Supplementary Material for "Strain and U driven phase transitions in monolayer intrinsic ferrovalley $NbIn_2As_2Se_2$"

Jiajun Zhu,[1] Heyun Zhao,[1] and Wanbiao Hu[1,2,3,4,*]

[1] *Yunnan Key Laboratory of Electromagnetic Materials and Devices, National Center for International Research on Photoelectric and Energy Materials, School of Materials and Energy, Yunnan University, Kunming, 650091, P. R. China*

[2] *Electron Microscopy Center, Yunnan University, Kunming 650091, P. R. China*

[3] *Southwest United Graduate School, Kunming 650092, P. R. China*

[4] *School of Engineering, Yunnan University, Kunming 650091, P. R. China*

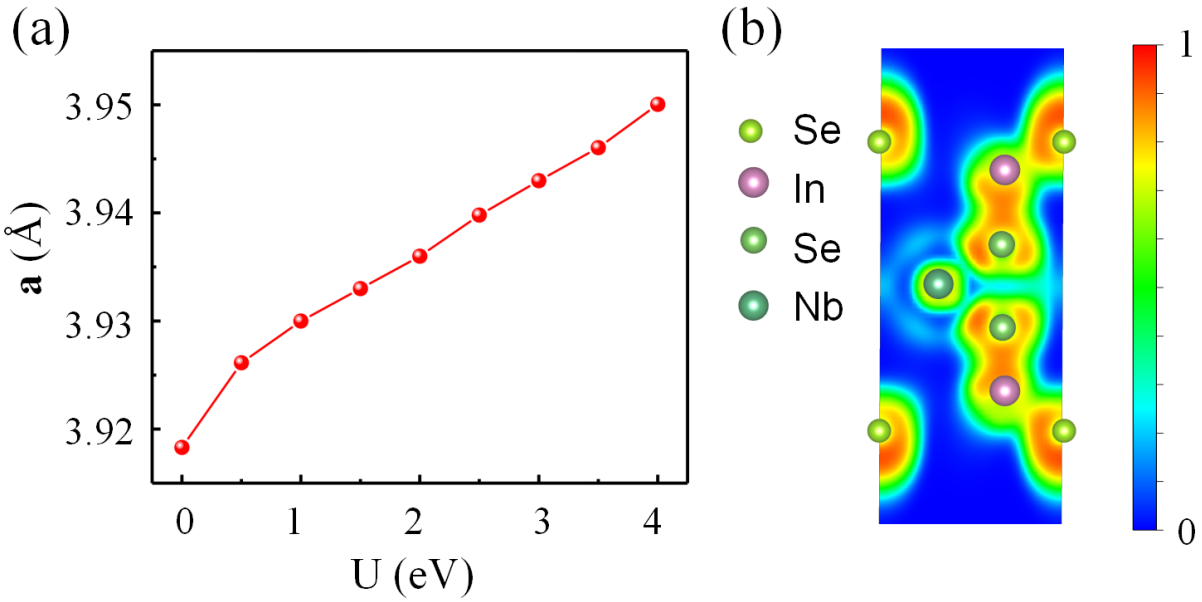


FIG. S1: (a) the lattice constants **a** as a function of U values. (b) Electron localization function in the (110) plane of monolayer $NbIn_2As_2Se_2$.

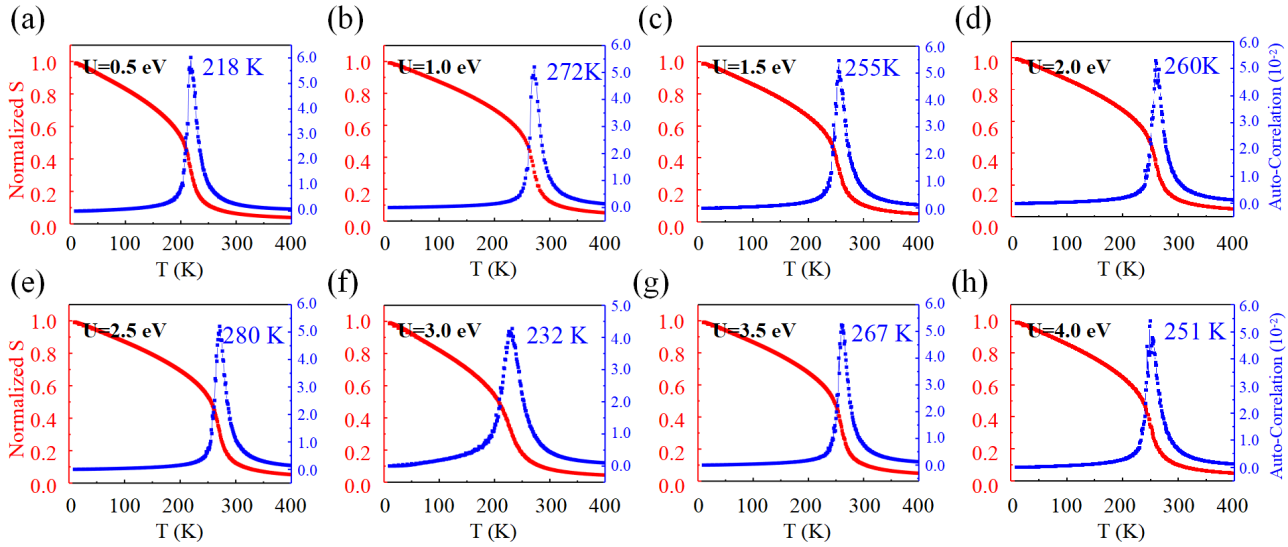


FIG. S2: The normalized magnetic moment (S) and auto-correlation as a function of temperature with U=0.5 eV (a), 1.0 eV (b), 1.5 eV(c), 2.0 eV(d), 2.5 eV(e), 3.0 eV(f), 3.5 eV(g) and 4.0 eV(h).

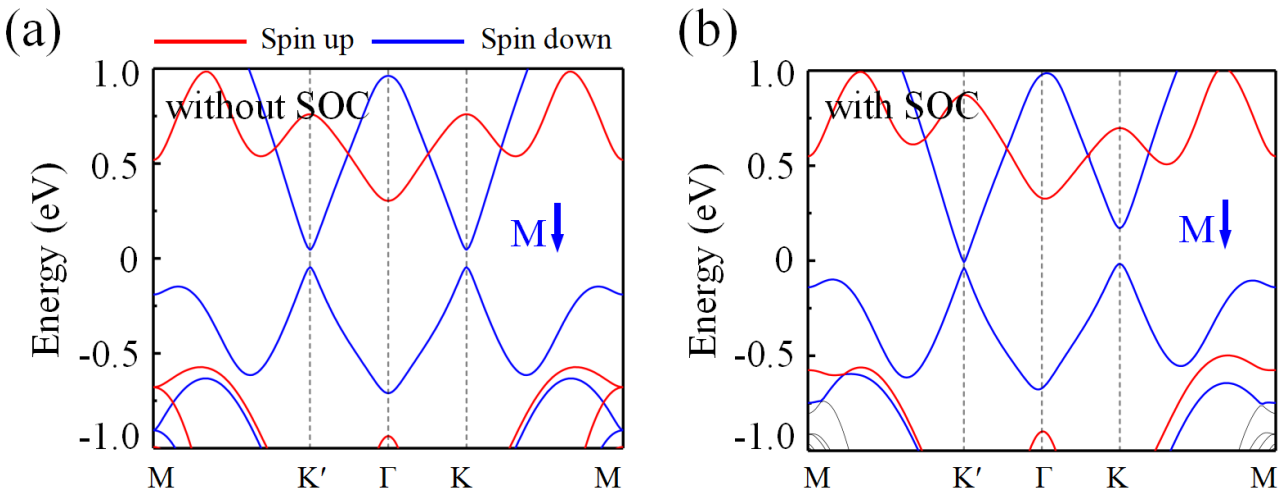


FIG. S3: The spin-polarized band structure of monolayer $NbIn_2As_2Se_2$ without(a)/with(b) SOC effect.

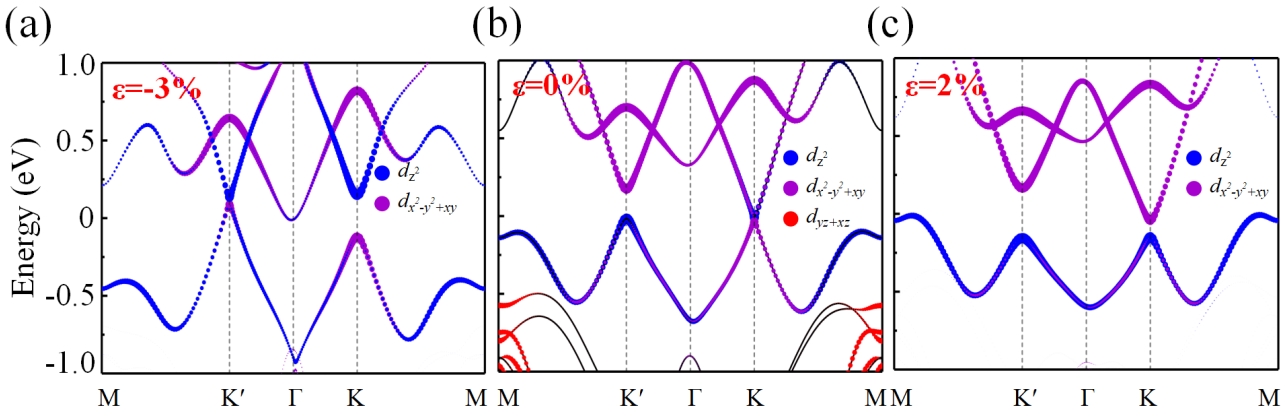


FIG. S4: The d orbit projection of monolayer $NbIn_2As_2Se_2$ at the ε = -3%(a), 0%(b) and 2%(c).

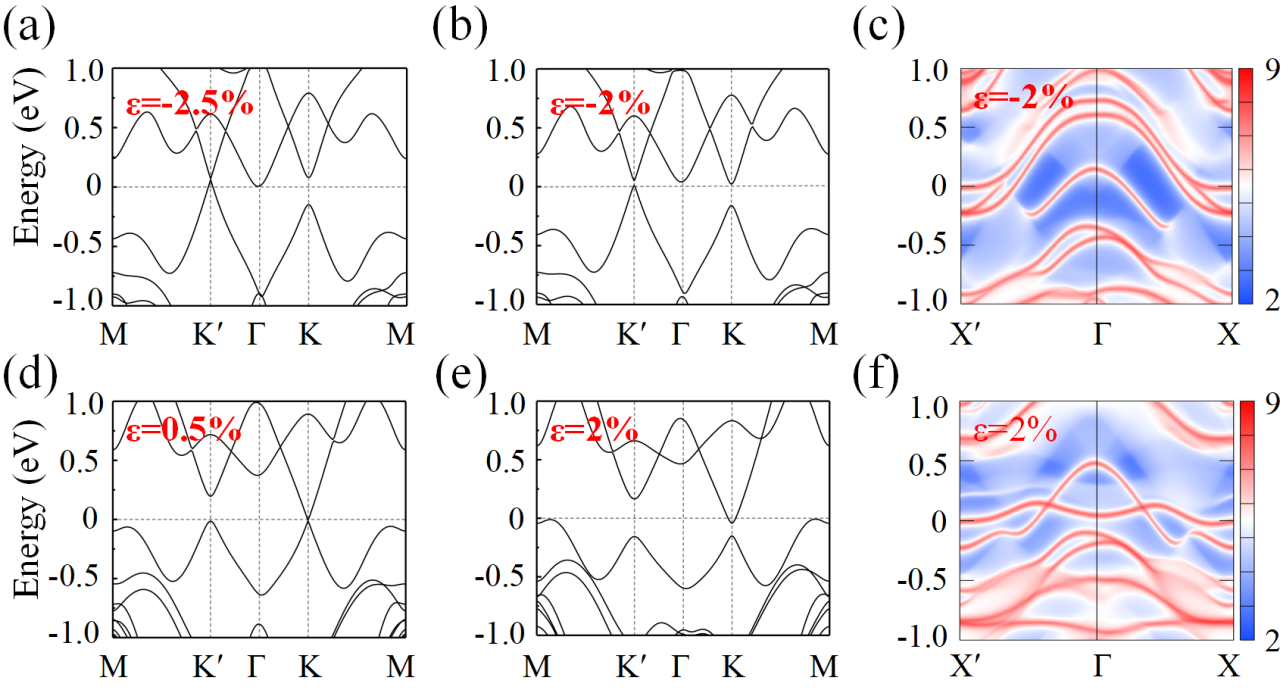


FIG. S5: The calculated band structure and the edges states of monolayer $NbIn_2As_2Se_2$ under U = 3 eV with different tensile strains.

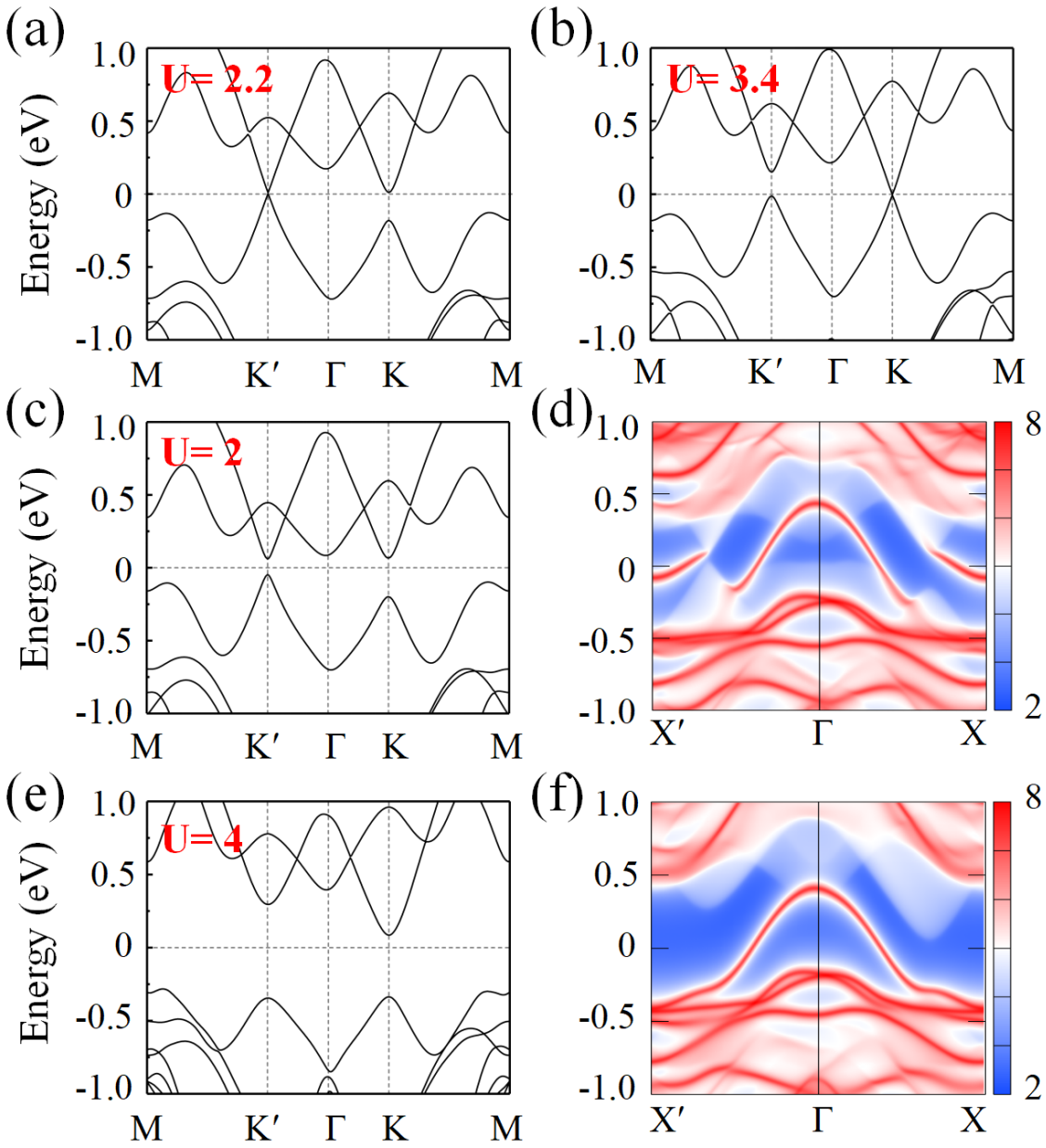
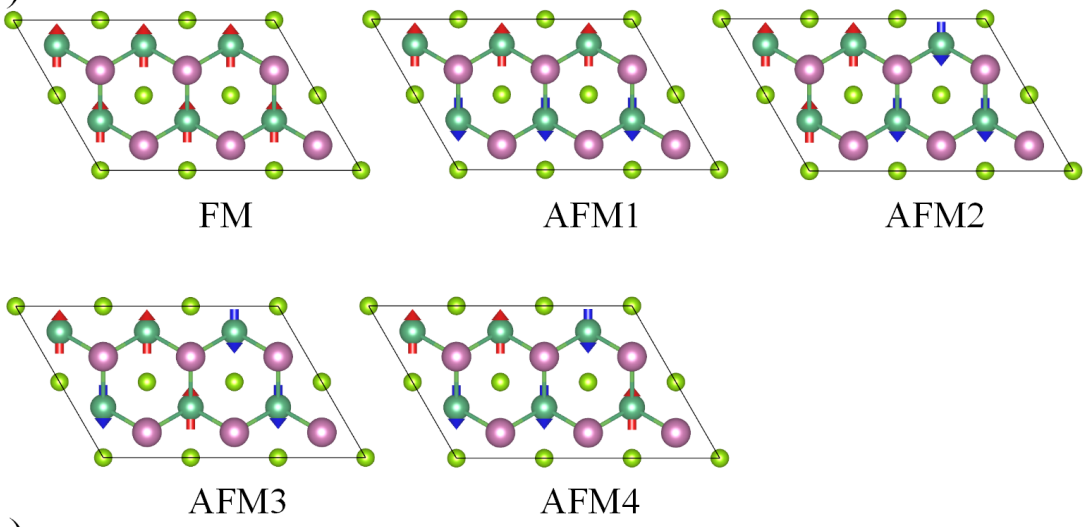


FIG. S6: The calculated band structure and the edges states of monolayer $NbIn_2As_2Se_2$ under varioous U values.

(a)



(b)

		FM	AFM1	AFM2	AFM3	AFM4
ΔE (meV)	PBE+U	0	231.64	179.71	179.72	233.27
	HSE06	0	697.34	294.55	245.32	828.84

FIG. S7: (a) The magnetic configuration diagram under the 3×2×1 supercell. (b) The total energies between different magnetic configurations and we define the total energy of ferromagnetism as zero. Calculate the energy $\Delta E = E_{FM} - E_{AFM}$ under U = 3 eV and the HSE06 method, where ΔE represents the energy difference, and E_{FM} and E_{AFM} denote the total energies of ferromagnetic and antiferromagnetic states, respectively.

We reconstructed the unit cell of monolayer $NbIn_2As_2Se_2$ into a 3×2×1 supercell to consider the magnetic configurations. After analysis, we found that there are four types of antiferromagnetic configurations, labeled as AFM1 to AFM4, which are presented in Fig. S7. To explore the magnetic ground state, we compared the energies between different magnetic configurations,as shown in Fig.S7(b). The calculation results indicate that the ferromagnetic state remains the ground state under U = 3 eV and the HSE06 method.

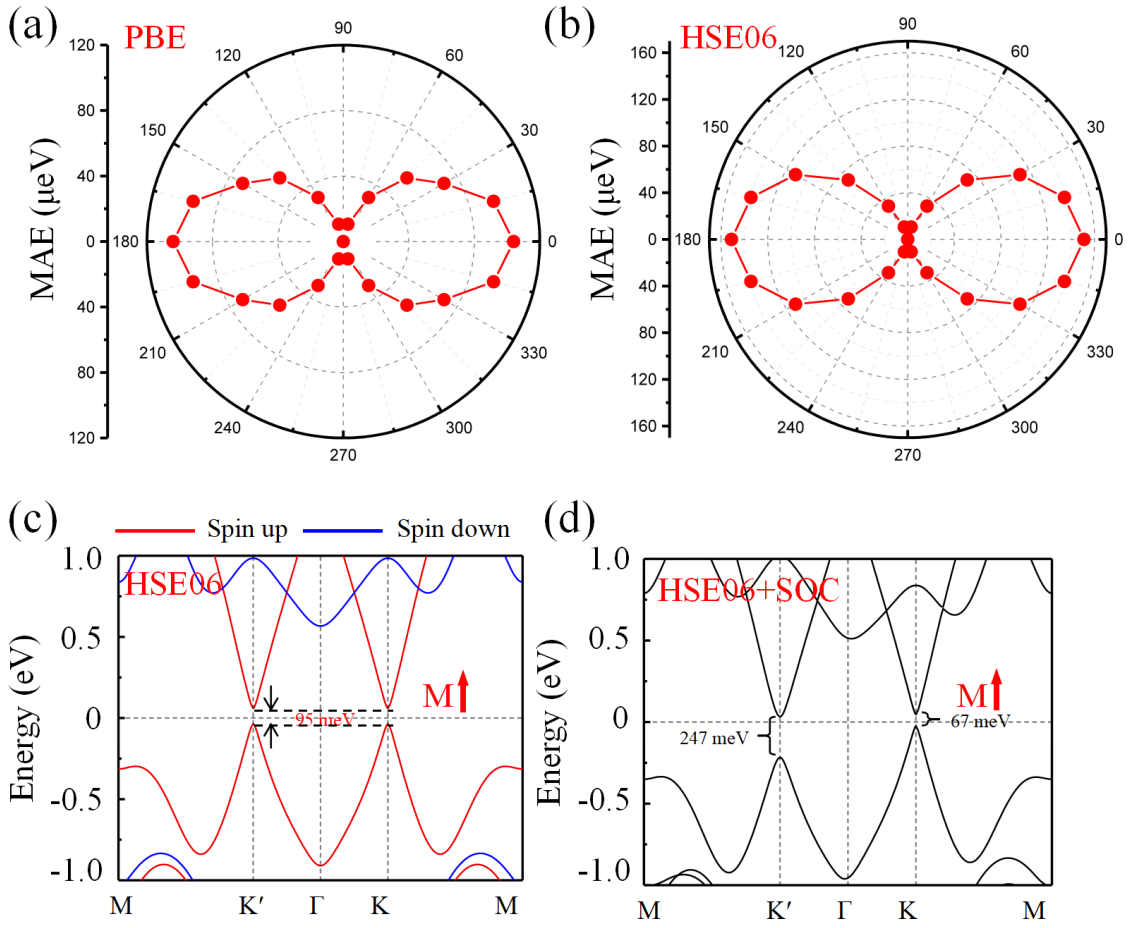


FIG. S8: The polar angel of magnetization dependence of the energy variation in *xoz* plane at U = 3 eV (a) and HSE06 (b), where we define the total energy of the magnetic direction along the **c**-axis as zero. The band structure of monolayer $NbIn_2As_2Se_2$ using the HSE06 method without SOC (c) and with SOC (d).

In Fig.S7, we have confirmed that the magnetic ground state of monolayer $NbIn_2As_2Se_2$ is ferromagnetic using both the PBE+U and HSE06 methods. Here, we continue to discuss MAE and have calculated the polar angel of magnetization dependence of the energy variation in *xoz* plane, where we define the total energy of the magnetic direction along the **c**-axis as zero. As shown in Fig. S9 , At U = 3 eV, the energy of the magnetic direction along the **c**-axis is lower than that of the magnetic direction within the plane by 104 eV, which is consistent with the main manuscript. Furthermore, after considering the more accurate HSE06 method, the energy of the monolayer $NbIn_2As_2Se_2$ with the magnetic direction along the c-axis remains lower. This result is consistent with the conclusion at U = 3 eV; however,

the energy of the magnetic direction out of the plane is lower than that in the plane by 151 eV.

We calculated the band structure of monolayer $NbIn_2As_2Se_2$ using the HSE06 method, as shown in Fig. S8. Without considering the spin-orbit coupling effect, the band structure is similar to the result at U = 3 eV, with both the CBM and VBM located at the K/K′ points. The band gap value is 95 meV, which is smaller than the 104 meV observed at U = 3 eV. The out-of-plane magnetism and spin-orbit coupling can break the valley degeneracy at the K/K′ points, as shown in Fig. S8(b). The valleys at the K/K′ points are no longer equal, with the band gap value at the K point being 67 meV, which is less than the band gap value at the K′ point of 247 meV. The trend of the band gap values is consistent with the main manuscript, showing that the band gap value at the K point is smaller than that at the K′ point.

The above analysis indicates that the results with U = 3 eV are similar to those obtained using HSE06, confirming that the conclusion for U = 3 eV is reliable.

* huwanbiao@ynu.edu.cn

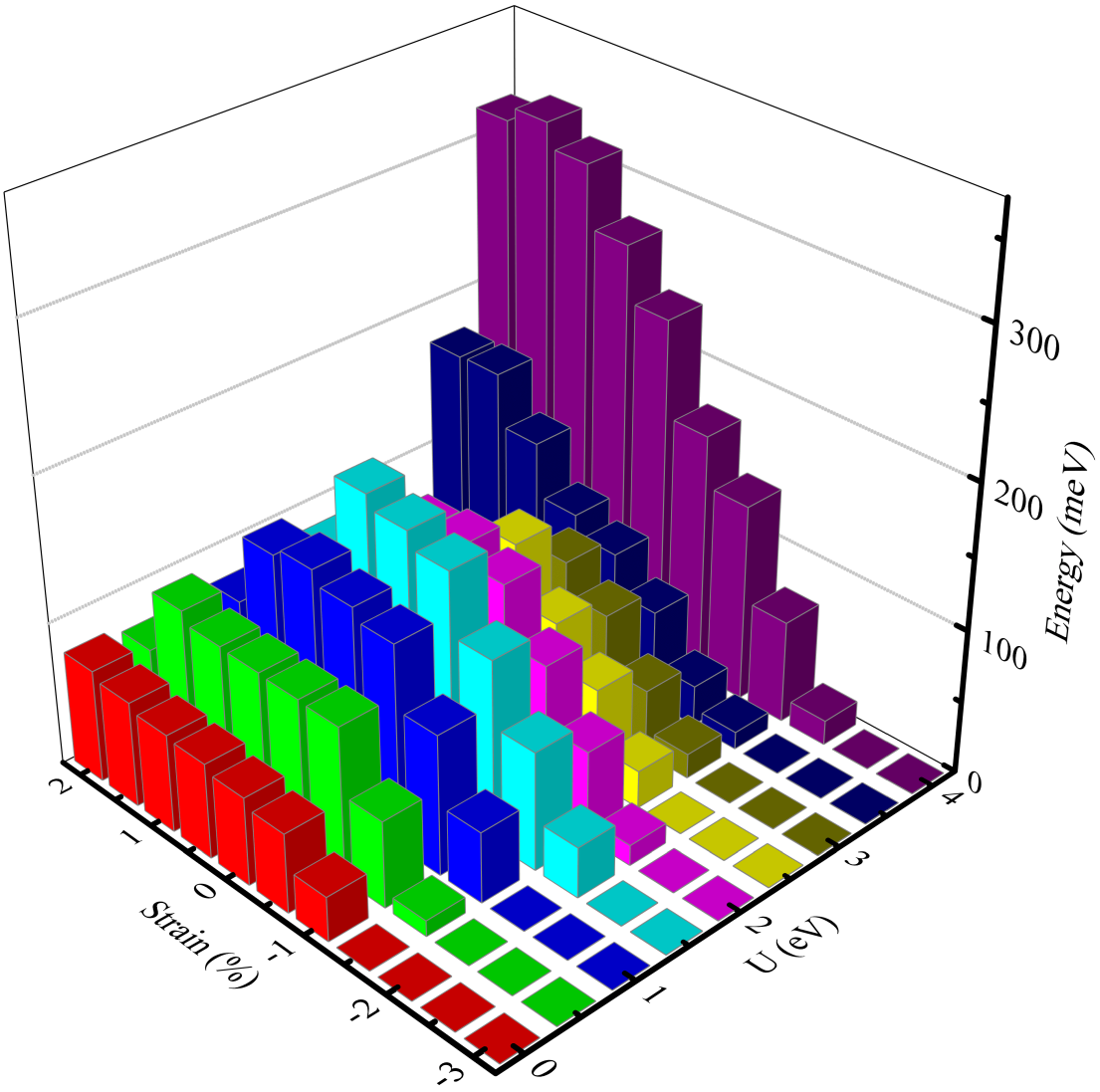


FIG. S9: The band gap diagram dependent on U values and biaxial strain.