

Supporting Information

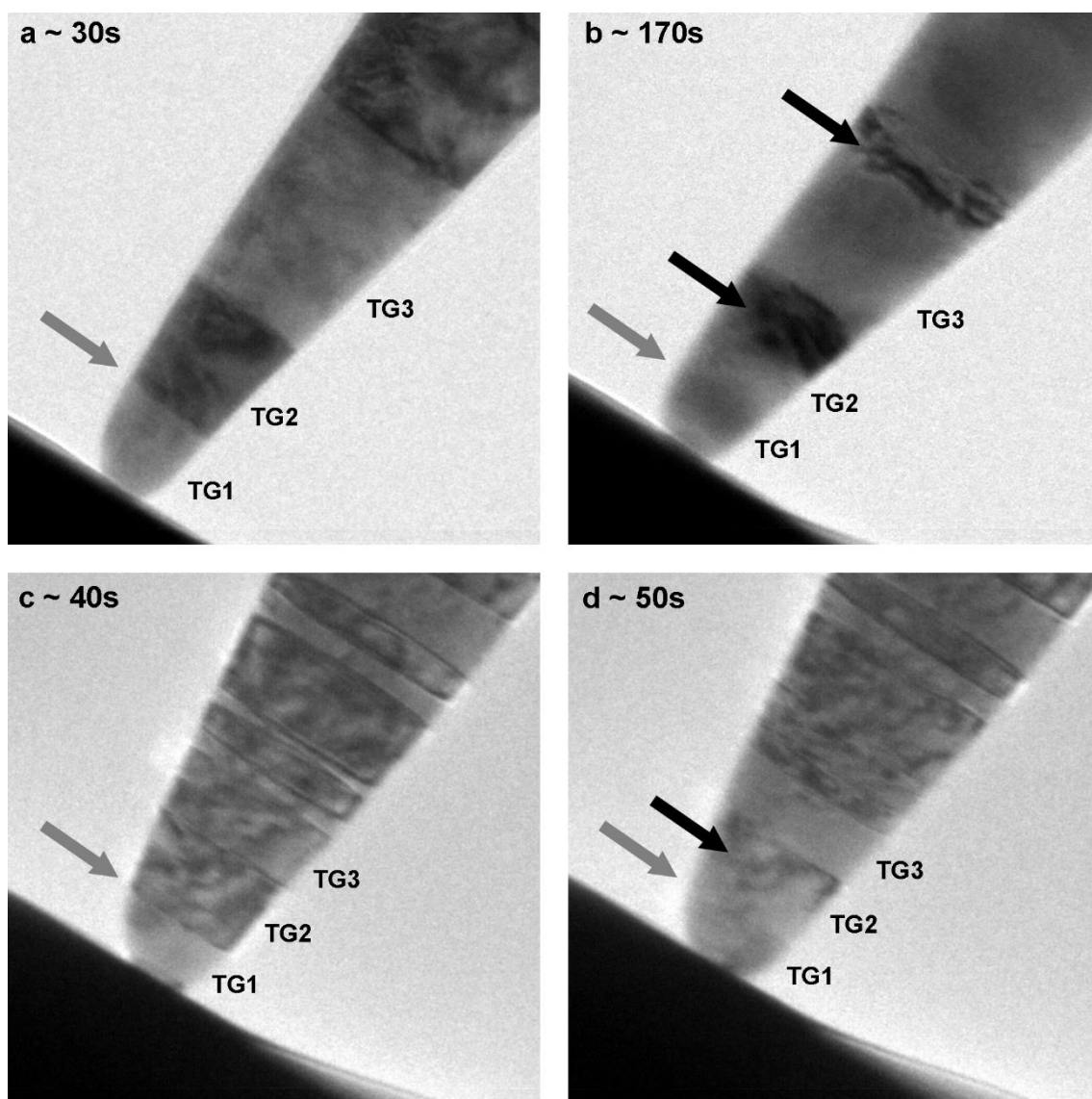


Figure S1. Captured *in-situ* TEM images of nanoscopic deformation of nt-Cu nanopillars: under compression at a medium strain rate of about 1.3×10^{-4} /s for different time: (a) about 30 s, (b) about 170 s (black arrows: dislocation activities, grey arrows: detwinning); under compression at an ultralow strain rate of about 2.3×10^{-5} /s for different time: (c) about 40 s, (d) about 50 s (black arrows: dislocation activities, grey arrows: detwinning).

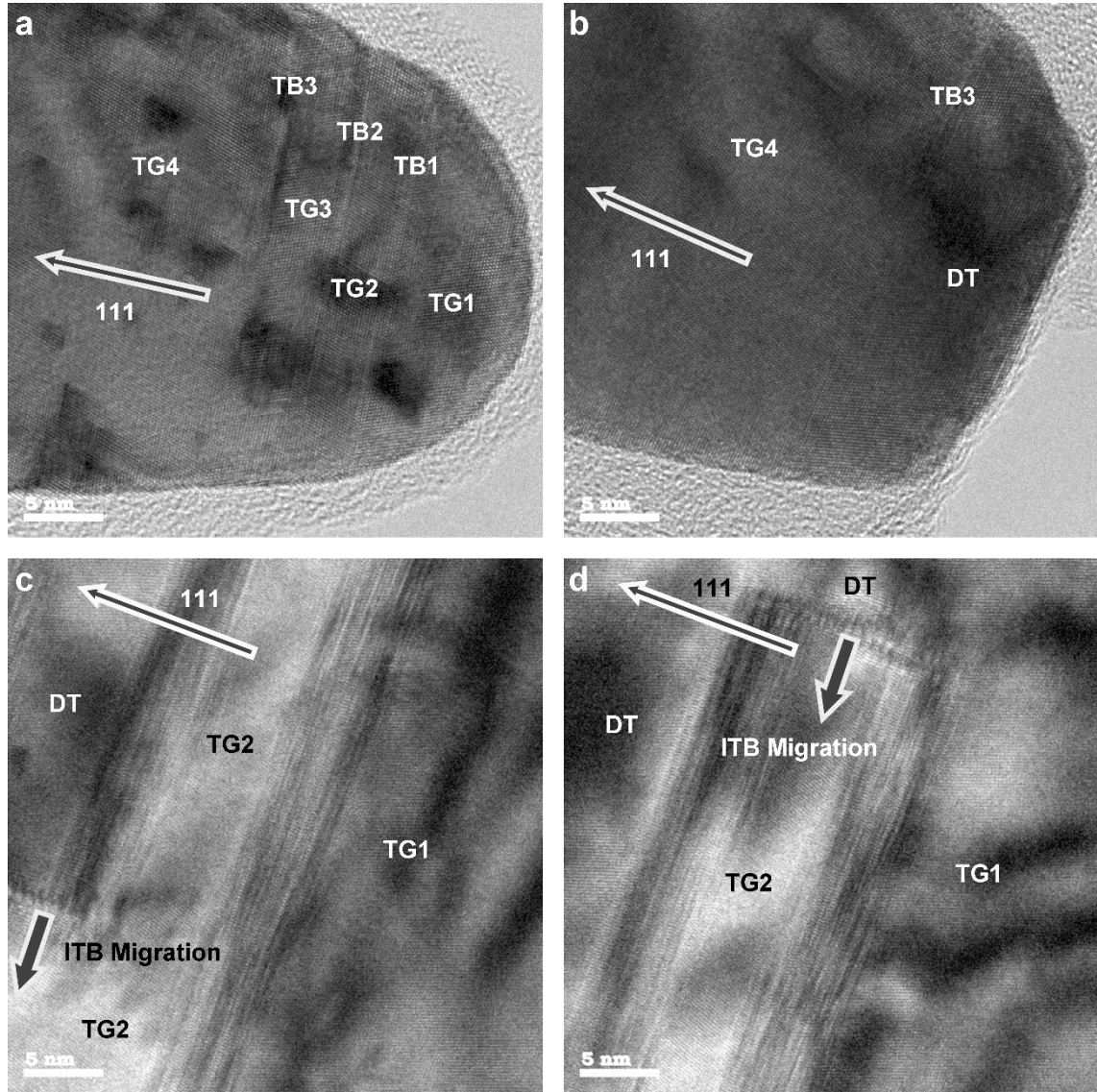


Figure S2. Original high-resolution TEM lattice images of nt-Cu nanopillars: at the pillar tip, (a) before and (b) after *in-situ* TEM compression (test interrupted at a slight strain) at a low strain rate of about 5.3×10^{-5} /s, showing $\{111\}$ TB migration (TB3, along the $\langle 111 \rangle$ direction) and detwinning (TG2 and TG3); several twin grains away from the pillar tip ((c) the left part and (d) the right part of the pillar sample), after *in-situ* TEM compression (test interrupted at a slight strain) at a low strain rate of about 4.5×10^{-5} /s, showing $\{1-21\}$ ITB migration (along the thick arrows) and detwinning (the right part of TG2; TG: twin grain, DT: detwinned).

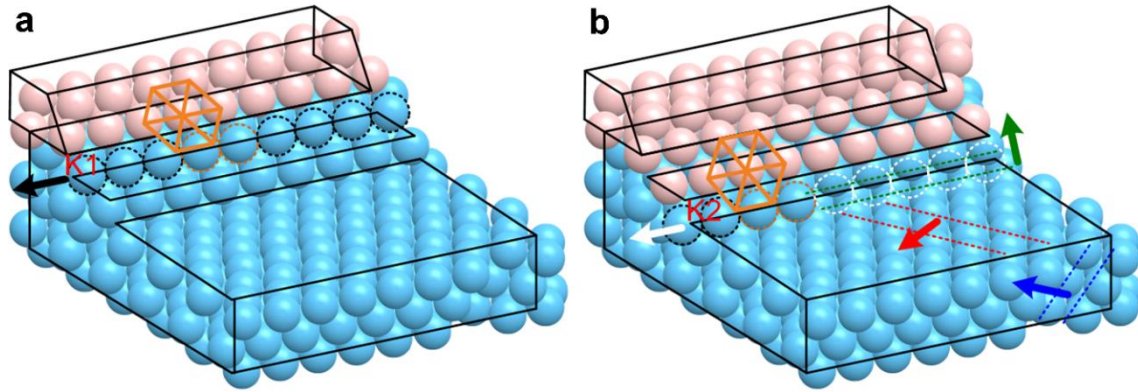


Figure S3. Schematic models of the kink-like motion of atoms and the interaction of kink with partial dislocation (referred to Figure 7c, top layers of atoms removed for clearer illustrations; blue atoms: below TB, pink atoms: above TB). (a) The $1/4 \langle 10-1 \rangle$ shift (black arrow) of a row of black-dash-circled blue atoms at kink K1 (atom by atom, *ex.* the movement of two orange-dash-circled blue atoms to the bottom-left and bottom-right sites of the orange hexagon) will align the row of blue atoms with the pink atoms to extend the close-packed $\{111\}$ plane above TB and hence detwin the row of atoms (the lattice below TB). (b) When an activated $1/6 \langle 21-1 \rangle$ partial dislocation (blue dashed lines and blue arrow) is incident to and interact with a $1/4 \langle 10-1 \rangle$ kink, the $1/6 \langle 21-1 \rangle$ partial dislocation will be dissociated into a $1/6 \langle 101 \rangle$ stair-rod (green dashed lines and green arrow) and a $1/6 \langle 11-2 \rangle$ glissile (red dashed lines and red arrow). The row of the dash-circled blue atom sites will be emptied by the $1/6 \langle 101 \rangle$ stair-rod to form a row of white-dash-circled vacancies at kink K2 (atom by atom, white-dash-circled). The $1/4 \langle 10-1 \rangle$ kink-like shift will cease operating (white arrow), and the $1/6 \langle 11-2 \rangle$ glissile will alternatively dominate the detwinning process.