Electronic and Optical Properties of TiO_2 Solid-Solution Nanosheets for Bandgap Engineering: A Hybrid Functional Study

Yanyu Liu[1,2], Wei Zhou[1,2], and Naoto Umezawa[2]*

[1]Department of Applied Physics, Tianjin Key Laboratory of Low Dimensional Materials Physics and Preparing Technology, School of Science, Tianjin University, Tianjin 300072, P.R. China

[2]International Center for Materials Nanoarchitectonics (MANA), National Institute for Materials Science, 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan

*To whom correspondence should be addressed.

E-mail: weizhou@tju.edu.cn. Phone: +86 (0)22 27408599 Fax: +86 (0)22 27406852.

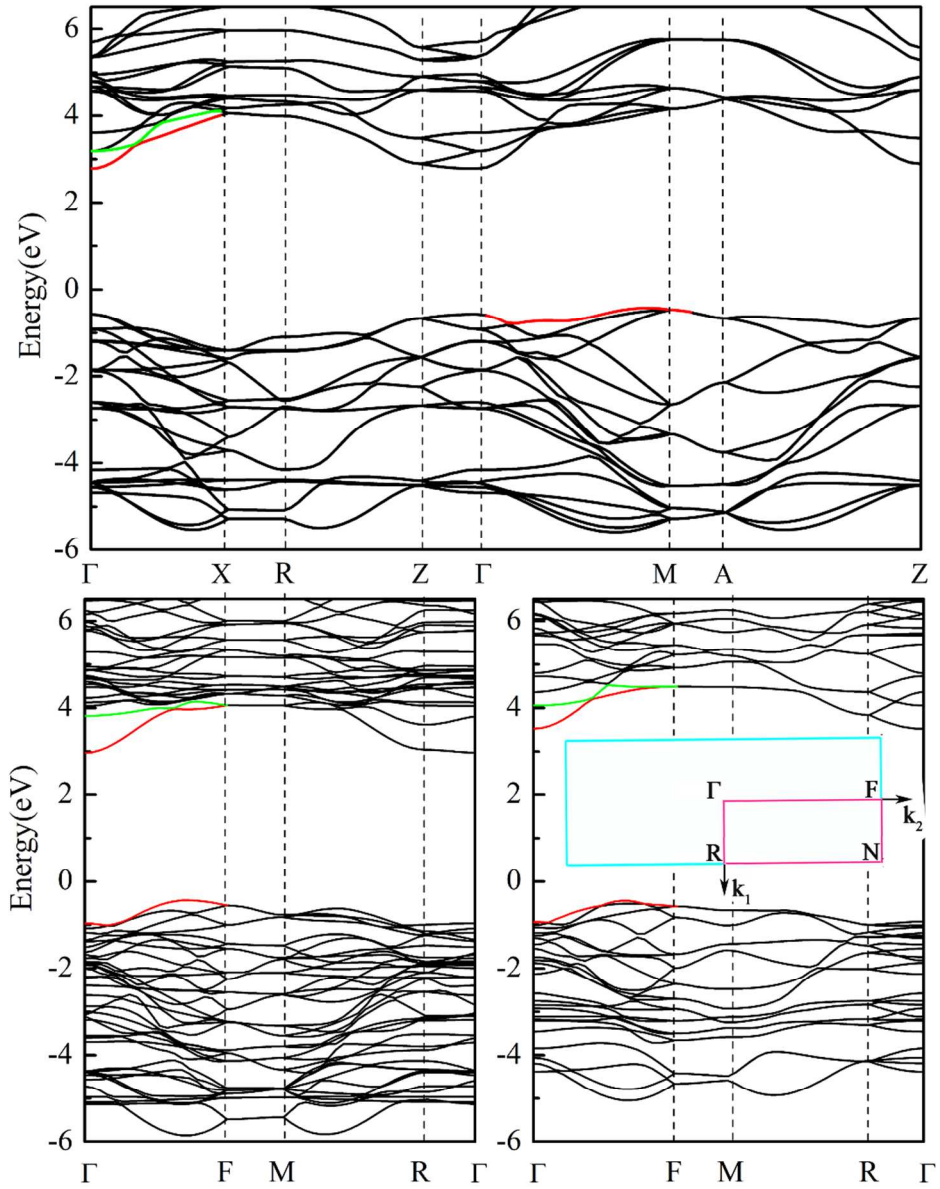

Figure S1. Band structures of the (a) bulk TiO_2, (b) TiO_2 nanosheet with three atomic layers from HSE06. Inset: First Brillouin zone of TiO_2 nanosheets, (c) TiO_2 nanosheets with two atomic layers (the bulk TiO_2, the coordinates of the symmetric k-points: Γ (0 0 0), X(0 0.5 0), R(0 0.5 0.5), Z(0 0 0.5), M(0.5 0.5 0), A(0.5 0.5 0.5) for the TiO_2 nanosheets: Γ (0 0 0), F(0 0.5 0), N(0.5 0.5 0), R(0.5 0 0)).

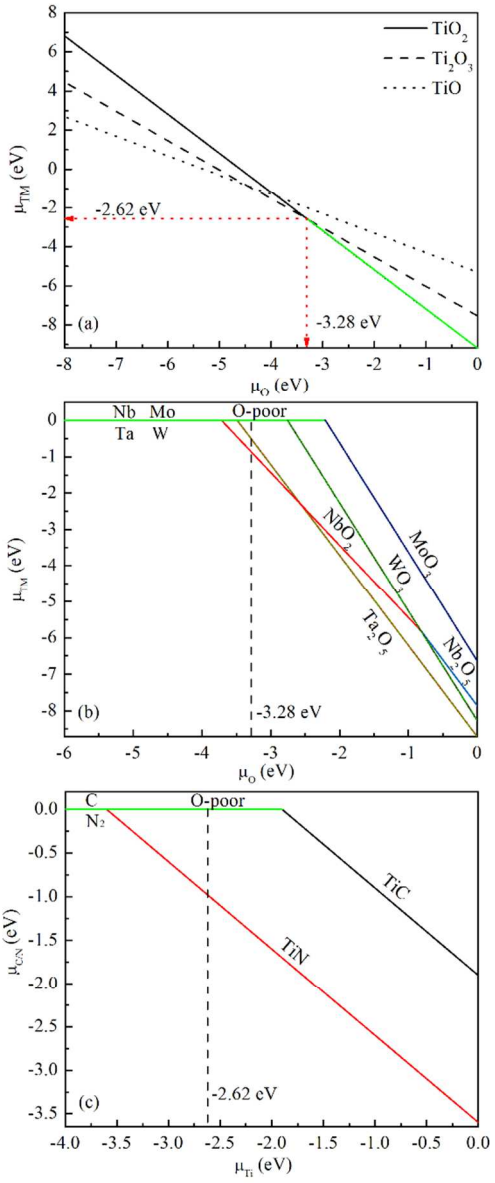

Figure S2. (a) Visualization of the three conditions described by eqn (2)–(4). These conditions are simultaneously satisfied in -3.28 eV ≥ μ_{O} ≥0 eV and -9.18 eV ≥ μ_{Ti} ≥ -2.62 eV as indicated by a green bold line. (b) the relationship between the transition metal chemical potentials μ_{TM} and the O chemical potential μ_{O}, as derived from the equilibrium growth of the host: $\mu_{\mathrm{TM}} = \left[H\left(\mathrm{TM}_x\mathrm{O}_y\right) - y\mu_{\mathrm{O}} \right] / x$, μ_{O} at -3.28 eV and 0 eV represent the O-poor and O-rich condition, respectively. (c) the relationship between the C and N chemical potentials $\mu_{\mathrm{C/N}}$ and the Ti chemical potential μ_{Ti}, as derived from the equilibrium growth of the host: $\mu_{\mathrm{C}} = H\left(\mathrm{TiC}\right) - \mu_{\mathrm{Ti}}$ and $\mu_{\mathrm{N}} = H\left(\mathrm{TiN}\right) - \mu_{\mathrm{Ti}}$, μ_{Ti} at -2.62 eV and -9.18 eV represent the O-poor and O-rich condition, respectively.

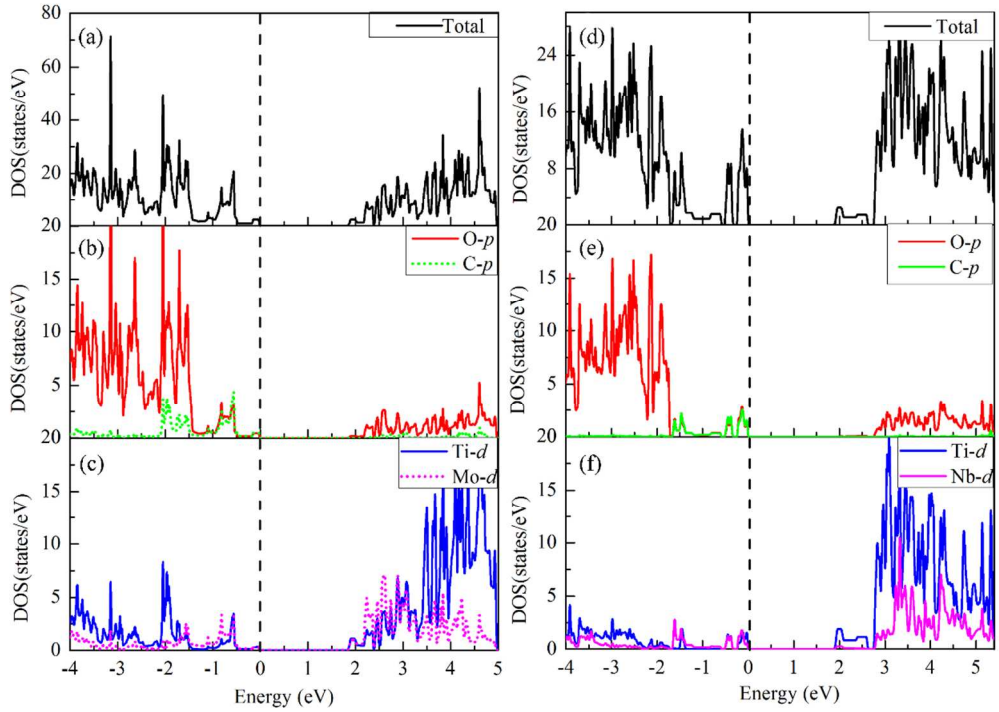

Figure S3. Total and projected DOS for (a)-(c) $(TiO_2)_{2/3}(MoOC)_{1/3}$ and (d)-(f) $(TiO_2)_{2/3}(Nb_2O_3C)_{1/3}$ solid solutions from HSE06.

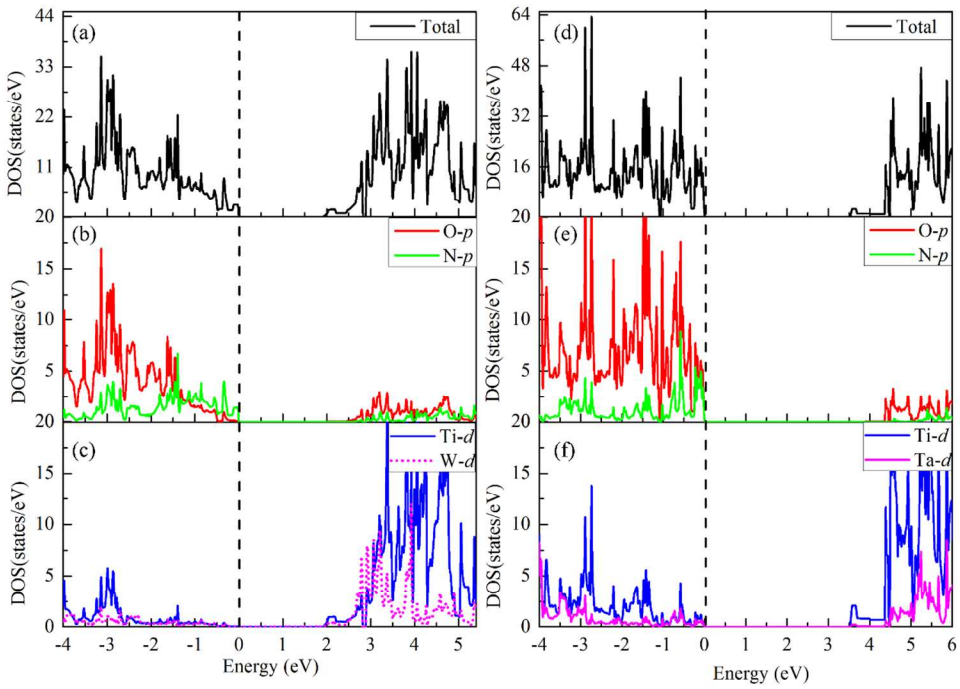

Figure S4. Total and projected DOS for (a)-(c) $(TiO_2)_{2/3}(WN_2)_{1/3}$ and (d)-(f) $(TiO_2)_{2/3}(TaON)_{1/3}$ solid solutions from HSE06.

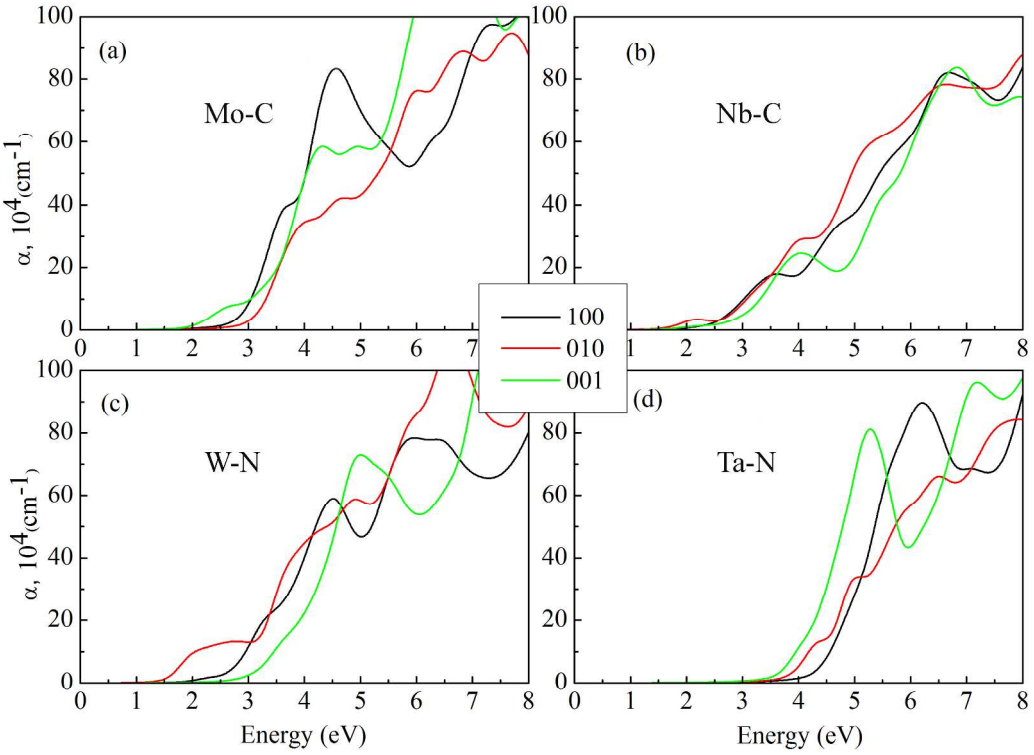

Figure S5. Optical properties of (a) $(TiO_2)_{2/3}(MoOC)_{1/3}$, (b) $(TiO_2)_{2/3}(Nb_2O_3C)_{1/3}$, (c) $(TiO_2)_{2/3}(WN_2)_{1/3}$ and (d) $(TiO_2)_{2/3}(TaON)_{1/3}$ nanosheets from HSE06.