# Multiscale Modeling and Machine Learning Design Optimization of Nanomaterials for Sustainability

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Thesis Committee:
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# Motivation

## A Simple Question: What are *Good* Materials?

- Hard
- High Strength
- High Thermal Conductivity
- Corrosion Resistant
- …

Credit: AP

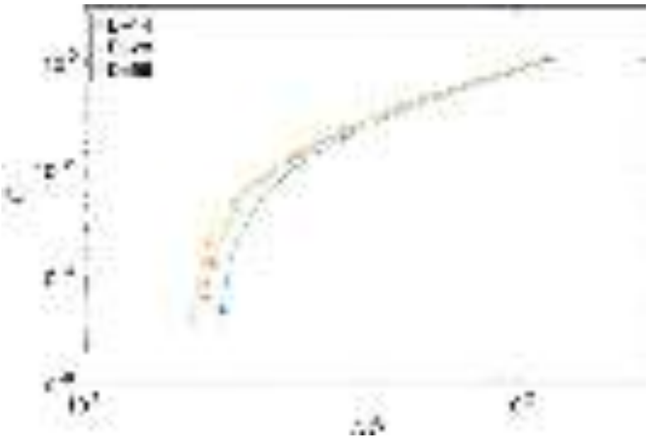
- Soft
- Breathability
- Elasticity
- …

Credit: *UConn Today*, 2018

**Structures**

**Computations**

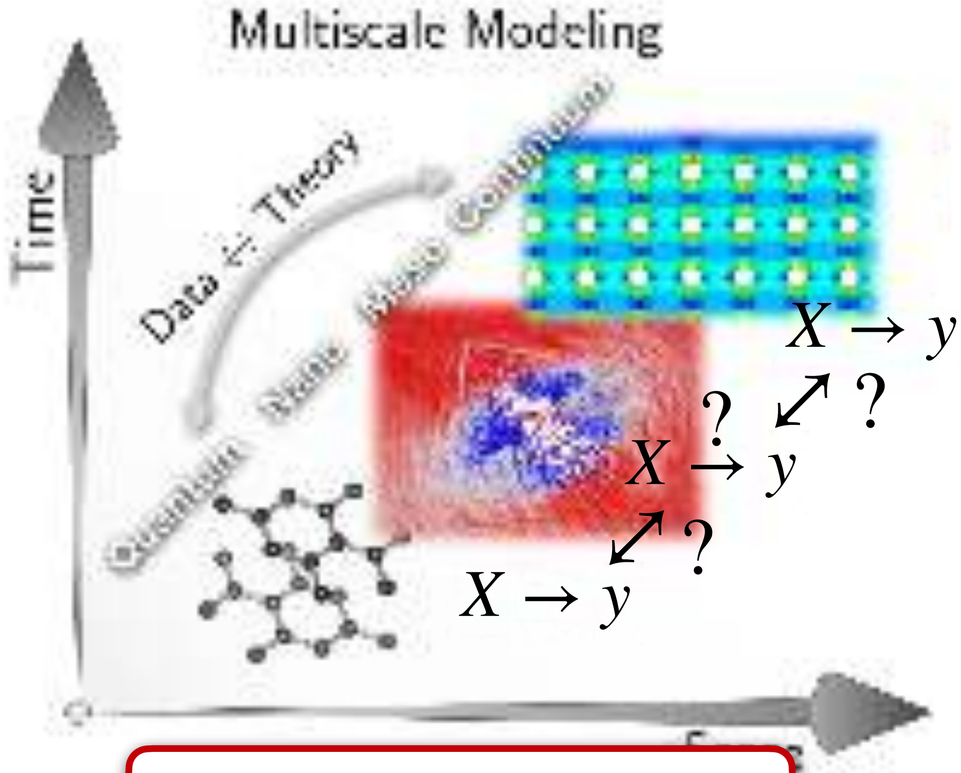
$$X \xrightarrow{\text{Model}} y$$

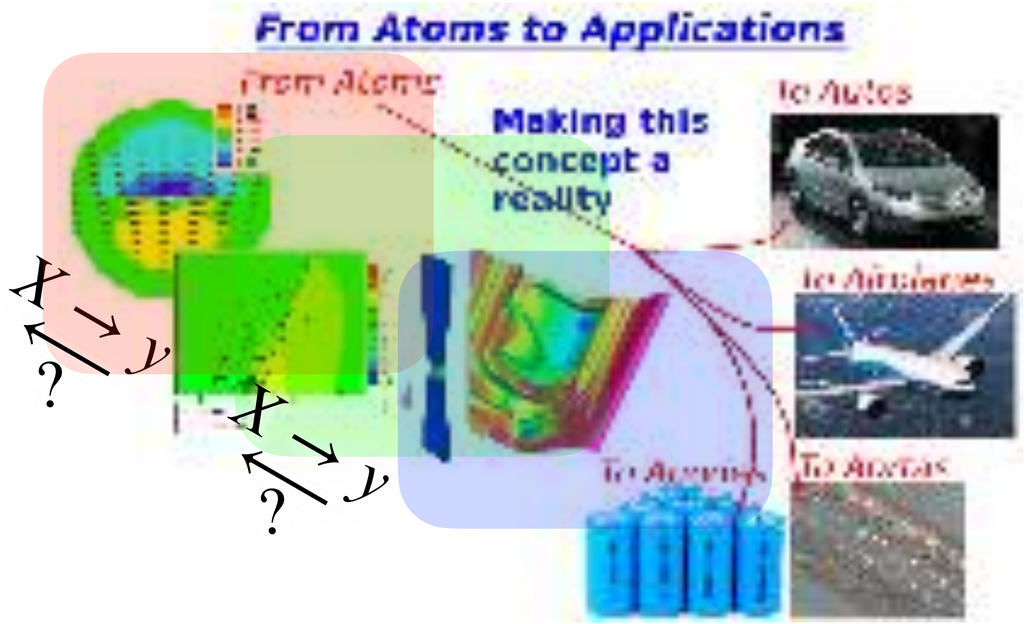


Credit: Bauer et al. *PRF*, 2019

# Motivation

## Follow-Up Questions: How to *Understand & Design* Good Materials?




Credit: *Laborat[illegible]ng*, EPFL

Understa[illegible]nulations

Designi[illegible]ications

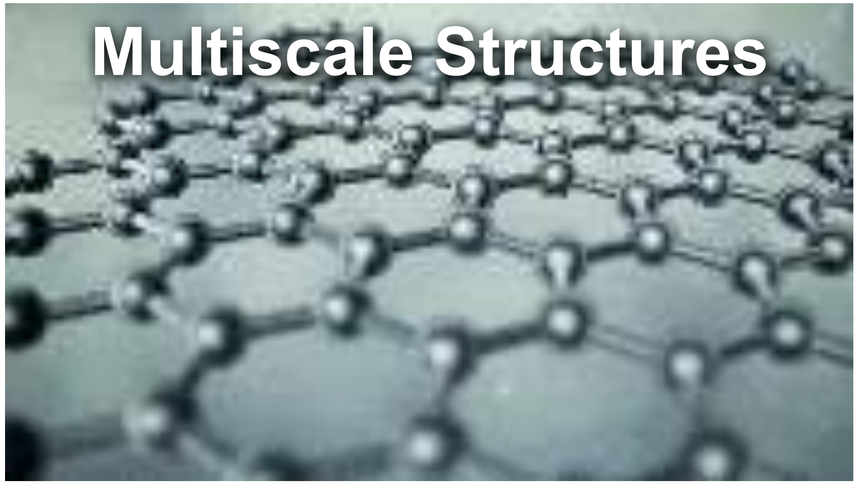
**PART I**

**PART II & III**

# Part I: Multiscale Mechanics of Graphene

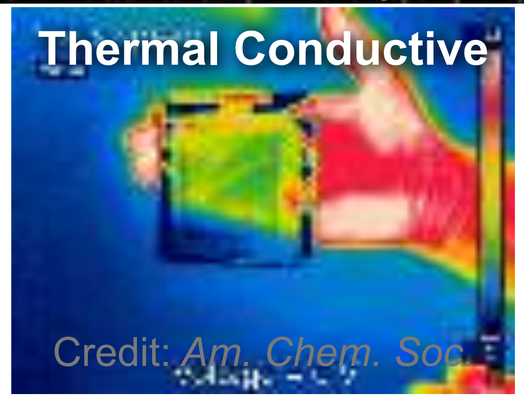
**Graphene**

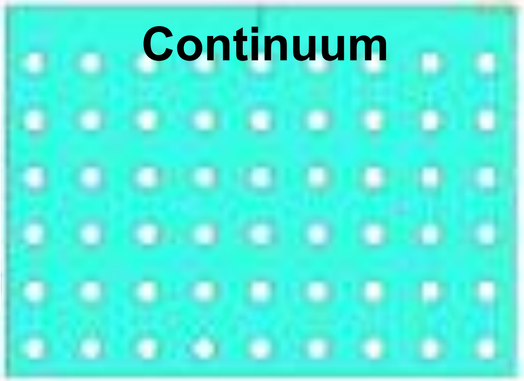
"*A wonder material*"

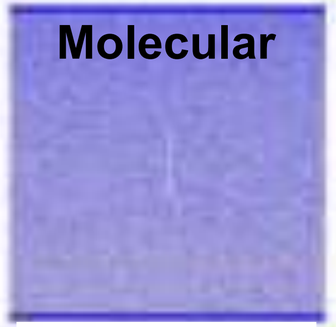



Credit: *SciTechDaily.com*






***Ab initio***

Credit: *gfycat.com*

# Part I: Multiscale Mechanics of Graphene

*Begin the research by **asking the question** from the **multi-scale** perspective*

**Question I:** What's the effect of empirical interatomic potentials in modeling & computations?

**Question II:** Can we benchmark empirical potentials with machine learning potentials? If yes, what's the differences?

**Question III:** Can we verify molecular dynamics simulations from mechanics theory? If yes, how?

https://doi.org/10.1142/S1758825123500448

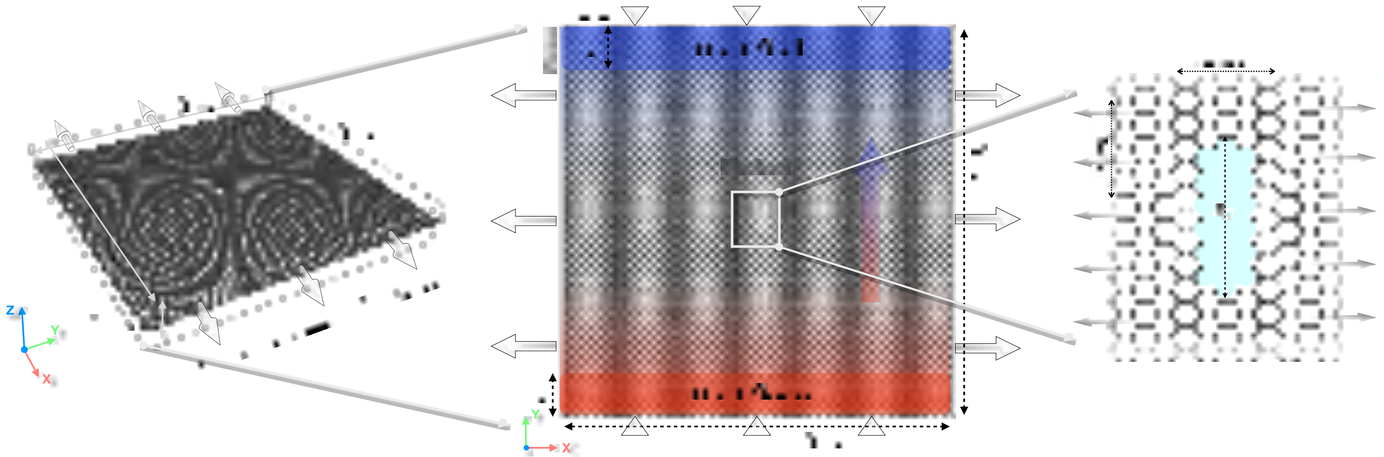
**Zhai** and Yeo, *International Journal of Applied Mechanics (In Press)*, 2023

**Zhai** and Yeo, *Molecular ML Conference (MIT, Cambridge, MA)*, 2022

# Part I: Multiscale Mechanics of Graphene

**Question I:** What's the effect of empirical molecular potentials in modeling?

- A three-dimensional simulation box with full PBC of applied strain rate ($10^9 s^{-1}$) loading implemented in LAMMPS. [1]
- A thermal gradient was applied in the Y direction, including the heat source (bottom) and heat sink (upper side).
- A defect (slit) is set in the center of the graphene layer of different lengths. [1]



[1] Zhao and Aluru, *J. Appl. Phys.*, 2010

**Zhai** and Yeo, *International Journal of Applied Mechanics (In Press)*, 2023
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# Part I: Multiscale Mechanics of Graphene

**Question I:** What's the effect of empirical molecular potentials in modeling?

## Empirical Potentials

- Optimized Tersoff potential

$$b_{ij}^{\text{Tersoff}} = f^{\text{Tersoff}}\left(b_{ij}^{\text{Tersoff}} + b_{ji}^{\text{Tersoff}}\right)$$

- REBO

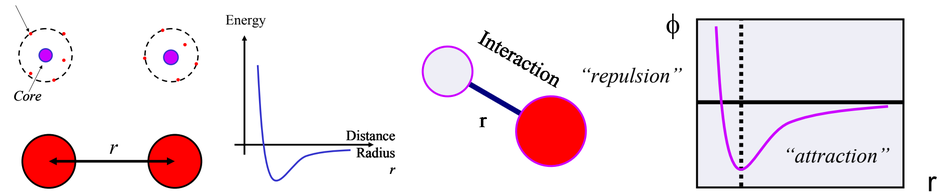
$$b_{ij}^{\text{REBO}} = f^{\text{REBO}}\left(b_{ij}^{\text{REBO}} + b_{ji}^{\text{REBO}}\right)$$

- AIREBO

$$E^{\text{AIREBO}} = E^{\text{REBO}} + E^{\text{LJ}} + E^{\text{Torsion}}$$

- AIREBO-M

$$E^{\text{AIREBO-M}} = E^{\text{REBO}} + E^{\text{Morse}} + E^{\text{Torsion}}$$




**Credit**: Buehler, *MIT DSpace*, 2006

## Characteristics

- Weighting function for cutoff
- Non-bonded systems
- Physical intuition: attraction + repulsion
- Exponential relation to interatomic radii

**Zhai** and Yeo, *International Journal of Applied Mechanics (In Press)*, 2023

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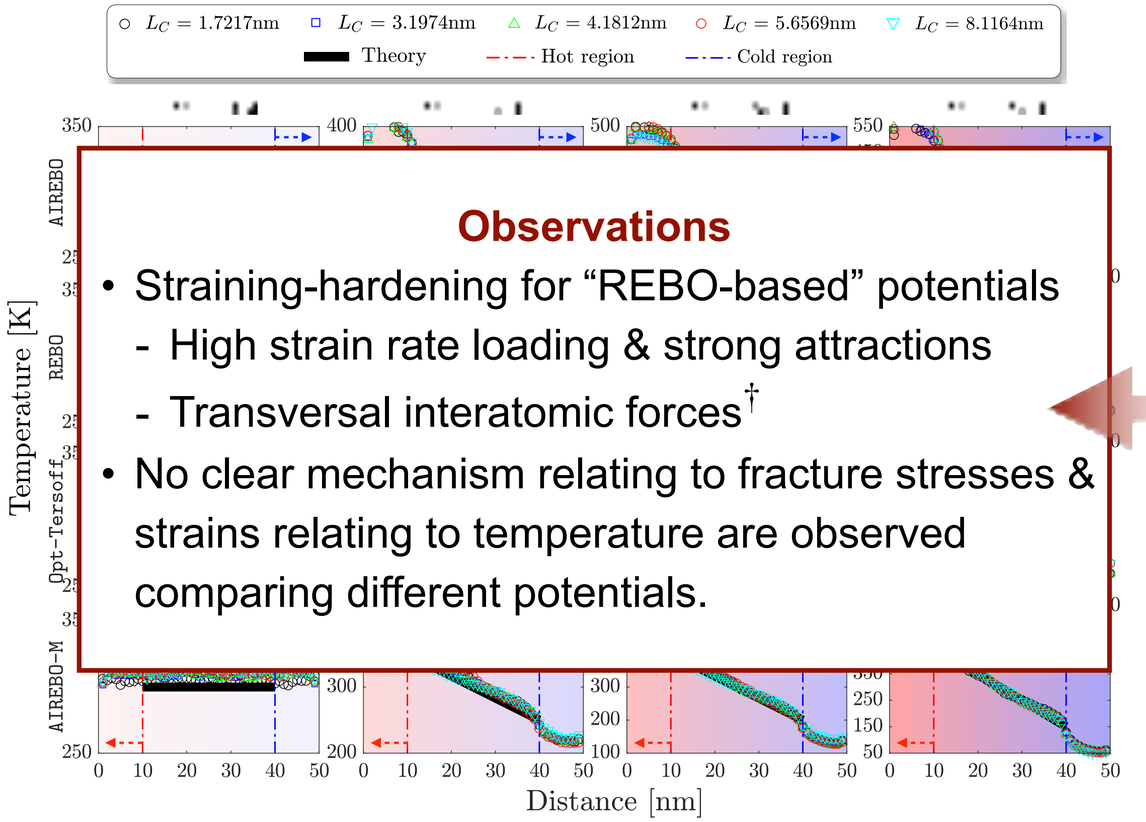
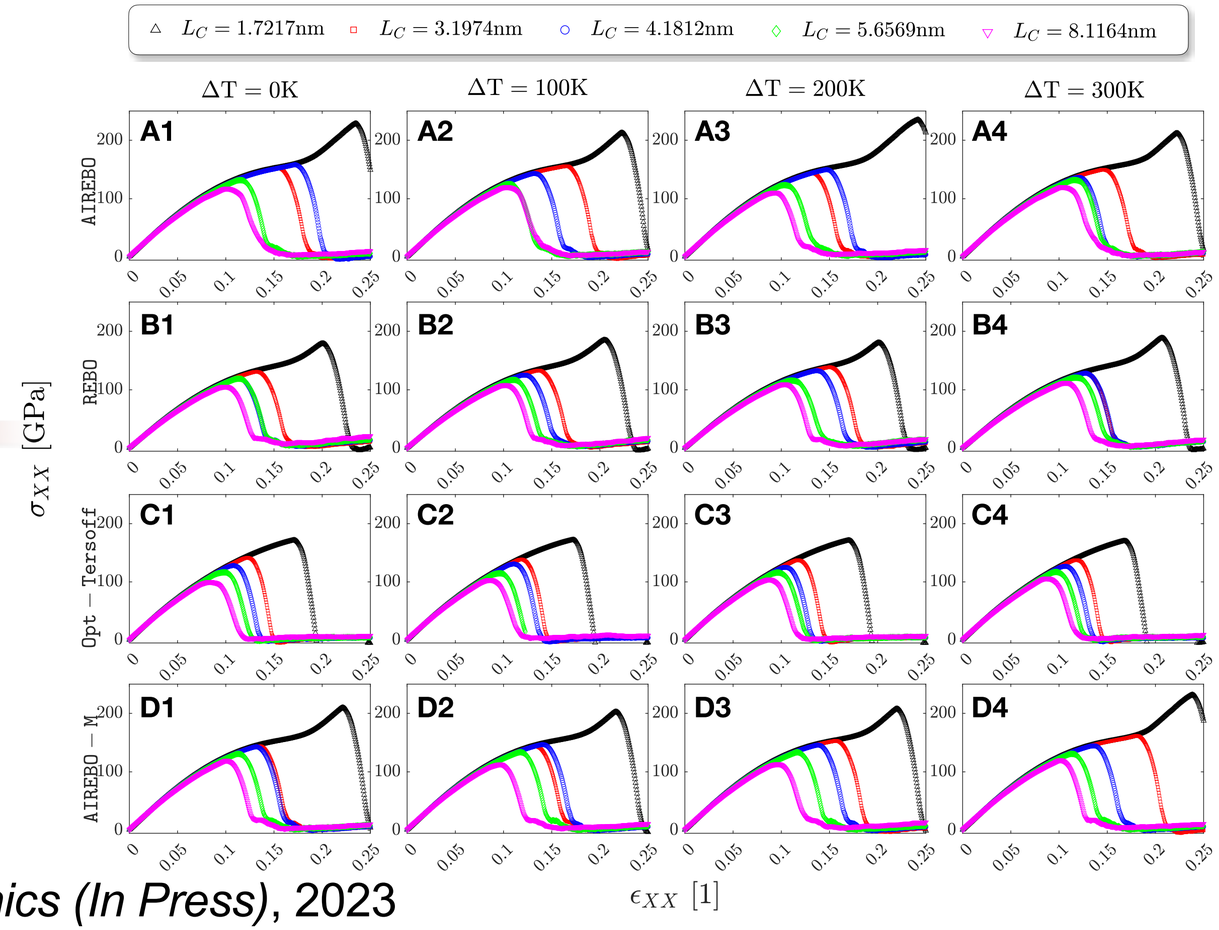
# Part I: Multiscale Mechanics of Graphene

**Question I:** What's the effect of empirical molecular potentials in modeling?




**Observations**

- Straining-hardening for "REBO-based" potentials
  - High strain rate loading & strong attractions
  - Transversal interatomic forces†
- No clear mechanism relating to fracture stresses & strains relating to temperature are observed comparing different potentials.

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# Part I: Multiscale Mechanics of Graphene

## **Question II:** Can we benchmark empirical & ML potentials? Differences?

J. Behler

M. Parrinello

PRL 98, 146401 (2007) PHYSICAL REVIEW LETTERS week ending 6 APRIL 2007

**Generalized Neural-Network Representation of High-Dimensional Potential-Energy Surfaces**

Jörg Behler and Michele Parrinello

*Department of Chemistry and Applied Biosciences, ETH Zurich, USI-Campus, Via Giuseppe Buffi 13, CH-6900 Lugano, Switzerland*

(Received 27 September 2006; published 2 April 2007)

The accurate description of chemical processes often requires the use of computationally demanding methods like density-functional theory (DFT), making long simulations of large systems unfeasible. In this Letter we introduce a new kind of neural-network representation of DFT potential-energy surfaces, which provides the energy and forces as a function of all atomic positions in systems of arbitrary size and is several orders of magnitude faster than DFT. The high accuracy of the method is demonstrated for bulk silicon and compared with empirical potentials and DFT. The method is general and can be applied to all types of periodic and nonperiodic systems.

DOI: 10.1103/PhysRevLett.98.146401 PACS numbers: 71.15.Pd, 61.50.Ah, 82.20.Kh

### Machine-Learned Potentials

- Theoretical Formulation

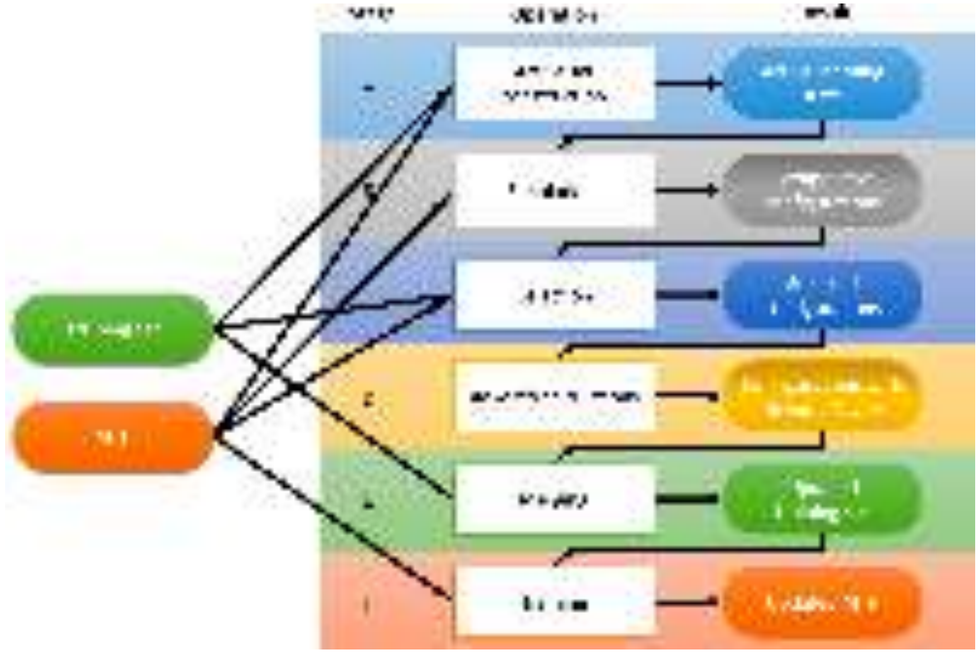
$$G_i^R = \sum_{j \neq i}^{all} \mathcal{F}_R(r_{ij}) f_c(r_{ij}),$$

$$G_i^A = \sum_{j,k \neq i}^{all} \mathcal{F}_A(r_{ij}, r_{ik}, r_{jk}) f_c(r_{ij}) f_c(r_{ik}) f_c(r_{jk})$$

→ [illegible]

### Benchmarked ML Potentials

- Machine-Learning Interatomic Potential [2]



**Zhai** and Yeo, *International Journal of Applied Mechanics (In Press)*, 2023

**Zhai** and Yeo, *Molecular ML Conference (MIT, Cambridge, MA)*, 2022

[2] Novikov et al., *Mach. Learn.: Sci. Tech.*, 2020

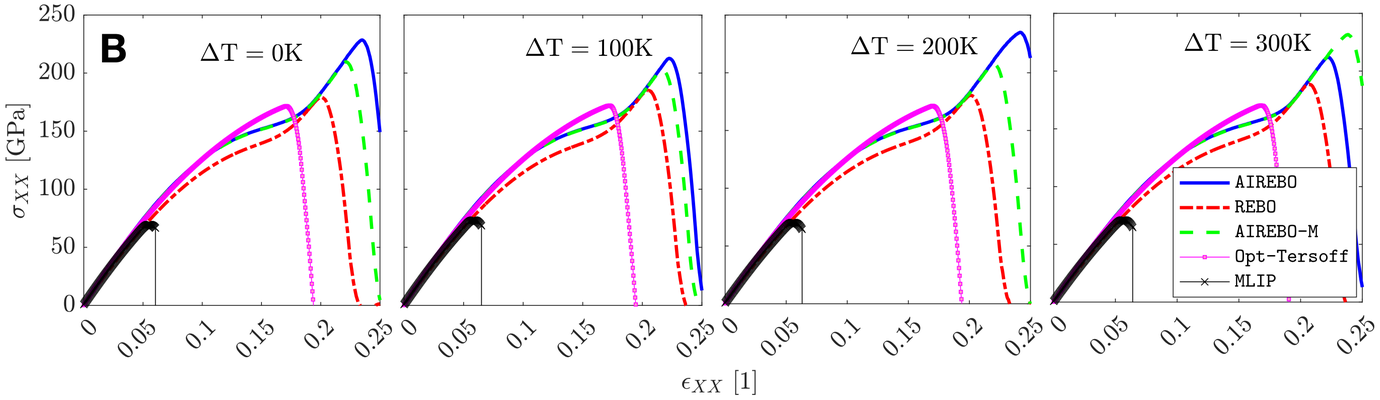
[3] Wen and Tadmor, *npj Comp. Mat.*, 2020

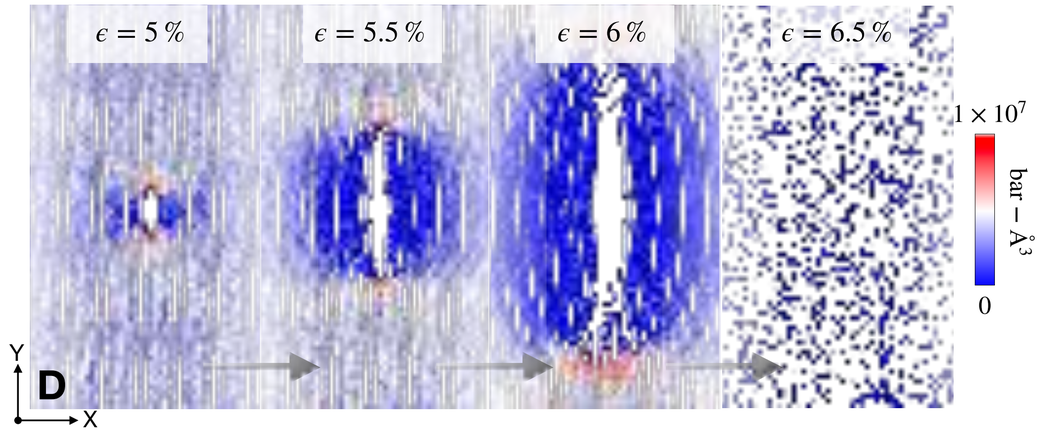
# Part I: Multiscale Mechanics of Graphene

**Question II:** Can we benchmark empirical & ML potentials? Differences?

**Observations**

- MLIP severely underestimate the fracture stress compared w/ empirical potentials.
  - Long-range interactions not captured in the *ab initio* training data.
- MLIP is incapable of simulating post-fracture behavior.
- MLIP does not capture the temperature effect in stress-strain responses.




**Zhai** and Yeo, *International Journal of Applied Mechanics (In Press)*, 2023

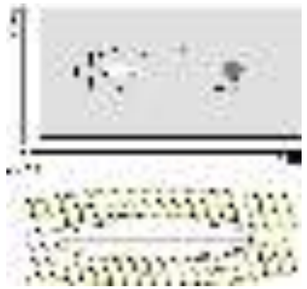
**Zhai** and Yeo, *Molecular ML Conference (MIT, Cambridge, MA)*, 2022

# Part I: Multiscale Mechanics of Graphene

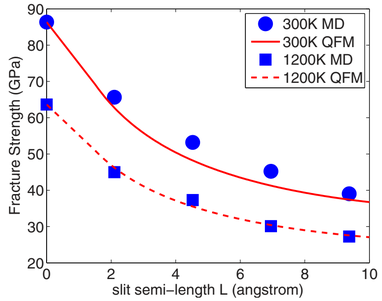
**Question III:** Can we verify MD simulations from *Mechanics*? If yes, how?

N. Pugno R. Ruoff

Credit: Università di Trento and Wikipedia



Ippolito et al., 2006




Zhao & Aluru, 2010

## Quantized Fracture Mechanics

- The fracture stress derived for QFM:

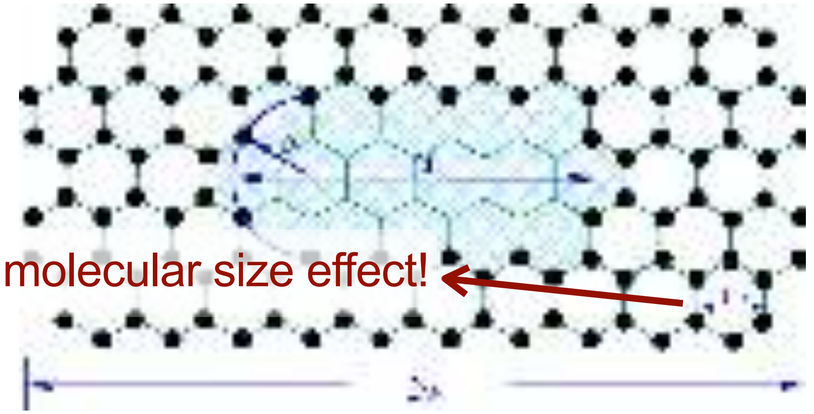
$$\Sigma = \frac{L_C}{2} \qquad \sqrt{\pi L_0}$$

→ "Local Effect"

## Linear Elastic Fracture Mechanics

- Relation between fracture stress & intensity factor:

$$\sigma_f = \frac{K_{IC}}{\sqrt{\pi a}}$$



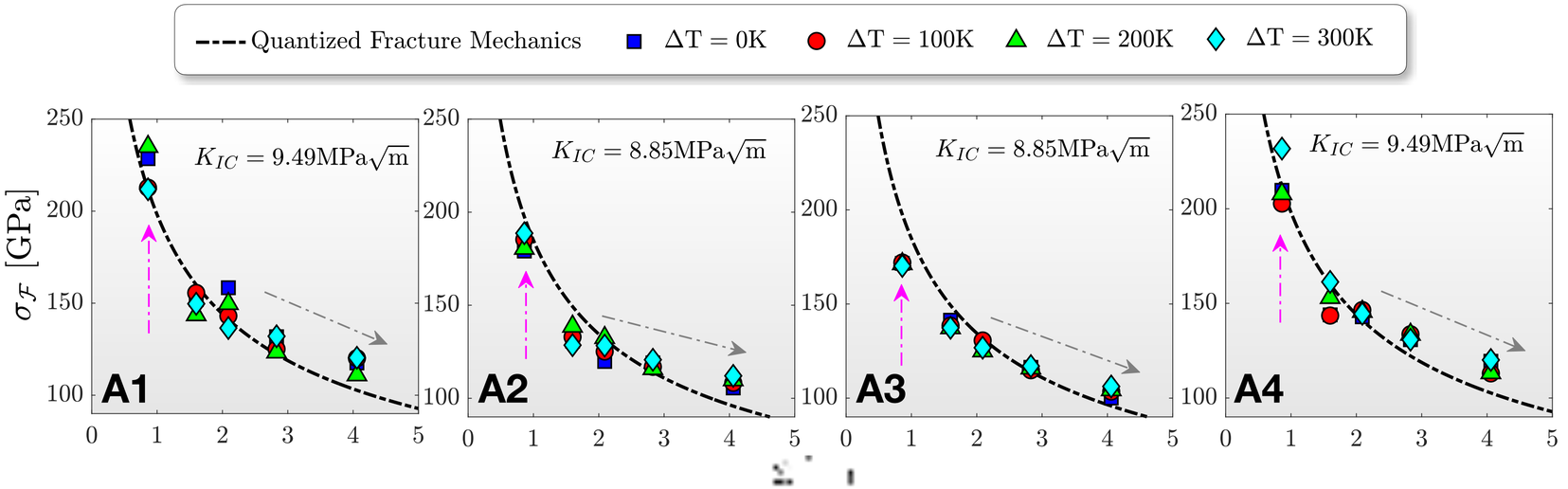
$L_0$ includes molecular size effect!

**Zhai** and Yeo, *International Journal of Applied Mechanics (In Press)*, 2023

**Zhai** and Yeo, *Molecular ML Conference (MIT, Cambridge, MA)*, 2022

[4] Pugno & Ruoff, *Philo. Mag.*, 2012

**Question III:** Can we verify MD simulations from *Mechanics*? If yes, how?




**Observations → Verifications**

- Molecular dynamics simulations data fitted well to QFM and the fitted $K_{IC}$ matches experimental observations. [5]
- From both QFM & MD, one observes with smaller initial defect the fracture stress increases nonlinearly.

**Zhai** and Yeo, *International Journal of Applied Mechanics (In Press)*, 2023
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[5] Zhang *et al.*, *Nat. Comm.*, 2014

# Part I: Multiscale Mechanics of Graphene

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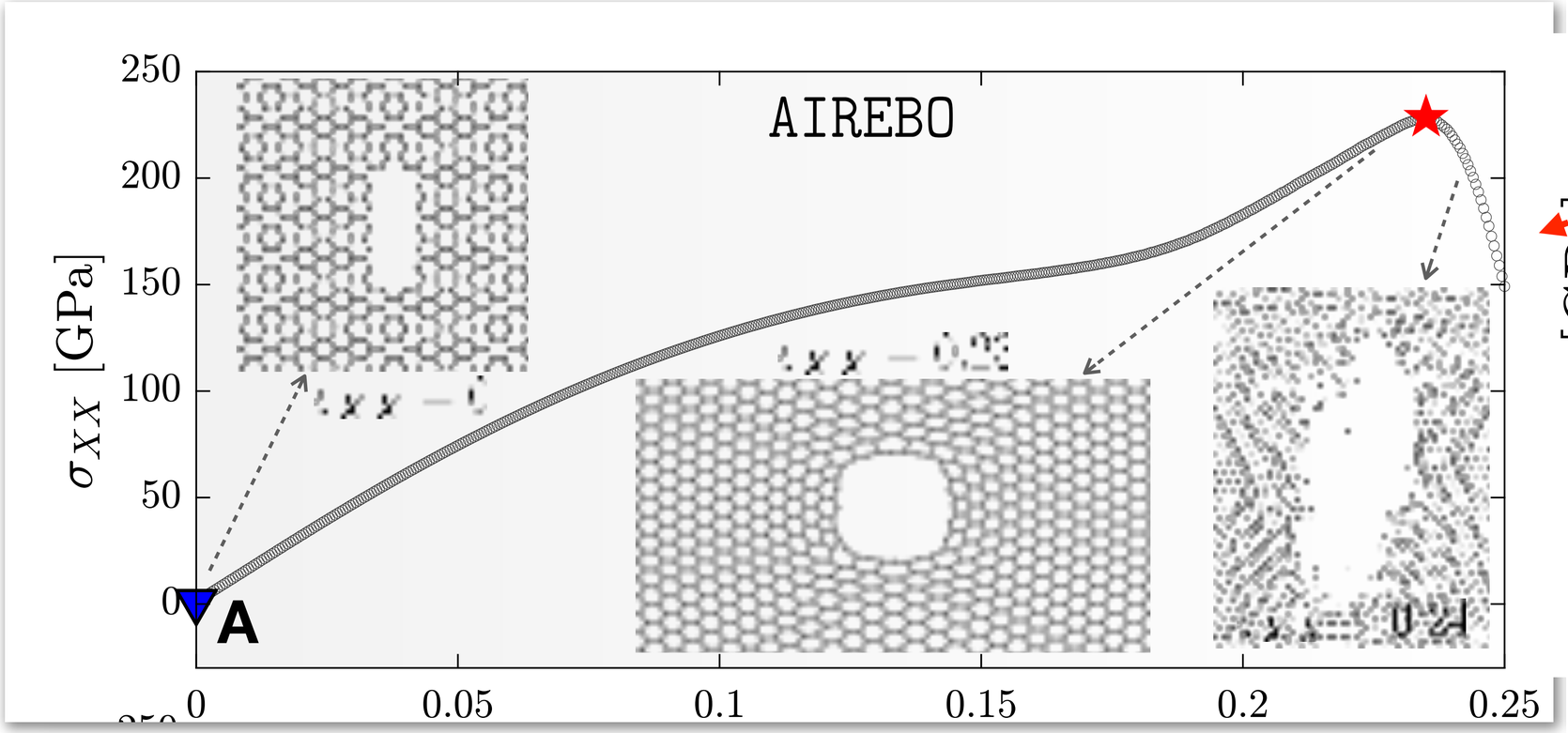


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**Zhai** and Yeo, *Molecular ML Conference (MIT, Cambridge, MA)*, 2022

**Chemo-mechanics**

- Strain-hardening for "REBO-based" potentials from Q. 1.
  - Larger crack vacuum area
  - "Brittle-like" fracture
- Nonlinear fracture stress increase predicted from QFM
  - †Transversal interatomic energy
  - Potential models characterization
  - Higher fracture strain

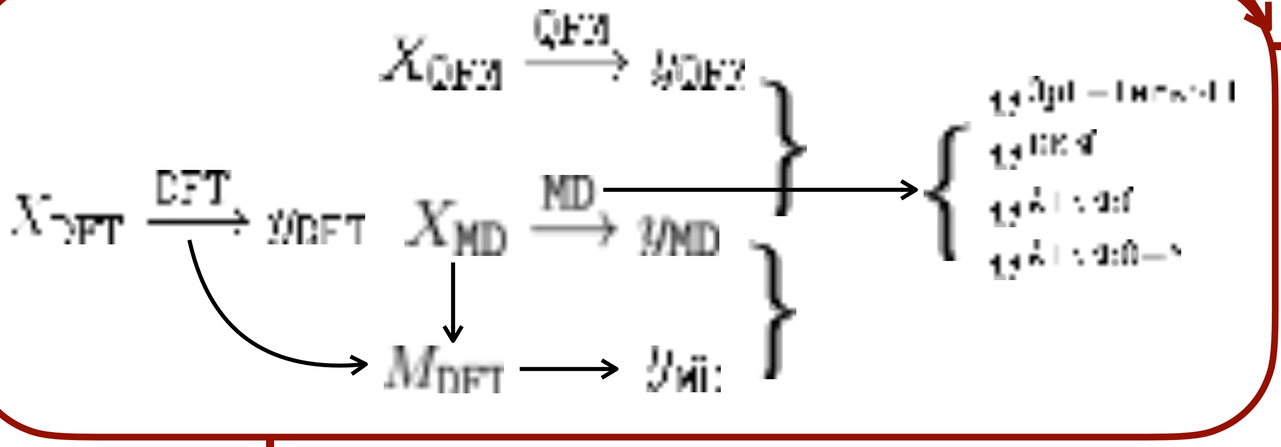
# Outline

## Forward Problem

$$X \xrightarrow{M} y$$

## Multiscale Modeling

$$X_\alpha \xrightarrow{M_\alpha} y_\alpha \Leftrightarrow X_\beta \xrightarrow{M_\beta} y_\beta$$



# Outline

## Forward Problem

$$X \xrightarrow{M} y$$

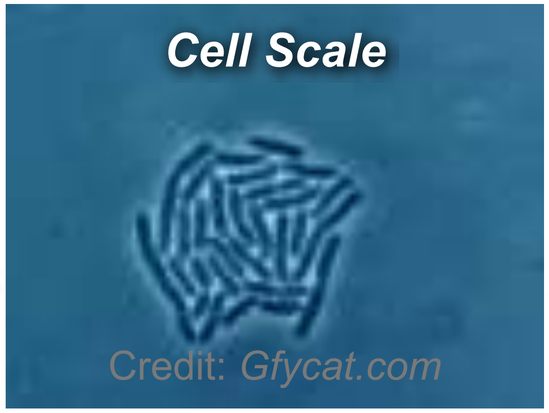
## Multiscale Modeling

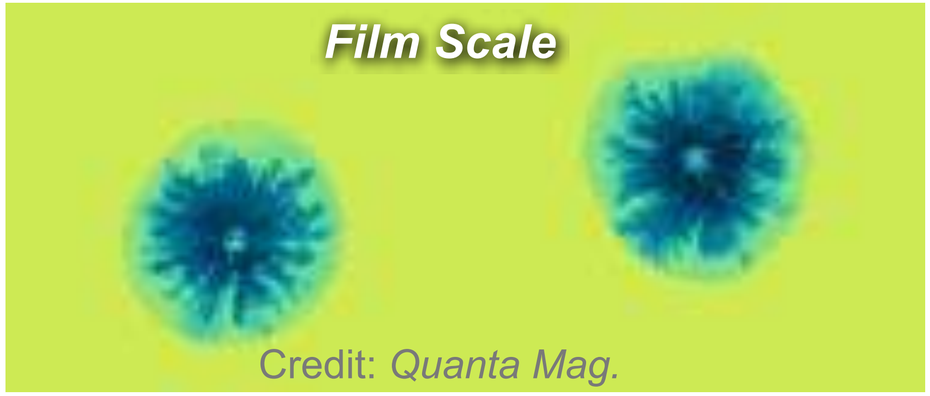
$$X_\alpha \xrightarrow{M_\alpha} y_\alpha \Leftrightarrow X_\beta \xrightarrow{M_\beta} y_\beta$$

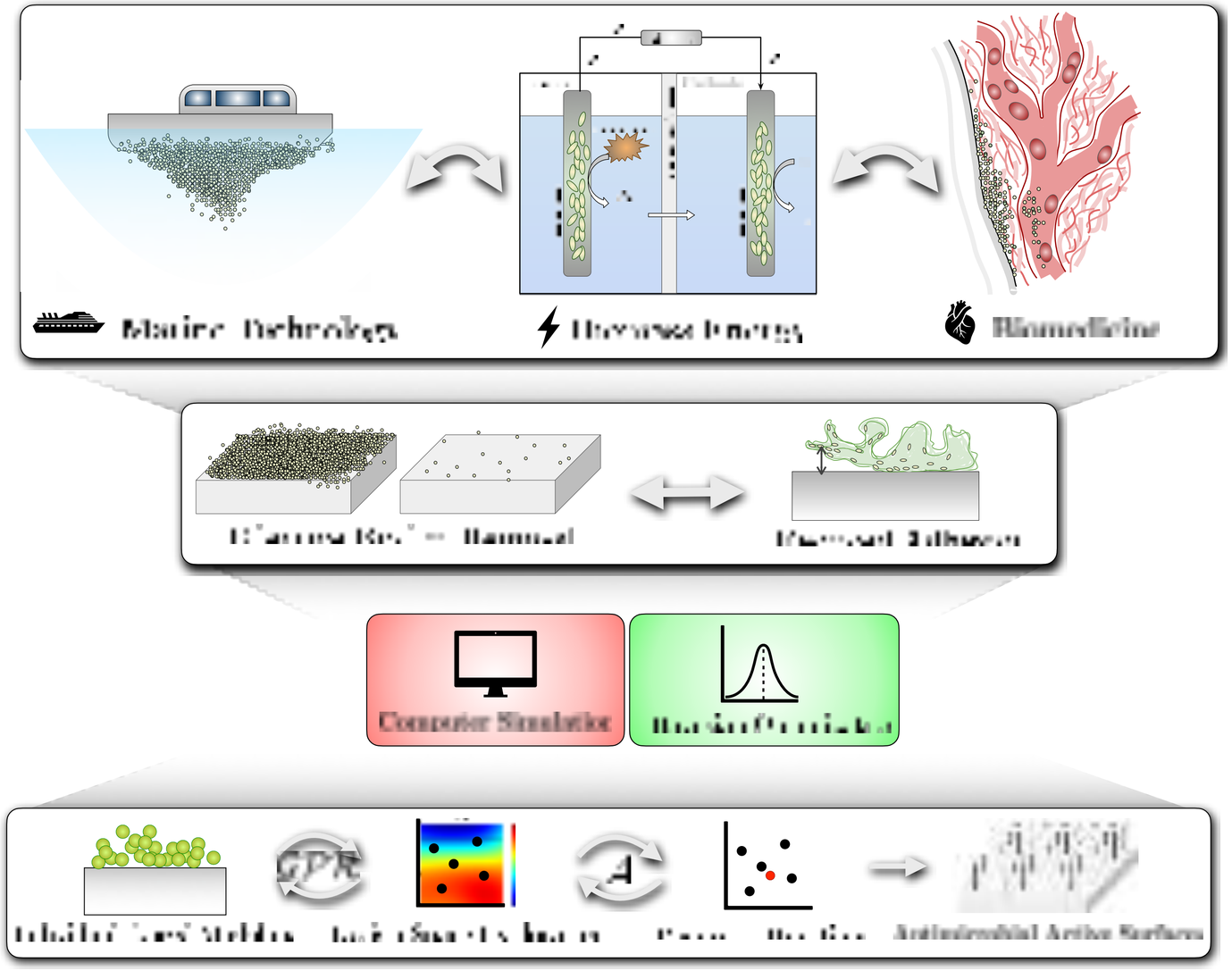
# Part II: Designing Antibiofilm Surfaces

**Biofilm**

*"A global crisis"*

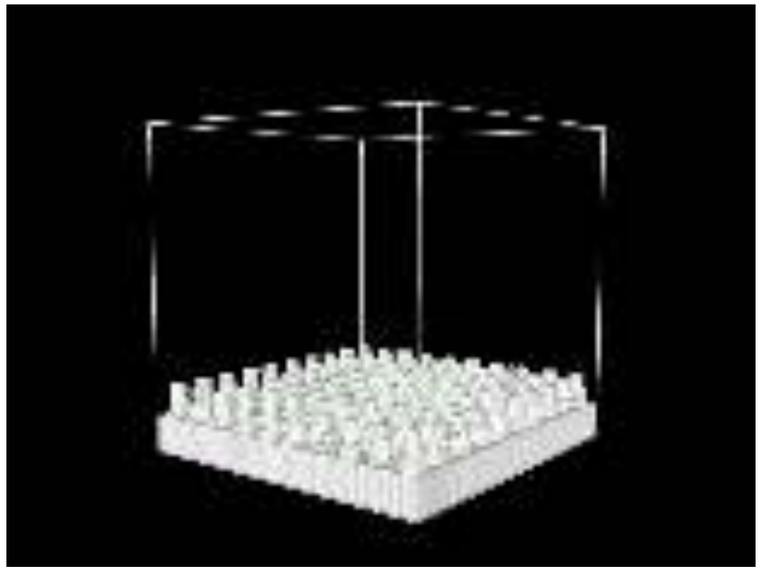


# Part II: Designing Antibiofilm Surfaces

*Begin the research by **asking the question** from the **scale & design** perspective*



**Question I:** How to remove biofilm → how to simulate biofilm formulation and removal process?

**Question II:** How to automate the design process digitally?

**Question III:** What's the biomechanics behind the optimization and designed antimicrobial surfaces?


https://doi.org/10.1021/acsbiomaterials.2c01079

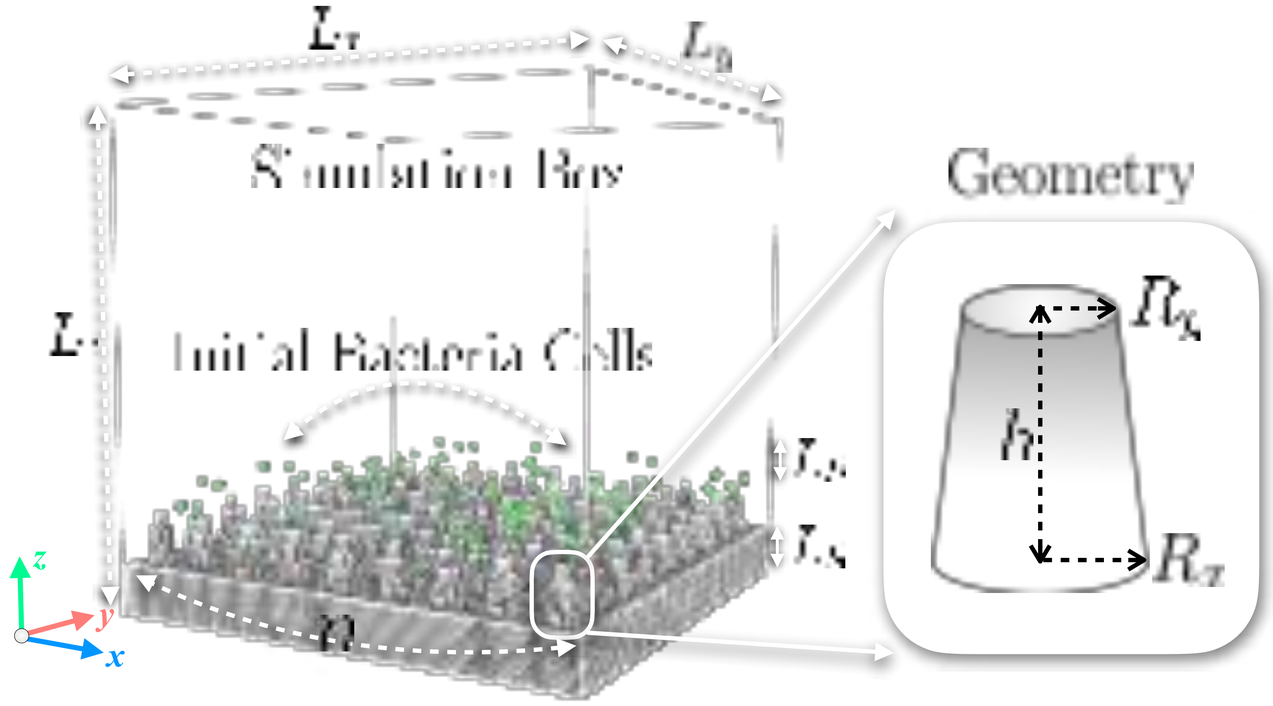
**Zhai** and Yeo, *ACS Biomaterials Science & Engineering*, 2023, 9, 1, 269–279

**Zhai**, *Sibley Graduate Research Symposium*, 2022

# Part II: Designing Antibiofilm Surfaces

**Question I:** How to simulate biofilm formulation and removal process?

- A cubic simulation box with sizes (LAMMPS) of $L_x = L_y = L_z = 4 \times 10^{-5}\text{m}$.
- Geometry is defined by four design variables $R_{bottom}$, $R_{top}$, $h$, and $n$.
- Initial bacteria cells are randomly generated in a defined area $L_{\mathcal{B}}$, above the substrate $L_{\mathcal{S}}$.
- Four physical scenarios are considered: pure growth, shear-off, vertical and lateral vibrations [6].




[6] Gu *et al.*, *Nat. Comm.*, 2020

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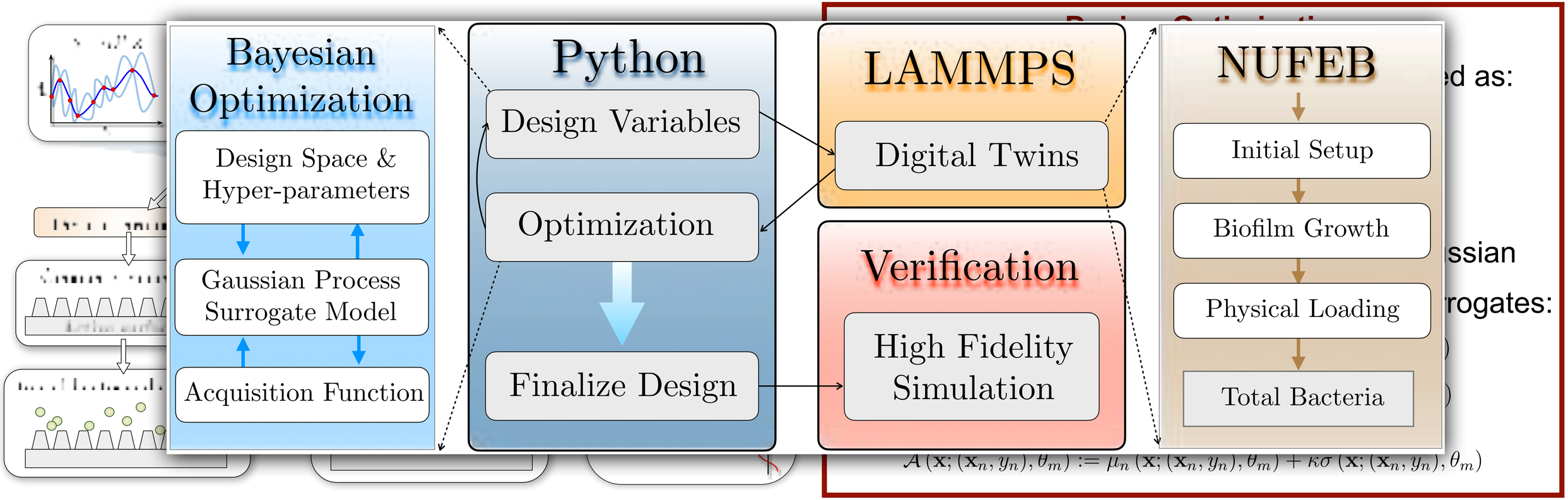
# Part II: Designing Antibiofilm Surfaces

**Question II:** How to **automate** the design process **digitally**?

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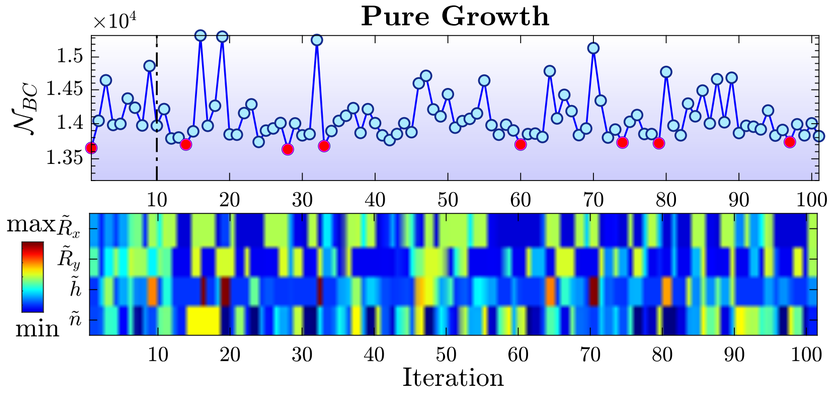


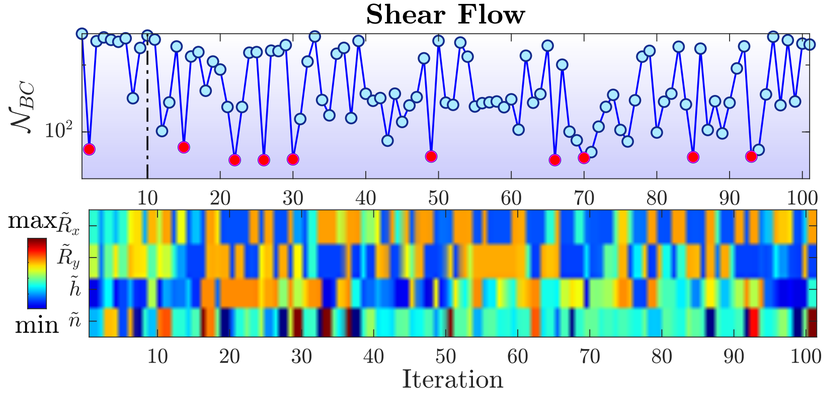


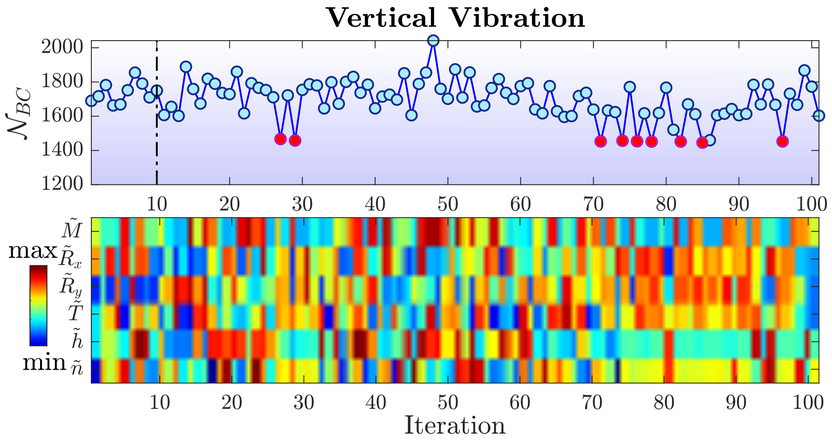


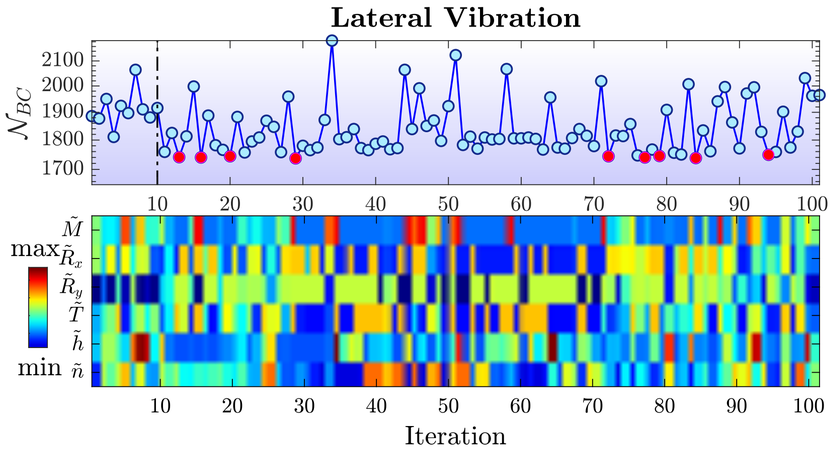


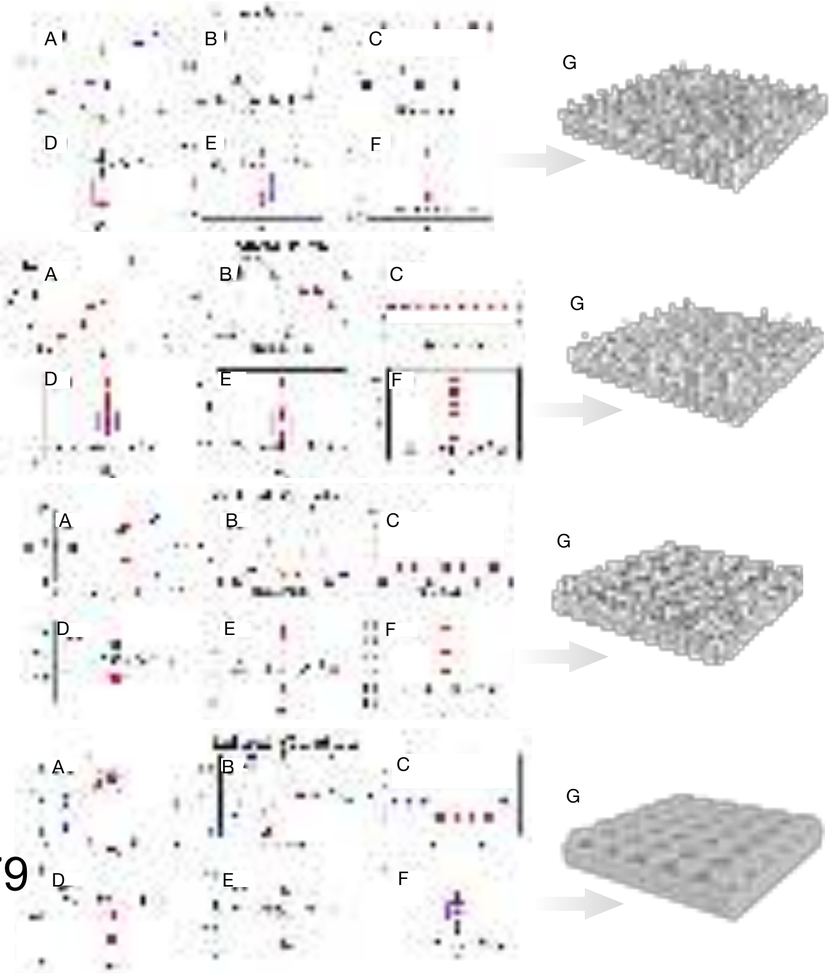


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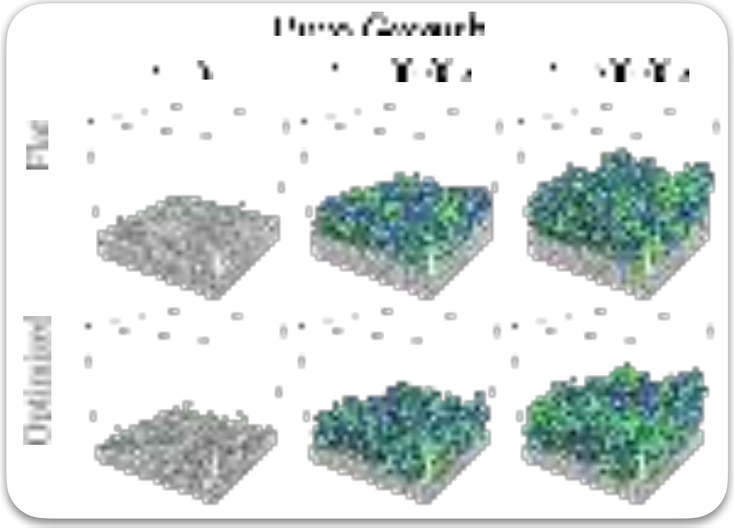
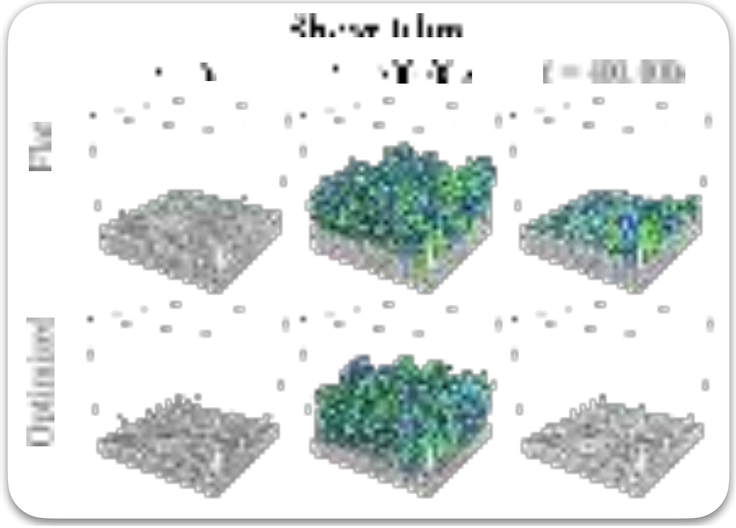
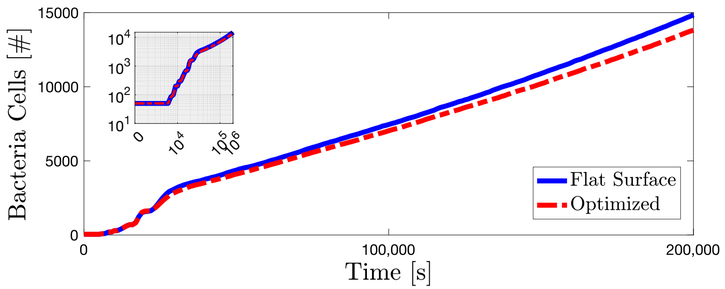
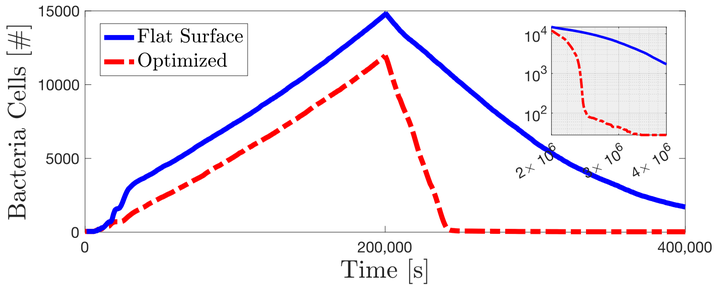
# Part II: Designing Antibiofilm Surfaces

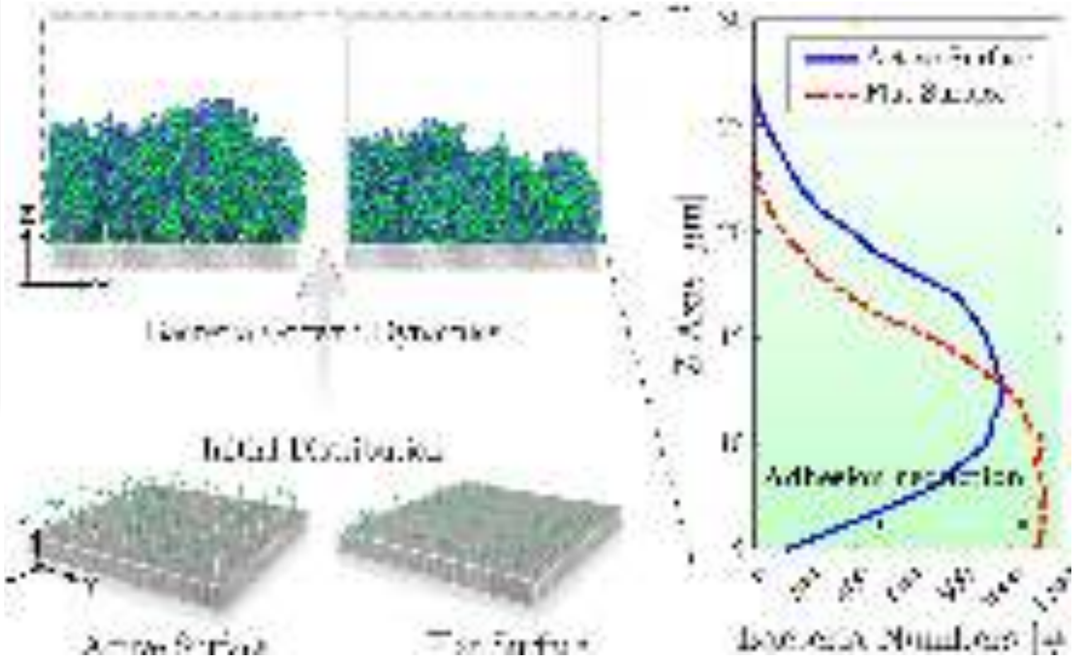
**Question III:** What's the biomechanics of the antimicrobial surfaces?







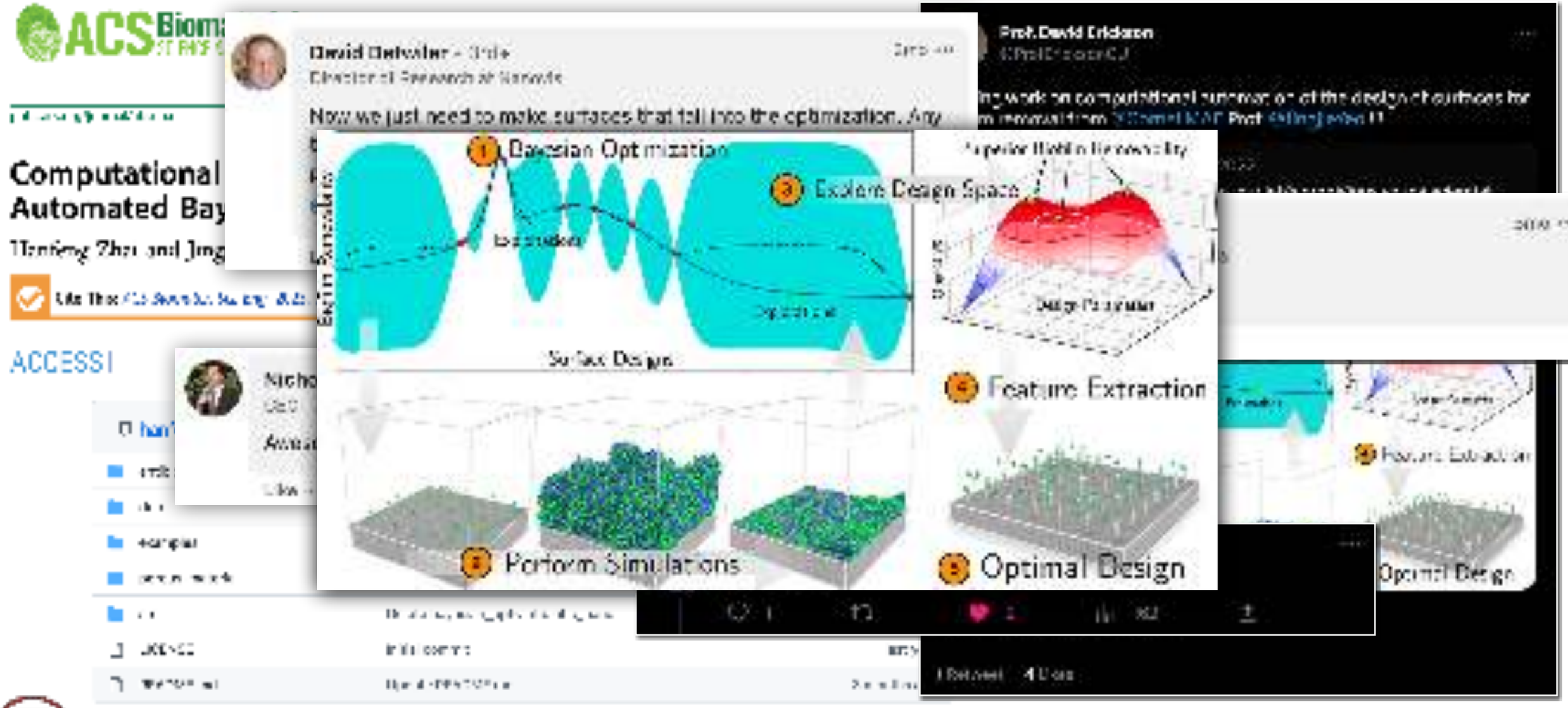



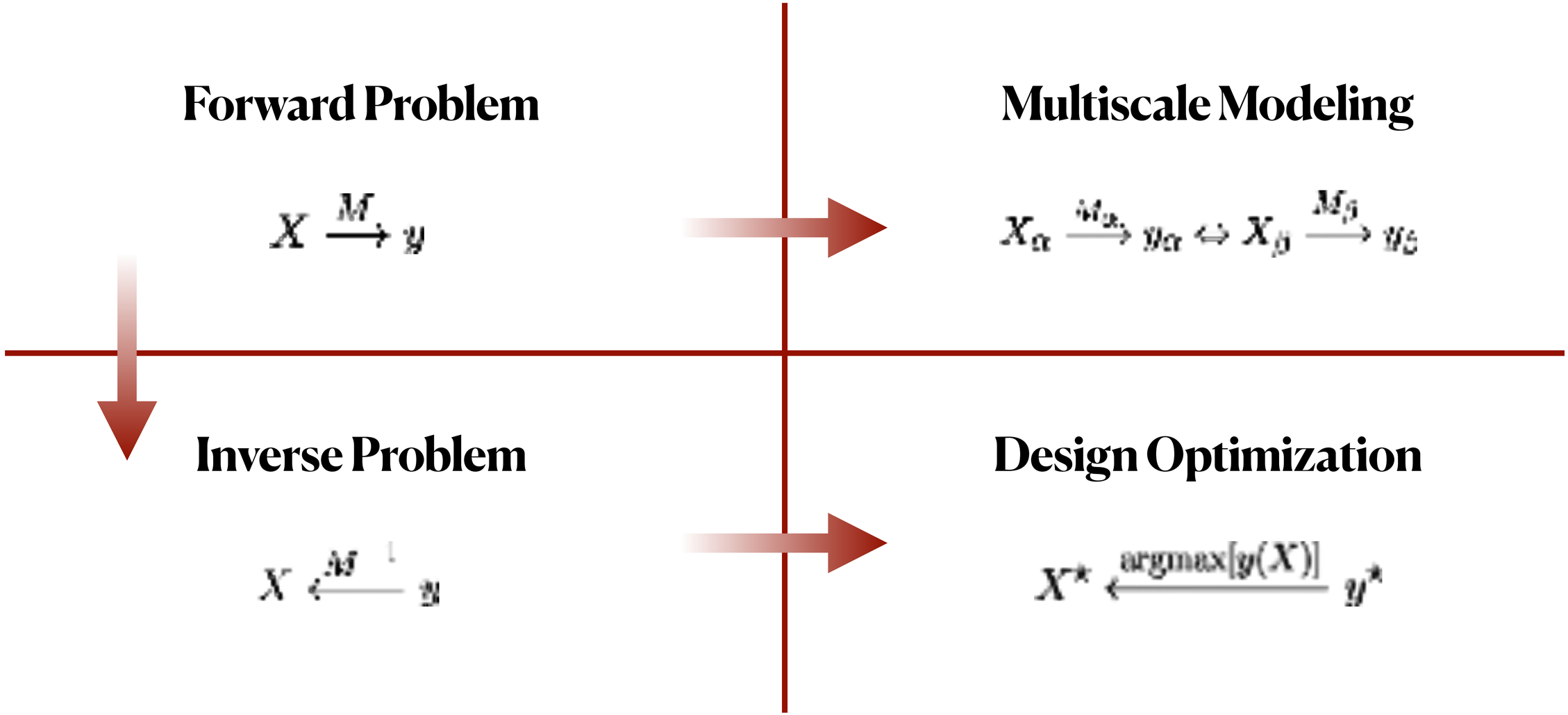
**Zhai** and Yeo, *ACS Biomaterials Science & Engineering*, 2023, 9, 1, 269–279

**Zhai**, *Sibley Graduate Research Symposium*, 2022

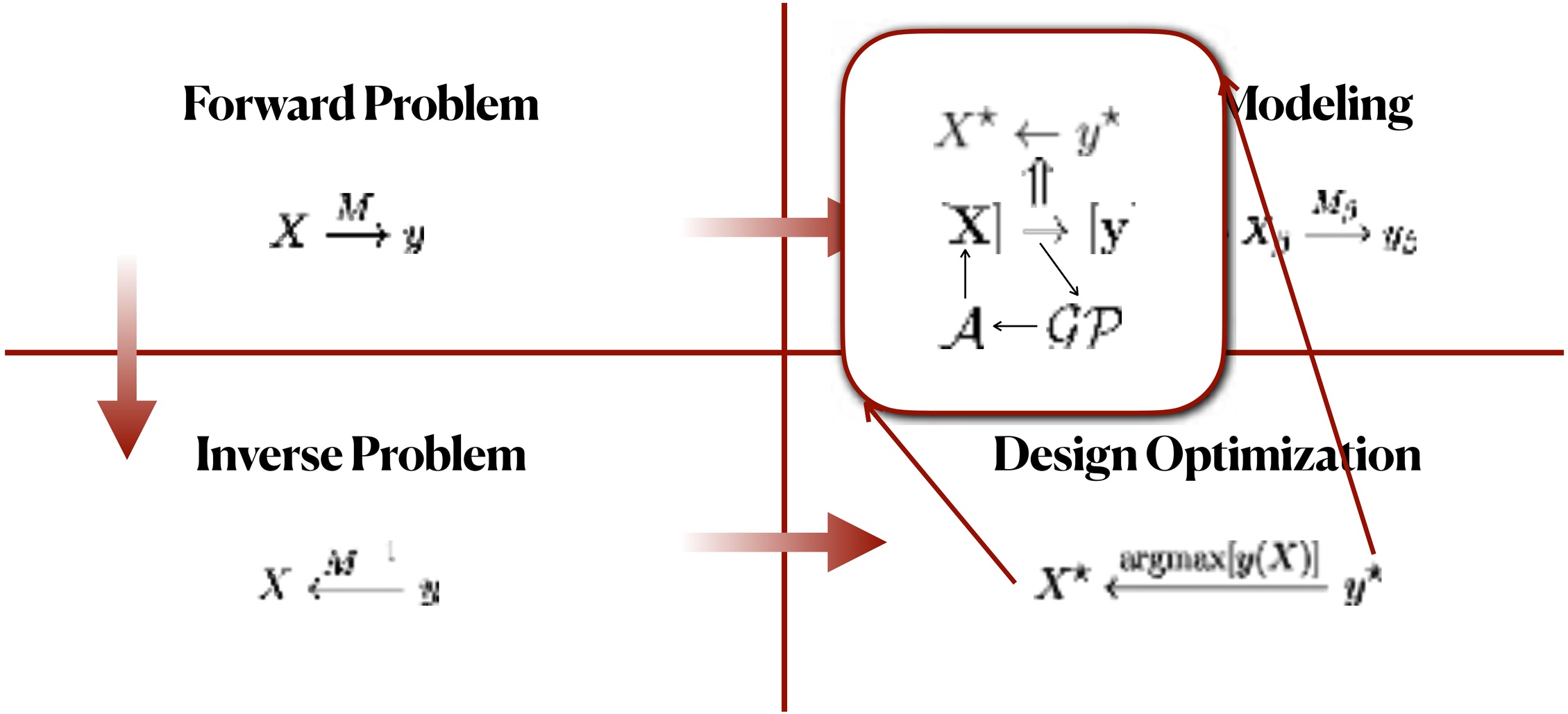
# Part II: Designing Antibiofilm Surfaces



# Outline


Forward Problem
$X \xrightarrow{M} y$
Multiscale Modeling
$X_\alpha \xrightarrow{M_\alpha} y_\alpha \leftrightarrow X_\beta \xrightarrow{M_\beta} y_\beta$
Inverse Problem
$X \xleftarrow{M^{-1}} y$
Design Optimization
$X^* \xleftarrow{\text{argmax}[y(X)]} y^*$

# Outline


Forward Problem
$X \xrightarrow{M} y$
Inverse Problem
$X \xleftarrow{M^{-1}} y$
$X^\star \leftarrow y^\star$
$\Uparrow$
$X] \Rightarrow [y$
$\mathcal{A} \leftarrow GP$
Modeling
$X_3 \xrightarrow{M_3} y_3$
Design Optimization
$X^\star \xleftarrow{\text{argmax}[y(X)]} y^\star$

# Outline

## Forward Problem

$$X \xrightarrow{M} y$$

## Multiscale Modeling

$$X_\alpha \xrightarrow{M_\alpha} y_\alpha \leftrightarrow X_\beta \xrightarrow{M_\beta} y_\beta$$

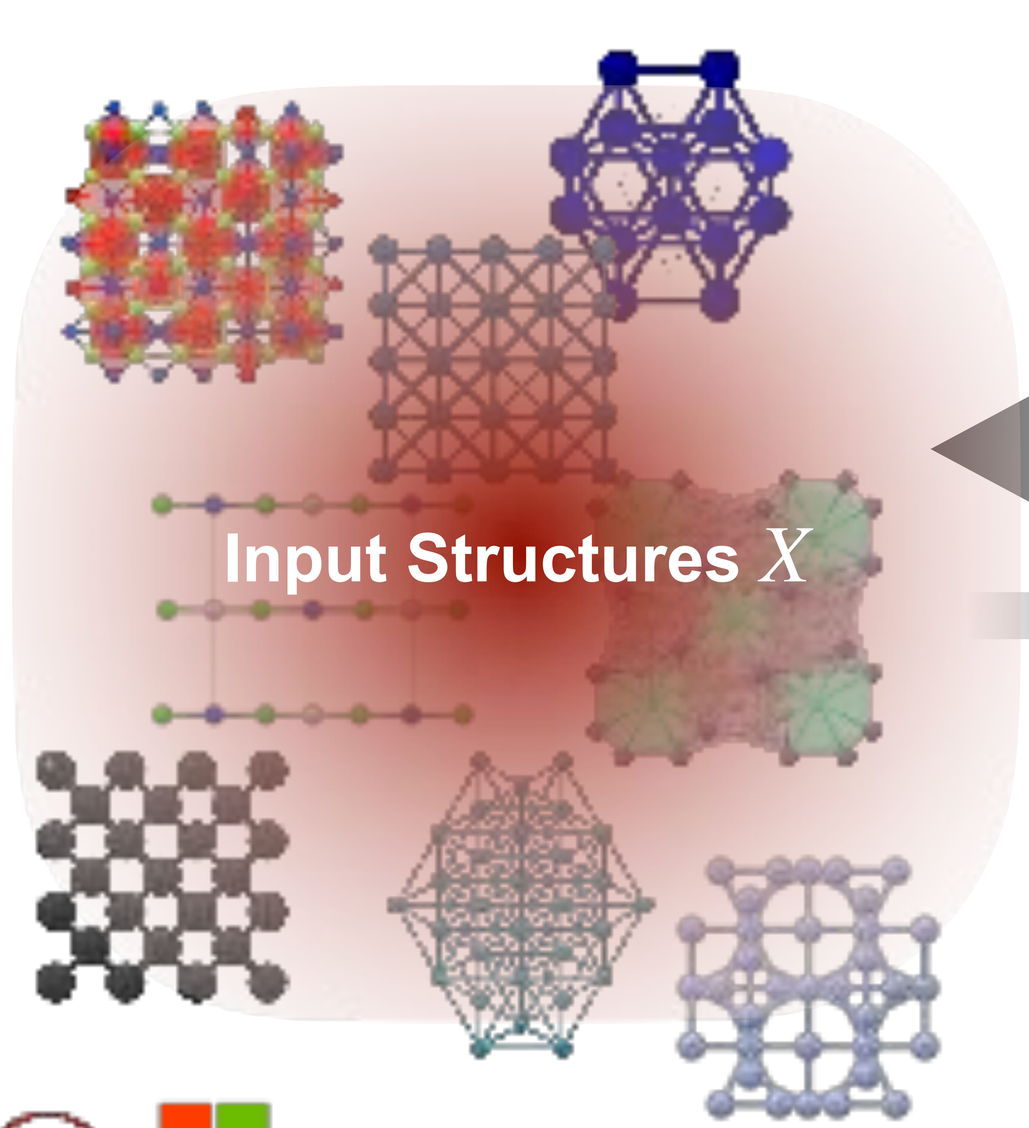
## Inverse Problem

$$X \xleftarrow{M^{-1}} y$$

## Design Optimization

$$X^* \xleftarrow{\text{argmax}[y(X)]} y^*$$

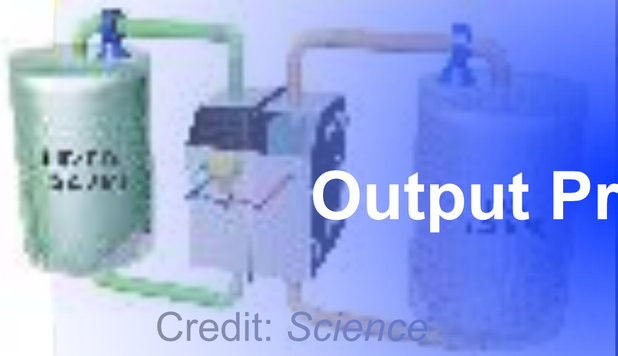
# Part III: Benchmarking Optimization Algorithms



**Input Structures $X$**

Credit: *CNN*

Credit: *Forbes*



Credit: *Science*

**Output Properties $y$**

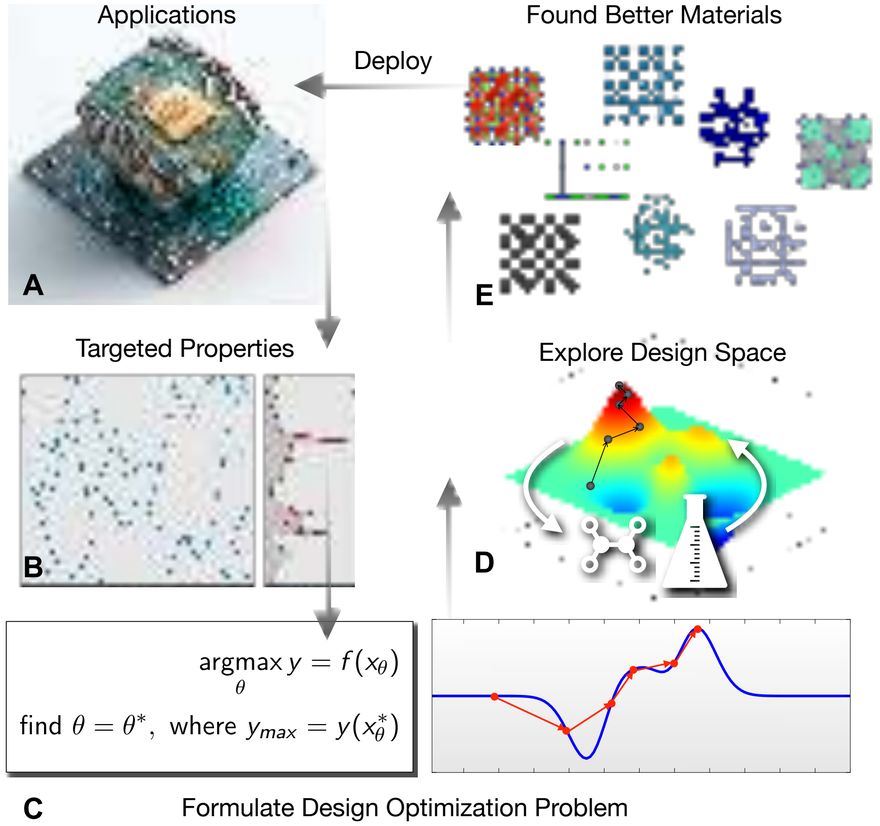
Credit: *Nature Portfolio*

Credit: *CNN*

Credit: *The New York Times*

# Part III: Benchmarking Optimization Algorithms

*Begin the research by **asking the question** from the **design** perspective*




**Question I:** How to design molecular materials digitally?

**Question II:** How to benchmark different optimization methods? What's the differences?

**Question III:** What's the extracted materials from the optimizations? Does they obey real-world scenarios?

**Zhai,** Hao, & Yeo, *Unpublished*, 2023.

# Part III: Benchmarking Optimization Algorithms

**Question I:** How to design molecular materials digitally?

## Design Optimization

- The design optimization problem is formulated as:

$$\underset{n_{atom},\,\xi_n,\,\eta}{\arg\max}\ \mathscr{J} = K - E_{Fermi},$$

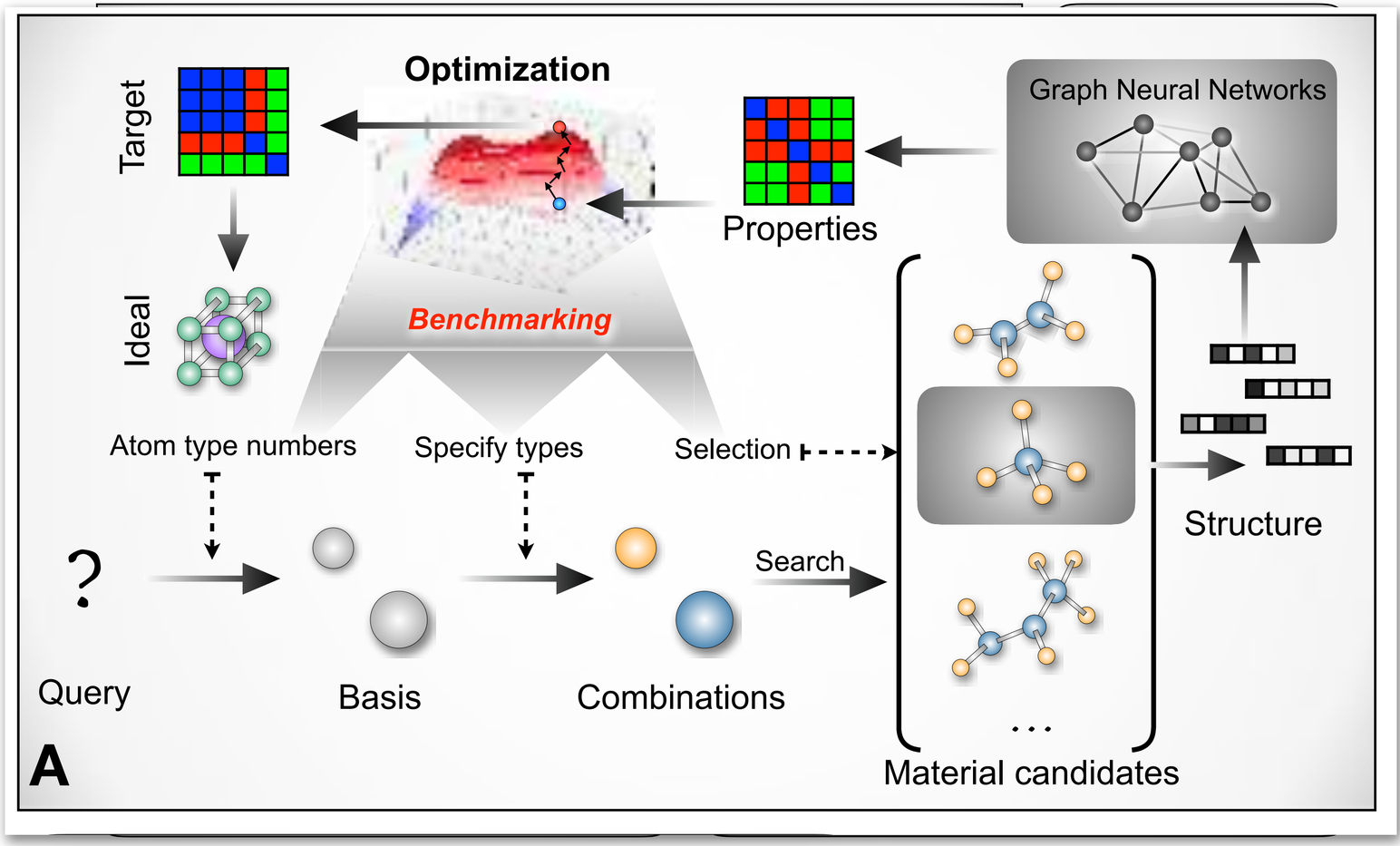
$$\text{where}\quad K, E_{Fermi} = MEGNet(\mathscr{G}_\Theta),$$

$$\rightarrow \mathscr{G} = \Omega(n_{atom},\,\xi_n,\,\eta);\ \Theta = [n_{atom}, \xi_n, \eta]$$

$$\text{subject to}\quad n_{atom} \in [1,4] \text{ or } \equiv 1,\ \xi_n \in [0,100],\ \eta \in [0,100]$$

- The automation is connected via MEGNet:

$$\overbrace{\underbrace{(MEGNet(\Theta_{n+1}) \leftarrow \Theta_{n+1})}_{\text{exploration}}}^{\text{evaluation}} \underset{\text{feedback}}{\overset{\text{search}}{\rightleftharpoons}} \overbrace{\underbrace{IOA(\Theta_n, MEGNet(\Theta_n); \mathbf{P}_{IOA})}_{\text{exploitation}}}^{\text{evaluation}}$$



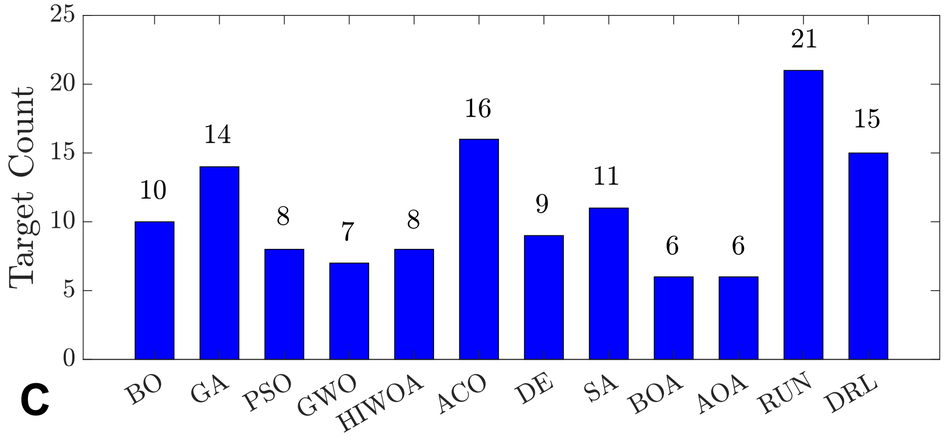

**Zhai,** Hao, & Yeo, *Unpublished*, 2023.

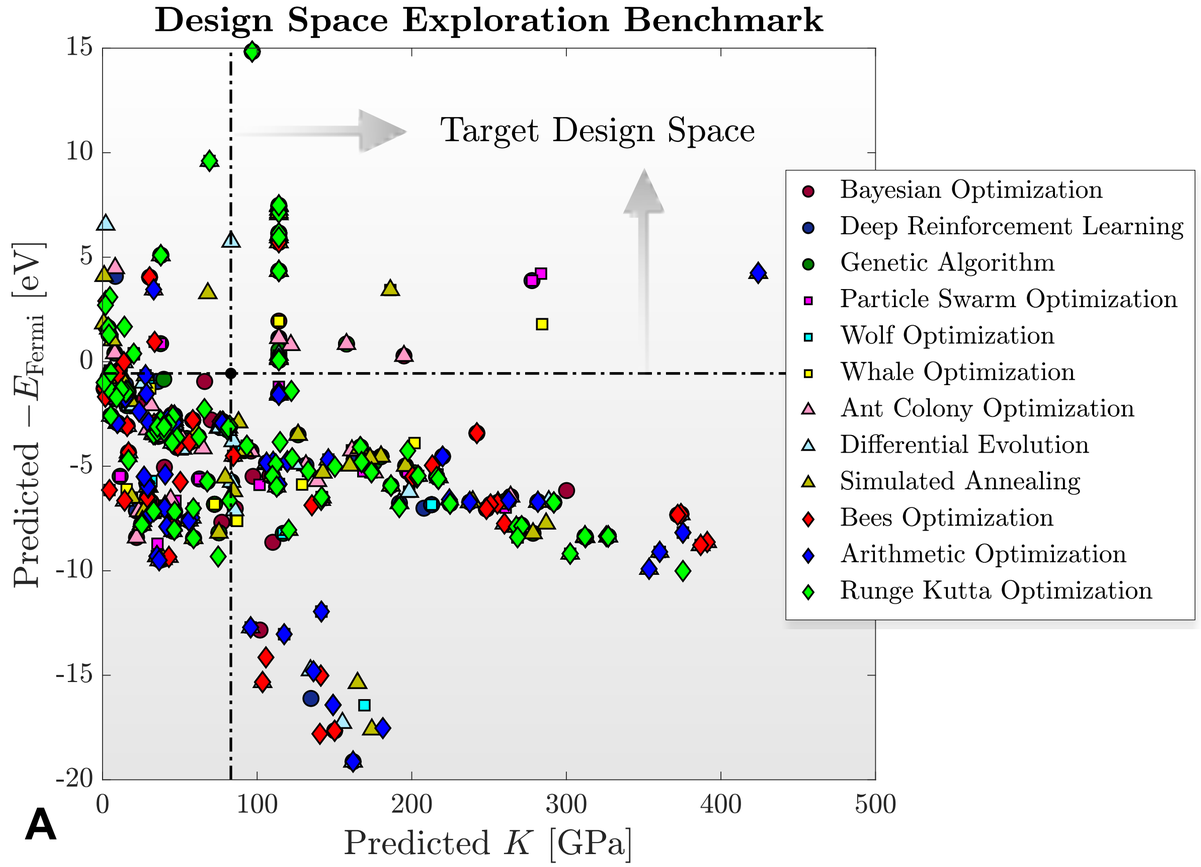
# Part III: Benchmarking Optimization Algorithms

**Question II:** How to benchmark different optimization methods? Differences?

**Observations**

- The RUN algorithm outperforms the result optimization methods in material count in the “target design space”.
- GA, ACO, and DRL are generally good in single-element molecule design.



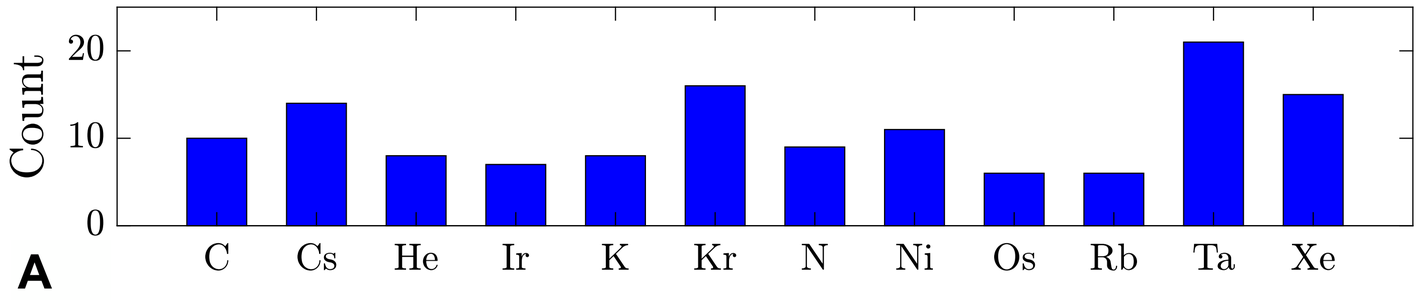

**Zhai,** Hao, & Yeo, *Unpublished*, 2023.

# Part III: Benchmarking Optimization Algorithms

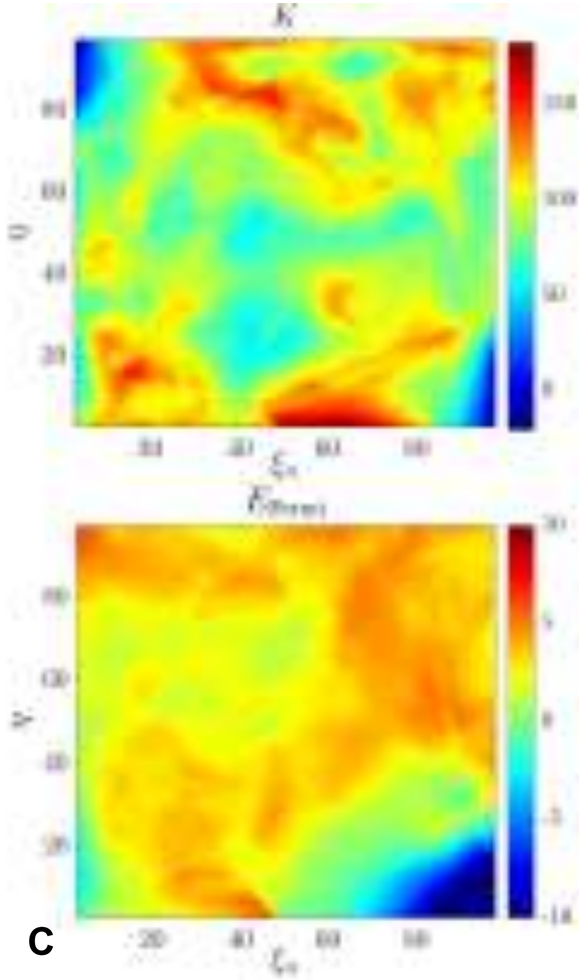
**Question II:** How to benchmark different optimization methods? Differences?

**Observations**

- Ta is the most evaluated molecule among 12 optimization methods.
- Design space is highly non-convex.




A



C

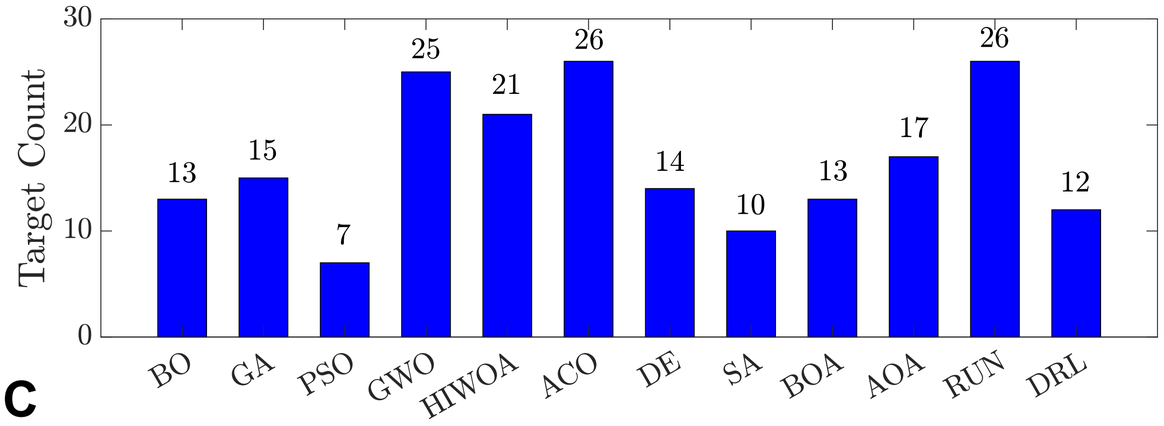
**Zhai,** Hao, & Yeo, *Unpublished*, 2023.

# Part III: Benchmarking Optimization Algorithms

**Question II:** How to benchmark different optimization methods? Differences?

**Observations**

- GWO, HIWOA, ACO, and RUN stand out for target design space material counts.
- DRL didn't successful learn the policy (per se).

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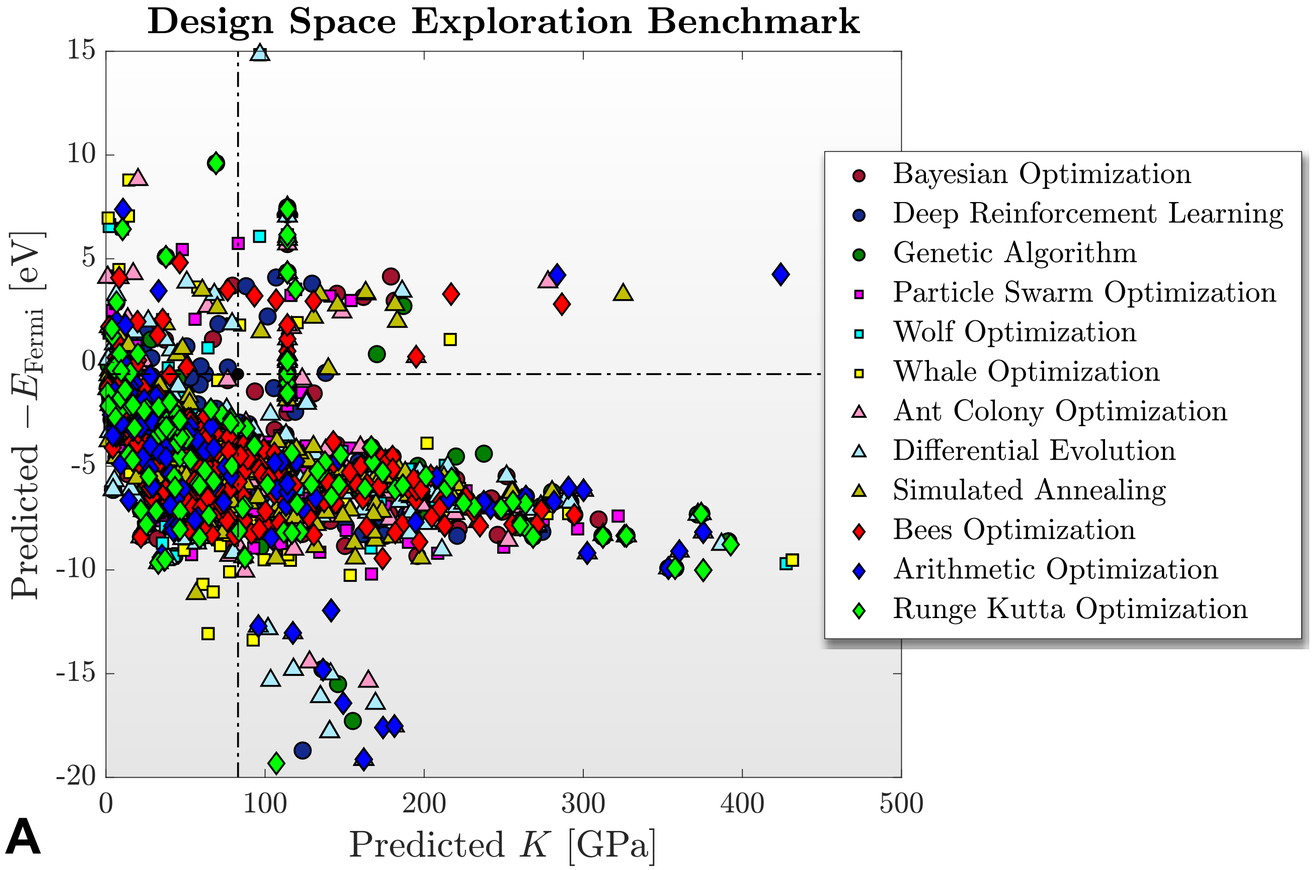



A

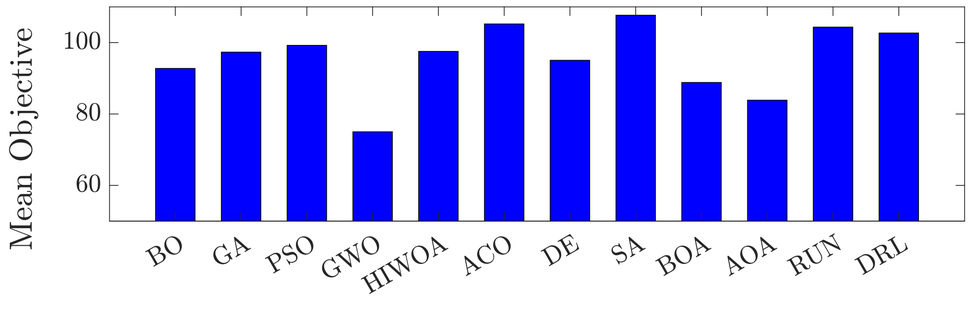
**Zhai,** Hao, & Yeo, *Unpublished*, 2023.

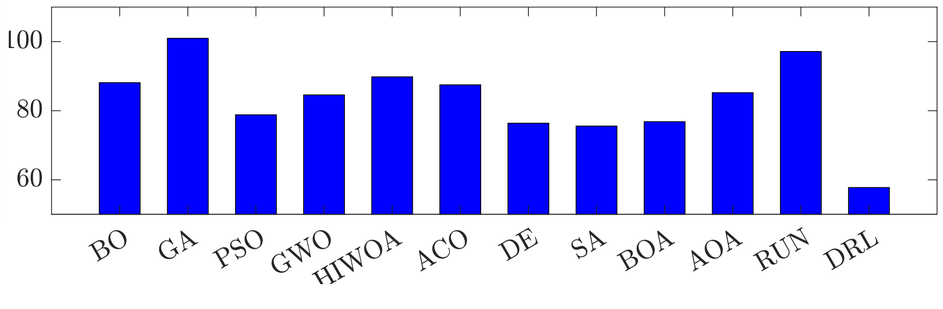
# Part III: Benchmarking Optimization Algorithms

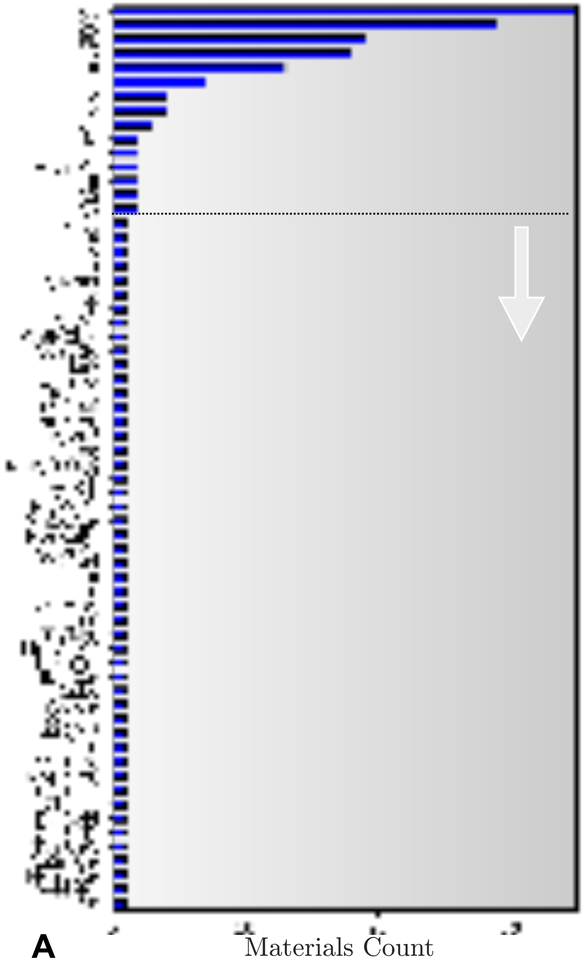
**Question II:** How to benchmark different optimization methods? Differences?

**Observations**

- Cs is the most evaluated material among all the optimization methods.
- $Hf_2InMo$ is the most evaluated multi-element chemical compound.
- ACO, SA, RUN, and DRL: higher mean objective values for single-element materials; GA and RUN: multi-element mat.




**Zhai,** Hao, & Yeo, *Unpublished*, 2023.

# Outline

## Forward Problem

$$X \xrightarrow{M} y$$

## Multiscale Modeling

$$X_\alpha \xrightarrow{M_\alpha} y_\alpha \leftrightarrow X_\beta \xrightarrow{M_\beta} y_\beta$$

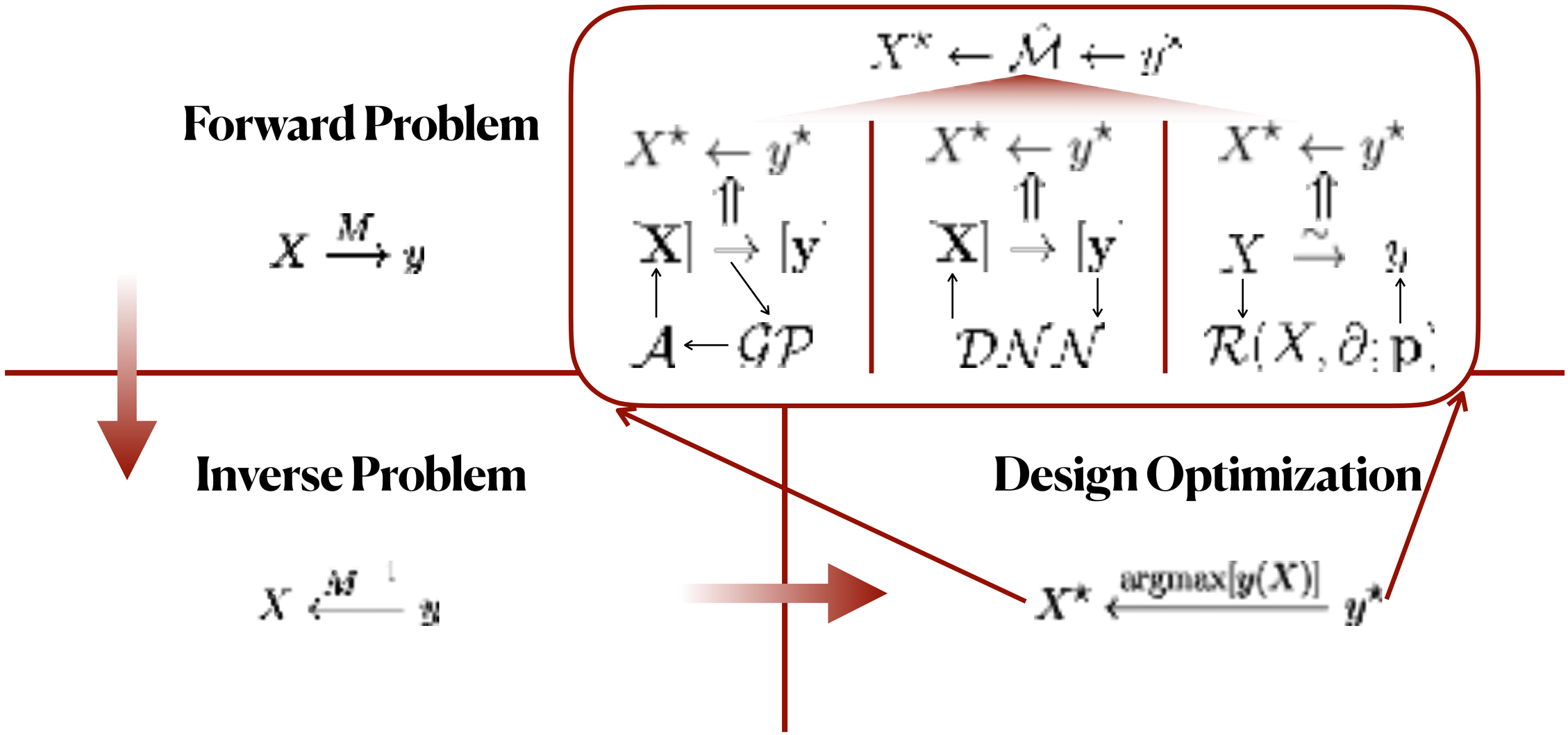
## Inverse Problem

$$X \xleftarrow{M^{-1}} y$$

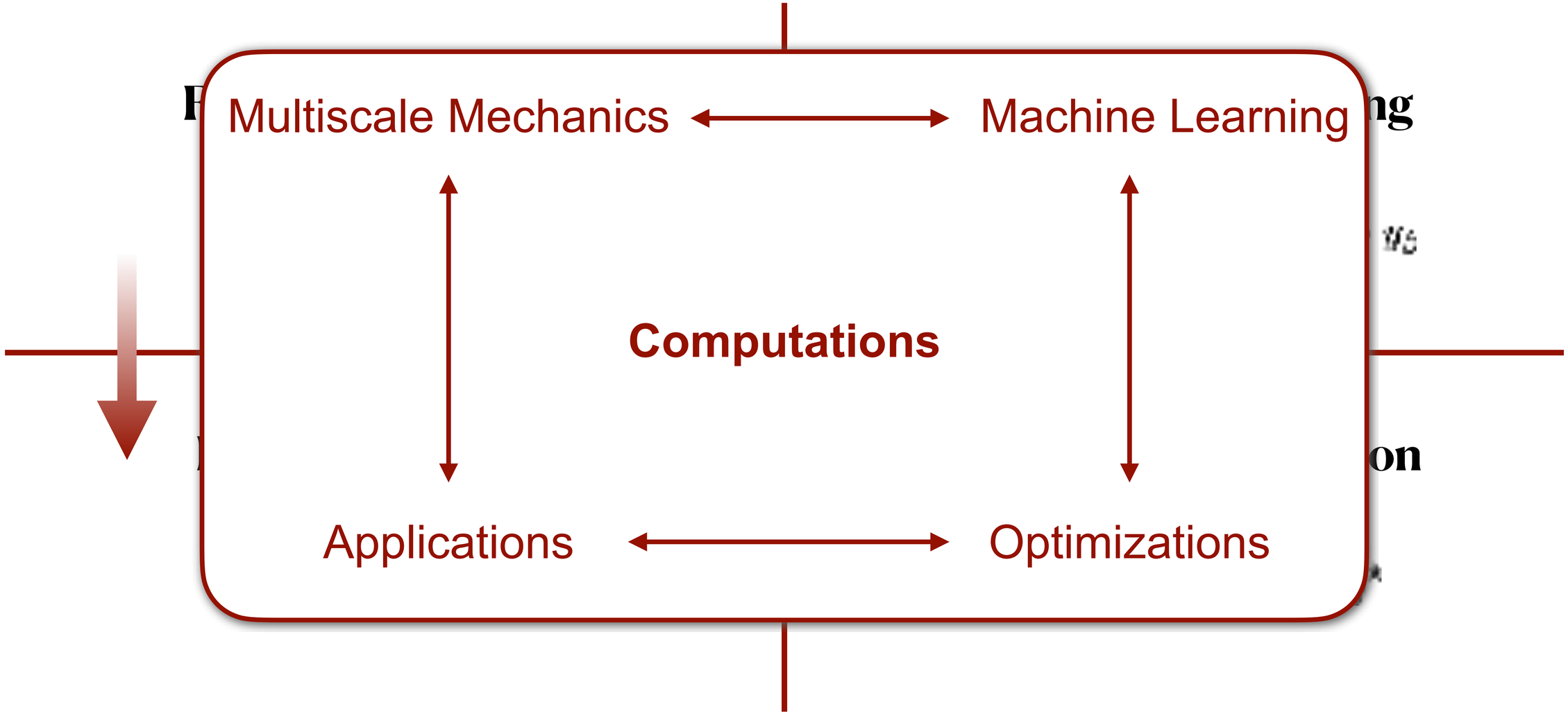
## Design Optimization

$$X^* \xleftarrow{\text{argmax}[y(X)]} y^*$$

# Outline


Forward Problem
$X \xrightarrow{M} y$
Inverse Problem
$X \xleftarrow{M^{-1}} y$
$X^* \leftarrow \hat{\mathcal{M}} \leftarrow y^*$
$X^* \leftarrow y^*$
$\mathbf{X}] \rightarrow [y$
$\mathcal{A} \leftarrow \mathcal{GP}$
$X^* \leftarrow y^*$
$\mathbf{X}] \rightarrow [y$
$\mathcal{DNN}$
$X^* \leftarrow y^*$
$X \xrightarrow{\sim} y$
$\mathcal{R}(X, \partial; \mathbf{p})$
Design Optimization
$X^* \xleftarrow{\text{argmax}[y(X)]} y^*$

# Outline


Multiscale Mechanics
Machine Learning
Computations
Applications
Optimizations

# Summary

## Fall 2021

- Foundations of Solid Mechanics
- Multidisc. Design Optimization
- Adaptive and Learning Systems
- Seminars & Colloquiums

## Spring 2022

- Computational Materials Scien.
- Multiscale Computational Mech.
- Bas. Programming Python
- Seminars & Colloquiums

## Fall 2022

- Math. Modeling of Systems
- Principles of Large-Scale ML (V)

## Spring 2023

- Non. Finite Element Analy.: Solids
- Inverse Problems: Theory & Appl.

# Summary

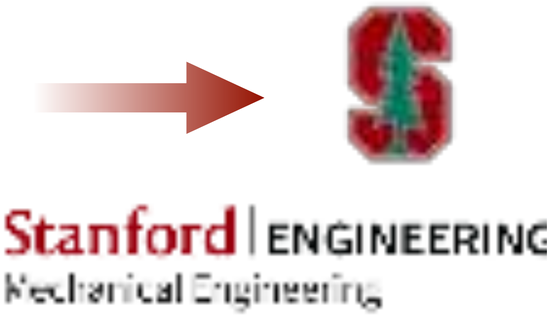
## Publications

- Zhai & Yeo, *ACS Biomater. Sci. Eng.* 2023, **9**, 1, 269–279
- Zhai & Yeo, *Int. J. Appl. Mech.*, 2023 (In Press)

  - Wang et al., Submitted
  - Zhai, Hao, & Yeo, In preparation
  - Zhai & Yeo, In preparation
  - …

## Presentations

- MIT Molecular ML Conference
- Sibley Graduate Research Symposium
- MSE Graduate Research Symposium
- ELMI Monthly Meeting
- ELMI Research Symposium
- MAE PhD Visit Day
- Sobhani Lab Group Meeting




# Acknowledgement


Prof. Jingjie Yeo

Prof. Derek Warner

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TACC

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