UNIVERSITY OF LIVERPOOL

LEVERHULME TRUST

The Leverhulme Research Centre for Functional Materials Design

Geometry-Informed Machine Learning for Materials Science

Winter School on Robotics and AI for Materials Discovery

Miloslav Torda

3 December 2024

CCDC diamond Hartree Centre Science & Technology Facilities Council Imperial College London ICNS King Abdullah University of Science and Technology UNIVERSITY OF Southampton

Days 1 - 10
Teach yourself variables, constants, arrays, strings, expressions, statements, functions,...

Days 11 - 21
Teach yourself program flow, pointers, references, classes, objects, inheritance, polymorphism, ...

Days 22 - 697
Do a lot of recreational programming. Have fun hacking but remember to learn from your mistakes.

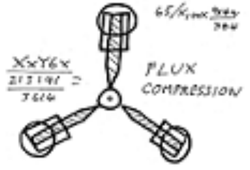
Days 698 - 3648
Interact with other programmers. Work on programming projects together. Learn from them.

Days 3649 - 7781
Teach yourself advanced theoretical physics and formulate a consistent theory of quantum gravity.

Days 7782 - 14611
Teach yourself biochemistry, molecular biology, genetics,...

Day 14611
Use knowledge of biology to make an age-reversing potion.

Day 14611
Use knowledge of physics to build flux capacitor and go back in time to day 21.



Day 21
Replace younger self.

As far as I know, this is the easiest way to "Teach Yourself C++ in 21 Days".

Outline

1. Why think about machine learning geometrically?

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2. Case Study: Molecular Crystal Structure Prediction.

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2. Case Study: Molecular Crystal Structure Prediction.

3. Closing remarks.

1. Why think about machine learning geometrically?

Geometry of the Linear Regression Model

N measurements. $(y_i, x_i^1, \ldots, x_i^k)$ $(i = 1, \ldots, N)$

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Linear Algebra

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$$\underbrace{\begin{pmatrix} y_1 \\ y_2 \\ \vdots \\ y_N \end{pmatrix}}_{\boldsymbol{y}} = \underbrace{\begin{pmatrix} 1 & x_1 & \cdots & x_1^k \\ 1 & x_2 & \cdots & x_2^k \\ \vdots & \vdots & \ddots & \vdots \\ 1 & x_N & \cdots & x_N^k \end{pmatrix}}_{\mathbf{F}} \underbrace{\begin{pmatrix} \theta_0 \\ \theta_1 \\ \vdots \\ \theta_k \end{pmatrix}}_{\boldsymbol{\theta}} + \underbrace{\begin{pmatrix} \varepsilon_1 \\ \varepsilon_2 \\ \vdots \\ \varepsilon_N \end{pmatrix}}_{\boldsymbol{\varepsilon}}$$

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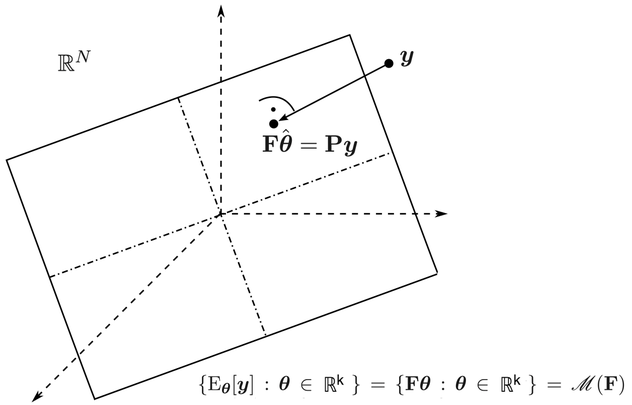

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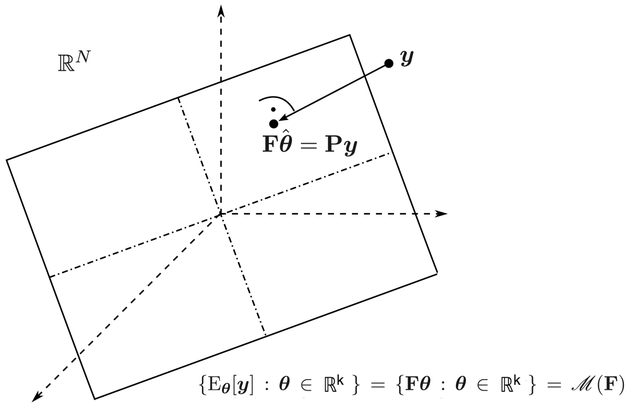

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Learning means finding values of θ from measurements

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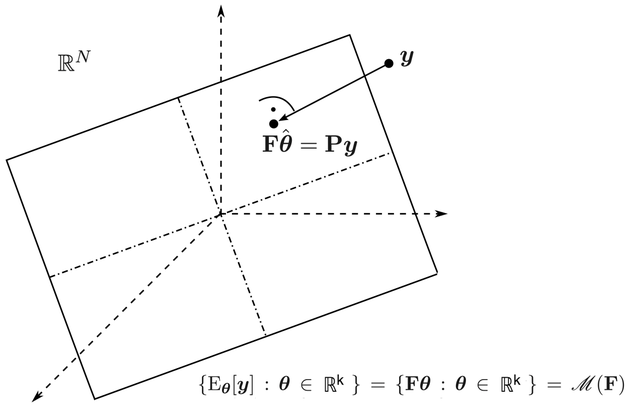

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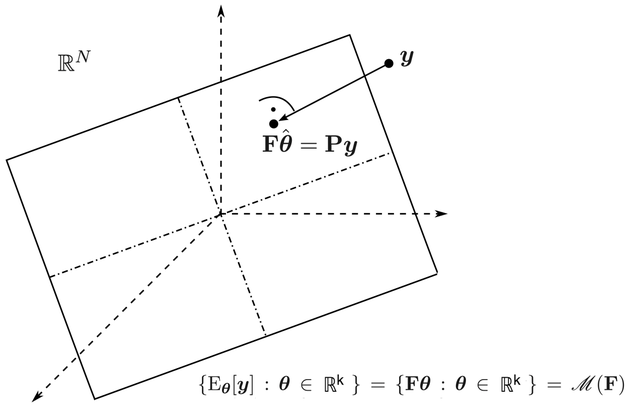

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Example. $\mathbf{y} = \begin{pmatrix} 1 \\ 1 \\ \vdots \\ 1 \end{pmatrix} \theta_0 + \epsilon$ $\qquad \hat{\theta}_0 = \frac{1}{N} \sum_{i=1}^{N} \mathbf{y_i}$

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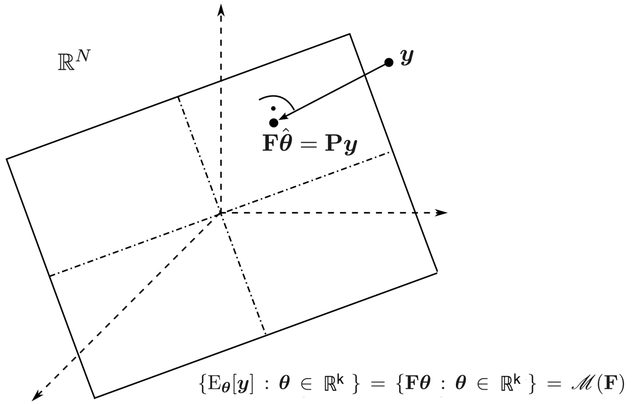

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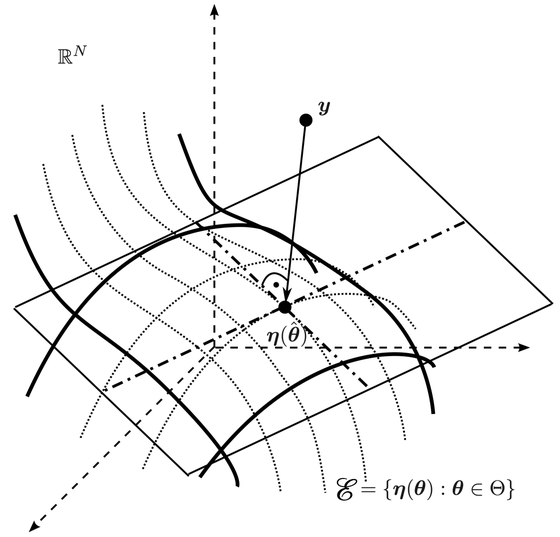

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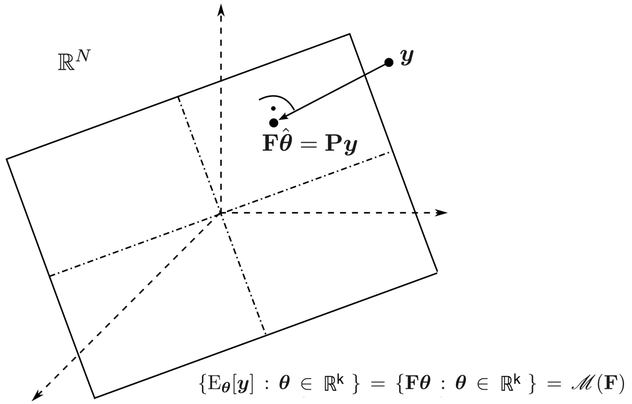

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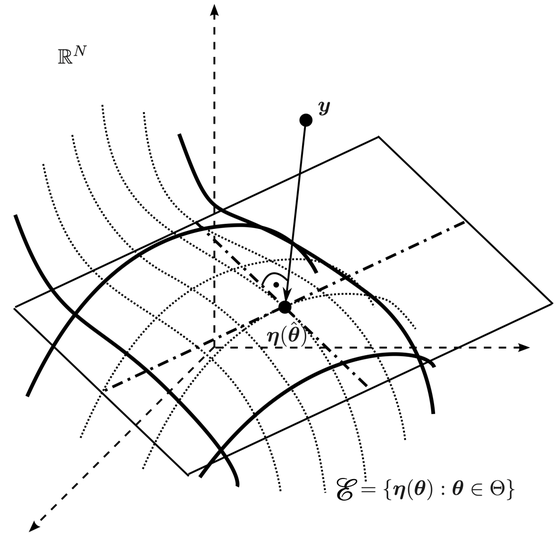

However, the underlying idea is the same.

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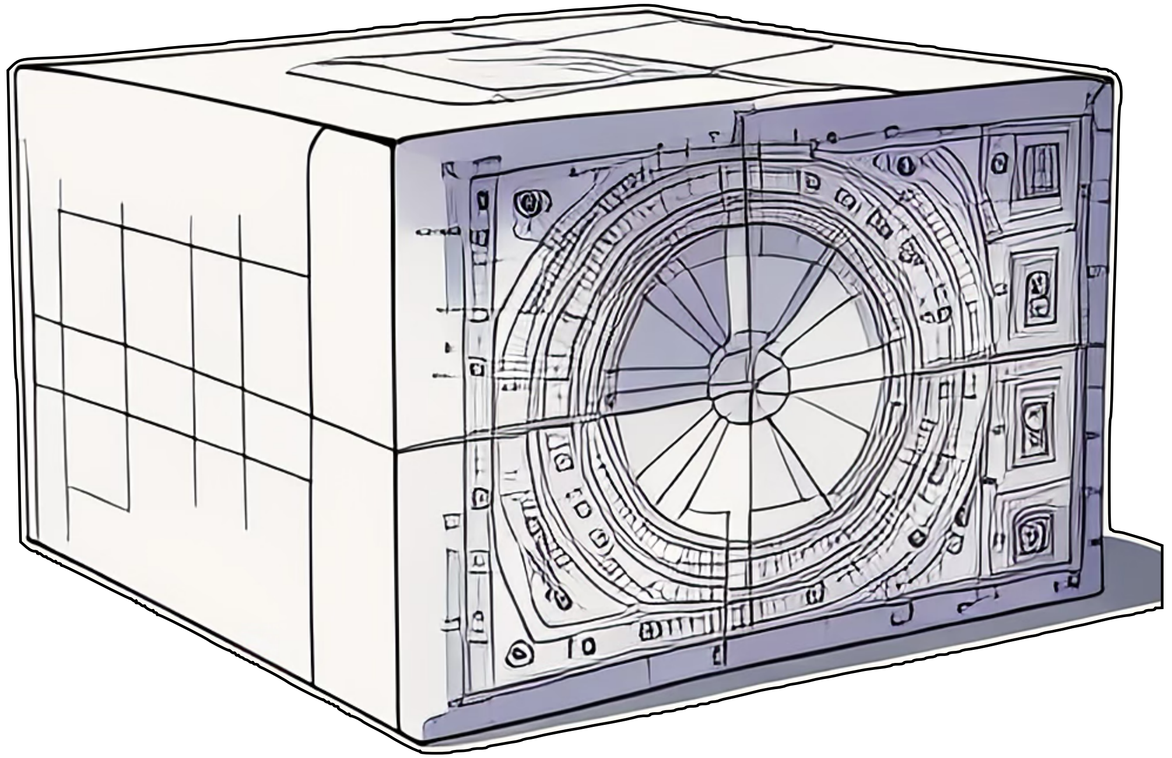

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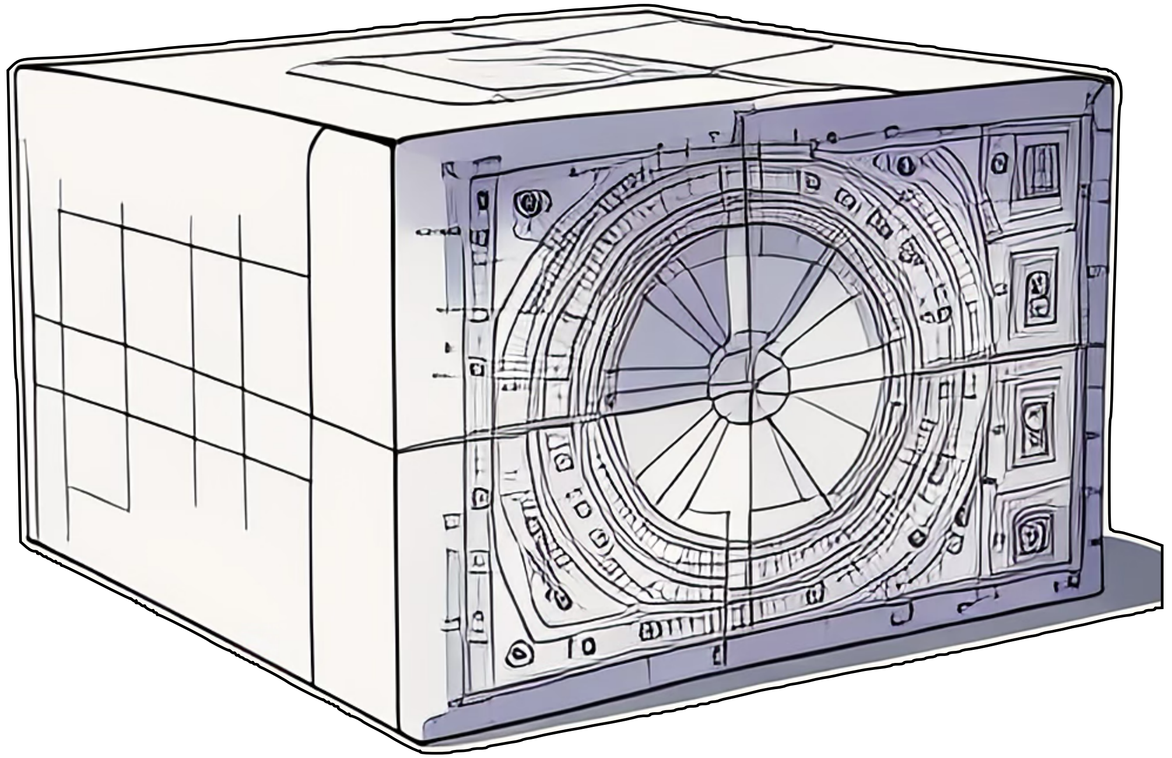

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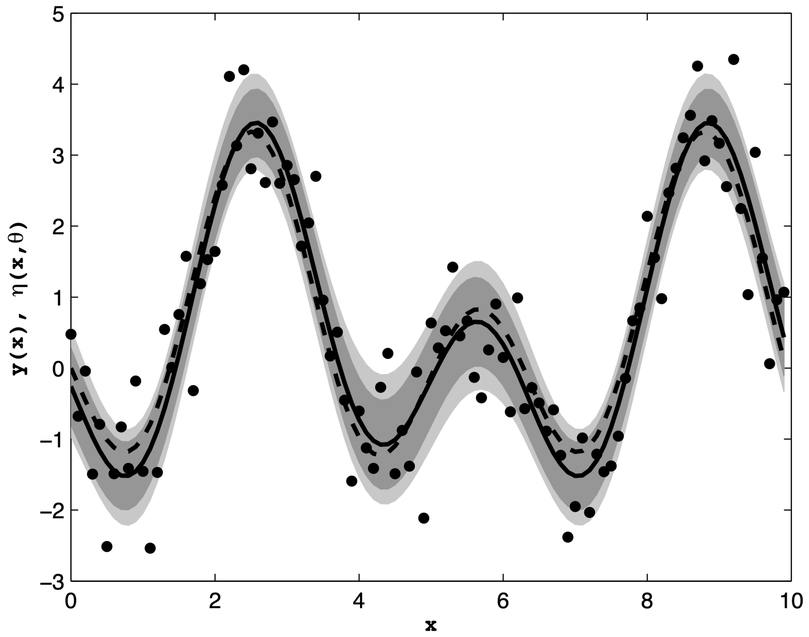
Trigonometric regression

Model 1. $y = 0.4514 + 0.6077\sin(x) - 1.2837\cos(x) - 1.4938\sin(2x) + 0.5741\cos(2x)$

Model 2. $y = 0.4521 + 0.6065\sin(x) - 1.2916\cos(x) - 1.4803\sin(2x) + 0.5728\cos(2x) + 0.0155\sin(3x) + 0.0584\cos(3x)$



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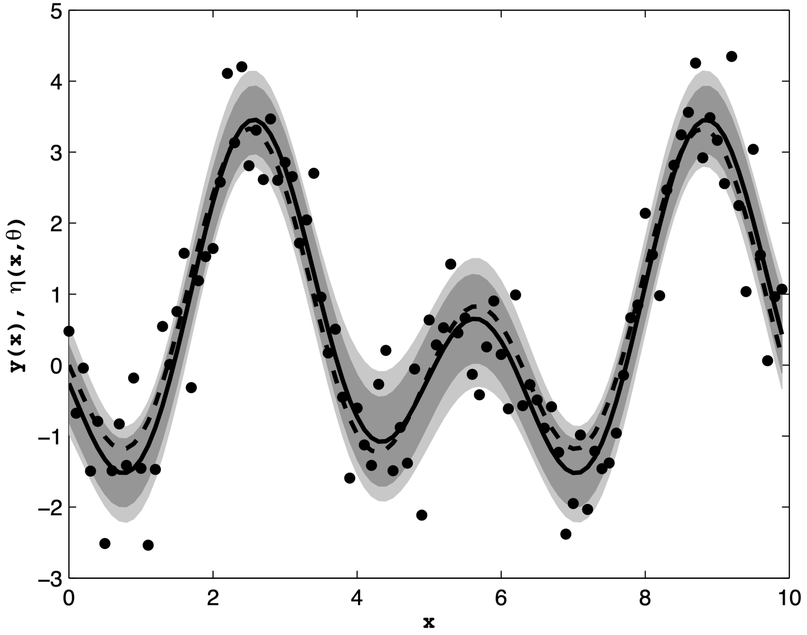
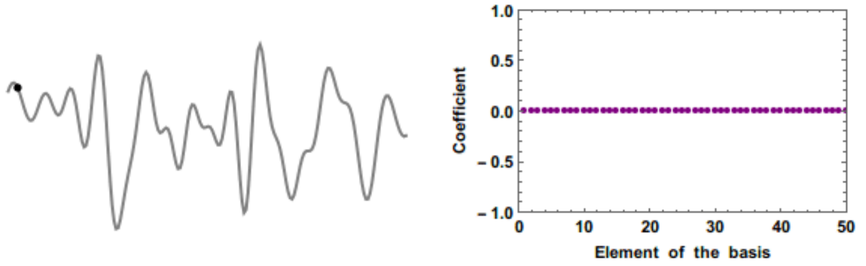


Example. Compressed Sensing / Sparse Sampling



Jacopo Bertolotti, CC0, via Wikimedia Commons, https://commons.wikimedia.org/wiki/File:Orthogonal_Matching_Pursuit.gif

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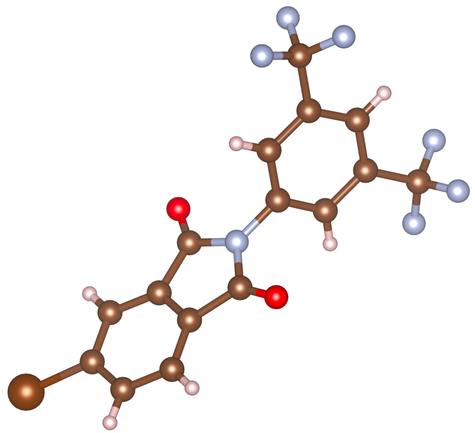
2. Case Study: Molecular Crystal Structure Prediction.

Physical problem. Predict the arrangement of molecules within a crystalline solid based only on the chemical composition of the target molecule from first-principles (i.e. thermodynamics, quantum mechanics)



Chemical composition of the target molecule

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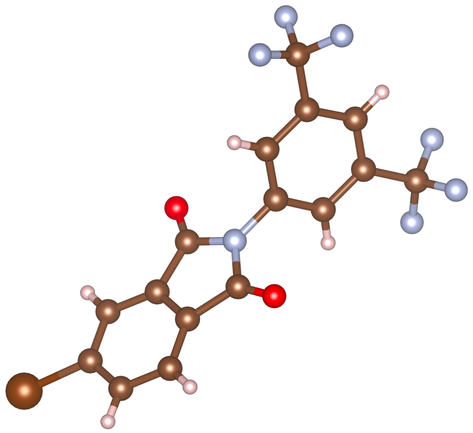

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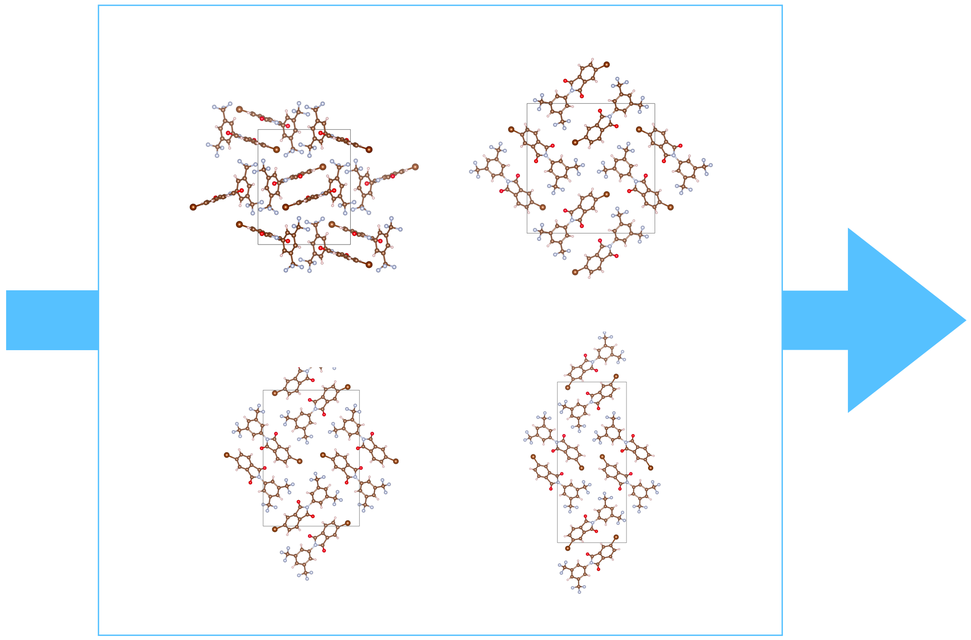

Force-field based structure generation and filtering

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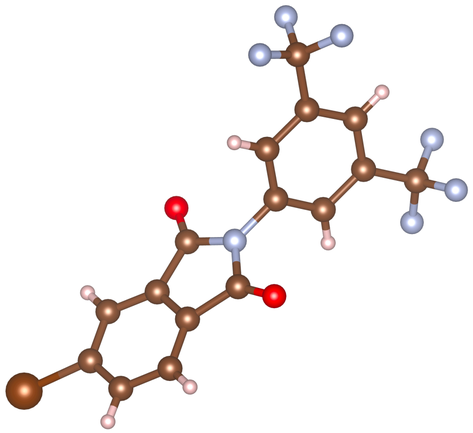

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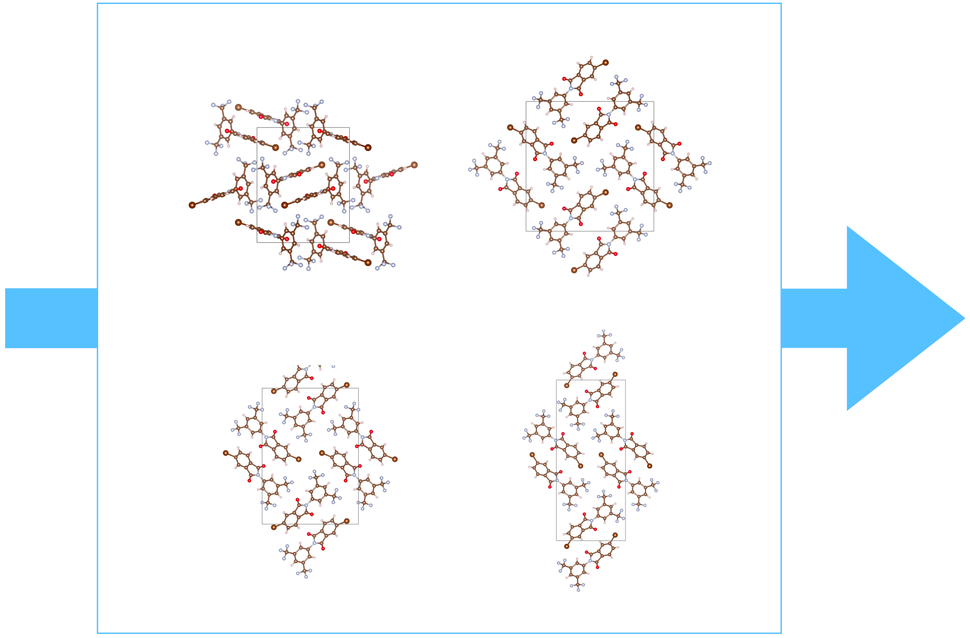

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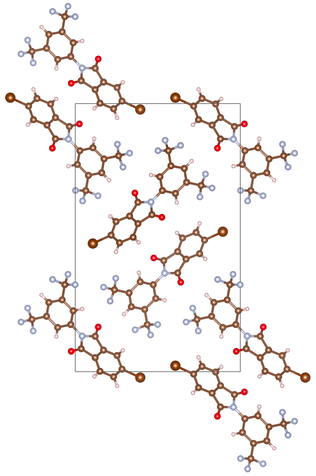

High-Accuracy DFT Geometry Refinement and Final Structure Assessment

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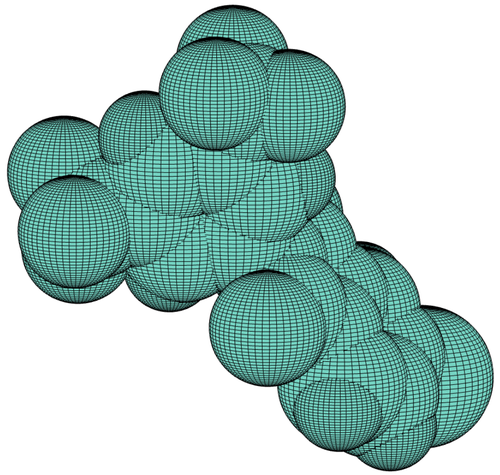
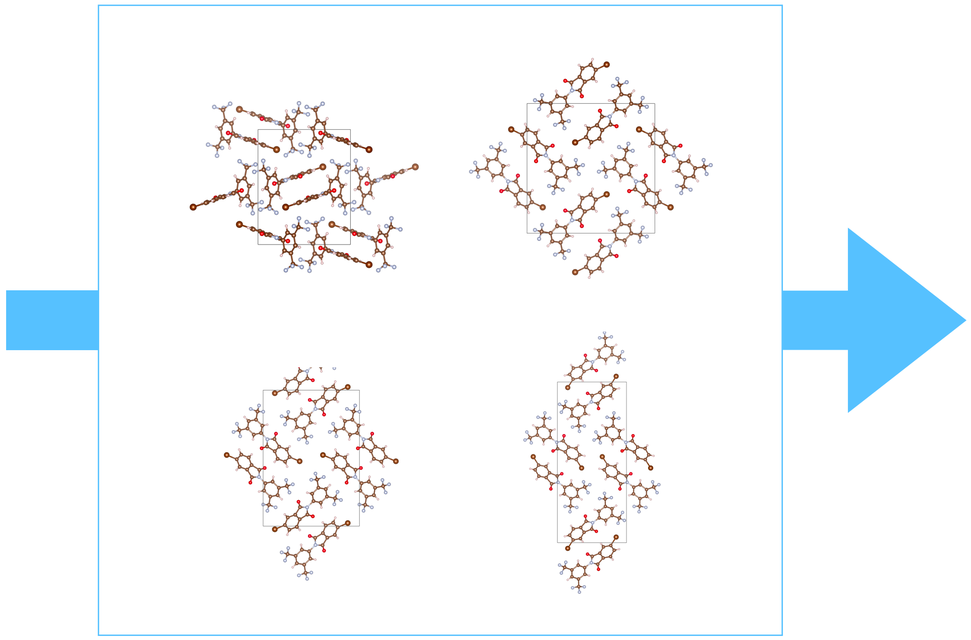


Geometric Representation of a Molecule



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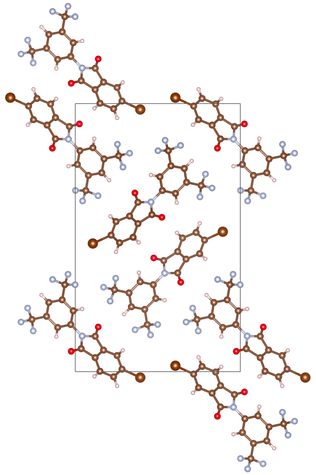

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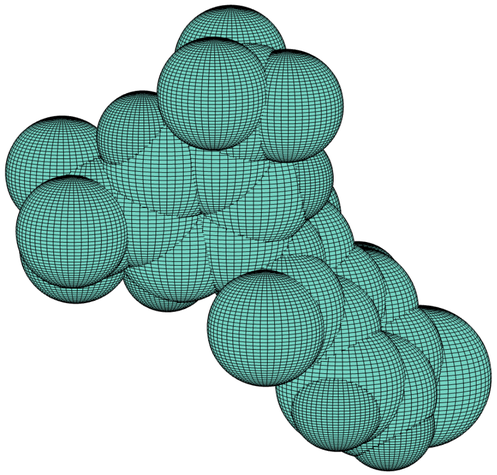

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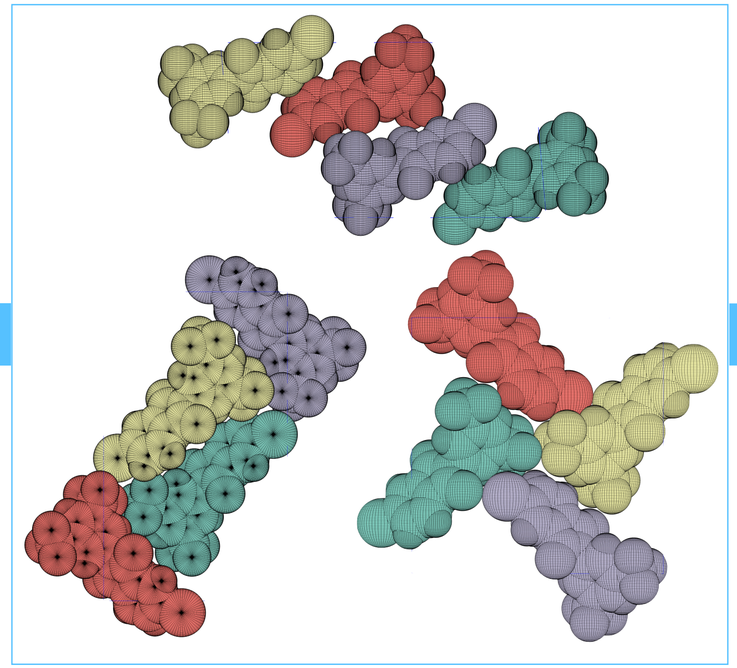
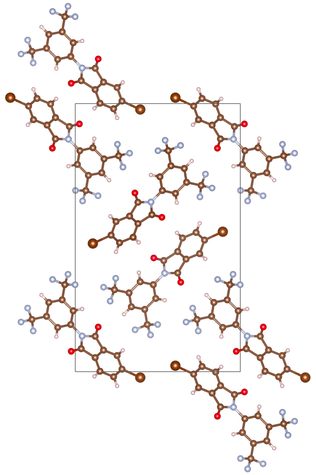


Geometric-Inductive-Biased Structure Generation



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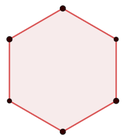
Rigid molecule with 6 general atoms. Configuration of molecule $\xi = \begin{pmatrix} c_1 \\ c_2 \\ \delta \end{pmatrix}$



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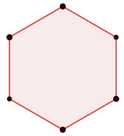
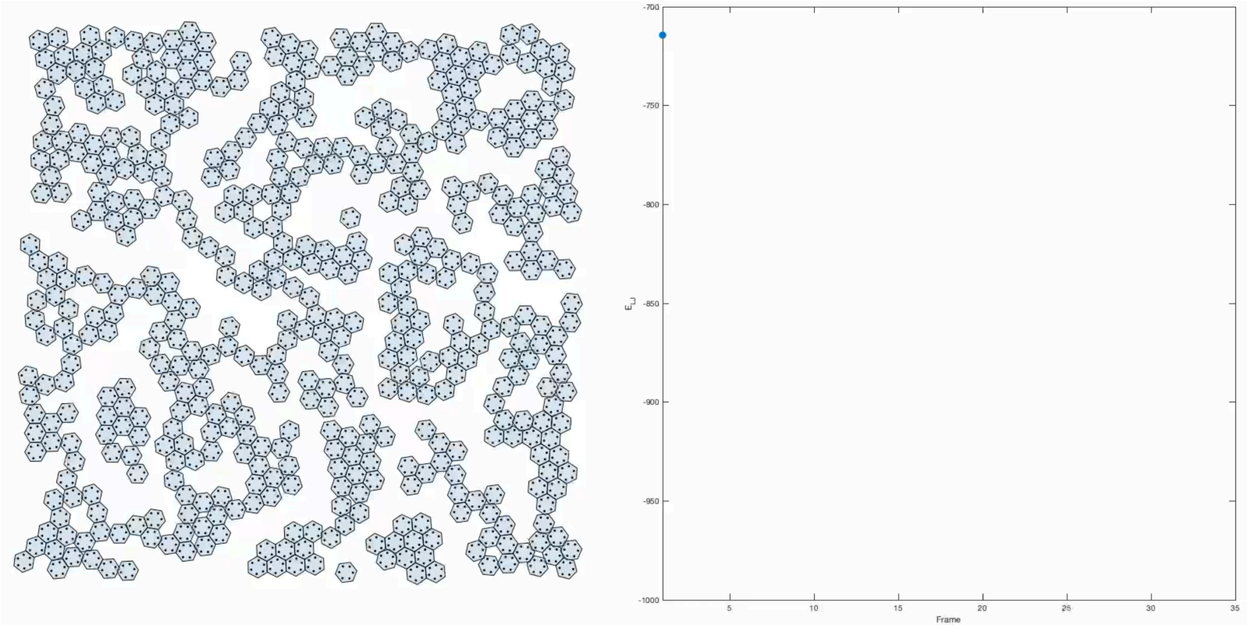
A system of 500 model molecules in a square box.





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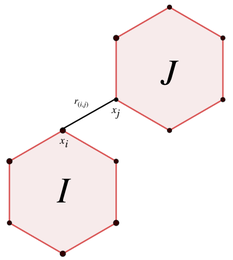
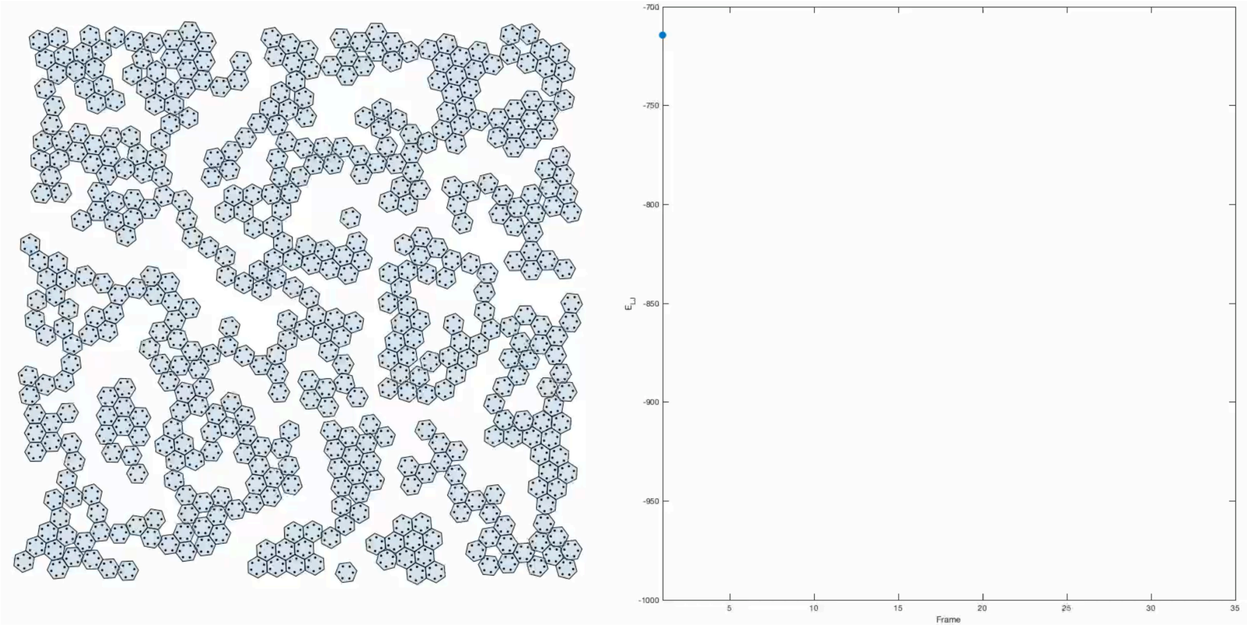
Intermolecular energy: $E(I,J) = \frac{1}{2}\sum_{i\in I}\sum_{j\in J}\left[\left(\frac{1}{r_{(i,j)}}\right)^{12} - \left(\frac{1}{r_{(i,j)}}\right)^{6}\right]$ $r_{(i,j)} = \|x_i - x_j\|$, $x_* \in \mathbb{R}^2$





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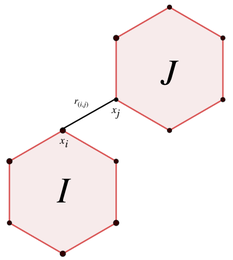

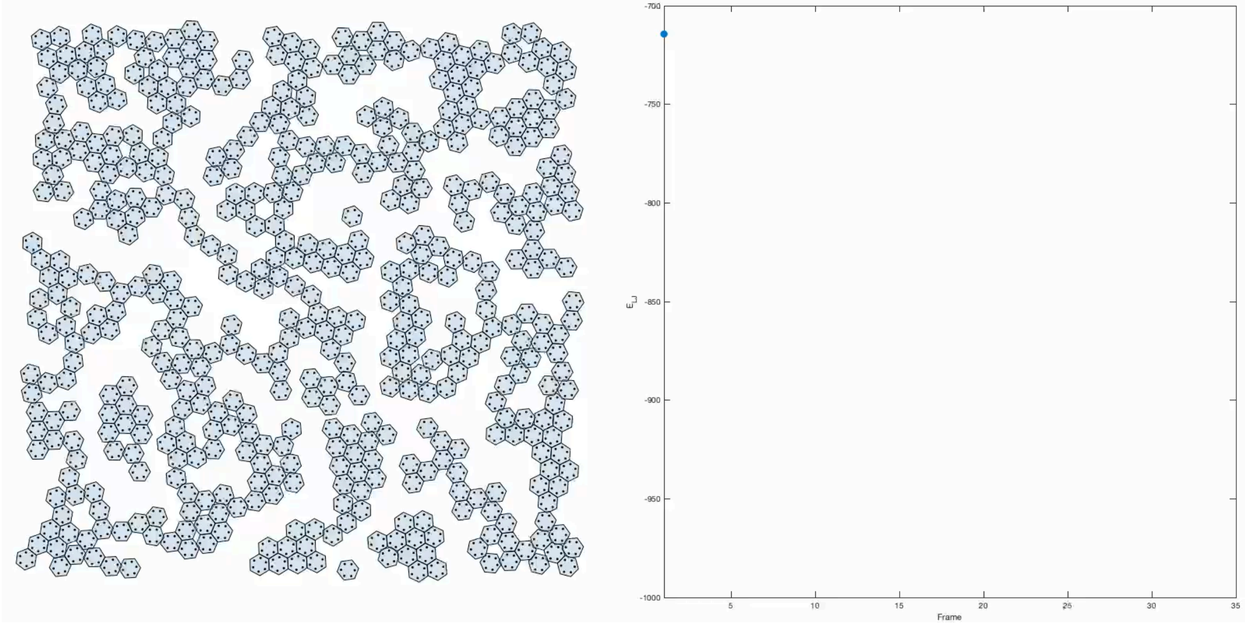

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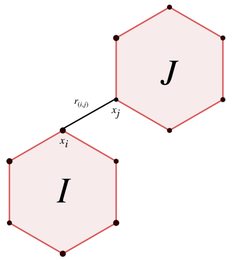

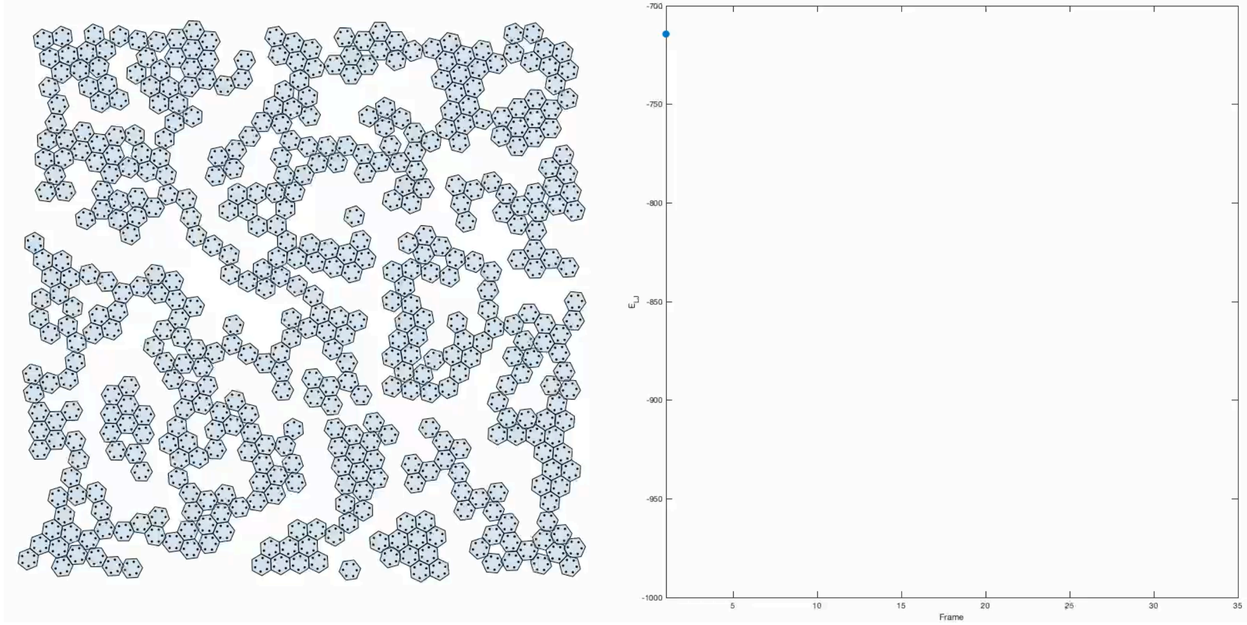

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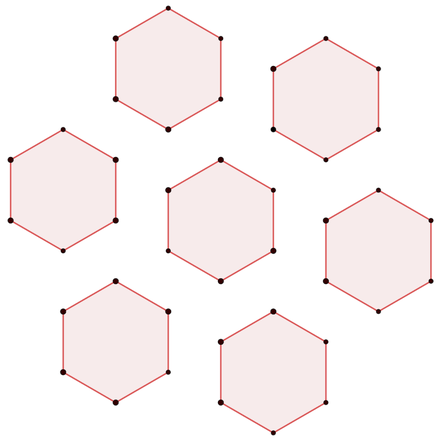

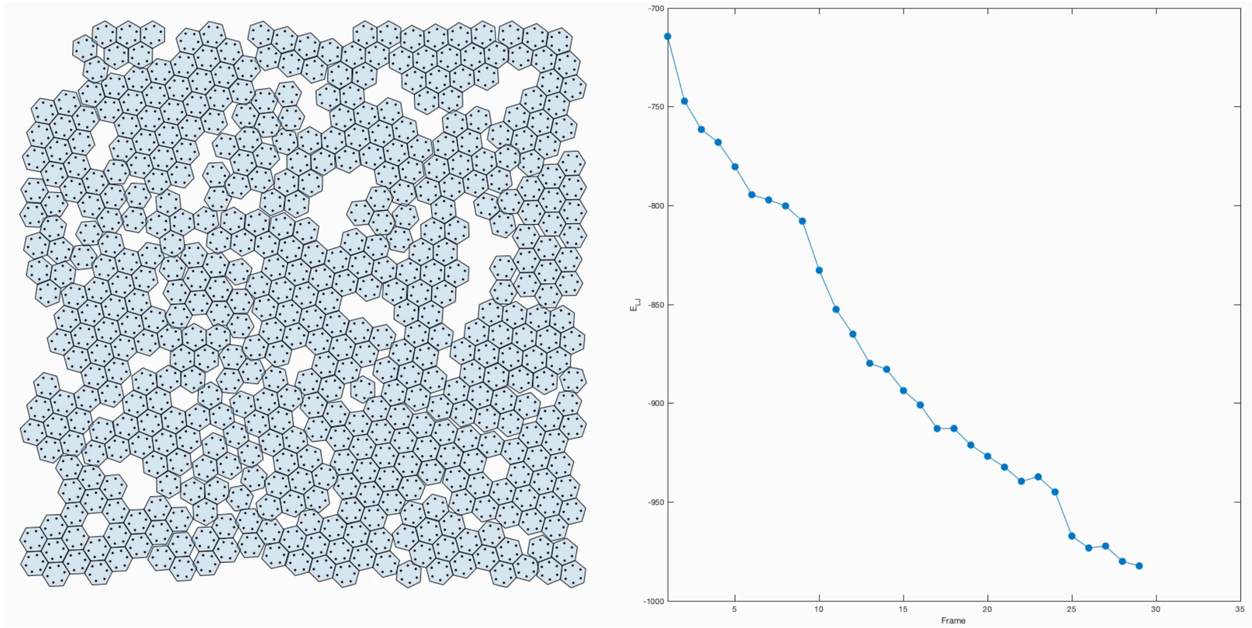

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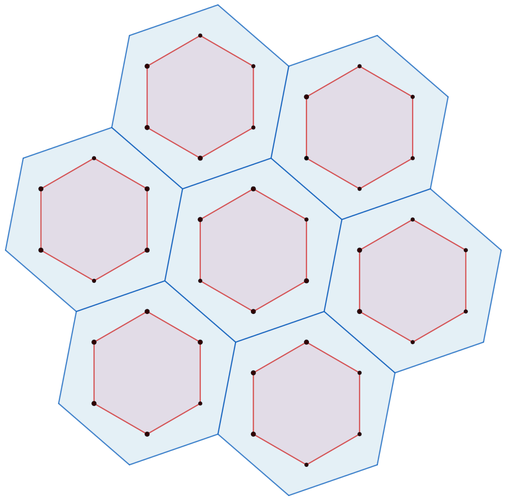

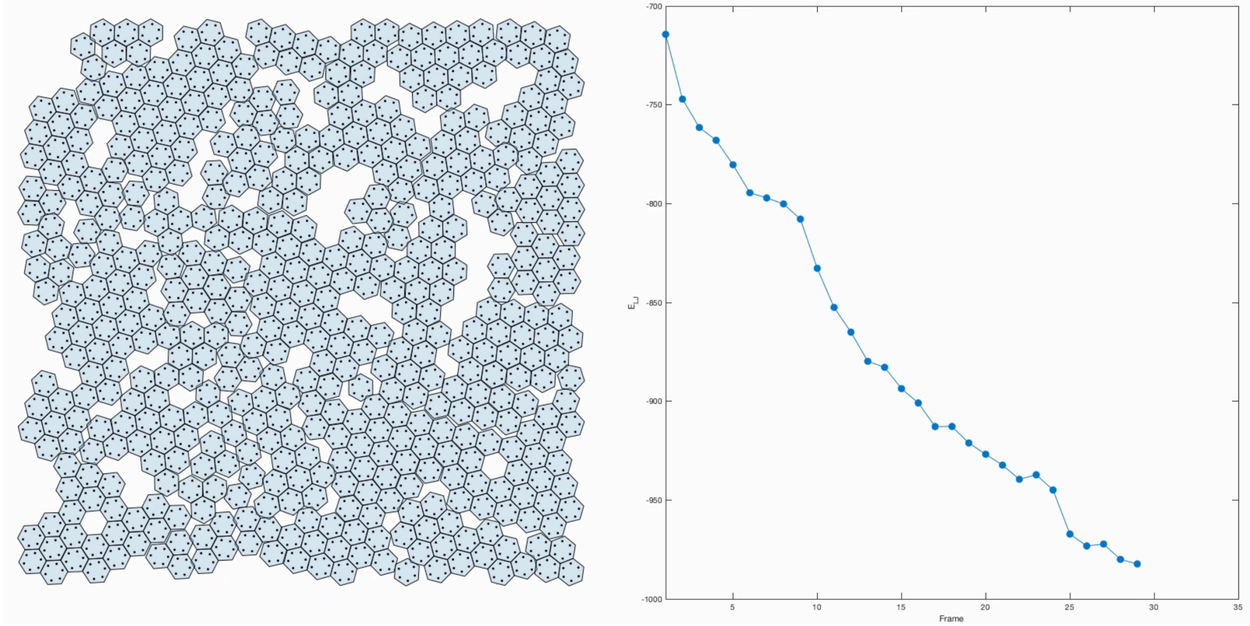

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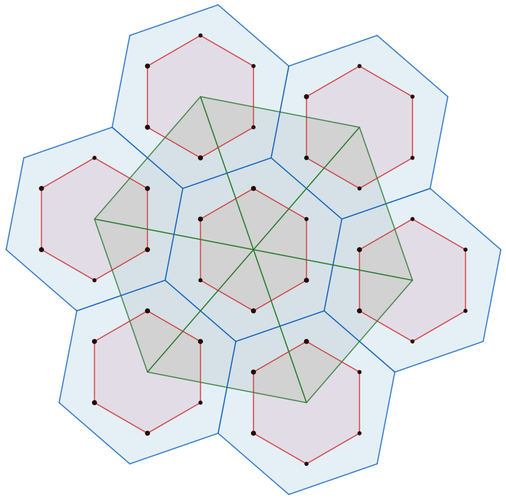

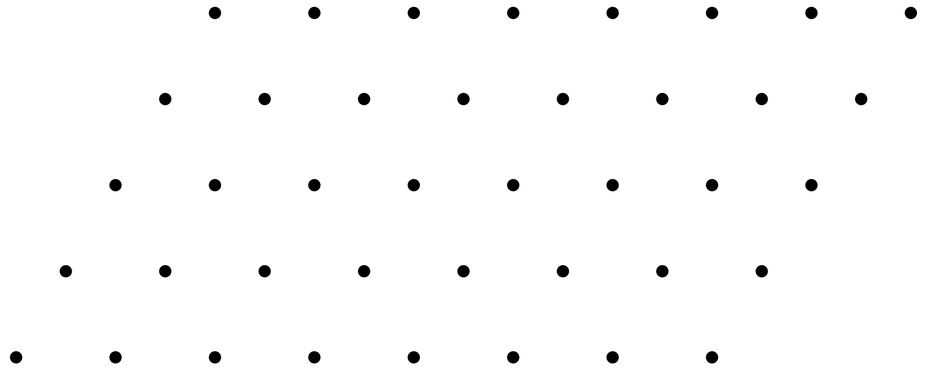

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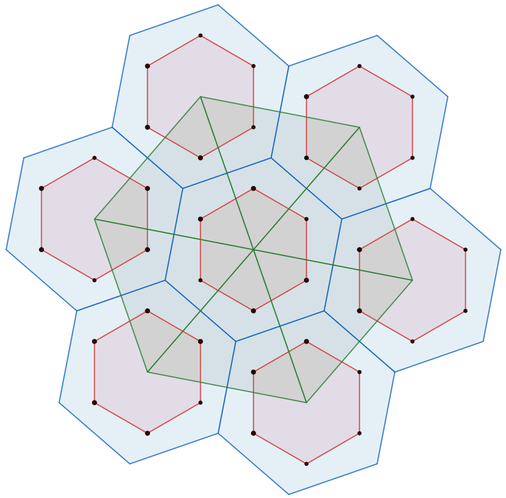

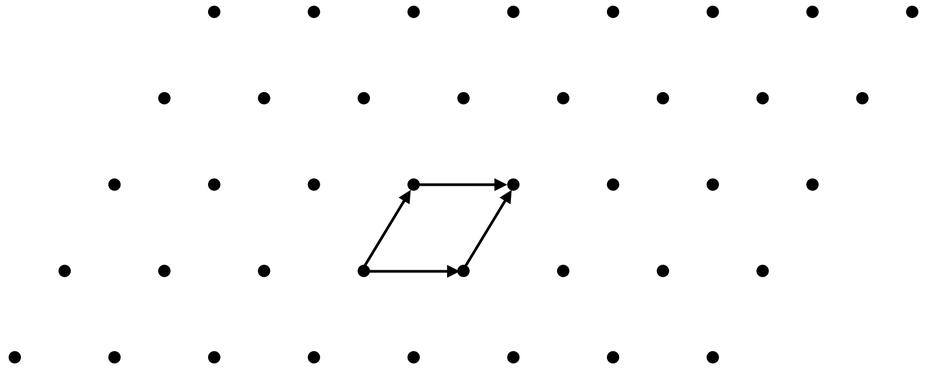

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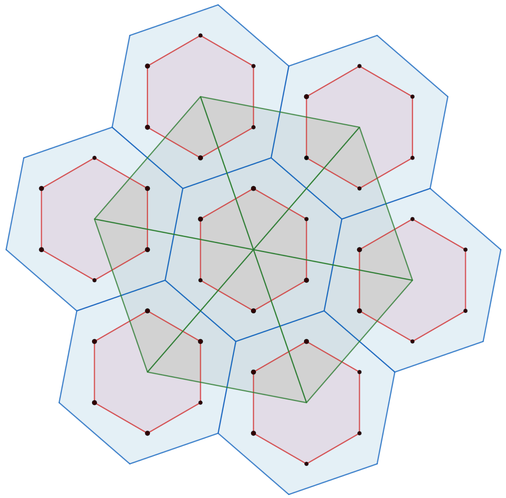

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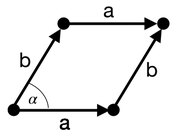

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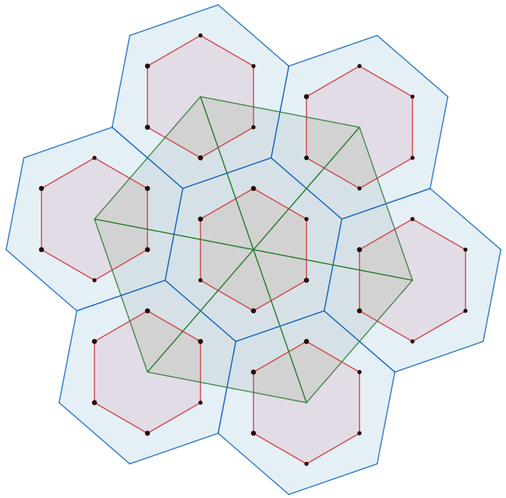

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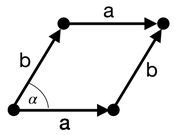

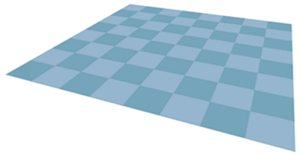

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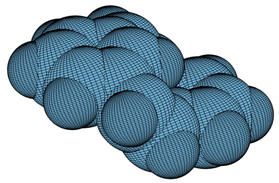
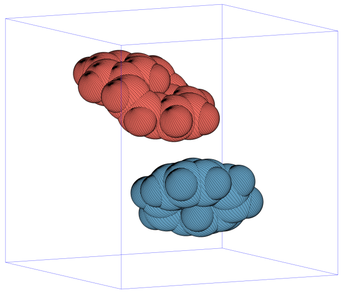
As usual, things are more complicated.

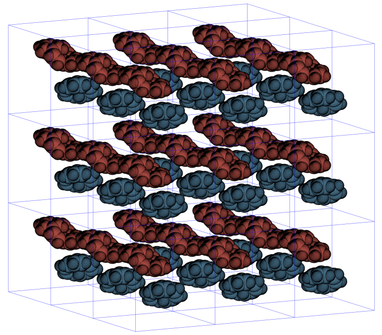
Close - Packing Principle

More symmetry than a simple lattice translation

Molecule as a collection of Van der Waals spheres







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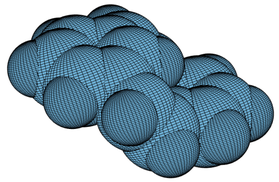

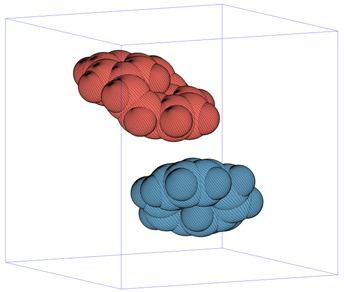

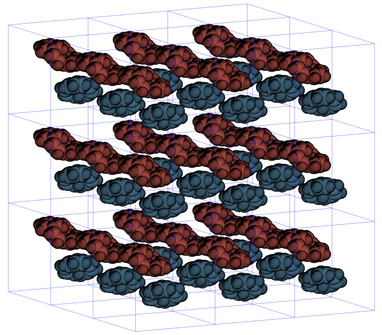

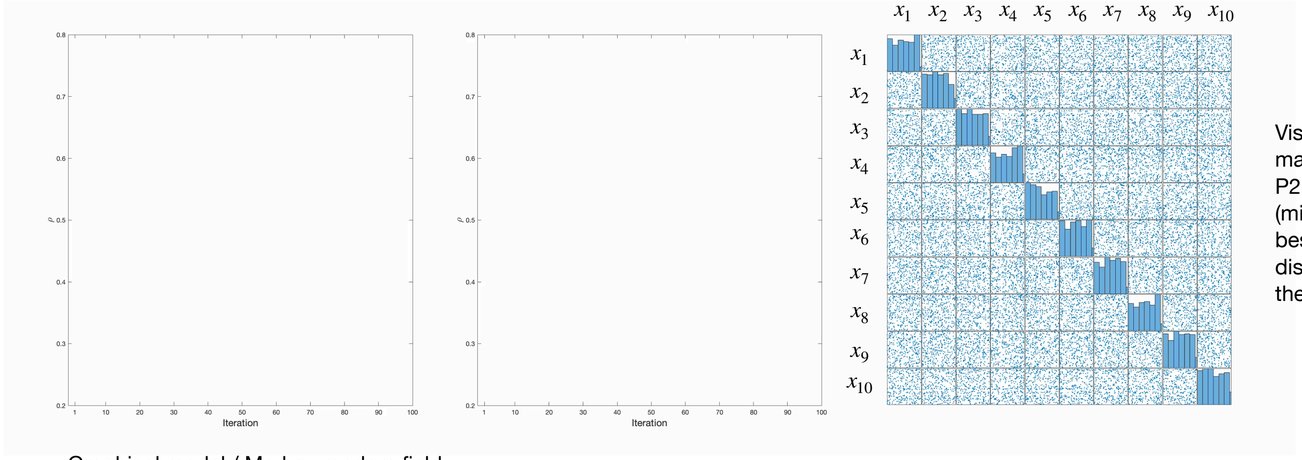

Visualization of the MCYPDE packing maximization run for the space group $P2_1$ (Left) Maximum packing density; (middle) average packing density of N-best packings at $i-th$ iteration; (right) distribution of the packing generation at the iteration.

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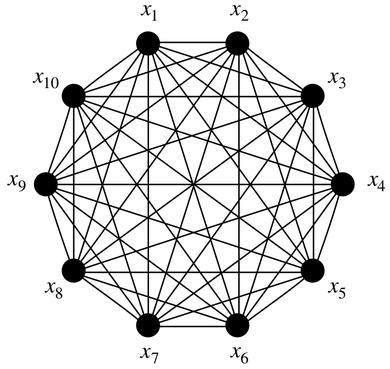

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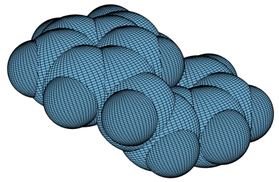

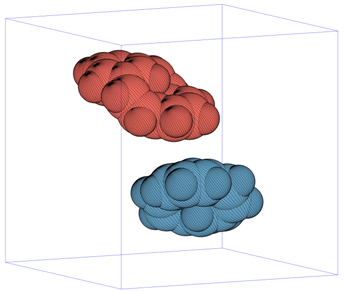

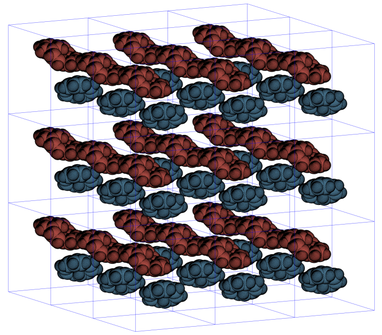

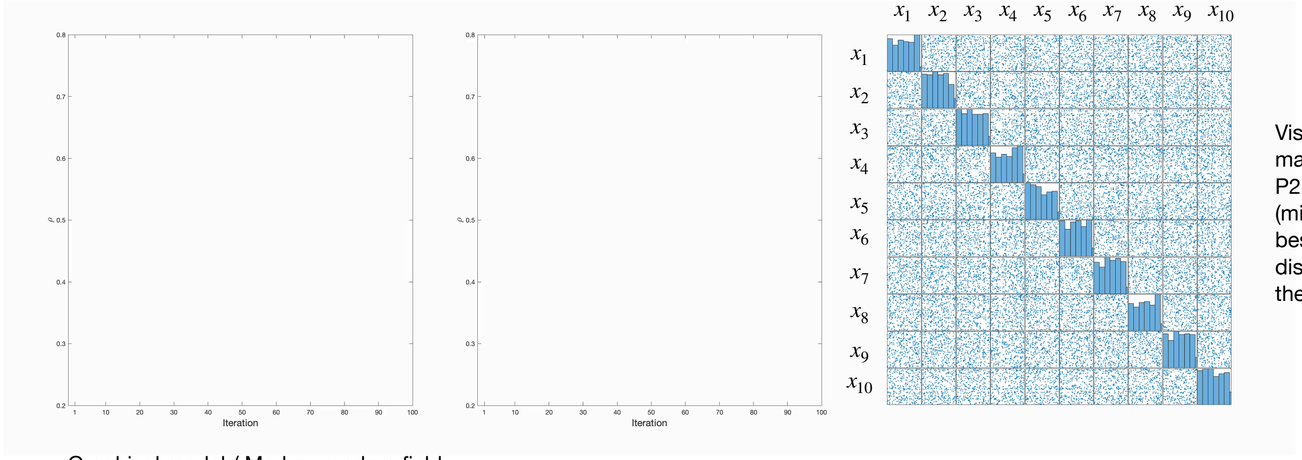

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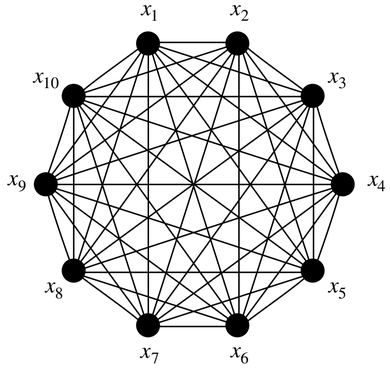

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Each node exchanges its belief about the expected value through branches.

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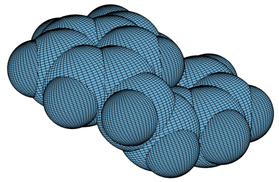

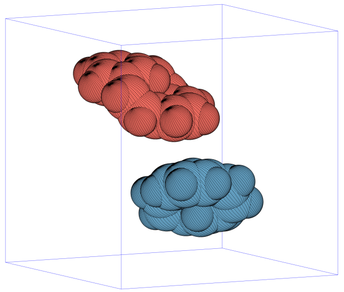

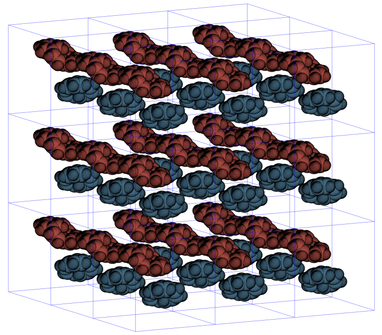

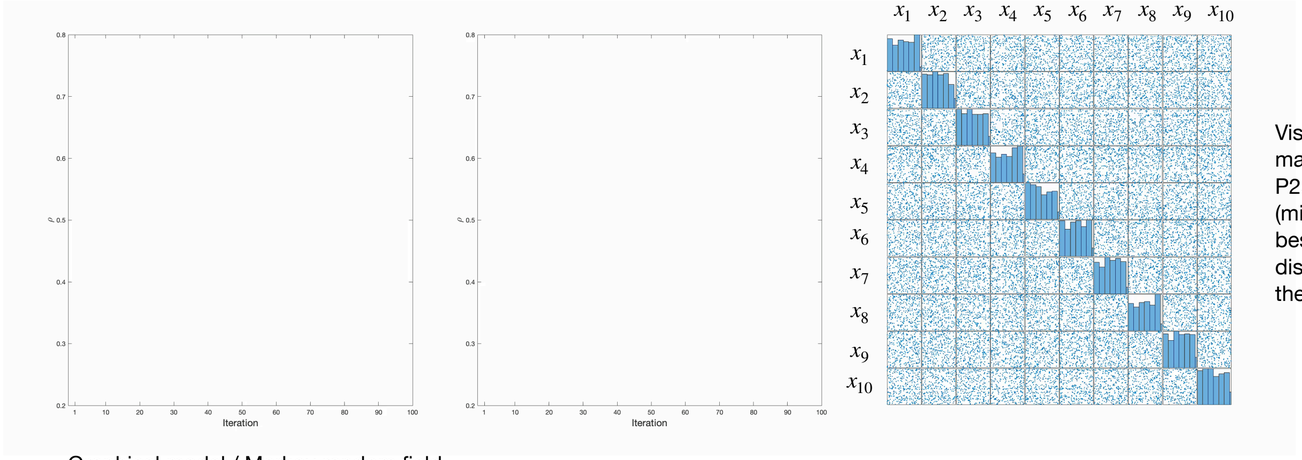

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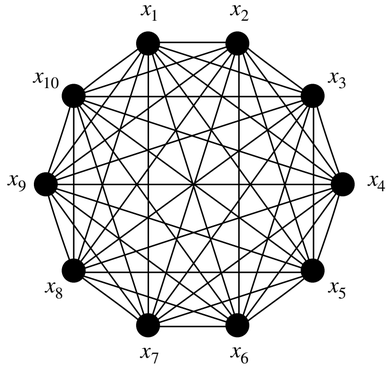

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Maximization of Multi-Information/Total Correlation

- A measure of stochastic dependence in complex systems.
- Generalization of the **Infomax Principle:**
- A rule for training Artificial Neural Networks.
- The algorithm learns the optimization landscape given by the molecular Close-Packing.

$$\max_{\{f \in \mathcal{F} \mid O = f(I)\}} MI(I;O)$$

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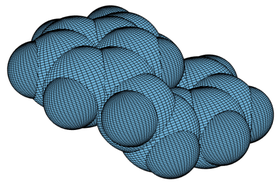

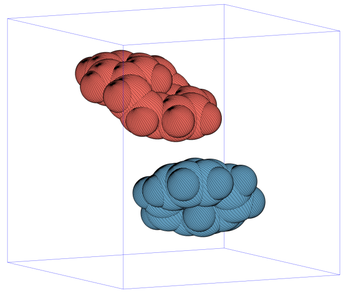

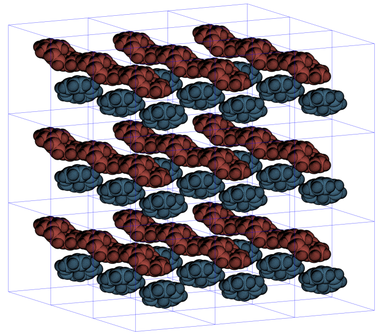

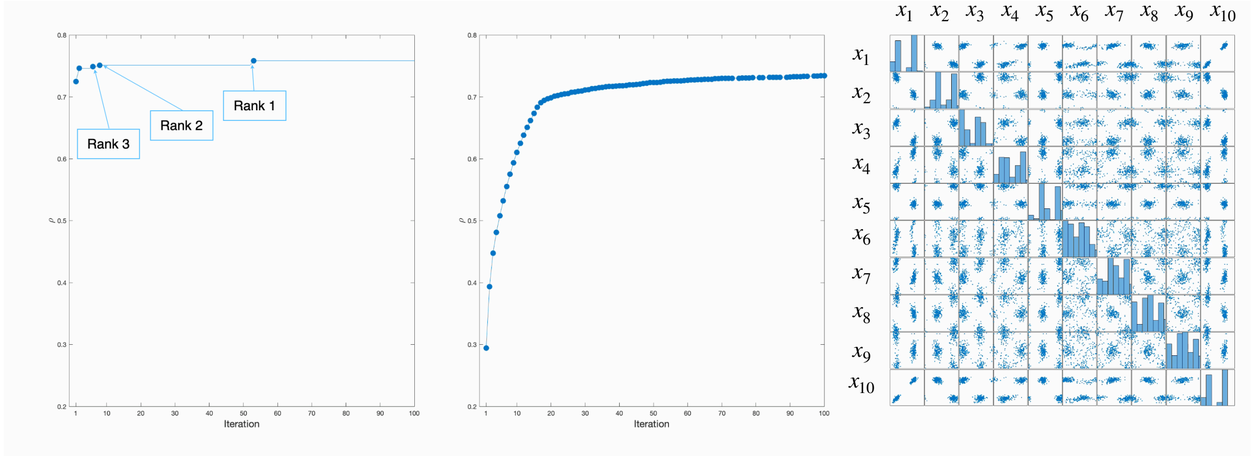

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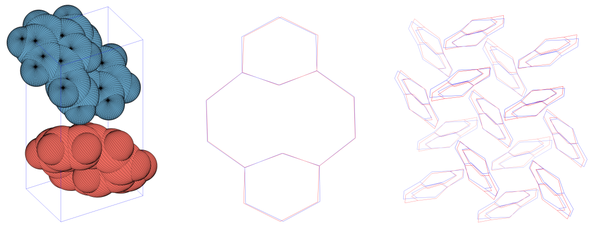
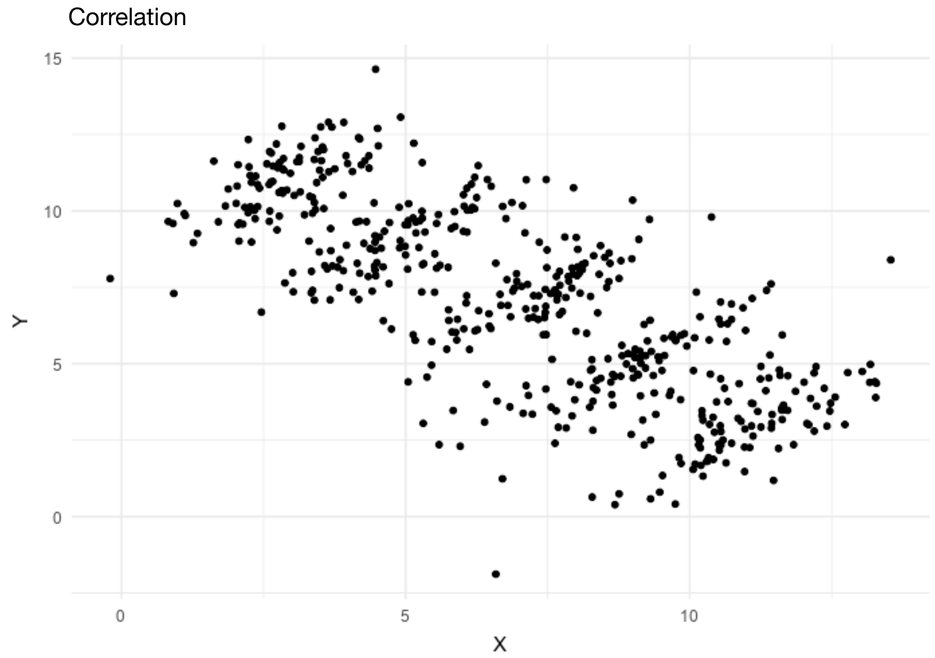
Reference	Comparison	Molecules in Common	RMS	ρ
Ground state	Rank 1	30 out of 30	0.247	0.7584
Ground state	Rank 2	8 out of 30	1.790	0.7514
Ground state	Rank 3	10 out of 30	0.756	0.7510



Visualization of the packing match to the CSP lowest energy structure (Left) A single unit cell displayed in a space-filling representation, where colors indicate symmetry operations modulo lattice translations. (Middle and right) Overlay in a wireframe representation: the global energy minimum (blue) and the matching packing (red). (Middle) Asymmetric unit of the configuration; (right) a 15-molecule cluster.

3. Closing remarks.

Simpson’s paradox



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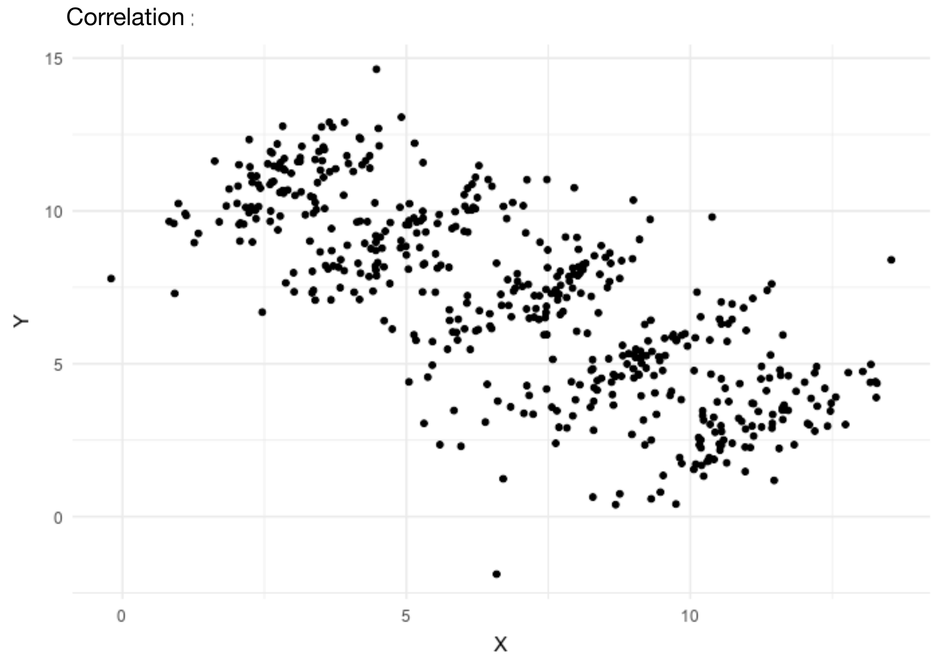
3. Closing remarks.

Simpson’s paradox



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Correlation does not imply causation.

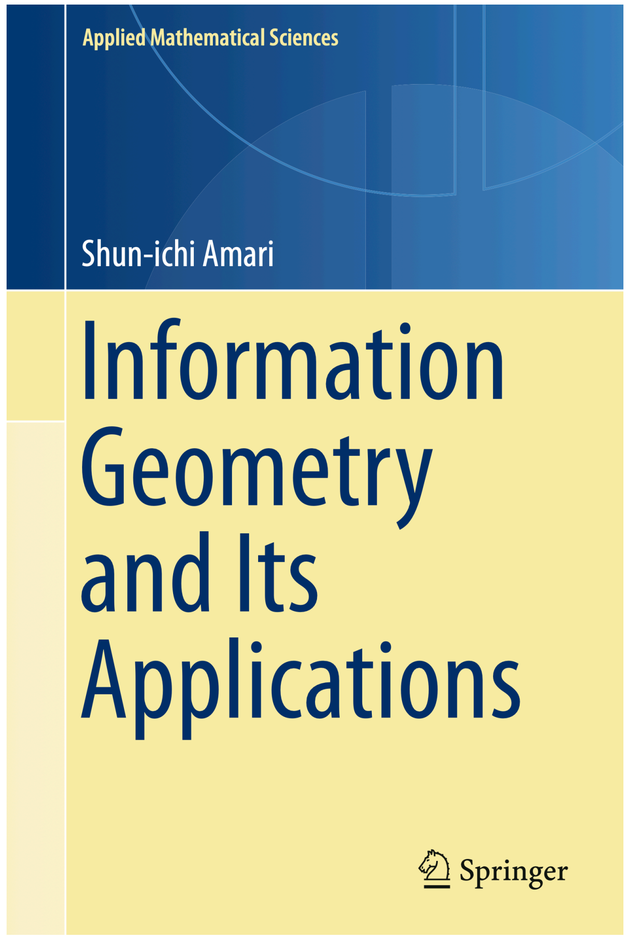
3. Closing remarks.

No free lunch theorem

"state[s] that any two optimization algorithms are equivalent when their performance is averaged across all possible problems”.

Wolpert, D.H.; Macready, W.G. (2005). "Coevolutionary Free Lunches". IEEE Transactions on Evolutionary Computation. 9 (6): 721–735.

3. Closing remarks.



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