Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C. This journal is © The Royal Society of Chemistry 2022



FIG. 1. (Color online) Phonon spectrum of  $Fe_2Br_2$  monolayer by using GGA+U (U=2.5 eV).



FIG. 2. (Color online)(Top) Energy and temperature variations with the increase of the time and (Bottom) final structures (top view (a) and side view (b)) after 8 ps at 300 K for Fe<sub>2</sub>Br<sub>2</sub> monolayer by using GGA+U (U=2.5 eV).



FIG. 3. (Color online) Angular dependence of the Young's modulus (Left) and Poisson's ratio (Right) of Fe<sub>2</sub>Br<sub>2</sub> monolayer by using GGA+U (U=2.5 eV).



FIG. 4. (Color online) The Fe/Br-element and Fe-d-orbital characters of energy bands of Fe<sub>2</sub>Br<sub>2</sub> monolayer by using GGA+U (U=2.5 eV).



FIG. 5. (Color online) Topological edge states of  $Fe_2Br_2$  monolayer by using GGA+U (U=2.5 eV) with only considering SOC of Fe/Br (a/b).



FIG. 6. (Color online) The energy band structures of  $\text{Fe}_2\text{Br}_2$  monolayer by using GGA+U (U=2.5 eV) with five different  $a/a_0$  values.



FIG. 7. (Color online) Topological edge states of  $Fe_2Br_2$  monolayer by using GGA+U (U=2.5 eV) at 0.96 and 1.04 strains.



FIG. 8. (Color online) The energy band structures of  $Fe_2Br_2$  monolayer by using HSE06 (Left) and HSE06+SOC (Right). The black (red) lines represent the band structure in the spin-up (spin-down) direction without SOC.