

Epitaxially defined Luttinger liquids on MoS₂ bicrystals

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ABSTRACT

A mirror twin boundary (MTB) in a transition metal dichalcogenide (TMD) monolayer can host one-dimensional electron liquid of a topological nature with tunable interactions. Unfortunately, electrical characterization of such boundaries has been challenging due to the paucity of samples with large enough size and high quality. Here, we report the conductance measurements of individual MTBs in epitaxially grown monolayer molybdenum disulfide (MoS_2) bicrystals that are tens of micrometers long. These MTBs exhibit power-law behaviors of conductance as a function of temperature and bias voltage up to room temperature, consistent with electrons tunneling into a Luttinger liquid. Transport measurements of two distinct types of MTBs reveal the critical role of the atomic-scale defects. This study demonstrates that MTBs in TMD monolayers provide an exciting new platform for studying the interplay between electronic interactions and topology.

A mirror twin boundary (MTB) in a transition metal dichalcogenide (TMD) monolayer is a one-dimensional (1D) grain boundary that separates two intersecting domains with 180° in-plane rotation [1-3]. Density functional theory calculations predict that MTBs in a TMD monolayer should host 1D electron liquids [3] of topological origin and thus provide an interesting system for investigating the interplay between electronic correlation and topology [4,5]. Although scanning tunneling microscopy (STM) studies of a short (~ 10 nm) MTB segment [6-10] and an angle-resolved photoemission spectroscopy study of a dense MTB network [11] have provided a glimpse of correlated electron states in these 1D boundaries, sample limitations have posed an outstanding challenge for electron transport experiments on MTBs. Compared with STM measurement that is local in nature, transport measurement captures the physics at mesoscopic scale. Moreover, fabricating MTB solid-state devices and performing transport measurements on a scalable MTB material platform will not only enable interrogation of exotic correlation and topological phenomena, but also open the avenue for engineering MTBs into electronic devices [12].

In this work, we epitaxially grow monolayer molybdenum disulfide (MoS_2) bicrystals on a c-plane sapphire substrate. These bicrystals possess MTBs of tens of micrometers in length at the intersections of two MoS_2 monolayer single crystals. We identify two types of MTBs: type I, in which the MTB is nearly straight along the whole intersection of the two crystals, and type II, in which the MTB makes frequent 60° turns, producing a zigzag structure. Field-effect transistor devices based on type I and type II MTBs exhibit distinct conductance behaviors: type I MTBs exhibit Coulomb blockade at cryogenic temperatures, indicative of fewer defects along the 1D channel, while type II MTBs do not. Both type I and type II MTBs show power-law-type behaviors of tunneling conductance as a function of temperature and bias voltage up to room temperature,

consistent with electrons tunneling into Luttinger liquids. The transport behaviors of type I and type II MTBs are characterized by distinct power-law exponents, however, pointing to the role of point defects at the sharp turns in producing large momentum scattering and valley pseudospin mixing for type II MTBs.

Epitaxial growth of MoS₂ bicrystals

We performed large-area growth of monolayer MoS₂ bicrystals using metal-organic chemical vapor deposition [12] (see Supplemental Material [13] for details). Epitaxial growth of MoS₂ monolayer crystals on a *c*-plane sapphire allows for two crystallographic variants of triangular facets which are 180° rotated from each other [Fig. 1(a)] [12]. By establishing S-vapor rich growth conditions, we realize the formation of S-terminating edges at each triangular facet. When two facets merge to form MoS₂ bicrystals, the resulting MTB hosts two columns of S-chains with a 4|4E sublattice, where each S chain along the 1D boundary is a reflection copy of the other with a half-unit cell shift [Fig. 1(b)]. The macroscopic shape of the bicrystal is either a rhombus or a bowtie. In the rhombus type, where two triangular facets meet at the edges, a straight MTB (type I) is formed, which extends tens of micrometers in length. In the bowtie type, where two triangular facets meet at the vertexes, a zigzag MTB (type II) is formed (see also Supplemental Material Fig. S1 [13]). Figure 1(c) and 1(d) show dark-field transmission electron microscopy (DF-TEM) images of the two MTB types. In the zigzag MTBs, the straight 4|4E MTB segments, whose average length is ~60 nm [Fig. 1(e)], are connected by a series of 60° turns. In each turn, we typically find one or two point defects, identified as 5|7 or 4|8 rings. Associated with these point defects, we occasionally observed 4|4P sublattices. However, the 4|4P sublattices extend no more than 2-3 nm before they gradually transition to the usual 4|4E sublattice in the straight segments.

In the following transport characterizations, we consider the effects of 4|4P sublattices as a part of the point-defect-induced changes.

Transport characterizations of type I (straight) and type II (zigzag) MTBs

We use conventional dry transfer method [20] to encapsulate a monolayer MoS₂ bicrystal with top and bottom boron nitride (hBN) layers. We add a graphite layer as electrostatic gate and lithographically define gold/titanium electrodes to complete field-effect transistor [Fig. 2(a), see Supplemental Material [13] for detailed fabrication procedures].

The dashed lines in Fig. 2(b) illustrate the room-temperature drain-source current I as a function of gate voltage (V_G) obtained from a control device that does not contain an MTB. It exhibits a n -type transistor behavior [21], as expected for a single-crystal MoS₂ monolayer device. The device **D1** containing a 200-nm-long type I MTB shows a markedly different behavior. The current in **D1** extends deep into the negative V_G region and remains only weakly dependent on V_G , signaling the MTB's metallic character. A 220-nm long type II MTB transistor (**D2**) also exhibits measurable currents at negative V_G , but the current is strongly dependent on V_G , eventually turning off at very negative gate voltages [Fig. 2(c)]. In addition, the magnitude of the current in **D2** is generally much smaller than that of **D1**. All the transistors with the hBN passivation show negligible hysteresis with V_G . Details about the transport measurements are discussed in Supplemental Material [13].

When the temperature falls below $T = 70$ K, the current of **D1** exhibits dramatic oscillations as a function of V_G . Figure 2(d) shows the differential conductance (dI/dV) map as a function of both V_G and bias voltage V measured from **D1** at 5.0 K. There is a clear modulation of the conductance

gap (purple region) as a function of V_G at cryogenic temperatures, the hallmark of Coulomb blockade. This phenomenon is a consequence of the finite energy required to add (remove) a single electron to (from) the finite-sized type I MTB [22]. The aperiodic pattern in Fig. 2(d) suggests there are multiple weakly coupled electron islands in series in this device [22-24]. An order-of-magnitude estimate of the geometrical capacitance gives a charging energy of ~ 26 meV for the 200-nm-long MTB [25-27]. The overall addition energies extracted from Fig. 2(d) are much larger than this estimate, consistent with the fact that the device consists of smaller electron islands in series. The formation of multiple electron islands may be due to occasional defects along the type I MTB.

As shown in Fig. 2(e), **D2** does not exhibit conductance oscillations as a function of V_G at 5.0 K, but instead the conductance gap gradually increases as V_G becomes more negative. The absence of conductance gap modulation is related to the frequent 60° turns in the type II MTB with substantial amounts of point defects in series. In this case, the metallic character of MTB is compromised by these atomic-scale defects, and the conduction is eventually turned off.

As the temperature increases from 5.0 K to room temperature, the zero-bias conductance of **D1** gradually increases with temperature, and the device behavior changes from Coulomb blockade to a linear trend in the log-log plot above $T \sim 70$ K, suggesting a power-law-like behavior [Fig. 3(a)]. Such a crossover is reminiscent of the transport characteristics in carbon nanotube field-effect transistors changing from Coulomb blockade to Luttinger liquid physics [28,29]. Out of the blockade regime, we extract a slope of the linear trend of 0.9 (guided by the dashed line) over the

temperature range of 70 to 297 K. In contrast, the zero-bias conductance of **D2** follows a power-law behavior over the entire temperature range with an exponent of 2.1 [Fig. 3(b)].

The power-law-like behaviors of conductance with temperature strongly suggest the formation of Luttinger liquids (correlated electronic states in 1D) on the MTBs. In a Luttinger liquid with tunnel contacts to metal electrodes, the tunneling density of states is suppressed following a power-law relationship with energy [22]. In our transport experiment, the energy comes from both temperature ($k_B T$) and bias voltage (eV). As a result, when the bias becomes dominant ($eV/k_B T \gg 1$), the differential conductance should also exhibit a power-law behavior as a function of V , with the same power-law exponent as with temperature. In fact, the tunnel differential conductance should follow

$$\frac{dI}{dV} = AT^\alpha \cosh\left(\gamma \frac{eV}{2k_B T}\right) \left| \Gamma\left(\frac{1+\alpha}{2} + \gamma \frac{ieV}{2\pi k_B T}\right) \right|^2, \quad (1)$$

where α is the exponent and with A and γ as fitting parameters [28,30] (see a discussion of γ in Appendix A). This equation suggests that in a scaled differential conductance-scaled bias plot ($dI/dV/T^\alpha$ vs. $eV/k_B T$), data collected at different temperatures and biases should collapse to a single curve.

Figure 3(c) presents the $dI/dV/T^\alpha$ vs. $eV/k_B T$ plot for temperatures above 70 K (the Coulomb blockade-Luttinger liquid crossover) for **D1**. In the high-bias regime ($eV/k_B T \gg 1$), the differential conductance shows the expected power-law-like exponent of 0.9 with bias voltage. Additionally, the data from all measured temperatures collapse to a single curve, consistent with the universal scaling law in Eq. 1. Using the exponent α , it is possible to infer the electron-electron interaction strength, i.e., the Luttinger parameter g . For a non-interacting system, $g = 1$, and for a correlated

state with repulsive interactions, $0 < g < 1$. In our MTB devices with valley-chirality locking (see Fig. 4 below), because electrons tunnel into and out of the Fermi-liquid metal electrodes, we reach $\alpha = (g^{-1} + g - 2)/4$ (see Supplemental Material [13] for details). The exponent of 0.9 corresponds to $g = 0.19 \pm 0.01$, indicating strong electronic correlations in our MTB. We note that Fig. 3(c) does not include data below 70 K because Coulomb blockade strongly suppresses the conductance in the low-bias regime. In the high-bias regime, where the bias overcomes the Coulomb blockade and becomes the energy-dominant term, the differential conductance shows a power-law-like behavior as a function of bias regardless of the temperature (Supplemental Material Fig. S2 [13]).

The above measurements were performed at $V_G = 0.42$ V, where the blockade effect is less significant [red dot in Fig. 2(d)]. We performed similar analysis at different gate voltages. We find that at gate voltages where the blockade is relatively weak, the power-law exponents are between $0.7 \sim 0.9$ with no obvious gate dependence as long as the temperature is above the Coulomb blockade-Luttinger liquid crossover. At gate voltages where the blockade is strong, we do not observe clear Luttinger liquid behavior at our measured temperatures. We discuss examples from these two regimes in Supplemental Material Fig. S3 [$V_G = -0.87$ V, pink dot in Fig. 2(d)] and S4 [$V_G = -1.56$ V, green dot in Fig. 2(d)] [13], respectively.

For the type II MTB device **D2**, the zero-bias conductance at $V_G = 0$ V exhibits a single power law from 5 to 297 K [Fig. 3(b)], with an exponent of 2.1. The scaled differential conductance-scaled bias plot ($dI/dV/T^\alpha$ vs. $eV/k_B T$) at different temperatures and biases [Fig. 3(d)] again collapse into a single universal curve. The agreement between the temperature exponent and the bias exponent

(2.0) provides strong evidence that the electrons in the type II MTB form a Luttinger liquid with the parameter $g = 0.099 \pm 0.01$. The smaller Luttinger parameter compared to that of **D1** indicates a stronger electron-electron interaction effect caused by the more frequent scattering slowing down the Fermi velocity (see Discussion below). We also fabricated a 560-nm-long type II MTB device **D3** where the power-law exponent is 3.2, which is larger than that of the 220-nm-long **D2** (Supplemental Material Fig. S5 [13]). This indicates an even smaller Luttinger parameter in **D3**, as the scattering increases with longer channel.

We also investigated the Luttinger liquid behaviors at different gate voltages for **D2**. Supplemental Material Figure S6 [13] shows that the device conductance follows a power-law behavior as a function of T , and the exponent increases as V_G is more negative. At more positive V_G , the device behavior deviates from the power law: this is because at high doping the device conduction is dominated by the bulk crystal and thus the Luttinger liquid model cannot describe the device behavior adequately. Supplemental Material Figure S7 [13] shows the scaled conductance plots at $V_G = -3$ V and $V_G = 8$ V.

Discussion

The experimental observations point to the significant role of atomic-scale defects in the transport behaviors. The metallic nature of the MTB originates from its nontrivial topology. Because of the inversion symmetry breaking in MoS₂ monolayer, the K and K' valleys exhibit opposite valley Chern numbers of $\pm 1/2$ [15,31]. Therefore, an MTB produces a valley Chern number change of ± 1 across the boundary, producing one pair of valley-projected counter-propagating states along the MTB [32]. Using the first-principles calculations (see Supplemental Material [13] for details),

we obtain the relaxed MoS₂ bicrystal structure with one MTB [Fig. 4(a)] and its electronic band structure [Fig. 4(b)]. As shown in Fig. 4(b), such a structure features in-gap states. The calculated partial densities of the in-gap states reveal their association with the MTB, as illustrated in Fig. 4(a). The MTB band shown in Fig. 4(b) has opposite velocities at the projected K and K' valleys, indicating valley-chirality locking.

The intervalley scattering from atomic-scale scatterers prevents the valley from being a good quantum number and can localize the 1D MTB states [33-35]. This leads to a decrease in Fermi velocity or the degradation of the 1D states. As illustrated phenomenologically in Fig. 4(c), the increase of atomic-scale scattering causes the MTB band to be more localized (less dispersive), eventually producing an energy gap near the bulk valence band. For the type I MTB with a small number of scatterers [Fig. 4(c) left], the Fermi velocity (slope of the dispersion) is insensitive to the Fermi level position, and thus the power-law exponent, a measure of the electron interaction strength, shows minimal gate dependence. For the type II MTB with more scatterers arising from the frequent 60° turns [Fig. 4(c) right], the Fermi velocity continuously decreases as the Fermi level is gate tuned to deep in the bulk gap. The lower velocity enhances the electron interaction effect, which accounts for the observed larger power-law exponent as the gate becomes more negative.

The scattering analysis is fully consistent with our experimental results on **D1**, **D2** and **D3**. As the atomic-scale scattering increases from **D1** to **D2** to **D3**, the Fermi velocity decreases. This enhances the interactions between electrons and therefore the Luttinger parameter decreases from **D1** to **D2** to **D3**. According to the relationship between Luttinger parameter and the power-law exponent,

the exponents for these three devices should increase from **D1** to **D2** to **D3**, which indeed agree with our experimental findings.

In Appendix B, we include further discussion regarding the previous literature on STM studies on MTBs, scattering mechanisms in carbon nanotube, and 1D topological states in bilayer graphene domain walls.

Outlook

This work demonstrates that MTB hosts various novel physical phenomena, including single-electron transport, correlated electron states, and nontrivial band topology. These properties may be utilized in building novel electronic devices. By engineering the electrical contacts, the nontrivial topology may lead to dissipationless transport at room temperature because of the large bandgap in MoS₂. While this work features electrons tunneling into the 1D MTB channel due to large contact resistance, future improvements in contacts will enable discovery of more exotic physics in MTBs, such as the spin Kondo effect [36]. The spin-charge separation, another important feature of Luttinger liquid, could also provide additional insight of the MTB behaviors and can be the subject of future transport experiment with more advanced settings [4,5,37,38]. Finally, our growth method can be easily scaled up, providing a pathway to produce MTB circuits.

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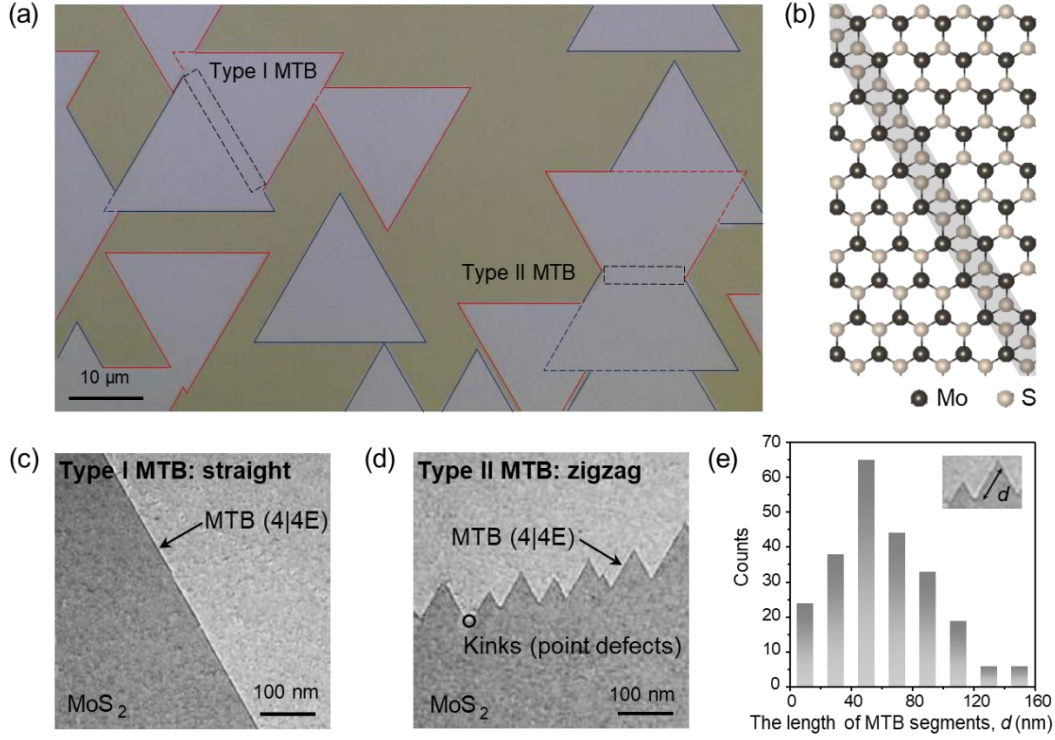


FIG. 1. Two types (straight and zigzag) of MTBs imbedded in MoS₂ monolayer bicrystals.

(a) Optical image of MoS₂ monolayer bicrystals grown on *c*-plane sapphire. Blue and red lines indicate the two variants of MoS₂ monolayers. Black dashed boxes indicate the locations of MTBs.

(b) Atomic structure of the 4|4E MTB (dark shade).

(c),(d) DF-TEM images obtained from a rhombus-shape bicrystal (c) and a bowtie-shape bicrystal (d), showing the type I straight MTB and type II zigzag MTB, respectively.

(e) Statistical distribution of the length of MTB segments in type II MTBs. The average length of MTB segments is 61.6 nm.

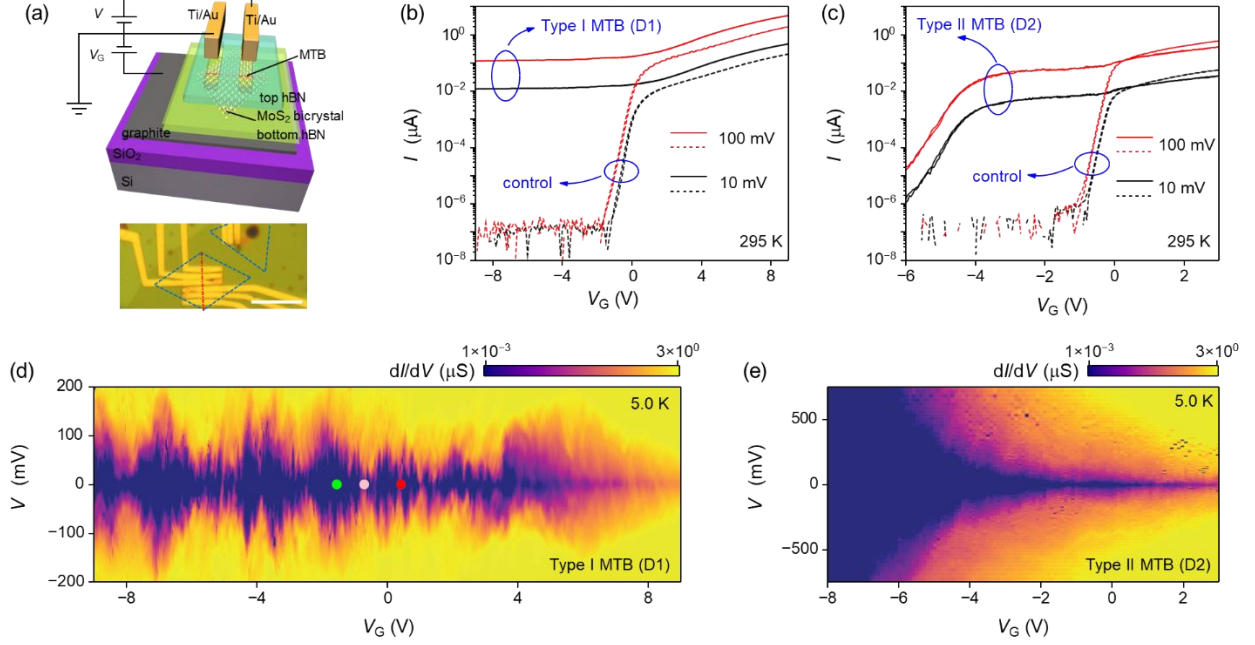


FIG. 2. Transport properties of MTB devices. (a) Top: a schematic showing the MTB device structure. Bottom: optical image of the type I MTB devices and the control devices, where monolayer MoS₂ flakes are outlined by the blue dashed lines and the MTB by the red dash-dotted line. Scale bar: 10 μm . (b),(c) Transistor drain-source current I as a function of V_G for **D1** (b) and **D2** (c), together with the control devices, at room temperature. V_G sweeps from negative maxima to positive maxima and then back. The legends show the applied biases V . (d),(e) dI/dV - V - V_G maps for **D1** (d) and **D2** (e) at 5.0 K. In (d), red ($V_G = 0.42$ V) and pink dots ($V_G = -0.87$ V) denote gate voltages where the blockade is less significant, while the green dot ($V_G = -1.56$ V) denotes a gate voltage with strong blockade.

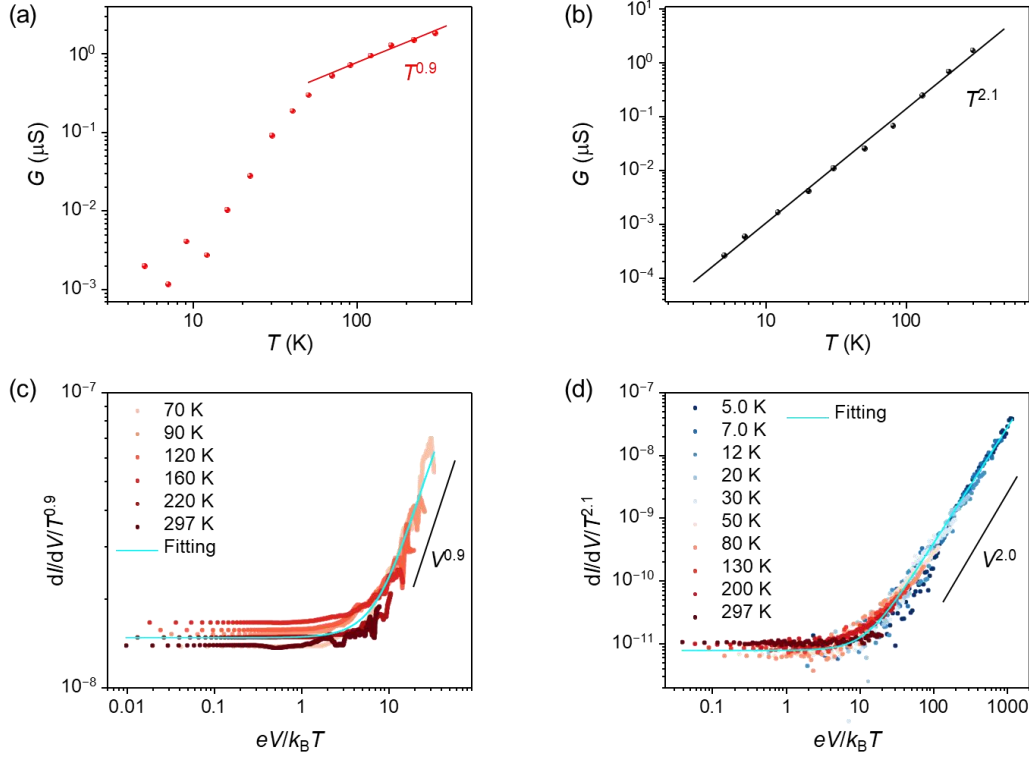


FIG. 3. Luttinger liquid behaviors in MTBs. (a),(b) Zero-bias conductance G versus temperature T of **D1** (a) and **D2** (b). For **D1**, above a crossover temperature, G follows a $T^{0.9}$ trend. G is rapidly suppressed below the crossover temperature. For **D2**, a single $T^{2.1}$ power law is observed. (c),(d) The scaled conductance versus the scaled bias plots of **D1** (c) and **D2** (d) for temperatures in the power-law regime. All data collapse to single curves, showing $V^{0.9}$ and $V^{2.0}$ power laws at high biases for **D1** and **D2**, respectively. The linear-linear plots of (c) and (d) are available in Supplemental Material Fig. S8 [13].

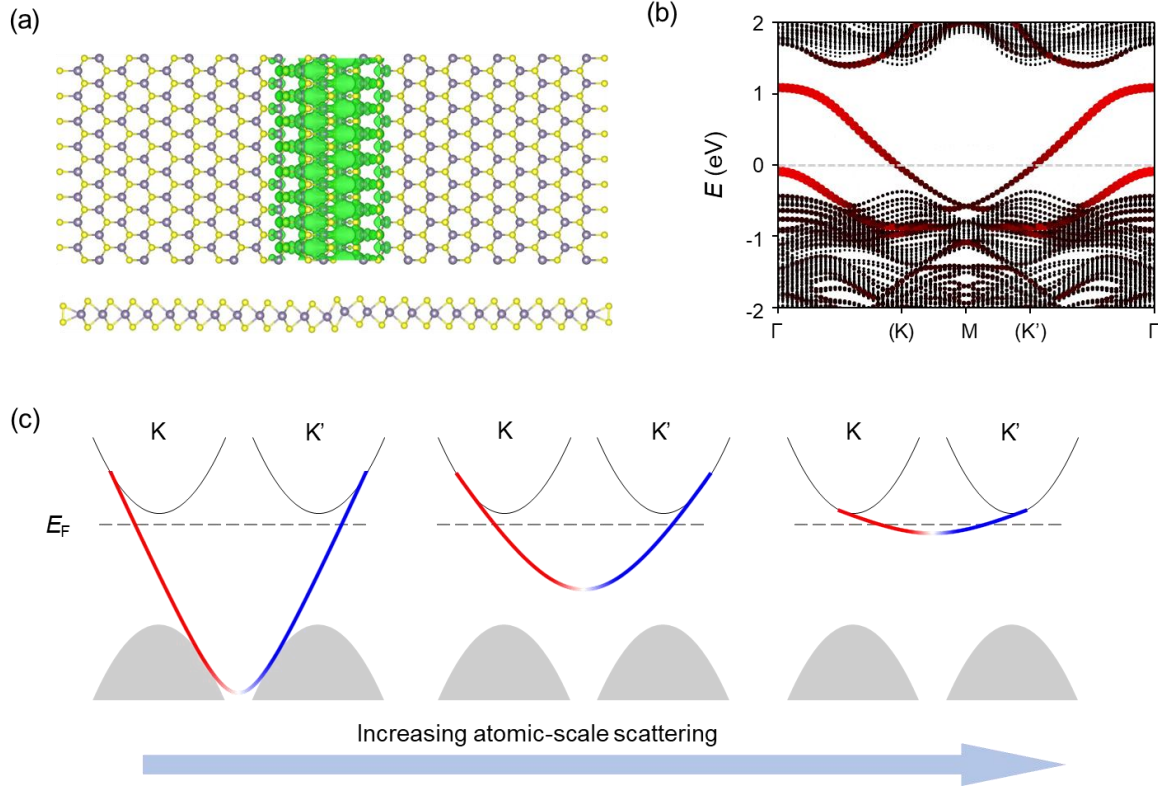


FIG. 4. Topological origin of the metallic MTB and the role of atomic-scale scatterers. (a) Top and side views of the calculated relaxed crystal structure of a MoS_2 bicrystal with one MTB. The in-gap states are associated with the MTB (green). (b) The calculated electronic band structure of the MoS_2 bicrystal with one MTB in (a), featuring valley-chirality locked 1D band in the bulk gap (red dots). (c) Schematics of the effect of atomic-scale scattering on altering the MTB bands. The MTB bands of the left and right chiralities are represented by the red and blue curves around the K and K' valleys, respectively.

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End Matter

Appendix A: The discussion of the fitting parameter γ .

In Eq. 1, γ is the ratio of the voltage drop across the most resistive tunneling junction to the total bias voltage V . Consequently, $1/\gamma$ gives the minimum number of junctions in the device. As shown in Fig. 3(c), in the best fitting of our observations to Eq. 1, we find γ to be ~ 0.3 . The minimum number of tunneling junctions is therefore $[1/\gamma] = 4$. With two tunneling junctions to the metal electrodes, this suggests that the MTB in **D1** consists of approximately $4 - 2 = 2$ junctions, corresponding to three Luttinger liquid segments and that there are tunneling events between them. This is consistent with the aperiodic Coulomb blockade [Fig. 2(d)], which suggests that the MTB in **D1** contains multiple Coulomb-blockade regions in series.

The data in Fig. 3(d) can also be fitted using Eq. 1, with $\gamma \sim 0.25$, suggesting there are at least 4 Luttinger liquid segments in this device. As the type II MTB exhibits a zigzag structure, most likely it is the sharp turns that segmentize the MTB. A Luttinger liquid fitting on the 560-nm-long type II MTB device **D3** yields $\gamma \sim 0.11$ (Supplemental Material Fig. S5 [13]), indicating that the number of junctions, $1/\gamma$, is approximately proportional to the device length, as expected from the sharp turns segmentizing the MTB.

Appendix B: The discussion of relevant literature.

The discussion of defect scattering in the main text accounts for the difference between the Luttinger parameters extracted from our transport experiments and the previously reported values (0.5 ± 0.1 in MoS₂ MTB [8] and 0.54 ± 0.03 in MoSe₂ MTB [10]) from local STM studies. In an STM experiment, the conductance occurs via the electron tunneling into the bulk of a single segment of Luttinger liquid from an STM tip. As a result, the Luttinger parameters obtained in these experiments are intrinsic material properties because no scattering is involved in this process. In our transport experiments, we treat the structural detail of an MTB device as a black box and probe its effective Luttinger parameter. Because the scattering slows down the Fermi velocity, the effective Luttinger parameter becomes smaller. As such, the more defective type II MTBs exhibit smaller g than those in type I MTBs, and both are smaller than those obtained in STM measurements.

The role of scatterers in MTB is reminiscent of a previous study of carbon nanotubes [39]. The transport properties of metallic tubes are not very sensitive to long-range scatterers because of the large-momentum separation between the two valleys. Nevertheless, atomic-scale scatterers can significantly modify the transport properties. One critical difference between the carbon nanotubes and MTBs is that both chiralities are present at each valley of the metallic carbon nanotubes, while the valley-chirality locking causes MTBs to be immune to intra-valley backscattering.

The 1D topological states similar to our MTB states were previously studied in bilayer graphene with a layer-stacking or electric-field domain wall [33,40-43]. In that case, however, an external

electric field is required to break the inversion symmetry and produce a valley Chern number. Additionally, the electric field-induced gap in bilayer graphene is small, and the 1D electronic interaction effects such as Luttinger liquid behavior [4,5] and Coulomb blockade have not been observed, even at low temperature. In our MoS₂ bicrystal, the gap is intrinsic with a magnitude of ~ 2 eV, permitting the Luttinger liquid behavior on MTB to persist up to room temperature.