

Tuning Frictional Properties of Kirigami Altered Graphene Sheets using Molecular Dynamics and Machine Learning

Designing a Negative Friction Coefficient

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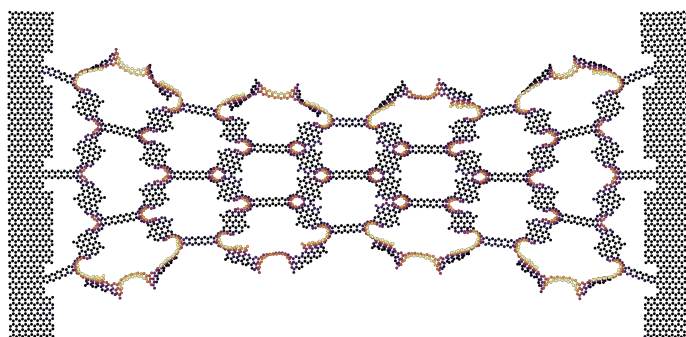
UNIVERSITY OF OSLO

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Abstract

Abstract.

Acknowledgments

Acknowledgments.

List of Symbols

F_N Normal force (normal load)

Acronyms

MD Molecular Dynamics. 2, 3, 9, 10

ML Machine Learning. 2, 3, 10

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Chapter 1

Introduction

Structure of Motivation section:

1. Introduce and motivate friction broadly.
2. Motives for friction control using a grasping robot as example.
3. Analog to gecko feet where adhesive properties are turned on and off.
4. Interest in origin of friction through nanoscale studies which further motivates the use of MD.
5. Intro to metamaterials and the use of kirigami designs,
6. How to optimize kirigami designs with reference to Hanakata and motivating the use of ML.
7. Out-of-plane buckling motivates the use of kirigami for frictional properties.

Does some of the latter paragraphs belong to the approach section?

1.1 Motivation

Friction is a fundamental force that takes part in most of all interactions with physical matter. Even though the everyday person might not be familiar with the term *friction* we recognize it as the inherent resistance to sliding motion. Some surfaces appear slippery and some rough, and we know intuitively that sliding down a snow covered hill is much more exciting than its grassy counterpart. Without friction, it would not be possible to walk across a flat surface, lean against the wall without falling over or secure an object by the use of nails or screws [p. 5] [1]. It is probably safe to say that the concept of friction is integrated in our everyday life to such an extent that most people take it for granted. However, the efforts to control friction dates back to the early civilization (3500 B.C.) with the use of the wheel and lubricants to reduce friction in translational motion [2]. Today, friction is considered a part of the wider field *tribology* derived from the Greek word *Tribos* meaning “rubbing” and includes the science of friction, wear and lubrication [2]. The most compelling motivation to study tribology is ultimately to gain full control of friction and wear for various technical applications. Especially, reducing friction is of great interest as this has tremendous advantages for energy efficiency. It has been reported that tribological problems have a significant potential for economic and environmental improvements [3]:

“On global scale, these savings would amount to 1.4% of the GDP annually and 8.7% of the total energy consumption in the long term.” [4].

On the other hand, the reduction of friction is not the only sensible application for tribological studies. Controlling frictional properties, besides minimization, might be of interest in the development of a grasping robot where a finetuned object handling is required. While achieving a certain “constant” friction response is readily obtained through appropriate material choices during manufacturing, we are yet to unlock the capabilities to alter friction dynamically on the go. One example from nature inspiring us to think along these lines are the gecko feet. More precisely, the Tokay gecko has received a lot of attention in scientific studies aiming to unravel the underlying

mechanism of its “togglable” adhesion properties. Although geckos are able to produce large adhesive forces, they retain the ability to remove their feet from an attachment surface at will [5]. This makes the gecko able to achieve a high adhesion on the feet when climbing a vertical surface while lifting it for the next step remains relatively effortless. For a grasping robot we might consider an analog frictional concept of a surface material that can change from slippery to rough on demand depending on specific tasks.

In the recent years an increasing amount of interest has gone into the studies of the microscopic origin of friction, due to the increased possibilities in surface preparation and the development of nanoscale experimental methods. Nano-friction is also of great concern for the field of nano-machining where the frictional properties between the tool and the workpiece dictates machining characteristics [3]. With concurrent progress in computational power and development of Molecular Dynamics (MD), numerical investigations serve as an extremely useful tool for getting insight into the nanoscale mechanics associated with friction. This simulation based approach can be considered as a “numerical experiment” enabling us to create and probe a variety of high complexity systems which are still out of reach for modern experimental methods.

In materials science such MD-based numerical studies have been used to explore the concept of so-called *metamaterials* where material compositions are designed meticulously to enhance certain physical properties [6][7][8][9][10][11]. This is often achieved either by intertwining different material types or removing certain regions completely. In recent papers by Hanakata et al. [6](2018) [7](2020) numerical studies have showcased that mechanical properties of a graphene sheet, in this case yield stress and yield strain, can be altered through the introduction of so-called *kirigami* inspired cuts into the sheet. Kirigami is a variation of origami where the paper is cut additionally to being folded. While these methods originate as an art form, aiming to produce various artistic objects, they have proven to be applicable in a wide range of fields such as optics, physics, biology, chemistry and engineering [12]. Various forms of stimuli enable direct 2D to 3D transformations through folding, bending, and twisting of microstructures. While original human designs have contributed to specific scientific applications in the past, the future of this field is highly driven by the question of how to generate new designs optimized for certain physical properties. However, the complexity of such systems and the associated design space makes for seemingly intractable problems ruling out analytic solutions.

Earlier architecture design approaches such as bioinspiration, looking at gecko feet for instance, and Edisonian, based on trial and error, generally rely on prior knowledge and an experienced designer [9]. While the Edisonian approach is certainly more feasible through numerical studies than real world experiments, the number of combinations in the design space rather quickly becomes too large for a systematic search, even when considering the simulation time on modern day hardware. However, this computational time constraint can be relaxed by the use of machine learning (ML) which have proven successful in the establishment of a mapping from the design space to physical properties of interest. This gives rise to two new styles of design approaches: One, by utilizing the prediction from a trained network we can skip the MD simulations all together resulting in an *accelerated search* of designs. This can be further improved by guiding the search accordingly to the most promising candidates, as for instance done with the *genetic algorithm* which suggest new designs based on mutation and crossing of the best candidates so far. Another, even more sophisticated approach, is through generative methods such as *Generative Adversarial Networks* (GAN). By working with a so-called *encoder-decoder* network structure, one can build a model that reverses the prediction process. That is, the model predicts a design from a set of physical target properties. In the papers by Hanakata et al. both the *accelerated search* and the *inverse design* approach was proven successful to create novel metamaterial kirigami designs with the graphene sheet.

Hanakata et al. attributes the variety in yield properties to the non-linear effects arising from the out-of-plane buckling of the sheet. Since it is generally accepted that the surface roughness is of great importance for frictional properties it can be hypothesized that the kirigami cut and stretch procedure can also be exploited for the design of frictional metamaterials. For certain designs we might hope to find a relationship between stretching of the sheet and frictional properties. If significant, this could give rise to a variability of the friction response beyond manufacturing material choice. For instance, the grasping robot might apply such a material as artificial skin for which stretching or relaxing of the surface could result in a changeable friction strength; Slippery and smooth when in contact with people and rough and firmly gripping when moving heavy objects. In addition, a possible coupling between stretch and the normal load through a nanomachine design would allow for an altered friction coefficient. This invites the idea of non-linear friction coefficients which might in theory also take on negative values given the right response from stretching. The latter would constitute an extremely rare property. This has (only?) been reported indirectly for bulk graphite by Deng et al. [13] where the friction kept increasing during the unloading phase. [Check for other cases and what I can really say here.](#)

To the best of our knowledge, kirigami has not yet been implemented to alter the frictional properties of a nanoscale system. In a recent paper by Liefferink et al. [14](2021) it is reported that macroscale kirigami can be used to dynamically control the macroscale roughness of a surface through stretching which was used to change the frictional coefficient by more than one order of magnitude. This supports the idea that kirigami designs can in fact be used to alter friction, but we believe that taking this concept to the nanoscale regime would involve a different set of underlying mechanisms and thus contribute to new insight in this field.

1.2 Goals

In this thesis we investigate the possibility to alter and control the frictional properties of a graphene sheet through application of kirigami inspired cuts and stretching of the sheet. With the use of MD simulations we evaluate the friction properties under different physical conditions in order to get insight into the prospects of this field. By evaluating variations of two kirigami inspired patterns and a series of random walk generated patterns we create a dataset containing information of the frictional properties associated with each design under different load and stretch conditions. We apply ML to the dataset and use an accelerated search approach to optimize for different properties of interest. The subtask of the thesis are presented more comprehensively in the following.

1. Define a sheet indexing that allows for an unique mapping of patterns between a hexagonal graphene lattice representation to a matrix representation suited for numerical analysis.
2. Design a MD simulation procedure to evaluate the frictional properties of a given graphene sheet under specified physical conditions such as load, stretch, temperature etc.
3. Find and implement suitable kirigami patterns which exhibit out-of-plane buckling under tensile load. This includes the creation of a framework for creating variations within each pattern class. Additionally create a procedure for generating different styles of random walk patterns.
4. Perform a pilot study of a representative subset of patterns in order to determine appropriate simulation parameters to use for the further study along with an analysis of the frictional properties shown in the subset.
5. Create a dataset consisting of the chosen kirigami variations and random walk patterns and analyse data trends.
6. Train a neural network to map from the design space to physical properties such as mean friction, maximum friction, contact area etc. and evaluate the performance.
7. Perform an accelerated search optimizing for interesting frictional properties using the ML model. This should be done both through the pattern generation procedures and by following a genetic algorithm approach.
8. Use the most promising candidates from the accelerated search to investigate the prospects of creating a nanomachine setup which exhibits a negative friction coefficient.
9. Study certain designs of interest with the scope of revealing underlying mechanism. This includes simple correlation analysis but also a visualization of feature and gradient maps of the ML network.

Is the list of subtask too specific? Some of the details here might be better suited for the thesis structure section.

1.3 Contributions

What did I actually achieve

1.4 Thesis structure

How is the thesis structured.

Part I

Background Theory

Part II

Simulations

Chapter 2

Pilot study

Having defined our system, we carry out an initial study of the numerical approach. This includes an analysis of how to define and measure the frictional properties of interest, and an investigation of the main parameters governing the numerical solutions. From this point of view we decide on a suitable set of parameters for the remaining study. Additionally, we investigate the frictional behaviour under the variation of load and stretch for a selected set of configurations which serves as a baseline for later comparison.

2.1 Friction simulation parameters

The MD simulation is governed by a small set of parameters, some which are related directly to the numerical aspects of the simulation and other to the physical conditions in the simulation. Thus, we differentiate between the two main categories: 1) *Physical*, parameters which alter the physical conditions of the “numerical experiment” and are expected to effect the frictional behaviour. 2) *Numerical*, parameters which are related more closely to the numerical procedure itself, expected to influence the simulation dynamics, which should be chosen to ensure the most stable results. For the purpose of creating the machine learning dataset most of these parameters will be kept constant with only a subset of the physical parameters being varied. The parameters are summarized in Table 2.1 with the grey shaded area denoting the parameters which we will vary for the dataset. Due to the great number of parameters it is unreasonable to make an exhaustive search of all parameters, in order to choose the final settings. Instead, we take a basis in the parameters used in similar studies [SOURCES](#) and adjust them as we carry out the analysis of the simulation results. Thus, we start at values most representative for other similar simulations and adjust according to the stability of the results and the computation time. Since we are going to introduce a lot of complexity to the system, through the cut and stretch deformation, we are less strict about meeting other parameter criterias (does this make sense). The final parameter choice is shown in Table 2.1 which we be the default values in the following study, i.e. when nothing is stated explicitly we will use these parameters. As we analyse the data we will provide further arguments for specific choices made.

Table 2.1: Parameters of the numerical MD simulation for measuring friction. The values correspond to the final choice used for the dataset. The shaded area denote the parameters varied in the ML dataset.

Category	Parameter	Value	Description
Physical	T	300 K	Temperature.
	v_{slide}	20 m/s	Sliding speed for the sheet translation.
	K	inf	Spring constant for the coupling between the virtuel atom and the sheet pull blocks.
	Scan direction	$(x, y) = (0, 1)$ (zigzag direction)	The direction for which we translate the sheet.
	Sheet configuration	Contiguous	Binary mapping describing which atoms are removed (0) and which is still present (1) in the graphene sheet.
	Stretch amount	0% - rupture	The relative stretch of the sheet.
	F_N	[0.1, 10] nN	Applied normal force to the pull blocks.
Numerical	dt	1 fs	Integration timestep.
	t_R	15 ps	Relaxtion time before strething.
	Pauses	5 ps	Relaxtion pauses after stretch, and during the normal load phase (before translating the sheet).
	Stretch Speed	0.01 ps^{-1}	The rate of stretching for the sheet.
	Slide distance	400 Å	How far to translate the sheet.
	Sheet size	$130.029 \times 163.219 \text{ Å}$	Spatial 2D size of the sheet.
	Pull block size	$2 \times 130.029 \times 15.183 \text{ Å}$	Spatial 2D size of the pull blocks.

2.2 Single friction simulation analysis

We begin by assessing the raw data for a single friction simulation using the default parameters shown in ?? for a non-cut sheet with no stretch applied and a normal force of 1 nN.

2.2.1 Force oscillations

We evaluate the friction force by force acting on the sheet from the substrate. We initially look at the force friction force F_{\parallel} parallel to the drag direction as seen in Fig. 2.1. The sample rate is $10 \text{ ps}^{-1} = 100 \text{ timesteps}^{-1}$ for which each sample is the mean value of the 100 timesteps preceding the given sample interval. We observe immediately that the data carries oscillations on different time scales which matches the expectations for sliding between periodic surfaces. By applying a savgol filter to the data with a polyorder of 5 and window length of 150 timesteps, corresponding to a sliding distance of 3 \AA or a time window of 15 ps, we can qualitatively point out at least two different frequencies of oscillation. During the first 10 \AA of sliding in Fig. 2.1a we see roughly three waves on the savgol filter corresponding to a relative high frequency, while on the scale of 100 \AA of sliding in Fig. 2.1b the same savgol filter reveals a lower frequency in addition, creating the visual pattern of a wavepacket.

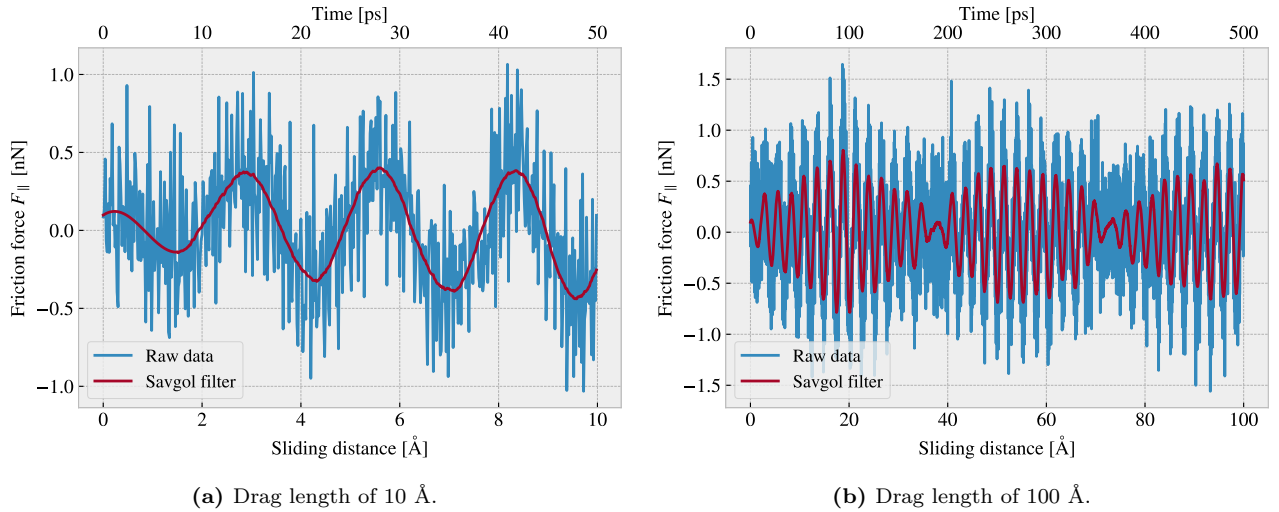


Figure 2.1: Friction force F_{\parallel} with respect to the drag direction between (full) sheet and substrate versus sliding distance. The sliding distance is measured by the constant movement of the virtual atom and not the COM of the sheet. However, we expect these measures to be fairly identical due the fact that the pull blocks is rigidly coupled to the virtual atom. The red line represents a savgol filter with window polyorder 5 and window length of 150 timesteps (corresponding to a sliding distance of 3 \AA or a time window of 15 ps).

By performing a Fourier Transform (FT) on the data we can quantify the leading frequencies as seen in Fig. 2.2a. By plotting the two most dominant frequencies $f_1 = 0.0074 \text{ ps}^{-1}$ and $f_2 = 0.0079 \text{ ps}^{-1}$ as $\sin(2\pi f_1) + \sin(2\pi f_2)$ we find a qualitatively convincing fit to the observed wavepacket shape as seen in Fig. 2.2b. By using the trigonometric identity

$$\begin{aligned}\sin(\alpha + \beta) &= \sin(\alpha) \cos(\beta) + \cos(\alpha) \sin(\beta), \\ \sin(\alpha - \beta) &= \sin(\alpha) \cos(\beta) - \cos(\alpha) \sin(\beta),\end{aligned}$$

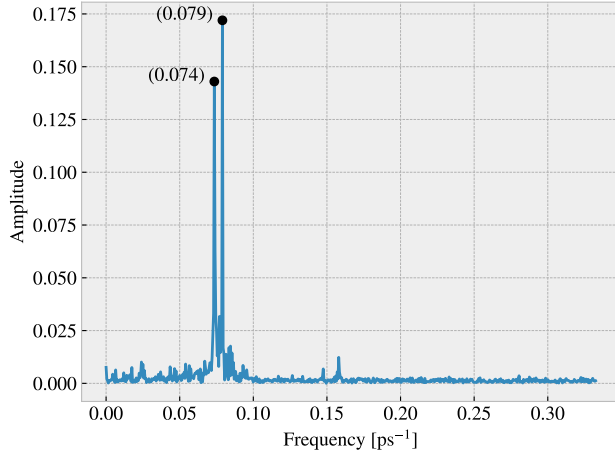
and decomposing $f_1 = a - b$, $f_2 = a + b$ we can rewrite the sine sum as the sinusoidal product

$$\begin{aligned}\sin(2\pi f_1) \sin(2\pi f_2) &= \sin(2\pi(a - b)) \sin(2\pi(a + b)) \\ &= \sin(a) \cos(b) + \cancel{\cos(2\pi a) \sin(2\pi b)} + \sin(2\pi a) \cos(2\pi b) - \cancel{\cos(2\pi a) \sin(2\pi b)} \\ &= 2 \sin(2\pi a) \cos(2\pi b),\end{aligned}$$

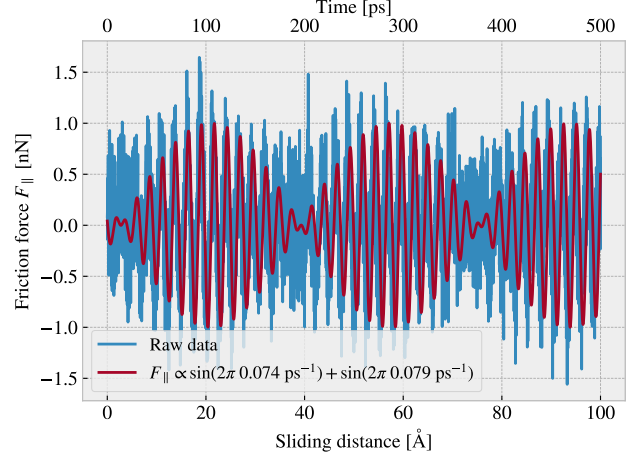
with

$$\begin{aligned} a = \frac{f_1 + f_2}{2} &= 0.0763 \pm 0.0005 \text{ ps}^{-1}, & b = \frac{f_2 - f_1}{2} &= 0.0028 \pm 0.0005 \text{ ps}^{-1}, \\ &= 0.381 \pm 0.003 \text{ \AA}^{-1}, & &= 0.014 \pm 0.003 \text{ \AA}^{-1}, \end{aligned}$$

where the latter frequency is denoted with respect to the sliding distance. This makes us recognize the high oscillation frequency as a and the low frequency as b . The faster one has a period of $T_a = 2.62 \pm 0.02 \text{ \AA}^1$. This corresponds well with the magnitude of the lattice spacing and especially that of graphene at 2.46 \AA as expected theoretically (make reference to theory section?). We also take note of the longest period $T_b = 71 \pm 15 \text{ \AA}^{-1}$ which will be relevant for the evaluation of measurement uncertainty in section Sec. 2.3.



(a) FT result shown for a reduced frequency range.



(b) Two most dominant frequencies applied to the data from Fig. 2.1b

Figure 2.2: Fourier transform (FT) analysis of the full friction force data (all 400 \AA sliding distance) shown in Fig. 2.1. (a) shows the two most dominant frequency peaks. Note that no significant peaks was found in a higher frequency than included here. (b) shows a comparison between the raw data and the wavefunction corresponding to the two peaks in figure (a).

2.2.2 Decompositions

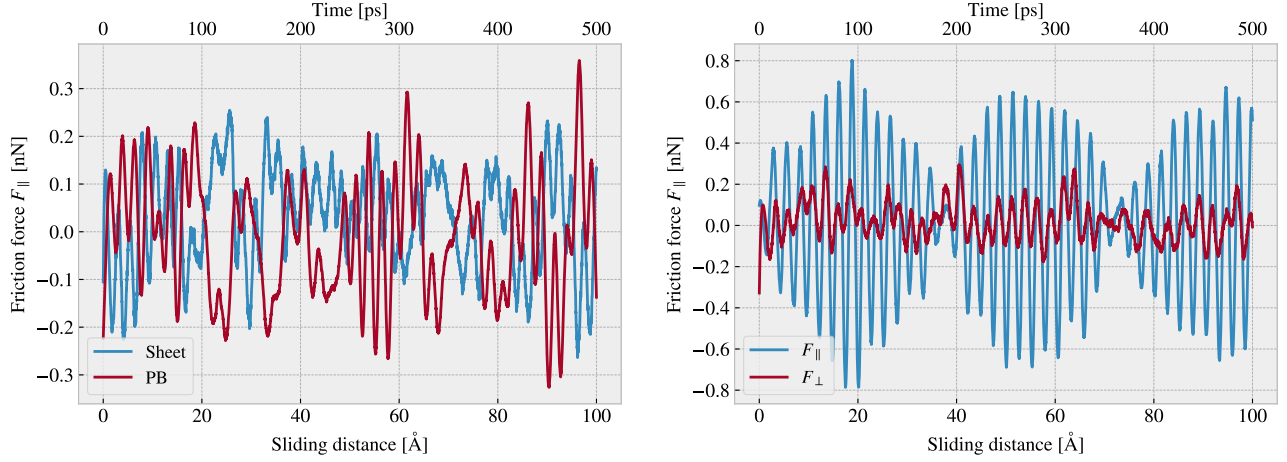
In the previous analysis we have looked only at the friction force for the full sheet, including the pull blocks which is locked off during sliding, and with respect to the drag direction. This represents our choice measurement which we will adress in the following.

Due to the fact that we are only applying cuts to the inner sheet (excluding the pull blocks), it might seem more natural to only consider the friction on that part. If the desired frictional properties can be achieved by altering the inner sheet one can argue that any opposing effects from the pull blocks can be mitigated by scaling the relative size between the inner sheet and the pull blocks. However, when looking at the time series of the friction force decomposed with respect to the inner sheet and pull block region (see figure Fig. 2.3a), we observe the friction force arising from those parts are seemingly antisymmetric. That is, the distribution of the fricitonal pull from the substrate on the sheet is oscillating between the inner sheet and the pull block. Keeping in mind that normal force is only applied to the pull blocks we might take this as an integrated feature of the system which does not nessecary dissapear when changing the spatial ratio between inner sheet and pull block. Any interesting friciton properties might depend on this internal distribution of forces. Hence, we hedge our bets and use the full sheet friction force as a hollistic approach to this measurement problem.

Similar we might question the decision of only considering the frictional force projected onto the sliding direction as we are neglecting the “side shift” induced during the slide phase. In Fig. 2.3b we see the decomposition into force components parallel F_{\parallel} and perpendicular F_{\perp} to the slide direction respectively. We see that the most dominant trends is projected into the parallel component. If we want to include the perpendicular component as

¹The uncertainty Δy is calculated as $\Delta y = \left| \frac{\partial y}{\partial x} \Delta x \right|$ for uncertainty Δx and $y(x)$

well we would have to evaluate the friction as the length of the force vector for which we would lose the sign of the force direction. Hence, we would only get a positive contributin which would not be able to capture the change between resisting and assisting the sliding during stick-slip motion. One option to accommodate this is by using the vector length but keeping the sign from the projection parallel to the sliding direction. However, we omit such compromises as this might make analysis interpration more difficult, and we use only the parallel component going forward.



(a) Decomposition into group inner sheet (sheet) and pull blocks (PB). (b) Decomposition into parallel (F_{\parallel}) and perpendicular (F_{\perp}) to drag sliding direction.

Figure 2.3: Friction force decomposition on the data shown in Fig. 2.1 with applied savgol filters similar to that of Fig. 2.1b with window polyorder 5 and window length of 150 timesteps (corresponding to a sliding distance of 3 Å or a time window of 15 ps).

2.2.3 Center of mass path

From the previous observations of the friction force time series we see evidence of a stick-slip behaviour. Specially, we see in Fig. 2.3b that this might be the case both parallel and perpendicular to the sliding direction. By looking at the x, y -position for the sheet center of mass (COM) we observe the stick-slip motion manifested as a variation in COM speed combined with a side to side motion as shown in figure Fig. 2.4a. In an attempt to increase the magnitude of the slips we evaluate a similar simulation with spring constant $K = 30$ N/m (see figure Fig. 2.4b) in contrast to that of an infinte spring constant. While the maximum slip speed stays within a similar order of magnitude the slip length in the sliding direction is increased along with the side to side motion. Note that the axis scale is different between Fig. 2.4a and Fig. 2.4a. However, in both cases we observe that the side to side motion is associated with a low speed, meaning that is more reminiscent of a “slow” creep alignment with the substrate than a slip.

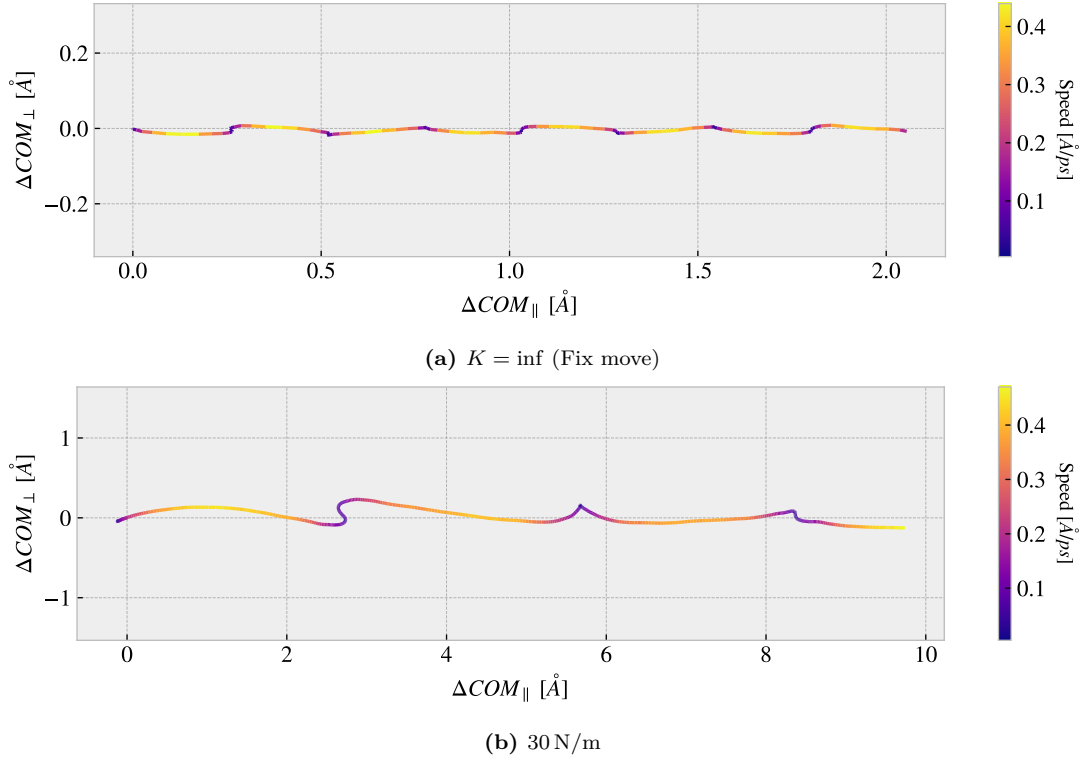


Figure 2.4: Center of mass position relative to the start of the sliding phase in terms of the direction parallel to the sliding direction ΔCOM_{\parallel} and the axis perpendicular to the sliding direction ΔCOM_{\perp} . The colorbar denotes the absolute speed of the COM.

2.3 Defining metrics for dynamic and static friction

In order to evaluate the frictional properties of the sheet we reduce the comprehensive friction force time series addressed in section Sec. 2.2 into single metrics describing the dynamic and static friction. The natural choice is to use the mean and max values of the time series.

2.3.1 Dynamic friction

For the dynamic friction measurement we take the mean value of the latter half of the dataset to ensure that we are sample from a stable system. For a full sliding simulation of 400 Å we thus base our mean value on the latter 200 Å of sliding. In Fig. 2.5a we have shown the friction force of the first 10 Å of sliding together with a running mean with window length of 5 Å corresponding to 50% the data length. This is merely done to illustrate the sampling procedure and by only using a 10 Å sliding distance the final mean estimate (indicated with a dot) takes a negative value due to the specific cut-off of the few oscillation captured here. Nonetheless, one approach to quantify the uncertainty of the final mean estimate is to consider the variation of the running mean preceding the final mean value. The more the running mean fluctuates the more uncertainty associated with the final estimate. However, only the running mean “close” to the ending should be considered, since the first part will rely on data from the beginning of the simulation. From the Fourier analyse in section Sec. 2.2.1 we found the longest significant oscillation period to be $\sim 71 \text{ Å}^{-1}$ corresponding to $\sim 35\%$ of the running mean window consisting of 200 Å of sliding when including all the data. Hence, we use the standard deviation of the final 35% of the running mean to approximate the uncertainty of the final mean value, and we estimate the relative error by dividing the standard deviation by the final mean value. In Fig. 2.5b we showcase a running standard deviation of a window length 35% the running mean window in Fig. 2.5a for the illustrative case of a total 10 Å slide. The final uncertainty value is marked by a dot, and we see as expected that we get a high relative error of $\sim 257\%$ which corresponds well with the short sampling period and the mean value taking an unphysical negative value.

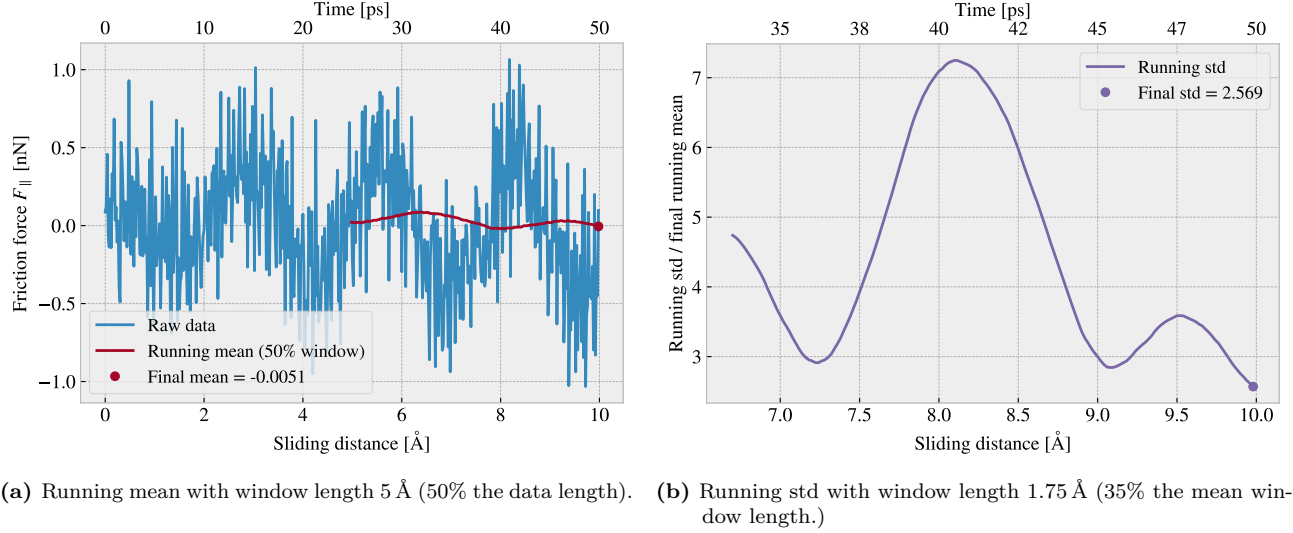


Figure 2.5: Running mean and running standard deviation (std) on the friction force data from a 10 Å of sliding simulation. The running mean window is 50% the data length while the running std window is 35% the running mean window length.

When including the full dataset of 400 Å of sliding, such that std window actually matches with the longest period of oscillations expected from the data, we get a final relative error of $\sim 12\%$ as shown in fig Fig. 2.6. This is arguable just at the limit for an acceptable error, but as we shall see later (Make a reference to fig or sec) this high relative error is mainly connected to the cases of low friction. When changing the simulation parameters, such that the mean friction evaluates to considerable higher values, the relative error drops to the order (put in numbers). One interpretation of this finding is simply that the oscillations in the running mean is somewhat independent of the magnitude of the friction. In that case, the relative error will spike for the low friction cases.

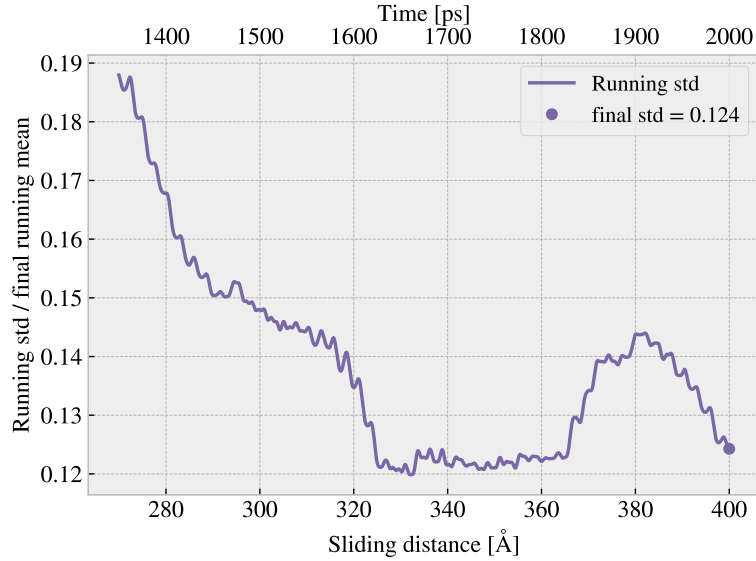


Figure 2.6: Running standard deviation (std) for a full 400 Å sliding simulation. The running std window is 70 Å (35% the running mean window of 50% the data length).

2.3.2 Static friction

The max value is the most obvious choice for addressing the static friction, even though that the definition of the static friction is a bit vague. When considering the friction force time series in Fig. 2.1 we observe that the

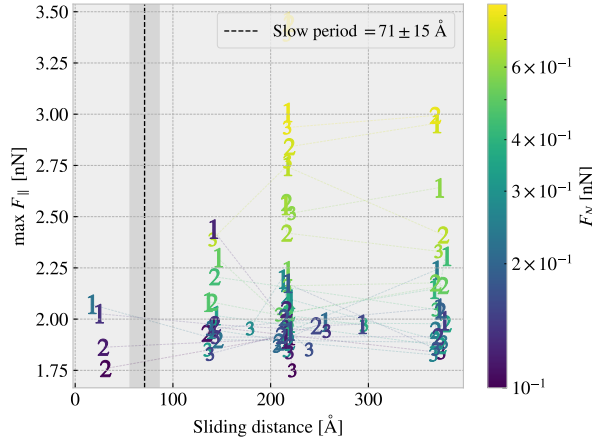


Figure 2.7: Distribution of top three max friction force peaks for 30 uniformly sampled normal forces $F_N \in [0.1, 10]$ nN. The dotted line and the grey area marks the slowest significant oscillation period found in the data and thus marking a dividing line for whether a peak falls within the “beginning” of the sliding simulation.

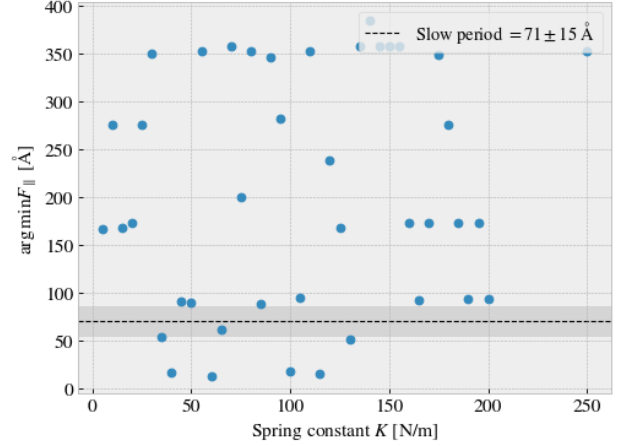


Figure 2.8: Sliding displacement for the max friction peak to appear as a function of spring constant. **Fixmove is tmp mapped to K = 200 here without any discontinuous lines.**

stick-slip oscillations increase in magnitude toward a global peak at ~ 20 Å. Thus, we could identify this peak as the static friction force, but the global max does in fact rarely fall within the first part of the sliding. In Fig. 2.7 we investigate the top three max value, at which sliding distance they occur and at what magnitude, for 30 uniformly sampled normal forces in the interval $[0.1, 10]$ nN. It is immediately clear that only few of the peaks falls within the “beginning” of the simulation defined by the slowest significant oscillation period of 71 ± 15 Å. In fact only 2/30 global values and 4/90 top three values can be associated to the start of the sliding by this definition. Thus, this result suggests that the max value cannot be used as a reliable measure for the static friction either due to its lack of presence or due to the simulation setup procedure. For a more typical evaluation of the static friction force one would increase force slowly until the first slip significant slip is recorded (a series of precursors is expected to precede this). In our simulations we drag the sheet relatively fast in a rigid manner which might be the reason for the lacking the static friction. Bonelli et al. [15] reported that the stick-slip behaviour was only presented when using a relatively soft spring. Thus, by changing the spring constant we investigate possibility to observe a static friction (I kind of interchanged stick-slip and static friction in this argument, but I still think it can be used to argue for doing the test...) response within the framework of our simulation procedure as shown in Fig. 2.8. However, the results do not indicate any implications that a recognizable domain exist for which the static friction response would be reliable. Hence, we will base the final assessment on frictional properties purely on the dynamic friction force.

2.3.3 Pressure reference for normal load domain

Find place to put this.

In order to relate the magnitude of the normal force in our friction measurement we will use the pressure as a reference. We will use the pressure underneath a stiletto shoe as a worst case for human pressure execution underneath the shoes. From (source 1) it is reported that the diameter of a stiletto heeled shoe can be less than 1 cm. Hence a 80 kg man² standing on one stiletto heel (with all the weight on the heel) will result in a pressure

$$P = \frac{F}{A} = \frac{mg}{r^2\pi} = \frac{80 \text{ kg} \cdot 9.8 \frac{\text{m}}{\text{s}^2}}{\left(\frac{1 \times 10^{-2} \text{ m}}{2}\right)^2 \pi} = 9.98 \text{ MPa}$$

²Yes, a man can certainly wear stiletto heels.

While this is in itself a spectacular realization that is often used in introductory physics courses (source 2) to demonstrate the rather extreme pressure under a stiletto heel (greater than the foot of an elephant) (how many Atmos?) this serves as a reasonable upperbound for human executed pressure. With a full sheet area of $\sim 21 \times 10^3 \text{ \AA}^2$ we can achieve a similar pressure of $\sim 10 \text{ MPa}$ with a normal force of

$$F_N = 10 \text{ MPa} \cdot 21 \times 10^{-17} \text{ m}^2 = 2.10 \text{ nN}$$

Of course this pressure might be insufficient for various industrial purposes, but with no specific procedure in mind this serves as a decent reference point. Notice that if we consider a human foot with ares 113 cm^2 the pressure drops to a mere 70 kPa corresponding to $\sim 0.01 \text{ nN}$.

2.4 Out of plane buckling

The out of plane buckling is the main motivation for applying the kirigami inspired cuts to the sheet. Thus, we perform a stretch simulation in a low temperature $T = 5 \text{ K}$ vacuum in order to verify that the chosen cut configurations do in fact contribute to a significant out of plane buckling when stretched. For the non-cut, popup and honeycomb configuration we assess the movement in the z-direction (perpendicular to the plane) during the stretch, which we visualize by the min and max z-value along with the atom count quartiles 1%, 10%, 25%, 50% (median), 75%, 90% and 99% as shown in figure Fig. 2.9. We observe that the popup and honeycomb pattern buckles considerable out of plane during the stretch in comparison to the non-cut sheet which only exhibit minor buckling of $\sim 2 \text{ \AA}$ which is on the same order as the atomic spacing in the sheet. We also notice that the popup pattern buckles more in consideration to the min and max peaks while the 1%, 99% quartiles is on the same magnitude as the honeycomb. By looking at the simulation visualization ([include OVITO figures for vacuum stretch as well?](#)) we can conclude that this is mainly due to the fringes of the sheet “flapping” around.

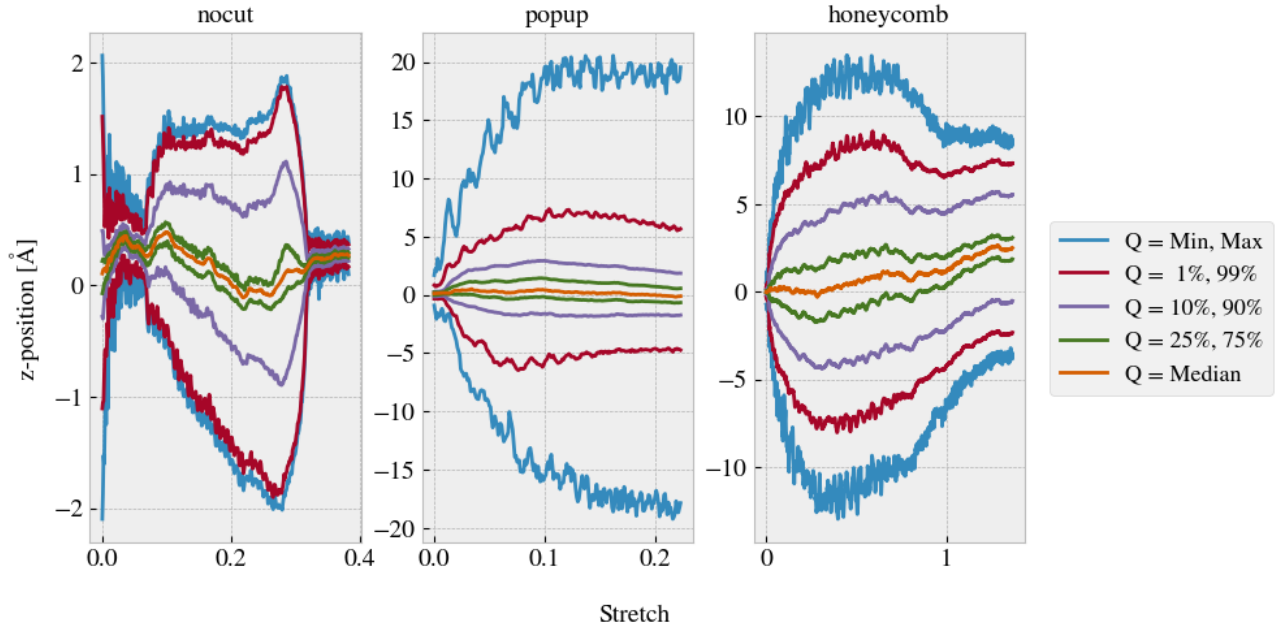


Figure 2.9: Out of plane buckling during stretch of sheets in vacuum at $T = 5 \text{ K}$. Reading from left to right the vacuum rupture stretch are 0.38, 0.22 and 1.37. [perhaps use a color scale instead of the standard color cycles here.](#)

The next step is to verify that the buckling will lead to a significant altering of the contact area when the sheet is in put in contact with the substrate. We investigate this by simulating the stretch at the default temperature $T = 300 \text{ K}$ with the presence of contact forces between the sheet and substrate. Note that no normal load is applied as the sheet and substrate is sufficiently attracted by the LJ potential. Selected frames from the simulation is shown in appendix ???. We assess the contact area by the relative amount of atoms in the sheet within chemical range of the substrate. The cut-off for this interaction is 4 \AA corresponding to $\sim 120\%$

the LJ equilibrium distance. Since the contact area is usually calculated as the amount of atoms in contact multiplied with an associated area for each contact this feature is taken to be proportional to the contact area. The relative amount of bonds as a function of stretch for the various configurations is shown in figure Fig. 2.10 which clearly indicates a drop in contact area as the cutted sheets are stretched.

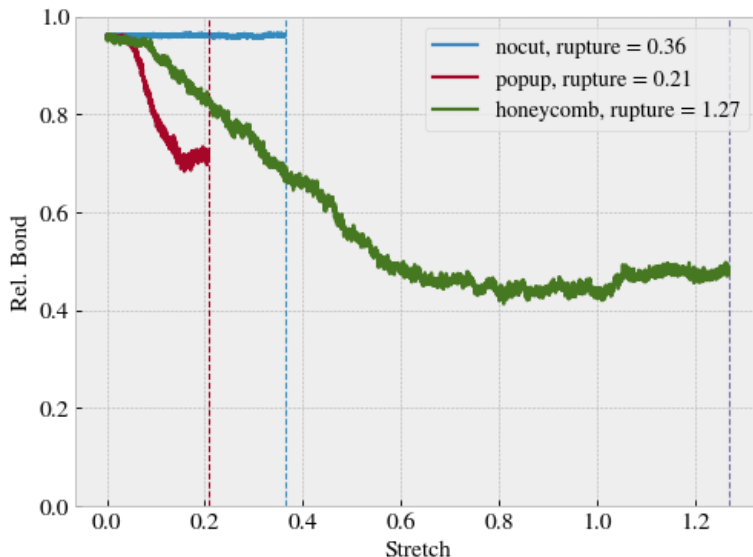
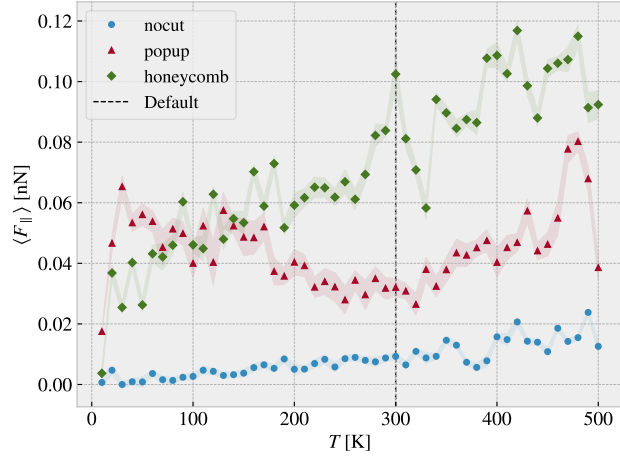


Figure 2.10: Contact vs. stretching of the sheet, where the contact is measured by the relative amount atoms in the sheet within chemical interaction range to the substrate. The cut-off for this interaction range is 4 Å corresponding to $\sim 120\%$ the LJ equilibrium distance. $T = 300$ K

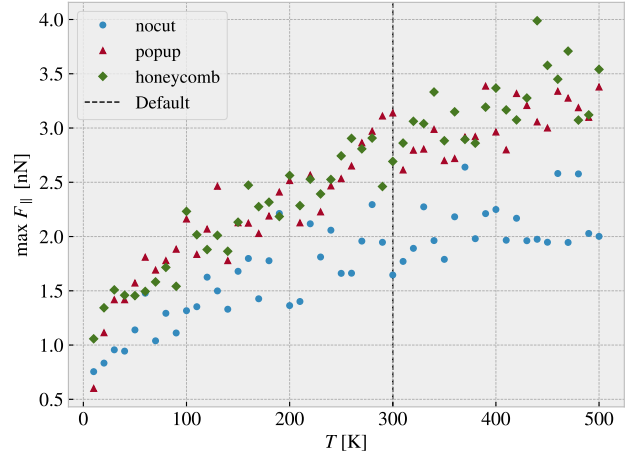
Compare figure Fig. 2.10 to that of figure Fig. 2.15 where multiple simulations constitute the stretch-contact curve.

2.5 Investigating selected parameters

We investigate the importance of the physical variables T , v_{slide} and K (make plots for scan angle as well?) and the choice of timestep dt . This is done partly to understand how the dependencies relate to theoretical, numerical and experimental results, and partly to understand how these parameter choices define the regime for our multi configurational search. We use the default parameters in ?? with exception of the single parameter of interest which is varied in a reasonable range of the default choice. In Fig. 2.11-Fig. 2.14 the dynamic friction estimate and the max friction force is shown as a function of T , v_{slide} , K and dt respectively. For the dynamic friction estimate the absolute error is denoted by a shaded error which linearly connects the points.

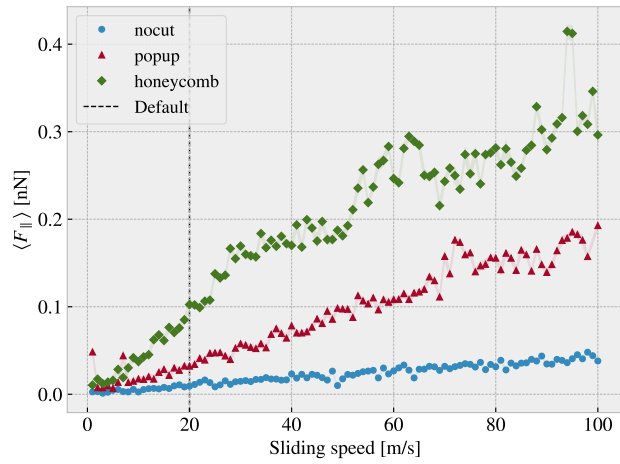


(a) Dynamic friction force estimate.

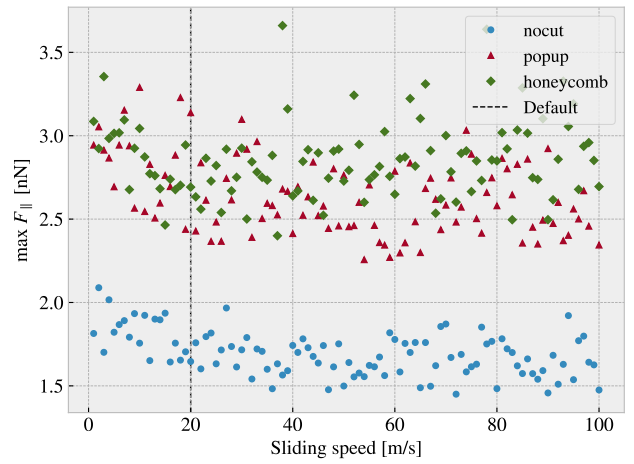


(b) Max friction

Figure 2.11: Temperature.



(a) Dynamic friction force estimate.



(b) Max friction

Figure 2.12: Sliding speed

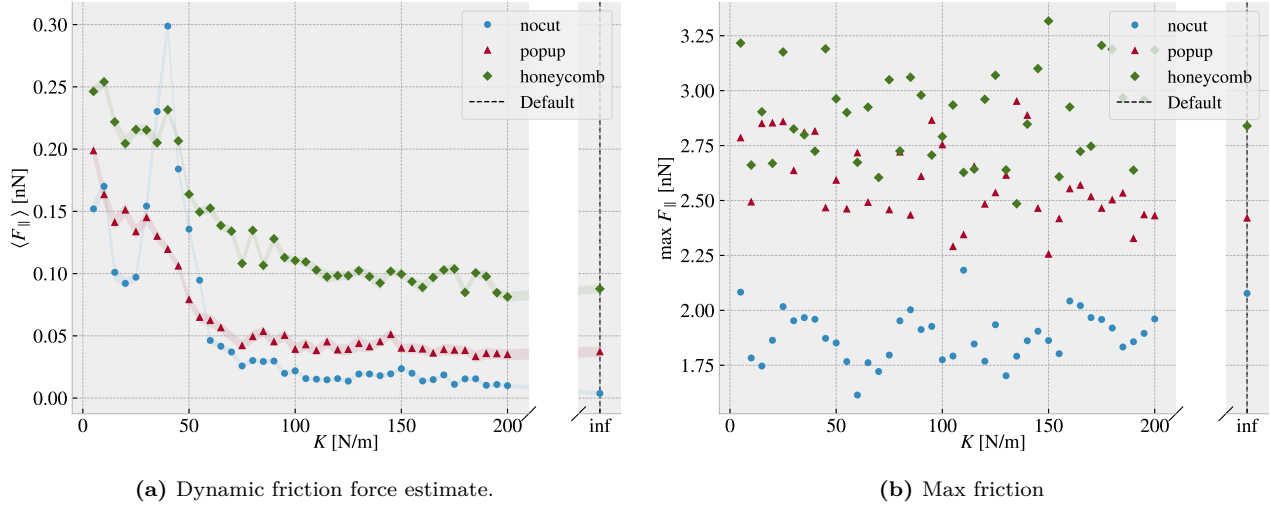


Figure 2.13: Spring constant

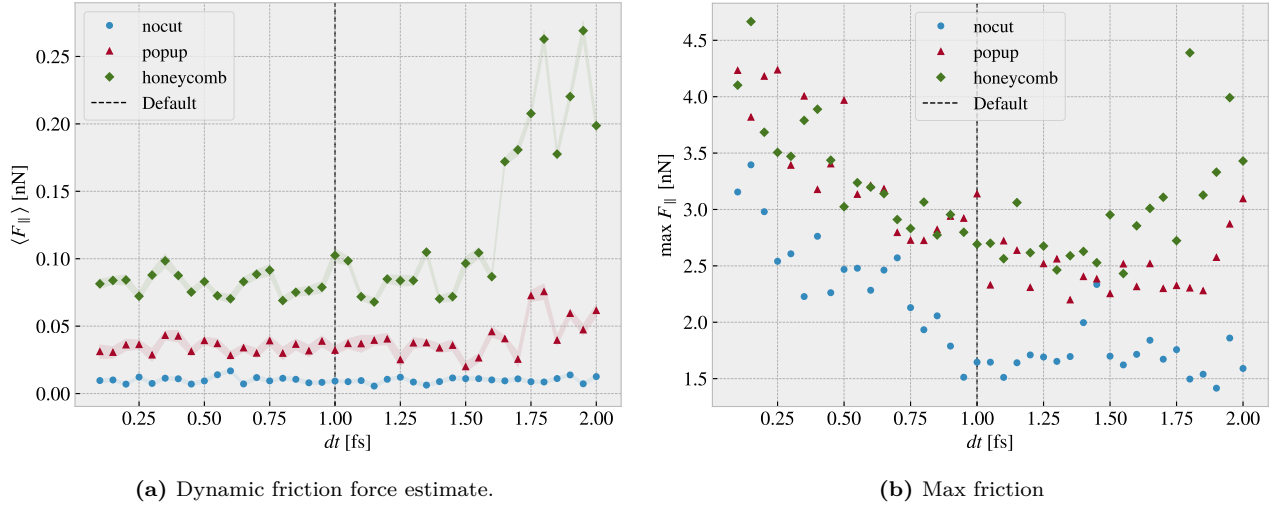


Figure 2.14: Timestep

Quick thoughts:

- Temperature: We do clearly not see the $1/T$ temperature decrease. The non-cut sheet seems to showcase a linear relationship which is also somewhat present for the honeycomb which matches some of the findings in other MD simulations. For the popup we do see a local decrease at low temperatures which flip at around the default $T = 300$ K temperature. The max friction peaks seem to increase with temperature as well indicating that the peaks might be associated with thermal fluctuations rather than actual stick-slip behaviour. This supports the finding that the static friction response is not significantly present in these simulations.
- Velcotiy: Considering the non-cut sheet first the velocity dependency is seemingly linear which deviates from the expected logarithmic trend. For the cutted configurations we find some peaks which might indicate the presence of resonance frequencies. The cutted sheet might be closer to a logarithmic trend, but this is not spot on either. The max friction seems to decrease slightly with small velocities and then stay rather constant. This can probably be explained by the reduced time to stick between stick slip.
- Spring constant: On all three configurations the dynamic friction decreases with an increasing spring constant. The best explanations might be due to the lack of freedom to “get stuck” in incommensurable

configurations. We also notice that the friction varies a lot at lower spring constants supporting the choice of having a stiff spring for stability reasons. Especially the non-cut sheet peaks at $K = 40 \text{ N/m}$. The max friction seem to be constant with K .

- dt : The dynamic friction is relatively stable around the default choice of $dt = 1 \text{ fs}$. However, the fluctuations with respect to dt is more significant for popup pattern and even more for the honeycomb pattern. This indicates that the more complex dynamics of the simulation is more sensitive to the timestep. We might interpret this information as an additional measure of uncertainty. The maximum friction decreases with increasing timestep which can be asserted a statistical interpretation: Higher peaks will be captured by the high resolution of a low dt and vice versa. The high max values towards the point of $dt = 2 \text{ fs}$ is most likely due to the approach of instability in the simulation as seen more clearly for the dynamic friction evaluation.

2.6 Normal force and stretch dependencies

Till this point we have only changed variables one by one to investigate single dependencies. We now advance the study to a simultaneous variation of stretch and normal force.

Explain how the stretch is uniformly sampled within equally divided intervals and the normal force is actually uniformly sampled in a given range. Argue that the first might be approximately uniformly distributed for large numbers.

Talk about rupture test also. Maybe in the theory/method section under numerical procedure: Before simulating a rupture test is perform to determine under what stretch the sheet ruptures. This is a slightly higher threshold than when applied normal load and sliding along the substrate.

2.6.1 Contact area

??

We reproduce the contact area investigation of Fig. 2.10 with the modification that the contact count is measured as an average of the latter 50% of the sliding simulation at a non-zero applied normal load. The results are shown in Fig. 2.15 with 30 attempted (some rupture) stretch (pseudo) uniformly distributed stretch between 0 and the rupture point and 3 uniform distributed normal loads in the interval $[0.1, 10] \text{ nN}$.

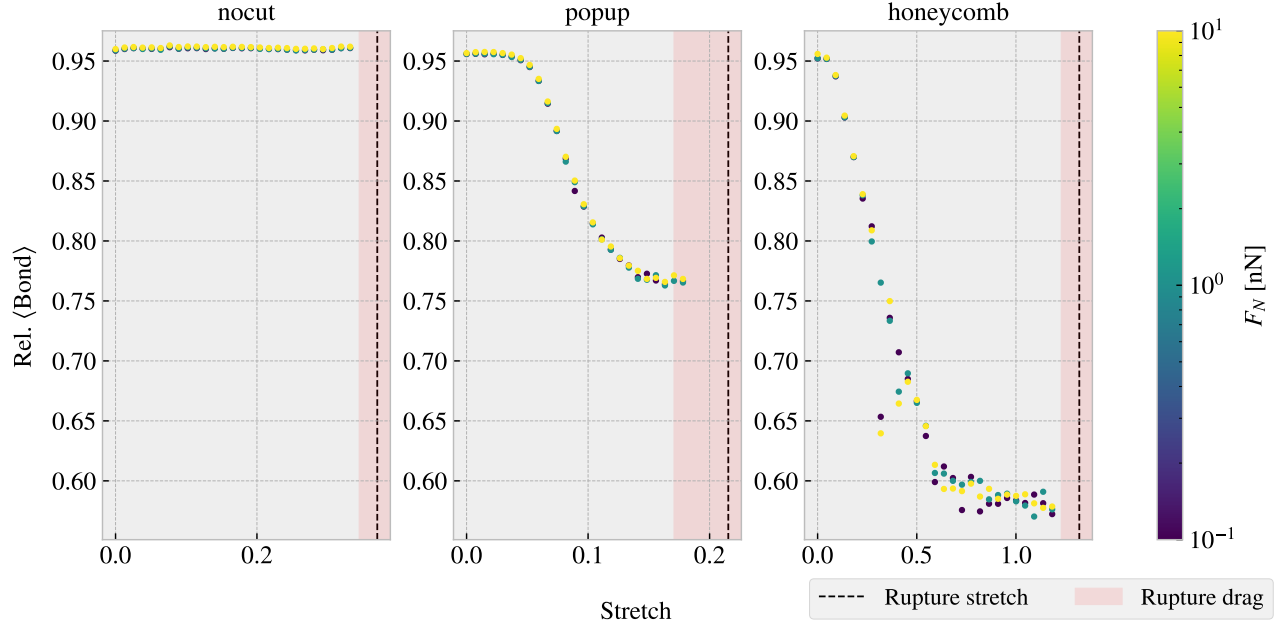
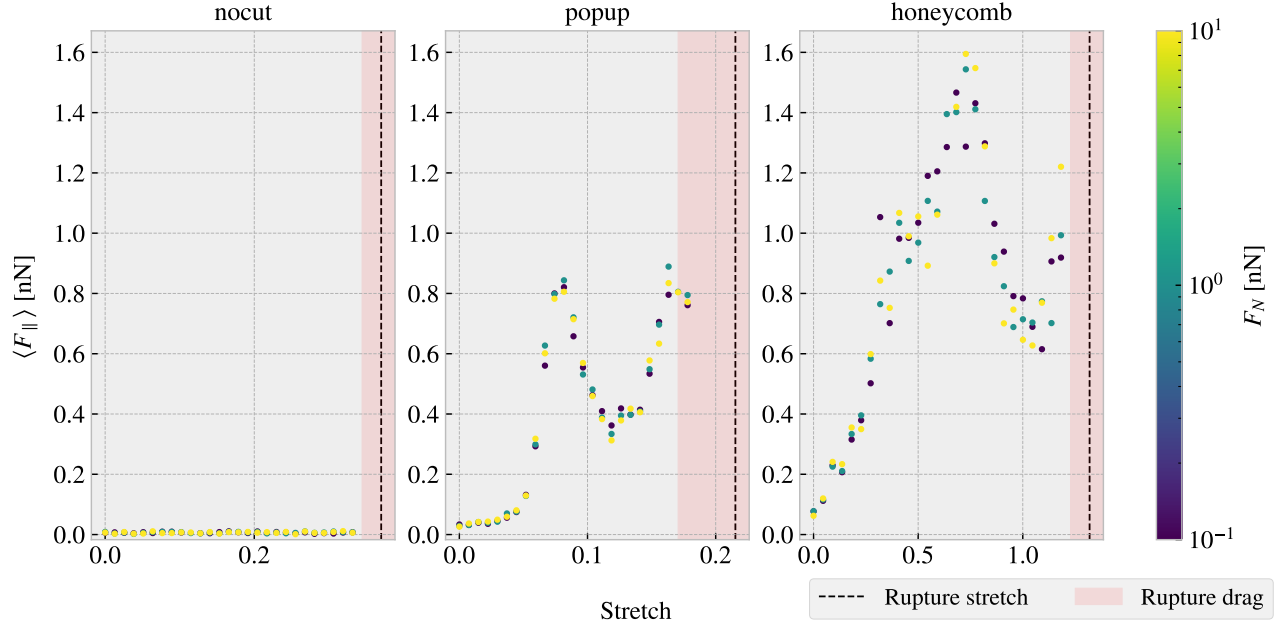


Figure 2.15: Average relative amount of bonds between the sheet and the substrate defined by the cut-off distance of 4 Å. The average is taken over the latter half of the sliding phase. The red shade denotes the stretch range where ruptures occur at certain normal loads under sliding while the black-dotted line represent the rupture point due to stretching (rupture test)

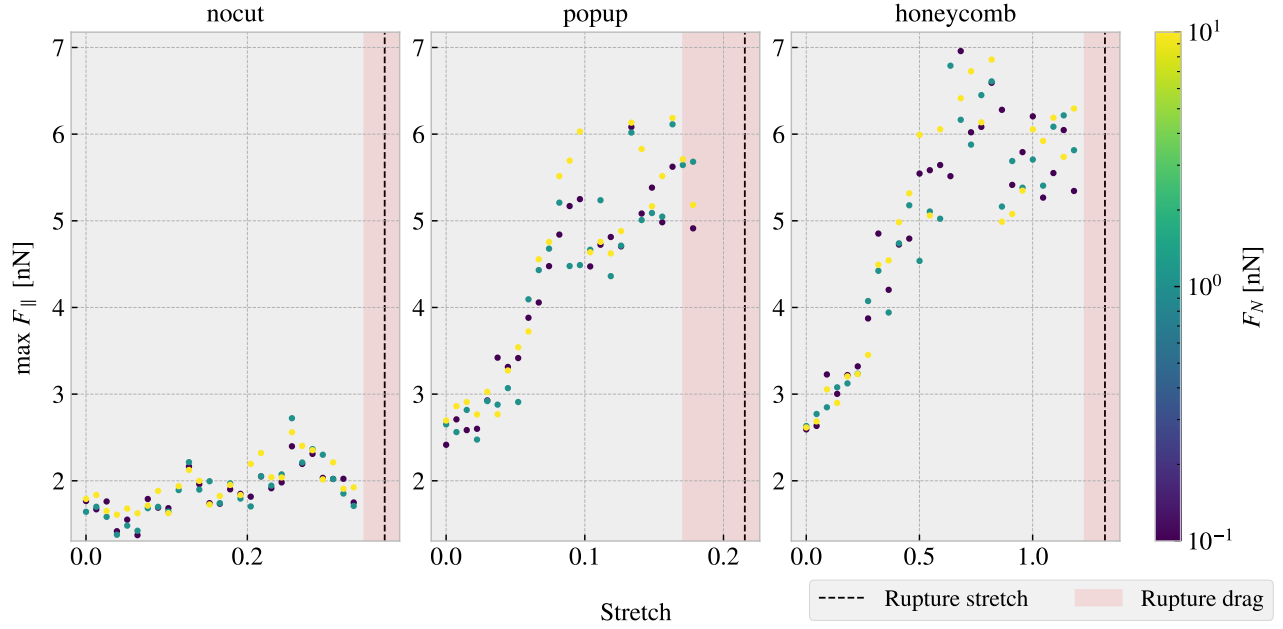
From Fig. 2.10 we observe a significant decrease in the contact due to stretching of the cut configurations in contrast to the non-cut which stays roughly constant. This is reminiscent of the non-sliding stretch vs. contact curve shown in Fig. 2.10. Given these results, theoretically one would expect the dynamic friction to decrease with stretch for the cut configurations.

2.6.2 Stretch

We make a similar analysis as done in the previous section ?? with the substitution of friction force instead of contact (The data is taken from the same simulations runs). The dynamic friction force (put uncertainty here even though that it is quite low?) and the max friction is shown in Fig. 2.16a and Fig. 2.16 respectively.



(a) Dynamic friction estimate.



(b) Max friction

Figure 2.16: CAPTION

From Fig. 2.16a we find to our surprise that the dynamic friction increase with stretch for the cut configurations despite a simultaneous decrease in contact area as shown in figure Fig. 2.15. This suggests that the amount of chemical bonding atoms is not the dominant mechanism for the friction of this system. Instead, we might point to a mechanism more mechanical of nature associated to phonon excitations. When the cut sheet is stretched the stress (show stress maps somewhere or not necessary?) might induce a certain distribution and magnitude of point pressures to favor energy dissipation. Nonetheless, the results showcase a strong coupling between stretch and friction force, also for the max friction force, which is beyond the expectations at this stage of the study. The non-cut configuration does not show significant dependency on the stretch which reveal that this effect is only present when combining cut and stretch and not purely by stretching the sheet.

By considering the increase in dynamic friction towards the first peak we get a relative friction increase and increase vs. stretch ratios as described in Table 2.2. While the honeycomb force increase towards the first peak is approximately linear the popup exhibits seemingly exponential growth which yield a slope on the order ~ 30 nN.

Table 2.2: (stretch, dynamic friction) coordinates from Fig. 2.16a at start and the first peak respectively used to approximate the relative increase in friction force and the ratio for friction increase vs. stretch for said range. In practice the latter ratio denotes the slope of a forced linear trend.

Configuration	Start	First peak	Relative increase	Friction force vs. stretch ratio [nN]
Popup	$\sim (0, 0.03)$	$\sim (0.082, 0.83)$	27.7	9.76
Honeycomb	$\sim (0, 0.07)$	$\sim (0.728, 1.57)$	22.4	2.06

Additionally, we notice that both the popup and honeycomb also exhibits stretch ranges where the dynamic friction force decrease with increasing stretch. Qualitatively we assign the slope to be on the same order of magnitude as those towards the first peak. This is useful for the prospect of taking advantage of this phenomenon as we can essentially achieve both higher and lower friction for increasing stretch for different starting points.

2.6.3 Normal force

Main take away from this section should be that the normal force does not really change the friction much; The friction coefficient is extremely low, but I'm not sure how well the linear fits are (whether they are linear or sublinear). Not sure if I should do a linearly increasing normal force for better linear plots?

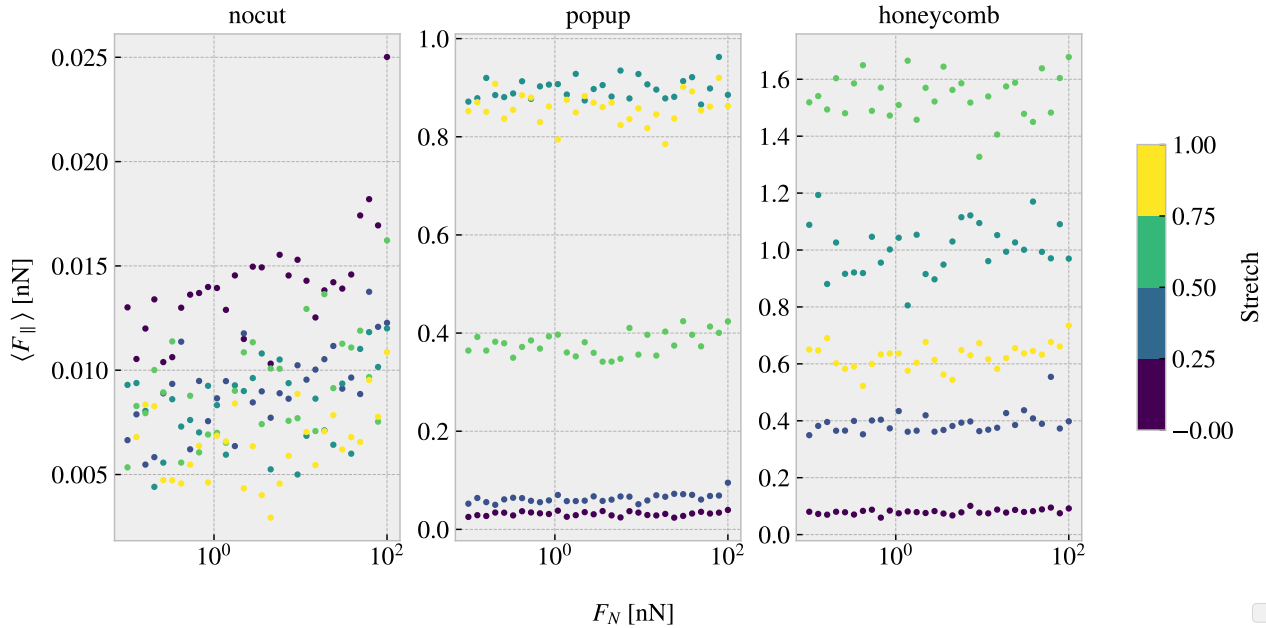


Figure 2.17: ...

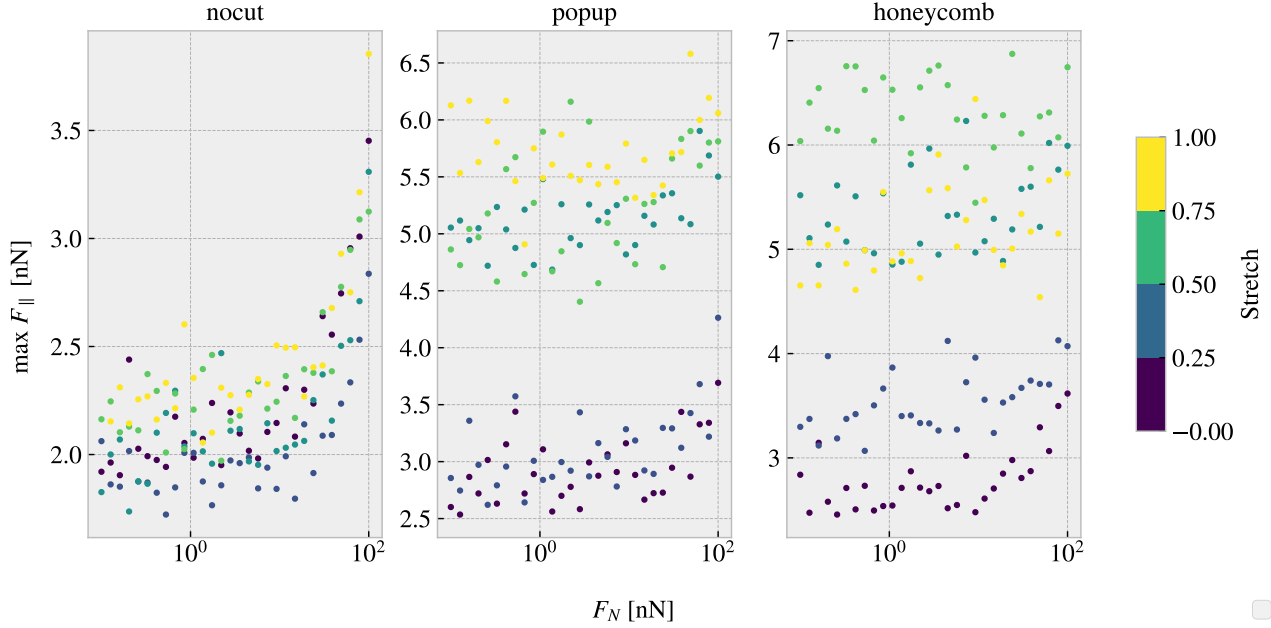


Figure 2.18: Colorbar is only fitted for the right plot (honeycomb)... this should be fixed. Should I have run a linear distribution of FN so I could plot it linear here also...?

Table 2.3: Mean friction coeff

nocut	$0.00009 \pm 1 \times 10^{-5}$	$0.00005 \pm 1 \times 10^{-5}$	$0.00004 \pm 1 \times 10^{-5}$	$0.00005 \pm 2 \times 10^{-5}$	
popup	$0.00005 \pm 3 \times 10^{-5}$	$0.00024 \pm 5 \times 10^{-5}$	$0.0002 \pm 2 \times 10^{-4}$	$0.0005 \pm 1 \times 10^{-4}$	$0.0003 \pm 2 \times 10^{-4}$
honeycomb	$0.00013 \pm 6 \times 10^{-5}$	$0.0006 \pm 3 \times 10^{-4}$	$0.0004 \pm 6 \times 10^{-4}$	$0.0007 \pm 6 \times 10^{-4}$	$0.0009 \pm 3 \times 10^{-4}$

Table 2.4: Max friction coeff

nocut	$0.0139 \pm 9 \times 10^{-4}$	$0.0083 \pm 7 \times 10^{-4}$	$0.010 \pm 1 \times 10^{-3}$	$0.0105 \pm 9 \times 10^{-4}$	
popup	$0.007 \pm 2 \times 10^{-3}$	$0.010 \pm 2 \times 10^{-3}$	$0.007 \pm 2 \times 10^{-3}$	$0.009 \pm 3 \times 10^{-3}$	$0.006 \pm 2 \times 10^{-3}$
honeycomb	$0.010 \pm 1 \times 10^{-3}$	$0.007 \pm 2 \times 10^{-3}$	$0.007 \pm 3 \times 10^{-3}$	$0.000 \pm 3 \times 10^{-3}$	$0.004 \pm 3 \times 10^{-3}$

One theory for the low friction coefficient might dependent on the fact that the normal force is only applied on the pull blocks. Especially with the cutted sheet the tension drops such that the effective normal force on the inner sheet is not changing very much. By this theory the friction force vs. normal force on the pull blocks should look a bit more like expected and we might make some plots of those to check

When looking at the graphs for the PB the max friction is visually textbook linear, while the mean friction is a bit more linear but also with negative coefficients...

2.7 Computational cost

Talk about the computational cost of different choices. How does computation time scale with drag speed, dt and maybe T and K as well. One could also mention scaling with system size.

Show how the number of cores per simulation scale to argue that running on just one core (maybe 4) is smart for the next step of many simulations.

Mention the trouble with GPU to show that this was considered, and in fact this was the reason for choosing the Tersoff potential over the AIREBO which is perhaps more common these days...

Appendices

Appendix A

Appendix B

Appendix C

Bibliography

- ¹E. Gnecco and E. Meyer, *Elements of friction theory and nanotribology* (Cambridge University Press, 2015).
- ²Bhusnan, “Introduction”, in *Introduction to tribology* (John Wiley & Sons, Ltd, 2013) Chap. 1, 1–?
- ³H.-J. Kim and D.-E. Kim, “Nano-scale friction: a review”, *International Journal of Precision Engineering and Manufacturing* **10**, 141–151 (2009).
- ⁴K. Holmberg and A. Erdemir, “Influence of tribology on global energy consumption, costs and emissions”, *Friction* **5**, 263–284 (2017).
- ⁵B. Bhushan, “Gecko feet: natural hairy attachment systems for smart adhesion – mechanism, modeling and development of bio-inspired materials”, in *Nanotribology and nanomechanics: an introduction* (Springer Berlin Heidelberg, Berlin, Heidelberg, 2008), pp. 1073–1134.
- ⁶P. Z. Hanakata, E. D. Cubuk, D. K. Campbell, and H. S. Park, “Accelerated search and design of stretchable graphene kirigami using machine learning”, *Phys. Rev. Lett.* **121**, 255304 (2018).
- ⁷P. Z. Hanakata, E. D. Cubuk, D. K. Campbell, and H. S. Park, “Forward and inverse design of kirigami via supervised autoencoder”, *Phys. Rev. Res.* **2**, 042006 (2020).
- ⁸L.-K. Wan, Y.-X. Xue, J.-W. Jiang, and H. S. Park, “Machine learning accelerated search of the strongest graphene/h-bn interface with designed fracture properties”, *Journal of Applied Physics* **133**, 024302 (2023).
- ⁹Y. Mao, Q. He, and X. Zhao, “Designing complex architected materials with generative adversarial networks”, *Science Advances* **6**, eaaz4169 (2020).
- ¹⁰Z. Yang, C.-H. Yu, and M. J. Buehler, “Deep learning model to predict complex stress and strain fields in hierarchical composites”, *Science Advances* **7**, eabd7416 (2021).
- ¹¹A. E. Forte, P. Z. Hanakata, L. Jin, E. Zari, A. Zareei, M. C. Fernandes, L. Sumner, J. Alvarez, and K. Bertoldi, “Inverse design of inflatable soft membranes through machine learning”, *Advanced Functional Materials* **32**, 2111610 (2022).
- ¹²S. Chen, J. Chen, X. Zhang, Z.-Y. Li, and J. Li, “Kirigami/origami: unfolding the new regime of advanced 3D microfabrication/nanofabrication with “folding””, *Light: Science & Applications* **9**, 75 (2020).
- ¹³Z. Deng, A. Smolyanitsky, Q. Li, X.-Q. Feng, and R. J. Cannara, “Adhesion-dependent negative friction coefficient on chemically modified graphite at the nanoscale”, *Nature Materials* **11**, 1032–1037 (2012).
- ¹⁴R. W. Liefferink, B. Weber, C. Coulais, and D. Bonn, “Geometric control of sliding friction”, *Extreme Mechanics Letters* **49**, 101475 (2021).
- ¹⁵F. Bonelli, N. Manini, E. Cadelano, and L. Colombo, “Atomistic simulations of the sliding friction of graphene flakes”, *The European Physical Journal B* **70**, 449–459 (2009).