



Supporting Information

for

Atom-scale contributions to nanofriction response in transition metal dichalcogenides

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Beilstein J. Nanotechnol. **2026**, *17*, 1330–1347. doi:10.3762/bjnano.17.92

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Structural Information

The geometry of each hetrostructure used in the molecular dynamics simulations with a total number of 360 atoms (144 TM, 36 M, 72 X, 108 Si) is provided in what follows in POSCAR format.

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      9.093929255     13.124079933      16.812799871
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Au/MoSe2/Si

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Au	Mo	Se	Si
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Cartesian

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Au	W	S	Si
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Cartesian

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Au/WSe2/Si

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Au	W	Se	Si
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Cartesian

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Ag/MoS₂/Si

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Ag	Mo	S	Si
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Ag/MoSe2/Si

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Ag	Mo	Se	Si
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Ag/WSe2/Si

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Ag	W	Se	Si
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Frictional Properties

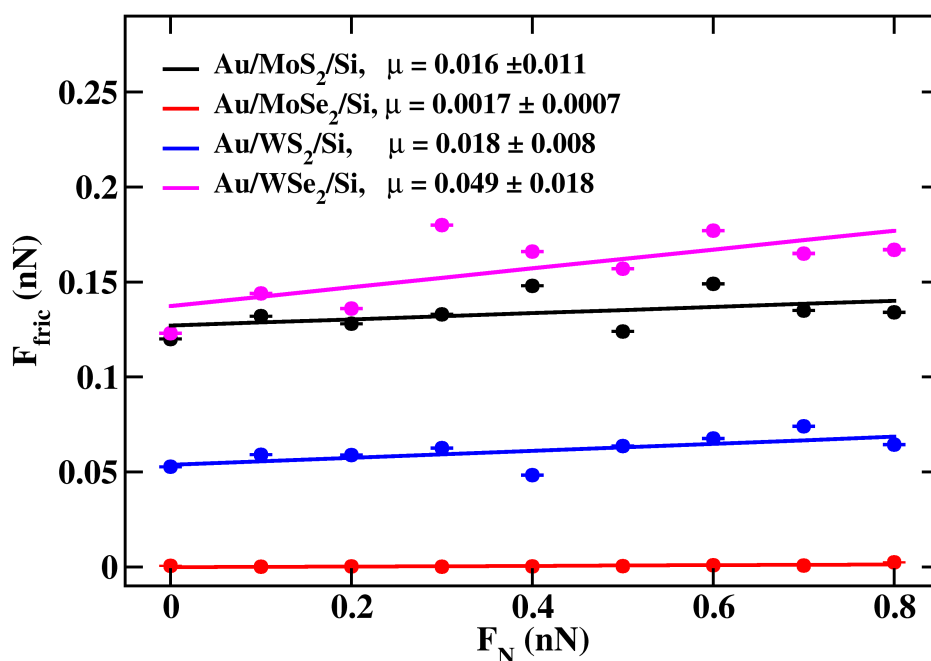


Figure S1: Linear fit of average friction force as a function of load with a tip velocity of 3 m/s.

Figure S1, Figure S2 and Figure S3 show friction force as a function of normal load for four Au/MX₂/Si systems except Au/MoS₂/Si which is presented in the main manuscript, with linear fits used to extract the coefficient of friction μ . Most of the systems exhibit nonmonotonic nature with loads and extraction of μ from a linear fit results in large errors. This same trend is observed in Ag/MX₂/Si systems as shown in Figure S6, Figure S7, Figure S8 and Figure S9.

Table S1, Table S2, Table S3, and Table S4 summarize the fitted values of the asymptotic friction coefficient (μ_∞) and the adhesion force parameter (A), along with their corresponding absolute errors, which are on the order of 10^{-4} . The TM/MoS₂/Si and TM/WSe₂/Si systems exhibit the highest values of the adhesion parameter A among all the considered systems and across all sliding velocities. This

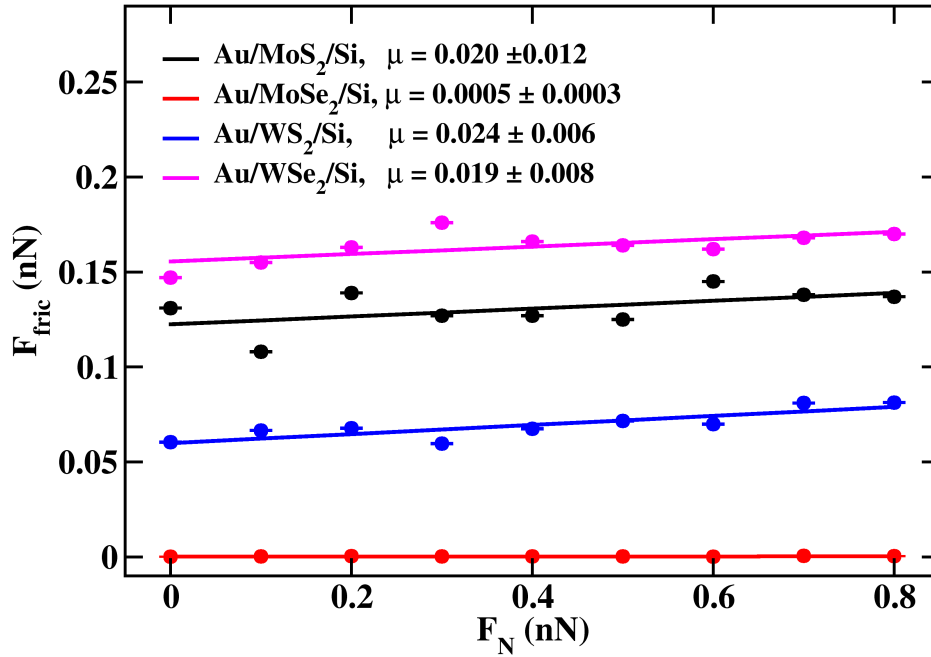


Figure S2: Linear fit of average friction force as a function of load with a tip velocity of 4 m/s.

behaviour indicates enhanced interfacial adhesion, reflecting stronger adhesive interactions between the contacting surfaces relative to the other systems studied.

The friction force profile for the Au/MoSe₂/Si system is shown in Figure S4. In this case, both lateral slip and zigzag behaviour are suppressed and the tip moves along *y*-direction. This is also observed in the Fourier transform spectra of Au/MoSe₂/Si where the second and third peaks, representing the lateral and zig-zag motions, are absent (Figure S5).

Figure S10 shows the spatial Fourier transform of the frictional force in the Ag/MX₂/Si and Au/MX₂/Si systems. In the Au/WSe₂/Si case, the first peak dominates than the Ag/WSe₂/Si system which leads to higher frictional force.

Figure S11 shows the Fourier transform of friction force as a function of different velocity for Au/MoS₂/Si system. As we can see that the peak positions are identical for all the velocity confirming that the lateral slip and zig-zag motions are intrinsic features of the potential energy surface topology rather than artifacts of increasing sliding velocity.

Figure S12 shows the calculated μ' over each load as a function load for Au/MX₂/Si system with tip velocity of 2 m/s. The plot shows the decreasing trend of μ' by increasing the load, indicating a departure from the classical behaviour at the nanoscale.

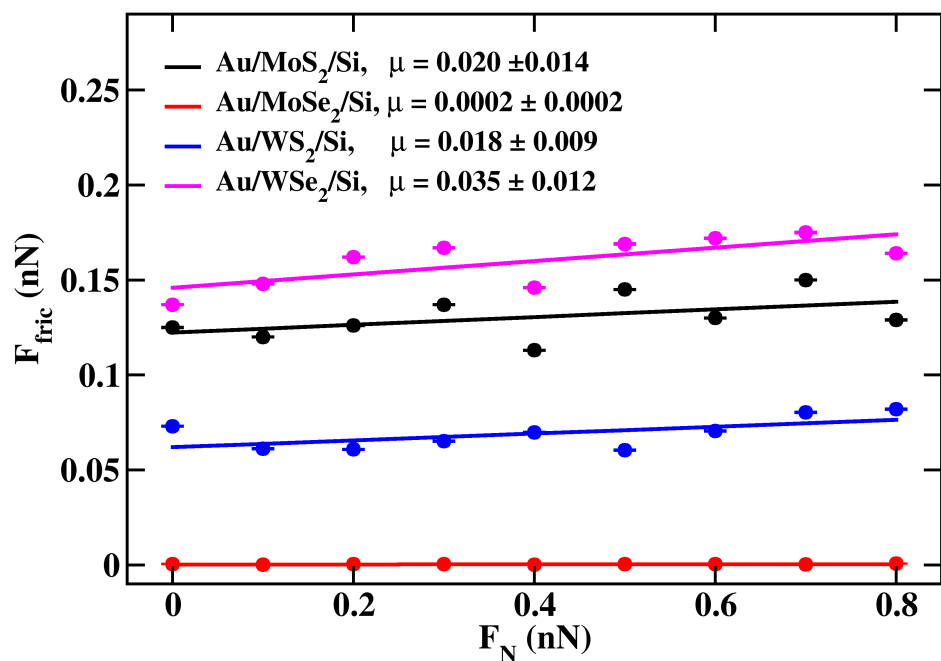


Figure S3: Linear fit of average friction force as a function of load with a tip velocity of 5 m/s.

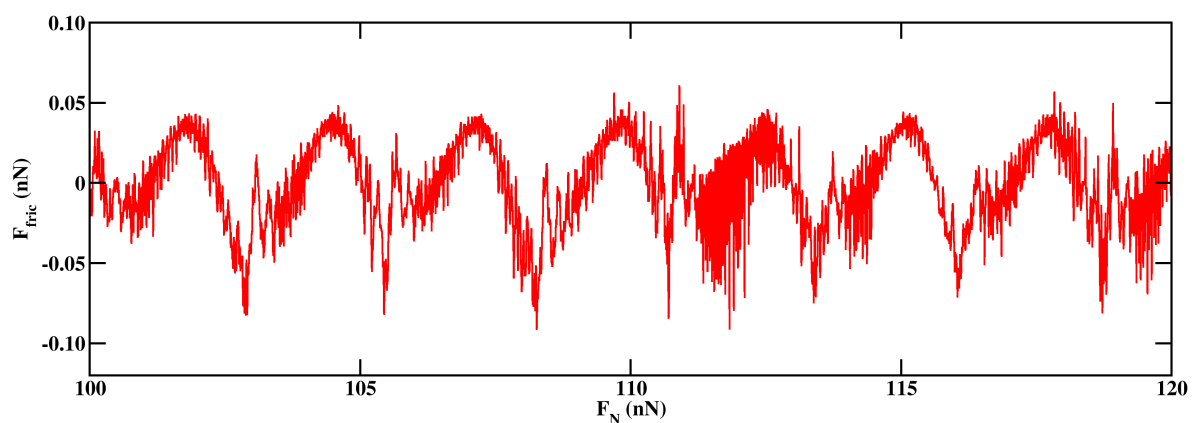


Figure S4: Friction force profile of Au/MoSe₂/Si where tip moving with velocity 2 m/s for load of 0 nN.

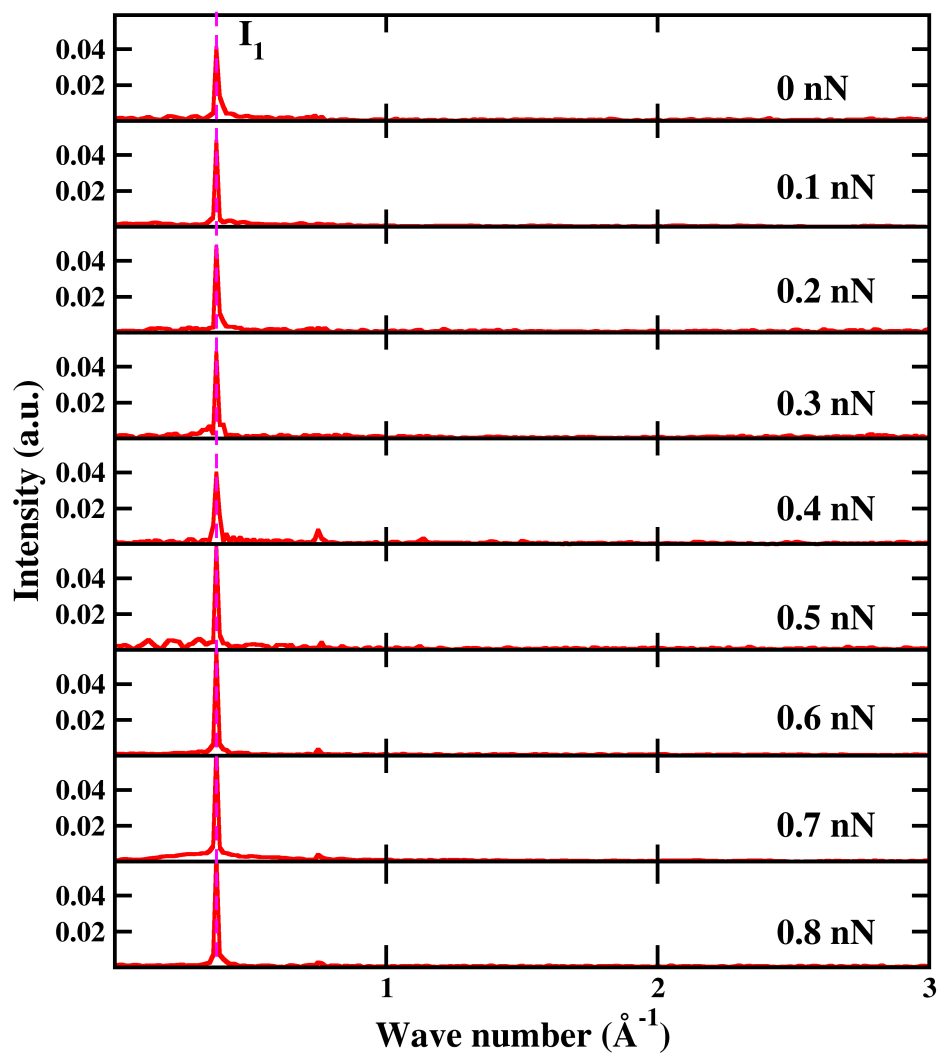


Figure S5: Spatial Fourier transform of Au/MoSe₂/Si with varying loads at velocity 2 m/s.

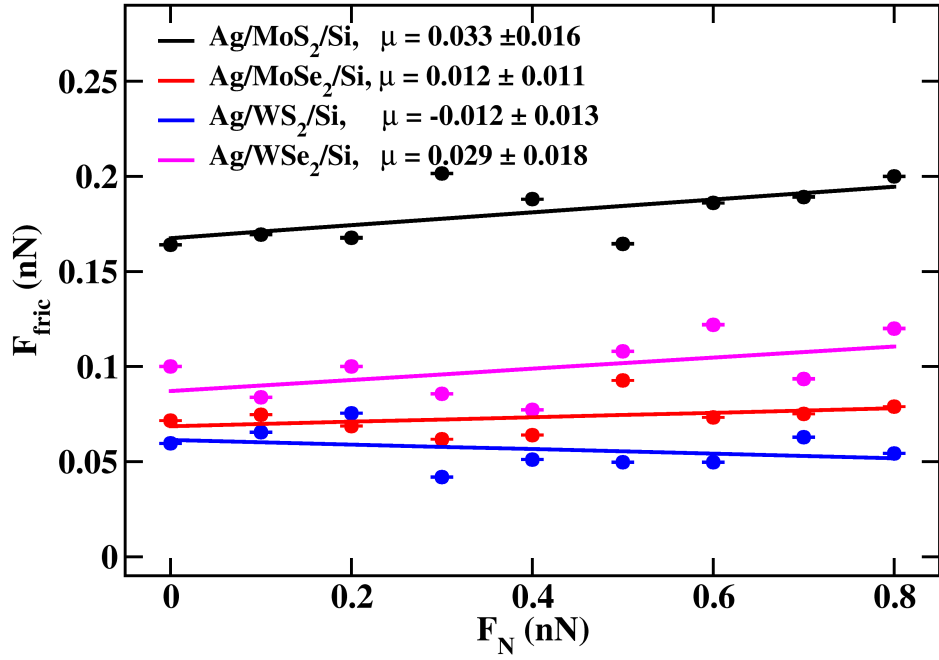


Figure S6: μ extracted from a linear fit on the average friction force as a function of load with a tip velocity of 2 m/s.

Table S1: Fitting parameters extracted from simulations with tip velocity equal to 2 m/s.

Monolayer	Substrate	$\mu_{\infty}(10^{-4})$	$A(10^{-4} \text{ nN})$
MoS ₂	Au	415 ± 4	997 ± 2
MoSe ₂		4.0 ± 0.4	5.2 ± 0.2
WS ₂		229 ± 4	569 ± 2
WSe ₂		894 ± 4	1046 ± 2
MoS ₂	Ag	292 ± 5	1707 ± 3
MoSe ₂		87 ± 3	704 ± 2
WS ₂		-63 ± 4	571 ± 2
WSe ₂		555 ± 4	717 ± 2

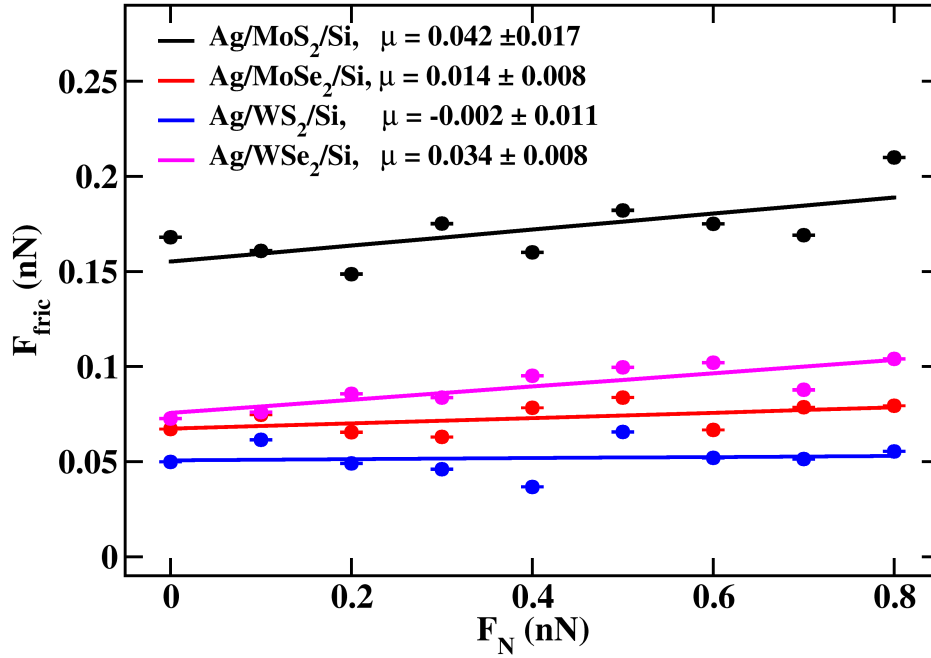


Figure S7: μ extracted from a linear fit function on the average friction force as a function of load with a tip velocity of 3 m/s.

Table S2: Fitting parameters extracted from simulations with tip velocity equal to 3 m/s.

Monolayer	Substrate	$\mu_{\infty}(10^{-4})$	$A(10^{-4} \text{ nN})$
MoS ₂	Au	79 ± 4	1317 ± 2
MoSe ₂		20.1 ± 0.4	-3.3 ± 0.2
WS ₂		167 ± 4	547 ± 2
WSe ₂		367 ± 5	1447 ± 3
MoS ₂	Ag	565 ± 5	1474 ± 3
MoSe ₂		109.2 ± 0.3	688 ± 2
WS ₂		20 ± 4	506 ± 2
WSe ₂		336 ± 3	770 ± 2

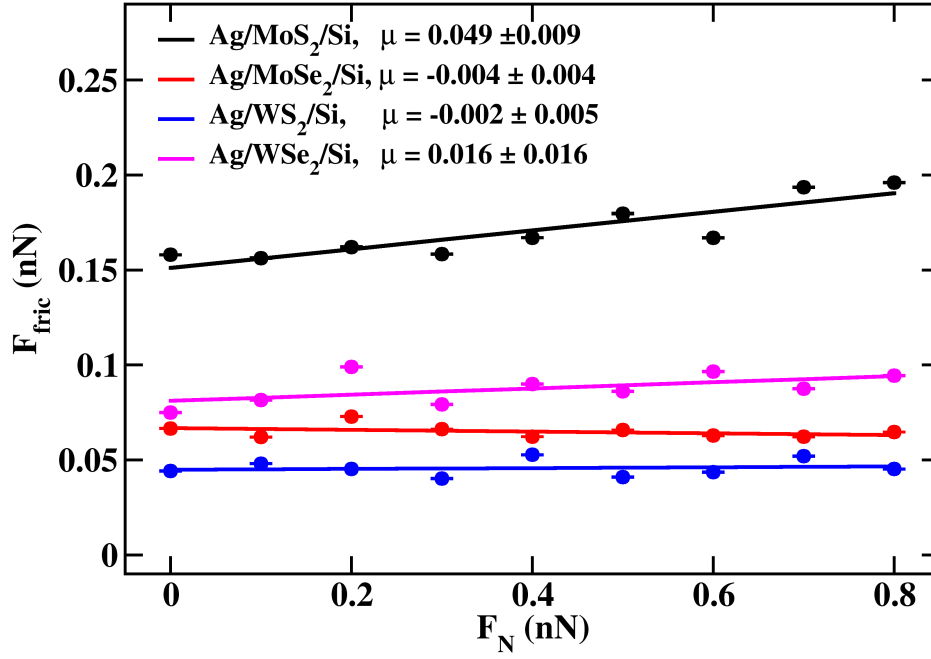


Figure S8: μ extracted from a linear fit function on the average friction force as a function of load with a tip velocity of 4 m/s.

Table S3: Fitting parameters extracted from simulations with tip velocity equal to 4 m/s.

Monolayer	Substrate	$\mu_{\infty}(10^{-4})$	$A(10^{-4} \text{ nN})$
MoS ₂	Au	323 ± 4	1164 ± 2
MoSe ₂		4.6 ± 0.4	0.9 ± 0.2
WS ₂		237 ± 5	599 ± 2
WSe ₂		113 ± 5	1603 ± 2
MoS ₂	Ag	563 ± 5	1464 ± 3
MoSe ₂		-12 ± 2	693 ± 2
WS ₂		7 ± 3	457 ± 2
WSe ₂		152 ± 3	818 ± 2

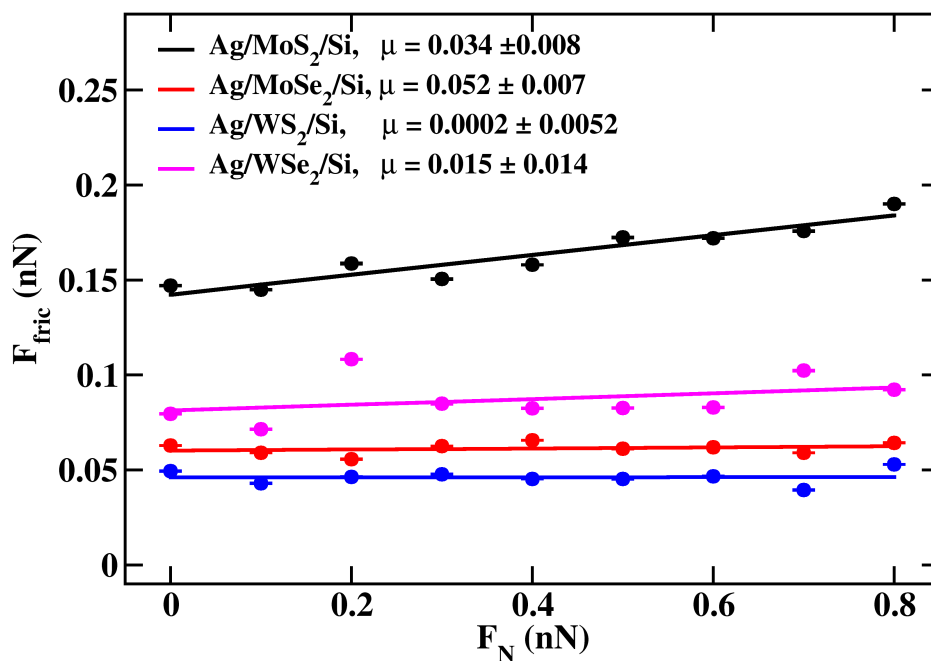


Figure S9: μ extracted from a linear fit function on the average friction force as a function of load with a tip velocity of 5 m/s. Ag/WS₂/Si and Ag/WSe₂/Si systems shows comparatively higher errors than other systems.

Table S4: Fitting parameters extracted from simulations with tip velocity equal to 5 m/s.

Monolayer	Substrate	$\mu_{\infty}(10^{-4})$	$A(10^{-4} \text{ nN})$
MoS ₂	Au	224 ± 4	1208 ± 2
MoSe ₂		2.9 ± 0.4	2.0 ± 0.2
WS ₂		229 ± 4	569 ± 2
WSe ₂		262 ± 5	1510 ± 3
MoS ₂	Ag	608 ± 5	1373 ± 3
MoSe ₂		44 ± 3	597 ± 2
WS ₂		50 ± 4	436 ± 2
WSe ₂		220 ± 4	758 ± 2

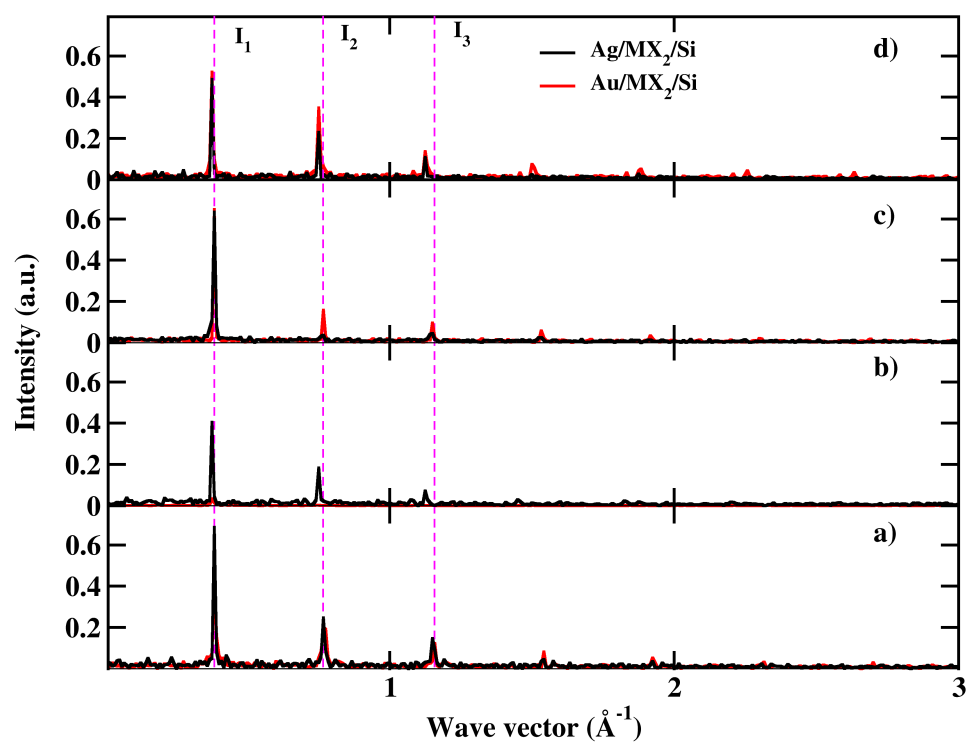


Figure S10: Spatial Fourier transform of friction force with monolayer a) MoS₂, b) MoSe₂, c) WS₂ and d) WSe₂ at different substrates.

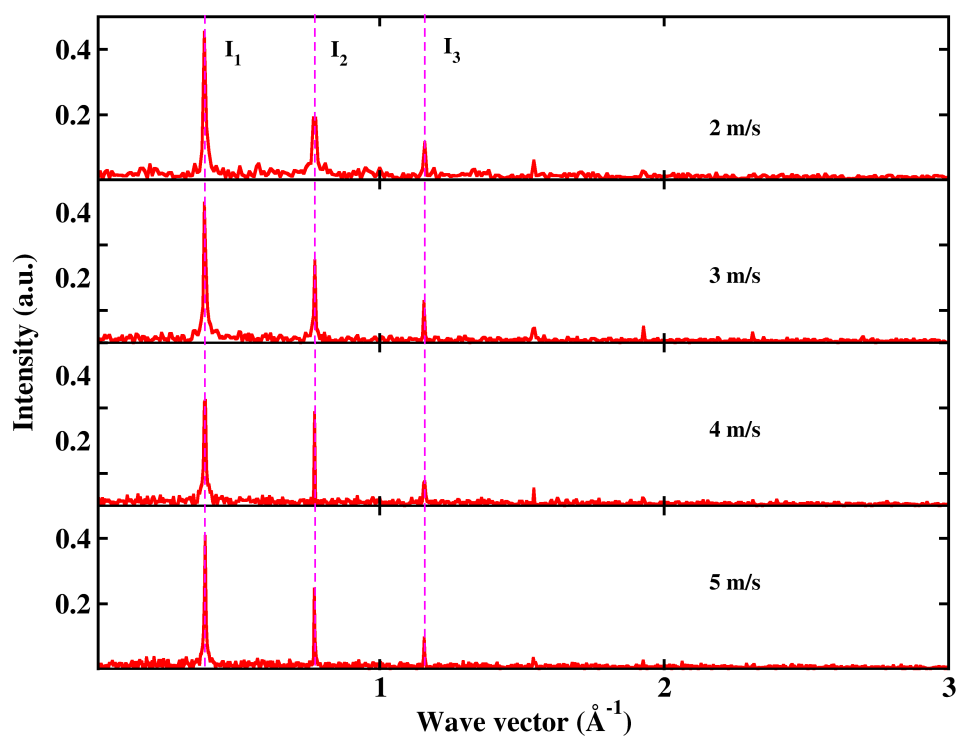


Figure S11: Spatial Fourier transform of friction force of Au/MoS₂/Si under the load of 0.4 nN with different velocity

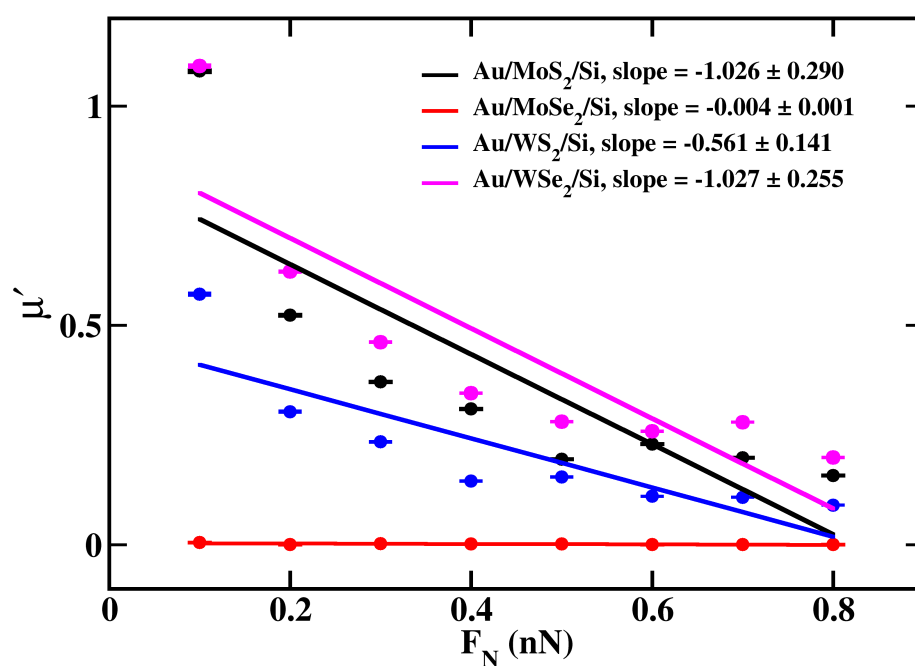


Figure S12: Calculated μ' over each load plotted as a function of load for the Au/MX₂/Si system with tip velocity of 2 m/s.