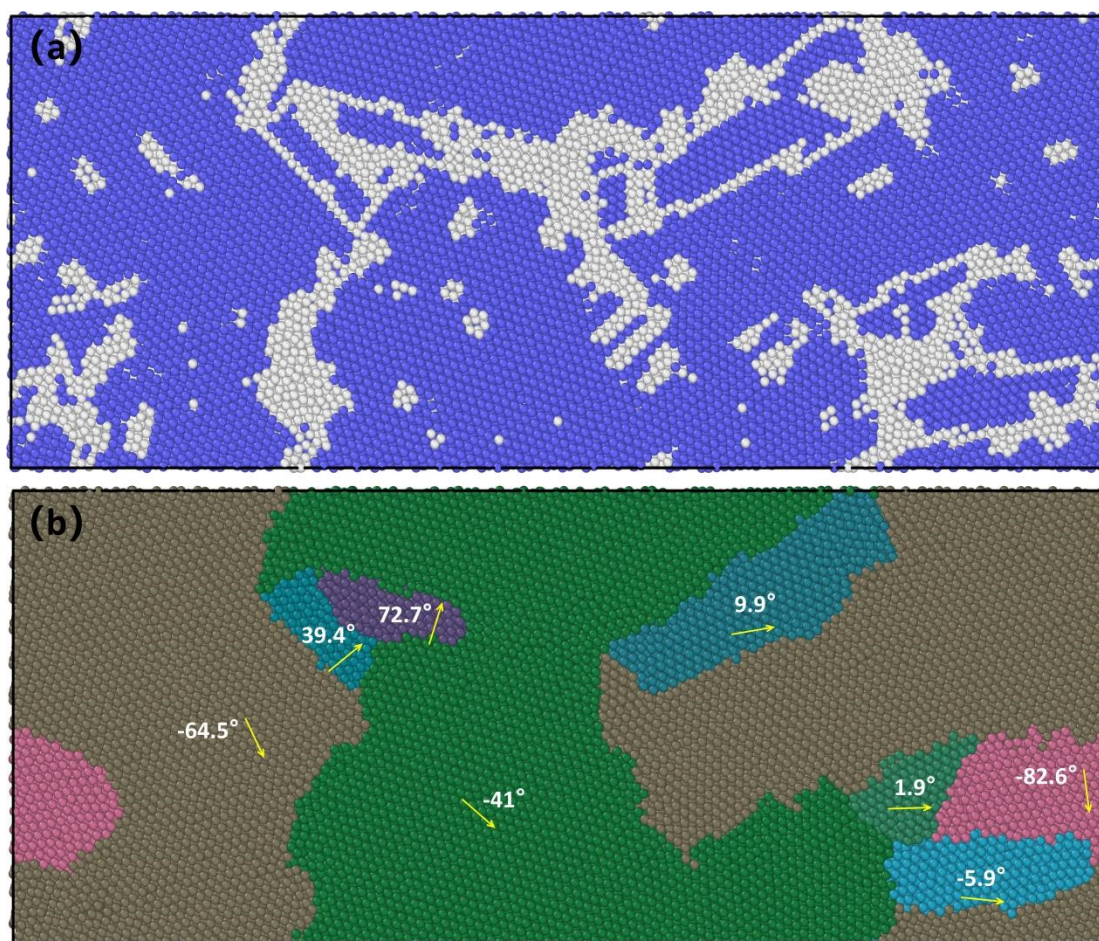
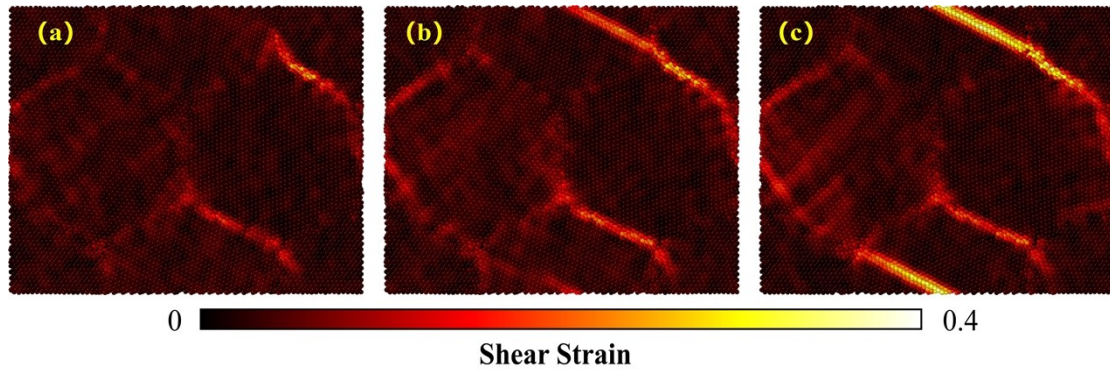


## Supplementary Materials



**Fig. S1.** (a) Snapshot of the atomic structure and (b) Snapshot of grain delineation in the polycrystalline microstructure of the NC U-12Mo alloy with a grain size of 12 nm at a strain level of 50%. A comparison of the orientations of different regions relative to the X axis with those in the undeformed microstructure (see Fig. 1) indicate that significant rotations about [110] have taken place during the deformation. The sample contains two larger grains and six smaller grains. Among them, the two larger grains together occupy 83.2% of the system, with lattice orientations of  $-41^\circ$  and  $-64.5^\circ$  (i.e.,  $41^\circ$  and  $64.5^\circ$  rotated in the clockwise direction, respectively).



**Fig. S2.** Snapshots of shear strain of NC U-12Mo alloy with grain size of 12 nm during deformation at 300 K and  $1 \times 10^8 \text{ s}^{-1}$  strain rate at strains of (a) 3.6%, (b) 4.2 and (c) 4.3%, respectively. The upper grain boundary where grain 2 is in contact with grain 3 bears a large shear strain. Subsequently, at 4.2% strain, the first twin appears in grain 4, which emits from the grain boundary triple junction as a way of releasing the stress.