Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary Information

Spin splitting and reemergence of charge compensation in monolayer WTe_2 by 3*d* transition-metal adsorption

Yan Song,[a] Xiaocha Wang[b] and Wenbo Mi*[a]

[a]Tianjin Key Laboratory of Low Dimensional Materials Physics and Preparation Technology, School of Science, Tianjin University, Tianjin 300354, China

[b]School of Electronics Information Engineering, Tianjin University of Technology, Tianjin 300384, China

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*Author to whom all correspondence should be addressed.

E-mail: miwenbo@tju.edu.cn

S1. Band folding of monolayer WTe_2

The relation between wave vectors $\boldsymbol{k}$ and $\boldsymbol{k}'$ can be obtained via the translational symmetry operation, which is given by

$$\boldsymbol{k}' = k \pm n\boldsymbol{b}_1' \pm m\boldsymbol{b}_2' \tag{1}$$

where $\boldsymbol{k}$ and $\boldsymbol{k}'$ represent the vectors of the primitive cell and the supercell respectively, $\boldsymbol{b}_1'$ and $\boldsymbol{b}_2'$ are the reciprocal-lattice vectors of the supercell, n and m are the integer numbers. In monolayer WTe_2, the basic vectors of the primitive cell $\boldsymbol{a}_1$ and $\boldsymbol{a}_2$ as indicated by the red arrows in Fig. 1(a) are expressed as

$$\boldsymbol{a}_1 = (a_1, 0), \boldsymbol{a}_2 = (0, a_2) \tag{2}$$

where a_1 and a_2 are the lattice constants of the primitive cell. The basic vectors of the supercell $\boldsymbol{a}_1'$ and $\boldsymbol{a}_2'$ as indicated by the blue arrows in Fig. 1(a) are expressed as

$$\boldsymbol{a}_1' = (3a_1, 0), \boldsymbol{a}_2' = (0, 2a_2) \tag{3}$$

Thereby, the reciprocal-lattice vectors for the primitive cell and the supercell are obtained, respectively

$$\boldsymbol{b}_1 = (\frac{2\pi}{a_1}, 0), \boldsymbol{b}_2 = (0, \frac{2\pi}{a_2}) \tag{4}$$

$$\boldsymbol{b}_1' = (\frac{2\pi}{3a_1}, 0), \boldsymbol{b}_2' = (0, \frac{\pi}{a_2}) \tag{5}$$

Hence, based on the reciprocal-lattice vectors, the coordinates of the high symmetry points of the primitive cell are expressed as

$$\Gamma = (0,0), X = (\frac{\pi}{a_1}, 0) \tag{6}$$

$$Y = (0, \frac{\pi}{a_2}), S = (\frac{\pi}{a_1}, \frac{\pi}{a_2}) \tag{7}$$

The high symmetry points of the supercell are expressed as

$$\Gamma' = (0,0), X' = (\frac{\pi}{3a_1}, 0) \tag{8}$$

$$Y' = (0, \frac{\pi}{2a_2}), S' = (\frac{\pi}{3a_1}, \frac{\pi}{2a_2}) \tag{9}$$

Under equation (1), we get

$$X - \boldsymbol{b}_1' = X' \tag{10}$$

$$Y - \boldsymbol{b}_2' = \Gamma' \tag{11}$$

$$S - \boldsymbol{b}_1' - \boldsymbol{b}_2' = X' \tag{12}$$

Hence, X and S points of the primitive cell are folded to X' point of the supercell and the Y point is folded to Γ' point.

S2. Effects of SOC and Coulomb interaction on band structure of Mn-WTe_2

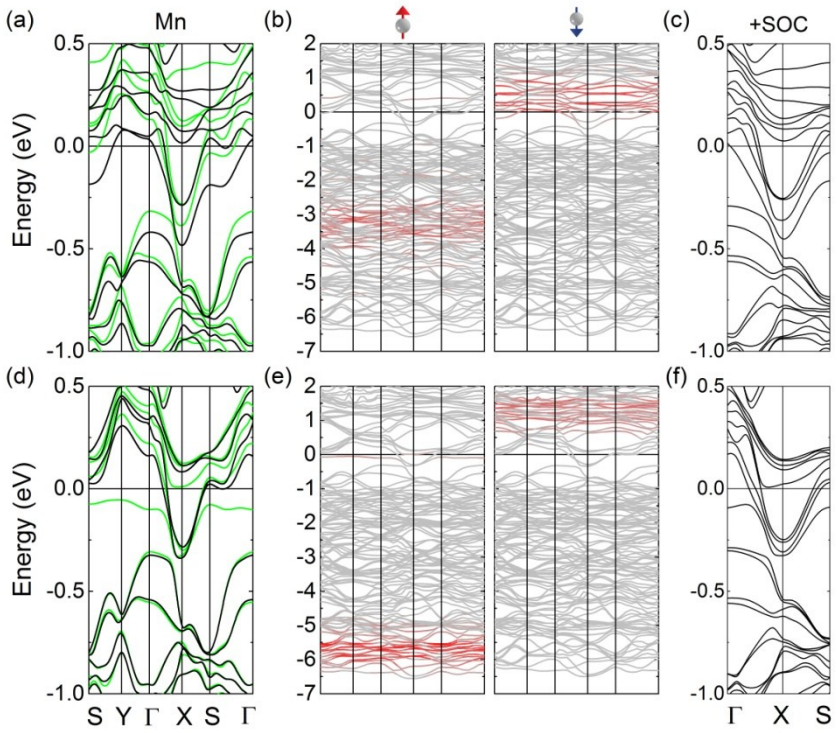

Fig. S1 Band structures of Mn-adsorbed monolayer without (a)-(c) and with (d)-(f) Coulomb interaction. Green and black are used to denote the spin up and spin down bands. Red represents the Mn component (red: 100%, light gray: 0%). (d) and (h) are bands with inclusion of SOC.

S3. Effects of SOC and Coulomb interaction on charge density of Mn-WTe_2

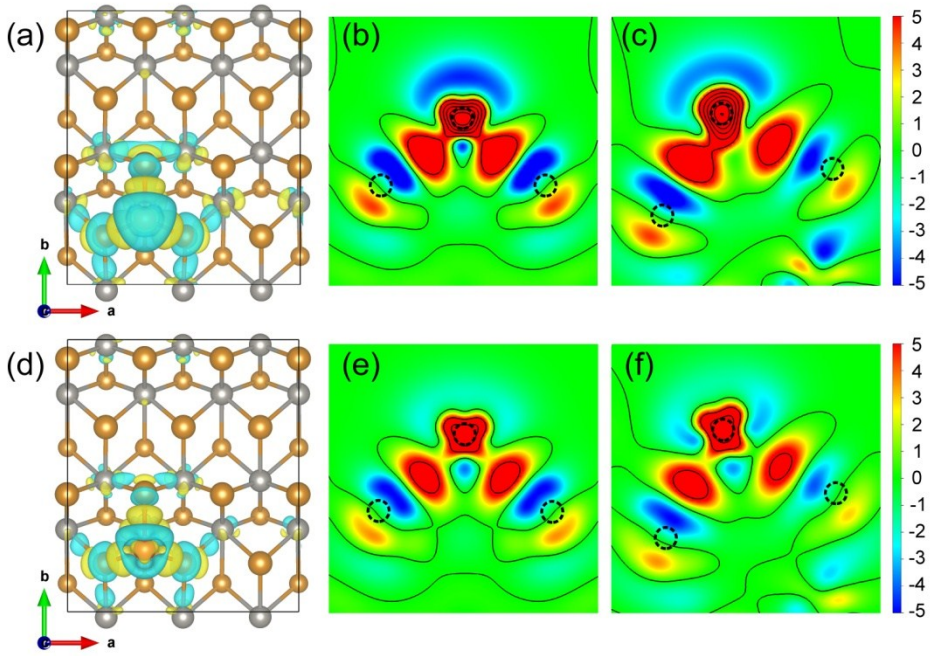

Fig. S2 Top views and 2D contours of the charge density difference for Mn-adsorbed WTe_2 without (a)-(c) and with (d)-(f) Coulomb interaction. Yellow (blue) region in (a) and (d) represents net charge gain (loss). In 2D contours, charge gain and loss are depicted as the color scale.

S4. Effects of SOC and Coulomb interaction on band structure of Ni-WTe_2

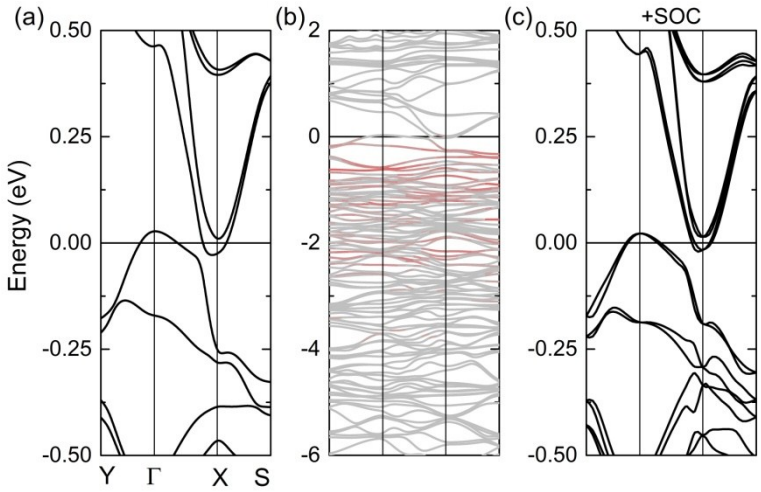

Fig. S3 Band structures of Ni-adsorbed WTe_2 with Coulomb interaction. Red represents the Ni component (red: 100%, light gray: 0%).

S5. Orbital-resolved density of states for pristine monolayer WTe_2

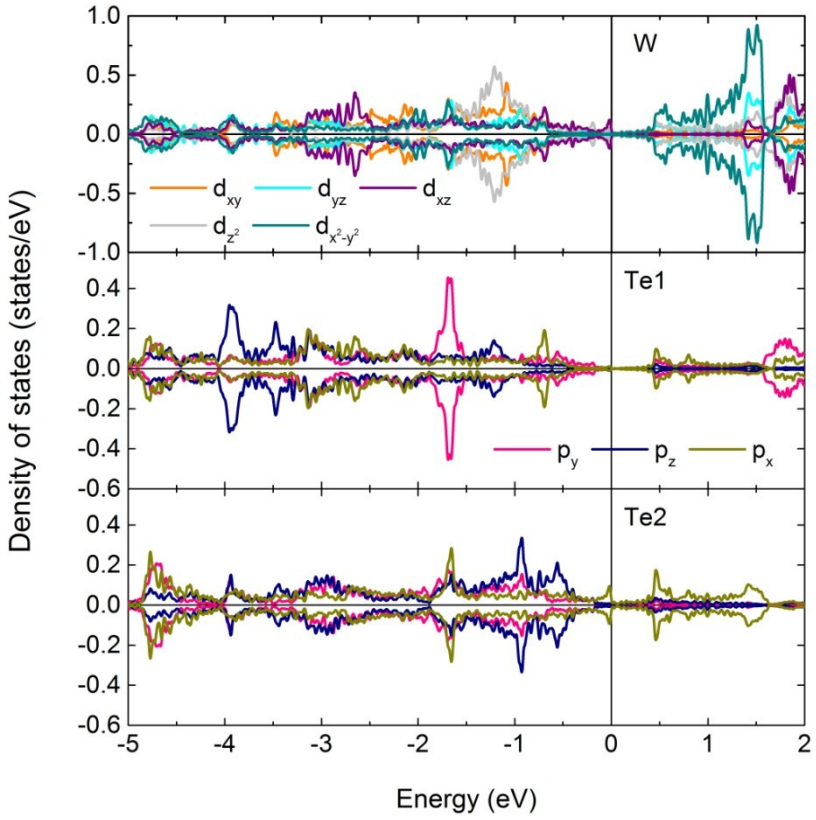

Fig. S4 Orbital-resolved density of states for pristine monolayer WTe_2.