Supporting Information:

Friction between van der Waals solids during lattice directed sliding

Paul E. Sheehan*†‡ and Charles M. Lieber‡

† US Naval Research Laboratory, Code 6177; Washington, DC 20375;
paul.sheehan@nrl.navy.mil

‡ Department of Chemistry and Chemical Biology, Harvard University; Cambridge, MA 02138

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Materials and Methods.

Growth of MoSe$_2$ crystals.

The molybdenum diselenide crystals were prepared in two steps. First, one equivalent of Mo and two equivalents of Se with a total mass of 10g were sealed in a quartz ampoule under vacuum and reacted at 750° C for seventy hours. Analysis by powder x-ray diffraction indicated it was of the correct structure (Figure S1). Second, single crystals of MoSe$_2$ were grown via chemical vapor transport. Three grams of the powder were ground and resealed at one end of a 30 cm long quartz tube with 60 µL of bromine which acts as transport agent. A temperature gradient of 80 °C (i.e., 995—915 °C) was applied along the length of the tube to enhance transport. After two weeks, the quartz tube was removed and the crystals collected. The plate-like crystals had lengths and widths from 3 to 10 mm.

Growth of MoO$_3$ nanocrystals.

The samples consisted of MoO$_3$ nanocrystals thermally grown on molybdenum dichalcogenide substrates. The natural molybdenite (MoS$_2$) was obtained from the collections of the Peabody Museum (Harvard University, Cambridge, MA). From this, a square sample approximately 6 mm wide was cleaved with a razor, and then its surface was cleaned by removal of the topmost layers using adhesive tape. The surfaces of the 3-10 mm long MoSe$_2$ crystals were cleaned in the same way. For both samples, nanocrystals of MoO$_3$ were formed by oxidizing these molybdenum dichalcogenide substrates in a tube furnace following a previously developed method. Briefly, a quartz tube within a tube furnace and connected to an O$_2$ gas supply heated a one inch long ceramic boat to 480° C for several hours to equilibrate the boat temperature as determined by a thermocouple. To oxidize the samples, the downstream end cap of the tube would be removed, the boat quickly pulled to the open end of the tube, a freshly cleaved crystal placed in its center, and then the boat would be returned to the center of the furnace. This enabled the crystal insertion with only a ~ 3 °C drop in temperature and within a ~10 second period. After five minutes, the oxidized crystal would be removed. Nanocrystals formed this way were previously extensively characterized by Kim et al. using TEM, XPS, and AFM to
determine that they were MoO$_3$ with lattice values of $a = 3.966$ Å and $c = 3.696$ Å close to the bulk MoO$_3$ ($a = 3.963$ Å, $c = 3.696$ Å).$^4$

**Selection of MoO$_3$ nanocrystals.**

MoO$_3$ nanocrystals were chosen for measurements based on several requirements. First, the nanocrystals were at most a few monolayers thick so that they can be machined efficiently with the AFM probe tip.$^3,^5$ Second, the nanocrystals must have sufficient space (>300 nm) between them to allow for movement during friction measurements. Finally, the exposed substrate should be unoxidized and relatively free of etch pits since these could affect intrinsic motion of the nanocrystals on the molybdenum dichalcogenide surfaces.

**Complementary measurements of shear stress.**

Two experimental configurations in addition to that described in the main text were used to determine shear stress values (Figure S2 and S3). In each, the molybdenum dichalcogenide single crystal was aligned such that one of axes was parallel to the 90º scan direction, the direction of maximum lateral force sensitivity. In the first configuration, the minimum normal load required to move the nanocrystal was measured. Below that critical load the AFM tip will move up and over the crystal; above that load the nanocrystal will slide. To determine this critical value the load on the tip was slowly increased until the nanocrystal just started to move (“normal force to budge” in Figure S3). By measuring the angle of contact between the probe and nanocrystal, the component of the force parallel to the substrate, the lateral force, could be calculated (Figure S2). The contact angle was obtained by imaging the tip’s profile on an atomically-sharp SrTiO$_3$ (305) surface. Since the substrate is sharper than the probe, this has the effect of imaging the tip instead of the surface.$^6$ With this profile the angle between the tip and the nanocrystal at the point of contact was determined, and hence the lateral force exerted from $F_l = F_n \sin \theta$ (see inset of Figure S2). The second configuration was similar to the first in that the normal load was increased until the crystal just moved; however, in this case the peak lateral force was measured by the torsion of the AFM cantilever. Notably, all methods for measuring the requisite sliding force yielded a finite value of the y-intercept, which could be consistent with several phenomena including the edges of the MoO$_3$ nanocrystal interacting differently with the MoX$_2$ surface or buckling of the MoO$_3$ nanocrystal due to the force of the AFM probe.$^7$
Figure S1. Powder x-ray spectra of MoSe₂. 10 separate runs were averaged for this particular spectrum. The vertical lines are the reference data⁸ and are consistent with phase pure MoSe₂.
Figure S2. The lateral force, $F_l$, required to move MoO$_3$ nanocrystals along their preferred direction of travel. The normal force on the tip was slowly increased until the nanocrystal budged. Then the lateral force was calculated from $F_l = F_n \sin \theta$ where $F_n$ is the applied normal load and $\theta$ is the measured angle of contact. The interfacial shear, $\tau$, is 0.52±0.03 MPa.
Figure S3. Lateral force, $F_L$, measured by three different methods. The shear strength, $\tau$, of the MoO$_3$ / MoS$_2$ interface was determined in three distinct configurations from plots of the lateral force, $F_L$ versus MoO$_3$ nanocrystal area. The open diamonds show the data from the main text. The filled circles are the same as Figure S2 and show the value determined by converting the normal force to a lateral force. The final value shows the values for when lateral forces are measured directly by the torsion of the AFM probe cantilever. All three methods yielded similar values and showed an offset in the required force, which could correspond to buckling of the edge of the MoO$_3$ nanocrystal or a stronger interaction between the nanocrystal’s edges and the MoS$_2$ substrate.
Figure S4. Summary of friction decay time constants. The friction decay time constants for MoO$_3$ on MoS$_2$ (blue histogram bars) and MoSe$_2$ (orane histogram bars). The decay rate did not correlate to the dimensions of the MoO$_3$ nanocrystal used for areas from 12,000 – 60,000 nm$^2$.