Supplementary Materials

1. Evaluation of simulation box size

In the study, the substrate has a dimension of 120a×120a×60a along x-[100], y-[010], and z-[001] crystal orientations, and a is the lattice constant. In the open literature, we found that many models are as large as or smaller than our model. For example, in the nanoscratch simulation of Fe [1], the box size is 100a×100a×50a; and for Cu [2], the box size is 70a×50a×33a. These studies can obtain the main features of friction and wear in the related materials. Particularly, in the nanoscratching of Al [3], the simulation results showed that the friction coefficient becomes relatively stable as the thickness is larger than ~16a.

Furthermore, we also discussed two other larger model sizes, i.e., 180a×120a×60a and 120a×120a×80a. As shown in Figure S1, a larger box would not alter the main friction behaviors in this study. Thus, the model size used in this study is considered large enough.

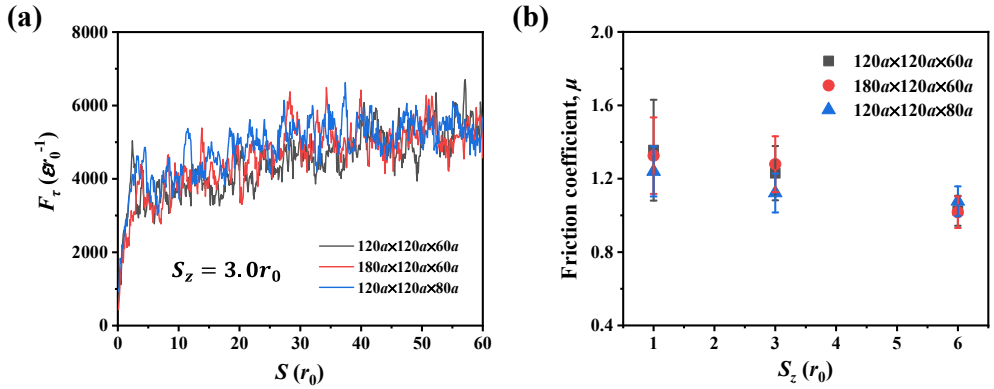

Figure S1. Evaluation of simulation box size: (a) the tangential forces for three models, and (b) the friction coefficient at three scratch depths.

2. Reliability of simulation results

To demonstrate the reliability of simulation results, we performed another two sets of nanoscratch simulations. As shown Figure S2, it was confirmed that there is little difference in the concerned results over a wide range of scratch depths.

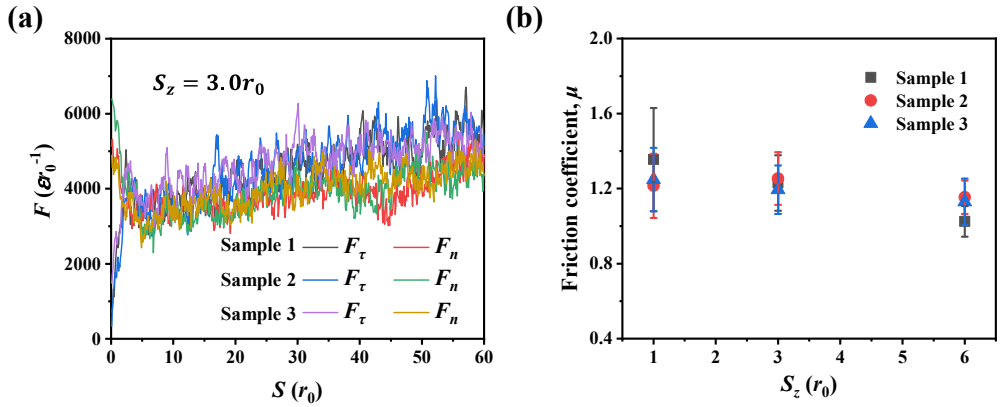

Figure S2. (a) Tangential force and normal force at the depth of 3.0r_0 in three samples, and (b) the friction coefficient at different scratch depths.

References

[1] Y. Gao, H.M. Urbassek, Scratching of nanocrystalline metals: A molecular dynamics study of Fe. *Applied Surface Science* **2016**, *389*, 688-695.

[2] J. Li, Q. Fang, Y. Liu, L. Zhang, Scratching of copper with rough surfaces conducted by diamond tip simulated using molecular dynamics. *The International Journal of Advanced Manufacturing Technology* **2014**, *77*, 1057-1070.

[3] T. Junge, J.-F. Molinari, Plastic activity in nanoscratch molecular dynamics simulations of pure aluminium. *International Journal of Plasticity* **2014**, *53*, 90-106.