

Moiré modulated quantum spin liquid candidate 1T-TaSe₂

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Quantum spin liquids are quantum phases of matter featuring collectively entangled states and emergent fractional many-body excitations. While methods exist to probe three-dimensional quantum spin liquids experimentally, these techniques lack the sensitivity to probe two-dimensional quantum spin liquids. This seriously hampers the study of potential monolayer quantum spin liquid candidates such as α -RuCl₃ and 1T-TaSe₂. Scanning tunneling microscopy (STM) and spectroscopy (STS) have recently been suggested as promising probes of the quantum spin liquid state, as they can access the spinon spectrum through inelastic tunneling spectroscopy (IETS). In this work, we employ this approach on the quantum spin liquid candidate material 1T-TaSe₂ and directly measure its low-energy inelastic excitations. We observe the emergence of a $\sqrt{3} \times \sqrt{3}$ reconstruction driven by the substrate, equivalent spectroscopy across all spin sites and coexistence of zero and finite energy excitations. We show that these observations are consistent with a modulated $\sqrt{3} \times \sqrt{3}$ spin liquid ground state. Our results demonstrate that IETS provides a powerful route to obtain atomic-scale insight into the magnetic excitations of two-dimensional materials, allowing to explore the effects of moiré modulations on potential quantum liquid phases.

Quantum spin liquids (QSLs) are exotic states of matter that arise in interacting quantum spin systems that do not order even at very low temperatures. They are predicted to exhibit exotic phenomena including fractionalized excitations and topological order.^{1–4} Beyond their fundamental appeal, QSLs have been proposed for applications in fault-tolerant quantum computation⁵, and doped QSLs may provide a route to high temperature unconventional superconductivity⁶. However, the characterization of QSLs remains challenging even in the bulk, and many open questions remain.^{1,4,7,8} The magnitude of the measured signal with the most used experimental probes, inelastic neutron scattering, NMR, μ SR, and thermal Hall transport^{9–11}, depends on the sample volume and the sensitivity is insufficient for two-dimensional (2D) systems. Consequently, studying QSL candidates in two dimensions remains an open challenge.

Two-dimensional QSL candidate materials arise in magnetically frustrated systems, with frustrated interactions in α -RuCl₃ and geometric frustration in 1T-TaS₂ being prominent examples. In bulk α -RuCl₃, inelastic neutron scattering has revealed a continuum of spin excitations, a signature of a proximal QSL phase,¹² featuring potential chiral modes.¹³ Theoretical work has indicated that monolayer α -RuCl₃ might be even closer to an intrinsic Kitaev-type QSL than the bulk material.^{14,15} In 1T-TaS₂, charge density wave reconstruction results in a 13 Ta atom unit cell that hosts an unpaired spin.^{2,16} Nuclear magnetic resonance, neutron scattering and muon spin resonance experiments have shown zero static magnetic moments down to 20 mK^{10,11,17} with a residual linear term in thermal conductivity, which implies the possible presence of a QSL state^{18,19}. In a closely related 2D system, monolayer 1T-TaSe₂, STM-based quasiparticle interference data and Kondo effect provide evidence of the potential presence of a spinon Fermi surface with gapless spinon excitations.^{20,21} 2D QSL candidates should

benefit from their ultimate tunability via gating, moiré patterns, and strain.²² The presence of proximal QSL phases in these systems highlights the need for new tools specialised in probing 2D materials.

Inelastic electron tunnelling spectroscopy (IETS) provides an experimental strategy to probe two-dimensional quantum spin liquids^{23–26}, analogously to magnon excitations in a ferromagnet^{27,28} or spin-flip excitations in isolated spins^{29,30}. In this work, we probe the spin liquid candidate monolayer 1T-TaSe₂ by IETS with a scanning tunneling microscope (STM). The low-energy IETS spectra show symmetric features arising from many-body excitations inside the Mott gap of monolayer 1T-TaSe₂. These excitations have a $\sqrt{3} \times \sqrt{3}$ modulation in real space and they stem from a substrate-induced moiré reconstruction of the many-body ground state in 1T-TaSe₂, featuring zero energy and finite energy excitations. Using many-body methods and auxiliary fermions, we show that the observed spectroscopy is consistent with a moiré-modulated QSL state. Our work shows that IETS is a powerful tool for probing many-body excitations in 2D materials at the atomic scale and points towards possibilities of moiré engineering of QSL states.

Moiré modulation in 1T-TaSe₂

Monolayer 1T-TaSe₂ films were grown on freshly cleaved highly oriented pyrolytic graphite (HOPG) substrates via molecular beam epitaxy (MBE) (details in the methods section).^{31,32} 1T-TaSe₂ layer contains one Ta atomic layer sandwiched between a pair of Se atomic layers, with each Ta atom coordinated by six Se atoms (Figure 1a). The Se cage forms an octahedron in the 1T phase. Figure 1b shows a large-scale topography of the MBE-grown TaSe₂ islands, which are predominantly 1T phase with high crystallinity and ultra-clean surfaces.

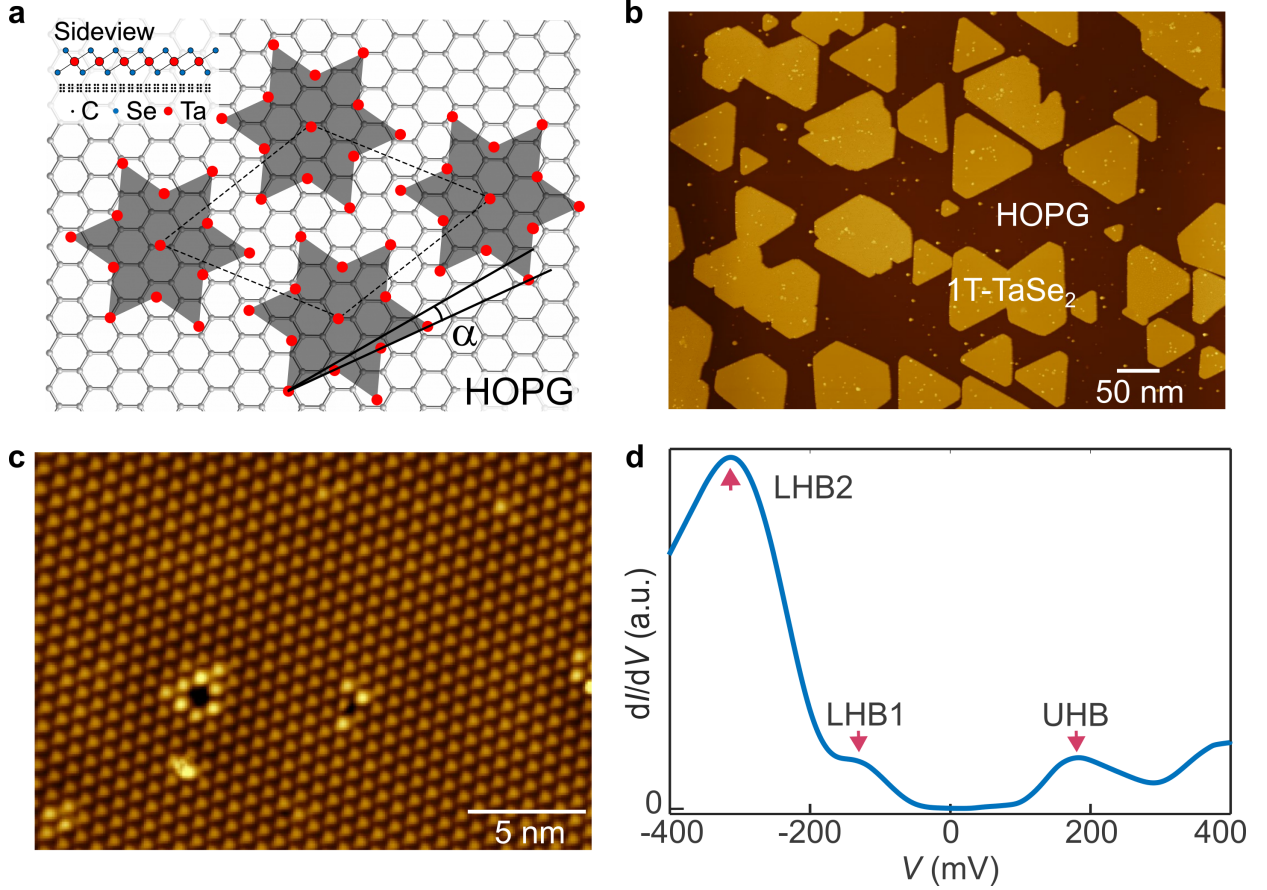


FIG. 1. CDW and Mott insulator state in monolayer 1T-TaSe₂ grown by MBE and characterized by STM at 5 K. **a**, Schematic of monolayer 1T-TaSe₂ crystal and the star-of-David structure. The red, blue and black atoms represent Ta, Se and carbon atoms. The angle between HOPG and 1T-TaSe₂ determines the moiré structure. **b**, A large-scale STM image of 1T-TaSe₂ monolayer islands on HOPG substrate. $V = 1$ V, $I = 20$ pA. **c**, A high-resolution STM topography image of the CDW superlattice. $V = -0.5$ V, $I = 50$ pA. **d**, Representative tunnelling spectrum on 1T-TaSe₂. Arrows indicate the two lower Hubbard bands (LHB1 and LHB2) and the upper Hubbard band (UHB).

The islands are nearly aligned with the HOPG substrate; the relative orientation between the atomic lattices of TaSe₂ and HOPG varies between 1° and 3° (see SI Figure 2). Figure 1c shows a zoomed-in STM image, where the charge density wave (CDW) modulation is visible as a triangular lattice. The Fourier transformed (FT) image (SI Figure 1) confirms the CDW structure of $\sqrt{13} \times \sqrt{13}$ times atomic lattice with an angle of 13.9° between the atomic and CDW lattices.^{31,32} Tunneling spectroscopy in Figure 1d shows the lower (-130 mV) and the upper Hubbard bands (180 mV) separated by the Mott gap, consistent with previous works.^{31,32} The orbital textures at different energies can be resolved via dI/dV maps (SI Figure 3), which are consistent with previous findings.³¹

The different 1T-TaSe₂ monolayer islands have slightly different orientations with respect to the HOPG substrate, and this results in different moiré periodicities on the sample. When the angle between the two atomic lattices is 1°, the moiré pattern forms a $\sqrt{3} \times \sqrt{3}$ reconstruction with respect to the CDW (later written as $\sqrt{3}$ for simplicity), as shown in Figure 2a-b. Fourier transforms

(FTs) of the topographies clearly show the peaks corresponding to the moiré pattern at the K and K' points of the CDW Brillouin zone (see Fig. 2b). This result agrees well with the simulation when the lattice angle difference between the 1T-TaSe₂ and HOPG is at 1° (Figure 2c). As the lattices rotate with respect to each other, the moiré shifts from the originally commensurate $\sqrt{3}$ modulation to an incommensurate superlattice, manifesting a topographically domain-like structure (Figure 2d). The FT peaks of the moiré (Figure 2e) also rotate slightly from the K and K' points of the CDW Brillouin zone, forming a trimer-like structure with the moiré peaks from the second CDW Brillouin zone. The simulation with 2° lattice rotation shows comparable features (Figure 2f) to the experiments. When the lattice rotation mismatch increases to 3°, the moiré modulation becomes more obvious in the STM images (Figure 2g) and the corresponding FT peaks are further separated compared to the rotation 2° (Figure 2h). The simulation at this angle in Figure 2i also matches the experimental results. More systematic simulation results of the angle-dependent moiré evolution of

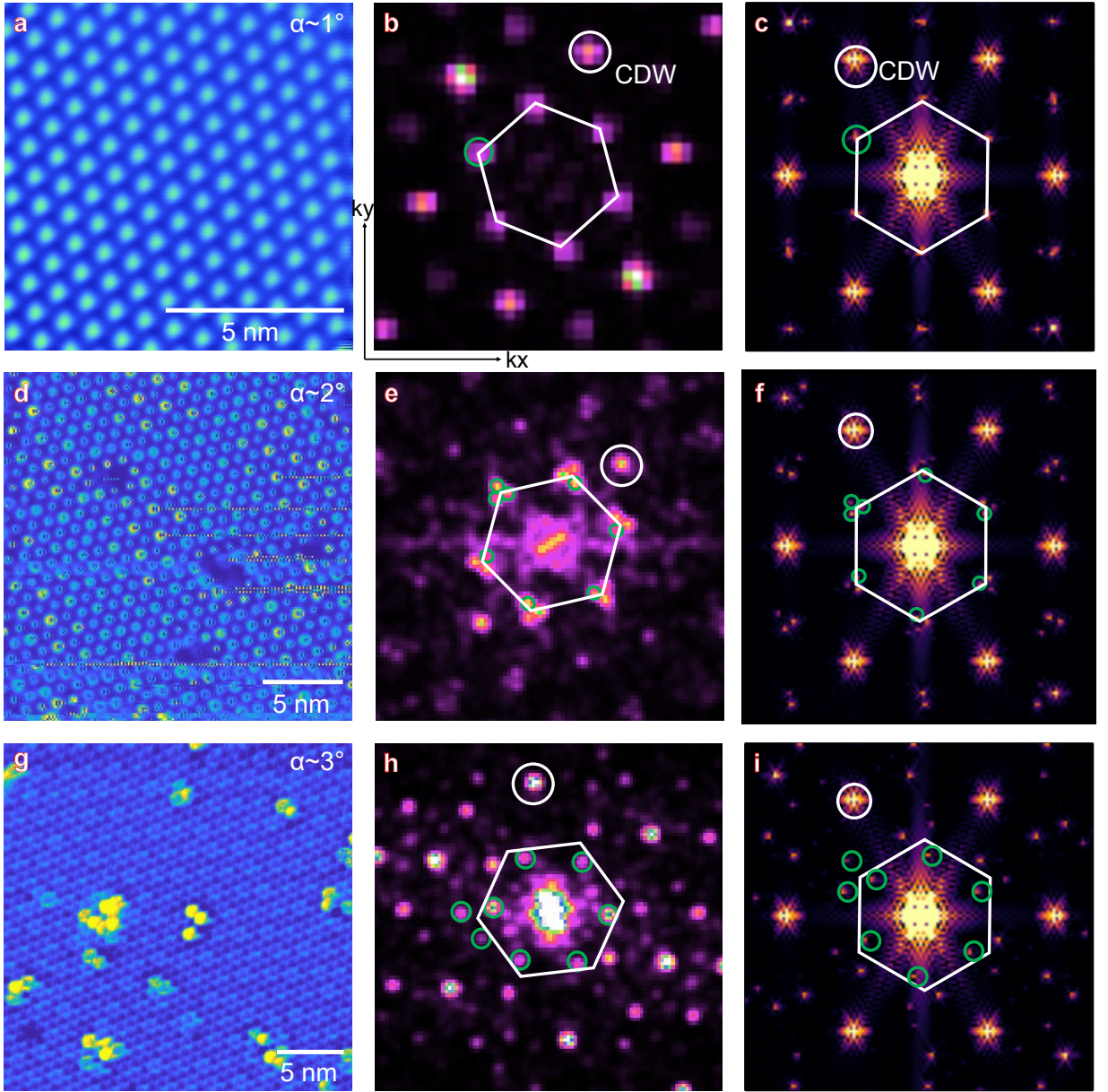


FIG. 2. Moiré modulation in 1T-TaSe₂ on HOPG substrate at different lattice orientations. **a,d,g**, dI/dV maps (extracted from grid data, at biases around LHB1) on different 1T-TaSe₂ islands with atomic lattices rotated by 1°, 2°, and 3° relative to the HOPG lattice. Besides CDW superlattices, new domain-like modulations are observed on 2° and 3° islands. **b,e,h**, FT images of the corresponding dI/dV maps. The white circles highlight one of the CDW peaks and the hexagons highlight the CDW Brillouin zone. At or near the K and K' points of the CDW Brillouin zone, new sets of peaks highlighted by green circles correspond to the moiré pattern. **c,f,i**, Simulated FT images of moiré with relative angles around 1°, 2°, and 3° from HOPG.

the topographies and the corresponding FTs are shown in SI Figure 4. Locally, even when the rotational alignment is broken, the modulation periodicity is still very close to $\sqrt{3}$. However, the phase of this modulation w.r.t. the CDW lattice shifts over longer length scales.

Low energy excitation spectra of 1T-TaSe₂

We probed many-body excitations in monolayer 1T-TaSe₂ by low-energy IETS measurements at 5 K. Inelastic tunnelling on magnetic systems causes spin-flip processes ($\Delta S = 1$), and these additional tunneling channels contribute to the measured dI/dV , which can then be used to probe the fundamental magnetic excitations of the sample.^{30,33,34} The red curve in Figure 3a is a typical

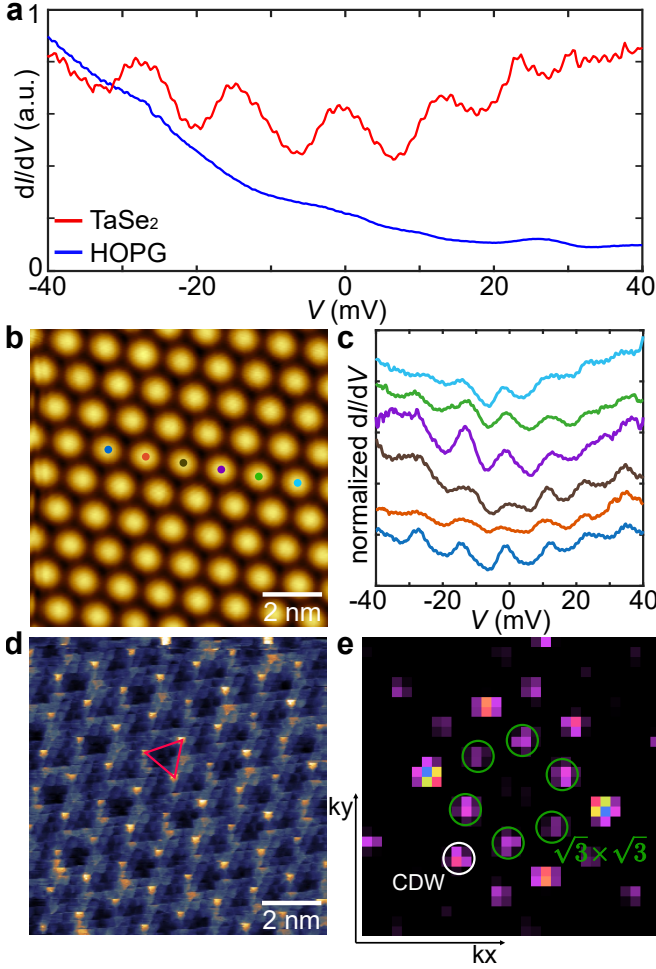


FIG. 3. Low energy inelastic excitations in 1T-TaSe₂. **a**, In-gap inelastic excitations of monolayer 1T-TaSe₂ (red line). Spectrum obtained with the same tip on HOPG as a comparison (blue line). **b**, STM image over several $\sqrt{3}$ moiré cells. $V = -0.5$ V, $I = 100$ pA. **c**, dI/dV at neighbouring CDW centers indicated in panel b. **d**, A detailed constant-height dI/dV map of $\sqrt{3}$ moiré modulation. $V = -15$ mV, $I = 400$ pA. Red triangles highlight the moiré trimerization. **e**, FT of the data in panel d showing the $\sqrt{3} \times \sqrt{3}$ reconstruction.

IETS spectrum collected on monolayer 1T-TaSe₂ and it shows a zero-bias peak and two pairs of symmetric peaks associated with many-body excitations. The blue curve in Figure 3a is acquired on the HOPG substrate with the same tip and it shows no bias-symmetric features as expected. We recorded several IETS spectra at 6 neighboring CDW centers, whose positions are indicated in Figure 3b, and the corresponding spectra are shown in Figure 3c (further data in SI Section 3A and Figure S7). The spectroscopy taken at the different CDW sites consistently shows 5 resonant peaks, including a zero-energy peak, across the different unit cells, so the moiré does not break a symmetry that would lead to inequivalent excitations in the different CDW sites.

Figure 3d shows a constant-height dI/dV map at a

bias of $V = -15$ mV corresponding to one of the resonances in the dI/dV spectra (other low-bias constant-height DOS maps are shown in the SI Figure S8). The dI/dV intensity shows the spatial variation of the intensity of the low-bias spectral features, and it is essentially equal in the middle of the CDW sites. However, there is a clear modulation in between the sites that reflects the $\sqrt{3}$ modulation. This is better visualized in the FT of the map shown in Fig. 3e. This implies a $\sqrt{3} \times \sqrt{3}$ modulation of the interactions between the spins $1/2$ of the CDWs, and therefore will influence the magnetic ground state of the system. These experimental observations put constraints on the symmetry of the model that we will use to interpret the low energy excitations. The modulation corresponds to a trimerization between the sites (highlighted by the red triangle in Fig. 3d), where the 3 sites are equivalent.

Understanding the low energy excitations in 1T-TaSe₂

We now proceed to interpret the low-energy excitations observed in our experiment. 1T-TaS₂ and 1T-TaSe₂ monolayers are potential two-dimensional QSL candidates^{2,10,11,17–20}, due to the intrinsic frustration of the triangular lattice formed by the CDW. Both 1T-TaSe₂ and 1T-TaS₂ monolayers are correlation-driven Mott insulators and show zero net magnetization down to low temperatures^{31,35,36}. The low-energy in-gap excitations thus carry information about the magnetic ground state of 1T-TaSe₂. In particular, inelastic excitations of a Mott state reflect $S = 1$ spin excitations associated with the spin structure factor $A(\omega) = \langle \Omega | S_n^- \delta(\omega - H + G_\Omega) S_n^+ | \Omega \rangle$ of the ground state. Purely magnetic inelastic excitations (e.g. spin-flip excitations) would result in a monotonically increasing dI/dV signal (as $dI/dV \propto$ integrated inelastic spectral function). This is clearly in contrast to our experiments, suggesting the presence of an additional channel, potentially a Kondo contribution³⁰. This would indicate that the zero-bias feature arises from Kondo coupling between the magnetic moments hosted in the CDW unit cells and the underlying HOPG substrate. The finite-bias features then arise from the Kondo-assisted magnetic inelastic excitations of the 1T-TaSe₂ layer.^{37–42} While we cannot directly rule out a magnetically ordered ground state solely on inelastic spectroscopy, the presence of a zero-bias Kondo feature suggests that an ordered state is not present (see SI Section S5 and Fig. S11 for discussion on a moiré modulated magnet). In contrast, QSL ground state can coexist with Kondo screening from the HOPG metallic states^{43,44}, a scenario compatible with our observations. An alternative interpretation that could explain the zero-bias resonance would be a paramagnetic state, where each spin $1/2$ would behave as an independent impurity coupled to the metallic substrate. However, the higher-bias resonances would not be present in this sce-

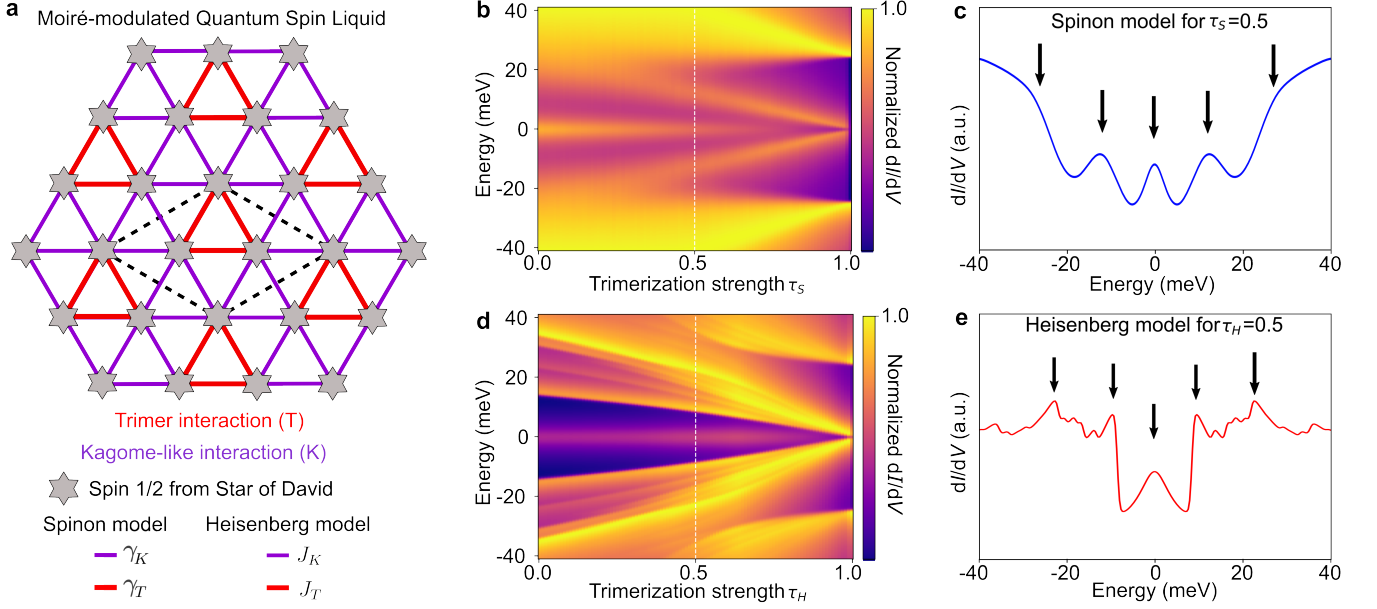


FIG. 4. Theoretical modeling of a moiré-modulated QSL state in 1T-TaSe₂. **a**, Sketch of the spinon and Heisenberg models considered for a moiré-modulated QSL. The grey stars represent localized spins 1/2. The moiré introduces an effective $\sqrt{3} \times \sqrt{3}$ modulation on the triangular lattice, creating a trimerization of the spins 1/2. Spinon and many-body models with two inequivalent coupling parameters in the TaSe₂ lattice. **b,d** Simulated dI/dV as a function of energy and trimerization strength values for the spinon and Heisenberg models. **c,e** Simulated dI/dV with $\tau_S = \tau_H = 0.5$ for spinon model and many-body models, respectively.

nario (see SI Section S13 for a discussion on other alternatives). All this suggests that the observed excitations may stem from a QSL state.

Our experimental observations within the framework of a modulated QSL are rationalized by a Hamiltonian for the CDW spin sites taking the form $H = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$, where the antiferromagnetic exchange coupling J_{ij} follows the $\sqrt{3} \times \sqrt{3}$ experimental modulation created by the moiré (Figure 3d) and each $\vec{S}_n$ is localized in a 13-Ta star of the CDW. Figure 4a shows a schematic of the Hamiltonian model for the modulated Heisenberg magnet. The moiré introduces $\sqrt{3} \times \sqrt{3}$ modulation on the first neighbor magnetic exchange interactions, J_T couples the spins 1/2 in trimers and J_K provides a kagome-like interaction in the limit of $J_T = 0$ (see Fig. S9 for this limit in the SI). We define the strength for the trimerization as $\tau_H = 1 - J_K/J_T$ ($\tau_H = 1$ in the limit of full trimerization). In a QSL state, the spin Hamiltonian can be solved with an auxiliary fermion Abrikosov replacement¹ $S_n^\alpha = \frac{1}{2} \sum_{s,s'} \sigma_{s,s'}^\alpha f_{n,s}^\dagger f_{n,s'}$ with the constraint $\sum_{s,s'} f_{n,s}^\dagger f_{n,s'} = \mathcal{I}$. Using the previous replacement on the spin Hamiltonian with a fixed point approximation leads to an effective spinon Hamiltonian of the form $H = \sum_{ij,s} \gamma_{ij} f_{i,s}^\dagger f_{j,s}$, with $\gamma_{ij} = \gamma_K, \gamma_T$, inheriting the $\sqrt{3} \times \sqrt{3}$ moiré modulation and the trimerization strength as $\tau_S = 1 - \gamma_K/\gamma_T$ (Figure 4a). In the QSL state, the spin structure factor $A(\omega, n)$ reflects two-spinon excitations, and as a result, an effective self-convolution of the spinon spectral function²³. The simulated dI/dV ob-

tained is obtained from including the spin-flip assisted tunneling together with a Kondo contribution (see SI Section S6 for details). We shown in Fig. 4b the simulated dI/dV from the spinon model as a function of the trimerization strength τ_S including a Kondo contribution. As a complementary benchmark, we show in Fig. 4d the siulated dI/dV obtained from the exact solution of the quantum Heisenberg model for a finite lattice of 2D spin clusters as a function of the trimerization strength τ_H . As shown in Figs. 4b and 4d, the ratio of trimerization has a strong effect on the peaks of the simulated dI/dV. From Figures 4c and 4e, at strong moiré modulation (τ_S and $\tau_H \sim 0.5$), the simulated dI/dV for both models match quite well with the experimental data (Figure 3a). These findings thus suggest that the experimental excitation spectrum in 1T-TaSe₂ is consistent with a moiré-modulated QSL model. In particular, the five resonant peaks are well captured by both the spinon and quantum Heisenberg models, pointing out the crucial effect of the moiré modulation.

CONCLUSIONS

In this work, we have shown an emergence of a moiré induced modulation in the QSL candidate 1T-TaSe₂. This moiré pattern plays an important role in modulating the low-energy inelastic excitations. Specifically, the inelastic modes feature a zero bias peak coexisting with

higher energy excitations, and equivalent spectroscopy on all sites forming the $\sqrt{3} \times \sqrt{3}$ reconstructed structure. We demonstrate that these experimental observations are consistent with a $\sqrt{3} \times \sqrt{3}$ -modulated QSL ground state in 1T-TaSe₂. Our work provides a strategy to explore QSL phases with STM, taking advantage of the signatures that moiré modulations can imprint on the excitations of these highly entangled quantum phases.

METHODS

Sample preparation

1T-TaSe₂ was grown by molecular beam epitaxy (MBE) on highly oriented pyrolytic graphite (HOPG) under ultra-high vacuum conditions (UHV, base pressure $\sim 1 \times 10^{-10}$ mbar). HOPG crystal was cleaved and subsequently out-gassed at $\sim 600^\circ\text{C}$ in UHV. High-purity Ta was evaporated from an electron-beam evaporator. Se was evaporated from a Knudsen cell using Se powder (99.9%, Merck). Before growth, the flux of Ta was calibrated on an Au(111) at ~ 1 monolayer per hour. The sample was grown in a Se background pressure of $\sim 1 \times 10^{-8}$ mbar and the growth duration was 30 minutes. Before the growth, the HOPG substrate temperature was stabilized at $\sim 550^\circ\text{C}$.

STM measurements

After the preparation, the sample was inserted into the low-temperature STM (Createc LT-STM) connected to the same UHV system, and subsequent experiments were performed at $T = 5$ K. STM images were taken in

the constant-current mode. dI/dV spectra were recorded by standard lock-in detection while sweeping the sample bias in an open feedback loop configuration, with a peak-to-peak bias modulation specified for each measurement and at a frequency of 757 Hz. For IETS measurement, the modulation is 10mv.

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CONTRIBUTIONS

Z.W., R.D., and P.L. initiated and conceived this project. Z.W., M.A., B.G.A., R.D., and A.C. carried out the STM/STS measurements. Z.W., B.G.A. and M.A. performed the sample growth. A.O.F., A.V.M., and J.L.L. performed the theoretical modelling. R.D. and Y.D. performed coding and data analysis. Z.W. wrote the manuscript with feedback from all the co-authors.

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