



Master's thesis

# Predicting Frictional Properties of Graphene Kirigami Using Molecular Dynamics and Neural Networks

Designs for a negative friction coefficient

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Computational Science: Materials Science  
60 ECTS study points

Department of Physics  
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Spring 2023





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# Predicting Frictional Properties of Graphene Kirigami Using Molecular Dynamics and Neural Networks

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# Abstract

Various theoretical models and experimental results propose different governing mechanisms for friction at the nanoscale. We consider a graphene sheet modified with Kirigami-inspired cuts and under the influence of strain. Prior research has demonstrated that this system exhibits out-of-plane buckling, which could result in a decrease in contact area when sliding on a substrate. According to asperity theory, this decrease in contact area is expected to lead to a reduction of friction. However, to the best of our knowledge, no previous studies have investigated the friction behavior of a nanoscale Kirigami graphene sheet under strain. Here we show that specific Kirigami designs yield a non-linear dependency between kinetic friction and the strain of the sheet. Using molecular dynamics simulations, we have found a non-monotonic increase in friction with strain. We found that the friction-strain relationship does not show any clear dependency on contact area which contradicts asperity theory. Our findings suggest that the effect is associated with the out-of-plane buckling of the graphene sheet and we attribute this to a commensurability effect. By mimicking a load-strain coupling through tension, we were able to utilize this effect to demonstrate a negative friction coefficient on the order of  $-0.3$  for loads in the range of a few nN. In addition, we have attempted to use machine learning to capture the relationship between Kirigami designs, load, and strain, with the objective of performing an accelerated search for new designs. Although this approach yielded some promising results, we conclude that further improvements to the dataset are necessary in order to develop a reliable model. We anticipate our findings to be a starting point for further investigations of the underlying mechanism for the frictional behavior of a Kirigami sheet. For instance, the commensurability hypothesis could be examined by varying the sliding angle in simulations. We propose to use an active learning strategy to extend the dataset for the use of machine learning to assist these investigations. If successful, further studies can be done on the method of inverse design. In summary, our findings suggest that the application of nanoscale Kirigami can be promising for developing novel friction-control strategies.



# Acknowledgments

The task of writing a master's thesis is a demanding and extensive project which I could not have done without the support of many good people around me. First of all, I want to thank my supervisors Henrik Andersen Sveinsson and Anders Malthe-Sørensen for the assistance in this thesis work. I am especially grateful for the weekly meetings with Henrik and the inspiring discussions had as we unraveled the discoveries related to the topic of this thesis. I remember that I initially asked for an estimate of how much time he had available for supervision and the answer was something along the lines of "There are no limits really, just send me an email and we figure it out". This attitude captures the main experience I have had working with Henrik and I am profoundly grateful for the time and effort he has put into this project. I hope that he did not regret this initial statement too much, because I have certainly been taken advantage of it. I also want to thank Even Marius Nordhagen for technical support regarding the use of the computational cluster. In that context, I also want to acknowledge the Center for Computing in Science Education (CCSE) for making these resources available.

I would like to express my gratitude to all the parties involved in making it possible for me to write my thesis from Italy. I am particularly grateful for the flexibility shown by my supervisors and for the support of Anders Kvellestad, who allowed me to work remotely as a group teacher. I would also like to thank Scuola Normale Superiore for providing me with access to their library.

I realize that it is a commonly used cliché to express gratitude for the support of loved ones. However, I want to highlight the exceptional role played by my fiancé, Ida, who deserves the main credit for enabling me to maintain a healthy state of mind. She has provided me with a solid foundation for a fulfilling life that enables me to pursue secondary objectives, such as an academic career. I look forward to spending the rest of my life with you.

In this thesis, I have used the formal pronoun "we" mainly as a customary habit related to the formalities of scientific writing in a team. Nonetheless, I have realized that this usage is more fitting as I have not been working alone on this project. I have received support all the way from colleagues and friends at the University of Oslo, my family residing in Denmark, and my life partner who slept beside me every night here in Italy. They are the "good people around me" who have made this thesis possible.



# Acronyms

**GAN** Generative Adversarial Networks.

**MD** Molecular Dynamics.

**ML** Machine Learning.



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# Chapter 1

## Introduction

### 1.1 Motivation

Friction is the force that prevents the relative motion of objects in contact. In our everyday life, we recognize it as the inherent resistance to sliding motion. Some surfaces appear slippery and some appear 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 [1, p. 5]. It is probably safe to say that the concept of friction is integrated into our everyday life to such an extent that most people take it for granted. However, the efforts to control friction date 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”. It 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, the reduction of friction is of great interest since this can be utilized to improve energy efficiency in mechanical systems with moving parts. Hence, it has been reported that tribological problems have a significant potential for both 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 finetuned object handling is required. While achieving a certain “constant” friction response is readily obtained through appropriate material choices, we are yet to unlock the full capabilities to alter friction dynamically on the go. One example from nature inspiring us to think along these lines is 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 “toggable” adhesion properties. Although the gecko can produce large adhesive forces, it retains the ability to remove its 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 them 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; slippery and smooth when interacting with people and rough and firmly gripping when moving heavy objects.

In recent years an increasing amount of interest has gone into the studies of the microscopic origins 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 dictate machining characteristics [3]. With concurrent progress in computational capacity and development of Molecular Dynamics (MD), numerical investigations serve as an invaluable 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 the material compositions are designed meticulously to enhance certain physical properties [6–11]. This is often achieved either by intertwining different material types or removing certain regions completely. In recent papers by Hanakata et al. [6, 7], numerical studies have showcased that the mechanical properties of a graphene sheet, 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 the 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<sup>1</sup> problems ruling out analytic solutions.

Earlier 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 computation time on modern-day hardware. However, this computational time constraint can be relaxed by the use of machine learning (ML) which has been 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 altogether resulting in an *accelerated search* of designs. This can be further improved by guiding the search according to the most promising candidates. For instance, as done with the *genetic algorithm* based on mutation and crossing. Another more sophisticated approach is through generative methods such as *Generative Adversarial Networks* (GAN) or diffusion models. The latter is being used in state-of-the-art AI systems such as OpenAI's DALL-E2 [13] or Midjourney [14]. By working with a so-called *encoder-decoder* network structure, one can build a model that reverses the prediction process. This is often referred to as *inverse design*, where the model predicts a design based on physical target properties. In the papers by Hanakata et al. [6, 7] 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. attribute the variation in mechanical 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 Kirigami-induced out-of-plane buckling can also be exploited for the design of frictional metamaterials. For certain designs, we might hope to find a relationship between the stretching of the sheet and frictional properties. If significant, this could give rise to an adjustable friction beyond the point of manufacturing. 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.

In addition, the Kirigami graphene properties can be explored through a potential coupling between the strain and the normal load, through a nanomachine design, with the aim of altering the friction coefficient. This invites the idea of non-linear friction coefficients which might in principle also take on negative values. This would constitute a rarely found property which is mainly observed for the unloading phase of adhesive surfaces [15] or in the loading phase of particular heterojunction materials [16, 17].

To the best of our knowledge, Kirigami has not yet been implemented to alter the frictional properties of a nanoscale system. However, in a recent paper by Liefferink et al. [18] it is reported that macroscale Kirigami can be used to dynamically control the macroscale roughness of a surface through stretching. They reported that the roughness change led to a changeable frictional coefficient by more than one order of magnitude. This supports the idea that Kirigami designs can be used to alter friction, but we believe that taking this concept to the nanoscale would involve a different set of governing mechanisms and thus contribute to new insight in this field.

## 1.2 Goals

In this thesis, we investigate the prospects of altering the frictional properties of a graphene sheet through the application of Kirigami-inspired cuts and stretching of the sheet. With the use of molecular dynamics (MD) simulations, we evaluate the frictional properties of various Kirigami designs under different physical conditions. Based on the MD results, we investigate the possibility to use machine learning for the prediction of frictional properties and subsequently using the model for an accelerated search of new designs. The main goals of the thesis can be summarized as follows.

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<sup>1</sup>In computer science we define an *intractable* problem as a problem with no *efficient* algorithm to solve it nor any analytical solutions. The only way to solve such problems is the *brute-force* approach, simply trying all possible combinations, which is often beyond the capabilities of computational resources.

1. Design an MD simulation procedure to evaluate the frictional properties of a Kirigami graphene sheet under specified physical conditions.
2. Develop a numerical tool to generate various Kirigami designs, both by seeking inspiration from macroscale designs and by the use of a random-walk-based algorithm.
3. Investigate the frictional behavior under varying strain and load for different Kirigami designs.
4. Develop and train a machine learning model to predict the MD simulation results and perform an accelerated search of new designs with the goal of optimizing certain frictional properties.

### 1.3 Contributions

By working towards the goals outlined above (Sec. 1.2), I have discovered a non-linear relationship between the kinetic friction and the strain for certain Kirigami patterns. This phenomenon was found to be associated with the out-of-plane buckling of the Kirigami sheet but with no clear relationship to the contact area or the tension in the sheet. I found that this method does not provide any mechanism for a reduction in friction, in comparison to a non-cut sheet. However, the straining of certain Kirigami sheets allows for a non-monotonic increase in friction. The relationship to normal load was proven negligible in this context and I have demonstrated that a coupled system of load and strain (through sheet tension) can exhibit a negative friction coefficient in certain load ranges. Moreover, I have created a dataset of roughly 10,000 data points for assessing the employment of machine learning and accelerated search of Kirigami designs. I have found, that this approach might be useful, but that it requires an extended dataset in order to produce reliable results for a search of new designs.

During my investigations, I have built three numerical tools, in addition to the usual scripts for data analysis, which are available on Github [19]. The tools are summarized in the following.

- I have written a LAMMPS-based [20] tool for simulating and measuring the frictional properties of a graphene sheet sliding on a substrate. The code is generally made flexible with regard to the choice of sheet configuration, system size, simulation parameters and MD potentials, which makes it applicable for further studies on this topic. I have also built an automated procedure to carry out multiple simulations under varying parameters by submitting jobs to a computational cluster via an ssh connection. This was done by adding minor additions to the Python package developed by E. M. Nordhagen [21].
- I have generated a Python-based tool for generating Kirigami patterns and exporting these in a compatible format with the simulation software created. The generation of molecular structures is done with the use of ASE [22]. Our software includes two classes of patterns inspired by macroscale designs and a random walk algorithm which allows for a variety of different designs through user-defined biases and constraints. Given our system size of choice, the first two pattern generators are capable of generating on the order of  $10^8$  unique designs while the random walk generator allows for significantly more.
- I have built a machine-learning tool based on Pytorch [23] which includes setting up the data loaders, a convolutional network architecture, a loss function, and general algorithms for training and validating the results. Additionally, I have written several scripts for performing grid searches and analyzing the model predictions in the context of the frictional properties of graphene.

All numerical implementations have been originally developed for this thesis except for the libraries mentioned above along with common Python libraries such as Numpy and Matplotlib.

### 1.4 Thesis structure

The thesis is divided into two parts. In Part I we introduce the relevant theoretical background, and in Part II we present the numerical implementations and the results of this thesis.

Part I contains a description of the theoretical background related to Friction (??), Molecular Dynamics (??) and Machine Learning (??). In ?? we formulate our research questions in the light of the friction theory.

In Part II, we begin by presenting the system in ?? which includes a definition of the main parts of the system and the numerical procedures related to the MD simulation. Here we also present the generation of Kirigami designs. In ?? we carry out a pilot study where we evaluate the simulation results for various physical conditions and compare a non-cut sheet to two different Kirigami designs. In ??, we further explore



the Kirigami patterns through the creation of a dataset and the employment of machine learning and an accelerated search for new designs. In ??, we use the results from the pilot study to demonstrate the possibility to achieve a negative friction coefficient for a system with coupled load and strain. Finally, in Chapter 2, we summarize our results and provide an outlook for further studies. Additional figures are shown in ??, ?? and ??.

# Part I

# Background Theory





## Part II

# Simulations





# Chapter 2

## Summary

In this thesis, we have studied the nanoscale friction of a Kirigami graphene sheet under the influence of strain using molecular dynamics (MD) simulations. We have developed a numerical tool for generating diverse Kirigami designs which we have utilized to create a dataset for the frictional behavior depending on Kirigami pattern, strain, and load. Our findings suggest that the frictional behavior of a Kirigami sheet is highly dependent on the geometry of the pattern and the strain conditions. We have observed that the out-of-plane buckling can be associated with a non-monotonous friction-strain relationship which can be utilized to demonstrate a negative friction coefficient in a system with coupled load and strain. Moreover, we have investigated the possibility to use machine learning on this dataset and we have attempted an accelerated search for the optimization of various friction properties. Our findings imply that machine learning can be feasible for this approach, but additional data is required to establish a more reliable foundation for the prediction on new Kirigami patterns. In this chapter, we will provide a summary of our findings and draw conclusions based on the results obtained. Finally, we will suggest some topics for further research.

### 2.1 Summary and conclusions

#### 2.1.1 Designing an MD simulation

We have designed an MD simulation for the examination of friction for a graphene sheet sliding on a silicon substrate. The key system features were the introduction of the pull blocks, defined as the end regions of the sheet with respect to the sliding direction, which was utilized for applying normal load and sliding the sheet. The pull blocks were made partly rigid and used to employ a thermostat as well. Through an analysis of the friction forces retrieved from the sliding simulations, we have established a standardized metric for kinetic friction. In particular, we measured the force exerted by the substrate on the full sheet, including the pull blocks, with respect to the sliding direction. We determined the kinetic friction as the mean value of the friction force for the last half of the simulation. The uncertainties were estimated based on the fluctuations in the running mean. We found that the assessment of static friction was ambiguous for our simulation and did not pursue this further. From the analysis of the force traces, friction force vs. time, we identified the friction behavior in our default parameter simulations as being in the smooth sliding regime. This is attributed to the choice of a relatively high sliding speed ( $v = 20 \text{ m/s}$ ) and infinitely stiff springs ( $K = \infty$ ) for the tethering of the sheet. This was supported by a demonstration of a transition to the stick-slip regime through the use of softer springs ( $K = 10 \frac{\text{N}}{\text{m}}$ ) and a decrease in sliding speed ( $v = 1 \frac{\text{m}}{\text{s}}$ ). By conducting a more systematic investigation of the effects of temperature, sliding speed, spring constant and timestep, we identified a set of default values based on numerical stability and computational cost. During this process, we aimed to select the variables that would maintain relatively stable measures for friction with moderate perturbations around these default values. We found that friction increased with temperature which is in disagreement with the Prandtl–Tomlinson model and most experimental results. However, this agrees with the predictions from the 2D Frenkel–Kontorova model and other MD studies which attribute this to ballistic sliding. In the absence of clear indications regarding an appropriate temperature choice, we opted for the standard choice of room temperature ( $T = 300 \text{ K}$ ). Furthermore, we found friction to increase with sliding speed as expected from both experimental and numerical studies. We found an approximately viscous friction  $F_{\text{fric}} \propto v$  as predicted by the Prandtl–Tomlinson model, with some signs of phonon resonance at certain sliding speeds which aligns with the predictions from the Frenkel–Kontorova models. We chose a rather high sliding speed of  $20 \text{ m/s}$  mainly for the consideration of computational costs. For the spring constant, we found decreasing friction with increasing stiffness of the springs. This is associated with the transition from a stick-slip-influenced



regime toward smooth sliding and can be attributed to an underlying change in commensurability. This is predicted by the Frenkel-Kontorova models and supported by both numerical and experimental results. The choice of an infinitely stiff spring was made from an assessment of the variation in friction with perturbations in the spring constant value. Finally, we confirmed that a timestep of 1 fs provides reasonable numerical stability. However, based on fluctuations with timestep we find that the uncertainty in the simulations might be higher than first estimated. For the non-strained Kirigami sheet, these fluctuations were on the order of  $\pm 0.017 \text{ nN}$  for the evaluation of the kinetic friction.

### 2.1.2 Generating Kirigami patterns

In order to investigate the effects of Kirigami design we have created a numerical tool for generating various patterns. By defining an indexing system for the hexagonal lattice structure we were able to define the Kirigami cut patterns as a 2D binary matrix for numerical implementation. We have selected two macroscale designs, which we denote the *Tetrahedron* and *Honeycomb* patterns, based on their ability to exhibit out-of-plane buckling when subjected to strain. By digitalizing the designs for the hexagonal graphene lattice, we found that the characteristic design features can be translated to the nanoscale, as we observed similar out-of-plane buckling in MD simulations. Through our numerical tool, we were able to create an ensemble of perturbed variations of these patterns which yielded approximately  $1.35 \times 10^5$  and  $2.025 \times 10^6$  unique configurations for the Tetrahedron and Honeycomb patterns respectively. When considering the possibility to translate the periodic patterns on the sheet, the number of unique configurations can be increased by approximately a factor of 100. To introduce some random design features, we have developed a tool for generating Kirigami patterns based on random walks. The tool includes mechanisms such as bias, avoidance of existing cuts, preference for maintaining a direction, and procedures for repairing the sheet for simulation purposes. In general, we found that the numerical tools for creating Kirigami designs surpassed our computational capabilities for conducting MD simulations with various loads and strains for each of the designs. Our MD-based dataset only utilized a subset of configurations with 9660 data points based on 216 Kirigami configurations (Tetrahedron: 68, Honeycomb: 45, Random walk: 100, Pilot study: 3). Hence, we argue that the Kirigami generative tool can be valuable for further studies on an extended dataset.

### 2.1.3 Friction control using Kirigami design and strain

We have investigated the frictional behavior of the Tetrahedron and Honeycomb patterns in comparison to a non-cut sheet under various strains and loads. Initially, we observed that straining the Kirigami sheets in a vacuum resulted in an out-of-plane buckling. When adding the substrate to the simulation, this translated into a decreasing contact area with strain. We found the Honeycomb sheet to exhibit the most significant buckling with a corresponding reduction of relative contact to approximately 43%, whereas the non-cut sheet did not produce any significant buckling in comparison. For the Kirigami sheets, we found that friction initially increased with strain, which made for increasing friction with decreasing contact area. As the strain continued to increase the friction-strain curve exhibited highly non-linear trends with strong negative slopes as well (see ??). During the full strain, the contact area was decreasing monotonically except for a slight increase just before rupturing. These results contradict the asperity theory hypothesis of decreasing friction with decreasing contact area, which is also supported by the predictions of the 2D Frenkel-Kontorova model suggesting increasing friction with (contacting) system size. Thus, we conclude that the contact area is not a governing mechanism for the friction-strain relationship observed. From the investigation of the friction-load relationship, we found that both friction and contact area increased slightly with load, but we were not able to find significant evidence of any linear relationship between friction and contact area as predicted by asperity theory. The non-cut sheet did not show any dependency on the strain as both the friction and the contact area remained constant with strain. Thus our findings suggest that a changing contact area and the strain-induced friction effects might be associated with an underlying mechanism related to the buckling of the sheet. When analyzing the independent effect of the non-strained Kirigami sheets, we found a slight increase in friction between the non-cut sheet and the Kirigami sheets. However, this increase was one order of magnitude lower than the friction changes induced by the strain in combination. Therefore, we can conclude that the observed friction behavior cannot be attributed solely to the effects of the non-strained Kirigami sheet or the tension induced by the strain in a non-cut sheet. When considering the friction dependency with load, we generally found a weak dependency corresponding to a friction coefficient on the order of  $10^{-4}$ – $10^{-5}$  even though we could not confirm any clear relationship. This is best attributed to the superlubric state of the graphene sheet as seen in other studies as well. The slope of the friction-load curve was not considerably affected by the straining of the Kirigami sheet which led us to the conclusion that strain-induced effects are dominant in comparison to any load-related effects.

When considering our findings in light of previous related results, we find it plausible that the governing mechanism for the observed friction effects is related to commensurability as predicted by the Frenkel-Kontorova models. Since we have observed extremely low friction in our system, we argue that the non-deformed sheet corresponds to an incommensurable configuration. This is supported by the fact that we were not able to lower the friction below the original starting point. Additionally, this also aligns with the observation that the introduction of the non-strained Kirigami designs increased friction slightly. Since the Kirigami designs correspond to the removal of atoms from the sheet, this can be hypothesized to relax the incommensurability to some extent. When the Kirigami sheet buckles during stretching, it allows for a considerable rearrangement of the atoms in contact with the substrate. Hence, it may transition in and out of commensurable configurations, which could explain the non-monotonous trend for the friction-strain curve. One way to test this hypothesis is to alter the simulation conditions such that the non-strained sheet starts in a commensurable phase. This might be achieved by a softening of the springs for tethering and a lowering of the sliding speed since this was found to yield stick-slip behavior which can be associated with a commensurable phase. Another way is to reorient the sheet or change the sliding direction as reported in both numerical and experimental results. Then we might find a transition from a commensurable to an incommensurable case during the initial straining which would result in a lowering in friction with respect to the starting point.

#### 2.1.4 Capturing trends with machine learning

By utilizing the numerical tool for generating Kirigami designs, we have created a MD-based dataset for the frictional behavior depending on Kirigami design, load and strain. The dataset reveals some general correlations with mean friction, such as a positive correlation to strain (0.77) and porosity (0.60), and a negative correlation to contact area (-0.67). These results align with the findings in the pilot study, suggesting that the change in friction is associated with cuts in the sheet (porosity) and a changing contract area representing out-of-plane buckling.

By defining the friction property metrics:  $\min F_{\text{fric}}$ ,  $\max F_{\text{fric}}$ ,  $\max \Delta F_{\text{fric}}$  and  $\max \text{drop}$  (maximum decrease in friction with strain), we investigated the top design candidates within our dataset. From these results, we found no indication of the possibility to reduce friction with the Kirigami approach since the non-cut sheet provided the overall lowest friction. Furthermore, among the top candidates, we found that a flat friction-strain profile is mainly associated with little decrease in the contact area and vice versa. These observations are consistent with the results of the pilot study and support the hypothesis that commensurability plays a key role in governing the behavior of the system. In terms of the maximum properties, we observed an improvement compared to the values obtained in the pilot study, with the Honeycomb patterns exhibiting the highest scores. This indicates that the dataset contains relevant information for optimizing these properties since it includes examples of design improvements.

For the machine learning investigation, we have implemented a VGGNet-inspired convolutional neural network with a deep “stairlike” architecture: C32-C64-C128-C256-C512-C1024-D1024-D512-D256-D128-D64-D32, for convolutional layers  $C$  with the number denoting channels and fully connected (dense) layers  $D$  with the number denoting nodes. The final model contained  $1.3 \times 10^7$  parameters and was trained using the ADAM optimizer for a cyclic learning rate and momentum scheme. We trained the network for a total of 1000 epochs while saving the best model during training based on the validation score. The model validation performance gives a mean friction  $R^2$  score of  $\sim 98\%$  and a rupture accuracy of  $\sim 96\%$ . However, we got lower scores for a selected subset of the Tetrahedon ( $R^2 \sim 88.7\%$ ) and Honeycomb ( $R^2 \sim 96.6\%$ ) patterns based on the top 10 max drop property scores respectively. The scores obtained were lower, even though the selected configurations were partly included in the training data and the hyperparameter selection favored the performance on this selected set. Thus we conclude that these selected configurations, associated with a highly non-linear friction-strain curve, represent a bigger challenge for machine learning modeling. One interpretation is that these involve the most complex dynamics and perhaps that this is not readily distinguished from the behavior of the other configurations which constitute the majority of the dataset. By evaluating the ability of the model to rank the dataset based on property scores, we found that it was able to effectively represent the top three scores for the maximum categories. However, the ranking for the minimum friction property was lacking, which we attribute to the requirement of higher prediction precision that the model did not meet. To obtain a more accurate evaluation of the model’s performance, we generated a test set using MD simulations for some of the random-walk-based suggestions obtained from the accelerated search. The results showed a significantly worse performance compared to the validation set, with a two-order of magnitude higher loss and a negative  $R^2$  score for the mean friction property. The negative  $R^2$  score suggests that the model’s predictions were worse than simply predicting the mean value of the true data. However, by reevaluating the hypertuning and choosing solely based on validation loss,



we still found poor results on the test set. This suggests, that the inadequate performance is not solely due to a biased hypertuning process, but rather because our original dataset did not cover a sufficiently diverse range of Kirigami configurations. The validation scores indicate that the use of machine learning is a feasible approach for addressing this problem, as we were able to identify some general trends in the data. Nonetheless, from the test scores, it is evident that further improvements to the dataset are necessary in order to develop a reliable model.

### 2.1.5 Accelerated search for Kirigami patterns

Using the machine learning model we performed two types of accelerated search. One by evaluating the property scores of an extended dataset and another with the use of the genetic algorithm approach. For the extended dataset search, we used the developed pattern generators to generate  $1.35 \times 10^6$  Tetrahedron,  $2.025 \times 10^7$  Honeycomb and  $10^4$  Random walk patterns. The search results for the minimum friction property indicate a preference for low cut density. This aligns with the overall observation that the dataset does not provide any suggestions for a further reduction in friction. The search for the maximum properties resulted in some minor score increases, but the suggested candidates were mainly overlapping with the original dataset. By investigating the sensitivity to translations of the Tetrahedron and Honeycomb patterns, we observed significant variations in the model predictions with only minor translations. This might be attributed to a physical dependency since these translations affect the edge of the sheet. However, given the poor model performance on the test set, we believe it is more likely that this variation is due to an insufficiency in the model caused by the limitations of the dataset.

In our investigation of the genetic algorithm approach, we used a starting population that was based on the results from the extended dataset accelerated search, as well as some randomly generated initializations with different porosity values. However, this approach did not provide any noteworthy indication for new design structures worth more investigation. In general, the initialization of the population itself proved to be a more promising strategy than the genetic algorithm. We acknowledge that further effort could potentially yield useful results with the genetic algorithm approach. However, we believe that the current lack of promising results can be attributed to the uncertainty of the model which where the reason for not pursuing this any further.

By considering the Grad-CAM explanation method, we observed that the model predictions were often substantially reliant on the top and bottom edges of the Kirigami configurations. This was unexpected since these edges are not true edges but are connected to the pull blocks used in the simulation. Despite the model's uncertain predictions, we speculate that this may be due to the thermostat effects from the pull blocks. Therefore, we note this as a feature worth investigating in the simulations.

### 2.1.6 Negative friction coefficient

Based on our initial investigations of the Kirigami sheet, we have discovered a highly non-linear friction-strain relationship. By proposing a linear coupling between load and strain with ratio  $R$ , we found that these results suggest the possibility to utilize the negative slope on the friction-strain curve to achieve a negative friction coefficient. Based on the local decrease in friction, using the Tetrahedron (7, 5, 1) and Honeycomb (2, 2, 1, 5) patterns from the pilot study, we estimate that the average coefficient within this range will be  $-R \cdot 12.75 \text{ nN}$  for the Tetrahedron pattern and  $-R \cdot 2.72 \text{ nN}$  for the Honeycomb pattern.

To investigate this hypothesis, we conducted a simulation with a coupling between load and sheet tension, mimicking a nanomachine attached to the sheet, using the Kirigami configurations from the Pilot study. We observed that the non-linear behavior in the friction-strain curve also translated into a non-linear friction-load relationship for the coupled system. Additionally, we found that the Honeycomb pattern exhibited a non-linear strain-tension curve which resulted in an almost discontinuous increase in friction for the initial increase in load. We attribute this feature to an unfolding process visually confirmed from the simulation frames. For the coupled system with a load-to-tension ratio of  $T = 6$ , we found regions in the friction-load curve with significant negative slopes. By considering the maximum and minimum points for these regions we estimated the average friction coefficient to be  $-0.31$  in the load range  $F_N = [4.65, 6.55] \text{ nN}$  for the Tetrahedron pattern and  $-0.38$  in the range  $F_N = [0.71, 4.31] \text{ nN}$  for the Honeycomb pattern. These results can be scaled by adjusting the load-to-tension ratio  $T$ . By neglecting any independent effects from load, these results suggest that a negative friction coefficient on the order of  $\sim -0.3$  corresponds to the general coefficient  $-0.05T$ .

Based on our investigations, we have found that the combination of Kirigami cuts and strain has significant potential for controlling friction. Specifically, we have demonstrated that by enforcing a coupling between load and strain through tension, it is possible to achieve a negative friction coefficient. Therefore, we believe that this approach could be promising for developing novel friction-control strategies.

## 2.2 Outlook

In this thesis, we have demonstrated that certain Kirigami designs exhibit non-linear friction behavior with strain. This discovery was made through an exploration of different designs, which invites further investigation into the underlying mechanisms of this phenomenon. To this end, it would be valuable to choose only one or two selected designs, such as the Tetrahedron and Honeycomb patterns, and perform an extended study of the effects of various physical parameters on the friction-strain curve.

First of all, we suggest an investigation of how the friction-strain curve depends on temperature, sliding speed, spring constant, and loads for an increased range  $F_N > 100nN$ . This is especially interesting in the context of physical conditions leading to a stick-slip behavior since our study takes a basis in smooth sliding friction. Moreover, it would be valuable to verify that the choices for relaxation time, pauses, interatomic potentials and substrate material are not critical for the qualitative observations found for the friction-strain relationship. Especially, the Adaptive Intermolecular Reactive Empirical Bond Order (AIREBO) potential for the modeling of the graphene sheet might be of interest. In this context, it might also be useful to investigate the effects of excluding adhesion from the simulations.

In order to investigate the hypothesis of commensurability as a governing mechanism, we suggest an investigation of the friction-strain curve for different scan angles. If commensurability is an important factor, we hypothesize that the friction-strain curve will exhibit different qualitative shapes at varying scan angles. Additionally, it may be interesting to investigate the friction-strain relationship under a uniform load on the entire sheet to gain insight into how the loading distribution affects the out-of-plane buckling and associated commensurability effect.

Another topic worth exploring is the impact of scale and edge effects. This includes an investigation of scaling the ratio of the sheet area to the sheet edge length. However, the machine learning predictions also suggest that it may be valuable to study Kirigami-induced edge effects as we translate the patterns on the sheet. With this regard, we would also suggest a more detailed study of the effect of the thermostat in the pull blocks which is suggested to have a possible impact when judging from the Grad-CAM analysis.

Regarding the machine learning approach, our findings indicate that there is a significant need to expand the dataset. In order to get more insight into this issue one could use unsupervised clustering techniques like the t-Distributed Stochastic Neighbor Embedding (t-SNE) to visualize the distribution of Kirigami configurations in the dataset. Another valuable approach is the active learning method similar to that used by Hanakata et al. [6]. That is, we extend the dataset using the top candidates of a machine learning-driven accelerated search and repeat the process of training the model, searching for new candidates and extending the dataset. This provides a direction for the extension of the dataset which could lead to a more efficient approach to address the dataset problem. We note that one can also create a dataset based on a fixed Kirigami design and vary the physical parameters to support the investigations mentioned above. For both variations, we believe that the results could benefit from a consideration of more advanced model architectures and machine learning techniques. For instance, we suggest increasing the receptive field in the convolutional part of the model, by the use of larger strides or dilated convolution. If we can develop a reliable machine learning model, it would invite further studies of inverse design methods such as GAN or diffusion models.





# Appendices





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