

Electronic structure of monolayer CrTe₂: An antiferromagnetic two-dimensional van der Waals material

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Magnetic van der Waals materials are an important building block to realize spintronic functionalities in heterostructures of two-dimensional (2D) materials. However, establishing their magnetic and electronic properties and the interrelationship between the magnetic ground state and electronic structure is often challenging because only a limited number of techniques can probe magnetism and electronic structure on length scales of tens to hundreds of nanometers. Chromium chalcogenides are a class of 2D magnetic materials for which a rich interplay between structure and magnetism has been predicted. Here, we combine angle-resolved photoemission and quasiparticle interference imaging to establish the electronic structure of a monolayer of CrTe₂ on graphite. From a comparison of model calculations with spectroscopic mapping using angle-resolved photoemission spectroscopy and scanning tunneling microscopy we establish the magnetic ground state and the low-energy electronic structure. We demonstrate that the band structure of monolayer CrTe₂ is captured well by density functional theory (DFT) in a DFT + *U* framework when a Coulomb repulsion of *U* = 2.5 eV is accounted for.

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I. INTRODUCTION

The discovery of magnetic two-dimensional (2D) van der Waals materials [1,2] has been a milestone in our ability to create heterostructures with spintronic functionalities. Being able to stack materials with strong spin-orbit coupling and magnetic order or monolayers of superconducting and magnetic materials is a prerequisite to creating spin-valve structures and a route to stabilizing unconventional superconducting order parameters [3]. The chromium tellurides are a family of 2D magnetic materials which show magnetism at room temperature [4,5] for which density functional theory (DFT) predicts a wide range of magnetic ground states dependent on structural details [6,7], including noncollinear magnetic orders [8] and topological magnetism [9,10]. The family also hosts a range of structurally similar compounds in which ferromagnetic and antiferromagnetic ground states are stabilized with comparatively small changes in the stoichiometry [11], an effect also seen in other correlated materials [12]. This range of magnetic ground states makes them an ideal platform to tailor the magnetic order for specific applications. The localized nature of the Cr *d* orbitals suggests that electronic correlation effects beyond DFT are important [13].

Determining their influence on the properties and assessing the validity of the predictions require detailed knowledge of the electronic structure.

Techniques that are able to detect and characterize magnetism in monolayer materials are limited: Magnetization measurements are typically not sufficiently sensitive to pick up a signal from monolayer materials, and neutron scattering requires sample masses on the order of grams. This calls for microscopic techniques that can probe both the magnetism and the underpinning electronic structure on submicron length scales and in atomically thin materials. Spin-polarized scanning tunneling microscopy (STM) is one such technique and can detect magnetism and magnetic order with atomic resolution [14,15]. The ability to relate magnetic properties to the details of the electronic structure—crucial to developing a microscopic understanding and theories that can realistically describe the magnetism—additionally requires the ability to establish the low-energy electronic structure, determined using momentum space techniques. Angle-resolved photoemission spectroscopy (ARPES) is the most commonly used technique for this. Significant progress has been made towards reducing the probing light spot size. Nonetheless, when integrating over multiple flakes or islands, rotational disorder or the presence of multiple domains on small length scales can still make the interpretation challenging.

Here, we demonstrate that the combination of ARPES and quasiparticle interference (QPI) imaging of the 2D van der Waals magnet CrTe₂ enables the determination of the detailed low-energy electronic structure. This is facilitated by a comparison of continuum local density of states calculations for realistic modeling of the QPI and the unfolded spectral function for ARPES with DFT calculations, enabling a

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detailed account of the electronic structure. We show that the electronic structure exhibits clear fingerprints of the magnetic configuration, which depends sensitively on the in-plane lattice constant, as shown by theory [6,7,13,16,17] and experiment [18]. The comparison between calculations and experiment demonstrates that the electronic structure is captured well once a Coulomb repulsion of 2.5 eV for the d orbital of Cr is included in a DFT + U framework.

II. METHODS

A. Growth

To grow monolayer films of CrTe_2 by molecular beam epitaxy (MBE), we employ an optimized growth procedure that uses Ge as a sacrificial species to achieve large-scale growth of monolayer CrTe_2 [19]. Because of the close proximity of the growth windows for CrTe_2 , Cr_2Te_3 , and other intercalated Cr_xTe_y compounds, careful control of the growth temperature is crucial [11]. While monolayer CrTe_2 forms with a lattice constant of around 3.7 Å [11,18,20], thicker films have a lattice constant closer to 3.8 Å, suggesting the formation of the self-intercalated compound Cr_xTe_y [5,11,18]. Here, the growth of CrTe_2 is optimized by *in situ* cross-checking of the lattice constant using reflection high-energy electron diffraction (RHEED).

Monolayer CrTe_2 was synthesized on highly oriented pyrolytic graphite (HOPG) and natural graphite substrates using a DCA R450 molecular beam epitaxy (MBE) system, with a base pressure of approximately 1×10^{-10} mbar. Chromium and tellurium were evaporated from effusion cells at temperatures of 1025 °C and 425 °C, respectively, achieving a Cr-Te flux ratio of approximately 1:200. Before the film growth, the substrates were outgassed in the growth chamber at 800 °C for approximately 25 min. The monolayer CrTe_2 was grown with a substrate temperature of 400 °C for 1 h and subsequently cooled in the presence of Te. To enhance nucleation, we coevaporated small quantities of Ge from an electron beam evaporator during the growth. The incident flux of Ge ions at the substrate surface enhances the nucleation of the epilayer, promoting higher growth rates and improving uniformity of the grown layer [11,19]. *In situ* RHEED was used to monitor surface quality and phase formation throughout the growth process.

We find an in-plane lattice constant of 3.55 ± 0.15 Å for CrTe_2 [compare Fig. 1(a)], slightly smaller than but consistent with the lattice constant of 3.67 ± 0.02 Å found by RHEED [Fig. 1(b)]. Longer growth times result in films with a slightly larger lattice constant, more consistent with Cr_xTe_y variants with larger x compared to CrTe_2 and as found in ferromagnetic Cr_xTe_y films [11]. The linecut in Fig. 1(c) shows that the height of the islands is consistent with monolayer CrTe_2 , which for the first layer has an apparent height of about 1 nm. Although larger than the c parameter of the bulk CrTe_2 of 6.10 Å [21], it is consistent with measurements [18,20] and DFT calculations.

B. DFT calculations

We performed structural relaxation of monolayer CrTe_2 and a graphene layer to estimate the theoretical step height

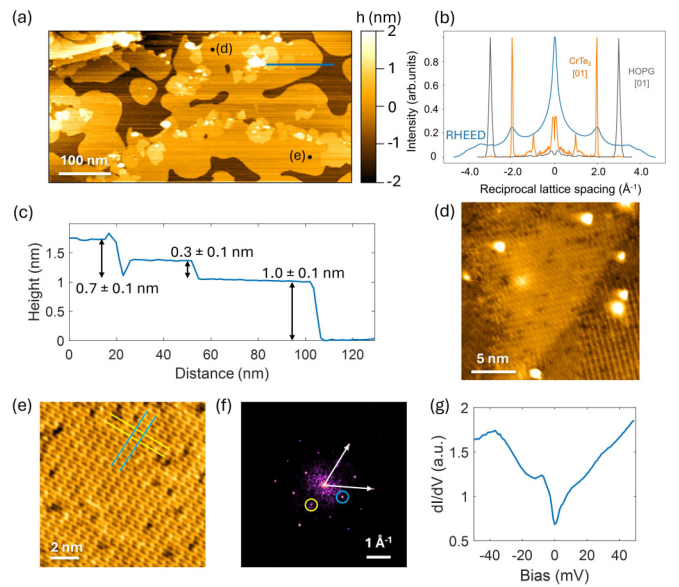


FIG. 1. Growth, structure, and morphology of CrTe_2 films. (a) Large-scale topography of CrTe_2 growth on highly oriented pyrolytic graphite (tunneling set point $V_s = 900$ mV, $I_s = 60$ pA), (b) Linecut from the RHEED pattern (blue) with a linecut through the Fourier transformations (FTs) of topographic images of CrTe_2 (orange) and HOPG (gray) from STM. (c) Linecut along the line shown in the topography in (a) ($V_s = 900$ mV, $I_s = 60$ pA). (d) Topography on a CrTe_2 island showing domains of zigzag/stripe order ($V_s = 200$ mV, $I_s = 200$ pA). (e) Atomic-resolution image of a single domain of the zigzag order ($V_s = 250$ mV, $I_s = 250$ pA) and (f) the corresponding Fourier transformation, with atomic peak positions marked by the white arrows. The stripe and zigzag modulations are shown in yellow and blue. (g) Differential conductance spectrum $g(V)$ of monolayer CrTe_2 ($V_s = 50$ mV, $I_s = 100$ pA, $V_m = 0.8$ mV, average of 256 spectra from a spectroscopic map).

using QUANTUM ESPRESSO [22,23], using the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional [24], projector augmented wave pseudopotentials, and an energy cutoff of 40 Ry = 544 eV. The calculation includes a DFT-D3 correction that includes two- and three-body contributions to the dispersion energy to account for van der Waals interactions [25] but does not include magnetism or a Hubbard U , as they are not expected to significantly affect the structure. During the relaxation, the unit cell parameters and atomic positions are allowed to vary until the force on each atom is less than 1×10^{-3} Rydberg per atomic units = 0.01 eV/Å. For the relaxation, we used a $6 \times 6 \times 1$ k grid and a 30 Å vacuum layer to prevent interactions between adjacent layers.

The relaxed structure has a monolayer thickness of ~ 3 Å and a van der Waals gap of ~ 4 Å. We also estimate the electronic contribution to the island height measured with STM due to the different densities of states of the CrTe_2 and the graphite substrate. To this end, we performed a non-self-consistent field calculation on the relaxed structure with an energy cutoff of 40 Ry and a k grid of $16 \times 16 \times 1$. The density of states integrated over energy in real space is then simulated using the Tersoff-Hamann approximation [26]. This quantity is proportional to the tunneling current. We then calculate its average over the plane parallel to the layers and

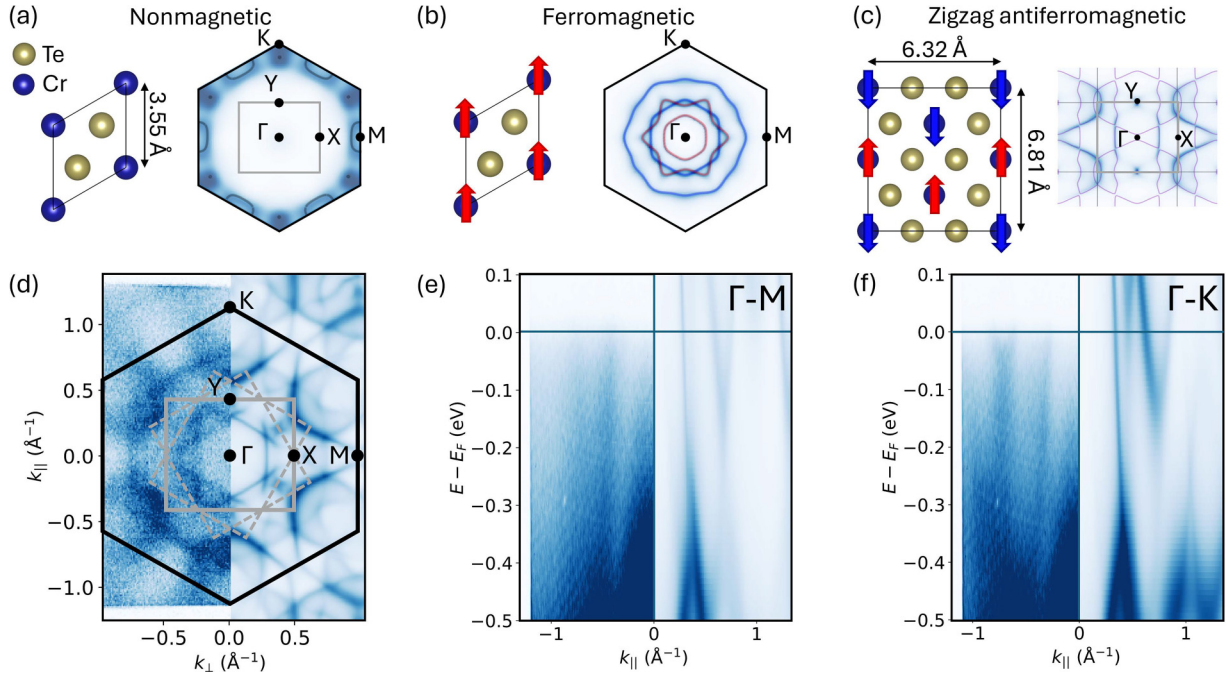


FIG. 2. Electronic structure from ARPES and DFT calculations. (a)–(c) Left: unit cell, right: Fermi surface overlaid on calculated spectral function for (a) nonmagnetic CrTe_2 , (b) ferromagnetic, and (c) zigzag antiferromagnetic (zz-AFM) order, obtained from DFT calculations for a freestanding monolayer. For the ferromagnetic case, (b), blue and red indicate spin majority and minority bands, respectively. The gray rectangle in (a) indicates the zz-AFM Brillouin zone (all calculations with $U = 2.5$ eV). (d) Left: ARPES constant energy contour acquired at the Fermi energy, averaged between $+25$ and -25 meV. Right: unfolded spectral function calculated for the zz-AFM order with an energy broadening of 100 meV and a shift of the chemical potential by 50 meV in the calculation. The spectral function is sixfold symmetrized to account for rotational domains of the zz-AFM order. The hexagonal Brillouin zone is shown in black; the zz-AFM Brillouin zone and its rotated copies are shown in gray. (e) and (f) Cuts along the Γ -M and Γ -K directions from the ARPES data (left half) and spectral function (right half) for the calculation for the zz-AFM order [again symmetrized and including the same shift as in (d)].

plot this as a function of distance from the layer surface. By comparing the distances at which a threshold value is reached between a CrTe_2 monolayer and a graphene layer, we estimate that the measured step height increases by ~ 3 Å. Combining these contributions gives a calculated height of 1 nm for monolayer CrTe_2 .

Further band structure calculations for different values of the Coulomb repulsion U and for projection onto tight-binding models for the QPI calculations were done using VASP with the same structure of a freestanding CrTe_2 layer as in the calculations with QUANTUM ESPRESSO. The calculations were performed using the PBE functional [27–32] with a plane wave energy cutoff of 600 eV and a $\mathbf{k}$ -point grid of $8 \times 8 \times 1$ and using $U = 2.5$ eV unless stated otherwise and $J = 0.3U$. For the continuum Green’s function calculation, we projected the band structure and wave functions onto localized orbitals using a modified version of WANNIER90 to obtain a tight-binding model as well as the localized wave functions to describe the vacuum overlap.

C. STM

STM measurements were performed in a home-built STM that operates in ultrahigh vacuum at a temperature of 1.6 K. Samples are transferred in a vacuum suitcase from the MBE system to the STM and are not usually exposed to a pressure of more than 5×10^{-9} mbar. The bias voltage V is applied to the

sample. Tunneling spectra are acquired in the open feedback loop condition after stabilizing the tip height at set point conditions (V_s, I_s), recording the differential conductance $g(V)$ using a lock-in amplifier with a lock-in modulation V_m added to the sample bias V . Samples are transported under vacuum to the STM. Visibility of both graphite and CrTe_2 in the same STM image allows calibration of the lattice constants between the two. The STM spectrum shown in Fig. 1 is an average of 256 spectra from a spectroscopic map. Cuts through the QPI map shown in Fig. 4(b) below are an average with a width of 5 pixels normal to the linecut. QPI in Fig. 3 below is symmetrized using horizontal and vertical mirror planes.

D. ARPES

ARPES measurements were performed at the nano-ARPES branch line of the I05 beamline at Diamond Light Source, United Kingdom. The light was focused to a spot of ~ 5 μm using a capillary mirror. The data presented in Figs. 2(d)–2(f) were acquired at a photon energy of 62 eV with both linearly horizontal (LH) and vertical (LV) polarizations and are plotted here as the sum of the two spectra measured with each polarization (LH + LV). The measurements were performed at temperatures of 40 and 90 K, well below the magnetic transition temperature of monolayer CrTe_2 [11]. The total energy resolution, including contributions from both the analyzer and the beamline, was

approximately 40 meV. For these measurements, the monolayer CrTe₂ was grown on a natural graphite substrate and transferred from the MBE to the ARPES system via a vacuum suitcase to prevent any exposure to air. Data analysis was performed using the PEAKS package [33].

E. QPI calculations

We performed continuum Green's function calculations using the St Andrews CALCQPI code [34–39], which uses the formalism developed by Hirschfeld and colleagues [40–42] to model the quasiparticle interference from a defect. The QPI was calculated on a real-space grid of 64×64 unit cells using 128×128 k points. For all QPI calculations, we used broadening $\eta = 5$ meV. Due to the two-dimensional nature of the sample, interlayer hopping, which is relevant in bulk samples [35,43], does not result in additional broadening here. The intensities of the QPI patterns depend on the precise impurity potential (compare Fig. 7 in Ref. [44] for calculations with different defect positions and Fig. 8 with positive potential).

F. Unfolded spectral function

To obtain the unfolded spectral function from a tight-binding model, following Ref. [45], we calculate

$$A(\mathbf{k}, \omega) = -\frac{1}{\pi} \sum_{\mu, \nu} e^{i\mathbf{k}(\mathbf{r}_\mu - \mathbf{r}_\nu)} \text{Im} G_0^{\mu\nu}(\mathbf{k}, \omega), \quad (1)$$

where μ and ν are orbital indices and $\mathbf{r}_\mu$ are the positions of orbital μ . For the unfolded spectra function, calculations were done on a $\mathbf{k}$ grid of 512×512 points for the ferromagnetic and nonmagnetic calculation distributed across the Brillouin zone; for the antiferromagnetic calculation, 128×128 points were considered on a uniform grid in the Brillouin zone.

III. RESULTS

A typical topographic image following the growth of CrTe₂ on HOPG is shown in Fig. 1(a), with monolayer high islands and only very low coverage of the second layer. The regions between the islands are the bare graphite substrate. Visibility of both graphite and CrTe₂ in the same image allows calibration of the lattice constants between the two, giving a lattice constant of 3.55 ± 0.15 Å for the CrTe₂ islands, slightly smaller than but consistent with the lattice constant found by RHEED for the CrTe₂ layer, which is 3.67 ± 0.02 Å [Fig. 1(b)].

The linecut in Fig. 1(c) shows that the height of the islands is consistent with monolayer CrTe₂, which for the first layer has an apparent height of about 1 nm. Although larger than the c parameter of the bulk CrTe₂, which is 6.10 Å [21], this height is consistent with previous STM and scanning transmission electron microscopy measurements [18,20]. We compare this to the apparent step height calculated using DFT, which gives a value for the height of a monolayer of 1 nm (see Sec. II B for details), in good agreement with the experimental value.

Images acquired on individual islands reveal a zigzag order superimposed on the atomic resolution [Figs. 1(d) and 1(e)], consistent with previous studies and suggesting a zigzag

antiferromagnetic (zz-AFM) order [18,20]. The magnetic ground state of monolayer CrTe₂ is predicted to depend sensitively on the lattice constant [6,7,13,16,17]. For the value measured here, the ground state is expected to be antiferromagnetic. The zigzag order was previously identified as an antiferromagnetic order from spin-polarized STM [18,20] with a Néel temperature of $T_N \sim 140$ K estimated from photoemission [11]. It has twice the periodicity of CrTe₂ and breaks the sixfold rotational symmetry [see Fig. 1(f)]. We observe multiple domains of the order within patches of ~ 20 nm [as shown in Fig. 1(d)]. An additional stripe order, with the same periodicity as the zigzag order but in the perpendicular direction, is also present. In agreement with previous studies [18,20], we attribute this order to a small distortion of the lattice (see Fig. 1 in Ref. [44]). Spatially averaged tunneling spectra recorded in the vicinity of the Fermi energy show that the islands are metallic [11], with a dip in the spectra close to zero bias and a peaklike feature at -40 mV [Fig. 1(g)].

To establish a detailed understanding of the electronic structure, we performed DFT + U calculations for nonmagnetic, ferromagnetic, and zigzag antiferromagnetic configurations of a freestanding monolayer of CrTe₂. For all the magnetic configurations investigated here (nonmagnetic, ferromagnetic, and antiferromagnetic), we find Fermi surfaces with multiple bands, predominantly of Te character, crossing the Fermi energy (Figs. 2(a)–2(c), all with $U = 2.5$ eV; see also Figs. 2(a), 2(b), and 2(f) in Ref. [44]). The electronic structure for the antiferromagnetic configuration exhibits significant differences from the ferromagnetic and nonmagnetic ones, not least because of the different symmetry of the Brillouin zone [shown in Fig. 2(a)].

The Fermi surface as measured by angle-resolved photoemission of monolayer CrTe₂ on natural graphite [Fig. 2(d)] confirms the metallic character. It has apparent sixfold symmetry and consists of a sharper inner hexagon surrounded by triangular features and a larger broader hexagon, both of which are centered around the Γ point. Around the K points in the hexagonal Brillouin zone there are broader pockets with high intensity. The last two features can also be seen in measurements taken with a higher photon energy (Fig. 3 in Ref. [44]), as well as some intensity at the corners of the inner hexagon.

Comparison with the calculations shows poor agreement with the nonmagnetic case because there are no features in the center of the Brillouin zone for such a calculation: The Fermi surface in Fig. 2(a) consists of small pockets around the M and K points, and the intensity of the calculated spectral function is highest around the zone edges. In the ferromagnetic case, all bands are centered around the Γ point [Fig. 2(b)], again inconsistent with the ARPES measurement, which also shows pockets enclosing the K points. The zz-AFM configuration, shown in Fig. 2(c), enlarges the CrTe₂ unit cell and therefore reduces the size of the Brillouin zone. For an improved comparison with ARPES, we unfolded the band structure obtained from DFT to calculate an unfolded spectral function (see Sec. II F for details), but neglecting, e.g., self-energy effects beyond an energy-independent broadening. The most notable effect of the unfolding is that the bands closest to the Γ point retain very little spectral weight: Their dominant spectral weight is in higher-order Brillouin zones.

The antiferromagnetic configuration also changes the symmetry of the unit cell from six- to twofold. The Fermi surface measured with ARPES shows clear hexagonal symmetry. However, because the $\sim 5 \mu\text{m}$ spot size of the incident beam is much larger than both the magnetic domain size and the island size [as shown in Figs. 1(d) and 1(a)], photoelectrons will stem from multiple islands on which the magnetic order forms independently, thus averaging over different domains of the zigzag antiferromagnetic order. To compare the DFT calculations and the ARPES data, we therefore account for the magnetic domains of the *zz*-AFM order by symmetrizing the calculation, adding copies of the unfolded spectral function rotated by 60° and 120° . Figure 2(d) shows the spectral function obtained after such a symmetrization (with $U = 2.5 \text{ eV}$) in comparison with the Fermi surface obtained from ARPES. The calculated spectral function shows very good agreement: The inner hexagon and triangles and the higher-intensity arcs near the *K* points are consistently reproduced in the calculations.

To assess the impact of electron correlations, we compared the data with calculations performed with a range of U values between 0 and 6 eV. While the precise value of U has relatively little influence on the band structure, there are notable differences between $U = 0 \text{ eV}$ and nonzero values of U . For example, an additional Van Hove singularity, which is not observed experimentally, is found below the Fermi energy at the Γ point for $U = 0 \text{ eV}$ (see Fig. 2 in Ref. [44]). The inclusion of the U term also causes the Cr-derived bands to be pushed away from the Fermi energy. The results of the spectral function calculations demonstrate that the inclusion of a nonzero U value improves the agreement with experiment (see Fig. 4 in Ref. [44] for comparisons with various U values). While key features of the ARPES data are recognizable for all values of U , their sizes and locations vary with the precise value. We find a U value of around 2 to 3 eV produces the results most consistent with the experimental data and use $U = 2.5 \text{ eV}$ for all comparisons and calculations. We also allow for an energy shift in the calculations for comparison with the data to account for charge transfer between the substrate and the CrTe₂ monolayer, which differs slightly for each U value used in the calculation. In the case of $U = 2.5 \text{ eV}$, the results of the calculations are shifted upwards in energy by 50 meV, resulting in good agreement with the ARPES measurements. In contrast, the nonmagnetic and ferromagnetic spectral functions with $U = 2.5 \text{ eV}$ [Figs. 2(a) and 2(b)] are very different from the experimental Fermi surface. Notably, while a U term is required to account for correlations on the Cr site, comparison of the DFT band structure with ARPES shows that no further renormalization of the band structure is required (compare Fig. 5 in Ref. [44]). This is likely a consequence of the states crossing the Fermi energy being dominantly Te-derived states.

This agreement of the calculated spectral function with ARPES for a *zz*-AFM configuration is supported by comparing linecuts through the symmetrized spectral function with the measured photoemission dispersions [Figs. 2(e) and 2(f)]. Holelike bands from the inner region of the Fermi surface and the broader outer hexagon are visible in both experiment and calculation. The features around the *K* points in Fig. 2(d) are much sharper in the spectral function calculation, but the band minimum near the Fermi energy in the Γ -*K* direction

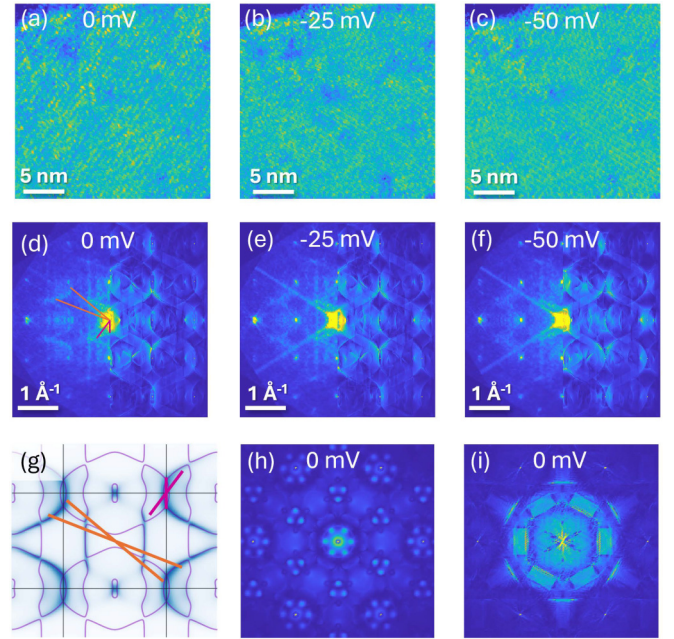


FIG. 3. Quasiparticle interference imaging of CrTe₂. (a)–(c) Real-space differential conductance images of spectroscopic map layers at (a) 0 mV, (b) -25 mV , and (c) -50 mV . (d)–(f) Left: Fourier transform of (a)–(c) symmetrized using two mirror planes (horizontal and vertical). Right: Fourier transforms of continuum local density of states (CLDOS) calculations at the same energies for a *zz*-AFM unit cell (with a negative scattering potential, $U = 2.5 \text{ eV}$, and including a 50 meV energy shift). Map set point: $V_s = 100 \text{ mV}$, $I_s = 200 \text{ pA}$, lock-in amplitude = 4 mV. The diagonal lines are artifacts of the Fourier transform. (g) Scattering vectors from (d) on a constant energy contour and unfolded spectral function calculation, both at the Fermi energy (with $U = 2.5 \text{ eV}$ and including a 50 meV energy shift). (h) and (i) Fourier transforms of CLDOS calculations at 0 meV with a positive scattering potential and $U = 2.5 \text{ eV}$ for a (h) nonmagnetic unit cell and (i) ferromagnetic unit cell.

[Fig. 2(f)] is broadly consistent with a similar feature in the measured ARPES data. Comparison with the spectral function and the band structure (Fig. 2 in Ref. [44]) confirms the holelike character of the bands.

We use QPI imaging to further refine the electronic structure. QPI allows selecting monodomain areas in the sample, such as the one shown in Fig. 1(e), and therefore facilitates disentangling the electronic structure within an individual domain. The QPI obtained from a spectroscopic map is shown in Fig. 3. In Figs. 3(a)–3(c), we show the real-space maps of the QPI, and in the left halves of Figs. 3(d)–3(f) their Fourier transforms (after symmetrization using horizontal and vertical mirror planes) are shown. The Fourier transform reveals four sharp peaks, suggesting a rectangular symmetry of the Brillouin zone and distinct patterns from the quasiparticle interference itself. The QPI patterns disperse strongly in energy, confirming their electronic origin. The right halves of Figs. 3(d)–3(f) show Fourier transforms of QPI patterns calculated using the continuum Green's function approach (see Sec. II E for details) based on the electronic structure with *zz*-AFM order using a Hubbard U term of 2.5 eV and an energy shift of 50 meV, as obtained from the comparison with

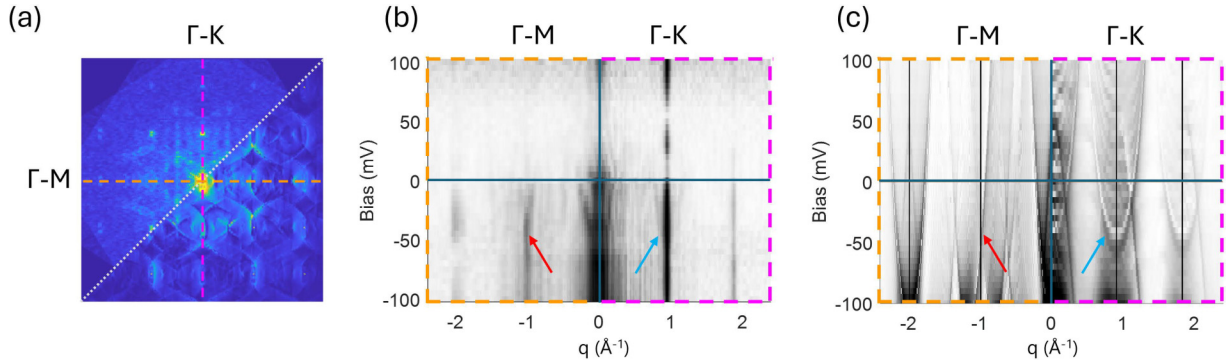


FIG. 4. Comparison of QPI and DFT. (a) QPI layer $\tilde{g}(\mathbf{q}, eV)$ (upper left half) and continuum local density of states (CLDOS) calculation (lower right half) based on the tight-binding model adjusted to the ARPES data. Pink and orange dashed lines show the high-symmetry directions of the Brillouin zone. (b) Cut through the measured STM data and (c) the CLDOS calculation using the optimized model for the electronic structure (see text) along the same path in reciprocal space, including an energy shift of 50 meV. Corresponding dispersions are marked with red and blue arrows. Map set point: $V_s = 100$ mV, $I_s = 100$ pA, lock-in amplitude = 4 mV. Cuts shown in (b) are averages of five slices around the center of the image.

the ARPES measurements. The choice of U value from this comparison is supported by the good agreement here between experimental and calculated QPI when compared with other U values (shown in Fig. 6 in Ref. [44]). The arcs in the Fourier transform are consistent with the shape of the zz -AFM Fermi surface, as indicated by the scattering vectors in Figs. 3(d) and 3(g). These regions of the Fermi surface also have the highest intensity in the calculated spectral function and correspond to the pockets around the K points of the hexagonal Brillouin zone in the ARPES measurements [Fig. 2(d)], enabling the direct relation of QPI and ARPES. We note that details of the scattering patterns also depend on the precise impurity potential (compare Fig. 7 in Ref. [44] for calculations with different defect positions and Fig. 8 with positive potential). The charge transfer is expected to be substrate dependent, providing a possible route to also tune the ground state. The magnetic ground state is the same between graphene on silicon carbide and HOPG as substrates [11,20].

For the quasiparticle interference patterns expected for the three ground states introduced in Fig. 2, the symmetry of the QPI patterns for the nonmagnetic and ferromagnetic states [Figs. 3(h) and 3(i)] is notably different compared to the zz -AFM state due to the symmetry of the magnetic order. This shows that comparing the simulated QPI not only enables us to determine the low-energy electronic structure but also provides information about the unit cell and hence magnetic order.

A detailed comparison of the experimental and calculated QPI for the zz -AFM order is shown in Fig. 4. We consider cuts along the high-symmetry directions, as indicated in Fig. 4(a), from both the experiment [Fig. 4(b)] and calculations [Fig. 4(c)]. Key features from the cuts can be seen in the calculations, as marked by arrows, and correspond to the arc features moving towards the Bragg peaks as a function of energy. The minimum of these features in the Γ - M direction in Fig. 4(c) corresponds to the minimum of the electronlike band in ARPES at about -100 meV [see Fig. 2(f)] and where the pockets around the K points in the ARPES data disappear and become incoherent. The calculations clearly show more and sharper features than observed experimentally. This is likely

a consequence of neglecting electronic correlation effects in the calculations, which in the experiment result in an energy-dependent finite lifetime and decoherence of the electronic states. We note that our calculations also account for scattering only from the Cr site, whereas in reality, there will also be defects on the Te sites resulting in different intensities of the scattering patterns (see Fig. 7 in Ref. [44] for comparison with scattering from a defect on the Te sites). Overall, however, key features are reproduced, with no band renormalization required, consistent with the analysis of the ARPES data shown above.

IV. DISCUSSION

The comparison between band structure calculations, QPI modeling, ARPES, and STM allows us to identify the electronic structure of the monolayer as that of zigzag antiferromagnetically ordered CrTe_2 and hence confirms the ground state also seen in spin-polarized STM studies of monolayer CrTe_2 [18,20]. This confirms the importance of the U term of $U = 2.5$ eV, lower than values derived from linear response methods [6,46], required to accurately model the electronic structure. The need for including this U term suggests that electron correlations cannot be neglected and that they also play an important role for the magnetic properties. Nonetheless, the relatively small U value which we find gives the optimum comparison to the experimental data points for the strong Cr-ligand hybridization, in line with other studies of Te-based 2D magnets [47,48]. The magnetic phase diagram with small structural distortions has already been found to be very rich from DFT calculations performed without inclusion of a U term [9]. This structural sensitivity of the magnetic ground state is further confirmed by the change from antiferromagnetism in monolayer CrTe_2 to ferromagnetism in the intercalated compound [11,18], highlighting the sensitivity and hence tuneability of the magnetic ground state with the in-plane lattice constant [18]. Our work establishes CrTe_2 as a promising platform to study the interplay between magnetism and correlations in two dimensions. Inclusion of spin-orbit coupling, which is sizable for the Te p bands, does not have

a significant impact on the electronic structure close to the Fermi energy here, so it can be neglected in the comparison between theory and experiment for the bands close to the Fermi energy (compare Fig. 9 in Ref. [44]). Our results further establish QPI as a tool to study the electronic structure of monolayer transition metal dichalcogenides and show that combination with continuum QPI calculations and comparison with ARPES allow extraction of detailed information about the low-energy electronic band structure in these systems. QPI is particularly useful here as a complementary tool to angle-resolved photoemission because of the possibility of selecting individual magnetic domains. The electronic structure we obtain is remarkably consistent with the prediction by DFT for a freestanding monolayer. While a non-negligible Coulomb repulsion U must be included, it is significantly smaller than required to describe the electronic states, e.g., in chromium oxides [49,50]. This is likely because the electronic states close to the Fermi energy have dominant Te character, whereas the Cr bands are pushed away from the Fermi energy by the Coulomb repulsion.

V. CONCLUSION

Through the combined application of angle-resolved photoemission and quasiparticle interference imaging, we can resolve the low-energy electronic structure of 2D magnetic systems. Detailed comparison with calculations shows that correlation effects are important in this system, reflected in a non-negligible U term required to obtain good agreement between calculations and experiment. This suggests that modeling of magnetism will also require correlation effects

to be taken into account, given the sensitivity of the magnetic ground state to structural details. The detailed understanding of the electronic structure opens the door to a designer approach to 2D magnetism. More generally, our work demonstrates how the combination of ARPES, QPI, and band structure calculations can be used to unravel the low-energy electronic structure and ground state of transition metal dichalcogenides beyond just 2D magnetic materials in exquisite detail, promising a detailed and deep understanding of their structure-property relationships.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [51].

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