Control of chiral orbital currents in a colossal magnetoresistance material

Yu Zhang1, Yifei Ni1, Hengdi Zhao1, Sami Hakani2, Feng Ye3, Lance DeLong4, Itamar Kimchi2,5* & Gang Cao1*

Colossal magnetoresistance (CMR) is an extraordinary enhancement of the electrical conductivity in the presence of a magnetic field. It is conventionally associated with a field-induced spin polarization that drastically reduces spin scattering and electric resistance. Ferrimagnetic Mn3Si2Te6 is an intriguing exception to this rule: it exhibits a seven-order-of-magnitude reduction in ab plane resistivity that occurs only when a magnetic polarization is avoided. Here, we report an exotic quantum state that is driven by ab plane chiral orbital currents (COC) flowing along edges of MnTe6 octahedra. The c axis orbital moments of ab plane COC couple to the ferrimagnetic Mn spins to drastically increase the ab plane conductivity (CMR) when an external magnetic field is aligned along the magnetic hard c axis. Consequently, COC-driven CMR is highly susceptible to small direct currents exceeding a critical threshold, and can induce a time-dependent, bistable switching that mimics a first-order ‘melting transition’ that is a hallmark of the COC state. The demonstrated current-control of COC-enabled CMR offers a new paradigm for quantum technologies.

A necessary characteristic of all known CMR materials is an alignment of magnetic spins that drastically reduces electron scattering and/or band gaps, and thus electric resistance. Mn3Si2Te6 is unique, in that the ab plane resistivity ρ is reduced by up to seven orders of magnitude only when a magnetic field, H, is applied along the magnetic hard c axis or when a saturated magnetic state is absent. By contrast, ρ decreases by no more than 20% when the magnetization is saturated by fields aligned with the magnetic easy a axis (or ab plane) (Fig. 1a,b). The data in Fig. 1a also show a large anisotropy field of at least 13 T, which is highly intriguing because magnetic anisotropy is a result of spin-orbit coupling, whereas the orbital angular momentum is zero for the Mn2+ (3d5) ion in Mn3Si2Te6. This new type of CMR has been confirmed, and signals the existence of a new quantum state yet to be identified and understood.

Herein we show that such a new state can be defined by its unconventional CMR, anisotropy and extraordinary response to application of small direct current (d.c.), I, including a first-order transition with bistable switching. The switching consists of an abrupt jump in voltage, V, which takes seconds or minutes to complete after application of a small current to the sample without further stimulus. The unusual time delay and voltage jump are strongly enhanced when H is oriented along the magnetic hard c axis (which favours CMR) but are otherwise absent. Application of H || c axis also induces a surprising d.c. tunneling behaviour that features ΔV/ΔI = 0. In short, the new state is highly conducting (CMR) and resilient when H || c axis, but is insulating and ‘melts’ (mimicking a solid-to-liquid phase transition) via a first-order, bistable switching when a small current exceeding a certain threshold is applied.

Taken separately, each key phenomenon (that is, CMR, anisotropy and bistable switching) defies conventional wisdom and models; the simultaneous occurrence of all these phenomena indicates that a new paradigm is required to understand the underlying state. Here, we argue all these exotic phenomena can be explained in terms of intra-unit-cell, ab plane COC that generate net c axis orbital magnetic moments (Mcoh) that couple with the ferrimagnetically ordered Mn spins (Mmn) (Fig. 1d,e).

Below the Curie temperature Tc = 78 K, the ab plane COC circulate along Te–Te edges of the MnTe6 octahedra, which produces net Mcoh oriented primarily along the c axis (Fig. 1d). The Mcoh arising from Te orbitals are necessarily coupled with Mmn, giving rise to an unusual spin-orbit effect that explains the observed large anisotropy field (Fig. 1b). Therefore, the observed magnetization results from both Mcoh and Mmn, and application of H || c axis can thereby amplify the ab plane COC and Mcoh that underpin the CMR. The COC state ψc is exceedingly sensitive to application of small I, and converts to a trivial state ψc via first-order, bistable switching when I > 2 mA. Coupling of the COC to the rigid-but-extended Te sublattice and Mmn allows ψc to remain metastable over long timescales set by the motion of Te atomic clusters (as opposed to only electrons/Mn spins, which respond on picosecond timescales). Consequently, the bistable switching or melting of ψc requires seconds or minutes to occur after application of a small I to the sample. Our observed T–I–H phase diagram (Fig. 1g) shows that ψc exists underneath a boundary sheet in T–I–H space that expands with increasing H || c axis.

COC have been investigated in previous studies of high-Tc cuprates, iridates, Kagome superconductors and magnets, a Kagome metal with a transport signature of loop current in a charge-ordered phase and moiré heterostructures. However, no macroscopic transport phenomena attributed to COC that coexist

1Department of Physics, University of Colorado at Boulder, Boulder, CO, USA. 2School of Physics, Georgia Institute of Technology, Atlanta, GA, USA. 3Neutron Scattering Division, Oak Ridge National Lab, Oak Ridge, TN, USA. 4Department of Physics and Astronomy, University of Kentucky, Lexington, KY, USA. 5E-mail: ikimchi3@gatech.edu; gang.cao@colorado.edu
with a long-range magnetic order (as observed in this work) have been reported. The signature of COC coupled with long-range magnetic order can be particularly subtle because mirror and time-reversal symmetries are already broken; nevertheless, effects of COC can be exotic and strong, as demonstrated in this work.

Mn₃Si₂Te₆ forms a trimer-honeycomb lattice (Fig. 1e) with a noncollinear magnetic structure in which the Mn spins lie predominantly along the a-b plane (Fig. 1f). Our recent neutron study indicates that the COC (Fig. 4g and Extended Data Fig. 1). The easy axis value, even for μ₀H > 0.05 T (red dashed line in Fig. 1b). By contrast, the c axis magnetization Mₖ cannot fully attain the a axis value, even for μ₀H > 13 T (blue dashed line in Fig. 1b). Such an unexpected anisotropy field suggests a new type of spin-orbit coupling is at play in Mn₃Si₂Te₆. Moreover, our measured average magnetic moments for Mn1 and Mn2 are 4.55 and 4.20 μB, respectively, which explain both the large magnitude and anisotropy of the CMR in Mn₃Si₂Te₆.

**Physical properties in magnetic fields**

The a axis resistivity ρₐ and the c axis resistivity ρₜ both rise rapidly below Tₐ (Extended Data Fig. 1a). The easy a axis magnetization Mₐ readily saturates to a value Mₛ = 1.56 μ₀Mn for μ₀H > 0.05 T (red dashed line in Fig. 1b). By contrast, the c axis magnetization Mₖ cannot fully attain the a axis value, even for μ₀H > 13 T (blue dashed line in Fig. 1b). Such an unexpected anisotropy field suggests a new type of spin-orbit coupling is at play in Mn₃Si₂Te₆. Moreover, our measured average magnetic moments for Mn1 and Mn2 are 4.55 and 4.20 μB, respectively, considerably smaller than 5.0 μB anticipated for Mn²⁺ (3d⁶)⁵. These data suggest a partial cancellation between Mₐ and Mₖ; accordingly, we infer Mₐ to be around 0.1 μB (Methods).

In addition, our magnetostriction data indicate that the a axis undergoes a significant expansion Δa/a > 0 when H||c axis but remains essentially unchanged when H||a axis (Fig. 1c); this indicates that a strong c axis magnetoelastic coupling favours an increase of orbital area associated with the COC (Extended Data Fig. 1b, c; Methods), as discussed below.
Properties with small currents

\(\rho_a\) at \(H = 0\) is reduced by up to six orders of magnitude when \(I\) is increased from 10 nA to 10 mA at low temperatures (Fig. 2a). \(T_C\) decreases rapidly with increasing \(I||a\) (Fig. 2b). Specifically, \(T_C\) decreases strongly from 83 K at 10 \(\mu\)A to 22 K at 1.8 mA, and eventually vanishes at 2 mA (Fig. 2b).

\(M_c\) behaves similarly with increasing \(I||a\), until magnetic order is suppressed abruptly for \(I > 2\) mA. The signature of \(T_C\) evolves into a sharper, first-order transition with increasing \(I\) until magnetic order is suppressed abruptly for \(I > 2\) mA. Note the magnitude of \(\rho_a\) in the vicinity of \(T_C\), remains essentially unchanged (Fig. 2b). This remarkable behaviour suggests small currents (<2 mA) barely affect spin scattering of electrons near \(T_C\), but weaken the spin exchange coupling that drives ferrimagnetic state.

Fig. 2 | Response of physical properties to d.c. currents and magnetic fields. a, \(a\)-axis resistivity \(\rho_a\) at various applied d.c. currents \(I\) for \(H = 0\). b, Enlarged plot of \(\rho_a\) in the outlined region in a. Dashed grey lines highlight the near-constant values of \(\rho_a\) at and just below \(T_C\). c, Temperature dependence of \(\rho_a\) at various d.c. currents \(I\) for \(\mu_o H = 14\) T. Note that the CMR is absent for \(I > 5\) mA and 10 mA. d, \(c\)-axis magnetization \(M_c\) at various d.c. currents \(I\) along the \(c\)-axis for \(\mu_o H = 0.5\) T. Note the first-order transition evident in the \(\rho_a\) and \(M_c\) data induced by \(I = 1.5\) and 1.8 mA. e, Change in \(\rho_a\) for \(\mu_o H = 14\) T and various values of applied \(I\). f, \(T_C\) at various values of \(I\) (generated from data in b and d). Note that both the CMR and \(T_C\) vanish for \(I > 2\) mA.

Fig. 3 | \(a\)-axis \(I-V\) characteristics. a, At various temperatures and \(H = 0\). b, Details of the outlined area in a; note the first-order transition at \(I_C\). c, \(I-V\) characteristics for \(T = 10\) K and \(H||c\) axis. d, Details of the outlined area in c. Note the regime where \(\Delta V/\Delta I = 0\) emerges for \(\mu_o H > 3\) T, and where \(V_N\) is approximately 0 for \(I < I_N\). e, \(I_C\) as a function of temperature (e) and magnetic field (f). Note that \(I_C\) is depressed with increasing temperature and eventually vanishes near \(T_C\), but is enhanced and remains sharp with increasing \(H||c\) axis.

\(I = 10\) nA
\(I = 0.1\) mA
\(I = 1.0\) mA
\(I = 10\) mA
\(I = 1.0\) mA
\(I = 0\)
The phase diagrams in Fig. 3e,f indicate that the first-order transition at $I_{\text{sf}}$ is followed by the second onset of NDR at $I = I_{\text{NDR}}$ (Extended Data Fig. 2).

The magnetic field dependence of the $I-V$ characteristic illustrates a new type of bistable switching (Fig. 3c). Upon the vanishing of $I_{\text{NDR}}$ at $H_{\text{az}} > 3$ T, an extraordinary region emerges with $\Delta V / \Delta I = 0$ for $0 < I < I_{\text{sf}}$ (Fig. 3d). Note that at $7$ T and $14$ T, $V = 0$ as $I$ increases from zero to $I_{\text{sf}}$, a behaviour strikingly similar to the d.c. Josephson effect although no superconducting state is involved here. This region disappears via an abrupt transition at $I_{\text{sf}}$, and a more resistive state emerges at $I_{\text{NDR}}$ (Fig. 3d).

The phase diagrams in Fig. 3e,f indicate that the first-order transition occurs at $I_{\text{sf}}$ as a function of temperature or magnetic field, respectively. Note that $I_{\text{sf}}$ behaves differently when $H || a$ axis (Extended Data Fig. 2a).

**Time-dependent bistable switching**

A particularly striking and unique feature of $\Psi_C$ is that the first-order, bistable switching requires a finite time (for example, seconds or minutes) and occurs without extra stimulus. The axis voltage $V_a$ measured as a function of time at $10$ K, is shown in Fig. 4a–c (Extended Data Fig. 3). Each measurement of $V_a$ starts at $t = 0$ when a constant $I_a$ is applied.

**Anomalous $I-V$ characteristic**

The $I-V$ characteristic for $H = 0$ exhibits a first-order transition characterized by a critical current $I_{\text{sf}}$ that separates two distinct states.

**Properties in fields and with currents**

The response of $\rho_a$ to changes drastically when $H || c$ axis (Fig. 2c), which restores the magnetic order otherwise suppressed by $I$, and simultaneously reduces $\rho_a$, leading to a metallic state below $T_C$ for $I \leq 2$ mA. However, for $I > 2$ mA, $H$ reduces $\rho_a$ only slightly, leaving a much weaker conducting state; the CMR is thus no longer present for $I > 2$ mA, despite the large $\rho_a$. $H_{\text{az}} = 14$ T.

Combining the data for $\rho_a(H = 0, b)$ in Fig. 2a and $\rho_a(H_{\text{az}} = 14$ T, $b)$ in Fig. 2c yields the magnetoresistance ratio as a function of $I$, $(\rho_b(14$ T, $b) - \rho_b(0, b))/\rho_b(0, b)$ at $10$ K in Fig. 2e. Clearly, the CMR occurs only when $I \leq 2$ mA and abruptly disappears when $I > 2$ mA, which closely tracks the current dependence of $T_C$ in Fig. 2f. While $H || c$ axis enables the CMR, $I > 2$ mA weakens and eventually suppresses the ferrimagnetic state, thereby recovering the $\Psi_C$ state without the CMR. The susceptibility to $H$ highlights the unique CMR and is even more evident in the $I-V$ characteristics.

$T_C$, which differs markedly from other systems. By contrast, $T_C$ shifts systematically to higher temperatures with increasing $H || c$ axis (Fig. 1a).

**Anomalous $I-V$ characteristic**

The $I-V$ characteristic for $H = 0$ exhibits a first-order transition characterized by a critical current $I_{\text{sf}}$ that separates two distinct states. In addition, there are two onsets of S-shaped negative differential resistance (NDR) $H_{\text{az}} (= I_{\text{sf}})$ and $H_{\text{az}} (> I_{\text{sf}})$ (Fig. 3a,b). The $I-V$ curve bends over at an onset of the first NDR regime at $I = I_{\text{NDR}}$, above which an increase in $I$ leads to a decrease in $V$. This first NDR regime ends at $I_{\text{sf}} = 1.90$ mA, where an abrupt increase in $V$ occurs, indicating a first-order transition (Fig. 3b). This transition at $I_{\text{sf}}$ is followed by the second onset of NDR at $I = I_{\text{NDR}}$ (Extended Data Fig. 2).

The magnetic field dependence of the $I-V$ characteristic illustrates a new type of bistable switching (Fig. 3c). Upon the vanishing of $I_{\text{NDR}}$ at $H_{\text{az}} > 3$ T, an extraordinary region emerges with $\Delta V / \Delta I = 0$ for $0 < I < I_{\text{sf}}$ (Fig. 3d). Note that at $7$ T and $14$ T, $V = 0$ as $I$ increases from zero to $I_{\text{sf}}$, a behaviour strikingly similar to the d.c. Josephson effect although no superconducting state is involved here. This region disappears via an abrupt transition at $I_{\text{sf}}$, and a more resistive state emerges at $I_{\text{NDR}}$ (Fig. 3d). The phase diagrams in Fig. 3e,f indicate that the first-order transition occurs at $I_{\text{sf}}$ as a function of temperature or magnetic field, respectively. Note that $I_{\text{sf}}$ behaves differently when $H || a$ axis (Extended Data Fig. 2a).

**Time-dependent bistable switching**

A particularly striking and unique feature of $\Psi_C$ is that the first-order, bistable switching requires a finite time (for example, seconds or minutes) and occurs without extra stimulus. The axis voltage $V_a$ measured as a function of time at $10$ K, is shown in Fig. 4a–c (Extended Data Fig. 3). Each measurement of $V_a$ starts at $t = 0$ when a constant $I_a$ is applied.
and continued until \( t = 180 \) s (or 1,800 s (Extended Data Fig. 3a)) has elapsed. For example, when \( t < 2.030 \) mA at \( t = 0 \), \( \Psi_c \) remains essentially constant for the time of measurements. A current increase of merely 0.25% (that is, from 2.030 to 2.035 mA) causes \( \Psi_c \) to spike abruptly at \( t = 9.6 \) s, with a voltage increase of \( \Delta V_c = 0.52 \) V or \( \Delta V_c / V_c = 30% \), and then immediately stabilizes at a constant value (Fig. 4a). Further, slight increases of \( t \) progressively shorten the delay time, but the change in voltage \( \Delta V_c \) remains essentially the same, at 0.52 V. This protocol indicates that only two discrete states exist, separated by a first-order switching that persists up to \( T_c \) (Extended Data Fig. 3b). On the other hand, when \( H || c \) axis (Fig. 4b), switching takes up to 114 s to occur at a significantly higher \( I = 3.98 \) mA with \( \mu_c H_c = 7 \) T, but involves a much larger voltage increase \( \Delta V_c \) of approximately 0.99 V or \( \Delta V_c / V_c \) of approximately 2.000% (Fig. 4b). By contrast, the abrupt bistable switching completely vanishes when \( H \parallel a \) axis (Fig. 4c), and is replaced by a gradual, continuous change in \( V_c \) with \( H \).

The stark contrast between data in Fig. 4b for \( H || c \) axis and Fig. 4c for \( H || a \) axis reinforces the following key points. First, application of \( H || c \) axis apparently strengthens \( \Psi_c \), via the \( c \) axis \( \Psi_{COC} \) (Fig. 4d), which interlocks rigidly with the Te sublattice and \( M_{\text{Mn}} \). It therefore takes a stronger \( I \) and a longer \( t \) for the charge carriers to alter the configuration of multiple atoms involved in conductive pathways. Second, the fact that this switching is always achieved via a first-order transition further emphasizes that there are key differences in the extended current paths and orbitals involved in the two states. Conversely, application of \( H || a \) axis, where the CMR is absent, tilts the \( a \) axis \( M_{\text{COC}} \) towards the \( c \) axis, which weakens \( \Psi_c \) (Fig. 4e). Note that the energy dissipated at each switching for \( H = 0 \) and \( \mu_c H_c = 7 \) T decreases rapidly with increasing \( I \), which is clearly inconsistent with Joule heating (Fig. 4f) (Methods).

Discussion and outlook

The COC, which couple to the ferrimagnetic state, originate from spontaneous breaking of time-reversal symmetry below \( T_c = 78 \) K and circulate along Te–Te edges of the MnTe octahedra, producing \( M_{\text{COC}} \) oriented primarily along the \( c \) axis (Methods). (Note that similar orbital moments of loop currents in cuprates have been discussed, but with no net moment\(^{20,21}\).) In the absence of \( H || c \) axis, the net circulation of the COC is zero as the COC can circulate both clockwise and counterclockwise (Figs. 1d, e and 4g). This results in a complicated configuration of disordered circulation domains that cause strong scattering and high resistance. Application of \( H || c \) axis favours only one circulation (that is, either clockwise or counterclockwise) and enlarges its domains (and concurrently reducing the domains with the opposite circulation).

As the overall conductivity is a sensitive function of the configuration of the COC domains, the alignment of the dominant circulation domains is crucial for the enhancement of \( \Psi_c \) by \( H || c \) axis, and the sharp reduction in electron scattering evident in the 10–1CMR (Fig. 1a, b).

Applying a nonequilibrium current \( I > 2 \) mA destabilizes \( \Psi_c \) in favour of \( \Psi_h \), as \( \Psi_h \) permits no net \( a \) axis current (Fig. 4g) (Extended Data Fig. 4; Methods). The resistivity is reduced with increased \( I \), but at the same time, the increased \( I \) also weakens and eventually suppresses the ferrimagnetic state and, consequently, \( \Psi_c \), leading to a first-order phase transition that destroys \( \Psi_c \) (Fig. 2a–d).

The rigid coupling of the COC (and \( M_{\text{COC}} \)) to the Te sublattice and \( M_{\text{Mn}} \) is a signature of \( \Psi_c \). The COC extend over multiple atomic sites with couplings that depend on the ionic positions and dictate the current strength. Therefore, \( \Psi_c \) entails a particularly strong coupling between the lattice, the \( c \) axis \( M_{\text{COC}} \) and \( M_{\text{Mn}} \), as well as the anisotropic magnetic-to-electronic coupling manifest in the magnetostriction (Fig. 1c; Methods). Such a rigid coupling in \( \Psi_c \) explains the current-driven first-order transition from \( \Psi_h \) to \( \Psi_c \). This coupling also enables \( \Psi_c \), to remain metastable over the long timescales that are set by Te atomic motion and bond lengths, or fluctuations/phonon effects.

Further probes (for example, optical control\(^{40}\) of \( \Psi_h \), particularly the orbital magnetization and its response to \( H || c \) axis, will yield more insights into the COC. For example, a Kerr or terahertz Kerr effect couples directly to electron current operators, but indirectly to spins via spin-orbit coupling\(^{41}\). A large, increasing Kerr effect with increasing \( H || c \) axis would provide further evidence for the existence of a strongly correlated COC state in MnS\(_{2}\)Te\(_{6}\).

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41586-022-05262-3.
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Methods

Experimental details
Single crystals of Mn$_3$Si$_2$Te$_6$ were grown using a flux method. Measurements of crystal structures were performed using a Bruker Quest ECO single-crystal diffractometer with an Oxford Cryosystem providing sample temperature environments ranging from 80 K to 400 K. Chemical analyses of the samples were performed using a combination of a Hitachi MT3030 Plus Scanning Electron Microscope and an Oxford Energy Dispersive X-Ray Spectroscopy (EDX). The measurements of the electric resistivity and $I$–$V$ characteristic were carried out using a Quantum Design (QD) Dynacool PPMS system having a 14-T magnet and a set of external Keithley meters that measure $I$ and $V$ with high precision. Magnetization measurements under applied electric current were performed using a QD MPMS-XL magnetometer with a home-made probe. The magnetoresistance was measured using a home-made dilatometer compatible with the QD Dynacool PPMS system. The dilatometer consists of four identical KYOWA, type KFL strain gauges forming a Wheatstone bridge with the sample mounted on one arm. The other three strain gauges act as compensators to cancel unwanted shifts in the strain gauges due to changes in temperature and/or magnetic field.

Further notes on the COC, CMR, crystal structure and bistable switching

The COC not only coexist with the long-range magnetic order but also produce their own nonzero net magnetization. COC discussed in other materials involve zero net circulation and zero net magnetic moment. By their observed symmetries, they necessarily entail an ‘antiferromagnetic’ pattern of clockwise and counterclockwise COC circulations. This distinction is key to what makes the COC of Mn$_3$Si$_2$Te$_6$ unique. It is important to note that CMR is described by a lowest-order tensor, unlike high-order, nonlinear electric response known as electronic magnetic–chiral anisotropy.

Mn$_3$Si$_2$Te$_6$ crystallizes in a trigonal space group P-31c (no. 163) with two inequivalent Mn sites, M1 and M2, forming a trimmer-honeycomb lattice (Fig. 1d,e). It orders ferrimagnetically at $T_c = 78$ K$^{20,21}$ with the Mn spins antiferromagnetically coupled along the $c$ axis\(^1\). Our recent neutron diffraction data show a noncollinear magnetic structure below $T_c$, in which the Mn spins lie predominantly within the $ab$ plane, but tilt within the $ab$ plane and cant towards the $c$ axis by 10° under ambient conditions (Fig. 1f).

Note that $\rho_c$ of Mn$_3$Si$_2$Te$_6$ does not follow an activation law and or a simple power law at low temperatures (Extended Data Fig. 1a), which differs from that reported in Seo et al.$^2$.

The CMR is enhanced (Extended Data Fig. 1b), whereas the saturated magnetization $M_i$ is reduced to 1.40 $\mu_B$/Mn in Mn$_3$(Si$_{1-x}$Ge$_x$)$_2$Te$_6$ (Extended Data Fig. 1c), which is further evidence that larger orbital moments of the COC state partially cancel the net magnetic moments. On the other hand, in Mn$_3$(Te$_{1-x}$Se$_x$)$_2$ (Extended Data Fig. 1c), the CMR is weakened (Extended Data Fig. 1b), but $M_i$ is enhanced up to 1.7 $\mu_B$/Mn (Extended Data Fig. 1c), which indicates that smaller orbital moments lie on the mixed-occupancy, chalcogenide sublattice, resulting in a weaker cancellation effect. It is also noteworthy that the Ge doping that enhances the CMR also expands the $ab$ plane, whereas Se doping that weakens the CMR shrinks the $ab$ plane (inset in Extended Data Fig. 1b).

$M_{\text{COE}}$ is estimated based on the total measured moments, 4.55 and 4.20 $\mu_B$ for Mn1 and Mn2, respectively\(^2\), which are significantly smaller than the expected 5 $\mu_B$. The reduced magnetic moments suggest partial cancellation between $M_{\text{m}}$ and $M_{\text{COE}}$. We infer that the upper limit of $M_{\text{COE}}$ is on the order of 0.10 $\mu_B$. We are mindful that it is always possible that the Mn local moments could be smaller than expected, thus less cancellation with $M_{\text{COE}}$ would be needed to produce the total observed moments.

The magnetostriiction is associated with $M_{\text{COE}}$ because the Lorentz force acts to expand an $ab$ plane COC circulating on the rigid Te–Te edges of MnTe$_6$ octahedra when $H||c$ axis. The net orbital magnetization, which equals the loop current times the orbital area, naturally increases due to increasing the orbital area. As such, the larger the orbital area or $ab$ plane, the larger $M_{\text{COE}}$ becomes; and a large $M_{\text{COE}}$ renders a larger cancellation, thus a smaller total, observed moment. This also explains the inverse correlation between the CMR and $M_{\text{COE}}$ illustrated in Extended Data Fig. 1b,c. Further highlighting the role of $M_{\text{COE}}$.

The rigid coupling of the COC to the Te sublattice and $M_{\text{COE}}$ is a signature of the new state. The COC extend over multiple atomic sites, with couplings that depend on the ionic positions and dictate the current strength. The strong coupling sets timescales that enable the COC state to remain metastable over seconds or minutes before undergoing a first-order transition to a trivial state when applied d.c. currents exceed certain critical values. Such a long timescale is rare, if not unprecedented. We infer that this transition may mimic a solid-to-liquid phase transition.

During solid melting, thermal energy is absorbed to break chemical bonds, without raising temperature. The temperature of the system rises abruptly only when all the bonds are broken, that is, the solid is completely melted. An analogy drawn here is that, during the COC melting, the applied d.c. currents circulate in the sample to break COC in every unit cell, without causing a voltage or resistance increase before all COC are completely melted. In any case, breaking the COC inevitably causes rearrangements of Te orbitals and, more generally, changing lattice properties and, consequently, a long delay for switching.

Further discussion on the COC

Below $T_c$, ferrimagnetic order is observed with a magnetic symmetry group $C2'/c'$ (no. 15.89, BNS setting), which allows certain configurations of Te orbital currents circulating within the unit cell. As we are particularly interested in the COC configurations with $c$ axis orbital moments, we focus on one subset of COC patterns that respect not only the magnetic group no. 15.89, but also the additional threefold rotational symmetry inherited from the full crystal space group P-31c (no. 163). There are four independent current patterns that preserve this combination of 15.89 plus threefold rotations. The resulting four-parameter COC state is depicted in Fig. 4g.

This symmetry-preserving COC state $\Psi_c$ yields currents circulating on octahedral top/bottom faces as well as around Mn2 and Si sites. The resulting magnetic moments are all oriented exactly along the $c$ axis, yielding a nonzero $M_{\text{COE}}$. There are four additional independent current patterns that preserve the magnetic group no. 15.89 but break threefold rotation symmetry; these patterns, denoted $\Psi$, are described below, and produce moments oriented within the $ab$ plane, coupling only weakly to $c$ axis fields. Henceforth, we focus on the $\Psi_c$ state that makes the primary contribution to the COC state and its response to $c$ axis fields.

The COC circulate along Te–Te octahedra edges and involve primarily Te orbitals. The Mn local moment pattern detected in neutron scattering linearly couples to the order parameter of $\Psi_c$ in a Landau-Ginzburg theory. Therefore, the COC and localized moments both contribute to the observed magnetic pattern and cannot be fully distinguished. As such, $\Psi_c$ can be viewed as a superposition of Mn and Te states that forms time-reversal-breaking multisite molecular orbitals.

Application of $H||c$ axis couples to $\Psi_c$ via the c axis orbital moments, which in turn drastically enhances the COC order parameter, therefore, the hopping matrix elements, and accommodates more current passing through the sample, leading to the observed CMR. Clearly, application of $H||c$ axis weakens the c axis orbital moments, thus reducing $\Psi_c$, and necessarily causes suppression of the CMR. On the other hand, applying uniform current $I > 2mA$ (along the a or c axes) weakens the COC order parameter, therefore destabilizing $\Psi_c$ in favour of the trivial state $\Psi$. This is expected because the COC configuration does not permit any net current because each of the four independent parameters that specify $\Psi_c$ forms a circulating current within the unit cell with no net uniform
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component (Fig. 4g). The resistivity is reduced with increased \( I \), but at the same time, the increased \( I \) also weakens and eventually suppresses the ferrimagnetic state and, consequently, \( \Psi_C \), leading to a first-order phase transition that destroys the COC state (Fig. 2).

Strong electron-lattice coupling is an important feature of \( \Psi_C \). The COC extend over multiple atomic sites, with couplings that depend on the ionic positions in the lattice and determine the current strengths. Therefore, \( \Psi_C \) entails a much stronger coupling between the lattice and the c axis magnetic moments, and associated anisotropic magnetoelastic coupling, compared with that due to conventional Mn\(^{2+}\)-site, spin-only moments. This property of \( \Psi_C \), is consistent with the large magnetostriction \( \Delta a/a \) when \( H \parallel c \) axis and the nearly negligible \( \Delta a/a \) when \( H \parallel a \) axis (Fig. 1c).

Note that these effects occur only below \( T_C \) where \( \Psi_C \) exists.

The strong coupling of COC to the lattice suggests that the current-driven transition from \( \Psi_C \) to \( \Psi_T \) should be strongly first order as observed (Figs. 1–4). The long timescale (seconds or minutes) for the bistable switching is consistent with a picture wherein melting \( \Psi_C \) involves changing lattice properties, thus allowing \( \Psi_T \) to remain metastable over long timescales set by ionic motion rather than just by fast electrons and/or Mn spins. The coupling between \( \Psi_C \) and the lattice increases when \( H \parallel c \) axis, thus the observed longer time delay, but significantly weakened when \( H \parallel a \) axis, leading to a mixed state with coexisting \( \Psi_C \) and \( \Psi_T \).

The full COC state is parametrized by eight independent COC. These include three of the COC of \( \Psi_C \) (red, blue and green in Fig. 4g) and the six COC shown in Extended Data Fig. 4 that do not constitute a linearly independent set of COC. One of the COC (purple in Fig. 4g) is in fact a linear combination of COC that circulate around Mn\(_1\) atoms (Extended Data Fig. 4a) and different COC from \( \Psi_C \) (blue in Fig. 4g).

The sum of the three COC circulating about Mn\(_1\) atoms (Extended Data Fig. 4a)) in equal magnitudes in fact gives the difference of two COC of \( \Psi_C \) (red minus blue in Fig. 4g). Moreover, the sum of the three COC in equal magnitudes circulating about the Mn\(_2\) atoms (Extended Data Fig. 4b) gives another difference of two COC of \( \Psi_C \) (blue minus twice purple). These linear combinations are allowed by the threefold rotational symmetry described above. In total, the COC state thus has the four parameters of \( \Psi_C \), six extra parameters symmetry allowed by the magnetic space group and two constraints relating the ten parameters. Indeed, the full COC state is thus parametrized by eight independent COC.

The unit vector normal to the plane about which each COC circulates in \( \Psi_C \) is exactly along the axis. The COC of this state clearly couple most strongly to applied fields in the c axis. By contrast, the COC of \( \Psi_T \) do not couple strongly to applied fields in the c axis. Instead, the orbital moments generated by the COC in the Mn\(_1\) plane point primarily in the \( ab \) plane, and they are (approximately) pairwise separated by 120°.

Their orientation is slightly nontrivial as the Mn\(_{Te}\) octahedra are not regular octahedra. The orbital moments generated by the COC in the Mn\(_2\) plane similarly are not well aligned with the c axis, and their orientation is also nontrivial.

Absence of Joule heating

Self-heating effects cause a continuous drift in local temperature; these effects are generally isotropic or diffusive and vary continuously with changing current. Such behaviour is ruled out in the present study by (1) the abrupt nature of the switching without any drifting, (2) the independence of the two discrete values of \( V_e \) on the magnitude of \( I \) (Fig. 4a,b), (3) the extremely anisotropic behaviour in (see Fig. 4b,c) and, moreover, (4) the electric energy \( W = V_e I \times t \) dissipated by the applied current at each switching, for \( H = 0 \) and \( \mu_0 H_c = 7 \) T, decreases rapidly with increasing \( I \), which is clearly inconsistent with Joule heating (Fig. 4f).

Data availability

The data that support the findings of this work are available from the corresponding authors upon request.

Acknowledgements

G.C. thanks M. Lee, R. Nandkishore, X. Chen, M. Hermele, D. Singh, D. Reznik, D. Dessau and N. Clark for useful discussions. I.K. thanks E. Berg, M. Mourigal, B. Uchoa, C. Varma and Z. Wang for useful discussions. This work is supported by National Science Foundation via grants no. DMR 1903888 and DMR 2204811. The theoretical part of this work is in part performed at Aspen Center for Physics, which is supported by National Science Foundation grant PHY-1607611. The work at the Spallation Neutron Source at the Oak Ridge National Laboratory is sponsored by the Scientific User Facilities Division, Office of Basic Energy Sciences, US Department of Energy.

Author contributions

Y.Z. conducted measurements of the physical properties and data analysis; Y.N. grew the single crystals, characterized the crystal structure of the crystals, measured magnetization with applied currents and contributed to the data analysis; H.Z. conducted measurements of crystal and physical properties including the magnetostriction and the data analysis; F.Y. determined the magnetic structure of Mn\(_{6}\)Si\(_6\)Te\(_6\), using neutron diffraction and contributed to the data analysis; G.H. contributed to the theoretical analysis including detailed configurations of chiral orbital currents presented in the figures, L.D. contributed to the data analysis and paper writing; I.K. proposed the theoretical argument, formed the theoretical discussion and contributed to paper writing. G.C. initiated and directed this work, analyzed the data, constructed the figures and wrote the paper.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41586-022-05262-3.

Correspondence and requests for materials should be addressed to Itamar Kimchi or Gang Cao.

Peer review information Nature thanks Lan Wang, Victor Yakovenko and Meng Wang for their contribution to the peer review of this work. Peer reviewer reports are available.

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Extended Data Fig. 1 | Resistivity at low temperatures for Mn₃Si₂Te₆ and transport and magnetic properties for Mn₃(Si₁₋ₓGex)₂Te₆, Mn₃Si₂(Te₁₋ₓSey)₆.

a, The temperature dependence of the a-axis resistivity ρₐ at low temperatures (data in brown), and ln (ρₐ) versus T⁻¹ (data in blue). Note that ρₐ does not follow an activation law and or a simple power law at low temperatures. b, c, The magnetic field dependence of the a-axis magnetoresistance ratio defined by ρₐ(H)/ρₐ(0) (b) and the easy a-axis magnetization Mₐ (c) for Mn₃(Si₁₋ₓGex)₂Te₆ (red), Mn₃Si₂(Te₁₋ₓSey)₆ (blue) and undoped compound (black), respectively. Inset in b schematic illustration of the unit cell expansion and contraction due to Ge doping (red) and Se doping (blue), respectively.
Extended Data Fig. 2 | Additional I-V characteristic. 

**a**, Comparison of the I-V characteristic at $H \parallel a$ axis and $H \parallel c$ axis: the $a$-axis I-V characteristic at 10 K for $H = 0$ (red), $\mu_0 H_a = 14$ T along the $a$ axis (green) and the $c$ axis (blue). Note that the regime where $\Delta V/\Delta I = 0$ emerges only when $H \parallel c$ axis. 

**b**, The I-V characteristic at various temperatures. Note the regime $\Delta V/\Delta I = 0$ persists up to 70 K. 

**c**, The I-V characteristic at $I \parallel c$ axis for various temperatures. Note that the I-V characteristic is qualitatively similar to that for $I \parallel a$ axis but the first-order switching at $I_c$ is weaker.
Extended Data Fig. 3 | Additional time-dependent bistable switching data. a, Time-dependent bistable switching at 10 K with 1,800 s elapsed. b, Time-dependent bistable switching at 50 K and $\mu_o H_{||c} = 14$ T. c, d, Time-dependent bistable switching for $I_{||c}$ axis at 10 K for $H = 0$ (c) and $\mu_o H = 14$ T (d).
Extended Data Fig. 4 | Chiral orbital current parameters of $\Psi$, in the Mn1 (a) and Mn2 (b) planes. a, Three independent currents (orange, purple, and cerulean) run along Te-Te bonds in the Mn1 plane and are symmetry allowed magnetic space group. b, Three more independent currents (cyan, magenta, and yellow) run along Te-Te bonds in the Mn2 plane. These currents are not linearly independent of the COC of Fig. 4g in the main text. The sum of the orange, purple, and cerulean COC in a gives rise to the difference of the red and blue COC of Fig. 4g in the main text. Moreover, the sum of the cyan, magenta, and yellow COC in b gives the difference of the blue and twice purple COC of Fig. 4g. Bonds with two arrows of the same colour indicate that the current magnitude is doubled on that edge. In total, the COC state is parametrized by eight independent loop currents.