

Title

Subtitle

by

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Title

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Abstract

Abstract.

Acknowledgments

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List of symbols?

Maybe add list of symbols and where they are used like Trømborg.

Introduction

0.1 Motivation

0.1.1 Friction

Friction is a fundamental force that takes part in almost all interactions with physical matter. Even though the everyday person might not be familiar with the term “friction” we would undoubtedly notice its disappearing. Without friction, it would not be possible to walk across a flat surface, lean against the wall or secure an object by the use of nails or screws (Static friction allows us to join objects together using screws [1][p. 5]). Similarly, we expect a moving object to eventually come to a stop if not supplied with new energy, and we know intuitively that sliding down a snow covered hill is much more exciting than its grassy counterpart. It is probably safe to say that the concept of friction is well 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]. Friction is 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 important motivation to study tribology is ultimately to gain full control of frictional and wear for various technical applications. Especially, reducing friction is of great interest as this has tremendous advantages regarding energy efficiency. It has been reported that the monetary value of tribological problems has 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].

The reduction of friction is not the only sensible application as a controlled increase in friction might be of interest in the development of grasping robots or perhaps breaking system (get some sourced examples maybe...).

To the best of my knowledge kirigami has not yet been implemented to alter the friction properties in a similar manner as done in this thesis.

0.1.2 Thesis

In this thesis we investigate the possibility to control the frictional properties of a graphene sheet by applying strategically positioned cuts to the sheet inspired by kirigami. Kirigami is a variation of origami where the paper is cut additionally to being folded. Hanakata et al. [5] has shown that kirigami inspired cuts on a graphene sheet can be used to alter the yield strain and yield stress of the sheet. They observed that the stretching of the cutted sheet induced a out-of-plane buckling which serves as a key observation for the motivation of this thesis. It is currently well established/believed that the friction between two surfaces is proportional to the real microscopic contact area (source here?). Hence, one can hypothesize that the buckling of the sheet will affect the contact area and consequently the frictional properties.

0.2 Approach

In the study by Hanakata et al. [5] they used a machine learning (ML) approach to overcome the complexity of the nonlinear effects arising from the out-of-plane buckling which made them successfully map the cutting patterns to the mechanical properties of yield and stress. The dataset used for the ML training was generated by molecular dynamics (MD) simulations for a limited set of cut configuration. By training the network the

MD simulaitions could effectively be skipped all together making for an accelerated search through new cut configurations for certain mechanical properties. By setting up a MD simulation that qunatifies the frictional properties of the graphene sheet we aim to make an analog study regarding the search for certain frictional properties.

We will take this on step further by creating a GAN network that utilises the latter network for creating an inverse design framework. That is, a network that takes frictional properties as input and return the corresponding cut configuration. By having such a tool we can execute a targeted search for exotic frictional properties. Particularly, we are interested in nonlinear and possibly even negative friction coefficients. Friction is essentially observed to increase with increasing load on the frictional surface, and we often describe this as having a positive friction coefficient. However, if we are able to couple the stretching of the sheet with friction we might be able to break this barrier for the coefficient. By imagining some nanomachine which translates downward pressure into either compression or expansion of the altered graphene, we could have a coupling between downward pressure and stretch of the sheet. In that case, a friction force depending on stretch could effectively be made to decrease with increasing load which would correspond to a negative friciton coefficient following this definition (formulate such that we do not imply free acceleration from nothing).

One of the features from inverse design, seperating it from the general class of ML approaches, is that we do not depend on trusting the ML predictions. While a standard neural network might be extremely efficient on a certain prediciton task we have usually no information on how these predictions are based. We say that the internal workings of the network is a black box beyond our capaacity of interpretation. However, for the inverse design problem we are prompted with a few promising design proposals which can immediately be tested in the MD simulations which we will regard as the most reliable predictor in this setting. Hence, if arriving at a successful design in alignment our search prompt, we can disregard any uncertainty in the network. In that case the remaining gap to bridge is that of the MD simulation and real life implementations.

0.3 Objective of the study

1. Design a MD simulation to evaluate the frictional properties of the grpahene sheet under different variations of cut patterns, stretching and loading, among other physical variables.
2. Train a network to replace the MD simulation completely.
3. (Variation 1) Do an accelerated search using the ML network for exotic frictional properties such as low and friction coefficients and a strong coupling between stretch and friction.
4. (Variation 2) Make a GAN network using the first network and predict cut configurations for some of the above mentiond frictional properties.
5. (If I have time) Make a nanomachine that couples load and stretch (perhaps just artificially without any molecular mechanism) to test the hypothesize of a negativ friction coefficient.

0.4 Contributions

What did I actually achieve

0.5 Thesis structure

How is the thesis structured.

Chapter 1

Background Theory and Method

Small introtext to motivate this chapter. What am I going to go over here.

1.1 Tribology - friction

Friction is a part of the wider field tribology which includes the study of friction, wear and lubrication between two surfaces in relative motion [1, p. 1]. In this thesis we will only concern ourselves with the first, wearless dry friction. Tribological systems take place across a broad range of time and length scales, ranging from geological stratum layers involved in earthquakes [3] to microscopic atomistic processes, as in the gliding motion of nanocluster of a nanomotor [6]. This vast difference in scale gives rises to different frictional mechanism being dominating at different scales. On a macro scale the system is usually inflicted to relatively high large loads and speeds leading to high contact stresses and wear. The micro-/nanoscale regime occupies the opposite domain operating under relatively small loads and speeds with negligible wear [3] [2, p. 5].

While macroscale friction is often reduced into a few variables such as normal load, material type, speed and surface roughness it is clear that micro-/nanoscale friction cannot be generalized under such a simple representation. On the micro-/nanoscale the tribological properties is dominated by surface properties which will introduce additional sensitivity to more variables such as temperature, humidity and even sliding history. The works of Bhushan and Kulkarni (1996)[7] showed that the friction coefficient decreased with scale, revealing the change in the mechanism of friction as the contact condition was altered, even though the materials used was unchanged.

The phenomenological descriptions of macroscale friction cannot yet be derived from the fundamental atomic principles, and bridging the gap between different length scales in tribological systems remains an open challenge [6]. Hence, the following sections will be organized into macro-, micro- and nanoscale representing the different theoretical understanding governing each scale. While our study of the graphene sheet is based on a nanoscale perspective the hypothesizing about application possibilities will eventually link back to the macroscale perspective. Thus spanning all three major scales in a brief becomes useful for a more complete interpretation of the findings of this thesis.

1.1.1 Macroscale

Our working definition of the *macroscale* is everything on the scale of visible everyday objects, which is usually denoted to millimeters 10^{-3} m and above. Most importantly, we want to make a distinction to the microscale, where the prefix indicates the size of micrometers m^{-6} , and hence we essentially assign everything larger than micro into the term macroscale¹.

1.1.1.1 Amontons' law

In order to start and keep a solid block moving against a solid surface we must overcome certain frictional forces F_{fric} [1]. The static friction force F_s corresponds to the minimum tangential force required to initiate the sliding

¹The width of a human hair is on the length scale 10^{-5} to 10^{-4} m which constitute a reasonable boundary between macro- and microscale which fit well with a lower bound of our perception capabilities.

while the kintec friciton force F_k corresponds to the tangential force needed to sustain such a sliding at steady speed. The work of Leonardo da Vinci (1452–1519), Guillaume Amontons (1663–705) and Charles de Coulomb (1736–1806) all contributed to the empirical law, commonly known Amontons' law, which is a common baseline for an introduction to friction. Amontons' law states the fricitonal forces is entirely independent of contact area and sliding velocity (at ordinary sliding velocities). Instead, it relies only on the normal force² F_N , acting perpendicular to the surface, and the material specific friction coefficient μ as

$$F_{\text{fric}} = \mu F_N. \quad (1.1)$$

The friction coefficient is typically different for the cases of static (μ_s) and kinetic (μ_k) friction, usually with values lower than one and $\mu_s \geq \mu_k$ in all cases [1, p. 6].

Allthough Amontons' law has been succesfull in the modelling of macroscale friction it has its limitations. For instance, it was later discovered that the staite friction is not independent of time. It depends on the so-called contact history with increasing friction as the logarithm of time of stationary contact [8]. For the kinetic friction the independency of sliding velocity dissapears at low velocities as thermal effects becomes important and for high velocities due to inertial effetc. [1, pp. 5-6].

While Amontons' law lacks any attempt to explain the phenomena of friction statistical mechanics provides an overall explanation by the concept of *equipartition of energy* [6]. A system in equilibrium has its kinetic energy uniformly distributed among all its degrees of freedom. When a macroscale object is sliding in a given direction it is clearly not in equilibrium since one of its degrees of freedom carriers considerable more kinetic energy. Thus, the system will have a tendency to transfer the kinetic energy to the remaining degrees of freedom as heat. This heat will dissipate to the surroundings and the object will slow down as a final result. Hence, friction is really just the tendency of going toward equilibrium energy equipartitioning among many interacting degrees of freedom. [6]

By including *statistical mechanics* into the discussion we have already taken a peek into lower scales as this field of science explain macroscale behaviour by microscopic interactions. In fact, this marks the limiting view of most macroscale theories: They lack fundamental explanations.

1.1.2 Microscopic scale

Going from a macro- to microscale perspective, a length scale of order $10^{-6}m$, it was realized that most surfaces is in fact rough [9]. The contact between two surfaces consist of numerous smaller contact point, so-called asperities, for which the friction between two such opposing surfaces involves interlocking of those asperities as visualized in figure 1.1. It is generally accepted that friction is caused by two mechanism: mechanical friction and chemical friction [3]. The mechanical friction is the “plowing” of the surface by hard particles or said asperities with an energy loss contributed to plastic deformations (allthough this assumption of plastic deformations has been critizised [10]). The chemical friction arrises from adhesion between microscopic contacting surfaces, (with energy loss contributed breaking and forming of bonds?).

1.1.2.1 Surface roughness - Asperity theories

Asperity theories are based on the observation that microscopic rough surfaces with contacting asperities, each with a contact area of A_{asp} , will have a true contact area $\sum A_{\text{asp}}$ which is found to be much smaller than the apperent macroscopic area A_{macro} [3]. The friction force was shown to be proportional to the true contact area as

$$F_{\text{fric}} = \tau \sum A_{\text{asp}},$$

where τ is an effective shear strength of the contacting bodies. This is in principle still compatible with Amontons' law (eq. (1.1)) if we find a linear relationship between the real contact area and the applied normal force F_N . In figure 1.1 we see a visualization on how the contact area might intuitively increase with normal force as the asperity tips is deformed into broader contact points.

²Normal force is often used interchangeably with the terms *load* and *normal load*.

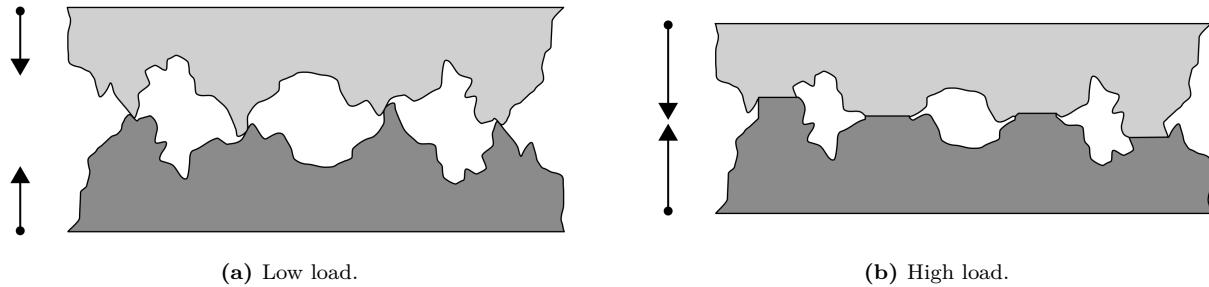


Figure 1.1: Qualitatively illustration of the microscopic asperity deformation under increasing load from frame a to b [11].

Many studies have focused on the individual single asperity contact to reveal the relationship between the contact area and F_N (13-15 from [9]). By assuming perfectly smooth asperities with radii of curvature from micrometers all the way down to nanometers continuum mechanics can be used to predict the deformation of asperities as normal force is applied. A model for non-adhesive contact between homogenous, isotropic, linear elastic spheres was first developed by Hertz (17 [9]), which predicted $A_{\text{asp}} \propto F_N^{2/3}$. Later adhesion effects were included in a number of subsequent models, including Maugis-Dugdale theory (18 from [9]), which also predicts a sublinear relationship between A_{asp} and f_N . Thus, the common feature of all single-asperity theories is that A_{asp} is a sublinear function of F_N , leading to a similar sublinear relationship for $F_{\text{fric}}(F_N)$, which fails to align with the macroscale observations modelled by Amontons' law (eq. (1.1)).

A variety of multiasperity theories has attempted to combine single asperity mechanics by statistical modelling of the asperity height and spatial distributions [10]. This has led to (in certain domains) a linear relationship between contact area and normal force. Unfortunately, these results are restricted in terms of the magnitude of the load and contact area, where multiasperity contact models based on the original ideas of Greenwood and Williamson [12] only predicts linearity at vanishing low loads, or Persson [13] which works for more reasonable loads but only up to 10-15 % of the macroscale contact area. However, as the load is further increased all multiasperity models predict the contact area to fall into the sublinear dependency of normal force as seen for single asperity theories [10].

1.1.3 Nanoscale - Atomic scale

Going from microscale to nanoscale on the order of 10^{-9} meter, it has been predicted that continuum mechanics will break down [14] due to atomic-scale surface roughness. In a numerical study by Mo et al. [9] it has been shown that the asperity area A_{asp} is in fact sublinear with F_N , but this is explained by the fact that not all atoms within the circumference of the asperity make contact with the substrate. By modelling the real area $A_{\text{real}} = N A_{\text{atom}}$, where N is the amount of atoms within the range of chemical interaction range and A_{atom} is the associated atom surface area, they found a consistent linear relationship between friction and real contact area. Without adhesive forces this lead to a similar linear relationship $F_{\text{fric}} \propto F_N$, while adding van der Waals adhesion to the simulation gave a sublinear relationship, even though the $F_{\text{fric}} \propto A_{\text{real}}$ was maintained.

This result shows that the relationship between contact area and friction is expected to be a dominant factor even at the atomic scale. However, considering the simulation setup of our numerical study, a flat sheet on a flat substrate, it is clear that we cannot rely directly on asperity theories even though they might be somewhat successful at the nanoscale. On the smallest possible scale, atomic scale, the surfaces do not have structural asperities. Instead, atomic level friction is being modelled as a consequence of the rough potential of the atomic landscape and real contact area defined in terms of atomic bonding.

Search for experimental and numerical studies of nanoflakes (ideally with graphene) to talk about expectations here.

1.1.3.1 Frenkel-Kontorova

The Frenkel-Kontorova (FK) model builds upon the time-honored PT model describing a point-like tip sliding over a space-periodic crystalline surface. The FK model extends this description to that of a 1D crystalline chain sliding on 1D crystalline surface as illustrated in 1.2.

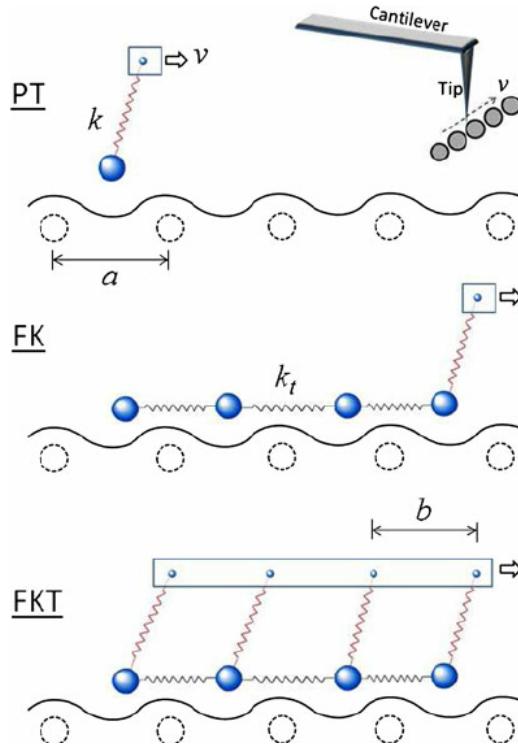


Figure 1.2: Temporary figure from https://www.researchgate.net/figure/illustrations-of-the-1D-PT-FK-and-FKT-models-Largefig1_257670317

The standard FK model consists of a 1D chain of N classical particles, representing atoms, interacting via a harmonic forces and moving in a sinusoidal potential as sketched in figure 1.3. The hamiltonian is

$$H = \sum_{i=1}^N \left[\frac{p_i^2}{2m} + \frac{1}{2}K(x_{i+1} - x_i - a_c)^2 + \frac{1}{2}U_0 \cos\left(\frac{2\pi x_i}{a_b}\right) \right], \quad (1.2)$$

where the first term $p_i^2/2m$ represents the kinetic energy with momentum p and mass m . The next term describes the harmonic interaction with elastic constant K , nearest neighbour distance $\Delta x = x_{i+1} - x_i$ and corresponding nearest neighbour equilibrium distance a_c . The final term represents the periodic substrate potential (external potential on site) with amplitude U_0 and period a_b . Different boundary choices can be made, with both free ends or periodic conditions being reasonable. For fixed ends the chain will never be able to slide off.

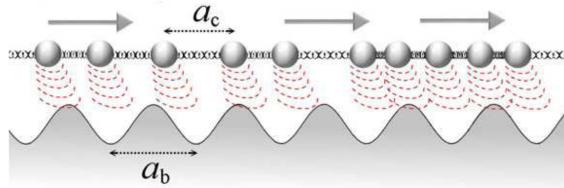


Figure 1. A sketch of the FK model, showing the two competing lengths: the average interparticle spacing and the lattice periodicity of the substrate.

Figure 1.3: Temporary figure from [6]

By applying an external force to the chain one can use the FK model to investigate static friction. It turns out that the sliding properties are entirely governed by its topological excitations referred to as so-called *kinks* and *antikinks*.

Commensurability We can describe the frictional behaviour in terms of commensurability, that is, how well thus the spacing of the atoms match the periodic substrate potential. We describe this by the winding number/coverage/length ratio $\theta = N/M = a_b/a_c$ where M denotes the number of minemas in the potential (in the range of the chain). A rational number for θ means that atoms align with minemas corresponding to a *commensurate* case. If θ is irrational the chain and substrate cannot fully align, and we denote this as an *incommensurable* case.

Let us begin with the simplest commensurate case of $\theta = 1$ where the spacing of the atoms matches perfectly the substrate potential periodicity, i.e. $a_c = a_b$, $N = M$. The ground state (GS) is the configuration where each atom fits in one of the substrate minema. By adding an extra atom we would need to shift over some atoms from this ideal state giving rise to a so-called kink excitation, i.e. two atoms will have to share the same potential corrugation as shown in figure 1.5. On the other hand, removing an atom from the chain results in a so-called antikink excitation where one potential corrugation will be left “atomless”. In order to reach a local minimum the kink (antikink) will expand in space over a finite length such that the chain undertakes (right word) a local compression (expansion). When applying a tangential force to the chain it is much easier for a kink to move along the chain than it is for non-excited atoms since the activation energy ϵ_{PN} for a kink displacement is systematically smaller (often much smaller) than the potential barrier U_0 . Thus, the motion of kinks (antikinks), i.e. the displacement of extra atoms (atom vacancies), is representing the fundamental mechanism for mass transport. In the ideal zero temperature case with a quasistatic increase in force all atoms would be put into an accelerating motion as soon as the energy NU_0 is present. However, in reality any thermal excitation would excite the system before this point is reached by the creation of kink-antikink pairs that would travel down the chain. For a chain of finite length these often occur at the end of the chain running in opposite direction. As a kink travels down the chain the atoms is advanced one atom spacing a_b along the substrate potential. This cascade of kink-antikink excitations is shown in figure 1.4

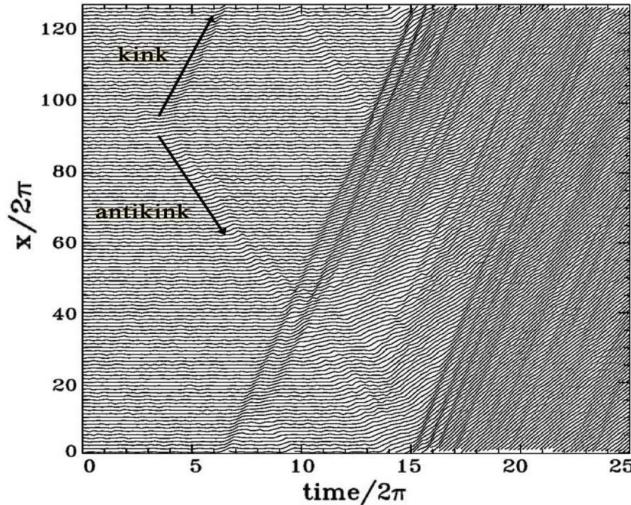


Figure 2. Time dependence of the atomic trajectories for the fully matched ($\theta = 1$) FK model at the (low-temperature) onset of depinning. Motion starts with the nucleation of a kink-antikink pair. The kink and the antikink depart in opposite directions cross the periodic boundary conditions, and collide quasielastically. A second kink-antikink pair forms in the wake of the initial kink. Further kink-antikink pairs are generated, with an avalanche-like increase of the kink-antikink concentration, eventually leading to a sliding state. Adapted from Ref. [2], Copyright (1997) by The American Physical Society.

Figure 1.4: Temporary figure from [6]

For the 2D case where an island is deposited on a surface, in our case the graphene sheet on the Si substrate, we generally expect the sliding to be initiated by kink-antikink pairs at the boundary.

For the case of incommensurability, i.e. $\theta = a_b/a_c$ is irrational, the GS is characterized by a sort of ‘‘staircase’’ deformation. That is, the chain will exhibit regular periods of regions where the chain is slightly compressed (expanded) to match the substrate potential, separated by kinks (antikinks), where the increased stress is eventually released through a localized expansion (compression) as illustrated in figure 1.5.



Figure 1.5: [Temporary figure from url`http://www.iop.kiev.ua/obraun/myreprints/surveyfk.pdf` p. 14. Incommensurable case ($\theta = ?$) where atoms sit slightly closer than otherwise dictated by the substrate potential for which this regularly result in a kink here seen as the presence of two atoms close together in one of the potential wells.]

The incommensurable FK model contains a critical elastic constant K_c , such that for $K > K_c$ (in an infinite chain?) the static friction F_s drops to zero making the chain able to slide at no energy cost. This can be explained by the fact that the displacement occurring in the incommensurable case will yield just as many atoms climbing up a corrugation as there are atoms climbing down. For an infinite chain this will exactly balance the interatomic forces making it non-resistant to sliding. Generally, incommensurability guarantees that the total energy (for $T = 0$) is independent of the relative position to the potential. However, when sliding freely a single atom will eventually occupy a maximum of the potential. When increasing the potential magnitude U_0 of softening the chain stiffness, lowering K , the possibility to occupy such a maximum is no longer present. This marks the so-called Aubry transition at the critical elastic constant $K = K_c(U_0, \theta)$ where the chain goes from a free sliding to a *pinned state* with a nonzero static friction. K_c is a discontinuous function of the ratio θ , due to the reliance on irrational numbers for incommensurability (? get plot of this?). The minimal value $K_c \approx 1.0291926$ in units $[2U_0(\pi/a_b)^2]$ is achieved for the golden-mean ratio $\theta = (1 + \sqrt{5}/2)$. Notice that the pinning is provided despite translational invariance due to the inaccessibility to move past the energy barrier which act as dynamical constraint. The Aubry transition can be investigated as first-order phase transition for which power laws can be defined for the order parameter. This is beyond the scope of this thesis as we merely are going to point to the FK model for the understanding of stick-slip behaviour and the concept of commensurability.

The phenomena of non-pinned configurations is named *superlubricity* in tribological context. Despite the misleading name this refers to the case where the static friction is zero while the kinetic friction is nonzero but reduced. For the case of a 2D sheet it is possible to alter the commensurability by changing the orientation of the sheet relative to the substrate. This has been shown for a graphene flake (single layer) sliding over a graphite surface (multiple layers) [[6] 45-47]. For incommensurability orientations the friction force was extremely weak (order 50 pN), compared to ELSE.

Kinetic friction In the FK model the kinetic friction is primarily caused by resonance between the sliding induced vibrations and phonon modes in the chain [15]. We can consider the case where all particles slide uniformly over the potential with velocity v^+ such that a so-called washboard frequency is induced with angular frequency $\Omega = 2\pi v^+ / a_b$. (The following is a bit shaky and should be double checked). The periodicity of the potential corresponds to a wavenumber of $q = 2\pi\theta$ (?) for phonons in the chain. The phonon dispersion relation for a 1D chain of atoms is given (source)

$$\omega = \sqrt{\frac{4K}{M}} \sin\left(\frac{k a_b}{2}\right)$$

for wavenumber k . Resonances will occur when the washboard frequency matches the phonon dispersion relation for $k = q$ or any of the harmonics $k = nq$, $n = 1, 2, 3, \dots$ yielding

$$\begin{aligned} n_1\Omega(v^+) &= n_2\omega(k = nq) \\ n_1 \frac{2\pi v^+}{a_b} &= n_2 \sqrt{\frac{4K}{M}} \sin(n\pi\theta a_b) \\ n_1 v^+ &= n_2 \sqrt{\frac{K}{M} a_b} \frac{\sin(n\pi a_c)}{\pi} \\ v^+ &\sim (?) \sqrt{\frac{K}{M} a_b} \frac{\sin(n\pi a_c)}{n\pi} \end{aligned}$$

for additional integers n_1 and n_2 (Doesn't really make sense to combine the integers like that, so go back and check [15]). Thus when the chain slides with a velocity around resonance, the washboard frequency can excite acoustic phonons which will dissipate to other phonon modes as well. At zero temperature the energy will transform back and forth between internal degrees of freedom and center of mass movement of the chain. Hence, at zero temperature this will speed up the translation decay (decay is synonymous for translational movement right?). However, for the more realistic case of non-zero temperature the substrate serves as a thermostat for which energy will dissipate from the chain to the substrate degrees of freedom giving rise to kinetic friction. A common way to model the non-zero temperature case is by the use of a Langevin thermostat, which simulates dissipation by adding a viscous damping force and thermal fluctuations by the addition of Gaussian random forces with variance proportional to the temperature (This is covered in more details in section 1.2.7.1). In combination, this gives rise to a kinetic friction that is both velocity and temperature dependent.

By extending the FK model into 2D [15] it can be shown numerically that the friction coefficient generally increases with increasing velocity and temperature respectively.

1.1.3.2 FK extension: Frenkel-Kontorova-Tomlinson (FKT)

Further model extensions. Important?

1.1.3.3 Graphene friction

Theory of friction experiment involving graphene.

1.1.3.4 Experimental procedures / Results?

- Atomic Force Microscope (AFM)

1.1.3.5 Summary of expected frictional properties

What frictional properties do we expect to see at a nanoscale. Numerical and experimental. Refer to theory for each element. Perhaps try to separate MD and experimental results from each other

It might not be easy to find experiments of graphene flakes on silicon substrate.

Qualitatively

1. Stick slip: We expect to see periodic stick-slip motion with period matching the lattice constant(s) [9]. In AFM and SFA experiments, the stick-slip motion tends to transition into smooth sliding when the speed exceeds $\sim 1\mu\text{ m/s}$ while in MD modelling the same transition is observed in the $\sim 1\text{ m/s}$ region [6]. This 6 order of magnitude discrepancy has been largely discussed in [141-144 [6]] in connection with the simplifications in MD simulations.
2. Static friction: Theoretically static friction is expected...
3. Orientation. Due to commensurability it is expected that the friction will vary along different orientations. In a numerical study by Zhu et al. [16] they found that a graphene flake sliding on a gold substrate exhibits the highest friction when sliding along the armchair direction. (We chose to slide along the zigzag direction more or less by random choice)

Variable dependence

1. Normal force: The general trend observed in AFM nanoscale friction is that friction force increase with normal load [1].
2. Velocity: Smooth kinetic friction generally increase with speed (velocity strengthening) [6]. E. Gnecco et al. [17] showed a logarithmic increase in mean friction with velocity (the tip of a friction force microscope and NaCl(100) at low velocity ($10^{-9} - 10^{-6}$ m/s)). If the scan velocity increases thermally activated processes becomes less important and beyond a critical value the friction forces becomes independent of velocity [1, p. 202]
3. Temperature: A decrease in friction as $1/T$ was observed by Zhao et al. in a series of AFM measurements on graphite in a wide temperature range (140-750 K) [1, source 351]
4. Ruan and Bhushan (1994) (source) found in an AFM study on graphite that the friction coefficient was around $\sim 0.01 - 0.03$
5. Contact area

Table 1.1: Quantitative nano friction dependence on various variables.

Variable	Dependency	Numerical studies	Experimental
Normal force F_N	F_f increasing F_N	MD simulations of amorphous carbon asperity on diamond substrate suggest a linear relationship to $F_f \propto F_N$ for nonadhesive contact and sublinear with van der waals adhesive forces [9]. Graphene flake?	The general trend observed AFM nanoscale friction is the friction force increase with normal load [1, p.200]. Various trends have been observed from linear to power trends.
Velocity v	$F_f \propto \ln v$		
Temperature T	$F_f \propto 1/T$		
Real contact area A	$F_f \propto A$		

1.2 Molecular Dynamics

Maybe also ‘‘Computer Simulations 7 of Nanometer-Scale Indentation and Friction’’ from [18]

Read [6][p. 18]

A promising compromise could possibly be provided by the so-called reactive potentials [120–122], capable of describing some chemical reactions, including interface wear with satisfactory computational efficiency in large-scale atomic simulations, compared to semi-empirical and first-principles approaches. [6]

- MD simulation (classical or ab initio)
- Basics of classical MD simulations: Integration and stuff
- Ab initio simulation (quantum mechanics, solving schrödinger)

1.2.1 Potentials

The choices of potentials used in the MD simulation is mainly based on the on [19] which have a somewhat similar MD friction simulation, the difference being that they impose a Si-tip on the graphene sheet supported by a Si-substrate where we impose drag the whole sheet upon the substrate. Nonetheless this serves as a good anchor for the methodology of the setup. The covalent bonds of C-C in graphene and Si-Si in the substrate is described by the Tersoff and Stillinger–Weber potentials, respectively. A typical 12-6 Lennard–Jones potential is used to describe the van der Waals adhesive interaction between graphene and the substrate.

1.2.1.1 General formulation of potentials...?

On a general note we can generalize the n-body potential as the expansion in orders of participating atoms as

$$E = \sum_i V_1(\mathbf{r}_i) + \sum_{\substack{i,j \\ i < j}} V_2(\mathbf{r}_i, \mathbf{r}_j) + \sum_{\substack{i,j,k \\ i < j < k}} V_3(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}_k) + \dots$$

where \mathbf{r}_n is the position of the n th particle and V_m is called an m -body potential [20]. The first one-body term corresponds to an external potential, followed by the two-body term, the three-body term and so on. The simplest model that includes particle interaction is the pair potential truncating the expansion after the two-body term. A general feature of the pair potentials is that they favor close-packed structures which is unsuited to describe covalent bonds that take more open structures. In particular, pair potentials are completely inapplicable to strongly covalent systems such as semiconductors [20]. In order to accommodate the description of covalent bonds the natural step is thus to include the next step of the expansion, the three-body terms, as we will see for the modeling of the graphene sheet C-C bonds and the Silicon sheet Si-Si bonds. For the interaction between the sheet and the substrate we can choose med a Lennard Jones pair potential describing the non-bonded van der Waals interaction.

1.2.2 Lennard Jones

This sections is based on [[21], [22], [23]].

The Lennard-Jones (LJ) model is probably one of the most famous pair potentials used in MD simulations. LJ models the potential energy between two non-bonding atoms based solely on interatomic distance r . The model accounts for attractive forces arising from dipole-dipole, dipole-induced dipole and London interactions, and repulsive forces that capture the hard core (is this safe to say?) of overlapping wave functions at small distances. Thus it is assumed neutrally charged atoms and was originally proposed for noble gases. The classical 12-6 version of the model (referring to the power law of the repulsive and attractive forces respectively) reads

$$E = 4\epsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right], \quad r < r_c, \quad (1.3)$$

where r is the interatomic distance with cut-off r_c , ϵ is the depth of the potential well and σ the distance where the potential is zero. By solving for the potential minimum ($dE/dr = 0$) we find the equilibrium distance to be $r_0 = \sigma 2^{1/6}$. This makes for an even clear interpretation of σ which effectively sets the equilibrium distance between atoms, i.e. the dividing line for which the net force is repulsive or attractive. While the LJ model in many ways is an oversimplified model that is insufficient in its description of ... (get source and concrete examples) it is commonly used as a model for intermaterial interactions (between moving object and substrate) in friction studies [[19], [24], [3]].

1.2.3 Stillinger weber

This section is based on [[25], [26]]

The stillinger weber potential takes the form of a three body potential

$$E = \sum_i \sum_{j>i} \phi_2(r_{ij}) + \sum_i \sum_{j \neq i} \sum_{k>j} \phi_3(r_{ij}, r_{ik}, \theta_{ijk}),$$

where r_{ij} denotes the distance between atom i and j and θ_{ijk} the angle between bond ij and jk . The summations is over all neighbours j and k of atom i within a cut-off distance $r = a\sigma$.

The two-body term ϕ_2 builds from the LJ model with the addition of an exponential cutoff term

$$\phi_2(r_{ij}) = A_{ij}\epsilon_{ij} \left[B_{ij} \left(\frac{\sigma_{ij}}{r_{ij}} \right)^{p_{ij}} - \left(\frac{\sigma_{ij}}{r_{ij}} \right)^{q_{ij}} \right] \exp\left(-\frac{\sigma_{ij}}{r_{ij} - a_{ij}\sigma_{ij}}\right). \quad (1.4)$$

The model parameters A , ϵ , B , σ , p , q and a comes with i, j indices to indicate that these parameters should be specified for each unique pair of atom types. However, in our case we will only provide a single value for each model parameter as we are exclusively dealing with Si-Si bonds. We see that the first term in eq. (1.4) is

reminiscent of the LJ model in eq. (1.3) while the last term effectively drives the potential to zero at $r = a\sigma$, which is thus the chosen cut-off distance for the potential evaluation. With the model parameters for the Si-Si modelling (see table 1.2) the cut-off becomes $\sim 3.8 \text{ \AA}$.

The three body term includes an angle dependency as

$$\phi_3(r_{ij}, r_{ik}, \theta_{ijk}) = \lambda_{ijk} \epsilon_{ijk} \left[\cos \theta_{ijk} - \cos \theta_{0,ijk} \right]^2 \exp\left(\frac{\gamma_{ij}\sigma_{ij}}{r_{ij} - a_{ij}\sigma_{ij}}\right) \exp\left(\frac{\gamma_{ik}\sigma_{ik}}{r_{ik} - a_{ik}\sigma_{ik}}\right), \quad (1.5)$$

where $\theta_{0,ijk}$ is the equilibrium angle. The first term of eq. (1.5) includes an angle dependency analog to a harmonic oscillator based on a cosine angle distance from the equilibrium angle. The final two terms act again as a cut-off function by driving the potential to zero at $r_{ij} = a_{ij}\sigma_{ij}$ and $r_{ik} = a_{ik}\sigma_{ik}$ respectively.

The parameters used for the Si-Si bond modeling is displayed in table 1.2 along with an interpretation of each model parameter.

Table 1.2: Parameters for the stilliner weber potential used for intermolecular interactions in the silicon substrate.

Parameter	Value	Description
ϵ	2.1683	Individual depth of the potential well for each atom type pair/tiplets.
σ	2.0951	Distance for which the individual pair interactions has zero potential (analog to the LJ model).
a	1.80	The individual cut-off distance for each atom type pair.
λ	21.0	The overall depth of the three-body potential well.
γ	1.20	The shape of the three-body cut-off terms.
$\cos(\theta_0)$	-1/3	Cosine of equilibrium angle.
A	7.049556277	The overall depth of the two-body potential well.
B	0.6022245584	Scales the repulsion part of the two-body term.
p	4.0	The power dependency for the repulsion part of the two-body term.
q	0.0	The power dependency for the attraction part of the two-body term.
tol	0.0	LAMMPS: Option to define a different cut-off than the theoretical of $r = a\sigma$. tol = 0 refers to the theoretical being used.

1.2.4 Tersoff

This section is based on [[27], [20]].

The tersoff potential abandon the idea of a general n -body form and attempts instead to build the model on a more physics informed approach; The more neighbours an atom has the weaker the bonds will be. Thus it introduces the bond order (bond strength), that is environment specific and decrease with increasing bond coordination (number of neighbours for a given atom). The potential energy is taken to have the form

$$E = \sum_i E_i = \frac{1}{2} \sum_{i \neq j} V_{ij},$$

$$V_{ij} = f_C(r_{ij}) [f_R(r_{ij}) + b_{ij} f_A(r_{ij})],$$

where the total potential energy is decomposed into a bond energy V_{ij} . The indices i and j run over the atoms of the system with r_{ij} denoting the distance between atom i and j . Notice that the sum includes all combinations of i, j where $i \neq j$ meaning that the same bond is double counted which is the reason for the additional factor 1/2. The reasoning behind comes from the asymmetry of the bond order $b_{ij} \neq b_{ji}$ leading to a $V_{ij} \neq V_{ji}$. The bond energy is composed of a repulsive term f_R , arising from overlapping wave functions, and an attractive term f_A associated with bonding. f_C is simply a smooth cut-off function to increase computational efficiency. b_{ij} represent the bond order, i.e. the strength of the bonds, which depends inversely on the number of

bonds, the bond angles (θ_{ijk}) and optionally the relative bonds lengths (r_{ij}, r_{jk}). Notice that an additional cut-off term a_{ij} was originally multiplied to f_R as a way of including terms that limit the range of the interactions to the first neighbour shell. These kind of limitations is already included in b_{ij} for the attractive term f_A but is often omitted for the repulsive term f_R , and we do so to by setting $a_{ij} = 1$.

The cut-off function f_C goes from 1 to 0 over a small interval range $R \pm D$ as

$$f_C(r) = \begin{cases} 1 & r < R - D \\ \frac{1}{2} - \frac{1}{2} \sin\left(\frac{\pi}{2} \frac{r-R}{D}\right) & R - D < r < R + D \\ 0 & r > R + D \end{cases}$$

which is continuous and differentiable for all r . R is usually chosen to include only the first neighbour shell. The repulsive and attractive terms f_R and f_A is modelled as an exponential function, similar to a morse potential,

$$\begin{aligned} f_R(r) &= A \exp(-\lambda_1 r), \\ f_A(r) &= -B \exp(-\lambda_2 r). \end{aligned}$$

The novel feature of the model lies in modeling of the bond order b_{ij} which includes three-body interactions by summing over a third atom $k \neq i, j$ within the cut-off $r_{ik} < R + D$ as shown in the following.

$$b_{ij} = (1 + \beta^n \zeta_{ij}^n)^{-\frac{1}{2n}} \quad (1.6)$$

$$\zeta_{ij} = \sum_{k \neq i, j} f_C(r_{ik}) g\left(\theta_{ijk}(r_{ij}, r_{ik})\right) \exp(\lambda_3^m (r_{ij} - r_{ik})^m) \quad (1.7)$$

$$g(\theta) = \gamma_{ijk} \left(1 + \frac{c^2}{d^2} - \frac{c^2}{[d^2 + (\cos \theta - \cos \theta_0)^2]} \right). \quad (1.8)$$

In eq. (1.8) $\zeta_{i,j}$ is an effective coordination and $g(\theta)$ captures angle dependency as it is minimized at the equilibrium angle $\theta = \theta_0$.

The parameters used to model the graphene C-C bonds is summarized in table 1.3

Table 1.3: Parameters for the tersoff potential used for intermolecular interactions in the graphene sheet

Parameter	Value	Description
m	3.0	Default (not used since $\lambda_3 = 0$)
γ	1.0	...
λ_3	0.0 \AA^{-1}	...
c	3.8049×10^4	Strength of the angular effect
d	4.3484	Determines the “sharpness” of the angular dependency
$\cos(\theta_0)$	-0.57058	Cosine of the equilibrium angle
n	0.72751	Power law exponent for the bond order dependency
β	1.5724×10^{-7}	...
λ_2	2.2119 \AA^{-1}	Decay of repulsion potential term
B	346.74 eV	Attractive potential term minimum at core ($r_{ij} = 0$).
R	1.95 Å	Center distance for cut-off
D	0.15 Å	Thickness of cut-off layers
λ_1	3.4879 \AA^{-1}	Decay of repulsion potential term
A	1393.6 eV	Repulsion potential term at core ($r_{ij} = 0$)

1.2.5 LAMMPS

1.2.6 Integration

Having defined a system of particles governed by interatomic potentials we need to move the system forward in time. By solving Newtons equations of motion we effectively do so by sampling the microcanonical ensemble

characterized by a constant number of particles N , volume V and energy E , hence denoted NVE. Newtons equaitons of motion read

$$m_i \frac{d^2 \mathbf{r}_i}{dt^2} = \mathbf{F}_i = -\nabla U_i \quad (1.9)$$

where i is the particle index and m_i its mass, $\mathbf{r}_i = (x_i, y_i, z_i)$ the position, t is time, $\nabla_i = (\frac{\partial}{\partial x_i}, \frac{\partial}{\partial y_i}, \frac{\partial}{\partial z_i})$ and U_i the potential energy. In system the potential energy is a function of the particle positions of nearby particles depending on the specefic potential in use. Since the forces defined by the potentials is conservative we expect the energy of the solution to be conserved. We redefine eq.(1.9) in terms of two coupled first order differential equations

$$\dot{\mathbf{v}}_i(t) = \frac{\mathbf{F}}{m_i}, \quad \dot{\mathbf{r}}_i(t) = \mathbf{v}_i(t), \quad (1.10)$$

where $\dot{x} = dx/dt$ (Newton's notation) and $\mathbf{v} = (v_x, v_y, v_z)$ is velocity. Numerically we can solve the coupled equations (eq.(1.10)) by integrating over discrete timnesteps. That is, we discretize the solution into temporal steps $t_k = t_0 + k \cdot \Delta t$ with time-step Δt .

1.2.6.1 Velocity Verlet

A common algorithm to integrate Newtons equation of motion (as formulated in eq.(1.10)) is the *velocity verlet*. We can derive the algorithm by the use of Taylor expansions. We begin by expanding the next-step position vector $\mathbf{r}_i(t + \Delta t)$ at time t

$$\mathbf{r}_i(t + \Delta t) = \mathbf{r}_i(t) + \dot{\mathbf{r}}_i(t)\Delta t + \frac{\ddot{\mathbf{r}}_i(t)}{2}\Delta t^2 + \mathcal{O}(\Delta t^3), \quad (1.11)$$

where $\ddot{\mathbf{r}} = d^2\mathbf{r}/dt^2$ and Δt^n is simply the relaxed notation for $(\Delta t)^n$. Similar we take the expansions of the next-step velocity vector $\mathbf{v}_i(t + \Delta t)$ at time t

$$\mathbf{v}_i(t + \Delta t) = \mathbf{v}_i(t) + \dot{\mathbf{v}}_i(t)\Delta t + \frac{\ddot{\mathbf{v}}_i(t)}{2}\Delta t^2 + \mathcal{O}(\Delta t^3). \quad (1.12)$$

Finnally, by taking the expansion of $\dot{\mathbf{v}}_i(t + \Delta t)$ we can eliminate the $\ddot{\mathbf{v}}_i$ -term in eq. (1.12) and simplify it as shown in the following.

$$\begin{aligned} \dot{\mathbf{v}}_i(t + \Delta t) &= \dot{\mathbf{v}}_i(t) + \ddot{\mathbf{v}}_i(t)\Delta t + \mathcal{O}(\Delta t^2) \\ \frac{\ddot{\mathbf{v}}_i(t)}{2}\Delta t^2 &= \frac{\Delta t}{2}(\dot{\mathbf{v}}(t + \Delta t) - \dot{\mathbf{v}}_i(t)) + \mathcal{O}(\Delta t^3) \\ &\Downarrow \\ \mathbf{v}_i(t + \Delta t) &= \mathbf{v}_i(t) + \dot{\mathbf{v}}_i(t)\Delta t + \frac{\Delta t}{2}(\dot{\mathbf{v}}_i(t + \Delta t) - \dot{\mathbf{v}}_i(t)) + \mathcal{O}(\Delta t^3) \\ &= \mathbf{v}_i(t) + \frac{\Delta t}{2}(\dot{\mathbf{v}}_i(t) + \dot{\mathbf{v}}_i(t + \Delta t)) + \mathcal{O}(\Delta t^3). \end{aligned} \quad (1.13)$$

By combining eq. (1.11) and eq. (1.13) and using Newton's second equation $\dot{\mathbf{v}} = \mathbf{F}_i(t)/m_i$ and $\mathbf{v} = \dot{\mathbf{r}}$ we arrive at the final scheme

$$\begin{aligned} \mathbf{r}_i(t + \Delta t) &= \mathbf{r}_i(t) + \mathbf{v}_i(t)\Delta t + \frac{\mathbf{F}_i(t)}{2m_i}\Delta t^2 + \mathcal{O}(\Delta t^3), \\ \mathbf{v}_i(t + \Delta t) &= \mathbf{v}_i(t) + \frac{\mathbf{F}_i(t) + \mathbf{F}_i(t + \Delta t)}{2m_i}\Delta t + \mathcal{O}(\Delta t^3). \end{aligned}$$

The scheme will give a local error of order Δt^3 corresponding to a global error of Δt^2 . One of the most popular ways to implement this numerically is as stated in the following steps.

1. Calculate $v_{k+\frac{1}{2}} = v_k + \frac{F_k}{2m} \Delta t$.
2. Calculate $r_{k+1} = r_k + v_{k+\frac{1}{2}} \Delta t$.
3. Evaluate the force $F_{k+1} = F(r_{k+1})$.
4. Calculate $v_{k+1} = v_{k+\frac{1}{2}} + \frac{F_{k+1}}{2m} \Delta t$

1.2.7 Thermostats

As we already mentioned above in Sec. 2, any kind of sliding friction involves mechanical work, some of which is then transformed into heat (the rest going into structural transformations, wear, etc.). The heat is then transported away by phonons (and electrons in the case of metallic sliders) and eventually dissipated to the environment [6].

Likewise all excitations generated in the simulations should be allowed to propagate in the system and disperse in the bulk of both sheet and substrate. Due to small simulation size this is likely to reflect back and ‘pile up’ unphysically. Thus in order to avoid continuous heating and attain a steady state the (Joule) heat must be removed at a steady state. This is very the viscous damping of the Langevin equations enter the picture. It can be difficult to set the value γ for the magnitude of this damping. The unphysical introduction of heat sink can be mitigated by some modifications he mention, which is kind of next level I guess.

1.2.7.1 Langevin thermostat

In order to control the temperature of the system we introduce the so-called Langevin thermostat. This is a stochastic thermostat that modifies Newton's equation of motion such that solution lies in the canonical ensemble characterized by a constant number of particles N , constant volume V and constant temperature T , hence denoted NVT. The canonical ensemble system is represented by the finite system being in contact with an infinite heat bath of temperature T . The NVT ensemble is equivalent to sampling a system in thermodynamic equilibrium where the weight of each microscopic state is given by the Boltzmann factor $\exp[-E/(k_B T)]$.

The Langevin equation is the modified version of Newton's second law for a Brownian particle. A Brownian particle is a small particle suspended in liquid, e.g. pollen or dust, named after Robert Brown (1773–1858) who was the first to observe its jittery motion. The Langevin equation describes this motion as the combination of viscous drag force $-\gamma \mathbf{v}$, where γ is a positive friction coefficient and \mathbf{v} the velocity vector, and a random fluctuation force \mathbf{R} . The Langevin equation reads

$$m \frac{d\mathbf{v}}{dt} = -\gamma \mathbf{v} + \mathbf{R} \quad (1.14)$$

where m is the particle mass. This effectively describes the particle of interest, the Brownian particle, as being suspended in a sea of smaller particles. The collision with these smaller particles is modelled by the drag force and the fluctuation force. We notice that if the fluctuation force is excluded eq. (1.14) becomes

$$m \frac{d\mathbf{v}}{dt} = -\gamma \mathbf{v} \Rightarrow \mathbf{v}_i(t) = v(0) e^{-\frac{\gamma t}{m}},$$

where the solution shows that the Brownian particle will come to a complete stop after a long time $\mathbf{v}_i(t \rightarrow \infty) \rightarrow \mathbf{0}$. This is in violation with the equipartition theorem

$$\frac{1}{2} m \langle v^2 \rangle_{eq} = \frac{k_B T}{2},$$

and hence the fluctuation force is necessary to obtain the correct equilibrium.

The following calculations are done in one dimension in order to simplify the notation. We describe the statistical nature of the collisions as a sum of independent momentum transfers

$$\Delta P = \sum_i^N \delta p_i$$

where ΔP denotes the change of momentum after N momentum transfers δp_i from the environment to the Brownian particle. We assume the first and second moments $\langle \delta p \rangle = 0$ and $\langle \delta p \rangle = \sigma^2$. When N is large the central limit theorem states that the random variable ΔP has a Gaussian distribution with $\langle P \rangle = 0$ and $\langle \Delta P^2 \rangle = N \sigma^2$. If we consider the momentum change ΔP over a discrete time Δt , where the number of collisions is proportional to time $N \propto \Delta t$, the corresponding fluctuation force $R = \Delta P / \Delta t$ will have a variance

$$\langle R^2 \rangle = \frac{\langle \Delta P^2 \rangle}{\Delta t^2} = \frac{N \sigma^2}{\Delta t^2} \propto \frac{1}{\Delta t}.$$

In a computer simulation we need to pick a random force $R(t)$ from a Gaussian distribution every time-step Δt . These forces will not be correlated as long as Δt is larger than the correlation time of the forces from the molecules which we will assume for this model (I think there exist corrections for this to refer to here). With this assumption we can write the correlation function as

$$\langle R(t)R(0) \rangle = \begin{cases} \frac{a}{\Delta t}, & |\Delta t| < \Delta t/2 \\ 0, & |\Delta t| > \Delta t/2, \end{cases} \quad (1.15)$$

where a is some strength of (...?). In the limit $\Delta t \rightarrow 0$ the correlation function becomes

$$\langle R(t)R(0) \rangle = a\delta(t), \quad (1.16)$$

where δ denotes the dirac delta function. This is valid for all spatial coordinates which will all be independent of each other. Since both the drag force and the fluctuation force originate from the molecular fluid, where the drag force $-\alpha v$ is velocity dependent it is reasonable to assume that fluctuation force is independent of velocity, i.e. $\langle R_i v_j \rangle = 0$ for all cartesian indices i and j .

In the following we will attempt justify the Langevin equaiton (why it is like it is) and determine the relationship between the drag coefficient γ and the random force R .

From the Langevin equation eq. (1.14) we can compute the velocity autocorrelation function (Move to appendix?). We do this in one dimension for simplicity. We begin by multiplying by $(e^{\gamma t/m})/m$

$$\dot{v}(t)e^{\gamma t/m} + \frac{\gamma}{m}v(t)e^{\gamma t/m} = \frac{F}{m}e^{\gamma t/m},$$

and integrate from $t = -\infty$. By the use of integration by parts on the latter term on the left hand side we calculate the velocity

$$\begin{aligned} \int_{-\infty}^t dt' \dot{v}(t')e^{\gamma t'/m} + \frac{\gamma}{m}v(t)e^{\gamma t/m} &= \int_{-\infty}^t dt' e^{\gamma t'/m} \frac{F(t')}{m} \\ \int_{-\infty}^t dt' \dot{v}(t')e^{\gamma t'/m} + \left(\left[v(t')e^{\gamma t'/m} \right]_{-\infty}^t - \int_{-\infty}^t dt' \dot{v}(t')e^{\gamma t'/m} \right) &= \int_{-\infty}^t dt' e^{\gamma t'/m} \frac{F(t')}{m} \\ v(t) &= \int_{-\infty}^t dt' e^{-\gamma(t-t')/m} \frac{F(t')}{m}, \end{aligned}$$

where $e^{-\gamma t/m}$ plays the role of a response function. We can then calculate the autocorrelation

$$\begin{aligned} \langle v(t)v(0) \rangle &= \int_{-\infty}^t dt_1 \int_{-\infty}^0 dt_2 e^{\frac{t-t_1-t_2}{m}} \frac{\langle F(t_1)F(t_2) \rangle}{m^2} \\ &= \int_{-\infty}^t dt_1 \int_{-\infty}^0 dt_2 e^{\frac{t-t_1-t_2}{m}} \frac{a\delta(t_1-t_2)}{m^2} \\ &= \int_{-\infty}^0 dt_2 e^{\frac{t-2t_2}{m}} \frac{a}{m^2} = \frac{a}{2m\gamma} e^{-\frac{\gamma t}{m}}, \end{aligned}$$

where we used eq. (1.16) and the fact that the integration commutes with the average (we are allowed to flip the order). By comparing this with the equipartition theorem we get

$$\begin{aligned} \frac{1}{2}m\langle v^2 \rangle &= \frac{k_B T}{2} \\ \frac{1}{2}m\langle v(0)v(0) \rangle &= \frac{a}{4\gamma} = \frac{k_B T}{2} \\ a &= 2\gamma k_B T \end{aligned}$$

We notice the appearance of γ meaning that the magnitude of the fluctuations increase both with friction and temperature. Further we can integrate the velocity over time to get displacement $x(t)$ and show that the variance (show this? In appendix maybe?) is

$$\langle x^2(t) \rangle = \frac{2k_B T}{\gamma} \left(t - \frac{m}{\gamma} \left(1 - e^{-\gamma t/m} \right) \right),$$

where for $t \gg m/\gamma$ only the t -term survives yielding

$$\langle x^2(t) \rangle = 2k_B T t / \gamma.$$

In 1D, the diffusion constant D is related to the variance as $\langle x^2 \rangle = 2Dt$, meaning that this represents the einstein relation $D = \mu k_B T$ with the mobility $\mu = 1/\gamma$.

when $t \ll m/\gamma$ we use the Taylor expansion $1 - e^{-x} \approx x - x^2/2$ for $x \ll 1$ to get

$$\langle x^2(t) \rangle = \frac{k_B T}{m} t^2$$

which exactly matches the thermal velocity

$$v_{\text{th}} \frac{\langle x^2(t) \rangle}{t^2} = \frac{k_B T}{m}$$

which follows from the equipartition theorem. The finite correlation time γ/m hence describe the crossover from the ballistic regime $\sqrt{\langle x^2(t) \rangle} \propto t$ to the diffusive regime $\sqrt{\langle x^2(t) \rangle} \propto \sqrt{t}$.

Introduce the fluctuation-dissipation theorem concept earlier since this is a motivation for the Langevin equation.

1.2.7.2 Implementing Langevin

The implementation of the Langevin equation into LAMMPS follows [28] and updates the force vector for each particle as

$$\begin{aligned} \mathbf{F} &= \mathbf{F}_c + \mathbf{F}_f + \mathbf{F}_r \\ &= -\nabla U - \gamma m \mathbf{v} + \sqrt{\frac{2k_B T m \gamma}{\Delta t}} \mathbf{h}(t) \end{aligned} \quad (1.17)$$

where \mathbf{F}_c is the conservative force computed via the usual inter-particle interactions described by the potential U , \mathbf{F}_f is the drag force and \mathbf{F}_r is the random fluctuation force where \mathbf{h} is a random vector drawn from a normal distribution with zero mean and unit variance. Notice that this generalized description of the Langevin equation deviates from the presentation in eq. (1.14) since we have added the conservative force \mathbf{F}_c , but also by the appearance of the mass in both the drag force and the fluctuation force due to the introduction of damping. It is beyond our scope to comprehend this. However, the fact that Δt now appears in the denominator for the random force variance $2k_B T m \gamma / \Delta t$ is due to the fact that we have discretized time. This is in agreement with the formulation in eq. (1.15). By applying eq. (1.17) we get the refined velocity verlet scheme

$$\begin{aligned} \mathbf{v}_i(t + \Delta t/2) &= \mathbf{v}_i(t) - \frac{\Delta t}{2} \left(\frac{\nabla_i U(t)}{m_i} + \gamma \mathbf{v}_i \right) + \sqrt{\frac{k_B T \gamma \Delta t}{2m_i}} \mathbf{h}_i \\ \mathbf{r}_i(t + \Delta t) &= \mathbf{r}_i(t) + \mathbf{v}_i(t + \Delta t/2) \Delta t \\ \mathbf{v}_i(t + \Delta t) &= \mathbf{v}_i(t + \Delta t/2) - \frac{\Delta t}{2} \left(\frac{\nabla_i U(t + \Delta t)}{m_i} + \gamma \mathbf{v}_i(t + \Delta t/2) \right) + \sqrt{\frac{k_B T \gamma \Delta t}{2m_i}} \mathbf{h}_i \end{aligned}$$

with new random vector \mathbf{h}_i for each particle and each update. Notice however, that LAMMPS only applies this scheme to the particle groups with the thermostat on.

1.2.7.3 MD limitations

1.3 Defining the system

1.3.1 Groups: Sheet, pullblocks and substrate

Include figure of system to point out thermo layers and freeze layers.

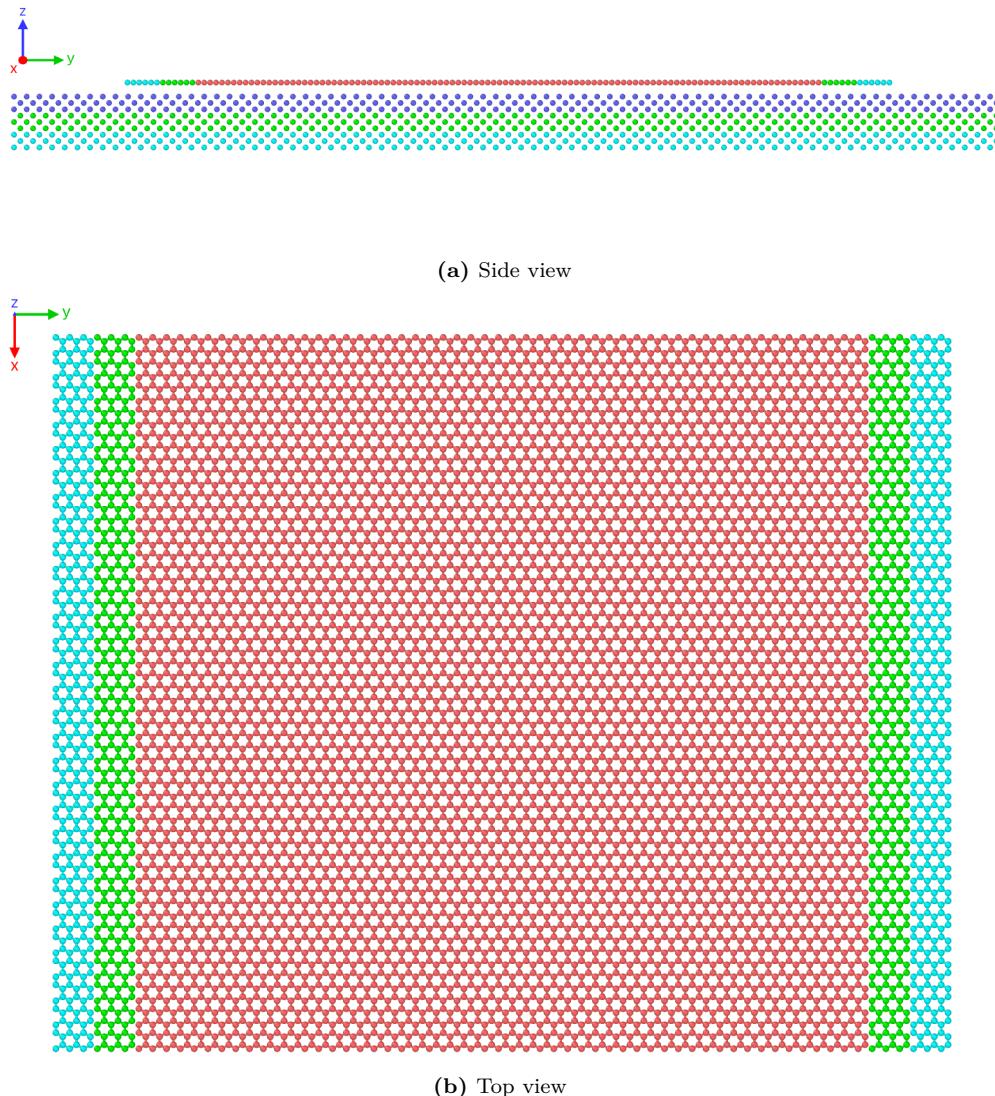


Figure 1.6: System. 27456 atoms in total: 7272 with thermostat (orange), 7272 is locked (light blue) and the remaining 12912 just with NVE. (Get better colors)

Table 1.4: System atom count and region division.

Region	Total	Sub region	Sub total	NVE	NVT	Locked
Sheet	7800	Inner sheet	6360	6360	0	0
		Pull blocks	1440	0	720	720
Substrate	19656	Upper	6552	6552	0	0
		Middle	6552	0	6552	0
		Bottom	6552	0	0	6552
All	27456			12912	7272	7272

The sheet dimensions is

Table 1.5: Sheet dimensions

Group	x, y -dim	dim [Å]	Area [Å ²]
Full sheet	$x_S \times y_S$	$130.029 \times 163.219 \text{ Å}$	$21,223.203$
Inner sheet	$x_S \times 81.40 \%_{y_s}$	$130.029 \times 132.853 \text{ Å}$	$17,274.743$
Pull blocks	$2 \times x_S \times 9.30 \%_{y_s}$	$2 \times 130.029 \times 15.183 \text{ Å}$	$2 \times 1,974.230$

1.3.1.1 Pressure reference

Find place to put this.

In order to relate the magnitude of the normal force in our friciton 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 man³ standing on one stiletto heels (with all the weight on the heel) will result in a pressure corresponding diameter of

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

While this is in itself a spectacular realization that is often used in introduction 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 $\sim 21 \times 10^3 \text{ Å}^2$ we can achieve a similar pressure of $\sim 10 \text{ MPa}$ with a normal force

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

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 area 113 cm^2 the pressure drops to a mere 70 kPa corresponding to $\sim 0.01 \text{ nN}$.

1.3.2 Creating sheets

We are going to create a 2D sheet graphene sheet.

1.3.2.1 Graphene

Graphene is a single layer of carbon atom, graphite is the bulk, arranged in a hexagonal lattice structure. We can describe the 2D crystal structure in terms of its primitive lattice vector and a basis. That is we populate each

³Yes, a man can certainly wear stiletto heals.

lattice site by the given basis and translate it to fill the whole plane by any linear combination of the lattice vectors

$$\mathbf{T}_{mn} = m\mathbf{a}_1 + n\mathbf{a}_2, \quad m, n \in \mathbb{N}.$$

For graphene we have the primitive lattice vectors

$$\mathbf{a}_1 = a \left(\frac{\sqrt{3}}{2}, -\frac{1}{2} \right), \quad \mathbf{a}_2 = a \left(\frac{\sqrt{3}}{2}, \frac{1}{2} \right), \quad |\mathbf{a}_1| = |\mathbf{a}_2| = 2.46 \text{ \AA}.$$

Notice that we deliberately excluded the third coordinate as we only consider a single graphene layer on not the bulk graphite consisting of multiple layers stacked on top of each other. The basis is

$$\left\{ (0, 0), \frac{a}{2} \left(\frac{1}{\sqrt{3}}, 1 \right) \right\}$$

It turns out that the spacing between atoms is equal for all pairs with an interatomic distance

$$\left| \frac{a}{2} \left(\frac{1}{\sqrt{3}}, 1 \right) \right| \approx 1.42 \text{ \AA}.$$

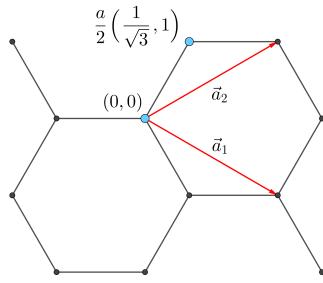
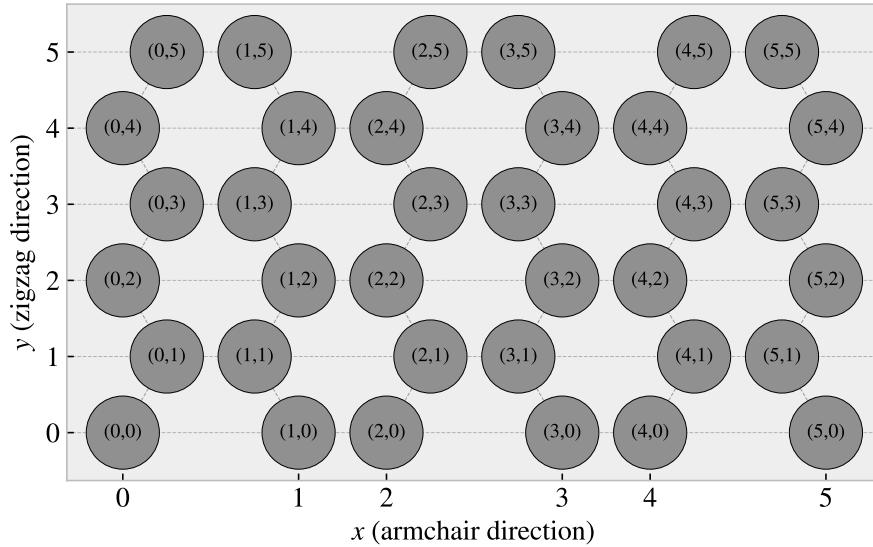


Figure 1.7: Graphene crystal structure with basis.

1.3.2.2 Indexing

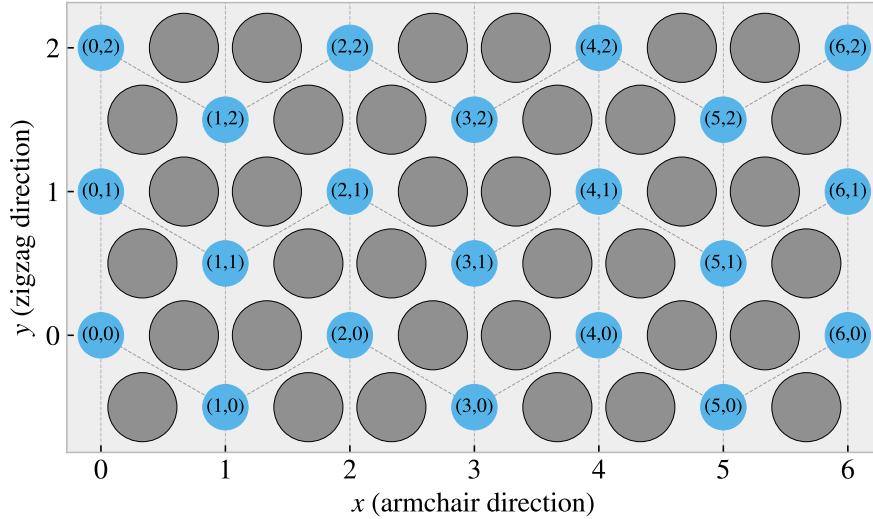
In order to define the cut patterns applied to the graphene sheet we must define an indexing system. We must ensure that this gives an unique description of the atoms as we eventually want to pass a binary matrix, containing 0 for removed atom and 1 for present atom, that uniquely describes the sheet. We do this by letting the x-coordinate point to zigzag chains and the y-coordinate to the position along that chain. This is illustrated in figure 1.8. Other solutions might naturally involve the lattice vectors, but as these only can be used to translate to similar basis atoms a unfortunate duality is introduced as ones need to include the basis atom of choice into the indexing system. With the current system we notice that locality is somewhat preserved. That is, atom (i, j) is in the proximity of $\{(i+1, j), (i-1, j), (i, j+1), (i, j-1)\}$, but only three of them are categorized as nearest neighbours due to the hexagonal structure of the lattice. While $(i, j \pm 1)$ is always nearest neighbours the neighbour in the x-direction flip sides with incrementing y-coordinate. That is the nearest neighbours (NN) is decided as

$$\begin{aligned} j \text{ is even} &\rightarrow \text{NN} = \{(i+1, j), (i, j+1), (i, j-1)\}, \\ j \text{ is odd} &\rightarrow \text{NN} = \{(i-1, j), (i, j+1), (i, j-1)\}. \end{aligned}$$

**Figure 1.8:** Graphene atom indexing

1.3.2.3 Removing atoms

As a mean to ease the formulation of cut patterns we introduce pseudo center element in each gap of the hexagonal honeycombs, see figure 1.9.

**Figure 1.9:** Graphene center indexing

Similar to the case of the indexing for the carbon atoms themselves the nearest neighbour center elements alternate with position, this time along the x-coordinate. Each center element has six nearest neighbours, in clock wise direction we can denote them: “up”, “upper right”, “lower right”, “down”, “lower left”, “upper left”. The “up” and “down” is always accessed as $(i, j \pm 1)$, but for even i the $(i + 1, j)$ index corresponds to the “lower right” neighbour while for odd i this corresponds to the “upper right” neighbour. This shifting applies for all left or right neighbours and the full neighbour list is illustrated in figure 1.10.

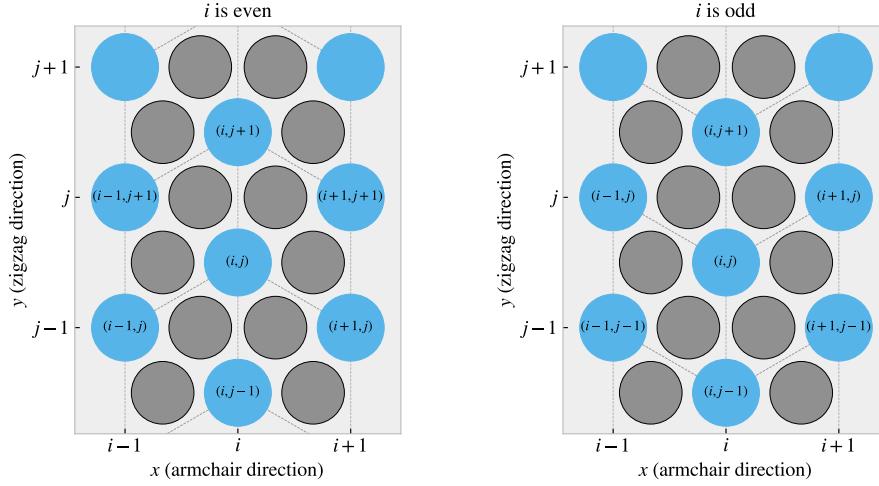


Figure 1.10: Graphene center elements directions

We define a cut pattern by connecting center elements into connected paths. As we walk element to element we remove atoms according to one of two rules

1. Remove intersection atoms: We remove the pair of atoms placed directly in the path we are walking. That is, when jumping to the “up” center element we remove the two upper atoms located in the local hexagon of atoms. This method is sensitive to the order of the center elements in the path.
2. Remove all surrounding atoms: We simply remove all atoms in the local hexagon surrounding each center element. This method is independent of the ordering of center elements in the path.

We notice that removing atoms using either of these rules will not guarantee an unique cut pattern. Rule 1 is the more sensitive to paths but we realize that, for an even i , we will remove the same five atoms following either of the following paths.

$$\begin{aligned}
 (i, j) &\rightarrow \underbrace{(i + 1, j + 1)}_{\text{upper right}} \rightarrow \underbrace{(i, j + 1)}_{\text{up}} \rightarrow \underbrace{(i + 1, j + 2)}_{\text{upperright + up}} \rightarrow \underbrace{(i + 1, j + 1)}_{\text{upper right}} \\
 (i, j) &\rightarrow \underbrace{(i + 1, j + 1)}_{\text{upper right}} \rightarrow \underbrace{(i + 1, j + 2)}_{\text{upperright + up}} \rightarrow \underbrace{(i, j + 1)}_{\text{up}}
 \end{aligned}$$

For rule 2 it is even more obvious that different paths can result in the same atoms being removed. This is the reason that we needed to define and indexing system for the atom position itself even though that all cuts generated manually will use the center element path as reference.

Illustrate some delete path?

1.3.3 Pull blocks

1.3.4 Kirigami inspired cut out patterns

1.3.4.1 Pop-up pattern

1.3.4.2 Honeycomb

1.3.4.3 Random walk

1.4 Fourier Transform (light)

Find out where to put this if necessary.

Fourier transform is a technique where we transform a function $f(t)$ of time to a function $F(k)$ of frequency. The Forward Fourier Transform is done as

$$F(k) = \int_{-\infty}^{\infty} f(t)e^{-2\pi i k x} dx$$

For any complex function $F(k)$ we can decompose it into magnitude $A(k)$ and phase $\phi(k)$

$$F(k) = A(k)e^{i\phi(k)}$$

Hence when performing a Forward Fourier transform on a time series we can determine the amplitude and phase as a function of frequency as

$$A(k) = |F(k)|^2, \quad \phi(k) = \Im \ln F(k)$$

1.4.1 Real life experimental procedures

From Introduction to Tribology, Second Edition, p. 526:

The surface force apparatus (SFA), the scanning tunneling microscopes (STM), and atomic force and friction force microscopes (AFM and FFM) are widely used in nanotribological and nanomechanics studies.

- Real life procedures to mimic in computation, for instance Atomic Force Microscopy (AFM) for friction measurements.
- Available technology for test of my findings if successful (possibilities for making the nano machine)

1.5 Machine Learning (ML)

- Feed forward fully connected
- CNN
- GAN (encoder + decoder)
- Genetic algorithm
- Using machine learning for inverse designs partly eliminate the black box problem. When a design is produced we can test it, and if it works we not rely on machine learning connections to verify its relevance.
- However, using explanations techniques such as maybe t-SNE, Deep dream, LRP, Shapley values and linearizations, we can try to understand why the AI chose as it did. This can lead to an increased understanding of each design feature. Again this is not dependent on the complex network of the network as this can be tested and verified independently of the network.

1.5.1 Feed forward network / Neural networks

1.5.2 CNN for image recognition

1.5.3 GAN (encoder + decoder)

1.5.4 Inverse design using machine learning

1.5.5 Prediction explanation

1.5.5.1 Shapley

1.5.5.2 Linearizations

1.5.5.3 LRP

1.5.5.4 t-SNE

Bibliography

- [1] E. Gnecco and E. Meyer, *Elements of Friction Theory and Nanotribology*. Cambridge University Press, 2015, 10.1017/CBO9780511795039.
- [2] Bhushan, *Introduction*, ch. 1, pp. 1–? John Wiley & Sons, Ltd, 2013.
<https://onlinelibrary.wiley.com/doi/pdf/10.1002/9781118403259.ch1>.
<https://doi.org/10.1002/9781118403259.ch1>.
- [3] H.-J. Kim and D.-E. Kim, *Nano-scale friction: A review*, .
- [4] K. Holmberg and A. Erdemir, *Influence of tribology on global energy consumption, costs and emissions*, .
- [5] P. 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** (Oct, 2020) 042006.
- [6] N. Manini, O. M. Braun, E. Tosatti, R. Guerra and A. Vanossi, *Friction and nonlinear dynamics*, *Journal of Physics: Condensed Matter* **28** (jun, 2016) 293001.
- [7] B. Bhushan and A. V. Kulkarni, *Effect of normal load on microscale friction measurements*, *Thin Solid Films* **278** (1996) 49–56.
- [8] J. H. Dieterich, *Time-dependent friction in rocks*, *Journal of Geophysical Research (1896-1977)* **77** (1972) 3690–3697, [<https://agupubs.onlinelibrary.wiley.com/doi/pdf/10.1029/JB077i020p03690>].
- [9] Y. Mo, K. T. Turner and I. Szlufarska, *Friction laws at the nanoscale*, .
- [10] G. Carbone and F. Bottiglione, *Asperity contact theories: Do they predict linearity between contact area and load?*, *Journal of the Mechanics and Physics of Solids* **56** (2008) 2555–2572.
- [11] W. Commons, *File:asperities.svg — wikimedia commons, the free media repository*, 2022.
- [12] J. A. Greenwood and J. B. P. Williamson, *Contact of nominally flat surfaces*, Dec, 1966. 10.1098/rspa.1966.0242.
- [13] B. N. J. Persson, *Theory of rubber friction and contact mechanics*, *The Journal of Chemical Physics* **115** (2001) 3840–3861, [<https://doi.org/10.1063/1.1388626>].
- [14] B. Luan and M. O. Robbins, *The breakdown of continuum models for mechanical contacts*, .
- [15] J. Norell, A. Fasolino and A. Wijn, *Emergent friction in two-dimensional frenkel-kontorova models*, *Physical Review E* **94** (04, 2016) .
- [16] P. Zhu and R. Li, *Study of nanoscale friction behaviors of graphene on gold substrates using molecular dynamics*, .
- [17] E. Gnecco, R. Bennewitz, T. Gyalog, C. Loppacher, M. Bammerlin, E. Meyer et al., *Velocity dependence of atomic friction*, *Phys. Rev. Lett.* **84** (Feb, 2000) 1172–1175.
- [18] B. Bhushan, *Nanotribology and nanomechanics*, *Wear* **259** (2005) 1–?
- [19] S. Li, Q. Li, R. W. Carpick, P. Gumbsch, X. Z. Liu, X. Ding et al., *The evolving quality of frictional contact with graphene*, .
- [20] J. Tersoff, *New empirical approach for the structure and energy of covalent systems*, *Phys. Rev. B* **37** (Apr, 1988) 6991–7000.
- [21] S. Corporation, “pair_style lj/cut command.”
- [22] X. Wang, S. Ramírez-Hinestrosa, J. Dobnikar and D. Frenkel, *The lennard-jones potential: when (not) to use it*, *Phys. Chem. Chem. Phys.* **22** (2020) 10624–10633.
- [23] R. Naeem, “Lennard-jones potential.”

- [24] Q. Zhang, D. Diao and M. Kubo, *Nanoscratching of multi-layer graphene by molecular dynamics simulations*, *Tribology International* **88** (2015) 85–88.
- [25] S. Corporation, “pair_style sw command.”
- [26] F. H. Stillinger and T. A. Weber, *Computer simulation of local order in condensed phases of silicon*, *Phys. Rev. B* **31** (Apr, 1985) 5262–5271.
- [27] S. Corporation, “pair_style tersoff command.”
- [28] T. Schneider and E. Stoll, *Molecular-dynamics study of a three-dimensional one-component model for distortive phase transitions*, *Phys. Rev. B* **17** (Feb, 1978) 1302–1322.