Supporting Information:

Friction between van der Waals solids during lattice directed sliding

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Materials and Methods.

Growth of MoSe$_2$ crystals.

The molybdenum diselenide crystals were prepared in two steps.[1] 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[2, 3] in a tube furnace following a previously developed method.[2] 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 nanocystals 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 then 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]
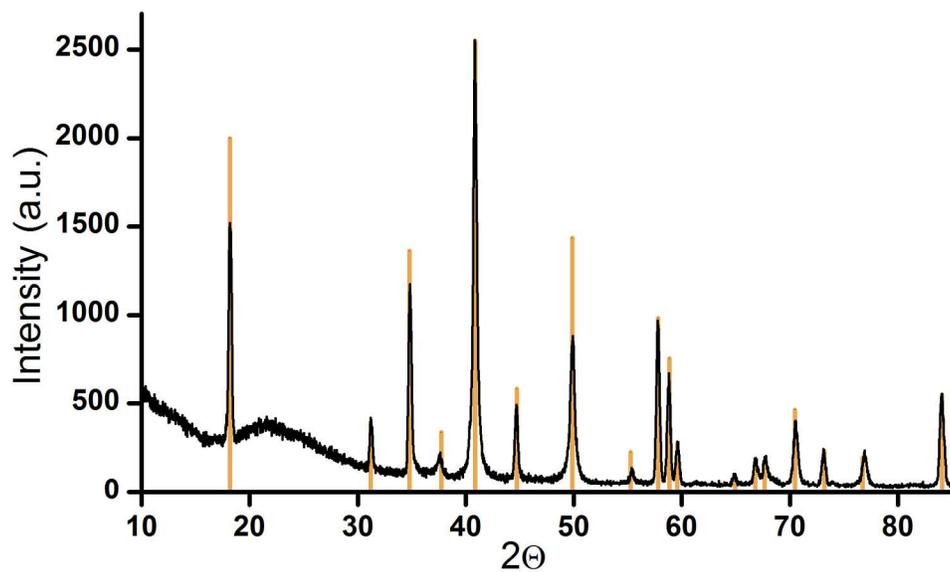
Figure S1. **Powder x-ray spectra of MoSe₂**. 10 separate runs were averaged for this particular spectrum. The vertical lines are the reference data,[8] and are consistent with phase pure MoSe₂.
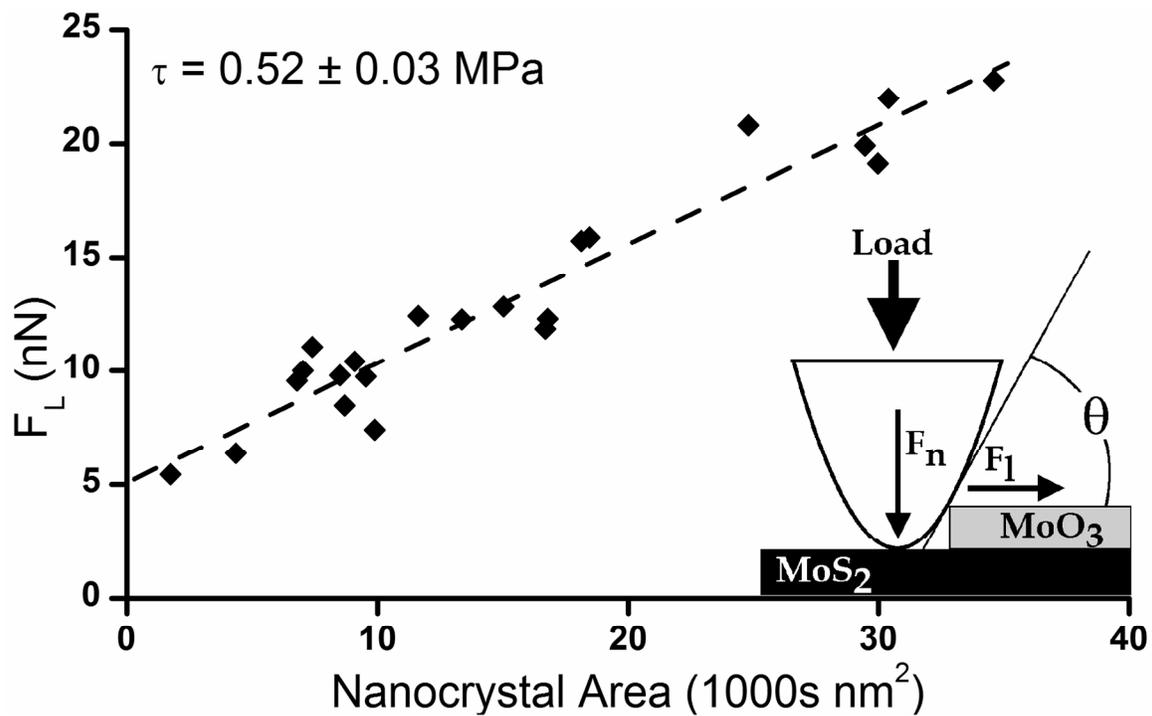
Figure S2. The lateral force, F_l, required to move MoO_3 nanocrystals along their preferred direction of travel. The normal force on the tip was slowly increased until the nanocrystal budged. Then the lateral force was calculated from $F_l = F_n sin\theta$ where F_n is the applied normal load and θ is the measured angle of contact. The interfacial shear, τ, is 0.52±0.03 MPa
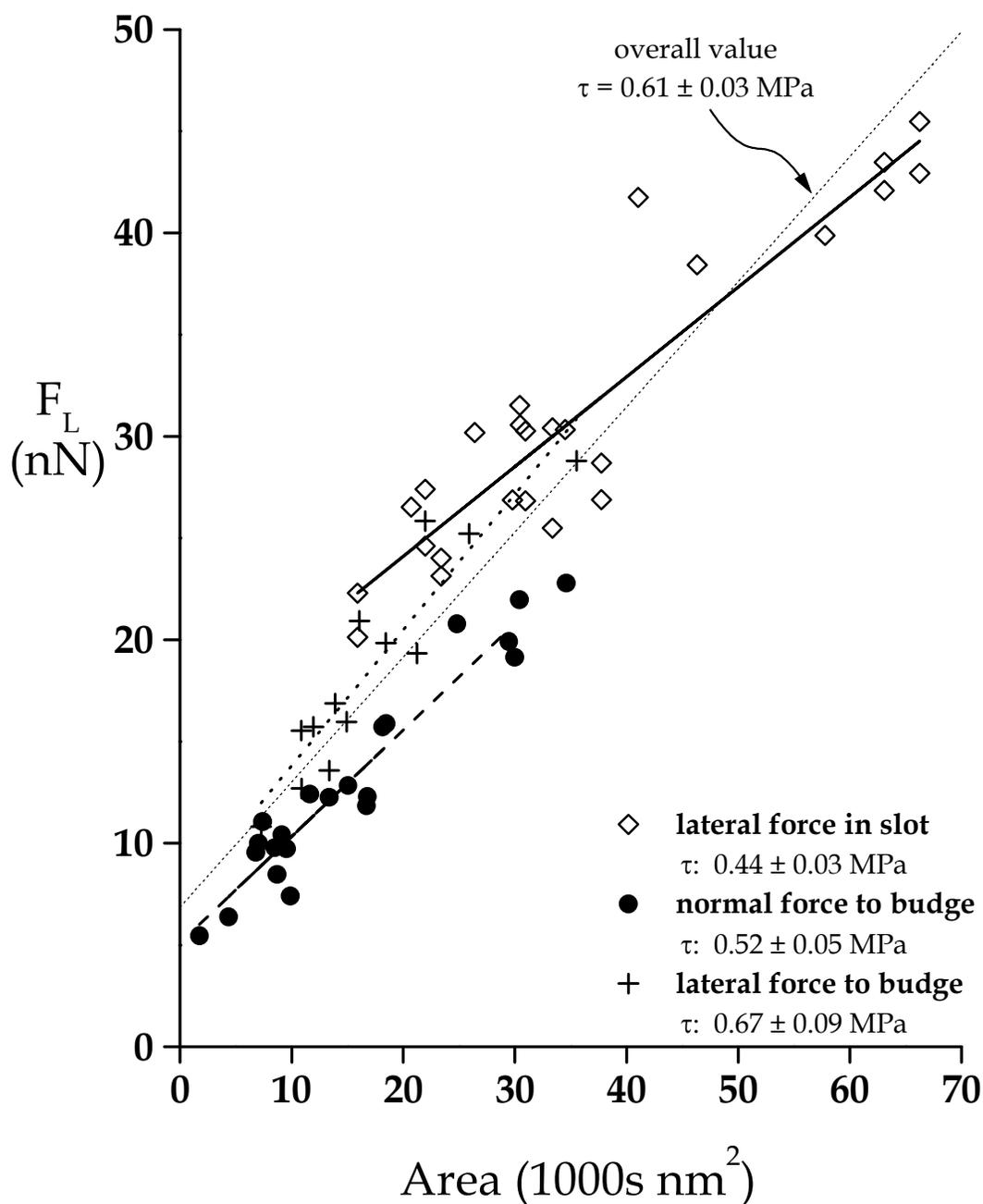
Figure S3. Lateral force, F_l, measured by three different methods. The shear strength, τ, of the MoO$_3$ / MoS$_2$ interface was determined in three distinct configurations from plots of the lateral force, F_L versus MoO$_3$ nanocrystal area. The open diamonds show the data from the main text. The filled 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.
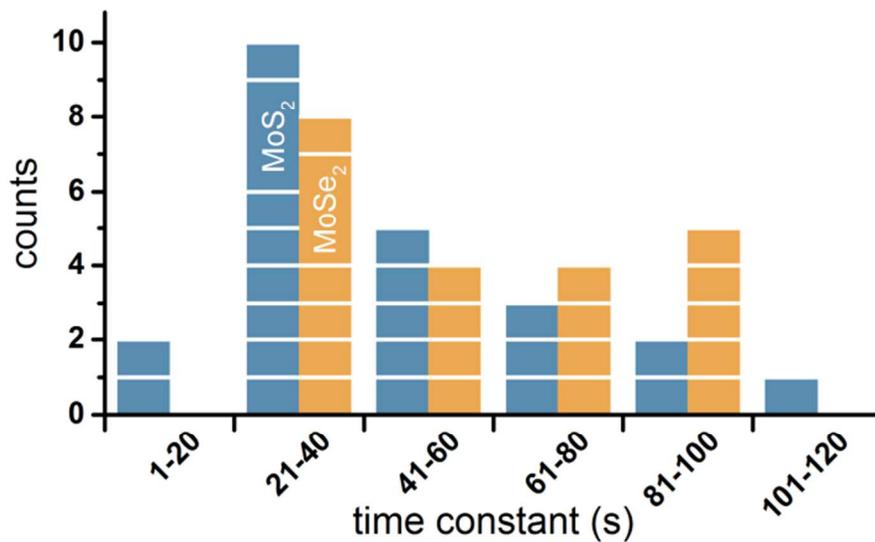
Figure S4. Summary of friction decay time constants. The friction decay time constants for MoO$_3$ on MoS$_2$ (blue histogram bars) and MoSe$_2$ (orane histogram bars). The decay rate did not correlate to the dimensions of the MoO$_3$ nanocrystal used for areas from 12,000 – 60,000 nm^2.

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