Interwining quantum order and non-trivial topology is at the frontier of condensed matter physics\(^1\text{-}^4\). A charge-density-wave-like order with orbital currents has been proposed for achieving the quantum anomalous Hall effect\(^5\text{-}^6\) in topological materials and for the hidden phase in cuprate high-temperature superconductors\(^7\text{-}^8\). However, the experimental realization of such an order is challenging. Here we use high-resolution scanning tunnelling microscopy to discover an unconventional chiral charge order in a kagome material, KV\(_3\)Sb\(_5\), with both a topological band structure and a superconducting ground state. Through both topography and spectroscopic imaging, we observe a robust 2 \(\times\) 2 superlattice. Spectroscopically, an energy gap opens at the Fermi level, across which the 2 \(\times\) 2 charge modulation exhibits an intensity reversal in real space, signalling charge ordering. At the impurity-pinning-free region, the strength of intrinsic charge modulations further exhibits chiral anisotropy with unusual magnetic field response. Theoretical analysis of our experiments suggests a tantalizing unconventional chiral charge density wave in the frustrated kagome lattice, which can not only lead to a large anomalous Hall effect with orbital magnetism, but also be a precursor of unconventional superconductivity.

The interdependence of geometry, correlations and topology is pivotal to many vexing questions of condensed matter. Advances in pushing this frontier forward directly contribute to our fundamental understanding of quantum matter and the application of quantum materials such as in quantum information science and energy relevant technology. Owing to the unusual lattice geometry, electrons in kagome lattice systems can experience a non-trivial energy spectrum and experimental mysteries suggest a striking yet unknown order phase. Here we perform STM experiments on KV\(_3\)Sb\(_5\), at 4.2 K to observe a CDW order with an unusual magnetic field response.

KV\(_3\)Sb\(_5\) has a layered structure with the stacking of a K\(_1\) hexagonal layer, Sb\(_1\) honeycomb lattice, V\(_3\)Sb\(_1\) kagome lattice and Sb\(_2\) honeycomb lattice. The structure consists of a K\(_1\) hexagonal layer, Sb\(_1\) honeycomb lattice, V\(_3\)Sb\(_1\) kagome lattice and Sb\(_2\) honeycomb lattice. The measured step height from topographic data is 2.3 Å, which is close to the bulk structural distance between the K and Sb layers. According to the crystalline symmetry, at a monolayer atomic step edge between the K and Sb layers, the upper step will be K while the lower is the Sb layer. Owing to the bonding length and geometry, the V and Sb layers can have stronger chemical bonding, and the material tends to cleave between the K and Sb layers. According to the crystalline symmetry, a monolayer atomic step edge between the K and Sb layers, the upper step will be K while the lower is the Sb layer. Owing to the bonding length and geometry, the V and Sb layers can have stronger chemical bonding, and the material tends to cleave between the K and Sb layers. According to the crystalline symmetry, a monolayer atomic step edge between the K and Sb layers, the upper step will be K while the lower is the Sb layer. Owing to the bonding length and geometry, the V and Sb layers can have stronger chemical bonding, and the material tends to cleave between the K and Sb layers.

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that have hexagonal and honeycomb surfaces. To study the nature of the 2×2 modulation, we focus on the Sb surface, which has a strong bonding with a kagome lattice and features large defect-free areas. We measure these areas at 4.2 K (Fig. 1d) and 80 K (Fig. 1d), which is just above the critical temperature of the speculated electronic ordering. It is clear that the 2×2 modulation disappears at 80 K. Through the Fourier transform of the Sb topographic data, we further visualize the existence of 2×2 modulation vector peaks at 4.2 K in Fig. 1f and their disappearance at 80 K in Fig. 1g.

For an electronic order, the states in the vicinity of the Fermi level involved in its formation can produce an energy gap, such as in the classic Peierls mechanism for CDW order. In our dI/dV data (I is the tunnelling current and V is the bias voltage), measuring the local density of states, we observe a gap-like feature on the Sb surface in Fig. 2a. This energy gap extends from −23 meV to +29 meV, with additional shoulders around ±10 meV, which could be due to the multi-orbital, multi-Fermi-surface nature of the material or anisotropy of the underlying order parameter. Moreover, we find this gap also disappears at 80 K in our measurement, attesting to a close relationship with the observed 2×2 modulation. Our first-principles calculation reveals that the low-energy states are from V 3d orbitals. Furthermore, the formation of V hexamers and trimers in the kagome lattice (as illustrated in the inset of Fig. 2b) with slightly reduced bond lengths than the original ones can reduce the total energy of the system by 32 meV, rendering it a promising candidate for the 2×2 superlattice modulation. Our calculation of the local density of states of the energy-optimized 2×2 superlattice structure also reveals an energy gap (Fig. 2b) around the Fermi level, which is of the same order of magnitude as the experimentally observed value. We further perturb the gap spectra by applying a magnetic field along the c axis up to 6 T, but we do not detect a strong field response of the gap structure (Fig. 2c). This observation is also consistent with a CDW gap.

For a CDW gap, it is also expected that across the energy gap, there should be an intensity reversal of the charge modulation. In the classic Peierls CDW scenario, negative voltage bias shows enhanced intensity over charge accumulation regions, whereas images of the same atomic area at positive bias show enhanced intensity over charge depleted regions. The dI/dV imaging in Fig. 2d,e shows that the maximum charge intensity at −30 meV turns to the lowest charge intensity at +30 meV despite the additional complexity of the modulation patterns. This observation thus demonstrates a type of charge intensity reversal. The charge modulation vector peaks are sharply evident in the Fourier transform of the spectroscopic imaging in Fig. 2f. This modulation vector (2×2) is non-dispersive within our momentum resolution, as revealed by the energy-resolved Fourier transform of the spectroscopic imaging in Fig. 2g. The non-dispersive feature demonstrates a static electronic order. The topography and spectroscopic imaging taken together strongly support a CDW order.

A further inspection of our high-resolution charge modulation vector peaks in the low-energy spectroscopic data reveals pronounced intensity anisotropy along different directions, as shown in Fig. 3a. We take the data at a surface defect-free region to study the intrinsic behaviour of CDW, as defects, particularly those inducing standing waves, can backscatter electrons and pin (the phase of) the CDW order. The observed anisotropy can be due to a chiral CDW order as observed and discussed in certain transition-metal dichalcogenides and high-temperature superconductors. The chirality can be defined as the counting direction (clockwise or anticlockwise) from the lowest to highest vector peaks. In the TiSe system, the chirality of the order can be manipulated by an optical field, which does not break time-reversal symmetry. In the current case, we find the chirality at the same atomic area can be switched by the magnetic field applied along the c axis for opposite directions, as shown in Fig. 3b,c. We have repeatedly observed the field switching effect at low energies (relevant to the CDW gap) for different samples. Data taken at certain high energies do not show the switching effect. It is possible that this is due to additional contributions in this multi-orbital material, which deserves future study. The magnetic field switching effect suggests a time-reversal symmetry breaking of the CDW order, which is also supported by our recent muon spin spectroscopy measurement. Under similar magnetic fields, muon spin spectroscopy measurement reveals a sharp enhancement of...
magnetic response below the charge ordering temperature, which will be published elsewhere.

We try to understand the origin of this unconventional CDW order. The low-energy band structure of KV$_3$Sb$_5$ consists of three nearly independent features (Fig. 4a): a quasi-two-dimensional electron pocket around the $\Gamma$ point formed by $p_z$ orbitals from Sb; a band exhibiting a van Hove singularity at the M point, formed by the $d_{xy}$ orbitals of V; and a pair of Dirac-cone-like bands near the M point, formed by the $d_{xz}/d_{yz}$ orbitals of V. The band at van Hove filling stemming from V $d_{xz}/d_{yz}$ orbitals can be most vital to the formation of the charge ordering.

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**Fig. 2 | Charge modulation observed via spectroscopic imaging.** a. Spatially averaged $dI/dV$ spectra for Sb surface taken at 4.2 and 80 K. The data is averaged over a defect-free area of 10 nm x 10 nm. b. First-principles calculation of the bulk local density of states considering the 2 x 2 superlattice modulation. The inset illustrates the charge order in the underlying V-based kagome lattice based on first-principles calculation, which forms hexamers and trimers (dark lines). c. Magnetic field perturbation of the $dI/dV$ spectra showing no detectable response. d,e. Atomically resolved $dI/dV$ imaging for the same Sb surface at energy $E = -30$ meV and $E = +30$ meV, respectively. The inset shows the underlying ordered kagome lattice inferred from the simultaneously obtained topographic image. f. The inset is the Fourier transform of the $dI/dV$ imaging at $-30$ meV, showing both charge order and lattice Bragg peaks. The main panel shows the intensity distribution along the Bragg peak direction. a = 5.5 Å is the in-plane lattice constant. g. Energy dependence of the charge order vector and lattice Bragg peaks, showing the non-dispersive nature of the charge order vector.

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**Fig. 3 | Magnetic response of the chiral charge order.** a-c. Spectroscopic 2 x 2 vector peaks taken at magnetic field $B = 0$ T, $+2$ T and $-2$ T, respectively. Data are taken on defect-free regions. The images are Fourier transforms of spectroscopic maps acquired on an Sb surface that is 30 nm x 30 nm in size at 10 mV. A circular region of the full Fourier-transformed image is shown for clarity, highlighting the six 2 x 2 vector peaks. The top and bottom panels are three-dimensional and two-dimensional presentations of the data. The chirality can be defined as the counting direction (clockwise or anticlockwise) from the lowest to highest pair vector peaks.
of CDW order, since the vector of the $2 \times 2$ superlattice charge modulation connects the van Hove singularities at M points and matches with the Fermiology of the $d_x^2-d_y^2$ band $^{33-35}$. Moreover, the kagome lattice at van Hove filling exhibits nested Fermi level eigenstates with unequal predominant sublattice occupancy $^{33}$ (Fig. 4a). Based on this sublattice interference mechanism $^{34}$, the predicted instability is a CDW order with relative angular momentum $^{35}$, which is consistent with a chiral charge order. Accordingly, we consider a triplet of chiral CDW order parameters $^{35} \Delta_{\sigma_1}, \Delta_{\sigma_2}$ and $\Delta_{\sigma_3}$ where $\Delta_{\sigma_i}, n = 1, 2$ and 3, are complex numbers whose absolute value approximately corresponds to the observed peak heights in Fig. 3 (Fig. 4a for more details). The observed magnetic field response suggests that the time-reversal symmetry is broken by the CDW order parameters. This is typically achieved if degenerate order parameter components acquire a complex relative phase. Under reversal of time, such a phase changes sign, and thus, if it is not 0 or $\pi$, the state breaks time-reversal symmetry. One of the natural phase choices for the triplet order parameters is then $\arg(\Delta_{\sigma_i}) = 2\pi n/3$. Applying this experimentally motivated choice of order parameters to a kagome model for the V $d_{xy}$ bands yields the energy gap and chiral charge ordering pattern as shown in Fig. 4b.

We further discuss the implications of the unconventional chiral CDW in light of the topological fermions and superconducting ground state of the system. As the Dirac bands of the V $d_{xy}$/$d_{yz}$ orbitals are also near M points, such an unconventional CDW order will open a topological energy gap $^{36}$ at the Dirac cones, thereby introducing a large Berry curvature (Fig. 4c-d). As a consequence, the system would exhibit an anomalous Hall effect. A $k_z$ (vertical momentum) integrated model estimation detailed in the Supplementary Information gives rise to a non-zero Berry curvature, the integration of which produces a giant anomalous Hall effect (right panel). The right panel plots the anomalous Hall conductance $\sigma_y$ as a function of energy $E$ at $k_z = 0$. $\sigma_y$ has a unit of $e^2/h$, where $e$ is the elemental charge and $h$ is Planck’s constant.

**Fig. 4 | Impact of time-reversal broken chiral charge order on the electronic structure.** a, Schematic of Fermi surfaces in the hexagonal Brillouin zone at $k_z = 0$. The wave vectors of the unconventional CDW are indicated, as well as the location at which Dirac nodal lines slightly above the Fermi energy cut the $k_z = 0$ plane. The V $d_{xy}$ Fermi surface, which is nested by the ordering wave vectors, has weight on distinct sublattices at each M point, giving rise to the sublattice interference mechanism. b, Two-dimensional model calculation of the impact of unconventional chiral CDW on the V $d_{xy}$ bands. The orange (blue) density of states is without (with) the CDW order parameter $\Delta$, which splits the van Hove singularity. The inset image shows the chiral charge pattern and associated orbital currents. The shaded area in the inset marks the 2 unit cell. c, Effective band structure of the nodal lines formed by the $d_{xy}/d_{yz}$ pattern and associated orbital currents. The shaded area in the inset marks the 2 unit cell. d, The introduction of a chiral time-reversal breaking CDW order parameter opens a topological gap around the Fermi level (left panel). This gap gives rise to a non-zero Berry curvature, the integration of which produces a giant anomalous Hall effect (right panel). The right panel plots the anomalous Hall conductance $\sigma_y$ as a function of energy $E$ at $k_z = 0$. $\sigma_y$ has a unit of $e^2/h$, where $e$ is the elemental charge and $h$ is Planck’s constant.
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Notes added in proof: With the acceptance of this work, we note that a similar chiral charge order is observed in RbV$_3$Sb$_5$ (ref. 38) and CsV$_3$Sb$_5$ (ref. 39), suggesting the ubiquitous chiral charge order in this family of kagome superconductor.

Online content
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Methods

Single crystals were grown with the same methods discussed ref. 23. Single crystals with size up to 2 mm × 2 mm were cleaved mechanically in situ at 77 K in ultra-high vacuum conditions, and then immediately inserted into the microscope head, already at 4He base temperature (4.2 K). More than 20 crystals were cleaved and studied in this research. For each cleaved crystal, we explore surface areas over 5 µm × 5 µm to search for atomic flat surfaces. Topographic images in this work were taken with the tunnelling junction set-up \( V = 100 \text{ mV} \) and \( I = 0.05 \text{ nA} \) for exploration of areas typically 400 nm × 400 nm. When we found atomically flat and defect-free areas, we took topographic images with the tunnelling junction set-up \( V = 100 \text{ mV} \) and \( I = 0.5 \text{ nA} \) to resolve the atomic lattice structure as demonstrated in the main paper. Tunnelling conductance spectra were obtained with an Ir/Pt tip using standard lock-in amplifier techniques with a lock-in frequency of 997 Hz and a junction set-up of \( V = 50 \text{ mV} \) and \( I = 0.5 \text{ nA} \), and a root mean square oscillation voltage of 0.3 mV. Tunnelling conductance maps were obtained with a junction set-up of \( V = 50 \text{ mV} \) and \( I = 0.5 \text{ nA} \), and a root mean square oscillation voltage of 5 mV. The magnetic field was applied with a zero-field cooling method. For field-dependent tunnelling conductance spectra, we ramped the field continuously from 0 T to 6 T with a 1 T per hour ramp rate, while simultaneously compensating the field-induced spatial drift of the tip position on the sample. For the field-dependent tunnelling conductance map, we first withdrew the tip away from the sample, and then slowly ramped the field to 2 T or −2 T. Then we reapproached the tip to the sample, found the same atomic area and then performed spectroscopic mapping at this magnetic field. In the main paper, we focus our study on clean regions away from impurities inducing standing waves.

Data availability

All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplementary Information. Additional data are available from the corresponding authors upon reasonable request.

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Author contributions

Y.-X.J., J.-X.Y. and N.S. conducted the STM experiments in consultation with M.Z.H.; B.R.O. and S.D.W. synthesized samples; B.R.O., J.R. and L.K. performed X-ray measurements; M.M.D., J.H., X.L., G.C., G.X., Z.W., R.T. and T.N. carried out the theoretical analysis in consultation with J.-X.Y. and M.Z.H.; Z.G., J.R., L.K., S.S.Z., I.B., Q.Z., M.S.H., T.A.C., D.M., M.L., Z.-J.C. and X.P.Y. contributed to the calibration of the measurement; Y.-X.J., J.-X.Y. and M.Z.H performed the data analysis and figure development and wrote the paper with contributions from all authors; M.Z.H. supervised the project. All authors discussed the results, interpretation and conclusion.

Competing interests

The authors declare no competing interests.

Additional information

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