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Magnetic Stability, Fermi Surface Topology, and Spin-Correlated Dielectric Response in Monolayer 1T-CrTe₂

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We have carried out density-functional theory (DFT) calculations to study the magnetic stability of both ferromagnetic (FM) and anti-ferromagnetic (AFM) states in monolayer 1T-CrTe₂. Our results show that the AFM order is lower in energy and thus is the ground state. By tuning the lattice parameters, the AFM order can transition to the FM order, in good agreement with experimental observation. We observe a commensurate Spin Density Wave (SDW) alongside the previously predicted Charge Density Wave (CDW), and attribute the AFM order to the SDW. The SDW order leads to distinct hole and electron Fermi pockets and a pronounced optical anisotropy, suggesting quasi-one-dimensional behavior in this material.

1 Introduction

The discovery of long-range magnetic order in two-dimensional (2D) CrI₃ down to the monolayer threshold in 2017 has ignited a surge of interest in exploring the magnetic characteristics of 2D materials.¹ This pioneering discovery, coupled with subsequent observations of 2D magnetism in various materials, has paved the way for a plethora of potential applications in the realm of spintronics. These applications are particularly enticing due to the potential of 2D materials to serve as energy-efficient alternatives to traditional electronic devices.² Some spintronics applications, to name a few, include spin valves and spin field-effect transistors.³ Additionally, 2D magnets have recently found applications in developing neuromorphic computing architectures.^{4,5}

To truly revolutionize next-generation spintronics with 2D magnetism, we need to find magnets with critical temperatures that are robust enough to withstand ambient conditions. A challenge lies in the fact that many experimentally synthesized 2D magnets exhibit critical temperatures significantly below room temperature, both in their ferromagnetic (FM) and antiferromagnetic (AFM) states. This has been a significant roadblock in realizing the full potential of these materials in practical applications. For instance, the FM magnet CrI₃ boasts a Curie temperature (T_C) of 45 K, while Cr₂Ge₂Te₆ has a T_C of approximately 66 K.^{1,6} On the higher end, FM order has been reported at room temperature in MnSe_x films but the various T_C experimental values have not been determined.⁷ Nonetheless, the T_C of MnSe₂ was theoretically estimated at 225 K which can be further increased by

applying strain to reach 330 K.⁸

On the other hand, AFM phases have been reported in the 2D phosphorus trisulfide magnets NiPS₃, FePS₃, and MnPS₃ with reported Néel temperatures (T_N) of 150 K, 118 K, and 78 K, respectively.^{9–11} More importantly, quasi-dimensional behavior was reported in the trisulfide magnets due to significant thermal and optical anisotropies^{12–15}. Also, the Van der Waals antiferromagnet CrSBr ($T_N \approx 132$ K) has been shown to be quasi-one-dimensional through anisotropies in effective mass and dielectric screening.¹⁶ These recent experimental observations suggest that the 2D transition metal antiferromagnets would be an excellent platform for studying spin-correlated quantum phenomena in low-dimensional materials down to near the one-dimensional limit.

1T-CrTe₂, which in its non-magnetic phase, crystallizes in the trigonal omega-structured $\bar{P}3m1$ space group, is an ideal candidate for spintronics applications. This is due to the fact that FM and AFM phases persisting up to room temperatures have been reported in 1T-CrTe₂.^{17,18} In the ferromagnetic state, CrTe₂ has been shown to have the lattice parameters $a_1 = a_2 = 3.81$ Å.¹⁸ While in the AFM order, CrTe₂ has been shown to have lattice parameters of $a_1 = 3.7$ Å and $a_2 = 3.4$ Å.¹⁹ This indicates that the lattice parameters and the lattice symmetry play a significant role in the realized magnetic phase.

Many theoretical studies based on density functional theory (DFT) have been dedicated to studying the magnetic order in CrTe₂. A switch between the AFM and FM phases in CrTe₂ monolayers due to strain and variations in lattice parameters have been predicted.^{20–23} Another DFT study has concluded that the monolayer and multilayered CrTe₂ up to 6 layers prefer an AFM ground state and that an AFM to FM transition occurs as the number of layers increases.²⁴ However, in these studies, the dynamical stability of the proposed FM and AFM orders with re-

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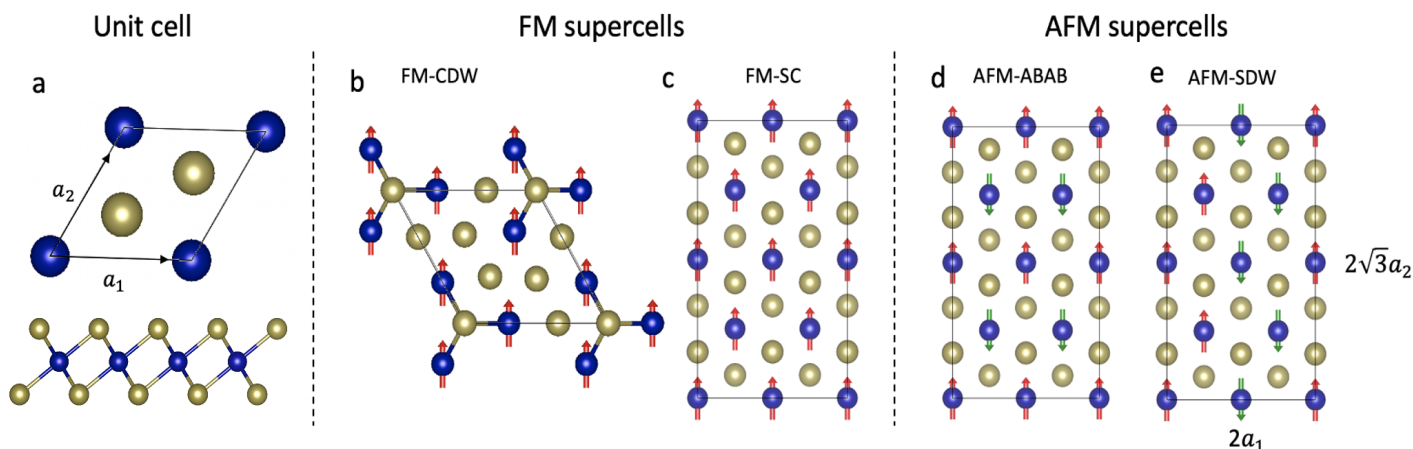


Fig. 1 The unit cell of 1T-CrTe₂ along with the supercells used in the DFT calculations. The Cr atoms are shown in blue while the Te atoms are shown in gold. a) The unit cell and the basis vectors a_1 and a_2 are shown on the top and a side view of the 1T phase is shown on the bottom. b) The supercell of the CDW phase (FM-CDW). c) The supercell of the FM phase (FM-SC). d) The supercell of the abab AFM order (AFM-ABAB). e) The supercell of the SDW phase (AFM-SDW).

spect to phonon dispersions was not considered. Interestingly, by considering the dynamical stability, the emergence of a charge density wave (CDW) state has been theoretically predicted.^{25,26} The emergence of this CDW order is not surprising as CDW states have been experimentally observed in other metal dichalcogenides.^{27–30} Furthermore, compounds that develop a CDW state can develop a spin density wave (SDW) state depending on the total spin value.^{29,31} And co-existing CDW and SDW phases have been experimentally observed in bulk Chromium.³²

Herein, we carried out DFT calculations to understand the contrasting magnetic behavior of 1T-CrTe₂ at the monolayer limit. Our calculations are not only motivated by understanding the peculiar FM and AFM phases of 1T-CrTe₂ but also the implications of the onset of long-range magnetic order on the dynamical, electronic, and optical properties. Hence, we started by constructing supercells corresponding to multiple suggested FM and AFM magnetic orderings in the literature and eliminated dynamically unstable supercells. We then varied the lattice parameters of the dynamically stable FM and AFM phases and compared them in terms of energetic favorability to obtain a phase diagram of the possible magnetic states. A major discovery in our research is that stable FM orders are associated with CDW phases, while stable AFM orders correspond to SDW phases. Moreover, we found that the Fermi surface corresponding to each stable phase evolves in a systematic manner. Finally, we showed that Fermi nesting features are responsible for stabilizing the AFM phase and lead to a highly anisotropic optical response suggesting quasi-one-dimensional behavior.

2 Computational Methods

We performed DFT calculations using the projected augmented wave (PAW) method as implemented in the Vienna ab initio Simulation Package (VASP).^{33,34} In our calculations, we adopted the Perdew-Burke-Ernzerhof (PBE)³⁵ flavor for the generalized-gradient exchange-correlation functional (GGA).

The Brillouin zone was sampled using a $9 \times 5 \times 1$ k -point grid

mesh³⁶ for the rectangular cells and a $9 \times 9 \times 1$ mesh was used for the parallelogram-shaped cell. Additionally, A 550 eV plane wave cutoff energy was used. A vacuum layer of more than 18 Å was applied along the z direction to minimize the interactions between images of layers. Each structure was relaxed till the Hellman-Feynman forces on each atom were less than 2×10^{-3} eV/Å and the energy convergence criterion was set to 10^{-8} eV.

In the unit cell, both the lattice parameters are equal in magnitude so that $a_1 = a_2$ as shown in Fig. 1. Previous DFT calculations have shown that this ferromagnetic unit cell is not dynamically stable and exhibits large imaginary phonon modes^{24,37} unless a Hubbard U parameter is used.³⁷ In our calculation, two FM states and two AFM states were considered using four different supercells, namely FM-SC, FM-CDW, AFM-ABAB, and AFM-SDW. The supercells have a rectangular shape and the size of each cell is $2a_1 \times 2\sqrt{3}a_2$ except for the parallelogram-shaped FM-CDW supercell having a size of $\sqrt{3}a_1 \times \sqrt{3}a_2$. The supercells and the orientations of the magnetic moments of the Cr atoms are shown in Fig. 1.

Naturally, the FM supercells have all magnetic moments oriented in the same direction. The magnetic moments' orientations in Fig. 1(e) are typically referred to as AFM-Zigzag since the magnetic moments of neighboring Cr chains parallel to the y -direction form a zigzag pattern. The magnetic moments' orientations alternate in the Cr atom chains parallel to the x -direction in Fig. 1(d) and hence the name AFM-ABAB.

For the computation of electronic band structure and density of states, we employed the Local Modified Becke-Johnson (LMBJ) meta-GGA functional. Default parameter values for α , β , e , σ , and r_s^{th} , as implemented in VASP, were utilized.³⁸ The choice of the LMBJ functional is due to its efficacy in accurately calculating band gaps of 2D materials. This reliable performance minimizes the likelihood of incorrect metallic predictions while maintaining a reasonable computational cost.^{39,40}

Table 1 The calculated lattice parameters, total energy E_t , and the energy compared to the ground state energy for AFM and FM states, respectively.

	Lattice Parameters		Magnetic Stability		Magnetic Moment
	a_1 (Å)	a_2 (Å)	E_t (eV/formula)	$E-E_{\text{ground}}$ (meV/formula)	μ_B/Cr
FM-SC	3.70	3.70	-16.26	20	2.75
FM-CDW	3.70	3.70	-16.28	0	2.72
AFM-ABAB	3.70	3.54	-16.30	40	2.67
AFM-SDW	3.59	3.60	-16.34	0	2.64

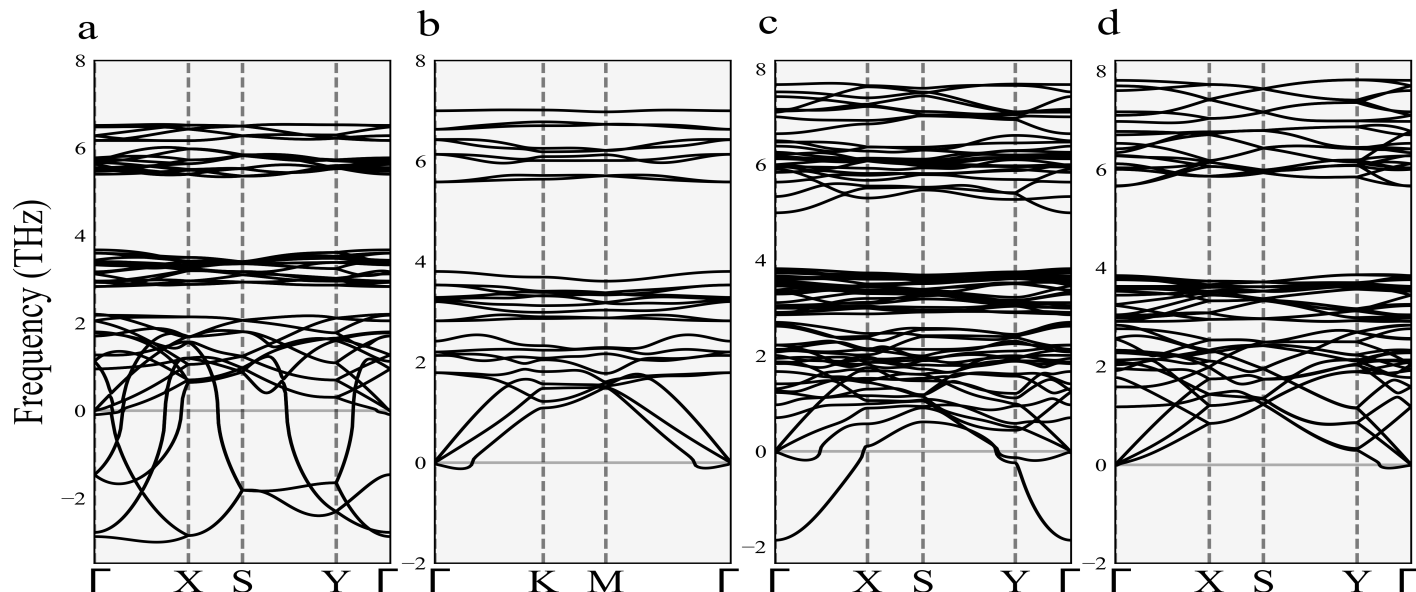


Fig. 2 The phonon band dispersions of the considered FM and AFM states. (a) FM-SC, (b) FM-CDW, (c) AFM-ABAB, and (d) AFM-SDW.

3 Results and Discussions

3.1 Magnetic & Dynamical Stability

Upon optimizing the lattice structures, we attained the optimized parameters for the four different supercells, summarized in Table 1 alongside total energies and magnetic moments per Cr atom.

By examining the FM structures, which have consistent lattice parameters of $a_1 = a_2 = 3.7$ Å, it was found that the FM-CDW state is a 20 meV lower in energy per formula than the FM-SC state. Furthermore, it is dynamically stable against FM-SC, as evidenced by the absence of large imaginary phonon modes in its phonon band dispersions (Fig. 2). The structure stability in the CDW phase is attributed to reconstruction of the atomic distances leading to two short and four-long Cr-Cr bonds. This aligns with previous literature which reported that the CDW is the ferromagnetic ground state in monolayer CrTe₂.²⁵

The AFM states, however, show differing lattice parameters. The AFM-ABAB supercell displays $a_1 = 3.7$ Å and $a_2 = 3.54$ Å, while the AFM-SDW showcases $a_1 = 3.59$ Å and $a_2 = 3.6$ Å. The AFM-SDW state, with 40 meV lower in energy per formula than the AFM-ABAB state, emerges as the energetically most favorable. This state also displays dynamic stability, unlike the AFM-ABAB supercell which shows large imaginary phonon modes. Therefore, the AFM-SDW state is the most stable AFM ground state of CrTe₂. The stability in this case is attributed to the reconstruction of the atomic distances so that the Cr-Cr bonds on the same diagonal are

equal in length. The slightly negative imaginary phonon modes in the phonon dispersions of the FM-CDW and AFM-SDW supercells are attributed to numerical instabilities at the long-range limit in the acoustic branches possessing the lowest frequency near the Γ point.^{41–44}

Based on our calculations, we concluded that among the considered FM and AFM configurations in this study, the AFM-SDW state is the magnetic ground state for the monolayer. This result is in agreement with the recent observation of Zigzag AFM order in the monolayer CrTe₂.^{19,45} Nonetheless, several studies also reported FM ordering in bulk and low-dimensional CrTe₂ albeit with lattice parameters that are larger than the reported lattice parameters of the AFM state.^{17,18,46–48}

Many DFT studies have explored how changes in lattice parameters can lead to a switch between FM and AFM states in various 2D magnetic materials.^{21,24,49,50} To further clarify the magnetic order of monolayer CrTe₂ and whether it is possible to tune its magnetic order, we perform self-consistent calculations with different lattice parameters varying between 3.4 Å and 3.8 Å using the FM-CDW and AFM-SDW supercells. This diagram can be interpreted as depicting the effect of different types of strain on the magnetic order.

The obtained energies with respect to the length of lattice vectors a_1 and a_2 are compared and a phase diagram of the magnetic transitions is obtained. Fig. 4 shows the preferred magnetic state according to the lattice parameter values. In the red region, the

AFM-SDW is more energy-efficient. In contrast, the FM-CDW is the preferred state in the blue region. It is necessary to point out that the only considered AFM phase is due to an SDW since it allows for the emergence of AFM order in metallic systems^{51–54}. This is also supported by the fact that this AFM-SDW phase is dynamically stable.

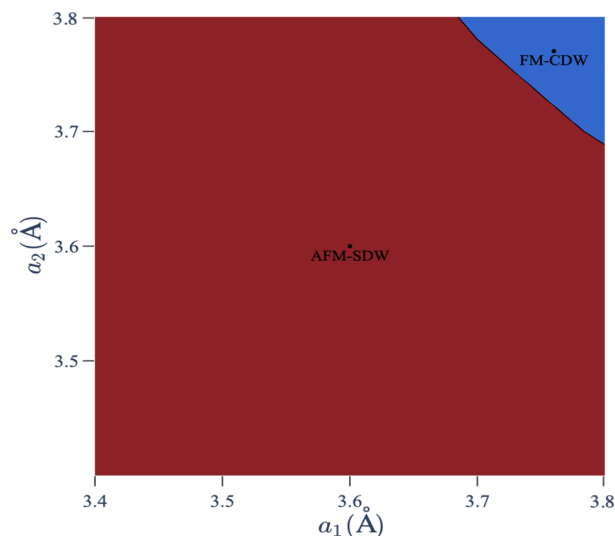


Fig. 4 The phase diagram of the switch between AFM to FM with respect to lattice constants a_1 and a_2 . The ticks on the y-axis and x-axis show the values for which the energy difference was calculated. The rest of the values were interpolated to produce the phase diagram.

This phase diagram is in excellent agreement with the reported lattice parameters of the monolayer. Specifically, in the AFM phase, the experimentally observed lattice parameters of the monolayer are $a_1 = 3.7 \text{ Å}$ and $a_2 = 3.4 \text{ Å}$.¹⁹ In the FM phase, the experimentally observed lattice parameters of the monolayer are $a_1 = a_2 = 3.81 \text{ Å}$.¹⁸ In the phase diagram we notice that the ferromagnetic phase tends to favor larger lattice parameters. The tendency of the ferromagnetic phase to prefer larger lattice parameters is intuitive to understand since neighboring magnetic moments align in parallel. In contrast, antiferromagnetic order, where magnetic moments can form alternating patterns, shows a tendency towards smaller lattice parameters.

3.2 Charge & Spin Density Waves

Commensurate and incommensurate density waves in chromium and its alloys have been subject to multiple extensive theoretical and experimental studies. Chromium's anti-ferromagnetic behavior below its Néel temperature is attributed to the formation of a SDW.^{55–62} More recently, a SDW in chromium has been observed in real space through spin-polarized scanning tunneling microscopy (SP-STM).³²

In non-metals, antiferromagnetism is usually addressed with spin exchange interaction models. Such models are successful at describing interacting localized magnetic moments in non-metals. However, the situation is more fluid in metals where itinerant electrons can lead to non-localized magnetic moments; when a metallic system develops a commensurate SDW, the system tran-

sitions into an AFM state⁶³ and multiple models have been proposed to explain SDW phases using both electron gas and tight-binding approximations.^{64–68} To show the structure of the aforementioned density waves, we plot the charge density of the FM-CDW supercell and the spin densities of the AFM-SDW supercell as shown in Fig. 3. These plots simulate STM (Scanning Tunneling Microscopy) and SP-STM (Spin-Polarized Scanning Tunneling Microscopy) images of the CDW and SDW phases, respectively. The simulated images were generated using a constant height scan of 3 Å with the exclusion of the effect of a bias voltage.

In the simulated STM image, the charge density forms hexagonal concentrations due to the overlap of three distinct incommensurate (with respect to the unit cell) CDW's. The directions of the three charge density waves are shown in Fig. 3(a) by vectors $\vec{a}, \vec{b}, \vec{c}$. The three vectors are of the same length and can form an equilateral triangle. $\vec{a}$ and $\vec{b}$ are exactly the first and second lattice vectors of the supercell while $\vec{c}$ is represented by the shorter diagonal of the supercell.

The incommensurate charge density waves modulate with a period of $\sqrt{3}|\vec{a}_1|$ along the directions of the three aforementioned vectors. It's worth noting that while the CDWs are incommensurate with respect to the unit cell, the supercell itself is commensurate having a $\sqrt{3} \times \sqrt{3}$ symmetry with respect to the unit cell. The switch to a CDW in phase in this material has been attributed to the activation of the 1.96 THz optical phonon mode.²⁵

The hexagonal peaks we observed have also been predicted in single-layer TMD compounds using Landau's Theory of CDW, where charge density serves as an order parameter.⁶⁹ Moreover, they were also reported experimentally in 1T-TaSe₂ using STM^{70,71}.

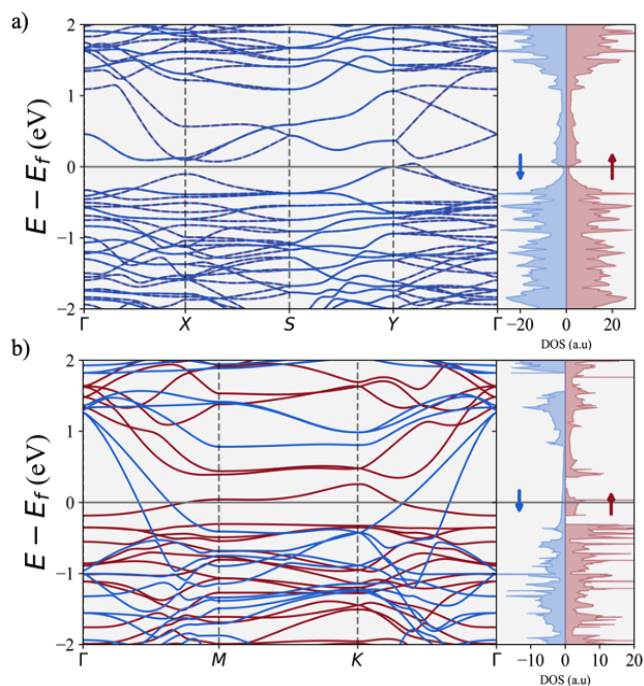


Fig. 5 Electronic band structures and DOS for the (a) AFM-SDW and (b) FM-CDW phases. Spin-up (red) and down (blue) are shown.

The electronic band structures and total density of states of the

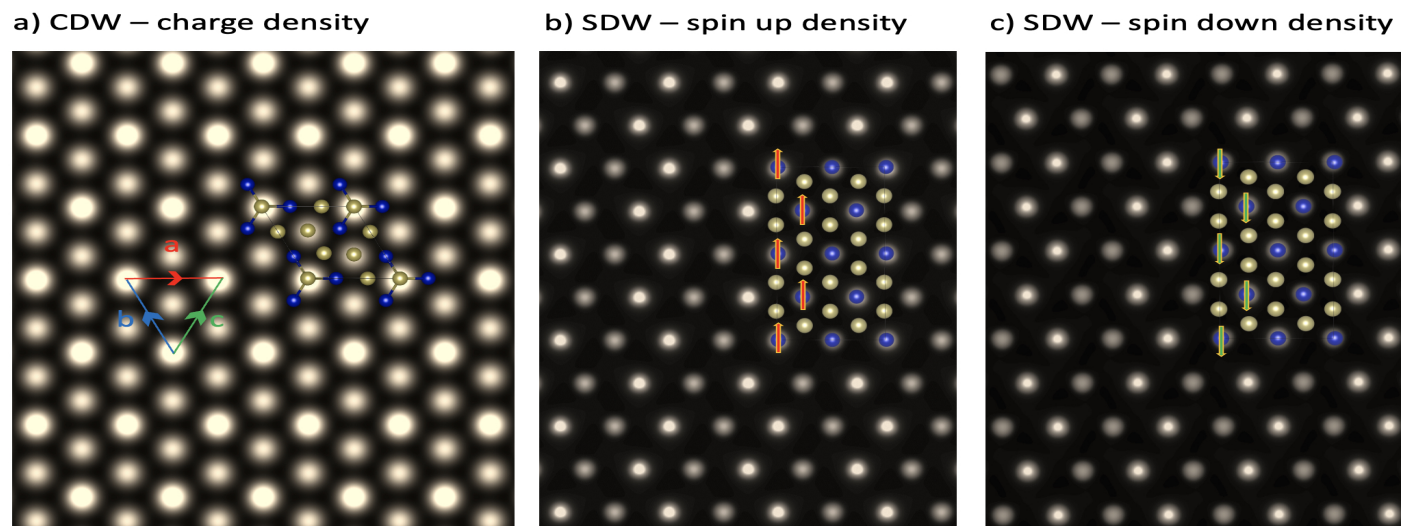


Fig. 3 Simulated STM and SP-STM images. a) STM of the incommensurate CDWs forming hexagonal concentrations. b) Simulated SP-STM image of the spin-up density waves. c) Simulated SP-STM image of the spin-down density waves.

FM-CDW phase are shown in Fig. 5. Consistent with the development of CDW, we notice an appreciable decrease in the density of states above the Fermi energy level compared to the band structures of the FM unit cell in Fig. S2. In the SDW phase, the SP-STM simulated images reveal distinct patterns for both spin channels, as shown in Fig. 3.

Unlike the CDW, the SDW phase features a commensurate density wave along the x -direction with a modulation period of $2|\vec{a}_1|$ and is responsible for the long-range AFM order. Notably, there is a noticeable decrease in the density of states near the Fermi energy, as seen in (b) of Fig. 5. This is not indicative of a Mott transition, which is typically characterized by a full gap due to strong electron-electron interactions. Instead, this decrease in the density of states near the Fermi level may be associated with a Slater transition since it coincides with a magnetic ordering transition due to the onset of the SDW, a characteristic feature of Slater transitions.⁷²

3.3 Evolution of The Fermi Surface

The Fermi surface's topology often reveals key characteristics of low-dimensional materials. This is especially true for systems with unique phases, which arise from the development of charge and spin density wave phases.^{74–83}

The system's exotic density wave phases emphasize how the Fermi surface's topology plays a role in its transitions. In Fig. 6, we examine the relationship between the Fermi surface and phase transitions. We start by considering the ferromagnetic unit cell and show its Fermi surface. As the transition into a CDW manifests, the Fermi surface reconstructs for both the spin-up and spin-down channels. Specifically, the Fermi surface of the spin-up electrons in the ferromagnetic CDW phase shows one less contour line indicating a reduction of the occupied electronic states at the Fermi level due to the onset of the CDW. Since this phase is also dynamically stable, it shows that the delicate interplay between electrons and phonons is ultimately necessary for stabilizing the

ferromagnetic order.

The second transition highlighted in the figure is that from the FM ordering to the AFM ordering due to the onset of the SDW. As this transition manifests, the Fermi surface topology changes drastically leading to the development of Fermi pockets. This kind of Fermi surface topology is typical in metallic systems exhibiting an AFM order and the theory behind the transition has been discussed extensively in the literature.^{53,54,84–86} As seen in Fig. 6 when the transition to an AFM occurs, the monolayer transitions from being a metal with a large Fermi surface to a metallic state exhibiting electron and hole pockets due to the development of the SDW order at the wavevector $\vec{k} = (\pi, \pi)$. While it is expected that the increase of the strength of the U parameter in a Hubbard-like model leads to this evolution from a larger Fermi surface to a "pocketed" Fermi surface,⁵⁴ our calculations demonstrate that such evolution can be observed without explicitly accounting for on-site repulsions.

The hole pockets are due to contributions above the Fermi energy along the Γ -S path while the electron pockets are due to contributions along the Γ -Y path. To better depict this, we have plotted the electronic band structures along these paths in Fig. 7. In the same figure, we also highlight the location of the expected nested hole pockets around $(\frac{\pi}{2}, \frac{\pi}{2})$ and symmetry-related points due to the nesting vector $\vec{k} = (\pi, \pi)$.⁸⁴

3.4 Anisotropic Optical Response

SDW phases typically arise in highly anisotropic metals.³¹ This highly anisotropic character presents itself in the optical response of quasi-one-dimensional chains and has been shown for the organic linear-chain compound (TMTSF)₂PF₆ exhibiting a SDW phase.⁸⁷ Of particular note is the large optical anisotropy resulting in linear dichroism, recently observed in zigzag antiferromagnets such as FePS₃ and NiPS₃.^{13–15}

To probe the optical properties of the SDW phase we performed DFT calculation in the independent Particle Approximation (IPA)

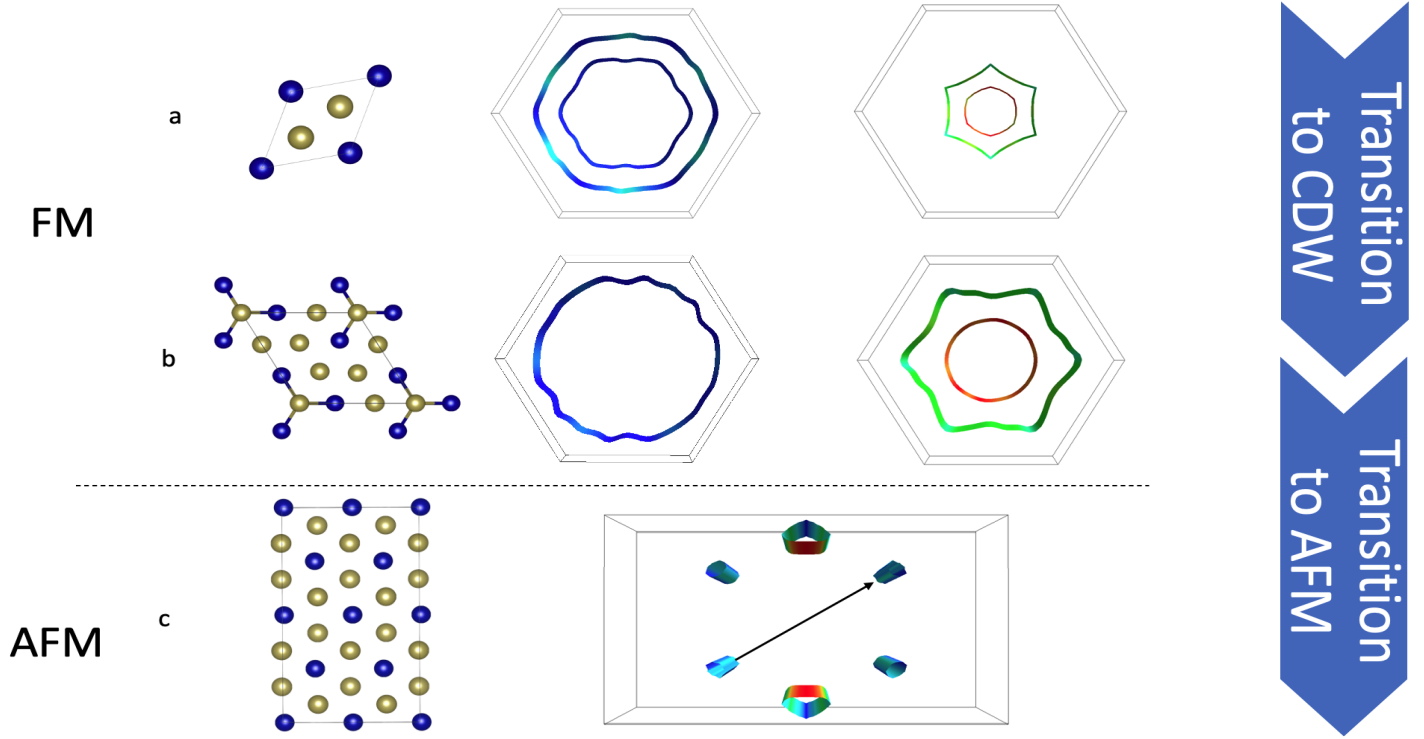


Fig. 6 The Fermi surfaces of the magnetic cells were generated by using FermiSurfer.⁷³ The colors indicate the Fermi velocity on an RGB scale so that the highest velocities are represented by red. (a,b) Spin-up and Spin-Down Fermi surfaces of the FM unit cell and the FM-CDW supercell, respectively. (c) Spin-degenerate Fermi surface of the AFM supercell. An example of the nesting vector connecting the hole pockets is also shown.

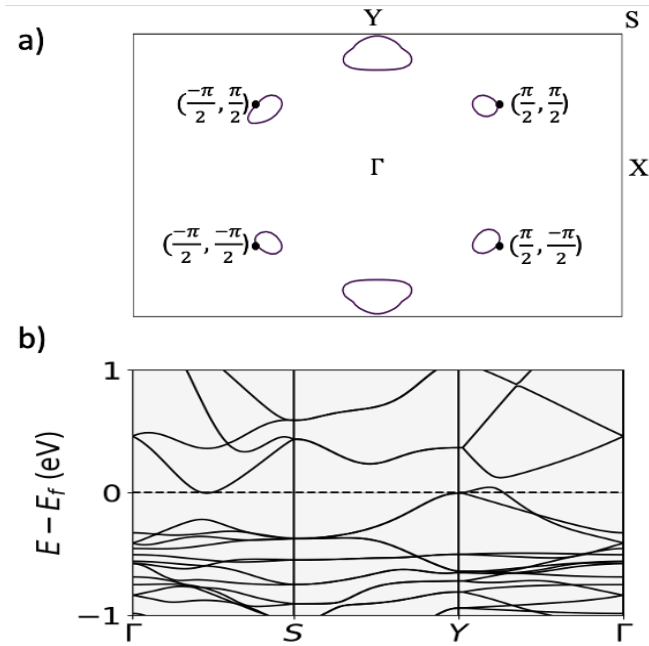


Fig. 7 a) The Fermi surface with hole and electron pockets on top. b) The band structures along the path relevant to the Fermi surface of the SDW.

image to obtain the complex dielectric tensor $\epsilon = \epsilon^1 + i\epsilon^2$.

The real part of the tensor element along the $(\alpha\beta)$ direction is given by the Kramers-Kronig transformation:

$$\epsilon_{\alpha\beta}^{(1)}(\omega) = 1 + \frac{2}{\pi} P \int_0^\infty \frac{\epsilon_{\alpha\beta}^{(2)}(\omega') \omega'}{\omega'^2 - \omega^2 + i\eta} d\omega'$$

where α and β represent the Cartesian coordinates (x, y, z) and P is the principle value.

The imaginary part of the tensor element is determined by summation over empty states as follows:

$$\begin{aligned} \epsilon_{\alpha\beta}^{(2)}(\omega) &= \frac{4\pi^2 e^2}{\Omega} \lim_{q \rightarrow 0} \frac{1}{q^2} \\ &\times \sum_{c,v,\mathbf{k}} 2w_{\mathbf{k}} \delta(\epsilon_{c\mathbf{k}} - \epsilon_{v\mathbf{k}} - \omega) \\ &\times \langle u_{c\mathbf{k}+\mathbf{e}_{\alpha}q} | u_{v\mathbf{k}} \rangle \langle u_{v\mathbf{k}} | u_{c\mathbf{k}+\mathbf{e}_{\beta}q} \rangle \end{aligned} \quad (1)$$

where c refers to conduction bands and v refers to valence bands while $u_{c\mathbf{k}}$ is the cell periodic part of the orbitals at the $\mathbf{k}$ -point.

We then plot the real and imaginary parts of the ϵ_{xx} and ϵ_{yy} to show the optical response along the x and y directions, respectively. The plotted results of these calculations, also shown in Fig. 8, reveal the presence of a pronounced optical anisotropy that is not present in the ferromagnetic phases of the unit cell or the CDW supercell as seen in Fig. S3 and Fig. S4. Specifically, along the y direction, the real (ϵ_{yy}^1) and imaginary (ϵ_{yy}^2) parts show large

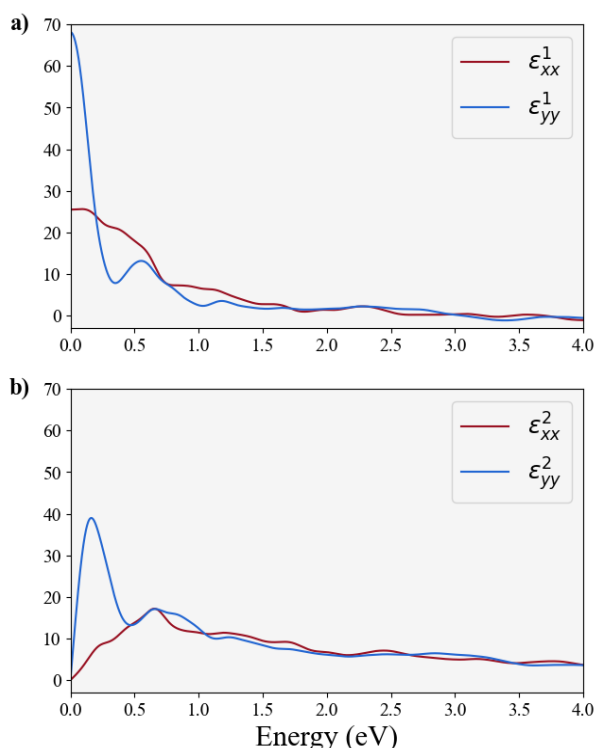


Fig. 8 Anisotropic optical response: The real and imaginary parts of the frequency-dependent dielectric tensor ϵ_{xx} and ϵ_{yy} , reflecting anisotropic conductivity in the quasi-one-dimensional system. The real part is shown in (a) and the imaginary part is shown in (b)

resonant responses near 0 eV. These resonances along the y direction are likely due to contributions from the larger electron pockets near the Fermi surface along the Γ -Y path. The anisotropic behavior indicates that the AFM state is quasi-one-dimensional due to the SDW causing the out-phase modulation along neighboring chains leading to the Zigzag pattern of the AFM order.

4 Conclusions

In this work, we have explored the intricate magnetic properties of 1T-CrTe₂ monolayers, a potential candidate for high-temperature intrinsic magnetism. By employing DFT calculations, we have investigated the magnetic and dynamic stability of both FM and AFM phases and their relationship with lattice parameters. We have found that charge and spin density waves are responsible for stabilizing the FM and AFM magnetic orders, respectively.

Our results suggest that the AFM state appears to be the ground state for 1T-CrTe₂ monolayers. We have also demonstrated that magnetic ordering can be influenced by adjusting the lattice parameters and hence allowing transitions between FM-CDW and AFM-SDW phases. This tunability of magnetism could potentially pave the way for the application of 1T-CrTe₂ in spintronic devices, where the manipulation of magnetic states is essential.

A key discovery of our study is the prediction of a commensurate spin density wave (SDW), which we believe is responsible for the AFM order. This SDW exhibits significant anisotropy, leading to quasi-one-dimensional behavior. Furthermore, we observed

that the anisotropy of this quasi-one-dimensional SDW extends to the material's optical response. This finding presents an intriguing direction for future research and suggests that understanding the relationship between magnetic and optical properties could lead to the development of novel magnetically tunable optoelectronic devices.

Conflicts of interest

The authors declare no conflicts of interest.

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Manuscript: Magnetic Stability, Fermi Surface Topology, and Spin-Correlated Dielectric Response in Monolayer 1T-CrTe₂

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Data Availability Statement:

The data for this study was generated using version 5.4.4 of VASP. The Python code used for data analysis is available at the following GitHub repository:

<https://github.com/rashigeek/CrTe2-paper>.