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Design and characterization of electrons in a fractal geometry

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The dimensionality of an electronic quantum system is decisive for its properties. In one dimension, electrons form a Luttinger liquid, and in two dimensions, they exhibit the quantum Hall effect. However, very little is known about the behaviour of electrons in non-integer, or fractional dimensions[1]. Here, we show how arrays of artificial atoms can be defined by controlled positioning of CO molecules on a Cu (111) surface[2–4], and how these sites couple to form electronic Sierpiński fractals. We characterize the electron wavefunctions at different energies with scanning tunnelling microscopy and spectroscopy, and show that they inherit the fractional dimension. Wavefunctions delocalized over the Sierpiński structure decompose into self-similar parts at higher energy, and this scale invariance can also be retrieved in reciprocal space. Our results show that electronic quantum fractals can be artificially created by atomic manipulation in a scanning tunnelling microscope. The same methodology will allow future studies to address fundamental questions about the effects of spin–orbit interactions and magnetic fields on electrons in non-integer dimensions. Moreover, the rational concept of artificial atoms can readily be transferred to planar semiconductor electronics, allowing for the exploration of electrons in a well-defined fractal geometry, including interactions and external fields.

Fractals have been investigated in a wide variety of research areas, ranging from polymers[5], porous systems[6], electrical storage[7] and stretchable electronics[8] down to molecular[5,9–11] and plasmonic[12] fractals. On the quantum level, fractal properties emerge in the behaviour of electrons under perpendicular magnetic fields; for example, in the Hofstadter butterfly[13] and in quantum Hall resistivity[14,15]. In addition, a multi-fractal behaviour has been observed for the wavefunctions at the transition from a localized to delocalized regime in disordered electronic systems[16–18]. However, these systems do not allow one to study the influence of non-integer dimensions on the electronic properties. Geometric electronic fractals, in which electrons are confined to a self-similar fractal geometry with a dimension between one and two, have been studied only from a theoretical perspective. For these fractals, a recurrent pattern in the density of states as well as extended and localized electronic states were predicted[19–22]. Recently, simulations of quantum transport in fractals revealed that the conductance fluctuations are related to the fractal dimension[23], and that the conductance in a Sierpiński fractal shows scale-invariant properties[24–26].

Here, we report how to construct and characterize, in a controlled fashion, a fractal lattice with electrons: the electrons that reside on a Cu(111) surface are confined to a self-similar Sierpiński geometry through atomic manipulation of CO molecules on the Cu(111) surface. The manipulation of surface-state electrons by adsorbates has been pioneered by Crommie et al.[27] and has been used to create electronic lattices 'on demand', such as a molecular graphene[2], an electronic Lieb lattice[3,28], a checkerboard and stripe-shaped lattice[29], and a quasiperiodic Penrose tiling[4]. We characterized the first three generations of an electronic Sierpiński triangle by scanning tunnelling microscopy and spectroscopy, acquiring the spatially and energy-resolved electronic local density of states (LDOS). These results were corroborated by muffin-tin calculations as well as tight-binding simulations based on artificial atomic *s*-orbitals coupled in the Sierpiński geometry.

The Sierpiński triangle with Hausdorff dimension $\log(3)/\log(2) = 1.58$ is presented in Fig. 1a. We define atomic sites at the corners and in the centre of the light blue triangles, as shown in Fig. 1b for the first generation $G(1)$[10,30]. $G(1)$ has three inequivalent atomic sites, indicated in red, green and blue, which differ by their connectivity. A triangle of generation $G(N)$ consists of three triangles $G(N-1)$, sharing the red corner sites. The surface-state electrons of Cu(111) are confined to the atomic sites by adsorbed CO molecules, acting as repulsive scatterers. Figure 1c shows the experimental realization of the first three generations of the Sierpiński triangle and Fig. 1d shows the relation with the artificial atomic sites. The distance between neighbouring sites is 1.1 nm, such that the electronic structure of the fractal will emerge in an experimentally suitable energy range[2].

Figure 1e presents the experimental LDOS at the red, blue and green atomic sites in the $G(3)$ Sierpiński triangle (indicated by the open circles in Fig. 1c). The differential conductance (dI/dV) spectra were normalized by the average spectrum taken on the bare Cu(111) surface, similar to ref. [2]. The onset of the surface-state band is located at $V = -0.45$ V. We focus on the bias window between -0.4 V and 0.3 V. Around $V = -0.3$ V the LDOS on the red, green and blue sites is nearly equal, whereas slightly above $V = -0.2$ V, the red sites exhibit a distinct minimum, while the green and blue sites show a considerably higher LDOS. At $V = -0.1$ V, the blue sites show a minimum, whereas the red and green sites exhibit a pronounced maximum in the LDOS. At $V = +0.1$ V, the blue sites show a larger peak in the differential conductance, whereas the green and red sites exhibit a smaller peak. The experimental LDOS is in good agreement with both the tight-binding (see Fig. 1f) and muffin-tin simulations (see Supplementary Information). This finding corroborates that our design leads to the desired confinement of the two-dimensional electron gas to the atomic sites of the Sierpiński geometry. In addition, it allows us to characterize the wavefunctions of the chosen Sierpiński geometry in detail.

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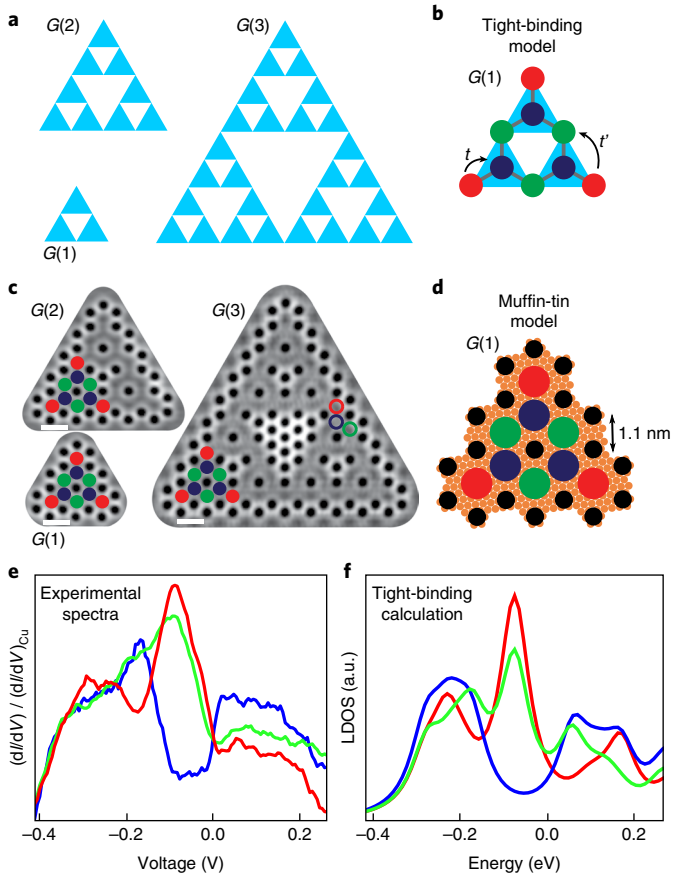

Fig. 1 | Geometry of the Sierpiński triangle fractal. a, Schematic of Sierpiński triangles of the first three generations $G(1)$–$G(3)$. $G(1)$ is an equilateral triangle subdivided into four identical triangles, from which the centre triangle is removed. Three $G(1)$ ($G(2)$) triangles are combined to form a $G(2)$ ($G(3)$) triangle. **b**, Geometry of a $G(1)$ Sierpiński triangle with red, green and blue atomic sites. t and t' indicate nearest-neighbour and next-nearest-neighbour hopping between the sites in the tight-binding model. **c**, Constant-current STM images of the realized $G(1)$–$G(3)$ Sierpiński triangles. The atomic sites of one $G(1)$ building block are indicated as a guide to the eye. Imaging parameters: $I = 1$ nA, $V = 1$ V for $G(1)$ and $G(2)$ and 0.30 V for $G(3)$. Scale bar, 2 nm. **d**, The configuration of CO molecules (black) on Cu(111) to confine the surface-state electrons to the atomic sites of the Sierpiński triangle. **e**, Normalized differential conductance spectra acquired above the positions of red, blue and green open circles in **c** (and equivalent positions). **f**, LDOS at the same positions, simulated using a tight-binding model with $t = 0.12$ eV, $t' = 0.01$ eV and an overlap $s = 0.2$. a.u., arbitrary units.

Figure 2 shows experimental wavefunction maps obtained at different bias voltages and a comparison with simulations using a tight-binding and muffin-tin model. In a thought experiment, we will discuss how electrons can be transported across the set-up between a source and a drain at arbitrary positions. At a bias voltage of −0.325 V, the red (R), green (G) and blue (B) sites all have a high LDOS, and this also holds between the sites. Hence, from a chemical perspective, this wavefunction has strong bonding character, yielding an excellent conductivity from source to drain along (R–B–G–B–R)-pathways. At $V = -0.2$ V, the red sites that connect the $G(1)$ triangles have a low amplitude: the wavefunction of the $G(3)$ triangle partitions into nine parts, each corresponding to a $G(1)$ triangle. The self-similar Sierpiński geometry thus leads to a subdivision of a fully bonding wavefunction delocalized over the $G(3)$ Sierpiński triangle at −0.325 V in self-similar $G(1)$ parts at −0.2 V, demonstrating self-similar properties of the LDOS itself. At the latter bias voltage, the conductivity along (R–B–G–B–R)-pathways suffers from the lower amplitude on the red sites (except the red corner sites). At $V = -0.1$ V, the LDOS shows a marked minimum on the blue sites and a peak at the green and red sites. From the tight-binding calculation, we find that the wavefunction has nodes on the blue sites, corresponding to a non-bonding molecular orbital from a chemical perspective. It is clear that the conductivity along the (R–B–G–B–R)-pathway mediated by nearest-neighbour hopping has vanished, and that electrons have to perform next-nearest-neighbour hopping between the red and green sites to propagate. These results connect with the theoretically calculated transmission of a Sierpiński carpet on a hexagonal lattice, which exhibits a gap in the conductivity although there is a high DOS in the system[23]. Finally, at $V = +0.1$ V, all blue sites in the $G(3)$ Sierpiński structure have a high amplitude, whereas the red and green sites exhibit a low amplitude. Again, the conductivity between source and drain is suppressed. We note that the LDOS maps of the three generations $G(1)$–$G(3)$ show the same features (see Supplementary Information), which is a consequence of the self-similarity of the geometry. We study this scale-invariance of the wavefunction in more detail with the box-counting method.

To determine whether the electronic wavefunctions inside the Sierpiński structure inherit the scaling properties of the Sierpiński geometry, we determine the fractal dimension of the wavefunction maps at different energies. We calculate the box-counting dimension (also called the Minkowski–Bouligand dimension) for both the experimental and simulated muffin-tin LDOS maps using $D = \lim_{r \to 0} \frac{\log N(r)}{\log(1/r)}$, with N the number of circles required to cover the contributing LDOS and r the radius of these circles. In this procedure, N is counted for various r, and subsequently the fractal dimension is given by the slope of the log–log plot for $N(r)$. The method is presented in Fig. 3a, and more details are given in the Supplementary Information. Figure 3b shows the box-counting dimension obtained experimentally (dark orange) and theoretically (light orange) for the wavefunction maps acquired at different energies (see, for example, Fig. 2). For comparison, we also show the dimension obtained from the wavefunction maps of a square lattice (dark and light blue, for the experiment and theory, respectively), realized in the same way and measured in the same energy window[3]. The difference between the experimental and simulated maps is ascribed to a more gradual contrast in the simulation, where also contributions of the tip density of states do not play a role. Fluctuations in the calculated dimension at higher energy occur due to the nodes that appear in the maps at higher energies. These nodes are not included as a part of the fractal set (LDOS amplitude is below the counting threshold) and therefore the calculated dimension is affected as the energy is increased. It can be clearly seen that the box-counting dimension of the Sierpiński triangle is close to the theoretical Hausdorff dimension 1.58 (orange solid line), whereas the square lattice has a dimension close to 2 (blue solid line). From these results, we conclude that the wavefunctions inherit the fractal dimension and therefore the scaling properties of the geometry to which they are confined, and that this dimension can be non-integer.

Finally, we show how the self-similarity of the wavefunction maps is reflected in momentum space. The Fourier-transformed wavefunction map at $V = -325$ mV (Fig. 4a) exhibits distinct maxima at $k = 1.9\,\mathrm{nm}^{-1}$ (turquoise), $k = 1.0\,\mathrm{nm}^{-1}$ (red) and $k = 0.5\,\mathrm{nm}^{-1}$ (yellow). These maxima correspond to the next-nearest-neighbour distances between the artificial atomic sites (see Fig. 1), the side of a $G(1)$ triangle, and the side of a $G(2)$ triangle in real space, respectively. We then transform parts of the Fourier map back into real space (Fig. 4b–d). The data inside the turquoise circle recover the full $G(3)$ Sierpiński triangle, as shown in Fig. 4b. Transforming the values inside the red

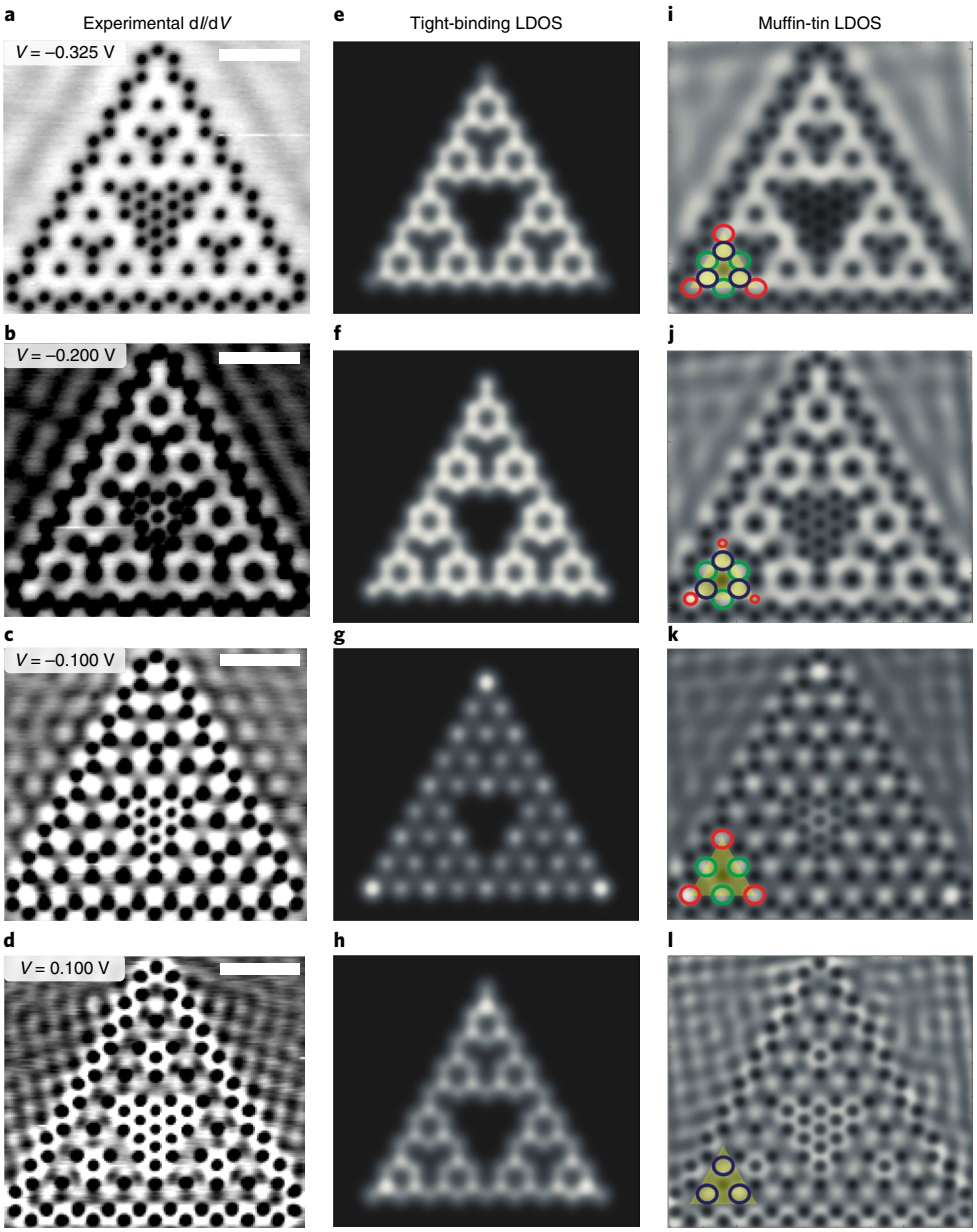

Fig. 2 | Wavefunction mapping. a–d, Differential conductance maps acquired above a $G(3)$ Sierpiński triangle at bias voltages −0.325 V, −0.200 V, −0.100 V and +0.100 V. Scale bar: 5 nm. **e–h**, LDOS maps at these energies calculated using the tight-binding model. **i–l**, LDOS maps simulated using the muffin-tin approximation. As a guide to the eye, a $G(1)$ building block is indicated, in which a larger radius of the circles corresponds to a larger LDOS at an atomic site, whereas no circle indicates a node in the LDOS.

circle, however, results in a Sierpiński triangle of generation 2, while the size is retained (see Fig. 4c). Analogously, transforming the data inside the yellow circle yield a first-generation Sierpiński triangle (Fig. 4d). This shows that the $G(3)$ wavefunction contains Fourier terms of the previous generations. The self-similar features of the Sierpiński triangle are thus inherently encoded in momentum space.

We have demonstrated a rational concept of building electronic wavefunctions with a fractional dimension from artificial atomic sites that couple in a controlled way. We discussed the wavefunctions that form by coupling the *s*-orbitals of artificial atoms in the single-electron regime. Although this study represents the simplest case, it already exhibits several aspects of fractal confinement. The emergent fractionalization of the wavefunction at the single-particle level has profound implications and opens a series of interesting questions for future investigation: Do electrons in $D = 1.58$ behave like Luttinger liquids? Do they exhibit the fractional quantum Hall effect in the presence of a strong perpendicular magnetic field, or is the behaviour hybrid between 1D and 2D? How does charge fractionalization manifest when the wavefunction is itself already fractional? Recent theoretical work already addresses parts of these questions and corroborates the potential of electrons in fractal lattices, showing that the Sierpiński carpet and gasket host topologically protected states in the presence of a perpendicular magnetic field[31]. Furthermore, the design of artificial-atom quantum dots coupled in a fractal geometry can also be implemented in semiconductor technology, thus making it possible to perform spectroscopy and transport experiments under controlled electron density. This would form a versatile platform to explore fractal electronics with several internal degrees of freedom, such as orbital type, Coulomb and spin–orbit interactions, as well as external electric and magnetic fields.

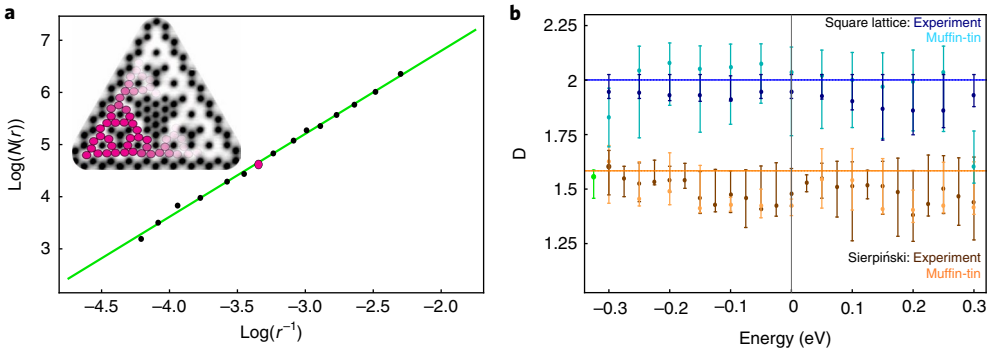

Fig. 3 | Fractal dimension of the Sierpiński wavefunction maps. a, The box-counting dimension of the wavefunction map acquired at $V = -0.325$ V is obtained from the slope of log(N) versus log(r^{-1}). The magenta dot indicates the radius r of the N circles used in the inset. Inset: Schematic of the box-counting method, where N circles with radius r cover the contributing experimental LDOS above the threshold of 45% at $V = -0.325$ V (see Supplementary Information for the determination of this threshold). **b**, Determination of the fractal dimensions of the LDOS of the $G(3)$ Sierpiński triangle (orange) and comparison with the 2D square lattice from ref. [3] (blue) for the experimental (dark) and muffin-tin (light) wavefunction maps. The solid lines indicate the geometric Sierpiński Hausdorff dimension ($D = 1.58$) and that of the square lattice ($D = 2$). The error bars represent the maximum of the error in determining the fractal dimension at different LDOS thresholds, which is between 45% and 65% (60% and 90%) for experiment (muffin-tin) and the error in determining the slope of the log–log plot as seen in **a**. The green result at −0.325 V is obtained from the slope in **a**. The fluctuations in the dimension are caused by nodes in the LDOS maps at different energies.

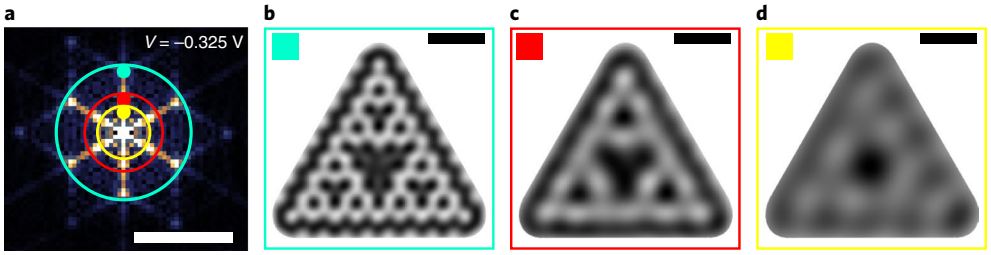

Fig. 4 | Fourier analysis of wavefunction maps. a, Fourier transform of the experimental differential conductance map at −0.325 V. The k-values outside the circles are excluded from the Fourier-filtered images in **b**–**d**. Scale bar: $k = 3$ nm^{-1}. **b**–**d**, Wavefunction map at −0.325 V after Fourier filtering, including merely the k-values within the turquoise (**b**), red (**c**) and yellow (**d**) circles indicated in **a**. Scale bar: 5 nm.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, statements of data availability and associated accession codes are available at https://doi.org/10.1038/s41567-018-0327-1

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Author contributions

S.N.K. did the calculations under the supervision of C.M.S. The experiments were performed by M.R.S. with contributions from S.E.F. and S.J.M.Z. under the supervision of I.S. and D.V. All authors contributed to the interpretation of the data and to the manuscript.

Additional information

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Methods

Scanning tunnelling microscope experiments. The scanning tunnelling microscopy and spectroscopy experiments were performed in a Scienta Omicron LT-STM system at a temperature of 4.5 K and a base pressure around 10^{-10}–10^{-9} mbar. A clean Cu(111) crystal, prepared by multiple cycles of Ar^+ sputtering and annealing, was cooled down in the scanning tunnelling microscope head. Carbon monoxide was leaked into the chamber at $p \approx 3.10^{-8}$ mbar for 3 min and adsorbed at the cold Cu(111) surface. A Cu-coated tungsten tip was used for both the assembly and the characterization of the fractal. The CO manipulation was performed in feedback at $I = 60$ nA and $V = 50$ mV, comparable to previously reported values[32,33], and was partly automated using an in-house-developed program. Scanning tunnelling microscopy was performed in constant-current mode. A standard lock-in amplifier was used to acquire differential conductance spectra ($f = 973$ Hz, modulation amplitude 5 mV r.m.s.) and maps ($f = 273$ Hz, modulation amplitude 10 mV r.m.s.) in constant-height mode. The Fourier analyses were performed using the software Gwyddion.

Tight-binding calculations. The atomic sites in the first three generations of the Sierpiński triangle[34] are modelled as *s*-orbitals, for which electron hopping between nearest-neighbour and next-nearest-neighbour sites is defined. The parameters used are $e_s = -0.1$ eV for the on-site energy, $t = 0.12$ eV for the nearest-neighbour hopping and $t'/t = 0.08$ for the next-nearest-neighbour hopping, similar to previously reported values[2]. Furthermore, we included an overlap integral $s = 0.2$ between nearest-neighbours and solved the generalized eigenvalue equation $H|\psi\rangle = E\mathcal{S}|\psi\rangle$, where $\mathcal{S}$ is the overlap-integral matrix. The LDOS is calculated at each specific atomic site and a Lorentzian energy-level broadening of $\Gamma = 0.8$ eV is included to account for bulk scattering. For the simulation of the LDOS maps, the same energy-level broadening was used and the LDOS at each site was multiplied with a Gaussian wavefunction of width $\sigma = 0.65a$, where $a = 1.1$ nm is the distance between two neighbouring sites.

Muffin-tin calculations. The surface-state electrons of Cu(111) are considered to form a 2D electron gas confined between the CO molecules, which are modelled as filled circles with a repulsive potential of 0.9 eV and radius $R = 0.55a/2$. The Schrödinger equation is solved for this particular potential landscape, and a Lorentzian broadening of $\Gamma = 0.8$ eV is used to account for the bulk scattering.

Box-counting method. The Minkowski–Bouligand[35] or box-counting method is a useful tool to determine the fractal dimension of a certain image, but has to be handled with care. In particular, as has been shown previously[36], the size of the boxes needs to be chosen within a certain radius. More specifically, the largest box should not be more than 25% of the entire image and the smallest box is chosen to be the point at which the slope starts to deviate from the linear regime in the $\log(N)$ versus $\log(1/r)$ plot. Redundant features such as the background Friedel oscillations were removed by applying a mask. Furthermore, the wavefunction maps are not binary, and therefore it is necessary to specify the threshold value above which the pixels are part of the fractal set. The threshold is a certain percentage of the maximum amplitude of the wavefunction map at a specific energy. The error introduced by the choice of the threshold is accounted for by performing the calculation procedure for several threshold percentages: between 45% and 65% for the experimental wavefunction maps, and 60%, 75% and 90% for the top, centre and bottom of the error bar for the simulated (muffin-tin) LDOS maps (see Supplementary Information for the differences in the LDOS for these thresholds). The differences in threshold between experiment and simulation arise because the maps from the simulation are more pronounced than the experimental ones. In addition, due to the dependence on the tip, the experimental maps cannot always be directly compared to each other (see Supplementary Information). Another error sets in by determining the slope of the log–log plot and specifying which radii are taken into account. In Fig. 3b, the error bars therefore show the maximum value of these two independent errors.

Data availability

All data is available from the corresponding authors upon reasonable request. The experimental data can be accessed using open-source tools.

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