Emergent Magnetism and Intrinsic Anomalous Hall Effect in KTaO$_3$ Two-Dimensional Electron Gases

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There has been intense recent interest in the two-dimensional electron gases (2DEGs) that form at the surfaces and interfaces of KTaO$_3$ (KTO), with the discovery of superconductivity at temperatures significantly higher than those of similar 2DEGs based on SrTiO$_3$ (STO). Here we demonstrate that KTO 2DEGs fabricated under conditions that suppress the superconductivity show emergent magnetism at low temperatures that can be tuned by a gate voltage, $V_g$. The signature of this magnetism is a zero-field transverse voltage that increases in magnitude with decreasing $V_g$, that is strongly asymmetric with respect to an applied perpendicular magnetic field, and that becomes hysteretic in field as the temperature is lowered. We argue that it is due to a finite Berry curvature of the KTO Ta bands in the presence of time-reversal symmetry breaking associated with ordered local moments, with unique features associated with the complex spin texture introduced by spin-orbit interactions in KTO.
Two-dimensional electron gases (2DEGs) in complex oxide heterostructures have seen a great deal of interest in the past two decades due to the variety of phenomena they exhibit. The most studied of these 2DEGs are those formed at the surface of SrTiO$_3$ (STO) (1–3). The phenomena seen in STO-based heterostructures include gate-tunable conductivity (4), superconductivity (5), spin-orbit interactions (6), spin-polarized transport (7, 8), quantum interference effects (9) and magnetism (10), among others. In the past few years, interest has turned to 2DEGs based on KTaO$_3$ (KTO). Bulk KTO is similar to STO in that it is a band insulator with a large gap (≈3.5 eV) (11), and it has a large dielectric constant that grows with decreasing temperature but saturates at low temperatures (12), signaling a transition to a quantum paraelectric phase (11). One significant difference between KTO and STO is the strength of the spin-orbit interactions, which are more than an order of magnitude larger than in STO (13).

Like STO, heterostructures of KTO and other materials such as LaAlO$_3$ (LAO) (14), LaTiO$_3$ (LTO) (15), EuO (16), or AlO$_x$ (17) have been shown to support a 2DEG whose properties can be tuned by a gate voltage $V_g$ (18) (19) (20). In particular, superconductivity was discovered recently in KTO based 2DEGs at temperatures significantly higher than those of STO based 2DEGs. Surprisingly, the highest transition temperatures $T_c \sim 2.2$ K have been found in KTO 2DEGs on (111) (14) oriented substrates, while 2DEGs on (110) substrates have $T_c \sim 1$ K (21), and those on (001) substrates show no superconducting transition or very low transition temperatures ($T_c \sim 50$ mK) (14) (22). This is in sharp contrast with STO based heterostructures, where superconductivity was first discovered in (001) oriented 2DEGs (5). Superconductivity is also found in (110) and (111) oriented STO heterostrucutres for some overlay materials such as LAO (23) (24), while it is absent for other materials like (La$_{0.3}$Sr$_{0.7}$)(Al$_{0.65}$Ta$_{0.35}$)O$_3$(LSAT) (25).

Another feature of STO based 2DEGs is the presence of magnetism, which was found to coexist with superconductivity (10) (26) (27). One manifestation of this intrinsic magnetism is
a hysteresis in the longitudinal and transverse magnetoresistance (MR) that is observed at low temperatures (10). The origin of this magnetism in STO based heterostructures is still not clear, although there has been some speculation that is associated with localized moments on Ti$^{3+}$ sites (28). Signatures of magnetism have also been reported in KTO based heterostructures, but these signatures are associated with the presence of known magnetic materials such EuO used as overlayers (16). Here we report longitudinal and transverse MR measurements on (001), (110) and (111) oriented AlO$_x$/KTO heterostructures fabricated under conditions that suppress superconductivity down to our lowest measurement temperatures. In particular, the transverse resistance shows remarkable behavior. It is finite even in zero applied magnetic field, the zero-field resistance growing in magnitude as $V_g$ and temperature $T$ are reduced; it is smallest in general for the (001) oriented samples, larger in the (110) oriented samples, and largest in the (111) oriented devices. This trend maps on to measurements of the transverse MR, which becomes increasingly asymmetric as $V_g$ is reduced, being most prominent again in the (111) oriented samples. Finally, at millikelvin temperatures, the MR becomes hysteretic, indicating the presence of long-range magnetic order in the devices. Density functional theory (DFT) calculations suggest that the transverse resistance is due to a charge Hall contribution associated with the finite Berry curvature of the KTO bands in the presence of time-reversal symmetry breaking arising from localized magnetic moments in the KTO.

The devices in this study were Hall bars with length 600 $\mu$m and width 50 $\mu$m patterned by photolithography followed by successive deposition and oxidation of thin layers of Al (details of the fabrication process can be found in Methods). Figure 1a shows a schematic of a Hall bar. Prior to spinning photoresist, the substrates were cleaned using standard cleaning procedures, but were not annealed at elevated temperatures. These devices do not show a superconducting transition down to our lowest measurement temperatures (see Supplementary Materials). To our knowledge, all KTO based devices that have shown superconductivity have been annealed
at elevated temperatures, either explicitly or as part of the sample growth process (19) (14) (20). In our case, substrates that were annealed but otherwise processed identically do go superconducting (differences between annealed and unannealed substrates are discussed in more detail in the Supplementary Materials). We focus here on measurements of the longitudinal and transverse low-frequency differential resistance (Fig. 1a) as a function of \( V_g \), \( T \) and applied external field \( H \). Details of the measurement can be found in Methods.

Figure 1b shows the longitudinal resistance \( R_L \) for \( H = 0 \) at \( T = 2.2 \) K as a function of \( V_g \) for Hall bars on (001), (110) and (111) oriented surfaces. \( R_L \) shows the same general trend for all three orientations: it increases with decreasing \( V_g \) and shows hysteresis with respect to \( V_g \). Similar gate voltage dependent behavior has been reported in KTO-based heterostructures (29), and in STO-based heterostructures (30). In contrast, the transverse resistance of the KTO heterostructures shows unusual behavior. Figure 1c shows the simultaneously measured transverse resistance \( R_T \) for the three crystal orientations as a function of \( V_g \) in zero external field. At \( V_g = 200 \) V, \( R_T \sim 10 \) Ω for the (001) and (110) oriented Hall bars. However, the (111) oriented Hall bar shows a much larger \( R_T \) at \( V_g \sim 200 \) V. More striking is the behavior of \( R_T \) as \( V_g \) is decreased. For all 3 orientations, \( |R_T| \) increases in magnitude as \( V_g \) is decreased, growing to a significant fraction of \( R_L \). In particular, there is a sharp increase in \( |R_T| \) for \( V_g \sim 100 \) V. This increase is most prominent in the (111) sample, but also present in the (001) and (110) samples. There is no corresponding feature in \( R_L \), suggesting the effect does not arise from a longitudinal contribution. A similar finite \( R_T \) is also observed in STO devices fabricated under similar conditions (31), but was smaller by an order of magnitude.

We now discuss the transverse and longitudinal MR of the devices. While anomalous behavior is seen in both \( R_T(H) \) and \( R_L(H) \), we focus on \( R_T(H) \) as it is easier to separate the anomalous contributions from the contribution due to the ordinary Hall effect. The hysteresis as a function of \( V_g \) shown in Fig. 1 is associated with glassiness in the system, and leads to
a logarithmic relaxation of the measured resistance over a span of many hours or longer when any external parameter is changed. To minimize the hysteresis, all the measurements reported here were taken after electrically "annealing" the sample by cycling $V_g$ repeatedly between 50 and 200 V. Even after such annealing, the resistances of the devices drift as a function of time. Consequently, the MR data shown below have been corrected for this drift as first described by Biscaris et al. for STO based devices (32). This correction is described in more detail in the Supplementary Materials.

Figure 2(a-c) shows $R_T$ for the (001), (110) and (001) oriented devices as a function of field $H_\perp$ applied perpendicular to the sample at different values of $V_g$ for $T \sim 2.2$ K (corresponding data for $\sim 5$ K are shown in the Supplementary). In order to be able to show data for all values of $V_g$ on the same graph, the respective zero-field value of $R_T$, which varies over a wide range as shown in Fig. 1, has been subtracted from each curve. At large $V_g$, $R_T(H_\perp)$ for all three orientations is linear, with a slope corresponding to an electron density of $n \sim 4 - 8 \times 10^{13}/\text{cm}^2$ based on a fit to a single band model, comparable to densities obtained by other groups (19) (14). As $V_g$ is decreased, $R_T(H_\perp)$ becomes increasingly asymmetric in $H_\perp$, with the slope of $R_T(H_\perp)$ for $H_\perp > 0$ becoming different from the slope for $H_\perp < 0$. In addition, the (001) and (111) devices show a change in curvature of $R_T(H_\perp)$ that occurs around $V_g \sim 100$ V (data on a finer voltage scale around this value of $V_g$ is shown in Supplementary Materials).

The asymmetry in the $R_T(H_\perp)$ data shown in Figs. 2(a-c) arises from an anomalous component that is symmetric in $H_\perp$. This can be seen if we decompose the data of Figs. 2(a-c) into components that are symmetric and antisymmetric in $H_\perp$, as shown in Figs. 2(d-i). The antisymmetric components of all three orientations are as expected for the conventional Hall effect contribution, with the slope of the (110) and (111) devices about twice that of the (001) device. All three orientations also show a symmetric component, but this varies in amplitude by orders of magnitude: it is a few ohms for the (001) device, a few tens of ohms for the (110)
device, and a few hundred ohms for the (111) device. Additionally, this symmetric component changes sign at low gate voltages, an effect which is qualitatively reproducible in subsequent experimental runs.

Even more striking are the data taken at millikelvin temperatures. Figure 3(a-c) shows $R_T(H_\perp)$ for all three orientations taken at $\sim 30$ mK. In addition to the asymmetry in $R_T(H_\perp)$, all three orientations show hysteretic behavior at lower gate voltages, taking the form of peaks in $R_T(H_\perp)$ at $\sim \pm 50$ mT, although this field value is slightly different for the different orientations. It is important to note that due to the large negative offset in $R_T$ demonstrated in Figure 1A, the hysteretic peaks represent a decrease in the magnitude of $R_T$. This hysteresis is most prominent in the (001) and (111) oriented devices; smaller but still evident in the (110) device. The hysteresis disappears for the (001) and (110) devices at larger $V_g$, where $R_T(H_\perp)$ is almost linear; it remains but decreases in amplitude for the (111) device up to $V_g = 200$ V. The (111) device also shows small, reproducible oscillations as a function of $H_\perp$ at larger fields. The hysteresis in the MR persists up to $T \sim 1.8$ K, while the oscillations disappear at around 800 mK (Supplementary Materials). Similar hysteresis is also observed in the longitudinal MR.

Hysteretic behavior in the MR is usually associated with long range ferromagnetic order in the system that interacts with the conduction electrons, with the fields at which the hysteresis occurs corresponding to the coercive field of the ferromagnet. To demonstrate that the hysteresis we observe does indeed arise from magnetic order, Fig. 3D shows the longitudinal MR of the (001) sample in perpendicular field at $V_g = 80$ V, with progressively larger values of an in-plane field $H_\parallel$ applied. It can be seen that by $H_\parallel \sim 100$ mT, the hysteresis in $H_\perp$ is completely suppressed, suggesting that once the ferromagnetic moments are fully aligned in plane, they no longer can give rise to hysteresis in the MR.

The presence of ferromagnetic order in our samples is surprising, as in contrast to the EuO/KTO samples studied by other groups where hysteretic behavior has been reported (16),
there is no intrinsically magnetic component in our samples. In the case of STO, it was proposed that Ti$^{3+}$ ions, one electron in an otherwise unoccupied $d$ orbital, could form local moments (28). Density functional theory (DFT) calculations show that in the presence of oxygen vacancies, Ta$^{4+}$ ions can similarly form local moments that give rise to a surface magnetism.

It is well known that such localized moments can give rise to anisotropic magnetoresistance (AMR) (33) and an anomalous Hall effect (AHE) (34). In conventional ferromagnets, for example, this leads to a contribution to the Hall effect that is proportional to the magnetization (AHE), and a contribution to the longitudinal MR that depends on the angle between the magnetization $M$ and the current (AMR). More recently, it has also been recognized that there is an intrinsic contribution to the AHE that arises from a Berry phase that develops in the presence of time reversal symmetry-breaking (34), such as would be introduced by surface magnetism of Ta$^{4+}$ ions. DFT calculations show that one does expect an anomalous Hall effect due to the intrinsic Berry phase contribution, which is modified due to the ferromagnetic ordering at lower temperatures. The signatures of these effects, however, are modified by the complex spin-orbit interactions in KTO.

In spin-orbit materials with simple band structure, the spin-orbit interaction locks the orientation of the spin $\sigma$ of the electron and its momentum $k$ (35). A similar situation occurs in KTO, except that the spin-orbit interaction depends strongly on the direction of $k$. For the (001) and (110) orientations, $\sigma$ remains in the plane of the 2DEG, while for the (111) orientation, $\sigma$ cant out of the plane, as has been noted by Bruno et al (13). The addition of oxygen vacancies accentuates this effect, as shown in Fig. 4. For the (111) oriented supercell shown in Fig. 4A, an added oxygen vacancy results in a change in charge density and net spin polarization shown in Fig. 4B and Fig. 4C. Similar results were obtained for the (001) oriented crystal and are discussed in Supplementary Materials. Additionally, DFT calculations of the Berry curvature of the lowest band give a finite anomalous Hall contribution for all orientations, consistent with
our observation of a large, zero-field transverse resistance at lower values of $V_g$ (Supplementary Materials).

At lower temperatures where the local magnetic moments order, the strong $k$ dependence of the spin-orbit interaction gives rise to a hysteretic AHE and AMR that depends on the crystal orientation, unlike the corresponding effects in conventional ferromagnets where they depend only on the angle between the magnetization (controlled by an external field) and the current. This is demonstrated experimentally in Figure 5, which shows the transverse resistance of the (001), (110) and (111) samples in an external magnetic field $H_{||}$ in the 2D plane at millikelvin temperatures. In contrast to the corresponding data in perpendicular field, here the (001) device shows almost no MR and no hysteretic behavior, while the (110) device, which showed a very small hysteretic response in perpendicular field, now shows a robust hysteretic MR. The (111) sample continues to show a hysteretic response, as might be expected from its complex spin-texture.

The transport signatures of these samples suggest a magnetic ordering despite a lack of intrinsically magnetic materials used in fabrication. We have shown that the magnetism is instead due to local moments on the Ta$^{4+}$ ions brought about by the same oxygen vacancies that host the 2DEG. These moments give rise to hysteretic behaviors below $T \sim 1.8$ K most prominently shown in Hall traces, which can tuned with applied gate voltage. These behaviors can be suppressed with the application of a parallel field, and coexist with both background oscillations in the (111) transverse traces and with a sizable transverse voltage that arises below $V_g = 100$ V. Further study is necessary to see if the magnetism coexists with the superconductivity observed in samples fabricated with similar processes that also rely on oxygen vacancies to generate the conducting gas.
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**Supplementary materials**

Materials and Methods

Supplementary Text
Figs. S1 to S9

References (36–41)
Figure 1: Sample schematic and back gate voltage annealing curves. A Representative diagram of the sample in a Hall bar geometry, where the back gate voltage, $V_g$, is applied between the conducting gas and the back of the KTaO$_3$ substrate. The current direction, the perpendicular applied magnetic field, and the contacts used for the longitudinal resistance $R_L$ and the transverse resistance $R_T$ are shown. B Semi-log plot of differential longitudinal resistance $R_L$ for each of the three crystal terminations vs $V_g$, measured during annealing sweeps conducted at 2.2K, in zero magnetic field. C Simultaneously measured differential transverse resistance $R_T$ during the same gate voltage anneal.
Figure 2: Hall curves as a function of gate voltage and crystalline orientation, measured at 2.2K. The first row shows transverse resistance, $R_T$ as a function of gate voltage for the A (001), B (110) and C (111) oriented samples respectively, for a field $\mu_0 H_\perp$ applied normal to the plane of the Hall Bar. D-F show the symmetric contribution of $R_T$ extracted from A-C for the same crystal orientations, demonstrating $V_g$-dependence and sign change of the low field peak. G-I show the antisymmetric contribution of $R_T$ extracted from A-C, from which carrier concentrations were extracted (see Supplementary Materials).
Figure 3: Hall curves and longitudinal magnetoresistance at $\sim 30$ mK. A Transverse Hall resistance, $R_T$, of the (001) sample as a function of gate voltage, demonstrating the growth of the hysteretic features and deviation from the expected linear curve. Data have been systematically shifted in $R_T$ for clarity. B Similar data for the (110) sample with even more pronounced low field features. C $R_T$ of the (111) sample, demonstrating hysteretic behavior, small background oscillations, and a large deviation from linear field dependence which changes dramatically with $V_g$. D The hysteretic peaks in the (001) sample at 80 $V_g$ are suppressed as a function of applied magnetic field $\mu_0 H_\parallel$ applied parallel to the plane of the Hall Bar.
Figure 4: **Charge and net spin polarization of a (111) KTO surface with an oxygen vacancy.**  
A The crystal structure of the supercell slab used to calculate the DFT results before the formation of an oxygen vacancy.  
B The change in spatial charge distribution at the surface when a single oxygen vacancy is created.  
C The resulting net spin polarization at the surface after the oxygen vacancy.
Figure 5: Parallel field Hall curves at $\sim30\text{mK}$ and $V_g = 200\text{V}$. A shows a very small planar Hall effect of the (001) sample while B shows a larger background planar Hall for the (110) sample with the emergence of hysteretic peaks. C shows the response of the (111) sample, with both hysteretic peaks at low fields and background oscillations at larger fields.
Supplemental Materials:
Emergent Magnetism and Intrinsic Anomalous Hall Effect in KTaO$_3$ Two-Dimensional Electron Gases

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Methods and Materials

Materials fabrication The KTaO$_3$ substrates used in this study were 5 mm x 5 mm x 0.5 mm single side polished crystals purchased from MSE Supplies LLC. For the samples described in the main text, the substrates were subjected to a normal cleaning regimen of 3 min ultrasonication in acetone, 3 min ultrasonication in DI water and 3 min ultrasonication in isopropanol before being spin coated with LOR-5A and S-1813 photoresists. The Hall bar patterns were exposed using a Suss MJB4 mask aligner before being developed in MF-319 developer. All three crystals were simultaneously metallized with 99.9995% aluminum in an in-house deposition chamber via electron-gun evaporation. Before deposition the surface was cleaned with 100 mTorr of oxygen plasma for one minute to remove residue from the lithography process. Deposition consisted of repeated steps of 1.5 nm aluminum deposition followed by a dwell period of 10 min. After the second and subsequent steps a partial pressure of oxygen was introduce into the chamber to ensure oxidation of the aluminum layers to amorphous aluminum oxide. Samples thus defined were then all wirebonded to a sample header for simultaneous measurement.
and cooled down in an Oxford MX100 crystostat equipped with a two-axis superconducting magnet.

**Measurement Description** Measurements were conducted using a low frequency AC lock-in technique with two PAR 124A lock-in amplifiers and an EGG 7260 digital lock-in amplifier. 100 nA of current was passed along the Hall bars sourced from home built current sources, and the resulting voltages were measured with similarly home built AD624 based pre-amplifier before being read by the lock-ins. The back gate voltage, $V_g$, was supplied by a Keithley KT2400 source equipped with a low pass filter and measured independently by a HP34401A digital multimeter.

**Density Functional Theory Details** All first principles calculations are based on density functional theory and were performed using the Quantum ESPRESSO software package (36). The calculations utilized the generalized gradient approximations (GGA) of Perdew-Burke-Ernzerhoff (PBE)(37). The lattice parameters and atomic positions for the calculations are taken from the Materials Project (38). Spin-orbit coupling is included in the calculations. In order to calculate the surface spectrum and spin-orbit texture, a Wannier tight-binding model is generated from the $p_{x,y,z}$ orbitals of the O atoms and the $d_{xy,yz,zx}$ orbitals of Ta using the Wannier90 software package (39). The surface spectra and spin-orbit texture is then calculated using the WannierTools software package(40). To examine the presence of emergent surface magnetism, leading to a finite zero field charge Hall effect, supercells were constructed for the (001) and (111) terminated crystals. Further details of the calculations can be found in Supplementary Text, under the Computational Methods Section.
Supplementary Text

Experimental Results

Temperature Dependence Resistance Resistance measurements as a function of temperature were taken for the reported samples after an initial gate voltage anneal cycle at $\sim 1.6K$. These results are reported in Figure S1 A, showing zero backgate voltage $R_L(T)$ for the two measured ranges from the cooldown steps on the dilution refrigerator. The resistance rises slightly from a minimum around 5K, before dipping or saturating at the lowest measured temperatures. Importantly, none of the three samples show superconducting transitions down to our lowest measurement temperature of 25mK.

Superconducting Sample Preparation To demonstrate that the lack of annealing is the main factor in the suppression of the superconductivity for the samples described in the main text, additional samples were made with all of the same processes, but with an added annealing step before lithography. This additional annealing step consisted of a 650C bake for two hours in atmosphere before ultrasonication in DI water for two more hours as described by Tomar et al. (41). Subsequent fabrication steps were identical to those described in Methods. The resulting samples were measured on a different cryostat, looking for a superconducting transition, which is shown for the (111) terminated samples in Figure S1 B. The transition was measured on three different Hall bars all on the same (111) crystal substrate, but measured simultaneously. The data shown are for initial cooldown, before any annealing sweeps, with 100V backgate applied, as prescribed by other superconducting papers.

Drift Correction As mentioned in the main text, it is well known that complex oxide samples drift over time whenever changes are made to significant experimental parameters such as back gate voltage. This drift is persistent over the course of hours to days, so is accounted for in these
Figure S1: **Comparison of the two sample preparations.** A Sheet resistance of the three crystal terminations of the samples described in the main text after annealing sweeps of $V_g$. B Superconducting transition for a (111) sample fabricated with a 650C annealing step and deionized water soak to prepare the KTO surface.

reported data by subtracting off a time dependent logarithmic form as described by Biscaras et al. In Figure S2 A we show a trace presented as measured, with the resistance drift on top of the changes from magnetoresistance, taken over the course of several hours. Figure S2 B shows the corrected data after subtracting off the time dependent contribution described by Biscaras et al. of the form $R(t) = R_0 + A \log(t + C)$. Here we fit with the constants $R_0 = 1040$, $A = 250$, and $C = 10000$, where in this case $R_0$ is not the residual resistance but the shift necessary to recover the appropriate resistance at the start of the run, which is important for calculating percent magnetoresistance. The results shown in Figure S2 B are then interpolated along 751 evenly spaced values of magnetic field, and averaged together into one representative trace, as seen in Figure S2 C. A zoomed in view of the center, showing the preservation of the hysteresis can be seen in Figure S3. All longitudinal traces were subjected to this treatment, each with their own fitting parameters. Drift was also seen in the transverse magnetoresistance, but only for the lowest gate voltages, and these data were removed from the reported results.
Figure S2: Demonstration of drift correction procedure for a longitudinal magnetoresistance trace taken at $\sim 30\text{mK}$ and $V_g = 200\text{V}$. A shows raw data demonstrating a time dependent drift of magnetoresistance after a change in gate voltage. B drift corrected data showing good agreement after multiple traces. C shows the resulting trace after the repeated sweeps were interpolated and averaged together.

because no reproducible Hall curve could be extracted.

**Carrier Concentration** Carrier concentration and carrier mobility were calculated from the slope of the antisymmetric part of the Hall resistance $R_H = \Delta R/\Delta B$. Assuming a single band model gives the Hall coefficient as $R_H = -1/ne$, where $e$ is the charge of the electron and $n$ is the areal density of the two dimensional conducting gas. The sign of the Hall coefficient, $R_H$, gives the sign of the carrier, which was confirmed to be electron-like. Our calculated carrier concentrations as a function of gate voltage are given in Figure S4 for each of the crystal terminations at measurement temperatures of $\sim 2.2\text{K}$ and $\sim 5\text{K}$. Both sets of results rely on an assumed single band model and break down at low gate voltages due to competing effects that lower the precision of the measurement. Importantly, these calculated carrier concentrations agree well with other works that explored the nature of the superconductivity in these gases (19) (14), suggesting that the suppression of the superconductivity is not due to a deviation in the density of the 2DEG. From these data we also can extract carrier mobility, which ranges from a few tens of cm$^2$ V$^{-1}$ s$^{-1}$ to a few hundred, also in line with what other groups have
Temperature Dependence of Hysteretic Magnetoresistance  A detailed study of the samples’ temperature dependent magnetoresistance was conducted to trace the evolution of the hysteresis with increasing temperature, and at a fixed gate voltage of $V_g = 200\, V$. These data revealed the temperature dependence of the three components of the magnetic transport signatures in the samples, as demonstrated in Figures S5. Figure S5 A shows the shifted Hall traces for the (111) sample, which has two critical temperatures, one for the background oscillations, which are suppressed by $\sim 800\, \text{mK}$, and one for the low field hysteresis, which is suppressed above $\sim 1.7\, \text{K}$. These data have been shifted from their measured values as there is a large, field-independent intrinsic Hall contribution which is subtracted off. They are also spread out in even intervals of resistance for clarity. The subtracted intrinsic Hall shift is plotted as a function of temperature in Figure S5 B. The measured data were limited by the available temperature range of the probe, but show a saturation as we approach the other reported results at $\sim 2.2\, \text{K}$.  

Figure S3: Demonstration of low external magnetic field drift correction procedure. A shows drift corrected data showing good agreement after multiple traces, while B shows the resulting smoothed data, demonstrating the fidelity of the procedure.
Figure S4: Gate voltage dependent carrier concentration. A-C show the extracted carrier concentrations measured at ∼2.2K for the (001), (110), and (111) terminated samples respectively. D-F show the same carrier concentrations but measured at ∼5K.
Figure S5: Temperature dependence of the low temperature Hall signatures. A shows the temperature dependence of the shifted Hall resistance for the (111) terminated sample from our base temperature of $\sim 30 \text{mK}$ to the highest stable temperature of $\sim 1.7 \text{K}$. B shows the temperature dependent intrinsic anomalous Hall offset of the (111), described as the average measured resistance of the Hall curves along the field range shown in A.

**Computational Methods**

**Surface 2DEG and Spin Texture** KTaO$_3$ belongs to spacegroup 221 with lattice parameter $a = 4.03 \ \text{Å}$. The lattice parameters and atomic positions are taken from the Materials Project(38). As described in Methods and Materials, all first principles calculations are based on density functional theory and were performed using the Quantum ESPRESSO software package(36). The calculations utilize the generalized gradient approximations (GGA) of Perdew-Burke-Ernzerhoff (PBE)(37). For bulk calculations, a plane-wave cutoff of 60 Ry is used and the primitive unit cell is sampled with a Monkhorst k-mesh of $9 \times 9 \times 9$. Spin-orbit coupling is included in the calculations. The calculated band structure resulting from a bulk unit cell is shown in Fig. S6.

In order to calculate the surface spectrum and spin-orbit texture, a Wannier tight-binding model is generated from the $p_{x,y,z}$ orbitals of the O atoms and the $d_{xy,yz,xz}$ orbitals of Ta using the Wannier90 software package(39). Following Ref. (13), the 2DEG at the surface can be
modeled through the introduction of a potential well at the surface in order to avoid the explicit introduction of symmetry breaking terms. The magnitude of this potential well is the only parameter tuned in the calculations and is fitted such that the results are in alignment with those of Ref. (13). The surface spectra and spin-orbit texture is then calculated using the WannierTools software package(40). The results of the surface spectra calculations for the three crystal terminations are shown in Fig. S7. Our calculations indicated a maximum Rashba coefficient, $\alpha_R \approx 2$ meVÅ, for the (111) and (110) surfaces, in line with what is found by Bruno et al (13) and $\alpha_R \approx 1$ meVÅ for the (001) surface. Similarly, calculations of the spin texture reveal an out of plane contribution for the (111) surface which is absent in the (001) and (110) samples, as shown in Fig. S8. These results also agree with those published by Bruno et al (13).

**Emergent Magnetism: The (001) Surface** To examine the presence of emergent surface magnetism, leading to a finite zero field charge Hall effect, a supercell is constructed of size $2 \times 1 \times 6$. The (001) surface is perpendicular to the $c$-axis of the supercell and a minimum of 20
Å of vacuum is added along the c-direction. We first focus on the (001) surface as it displays a zero field charge Hall conductivity while limiting computational expense. The terminations of the supercell are made symmetric. Multiple relaxations are performed, one for each oxygen vacancy that can be formed at the top layer. The relaxation calculations are performed using a plane-wave cutoff of 60 Ry, a charge cutoff of 550 Ry and the cell is sampled with a Monkhorst k-mesh of $2 \times 4 \times 1$, magnetization is considered in relaxation calculations and the bottom three layers are held fixed. Following relaxation the lowest energy structure is selected for a self consistent calculation. The self-consistent calculation is performed utilizing a Monkhorst k-mesh of $4 \times 8 \times 1$ followed by a non-self consistent calculation with a Monkhorst k-mesh of $8 \times 16 \times 1$. Following this, the density of states was calculated to determine spin-polarization and charge distribution. The simulation estimates a total magnetic moment of $0.56$ Bohr magneton for the full supercell with two surface Ta atoms. This result is consistent with the relative magnetude of the experimental results for the (001) sample. The process was repeated with no
oxygen vacancies to ensure the system converged to a non-magnetic state.

The partial charges of each atom were then compared for the systems with and without the charge vacancy. The atom which displays most significant change in partial charge is the Ta atom closest to the oxygen vacancy on the surface with a change in charge of approximately $0.5e$. This is visible in the density plot shown in Fig. (9(a)).

**Emergent Magnetism: The (111) Surface**  To study the emergent surface magnetism on the (111) surface, we construct a supercell with the $c$-axis along the [111] direction. It is further constructed such that the surface terminations are equivalent and there exists two Ta atoms on the surface. Again 20 Å of vacuum is added along the $c$-direction to limit finite size effects. The supercell is visible in Fig. 4A in the Main Text. The relaxation procedure listed above for a single oxygen vacancy on the surface is repeated using identical energy cutoffs and k-point grids. The simulation estimates a total magnetic moment of 6.67 Bohr magneton for the full supercell with two surface Ta atoms. Importantly, the total magnetic moment has increased
Figure S9: (a) Difference in charge density on the surface for the supercell with an oxygen vacancy at the center ($\rho_{\text{vacancy}}$) and without ($\rho_{\text{normal}}$). The dashed box outlines the location of the Ta atom nearest to the vacancy. The location of the oxygen vacancy at the center is evident by the increased values in that region. (b) The charge density inside the dashed box shown in (a). (c) Spin polarization on the surface of the supercell with an oxygen vacancy. Any significant contribution to spin-polarization is localized to the Ta atoms.
significantly from the 001 surface. We should note that the supercell used to model the (111) surface has 71 atoms as opposed to 35 atoms for the 001 surface, however, this still represents a significant qualitative increase and is in accordance with an increased zero-field charge Hall conductivity on the (111) surface. The process was repeated with no oxygen vacancies to ensure the system converged to a non-magnetic state.

The partial charges of each atom were then compared for the systems with and without the charge vacancy. The atom which displays most significant change in partial charge is the Ta atom on the surface, closest to the oxygen vacancy with a change in charge of approximately 0.5\(e\). This is visible in the density plot shown in Fig. 4 B.