

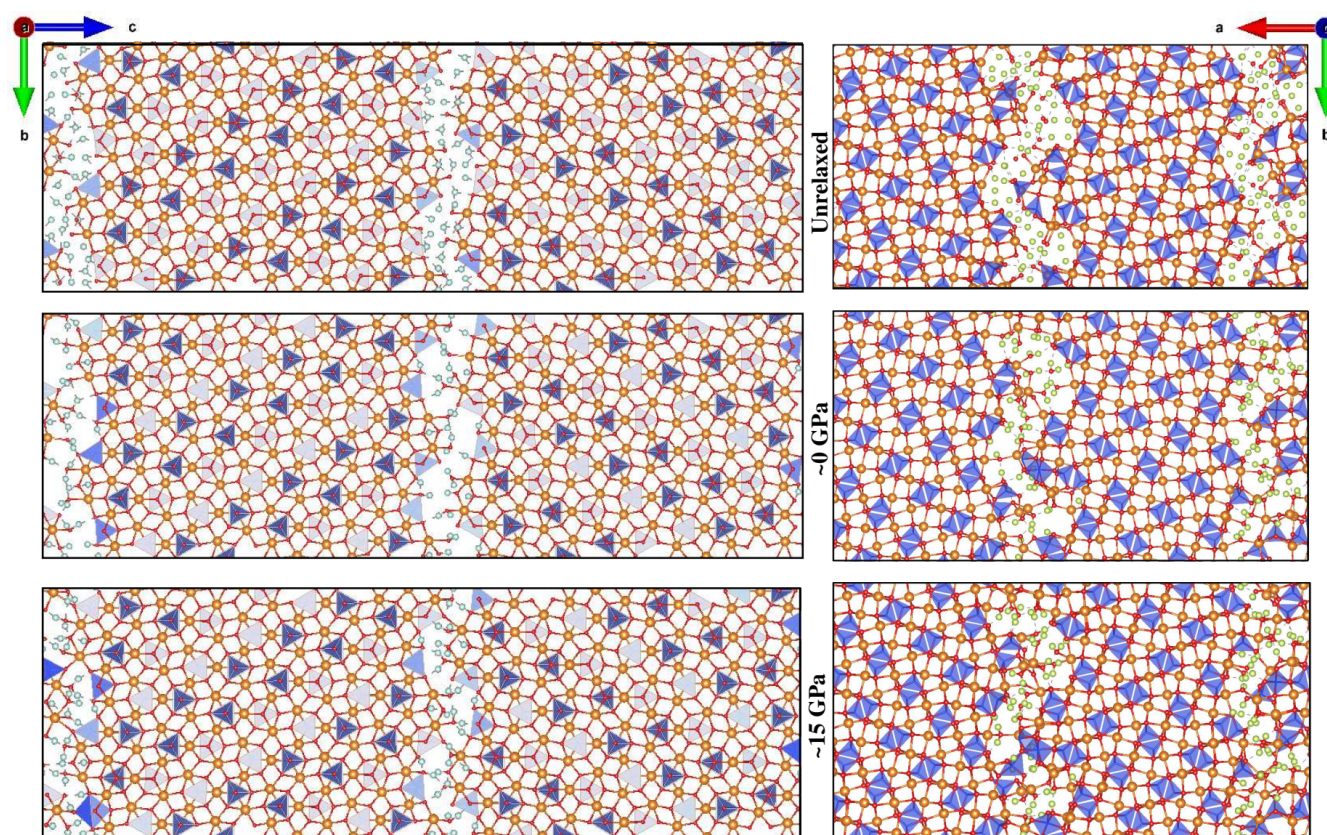
## Supplementary materials

# Hydration-driven stabilization and volume collapse of grain boundaries in $\text{Mg}_2\text{SiO}_4$ forsterite predicted by first-principles simulations

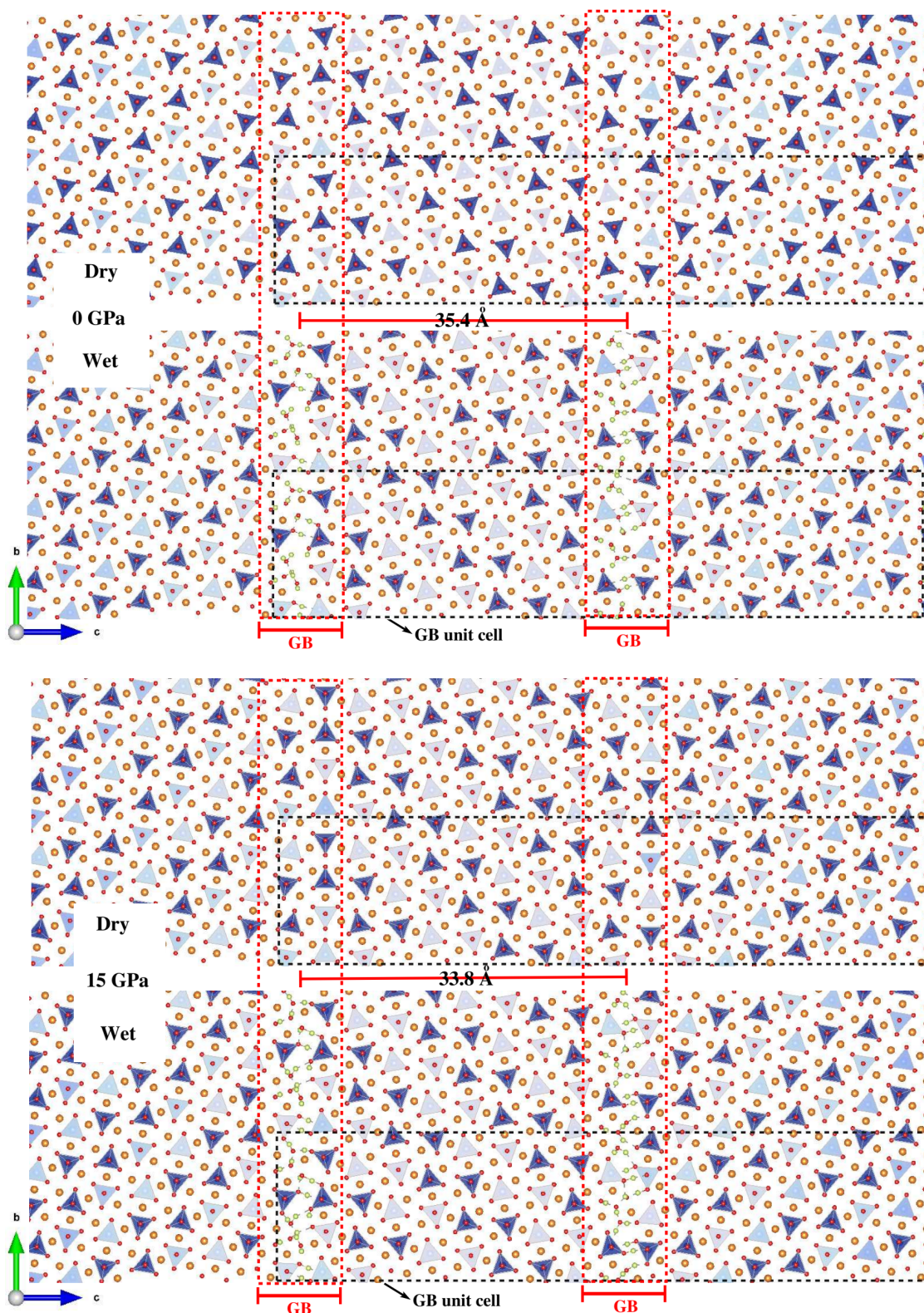
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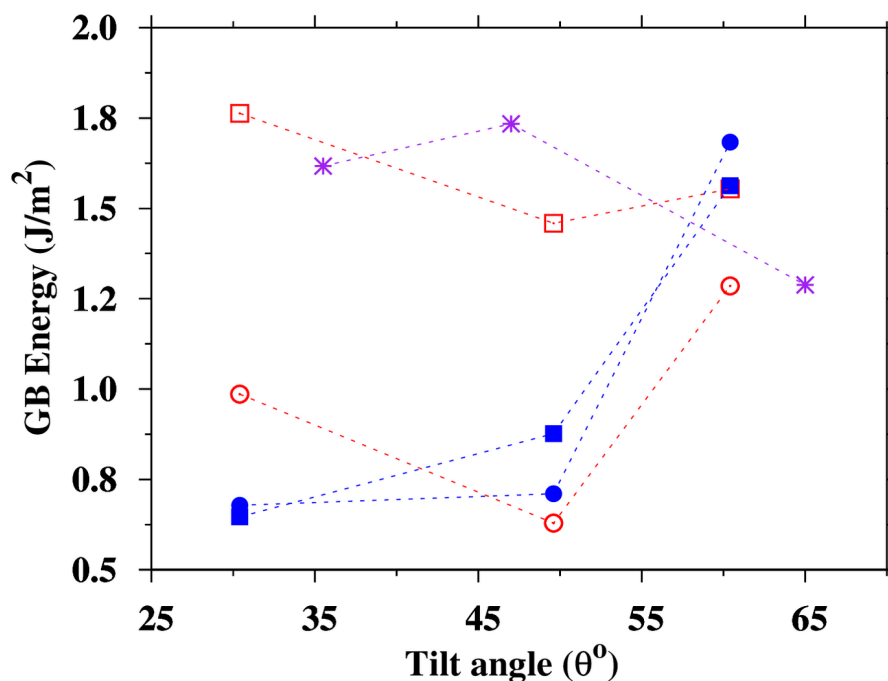
**Fig. S1:** Hydrous grain boundary structures for the stepped  $30.4^\circ$  (left) and stepped  $47.0^\circ$  (right) tilt, shown for unrelaxed and relaxed configurations at  $\sim 0$  and  $\sim 15$  GPa. Direct H-O ( $1^{\text{st}}$  NN) bonds ( $0 \leq \text{cutoff range} \leq 1.2 \text{ \AA}$ ) are represented as solid bi-color lines. The  $2^{\text{nd}}$  NN H-O bonds ( $1.2 < \text{cutoff range} \leq 2.1 \text{ \AA}$ ) are represented by dashed gray lines. Atomic colors brown, blue, red and light green correspond to Mg, Si, O and H, respectively. Si's are also shown as cation-anion polyhedra. To avoid the split edge view of the GB at the supercell edges and provide better visual clarity, a slight rigid shift is given along the  $c$  direction for  $30.4^\circ$  and  $a$  direction for  $47.0^\circ$ .



**Fig. S2:** Relaxed planar GB structures for  $49.6^\circ$  tilt, both in the anhydrous and hydrous setting, shown for 0 (top) and 15 (bottom) GPa. GB regions are marked by red dashed rectangles and GB unit cells are marked by black dashed rectangles. Atomic colors brown, blue, red and light green correspond to Mg, Si, O and H, respectively. Si's are shown as blue tetrahedra.



**Fig. S3:** Tilt angle dependence of the GB formation energy at 0 GPa for (011)/[100] type (30.4°, 49.6° and 60.4°) in planar (circles) and stepped (squares) configurations. The dry (water-free) and wet (hydrous) grain boundaries are represented by open and solid symbols, respectively. Also shown are the results for wet stepped (110)/[001] type (35.5°, 47.0° and 65.0°) GBs (asterisks).



**Fig. S4:** Pressure variation of volume change for crystalline, dry and hydrous grain boundaries (planar 30.4° tilt), and amorphous bulk H<sub>2</sub>O phase. Dotted lines mark the changes over the range 0-5 GPa.

