Pressure-induced isostructural electronic topological transitions in 2H-MoTe₂: X-ray diffraction and first-principles study

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Abstract. Synchrotron X-ray diffraction measurements on powder 2H-MoTe₂ (P6₃/mmc) upto ~ 46 GPa have been performed along with first-principles based density functional theoretical (DFT) analysis to probe the isostructural transition in low pressure regime and two electronic topological transitions (ETT) of Lifshitz-type in higher pressure regime. The low pressure isostructural transition at ~ 7 GPa is associated with the lattice parameter ratio c/a anomaly and the change in the compressibility of individual layers. The pressure dependence of the volume by linearizing the Birch–Murnaghan equation of state as a function of Eulerian strain shows a clear change of the bulk modulus at the ETT pressure of ~ 20 GPa. The minimum of c/a ratio around 32 GPa is associated with the linkage of electron pockets marked as second ETT of Lifshitz-type. We do not observe any structural transition up to the maximum applied pressure of ~ 46 GPa under quasi-hydrostatic condition.

Keywords: Lifshitz transition, Electronic topological transition, High pressure, Density-functional theory, Transition metal dichalcogenide, X-ray diffraction

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1. Introduction

Transition metal dichalcogenide (TMD) materials have attracted renewed interest following the recent discovery of Weyl semimetallic phases of both I- and II-types with the non-trivial electronic topological signatures [1, 2, 3, 4, 5, 6, 7]. Layered TMDs with the common formula MoX_2 , where X is a chalcogen element (S, Se, Te), consist of X-Mo-X sheets being stacked along c-direction and held together by van der Waals (vdW) interaction. The commonly studied structural form of TMDs is the semiconducting hexagonal phase (2H) where Mo atoms are trigonal-prismatically coordinated by the chalcogenide atoms [8]. One of the unique properties of these MoX_2 compounds is that they can exist in different structural forms (polymorphism) such as 2H, 1T, 1T', 3R and T_d -type lattices with different electronic properties [9, 10]. Few layer 1T'- MoTe_2 having monoclinic crystal structure with a bandgap [11] of ~ 60 meV is predicted to be a potential platform to study the $\mathbb{Z}_2$ -topological quantum devices [12]. In case of three-dimensional (3D) topological insulators (TIs) of A_2B_3 types (A=Bi, Sb; B=Se, Te), it is the rhombohedral phase [13, 14, 15] which holds $\mathbb{Z}_2$ -electronic topological signatures whereas for TMDs, it is the orthorhombic phase which shows non-trivial Weyl-topological signatures of type-II reflected in the observation of Fermi arcs on the surface [3, 5, 16], providing a potential route in realization of quantum spin Hall effect. Recently, superconductivity is observed in Td- MoTe_2 with $T_c = 8.2\text{K}$ at 11.7 GPa leading to a new pathway for the exploration of topological superconductivity [17, 18]. In addition, monolayer TMDs (direct band gap) have inequivalent valleys at the corners of the Brillouin zone which mark them as potential candidates for two-dimensional valleytronics [19, 20] as well as optoelectronics [21] at room-temperature [22, 23].

Tuning among different polytypes and thereby changing their electronic properties is attractive from application point of view such as making ohmic contacts by using homojunction [24] between the metallic and semiconducting phases of TMDs; varying the concentration of W in solid solution of $\text{Mo}_{1-x}\text{W}_x\text{Te}$ to induce semiconducting-2H to metallic-Td phase at room temperature [25]; and semiconducting-2H to metallic-1T' phase via charge carrier doping [26]. Applying pressure is one of the proven routes to induce structural polytypes as observed in 2H- MoS_2 ($P6_3/mmc$) for which, the onset of lateral shift of

the adjacent S–Mo–S layers is around ~ 20 GPa leading to a mixed phase of $2H_c$ (2H) and $2H_a$ structures with the $2H_c$ -phase being the dominant one [27] and thereby changes the pressure coefficients of the Raman modes [28]. The layer sliding transition in MoS₂ from $2H_c$ (semiconductor) to $2H_a$ (metal) completes at ~ 40 GPa [28]. Recently, it has been shown that the Mo-based ternary chalcogenide MoSSe (P6₃mc) also undergoes isostructural layer sliding transition from $2H'_c$ to a mixed phase of $2H'_c+2H'_a$ around 11 GPa [29]. However, it was shown [30] experimentally as well as theoretically that the similar compound MoTe₂ (P6₃/mmc) does not undergo structural transition upto the maximum reached pressure of 30 GPa. Even high pressure Raman and X-ray diffraction studies on MoSe₂ (P6₃/mmc) [31] confirm that there is no structural transition upto 36 GPa. Recent Raman spectroscopic measurements as well as density functional theoretical (DFT) calculations showed [30] that the bulk 2H-MoTe₂ undergoes two transitions as a function of pressure : at ~ 6 GPa and 16.3 GPa associated with semiconducting to semimetallic transition and electronic topological transition of Lifshitz-type, respectively. It is worth noting that ETT of Lifshitz-type is quite distinct in topological sense from the ETT of $\mathbb{Z}_2$ -type, involving dynamical corrections to the anomalous signatures of phonon modes [14].

For 2H-MoTe₂, we showed [30] that reliable Raman data from diamond anvil cell could not be captured using the methanol–ethanol (4:1) mixture as the pressure transmitting medium, and it was attributed to the adsorption of alcohols on the surface of MoTe₂ and the dissociation through thermodynamically favorable channels [32]. In this work, we report X-ray diffraction (XRD) studies on bulk 2H-MoTe₂ as a function of pressure using Ne gas as a pressure transmitting medium (PTM) to probe the changes in the lattice parameters around two phase transitions mentioned above where Raman modes showed changes in pressure coefficients [30]. The isostructural semiconducting to semimetallic transition coincides with a sharp decrease in the ratio c/a and the decrease of compressibility of individual layers around ~ 7 GPa. The ETT of Lifshitz type at ~ 20 GPa is shown to be accompanied by changes in bulk modulus. In addition to this, our study brings out another ETT of Lifshitz type around ~ 32 GPa associated with a minimum in c/a ratio and linkage of electron pockets at K and K₂ points in Brillouin zone.

2. Experimental and Computational Details

The sample of 2H-MoTe₂ [30, 33] was a gently pelletized powder obtained from crushing a single crystal and pressed between two glass slides. The lateral size of the sample was $\sim 50\text{-}60\ \mu\text{m}$, estimated from the microscope image, with a thickness less than $60\ \mu\text{m}$ (the measured thickness of the indented gasket). It was loaded along with a ruby chip inside a hole of diameter $\sim 150\ \mu\text{m}$ in a rhenium gasket inserted between the diamonds (of culet size $300\ \mu\text{m}$) of a symmetric diamond anvil cell (DAC). We confirmed that the sample thickness was well within the height of the indented gasket and it was almost in the center of the hole. High pressure X-ray diffraction experiments on MoTe₂ (P6₃/mmc, # 194) upto $\sim 46\ \text{GPa}$ were carried out using synchrotron radiation on beam line P02.2 [34] at PETRA-III (DESY), Hamburg, Germany. Ne gas was used as a pressure transmitting medium. The X-ray wavelength used for the experiment is $0.288\ \text{\AA}$. The sample to detector distance was calibrated using LaB₆. The pressure was determined using well known ruby fluorescence shift (the pressure uncertainty in our experiment is $\sim \pm 0.1\ \text{GPa}$). Only one small ruby ($\leq 10\text{-}12\ \mu\text{m}$) was loaded in the sample chamber for pressure calibration. DIOPTAS software [35] was used to integrate the Deby-Scherrer diffraction rings and to convert it into 1D pattern (intensity versus diffraction angle (2θ)). The lattice parameters (a and c) are derived from Rietveld refinements for all the XRD patterns using GSAS II software package [36]. At the end of every Rietveld refinements (after refining atomic coordinates), spherical harmonics (of order 8 to 10 with texture index ~ 1.8) were used to model the preferred orientation corrections. Our first-principles calculations are based on density functional theory (for details, see ref. [30]).

3. Results and Discussion

Rietveld analysis of XRD pattern at $0.3\ \text{GPa}$ is performed using GSAS II software package and plotted in Fig. 1(a) along with (hkl)-reflections. The Mo and Te atoms occupy 2b Wyckoff site and 4f Wyckoff site, respectively, same as for 2H-MoS₂ or MoSe₂ structure (P6₃/mmc) (see the inset of Fig. 1a). Fig. 1(b) represents corresponding two-dimensional XRD image at $0.3\ \text{GPa}$ without masking for better visualization. Fig. 2 shows three XRD patterns

taken at ~ 6.9 GPa, 26.1 GPa and 45.9 GPa. Upon compression of MoTe₂ upto ~ 46 GPa, all the XRD peaks shift to larger 2θ -values (smaller d-spacings, see Figs. S1 and S2 in Supplementary Materials [37]), as expected due to shrinkage of the 2H-MoTe₂ lattice. We do not observe any new peak(s) in the pressure range upto 46 GPa (see Fig. S1a).

Figure 1. (color online) – (a) Rietveld refinements algorithm used to match the patterns to P6₃/mmc crystal structure at 0.3 GPa with the GSAS II software package. The experimental data are indicated by red circles, the calculated pattern is the blue solid line, and the lower curve drawn in dark-green is the weighted difference between the observed and calculated pattern. The profile factor $R_p = 0.64\%$ and the weighted profile R-factor $R_{wp} = 0.99\%$. Inset shows the unit cell stacking of atoms along c -direction. (b) corresponding two-dimensional XRD image at 0.3 GPa.

The extracted lattice parameters (a , c) are plotted as a function of pressure in Figs. 3(a) and (b). The c -axis shows faster decrement up to ~ 7 GPa and then it slows down. The compression of c -axis is stronger than the a -axis as expected for an anisotropic layered material where stacked

Figure 2. (color online) – Rietveld refinement patterns to $P6_3/mmc$ crystal structure at (a) 6.9 GPa, (c) 26.1 GPa and (e) 45.9 GPa with the GSAS II software package. The profile R_p -factors (and R_{wp}) are for (a) 0.71 % (1.01 %), (b) 0.68 % (1.01 %) and (c) 0.69 % (1.02%). The corresponding two-dimensional XRD images at (b) 6.9 GPa, (d) 26.1 GPa and (f) 45.9 GPa are also shown.

layers along c are connected via weak van der Waals interactions. By fitting 3rd order Birch-Murnaghan equation of state [38, 39] (see Eq.(1)), the obtained compressibility's at ambient pressure (and pressure derivative of bulk modulus) for c -axis and a -axis are $\sim 4.7(1) \times 10^{-3} \text{ GPa}^{-1}$ ($B'_{0c}=25.9\pm2.8$) and $\sim 1.6(01) \times 10^{-3} \text{ GPa}^{-1}$ ($B'_{0a}=13.2\pm0.3$), respectively. The c -axis and a -axis

are reduced by $\sim 14.2\%$ and 10.3% , respectively over the whole pressure range of 46 GPa in agreement with the report by Zhao et al. [40]. Fig. 3(c) shows the variation of ratio c/a as a function of pressure displaying following changes: (i) sharp decrement of c/a by ~ 0.12 within the first 7 GPa (marked by an arrow); (ii) from 7 GPa, it goes to a minimum value around ~ 32 GPa followed by an increment in c/a (by ~ 0.03) upto the maximum applied pressure of 46 GPa. The sharp decrease in c/a at ~ 7 GPa in accordance with the reported [30] semiconducting to semimetallic transition is related to the decrease in interlayer spacing and hence induced semi-metallicity because of Te-orbitals overlap.

The normalized lattice constants along with the normalized unit cell volume are plotted in Fig. 3(d), and it is clear that there is no volume discontinuity upto 46 GPa. It is worth noticing from Fig. 3(d) that the separation between the normalized lattice constants decrease with the increase of pressure and the separation $(a/a_0 - c/c_0)$ is plotted in the inset of Fig. 3(b). Sharp increase in $a/a_0 - c/c_0$ upto 7 GPa, to a maximum in $a/a_0 - c/c_0$ around 32 GPa and then decrement upto 46 GPa corroborates with the c/a data.

Now, we present our DFT calculated lattice parameters (a , c) variations with pressure (solid blue lines in Figs. 3a and b). It is clear that compression of c -axis is faster than that for a -axis for first 8 GPa. From Fig. 3(c), we mark (shown by an arrow) another transition around 18 GPa where calculated c/a ratio goes to a minimum (discussed later in detail). The calculated normalized lattice parameters (see Fig. 3d) shows that the separation between a/a_0 and c/c_0 decreases with the increase of pressure in corroboration with our experimental observations.

The ratio of interlayer (Te-Te) to intralayer (Mo-Te) distance is plotted in Fig. 4(a); showing clearly that upto 7 GPa, the ratio decreases rapidly implying that the interlayer vdW distance reduces leading to quasi three-dimensional solid-network beyond 7 GPa. We mark another transition around 20 GPa where the ratio changes its slope. Here, it is worth to monitor the Te atom positions with pressure as only z -coordinate of Te position (z -Te) is allowed to vary (Mo atom is fixed) according to the constraints imposed on Wyckoff positions by space group $P6_3/mmc$. The individual layer compressibility (ILC)

[41] is related to z-Te atom and the rate of ILC ($-1/z(dz/dP)_{Te}$) is plotted in the inset of Fig. 4(a) clearly showing a rapid decrease upto 7 GPa and after that it varies very slowly in linear fashion upto ~ 20 GPa. This in-turn also indicates that the Te-Te orbitals start to overlap and beyond 7 GPa the Coulomb repulsion prevails to slow down the rate. Our marking of 7 GPa from XRD data for semiconductor to semimetal transition coincides with the recent high pressure transport measurements where it has been shown that the resistivity ratio (ρ_{300K}/ρ_{2K}) goes above 1 around 9 GPa [40] which is 2 GPa higher, probably due to the non-hydrostatic component of pressure transmitting medium (Daphne oil 7373) [40]. Our DFT calculated ratio of interlayer (Te-Te) to intralayer (Mo-Te) distance is plotted in Fig. 4(b) which brings the above mentioned two transitions at ~ 8 and 18 GPa, respectively. The calculated ILC (see inset of Fig. 4b) also marks the two transitions in agreement with our experimental observations. Both experimentally observed and calculated ILC show qualitatively similar trends along with its almost invariant behavior after ~ 32 GPa. The robustness of our XRD-refined z-Te is further confirmed by comparing with the DFT calculated values as a function of pressure (see Fig. S3 for details).

The P-V data (Fig. 5a) is fitted with the third-order Birch-Murnaghan (BM) equation of state [38, 39]:

$$P = \frac{3B_0}{2} \left[\left(\frac{V}{V_0} \right)^{-7/3} - \left(\frac{V}{V_0} \right)^{-5/3} \right] \left\{ \left[1 + \frac{3}{4}(B'_0 - 4) \right] \left[\left(\frac{V}{V_0} \right)^{-2/3} - 1 \right] \right\} \quad (1)$$

From the fitting up to 7 GPa (semiconductor-semimetal transition), the bulk modulus at ambient pressure B_0 is 31.4 ± 5.1 GPa, its pressure derivative $B'_0 = 11.1 \pm 3.4$ and the ambient pressure volume $V_0 = 150.56 \pm 0.37 \text{ \AA}^3$. From the fitting from 7 GPa to maximum pressure, those values are $B_0 = 57.3 \pm 2.6$ GPa, $B'_0 = 4.9 \pm 0.2$ and $V_0 = 146.73 \pm 0.56 \text{ \AA}^3$. The bulk modulus is higher in the metallic phase as compared to the semiconducting phase [40]. For DFT calculated P-V values and the corresponding fitted values (B_0 , B'_0 and V_0) are shown in Fig. 5(b). Around 8 GPa (semiconductor to semimetal transition), we couldn't get appreciable B_0 values from our DFT calculated PV-values : before 8 GPa, $B_0 = 58.8 \pm 11.8$ GPa and $V_0 = 141.94 \pm 1.86 \text{ \AA}^3$ (using 2^{nd} order BM equation) and after 8 GPa, $B_0 = 63.3 \pm 21.9$ GPa, $B'_0 = 5.3 \pm 1.3$ and $V_0 = 139.63 \pm 4.14 \text{ \AA}^3$ (using 3^{rd} -order BM equation).

Figure 3. (color online) – Here, points and solid lines are used for XRD and DFT calculated data, respectively. The pressure variations of lattice parameters a , c and c/a ratio are shown in (a), (b) and (c), respectively. The red solid lines in (a) and (b) represent fitting of 3rd order Birch-Murnaghan equation of state. Pressure dependent normalized lattice constants and the unitcell volume are plotted in (d). Error bars, less than the size of the data points, are also plotted. The inset of (b) shows the plot of $(a/a_0 - c/c_0)$ as a function of pressure.

Figure 4. (color online) – The ratio of interlayer (Te-Te) to intralayer (Mo-Te) distance is plotted as a function of pressure for (a) XRD refined and (b) DFT calculated values. Here, Rietveld refined coordinates are used to calculate the distances. Error bars, less than the size of the data points, are also plotted. Insets of (a) and (b) : rate of individual layer compressibilities (ILC) are plotted. The red dashed curves in (a) correspond to the fitting using $f(P)=p+qP+rP^2$, where p, q and r are constants. Three different regions are marked as I, II and III.

Figure 5. (color online) – (a) XRD refined and (b) DFT calculated P-V data are shown. The fitted lines using 3rd-order BM equation are shown in solid lines. Error bars, less than the size of the data points, are also plotted in (a). (c) XRD refined and (d) DFT calculated reduced pressure H vs. Eulerian strain (f) plots. The transition regions at which H changes its slope are marked in grey rectangular areas; the crystal structure remains the same up to maximum observed pressure of ~ 46 GPa. Marked LT represents Lifshitz transition. Here, regime I is excluded where trivial semiconductor-semimetallic transition occurred. Four different regions are marked as I, II, III and IV.

The ratio of interlayer (Te-Te) to intralayer (Mo-Te) distance and ILC both show a transition around 20 GPa. The transition at ~ 20 GPa coincides with the Lifshitz transition (LT) seen in our recent DFT studies [30] on 2H-MoTe₂ where ETT has been marked by the abrupt appearance of electron and hole pockets. Near ETT, no volume discontinuity (first derivative of Gibbs free energy) is expected but the changes in second derivatives of Gibbs free energy such as compressibility is expected to occur. Therefore, a better way to capture the signatures of subtle structural changes is to express Eq.(1) in terms of Eulerian strain (f) and reduced pressure (H) [38] :

$$H = B_0 + \frac{3}{2}B_0(B'_0 - 4)f \quad (2)$$

where, $f = \frac{1}{2}(X^2 - 1)$, $X = \left(\frac{V}{V_0}\right)^{-1/3}$ and reduced pressure $H = P/\{3f(1 + 2f)^{5/2}\}$.

The reduced pressure H scales linearly with Eulerian strain f . In absence of a transition, H is linearly dependent on f . Fig. 5(c) shows the slope change of H at ~ 20 GPa (marked as LT) where the changes of B_0 and B'_0 are also marked in Fig. 5(c) : $B_0 = 38.7 \pm 0.8$ GPa and $B'_0 = 7.7 \pm 0.3$ before the LT, $B_0 = 45.8 \pm 0.6$ GPa and $B'_0 = 5.7 \pm 0.1$ after the LT. Therefore, the discontinuity observed in H - f plot indicates that there are subtle structural modifications attributed to the electronic band structure modifications seen in DFT calculations on 2H-MoTe₂ [30]. Our calculated reduced pressure vs. Eulerian strain also confirms the ETT of Lifshitz-type around 18 GPa with another clear transition at ~ 34 GPa as shown in Fig. 5(d). As we know that there are no structural transitions or volume discontinuity for MoTe₂ [30] around 32 GPa (see Fig. 3d or Fig. 5a), therefore the possibility could be related to the changes in the electronic structures.

From our electronic structure calculations, we find that hole pocket appears at M-point at $P \sim 30$ GPa in addition to hole pockets seen at Γ and A points observed at 20 GPa [30] (Figs. 6 and 7). Further increase in pressure leads to an increase in the size of the hole pocket at M point. The bands forming electron pockets at K and K₂ points intersect (or cross) each other in the vicinity of K₂ point as shown in Fig. 6(a). At 30 GPa, this crossing of the band is just above the Fermi level which moves towards the Fermi level with increasing hydrostatic pressure. At 34 GPa, it is at Fermi level which leads to interesting change in Fermi surface topology. To probe this further,

Figure 6. (color online) – Electronic structure of 2H-MoTe₂ at (a) 30 GPa, (b) 32 GPa, (c) 34 GPa, (d) 36 GPa. The highlighted areas show the band crossing moves towards Fermi level with increasing hydrostatic pressure.

we investigate the evolution of the Fermi surface with pressure. At 34 GPa, the hole pockets are observed at M-point (see Fig. 7a, the blue pockets at the center of the sides of the hexagon) and the electron pockets are seen at K and K₂ points (Fig. 7, the purple pockets mark the electron pocket). We find the Fermi surface associated with the bands at K and K₂ points changes (topology of the Fermi surface) at an applied hydrostatic pressure of $P \sim 34$ GPa (see Figs. 7b-d). Since, the Fermi surface changes with applied pressure without breaking the structural symmetry, we assign it to a Lifshitz transition at P

~ 34 GPa. Therefore, our observed minimum in c/a ratio at 32 GPa (see Fig. 3c) is associated with the second Lifshitz transition. The electronic band structures at a few low pressures ($\sim 6, 12, 18$ and 24 GPa) are shown in Fig. S4 [37].

Figure 7. (color online) – Fermi surfaces at (a) 34 GPa of merged bands (all the bands crossing the Fermi levels). Panels (b), (c) and (d) show the electron pocket of Fermi surface changing with pressure at $P=32$ GPa, 34 GPa and 36 GPa respectively. Note that (b), (c) and (d) show the electron pockets at K and K_2 points with pressure. Note that purple color pockets show electron pockets whereas blue and red pockets (at the center and center of the sides of the hexagon) show hole pockets.

4. Conclusions

In summary, high pressure X-ray diffraction measurements have been performed along with first-principles based density functional theoretical analysis to show low pressure iso-structural and high pressure two electronic topological transitions of Lifshitz-type. Semiconducting to semimetallic

transition at ~ 7 GPa is related to the sharp decrease in c/a ratio and in rate of compressibility of individual layers. DFT-predicted electronic topological transition of Lifshitz-type at ~ 20 GPa is accompanied by the changes of bulk modulus as deduced in the reduced pressure vs. Eulerian strain plot. The minimum in c/a ratio at 32 GPa is associated with the second Lifshitz transition. We hope that our work will stimulate further high pressure transport studies as well as thermoelectric measurements to capture the anomalies near the Lifshitz transitions at ~ 20 and 32 GPa.

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Supplementary Materials – Pressure-induced isostructural electronic topological transitions in 2H-MoTe₂ : X-ray diffraction and first-principles study

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1. XRD PATTERNS OF MoTe_2 AT FEW ELEVATED PRESSURES

Figure S 1 – (color online)– (a) XRD patterns at a few elevated pressure, here linear scale is shown for intensity (y-axis). (b) For comparison, two XRD patterns at 4.9 GPa and 12.4 GPa are plotted : We do not observe appearance or disappearance of XRD lines between them. Also, there is no appreciable intensity variations of (100), (103) lines or any other intense lines [1].

We note that the layer-sliding isostructural 2H_c (semiconductor) to 2H_a (metal) transition is observed for MoS_2 [2] and MoSSe [3]. Here we rule out the possible isostructural 2H_c (semiconductor) to 2H_a (metal) transition around 7 GPa for MoTe_2 and the reasons are following: (1) We do not observe any a-lattice contraction or c-lattice expansion for MoTe_2 around 7 GPa as observed for MoS_2 [2] near isostructural semiconducting to semimetal transition; (2) No observations of appearance or disappearance of XRD lines around 7 GPa (see Fig. S1(b) for details) and no appreciable intensity variations among (100), (103) and (105) as observed for MoS_2 [2]; (3) No observations of discontinuity in the variations of d-spacings for intense (100), (103), (203) and (213) lines, see Fig. S2 for details. These observations from our XRD data are in agreement with previous report [4].

2. *d*-SPACINGS AS A FUNCTION OF PRESSURE

Figure S 2 – (color online)– *d*-spacings for (a) (100), (b) (103), (c) (203) and (d) (213) are plotted as a function of pressure. Third-order Birch-Murnaghan (BM) equation of state is fitted for all of them and shown in red solid lines showing continuous smooth variations with pressure.

3. COMPARISON OF $z\text{-Te}$ AS A FUNCTION OF PRESSURE

Figure S 3 – (color online)– Pressure evaluation of refined fractional z-coordinate of Te atom from XRD (error bars, less than the size of the data points, are also plotted) and compared with DFT calculated values.

Fig. S3 shows the comparison of pressure evaluation of fractional z-coordinate of Te atom between the Rietveld refined and DFT calculated values which are in well agreement confirming the robustness of our Rietveld refined methods.

4. ELECTRONIC BAND STRUCTURE OF 2H-MoTe₂ AT A FEW PRESSURE VALUES

Figure S 4 – (color online)– Electronic structure of 2H-MoTe₂ at (P =) (a) 6 GPa, (b) 12 GPa, (c) 18 GPa, (d) 24 GPa.

2H-MoTe₂ is an indirect bandgap semiconductor with a gap 0.57 eV (valance band maximum (VBM) at Γ and conduction band minimum (CBM) at the centre of the Γ -K path) (refer to Fig. 1(b,) [5]). With increasing pressure, this indirect bandgap reduces and closes at 8 GPa, marking semiconductor to semimetal transition. Further, the gap observed at K-point reduces and closes at 20 GPa leading to a change in the topology of Fermi surface. At 24 GPa, the bands at K-points crosses each below Fermi level.

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