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# 1 **Microscopic contributions to the deviation from Amontons friction** 2 **law**

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## 9 **Abstract**

10 We investigate the nanoscale friction behaviour of  $\text{MX}_2$  monolayers ( $\text{M} = \text{Mo}, \text{W}; \text{X} = \text{S}, \text{Se}$ ) on  
11  $\text{Au}(111)$  and  $\text{Ag}(111)$  substrates with a silicon tip using classical molecular dynamics simulations  
12 with machine-learning-based force fields. This approach enables an accurate description of tip-  
13 surface interactions and friction mechanisms at the atomic scale. We observe a pronounced non-  
14 monotonic dependence of the friction force on the applied normal load, indicating a breakdown of  
15 Amontons's law at the nanoscale. Analysis of lateral force signals and their spatial Fourier trans-  
16 forms reveals the coexistence of multiple sliding modes, including longitudinal sliding, lateral  
17 slip, and zig-zag motions. We show that the overall friction response is governed by the relative  
18 contributions of these motions. While the qualitative features of friction are largely substrate-  
19 independent, both the magnitude of friction and the balance between sliding modes depend sen-  
20 sitively on the substrate-monolayer combination. In particular,  $\text{Au}/\text{MoSe}_2/\text{Si}$  exhibits significantly  
21 reduced friction due to suppression of lateral slip motion. Our results indicate that the method is  
22 broadly applicable for probing nanoscale friction in related heterostructures.

## 23 **Keywords**

24 density functional theory; deep neural network; fourier transform; machine learning force field;  
25 molecular dynamics; nanofriction; nonmonotonic; transition metal dichalcogenides

## 26 **Introduction**

27 Friction arises whenever two contacting surfaces move relative to each other, leading to energy  
28 dissipation and material wear in mechanical systems. Unfortunately, liquid lubricants are ineffec-  
29 tive in reducing friction under extreme conditions, such as high normal load, high temperature and  
30 ultra-high vacuum [1]. In recent years, two-dimensional layered materials such as graphene, boron  
31 nitride and transition metal dichalcogenides (TMDs) have emerged as promising solid lubricants  
32 in achieving superlubricity on rough and worn surfaces in engineering applications [2-7]. Among  
33 them, TMDs stand out for their layered structure and weak van der Waals bonding between the lay-  
34 ers, [8,9] which facilitates interlayer shear, resulting in exceptionally low frictional properties in  
35 vacuum. However, the dynamic behaviour of the TMD layers with atomic detail during sliding is  
36 subject of continuous study, due to the complex phenomena occurring concurrently in tribologi-  
37 cal conditions. The most used and simple theoretical model of nanoscale friction is the Prandtl-  
38 Tomlinson model [10,11]. However, atomistic observation of lateral slips, dynamic deformation  
39 and thermal fluctuations is not well captured by this model; indeed, advanced analysis based on  
40 the Fourier Transform of the friction force helps to quantify these lateral movements properties  
41 [12,13].

42 The interaction between TMD layers and metallic substrates, together with the influence of a scan-  
43 ning probe tip, determines the overall nanoscale friction response. The atomically flat nature of  
44 metallic substrates helps to suppress out-of-plane fluctuations of the TMD layer, resulting in a  
45 more stable sliding interface and more reliable investigation of frictional mechanisms [14]. First-  
46 principles based calculations have provided valuable insights into the electronic and structural  
47 properties of these interfaces [15,16]; however, their application to large-scale friction simulations  
48 is limited due to high computational cost. Compared with first-principles calculations, classical

49 molecular dynamics (MD) is a technique capable of simulating dynamic and kinetic problems such  
50 as sliding processes and frictional behavior at larger scale and reduced computational load [17-19].  
51 MD simulations are often employed using empirical interatomic potentials, such as ReaxFF [20,21]  
52 and reactive empirical bond-order (REBO) potentials [19,22,23]; nevertheless, the parametrisation  
53 of such potentials is challenging and time demanding. Fortunately, with the development of  
54 machine learning (ML) in material science, this challenge has been well addressed [24]. Various  
55 machine learning force fields (MLFFs) have been developed, including the Gaussian approximation  
56 potential (GAP) [25], TensorMol [26], the neuroevolution machine learning potential (NEP) [27],  
57 and the deep neural network potential (NNP) [28]. Notably, neural networks possess the unique  
58 ability to theoretically represent unknown multidimensional real-valued functions with arbitrary  
59 precision by selecting appropriate network models. Recent research demonstrates that the NNP  
60 model matches the accuracy of quantum mechanics for both finite and extended systems, showcasing  
61 its size extensive nature [29,30]. At present, MLFFs have been successfully applied in diverse  
62 materials, including  $\text{TiO}_2$  [31],  $\text{Ga}_2\text{O}_3$  [32] and TMDs [29,33,34], making the NNP with the broad-  
63 est range of applications and the most promising prospects in current research.

64 In this work, we study the friction response of  $\text{MX}_2$  monolayers ( $\text{M} = \text{Mo}, \text{W}$  and  $\text{X} = \text{S}, \text{Se}$ ) de-  
65 posited on Au(111) and Ag(111) substrates by means of classical MD simulations at different loads  
66 and velocities of a scanning tip, employing MLFFs developed for this purpose. Our Fourier anal-  
67 ysis of the friction signal allows us to quantify and characterise the nanoscale contributions to the  
68 friction response. In particular, we examine the microscopic phenomena responsible for the break-  
69 down of Amontons's law as reflected in the mean friction force; this results in a nonmonotonic  
70 friction-load relationship and contributes to significant uncertainty in the extracted coefficient of  
71 friction from a linear fit. Significant contribution to such deviation originates from the tip's lateral  
72 motion, which also reduces the average friction force, except in the Au/MoSe<sub>2</sub>/Si system, as the  
73 lateral motion is missing from the force profile.

74 The article is organized as follows: we describe the computational methods in section Computa-

75 tional Details, while the results of the friction simulations of TMDs on different substrates are pre-  
76 sented in section Results and Discussion.

## 77 **Computational Details**

78 Density functional theory (DFT) based calculations for the structural optimisation of TM/MX<sub>2</sub>/Si  
79 systems (TM = Au, Ag; M = Mo, W and X = S, Se) are performed using the Vienna ab-initio sim-  
80 ulation package (VASP) [35]. The interactions between valence electrons and ionic cores are de-  
81 scribed using the projector augmented-wave (PAW) method [36]. The generalized gradient approx-  
82 imation (GGA) in the Perdew–Burke–Ernzerhof (PBE) form is employed to treat the exchange-  
83 correlation energy [37]. The electronic self-consistent field calculations are converged to within  
84 10<sup>-8</sup> eV, and the atomic positions are relaxed until the residual forces in each atom are less than  
85 10<sup>-2</sup> eV/Å. A 7 × 7 × 1 Monkhorst–Pack *k*-point mesh [38] is used for Brillouin zone sampling,  
86 and the plane-wave basis set is truncated at a kinetic energy cutoff of 500 eV. To accurately capture  
87 long-range dispersion interactions, the Grimme DFT-D3 correction [39] is incorporated to account  
88 for van der Waals forces between the layers. The ML potential is trained using the DeepMD-kit  
89 package with the DeepPot–SE (Deep Potential-Smooth Edition) model [40,41]. The model in-  
90 cludes an embedding network and a fitting network. The sizes of these networks are set to (25, 50,  
91 100) and (240, 240, 240), respectively. The cutoff radius is set to 8.0 Å and the descriptors decay  
92 smoothly from 0.5 to 8.0 Å. The initial learning rate is set to 0.001 at the beginning of the train-  
93 ing process to achieve a final value of 10<sup>-8</sup>. The total number of training batches are 10<sup>5</sup> for the  
94 training in the initial iterations and the active learning process. To automate the active learning pro-  
95 cess, the DP-GEN workflow is used [42]. All molecular dynamics simulations are performed using  
96 the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [43] package through  
97 the trained ML potentials. The optimised structure of each system is equilibrated at 300 K for 6  
98 ns with a time step of 1 fs by means of a Nosé–Hoover chain with three thermostats. To construct  
99 TM/MX<sub>2</sub>/Si interfaces, the TM substrate is cleaved from bulk Au and Ag as reported in Ref. 44  
100 and subsequently optimized within the DFT framework. Next, a TMD monolayer is placed on top

101 of the TM substrate in such a way to minimize the lattice mismatch of the resulting heterostructure.  
 102 A 30 Å vacuum slab orthogonal to the  $c$ -direction is added on top of the geometry to prevent any  
 103 interaction of the periodic replicas along the same direction; the geometry of the heterostructure is  
 104 then relaxed. During the optimization, the bottom layer of the TM substrate is fixed to better mimic  
 105 the experimental conditions; finally, a Si (111) tip is placed above the heterostructure and the whole  
 106 system is relaxed. In this last geometric optimization, the bottom TM layer kept fixed, while the  
 107 Si tip is constrained to relax only along the  $c$ -direction. An example of model geometry of the full  
 108 system is reported in Figure 1.

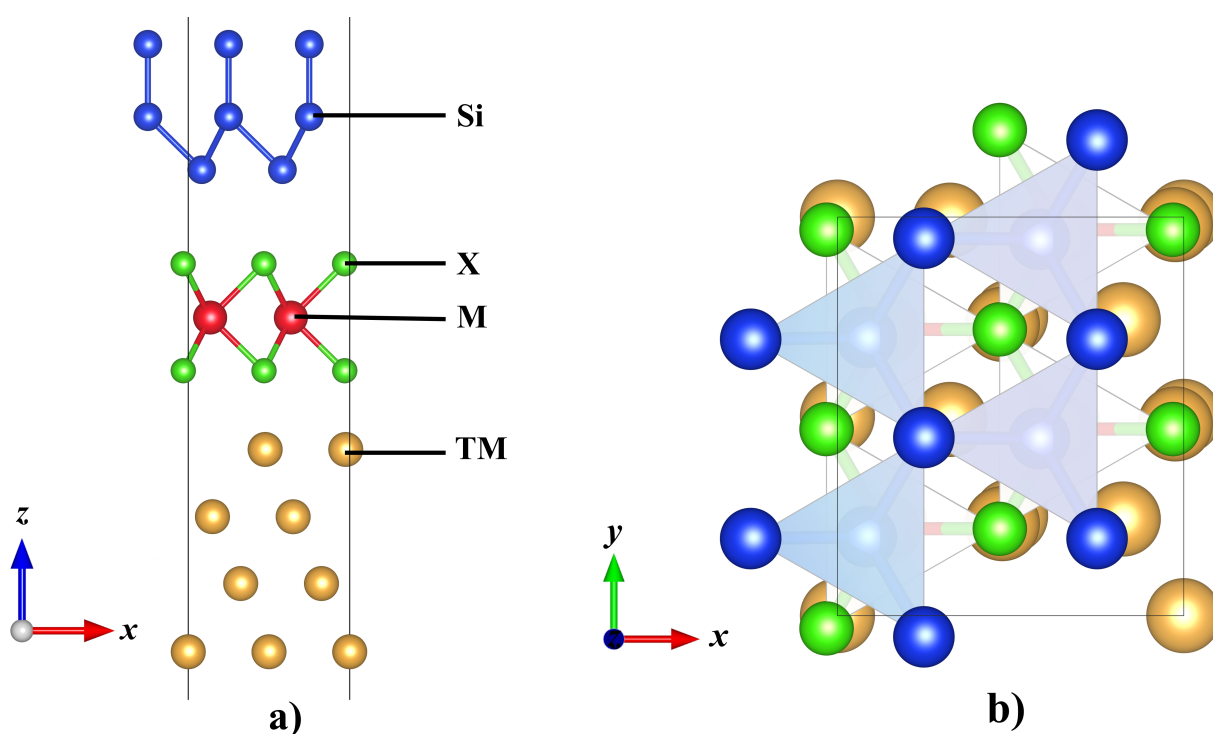


Figure 1: Initial setup to generate structure model of TM/MX<sub>2</sub>/Si. a) Front view and b) top view. The thin black lines indicating the unit cell.

## 109 Training of force field

110 After optimising the full Au/MX<sub>2</sub>/Si configuration, we proceed to construct the machine-learning  
 111 force field (MLFF) for the system. Once we obtain the optimised structure, the training data for the  
 112 MLFF are generated by perturbing the atom positions and cell size by at least 0.01 Å and 0.03 Å,  
 113 respectively. A total of ~400 configurations are generated to train the MLFF of all the systems and

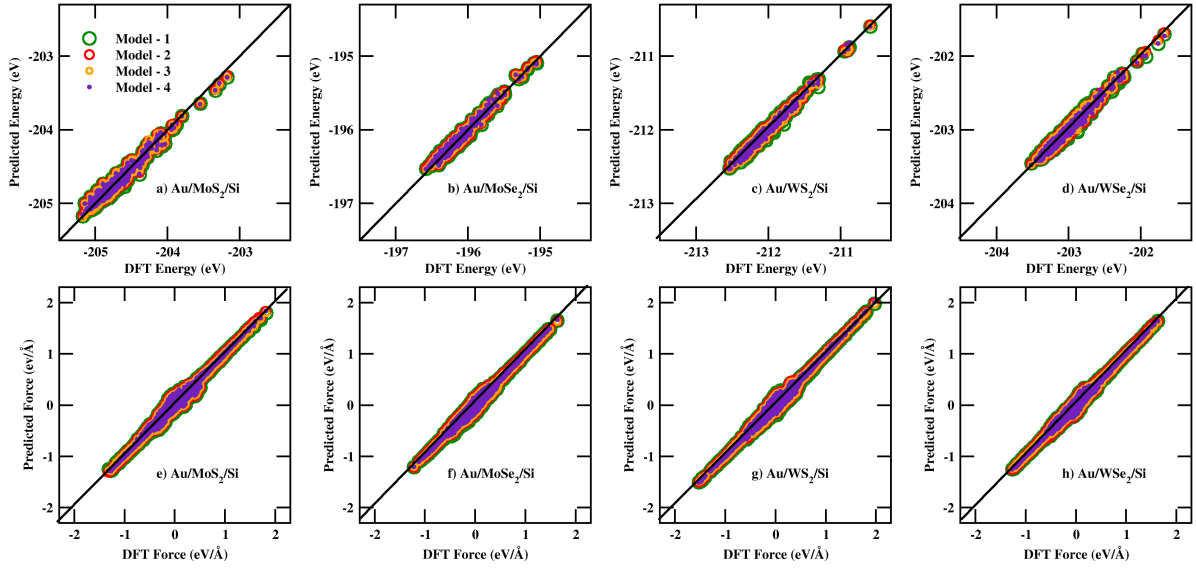


Figure 2: Energy (a–d) and force (e–h) comparisons between predicted and DFT results for Au/MX<sub>2</sub>/Si systems.

114 80 configurations are taken for the validation during the active learning process. For all configura-  
 115 tions, energy and forces are calculated with the selected DFT setup and a single point calculation,  
 116 that is at fixed atom geometry and lattice parameters. To effectively improve the MLFF, additional  
 117 data are generated using an active learning approach as proposed by Zhang et al. [42]. At each iter-  
 118 ation, four models are trained using the same input setup but starting with different random seeds.  
 119 To explore new configurations to enrich the dataset, deep potential molecular dynamics (DPMD)  
 120 simulations are performed with the initial models as reference, collecting the system’s atomic con-  
 121 figurations every 10 fs along the trajectories. DPMD simulations are performed for a duration of  
 122 500 ps. During the DPMD simulations, the force acting on each atoms in all configurations are  
 123 evaluated by all four models. The maximum standard deviation of the atomic forces  $\sigma_{max}$  is evalu-  
 124 ated as a criterion for the convergence of neural network training

$$125 \quad \sigma_{max} = \max_i \sqrt{\langle \|\mathbf{f}_i - \langle \mathbf{f}_i \rangle\|^2 \rangle} \quad (1)$$

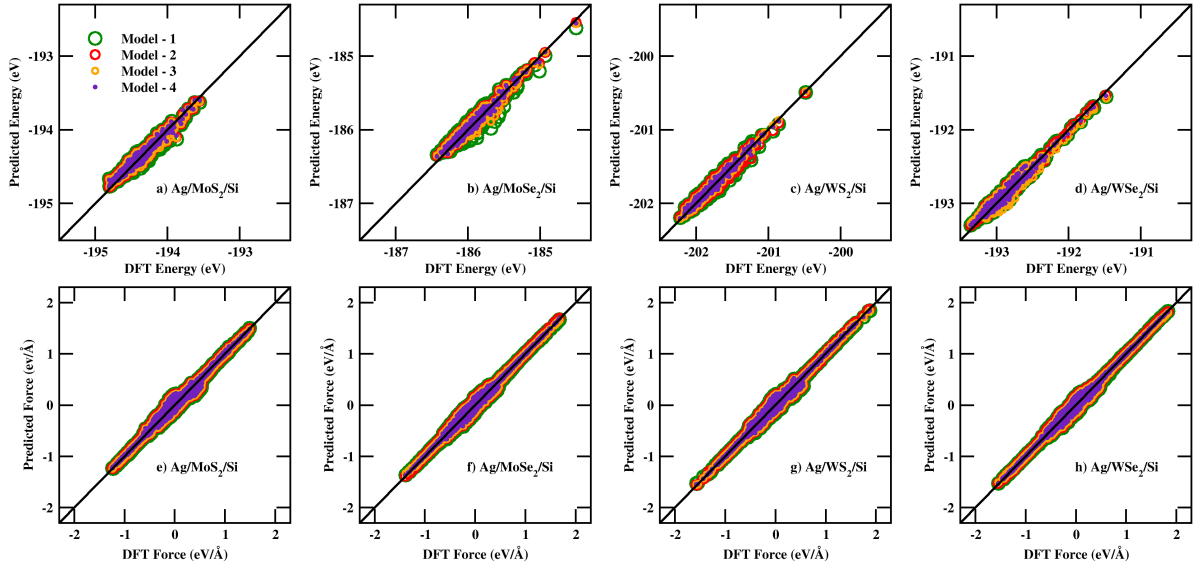


Figure 3: Energy (a–d) and force (e–h) comparisons between predicted and DFT results for Ag/MX<sub>2</sub>/Si systems.

126 where  $f_i$  is the force acting on  $i^{th}$  atom and  $\langle f_i \rangle$  is the average value taken from four models. A  
 127 structure is considered a candidate if the maximum standard deviation falls within the range of 0.05  
 128  $< \sigma_{max} < 0.15$ ; while structures with  $\sigma_{max} < 0.05$  are considered correct, those with  $\sigma_{max} > 0.15$   
 129 can be highly distorted configurations and are discarded. The selected structures are then subjected  
 130 to DFT calculations to obtain the corresponding energies and forces, and included in the dataset.  
 131 The training with four new models is repeated until the root mean square errors (RMSE) value is  
 132 reduced. The RMSE values of energy and forces for all the systems are presented in [Table 1](#). All  
 133 the final models have RMSE of energy and forces ranging from 0.92 to 1.16 meV/atom and 0.019  
 134 to 0.021 eV/Å, respectively. This range of RMSE values indicates that the MLFFs model is prop-  
 135 erly trained with values for the errors typical of surface and interface systems [33,34]. To validate  
 136 our MLFFs, we optimise the geometry of the heterostructures by using the trained MLFFs; we then  
 137 compute the RDF scalar products with the DFT optimised structure using the MAISE package [45],  
 138 in order to check if the two methods yield the same optimised geometry. This analysis shows that  
 139 the structures optimised with the two methods are almost identical, then confirming the reliabil-  
 140 ity of the MLFFs parametrisation. The same procedure is applied to the case of the Ag substrate.

141 The predicted energies and forces obtained from the machine-learning force field show very good  
 142 agreement with the reference DFT results (Figure 2 and Figure 3). This agreement further validates  
 143 the accuracy of the trained models.

Table 1: The RMSE value of all the trained models.

Monolayer	Substrate	Energy RMSE (meV/atoms)	Force RMSE (eV/Å)	RDF Scalar Product
MoS <sub>2</sub>	Au	1.5796	0.0213	0.904
MoSe <sub>2</sub>		1.0288	0.0204	0.892
WS <sub>2</sub>		0.9198	0.0191	0.946
WSe <sub>2</sub>		1.1787	0.0193	0.941
MoS <sub>2</sub>	Ag	1.33	0.0205	0.879
MoSe <sub>2</sub>		0.941	0.0196	0.878
WS <sub>2</sub>		0.989	0.0203	0.947
WSe <sub>2</sub>		1.298	0.0206	0.920

## 144 MD Setup for Friction Analysis

145 For the MD simulations, a  $3 \times 3 \times 1$  supercell of the optimised heterostructure was constructed  
 146 in order to minimise the effects of the system's finite size and to provide a sufficiently large con-  
 147 tact area for the sliding process (see Figure 4). The resulting simulation cell contains a total of 360  
 148 atoms (See the POSCAR data in Supporting Information File 1, Section I). The bottom layer of the  
 149 TM substrate is kept fixed throughout the simulation. This constraint prevents artificial translation  
 150 of the entire system and represents the bulk support typically present in experimental setups; the  
 151 remaining atoms are allowed to evolve dynamically during the simulation in the NVT ensemble at a  
 152 temperature equal to 300 K. To model the sliding process, all Si layers are treated as a rigid body to  
 153 simulate a rigid scanning tip. The tip is moved along the  $x$ -direction at constant velocities of 2, 3, 4  
 154 and 5 m/s, while there is no constraint on the motion along the  $y$ - and  $z$ -directions. During sliding,  
 155 normal loads of 0.0, 0.4, 0.6 and 0.8 nN are applied to the tip along the  $z$ -direction. Throughout

156 the simulation, the instantaneous forces acting on the tip and the atomic trajectories are recorded at  
 157 every 1 fs time step.

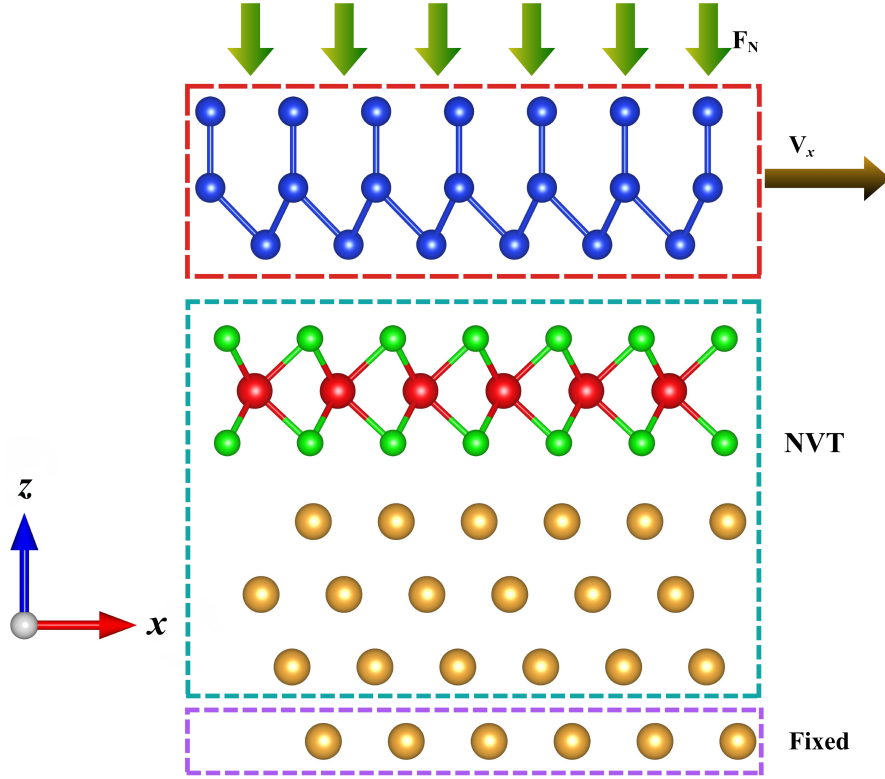


Figure 4: MD simulations setup.

## 158 Results and Discussion

### 159 Friction Force Analysis

160 We begin our study of the friction response by considering the friction force profile using 1 ns tra-  
 161 jectories under different applied normal loads, as shown in Figure 5. Interestingly, the overall shape  
 162 of the friction force profile remains nearly identical across all applied loads, indicating negligible  
 163 load dependence of the profile shape. We then calculate the average friction force acting on the Si  
 164 tip along the  $x$ -direction  $F_{fric}$ , by averaging over all the snapshots of the trajectory:

$$165 \quad F_{fric} = -\frac{1}{N_s} \sum_{i=1}^{N_s} \sum_{j=1}^{N_{tip}} F_{xj}(t_i) \quad (2)$$

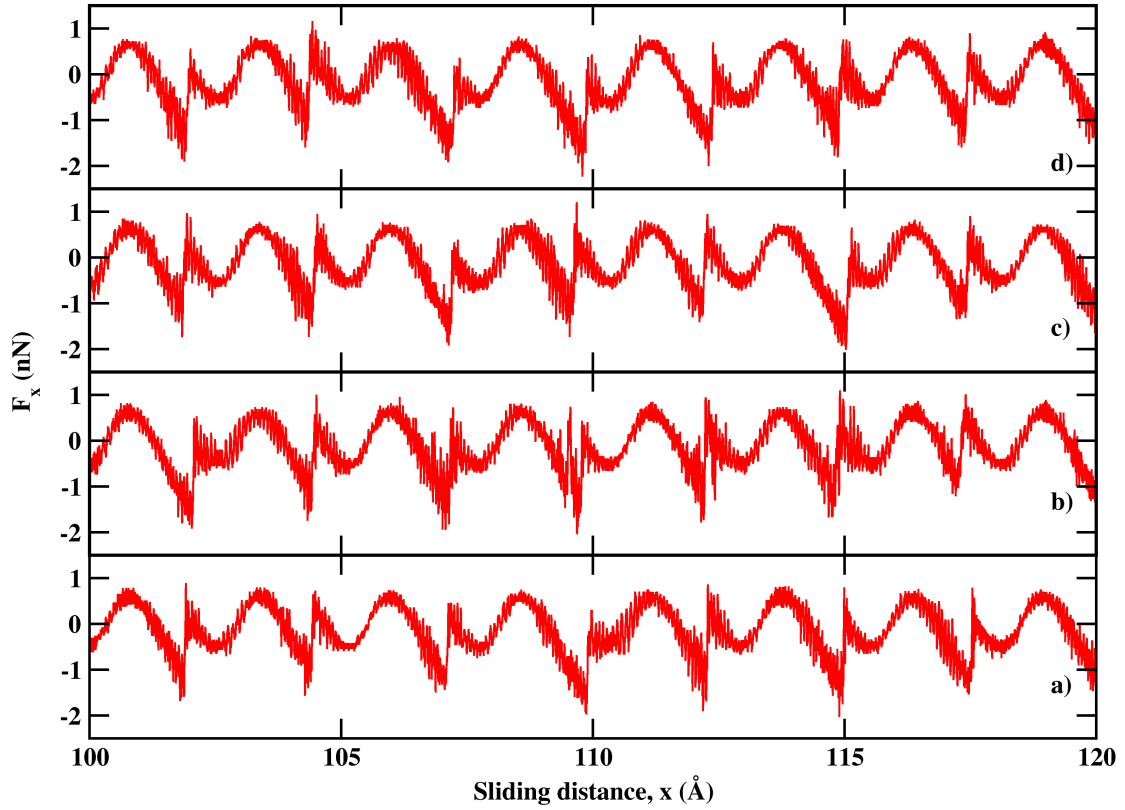


Figure 5: Friction force profile of Au/MoS<sub>2</sub>/Si with tip moving at 2 m/s for under loads of a) 0 nN, b) 0.4 nN, c) 0.6 nN and d) 0.8 nN.

166 where  $N_s$  is the number of time steps,  $N_{tip}$  is the number of atoms in the tip,  $F_{xj}$  is the force acting  
 167 on the  $j$ -th atom of the tip at the  $t_i$  time step along the sliding  $x$ -direction, and the negative sign  
 168 accounts for the fact that friction opposes the direction of motion. The coefficient of friction  $\mu$  is  
 169 then calculated according to Amontons's law with the following linearly fitting function:

$$170 \quad F_{fric} = \mu F_N + F_{fric}^0 \quad (3)$$

171 where  $F_N$  is the applied normal load and  $F_{fric}^0$  is the friction in the absence of load. The calcu-  
 172 lated average friction force at varying loads with different velocities for all the systems are reported  
 173 in Figure 6. Among all systems, Au/WSe<sub>2</sub>/Si system exhibits the highest friction force across all  
 174 velocities, followed by Au/MoS<sub>2</sub>/Si and Au/WS<sub>2</sub>/Si. Au/MoSe<sub>2</sub>/Si displays nearly negligible fric-

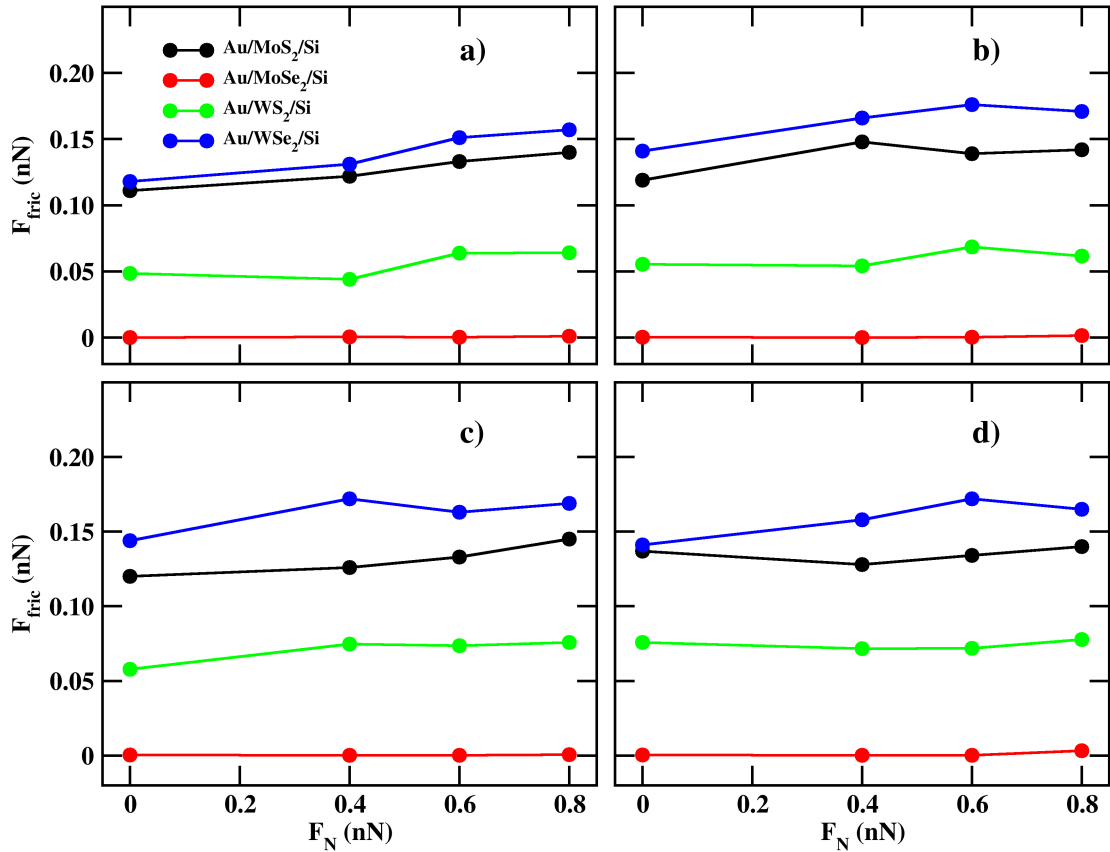


Figure 6: Average friction force as a function of applied load for all systems at tip velocities of (a) 2 m/s, (b) 3 m/s, (c) 4 m/s, and (d) 5 m/s.

175 tion force across all the applied normal loads. This behaviour is consistent with the corresponding  
 176 friction force profile, which will be discussed in detail below. In most cases, the friction–load rela-  
 177 tionship deviates from simple linear behaviour, exhibiting nonmonotonic trends irrespective of the  
 178 velocities. Looking at [Figure 7](#), which represents the fitted curve corresponding to [Figure 6a](#), the  
 179 force–fit analysis gives  $\mu$  values of 0.036, 0.023, 0.051, and 0.0009, with corresponding errors of  
 180 0.003, 0.014, 0.009, and 0.0004 for Au/MoS<sub>2</sub>/Si, Au/WS<sub>2</sub>/Si, Au/WSe<sub>2</sub>/Si, and Au/MoSe<sub>2</sub>/Si, re-  
 181 spectively. As a result, the extracted  $\mu$  shows relatively large errors, indicating that the assumption  
 182 of a linear friction–load relationship is no longer strictly valid. A similar trend is observed at other  
 183 velocities (see Supporting Information File 1, Section II, Figure S1, S2 and S3). This behaviour  
 184 suggests that in our systems, a simple proportional relationship between  $F_{fric}$  and  $F_N$  cannot fully

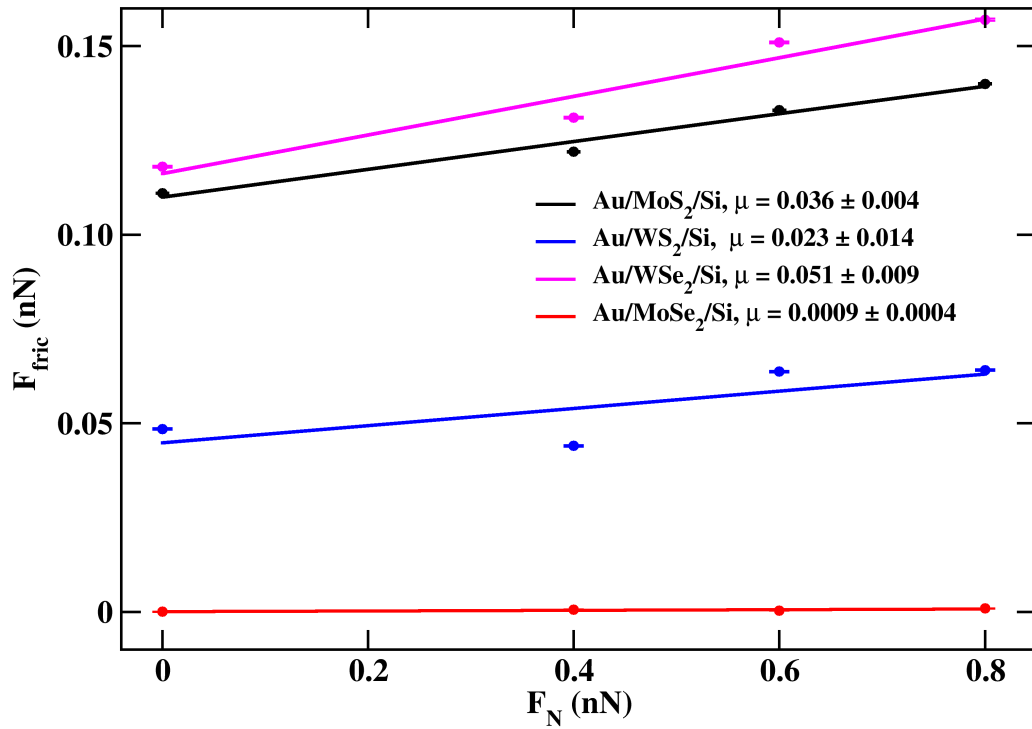


Figure 7: COF obtained from a linear fit of the average friction force as a function of loads for all systems at a tip velocity of 2 m/s.

185 describe the friction response. This behaviour has already been experimentally observed for an  
 186 Au/MoS<sub>2</sub>/Si monolayer grown on an Au surface [46].  
 187 To understand the origin of this nonmonotonic behaviour, we look at the detail of the friction force  
 188 on the tip during the sliding. Figure 8 shows the friction force on Si tip as a function of the slid-  
 189 ing distance for the case of Au/MoS<sub>2</sub>/Si system with a velocity of 2 m/s and normal load of 0 nN.  
 190 Apart from the sliding events along the sliding direction, some extra features can also be observed.  
 191 Analysis of the geometric sequence along the trajectory reveals that large sudden drops in the lat-  
 192 eral force correspond to slip events associated with the primary sliding motion along  $x$ -direction,  
 193 named as sliding. In contrast, abrupt increases in the force indicate lateral slip motion involving  
 194 transverse displacement along  $y$ -direction, named as lateral slip. Following these events, a gradual

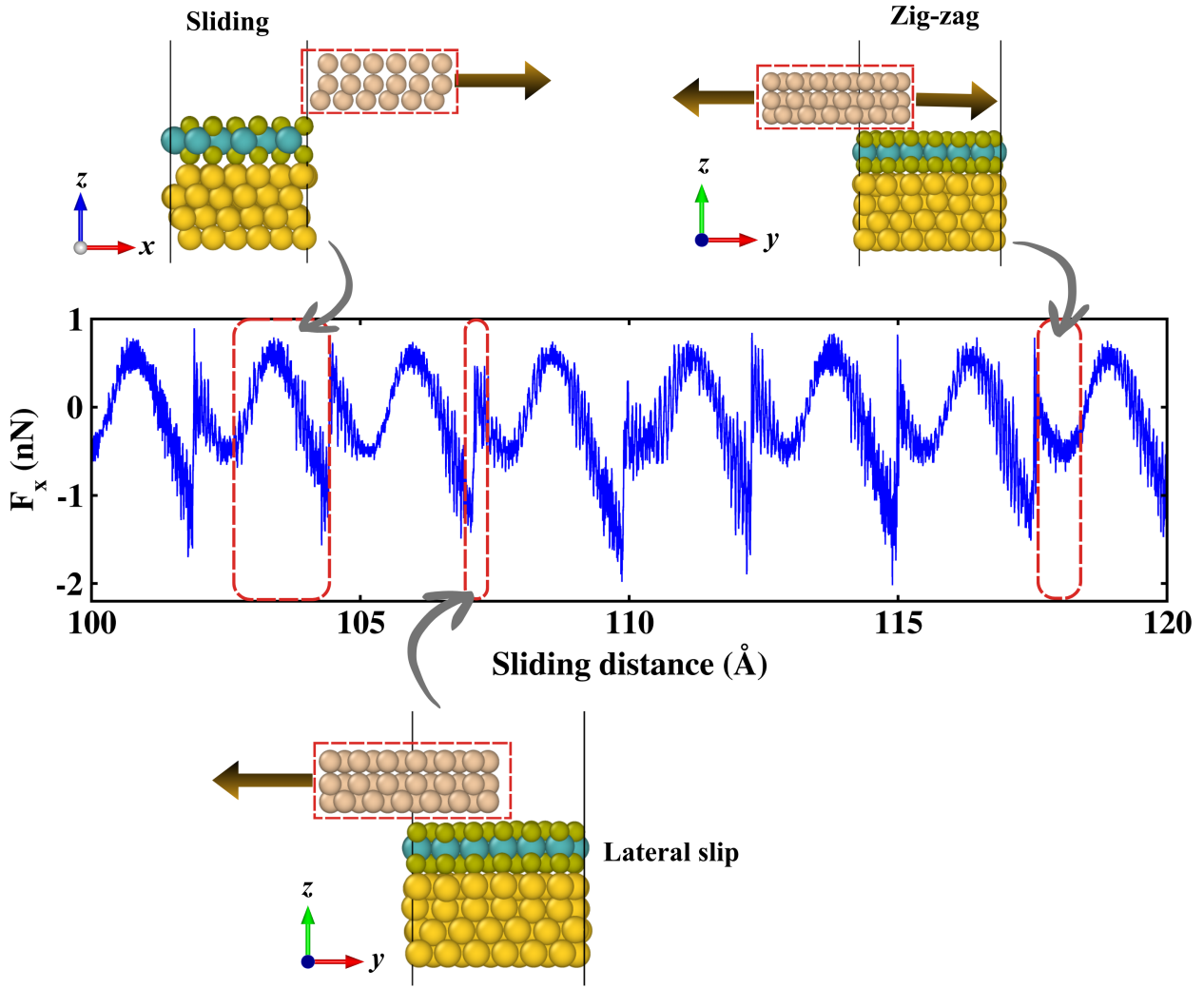


Figure 8: Friction force profile associated with the motion of the tip.

195 decrease in the force is observed, which corresponds to a zigzag-like motion of the tip as it relaxes  
 196 while moving laterally along  $y$ -direction, named as zig-zag. These observations confirm that the  
 197 tip does not follow a strictly one-dimensional sliding pathway; instead, the motion involves cou-  
 198 pled displacements along both  $x$  and  $y$  directions, leading to the different frictional behaviour along  
 199 the  $x$ -direction. The zig-zag events indicate that the system explores multiple local minima on the  
 200 interfacial potential energy surface during sliding; such lateral motion effectively modifies the in-  
 201 stantaneous contact configuration and the energy dissipation pathway, leading to fluctuations in  
 202 the lateral force along  $x$ -direction. Interestingly, these features in the force profile occur in most of  
 203 the cases except the system Au/MoSe<sub>2</sub>/Si. In the latter cases, the lateral slip and zig-zag part in the

204 force profile is missing which leads to a reduction in the average friction force, as shown in Sup-  
205 porting Information File 1 Figure S4. When two surfaces interact, the surface interactions create  
206 a corrugated potential energy landscape that governs the lateral motion during sliding. As the tip  
207 moves across this landscape, the motion may not remain strictly confined to the imposed sliding  
208 direction; instead, the system can explore neighbouring energy minima, resulting in transverse dis-  
209 placements and complex sliding pathways.

210 To quantitatively characterise these slip events, we perform a spatial Fourier transform of the forces  
211 experienced by the Si tip along the  $x$ -direction. This analysis provides insight into the nature of the  
212 forces that arise from different modes of tip motion, including sliding, lateral motion, and possible  
213 nonlinearities in the tip trajectory. Figure 9a displays the spatial Fourier transform of the average  
214 force experienced by the tip for the case of Au/MoS<sub>2</sub>/Si at different normal loads with velocity of  
215 2 m/s. Three magenta dotted lines indicate the main peaks at wave vectors  $k_1$ ,  $k_2$  and  $k_3$  with the  
216 respective intensities  $I_1$ ,  $I_2$  and  $I_3$ . In the Fourier transform, the intensity of a peak is proportional  
217 to the number of times the corresponding event occurs, while the inverse of the peak position gives  
218 the width of the events on a distance scale. In our simulations, the first peak at  $k_1$  corresponds to  
219 the sliding process along the  $x$ -direction, while the second and third peaks (at  $k_2$  and  $k_3$ , respec-  
220 tively) correspond to lateral zig-zag movement and lateral slip along the  $y$ -direction, respectively.

221 We do not consider other peaks, as their intensity is almost as small as the background noise corre-  
222 sponding to thermal fluctuations of the atomic motions. This is confirmed from the friction force  
223 profile where length of the events correlate to the peak positions. The Fourier transform spectra  
224 reveal clear correlation between the peak intensities and the average friction force. In particular,  
225 the overall ratio  $I_1/I_2$  of the first two peak intensities for all the systems increases as the mean fric-  
226 tion force increases (Figure 9b). This trend is observed for all systems across different velocities,  
227 except in the case of a Au/MoSe<sub>2</sub>/Si system; in fact, in this case only a single peak appears in the  
228 Fourier transform only at  $k_1$ , that is, the peak of the sliding, making it impossible to establish a cor-  
229 relation based on the ratio of peak intensities (see Supporting Information File 1, Section II, Figure  
230 S5). This trend suggests that when the number of lateral slip events increases, the effective friction

231 force decreases (Figure 9b). Since the friction force is evaluated under different normal loads, the  
232 relative contributions of these sliding modes vary with load. As a result, the mean friction force  
233 does not exhibit a strictly linear dependence on the applied load, which leads to the nonmonotonic  
234 behavior observed in the friction-load relationship, and contributes to the large uncertainty in the  
235 extracted coefficient of friction using Amontons's law.

## 236 **Effect of substrate on friction force**

237 To investigate the effect of the substrate on the lateral friction force, we analyse the trend of the  
238 force experienced by the tip as a function of sliding distance under different applied normal loads  
239 and different substrates, namely Au and Ag, as shown in Figure 10. We observe that the overall na-  
240 ture of the force signal—particularly the shape of the force profile—remains essentially unchanged  
241 as the load increases, provided the monolayer and velocity are kept constant; moreover, in the pres-  
242 ence of the Ag substrate, the average force on the tip follows a trend very similar to that observed  
243 for the Au substrate. Furthermore, we plot the average friction force as a function of the applied  
244 load for all systems and for all sliding velocities, in the presence of the Ag substrate, as shown in  
245 Figure 11. We found that the variation of friction force as a function of loads shows nonmonotonic  
246 trend and it is consistent with the case of Au substrate that we already have discussed before (see  
247 Figure 6). Notably, this behavior is consistently observed for all sliding velocities, indicating that  
248 the nonmonotonic features are largely independent of velocity. Overall, among all systems, the  
249 Ag/MoS<sub>2</sub>/Si system exhibits the highest friction force across all velocities, followed by Ag/WSe<sub>2</sub>/Si  
250 and Ag/MoSe<sub>2</sub>/Si, while the Ag/WS<sub>2</sub>/Si system shows the lowest friction force over the entire ve-  
251 locity range. In contrast, when considering the Au substrate, the Au/WSe<sub>2</sub>/Si system demonstrates  
252 the highest friction force at all velocities, followed by Au/MoS<sub>2</sub>/Si and Au/WS<sub>2</sub>/Si, as discussed in  
253 above paragraph. This indicates that the friction force can change significantly depending on the  
254 substrate. Furthermore, the calculated  $\mu$  (as shown in Supporting Information File 1, Section II,  
255 Figure S6, S7, S8 and S9) show quite large errors, arising from the nonmonotonic dependence of  
256 the friction force on the applied load. We already observed a similar trend in the case of the Au

257 substrate, suggesting that the overall nature of frictional behavior in our systems is largely inde-  
258 pendent of the substrate. [Figure 12](#) shows the three sharp peaks in FT at velocity 2 m/s for both  
259 Ag/MoS<sub>2</sub>/Si and Au/MoS<sub>2</sub>/Si systems, which have very similar profiles. We find that the positions  
260 of all three peaks remain nearly unchanged for different applied loads and different substrates, in-  
261 dicating that they are independent of loads and substrates. We also observe that the peak positions  
262 in the Fourier transforms shift as the monolayer material is varied, confirming that the forces ex-  
263 perience by the tip depend on the specific monolayer (Supporting Information File 1 Section II,  
264 [Figure S10](#)). In particular, the first peak intensities at varying loads are significantly higher with  
265 the Ag/MX<sub>2</sub>/Si systems compared to the case with Au/MX<sub>2</sub>/Si systems; this indicates that the fric-  
266 tion contribution along the sliding direction is greater when using the Ag substrate ([Figure 14a](#)).  
267 In contrast, the opposite trend is observed with the Au/WSe<sub>2</sub>/Si system, where the friction force  
268 is higher compared with the Ag/WSe<sub>2</sub>/Si ([Figure 14d](#)). This behaviour correlates with the Fourier  
269 transform results (which is shown in Supporting Information File 1, Section II, [Figure S10](#)), where  
270 the first peak intensity is higher leads to higher in peak intensity ratio ( see [Figure 13d](#)) in systems  
271 with the Au substrate. Overall, these findings suggest that the contribution of multiple sliding and  
272 lateral motion in the friction profile strongly influences the average friction force, depending on the  
273 specific monolayer and substrate combination.

274 Focusing to the intensity ratio of both Au/MX<sub>2</sub>/Si and Ag/MX<sub>2</sub>/Si system (see [Figure 13](#)), the vari-  
275 ation of the peak-intensity ratio follows a trend similar to that of the friction force (see [Figure 14](#)).  
276 In particular, the peak intensity ratios are higher in the case of the Ag substrate compared with the  
277 Au substrate, irrespective of the applied loads. Furthermore, the peak-intensity ratio exhibits a clear  
278 nonmonotonic dependence on the applied load, consistent with the nonmonotonic behaviour ob-  
279 served in the friction force, resulting in high errors in the estimation of the friction coefficient from  
280 a linear fit against the applied load. Therefore, the lateral motion of the tip plays an important role  
281 in determining the overall friction force and contributes to both the reduction of the average friction  
282 force and the emergence of its nonmonotonic behaviour.

## 283 **Conclusions**

284 In this study, we investigate the nanoscale friction behaviour of TM/MX<sub>2</sub>/Si systems (TM =  
285 Au, Ag; M = Mo, W; X = S, Se) using classical molecular dynamics simulations with machine-  
286 learning-based force fields trained on DFT data. Our results show that the mean friction force does  
287 not follow a strictly linear dependence on the applied normal load. Instead, a nonmonotonic fric-  
288 tion–load relationship is observed, which introduces significant uncertainty in determining the  
289 coefficient of friction based on Amontons’s law. Analysis of the spatial Fourier transform of lat-  
290 eral force signals reveals multiple spectral peaks, with their intensities strongly correlated with the  
291 average friction force. The presence of these peaks indicates that the tip motion is not limited to  
292 the primary sliding direction but also includes significant lateral contributions. Furthermore, the  
293 qualitative features of both the friction force profiles and the Fourier spectra remain similar across  
294 different substrates. While the magnitude of friction depends on the specific substrate material, the  
295 underlying mechanism responsible for the nonmonotonic behavior—arising from the coupling be-  
296 tween longitudinal sliding and lateral motion of the tip—remains essentially substrate-independent.  
297 Since our findings demonstrate that frictional forces are influenced by both the monolayer and the  
298 underlying substrate, these methods can be effectively employed to investigate nanoscale frictional  
299 behaviour in a wide range of heterostructures.

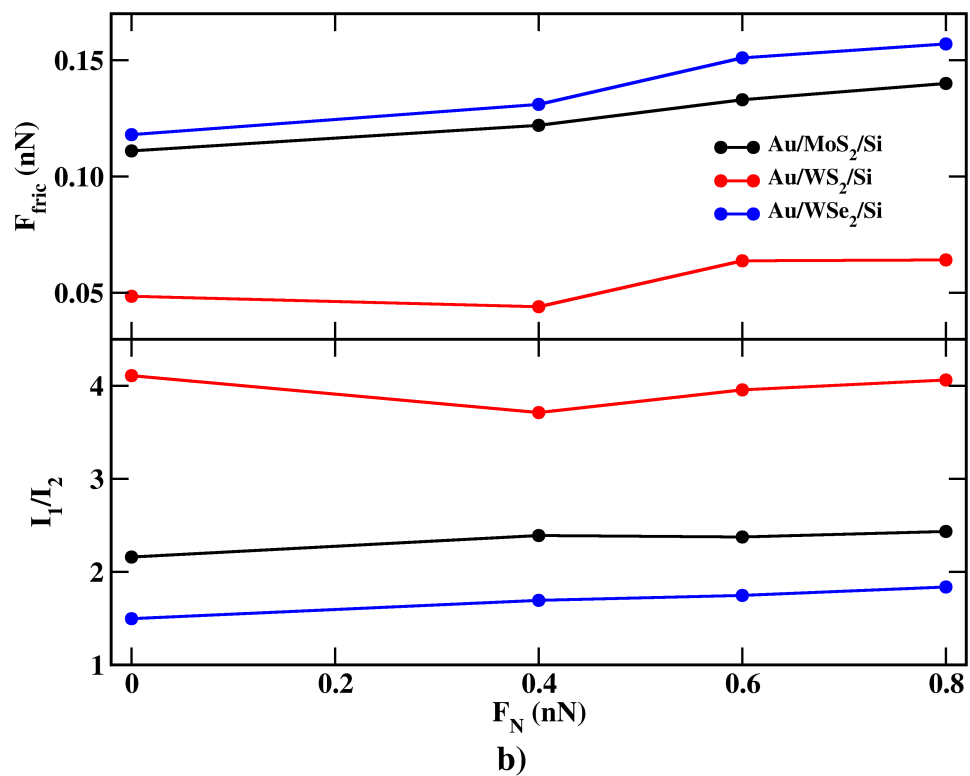
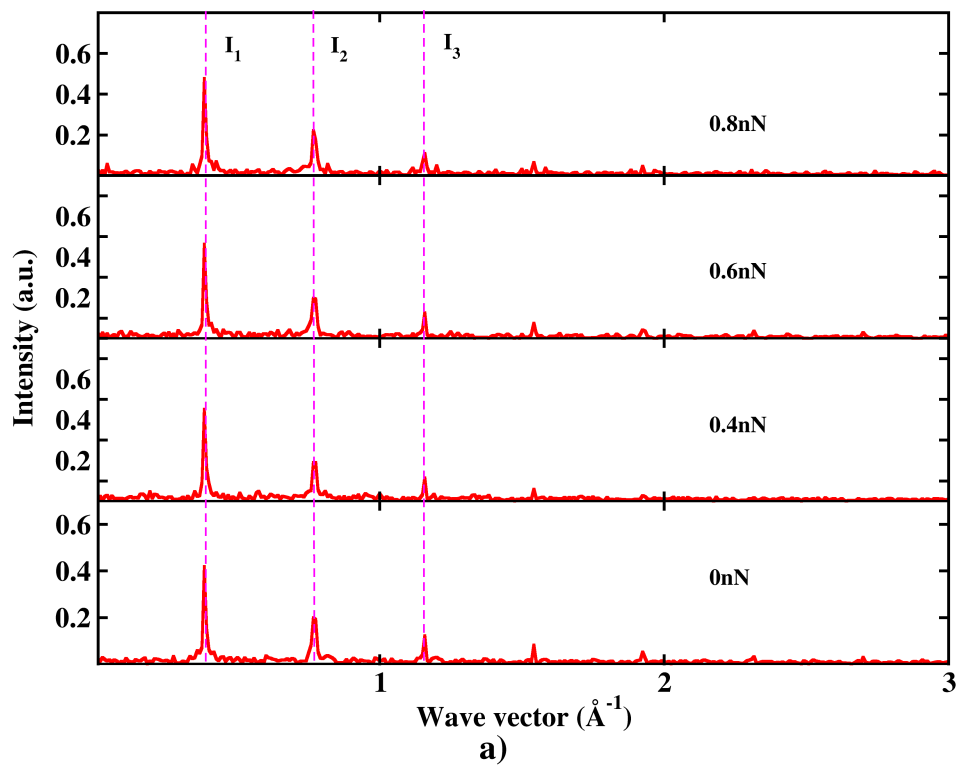


Figure 9: a) Spatial Fourier transform of Au/MoS<sub>2</sub>/Si with varying loads at velocity 2 m/s, b) Peak intensity ratio and their average friction force with varying loads for all the systems except Au/MoSe<sub>2</sub>/Si.

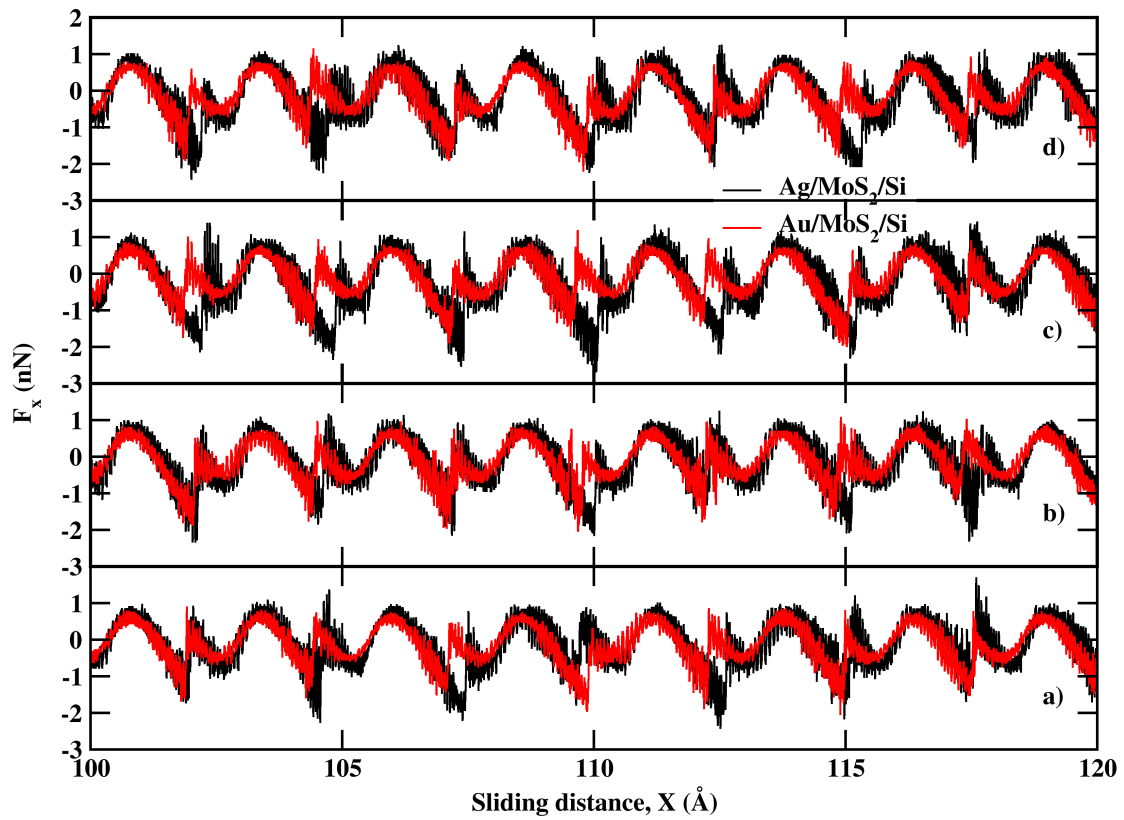


Figure 10: Comparison of friction force profile between Au/MoS<sub>2</sub>/Si and Ag/MoS<sub>2</sub>/Si as a function of displacement at loads a) 0 nN, b) 0.4 nN, c) 0.6 nN and d) 0.8 nN.

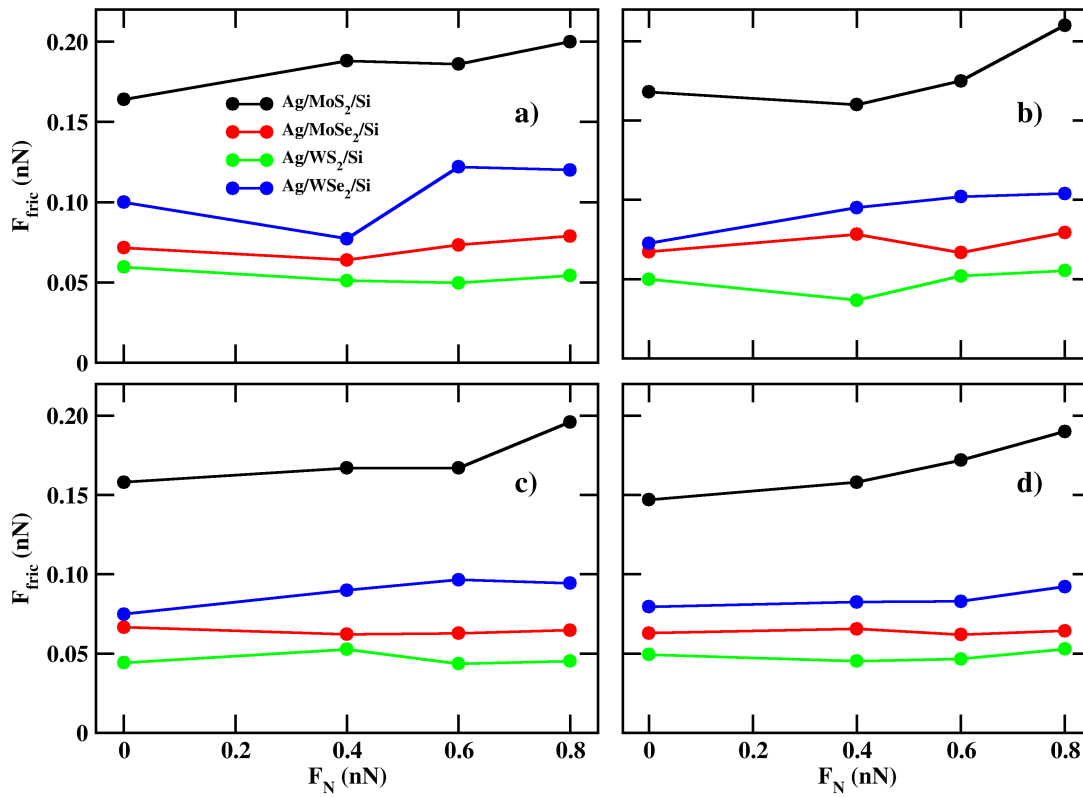


Figure 11: Average friction force as a function of applied loads for all the systems with tip velocities a) 2 m/s, b) 3 m/s, c) 4 m/s d) 5 m/s.

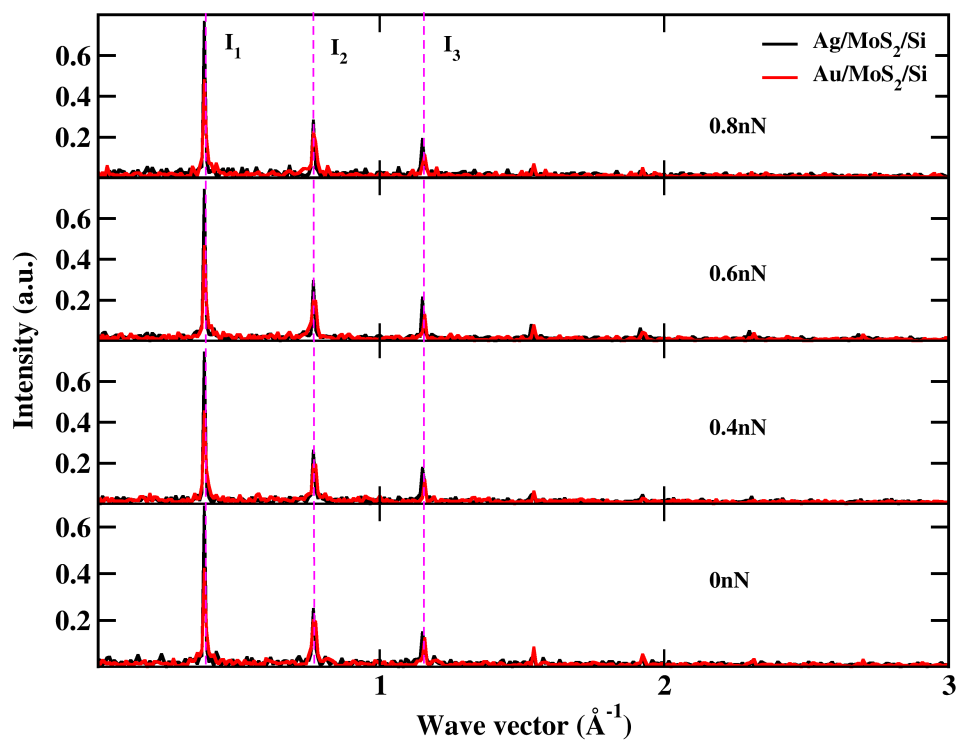


Figure 12: Fourier transform of the force on a tip varying with loads and different substrate for velocity 2 m/s.

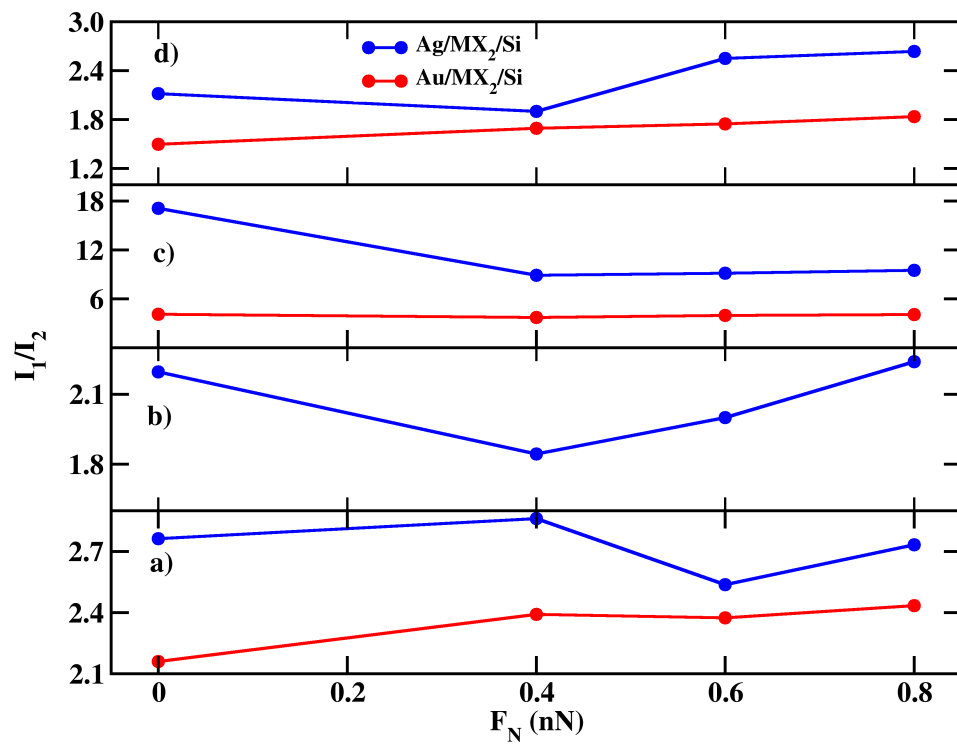


Figure 13: Ratio of the peak intensity  $I_1$  and  $I_2$  varying with loads and different substrate for velocity 2 m/s.

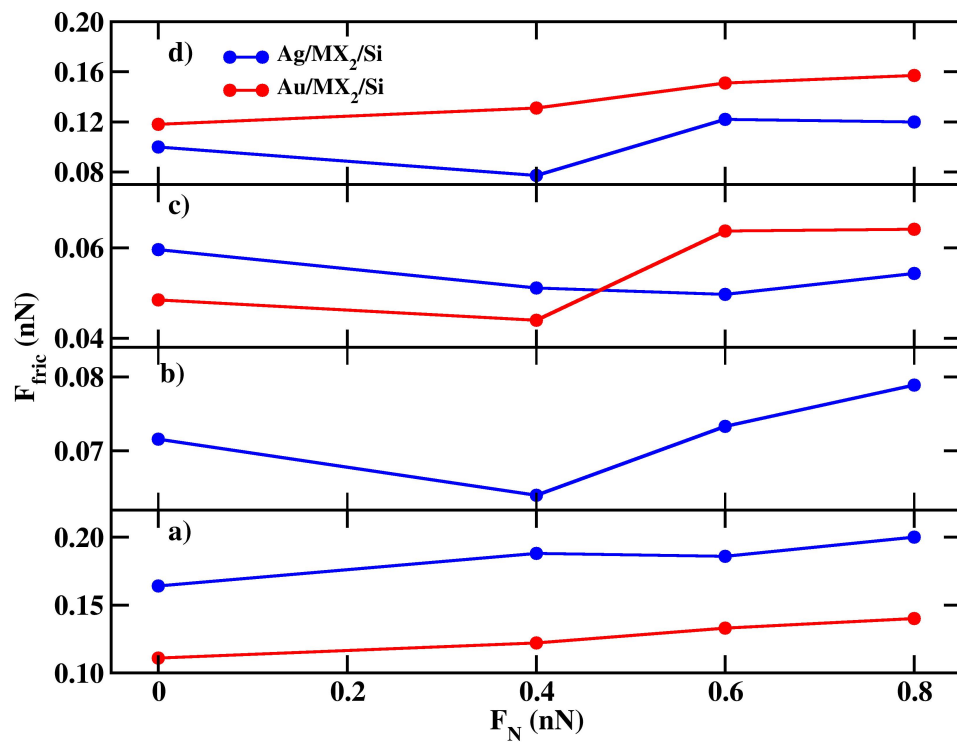


Figure 14: Average friction force, varying with loads and different substrate for velocity 2 m/s.

## 300 **Supporting Information**

301 Supporting information for Microscopic contributions to the deviation from Amontons friction law.

302 Supporting Information File 1:

303 File Name: [SI.pdf](#)

304 File Format: PDF

305 Title: Additional data and Figures

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