

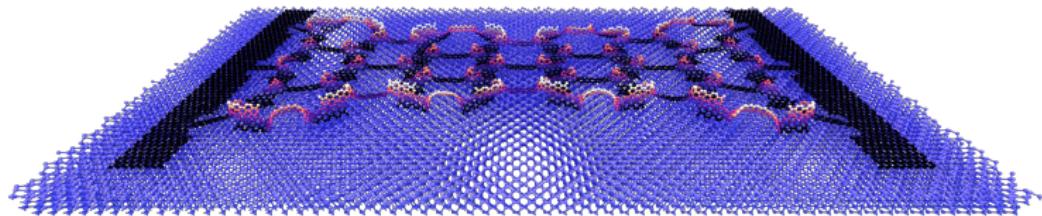
Predicting Frictional Properties of Graphene Kirigami Using Molecular Dynamics and Neural Networks

Designs for a negative friction coefficient

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Juni 02, 2023



Outline

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② Creating a graphene Kirigami system

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Kirigami

Molecular Dynamics

③ Pilot study

Friction metrics

Out-of-plane buckling

Friction-strain profiles

Negative friction coefficient

④ Kirigami configuration search

Machine learning

Accelerated search

⑤ Summary and outlook

- ① **Sheet modification:** Alter a graphene sheet using atomic scale cuts and stretching
- ② **Forward simulation:** Calculate the frictional properties of the sheet using MD simulations
- ③ **Accelerated search:** Use machine learning to replace the MD simulations and perform an accelerated search for new designs

Main research question

Can we control the friction of a nanoscale Kirigami sheet with pattern design and straining of the sheet?

Motivation

Kirigami

- Kirigami: Variation of origami with cuts permitted
- Designs: Macroscale → nanoscale

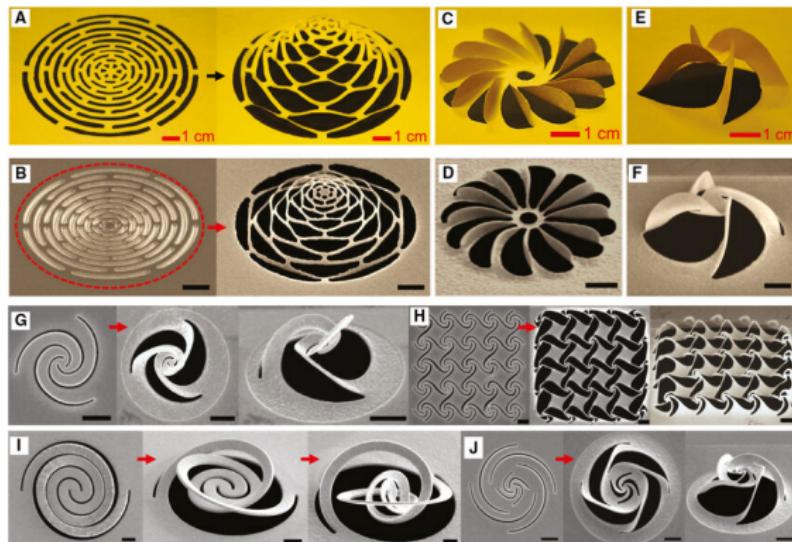
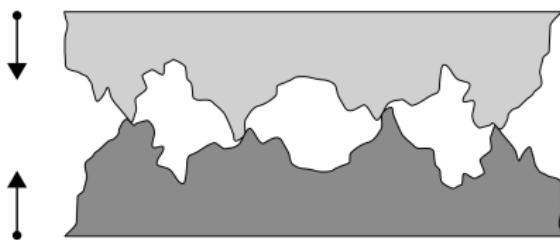


Figure: Example of macroscale Kirigami designs implemented on a microscale using a focused ion beam. Black scale bars: $1 \mu\text{m}$. Reproduced from [1].

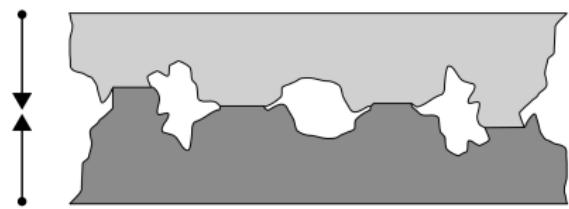
Motivation

Out-of-plane buckling

- Hanakata et al. [2, 3] found out-of-plane buckling with Kirigami designs
- Surface properties are predicted to be important for friction properties
 - Asperity theory: Contact area
 - Frenkel–Kontorova models: Commensurability



(a) Small load.

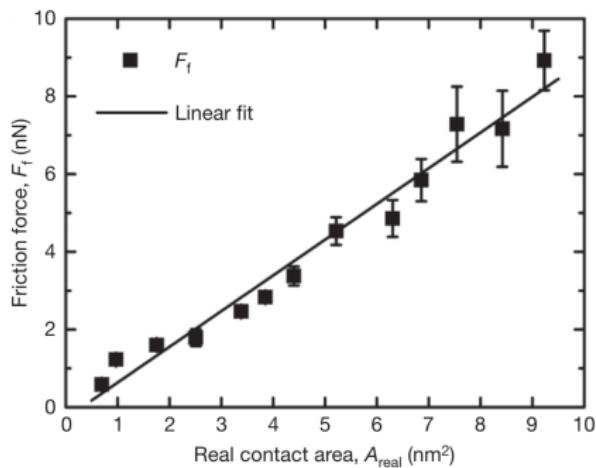


(b) High load.

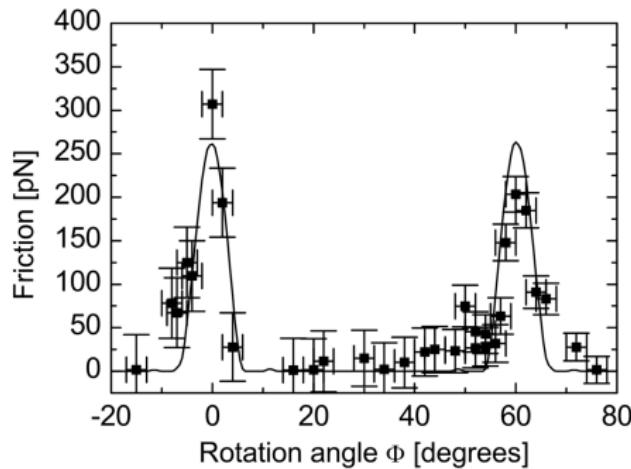
Figure: Qualitative illustration of microscopic asperity deformation under increasing load.
Reproduced from [4].

Motivation

Contact area and commensurability



(a) Numerical MD results using an amorphous carbon tip and a diamond sample. Reproduced from [5] with permission from Springer Nature.



(b) Experimental results of a graphene sheet sliding on graphite. Adapted from [6], reproduced from [7] with permission from the American Physical Society.

Creating a graphene Kirigami system

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System setup

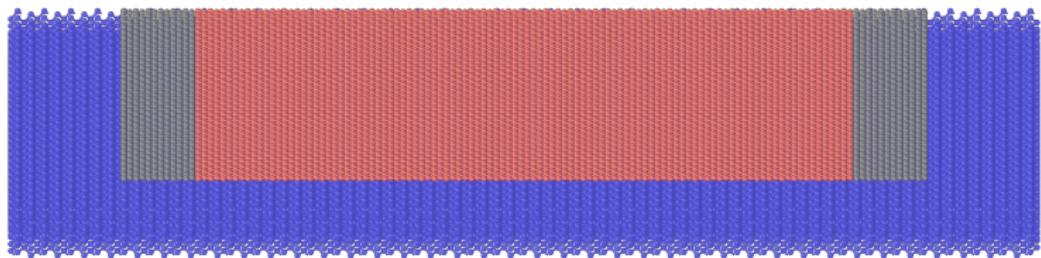


Figure: System of choice: Graphene sheet on a silicon substrate. Blue: Substrate, Red: Inner sheet, Grey: Pull blocks.

System setup

System size

Atoms $\sim 60,000$

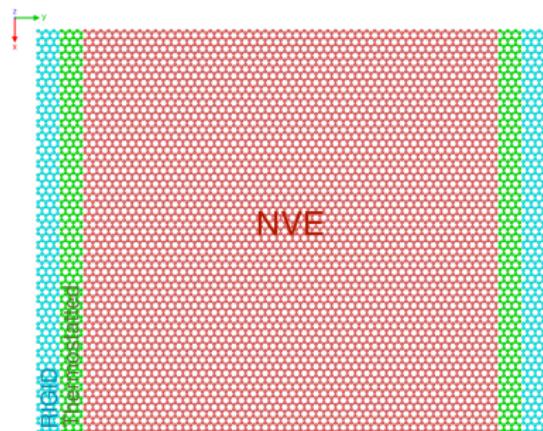
Sheet $\sim 130 \times 165 \text{ \AA}$

Regions

- Red: NVE
- Green: Thermostat NVT
- Blue: Rigid



(a) Side view.



(b) Top view.

Sheet Kirigami

Indexing

$$M \in \mathbb{Z}_2^{62 \times 106}, \quad \text{Combinations} = 2^{6572} = 10^{1978}$$

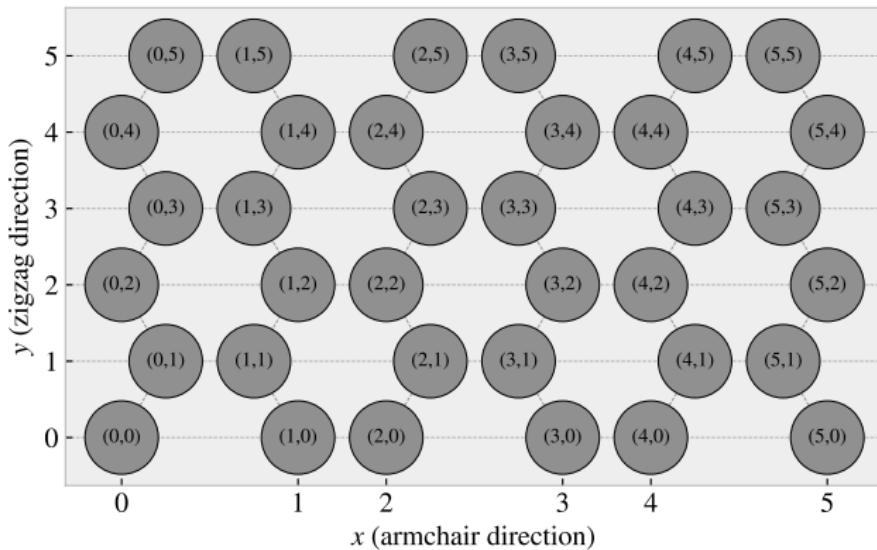


Figure: Graphene atom site indexing.

Sheet Kirigami

Indexing

$$M \in \mathbb{Z}_2^{62 \times 106}, \quad \text{Combinations} = 2^{6572} = 10^{1978}$$

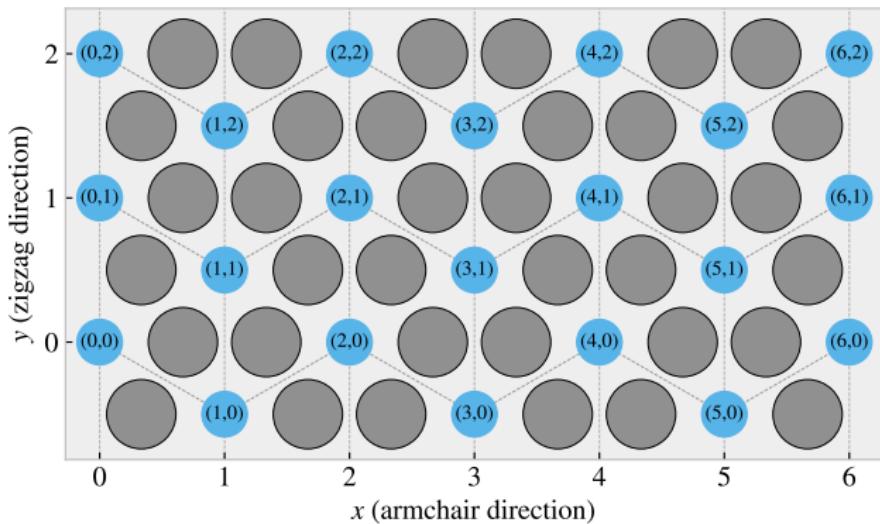
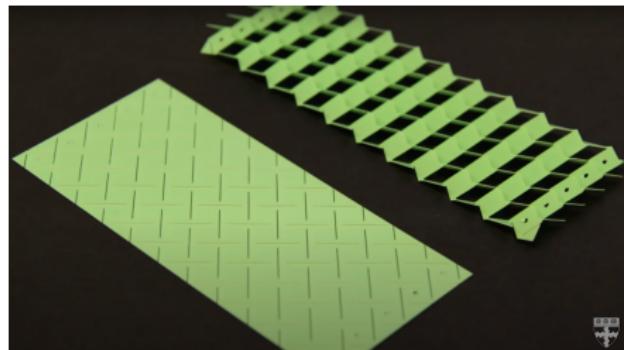


Figure: Graphene center element indexing.

Sheet Kirigami

Macroscale inspiration



(a) Tetrahedron: Alternating perpendicular cuts producing a tetrahedron-shaped surface buckling when stretched. Reproduced from [8].

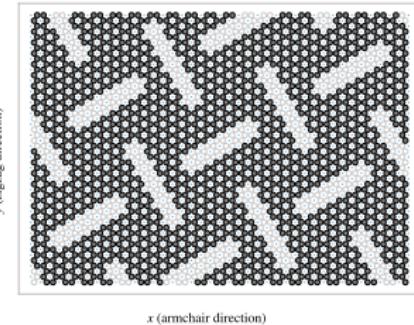
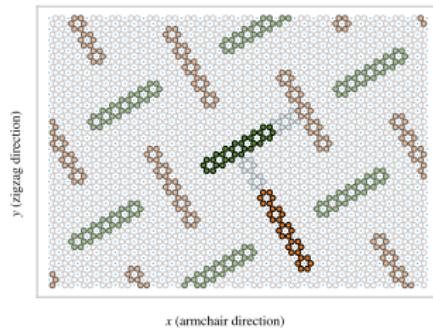


(b) Honeycomb: ScotchTM Cushion LockTM [9] producing a honeycomb-shaped surface buckling when stretched. Reproduced from [9].

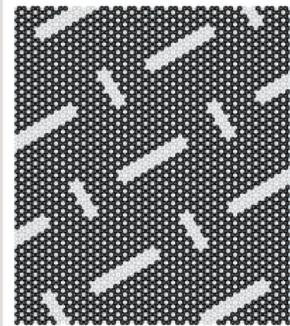
Figure: Macroscale kirigami cut patterns used as inspiration for the nanoscale implementation.

Sheet Kirigami

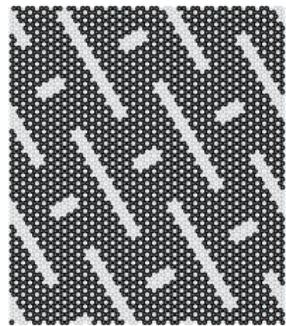
Tetrahedron patterns



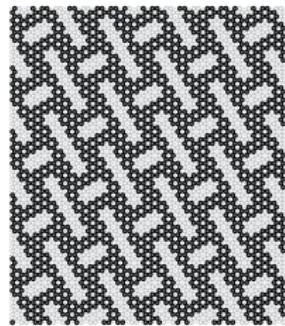
(9, 3, 4)



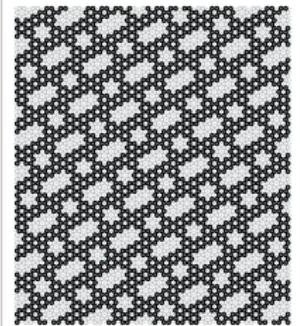
(3, 9, 3)



(3, 5, 1)



(3, 1, 1)

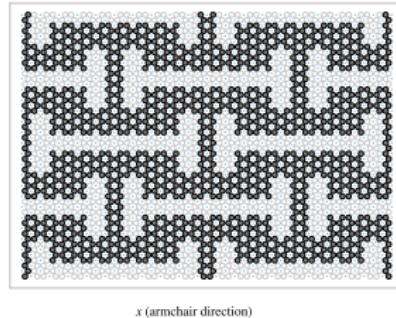
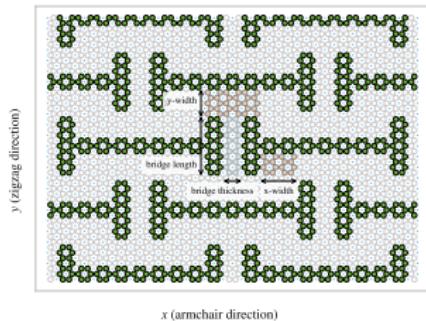


y (zigzag direction)

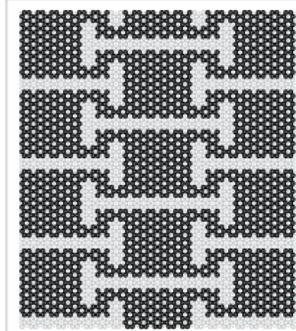
x (armchair direction)

Sheet Kirigami

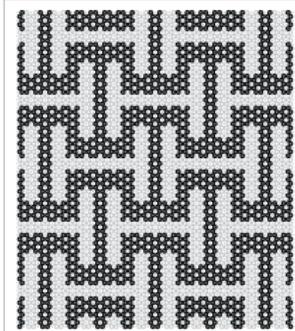
Honeycomb patterns



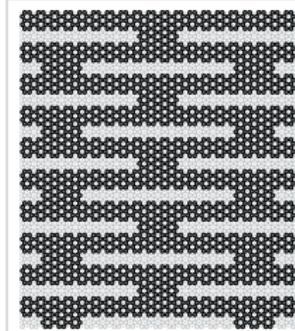
(1, 1, 5, 5)



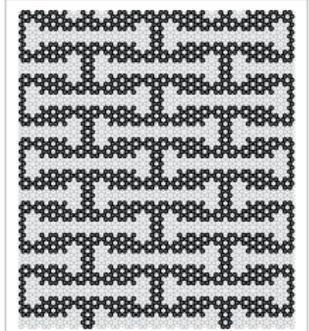
(1, 2, 1, 9)



(2, 2, 3, 1)

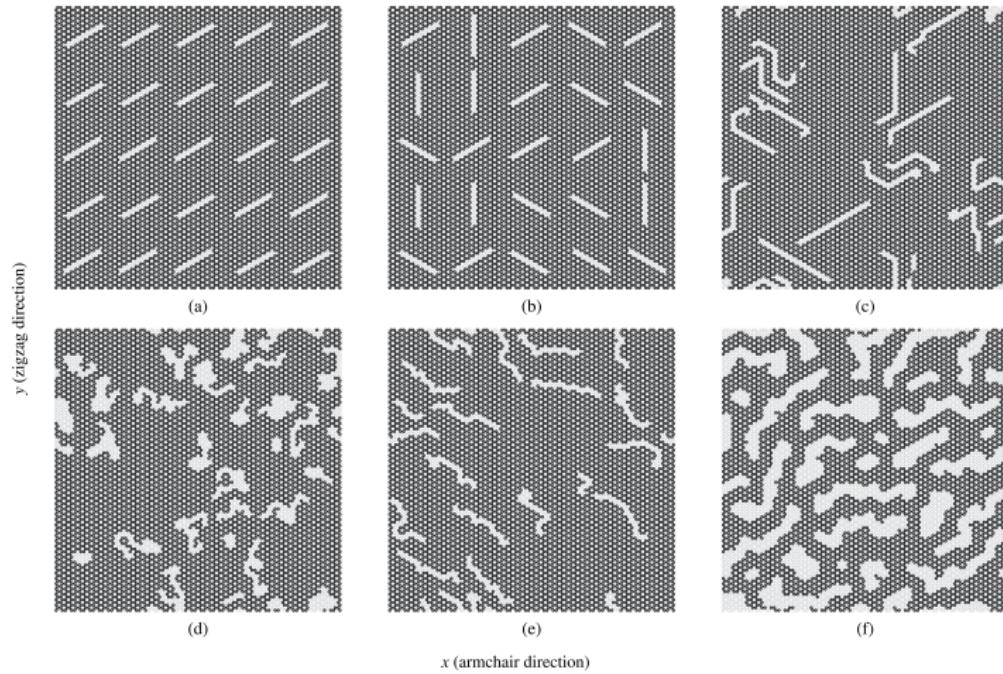


(2, 1, 1, 3)



Sheet Kirigami

Random walk patterns



Molecular Dynamics (MD)

- Newtons equations (NVE)

$$m_i \frac{d^2 \mathbf{r}_i}{dt^2} = \mathbf{F}_i = -\nabla U_i,$$

- Introducing temperature (NVT) with the Langevin equation

$$m_i \frac{d^2 \mathbf{r}_i}{dt^2} = \underbrace{-\nabla U_i}_{F_i} \quad \underbrace{-\alpha \mathbf{v}_i}_{\text{Drag}} \quad + \quad \underbrace{\mathbf{R}_i}_{\text{Fluctuation}},$$

$$\langle \mathbf{R} \rangle = 0, \quad \langle \mathbf{R}^2 \rangle = 2\alpha k_B T.$$

Pilot study

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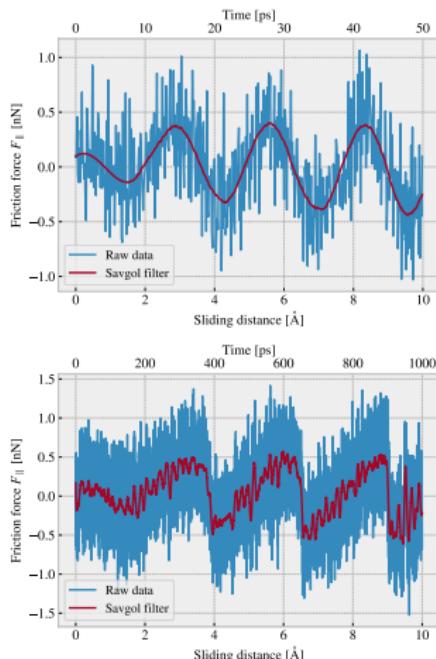
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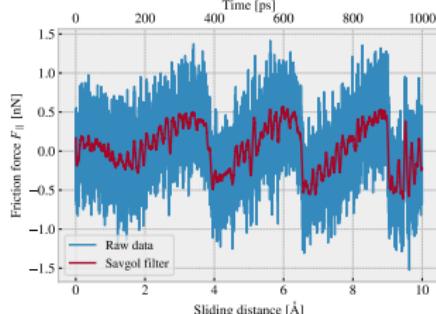
(a)

$K = \infty$,
 $v = 20 \text{ m/s}$
(10 Å sliding).



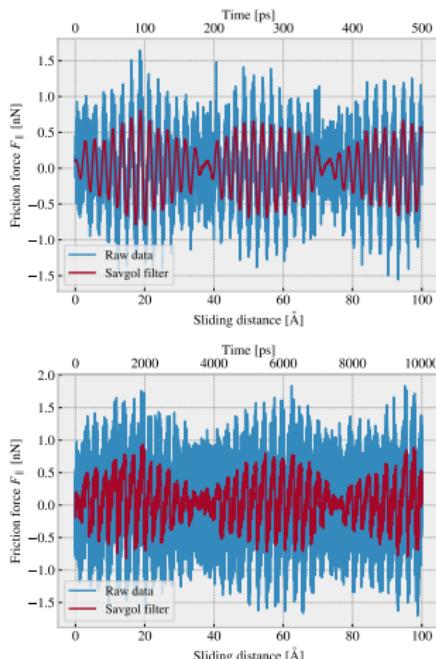
(c)

$K = 10 \text{ N/m}$,
 $v = 1 \text{ m/s}$
(10 Å sliding).



(b)

$K = \infty$,
 $v = 20 \text{ m/s}$
(100 Å sliding).



(d)

$K = 10 \text{ N/m}$,
 $v = 1 \text{ m/s}$
(100 Å sliding).

Figure: Friction force traces. The red line represents a Savgol filter.

Out-of-plane buckling

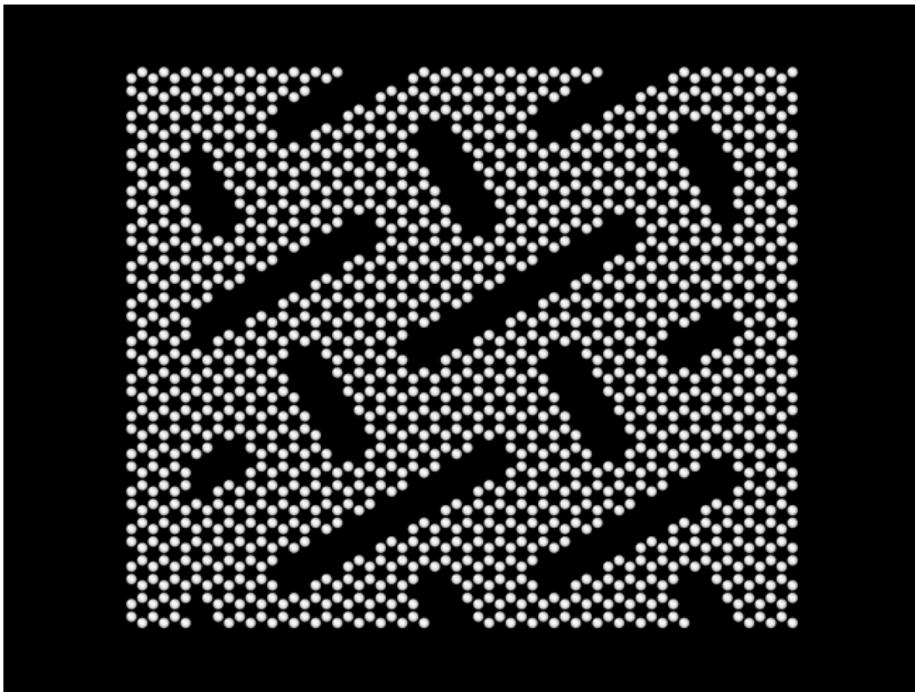


Figure: Kirigami sheet stretch in a vacuum.

Out-of-plane buckling

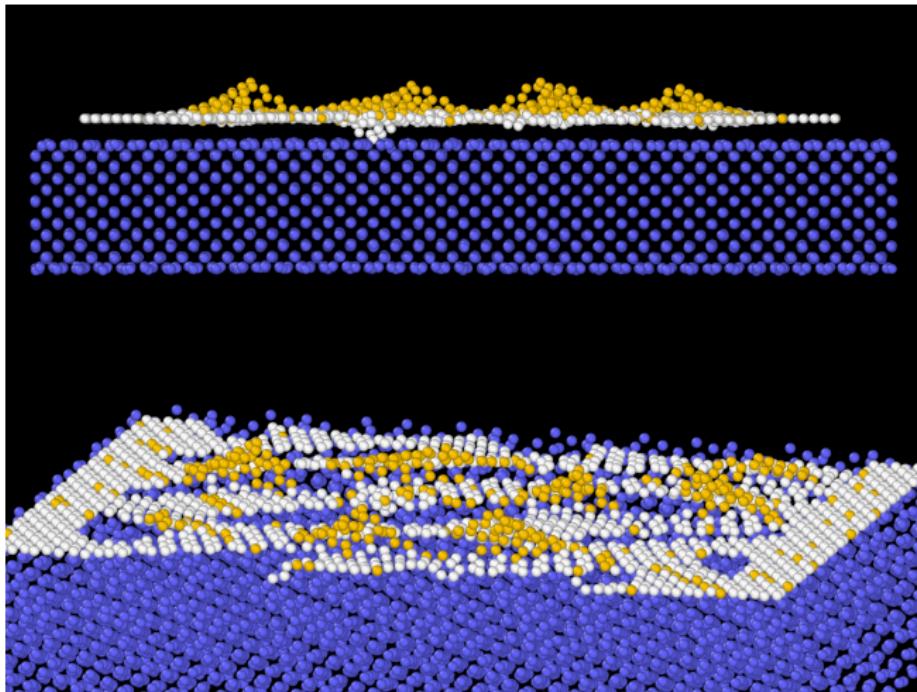


Figure: Kirigami stretch in contact with the substrate.

Out-of-plane buckling

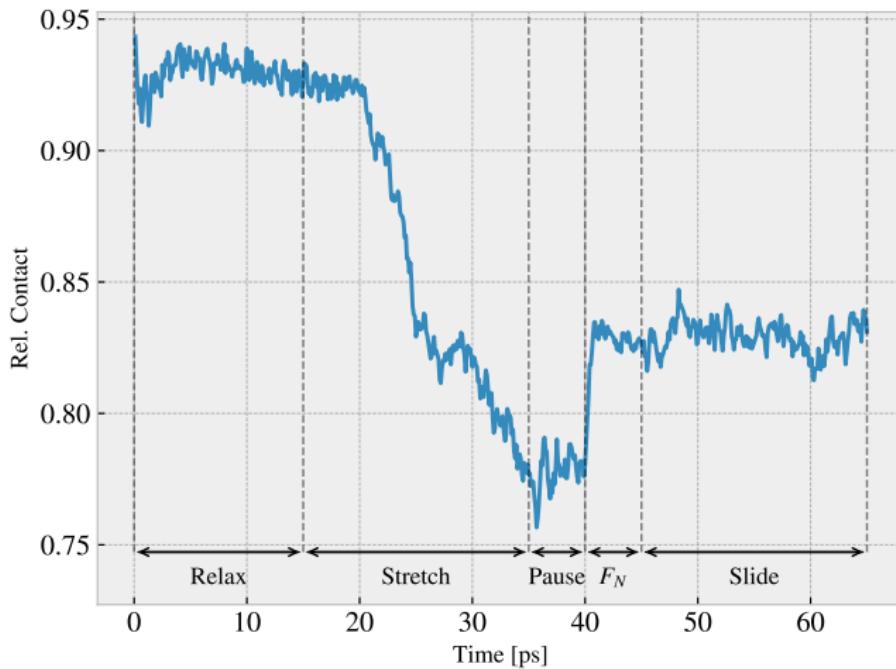


Figure: Contact area approximation: Number of C–Si bonds within a threshold distance of 110% the LJ interaction equilibrium distance.

Contact-strain profile

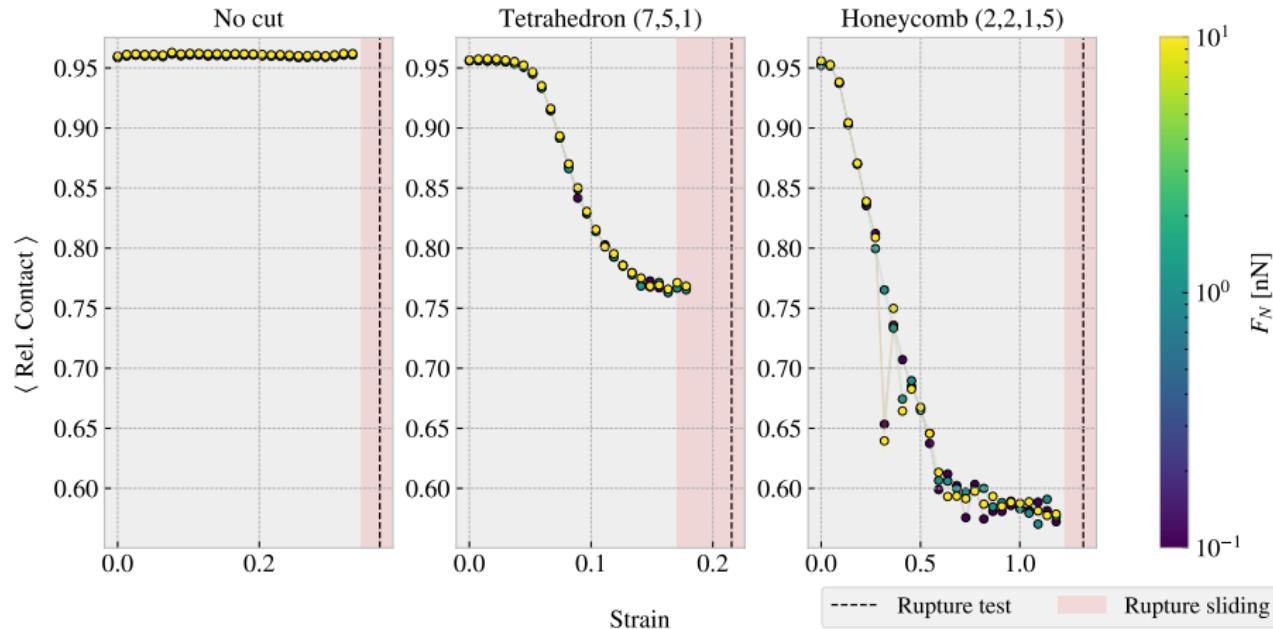


Figure: Contact-strain profile for $F_N = \{0.1, 1, 10\}$ nN.

Friction-strain profile

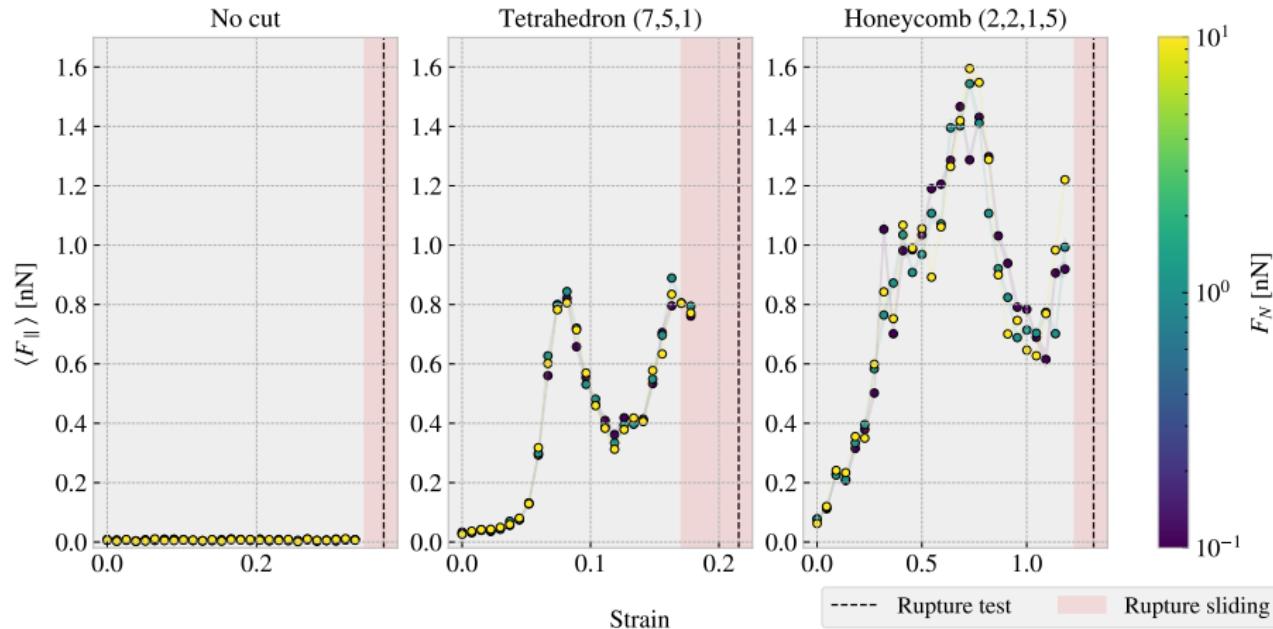


Figure: Friction-strain profile for $F_N = \{0.1, 1, 10\}$ nN.

Negative friction coefficient

Coupling of load to sheet tension

$$F_t = TF_N, \quad T = 6$$

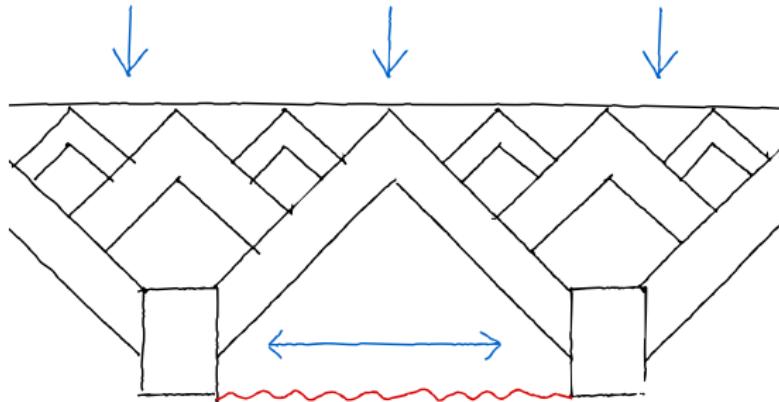


Figure: Working sketch for a nanomachine design translating applied load (from the top of the figure) to a straining of the graphene sheet (shown in red).

Negative friction coefficient

Tetrahedron results

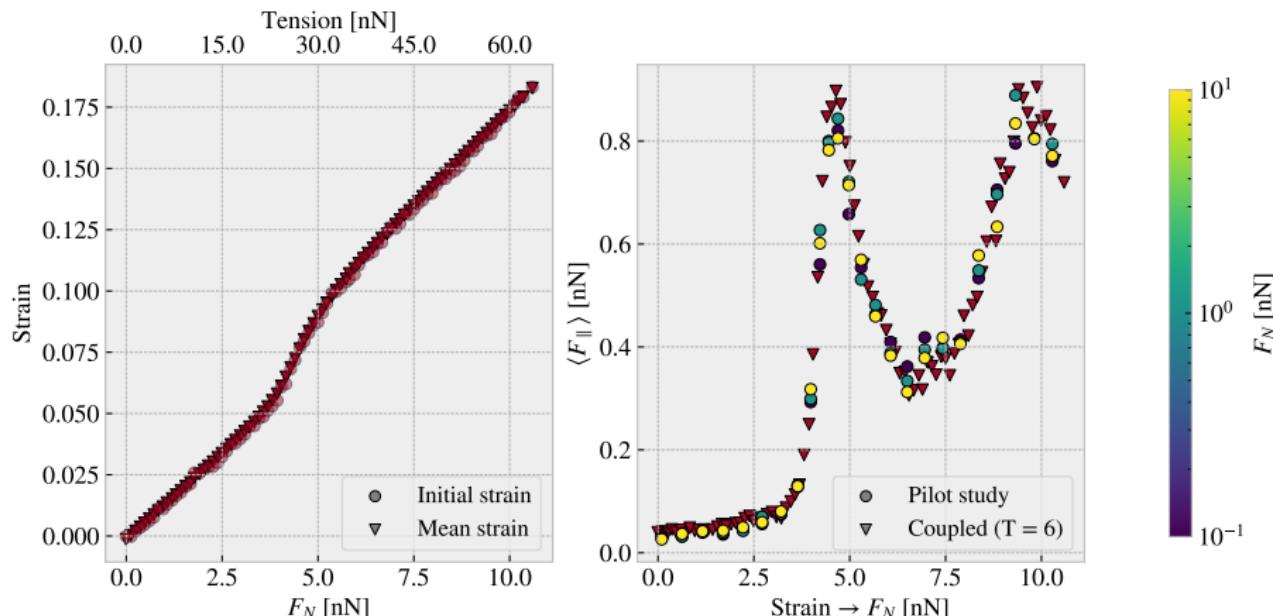


Figure: Tetrahedron (7, 5, 1)

Negative friction coefficient

Honeycomb results

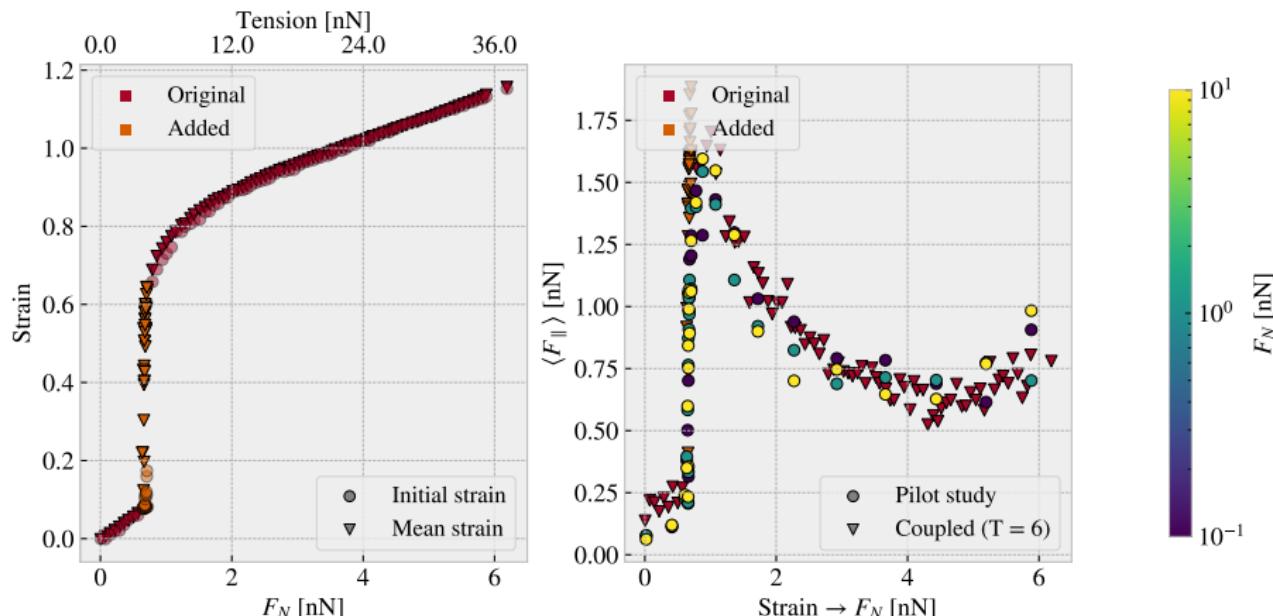


Figure: Honeycomb (2, 2, 1, 5)

Negative friction coefficient

Honeycomb deformations

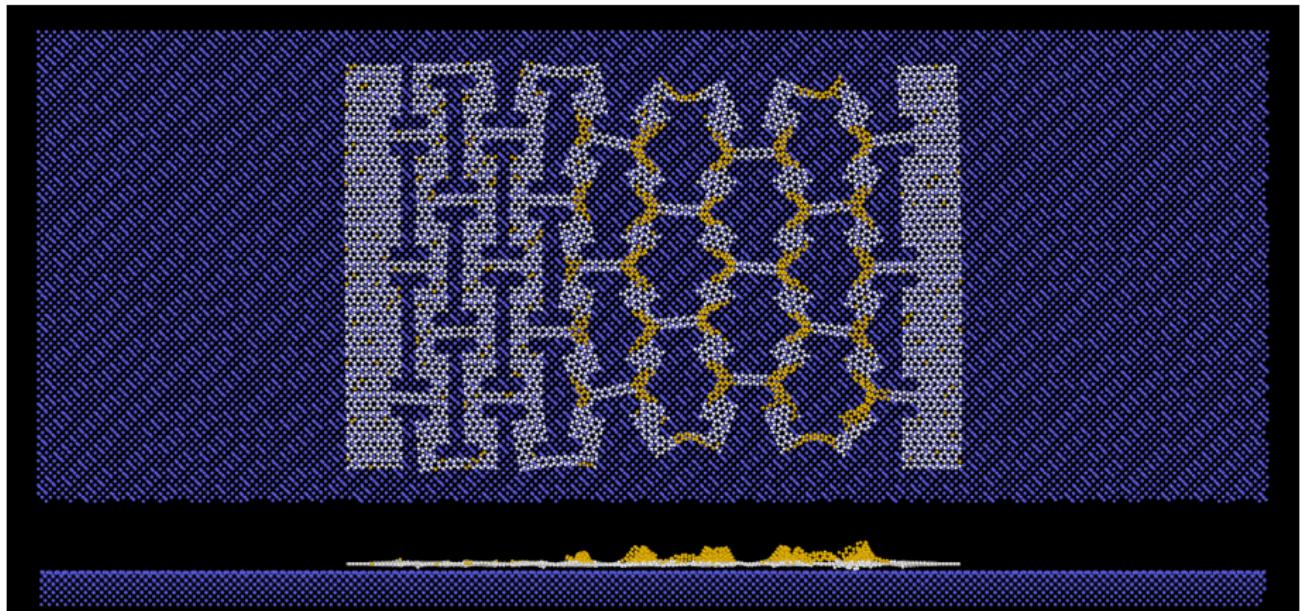


Figure: Honeycomb (2, 2, 1, 5) stretch.

Kirigami configuration search

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Dataset

Table: Summary of the generated data points in the dataset.

Type	Configurations	Data points	Ruptures
Pilot study	3	261	25 (9.58 %)
Tetrahedron	68	3015	391 (12.97 %)
Honeycomb	45	1983	80 (4.03 %)
Random walk	100	4401	622 (14.13 %)
Total	214 (216)	9660	1118 (11.57 %)

Dataset

Correlation

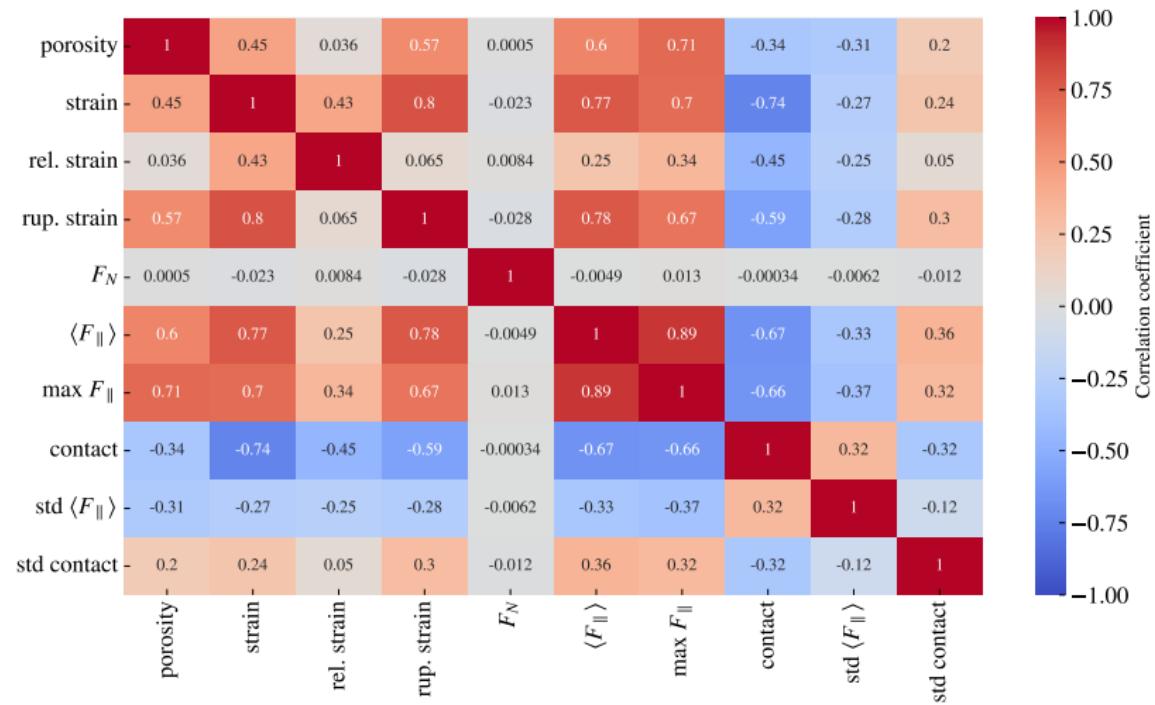


Figure: Pearson product-moment correlation coefficients.

Dataset

Properties of interest

Properties of interest

- (1) $\min F_{\text{fric}}$,
- (2) $\max F_{\text{fric}}$,
- (3) $\max \Delta F_{\text{fric}}$,
- (4) max drop.

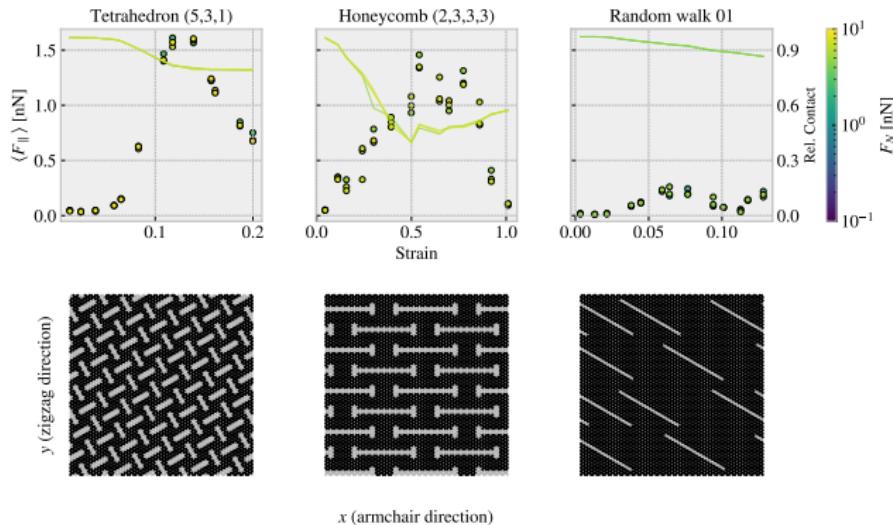


Figure: Max drop property. Best candidates in the dataset.

Machine learning

- Convolutional neural network
- Input: Kirigami configuration, strain and load
- Output: Mean friction, maximum friction, contact area, porosity, rupture, rupture strain

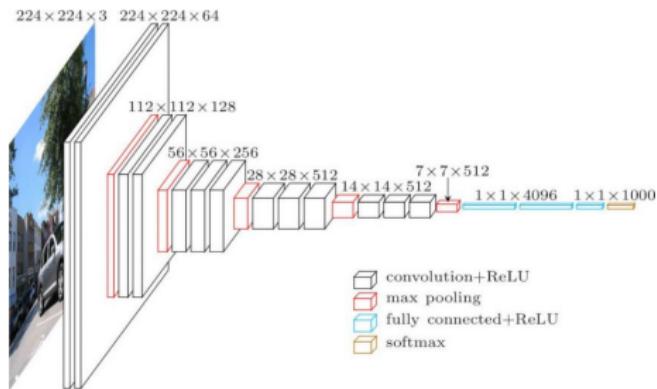


Figure: VGGNet-16 network convolutional network architecture. Reproduced from [10].

Machine learning

Model performance

Table: Evaluation of the final model performance.

	$R^2 [10^2]$		Abs. [10 ²]	Rel. [10 ²]	Acc. [10 ²]	
	Mean F_f	Max F_f	Contact	Porosity	Rup. Strain	Rupture
Validation	98.067	93.558	94.598	2.325	12.958	96.102
Tetrahedron	88.662	85.836	64.683	1.207	5.880	99.762
Honeycomb	96.627	89.696	97.171	1.040	1.483	99.111

Machine learning

Model performance

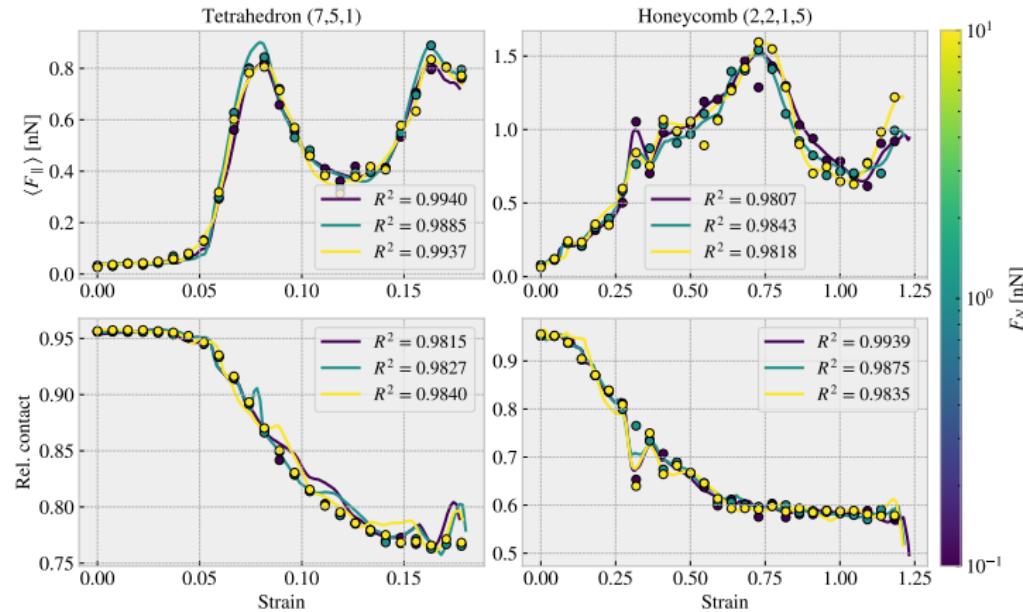


Figure: Visual evaluation of the final model predictions on the Tetrahedron (7,5,1) and Honeycomb (2,2,1,5) used in the pilot study.

Accelerated search

Extended dataset

- Tetrahedron: 1.35×10^5 configurations
- Honeycomb: 2.025×10^6 configurations
- Random walk: 10^4 configurations

Search Pred.	Tetrahedron	Honeycomb	Random walk
$\min F_{\text{fric}}$	-0.062	-0.109	-0.061
$\max F_{\text{fric}}$	1.089	2.917	0.660
$\max \Delta F_{\text{fric}}$	1.062	2.081	0.629
max drop	0.277	1.250	0.269

Original	Tetrahedron	Honeycomb	Random walk
$\min F_{\text{fric}}$	0.0067	0.0177	0.0024
$\max F_{\text{fric}}$	1.5875	2.8903	0.5758
$\max \Delta F_{\text{fric}}$	1.5529	2.0234	0.5448
max drop	0.8841	1.2785	0.1818

Accelerated search

Random walk candidates

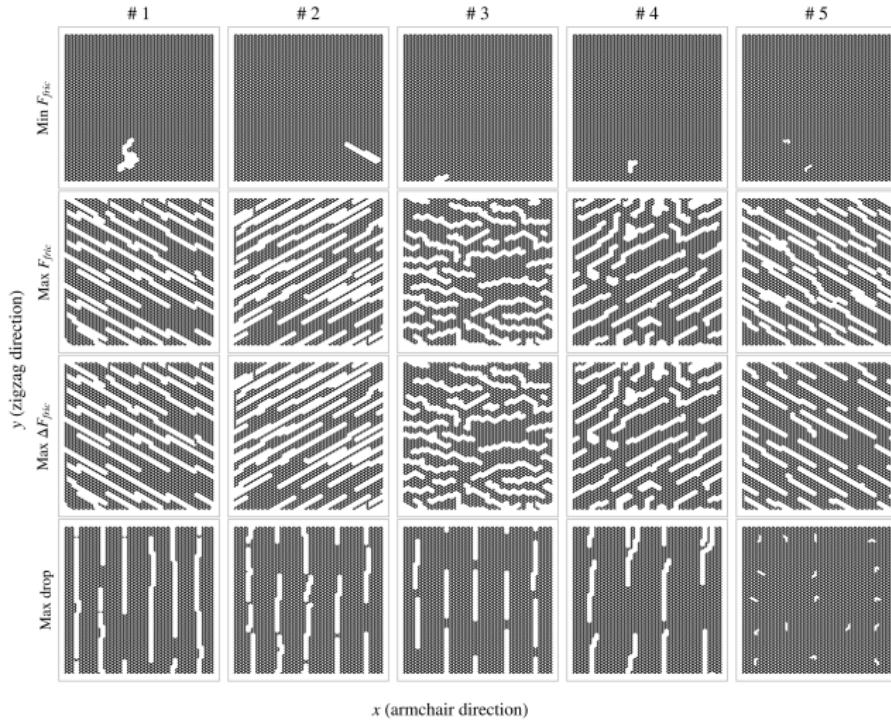
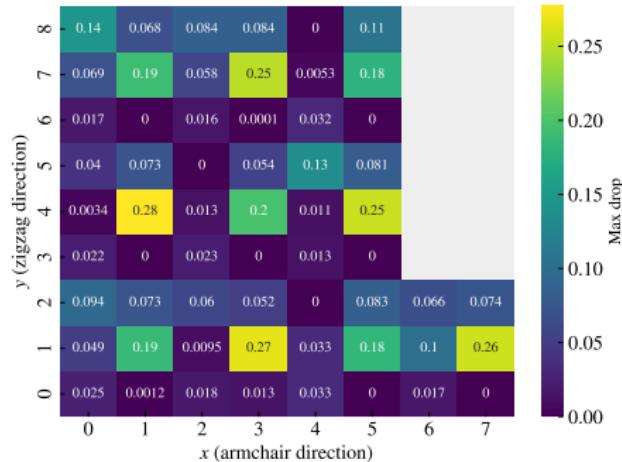


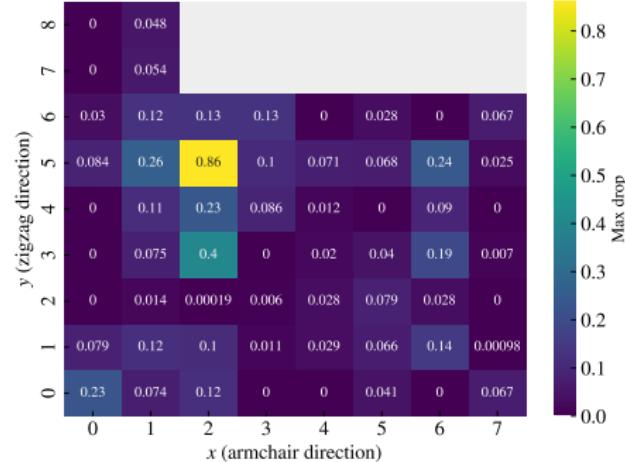
Figure: Top 5 candidates for the accelerated search using the Random walk generator.

Accelerated search

Translational variance



(a) Tetrahedron $(1, 7, 1)$. Rel. Std = 1.13



(b) Tetrahedron $(5, 3, 1)$. Rel. Std = 1.61

Figure: Machine learning prediction of the max drop property for all unique reference positions.

Accelerated search

Genetic algorithm

Genetic algorithm based on Markov chain probability.

- ① Rank configurations by fitness score.
- ② Calculate target states from the best candidates.
- ③ Assign a mutation probability based on ranking and target states the ranking.
- ④ Mutate and repeat.

Max drop property optimization → poor convergence

Accelerated search

Grad-CAM

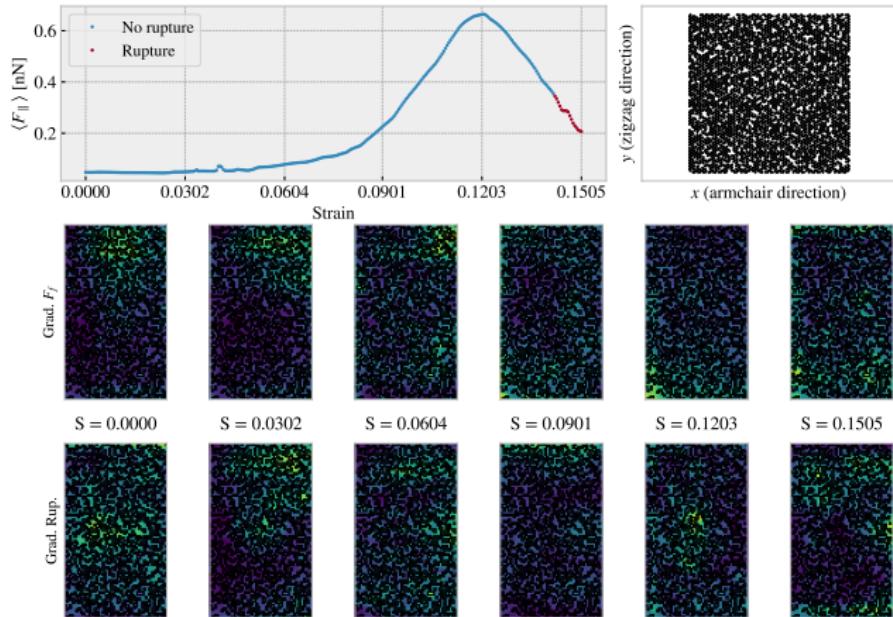


Figure: Genetic algorithm suggestion from a mixed porosity start. Top: Friction-strain curve and configuration. Bottom: Grad-CAM analysis

Accelerated search

Grad-CAM

Grad-CAM

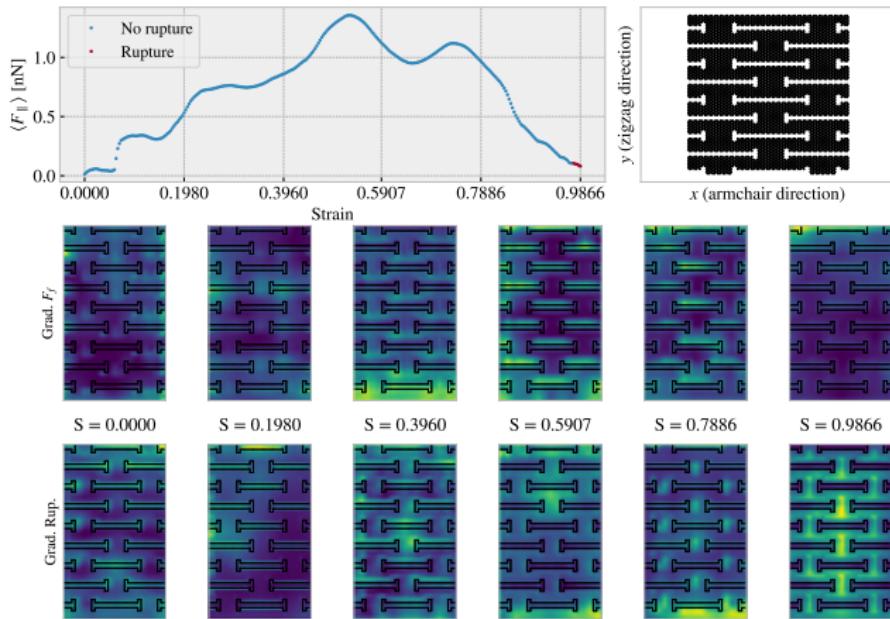


Figure: Honeycomb (3, 3, 5, 3), ref = (12, 0).

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Summary and outlook

Key findings

- Non-monotonous relationship between friction and strain
- The coupled system can be exploited to achieve a negative friction coefficient
- Machine learning is feasible but more data is needed

Further studies:

- Investigation of the underlying mechanism
 - Commensurability hypothesis can be investigated by varying scan angle
- Friction-strain relationship at different physical conditions: temperature, sliding speed, spring stiffness.
- Edge and thermostat effects.
- Improve dataset with active learning

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