Supplementary Materials for

Intersectional nanotwinned diamond-the hardest polycrystalline diamond by design

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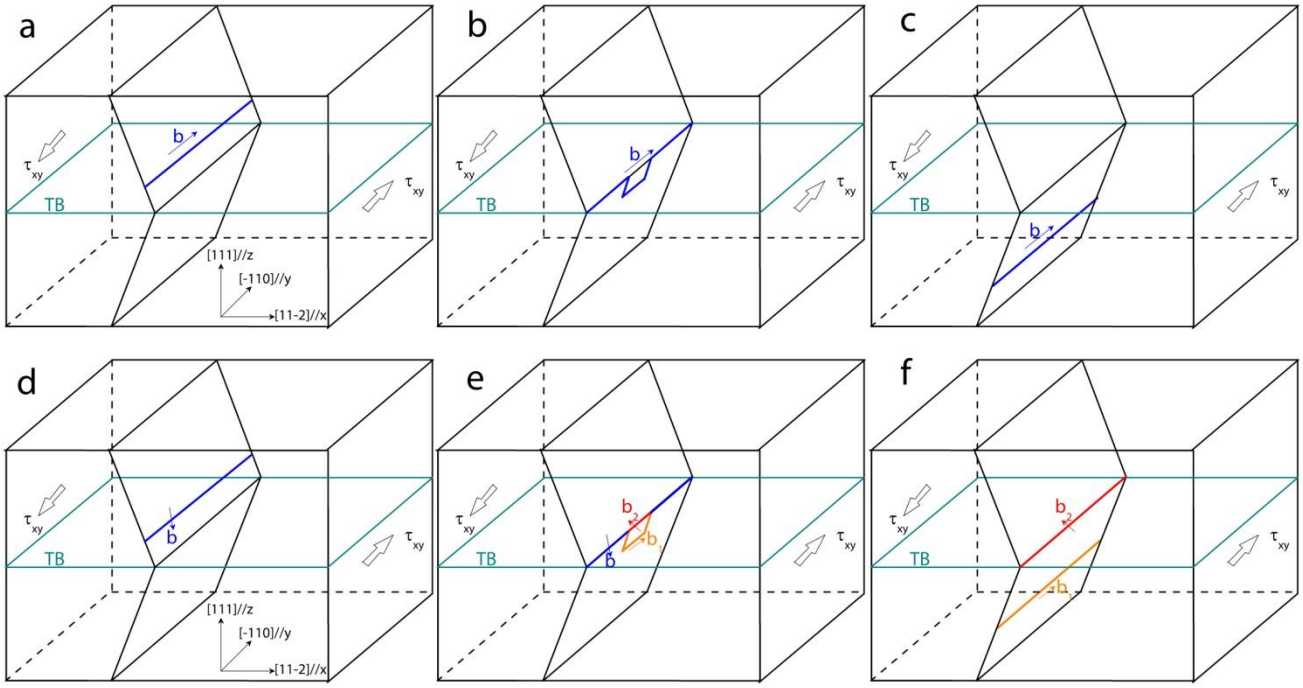
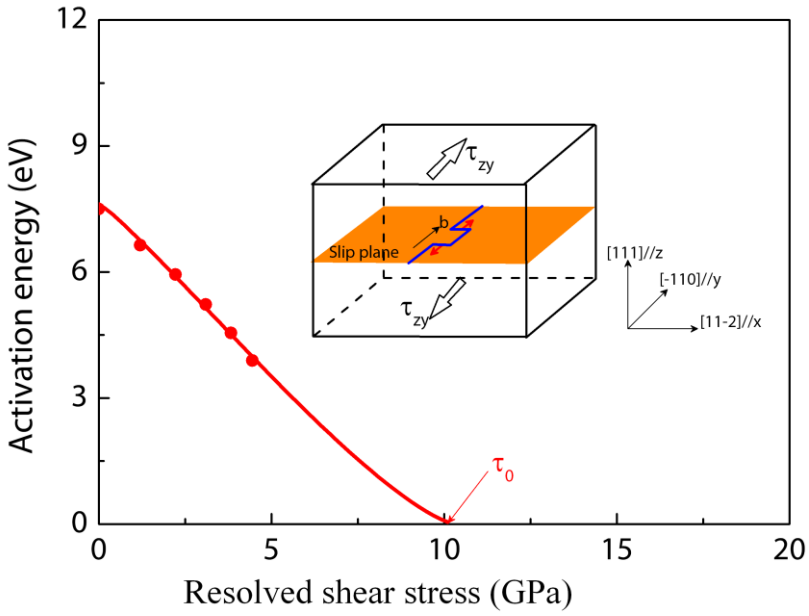
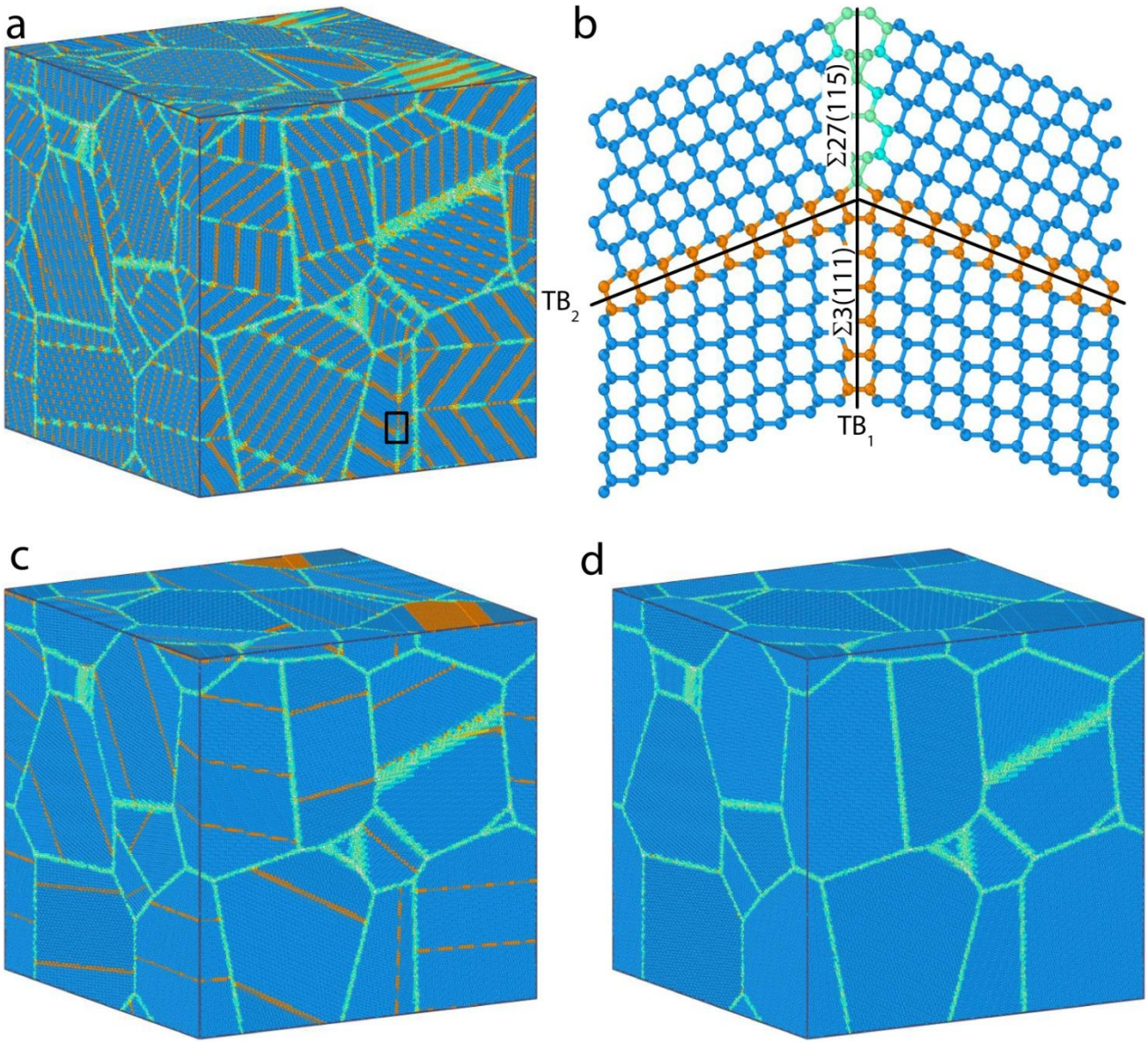


Supplementary Fig. 1 Schematic for shuffle-set 0 ° and 60 ° perfect dislocation reaction with TB. **a~c** Process of shuffle-set 0 ° perfect reaction with TB. **d~f** Process of shuffle-set 60 ° perfect reaction with TB. The dislocation reaction involves the formation of kinks, which migrate along the dislocation line.



Supplementary Fig. 2 Shear stress-dependent activation energy of dislocation motion in the diamond. The insert figure is schematic of shuffle-set 0° perfect dislocation motion in slip plane. The slip process involves kink formation and migration along the dislocation line. When the activation energy reaches zero, the corresponding shear stress is lattice friction stress τ_0.



Supplementary Fig. 3 Structure model of int-diamond, nt-diamond and ng-diamond. a Computational model for studying the yield strength of int-diamond. **b** The local enlarged drawing for the black rectangle in **a** showing detailed int-diamond structure. **c** Computational model for studying the yield strength of nt-diamond. **d** Computational model for studying the yield strength of ng-diamond.

Supplementary Table 1 The parameters used to calculate the critical resolved shear stress of slip transfer mode, confined layer slip mode, confined slip transfer mode I, and confined slip transfer mode II. τ_0 is the lattice function stress, in GPa; τ_{TB}^{0} is the barrier strength of shuffle-set 0° perfect dislocation reaction with TB, in GPa; τ_{TB}^{60} is the barrier strength of shuffle-set 60° perfect dislocation reaction with TB, in GPa; ν is Poisson's ratio; G is shear modulus, in GPa; b is the magnitude of Burgers vector, in nm; θ is the angle between slip plane and twin plane, in degree (°).

Parameters	τ_0	τ_{TB}^{0}	τ_{TB}^{60}	ν	G	b	θ
	10.3	19	48	0.078	540	0.25	70.5