (2.3 Å high) are seen (Fig.1d). The Fourier transform of both topographs show 3-fold symmetric Bragg peaks corresponding to the Kagome lattice structure (right inset of Fig.1c). The chemical nature of the surface termination in the topographic images is identified by carrying out DFT calculations of the surface electronic density (see Supplementary Note 1).

We find the cleaving plane to occur between the Pr-rich and Ir-rich layers with the oxygen atoms intact in each layer. DFT simulation of the STM surface termination further reveals an asymmetry in the surface electronic density between the Pr-Ir and Ir-Pr step heights that enables us to unambiguously identify terraces/islands in Fig.1d as Pr-rich/Ir-rich chemical composition. Regardless of the surface topology, however, the topographs show nanoscale inhomogeneity with picometer (pm) height variation (Fig.1c-g) corresponding to the variation in the integral local density of states (LDOS) in this stoichiometric material system. Spatial inhomogeneity is inherent to the complexity of correlated electron systems, such as manganites and cuprates, where various near-degenerate electronic phases can be continuously tuned at the nanoscale as a result of minute perturbations\(^\text{10}\).

The origin and nature of the spatial inhomogeneity is best visualized through spectroscopic imaging with the STM. Figure 2a-c show conductance (\(dI/dV\)) measurements on a 40 nm area at an energy of -10 meV below \( E_c \) at different temperatures (and different fields of view). Figure 2b, c corresponds to the same field of view of the topographs in Fig.1c, d, respectively. At all temperatures, a clear electronic inhomogeneity with nanometer length-scale, corresponding to relatively high and low density of states (at -10 meV) is observed. Quite remarkably, this nanoscale inhomogeneity is decoupled from the surface structural features (islands and step edges in Fig.1d) observed in the topograph suggesting the inhomogeneous puddles are extended to the bulk. For example, overlaying the unit-cell step and the Ir-rich islands seen in Fig.1d onto the conductance map of Fig.2c (gray lines) clearly reveals the absence of a link between the two (cross-correlation of -0.15), indicating the inhomogeneity not to be originating from the surface topographic structures and likely being electronic in nature. Moreover, the resemblance of the observed patterns in the conductance maps (Fig.2a-c) obtained from areas with very different topographic features (Fig.1c, d), further suggests that the electronic inhomogeneity is intrinsic to the bulk of the material.
To understand the origin of this electronic inhomogeneity, in Fig.2d-f we plot the dI/dV spectra, which corresponds to the LDOS, crossing two domains (black arrows in Fig.2a-c). At low temperature, the spectra reveal a sharp resonance right below the Fermi energy, originating from the orange region, that smoothly weakens and disappears before transitioning into a partial gap-like structure in the green region. Clearly, the overall spectral shape is very different in these two domains, indicating that the inhomogeneity corresponds to nanoscale phase separation of two different electronic states. As temperature is increased to 33 K, the spectra are broadened, yet maintain the same spatial behavior. By 45 K, thermal broadening takes over and only a hint of the resonance remains.

To obtain an unbiased and global description of the spatial phase separation, we use machine learning with K-means clustering algorithm that is based on the Euclidean separation of the data-space to spatially identify the different spectral clusters. This method effectively sorts spectra whose features are most distinct (see Supplementary Note 2). Figure 3a-c shows the results of the K-means, revealing three clusters in all the data. The overall K-means images are analogous to the STM conductance maps at a bias of -10 meV (see Supplementary Note 3). This is not true for all energies, as the STM set-point effect dramatically alters the maps at energies between the set-point bias and the Fermi energy. The K-means present the spatial phase separation originating from the entire spectral features (rather than a single energy) and independent of the experimental set-point effect (see Supplementary Note 2). Clearly, the spectra originating from the bright and dark domains represent two completely different electronic states with one showing a sharp resonance and overall higher density of states near E_F as compared to the dark regions that show a partial gap. The average of the spectra originating from the different clusters are plotted in Fig.3d-f and compared to individual STM spectra from corresponding regions (Fig.3g-i). While the K-means analysis does result in three clusters, two of these clusters (green and black) are qualitatively similar in their spectral lineshape, yet quite distinct from that of the orange cluster.

The sharp asymmetric resonance revealed in Figs.2, 3 at low temperatures is reminiscent of the Fano lineshape that originates from Kondo hybridization and is the hallmark of heavy fermion Kondo-lattice material systems. The nanoscale inhomogeneity of the Kondo hybridization in a stoichiometric material system, however, has not been observed in any of these Kondo lattice systems and may thus be a result of its competition with magnetic frustration on the Kagome lattice and its proximity to quantum criticality. In Pr_Ir_2O_7, Kondo physics is expected to originate from the Pr 4f electronic states through their hybridization with the Ir 5d itinerant electrons. To extract the Kondo temperature, we fit the data originating from the orange clusters to the Fano lineshape (see Fig.2 and Supplementary Note 4). The temperature-dependent resonance linewidths extracted from all the data yields a Kondo temperature with a Gaussian distribution centered at ~60 K, in good agreement with the resistivity minimum, providing spectroscopic evidence of inhomogeneous and frustrated Kondo-lattice in Pr_Ir_2O_7. The break-down of the Kondo-singlet formation of the f-moments in the metallic state (green-black phase) together with the absence of static magnetic order in Pr_Ir_2O_7 (as seen in neutron scattering and magnetization measurements) therefore indicate that the 4f local moments in the non-Kondo phase (green-black phase) exhibit self-screening through long-range entanglement among themselves and represent the anticipated MSL state that is in competition with the HFL. Note that, low temperature susceptibility measurements exclude spin glass formation scenario and indicate majority of spins to remain liquid.
Spontaneous emergence of multiphase electronic states, near critical points, forming nanoscale domains in an otherwise stoichiometric quantum material system is the hallmark of complexity in correlated electron systems where tiny perturbations of spin, charge, lattice, or orbital degrees of freedom can have a large response on the electronic ground state. Near continuous phase transitions, the spatial organization of the electronic states often follows scale-invariant fractal domain structure, where various physical quantities display power-law scaling over multiple decades. Following very recent work on the AFM NdNiO$_3$, as well as earlier work on cuprates, we study the pattern formation through the spatial organization of the electronic domains. To obtain a meaningful analysis of the geometric clusters and their critical exponents, we first carry out a high resolution spectroscopic imaging on the same field of view of Fig.1c to an area of 65 nm with a single pixel resolution of 0.25 nm.

Statistical analysis of domain geometry using the STM conductance maps at some energy, however, suffers from the experimental set-point effect which renders the integral of the conductance between the set-point bias and the Fermi energy to be spatially uniform. To overcome this technical issue, we use the K-means map which clusters the spectra based on their mathematical resemblance and independent of the set-point effect. To binarize the K-means map, the machine learning algorithm is run by a default two clusters (Fig.4a, which corresponds to the conductance map in Supplementary Fig.S13b). We use the box-counting method as well as the geometric domain distribution analysis (see Supplementary Note 7) to analyze the spatial patterns. Figure 4b displays the distribution of number of boxes (N) versus (box size)$^{-1}$ in a log-log plot, which is a measure of the fractal dimension. Here, N corresponds to the number of boxes that cover the domains. Over two-and-a-half decades, the distribution is linear and reveals a scale-invariant power law $N \propto \left(\frac{1}{\text{box size}}\right)^D$ with a fractal dimension $D = 1.49 \pm 0.02$ (Fig.4c). The fractal dimension is also obtained by analyzing the domains’ area (A), perimeter (P), and area distribution (D(A)), where the perimeter scales as $P \propto A^{0.5}$, and yields $D = 1.53 \pm 0.03$ (Fig.4c) in agreement with the box counting method. On the other hand, the domains’ area distribution scales as $D(A) \propto A^{-1}$ as the geometric clusters become near-critical. Fig.4e shows the power-law behavior of the distribution that persists for over two decades of scaling with the Fisher exponent $\tau = 1.62 \pm 0.16$ (Fig.4d). We further examine the scaling behavior by extracting the radius of gyration (R) of the domains, which is expected to display power-law behavior as a function of the perimeter (P) and area (A) near critical points. From Figure 4e,f, the power-law behavior is evident revealing the hull and volume critical exponents $d_h = 1.39 \pm 0.03$ and $d_v = 1.76 \pm 0.06$. Finally, following the earlier work, we calculate the pair-connectivity function for the map in Fig.4a for both clusters (see Supplementary Note 8). The pair-connectivity is a measure of the probability that two different sites separated by a distance $r$ on the map belong to the same cluster. Fig.4g shows the extracted data and fits to power-law with exponential cut-off $r^{-\eta} e^{-r/\chi}$, where $\eta$ and $\chi$ are the scaling exponent and the correlation length, respectively. The pair connectivity function displays a power law behavior for the black clusters (MSL; $\eta = 0.06$) with correlation length of 15 nm in contrast to the orange cluster (HFL) which dominates the map with a correlation length of over 1000 nm (larger than the map itself) indicating that the system is in proximity to a critical transition but is located on the HFL side of the phase diagram.
The power-law scaling over more than two and a half decades of the various physical quantities displayed in Fig.4 indicates a scale-invariant fractal geometry in proximity to a critical point. The values of the different extracted exponents, however, cannot be explained by the uncorrelated percolation theory indicating that strong electronic correlations originating from magnetic frustration are in play in the formation of the spatial geometric patterns.

Our work reveals a remarkable new route for tuning quantum phases at the nanoscale. It also raises the question of what microscopic physics allows such an exquisite tuning. We suggest that the sensitivity of this tuning is amplified by the dilute carrier nature of Pr$_2$Ir$_2$O$_7$. ARPES shows that the Fermi energy of the conduction electrons lies close to a quadratic band-touching\textsuperscript{4}. Using this band structure as an input, we show in Fig.4h that relatively small variation of the electron filling ($\mu/D$, where $\mu$ is the chemical potential and $D$ is the bandwidth), caused by the disorder potential, can produce a sizable change to the bare Kondo scale $T_K(\mu)$. This variation is amplified by the closeness of Fermi energy to the quadratic band-touching, in contrast to the generic case, where the Fermi energy is deep in the middle of a conduction-electron band (see Supplementary Note 9), which highlights the amplified ability of using the disorder potential to tune the ratio between the Kondo and RKKY energy scales and, hence, the balance between the HFL and MSL phases.

In conclusion, the findings in this work reveal the rich and complex behavior of emergent electronic states and their near-critical nature in a rare quantum material where magnetic frustration and Kondo quenching are considered on an equal footing. From the global phase diagram perspective (G-J\textsubscript{K}), our results indicate Pr$_2$Ir$_2$O$_7$ to be located near the delocalization-localization transition line (orange line in Fig.1a) between the frustration-induced spin liquid phase with Kondo-destroyed small Fermi surface that resembles the Ir $d$-electrons with quadratic band-touching and the Kondo-screened HFL phase with large Fermi surface incorporating the Pr $f$-electrons. The nanoscale pattern formation and the interweaved nature of the two electron phases, observed here, provide a valuable framework to the theoretical efforts in understanding the critical region of the global phase diagram. It also unifies the disparate experimental observations on Pr$_3$Ir$_2$O$_7$ as all resulting from the phase coexistence of HFL and MSL phases\textsuperscript{2-4}. Finally, it is important to state that the existence of interweaved domain boundaries between the HFL and MSL discovered in this work and the domain-wall states along these boundaries may play a relevant role in the emergence of the chiral spin-liquid and the anomalous Hall effect (AHE) observed at much lower temperatures\textsuperscript{5}. Indeed, theoretical calculations indicate that the AHE is significantly enhanced across the quantum critical point between MSL and HFL, where the Fermi surface suddenly reconstructs\textsuperscript{26}, which from our experiments occur at the domain boundaries.
Methods

Sample Preparation
Single crystals of Pr$_2$Ir$_2$O$_7$ of size approximately 1 cubic millimeter were grown using a flux method. Samples of roughly 1 x 1 x 1 mm$^3$ were attached to metallic plates using H74F epoxy. A conducting channel made of H20E silver conductive epoxy was formed from the plate to the side of a sample. An aluminum post was attached to the top surface of the sample perpendicular to the (111) cleaving plane using H74F epoxy.

Scanning Tunneling Microscopy
Samples are cleaved in situ in ultra-high vacuum at room temperature by knocking off the aluminum post. It is then immediately transferred to the STM where it is placed next to a Cu(111) crystal which is used to prep the PtIr tips before each experiment. They are both cooled down to the desired experimental temperature. The Cu is treated prior to tip preparation by several rounds of sputtering and annealing. STM topographs are taken in constant current mode, and the $dI/dV$ measurements are performed using a lock-in amplifier with a reference frequency of 0.921kHz.

Theoretical modeling
We describe the calculation to study the effect of tuning the local potential on the Kondo scale in the case of quadratic touching conduction-electron bands. Two conduction-electron bands ($c_1$ and $c_2$), each with a quadratic dispersion, touch at energy 0 (Fig. 4h, inset). The local potential is modeled in terms of an effective chemical potential, $\mu$. The bands interact with a spin-1/2 local moment by the Kondo coupling $J_K$. The model is solved in terms of saddle point equations that are exact in the limit of large $N$, where $N$ corresponds to generalizing the SU(2) spin symmetry to SU(N). In the calculation, $N$ is set to the physical value 2. We use the pseudo-fermion representation, in which the local moment is described by

$$ S = \frac{1}{2} f^\dagger \sigma f, $$

where $\sigma$ are the generators of the SU(N) group, along with the constraint by $\sum_\sigma f^\dagger f = N/2$. A Hubbard Stratonovich transformation is used to decouple the Kondo coupling term, which yields an effective hybridization:

$$ \sum_\sigma p f^\dagger_\sigma \left( B_1 c_{1,\sigma}(p) + B_2 c_{2,\sigma}(p) \right) + h.c. $$

Here, $B_1$, $B_2$ are the auxiliary fields for the decoupling, and become complex numbers in the large $N$ limit. We minimize the ground state energy of the effective model, subject to the pseudo-fermion constraint. The conduction-electron bands correspond to a pseudogap in the total density of states, with $\rho(E) \propto |E/D|^{1/2}$. We set the half bandwidth $D=1$; for each band, we normalize $\rho(E)$ so that its area is 1. For illustrative purpose, we focus on the Kondo coupling $J_K = 0.5D$. The Kondo energy scale is derived by calculating the shift of the energy due to the Kondo coupling:

$$ E_K^0 \equiv k_B T_K^0 = E(J_K) - E(J_K = 0). $$

Data availability
The authors declare that the main data supporting the findings of this study are available within the article and its Supplementary Information files. Extra data are available from the corresponding author upon request.

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Author Contributions
M.K. and J.F. performed the STM measurements. M.K., J.F. and M.G. performed the data analysis. Q.Y. and S.N. synthesized and characterized the materials. K.S. and M.L. performed the K-means analysis. S.H. and A.N.K. performed the DFT calculations. H.H., S.E.G., and Q.S. performed the theoretical calculations. P.A. designed the project and wrote the manuscript. All authors commented on the manuscript.

Competing Interests
The authors declare that they have no competing interests.

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Figures

Figure 1

Fig 1. (a) Global phase diagram of frustrated Kondo lattice. The dashed line represents delocalization-localization phase transition line between a HFL with large Fermi surface (P_L) and a MSL with small Fermi surface (P_S). (b) Top left and right images show a view of the (111) plane of the Pr-rich and Ir-rich surfaces, respectively, exposing the Kagome lattice structure, which represents the layer perpendicular to the [111] direction of the pyrochlore lattice. The bottom image shows a side view of two Pr-rich surfaces sandwiching an Ir-rich surface. Blue, green and red spheres correspond to Pr, Ir, and O atoms, respectively. (c) Topographic image (temperature of 33 K, set-point bias of 100 mV, set-point current of 500 pA) of the cleaved (111) surface showing an atomically resolved Kagome lattice structure. The left inset is a zoom-in of the black box in the main image, showing more clearly the atomic structure. The right inset is the Fourier transform showing 3-fold symmetric Bragg peaks. (d) Topograph (temperature of 45 K, set-point bias of 100 mV, set-point current of 500 pA) showing atomically resolved terraces with a step height corresponding to the unit cell in the [111] direction, with small islands observable on the terraces. (e) Linecuts of the topograph along the red and green lines in the left inset of (c). (f) Height derivative of the linecuts in (e) indicating no missing atoms. (g) Linecut along the blue line in the topograph in (d), showing the main terraces formed by a Pr-rich surface and the islands formed by the Ir-rich surface. The dashed horizontal lines represent the DFT step height from a Pr-rich to Ir-rich terraces.
Fig 2. (a)-(c) STM conductance maps at three different temperatures on different fields of view. Each map is 40 nm in size and corresponds to the local DOS at an energy of -10 meV below $E_F$. Set-point bias is 100 meV for all maps. Set-point current is 200 pA in (a) and 500 pA in (b, c). A clear electronic inhomogeneity with nanometer length scale is observed for all temperatures. The gray lines in (c) are an overlay of the unit cell step and islands from Figure 1(d) demonstrating no link between the electronic and structural features. (d)-(f) Spectra taken along the black dashed arrows shown in the conductance maps. Each arrow crosses from one domain into the other. The arrows alongside the spectra mirror the directions of the black arrows.
Fig 3. (a)-(c) K-means cluster maps for three temperatures corresponding to the maps shown in Fig.2(a-c). Each map exhibits three main clusters. (d)-(f) The average of all spectra in each of the clusters from the k-means map. (g)-(i) Individual spectra plotted from the orange and black areas of the conductance maps corresponding to the k-means maps. There is a clear difference between the spectra from these different areas at all three temperatures.
Figure 4

FIG. 4. (a) K-means cluster map forced using two clusters. The black clusters were used for the fractal analysis. (b) The fractal dimension analysis using a box-counting method resulting in fractal dimension $D = 1.49 \pm 0.02$. The data points are fitted by the least square method to the power law $N = \left( \frac{1}{\text{boxsize}} \right)^D$ where $N$ denotes the number of boxes of clusters. (c) Perimeter vs area of the clusters. Filled circles are excluded from the fit. Using $P = A^{0.72}$ gives a fractal dimension value of $D = 1.53 \pm 0.03$; this corresponds well with the box counting method. (d) Cluster area distribution with logarithmic binning, giving a fractal dimension value of $D = 1.53 \pm 0.03$. (e) Utilizing the radius of gyration for each cluster, $R_g = \sqrt{\langle (r - \langle r \rangle)^2 \rangle}$, where $r$ is summed over all points in that cluster, the equation $A = R_g^{d_v}$ gives a value of $d_v = 1.76 \pm 0.06$. (f) Similarly using $P = R_g^{d_h}$ gives a value of $d_h = 1.39 \pm 0.03$. (g) Pair connectivity function extracted from the black (black) and orange (orange) regions. The lines are fit to the function $y = r^{-1} e^{-r/\Delta}$, where $\eta$ and $\chi$ are the scaling exponent and the correlation length. (h) Kondo scale $T_K(\mu)$ normalized by $T_K(\mu_0 = -0.2D)$, as a function of chemical potential $\mu$ in case of a quadratic touching conduction electron bands. The Kondo scale is seen to be highly sensitive to slight variations of $\mu$ near the node (Fermi energy) in the band structure. The inset illustrates the two conduction-electron bands, each with a quadratic dispersion, touching at the Fermi energy.
Supplementary Material for “Discovery of nanoscale phase coexistence of heavy Fermi-liquid and metallic spin-liquid in geometrically frustrated Pr$_2$Ir$_2$O$_7$”

Supplementary Note 1

Overview of DFT studies on Pr$_2$Ir$_2$O$_7$

It has been shown experimentally that Pr$_2$Ir$_2$O$_7$ in its ground state (GS) is a metallic compound with no long-range magnetic order$^{1-7}$. Only two computational studies have been conducted at the density functional theory (DFT) level on characterizing the Pr$_2$Ir$_2$O$_7$ GS$^{8,9}$. Simulations with the the local density approximation (LDA)+U approach have shown that a paramagnetic semi-metallic state favored at $U_{\text{eff}} = 0$ becomes antiferromagnetic (AFM) semi-metallic at $U_{\text{eff}}$ values of $\sim$1.3 eV and eventually AFM insulating at $U_{\text{eff}} \sim 1.5-1.6$ eV$^8$, in disagreement with the experimental observations. More recently, Zhang et. al.$^9$ argued that the LDA+U approach is not a proper treatment for Pr$_2$Ir$_2$O$_7$ due to the intrinsic tendency of the +U correction to favor magnetic insulating states. The study relied on the LDA+DMFT (dynamical mean field theory)$^{10}$ approach and successfully reproduced the experimentally observed properties of the Pr$_2$Ir$_2$O$_7$ GS.

The primary focus of our DFT calculations was to (i) establish energetically favorable surface terminations in cleaved Pr$_2$Ir$_2$O$_7$ samples at zero temperature and (ii) simulate STM images of possible terminations to help interpret experimental observations. The significant computational cost of the LDA+DMFT treatment$^{11}$ makes it an impractical approach for the analysis of complex surfaces. In order to identify a suitable DFT flavor for this study, our starting point was the examination of Pr$_2$Ir$_2$O$_7$ GS properties within three commonly used DFT$^{12,13}$ approaches: the generalized gradient approximation (GGA) with the Perdew–Burke–Ernzerhof (PBE) functional$^{14}$, PBE+U$^{15,16}$, and Heyd–Scuseria–Ernzerhof (HSE) hybrid functionals$^{17}$.

Tests of DFT approximations

Our benchmarking included (i) a full optimization of the Pr$_2$Ir$_2$O$_7$ bulk structure with the PBE and PBE+U approaches; and (ii) comparison of the magnetic order in the PBE, PBE+U, and HSE treatments against the experimental observations. For the PBE (+U) treatments, we also examined the effect of the spin-orbit coupling (SOC) and found it to be insignificant for the relevant structural, magnetic, and stability properties of Pr$_2$Ir$_2$O$_7$. 
Supplementary Figure 1. (a) Relative energy of the ferromagnetic vs. non-magnetic bulk structures within various DFT treatments, and (b) average magnetic moment per Ir atom in the ferromagnetic bulk structure optimized with different DFT approaches.

All DFT calculations were based on the PBE parametrization of the generalized gradient approximation (GGA) and the projector augmented wave (PAW) potentials as implemented in VASP. A high energy cut-off of 500 eV and a dense k-point sampling of the Brillouin zone within the Monkhorst-Pack approach were employed to ensure the numerical convergence of the calculations. In the PBE+U treatment, we optimized the bulk GS with three values of $U_{\text{eff}}$: 1.2, 1.6, and 2.0 eV. For the HSE hybrid functional calculations, we used the HSE06 range-separated functional with 25% of short-range exact exchange interaction mixed, and chose to not downsample the Brillouin zone in the calculation of the exact non-local exchange term to achieve higher accuracy. As a common practice, the computationally demanding HSE calculations were performed for the structure fully optimized at the PBE level.

Supplementary Fig. 1a shows the energy difference between the Pr$_2$Ir$_2$O$_7$ bulk structure configurations with non-magnetic and collinear ferromagnetic (FM) initializations of atomic moments. Among the considered DFT flavors, only the PBE treatment correctly did not stabilize an (A)FM ordering, while the PBE+U and HSE approximations produced lower-energy states with significant FM moments on Ir atoms (Supplementary Fig. 1b). In the SOC non-collinear calculations, we extended the set of magnetic moment initializations with all-in-all-out and 2-in-2-out AFM configurations for Pr atoms in combination with various initial values for the Ir and O atoms and observed the same pattern: a non-magnetic state with PBE and non-zero magnetic moments on Ir atoms in PBE+U for all considered $U_{\text{eff}}$ values. We determined that the fully relativistic treatment had little effect on the structural parameters of the PBE-relaxed structure: inclusion of the SOC produced forces below 0.03 eV/Å and changed the lattice constant by less than 0.15%. Given this minor impact of the SOC on the properties relevant for the current study and the high computational cost of the fully relativistic treatment, we chose to rely on the PBE calculations in the simulation of Pr$_2$Ir$_2$O$_7$ surfaces.

### Surface structures and exfoliation energies

Full optimization of the cF88-Pr$_2$Ir$_2$O$_7$ structure with space group #227 at the PBE level resulted in lattice parameter $a = 10.512$ Å and Wyckoff positions $48f$ (0.4190,1/8,1/8) and $8a$ (1/8,1/8,1/8) for O, $16d$ (1/2,1/2,1/2) for Ir, and $16c$ (0,0,0) for Pr atoms. This structure was used to construct the slabs for comparing the surface stabilities.

As illustrated in Supplementary Fig. 2a, the bulk Pr$_2$Ir$_2$O$_7$ GS structure in the (111) direction consists of alternating Pr- and Ir-rich layers (which will be referred to as A and B layers). Each O is bonded to 3 metal
atoms within the layer and 1 metal atom in a neighboring layer, while the metal coordinations for bulk are PrO$_8$ and IrO$_6$. In isolated layers, the coordinations are (3PrO$_6$ & 1IrO$_6$) for A and (1PrO$_6$ & 3IrO$_4$) for B, so that one can assign (3xO+1xO$_3$)/3 = 8 O atoms to A layers and (1xO$_6$+3xO$_4$)/3 = 6 O atoms to B. Three relevant slab types can be constructed with the following surface terminations: [AB]$_n$, [AB]$_n$A, and [BA]$_n$B, where $n$ is the number of full formula units in the slab. The [AB]$_n$ slabs are non-symmetric but stoichiometric (Supplementary Fig. 2a), while [AB]$_n$A and [BA]$_n$B slabs are symmetric but non-stoichiometric (Supplementary Fig. 2b,c).

Supplementary Figure 2. Pr$_2$Ir$_2$O$_7$ (111) slabs are formed from Pr- and Ir-rich layers (A and B layers, respectively). (a) A non-symmetric stoichiometric [AB]$_2$ slab contains two formula units of the bulk structure with O, Ir, and Pr atoms shown in yellow, red, and blue, respectively. (b) A symmetric non-stoichiometric [AB]$_2$A slab includes two formula units of bulk (shown with grey color) and an A layer. (c) A symmetric non-stoichiometric [BA]$_2$B slab with two bulk units (shown with grey color) and an extra B layer.

The typical procedure in the calculation of surface energy is to construct slabs of the same termination and then calculate the surface energy as

$$E_{surf} = \frac{1}{2A}(E_{slab} - nE_{bulk})$$

in which $A$ is the surface area, $E_{slab}$ is the total DFT energy of the optimized slab, $n$ is the number of bulk formula units in the slab, and $E_{bulk}$ is the DFT energy of the bulk structure per formula unit. However, since the [AB]$_n$A and [BA]$_n$B slabs are non-stoichiometric, this equation cannot be used to determine the surface energy of the A and B terminations separately. Supplementary Fig. 3b shows the convergence of the combined (A+B) surface energy as a function of the number of [AB] units in the stoichiometric slabs. The results indicate that converged results for surface stability can be obtained for slabs with only two [AB] units (44 atoms). The value of 132 meV/Å$^2$ (6.3 eV/u.c. of (111) slab) can be attributed primarily to the break of 3 Ir-O and 4 Pr-O bonds per u.c. of the slab. Supplementary Fig. 3(b) also illustrates that both A and B surfaces undergo similar degrees of structural rearrangement, as the energy differences between relaxed and unrelaxed terminations are close to 10 meV/Å$^2$ for each side.
The stability of the individual A and B terminations can also be probed via the exfoliation of single layers. For the \( X = A \) and \( B \), the exfoliation energy \( E_{\text{exf}} \) is defined as:

\[
E_{\text{exf}} = \frac{1}{A} \left( E^X + E^{[AB]n} - E^{[AB]|nX|} \right)
\]

in which \( E^X \) is the energy of a single layer \( X \) and all energies are obtained after full optimization of the atomic positions in the slab. For each PBE calculation, we used the above-mentioned settings while the generated slab was separated with a 15 Å vacuum in the non-periodic direction to isolate the periodic boundary condition's effects in the DFT run. In geometry optimizations, all atoms were allowed to move while the lattice parameters were kept fixed. Moreover, to ensure that electric dipole effects do not affect our calculations, we ran a set of optimization with dipole correction included and observed no difference in optimized geometries and the energy shift was \( \sim 0.5 \text{ meV/atom} \).

Supplementary Fig. 3a shows the exfoliation energy for \([AB]_nA\) and \([BA]_nB\) for \( n \) up to 4. According to these calculations, the exfoliation energies for A and B layers are 158 and 62 meV/Å\(^2\), respectively. Note that their sum of 220 meV/Å\(^2\) is lower than the doubled value of the surface energy of \( 2 \times 132 = 264 \text{ meV/Å}^2 \), so it is more energetically favorable to lift two individual A and B layers than to create two new cuts through the middle of the sample.

It is important to note that transfer of top-most O atoms between A- and B-terminated surfaces during sample cleavage is expected to be uncommon because of unfavorable energetics. Our PBE calculations for stoichiometric \([AB]_3\) slabs revealed that changing the natural 4:3 ratio of O atoms covering the respective Pr- and Ir-rich surfaces to 3:4 (5:2) raises the total energy by 1.2 (3.6) eV. The PBE+SOC calculations produce similar 1.2 (3.8) eV penalties. Therefore, cleaved (111) surfaces are expected to retain the natural number of O atoms.

Supplementary Figure 3. (a) Exfoliation energy of A (Pr-rich) and B (Ir-rich) layers for peeling a single layer from the top of an \([AB]_n\) slab. (b) Total surface energy for \([AB]_n\) slab when the slab is unrelaxed (gray), only the B layer is relaxed (red), only the A layer is relaxed (blue), and the full slab is relaxed (black).

**STM simulation of Pr- and Ir-rich surfaces**

In order to simulate the constant-current STM images, we carried out an analysis of charge density isosurfaces for the A and B surface terminations. In each case, we optimized the slab at the PBE level with
the settings detailed above. Using the charge density and wave functions from this run, a secondary calculation was performed to obtain the partial charge density for the states within ±1 eV from the Fermi energy. Based on this data, we mapped the isosurfaces corresponding to various values of the charge density and obtained simulated STM images displayed in Supplementary Fig. 4.

An important observable in experimental STM images is the "step size" between surface regions defined by different terminations, such as the type-A and type-B layers. Corresponding values can be computed from the DFT charge density profiles and used to differentiate surface types observed in the experiment. Our DFT results in Supplementary Fig. 5 and Supplementary Table 1 show consistent values of the calculated height steps for a wide range of currents in constant-current STM imaging: 3.44 Å and 2.65 Å for going from layer A to B and from layer B to A, respectively. Taking into account the grid size used to output the charge density in VASP and the variations in the metal atom positions within layers, we estimated the numerical error in the step height evaluations to be 0.12 Å. As discussed in the main text, this information turned out to be in excellent agreement with the experimental measurements and allowed us to identify conclusively plateaus with Pr- and Ir-rich terminations.

Supplementary Figure 4. Simulated constant-current STM images for the Pr-rich (left) and Ir-rich (right) surfaces. The images demonstrate the relative depth of the 0.029 e Å⁻³ charge density iso-surface. Darker color indicates a deeper positioning of the charge density iso-surface.

Supplementary Figure 5. Schematic of the average step size for a given value of the charge density going from (a) layer A to B; and (b) layer B to A. The values were obtained from the analysis of the charge density profile as detailed in Table 1.

| charge density (e Å⁻³) | structures and step heights (Å) |
|------------------------|---------------------------------|
|                        | A to B                          |
|                        | B to A                          |
| 0.364                  | 3.44                            |
| 0.675                  | 3.44                            |
| 0.965                  | 3.45                            |

Supplementary Table 1. Step size of the charge density going from layer A to B and vice versa. The values were tabulated for three different values of the charge density obtained in our DFT calculations.
Supplementary Note 2

One base approximation of STM is that the conductance is roughly uniform over the measuring surface; this ensures that the conductance is proportional to the LDOS. This is enforced by maintaining the measured current above or below the Fermi energy at a constant value. In the case of Pr$_2$Ir$_2$O$_7$, however, this approximation breaks down, leading to an arbitrary scaling of spectra from one point to another on the surface. This renders the results of unsupervised machine learning unintelligible with unphysical bias.

The setpoint effect results in arbitrary modulations of the data samples

$$x_n(r) = \lambda(r)x_n^{\text{physical}}(r)$$

where the $x_n$ are D-dimensional data vectors and $\lambda(r)$ accounts for an unknown setpoint scaling. To remove the unphysical modulation caused by the setpoint effect, each spectra was re-scaled to have unit norm in data-space upon interaction with the K-means algorithm. This is a bit different from the typical data science pre-process such as standardization, in which each of the D elements per sample has zero mean and unit standard deviation. We make this choice of normalization to remove the overall modulation due to the form of the setpoint effect. Following this clustering, all spectra are mapped back to their unnormalized form.

This input data varies from being on a 64 by 64 grid to a 256 by 256 grid, with spectra sampled at either 101 or 11 energies, respectively.

All K-means calculations were implemented via SciKit-Learn's software package$^{26}$, written in Python 3.7, and coded in the Jupyter environment$^{27}$. We now give a brief review of the K-means framework.

The K-means algorithm sorts $N$ D-dimensional samples, $x_{nab}$, into $K$ clusters by separation distance in data-space. The distortion measure

$$J = \sum_n \sum_k r_{nk} \|x_n - \mu_k\|^2$$

gives the degree to which the clusters represent the original data. While in this paper we used D-dimensional Euclidean distance for our kernel, other options are certainly worth exploring in the future. The $r_{nk}$ are binary indicator variables which assign the $n$th sample to the $k$th cluster: $r_{nk} = 1$ if $k = \text{argmin}_j |x_n - \mu_j|^2$ and $k = 0$ otherwise; the $\mu_k$ are the cluster centers (means). We would like to determine the $r_{nk}$ and $\mu_k$ which minimize the distortion measure$^{28}$.

This can be done iteratively, first by picking some initial values of the $\mu_k$, often randomly assigned. Each iteration that follows has two phases: first assign each sample to the cluster with the most proximal mean, then recalculate the mean of each cluster. That is, we minimize J with respect to the $r_{nk}$ with the $\mu_k$ fixed, then minimize J with respect to $\mu_k$ with the $r_{nk}$ fixed. Iteration is continued until convergence is obtained and there is no change in cluster assignment; convergence is guaranteed, though convergence to a global minimum is not$^{28}$.

Phase 1: $r_{nk} = \begin{cases} 1 & \text{if } k = \text{argmin}_j |x_n - \mu_j|^2 \\ 0 & \text{otherwise} \end{cases}$; with $\mu_k$ fixed.

Phase 2: $\mu_k = \frac{\sum_n r_{nk} x_n}{\sum_n r_{nk}}$; with $r_{nk}$ fixed.
We derive the equation used for Phase 2 from: \( \frac{d}{d\mu_k} = 2 \sum_n r_{nk} (x_n - \mu_k) = 0.28 \)

The output of the K-means algorithm is a pre-determined number of clusters into which all the inputted spectra are sorted. We visualize these clusters in a cluster map, which shows the real-space distributions of each cluster. We also examine the means of each cluster.

The input to the K-means algorithm was purely spectra, with no spatial information. However, the output of the algorithm shows a strong tendency of the spectral clusters to define spatial clusters, which we explore with the pair connectivity function.
Supplementary Note 3

We compare the results of the K-means calculations, which result in a cluster map (Supplementary Fig. 6a), with the corresponding conductance map from which it was formed (Supplementary Fig. 6b). We see a strong correlation for energies near the resonance (the -20meV energy is shown in Supplementary Fig. 6b). We observe this correlation both visually, as many of the clusters in each map clearly match up, as well as by calculating the correlation between the two maps. We plot this correlation as a function of conductance map energy (inset of Supplementary Fig. 6b).

Supplementary Figure 6. (a) A 3-cluster K-means map. (b) A 256-pixel conductance map on which the K-means algorithm in (a) was run. This map shows the -20meV energy. Even by eye there is clearly a strong correlation between the conductance map and its corresponding K-means cluster map. The correlation is calculated to be 85%.

We see that there is a strong correlation at biases below the Fermi energy, with the strongest correlations being around -10 to -30 meV. Thus, the K-means cluster maps are analogous to the STM conductance maps at energies around -20meV. The fact that the correlation drops for energies away from -20meV is partially related to the STM set-point effect.
Supplementary Note 4

To fit the resonance in the spectra, we use the Fano lineshape equation, which models the Kondo resonance. The Fano equation is given as

\[ \frac{dI}{dV} \sim A \frac{(V - E + q)^2}{1 + \left(\frac{V - E}{\Gamma}\right)^2} + C + d \cdot V \]

where \(E\) is the energy of the resonance, \(\Gamma\) is the width of the resonance (half width at half max), \(A\) is related to the amplitude of the resonance, \(C + d \cdot V\) provides a linear background, \(V\) is the applied bias, and \(q\) is the asymmetry parameter of the Fano equation.

We start off by examining the average spectra of each of the three clusters. We know that the spectra which exhibit the resonance that we want to examine come from one of the three clusters shown in the K-means cluster maps. We then binarize the cluster maps so that the cluster which exhibits the resonance is separated from the two clusters which don’t have this resonance (Supplementary Fig. 7).

We then fit spectra from the cluster showing the resonance; we do this because the Fano lineshape model is based on the existence of a resonance. Trying to fit the model to a spectrum which doesn’t have this resonance is a meaningless exercise.

When we examine some spectra from this resonance cluster, we see that the magnitude of the resonance continuously weakens and often disappears near the boundaries of the cluster (Supplementary Fig. 8). This occurs as a result of the continuous phase transition, which happens when going from one domain to the other.
Three spectra taken from the cluster whose average shows the Kondo resonance. Clearly the first spectrum (left) strongly shows the resonance; the second (middle), taken from a domain edge, shows the resonance quite weakly, while the third (right) does not show the resonance. (d)-(f) Derivative of the spectra in (a)-(c), respectively. Clearly, the derivatives of the spectrum which show the resonance are quite different than the derivative of the spectra without.

In order to further remove spectra which do not have the resonance, we look at the first derivative of the spectra around the region where the resonance is seen; the resonance occurs around -10meV, so we examine a range of roughly -40 meV to 10 meV. When the resonance is present, the maximum value of the derivative is much larger than the average value of the derivative (Supplementary Fig. 8). Using this fact, we remove from our fitting any spectra where the maximum is not significantly (usually 5 to 10 times as much) larger than the average. Supplementary Fig. 9 shows the results of this filtering: we see the map from Supplementary Fig. 7b, and now the orange cluster is a more limited set of resonance spectra. This is the set to which we fit the resonance model. Additionally, many of the spectra which we remove this way, marked in green, are on the edges of domains, which indicates that the resonance is likely weaker on domain edges than at the center of the Kondo resonance domains.
Supplementary Figure 9. The map from Supplementary Fig. 7b, where the orange corresponds to the resonance areas and the black corresponds to the non-resonance areas. Now, the resonance clusters are filtered to remove as many non-resonance spectra as possible; these newly removed spectra are shown in green. Most of these are on domain edges. The remaining orange spectra are the ones we fit to the Fano lineshape.

We now fit our filtered data using the Fano lineshape between -40 meV to 10 meV to accurately capture the resonance peak. Each spectrum is fit to the Fano lineshape with a simple linear background. Supplementary Fig. 10 shows a representative spectrum along with its fit in the -40 meV to 10meV range. From each fit we extract histograms of the various parameters in the Fano equation.

Supplementary Figure 10. A spectrum (blue markers) showing the Kondo resonance along with the fit (red line) of the resonance to the Fano lineshape.
We perform this analysis for K-means cluster maps formed from data at both 10K and 33K. We represent each of the parameters of interest, $\Gamma$, $E$, and $q$, in the form of a histogram; we additionally calculate the amplitude, given as $A(1 + q^2)$. Supplementary Fig. 11 shows the histograms of these four quantities for both 10K and 33K. For these eight sets of data we calculate both the average value and the standard deviation. This standard deviation is calculated for each based off of the Gaussian which each histogram shows.

Supplementary Figure 11. (a)-(d) 10K data. (a) shows a histogram of the linewidth with an average of 7.4 ± 2.2 meV. This results in a Kondo temperature of 56K. (b) shows a histogram of the energy with an average of -7.6 ± 3.2 meV. (c) shows a histogram of the energy parameter $q$ with an average of 0.4 ± 0.9. (d) shows a histogram of the amplitude with an average of 0.001 ± 0.006. (e)-(h) 33K data. (e) shows a histogram of the linewidth with an average of 11.8 ± 1.9 meV. This results in a Kondo temperature of 64K. (f) shows a histogram of the energy with an average of -13.9 ± 3.2 meV. (g) shows a histogram of the asymmetry parameter $q$ with an average of -2.2 ± 1.0. (h) shows a histogram of the amplitude with an average of 0.04 ± 0.01. All errors are calculated as the standard deviation of the Gaussian shown.

Using the average value of the linewidth of this Kondo resonance, and a relationship between the linewidth and the Kondo temperature,

$$\Gamma = \sqrt{(\pi k_B T)^2 + 2(k_B T_K)^2}$$

we calculate the Kondo temperature. For the 10K dataset we get a Kondo temperature of 56 ± 20 K; for the 33K dataset we get a Kondo temperature of 64 ± 20 K. Since we form our range of Kondo temperatures from the standard deviation of the linewidths, the $T_K$ and the error here correspond to the range of Kondo temperatures through the entire Gaussian distribution.

Additionally, we attempt to perform this fitting analysis for K-means cluster maps from 45K data. As expected from the average spectra resulting from the 45K cluster maps, the spectra at this temperature exhibit a very weak resonance. Furthermore, since the background is unknown, it makes the fitting less reliable.
Supplementary Note 5

We now examine histograms of conductance maps for data at both 10K and 33K, specifically the conductance maps at the energy where we observe the resonance peak, which happens around -10 to -20meV. In Supplementary Fig. 12a we show a histogram of all conductance values from a conductance map taken at 10K and at an energy of -10meV. Similarly, in Supplementary Fig. 12c we show a histogram of all conductance values from a map taken at 33K and also at an energy of -10meV.

For each temperature, we start with the 2-cluster K-means map (binarized map). Using the two clusters from K-means we split the conductance map into the same two clusters. We then take each of the two clusters individually and create a histogram from the conductance values which form each cluster. Plotted in Supplementary Fig. 12b and 12d we show the overall histogram (as seen in Supplementary Fig. 12a and 12c) along with the two clusters’ individual histograms.

![Graphs showing histograms of conductance values at 10K and 33K](image)

Supplementary Figure 12. (a),(c) show the histograms of the entire set of conductance values taken from the conductance map at the resonance energy. (a) is at 10K and is taken from a -10meV conductance map and (c) is at 33K and is also taken from a -10meV conductance map. (b), (d) show the histograms in (a),(c), respectively, with the histograms of the two clusters overlaid.

The histograms in Supplementary Fig. 12a, c allow us to look at the density of states (the value of the conductance) near the resonance, which is how we define our clusters. We see in the histograms the
formation of a single Gaussian, rather than a binomial distribution, which indicates we have a continuous transition from the areas of low conductance to areas of high conductance.

Additionally, when we plot the density of states of the two clusters individually, we see that rather than being binomial there is a large overlap between the two, which supports the conclusion that one cluster transitions continuously into the other.
Supplementary Note 6

When analyzing the geometric clusters, one of the important factors to consider is the accuracy of those clusters. The resolution of the conductance maps taken has a major effect on how accurately we see these clusters. With too low of a resolution, smaller clusters are not seen, and this greatly affects the analysis. Seen in Supplementary Fig. 13a and 13b are two conductance maps, both taken on the same area of 65nm. The first map is taken with a 64x64 pixel resolution, while the second is taken with 256x256 pixel resolution.

Supplementary Figure 13. (a) A conductance map taken with 64x64 pixel resolution. (b) A higher resolution 256x256 pixel conductance map taken on the same area as the map in (a). (c) Zoomed-in image of the red box in (a). (d) Zoomed-in image of the red box in (b). Much more detail is clearly evident.
Looking at the map overall, many of the main clusters are seen in both maps. However, the details within these larger clusters are quite different between maps. This is because one pixel of the lower resolution map is equivalent to 16 pixels in the higher resolution map, thus allowing us to see much more detail. Supplementary Fig. 13c and 13d shows zoomed-in images from the red boxes in Supplementary Fig. 13a and b, allowing us to clearly see the difference in detail between the two resolutions.

The areas and boundaries of each cluster are much more detailed, clusters which appeared to be connected can be seen to actually be separate, and smaller clusters that can’t be visualized in the lower resolution map can be seen in the higher resolution map. This improved resolution of clusters and their boundaries is essential for any proper geometric cluster analysis.
Supplementary Note 7

As discussed in the main paper, we use a K-means cluster map for the analysis of the domains. In Supplementary Fig. 14 and 15 we first compare the conductance map and K-means cluster map of the same data set. Supplementary Fig. 14a and 15a show the conductance map and K-means maps, respectively, of the same dataset. Supplementary Fig. 14b shows the spectra taken from high and low areas of conductance from the conductance map in Supplementary Fig. 14a; Supplementary Fig. 15b shows the average spectra from each of the two clusters shown in the K-means map in Supplementary Fig. 15a.

Supplementary Figure 14. (a) Conductance map taken with high resolution. (b) Spectra from areas of high (orange) and low (dark green) conductance.

Supplementary Figure 15. (a) K-means cluster map from the dataset in Supplementary Fig. 14a. (b) Average spectra from the two clusters shown in a. The orange spectrum correspond to the orange clusters and the black spectrum to the black clusters.
The significant conclusion from this comparison is that the K-means map separates the spectra into two clusters corresponding to high and low conductance areas in the conductance map, and thus are used to form the domains which we investigate. Using K-means rather than the conductance map for determining domains has the additional benefit of clearly defining the boundaries of each cluster; unlike in the conductance map, where there is no clear way to determine the boundary of each cluster.

For the following analysis, we use two methods of limiting the clusters in the K-means map. The first way is to remove clusters of overly small and overly large area; this is shown in Supplementary Fig. 16b. The second method is to additionally remove clusters which cross the boundary of the image; this is shown in Supplementary Fig. 16c. There are two reasons why we want to limit the cluster areas. First, small clusters less than 5 pixels are just as likely to be random errors as they are to be a real cluster, so we eliminate them. Secondly, clusters with areas larger than 2000 pixels create much more variability and error in the fits, and often appear to be many clusters that are connected by just a single pixel, so we eliminate these as well.

Supplementary Figure 16. (a) K-means cluster map. (b) Map after having removed clusters with areas smaller than 5 pixels and larger than 2000 pixels. (c) Map after having removed both small and large clusters and additionally removing clusters crossing the boundary of the image.

While analyzing these domains we find that there is little difference between using the data in Supplementary Fig. 16b and Supplementary Fig. 16c. Thus we decide to limit our data by removing small and large clusters but leaving the boundary-crossing clusters intact.

The way that we implement this data limitation varies depending on the analysis method. For both the box-counting method and for analyzing the cluster area distribution, the input to our algorithms is the dataset where we have already removed the small and large area clusters (seen in Supplementary Fig. 16b). For the geometric domain analysis relating area, perimeter, and radius of gyration, we provide the full data (Supplementary Fig. 16a) and we then exclude the points from our fit which correspond to the large and small pixel areas in the dataset (excluded points shown by filled in circles in main paper figure).

The box-counting method is known to theoretically measure the dimension of fractal patterns. We use this idea to measure the fractal dimension of the clusters. We divide the 256-pixel map into a mesh of boxes. We vary this box size from 1 pixel up to 12 pixels, and for each different box size we count the number of boxes \( N \) that include part of a cluster. We then fit this data to the scale-invariant power law \( N \sim \left(\frac{1}{\text{boxsize}}\right)^D \), using the least square method, to find the fractal dimension \( D = 1.49 \pm 0.02 \).
Additionally, we analyze the fractal properties of these clusters using geometric domain methods. We apply an image region analysis software which looks at all the clusters and tells us both the area and perimeter of each. From these measurements we also calculate the radius of gyration,

\[ R_g = \sqrt{\langle (r - \langle r \rangle)^2 \rangle} \],

where \( r \) is summed over all the points in each individual cluster, for each cluster in the map.

A scale-invariant property similar to that of the box-counting method appears in the cluster area distribution: the cluster area \( A \) and the number of clusters \( N(A) \) follow the power law \( N(A) \sim A^\tau \). We take all the areas of the clusters, logarithmically bin them, count and normalize the numbers of clusters with areas within the logarithmic bins, and fit this data to the power law; we then get a Fisher exponent of \( \tau = 1.62 \pm 0.16 \).

Similar relationships exist relating the area and perimeter and relating the radius of gyration to the area and to the perimeter. Using \( P = cA^{D/2} \) gives a fractal dimension value of \( D = 1.53 \pm 0.03 \), using \( A = R_g^{d_v} \) gives a value for the volume fractal dimension of \( d_v = 1.76 \pm 0.06 \) and using \( P = R_g^{d_h} \) gives a value for the hull fractal dimension of \( d_h = 1.39 \pm 0.03 \).

All of the errors that we give for the various critical exponents come from calculating the 95% confidence value for each of the fittings we perform.
Supplementary Note 8

Without inputting any spatial information to the K-means calculation, the output spectral clusters manifest as spatially distinct domains. To understand the statistics of these spatial clusters, we use a tool of graph theory typical to the field of percolation called the pair connectivity function (PCF)\textsuperscript{29-31}, defined as $g_{\text{conn}}(r) = r^{-(d+2+\eta)}e^{-r/\xi}$. Using this we capture the spatial cluster size and near-transition power law behavior. Here $d$ is the dimensionality, $\eta$ is the connectivity, and $\xi$ is the correlation length.

For surface measurements $d = 2$, so $g_{\text{conn}}(r) = r^{-\eta}e^{-r/\xi}$. To compute $g_{\text{conn}}(r)$ from our experimental data we turn to an equivalent quantity: the likelihood that two points are connected, given that they belong to the same spectral cluster, and that the points are a distance $r$ apart. We know which points are in the same spectral clusters from the results of the k-means clustering. Once a sampling of $g_{\text{conn}}(r)$ has been constructed, the connectivity exponent and the correlation length are extracted by fitting this function to the extracted data.

To compute this likelihood of connection between two points, we sample the fraction

$$\frac{\text{Number of connected pairs}}{\text{Number of pairs}}$$

a thousand times at a given range. Repeating this for several values of $r$, we obtain the approximation

$$g_{\text{conn}}(r) = r^{-\eta}e^{-r/\xi} \approx \frac{\text{Number of connected pairs sampled}}{\text{Number of pairs sampled}}(r)$$

for each spectral cluster.
Supplementary Note 9

To understand the unique behaviour of the Kondo scale and its sensitivity to small perturbations of the chemical potential in the case of a quadratic band touching at the Fermi energy, we also consider, for comparison, a usual metallic system where the density of states is roughly a constant. We model this case in terms of two degenerate conduction-electron bands, each having a semi-elliptic density of state \( \rho(E) = \frac{2}{D^2 \pi} \sqrt{D^2 - E^2} \), with \( \mu \) not being too far away from \( E = 0 \).

Fig. 17 plots the corresponding result, \( \frac{T_K^0(\mu)}{T_K^0(\mu_0 = -0.2D)} \) as a function of \( \mu \). Compared with the quadratic band touching case, the Kondo scale is considerably less sensitive to the change in \( \mu \).

Supplementary Figure 17. Measure of the Kondo scale, \( \frac{T_K^0(\mu)}{T_K^0(\mu_0 = -0.2D)} \), as a function of the chemical potential for a metallic band with roughly constant density of states (red data points) and for a quadratic band touching (blue data points).
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