

## Description of Additional Supplementary Files

File Name: Supplementary Movie 1

Description: Dislocation nucleation assisted by surface diffusion in a  $\bar{1}$ -oriented 6.6-nm-diameter Ag NW at room temperature and a strain rate of  $\sim 10^{-3}$  s $^{-1}$ .

File Name: Supplementary Movie 2

Description: Pure displacive deformation in a  $\bar{1}$ -oriented 5.9-nm-diameter Pt NW at room temperature and a strain rate of  $\sim 10^{-3}$  s $^{-1}$ .

File Name: Supplementary Movie 3

Description: Molecular dynamics simulation of tensile straining of 6-nm-diameter Ag NW at 800 K and a strain rate of 10 $^5$  s $^{-1}$  showing dislocation nucleation assisted by long-range surface diffusion.

File Name: Supplementary Movie 4

Description: Molecular dynamics simulation of tensile straining of 6-nm-diameter Ag NW at 800 K and a strain rate of 10 $^5$  s $^{-1}$  showing dislocation nucleation assisted by localized diffusion of individual atoms at free surface in a random and chaotic way.

File Name: Supplementary Movie 5

Description: Molecular dynamics simulation of tensile straining of 20-nm-diameter Ag NW at 800 K and a strain rate of 10 $^5$  s $^{-1}$  showing dislocation nucleation with weak surface diffusion.

File Name: Supplementary Movie 6

Description: Molecular dynamics simulation of tensile straining of 6-nm-diameter Pt NW at 800 K and a strain rate of 10 $^5$  s $^{-1}$  showing surface dislocation nucleation and plastic flow with nearly no diffusional events.

File Name: Supplementary Movie 7

Description: Surface atom diffusion induced abnormal softening during plastic flow in a  $\bar{1}$ -oriented 13.1-nm-diameter Ag NW at room temperature and a strain rate of  $\sim 10^{-3}$  s $^{-1}$ . The movie is played at a speed of 3X.

File Name: Supplementary Movie 8

Description: Surface atom diffusion induced abnormal softening during plastic flow in a  $\bar{1}$ -oriented 30-nm-diameter Ag NW at a strain rate of  $\sim 10^{-3}$  s $^{-1}$ .