



# Long-lifetime spin excitations near domain walls in 1T-TaS<sub>2</sub>

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Spin chains in solid state materials are quintessential quantum systems with potential applications in spin-based logic, memory, quantum communication, and computation. A critical challenge is the experimental determination of spin lifetimes with the ultimate goal of increasing it. Local measurements by scanning tunneling microscopy (STM) have demonstrated the importance of decoupling spins from their environment, with markedly improved lifetimes in spin chains on the surfaces of band insulators. In this work we use low-temperature scanning tunneling microscopy to reveal long-lifetime excitations in a chain of spin-1/2 electrons embedded in a charge density wave Mott insulator, 1T-TaS<sub>2</sub>. Naturally occurring domain walls trap chains of localized spin-1/2 electrons in nearby sites, whose energies lie inside the Mott gap. Spin-polarized measurements on these sites show distinct two-level switching noise, as well as negative differential resistance in the  $dI/dV$  spectra, typically associated with spin fluctuations. The excitations show exceptionally long lifetimes of a few seconds at 300 mK. Our work suggests that layered Mott insulators in the chalcogenide family, which are amenable to exfoliation and lithography, may provide a viable platform for quantum applications.

spin chain | scanning tunneling microscopy | noise spectroscopy

Spin chains in one dimension (1D) have been used as prototypical systems to study quantum mechanical phenomena in lower dimensions due to their simplicity, integrability, and scalability. Their 1D nature can enhance correlation effects leading to exotic quasiparticle excitations due to spin–charge separation (1, 2), as well as Majorana edge states (3, 4) when coupled to a superconductor. One-dimensional spin chains thus provide a fertile playground for realizing new states of matter at the intersection of strong correlations, magnetism, topology, and superconductivity. Scanning tunneling microscopy and spectroscopy (STM/S) have provided unprecedented access and control at the atomic scale for the study of 1D spin chains compared to other ensemble averaging techniques like angle-resolved photoemission spectroscopy, neutron scattering, or magnetic susceptibility. Properties such as spin lifetimes, spin order, and the energetics of 1D spin chains have been explored in great detail using STM-based atomic manipulation (5–15). These studies have established that the substrate harboring the spin chain can affect the ground state configuration, spin–spin interactions, and the spin lifetimes dramatically (5). Long spin lifetimes have been observed on spin chains on insulating substrates and are highly desirable due to their applicability in quantum information technologies. More recently, optical studies of Mott insulating states in transition metal dichalcogenide moiré superlattices have shown unusually long spin lifetimes emerging from strong correlations (16). Mott insulators are therefore a new and unique platform to study spins in the presence of many-body interactions. In this work we use STM/S to probe the lifetime of quasi-1D spin chains trapped at the domain walls (DWs) in the layered Mott insulator, 1T-TaS<sub>2</sub>.

The insulator 1T-TaS<sub>2</sub> is an intriguing system that undergoes a low-temperature transition into a charge density wave (CDW) Mott insulator without magnetic order. The commensurate CDW (CCDW) phase that emerges below 200 K has CDW clusters that resemble the famous Star of David (SD) shape. In this work, we study cleaved 1T-TaS<sub>2</sub> single crystals measured at temperatures between 300 mK and 14 K. Fig. 1*A* shows a schematic of the CDW. Each SD cluster in the CDW phase consists of 12 Ta outer atoms on the SD that move toward a center Ta atom, giving a total of 13 Ta atoms per SD cluster. The SDs form a triangular lattice which is  $\sqrt{13} \times \sqrt{13}$  times larger than the unreconstructed lattice (17). Fig. 1*B* shows an STM image of a large area where each protrusion represents an SD cluster with Fig. 1*B*, *Inset*, showing a high-resolution image of the reconstructed S atoms (white dotted lines).

The CDW transition is accompanied by a metal–insulator Mott transition (18) which results in the occupation of a single unpaired electron per cluster that occupies a narrow Hubbard band below the Fermi energy. This picture comes from the well-understood band structure and charge arrangement in the insulating phase, as we describe here. Each of the 13 Ta<sup>4+</sup> sites in the cluster has one 5d electron (19). With

## Significance

There is an intense ongoing search for two-level quantum systems with long lifetimes for applications in quantum communication and computation. Much research has been focused on studying isolated spins in semiconductors or band insulators. Mott insulators provide an interesting alternative platform but have been far less explored. In this work we use a technique capable of resolving individual spins at atomic length scales, to measure the two-level switching of spin states in 1T-TaS<sub>2</sub>. We find quasi-1D chains of spin-1/2 electrons embedded in 1T-TaS<sub>2</sub> which have exceptionally long lifetimes. The discovery of long-lived spin states in a tractable van der Waal material opens doors to using Mott systems in future quantum information applications.

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The authors declare no competing interest.

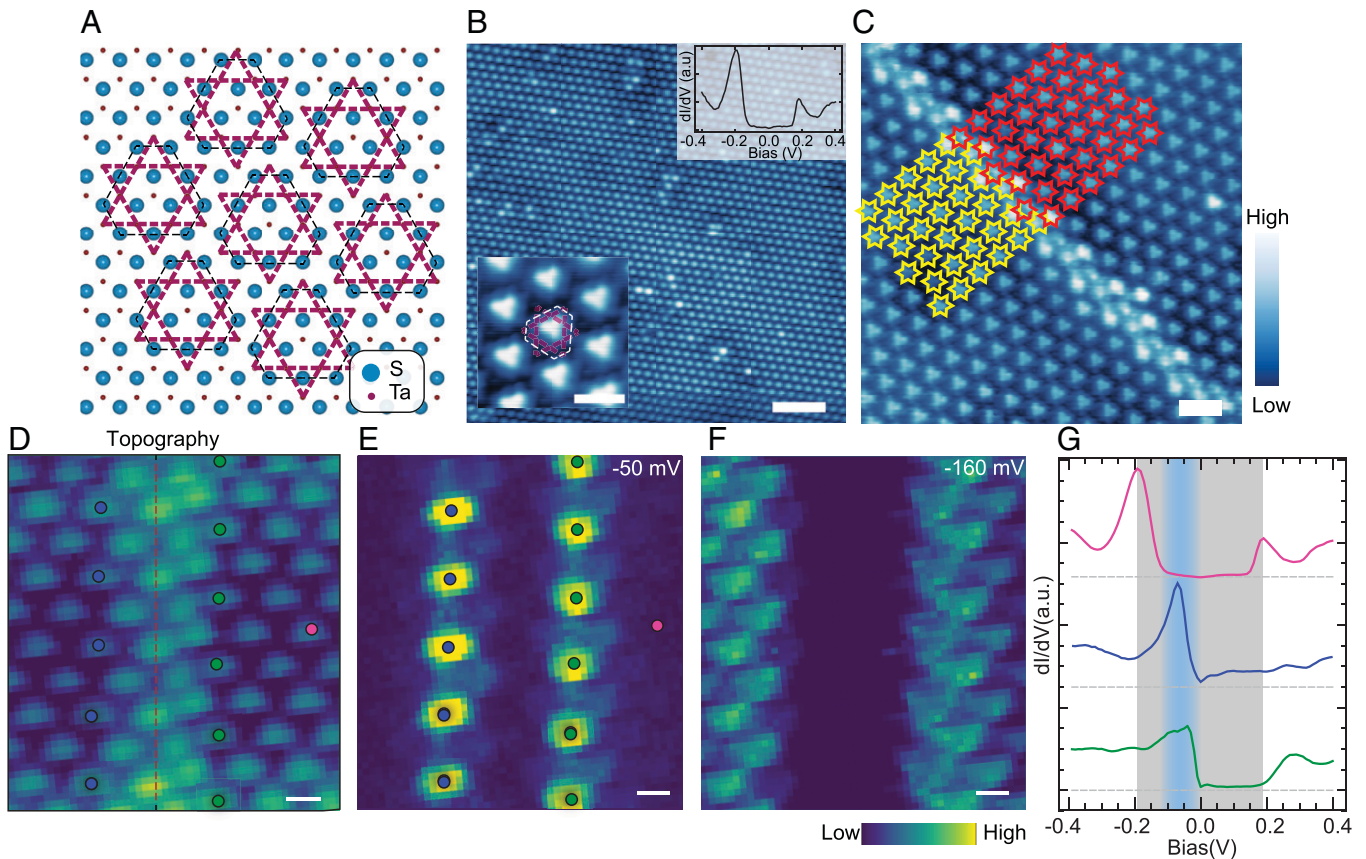
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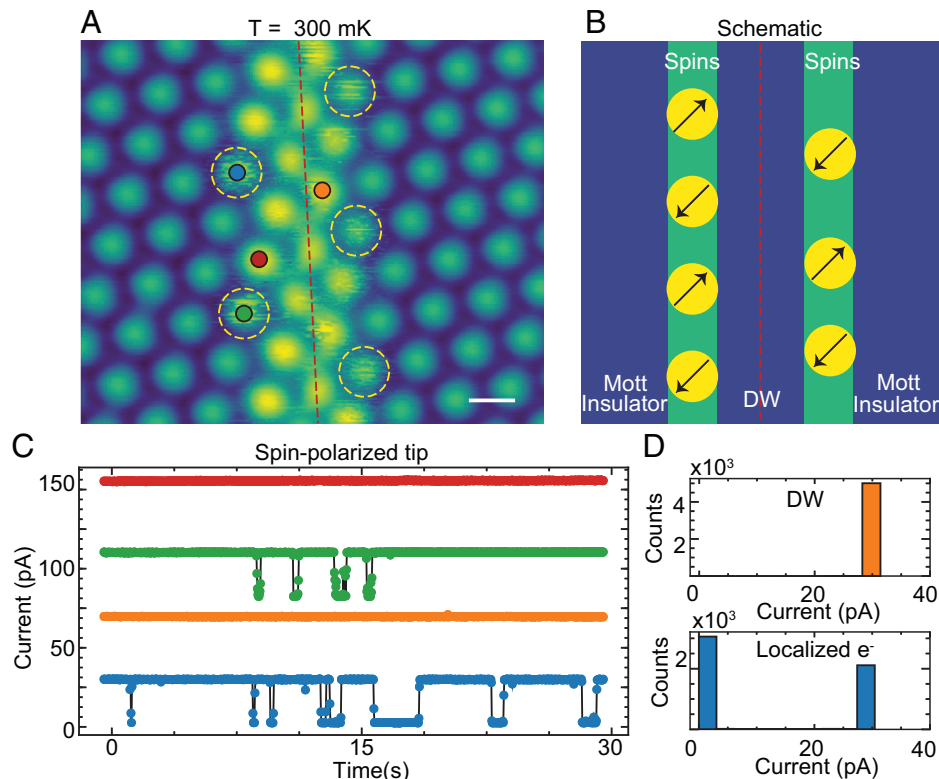
**Fig. 1.** CDW DW in 1T-TaS<sub>2</sub>: structure, morphology, and localized quasi-1D electrons near the DW. (A) Schematic of the SD reconstruction in monolayer TaS<sub>2</sub> showing the positions of the tantalum (smaller purple dots) and sulfur (blue dots) atoms. (B) Large area STM topography (50 nm × 50 nm) showing the  $\sqrt{13} \times \sqrt{13}$  reconstructed surface ( $V_s = 300$  mV,  $I_t = 30$  pA). (Scale bar, 5 nm.) (Bottom Inset) A zoomed in view of the reconstructed lattice with the SD overlay. (Scale bar, 1 nm.) (Top Inset) Typical  $dI/dV$  spectrum far from DWs. The two peaks around  $\pm 0.2$  eV are associated with the UHB and LHB, respectively. (C) STM topography (18 nm × 18 nm) of an area with the DW studied in this work. The SD clusters on the two sides of the DW are marked in yellow and red ( $V_{\text{bias}} = 400$  mV,  $I_t = 40$  pA). (Scale bar, 5 nm.) (D) Topography obtained simultaneously with the LDOS maps on a DW. Green/blue circles show the location of the localized spins. (E) LDOS map at  $-50$  mV. The green/blue colored dots label the location of the localized low-energy electrons. (F) LDOS map at  $-160$  mV (close to the energy of the LHB). The dark area reflects a suppression of the DOS due to band bending. (Scale bar in D–F, 1 nm.) (G)  $dI/dV$  spectra obtained on the localized electron sites (blue and green dots in E) and a far-off site (pink). The spectra shown are averaged over similar sites. The shaded gray area denotes the Mott gap. The shaded blue area denotes the energy of the localized electrons inside the Mott gap.

an odd number of electrons per cluster, band theory would predict that 1T-TaS<sub>2</sub> should be a metal. However, 1T-TaS<sub>2</sub> is instead an insulator (19, 20). Density functional theory calculations attribute this to the occurrence of a narrow, flat band that crosses the Fermi energy ( $E_F$ ) in the CCDW state, which splits into upper and lower Hubbard bands (UHB and LHB) due to a correlation-driven Mott transition (21). The Hubbard bands are located symmetrically about  $E_F$  at  $\pm 200$  meV inside the CDW gap ( $\pm 300$  meV), as shown in Fig. 1 B, Inset. We note that the LHB hosts only one electron per cluster, with the remaining electrons occupying a multiplicity of d bands several hundred meV below  $E_F$  (18, 21). While the existence of a single unpaired spin per cluster would normally suggest an antiferromagnetic ground state, no magnetic ordering has been observed even at the lowest temperatures, and a quantum spin liquid picture has been proposed to explain the absence of ordering (22).

A Mott insulator with a well-defined bulk gap and one unpaired spin per site is an ideal candidate to study the behavior of localized spins. Kondo physics has been observed, for example, in a similar CDW Mott insulator in proximity to a metal (23). Interestingly, previous STM studies have shown that the local potential generated by CDW DWs in 1T-TaS<sub>2</sub> can cause an energy shift of the bands as one approaches the DW (24–28) and trap localized charge. Fig. 1 C and D show

an image of one such DW. As shown in Fig. 1 G and *SI Appendix, Fig. S1*, the DW potential causes the energies of the UHB and LHB to shift as we move closer to the DW from either side and creates a quasi-1D potential trap for electrons. Fig. 1 E and F show STM  $dI/dV$  maps (or local density of states [LDOS] maps) in the vicinity of the DW. As shown in Fig. 1 E, the low-energy LDOS map within the bulk Mott gap reveals a pattern of bright clusters localized on two rows of SD sites on either side of the DW. This can also be seen in the integrated density of states inside the Mott gap (*SI Appendix, Fig. S2*).

We thus find that the low-energy occupied states near the DW are localized to two rows of SD clusters (marked by blue/green dots in Fig. 1 D and E and *SI Appendix, Fig. S2*) on either side of the DW, aligned in a quasi-1D pattern (henceforth dubbed “quasi-1D chains”). As marked by the blue/green dots in Fig. 1 D, the quasi-1D chains are not at the DW itself but are at nearby locations, at a distance of  $\sim 1.5$  nm from the DW. A combination of correlation effects, broken rotational and translational symmetry near the DW, and the strong potential provided by the SD clusters likely causes this charge localization. These trapped electrons in the quasi-1D chains are both energetically and spatially decoupled from the surrounding lattice as shown schematically in Fig. 2 B. In the bulk (i.e., far from the DW), each SD cluster is known to host one spin-



**Fig. 2.** Quasi-1D spin chains with two-level noise with a spin-polarized tip. (A) Topography of a DW obtained with spin-polarized tip at 300 mK. (Scale bar, 1 nm.) (B) Schematic of the electrons/spin chains along the DW inside the Mott gap. The red dashed line depicts the position of the DW. (C) Noise measurements, i.e., time traces of the tunnel current, obtained on the marked sites (as indicated in A). The time traces have been color-referenced to the clusters they were obtained on. The sweeps are offset by 30 pA for clarity. The blue and green sweeps show the presence of two-level telegraph noise on the SD cluster near the DW and an absence of it on the DW. The measurements were made with a spin-polarized tip, tunnel current  $I_t = 30$  pA, and  $V_{\text{Bias}} = 30$  mV with the feedback loop turned off. (D) The histogram plots of the current sweeps show a two-level distribution for the blue site (which is absent for the DW site).

1/2 electron at the energy of the LHB which is  $\sim 200$  meV below  $E_F$ . Near the DW, the energies of these electrons are shifted such that they fall inside the bulk insulating gap (Fig. 1G), causing them to be energetically decoupled from the surrounding clusters. The spatial decoupling can be seen in LDOS maps below the  $E_F$ . Nearby clusters on either side of the quasi-1D chains are not occupied at these energies (also see *SI Appendix*, Fig. S2). The quasi-1D chains are thus able to act as a subsystem within an insulator, mimicking a chain of spin-1/2 electrons (Fig. 2B). Free from the frustration of the bulk triangular lattice, and given the typical antiferromagnetic correlations in Mott systems, these sites may exhibit short-range spin order.

We next characterize this quasi-1D chain with spin-polarized STM. The spin sensitivity of the Cr tips was confirmed by imaging the antiferromagnetic stripes on FeTe (*SI Appendix*, Fig. S3). Our most striking finding is the existence of switching noise, which is localized to the clusters on the quasi-1D chain. This noise is visible in the current channel as shown in Fig. 2C and can be captured by noise spectroscopy, i.e., a measurement of the tunneling current as a function of time  $[I(t)]$  with the feedback off. From Fig. 2C (and *SI Appendix*, Figs. S4–S6) we see that the  $I(t)$  on the clusters making up the quasi-1D chain shows noise with characteristics of two-level switching with a time scale of the order of a few seconds at 300 mK. This is exemplified by the histogram of the current values which displays a bimodal distribution characteristic of the two-level behavior (see Fig. 2D and *SI Appendix*, Fig. S6, for better statistics).

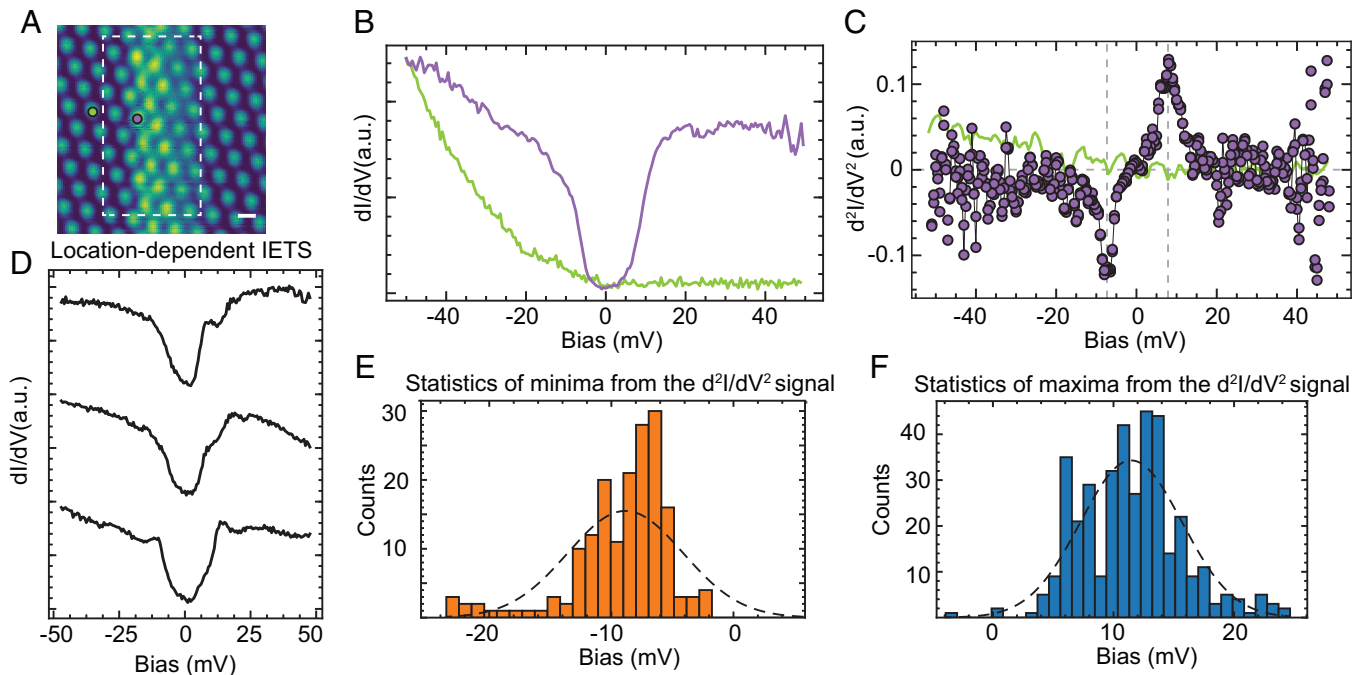
This immediately leads to the question of what might be responsible for this long-lifetime two-level switching. First, we can easily rule out tip effects as the source: the noise is site

dependent and occurs on specific clusters (green dots in Fig. 2A; also see *SI Appendix*, Fig. S4). We have confirmed the same phenomenology with 11 different tips on eight different samples on more than 50 DWs. Second, we can rule out structural fluctuations of the DW. In fact, the clusters nearest to the DW show far less or negligible noise as shown in Fig. 2C.

A critical hint to the origin of the switching noise is obtained by comparing our exceptionally long lifetimes to earlier STM experiments on chains of coupled spins arranged on a substrate through atomic manipulation. Several studies have been performed on such systems, and it is known that they show two-level noise with similar slow time scales (order of 100 ms or more) (8–12, 15). This suggests that the noise may arise from spin excitations. To further explore the possibility that spin flips are the source of the switching noise, we compare  $dI/dV$  spectra obtained with normal (W tips) and spin-polarized tips (Cr tips). Previous studies have shown that noise related to spin configuration changes is characterized by an inelastic signal in  $dI/dV$  spectra obtained with a normal tip (refs. 6, 7 and supplement of ref. 8) and a corresponding negative differential conductance (NDC) signal in  $dI/dV$  measured with a spin-polarized tip (30).

We first examine  $dI/dV$  spectra obtained with a normal tip. An inelastic signal is observed when tunneling electrons excite a bosonic mode, and the signature is a step-like increase in the  $dI/dV$  spectrum at the energy of the mode. The energy scale of the inelastic modes measured by previous STM studies on magnetic clusters and antiferromagnetic spin chains is typically a few meV to few tens of meV (refs. 6, 7 and supplement of ref. 8). In our system, we observe a clear inelastic signal on the SD clusters near the DWs that are seen as steps in the tunnel

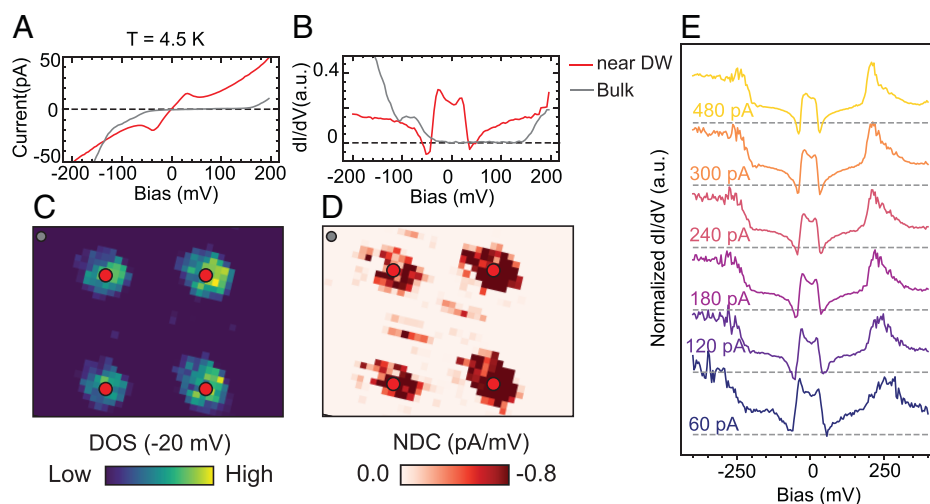




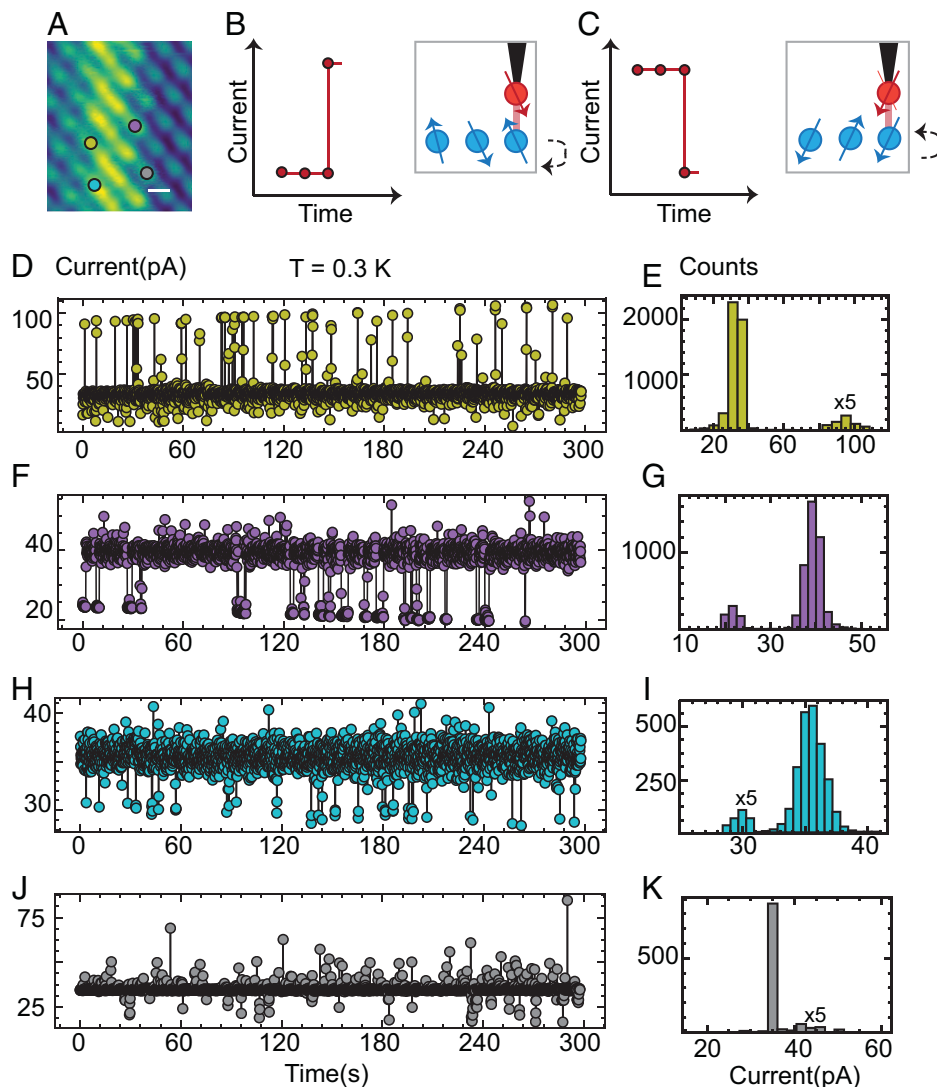
**Fig. 3.** Inelastic electron tunneling spectroscopy (IETS) signal in the  $dI/dV$  spectra at 300 mK obtained with a W-tip. (A) Topography of the DW showing clusters where the noise or  $dI/dV$  spectra in B were obtained. (Scale bar, 1 nm.) (B)  $dI/dV$  spectrum obtained on cluster with noise showing an inelastic signal ( $V_{\text{bias}} = 50$  mV,  $I_t = 50$  pA) (purple).  $dI/dV$  spectrum on a nearby cluster without noise which also does not show an inelastic signal. ( $V_{\text{bias}} = -70$  mV,  $I_t = 50$  pA) (green). (C) Derivative of the  $dI/dV$  signals showing the symmetric peak-dip feature at the energies associated with the inelastic mode (purple) but a featureless signal for the green curve. (D) IETS of different energies obtained at different locations near the DW shown in C. (E and F) Histogram of the energies of the peaks and dips of  $d^2I/dV^2$ , respectively, obtained around the DW shown by the dotted rectangle in A. The fitted Gaussian shows that the IETS signals at different locations near the DW follow a distribution around a mean value of 11 meV.

current on either side of the Fermi energy. These inelastic signatures are absent in spectra on neighboring clusters as well as in the bulk (Fig. 3B and *SI Appendix*, Fig. S7). The energy of these excitations can be determined by looking at the peak-dip energies in the derivative of the  $dI/dV$  signal ( $d^2I/dV^2$ ; Fig. 3C). In an antiferromagnetic spin chain the energy of the onset of the inelastic signal is a measure of  $J$ , the antiferromagnetic exchange interaction (or Heisenberg coupling). A statistical

analysis of the inelastic mode energies as determined by the peak and dip values near one particular DW is shown as a histogram in Fig. 3E and F. The mean mode energy for this DW is 11 meV, which is a similar order of magnitude compared to the previous studies on spin chains. We also find that the type and length of DWs have an effect on the average inelastic mode energy (*SI Appendix*, Fig. S8). This is expected since the details of the DW potential, length, and anisotropy in the exchange



**Fig. 4.** Spin-based NDC on the trapped electron sites measured with a spin-polarized tip. (A) Tunnel current obtained with a spin-polarized tip as a function of bias showing NDC at low energies near a DW. The locations of the spectra are indicated by gray and red circles in C and D. (B)  $dI/dV$  spectrum corresponding to A. The NDC manifests as negative values of the  $dI/dV$ . (C) Density of states (DOS) map near DW at  $-25$  mV showing the trapped electrons. (D) NDC map obtained with a spin-polarized tip near DW. A strong correlation between position of the electrons and strong NDC below  $E_F$  is evident (the details of obtaining the NDC map are in *SI Appendix*). (E) Normalized  $dI/dV$  spectra as a function of set point tunneling current obtained with a spin-polarized tip.  $V_{\text{bias}} = 400$  mV. The spectra have been normalized at 400 mV. The gray dashed line indicates the zero conductance. The NDC is small when the tip is far away (60 pA) but becomes successively stronger as the tip is moved closer to the sample (480 pA).



**Fig. 5.** Anticorrelated switching on nearby clusters measured with a spin-polarized tip. (A) Topography of the DW showing clusters where the noise spectra were obtained with the spin-polarized tip. (Scale bar, 1 nm.) (B and C) Schematic of the spin-polarized tunnel current when the spins flip from a state with a lower current to a higher current and vice versa. (D, F, H, and J) Current sweeps with feedback off obtained on clusters shown in C. All the sweeps were obtained at  $V_{\text{bias}} = 30$  mV,  $I_t = 30$  pA at  $T = 300$  mK. A small linear background has been subtracted from all of them that occurs due to piezo relaxation when the feedback is turned off. D and J show current behavior similar to the scenario in B, and F and H show current behavior similar to the scenario in C. (E, G, I, and K) Histograms of the current sweeps that show anticorrelated switching behavior between the two states for neighboring clusters. E and G and I and K show pairwise anticorrelation between the occupation of the up and down states. In J, the switching rate is much lower compared to D, F, and H possibly because the spin is located farther from the other spins.

couplings at each site determine possible ground-state and excited-state spin configurations (7, 29).

We next measure  $dI/dV$  spectra with a spin-polarized tip. In previous STM studies on spin chains, normal tips show an inelastic signal, while spin-polarized tips show pronounced NDC (30). NDC refers to the phenomenon where the conductance ( $dI/dV$ ) goes below zero for certain energies, signifying a decrease in tunnel current with increasing voltage. The NDC with spin-polarized tips was attributed to the tunneling electrons dynamically stabilizing an excited-state spin configuration (30). Observing NDC in this system is particularly important since the fluctuations smear out any static spin contrast. Fig. 4 A and B show  $I(V)$  and corresponding  $dI/dV$  on two different sites (marked in red/gray on the LDOS map in Fig. 4C). We find that the sites marked in red (corresponding to the clusters in the quasi-1D chain) show NDC with an energy scale comparable to inelastic excitations observed on similar DWs in our system. A spatial map of the NDC (see *SI Appendix*, Fig. S10,

for details) and a corresponding density of states map near a DW (Fig. 4 C and D) confirm that the NDC is sharply limited to the quasi-1D chain sites. Importantly, on moving the spin-polarized tip closer to the sample, the strength of NDC increases (Fig. 4E). This is as expected and demonstrates that the NDC is due to a coupling between the spin on the tip and the spins near the DW.

We have shown long-lived two-level switching noise in  $I(t)$ , inelastic signal in  $dI/dV$  with normal tips, and NDC in  $dI/dV$  with spin-polarized tips, all centered on clusters in the quasi-1D chain. This phenomenology is very similar to previous STM measurements on artificial spin chains. Taken together this suggests that the two-level noise in our system originates from spin flip excitations. The question that remains is whether the spin flips correspond to local excitations of a spin-1/2 electron on a single cluster or whether our signal is from collective behavior.

Let us first consider the possibility that spin flips of the local spin on the cluster right under the tip are responsible for the

observed switching noise. Earlier studies have shown that single spin flips of magnetic atoms may show long lifetimes (11) provided the spins involved are large ( $s = 2$ , for example). The spins in our case are expected to be spin-1/2, so individual spin flips are perhaps unlikely as the source of our noise. Another possibility is that the noise arises from coherent spin flips of the spin configuration of the chains (flipping of all spins from one direction to another for either ferromagnetically or antiferromagnetically coupled chains, for example). STM studies on different types of spin chains have shown that these occur at much slower time scales (8, 15). The antiferromagnetic exchange interaction in Mott insulators is given by  $J \sim \frac{4t^2}{U}$ , where  $U$  is the onsite repulsion and  $t$  is the bandwidth of the Hubbard band. Using  $U \sim 400$  meV and  $t \sim 70$  meV (from the measured  $dI/dV$  spectrum) for 1T-TaS<sub>2</sub>, we get an estimate of  $O(10)$  meV as the strength of the antiferromagnetic coupling between neighboring sites. Since the reduced symmetry of the low-energy electrons near the DW removes the frustration experienced by the local spins on the triangular lattice, it opens the possibility of short-range antiferromagnetic ordering of the local moments. In this scenario, the noise behavior may be attributed to the excitation of the ground-state spin configuration to one or more excited-state spin configurations. While detailed theoretical studies are required to pin down the allowed spin ground and excited states in this system, a few possible configurations based on the geometry of the DWs are shown in *SI Appendix, Fig. S11*.

While the observed noise fluctuations prevent us from obtaining spin contrast of the spin chains through topography, we find that the two-level noise does provide information on the spin configuration. As shown schematically in Fig. 5 *B* and *C*, when the spin on the tip is aligned to the spin on the sample, a spin flip on the sample would reduce the net spin-polarized tunnel current (while an antiparallel initial configuration would lead to an increase in the current upon spin flip). Shown in Fig. 5 *D*, *F*, *H*, and *J* are the noise on neighboring sites on the spin chain obtained with the same set point. We find that neighboring sites (for, e.g., Fig. 5 *D* and *H*) preferentially go from a lower (higher) current state to a higher (lower) current state. The histograms in Fig. 5 *E*, *G*, *I* and *K* illustrate this finding. This anticorrelated behavior of the switching serves as an alternate route to observing the spin contrast of the chains. The observed antiferromagnetic correlation between the neighboring sites is compatible with the dominance of Mott physics in this system.

The noise characteristics, the inelastic signal, and the NDC with spin-polarized tips are all characteristic of behavior observed in 1D spin chains and leave us with compelling evidence that DWs in TaS<sub>2</sub> host locally ordered quasi-1D spin states with long lifetimes. Although the bulk of 1T-TaS<sub>2</sub> has no long-range order, defects like DWs can pin spins locally into a short-range ordered state. These long-lived spin states hold promise for integration into devices for future spintronics-based

applications. Furthermore, strongly correlated spin chains in 1D are expected to exhibit spin-charge separation giving rise to quasiparticles such as spinons and chargons as have been observed in the quasi-1D Mott insulator SrCuO<sub>2</sub> (2). The DWs in TaS<sub>2</sub> have all the ingredients to exhibit spin-charge separation and could be prospective candidates to probe the existence of these exotic quasiparticles. Finally, rendering 1T-TaS<sub>2</sub> superconducting by gating or doping could provide another avenue to study spin chains coupled to a bulk superconductor and possible Majorana excitations.

## Methods

High-quality 1T-TaS<sub>2</sub> crystals used in this study were obtained from two separate groups. Both batches show similar phenomenology. The experiments were performed in two Unisoku STMs with base temperatures 300 mK and 4.3 K unless stated otherwise. The single crystals were cleaved at  $\sim 90$  K in ultra-high vacuum and immediately inserted into the STM. Spectroscopic measurements were performed by standard lock-in techniques. The spin-polarized tips were calibrated on Fe<sub>1+x</sub>Te, a known bicollinear antiferromagnet prior to their use. Noise spectroscopy was performed using a built-in module in the Nanonis package. The multigain preamplifier was set to a specific high frequency value ( $\sim 1.1$  kHz), and the sampling rate was controlled using the Nanonis module. The tip was placed over each cluster, and the current was recorded for a fixed amount of time.

**Data Availability.** All study data have been deposited in the Illinois Data Bank, [https://databank.illinois.edu/datasets/IDB-0883774?code=gFz\\_9HQomn0N51rNl1\\_Sr\\_Yc2Gii2XsYxADluXwOEMg](https://databank.illinois.edu/datasets/IDB-0883774?code=gFz_9HQomn0N51rNl1_Sr_Yc2Gii2XsYxADluXwOEMg).

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