Widely Tunable Berry Curvature in the Magnetic Semimetal Cr$_{1+\delta}$Te$_2$

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Magnetic semimetals have increasingly emerged as lucrative platforms hosting spin-based topological phenomena in real and momentum spaces. Cr$_{1+\delta}$Te$_2$ is a self-intercalated magnetic transition metal dichalcogenide (TMD), which exhibits topological magnetism and tunable electron filling. While recent studies have explored real-space Berry curvature effects, similar considerations of momentum-space Berry curvature are lacking. Here, the electronic structure and transport properties of epitaxial Cr$_{1+\delta}$Te$_2$ thin films are systematically investigated over a range of doping, $\delta$ (0.33 – 0.71). Spectroscopic experiments reveal the presence of a characteristic semi-metallic band region, which shows a rigid like energy shift with $\delta$. Transport experiments show that the intrinsic component of the anomalous Hall effect (AHE) is sizable and undergoes a sign flip across $\delta$. Finally, density functional theory calculations establish a link between the doping evolution of the band structure and AHE: the AHE sign flip is shown to emerge from the sign change of the Berry curvature, as the semi-metallic band region crosses the Fermi energy. These findings underscore the increasing relevance of momentum-space Berry curvature in magnetic TMDs and provide a unique platform for intertwining topological physics in real and momentum spaces.

1. Introduction

In recent years, fascinating new condensed matter phenomena have been found to arise from the interplay of spin degree of freedom of electrons with emergent geometric and topological effects.\cite{1,2} Prominent among these is the concept of Berry curvature $\Omega$, which arises from the geometric phase accumulated by electronic wavepackets traversing a closed loop.\cite{3,4} In crystalline solids, such Berry curvature can be interpreted as an effective magnetic field acting on moving electrons, and therefore manifests prominently in Hall transport experiments.\cite{1} For example, the quantization of its integral in the momentum space, a phenomenon known as band topology, results in quantized charge and spin Hall effects.\cite{5–8} On the other hand, magnetic materials exhibit rich manifestations of real- and momentum-space Berry curvature.\cite{9,10}
In particular, noncollinear and chiral spin structures give rise to real space Berry curvature, which results in the topological Hall effect (THE) for itinerant electrons. In contrast, the momentum space Berry curvature of Bloch wave functions of electronic bands contributes prominently to the anomalous Hall effect (AHE), which may result in its quantization and unconventional non-linear magnetic field dependence. The robustness of such k-space Berry curvature, its material tunability, and direct accessibility from experimental probes makes it attractive for electronic and optical applications.

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Prominent among magnetic TMDs is Cr$_{1+\delta}$Te$_2$ — a self-intercalated TMD with tunable valency (Cr$^{2+}$ (3d$^4$) to Cr$^{4+}$ (3d$^2$), assuming simple ionic crystal picture) – which therefore serves as a unique candidate for modulating electronic and magnetic properties over a wide range. Recent efforts have enabled a unique recipe to realize epitaxial thin films of Cr$_{1+\delta}$Te$_2$ over wide range of doping, with tunable magnetic properties. Here, by combining in situ angle resolved photoemission spectroscopy (ARPES), transport measurements, and density functional theory (DFT) calculations, we show that Cr$_{1+\delta}$Te$_2$ films form a unique magnetic and semi-metallic platform that hosts widely tunable Berry curvature near the chemical potential in the momentum space.

2. Results and Discussion

2.1. Basic Characteristics of Cr-Te Films

In this work, epitaxial Cr$_{1+\delta}$Te$_2$ thin films were grown by molecular beam epitaxy (MBE) using an established recipe which uniquely enables the doping level $\delta$ to be varied over a wide range ($0.3 < \delta < 0.8$). We have previously shown that Cr$_{1+\delta}$Te$_2$ thin films maintain the NiAs-type crystal structure.
of CrTe₂ (Figure 1a), while dopants Cr intercalate between the weakly coupled Te layers.[53] Note that regardless of our extensive efforts, growing single crystalline phase with $\delta < 0.3$ and $\delta > 0.8$ was not possible.[53] Figure 1b shows the corresponding evolution of Curie temperature, $T_C(\delta)$, estimated from magnetometry and a characteristic kink in resistivity measurements (see Figure S1, Supporting Information). Consistent with our previous report,[53] $T_C(\delta)$ increases from 160 K, and saturates to $\approx 350$ K for $\delta \gtrsim 0.5$.

2.2. Cr-Te Band Schematic

We turn to examine the expected effect of doping on the band structure of Cr₁₊δTe₂, whose bulk (lower) and surface (upper) Brillouin zones are shown in Figure 1c. The schematic low energy band structure of Cr₁₊δTe₂ – which is central to the key claims of this report – derived from ARPES experiments and DFT calculations is illustrated in Figure 1d (detailed in subsequent sections). Near $\Gamma$, there are three hole bands, which are denoted as HB₁Γ, HB₂Γ, and HB₃Γ for convenience. Meanwhile, near $M$, there are two hole bands (HB₁M, HB₂M) and one electron band (EBₐ). With increasing number of intercalated cations, the Fermi energy, $E_F$, is expected to correspondingly shift upwards. The wide range of tunable electron filling in Cr₁₊δTe₂, coupled with the semi-metallic character,[54,55] motivates us to explore the evolution of its Berry curvature in momentum space (Figure 1c,d), and its signature in band structure and transport measurements.

2.3. ARPES Measurements

The ARPES measurements are performed in situ, using an ultrahigh vacuum (UHV) cluster system that also connects the MBE system (Figure 2a). The ARPES measurements are used to establish the semi-metallic nature of the near $E_F$ band structure of Cr₁₊δTe₂ around the $\overline{M}$-point. Here, a

![Figure 2](image_url)

**Figure 2.** In situ angle resolved photoemission spectroscopy (ARPES) of Cr₁₊δTe₂ films ($\delta = 0.6$) at 15 K, with He-I light source ($h\nu = 21.2$ eV). a) Schematic of the ultra-high vacuum (UHV) cluster system used in this study, equipped with molecular beam epitaxy (MBE) growth system and ARPES using He based discharge lamp. b) ARPES mapping of near-$E_F$ spectral weight for $\delta = 0.6$ sample, representative of the Fermi surface, obtained by integrating between 0 meV and -75 meV from $E_F$. Black dashed lines are guide to the eyes. c-f Measured band dispersion for $\delta = 0.6$ along high-symmetry directions c–d) $\overline{K} - \overline{\Gamma} - \overline{K}$ and e–f) $\overline{K} - \overline{M} - \overline{K}$, respectively (He-I source). Here, (c, e) show the spectral intensity and (d, f) its second derivative respectively. Overlaid dashed white/grey lines bands indicate the identifiable bands (c.f. Figure 1d). Insets to (c, e) show respective momentum distribution curves (MDCs) at $E_F$, with black dots indicating band crossings.
semi-metallic band is characterized by an electron-like band branch at higher energies, and a hole-like band branch at lower energies coexisting and overlapping around the same momenta (see Figure 1d, dashed circle at $\mathbf{M}$). For the illustrative purpose of Figure 2b–f, we choose the $\delta \approx 0.6$ sample as its higher electron filling provides ARPES – sensitive to occupied states – with wider energy access to the $\text{Cr}_{1-x}\text{Te}_2$ band structure.

Figure 2b presents the $k$-resolved map of the Fermi surface, with clearly observable Fermi pockets at the $\mathbf{F}$ and $\overline{\mathbf{M}}$ points. To further understand the character of these Fermi pockets, energy dependent ARPES spectral weights measured across $\Gamma$ and $\mathbf{M}$ are shown in Figure 2c,d and Figure 2e,f, respectively. At $\mathbf{F}$ (Figure 2c,d), we observe three hole bands ($\text{HB}_{1\mathbf{F}}, \text{HB}_{2\mathbf{F}}, \text{HB}_{3\mathbf{F}}$), with the topmost –$\text{HB}_{1\mathbf{F}}$– crossing $E_{\text{F}}$, and producing a hole-like Fermi pocket (Figure 2b). Meanwhile, at $\overline{\mathbf{M}}$ (Figure 2e,f), we find an electron band ($\text{EB}_{\overline{\mathbf{M}}}$) crossing $E_{\text{F}}$ forms an electron-like Fermi pocket (Figure 2b). Notably, in addition to this ($\text{EB}_{\overline{\mathbf{M}}}$), two hole bands ($\text{HB}_{1\overline{\mathbf{M}}} \text{ and } \text{HB}_{2\overline{\mathbf{M}}}$) exist slightly below $E_{\text{F}}$ at $\overline{\mathbf{M}}$. Coexistence of electron and hole band branches at the $\overline{\mathbf{M}}$ point hosts the near $\mathbf{F}$ semi-metallic character in $\text{Cr}_{1-x}\text{Te}_2$.

Next, we have shown the doping evolution of the band structure by studying ARPES spectral intensities, along $\mathbf{K} - \Gamma - \overline{\mathbf{K}}$ and $\mathbf{K} - \mathbf{M} - \overline{\mathbf{K}}$ for samples with $\delta = 0.33, 0.4, 0.5,$ and 0.6. Consistent with the expected increase of electron filling with the increase of $\text{Cr}$ cations, all bands at $\mathbf{G}$ and $\overline{\mathbf{M}}$ are found to collectively shift downward relative to $E_{\text{F}}$ with increasing $\delta$ (Figure 3a–h). For $\delta < 0.5$ samples, the only occupied band – $\text{HB}_{3\overline{\mathbf{M}}}$ – is barely visible (Figure 3e,f). In contrast, for $\delta \geq 0.5$, both the bottom of the electron band EB$_{\overline{\mathbf{M}}}$ and top of hole band HB$_{3\overline{\mathbf{M}}}$ are clearly seen below $E_{\text{F}}$, in addition to the lower hole band (HB$_{2\overline{\mathbf{M}}}$) (Figure 3g,h). While intensity for HB$_{1\overline{\mathbf{M}}}$ between samples with $\delta = 0.5$ (Figure 3g) and $\delta = 0.6$ (Figure 3h) are largely different, weakly seen HB$_{3\overline{\mathbf{M}}}$ in sample with $\delta = 0.5$ (Figure 3g) is still in line with rigid-like band shift picture. Observation of rigid-like band shift is none trivial phenomenological finding, which capture essential doping evolution of electronic state for sample with $0.3 < \delta < 0.8$. As shown later, this finding plays a vital role to interpreting Hall effects.

To quantify the shift in band structure with doping, we further examine the evolution of various band features at $\mathbf{G}$ and $\overline{\mathbf{M}}$. By an appropriate choice of $\delta$-dependent $E_{\text{F}}$ shift, the dispersion of the measured bands across samples with varying $\delta$ (Figure 3a–h: markers) can be all collapsed onto a single dispersion, e.g. that of $\delta = 0.6$ sample (Figure 3d,h). The collapse of band dispersions for varying $\delta$ around $\mathbf{G}$ and $\overline{\mathbf{M}}$ is summarized in Figure 3i,j, respectively. The rigid-like shifts of energy bands thus observed at $\mathbf{G}$ and $\overline{\mathbf{M}}$ are defined as $\Delta E_{\mathbf{G}}$ and $\Delta E_{\overline{\mathbf{M}}}$, respectively, and their evolution with $\delta$ are plotted in Figure 3k. The observed linear dependence of $\Delta E_{\mathbf{G}}$ and $\Delta E_{\overline{\mathbf{M}}}$ on $\delta$ is consistent with the expected evolution of a quasi-2D band structure with parabolic dispersion. Linear fitting of the $\Delta E$ versus $\delta$ data in Figure 3k yields a slope of 1.54 eV energy shift per intercalated Cr atom, i.e., $\Delta E_{\mathbf{G}}/\delta \approx 1.54$ eV/atom. This relation is consistent with DFT calculation (see Figure S2, Supporting Information) and is especially useful for subsequent comparisons of experimental and theoretical Hall conductivities in the latter part of this report.

### 2.4. Anomalous Hall Effect Measurements

To visualize the manifestation of Berry curvature effects arising from the semi-metallic band component, we investigated the Hall transport properties of $\text{Cr}_{1-x}\text{Te}_2$ films, and their evolution with doping ($\delta = 0.33, 0.47, 0.51, 0.7$), magnetic field ($B$), and temperature ($T$). For a conventional magnetic material with magnetization $M$, the Hall resistivity is phenomenologically given as

$$\rho_{yx} (B) = \rho_{yx}^c (B) + \rho_{yx}^s (M (B))$$

Here, $\rho_{yx}^c$ and $\rho_{yx}^s$ are the ordinary and anomalous Hall resistivities respectively, wherein the latter incorporates effects of $k$-space Berry curvature.[9] First, $\rho_{yx}^c (B) \equiv R_{xy} B$, where $R_{xy}$ is the Hall coefficient, can be straightforwardly determined by a linear fit to $\rho_{yx}$ in the high field limit, $B \gg B_s$, where $B_s$ is the out-of-plane (OP) saturation field estimated from magnetometry measurements (see Figure S5, Supporting Information for raw data). The AHE resistivity $\rho_{yx}^s (B)$ obtained upon subtracting $\rho_{yx}^c$ from $\rho_{yx}$ is plotted in Figure 4a–c for a series of temperatures ($T/T_C \approx 0.9, 0.5, T = 2 K$) and doping. While the resulting AHE resistivity $\rho_{yx}^s (B)$ is typically proportional to the magnetization $M(B)$, the presence of chiral spin textures under intermediate fields may also induce a “topological” component that is non-monotonic in magnetization $M(B)$.[38-15] To avoid such complications, we choose to focus on the saturated magnitude of AHE resistivity by determining its value at $B \approx 40 \text{kOe} \gg B_s$. The saturated AHE resistivity, defined henceforth as $\rho_{yx}^s (B \geq B_s)$, is indicated by horizontal dashed lines in Figure 4a–c, and examined in detail subsequently.

As shown in Figure 4d,e, the AHE ($\rho_{yx}^s$) and the ordinary Hall coefficient ($R_{xy}$) show clear differences in their evolution with temperature and doping. As in Figure 4d, $R_{xy}(T, \delta)$ persistently presents a positive sign. While the quantitative interpretation of $R_{xy}$ is involved within multi-component Fermi surfaces,[66] the persistence of a positive $R_{xy}(T, \delta)$ indicates the dominant contribution of hole pockets at $\mathbf{G}$ and $\overline{\mathbf{M}}$. Meanwhile, $\rho_{yx}^s (T, \delta)$, in Figure 4e, exhibits multiple sign changes – both across doping and temperature. For example, with decreasing temperature, for $\delta = 0.33$, $\rho_{yx}^s$ changes from positive to negative, while for $\delta = 0.7$ it changes from negative to positive. Therefore, the sign of $\rho_{yx}^s$ is likely unrelated to $R_{xy}(T, \delta)$, and governed by a rich interplay of doping and temperature, which is indicative of corresponding variations in its constituent extrinsic and intrinsic AHE components.[9,57]

To separate extrinsic and intrinsic AHE, a widely accepted starting point is to consider the total anomalous Hall conductivity ($\text{AHC}$) $\sigma^\text{AHC} = \sigma^\text{AHC}_x + \sigma^\text{AHC}_y + \sigma^\text{AHC}_{\text{int}}$, arising from the linear combination of three independent conductivity channels: $\sigma^\text{AHC}_x = \sigma_x + \sigma_y + \sigma_{\text{int}}$. Here, $\sigma_x$ and $\sigma_y$ are extrinsic skew and side-jump scattering terms, while $\sigma_{\text{int}}$ is the intrinsic Karplus-Luttinger term.[9] Next, we note with respect to the established scaling relation ($\sigma^\text{AHC}_y$ versus $\sigma^\text{AHC}_{\text{int}}$) used to delineate these components that our sample is far from the region where intrinsic AHC dominates over extrinsic AHC components (see Figure S6, Supporting Information).[9] Thus, to separate intrinsic and extrinsic AHC components for our samples, we follow a phenomenological approach for disordered systems, adopted in several previous works.[5,57-65] For disordered systems, wherein extrinsic scattering is dominated by $T$-independent impurity scattering
relative to $T$-dependent phonon scattering, the following modified AHC scaling relation has been proposed:\cite{57}

$$\sigma_{\text{ly}} \propto \left( \alpha \sigma_{\text{xx},0}^2 + \beta \sigma_{\text{xx}}^2 \right) \sigma_{\text{xx}}^2 + b = \text{const.} \times \sigma_{\text{xx}}^2 + b \quad (2)$$

Here, $\alpha$, $\beta$ and $b$ represent contributions from skew scattering, side-jump, and intrinsic Karplus-Luttinger (Berry phase) derived terms, respectively. The extrinsic terms are scaled by the residual conductivity $\sigma_{\text{res}}$, which relates to the residual conductivity $\sigma_{\text{xx},0}$.

Figure 3. Doping Evolution of Measured band structure at 15 K with He-I light source ($h\nu = 21.2$ eV). a–h) Evolution of near-$E_F$ band dispersion along $\overline{K} - \overline{\Gamma} - \overline{K}$ and e–h) $\overline{K} - \overline{M} - \overline{K}$ (e, f show second derivative) for samples with $\delta = 0.33, 0.4, 0.5$ and 0.6 respectively (left to right). Overlaid markers denote fits to the bands, used in i–j) for comparisons. Black and gray dashed lines are guide to the eyes. i,j) Measured dispersions of the respective bands across all $\delta$ at $\overline{K} - \overline{\Gamma} - \overline{K}$ (i) and $\overline{K} - \overline{M} - \overline{K}$ (j), collapsed onto single curves by shifting all data points for each $\delta$ by a constant energy ($\Delta E_I(\delta)$ and $\Delta E_M(\delta)$, details in text). Black and gray lines in (i, j) are guide to the eyes. k) The variation of thus defined energy shifts $\Delta E_I$ and $\Delta E_M$ with $\delta$, with the overlaid line showing a linear fit (details in text).
resistivity $\rho_{xx0}$ arising from impurity-induced elastic scattering at the $T = 0$. Therefore, the intrinsic component ($b$) can be determined as the intercept of the plot of $A_{xy}^{sat}$ versus $\sigma_{xx}^2$, as previously shown for various material systems.[25,57–60]

Following the modified scaling relation (Equation (1)), the intercept $b (\equiv \sigma_{xx}^{exp})$ extracted from fits to the linear component of $A_{xy}^{sat}$ versus $\sigma_{xx}^2$ plots, shown in Figure 5a for the studied samples, were used to estimate the respective intrinsic AHCs. In this work, to transform from resistivity to conductivity, we use the relations $\sigma_{xy}^{exp} = \rho_{xx0} / \rho_{xx0}$, $\sigma_{xx} = 1 / \rho_{xx}$, given the small (<1%) contribution of transverse channel relative to the longitudinal channel. Note that the reliability of the modified scaling analysis is higher when the system is in disordered region with lower $\sigma_{xx}$ magnitude. Thus, to estimate intercept $b$ from $\sigma_{xy}^{exp}$ versus $\sigma_{xx}$ plots, we focus on region with lower $\sigma_{xx}$ magnitude, in particular for $\delta > 0.4$ (see Figure 5a). Figure 5b shows thus obtained doping dependent $\sigma_{xy}^{exp}$ (left axis), which is contrasted with the corresponding variation of temperature averaged $R_H$ (right axis). Crucially, the sign of $\sigma_{xy}^{exp}$ changes from positive to negative over $\delta \approx 0.33 – 0.47$, while the sign of $R_H$ remains persistently positive across doping. The expected origin of the measured sign change in $\sigma_{xy}^{exp}(\delta)$ is from the $k$-space Berry curvature evolution with doping (see Figure 5c), and should be captured within band structure calculations.

### 2.5. Comparison between Theory and Experiments

We turn to ab initio DFT calculations to understand the band evolution of Cr$_{1+\delta}$Te$_2$. First, we confirm rigid-like band shift via DFT calculations for different $\delta$ (see section A in SI). Notably, the band portions featuring avoided crossings shift collectively in energy by an amount consistent with $\Delta E_F / \Delta \delta \approx 1.54$ eV/atom estimated previously from ARPES experiments (Figure 3k). Since our intercalant Cr is disordered, it is reasonable to rely on the CrTe$_2$ band structure as a parent electronic state to explain the $k$-space evolution of intercalated TMD Cr$_{1+\delta}$Te$_2$, and thereby avoid modeling the complications of intercalated Cr atoms (see section A in SI). In spite of this simplified approach,
for $\delta = 0.6$ for ARPES is set to $\sim 1.9$ eV higher than the calculated $E - E_F$(cal.) for DFT (compared for all dopings in Figure S7, Supporting Information). Such a shift between ARPES and DFT is consistent in sign with the expected filling of electrons with increasing $\delta$, and in magnitude with the reported trends toward an underestimation of $E - E_F$(calc.) in ab initio calculations of this system\cite{57} (see supporting information, Section A for validation of this reasonable shift from the view point of valency of Cr).

Since our ARPES results consistently evidence a rigid-like shift of the Cr$_{1-x}$Te$_2$ band structure with increasing $\delta$, we compare $\sigma_{xy}^{\text{expt}}(\delta)$ with the theoretical value of $\sigma_{xy}^{\text{DFT}}(E)$ obtained for the parent CrTe$_2$. Here, $\sigma_{xy}^{\text{DFT}}(E)$ is calculated by integrating the total Berry curvature $\Omega_{xy}(k)$ across the whole Brillouin zone (see method section and Equations (3–5) therein). Notably, the one-to-one comparison between $\sigma_{xy}^{\text{expt}}(E)$ and $\sigma_{xy}^{\text{DFT}}(\delta)$, shown in Figure 6c, is enabled by combining the $E_F$ reference point between ARPES and DFT (Figure 6a,b) with the measured $\Delta E_F/\Delta \delta$ (Figure 3k). We observe excellent qualitative agreement between the transport and DFT results across $\delta$. In particular, the calculations reproduce the sign change from negative to positive around $\delta = 0.5$ (Figure 6c; shaded area).

Importantly, the calculations further provide valuable insights on the $k$-space origin of the AHE sign change observed in DFT and transport experiments. In particular, understanding energy evolution of Berry curvature between 1.5 eV and 1.9 eV is essential due to sign flip of $\sigma_{xy}^{\text{DFT}}(E)$ (see horizontal arrows and shaded area in Figure 6c). We first examine in Figure 6d the $k_z$-dependence of band dispersion along $\overline{K} - \overline{M} - \overline{K}$, which exhibits – in each case – a characteristic nearly degenerated band region with a spin-orbit coupling (SOC)-driven gap (dashed circles). Notably, this band region disperses substantially with $k_z$, consequently widening the energy window hosting prominent Berry curvature distribution (to $\approx 0.2$ eV). Next, the role of this band region in the sign change of $\sigma_{xy}^{\text{DFT}}$ is elucidated by examining the $k_z$-integrated total Berry curvature, $\int \text{d}k \Omega_{xy}(k)$, shown in Figure 6e,f for two different energies $E - E_F$(calc.) = 1.5 eV (c.f. expt. $\delta = 0.3$) and 1.9 eV (c.f. expt. $\delta = 0.6$) on either side of the sign change (Figure 6c; horizontal arrows). Notably for 1.9 eV, we observe, around the $\overline{M}$ point (Figure 6f: vertical arrows), a prominent region hosting large positive $\int \text{d}k \Omega_{xy}(k)$, with momenta consistent with that hosting the SOC-driven gap (Figure 6d: dashed circle). Contrastingly, for 1.5 eV (Figure 6e), this particular source of total Berry curvature is absent around $\overline{M}$ point. Therefore, from Equation (4) in method section ($\sigma_{xy}^{\text{DFT}}(E) \approx - \int \text{d}k \Omega_{xy}(k)$), we can conclude that $\overline{M}$ point prominent positive Berry curvature at 1.9 eV is the leading cause of the sign change of $\sigma_{xy}^{\text{DFT}}$ from positive to negative with increasing $\delta$. Here, Weyl points (prominent sources of Berry curvature in other works\cite{19,24}) – are also found in our DFT calculations for CrTe$_2$ (see supporting information Section C). However, their energies ($E - E_F$(calc.) $> 2$ eV), are well above $E_F$ even for the highest doped sample. Thus, we can ignore the role of Weyl physics in our AHE results.

We expect that quantitative difference between $\sigma_{xy}^{\text{DFT}}$ and $\sigma_{xy}^{\text{expt}}$ may arise mainly from two sources. One is the effect of disorder. The longitudinal conductivities of these samples, $\sigma_{xx} \approx 10^5 - 10^6$ S/cm, i.e., on the verge of bad metal regime, where coherent skew scattering and side jump mechanisms are as we show hereafter, the observed sign change of AHE can be explained simply by the filling change of the electronic state of parent CrTe$_2$.

We first show matching of the energy level between ARPES measurements and DFT calculations. Figure 6a,b shows the DFT band structure for CrTe$_2$ with varying $k_z$ overlaid on the ARPES data for $\delta = 0.6$. First, the DFT calculations match the key qualitative features seen by ARPES, e.g., the electron and hole bands at $\Gamma$ and $\overline{M}$, and notably the semi-metallic band feature at $\overline{M}$. Second, the DFT and ARPES band structures match quantitatively – with particular focus on the bottom of the electron band at $\overline{M}$ (see arrow in Figure 6b) – when the measured $E_F$.
prominent. On the other hand, the intrinsic Berry phase contribution, is naively expected to be intact under disorder, although its reduction has been also reported experimentally.\cite{19,31} This suggests that Cr$_{1+\delta}$Te$_2$ forms a valuable platform for future explorations of the controversial effect of disordered transport phenomena in magnets. The other source relates to the impact of non-rigid band effects. The progressive intercalation of Cr may modify the band structure and associated Berry curvature for Cr$_{1+\delta}$Te$_2$ with respect to the parent CrTe$_2$, in addition to shifting the Fermi level. Regardless, it should be noted that the dominant role of the large Berry curvature around the $M$ point across the critical doping – which is central to observed AHC sign change in this work – is qualitatively supported even if we use $\delta = 1$, i.e., CrTe as the parent band structure (see section B in Supporting Information for details).

As an aside, the observed consistency of band structure between spin-resolved DFT calculations and ARPES experiments also provides a viable explanation for the saturation of $T_C$ with increasing doping noted here (Figure 1b) and in previous works.\cite{53} As shown in Figure 6d, the upper branch of the semi-metallic band region, which hosts large Berry curvature, derives from minority spins (blue curve), and becomes occupied above $\delta \approx 0.5$. Since $T_C$ is proportional to the net number of spins within conventional itinerant ferromagnets,\cite{61} the transition from

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**Figure 6.** Momentum-Space Analysis of Berry Curvature Effects. a–b) Comparison between ARPES-measured band dispersion for Cr$_{1+\delta}$Te$_2$ with $\delta = 0.6$ (shown in grayscale) with DFT-calculated band structure for CrTe$_2$ (energy shifted by 1.9 eV, overlaid as lines) along a) $\overline{K} - \overline{\Gamma} - \overline{K}$ and b) $\overline{K} - \overline{M} - \overline{K}$. Left and right axes are $E - E_F$ for DFT and ARPES respectively. The $E_F$ for ARPES data ($\delta = 0.6$) is set to 1.9 eV higher than that for DFT data ($\delta = 0$). DFT-calculated bands comprise major (red) and minor (blue) spin orientations, and results for varying $k_z$ are all overlaid for comparison with ARPES data, which lack $k_z$ resolution. c) Comparison between DFT-calculated $\sigma^{\mathbf{m}}_\mathbf{E}(E)$ (grey line, from Equation (4) in method section, details in text) and measured $\sigma^{\mathbf{m}}_\mathbf{E}(\delta)$ (black dots, from Figure 5b). One-to-one comparison between calculations (E) and experiments (δ) is enabled by matching the Fermi energies in (a)-(b), together with the measured $\Delta E_F/\Delta \delta$ (Figure 3k, details in text). Horizontal arrows indicate the energies used for (e) and (f) respectively. d) The $k_z$ resolved dispersion of the DFT calculated semi-metallic band region near $E_F$ along $\overline{K} - \overline{M} - \overline{K}$ (c.f. collated in (b)). Colors indicate spin orientation (c.f. (a)), while dashed circle highlights spin-orbit coupling (SOC) driven opening of a band gap, hosting large $k$-space Berry curvature. e–f) Color plots of the distribution of calculated Berry curvature, $\int \text{d}k \Omega_m(k)$, across the $k_x - k_y$ plane at e) $E - E_F^{\text{calc}} = +1.5$ eV and f) $+1.9$ eV (horizontal arrows in (c)). Arrows highlight a region near $M$-point, hosting a large Berry curvature contribution within (f).
majority to minority spins can be qualitatively expected to produce the observed reduction of slope of $T_C(\delta)$ from $\delta = 0.3 - 0.4$ (steep slope) to $\delta \approx 0.5 - 0.7$ (gentle slope) (details in supporting information Section D). Moreover, the collective picture on the observed band dependence of magnetism of Cr$_{1+\delta}$Te$_2$ band structure with $\delta$ provides further validity to our overall interpretation of the k-space evolution of AHE in this system.

3. Summary & Impact

In summary, complementary electronic structure and transport studies enable us to reveal the existence of a characteristic semi-metallic band region with prominent Berry curvature in the self-intercalated TMD Cr$_{1+\delta}$Te$_2$. With varying doping, ARPES measurements capture the rigid-like shift of the electronic band structure, realizing tunable Berry curvature at the chemical potential. Consistently, transport measurements reveal a doping-dependent sign flip in the intrinsic anomalous Hall conductivity whose k-space Berry curvature origin is elucidated via DFT calculations. The confluence of a prominent semi-metallic band region and flexibility in accommodating intercalated atoms render Cr$_{1+\delta}$Te$_2$ a unique TMD platform for tailoring k-space Berry curvature.

The confluence of a prominent semi-metallic band region and ability to accommodate intercalants over a wide range renders Cr$_{1+\delta}$Te$_2$ as a unique TMD platform for tailoring k-space Berry curvature. Our findings also point to the potential of Cr$_{1+\delta}$Te$_2$ serving as a model system for investigating exotic phenomena intertwining Berry curvature physics between real and momentum spaces. The presence of a large k-space Berry curvature at the chemical potential is expected to lend itself to substantial tunability and redistribution, even in the presence of magnetic perturbations corresponding to moderate energy scales. Particularly intriguing is the region with $\delta = 0.5$ and above (Figure 6c), where the interplay of myriad magnetotransport phenomena may result in the deviation of $\sigma^{\nu}_{\alpha \beta}$ versus $\sigma^{\nu}_{\alpha \alpha}$ from linearity (Figure 5a). Interestingly, this critical doping range concomitantly hosts change of magnetic anisotropy from out-of-plane to in-plane. One possible cause for these intriguing effects may be the enhanced magnetic fluctuation/frustration, which may manifest itself in additional contributions to Hall transport measurements. In this regard, a recent study points to such magnetic fluctuation/frustration resulting in enhanced thermopower around $\delta \approx 0.5$. Our work opens the door for probing and tailoring the interplay of rich, intertwined magnetic effects in real- and momentum-space for future experimental and theoretical studies in Cr$_{1+\delta}$Te$_2$.

3.2. Transport and Magnetometry Measurements

Resistivity and Hall transport measurements were performed with a Quantum Design PPMS® Dynacool system. Both the longitudinal and transverse resistivity were measured simultaneously in the presence of a magnetic field to obtain the AHE results reported in this work. Meanwhile, magnetization measurements were performed using a Quantum Design MPMS® 3 system.

3.3. In Situ Photoemission Spectroscopy (ARPES)

ARPES measurements were conducted on samples transferred in pristine condition from the adjacent, UHV-connected MBE system, i.e., without any ambient exposure or capping layer. The DA30HL (Scienta Omicron™) electron analyzer was used to study the photoexcited electrons with instrumental energy resolution $< 10$ meV. All ARPES data shown in this report were obtained at 15 K, using He I light source ($\nu = 21.2$ eV), which provides higher energy resolution and signal-to-noise ratio comparing to He II light source ($\nu = 40.8$ eV). We note that observing qualitatively similar Fermi pockets were also seen, using He II light source ($\nu = 40.8$ eV) regardless of much lower intensity (see Figure S12, Supporting Information). In this work, the photon energy-dependence of $k_z$ is not expected to significantly affect the key results, namely the observation of semi-metallic band at the M-point and the rigid-like shift of all near-$E_F$ bands with doping, $\delta$. The weaker spectral intensity around $\mathbf{M}$ relative to that around $\mathbf{\Gamma}$ likely arises from orbital dependent cross-section, since at $\sim 21$ eV, the photoionization cross-section for $d$ orbitals (near $\mathbf{M}$) is weaker than that of $p$-orbitals (near $\mathbf{\Gamma}$).

3.4. DFT Band Calculations

First principles DFT calculations were performed using an established framework utilizing the generalized gradient approximation (GGA) in the Perdew-Burke-Ernzerhof (PBE) form. Projector-augmented-wave wave functions with energy cut-off of 500 eV were used within the Vienna ab-initio simulation package (VASP). The structures were optimized until the residual forces were less than $10^{-3}$ eV/Å, and the self-consistency criteria for convergence was set to $10^{-6}$ eV. Γ-centered Monkhorst-Pack grids of size $12 \times 12 \times 8$, and on-site Coulomb repulsion energy, $U = 2$ eV were used for CrTe$_2$, and the band structure was obtained from the PBE calculations. In these calculations, the spin orientation is fully aligned along the out-of-plane (OP) direction, consistent with characterization studies performed in our previous work, the film thickness was measured using atomic force microscopy, crystallinity was confirmed by X-ray diffraction (XRD) and cross-sectional scanning transmission electron microscopy (STEM), and doping $\delta$ was determined by combining EDS with XRD lattice constants. The thickness of all samples used in this study was $\approx 100$ nm.
the intended comparison with anomalous Hall signal from the fully saturated, OP magnetic state.

3.5. Anomalous Hall Conductivity Calculations

For simplicity, we focus on $\sigma_{xy}^{\text{rash}}(E)$ from the parent $\text{CrTe}_2$ whose comparison with $\sigma_{xy}^{\text{anom}}(\delta)$ is justified given the consistency of band structures and observed rigid band shift with doping (see main text for details). A Hamiltonian derived from maximally localized Wannier functions was obtained using the WANNIER90 package[69] and the anomalous Hall conductivity was calculated using WannierTools.[70] The following expressions were used to calculate the Berry curvature ($\Omega_{xy}$), total Berry curvature ($\Omega_{xy}^{\text{total}}$) and anomalous Hall conductivity ($\sigma_{xy}^{\text{anom}}$) respectively from the Hamiltonian $\hat{H}$ derived from maximally localized Wannier functions.[71]

$$\Omega_{n,xy}(k) = -2\text{Im} \sum_{m,n} \left( u_n^* \left( \frac{\partial \hat{H}}{\partial k_x} \right) u_m \left( \frac{\partial \hat{H}}{\partial k_y} \right) u_n \right)$$  \hspace{1cm} (3)

$$\Omega_{xy}(k) = \sum_n f_n(k) \Omega_{n,xy}(k)$$  \hspace{1cm} (4)

$$\sigma_{xy}^{\text{rash}} = \frac{e^2}{h} \int_{BZ} \frac{dk}{(2\pi)^2} \Omega_{xy}(k)$$  \hspace{1cm} (5)

where the $f_n(k)$ is the occupation factor defined as $f_n(k) = \delta(E_\text{F} - E_n(k)))$, $E_n(k)$ and $u_n$ are $n$th eigenvalue and eigenstate, respectively. Because Fermi energy ($E_\text{F}$) is adjustable in our $\text{CrTe}_2$ system, $\sigma_{xy}^{\text{rash}}$ is expressed as $\sigma_{xy}^{\text{anom}}(E)$.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available on request from the corresponding author. The data are not publicly available due to privacy or ethical restrictions.

Keywords

anomalous Hall effects, ARPES, Berry curvature, low dimensional materials, magnetic semimetals

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