Two-dimensional electron gases (2DEGs) at transition-metal oxide (TMO) interfaces, and boundary states in topological insulators, are being intensively investigated. The former system harbors superconductivity, large magneto-resistance, and ferromagnetism. In the latter, honeycomb-lattice geometry plus bulk spin-orbit interactions lead to topologically protected spin-polarized bands. 2DEGs in TMOs with a honeycomb-like structure could yield new states of matter, but they had not been experimentally realized, yet. We successfully created a 2DEG at the (111) surface of KTaO₃, a strong insulator with large spin-orbit coupling. Its confined states form a network of weakly-dispersing electronic gutters with 6-fold symmetry, a topology novel to all known oxide-based 2DEGs. If those pertain to just one Ta-(111) bilayer, model calculations predict that it can be a topological metal. Our findings demonstrate that completely new electronic states, with symmetries not realized in the bulk, can be tailored in oxide surfaces, promising for TMO-based devices.
direction. Furthermore, by analyzing the symmetries of this electronic structure, we demonstrate that confining this novel 2DEG to a bilayer of Ta atoms, hence a honeycomb lattice, will lead to a topological metal.

**Results**

**Basic considerations: structure of KTaO₃ along the [111] direction.** Figure 1(a) presents the crystal structure of KTaO₃. Along the [111] direction, the system has 3-fold symmetry, as any cubic system, and consists of alternating layers of K and TaO₃. There are three different Ta-(111) layers per unit cell, labelled Ta-I, Ta-II and Ta-III. Figure 1(b) shows the Ta atoms in the 3D structure seen from the [111] direction. Arrows represent the lattice vectors of a 2D unit cell in the (111) plane, which corresponds to a centered hexagonal network with 6-fold symmetry. When only two layers are considered (Ta-I and Ta-II, for example), the lattice formed by nearest neighbors is a honeycomb lattice (thick black lines), similar to the bilayer structure proposed in Ref. 20. As we shall see later, our ARPES data are consistent with the formation of a 2DEG with the periodicity and the symmetry of an unreconstructed (111) plane.

**Experimental electronic structure.** Figure 2(a) shows the Fermi surface (color map) of the 2DEG measured at the (111) surface of KTaO₃, pictured in Fig. 2(b). Experimental details of the extraction of this Fermi surface are given in the Supplementary Material. The black hexagons in figure 2(a) represent the Brillouin zone of the unreconstructed (111) surface. Note that the observed Fermi surface has a well defined 6-fold symmetry and a periodicity corresponding to that of the unreconstructed KTO-(111) layers. By Bloch theorem, we infer that the 2DEG is formed by itinerant conduction electrons experiencing an in-plane potential compatible with unreconstructed (111) layers. In turn, this strongly suggests that, while one might expect that the highly-polar KTO-(111) surface reconstructs, the observed 2DEG is formed by electrons confined in unreconstructed subsurface layers.

More specifically, the Fermi surface of Fig. 2(a) consists of 6 branches extending out from Γ to the six M points. From the area enclosed by this Fermi surface, we obtain a carrier density \( n_{2D} \sim 10^{14} \text{ cm}^{-2} \). The observation of such a large Fermi surface fully supports the quasi-2D (i.e., confined) character of the electron gas. Indeed, if the Fermi surface was a cross-section of an hypothetical 3D Fermi surface, the ensuing electron density would be, by simple scaling, \( n_{2D} \sim n_{3D}^{1/3} \sim 10^{23} \text{ cm}^{-2} \). For such density, only one order of magnitude smaller than that of pure gold, the sample would be highly conducting and its aspect would be mirror-like, in stark contrast with the transparent character of the bulk crystal shown in figure 2(b), indicative of an insulating state.

Different energy-momentum intensity maps across the Fermi sea provide valuable complementary information about the 2DEG at the (111) surface of KTO. Figures 2(c, d) show the dispersion along the (110) direction, dashed green line in figure 2(a), in the form of, respectively, intensity map and stack of momentum distribution curves (MDCs). This dispersion indicates that the Fermi surface branches are formed by electron-like bands coming close to each other near Γ. The effective mass of these electron-like bands, deduced from a parabolic fit (red dashed lines), is \( m_{110}^* \approx 0.3m_e \) (\( m_e \) is the free electron mass). Furthermore, as shown by the stack of energy distribution curves (EDCs) in figure 2(e), the bottom of each of these electron bands is essentially non-dispersive along the ΓM direction. Accordingly, the Fermi surface forms a star with six open branches, each branch being composed of two quasi-parallel sheets. In other words, the electronic structure of the 2DEG at the (111) surface of KTO consists of a network of weakly dispersing “electron gutters”.

Since we observe only the first subband (\( E_F \)) of the 2DEG, we can only estimate an upper limit for the extension \( L \) of the 2DEG. We make the reasonable assumptions that \( E_1 \approx -V_0 \), where \( V_0 \) is the depth of the confining potential well, and that the next quantum level (\( E_2 \)) of the same nature as the measured subband lies at an energy \( E_2 - E_1 \approx V_0 \) at or above \( E_F \). The result, detailed in the Supplementary Material, is \( L < 16 \text{ Å} \), or equivalently, \( L < 7 \text{ Ta-layers along (111)} \).

Note, from figures 2(c–e), that there is an intense background below \( E = -260 \text{ meV} \) trailing behind the bands forming the 2DEG. The most likely origin of this background is inelastic scattering of the electrons off impurities or disorder. This can be either in-plane scattering, which will broaden the spectral lines, or scattering of the electron off impurities or disorder. This can be either in-plane scattering, which will broaden the spectral lines, or scattering of the electron off impurities or disorder. This can be either in-plane scattering, which will broaden the spectral lines, or scattering of the electron off impurities or disorder. This can be either in-plane scattering, which will broaden the spectral lines, or scattering of the electron off impurities or disorder. This can be either in-plane scattering, which will broaden the spectral lines, or scattering of the electron off impurities or disorder.

Figure 1 | KTaO₃ crystal in real space: schematics of the structure along the [111] direction. (a) Crystal structure of KTaO₃. Dashed triangles indicate the three Ta-(111) layers in the unit cell, labeled Ta-I, Ta-II and Ta-III. Ta atoms lying in the same (111) plane have the same shading color (green, blue or red). Note that Ta atoms in consecutive Ta-(111) layers are nearest neighbors. (b) View along the [111] direction of the Ta atoms alone. Lines connecting Ta atoms represent the edges of the 3D unit cell. Thick black lines connect nearest neighbors in layers Ta-I and Ta-II, forming a honeycomb lattice. Curved double arrows represent electron hopping between nearest neighbors. Straight black arrows are the lattice vectors of a 2D unit cell in the (111) plane. Such 2D unit cell is independent of the number of Ta-layers considered.
behind the broadened parabolic quasi-particle peak. As this background is extrinsic to the electronic structure, it has no effect on the quasi-particle spectral function, in agreement with the observation, noted earlier, that the experimental effective mass of the 2DEG’s band is essentially the same as the one given by non-correlated tight-binding calculations (Supplementary Material and Ref. 13).

Discussion
The observed electronic structure can be understood in terms of electron hopping between neighboring Ta layers. To see how, we modeled the 2DEG using a tight binding (TB) hamiltonian on a Ta-(111) bilayer with the hopping parameters determined from our previous study of KTO-(001)\textsuperscript{13}—see details in the Supplementary Material.
2D metal topological 2DEG at the KTO (111) surface would correspond to a salient feature of the observed Fermi surface. They can be intuitively understood from the directional overlaps between $t_{2g}$ orbitals, while dashed green and violet lines illustrate the channels created by the $d_{xy}$ orbitals, respectively.

Figure 3 | Comparison of experimental and tight-binding Fermi surfaces of the 2DEG at the (111) surface of KTaO$_3$. (a) Experimental Fermi momenta from the original data after symmetrization (blue open circles) compared to the Fermi surface from a two-layer tight-binding calculation (red lines). The Fermi momenta were determined from the peak positions of the MDCs at $E_F$, using Lorenzian fits. The inner quasi-circular Fermi surface found in the calculations, of radius $\sim 0.15$ Å$^{-1}$, is difficult to compare with the data, due to the experimental resolution. (b) Directional paths, in real space, created by the different $t_{2g}$ orbitals, $d_{xy}$, $d_{xz}$, and $d_{yz}$ on a (111) bilayer of Ta atoms. The orange solid lines illustrate hopping channels created by the $d_{xy}$ orbitals, while dashed green and violet lines illustrate the channels created by the $d_{xz}$ and $d_{yz}$ orbitals, respectively.

Material. Indeed, as schematically indicated by the double arrows in figure 1(b), and in Ref. 20, the largest hopping occurs between the Ta atoms of consecutive (111) layers, as they correspond to nearest neighbors along [001] in the cubic 3D lattice. In Figure 3(a) we compare the experimental ARPES Fermi surface to the calculated Fermi surface. We observe that the model correctly gives the bandstructure of the weakly dispersing electron gutters. The comparison between the data and calculations close to the $\Gamma$ point is more difficult due to the experimental resolution. We checked that including extra Ta layers or considering the hopping of electrons between Ta atoms on the same (111) layer, which are next-nearest neighbors, will mainly change the computed band-structure around the $\Gamma$ point, but not the structure of the gutters, so that the comparison above remains valid.

The weakly dispersing gutters along the $\Gamma M \Gamma$ lines are the most salient feature of the observed Fermi surface. They can be intuitively understood from the directional overlaps between $t_{2g}$ orbitals in neighboring Ta sites, which give rise to large nearest-neighbor hopping amplitudes along the [001] (and equivalent) directions. When seen from the [111] direction, these appear as directional zig-zag chains at 120$^\circ$ from each other, as depicted in Fig. 3(b) for two consecutive Ta (111) layers. Each set of parallel hopping chains on the 2D lattice will lead to open weakly dispersing Fermi sheets in reciprocal space, i.e., to the observed gutters.

Our results demonstrate that a 2DEG with hexagonal symmetry can be easily created at the (111) surface of KTO. As an outlook for near-future investigations, an exciting feature arises when such a 2DEG is confined to a Ta-(111) bilayer, which forms a honeycomb lattice –as seen from the lattice formed by layers Ta-I and Ta-II in figure 1(b). In this case, following Ref. 27, one can demonstrate (details in the Supplementary Material) that the electron wave-function of the ground state has an odd parity at one of the $\bar{M}$ points (and its time-reversal partner), but an even parity at all other points, implying a non trivial topological index $Z_2 = 1$. In this case, the 2DEG at the KTO (111) surface would correspond to a topological 2D metal, similar to some of the non-trivial states predicted in Ref. 20, with the 2DEG itself corresponding to the “bulk” states, and the non-trivial states appearing at the 1D edges of the sample.

From a broader perspective, our results show that different surface orientations can be used to tailor Fermi seas and to radically change the microscopic state, hence the physical properties, of oxide perovskite-based 2DEGs. Of particular interest is the prospect for engineering 2DEGs of strongly correlated insulators on honeycomb lattice-structures, which could lead to correlated topological and edge-states with new physical properties not shown by semiconducting topological insulators. Thus, in perspective, our results provide some initial bridges between the fields of topological matter, correlated electrons and oxide electronics.

Methods

The angle resolved photoemission spectroscopy (ARPES) experiments were done at the Synchrotron Radiation Center (SRC, University of Wisconsin, Madison) and at the Synchrotron SOLEIL (France), using linearly polarized photons in the energy range 20–100 eV and Scienta R4000 detectors with vertical slits (henceforth the $k_y$ direction). The momentum and energy resolutions were 0.25$^\circ$ and 15 meV, respectively. The mean diameter of the incident photon beam was smaller than 50 µm (SOLEIL) and about 150 µm (SRC).

The samples studied were undoped transparent high-grade laser crystals of KTaO$_3$ (SurfaceNet, GmbH), of purity 99.9995%, grown by a modified top seeded solution growth (TSSG) using KO$_2$ as solvent. The estimated amount of oxygen vacancies is less than 10 ppm, being mostly concentrated in the seeding area, which was not used for the final cut crystals. Similarly, the estimated amount of defects is less than 100 cm$^{-3}$.

The crystals were mounted with [110] $k_y$ and fractured in-situ along the (111) surface at 25 K (SRC) and 10 K (SOLEIL), in pressure lower than $6 \times 10^{-11}$ Torr. The fractured surfaces had homogeneous electron photo-emittance over a large area. The results have been reproduced in 6 different cleaves.

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**Author contributions**

Project conception: A.F.S.-S.; ARPES measurements: C.B., T.R., F.F., F.B. and A.F.S.-S.; infrastructure for ARPES experiments at Soleil: F.B., A.T.I. and P.L.F.; sample characterizations: M.B., A.B., T.M. and P.L.; data analysis, interpretation, calculations: C.B., A.F.S.-S., M.G., O.H.C. and M.J.R.; writing of the manuscript: A.F.S.-S., with input from C.B., M.G. and M.J.R. All authors discussed extensively the results and the manuscript.

**Additional information**

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