

BRIDGING QUANTUM MECHANICS AND MOLECULAR DYNAMICS WITH ARTIFICIAL NEURAL NETWORKS

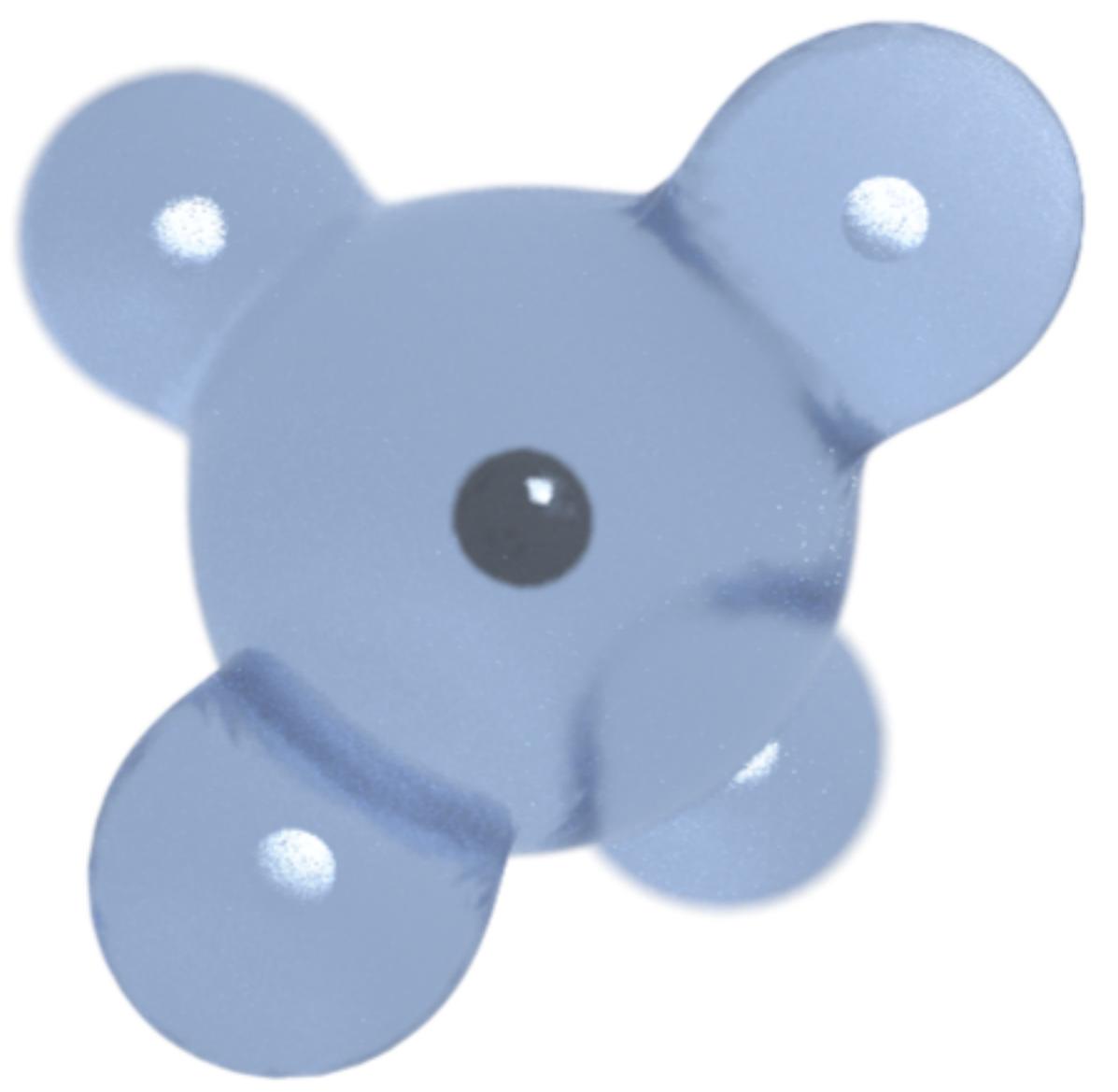
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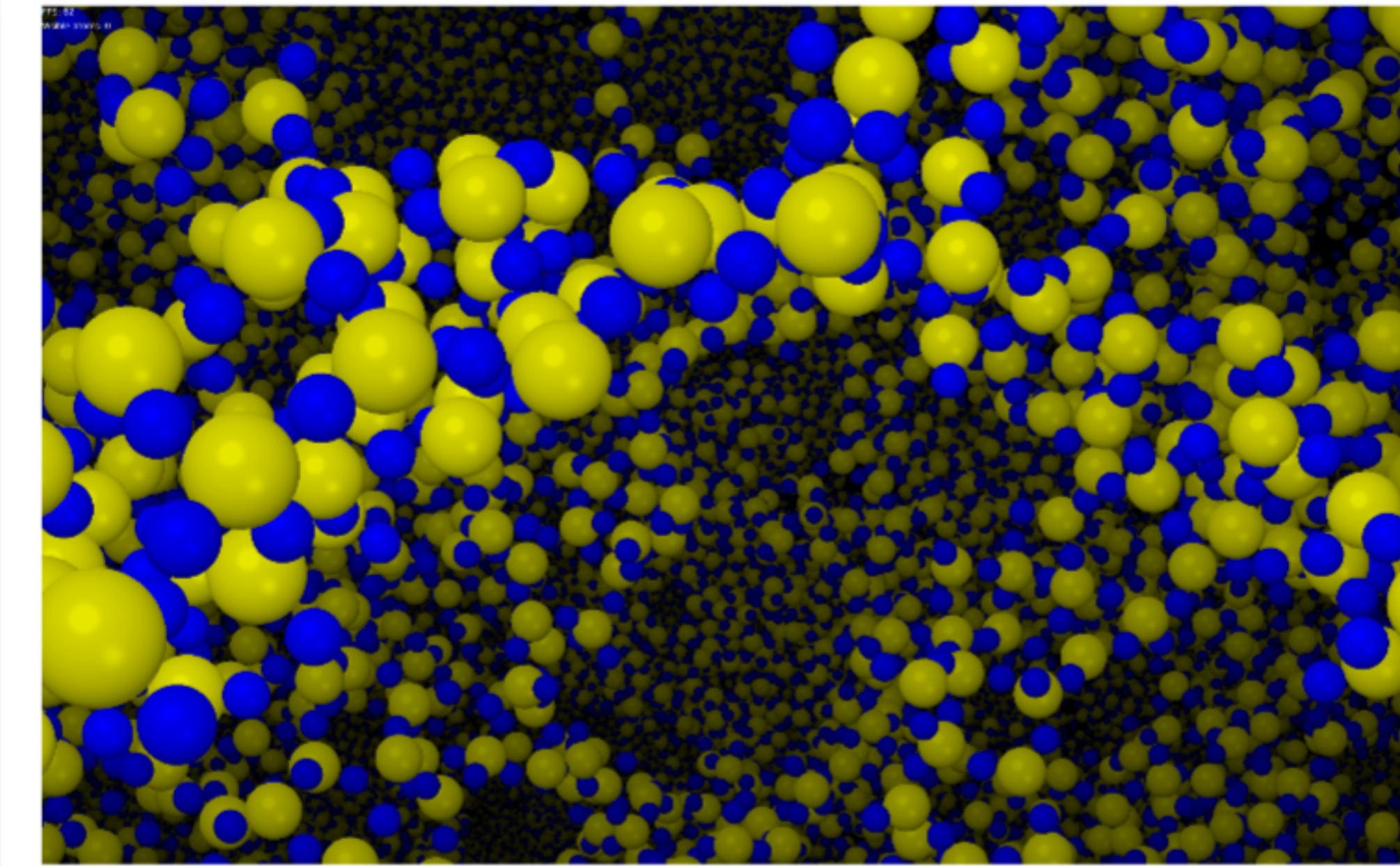
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Quantum Mechanics



energy,
force,
electron density

Molecular Dynamics



temperature,
pressure,
permeability

Problem

detailed calculations = expensive
many atoms = expensive

Goal

Build a bridge between the two scales.
Maximize both quality and quantity.

How?

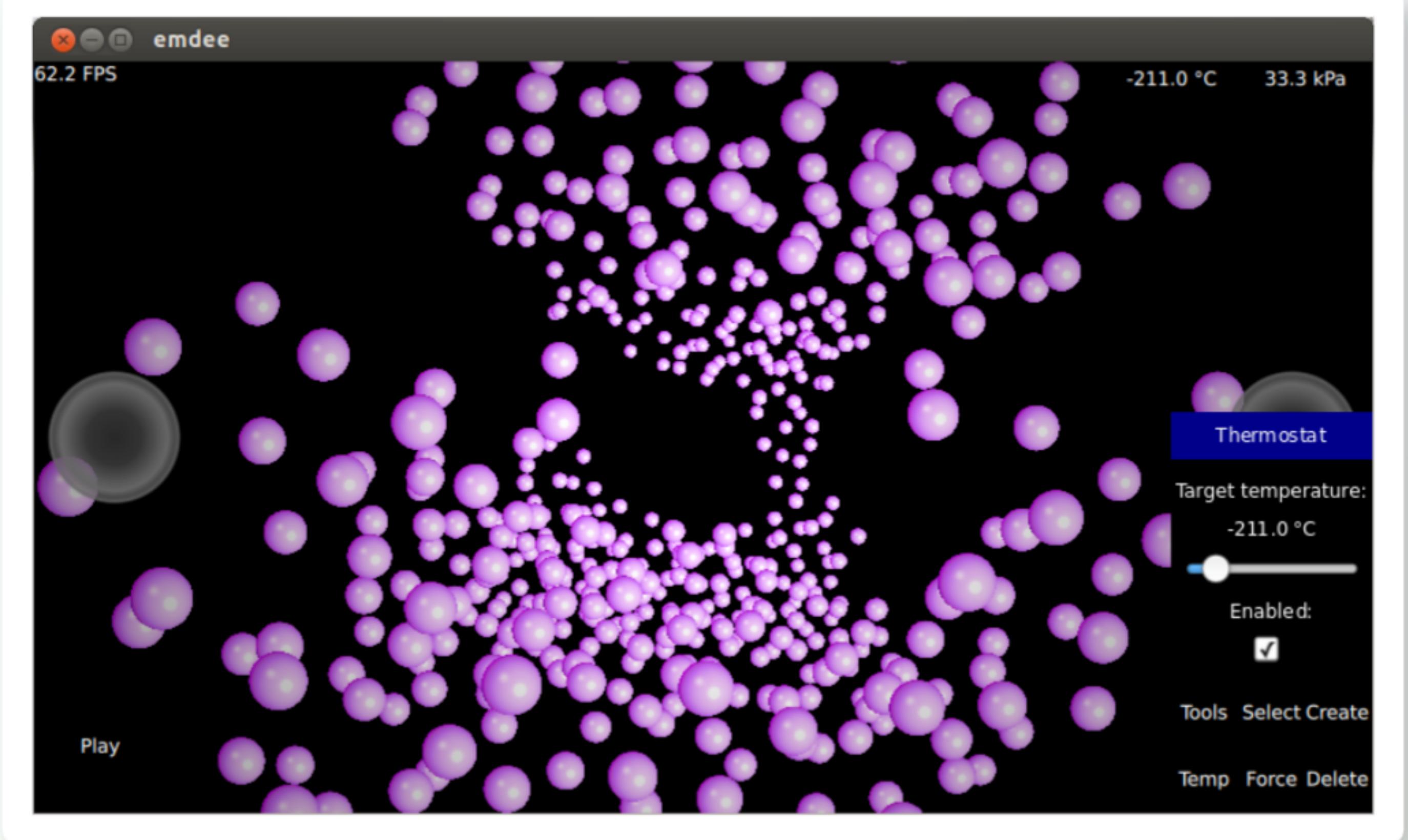
details + many atoms = unaffordable

Machine learning
with artificial neural networks.

MOLECULAR DYNAMICS

Molecular Dynamics

Classical potentials
and newtonian mechanics

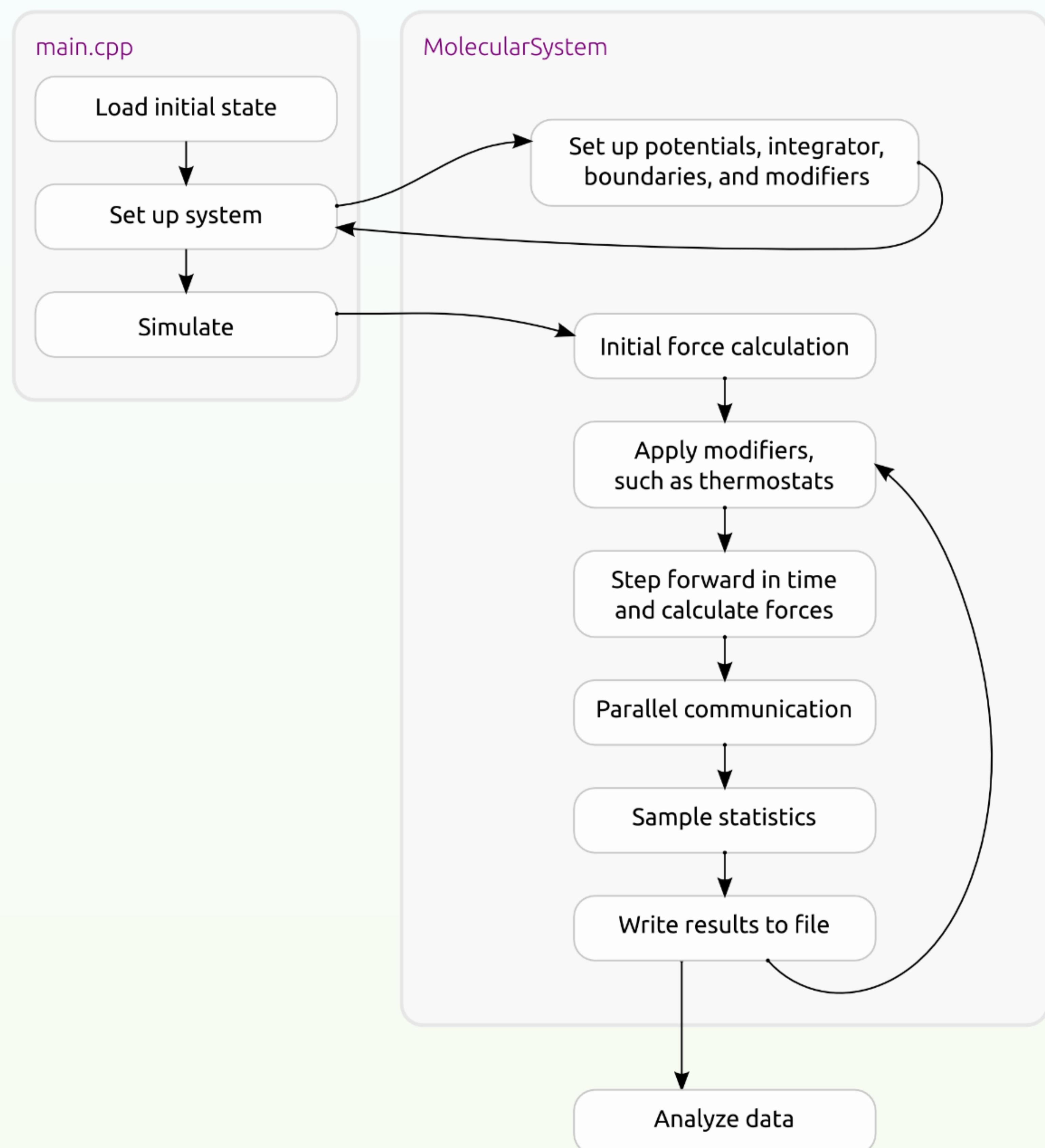


Atoms defined by
position, velocity and type.
Step-wise time-integration.
Forces from potential:

$$F = -\nabla V$$

Molecular Dynamics Implementation

General code.
Built-in selection of potentials.
Extensible.
Performance-centric.



Classical Potentials

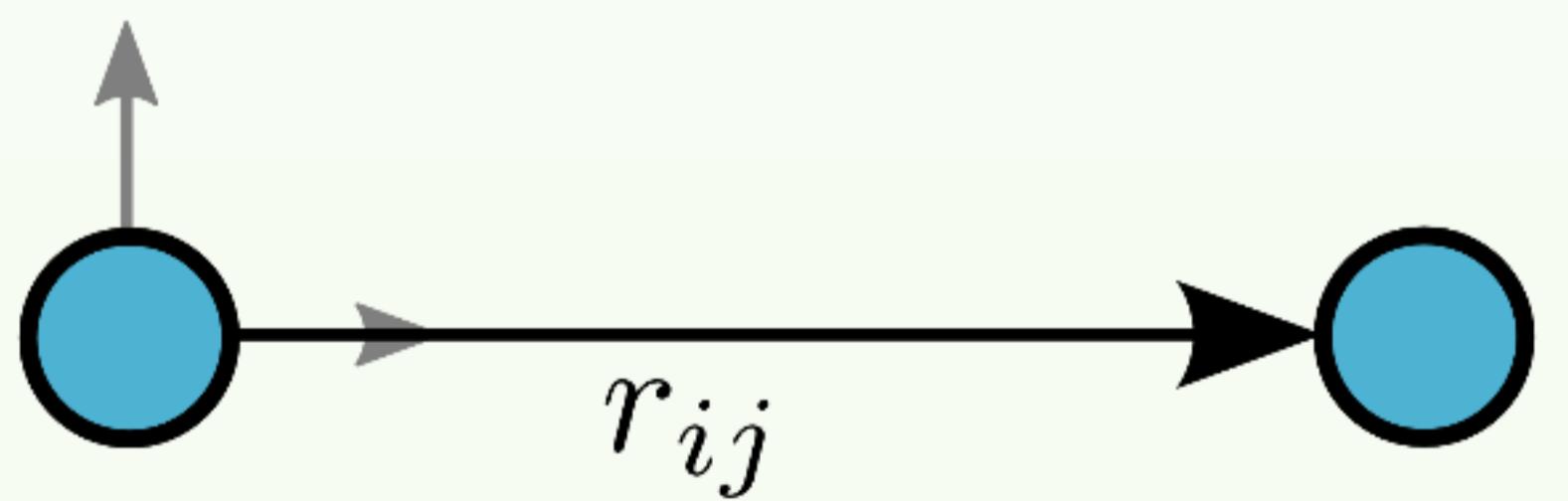
$$V(\mathbf{r}) = \sum_i V_1(\mathbf{r}_i) + \sum_{i < j} V_2(\mathbf{r}_i, \mathbf{r}_j) + \sum_{i < j < k} V_3(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}_k) + \dots$$

How Do We Define a Potential?

From experiments
and/or quantum mechanics

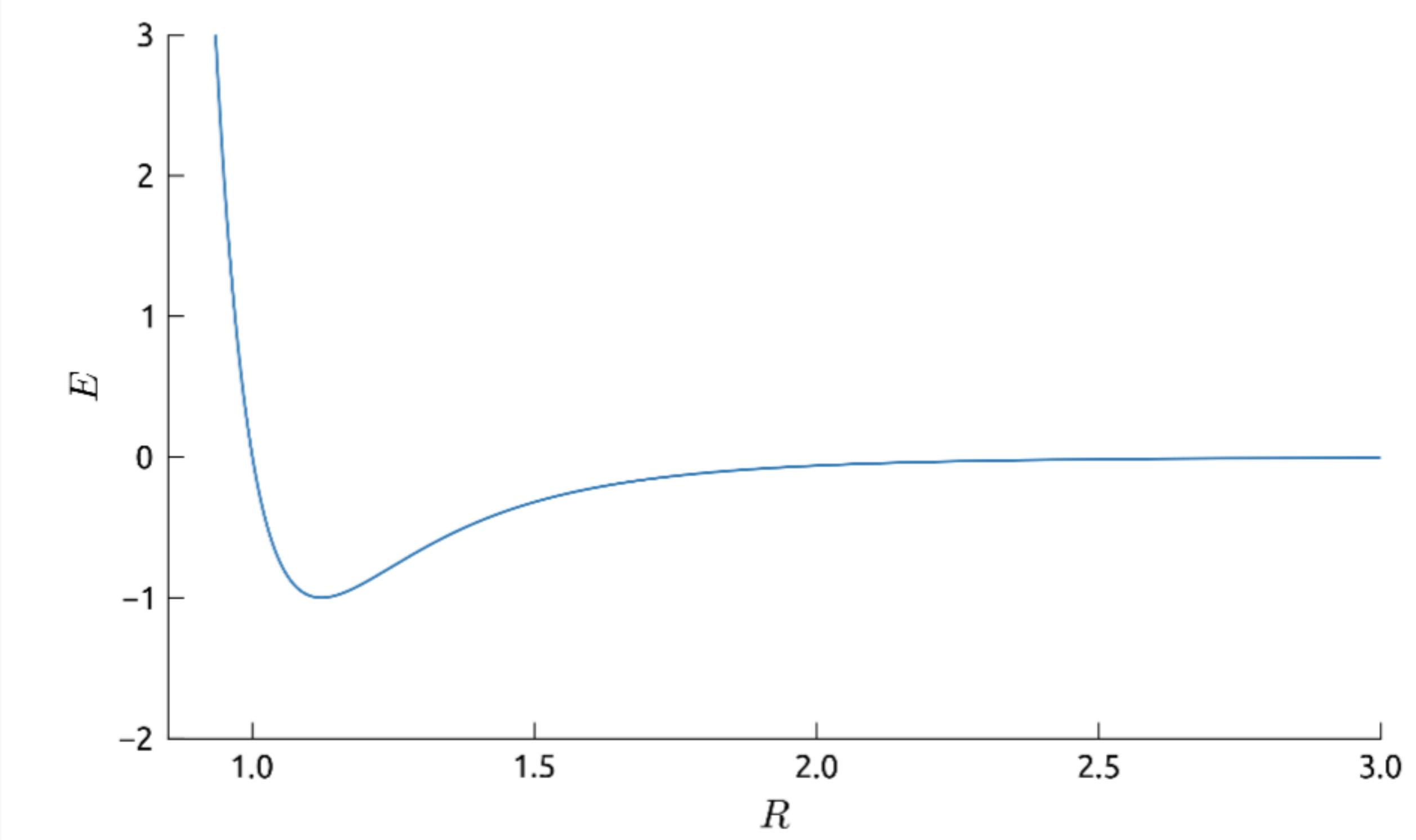
Two-Body Terms

$$V_2(\mathbf{r}_i, \mathbf{r}_j) = V_2(r_{ij})$$



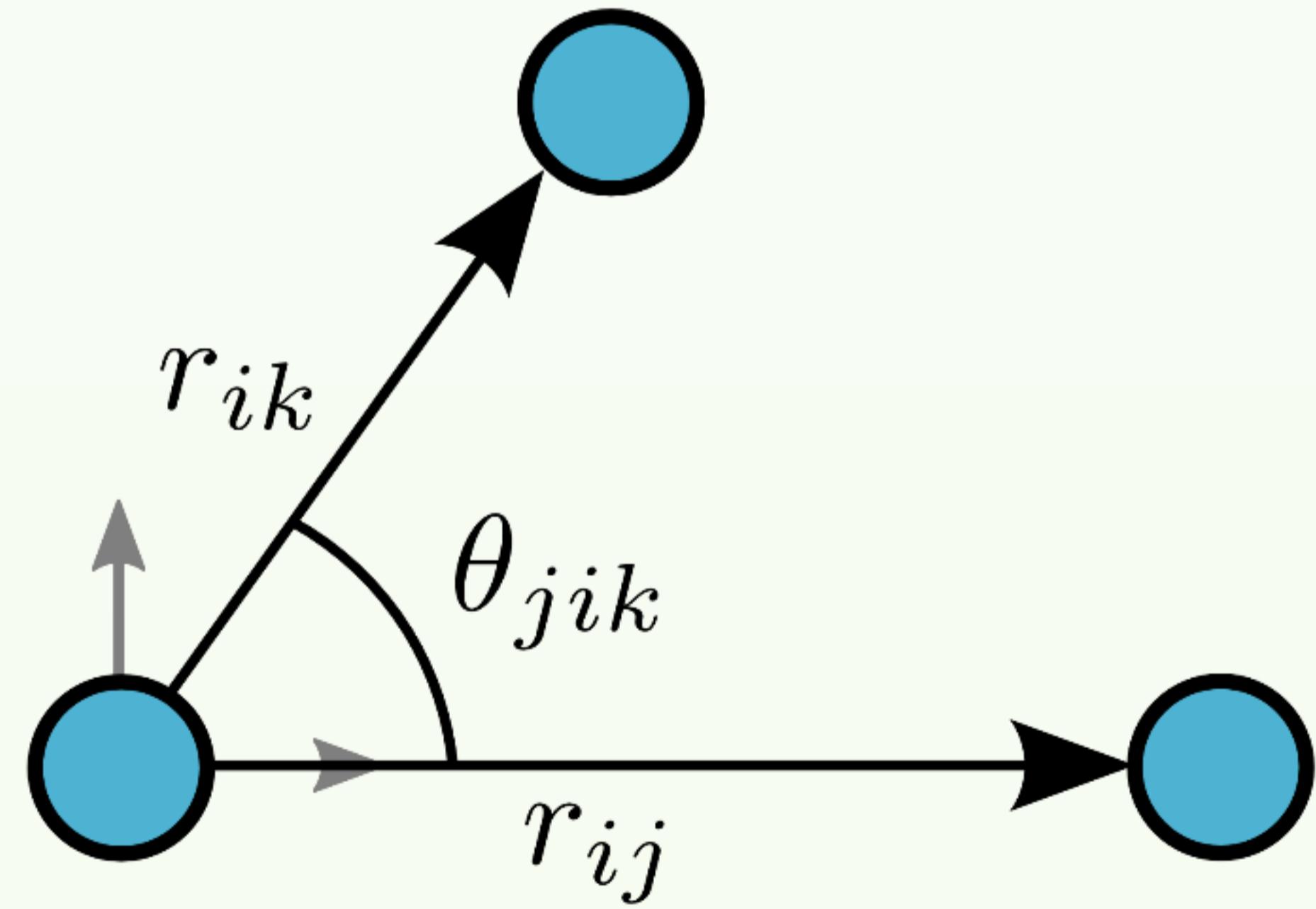
Lennard-Jones

$$V_{\text{LJ}}(r) = 4\epsilon \left(\frac{\sigma^{12}}{r^{12}} - \frac{\sigma^6}{r^6} \right)$$



Three-Body Terms

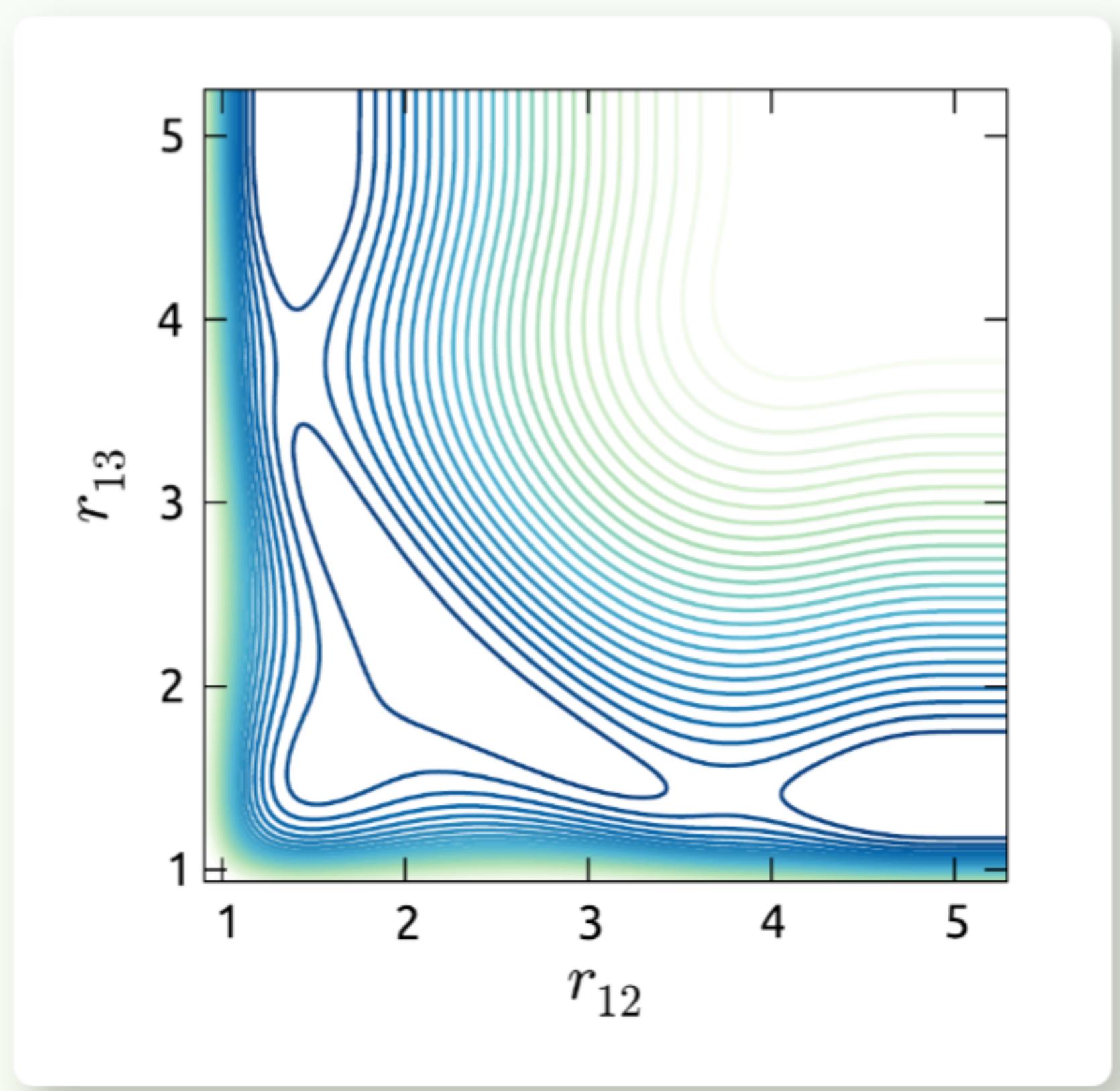
$$V_3(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}_k) = V_3(r_{ij}, r_{ik}, \theta_{jik})$$



Kohen-Tully-Stillinger

$$h(r_{ji}, r_{ik}, \theta_{ijk}) = \begin{cases} \lambda_{ijk} a \exp\left(\frac{\gamma_{ij(k)}^3}{r_{ji} - r_0} + \frac{\gamma_{(i)jk}}{r_{jk} - r_0}\right), & r_{ij}, r_{ik} < r_0 \\ 0 & \text{otherwise.} \end{cases}$$

$$a = (1 + \mu_{ijk} \cos \theta_{ijk} + \nu_{ijk} (\cos \theta_{ijk})^2)$$



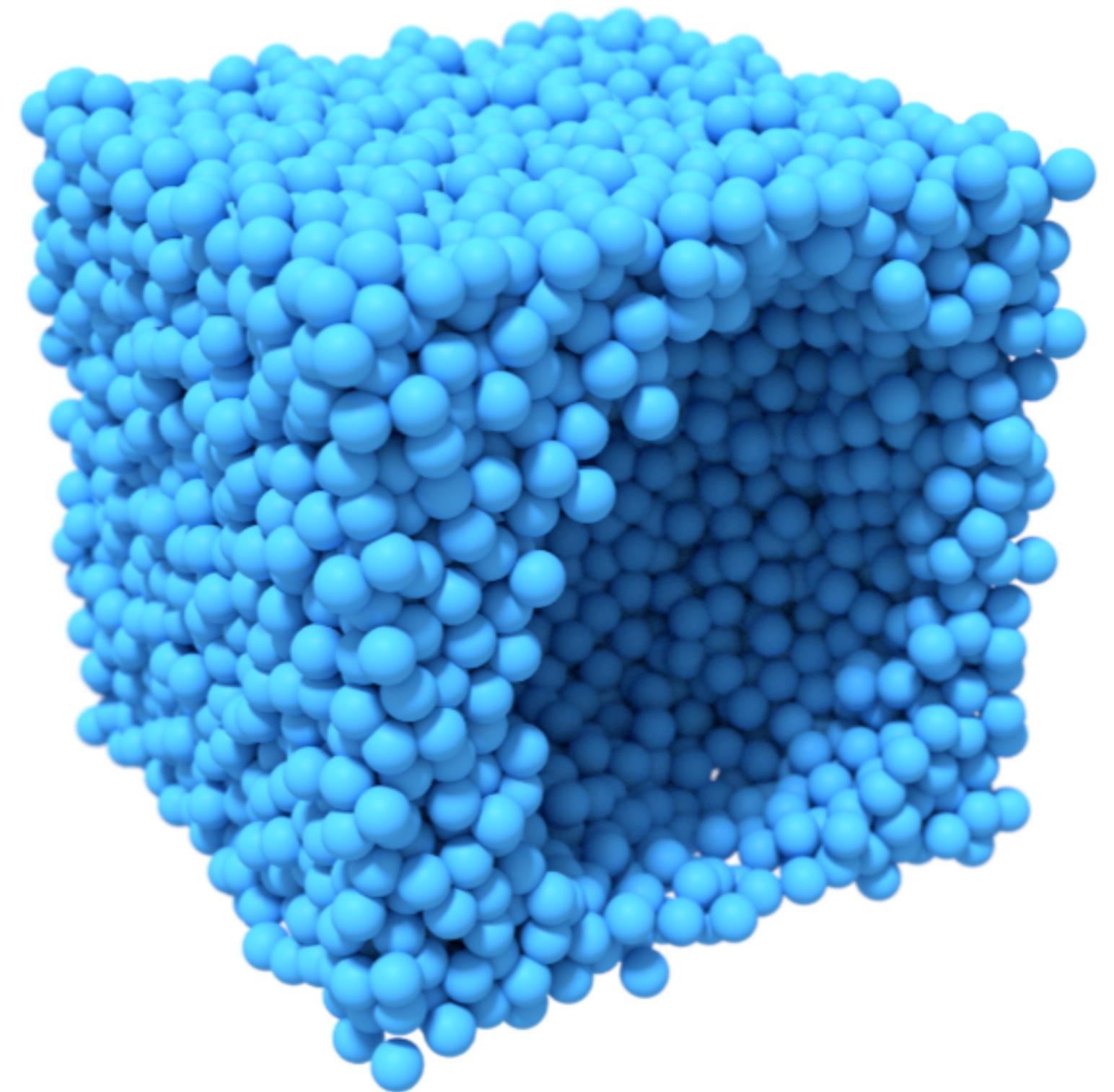
$$V_2 + V_3$$

Advanced Potentials: ReaxFF (9+ terms)

$$V = V_{\text{bond}} + V_{\text{over}} + V_{\text{under}} + V_{\text{penalty}} + V_{\text{valence}} + \\ V_{\text{torsion}} + V_{\text{conj}} + V_{\text{van der Waals}} + V_{\text{Coulomb}}$$

Demo: Argon Crystallization

Boiling argon
cooled slowly.



Lennard-Jones potential.

Phase change at 90K (experimental: 84 K).

COMPUTATIONAL QUANTUM MECHANICS

Computational Quantum Mechanics of Atomic Systems

Solving the Schrödinger equation
for nuclei and electrons.

Schrödinger equation

$$H\Psi = E\Psi$$

Hamiltonian

$$\begin{aligned} H = & - \sum_i^N \frac{1}{2} \nabla_{r_i}^2 - \sum_n^{N_n} \frac{1}{2m_p} \nabla_{R_i}^2 + \sum_{n < m}^{N_n} \frac{Z_n Z_m}{|\mathbf{R}_n - \mathbf{R}_m|} \\ & - \sum_i^N \sum_n^{N_n} \frac{Z_n}{|\mathbf{r}_i - \mathbf{R}_n|} + \sum_{i < j}^N \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|} \end{aligned}$$

Born-Oppenheimer

$$H = -\sum_i^N \frac{1}{2} \nabla_{r_i}^2 - \frac{1}{2} \sum_i^N \sum_n^{N_n} \frac{Z_n}{|\mathbf{r}_i - \mathbf{R}_n|} + \sum_{i < j}^N \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}$$

Nuclei configuration is fixed.
Argued for by mass difference.

Slater Determinant

$$\Psi = \frac{1}{\sqrt{N!}} \begin{vmatrix} \psi_1(\mathbf{x}_1) & \psi_1(\mathbf{x}_2) & \cdots & \psi_1(\mathbf{x}_N) \\ \psi_2(\mathbf{x}_1) & \psi_2(\mathbf{x}_2) & \cdots & \psi_2(\mathbf{x}_N) \\ \vdots & \vdots & \ddots & \vdots \\ \psi_N(\mathbf{x}_1) & \psi_N(\mathbf{x}_2) & \cdots & \psi_N(\mathbf{x}_N) \end{vmatrix}$$

Single-particle wave functions.
No correlations apart from exchange.

Hartree-Fock Method

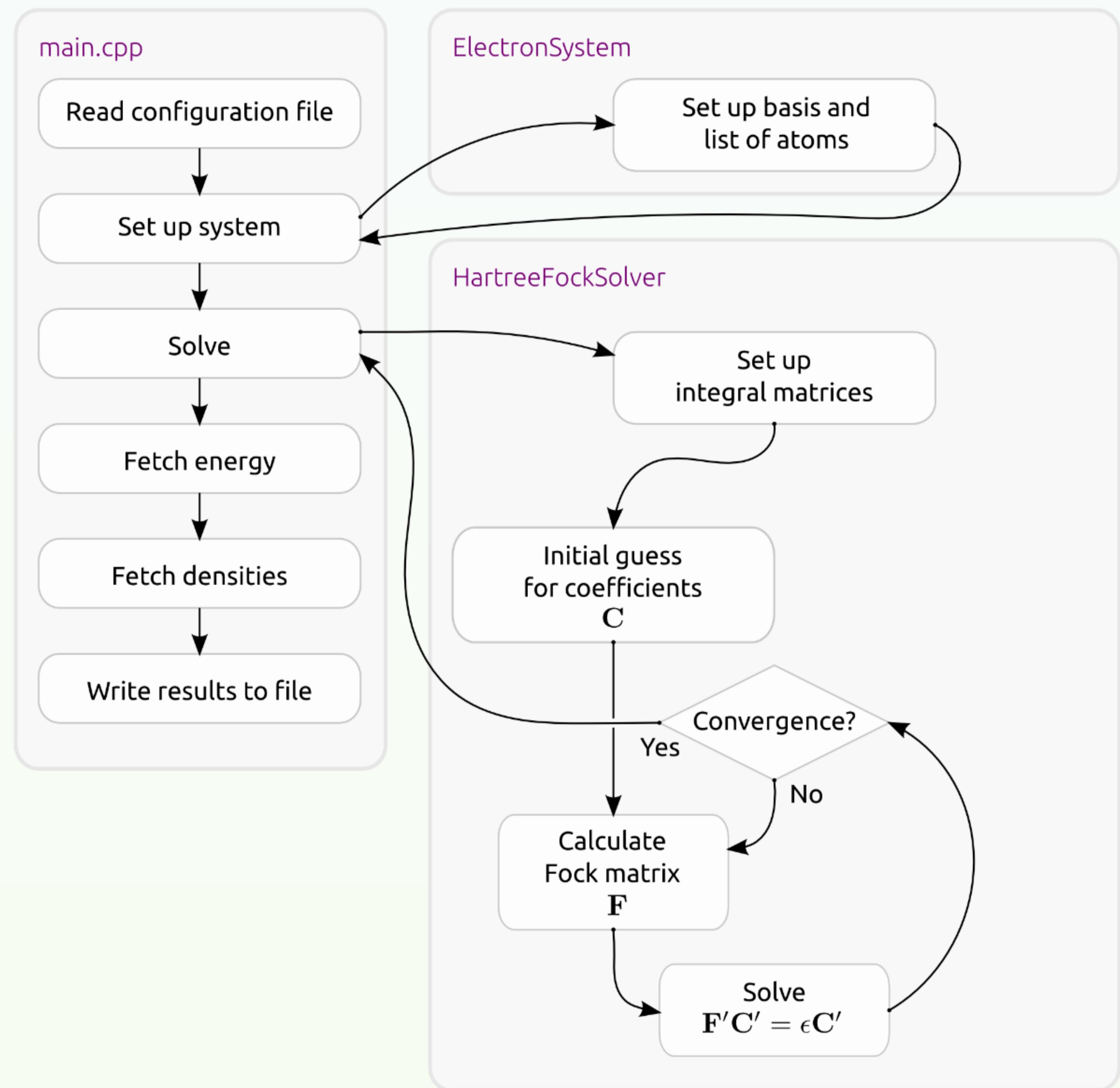
Self-consistent.
Iterative scheme.

Hartree-Fock Equations

$$\mathcal{F}\psi_k = \epsilon_k\psi_k$$

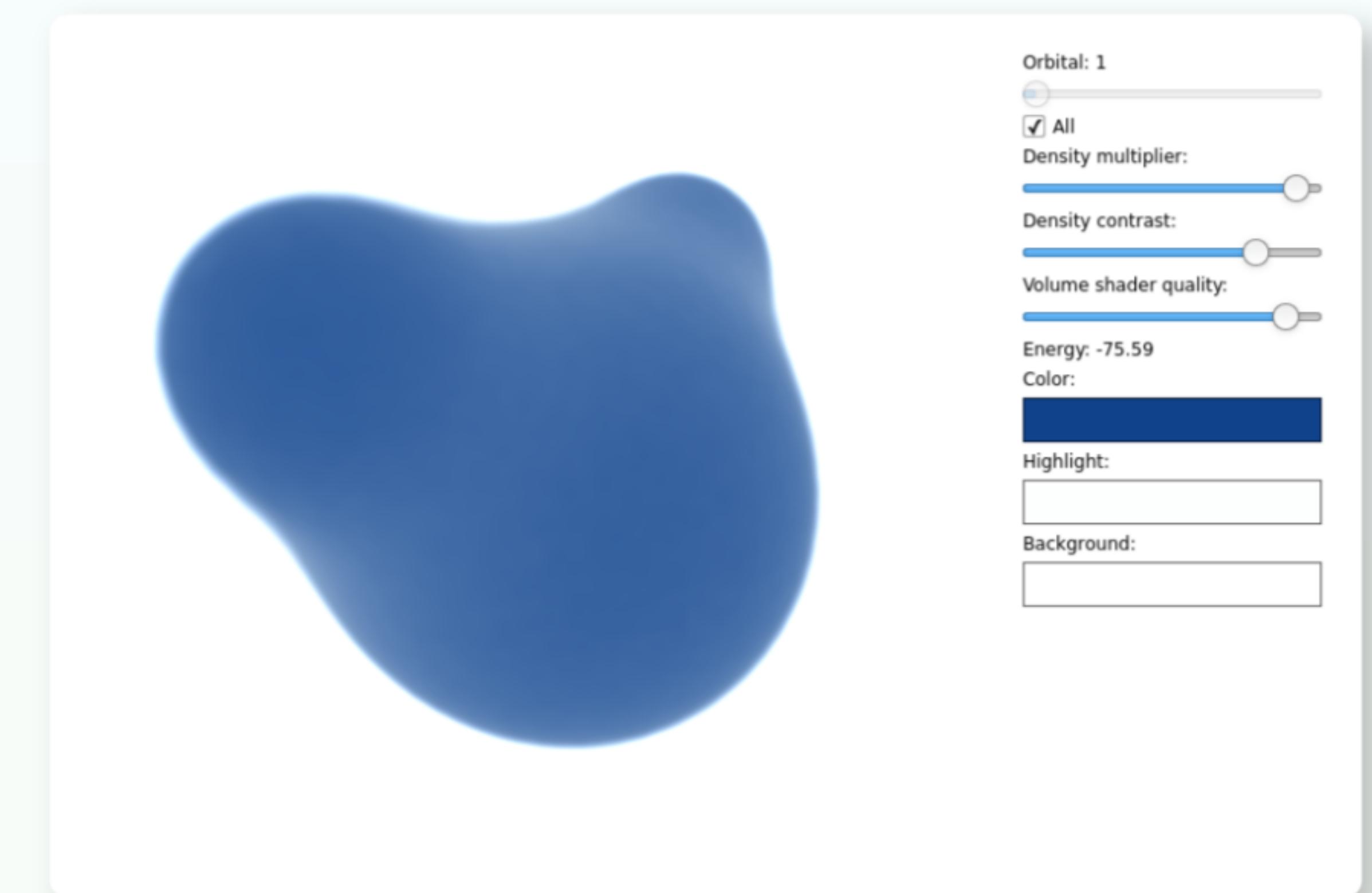
$$\phi_k(\mathbf{r}) = \sum_p^M C_{pk} \varphi_p(\mathbf{r})$$

$$\mathbf{F}\mathbf{C}_k = \epsilon_k \mathbf{S}\mathbf{C}_k$$

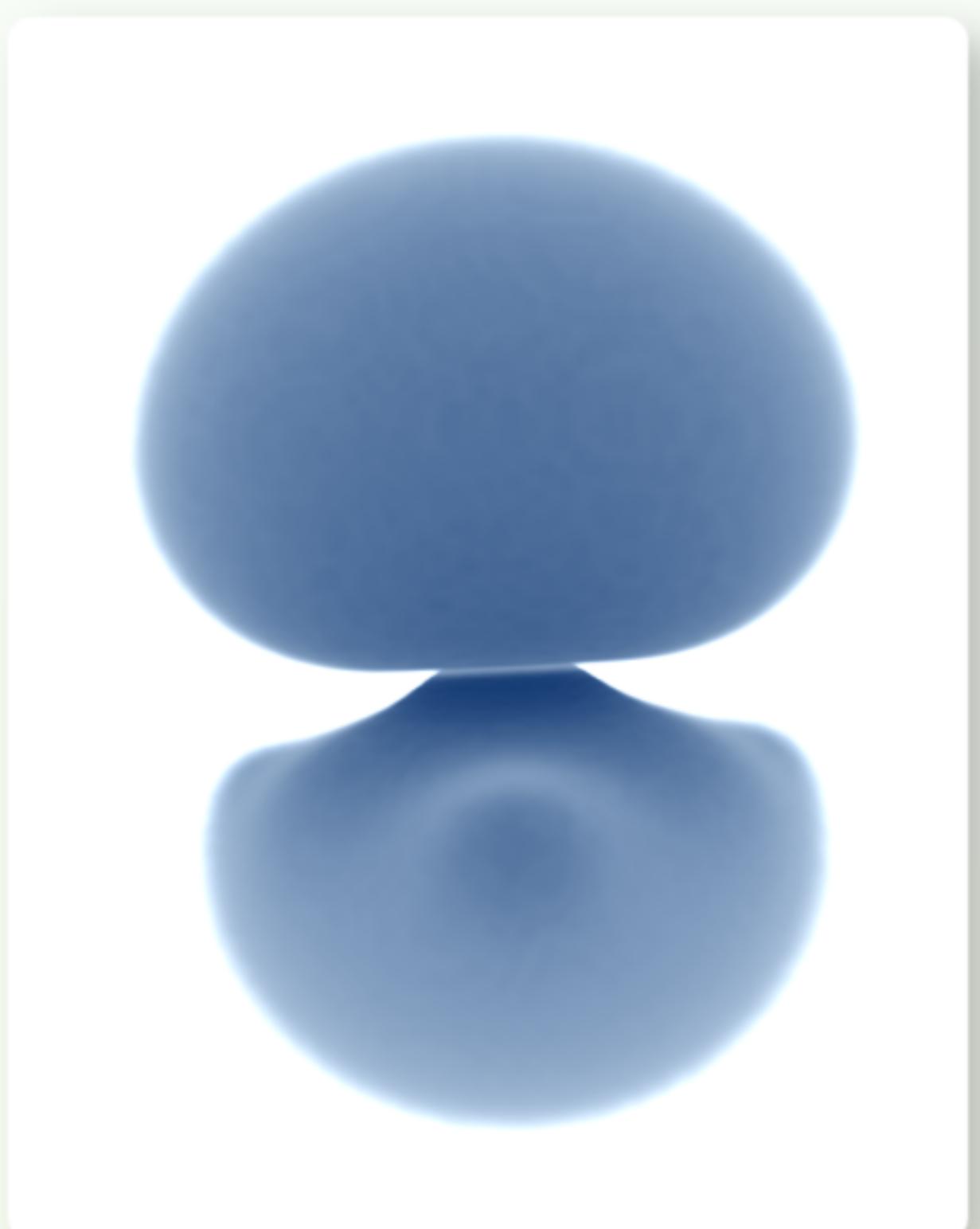


Electron Density

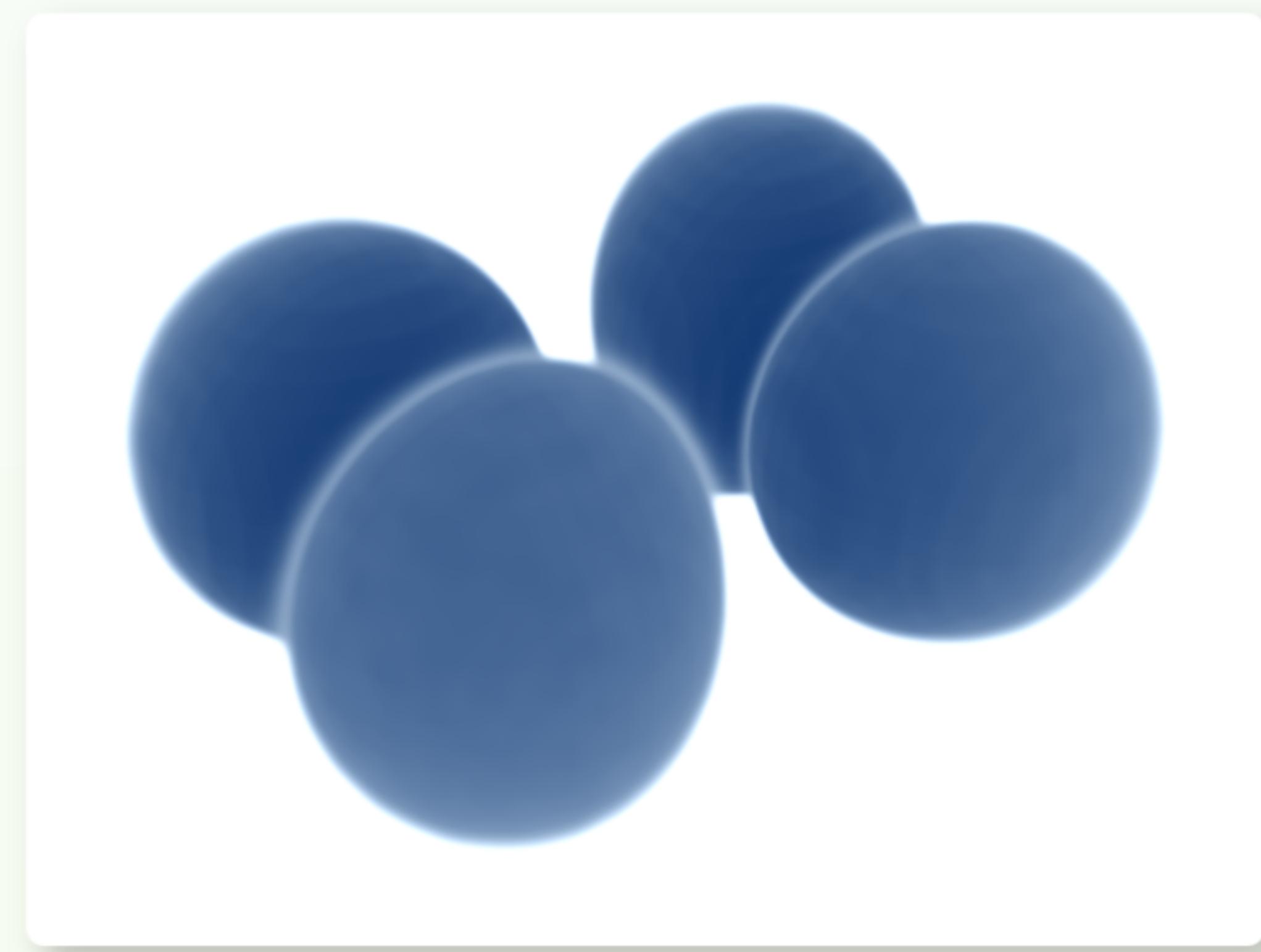
Demo!



H_2O



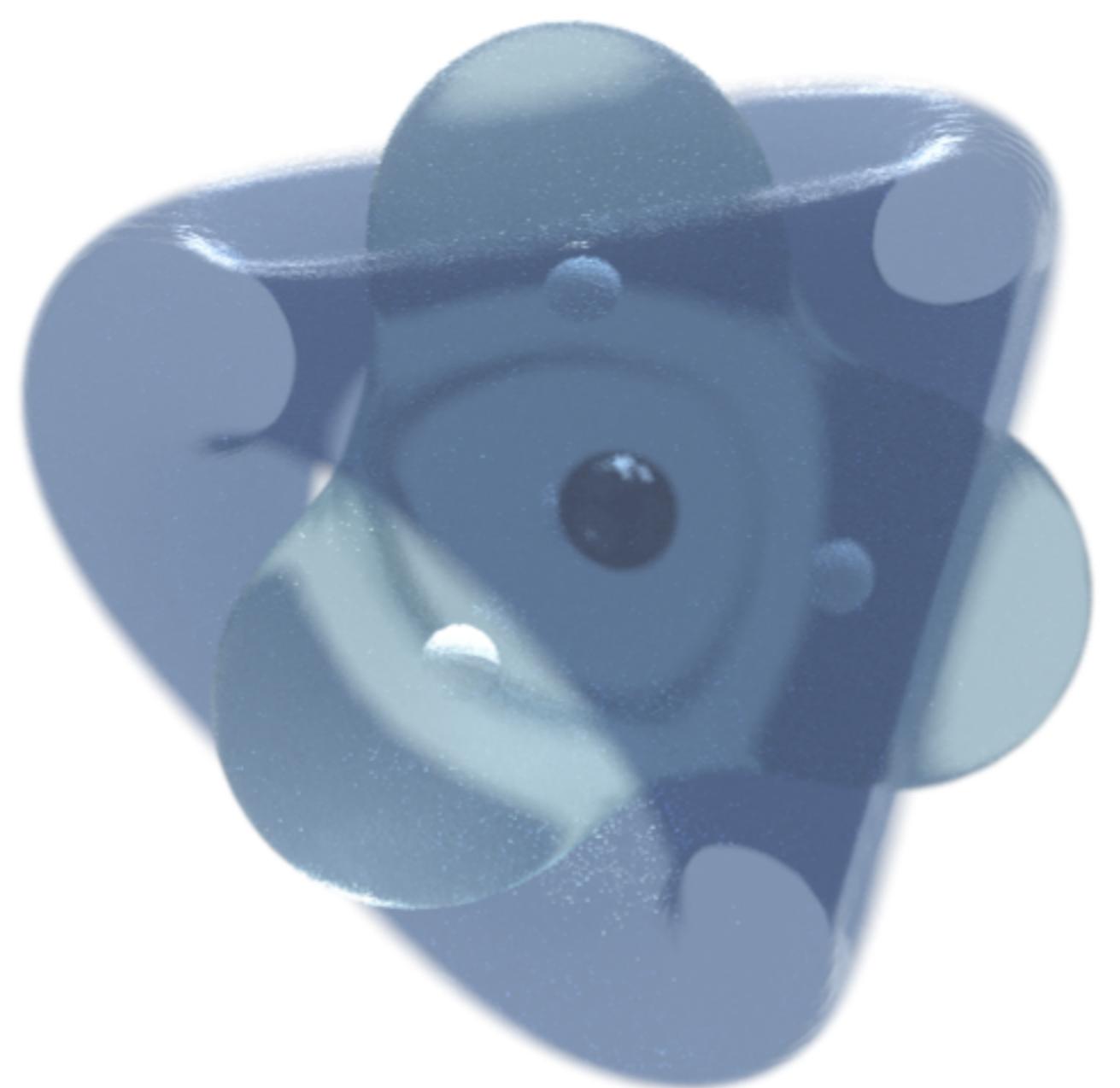
NH_3



O_2

Electrostatic Potential

Shows electro- and nucleophilic areas



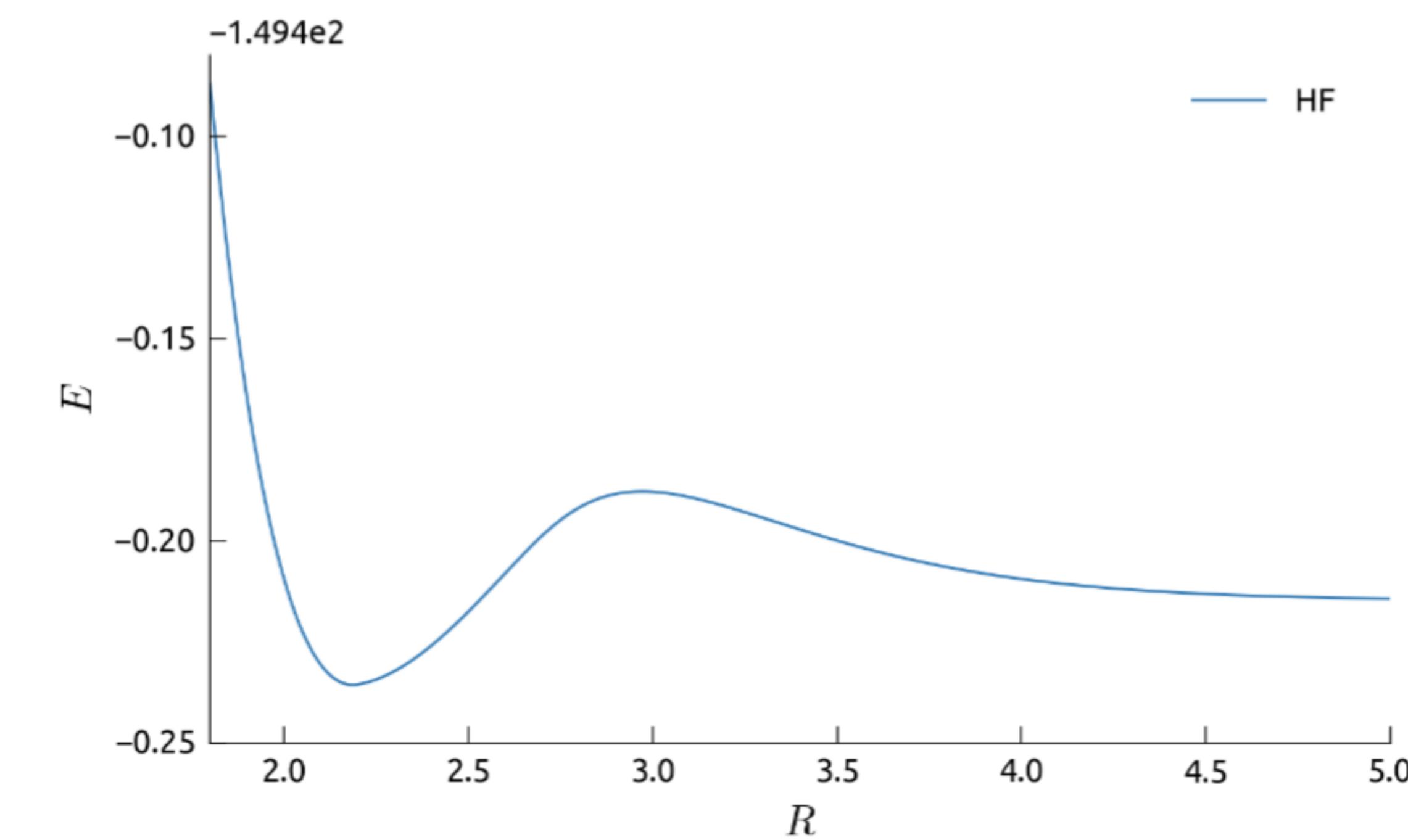
Path to Molecular Dynamics

Electronic energy in quantum mechanics corresponds to potential energy in molecular dynamics.

$$V_{\text{MD}} = E_{\text{QM}}$$

Form of the Potential?

Search and fitting can be laborious.



ARTIFICIAL NEURAL NETWORKS

Neural Networks

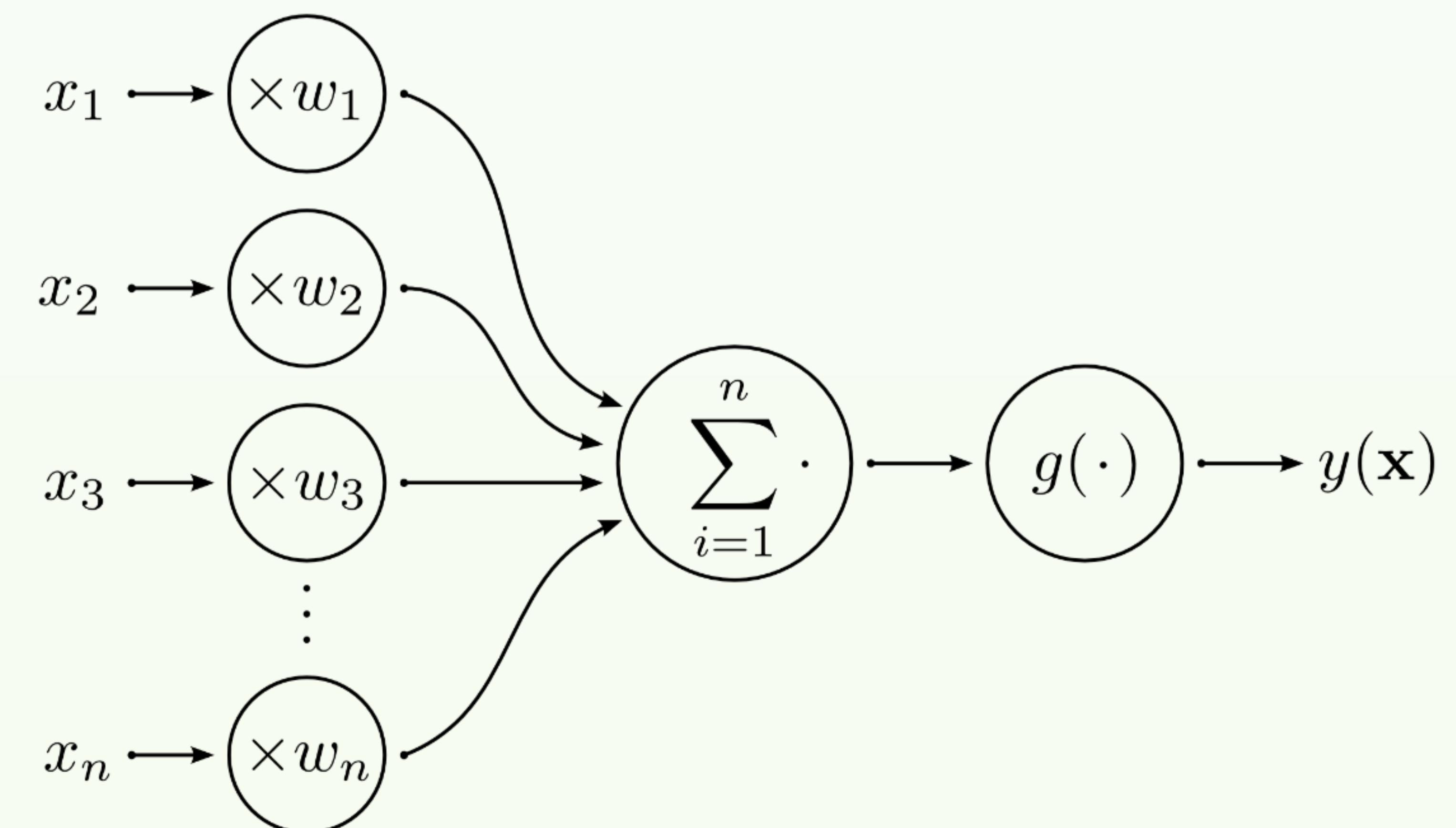
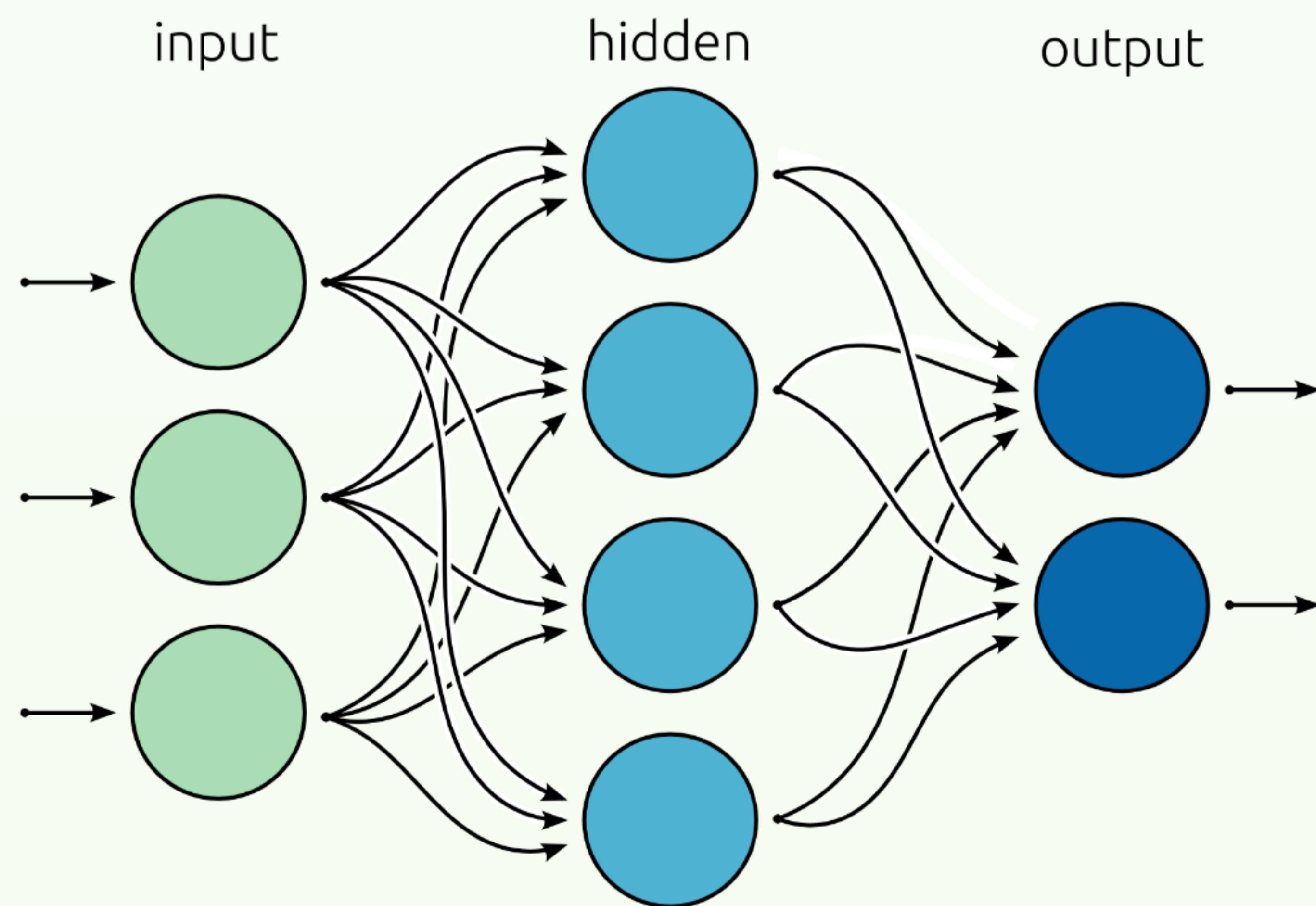
Neuron

Weighted connections between neurons.

Input is summed and evaluated.

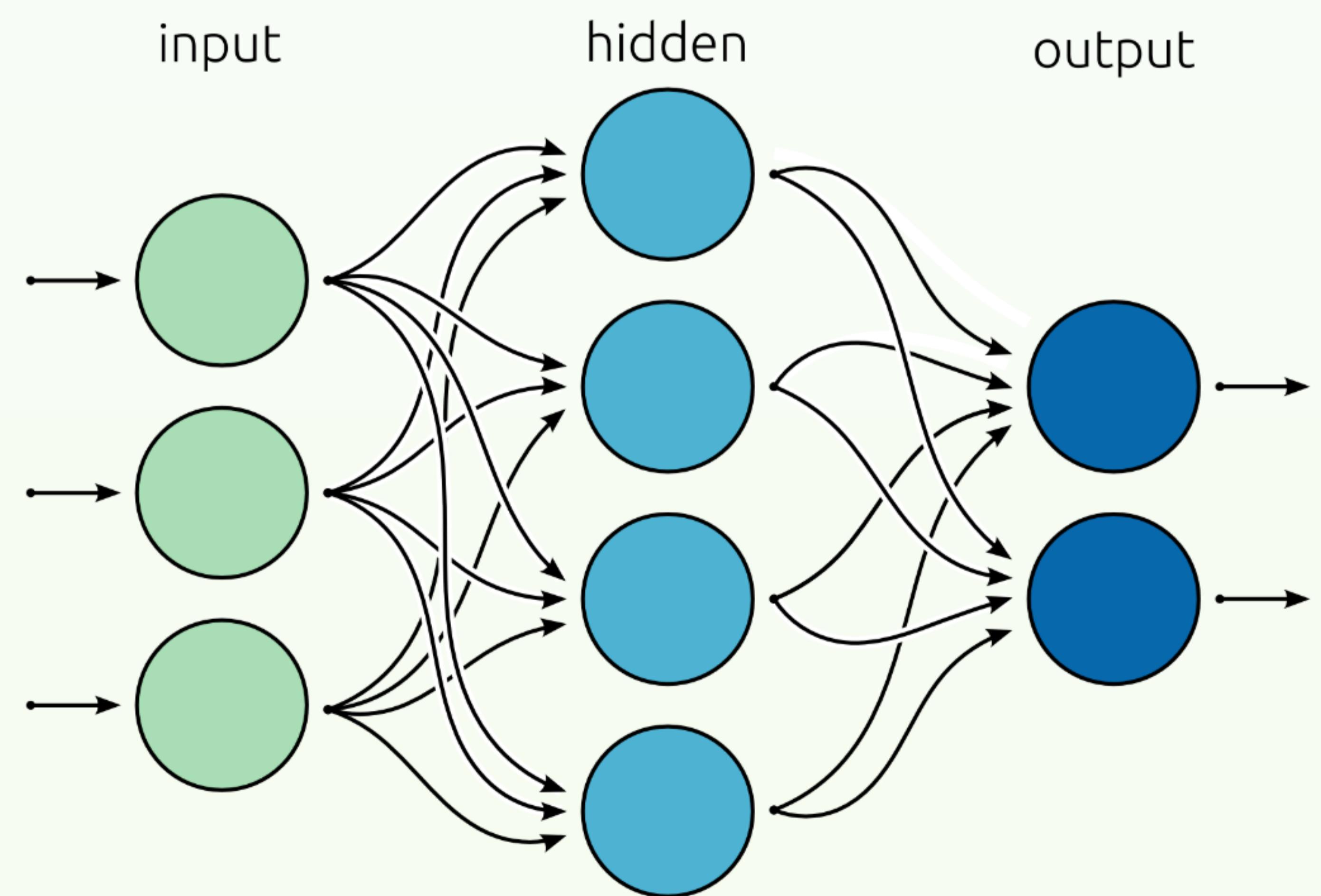
Output passed to following layers.

$$y(\mathbf{x}) = g \left(\sum_{i=1}^n w_i x_i \right)$$



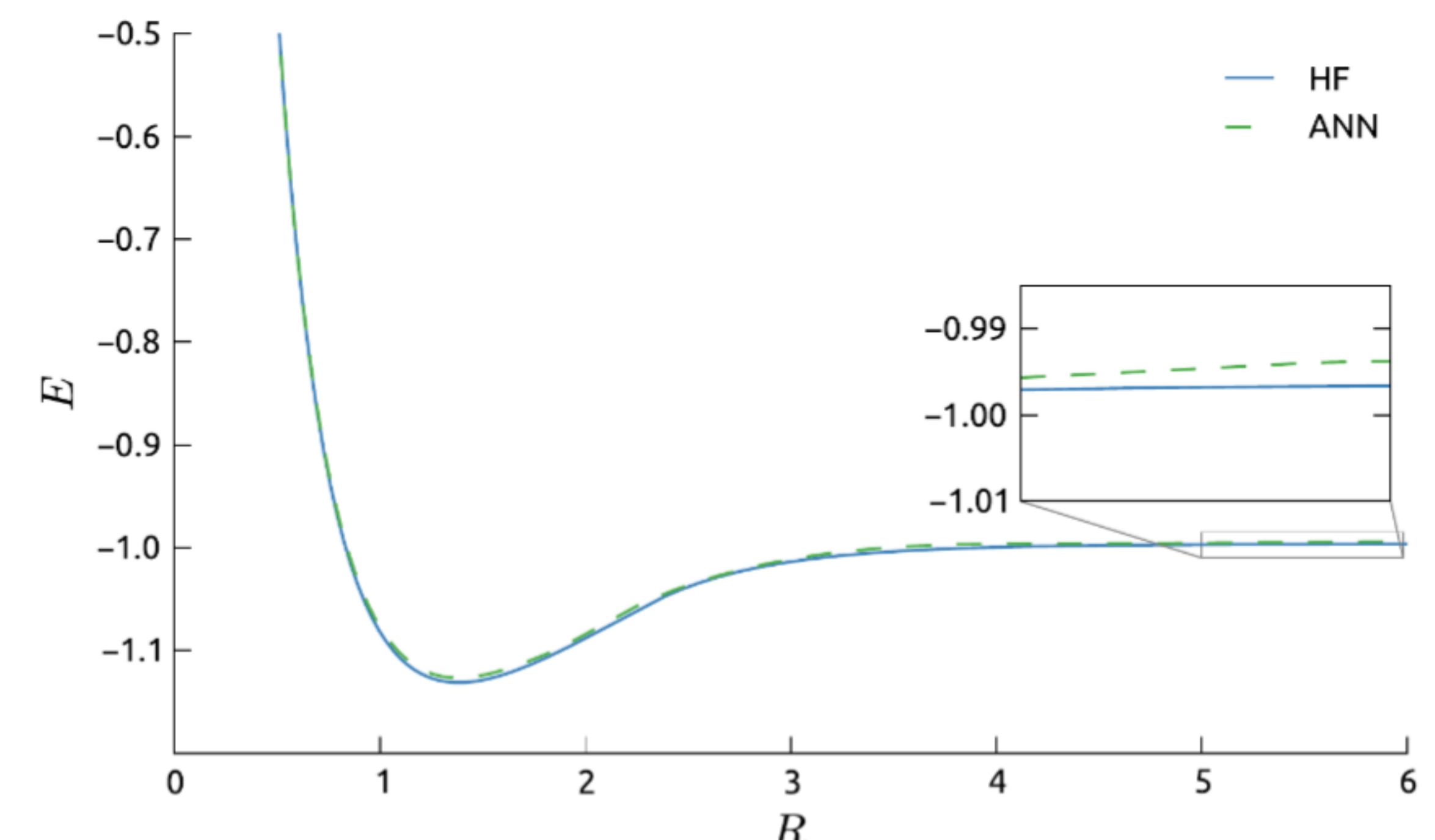
Training With Backpropagation

- Load training data.
- Run network.
- Compare retrieved and desired output.
- Propagate error and derivative backwards.
- Adjust weights.

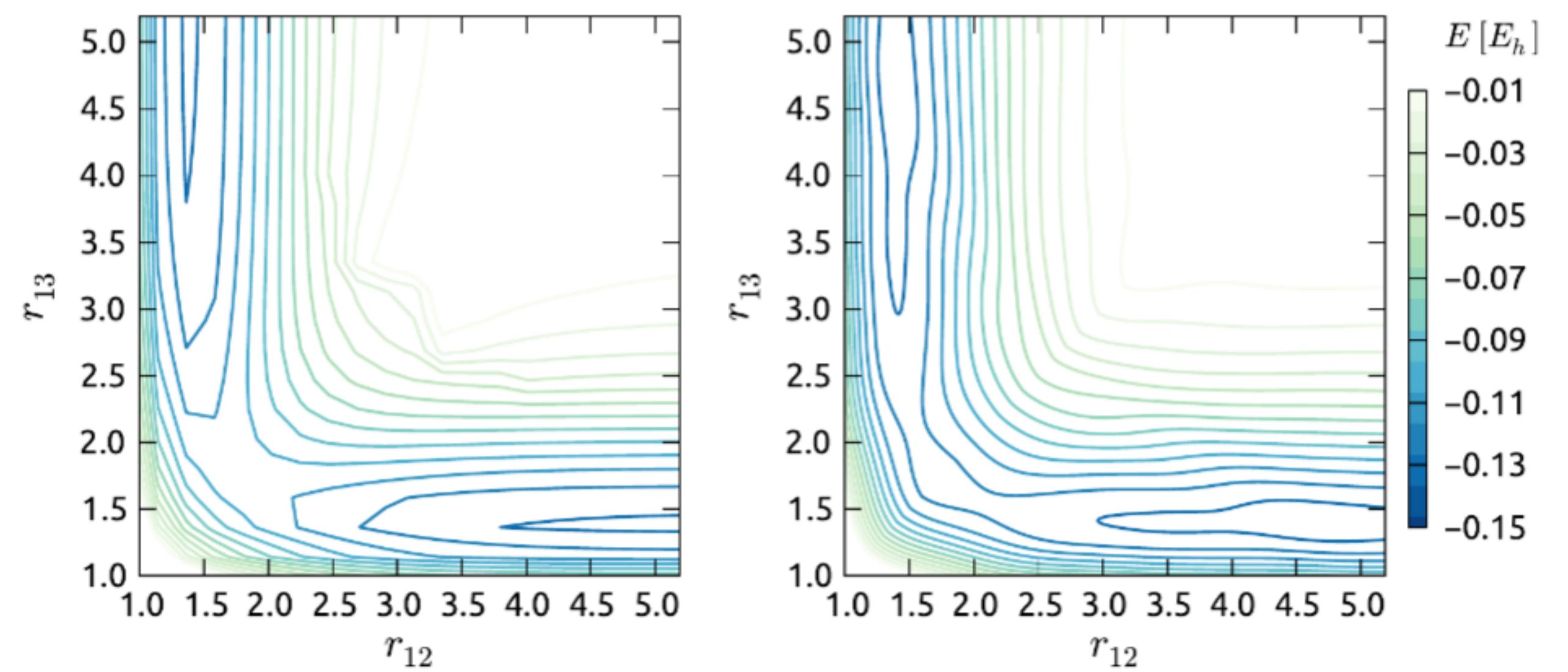


Approximating Potentials

- Calculate energy for configurations.
- Train network with Fast Artificial Neural Network Library (FANN).
- Plot result.



H_2



(a) HF

(b) ANN

H_3

HYDROGEN MOLECULES: RESULTS OF THE COMPLETE WORKFLOW

Results of the Complete Workflow

Performance

Hydrogen molecules!

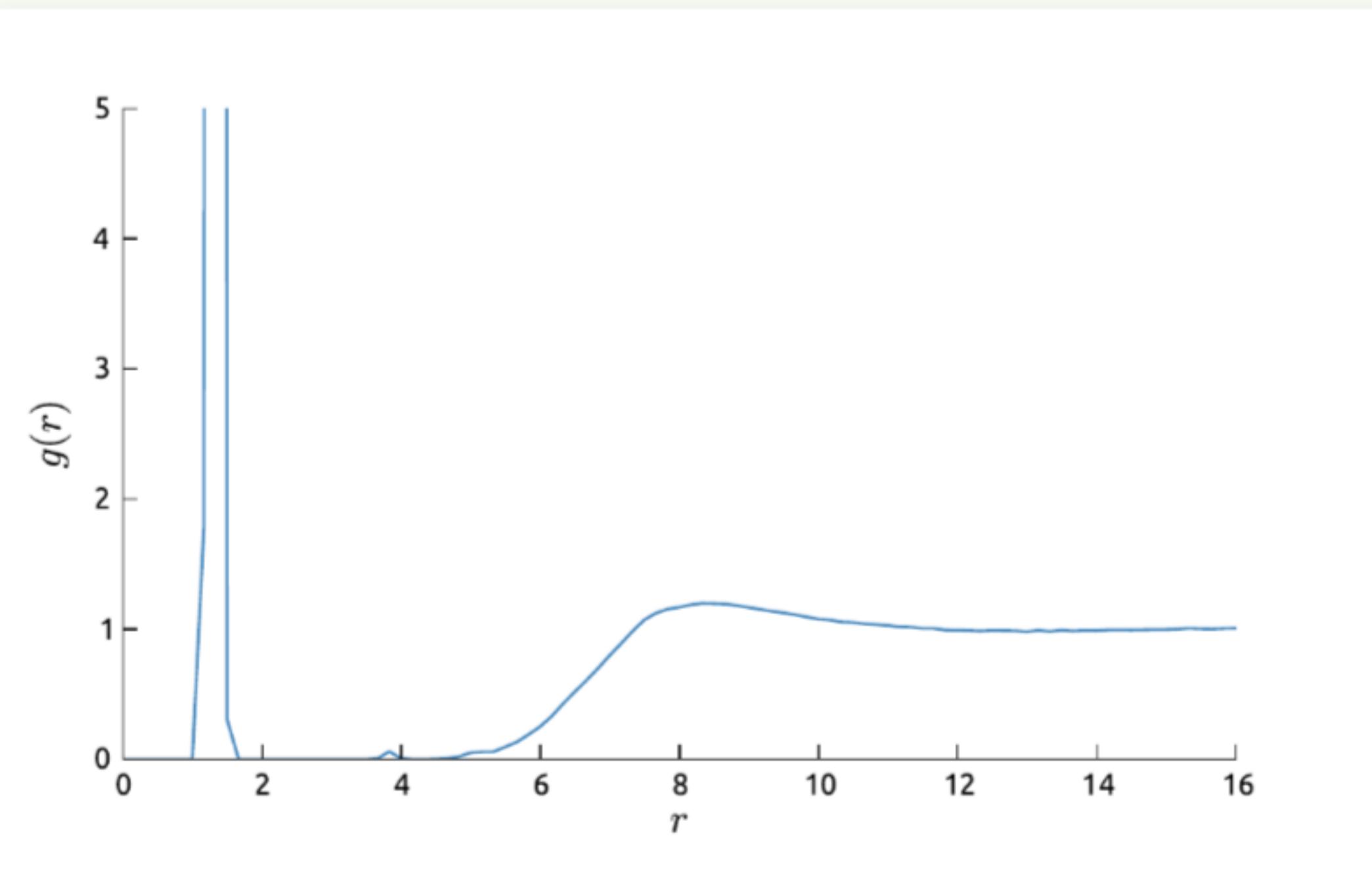
10 - 20 times as expensive.



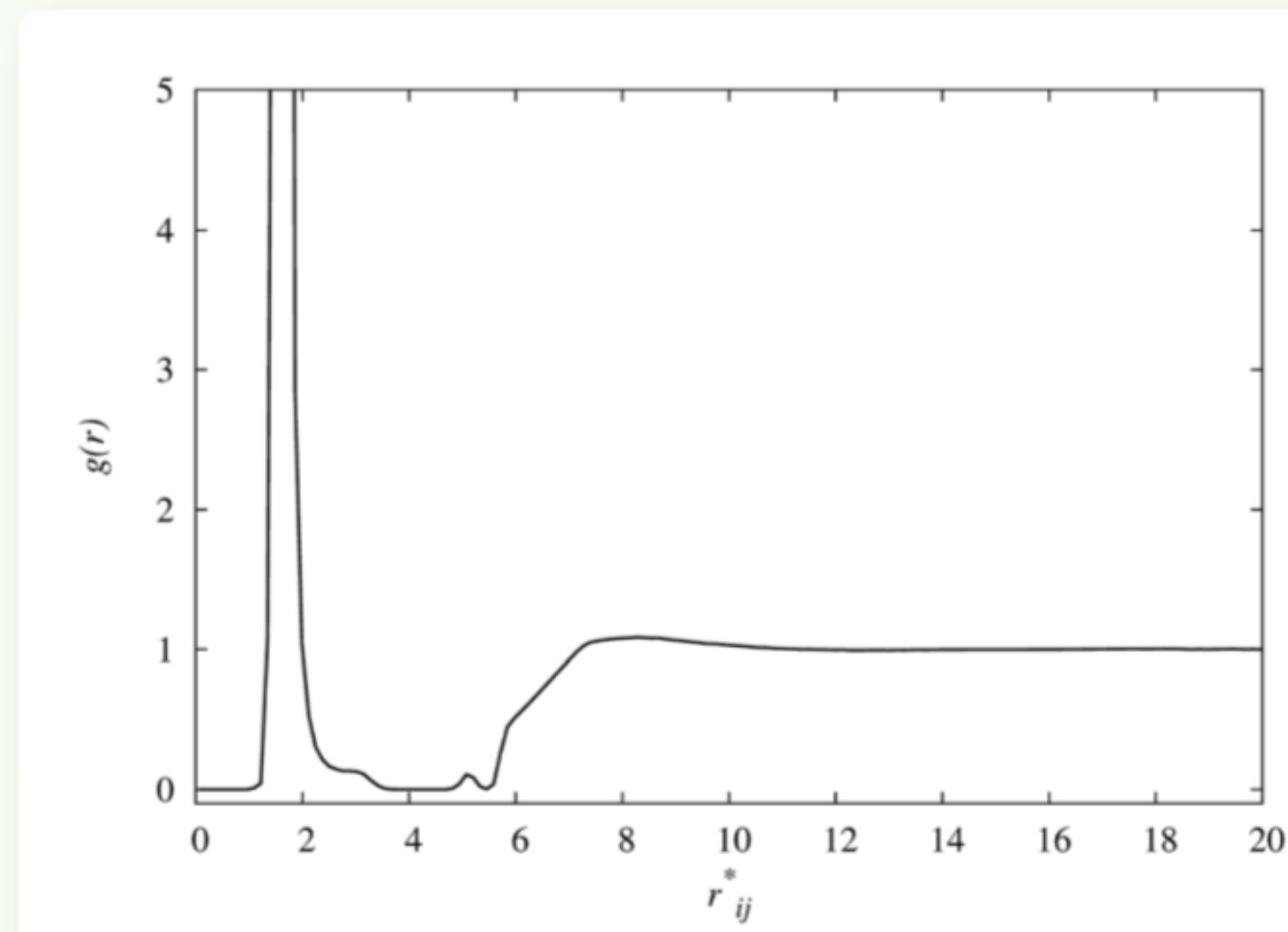
Hydrogen Dissociation

Good correspondence
with study by Skorpa et. al.

$g(r)$ for ANN at 156 K



$g(r)$ for Kohen-Tully-Stillinger at 156 K



Conclusion

- Potentials for MD from quantum.
- Macroscopic properties from MD.
- Neural networks for potential approximation.
- Less person-hours, more computation time.

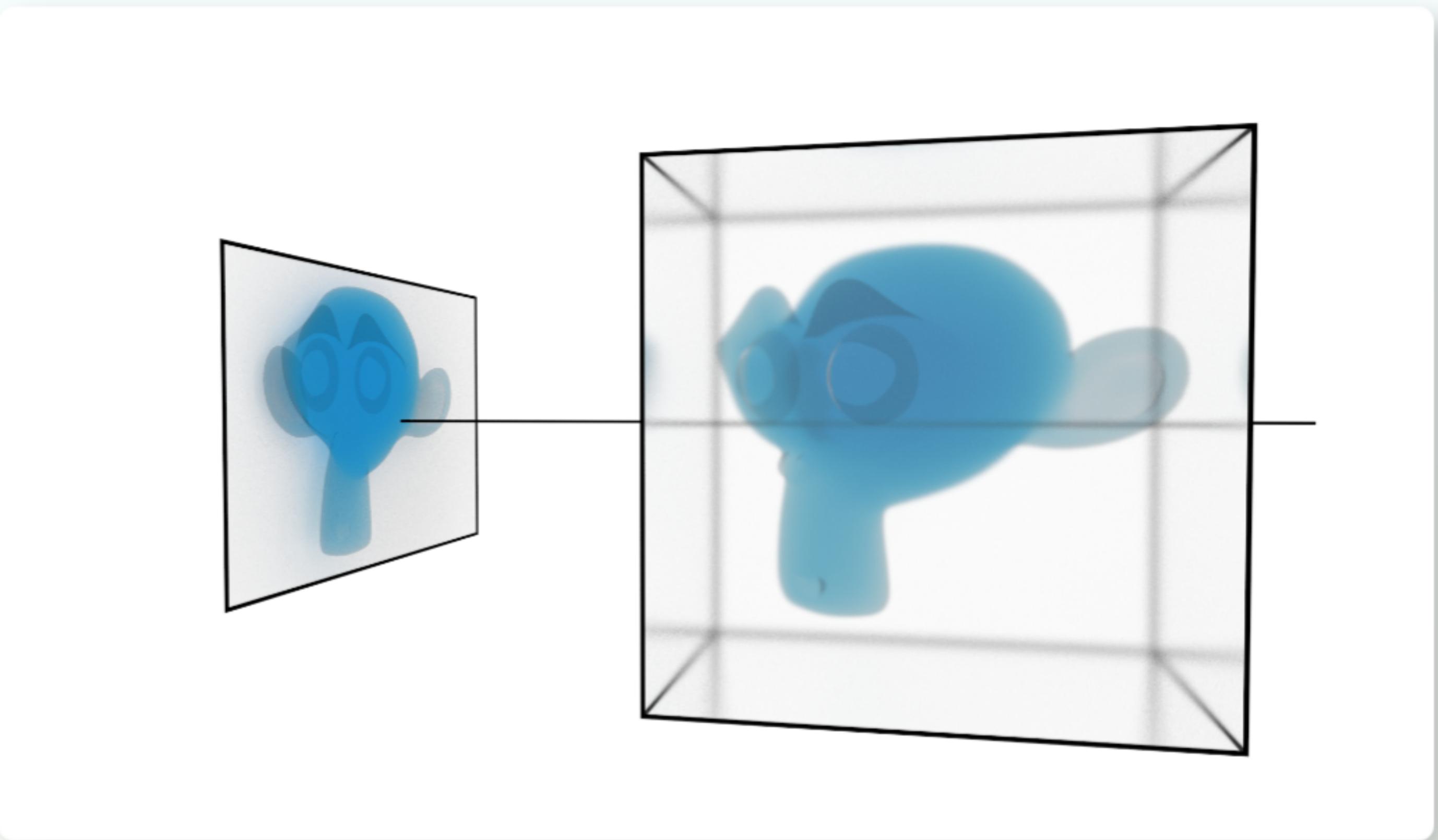
Future Possibilities

- Test cutoff effects.
- Better quantum computations.
- Better neural networks.
- More advanced systems.

VISUALIZATION

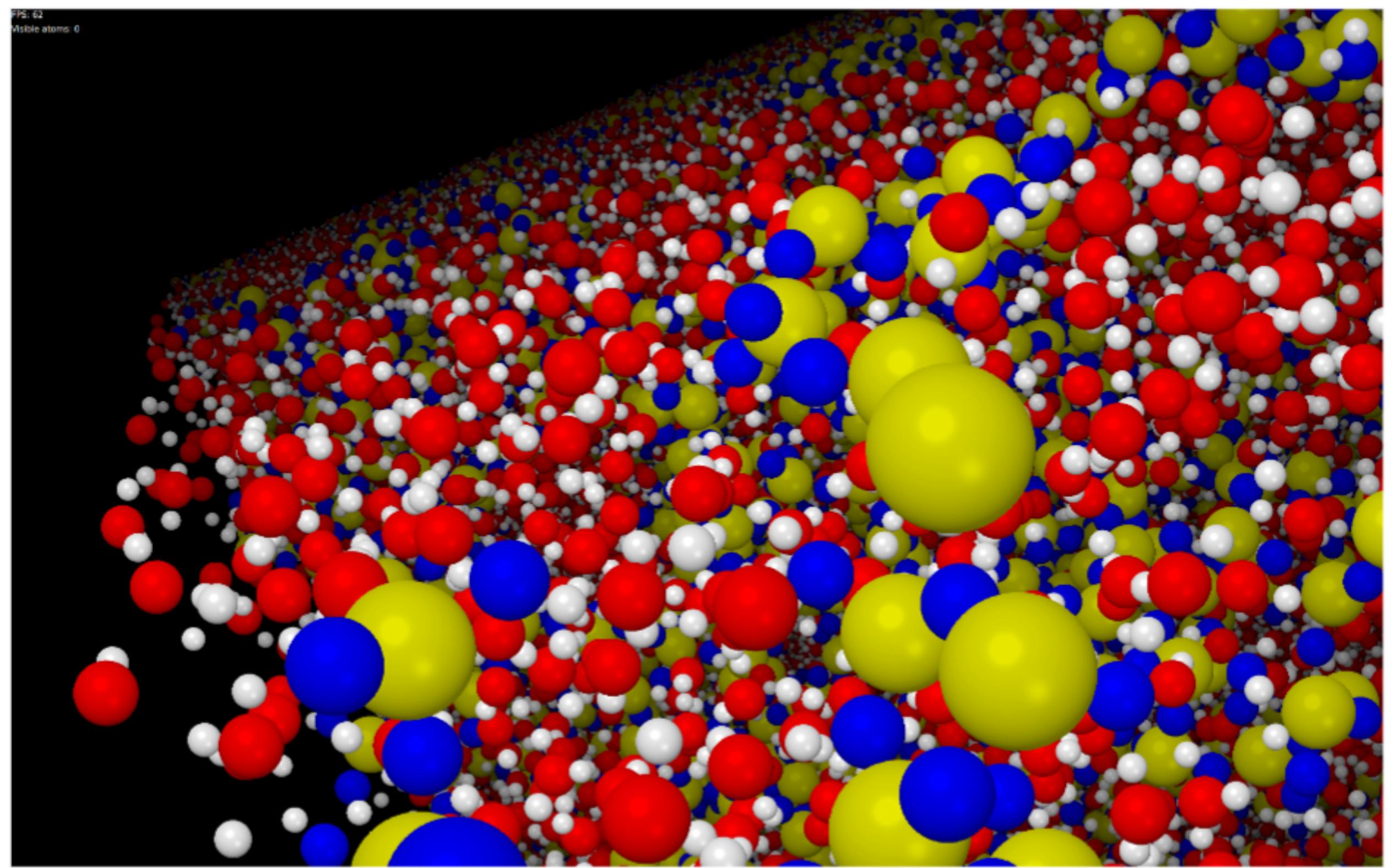
Volume Rendering of Density

Rays are traced through a cube of volume data. Values of the data are accumulated and define the intensity and color of a pixel.



Demo: Virtual Reality

Millions of Atoms



Thank you for your attention!