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Supplementary Materials for

Atomic-scale insights into the interfacial instability of superlubricity in hydrogenated amorphous carbon films

Xinchun Chen, Xuan Yin, Wei Qi, Chenhui Zhang*, Junho Choi, Sudong Wu, Rong Wang, Jianbin Luo*

*Corresponding author. Email: chzhang@tsinghua.edu.cn (C.Z.); luojb@tsinghua.edu.cn (J.L.)

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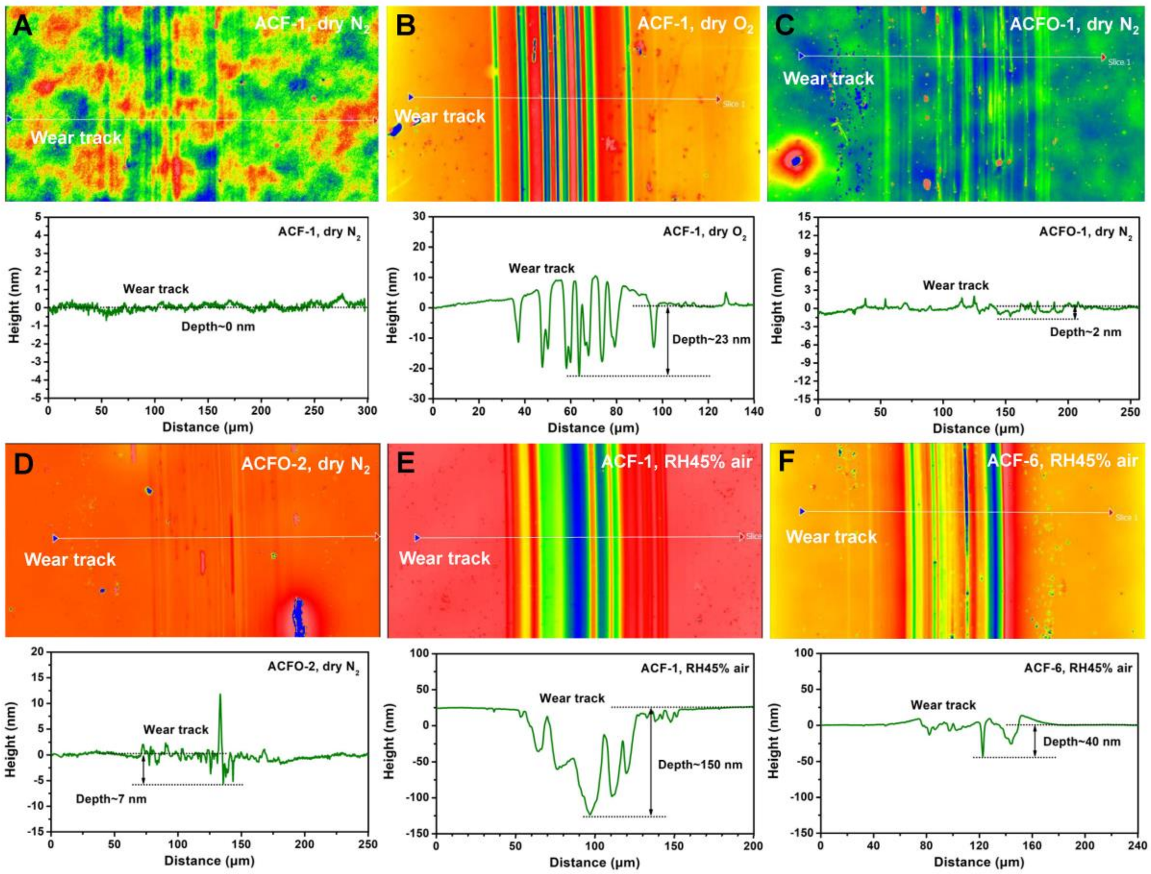

Fig. S1. Surface morphologies and cross sections of the wear tracks produced on amorphous carbon film surfaces after the friction tests as shown in Fig. 1 (G and H).

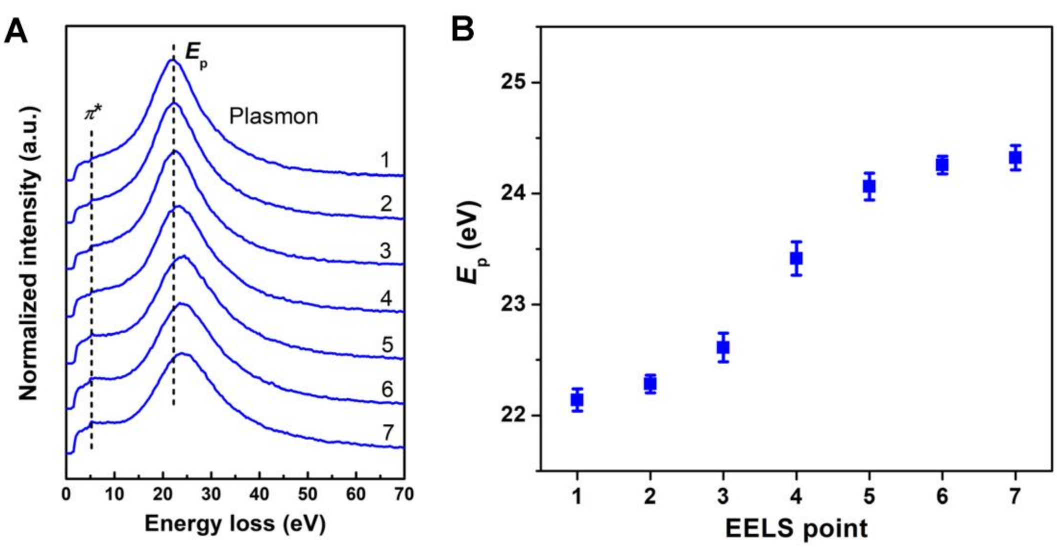

Fig. S2. (**A**) **Evolution of EELS low-loss spectra recorded across the shallow shear band as marked in Fig. 4F and (B) the derived E_p values through peak fitting.**

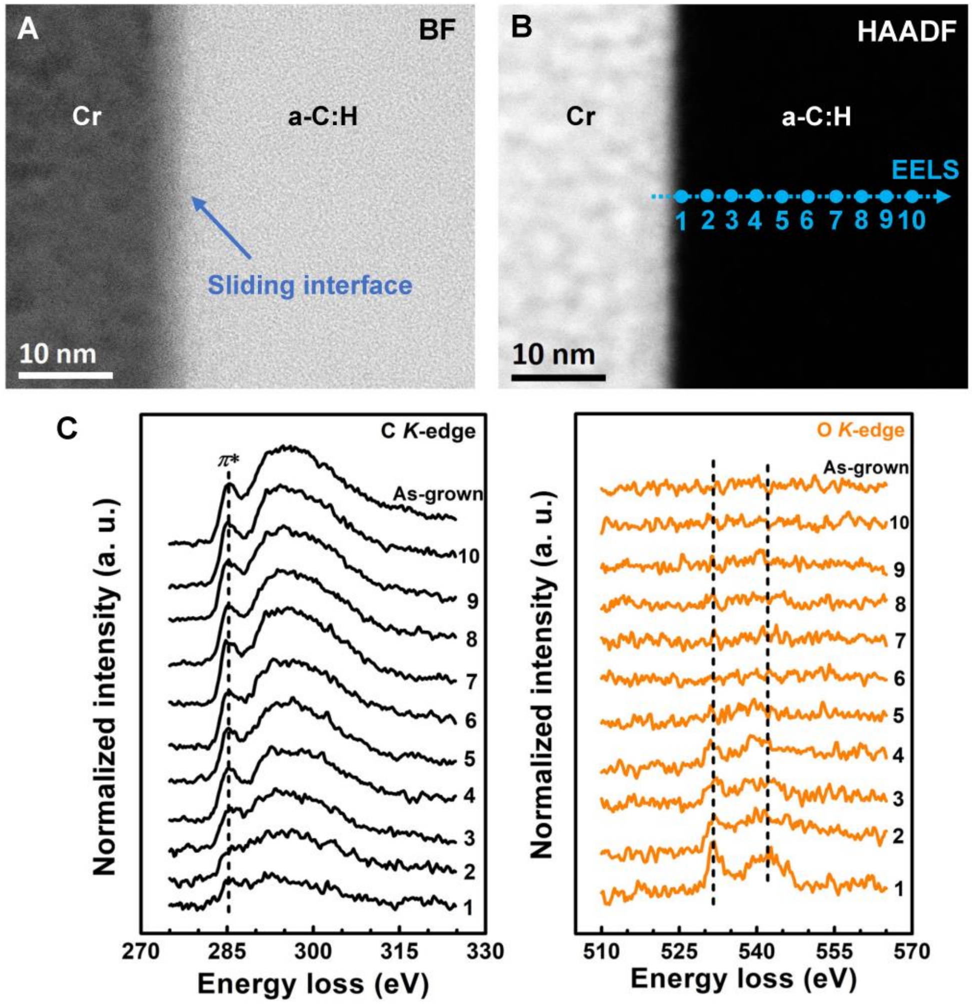

Fig. S3. STEM and EELS characterization of the wear track formed on a-C:H film surface after sliding against bare SUJ2 steel ball in humid air (ACF-1, Fig. 1H). (**A**) and (**B**) STEM-BF and STEM-HAADF images showing the morphologies of the sliding interface in the wear track. (**C**) The EELS C-*K* and O-*K* core edges recorded across the sliding interface as marked in (**B**), which indicate the oxidization of the film surface with an affected depth of 10 nm (EELS points 1-5).

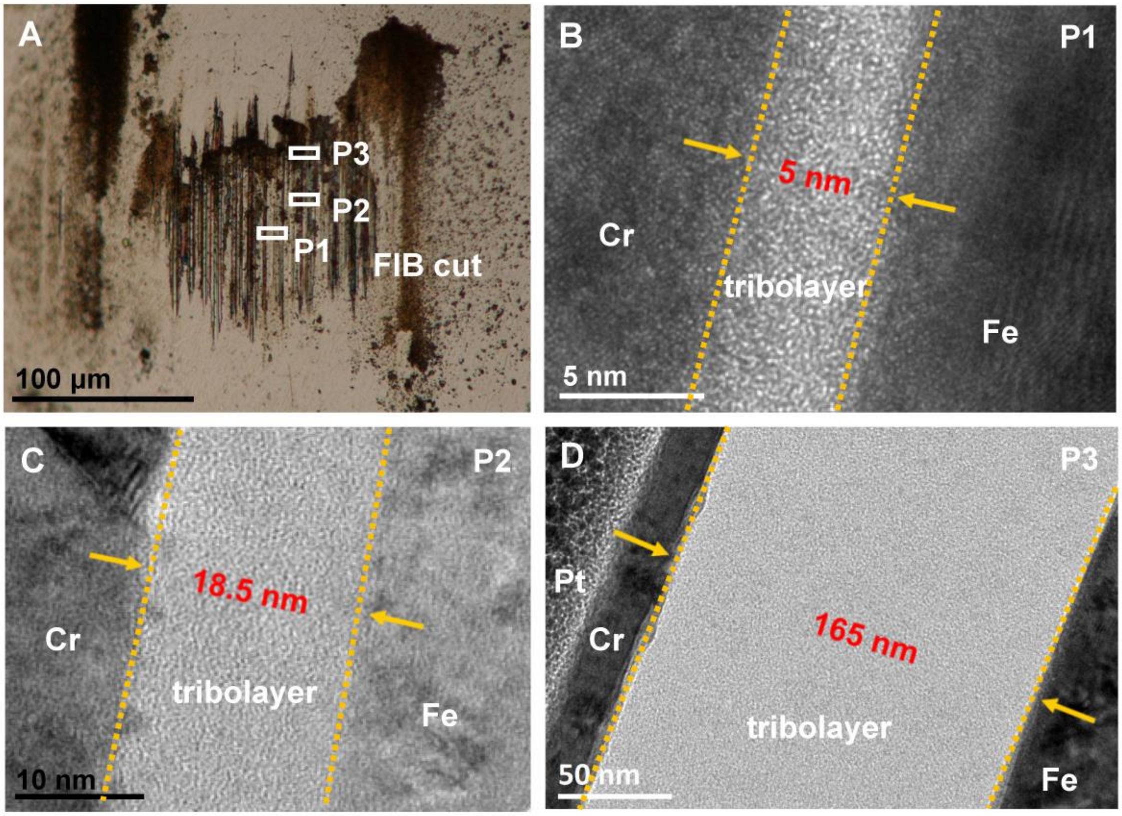

Fig. S4. FIB slicing and TEM characterization confirming the variable thicknesses of the tribolayer in different contact positions of the wear scar formed on bare steel ball surface after sliding against a-C:H:Si film in humid air (ACF-6, Fig. 1H). Confirming the variable thicknesses of the tribolayer in different contact positions of the wear scar. (**A**) Three representative FIB-cutting positions including the scar center (P1), the scar edge (P3) and the position between them (P2). (**B-D**) HRTEM images indicate the thicknesses of the tribolayer in P1, P2 and P3 are 5, 18.5 and 165 nm, respectively.

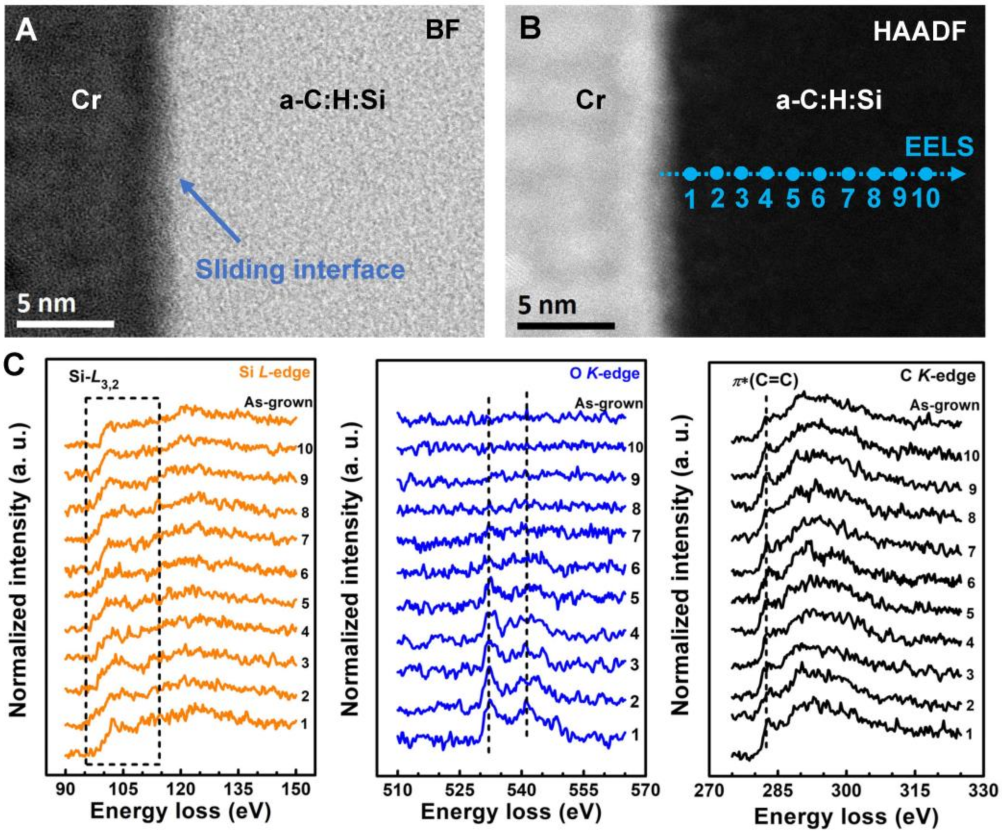

Fig. S5. STEM and EELS characterization of the wear track formed on a-C:H:Si film surface after sliding against bare SUJ2 steel ball in humid air (ACF-6, Fig. 1H). (**A**) and (**B**) STEM-BF and STEM-HAADF images showing the morphologies of the sliding interface in the wear track. (**C**) The EELS Si-*L*, O-*K* and C-*K* core edges recorded across the sliding interface as marked in (**B**), which indicate the formation of a thin silica-like oxidized layer (~5 nm, EELS points 1-5) on the film surface.