Supplemental Material for

Effect of carrier concentration on the electronic structure and magnetothermal properties of two-dimensional VTe_2 monolayer with high Curie temperature

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Abstract: Two-dimensional (2D) magnetic transition metal dichalcogenides have unique electronic properties, ferromagnetism, and tunable properties in low-dimensional systems. In this paper, the structural, electronic, and magnetic properties of the VTe_2 monolayer under different carrier concentrations were investigated using first-principles calculations and Monte-Carlo (MC) simulations. It is found that, by introducing a suitable number of electrons, the VTe_2 monolayer can undergo a transition from a semiconductor to a half-metal state, with 100% spin polarization. The magnetocrystalline anisotropy energy is up to 1855.62 μeV in the *z*-axis direction, which is conducive to maintaining ferromagnetic order above room temperature.

Especially, the easy magnetic axis can undergo an in-plane to out-of-plane transition when doped with a small number of holes. In addition, doping can sensitively enhance or weaken the ferromagnetic exchange coupling strength. The magnetothermal results show that the Curie temperature of the VTe_2 monolayer is 547 K in the absence of size effect, and can be further increased to 574 K when holes doping reach 1.825×10^{13} cm^{-2} (0.02 holes per atom). Increasing magnetocrystalline anisotropy and magnetic field can also make the Curie temperature larger. Our results suggest the potential applications of VTe_2 in spintronics and provide a deeper understanding of the modulation mechanism.

Keywords: Electronic structure; Ferromagnetic; Magnetothermal; First-principles calculation; Monte Carlo simulations

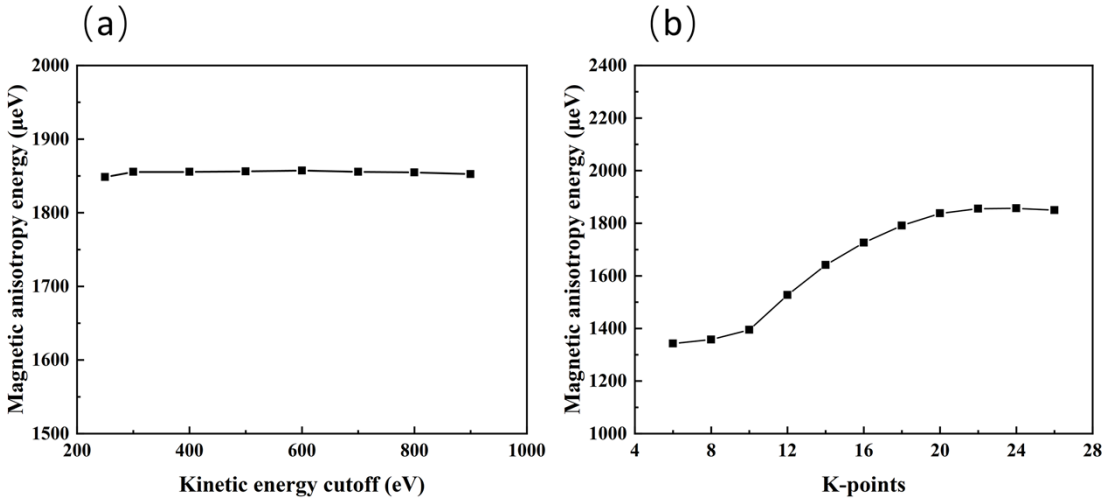

Fig. S1 (Color online) (a and b) The MAE of VTe_2 changes with the ENCUT and *k*-points.

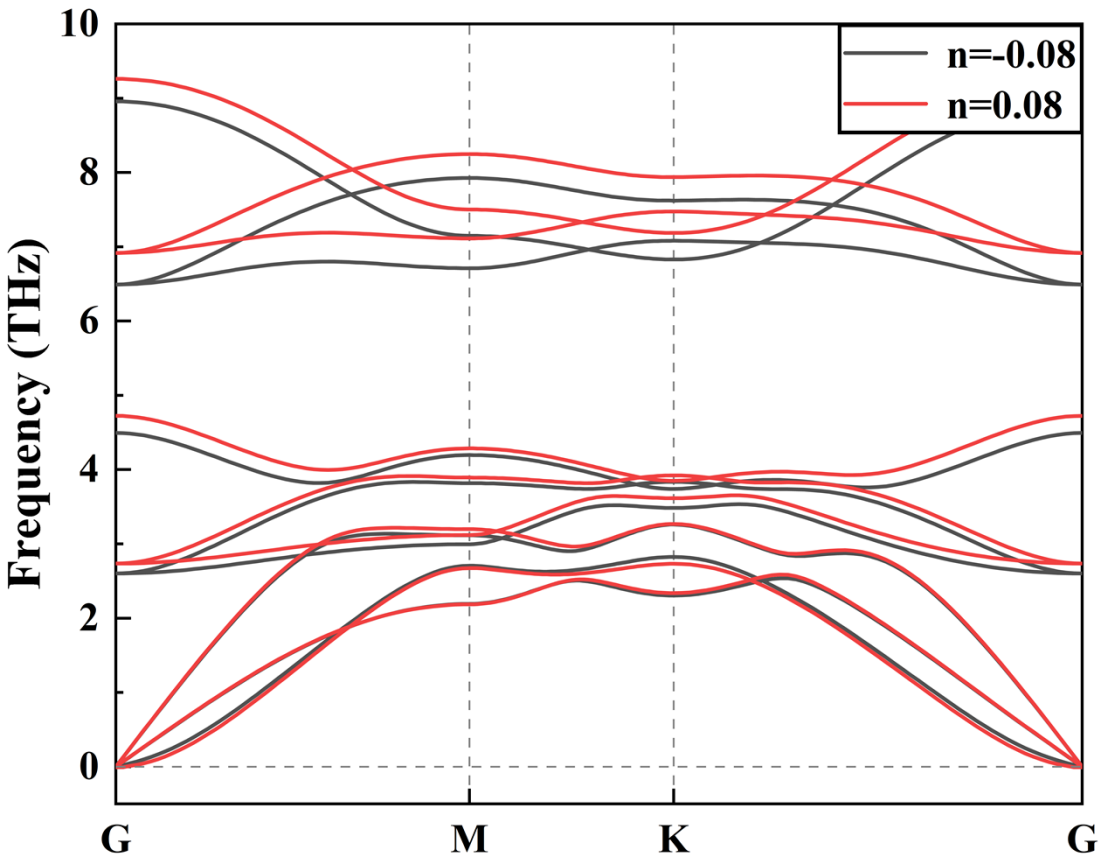

Fig. S2 (Color online) The phonon spectrum when the carrier concentration reaches 7.293×10^{13} cm^{-2} (0.08 electrons/holes per atom).

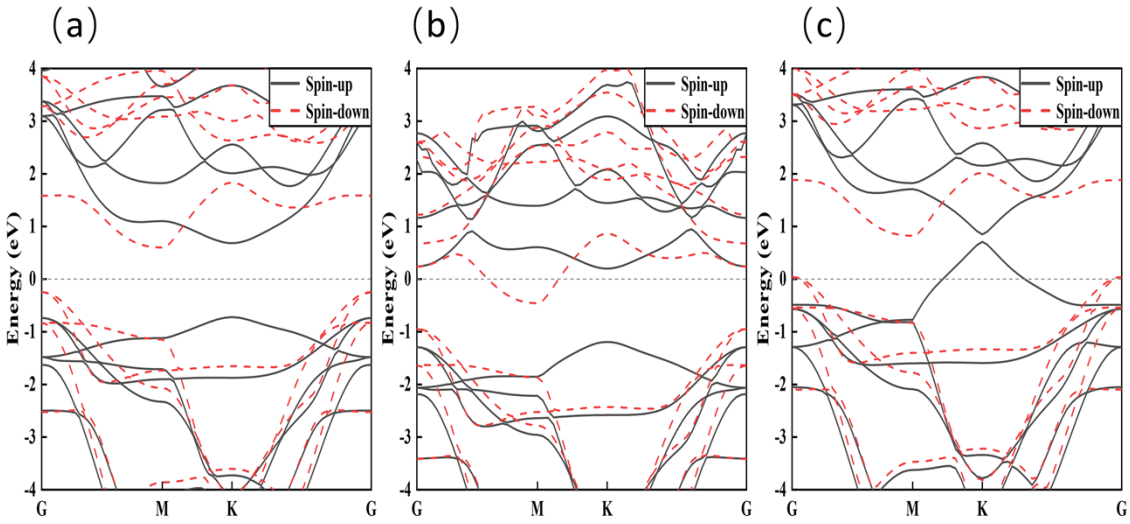

Fig. S3 (Color online) (a) Atom-resolved band structure of the VTe_2 monolayer at HSE06. (b and c) Atom-resolved band structure of the VTe_2 monolayer at the carrier concentrations reach 7.293×10^{13} cm^{-2} (0.08 electrons and holes per atom) under HSE06.

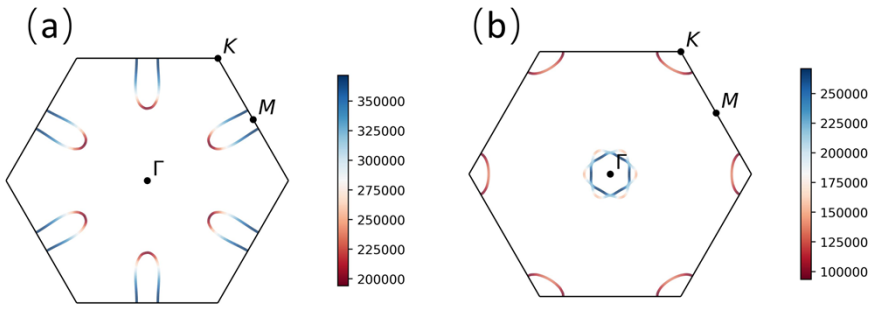

Fig. S4 (Color online) The Fermi surface when the carrier concentration reaches 3.647×10^{13} cm^{-2} (0.04 electrons/holes per atom).

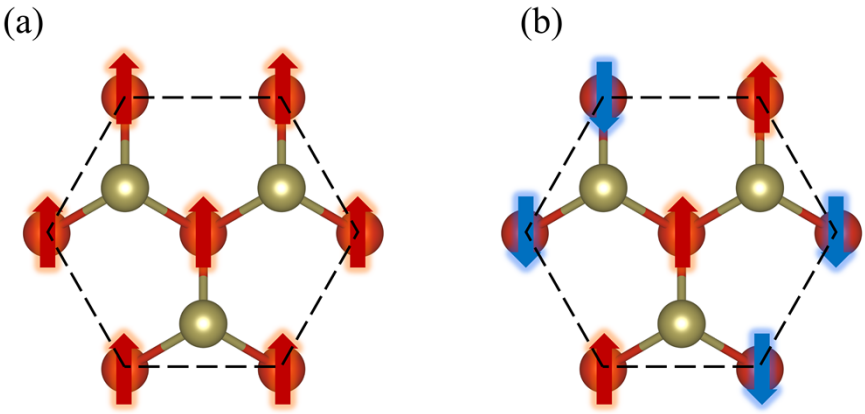

Fig. S5 (Color online) (a and b) The ferromagnetic configuration and antiferromagnetic configuration of VTe_2 monolayer. The ferromagnetic and antiferromagnetic states can be written $H_{FM} = -6 \times 9 \times \frac{1}{2} JS_0^2 - DS_0^2 - hS_0^2$ and $H_{AFM} = (-2 \times 9 \times \frac{1}{2} JS_0^2 + 4 \times 9 \times \frac{1}{2} JS_0^2) - DS_0^2 - hS_0^2$. The factor of 1/2 is included because the nearest V-V interaction is shared between two adjacent unit cells.

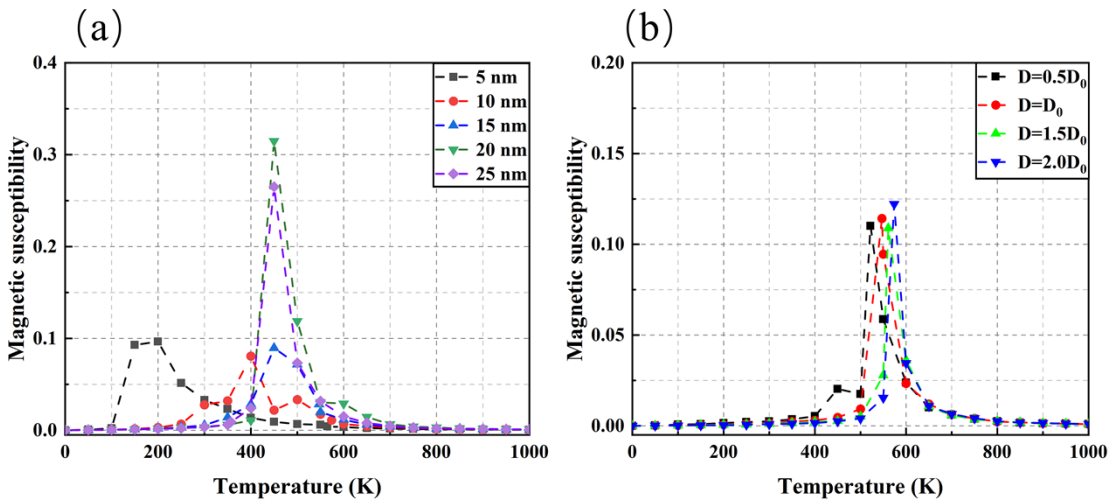

Fig. S6 (Color online) (a and b) Variation of magnetic susceptibility with temperature under different size effect and crystal field.