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

Designs for a negative friction coefficient.

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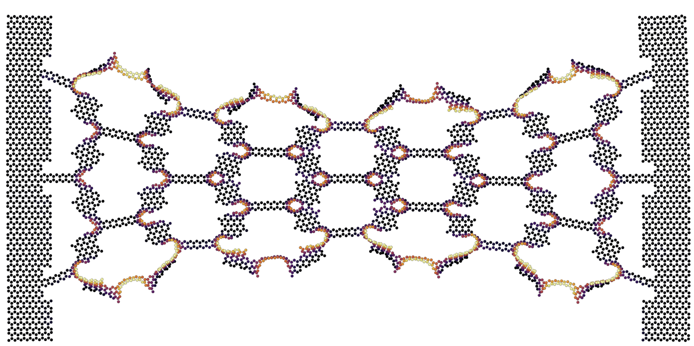

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Abstract

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Acknowledgments

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List of Symbols

F_N Normal force (normal load)

Acronyms

LJ Lennard-Jones. 8

MD Molecular Dynamics. 5, 6, 7

ML Machine Learning. 7

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Part I

Background Theory

Part II

Simulations

Chapter 1

Summary

The work presented in this thesis covers several topics (find a better opening line). We have created an MD simulation which enabled us to study the frictional behavior of a graphene sheet sliding on a Si substrate. In addition, we have created a numerical framework for creating Kirigami design patterns and introducing these into the friction simulations. This was used to study the effects of the out-of-plane buckling induced by a selected pair of Kirigami designs in comparison to a non-cut sheet under the influence of strain. Further, we have created a dataset of various Kirigami designs for the scope of investigating the possibilities with Kirigami design. We have investigated the possibility to use machine learning on this dataset and attempted an accelerated search. Finally we look into the prospects of achieving a negative friction coefficient for a system with coupled load and stretch. In this chapter we will summarize the findings and draw some conclusions. We will also provide some topics for further research.

1.1 Summary and conclusions

1.1.1 Design MD simulations

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. Similarly, we divided the substrate into three parts with the lowermost part being rigidly locked in place, the middle part reserved for a thermostat and the upper part remaining free from further modifications. By an analysis of the friction forces from a sliding simulation we settled on a standardization of the kinetic friction metric as the mean value of the last half of the sliding simulation. This was measured with respect to the sliding distance and for the full sheet (including the pull blocks). We defined the uncertainty of the measurements through the standard deviation of 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 identify the friction behavior in our simulation domain as being in the smooth sliding regime. This is further supported by a demonstration of a transition to stick-slip behavior for softer springs and lower sliding speed which points toward our system being in the ballistic sliding regime. By conducting a more systematic investigation of the effects of temperature, sliding speed, spring constant and timestep, we settled on standardized choices based on numerical stability and computational cost. We found that friction increased with temperature which disagrees with experimental and theoretical considerations but matches other MD. This is attributed to simulation conditions corresponding to a ballistic regime. We choose a room temperature 300 K as a standard choice. We found friction to increase with velocity as expected with some signs of phonon resonance sliding speeds as well. We chose a rather high velocity of 20 m/s mainly for the consideration of computational cost. For the spring constant, we found decreasing friction with increasing stiffness of the springs which is associated with the transition from a stick-slip-influenced regime toward smooth sliding. The choice of an infintely stiff spring was made from a stability assessment. Finally, we confirmed that a timestep of 1 fs provides reasonable numerical stability. However, some fluctuations were observed, especially for the Honeycomb pattern, which might be interpreted as a sign of a higher uncertainty than expected on the order of ± 0.017 nN.

1.1.2 Design Kirigami framework

We have desgined a numerical framework for creating Kirigami designs. By defining an indexing system for the hexagonal lattice structure we were able to define the Kirigami designs as 2D matrix for numerical implementation. We digitalized two different macroscale designs, which we named the *Tetrahedron* and *Honeycomb* pattern respectively, that successfully produced out-of-plane buckling when stretched. Through a numerical framework we could create an ensemble of perturbed variations which gave approximately 135k configurations for the Tetrahedron pattern and 2025k patterns for the size of the sheet used in our study. When considering the possibility to translate the patterns this gave roughly a factor 100 more of unique pertubations. We also created a framework for creating Kirigami designs through a random walk. This was further controlled by introducing features such as bias, avoidance of existing cuts, preference to keeping a direction and procedures to repair the sheet for simulation purposes. The capabilities of the numerical framework for generating Kirigami designs was far larger than the capabilities for producing MD designs within the time constraint of this thesis. Thus we believe that this contains the possibility to benefit more extended studies and for the creation of a larger dataset.

1.1.3 Control friction using Kirigami

We have investigated the friction behavior of a non-cut sheet in comparison to the Tetrahedron and Honeycomb pattern under various strain and load. Initially we observed that straining the Kirigami sheet resulted in a out-of-plane buckling in vaccum. When adding the substrate to the simulation this translated to a reduced contact area with strain. We find the Honeycomb sheet buckled the most resulting in a reduction of the contact area to approximately 43%. The non-cut sheet did not produce any significant buckling in comparison. However, when considering the friction effects we found that friction generally increased with strain with contradicts the asperity theory hypothesis of decreasing friction with decreasing contact area. Moreover, the friction-strain curve exhibited highly non-linear trends with strong negative slopes as well as shown in **??**. The non-cut sheet did not reveal any significant dependency on the strain. This led us to the conclusion that the contact area nor tension alone (for a non-cut sheet) can regarded as a dominant mechanism for friction in the graphene sheet system. When considering the dependency with load we generally found a weak dependency which canassociated to friction coefficients 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 on the substrate. The slope of the friction-load curves was also not affected by the straining of the Kirigami sheet which led us to the conclusion that the strain-induced effects are dominant in comparison to load-related effects. By proposing a linear coupling between load and strain with ratio R we find that the results allow for negative friction coefficients for the Tetrahedron pattern on $-R 12.75\,\mathrm{nN}$ and Honeycomb $-R \cdot 2.72\,\mathrm{nN}$

1.1.4 Capture trends with ML

With the use of MD simulations, we have generated an extended dataset of 9660 data points based on 216 Kirigami configurations (Tetrahedron: 68, Honeycomb: 45, Random walk: 100, Pilot study: 3) under various strains and normal loads. 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 from the pilot study suggesting that these features are relevant, but not necessarily the cause, of the observed phenomena. By defining the friction property metrics: $\min F_{\text{fric}}$, $\max F_{\text{fric}}$, $\max \Delta F_{\text{fric}}$ and max drop (maximum decrease in friciton with strain), we investigated the top candidates within our dataset. From these results, we found no incentive of the possibility to reduce friction with the Kirigami approach since the non-cut sheet provided the lowest overall friction. Regarding the maximum properties, we found an improvement from the original pilot study values and with the Honeycomb pattern producing the highest scores. This suggests that the data contains some relevant information for optimization with respect to these properties. 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.

For the machine learning investigation, we have implemented a VGGNet-16-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 contains 1.3×10^7 and was trained using the ADAM optimizer for a cyclic learning rate and momentum scheme for 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 ($R^2 \sim 96.6$) pattern based on the top 10 max drop scores respectively. These scores were lower despite the fact that the selected set was partly included in the training data as well and the fact that 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 challennge for machine learning prediction. 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 constitutes the majoriry of the data set. By evaluating the ability for the model to rank the dataset according to the property scores we found in general a good representation of the top 3 scores for the maximum categories, while the minimum friction property ranking was lacking. We attribute this latter observation to a higher need for precision in order to rank the lowest friction values properly which the model did not possess.

In order provide a more true evaluation of the model performance we created a test set based on MD simulations for an extended Random walk search. This test revealed a significantly worse performance than seen for the validation set with a two-order of magnitude higher loss and a negative friction mean R^2 score which corresponds to the prediction being worse than simply guessing on a constant value based on the true data mean. However, by considering one of the early hypertuning choices, regarding architecture complexity, we evaluated the model when prioritizing mainly for the lowest validation loss. This gave similar performance on the test set which indicates that it is not simply a product of a biased hypertuning process, since we based our choices on the selected confgiruation set (which overlapped with the training data). Instead, it points to the fact that our original dataset did not cover a wide enough configuration distribution to accurately capture the full physical complexity of the Kirigami friction behavior.

1.1.5 Accelerated search

Using the ML 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 $135\,\mathrm{k} \times 10$ Tetrahedon, $2025\,\mathrm{k} \times 10$ Honeycomb and $10\,\mathrm{k}$ Random walk patterns. For the minimum friction property, the search suggests a favoring of a low cut density (low porosity) which aligns with the overall idea that the dataset does not provide an incentive for further friction reduction. The maximum properties resulted in some minor score increases but the suggested candidates were overlapping with the original dataset. By investigating the sensitivity to translation of the Tetrahedron and Honeycomb patterns we found that the model predictions varied drastically with pattern translation. This can be attributed to a physical dependency since the edge of the sheet is effected by this translation. However, due to the poor model performance on the test set, we find it more likely to be a model insufficienty arising from a lacking training dataset.

For the genetic algorithm approach, we investigated the optimization for the max drop property with respect to starting population based on the result from the extended dataset accelerated search, and some random noise initializations with different porosity values. This approach did not provide any noteworthy incentive for new design structures worth more investigation. In general, the initialization of the population itself proved to be a more promising strategy tha the genetic algroithm. However, this is highly affected by the uncertainty of the model predictions, and thus we did not pursue this any further. By considering the Grad-CAM explanation method we found that the model predictions sometimes seem to pay considerable attention to the top and bottom edge of the configurations. This is surprising since these are not true edges but are connected to the pull blocks in the simulation. Despite the uncertainties in the predictions, we argue that this might be attributed to thermostat effects from the pull blocks and thus we note this as a feature worth more studying.

1.1.6 Negative friction coefficient

By enforcing a coupling between load and stretch, mimicking a nanomachine attached to the sheet, we investigated the load curves arising from loading of the Tetrahedron $(7, 5, 1)$ and Honeycomb $(2, 2, 1, 5)$ pattern from the pilot study. The non-linear trend observed for increasing strain carried over to the coupled system as well producing a highly non-linear friction-load curve. This demonstrates a negative friction coefficient say something about the values.

1.2 Outlook / Perspective

Having successfully demonstrated a non-linear effect on friction with increasing strain of the sheet our results invite a series of further studies to investigate this relation. First of all, it would be valuable to investigate how the friction-strain curve depend on temperature, sliding speed, spring constant, and on load for an increased range $F_N > 100nN$. This is especially interesting in the context up conditions leading to a stick-slip behvaior as our results were carried in the smooth sliding. Moreover, it would be important to verify that the choices for relaxation time and pauses are not critical for the qualitative observations as well as trying a different interatomic potential for the graphene and perhaps an entirely different substrate material. Especially the Adaptive Intermolecular Reactive Empirical Bond Order (AIREBO) potential for the modeling of the graphene sheet might be of interest. The effects from excluding adhesion (the LJ interaction) can also be useful for the investigation of the observed phenomena.

In order to get a better understanding of the underlying mechanism for the friction-strain relationship we might investigate commensurability effects further by varying the scan angle. we might also consider investigating the friction-strain relationship under a uniform load to get insight into whether the loading distribution is of importance. Another topic worth studying is the relation to scale. Thus it would be interesting to study size effects but also further look into edge effects by translating the pattern. With this regard, we would also suggest a more detailed study of the effect from the thermostat in the pull blocks which is suggested to have a possibly importance by judging from the machine learning model Grad-CAM analysis.

For machine learning, we can either try to extend the data set to resolve the issue of the model not being generalized enough. We can also create a dataset for a single kirigami design and include some of the mentioned physical variables above and attempt to use machine learning for unraveling these relations. In that context we would advice for as more detailed investigation of machine learning techniques. If succesfull this would invite a study of inverse design methods such as GAN or diffusion models.

- How is this behavior effected by scaling?
- How does the distirbution of normal load effect the Kirigami friction behavior?
- Things to vary: load range, scan directions, adhesive forces, longer relaxation time, different potential (AIREBO)
- Investigate if the contact area is effecting the friciton non-linear by turning of friction force for atoms corresopnding to those that lift of from the sheet during the out-of-plane buckling.
- Investigations of commensurability effects.
- Study dependency of translation of the patterns as suggested by the ML results.
- Investigate effects from pull blocks...
- Investigate effects from the thermostat since the top and bottom edges was shown interest by the model prediction.

Appendices

Appendix A

Appendix A

Appendix A

Appendix B

Appendix B

Appendix C