Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supplementary Information:

Auxetic, Flexible, and Strain-Tunable Two-Dimensional th-AlN with Anisotropic High Carrier Mobility for Photocatalytic Visible Light Water Splitting

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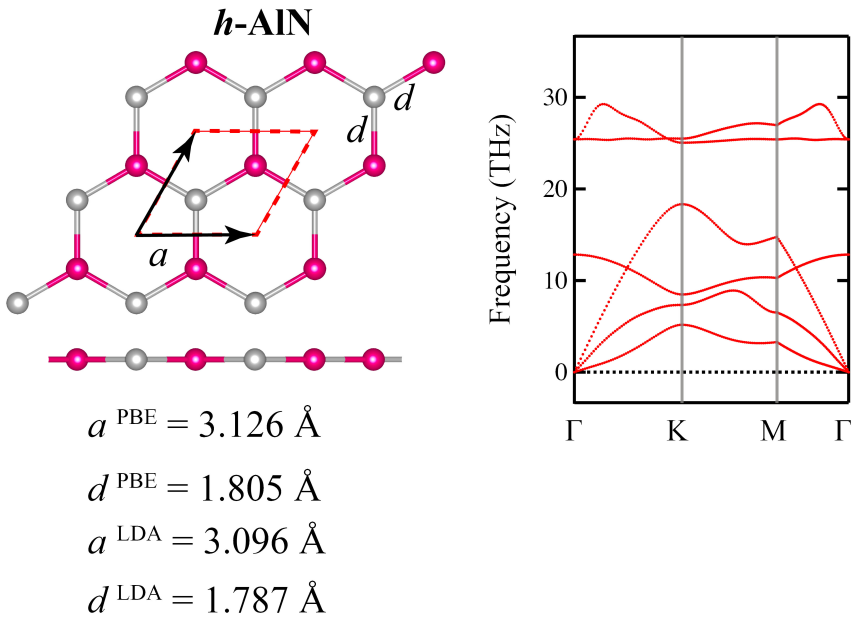

Fig. S1: Optimized atomic configurations (left panel) and phonon band structure (right panel) of hexagonal AlN (h-AlN). The value of lattice constant a and bond length d obtained from the LDA and PBE functional level is depicted in the figure.

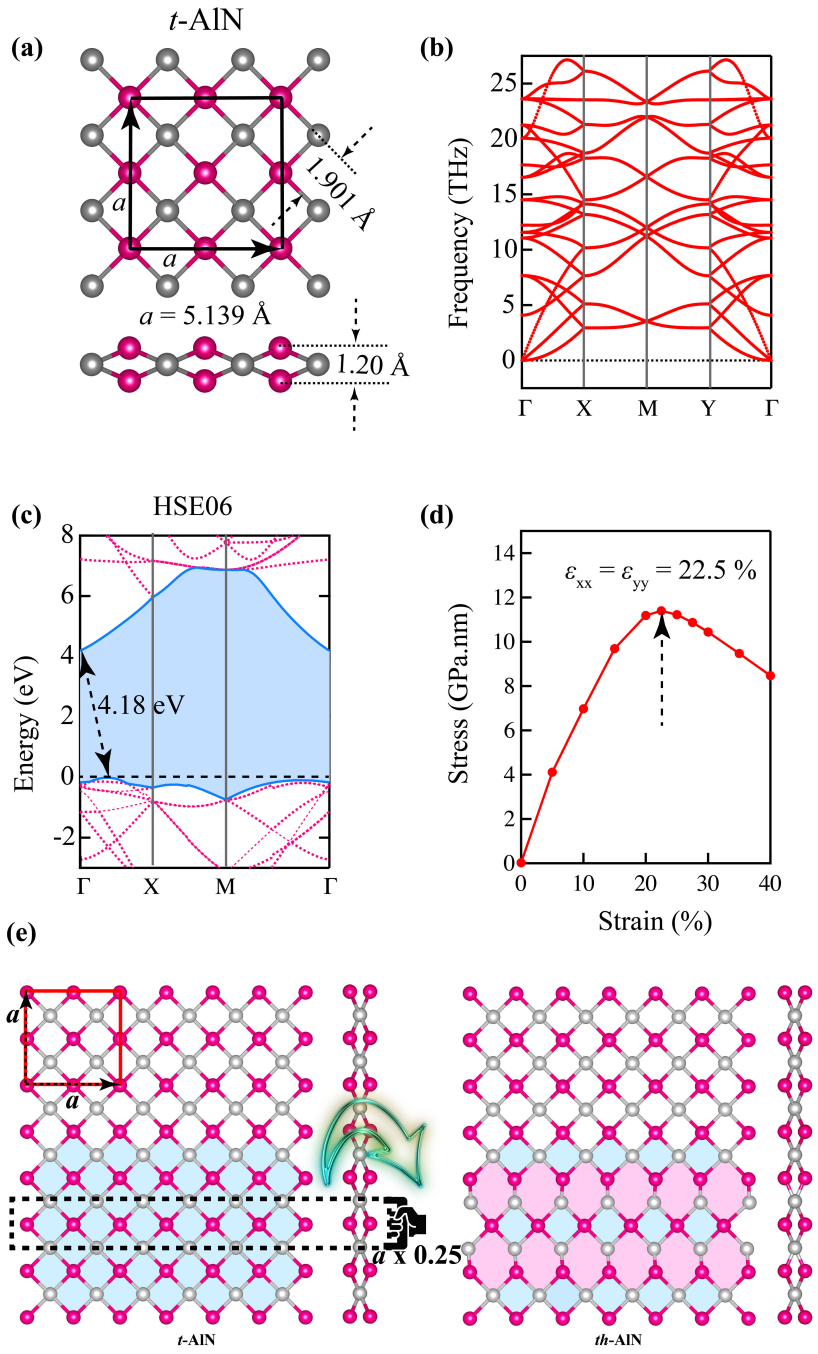

Fig. S2: (a) Optimized atomic structure, (b) phonon band structure, (c) electronic band structure obtained from the HSE06 hybrid functional level, and (d) stress-strain relation for tetragonal aluminum nitride (named as *t*-AlN). From the electronic band structure, the Fermi level energy is set to zero (depicted by the black dashed line). The electronic band gap energy of *th*-AlN is found to be 4.18 eV. Silver and pink spheres represent Al and N atoms, respectively. According to the phonon band dispersions, all the phonon modes are positive frequencies, thus, confirming its dynamic stability. (e) Deriving *th*-AlN from 2D *t*-AlN.

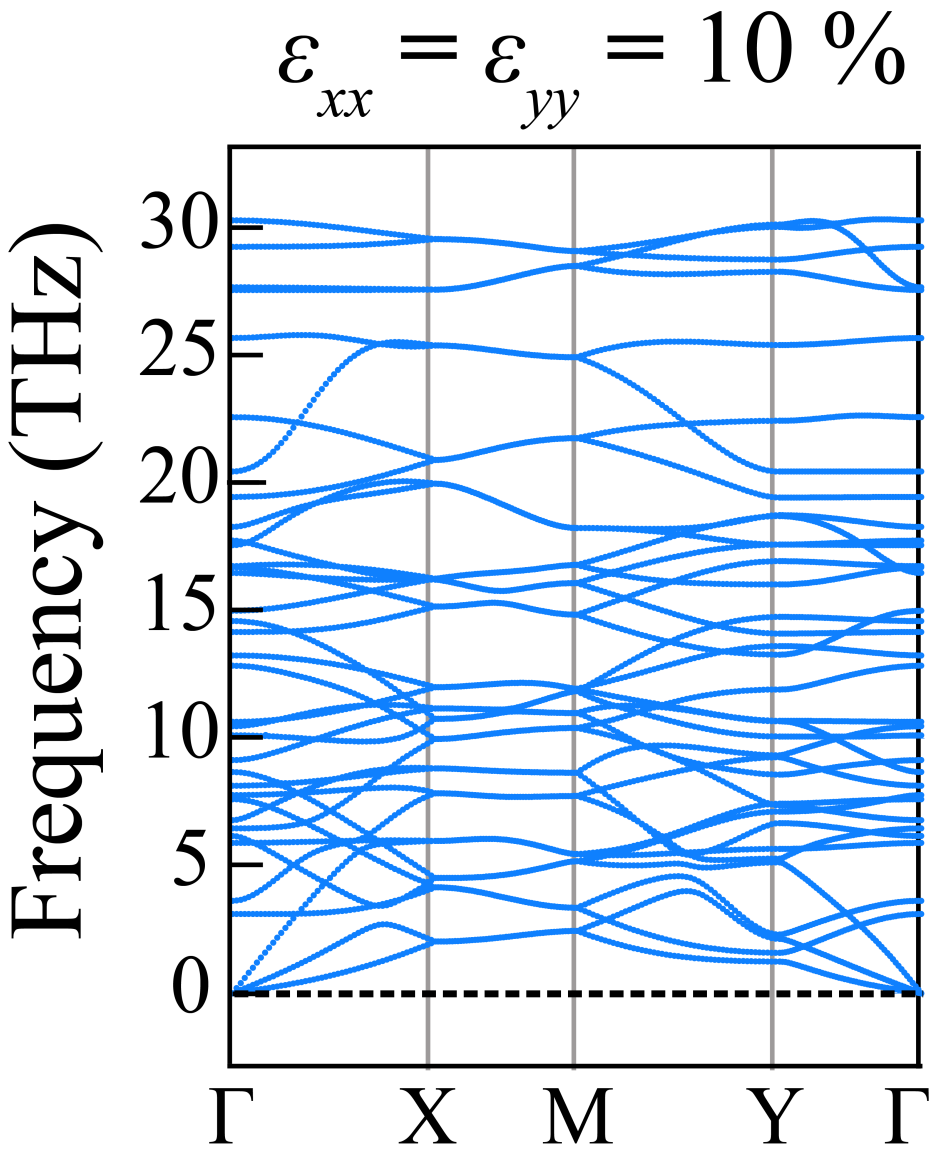

Fig. S3: Phonon dispersion curves of *th*-AlN when subjected to 10% equi-biaxial tensile strain.

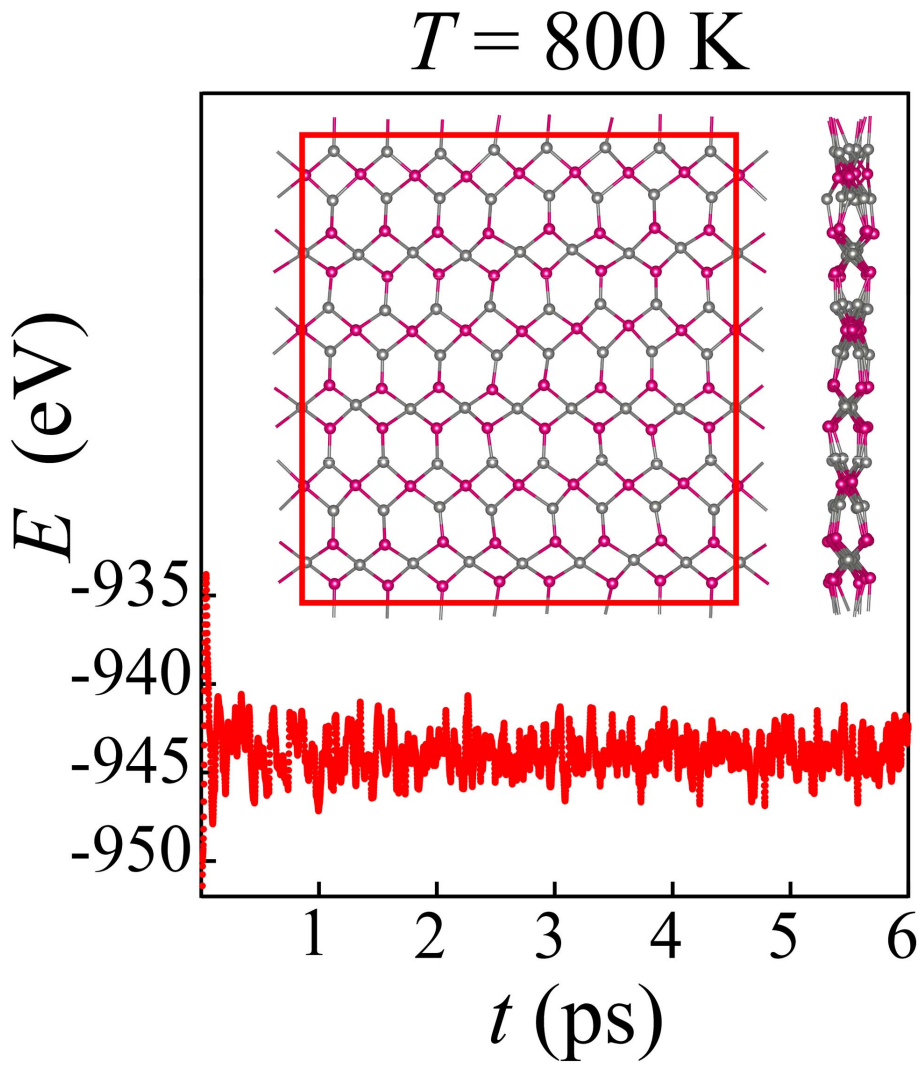

Fig. S4: Variation of total potential energy of *th*-AlN during the AIMD simulations at $T = 800$ K. The inset is snapshot of the atomic structures at the end of simulation. The silver and pink balls represent Al and N atoms, respectively.

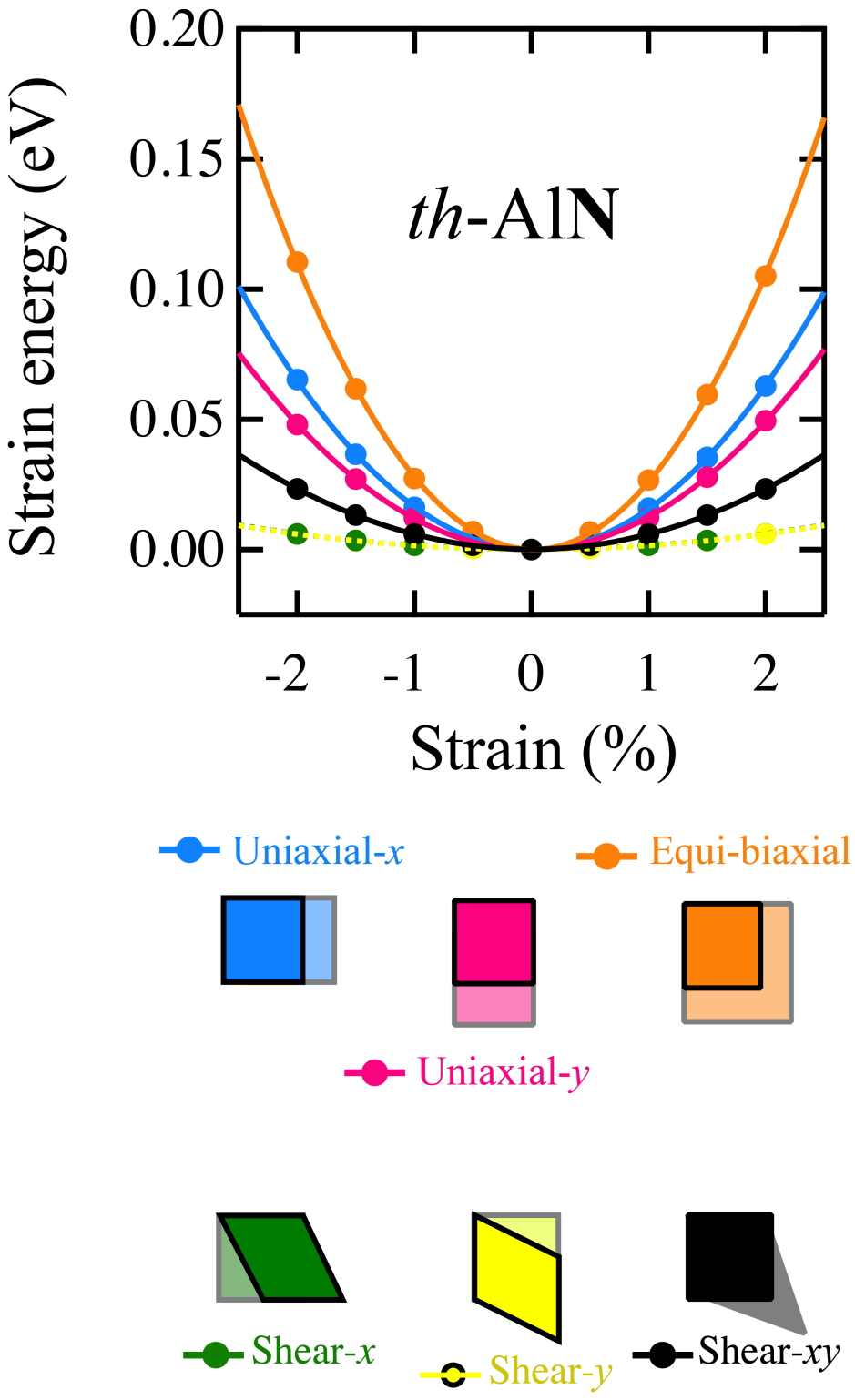

Fig. S5: Calculated strain energy of *th*-AlN with respect to various applied strains: uniaxial strain along the x- and y-directions, equi-biaxial strain, and shear strains.

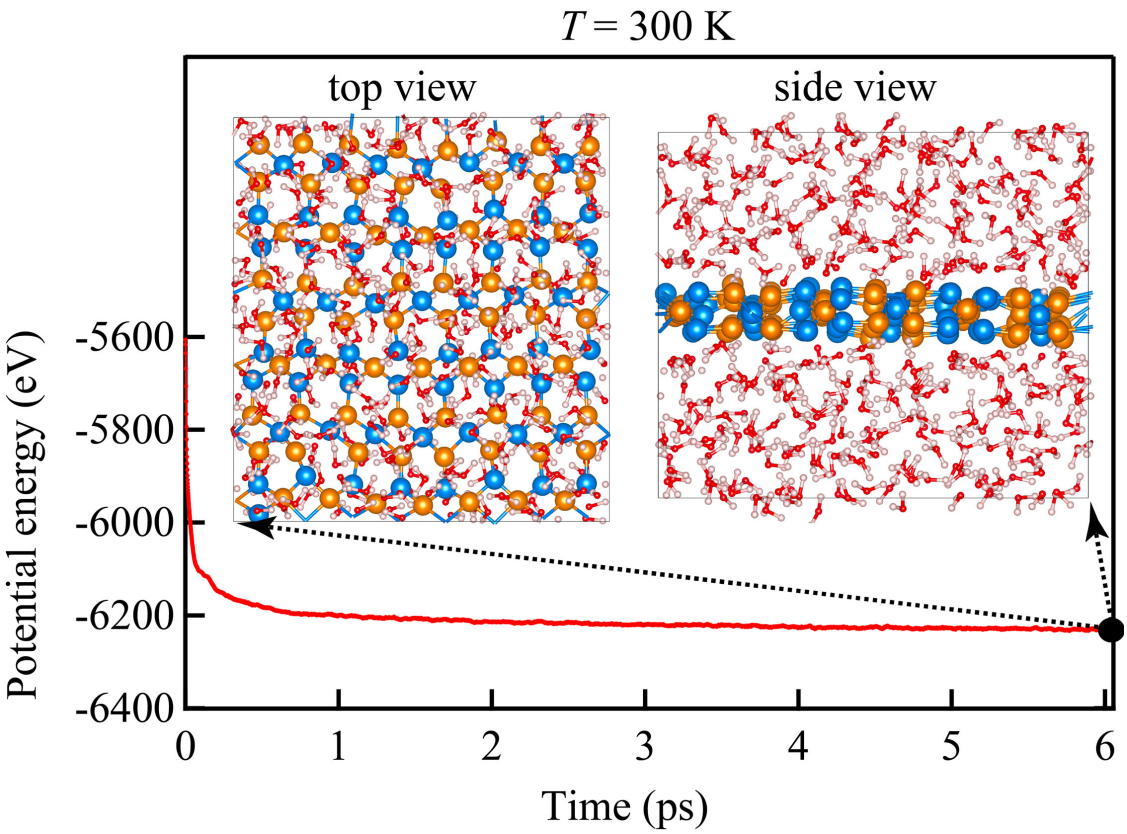

Fig. S6: Variation of total potential energy of *th*-AlN in the liquid water during the AIMD simulations at $T = 300$ K. The inset is snapshot of the atomic structures at the end of simulation. The blue, orange, red, and white balls represent Al, N, O, and H atoms, respectively.

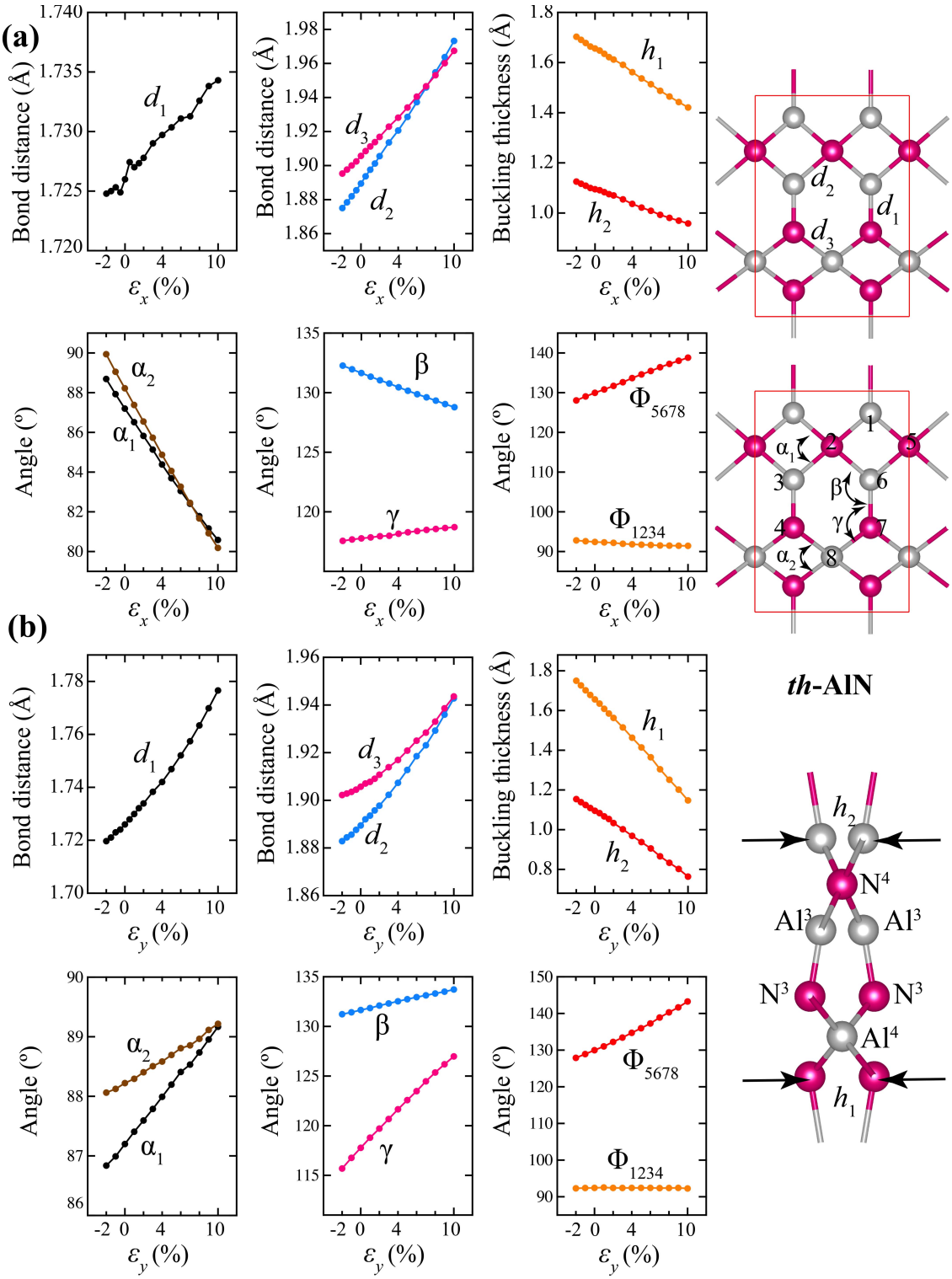

Fig. S7: The variation of bond length d_1, d_2, and d_3, buckling height h_1 and h_2, bond angles, and dihedral angles for *th*-AlN with respect to uniaxial-x strain (ε_x) and uniaxial-y strain (ε_y).

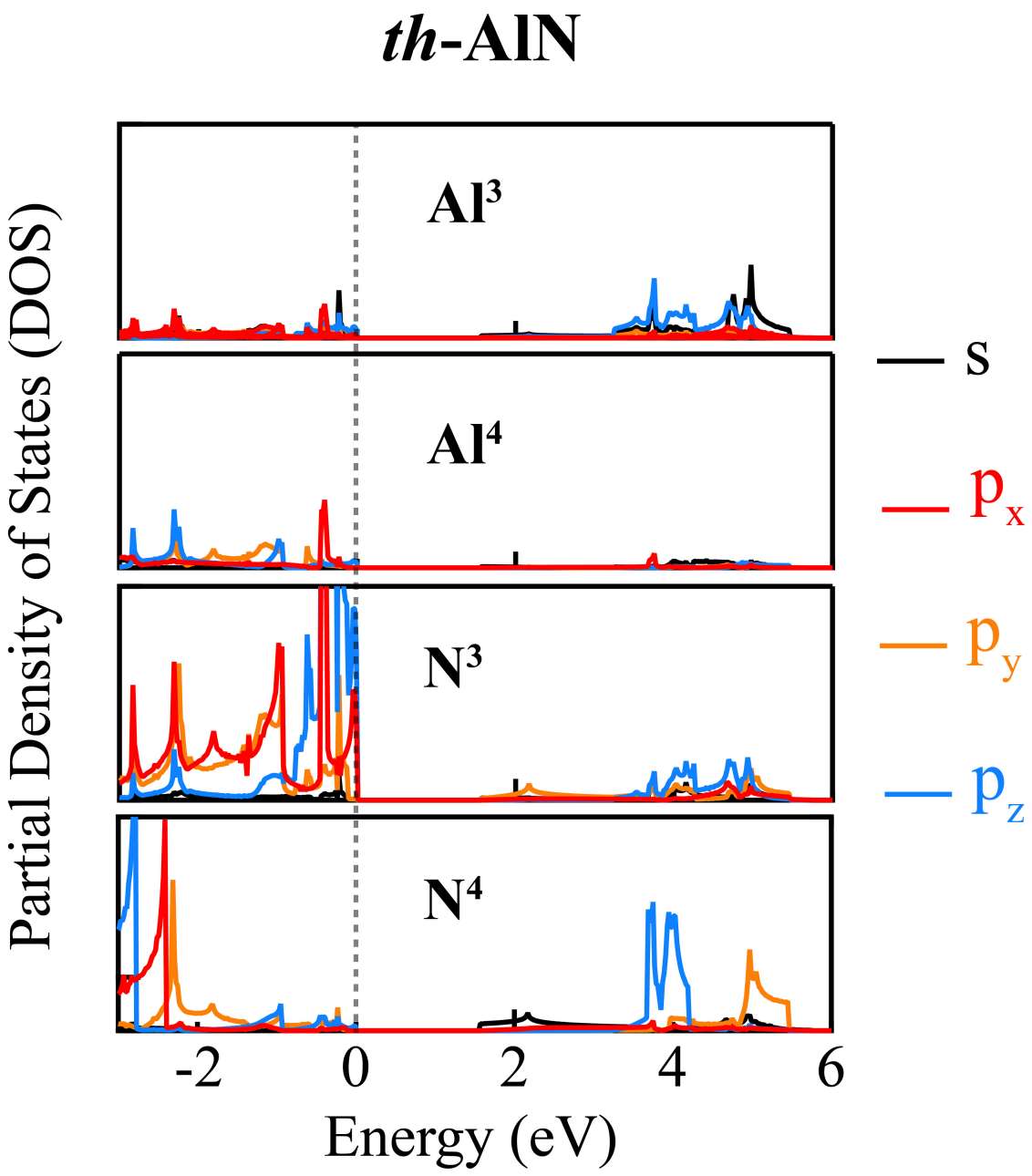

Fig. S8: Electronic density of states (DOS) of *th*-AlN. Here, black, pink, orange, and sky blue represent s, p_x, p_y, and p_z orbitals, respectively.

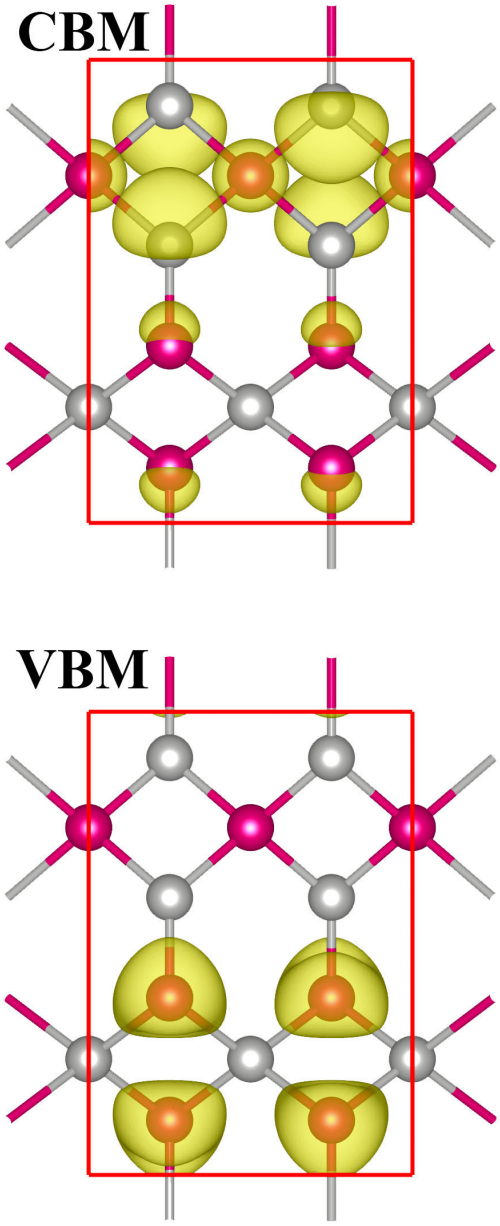

Fig. S9: Charge density of band edges (CBM and VBM) of *th*-AlN. Here, iso-surface of the charge density (yellow color lobes) is 0.005 e/A^3. The silver and pink balls represent Al and N atoms, respectively.

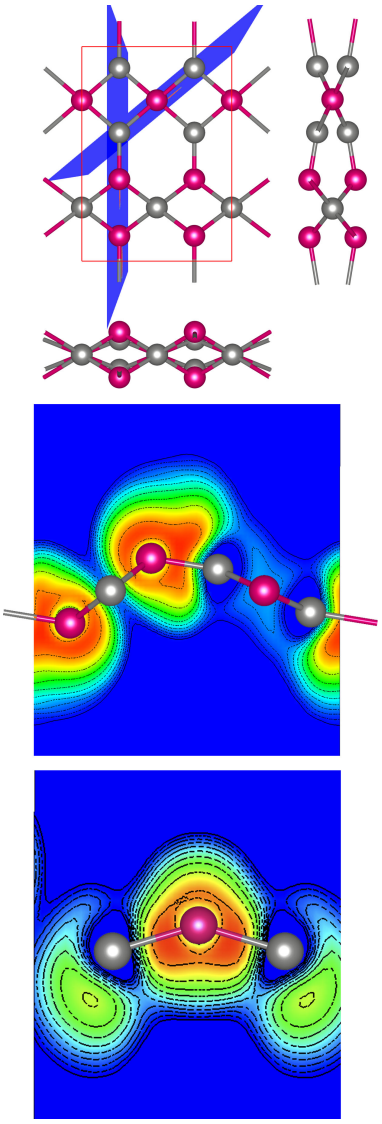

Fig. S10: Electron local function (ELF) of *th*-AlN. The specific 2D-slice projections are chosen to show the bonds between 3-fold coordinated Al and 4-fold coordinated N atoms (Al^3-N^4) as depicted in the middle panel and between 3-fold coordinated Al and N atoms (Al^3-N^3) as depicted in the lower panel. The ELF is dimensionless, and has a range from 0 (in blue color) to 1 (in red color). ELF=1 means highly localized and bounded electrons, while ELF=0 means lack of electron.

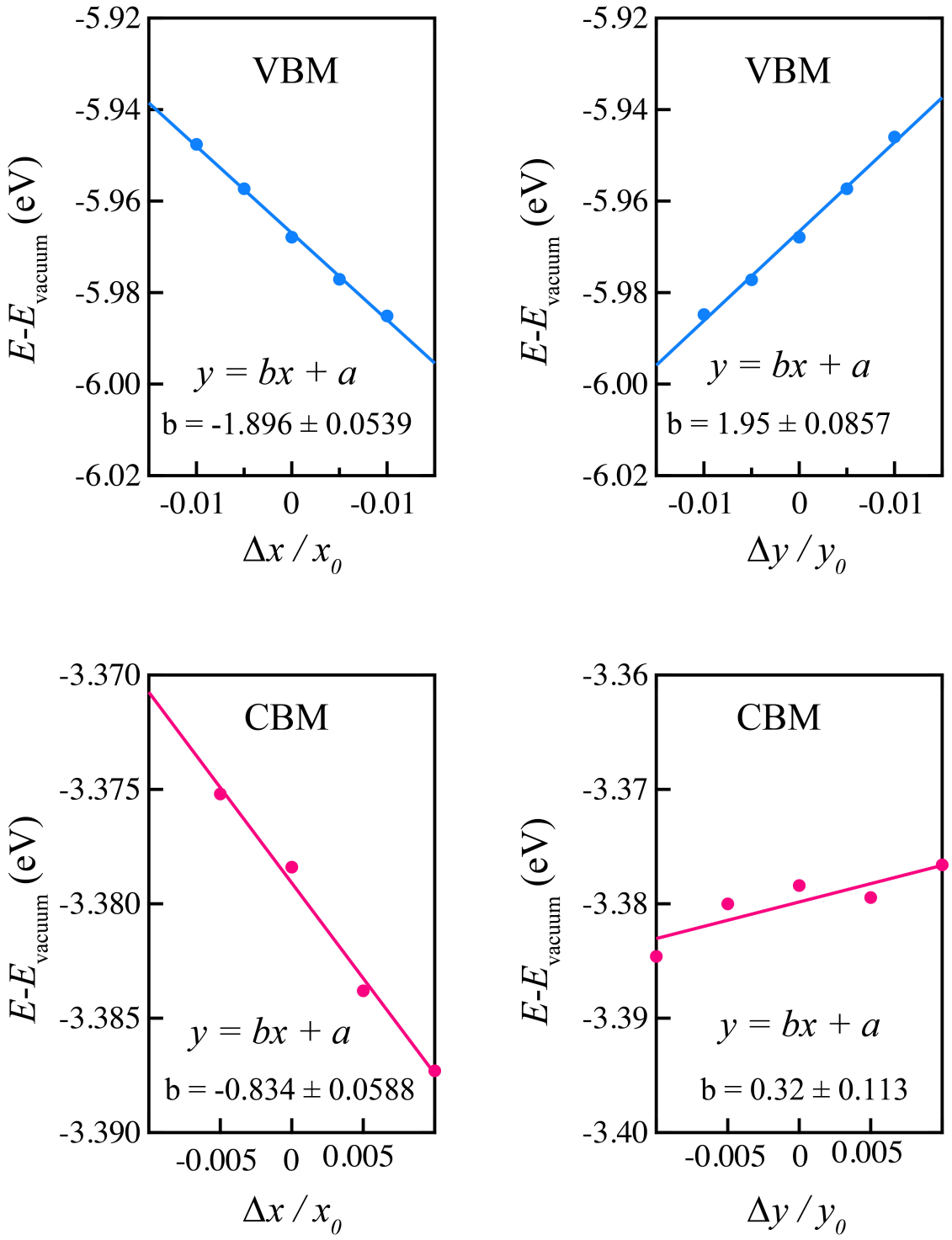

Fig. S11: Dependence of total energy (calculated by the HSE06 functional) of band edges (CBM and VBM positions) *versus* the vacuum level as a function of applied uniaxial strains along the transport direction for *th*-AlN.