The Weyl semimetal WTe$_2$ has shown several correlated electronic behaviors, such as the quantum spin Hall effect, superconductivity, ferroelectricity, and a possible exciton insulator state, all of which can be tuned by various physical and chemical approaches. Here, a new electronic phase in WTe$_2$ induced by lithium intercalation is discovered. The new phase exhibits an increasing resistivity with decreasing temperature and its carrier density is almost two orders of magnitude lower than the carrier density of the semimetallic T$_d$ phase, probed by in situ Hall measurements as a function of lithium intercalation. The theoretical calculations predict the new lithiated phase to be a potential charge density wave (CDW) phase with a bandgap of $\approx 0.14$ eV, in good agreement with the in situ transport data. The new phase is structurally distinct from the initial T$_d$ phase, characterized by polarization-angle-dependent Raman spectroscopy, and large lattice distortions close to 6% are predicted in the new phase. This finding of a new gapped phase in a 2D semimetal demonstrates electrochemical intercalations close to 6% are predicted in the new phase. This finding of a new phase is structurally distinct from the initial T$_d$ phase, characterized by polarization-angle-dependent Raman spectroscopy, and large lattice distortions close to 6% are predicted in the new phase. This finding of a new gapped phase in a 2D semimetal demonstrates electrochemical intercalations close to 6% are predicted in the new phase. This finding of a new gapped phase in a 2D semimetal demonstrates electrochemical intercalations close to 6% are predicted in the new phase. This finding of a new gapped phase in a 2D semimetal demonstrates electrochemical intercalations close to 6% are predicted in the new phase. 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consistent with the formation of a $2 \times 2$ folding of the Brillouin zone. Concomitantly, lithiation stabilizes the $T_d'$ phase, which is a $2 \times 2$ superlattice of the $T_d$ phase involving a distortion of the W chain along $a$-axis where the W–Te–W bond lengths are shortened and elongated in their successive arrangements in contrast to the identical W–Te–W bond lengths in the $T_d$ phase. Overall, the $a$ and $c$ lattice parameters are strained by 4.9% and 5.84% respectively when WTe$_2$ changes from the $T_d$ to the $T_d'$ phase. Thus, we report the first experimental evidence of a gapped phase, potential CDW, in WTe$_2$ enabled by lithium intercalation.

2. Results and Discussions

2.1. Lithium Intercalation Induced Phase Transition

For lithium intercalation, we construct a WTe$_2$ flake electrochemical cell using liquid or polymer gel electrolyte with capabilities of in situ Raman and in situ transport measurements (Figure 1a, Experimental Section). The exfoliated WTe$_2$ flakes have a lateral size distribution of 20–50 µm and a thickness range of 30–100 nm (>50 layers) (Figure S1, Supporting Information). By controlling the electrochemical intercalation voltage ($V_{EC}$) between the WTe$_2$ flake (cathode) and lithium metal (anode), lithium ions can be controllably inserted into the van der Waals gaps of the WTe$_2$ flake (Figure 1b). Lower values of $V_{EC}$ represent more lithium ions intercalated into WTe$_2$ flakes.

A new structural phase emerges with distinct Raman modes at $V_{EC}$ of 0.8 V vs Li$^+/Li$ in a polymer cell (Figure 1c,d, Figure S2, Supporting Information) or at $V_{EC}$ of 1.2 V vs Li$^+/Li$ in a liquid cell (Figure S3, Supporting Information). In the low frequency region (<50 cm$^{-1}$) of the Raman spectrum where interlayer vibration modes are active, the shear mode of the $T_d$ phase[14] at $\approx 8$ cm$^{-1}$ splits into two peaks at 6 and 13 cm$^{-1}$, as illustrated in Figure 1c. In the high frequency region (>50 cm$^{-1}$) where intralayer phonon modes are populated, several Raman modes with distinct peak positions appear (Figure 1d). These new Raman peaks do not match those observed in the initial $T_d$, $1T'$, or 2H phases of WTe$_2$ (Table S1, Supporting Information).

The Raman spectrum of the new phase has two key characteristics: increased number of Raman active modes compared to the $T_d$ phase and no discernible patterns in the peak shifts of the Raman modes relative to the $T_d$ phase. Therefore, the new Raman modes are not a result of strain or electron doping, but a manifestation of phase transition to a new phase with atomic configurations distinct from the $T_d$ phase. Recently, Muscher

Figure 1. Phase transition in $T_d'$-WTe$_2$ induced by lithium intercalation. a) Schematic of a lithium-ion electrochemical cell with capabilities of in situ Raman and in situ Hall measurements. $V_{EC}$ represents the electrochemical intercalation voltage applied between the Li (silver rectangle) and WTe$_2$ (green rectangle). Adapted under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://creativecommons.org/licenses/by/4.0).[20] Copyright 2021, The Authors, published by Wiley-VCH. b) Side-view of lithium intercalation in the van der Waals gaps of $T_d'$-WTe$_2$ (W: gray, Te: gold, Li: green). c,d) Low-frequency (c) and high-frequency (d) Raman spectra of the $T_d'$ phase (black, bottom) and lithiated $T_d'$ phase (red, top). The insets in (c) are the corresponding optical images of the WTe$_2$ device before (bottom) and after (top) the phase transition. Scale bars: 10 µm. The red circles and black squares in (c) mark the positions of Raman active modes in $T_d$ and the new phase, respectively. The black vertical dashed lines in (c) and (d) are visual guides for the positions of the Raman modes in the $T_d$ phase.
et al. reported a new lithiated Td′-WTe2 phase whose Raman modes match our experimental data (Table S1, Supporting Information). Therefore, we identify the new lithiated phase in this work to be the same as that reported by Muscher et al. and henceforth refer to it as the lithiated Td′ phase (Td′ when referring to the structure without any Li atoms). Optically, when WTe2 is in the lithiated Td′ phase, dark streaks appear along the crystallographic axis b, perpendicular to the direction of the W–W chain, as depicted in the inset of Figure 1c. A closer scrutinization of the streaks via scanning electron microscopy (Figure S4, Supporting Information) indicates them to be wrinkles, suggesting the cause of the dark streaks to be an anisotropic strain induced during the phase transition from Td to Td′.

A representative evolution of the high-frequency Raman peaks as VEC is lowered from open-circuit voltage (OCV) to 0.4 V vs Li+/Li in a polymer cell is shown in Figure 2b with corresponding optical images in Figure 2a. Three distinct stages take place in the course of lithium intercalation. First, WTe2 remains in the Td phase when OCV > VEC > 0.8V vs Li+/Li with negligible changes in the Raman spectra and the optical contrast. In the second stage, when 0.8 V > VEC > 0.4 V vs Li+/Li, WTe2 is in the new lithiated Td′ phase characterized by the distinct Raman modes (VEC = 0.8 V vs Li+/Li between 3000 s to 10 000 s, pink shaded region), the resistance continues to increase and eventually saturates at 7032 Ω, a nearly twenty times increase in resistance from the pristine Td phase. The saturated resistance indicates a completion of the phase transition. At VEC of 0.4 vs Li+/Li, WTe2 starts to break down and becomes amorphous,
accompanied with an irreversible sharp increase in resistance (purple shaded region).

For further characterization of the crystal structure and the electronic band structure of the lithiated Td′ phase, we performed in situ angle-resolved Raman spectroscopy and in situ magneto-transport measurements, which we discuss in the following sections.

2.2. In Situ Angle-Resolved Raman Spectroscopy of the Lithiated Td′ Phase

To characterize the crystal symmetry of the new phase, we performed in situ angle-resolved Raman spectroscopy, as presented in Figure 3 (full spectra in Figure S5, Supporting Information). As a function of the angle θ between the polarization direction of the incident laser and the vertical dashed line shown in Figure 3a, we acquired the angle-dependent Raman spectra of the pristine Td phase (Figure 3b) and the lithiated Td′ phase (Figure 3e). The lithiated Td′ phase has more Raman active modes, albeit with weaker intensities, than Td in the range from 50 to 220 cm−1, suggesting a lower crystal symmetry of the Td′ phase. Additionally, the symmetry of the prominent Raman modes is two-fold in the lithiated Td′ phase, such as the peaks centered at 80 and 210 cm−1, while the symmetry of the Raman modes is mostly four-fold in the Td phase, e.g., peaks at 75, 130, and 214 cm–1. The pole graphs of the Raman modes near 210 cm−1 clearly show the four-lobed and two-lobed nature of the Raman modes in the Td and lithiated Td′ phase, respectively (Figure 3c,f).

2.3. In Situ Magneto-Transport Measurements during Lithium Intercalation

The twenty-fold resistance increase of WTe2 from the initial Td phase to the lithiated Td′ phase suggests there may be a significant change in the electronic band structure of WTe2 in the new Td′ phase. To answer this, we carried out in situ magneto-transport measurements of a WTe2 flake as a function of Li intercalation.

Prior to intercalation, magneto-transport of a pristine WTe2 flake was measured at 2 K, which showed clear Shubnikov–de Haas oscillations in high magnetic fields (Figures S6 and S7, Supporting Information), suggesting a high crystal quality of WTe2. Fitting the $R_{xx}$ and $R_{xy}$ to a two-band transport model\(^{[16,17]}\) showed that the pristine WTe2 flake has an almost equally compensated electron and hole density at 2 K ($n_e = 2.11 \times 10^{20}$ cm$^{-3}$; $n_h = 1.97 \times 10^{20}$ cm$^{-3}$; Figure S6, Supporting Information). The nearly balanced electron and hole population indicates that the Fermi level ($E_F$) is close to zero in pristine WTe2 intersecting the conduction band and valence band equally at 2 K, as predicted by DFT\(^{[16]}\) and visualized in Figure 4a. At 300 K, transport data of the pristine WTe2 flake suggests that electrons are populated more than holes (Figure S8f,g, Supporting Information) and thereby conduction is primarily carried out by electrons at room temperature.

During intercalation, both $R_{xx}$ and $R_{xy}$ were measured in the magnetic field of 3 T at 300 K as $V_{EC}$ was swept from OCV to 0.8 V, as displayed in Figure 4c,d. In accordance with the Raman observation that new Raman modes from Td′ emerge when $V_{EC}$
stays at 0.8 V for 30 min, the red vertical line in Figure 4c,d marks the phase transition point from the Td to the lithiated T′d phase. When WTe2 stays in Td during OCV > VEC > 0.8 V vs Li+/Li, Rxy remains almost unchanged and decreases marginally at 1.0 V vs Li+/Li from 32 to 28 Ω, likely due to electron doping from Li. At VEC of 0.8 V vs Li+/Li, Rxx rises immediately and saturates at 2620 Ω after 50 min at 0.8 V vs Li+/Li, indicating a completion of phase transition from Td to lithiated T′d phase. The increase of Rxx measured with four probes agrees with the two-terminal measurement shown in Figure 2c.

Unlike the steady increase of Rxx at VEC = 0.8 V vs Li+/Li, the Hall resistance Rxy shows a plateau that lasts about 30 min before it starts to increase. According to the in situ Raman data, WTe2 stays as Td phase during the first 30 mins at VEC = 0.8 V. Therefore, the plateau region of Rxy suggests a potential intermediate WTe2 phase, which does not have drastic lattice rearrangements that can be detected by Raman spectroscopy but possesses a distinct electron band structure from that of Td. After the plateau, Rxy increases and eventually saturates at 45 Ω in the new T′d phase. After removing the contribution from Rxx due to the imperfect alignment of the Hall electrodes (see the Experimental Section), the plateau region persists in the corrected Rxy, but the sign changes from positive to negative in the new T′d phase, suggesting a hole-dominated transport (Figure S9, Supporting Information). Figure 4e shows the Hall carrier density (nHall) determined from the corrected Rxy. When OCV > VEC > 0.8 V vs Li+/Li, nHall remains unchanged at 7 × 10¹⁵ cm⁻² as WTe2 is in the Td phase. Assuming the thickness of the flake to be between 50 to 100 nm from the Raman characterization and the color of the flake (Figure S1, Supporting Information), the corresponding 3D carrier density falls in the range of 0.76–1.5 × 10¹⁴ cm⁻³. The spikes seen in nHall correspond to the regions where the differences of Rxy values between 3 and 0 T are very small, and therefore are artifacts and not physically meaningful. At the onset of VEC = 0.8 V, nHall falls considerably to 9.1 × 10¹³ cm⁻² and remains at this density. After ~30 min at VEC = 0.8 V, WTe2 is in the lithiated T′d phase and nHall changes the sign with a further reduced hole carrier density of 1.9 × 10¹⁴ cm⁻².

The observed characteristics of Rxx and Rxy are reproduced in the second round of lithium intercalation (Figure S8a–c, Supporting Information) and in a different Hall bar device (Figure S10, Supporting Information), confirming the reversibility of the phase transition and ruling out the possibilities of electrical contact degradation for the observation. We also confirm that the electrical contacts remain unaffected by intercalation as the magneto-transport behaviors remain unchanged between pristine state and deintercalated state (Figure S8f,g, Supporting Information). Additionally, X-ray photoelectron spectroscopy (XPS) of the deintercalated device shows no significant change from the pristine state, except a slight electron doping effect from residual lithium as evidenced by a 0.4–1.0 eV downshift of the Te 3d binding energy (Figure S11, Supporting Information). Therefore, the increase of resistance and accompanying reduction of carrier density exclusively originate from the change in the electronic band structure.
structure as WTe₂ changes to the new lithiated Td’ phase from the Td phase.

The reduced carrier density and the change of the carrier type suggest a potential gap opening in initially semimetallic WTe₂. To test this hypothesis, temperature-dependent $R_{xx}$ were acquired at pristine state (Figure S12, Supporting Information) as well as at various stages of intercalation by first halting the intercalation through rapid quenching to 200 K, as shown in Figure 4b. At OCV > $V_{EC}$ > 0.8 V vs Li⁺/Li, lithiated WTe₂ remains in the Td phase and $R_{xx}$ vs $T$ curves show a metallic transport with decreasing resistance at lower temperatures. In contrast, at $V_{EC}$ ≤ 0.8 V vs Li⁺/Li, the lithiated WTe₂ behaves like a semiconductor with resistance increasing with decreasing temperature. This suggests that the new lithiated Td’ phase has a bandgap, in contrast to the starting semimetallic Td phase, as illustrated in Figure 4a. We note that the temperature range was limited to 150 K for transport measurements due to the sudden volume change of the polymer electrolyte at that temperature, which breaks the devices. The lithiated phase was measured down to 2 K using a poly(ethylene oxide) (PEO)–lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) polymer electrolyte mixture, as shown in Figure S13 (Supporting Information).

We carried out DFT calculations of the electronic band structure of the Td and Td’ phases (Figure 5a,b) to elucidate the origin of the dramatic differences in the electron transport of these phases. The lithiated Td’ phase has a simple monoclinic conventional unit cell with twice the number of atoms of the orthorhombic Td unit cell, as illustrated in Figure 5a,b. Detailed crystallographic information of Td and lithiated Td’ is summarized in Tables S2 and S3 (Supporting Information). The computed band structure for pristine Td phase in Figure 5c shows that the electron pocket is slightly larger than the hole pocket, consistent with our magneto-transport data of pristine Td state. Moreover, the band structure of the lithiated Td’ phase in Figure 5d confirms that there is a gap opening in the lithiated Td’ phase with a direct gap of 0.66 eV and an indirect gap of 0.14 eV along the $\Gamma$–$X$ direction. The proximity of the Fermi level to the valence band maximum further suggests a hole-dominant transport behavior in lithiated Td’ phase, which confirms the finding of electron to hole transport transition in our in situ magneto-transport measurements.

### 2.4. Mechanism of the Phase Transition

The crystal structure of the lithiated Td’ phase was recently discovered using single crystal X-ray crystallography by Muscher et al., characterized by a subgroup of four distinct in-plane W–W bond distances along crystal $a$ axis, as shown in Figure S14 (Supporting Information).[13] Remarkably, a crystal structure with the same in-plane W–W chain distortion was theoretically predicted to be a 2x2 CDW in monolayer WTe₂.

![Figure 5. a,b) In-plane and cross-sectional atomic structures of Td-WTe₂ (a) and lithiated Td’-Li0.5WTe₂ (b) with W atoms in gray, Te atoms in gold, and Li atoms in green. Unit cells are highlighted with orange dashed lines. c,d) DFT electronic band structures of Td-WTe₂ (c) and lithiated Td’-Li0.5WTe₂ (d) along a high-symmetry K-path in Brillouin zone, as shown in the insets.[21] reveal their semimetallic and semiconducting nature, respectively. The horizontal black dashed lines in (c) and (d) mark the Fermi level for each phase.](image-url)
under electron doping at a level of 0.5 electrons per unit cell (\(9.04 \times 10^{13}\) e cm\(^{-2}\)).[11,12] This strongly suggests that the new phase we observe coincides with the predicted \(2 \times 2\) CDW and that the CDW formation is driven by the extremely high levels of electron doping from Li intercalants.

To confirm the nature of the new phase in bulk and explore the mechanism driving the phase transition, we computed the phonon dispersion of the lithiated T\(_d\) phase in bulk as shown in Figure 6. Our Bader charge\(^{[18]}\) analysis suggests that each Li atom dopes \(\approx 0.85\) e to the Te atoms. Accordingly, we computed the phonon dispersion of the lithiated T\(_d\) phase for two concentrations of electronic doping: 1 Li atom/unit cell (T\(_d\)\(_{\text{Li0.25WTe2}}\)) in Figure 6b and implicit doping at a concentration equivalent to the level of doping by Li in the lithiated T\(_d^\prime\) phase (T\(_d^\prime\)\(_{\text{Li0.5WTe2}}\)) in Figure S15 (Supporting Information). The phonon dispersion of the pristine T\(_d\) phase in Figure 6a shows a stable structure. Upon introduction of 1 Li atom per unit cell, the phonon branch along the X-S-Y path in the Brillouin zone becomes soft, suggesting the instability of the lattice and the possible formation of a CDW, similar to the picture of the CDW phase diagram predicted in monolayer WTe\(_2\).[11] A similar softening is also observed in the case of the implicitly doped T\(_d\) in Figure S15 (Supporting Information), indicating that high level of electron doping alone leads to lattice instability. On the contrary, the lithiated T\(_d^\prime\) phase (T\(_d^\prime\)\(_{\text{Li0.5WTe2}}\)) shows a stable phonon dispersion in Figure 6c while the removal of Li atoms in the T\(_d^\prime\) phase softens the phonon modes along the X–H\(_1\)–C–H–Y path in Figure 6d. Energetically, the phase transition from T\(_d\) to lithiated T\(_d^\prime\)\(_{\text{Li0.5WTe2}}\) is \(-0.62\) eV, whereas it is \(-0.36\) eV going from T\(_d\) to lithiated T\(_d^\prime\)\(_{\text{Li0.5WTe2}}\).

3. Conclusion

A new gapped phase is discovered in semimetallic T\(_d\)\(_{\text{WTe2}}\), stabilized by an exceptionally high level of electron doping from lithium intercalation. We identified the structure of the new phase to be a lithiated T\(_d^\prime\) phase by in situ angle-resolved Raman spectroscopy. A bandgap opening in the new phase is suggested by in situ transport measurements that show an increasing resistance with decreasing temperature and a much-reduced carrier density. Our DFT calculations show that this new lithiated T\(_d^\prime\) phase has an indirect gap of 0.14 eV and a direct bandgap of 0.66 eV. Furthermore, the structure of the lithiated T\(_d^\prime\) phase is identical to the unit cell of the predicted \(2 \times 2\) CDW in WTe\(_2\) monolayer by electron doping,[11] and phonon dispersion calculations show that lithiation induces phonon softening in the T\(_d^\prime\) phase at various electron doping levels. Therefore, we identify this new lithiated T\(_d^\prime\) phase to be a potential \(2 \times 2\) CDW in WTe\(_2\). A careful crystallographic study of a single WTe\(_2\) device via in situ diffraction is necessary to directly verify the nature of CDW.

It is notable that the new lithiated T\(_d^\prime\) phase lands at the high electron doping end in the electronic phase diagram of WTe\(_2\), past the superconducting phase of WTe\(_2\).[13] So far, superconductivity in WTe\(_2\) has been observed in monolayers by gating\(^{[7]}\) or in the bulk by pressure.\(^{[10]}\) Since electron doping in bulk WTe\(_2\) requires chemical doping rather than electrical field gating, lithium intercalation can be used to dope “bulk-like”
WTe$_2$ flakes to potentially induce superconductivity as lithium intercalants present in every van der Waals gap can dop every WTe$_2$ layer. Lastly, we envision that the exceptional electron doping using lithium intercalation can also introduce CDW in other 2D transition metal dichalcogenide systems that are not known to host CDW in their intrinsic state, such as 1T-MoTe$_2$. Thus, the greatly expanded electronic and structural phase diagram of 2D materials accessible using lithium intercalation will stimulate the research for novel quantum phases, such as topological superconductivity. Additionally, the sharp switch of electrical resistance of layered WTe$_2$ by controllable tuning of electro-chemical intercalation voltage projects prospective applications in resistive memory.

4. Experimental Section

Device Fabrication: The WTe$_2$ flakes were mechanically exfoliated from bulk WTe$_2$ crystals purchased from 2D Semiconductors. WTe$_2$ flakes with desired lateral size and thickness were wet transferred onto SiO$_2$/Si wafer with the assistance of KOH.[23] The thickness of the flake was determined by a Cypher ES atomic force microscope from Asylum Research. For the device fabrication, electrodes were written by electron beam lithography (Nanofabrication G4 FIB-SEM) at a voltage of 30 kV and a current of 1.6 nA with a dose of 410 $\mu$C cm$^{-2}$. The developed devices were deposited with 15 nm Cr and 200 nm Au using a thermal evaporator (MBraun EcoVap) at a pressure of 10$^{-7}$ mbar. The devices were transferred into an argon glovebox with an O$_2$ and H$_2$O level below 1 ppnm right after the lift-off to minimize the oxidation.

Electrochemical Intercalation Cell Fabrication: All the intercalation cells discussed in the main text adopted a planar cell configuration with both the WTe$_2$ electrode and lithium electrode on a piece of transparent glass slide or SiO$_2$/Si wafer. The detailed fabrication steps are described in our previous research papers.[22,24] For the liquid cells, the liquid electrolyte of 1 M LiPF$_6$ (EC/DEC, v:v = 50:50, battery-grade, Sigma-Aldrich) was injected into the cell to submerge the lithium and the device. For polymer cells, 0.227 g 1 M LiPF$_6$ solution was premixed with 0.475 g poly(ethylene glycol) methyl ether methacrylate (PEGMA) and 1.145 g bisphenol A ethoxylate dimethacrylate (BEMA) in a tarnished glass vial and stirred overnight inside the glovebox. Then, 46 mg of photoinitiator, bisphenol A ethoxylate dimethacrylate (BEMA) in a tarnished glass vial were carried out by first freezing the electrochemical intercalation through a rapid cooling to 200 K at a rate of 10 K min$^{-1}$, at which point the intercalation current running through the cell gradually goes to zero. Then the system was gradually cooled to 150 K at a rate of 2 K min$^{-1}$.

Post Mortem Characterizations: Intercalation devices were disassembled by physically removing the liquid or polymer electrolyte by a razor blade. Then the device was immersed in isopropanol to further remove the residual electrolyte. The cleaned device was characterized by Raman spectroscopy (Horiba LabRAM HR Evolution Spectrometer; 633 nm) and SEM (Helios G4 FIB-SEM) with a stage tilting angle of 50° at a voltage of 5 kV and a current of 25 pA. XPS of deintercalated device and exfoliated flake was performed on a PHI VersaProbe II Scanning XPS Microprobe with an Al K$_\alpha$ monochromatic X-ray source. A beam spot with the diameter of 20 μm was used to obtain XPS data only from the device, assisted by scanning X-ray induced secondary electron imaging. All of the XPS spectra were calibrated using a carbon 1s peak located at 284.50 eV.

Ab Initio Calculations: Density functional theory (DFT) calculations were carried out in a plane-wave basis set within the projector augmented wave (PAW) approach[25] as implemented in the Quantum Espresso software package.[26] The exchange-correlation was treated at the generalized gradient approximation (GGA) level of Perdew, Burke, and Ernzerhof (PBE).[27] The van der Waals interactions were accounted for using Grimme’s D3 correction with Becke–Johnson damping.[28] A kinetic energy cut-off of 1040 eV was used for the expansion of the plane-wave basis in all the calculations, and the Brillouin zone was sampled using Gamma-centered Monkhorst–Pack[29] k-point meshes of $32 \times 18 \times 8$.
and $16 \times 18 \times 8$ for the $T_d$ (12 atoms) and $T_d'$ (24 atoms) unit cells respectively. The unit cells were relaxed until the total energy and the force on each atom converged to within 0.02 meV/atom$^{-3}$ and 0.03 eV Å$^{-3}$, respectively. The lithiated $T_d$ phase was relaxed with the volume fixed to that of the pristine $T_d$ phase to prevent a doping-induced phase transition. Spin–orbit coupling with noncolinear magnetization was used for all band structure calculations. Phonopy was used for the calculation of phonon bands, with supercell sizes of $2 \times 2 \times 2$ and $2 \times 2 \times 1$ for $T_d$ and $T_d'$-$\text{WTe}_2$ phases, and $q$-meshes of $8 \times 4 \times 2$ and $4 \times 4 \times 4$ were used respectively. For implicitly doped $T_d$, a $2 \times 2 \times 1$ supercell with a $q$-mesh of $4 \times 4 \times 4$ was used.

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 from the corresponding author upon reasonable request.

Keywords

charge density waves, electron doping, lithium intercalation, phase transitions, tungsten ditelluride

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