AI + Materials Science

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Website: ai4s.lab.westlake.edu.cn/course

Image from: DeepMind

Outlines

- Intro to Materials Science
    - Crystalline Materials
    - Structure-Property Relationship
    - DFT Calculation
- AI in Materials Science
    - Artificial intelligence ab initio (AI^2) methods
    - Directly Bridging
    - Inverse Problem
    - AI as a Powerful Assistance
    - Outside and Beyond Crystals

What are Materials?

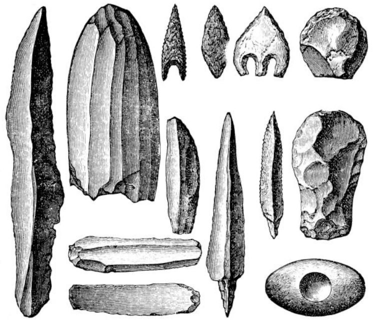
- Fundamental building blocks of modern life
- Every piece of solid thing you can touch around you

- The history of human civilization is also the history of how humans exploit different kinds of materials

Stone Age
3.4 to 1 million years ago

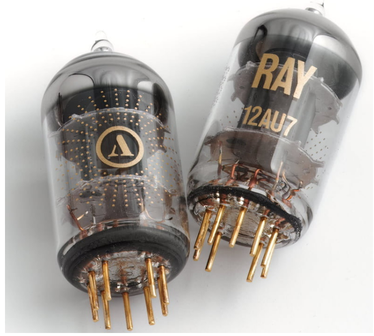
Bronze Age
3300 BC to 1200 BC

Iron Age
1200 BC to c. 550 BC



Why do we Care about Materials?

- **Necessity**:
  - Everything is made out of something
  - Materials selection is critical to design and performance
- **Ambition**:
  - New materials = new opportunities
  - Ex: transistor to integrated circuit
- **Fear**:
  - Most failures are materials failures
  - Ex: aircraft accident due to material fracture



Vacuum Tube

MOSFET

Classification of Materials: Two Categories

Fundamental difference: Long-range Order of Atoms

- **Crystalline Materials**:
  - Atoms are arranged in a **periodic pattern** repeated throughout the material
  - Ex: diamond, most of semiconductors and metals


- **Amorphous Materials**:
  - No systematic and regular pattern of atom arrangements
  - Ex: glass, plastic, rubber


Generally speaking:

- Most of crucial functional applications rely on crystalline materials

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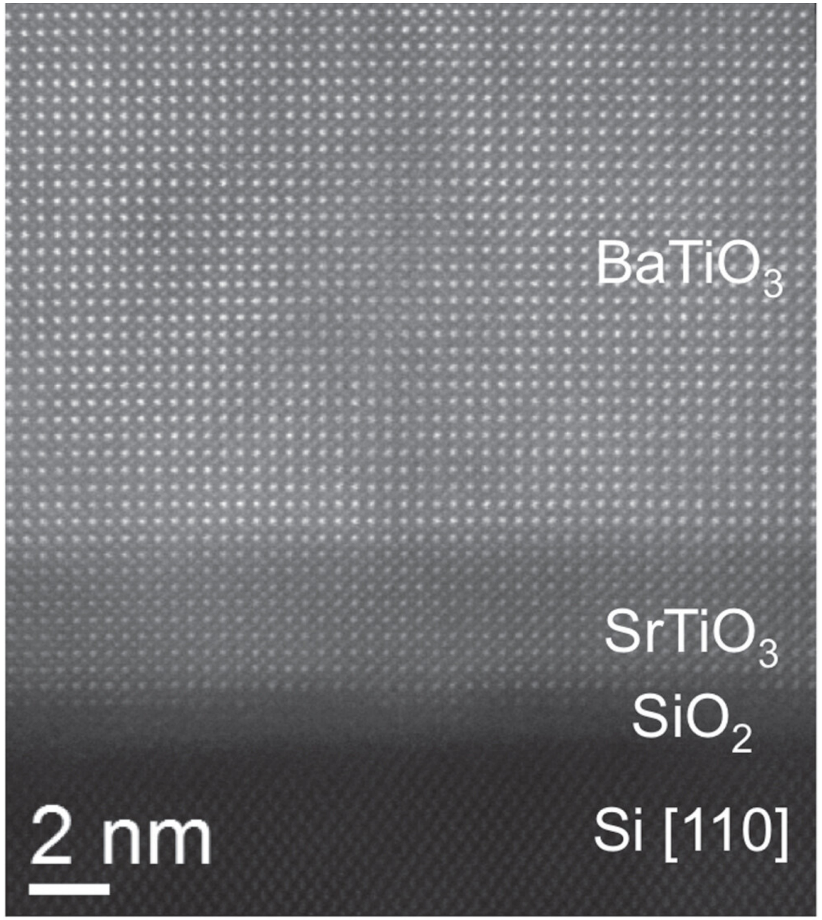
Intro to Materials Science

Crystalline Materials

Crystalline Materials:

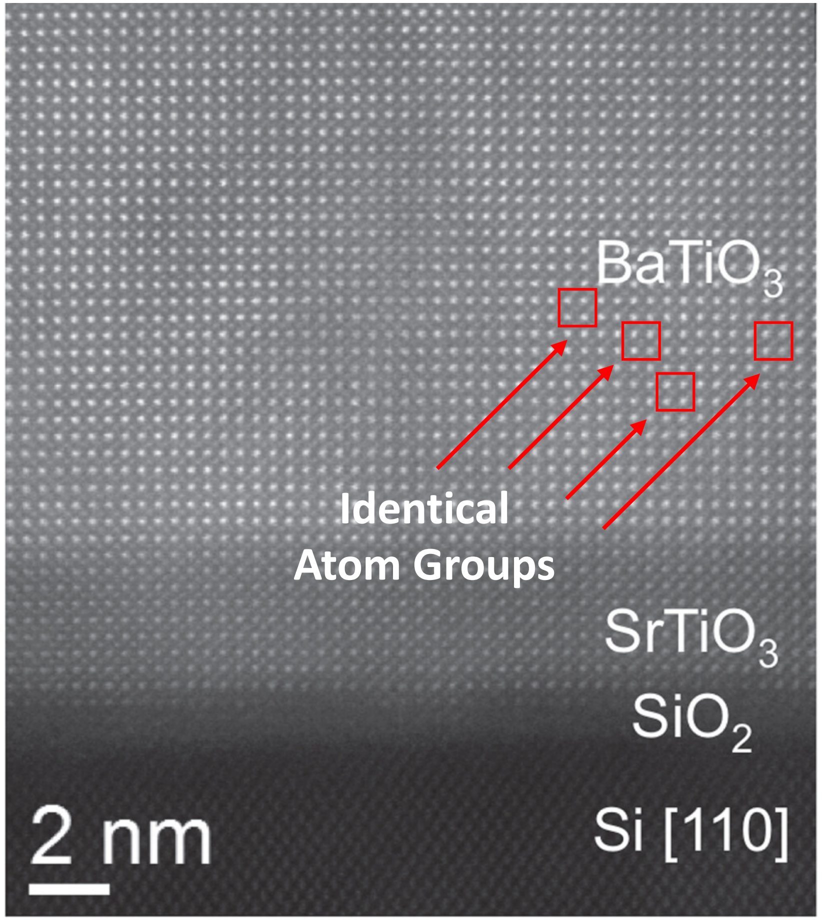


Atomic Resolution TEM HAADF Image of Crystalline Material [1]

- Periodic repeating pattern of atoms can be clearly observed
- But how to properly describe such structures?

[1] Mazet, L., et al. (2015). A review of molecular beam epitaxy of ferroelectric $BaTiO_3$ films on Si, Ge and GaAs substrates and their applications. *Science and Technology of Advanced Materials*, *16*(3).

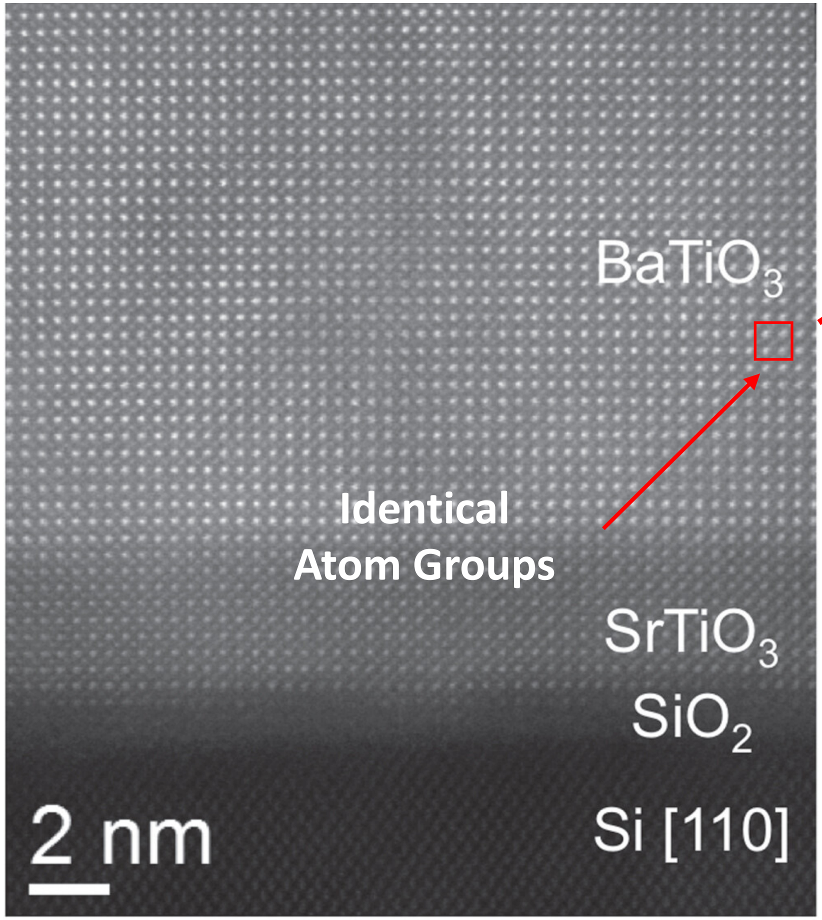
Crystalline Materials:



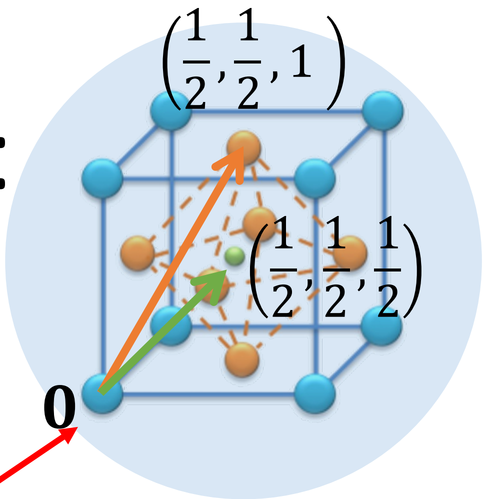
Atomic Resolution TEM HAADF Image of Crystalline Material

- Fundamental Truth:
  - The crystal consists of identical atom groups as **minimal repeating units**
  - The crystal is constructed by duplicating its repeating unit following a **certain pattern**

Crystalline Materials:



Atomic Resolution TEM HAADF Image
of Crystalline Material



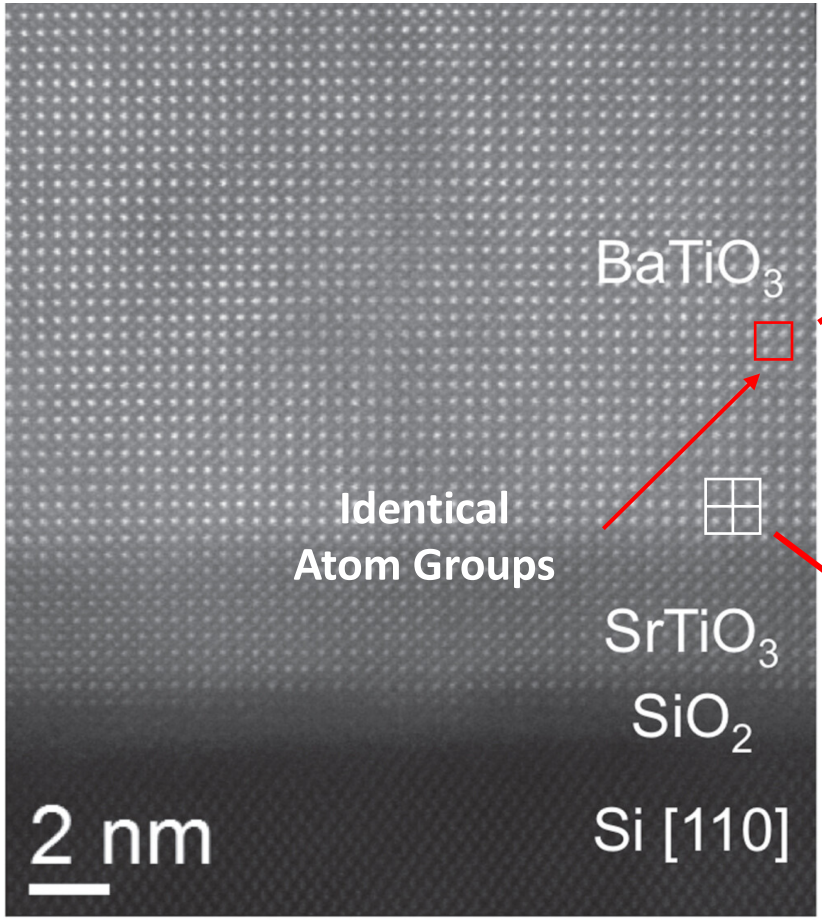
- **Minimal repeating unit:**
  - **Unit Cell**
  - The position of each atom is described by a fractional coordinate in the cell:
  - Ex: Ti atom at the body center

$$\overrightarrow{\mathbf{Ti}} = \left[\frac{1}{2},\frac{1}{2},\frac{1}{2}\right]^{\mathrm{T}}$$

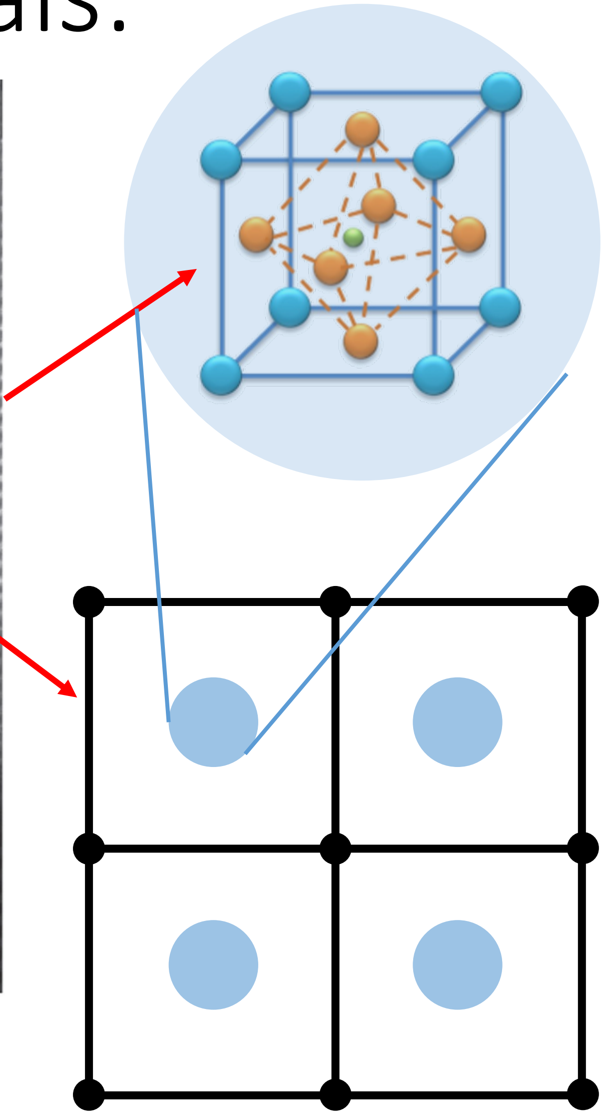
  - Ex: O atom at the top center

$$\vec{\mathbf{O}} = \left[\frac{1}{2},\frac{1}{2},1\right]^{\mathrm{T}}$$

Crystalline Materials:



Atomic Resolution TEM HAADF Image of Crystalline Material

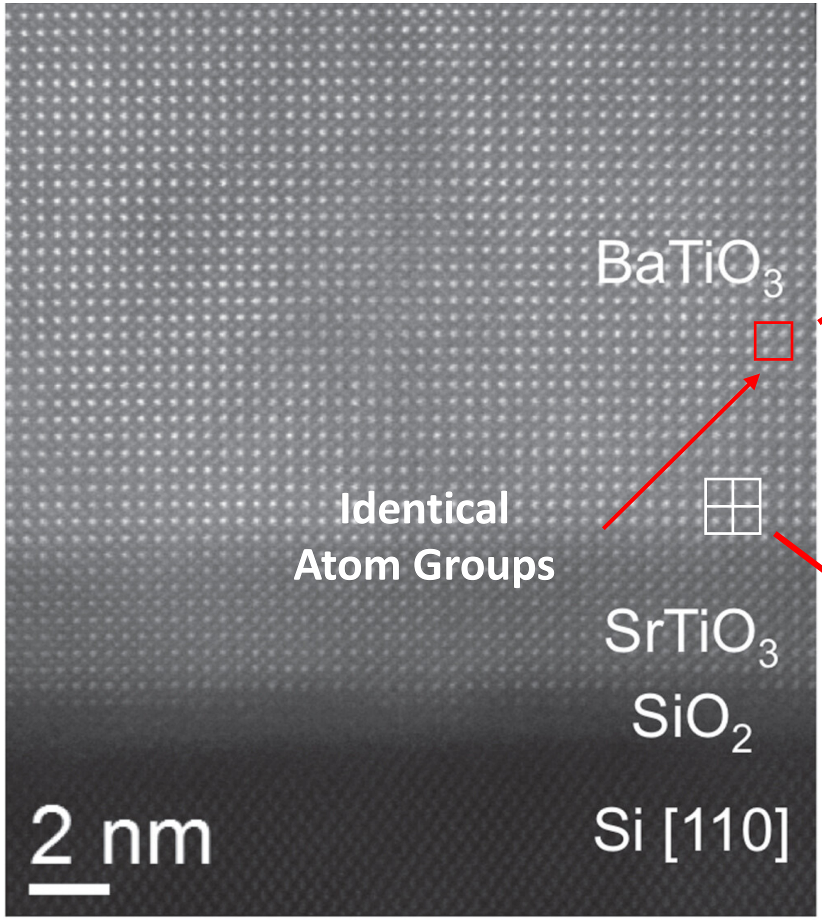


- Minimal repeating unit:
  - **Unit Cell**

Each **Unit Cell** can be **abstracted** as a **blue dot** in the **Lattice Grid**

- Repeating pattern:
  - **Lattice Grid**

Crystalline Materials:



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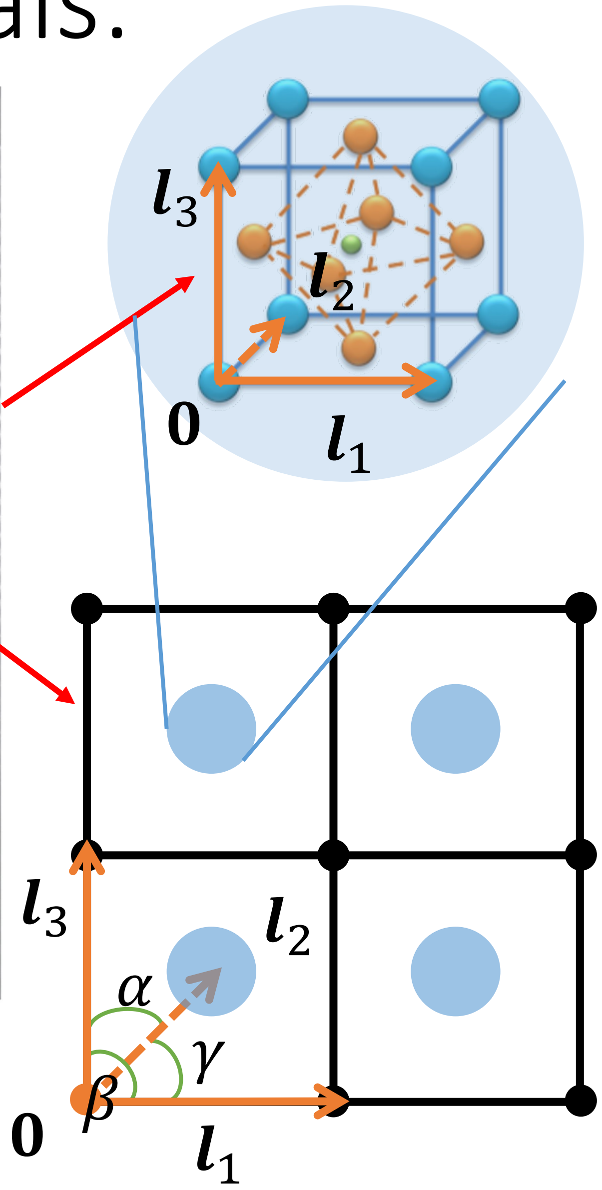


Lattice Grid:

- Similar to the Cartesian coordinate system, defined by a set of basis Lattice Vectors:

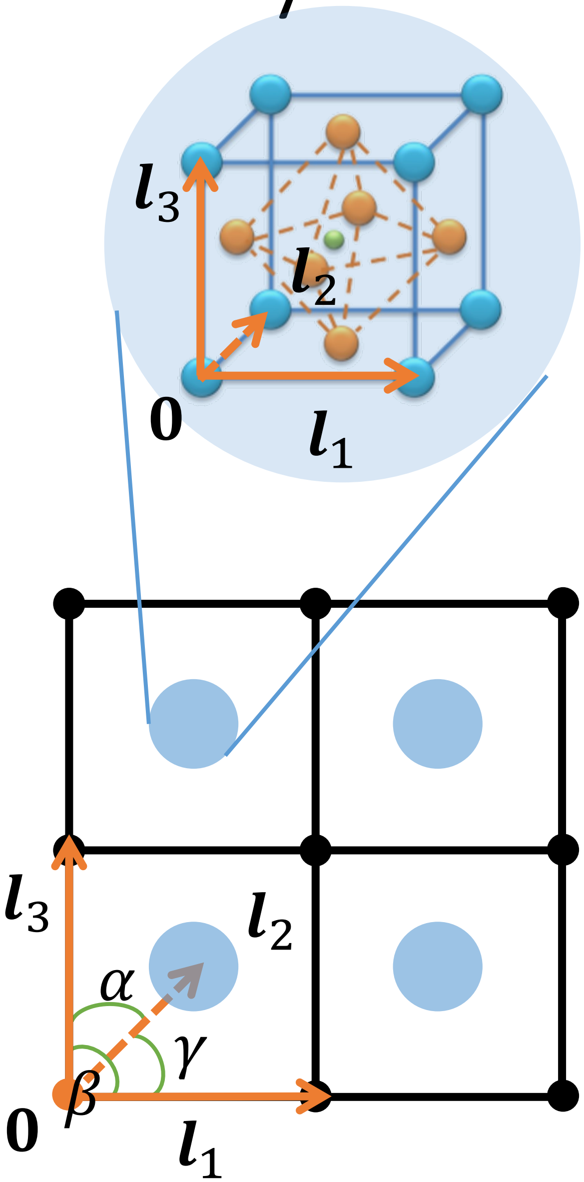
$$\boldsymbol{l}_1, \boldsymbol{l}_2, \boldsymbol{l}_3$$

- The angles between lattice vectors is defined as lattice angles:

$$\alpha, \beta, \gamma$$

- Together, they are defined as **Lattice Constant**

Crystalline Materials:



- In our example: Cubic $BaTiO_3$

$$||\vec{\boldsymbol{a}}|| = \left|\left|\vec{\boldsymbol{b}}\right|\right| = ||\vec{\boldsymbol{c}}|| = 4.01\ \text{Å}$$

$$\alpha = \beta = \gamma = 90°$$

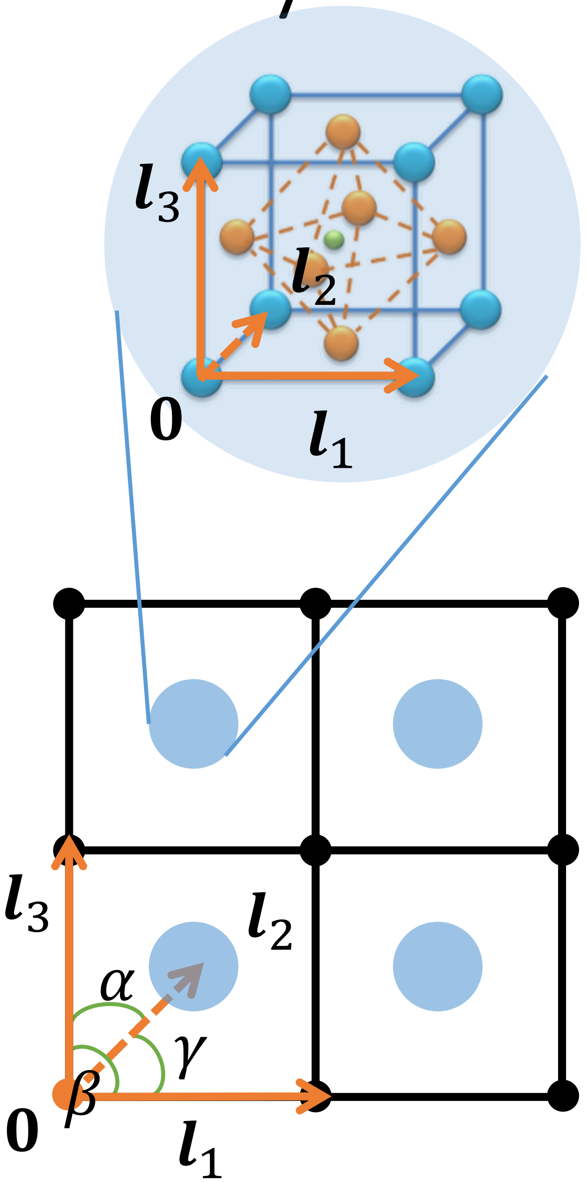
  - Ps. $1\ \text{Å} = 0.1\ \text{nm}$
- Mathematically, structure of a crystalline material $\boldsymbol{\mathcal{M}}$ can be represented as:

$$\boldsymbol{\mathcal{M}} = (A, \boldsymbol{X}, \boldsymbol{L})$$

  - $A \in \mathbb{Z}^n$: vector of atomic numbers of n atoms in the **Unit Cell**
  - $\boldsymbol{X} = [\boldsymbol{x}_1, \boldsymbol{x}_2, \dots, \boldsymbol{x}_n] \in \mathbb{R}^{3\times n}$: tensor of fractional atom coordinates
  - $\boldsymbol{L} = [\boldsymbol{l}_1, \boldsymbol{l}_2, \boldsymbol{l}_3] \in \mathbb{R}^{3\times 3}$: tensor of **Lattice Vectors**

- Given the **Unit Cell**, repeat along the **Lattice Vectors**, you are now able to reconstruct the **ENTIRE** crystal!

Crystalline Materials: $\mathcal{M} = (A, \boldsymbol{X}, \boldsymbol{L})$



- $A \in \mathbb{Z}^n$: vector of atomic numbers of n atoms in the **Unit Cell**
- $\boldsymbol{X} = [\boldsymbol{x}_1, \boldsymbol{x}_2, \ldots, \boldsymbol{x}_n] \in \mathbb{R}^{3\times n}$: tensor of fractional atom coordinates
- $\boldsymbol{L} = [\boldsymbol{l}_1, \boldsymbol{l}_2, \boldsymbol{l}_3] \in \mathbb{R}^{3\times 3}$: tensor of **Lattice Vectors**
- Crystal structure $\mathcal{M}$ has properties as follow:
  - **Real space** coordinates $\boldsymbol{C}$ of atoms can be obtained as:

$$\boldsymbol{C}^{\mathrm{T}} = \boldsymbol{X}^{\mathrm{T}} \cdot \boldsymbol{L}$$

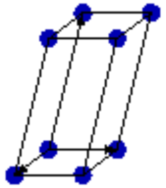
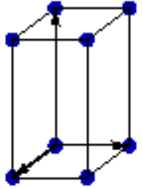
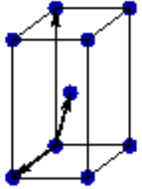
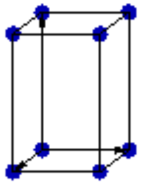
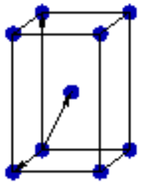
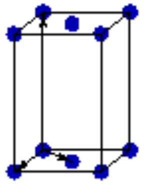
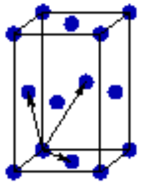
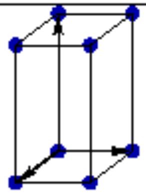
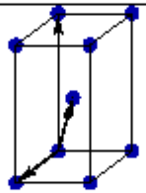
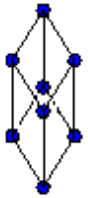
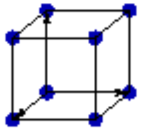
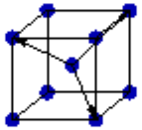
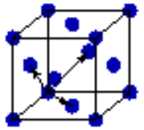
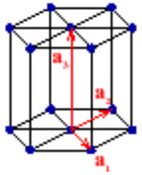
  - Periodicity:

$$\boldsymbol{x}_i' = \boldsymbol{x}_i + \boldsymbol{t} \cdot \boldsymbol{L}$$
$$\boldsymbol{t} \in \mathbb{Z}^3$$

  - coordinates of atoms with same index i in the periodic expanded space can be obtained by adding a combination of integer multiple lattice vectors

Crystal Structures:

- 7 crystal systems and 14 Bravais lattices
- spatial arrangement of atoms determines the material properties

Bravais lattice	Parameters	Simple (P)	Volume centered (I)	Base centered (C)	Face centered (F)
Triclinic	$a_1 \neq a_2 \neq a_3$ $\alpha_{12} \neq \alpha_{23} \neq \alpha_{31}$				
Monoclinic	$a_1 \neq a_2 \neq a_3$ $\alpha_{23} = \alpha_{31} = 90°$ $\alpha_{12} \neq 90°$				
Orthorhombic	$a_1 \neq a_2 \neq a_3$ $\alpha_{12} = \alpha_{23} = \alpha_{31} = 90°$				
Tetragonal	$a_1 = a_2 \neq a_3$ $\alpha_{12} = \alpha_{23} = \alpha_{31} = 90°$				
Trigonal	$a_1 = a_2 = a_3$ $\alpha_{12} = \alpha_{23} = \alpha_{31} < 120°$				
Cubic	$a_1 = a_2 = a_3$ $\alpha_{12} = \alpha_{23} = \alpha_{31} = 90°$				
Hexagonal	$a_1 = a_2 \neq a_3$ $\alpha_{12} = 120°$ $\alpha_{23} = \alpha_{31} = 90°$				

Intro to Materials Science

Structure-Property Relationship

The Essence of Materials Science

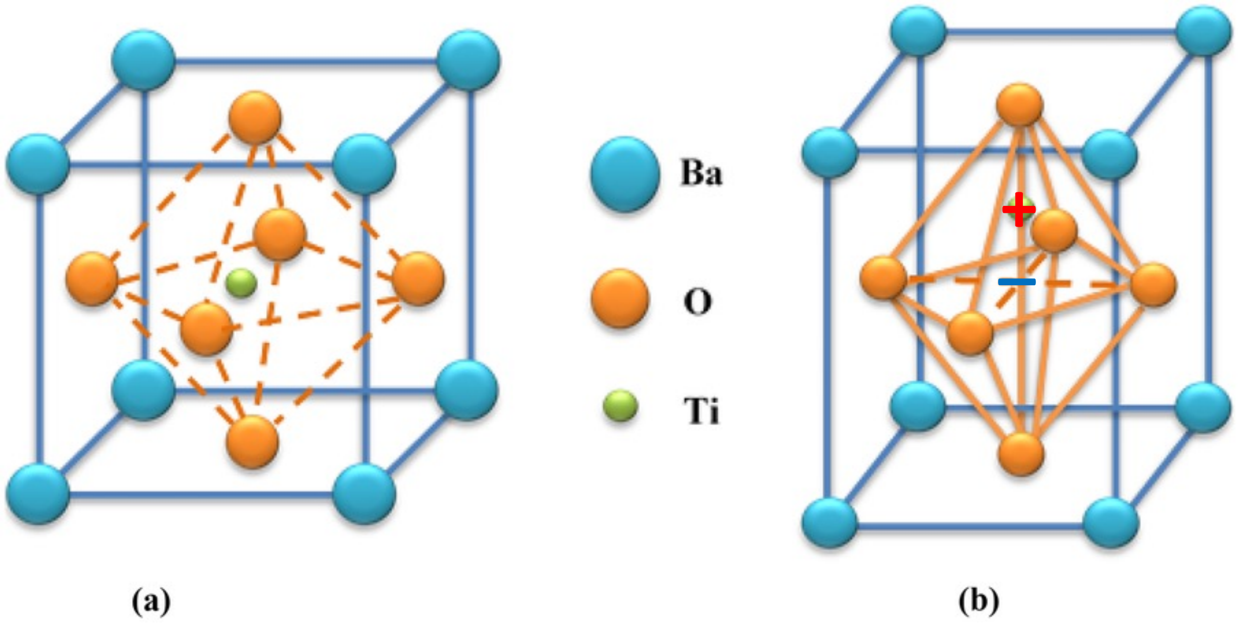
- The fundamental research subject can be summarized as:

Structure-Property Relationship

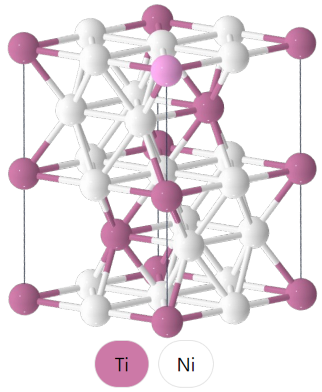
- Structure:
  - **Composition**: kind and fraction of atoms present
  - **Microstructure**: how those atoms are arranged in the materials
- Property:
  - **Isotropic**: orientation-irrelevant, e.g. density, heat capacity, melting point
  - **Anisotropic**: e.g. polarity, magnetization, piezoelectricity

Structure-Property Relationship

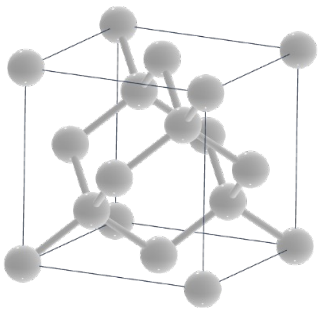
- Ex: Piezoelectric ceramics $BaTiO_3$
- Structure:
  - **Composition**: Ba:Ti:O = 1:1:3
  - **Microstructure**: described as Fig.a
- Property:
  - **Isotropic**:
    - density = 6.02 g/cm^3,
    - melting point = 1,625 °C
  - **Anisotropic**:
    - piezoelectricity along the polarization (z-axis in Fig.b)



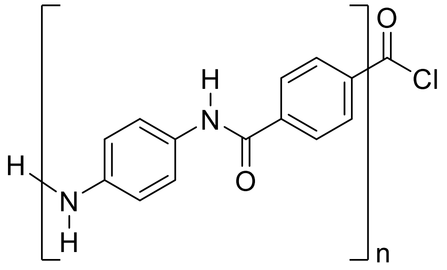
Structure-Property Relationship



Nitinol



Si



Kevlar

Determines

Shape-memory Alloy

Phase-transfer Temperature

Stiffness

Mechanical Strength

Semiconductor

Band-gap

Carrier Mobility

Thermostability

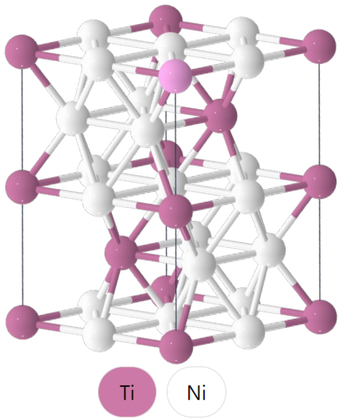
Bullet-proof Fiber

Glass-transition Temperature

Degradation Rate

……

Experimental Approaches



Structure

Preparation

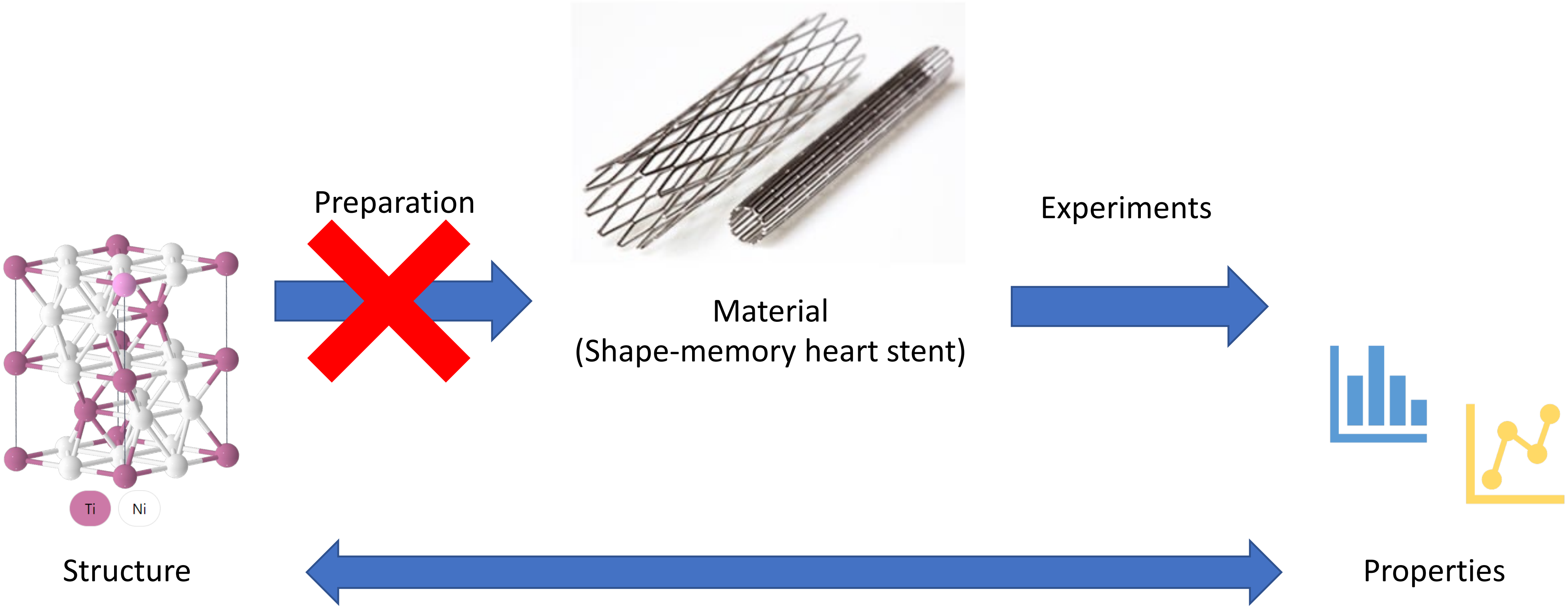
Material
(Shape-memory heart stent)

Experiments

Properties

Structure-Property
Relationship

Experimental Approaches



Structure

Preparation

Material
(Shape-memory heart stent)

Experiments

Properties

Structure-Property
Relationship

Experimental Approaches

$\mathrm{Ni_xTi_{1-x}}$

Composition

Preparation

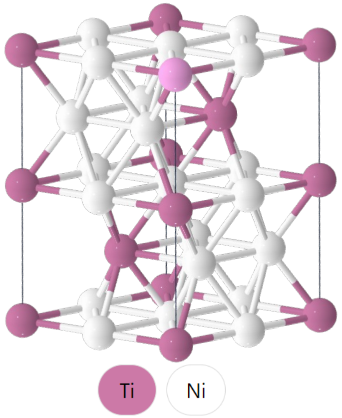
Material
(Shape-memory heart stent)

Experiments

Properties

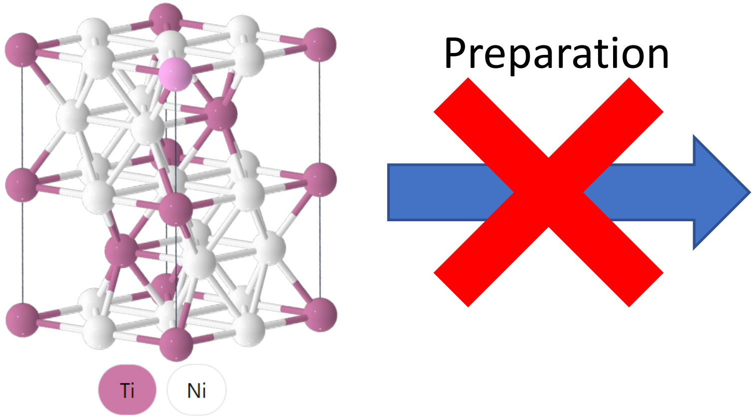
Structure-Property
Relationship

Characterization



Structure

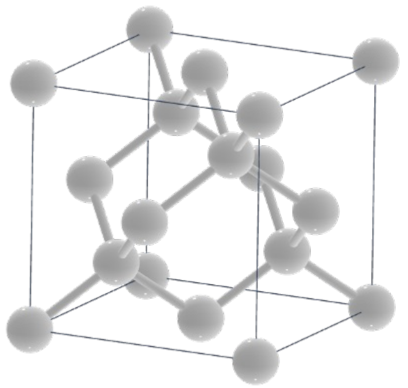
Experimental Approaches



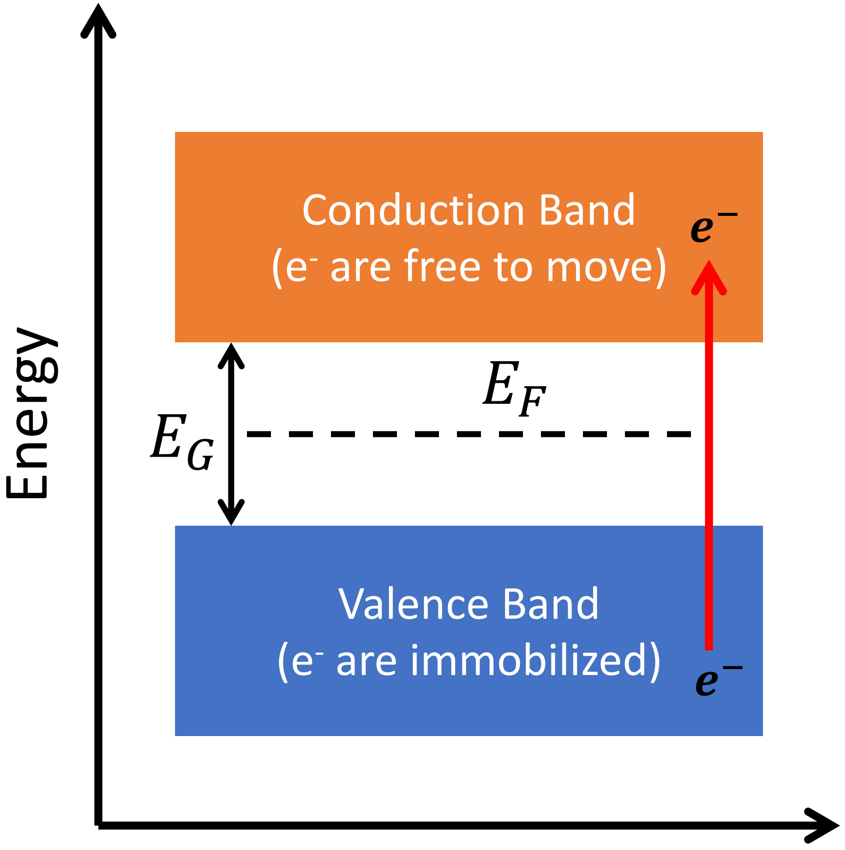
Structure

- Limitations:
  - Desired structure can not be prepared with precise, atomic-level control
  - Property of many structures worth studying is unable to be determined experimentally
  - High experimental costs and low efficiency

Theoretical Approaches



Si



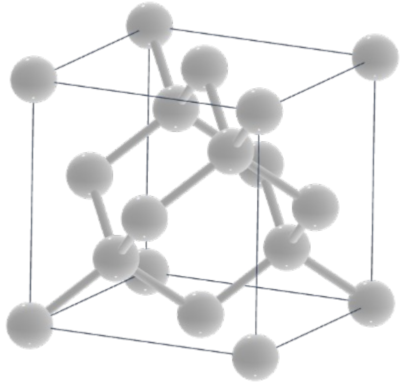
Band Structure of
Intrinsic Semiconductor (Si)

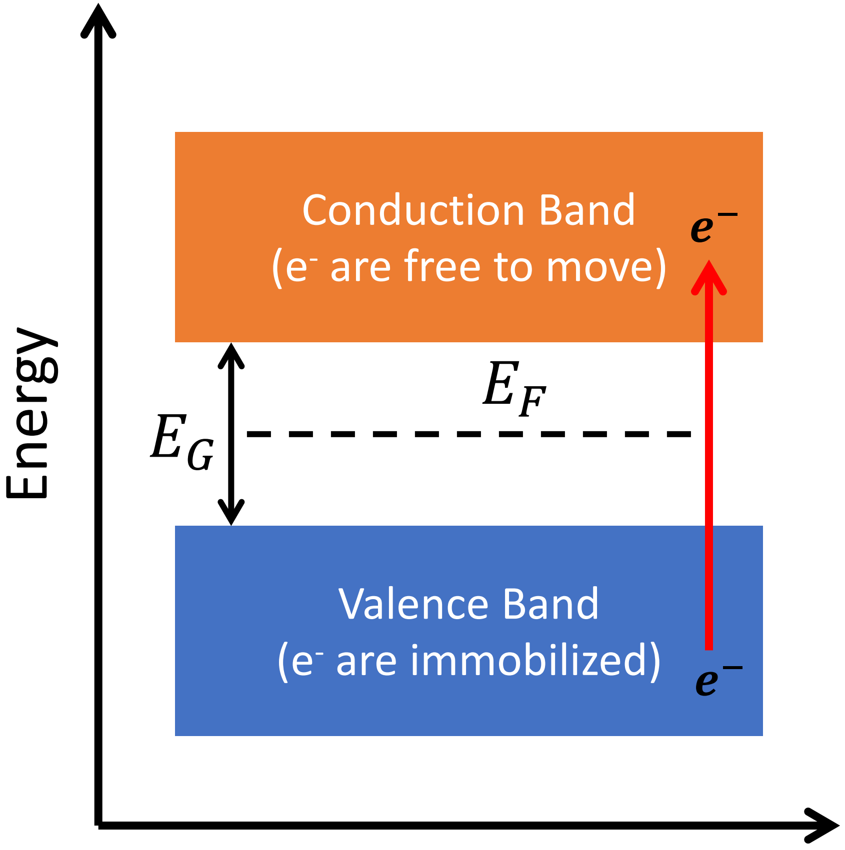
- The valence band of Si is fully filled and the conduction band is empty, preventing the electrons from traveling through the material
- The electrons need to be excited across the band gap by thermal or external energy
- Therefore, the conductivity of Si at temperature T can be approximated:

$$\sigma \approx N_0 e \mu_e \exp \frac{(E_G)}{kT}$$

- Here N_0 is number of electrons per unit volume, μ_0 is the carrier mobility of electron

Theoretical Approaches


Si



Band Structure of
Intrinsic Semiconductor (Si)

- Limitations:
  - Lots of assumptions and approximations have to be applied to the theory
  - Ignored some of the complex micro-scale interactions
  - Limited application scenarios

Intro to Materials Science

DFT Calculation

Computational Approaches

- Can we try to compute the material properties purely from its structures?
  - *Ab initio* Calculations: from electrons to properties
- Wave Function of electron:

$$\Psi(\boldsymbol{r}, t)$$

- Probability Density:

$$\Psi^*(\boldsymbol{r}, t)\Psi(\boldsymbol{r}, t) = |\Psi(\boldsymbol{r}, t)|^2$$

- Schrödinger Equation:

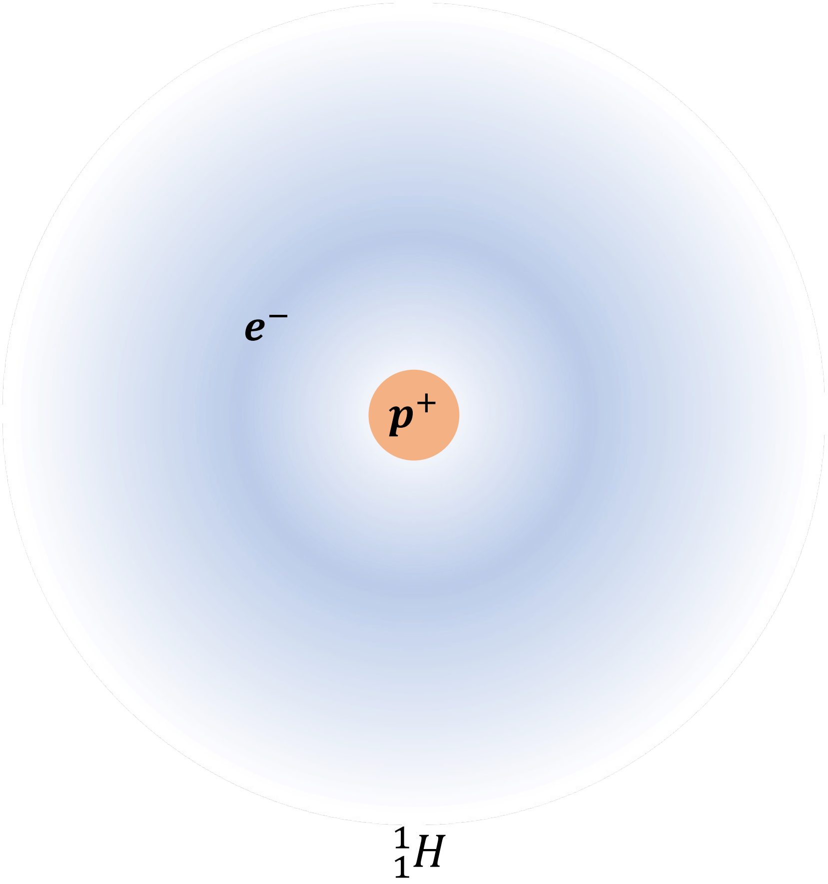
$$\hat{H}\Psi(\boldsymbol{r}, t) = i\hbar\frac{\partial}{\partial t}\Psi(\boldsymbol{r}, t)$$

- where the Hamiltonian operator is:

$$\hat{H} = -\frac{\hbar^2}{2m}\nabla^2 + \hat{V}$$

- Time-Independent Schrödinger Equation:

$$\hat{H}\psi(\boldsymbol{r}) = E\psi(\boldsymbol{r})$$
$$E_1, \psi_1;$$
$$\dots$$



Ab initio Calculations: Schrödinger equation

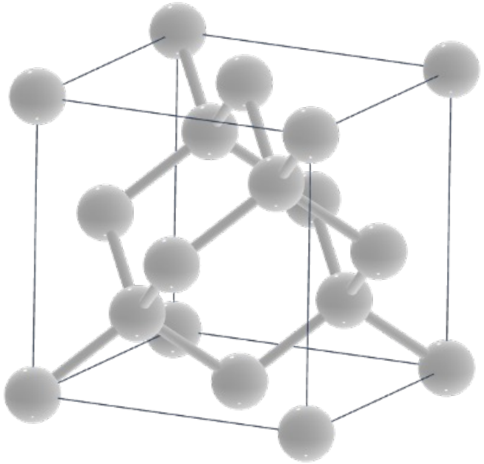
- Works fine for single electron system: H atom, hydrogenic ions (He^+, Li^{2+})
- Directly solving the many-electron Schrödinger equation is currently computationally impossible, with complexity exponential to N, the number of electrons in the system
- Schrödinger Equation:

$$\hat{H}\Psi(\boldsymbol{r},t) = i\hbar\frac{\partial}{\partial t}\Psi(\boldsymbol{r},t)$$

- Many-Body Calculation:

$$\hat{H}\psi(\boldsymbol{R}) = [\hat{T} + \hat{V} + \hat{U}]\psi(\boldsymbol{R}) = E\psi(\boldsymbol{R})$$

$$\left[\sum_{i=1}^{N}\left(-\frac{\hbar^2}{2m_i}\nabla_i^2\right) + \sum_{i=1}^{N}V(\boldsymbol{r}_i) + \sum_{i\neq j}^{N}U\left(\boldsymbol{r}_i,\boldsymbol{r}_j\right)\right]\psi = E\psi$$



Si

112 e^- per unit cell

Ab initio Calculations: Hartree-Fock Method

- Hartree-Fock Method (HF): Self-Consistent Field (SCF)
- Assuming each electron is in **an average potential field** generated by all the other electrons:

$$\begin{tikzcd}
U(\boldsymbol{r}_i, \boldsymbol{r}_j) = V(\boldsymbol{r}_i) = V(\boldsymbol{R}_{j \neq i}) \arrow[d] \\
\widehat{H}_i^{(n-1)} \psi_i^{(n)}(\boldsymbol{r}_i) = E_i \psi_i^{(n)}(\boldsymbol{r}_i) \arrow[u, bend right, "n \mathrel{+}= 1"'] \arrow[d, "\text{converge}"] \\
\psi_i^{(n)}
\end{tikzcd}$$

- then:
- Hohenberg-Kohn (HK) Theorems:
- Electron Density:

$$\rho_0(\boldsymbol{r}) = N \int d\boldsymbol{r}_1 \dots \int d\boldsymbol{r}_N \, \psi_0^*(\boldsymbol{R}) \psi_0(\boldsymbol{R})$$

- Total Energy:

$$E_0(\rho_0) = \psi_0^*(\rho_0) \widehat{H} \psi_0(\rho_0)$$

- HK Theorems proof: https://people.chem.ucsb.edu/metiu/horia/OldFiles/115C/KH_Ch4.pdf

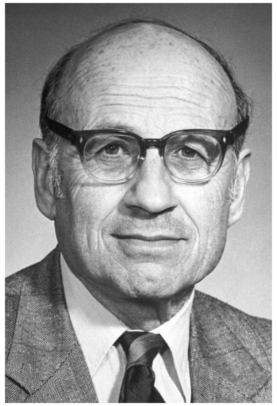
Ab initio Calculations: DFT

- Density Functional Theory (DFT):
  - Assumes that the property of the system is determined by electron density ρ
  - Treats the **energy as the functional of electron density** (3 variables), instead of trying to solve the entire wave function in the Schrödinger Equation (Nx3 variables)
  - The target of DFT calculation is to solve the Kohn-Sham equation:

$$\left(-\frac{\hbar\nabla^2}{2m}+v_{\mathrm{KS}}[\rho](\boldsymbol{r})\right)\varphi_i(\boldsymbol{r})=\varepsilon_i\varphi_i(\boldsymbol{r})$$

  - Where ρ is defined in terms of the Kohn-Sham wave functions φ_i:

$$\rho(\boldsymbol{r})=\sum_i|\varphi_i(\boldsymbol{r})|^2$$



Walter Kohn

Lu Jeu Sham

Ab initio Calculations: DFT

The Nobel Prize in Chemistry 1998

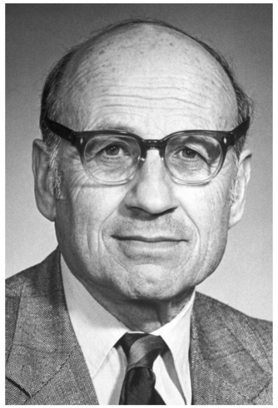


Photo from the Nobel Foundation archive.

Walter Kohn

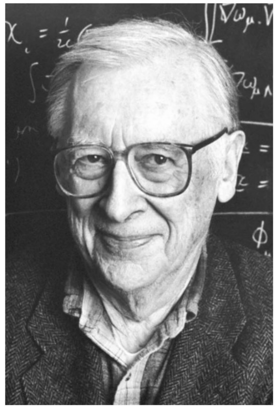


Photo from the Nobel Foundation archive.

John A. Pople

- Density Functional Theory (DFT):
- The Kohn-Sham potential v_{KS} consists of 3 terms:
  - External potential v_{ext}: generated by the nucleus
  - The Hartree potential v_{Hartree}: the Coulomb repulsion between electrons
  - Exchange-correlation potential v_{xc}: complex quantum mechanical interactions

$$v_{\mathrm{KS}}[\rho](\boldsymbol{r}) = v_{\mathrm{ext}}(\boldsymbol{r}) + v_{\mathrm{Hartree}}[\rho](\boldsymbol{r}) + v_{\mathrm{xc}}[\rho](\boldsymbol{r})$$

Density Functional Theory (DFT): Step 1

Similar to the H-F method, DFT applies self-consistency (SC) calculation:

1. Initialize electron density $\rho_0(\boldsymbol{r})$ to start the iterative procedure
    - In principle, any random positive function normalized to the total number of electrons is acceptable
    - A reasonable initiation can speed up convergence, e.g.:

$$\rho_0(\boldsymbol{r}) = \sum_{\alpha} \rho_\alpha(\boldsymbol{r} - \boldsymbol{R}_\alpha)$$

    - Where $\boldsymbol{R}_\alpha$ and ρ_α represents the position and atomic density of the nucleus

Density Functional Theory (DFT): Step 2

2. Calculate Kohn-Sham potential $v_{\mathrm{KS}} = v_{\mathrm{ext}} + v_{\mathrm{Hartree}} + v_{\mathrm{xc}}$:
   - The **external potential** v_{ext} is typically the sum of nuclear potentials:

$$v_{\mathrm{ext}}(\boldsymbol{r}) = \sum_{\alpha} v_{\alpha}(\boldsymbol{r} - \boldsymbol{R}_{\alpha})$$

   - Where v_{α} can be the Coulomb potential with the nuclear charge Z_{α}:

$$v_{\alpha}(\boldsymbol{r}) = -\frac{Z_{\alpha}}{r}$$

   - Or use other predefined pseudo-potentials for v_{α} depending on systems and tasks

Density Functional Theory (DFT): Step 2

2. Calculate Kohn-Sham potential $v_{\text{KS}} = v_{\text{ext}} + v_{\text{Hartree}} + v_{\text{xc}}$:
    - The **Hartree potential** v_{Hartree} can be calculated by either direct integration:

$$v_{\text{Hartree}}(\boldsymbol{r}) = \int \frac{\rho(\boldsymbol{r}')}{|\boldsymbol{r} - \boldsymbol{r}'|} d^3\boldsymbol{r}'$$

    - Or solving Poisson's equation:

$$\nabla^2 v_{\text{Hartree}}(\boldsymbol{r}) = -4\pi\rho(\boldsymbol{r})$$

Density Functional Theory (DFT): Step 2

2. Calculate Kohn-Sham potential $v_{\mathrm{KS}} = v_{\mathrm{ext}} + v_{\mathrm{Hartree}} + v_{\mathrm{xc}}$:
   - The **exchange-correlation potential** v_{xc} consists of all the other non-classical interactions between electrons, defined as functional derivative of exchange-correlation energy:

$$v_{\mathrm{XC}}[\rho](\boldsymbol{r}) = \frac{\delta E_{\mathrm{xc}}}{\delta \rho(\boldsymbol{r})}$$

   - The exchange-correlation energy E_{xc} can be calculated with different designs and approximations, for example the simplest local-density approximation (LDA):

$$E_{\mathrm{xc}} = \int d^3\boldsymbol{r}\ \varepsilon^{\mathrm{HEG}}\big(\rho(\boldsymbol{r})\big)$$

   - Where $\varepsilon^{\mathrm{HEG}}(\rho)$ is the exchange-correlation energy per unit volume of homogeneous electron gas (HEG) of a given density ρ, which can be tabulated for different densities

Density Functional Theory (DFT): Step 3

3. Solving the Kohn-Sham equation:

$$\left(-\frac{\hbar\nabla^2}{2m} + v_{\mathrm{KS}}[\rho](\boldsymbol{r})\right)\varphi_i(\boldsymbol{r}) = \varepsilon_i\varphi_i(\boldsymbol{r})$$

$$\widehat{H}_{\mathrm{KS}}\varphi_i(\boldsymbol{r}) = \varepsilon_i\varphi_i(\boldsymbol{r})$$

- Where $\widehat{H}_{\mathrm{KS}} = -\frac{\hbar\nabla^2}{2m} + v_{\mathrm{KS}}[\rho](\boldsymbol{r})$ is the Kohn-Sham Hamiltonian operator
- Apply the Hamiltonian operator to the basis set $\{\phi_i(r)\}$ to obtain Hamiltonian matrix H_{KS}:

$$H_{ij} = \int \phi_i^*(\boldsymbol{r})\widehat{H}\phi_j(\boldsymbol{r})d\boldsymbol{r}$$

Density Functional Theory (DFT): Step 3

3. Solving the Kohn-Sham equation:

- The Hamiltonian matrix $\boldsymbol{H}_{\mathrm{KS}}$ is an $N \times N$ symmetrical matrix
- N is the total number of basis functions, with the basic assumption in DFT that the Kohn-Sham wave function is the **linear combination** of the basis set $\{\phi_i(r)\}$:

$$\varphi_i(\boldsymbol{r}) = \sum_j c_{ij}\phi_j(\boldsymbol{r})$$

- Where c_{ij} is the are the coefficients of the basis functions in the expansion of the ith Kohn-Sham wave function φ_i
- Thus, each element H_{ij} in the H_{KS} describes the interaction between basis ϕ_i and ϕ_j

Density Functional Theory (DFT): Step 3

3. Solving the Kohn-Sham equation:

- Once the Kohn-Sham Hamiltonian matrix $\boldsymbol{H}_{\mathrm{KS}}$ is constructed, the Kohn-Sham equations turned into an eigenvalue problem:

$$\boldsymbol{H}_{\mathrm{KS}}\boldsymbol{C} = \boldsymbol{E}\boldsymbol{C}$$

- where $\boldsymbol{C}$ is the matrix of coefficients, and $\boldsymbol{E}$ is the diagonal matrix of eigenvalues (orbital energies)
- Then, the Kohn-Sham wave functions can be constructed using $\boldsymbol{C}$ and basis set:

$$\varphi_i(\boldsymbol{r}) = \sum_j c_{ij}\phi_j(\boldsymbol{r})$$

Density Functional Theory (DFT): Step 4&5

4. Calculate new electron density from obtained wave functions:

$$\rho_1(\boldsymbol{r}) = \sum_i |\varphi_i(\boldsymbol{r})|^2$$

5. Compare $\rho_1(\boldsymbol{r})$ and initial guess $\rho_0(\boldsymbol{r})$:

- If the difference between two densities is lower than a user-defined criterion η:

$$|\rho_1(\boldsymbol{r}) - \rho_0(\boldsymbol{r})| < \eta$$

- The calculation is considered as **self-consistent**
- With correctly calculated form of wave functions and Hamiltonian matrix, several observables can be evaluated, including total energy, band structure, conductivity, etc.

Density Functional Theory (DFT): Iteration

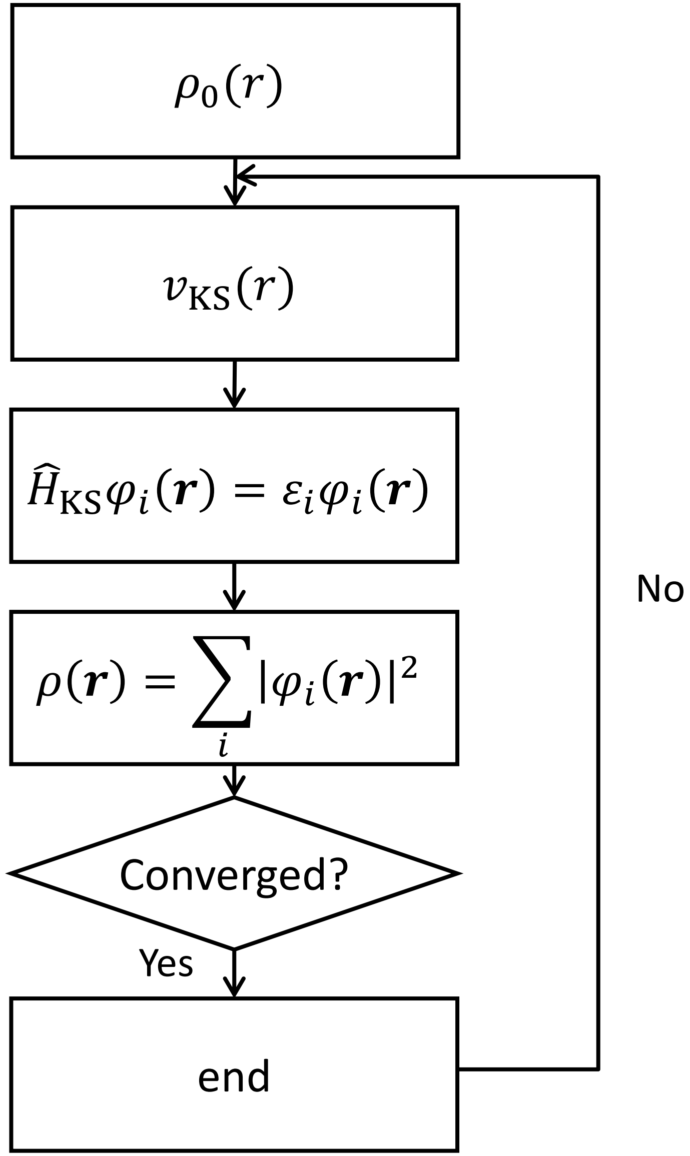
6. If the convergence criterion has not been reached:

- A new iteration begins with $\rho_0'(\boldsymbol{r})$ of several different options:
  - A random **guess** again
  - Use the output of the previous cycle: always leads to **instabilities**
  - **Mix** the last output and the original input:

$$\rho_0'(\boldsymbol{r}) = \beta\rho_0(\boldsymbol{r}) + (1-\beta)\rho_1(\boldsymbol{r})$$

  - The mixing parameter β is typically chosen to be around 0.3
  - Other mixing strategies similar to the one above
- Repeat step 2 to 5 until converged, highly dependent on calculation conditions and hyperparameters

Density Functional Theory (DFT): Iteration



$\rho_0(r)$

$v_{\mathrm{KS}}(r)$

$\hat{H}_{\mathrm{KS}}\varphi_i(\boldsymbol{r}) = \varepsilon_i\varphi_i(\boldsymbol{r})$

$\rho(\boldsymbol{r}) = \sum_i |\varphi_i(\boldsymbol{r})|^2$

Converged?

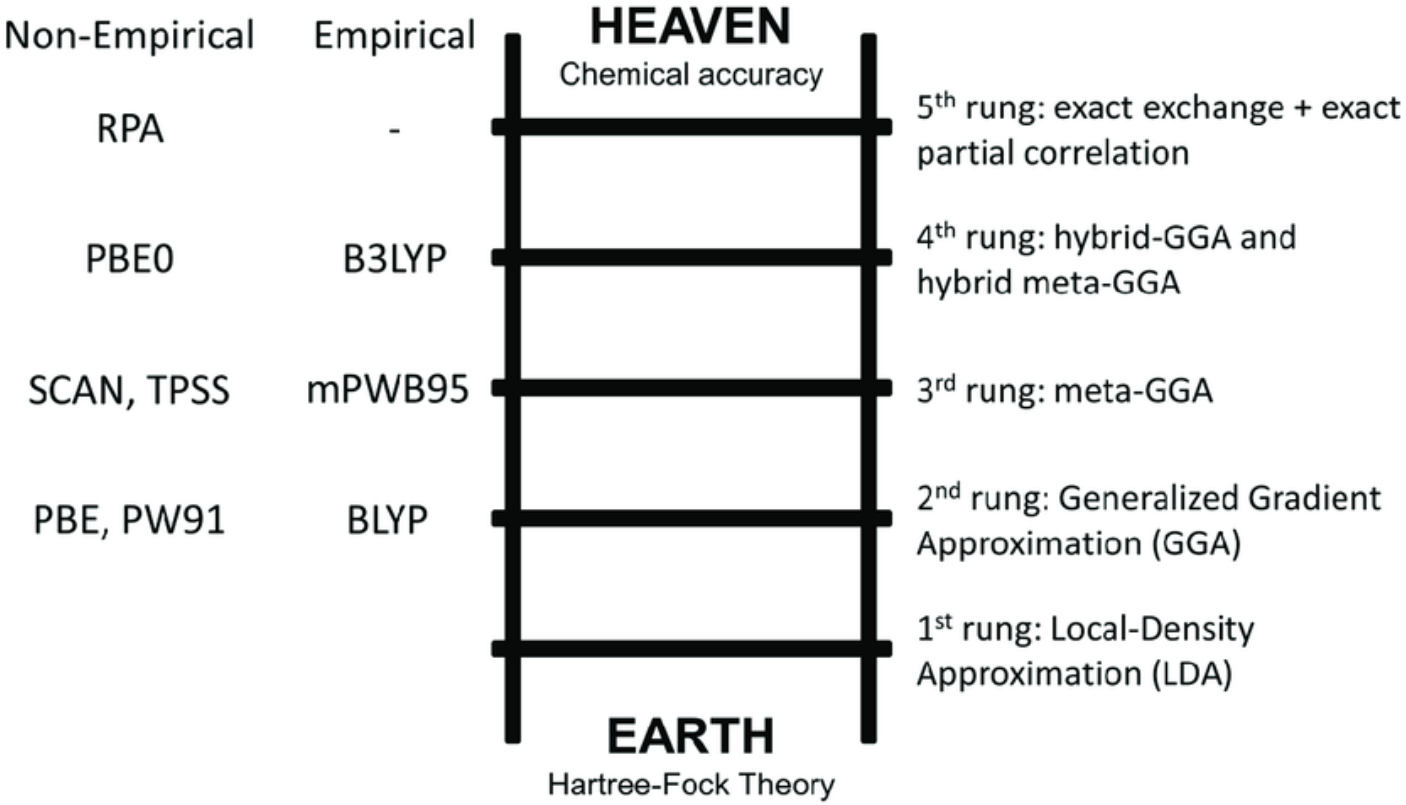
Yes

No

end

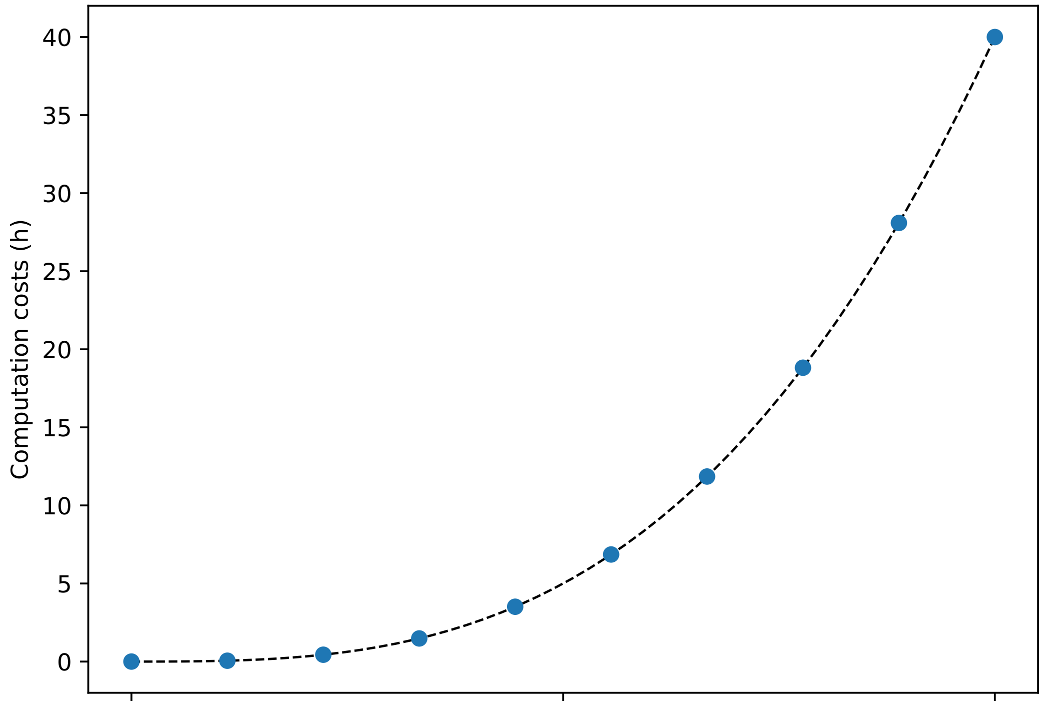
Density Functional Theory (DFT): Limitations

- Jacob’s Ladder:
  - The **accuracy** of DFT calculation mainly depends on the choice of **exchange-correlation functional**
  - More complexity brings **better accuracy**, but also **higher expenses**

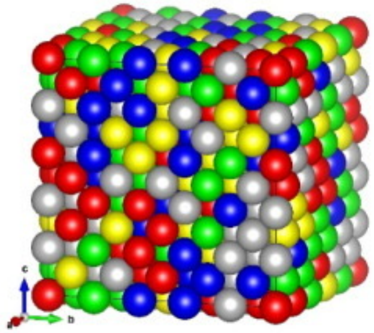


Density Functional Theory (DFT): Limitations

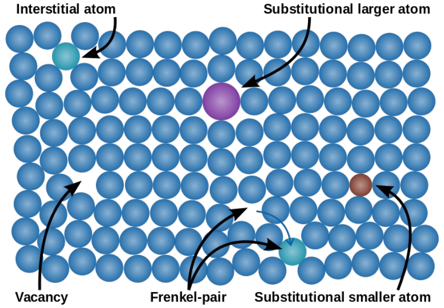
- DFT calculation is still computationally expensive
- Especially for large-scale structures consists 10^3 atoms



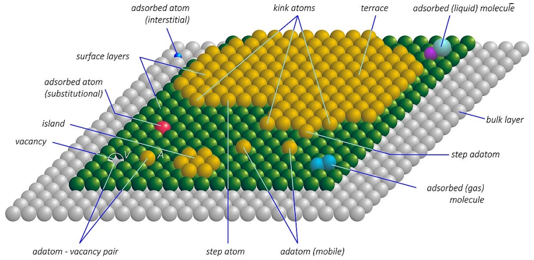
Complexity: $O(N^3)$



Alloy



Defects



Surface

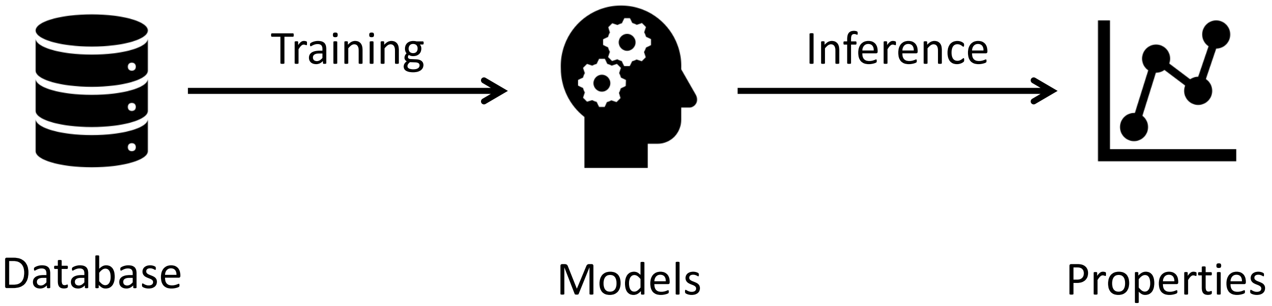
Outlines

- Intro to Materials Science
  - Crystalline Materials
  - Structure-Property Relationship
  - DFT Calculation
- AI in Materials Science
  - Artificial intelligence ab initio (AI^2) methods
  - Directly Bridging
  - Inverse Problem
  - AI as a Powerful Assistance
  - Outside and Beyond Crystals

AI in Materials Science

Artificial intelligence ab initio (AI^2) methods

Deep Learning Approaches: Data Driven



Database

Training

Models

Inference

Properties

MATERIALS PROJECT

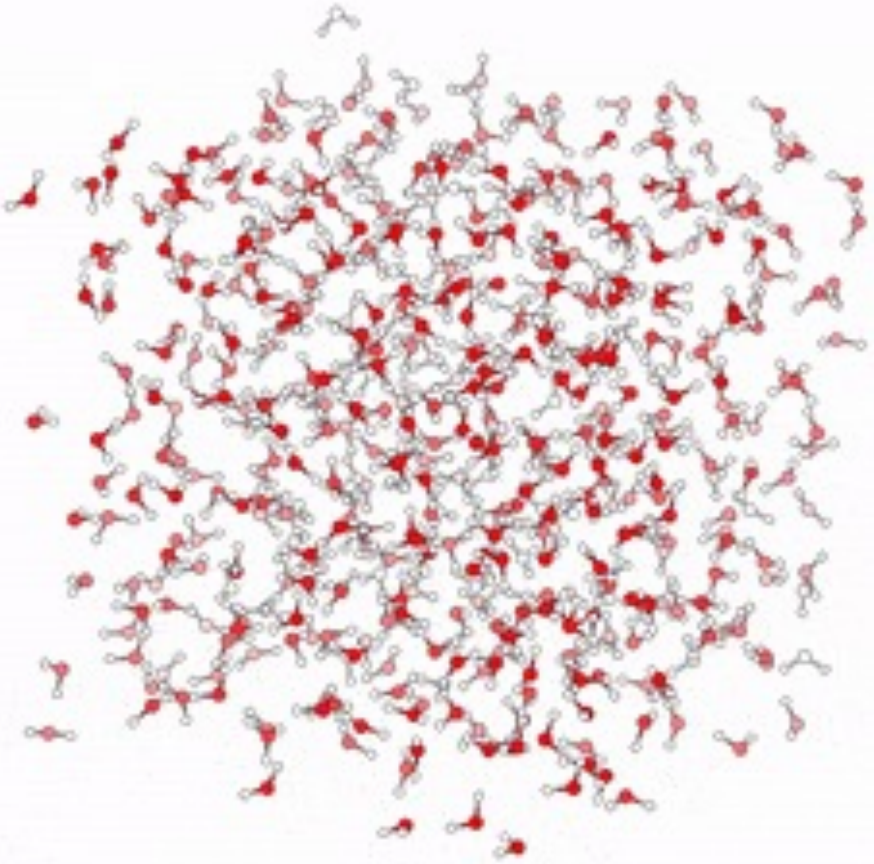
MATERIALSCLOUD

OQMD
The Open Quantum
Materials Database

ICSD

Deep Learning Approaches: DPMD

Deep Potential Molecular Dynamics (DPMD):

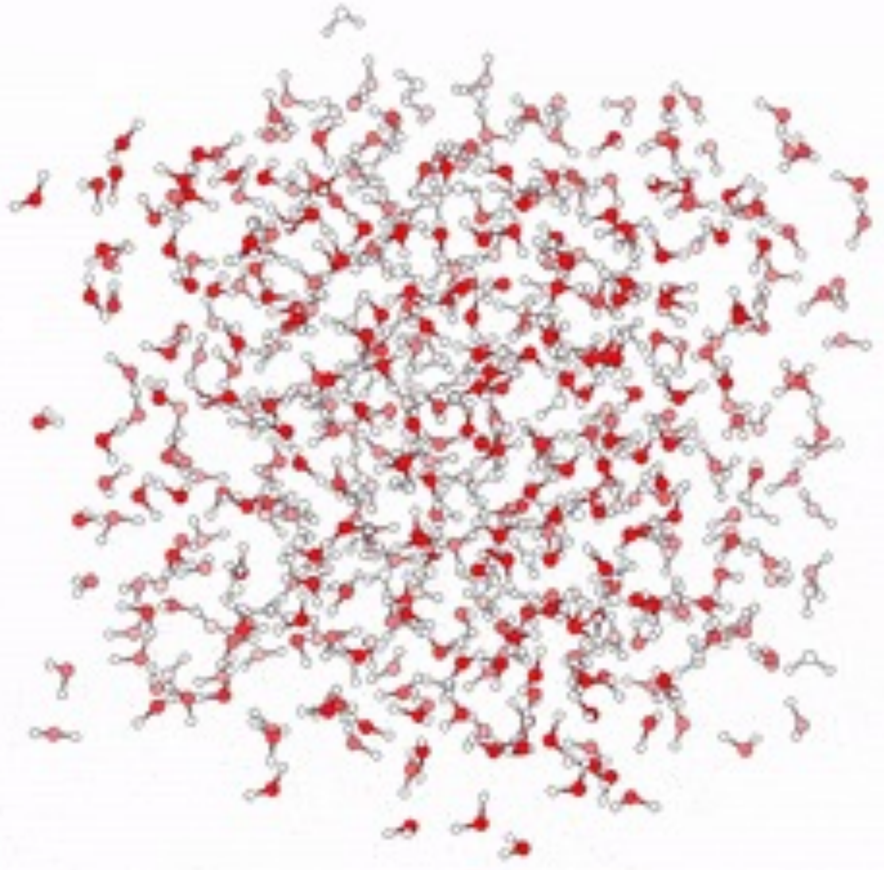


- In each frame of the **ab initio molecular dynamics** (AIMD) simulation, the **potential surface** $E(\boldsymbol{R}_1, \boldsymbol{R}_2, \dots, \boldsymbol{R}_N)$ of the structure is determined through **Quantum Mechanics** (QM) calculation, where the $\boldsymbol{R}_i$ is the coordinates of the atom i
- The **forces** on each atom can be calculated as the negative derivative of potential to the coordinates:

$$\boldsymbol{F}_i = -\nabla E = -\left(\frac{\partial E}{\partial x_i}, \frac{\partial E}{\partial y_i}, \frac{\partial E}{\partial z_i}\right)$$

Deep Learning Approaches: DPMD

Deep Potential Molecular Dynamics (DPMD):



- MD computes the half-step velocities of the atoms according to the forces and simulation time step Δt:

$$\boldsymbol{v}_i\left(t+\frac{\Delta t}{2}\right)=\boldsymbol{v}_i(t)+\frac{\Delta t}{2}\boldsymbol{a}_i(t)$$

- Where $\boldsymbol{a}_i(t)=\frac{\boldsymbol{F}_i(t)}{m_i}$ is the acceleration
- At last, MD computes the atom coordinates and full-step velocities, generating a new frame:

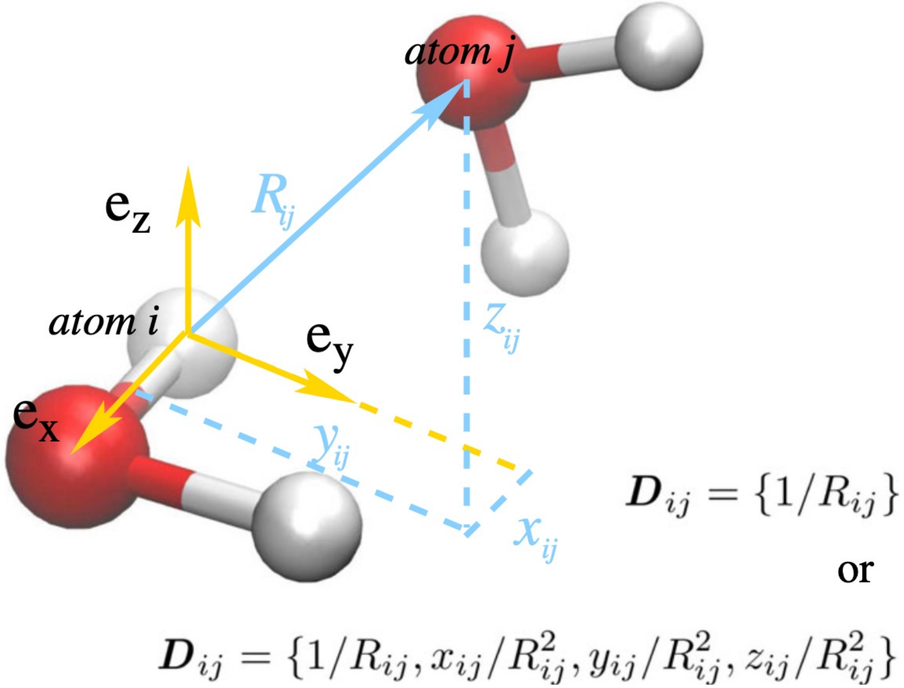
$$\boldsymbol{R}_i(t+\Delta t)=\boldsymbol{R}_i(t)+\Delta t\boldsymbol{v}_i\left(t+\frac{\Delta t}{2}\right)$$

$$\boldsymbol{v}_i(t+\Delta t)=\boldsymbol{v}_i\left(t+\frac{\Delta t}{2}\right)+\frac{\Delta t}{2}\boldsymbol{a}_i(t+\Delta t)$$

- Since each iteration involves QM calculation, AIMD can be **extremely time and energy consuming**

Deep Learning Approaches: DPMD

Deep Potential Molecular Dynamics (DPMD) [1]:



- In the DPMD framework, a set of **local orthogonal unit bases** $\{\boldsymbol{e}_x, \boldsymbol{e}_y, \boldsymbol{e}_z\}$ centered in the target atom i is constructed:
    - $\boldsymbol{e}_x$: parallel to the O-H bond
    - $\boldsymbol{e}_z$: perpendicular to H-O-H water molecule plane
    - $\boldsymbol{e}_y$: $\boldsymbol{e}_z \times \boldsymbol{e}_x$
- The relative coordinate of a neighboring atom j can be determined:

$$\boldsymbol{R}_{ij} = x_{ij}\boldsymbol{e}_x + y_{ij}\boldsymbol{e}_y + z_{ij}\boldsymbol{e}_z$$

- The input coordinates of the Neural Network (NN) can be:
    - With both radial and angular information:

$$\boldsymbol{D}_{ij} = \{1/\boldsymbol{R}_{ij}, x_{ij}/\boldsymbol{R}_{ij}^2, y_{ij}/\boldsymbol{R}_{ij}^2, z_{ij}/\boldsymbol{R}_{ij}^2,\}$$

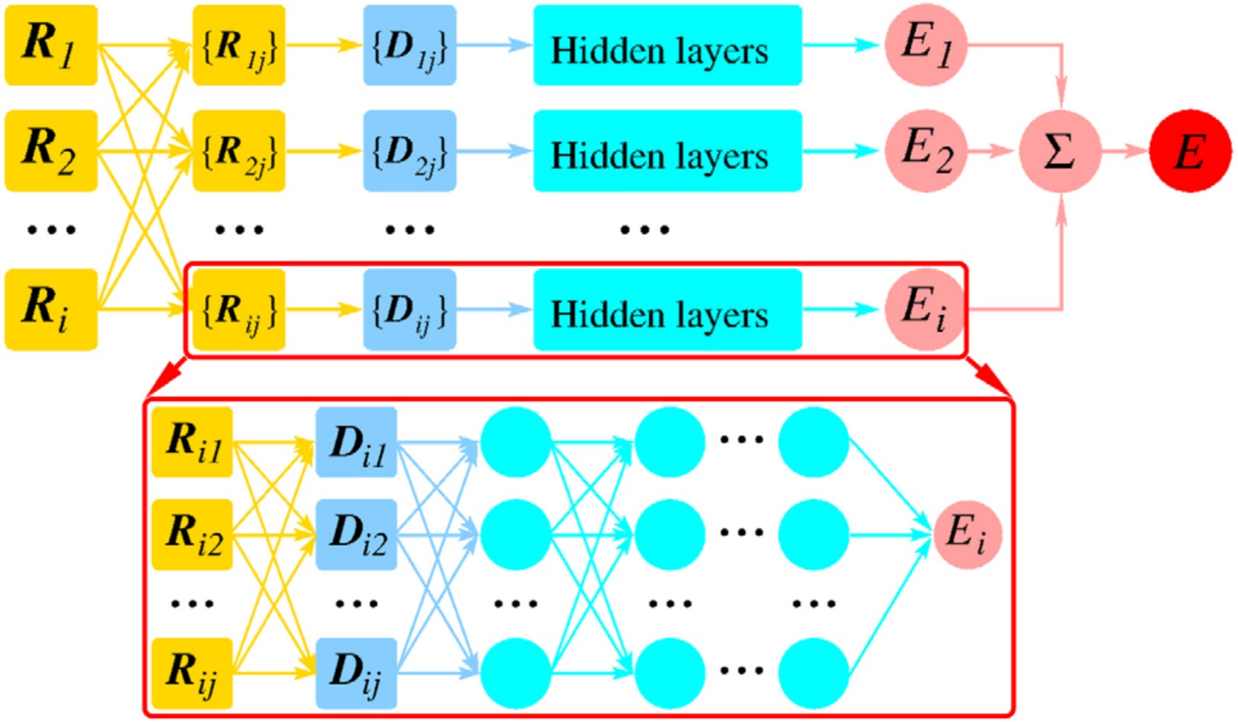
    - Or with radial information only:

$$\boldsymbol{D}_{ij} = \{1/\boldsymbol{R}_{ij}\}$$

[1] Zhang, Linfeng, et al. "Deep potential molecular dynamics: a scalable model with the accuracy of quantum mechanics." *Physical review letters* 120.14 (2018): 143001.

Deep Learning Approaches: DPMD

Deep Potential Molecular Dynamics (DPMD):



D_{ij} of empty neighbors are set to 0

- In the DPMD workflow, the relative coordinates $\{\boldsymbol{R}_{ij}\}_{i=1,2,\ldots}$ are first computed from absolute coordinates $\boldsymbol{R}$, then converted into $\{\boldsymbol{D}_{ij}\}_{i=1,2,\ldots}$
- The NN outputs the energy of each atom E_i and sum them together to obtain the total energy E
- The loss function is defined as a multi-task loss:

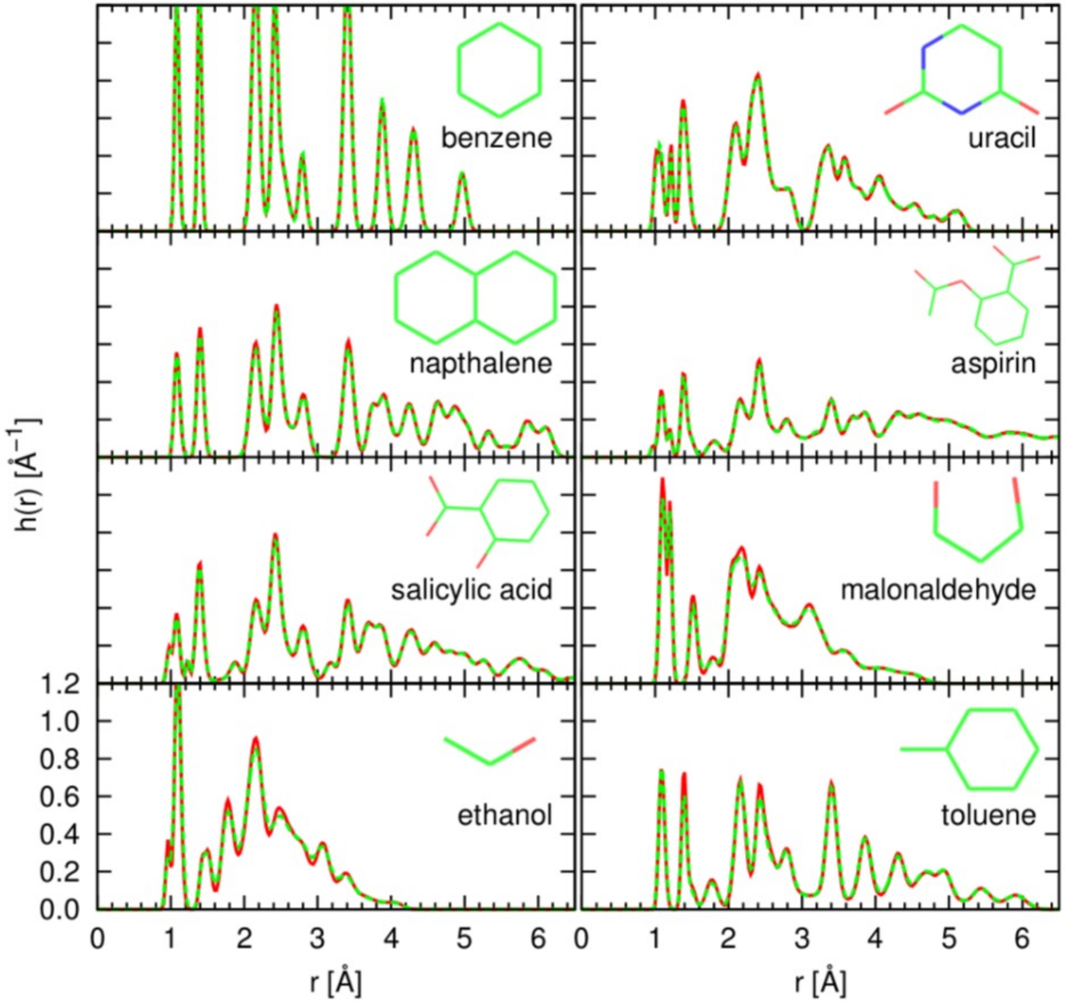
$$L = p_\epsilon \Delta\epsilon^2 + \frac{p_f}{3N}\sum_i |\Delta \boldsymbol{F}_i|^2 + \frac{p_\xi}{9}\left|\left|\Delta\xi\right|\right|^2$$

- Consists of 3 terms:
  - Energy per atom ϵ
  - Force on each atom $\boldsymbol{F}_i$,
  - Virial tensor ξ (measures internal stress)
- Where factors p are loss weight

Deep Learning Approaches: DPMD

Deep Potential Molecular Dynamics (DPMD):

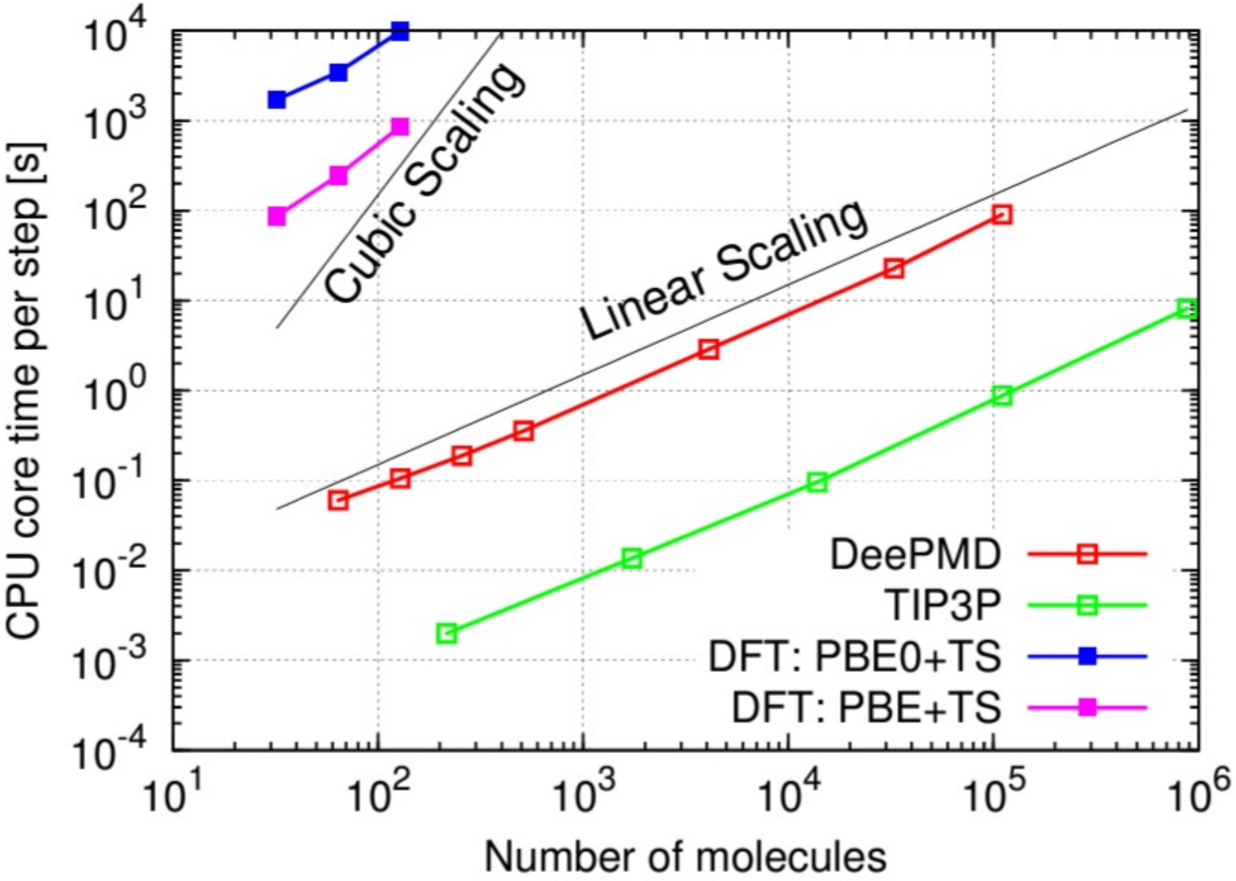
- **Relaxation**: structures are updated iteratively through AIMD, evolving from an **unstable structure** to a **balanced state**
- Comparing the **interatomic distance** distribution after relaxation via both **AIMD** and **DPMD**, the obtained balanced structures fit quite well
- Recall that the **NN does not directly predict the interatomic distance**, exhibiting the **convincing accuracy** of the deep potential force field



Deep Learning Approaches: DPMD

Deep Potential Molecular Dynamics (DPMD):

- Since the time complexity of **DPMD** is $O(N)$, it tremendously **lowers the computation costs**, especially for larger systems

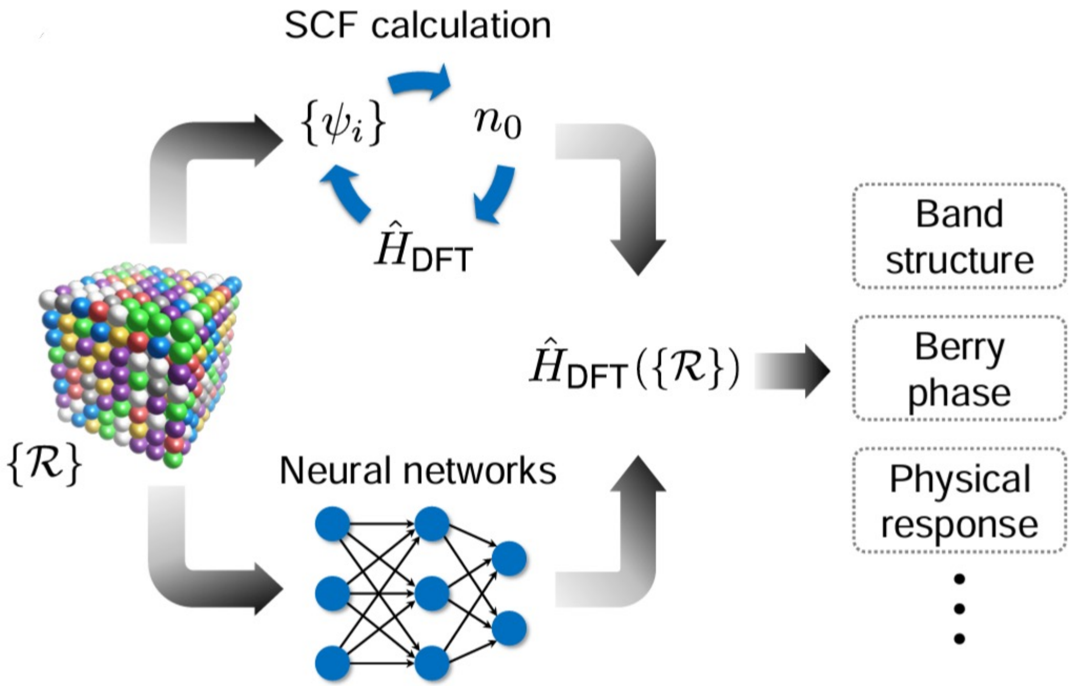


Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH) [1]:

Tips about DFT Hamiltonian $\widehat{H}_{\mathrm{DFT}}$:

- Recall that $\widehat{H}_{\mathrm{DFT}}$ describes **the interactions between basis functions** $\{\phi_i\}$
- $\widehat{H}_{\mathrm{DFT}}$ is determined by the structure $\{\boldsymbol{\mathcal{R}}\}$
- Many observables are deterministically computed from $\widehat{H}_{\mathrm{DFT}}$
- $\widehat{H}_{\mathrm{DFT}}$ has to be obtained through computational expensive SCF calculation



[1] Li, He, et al. "Deep-learning density functional theory Hamiltonian for efficient ab initio electronic-structure calculation." *Nature Computational Science* 2.6 (2022): 367-377.

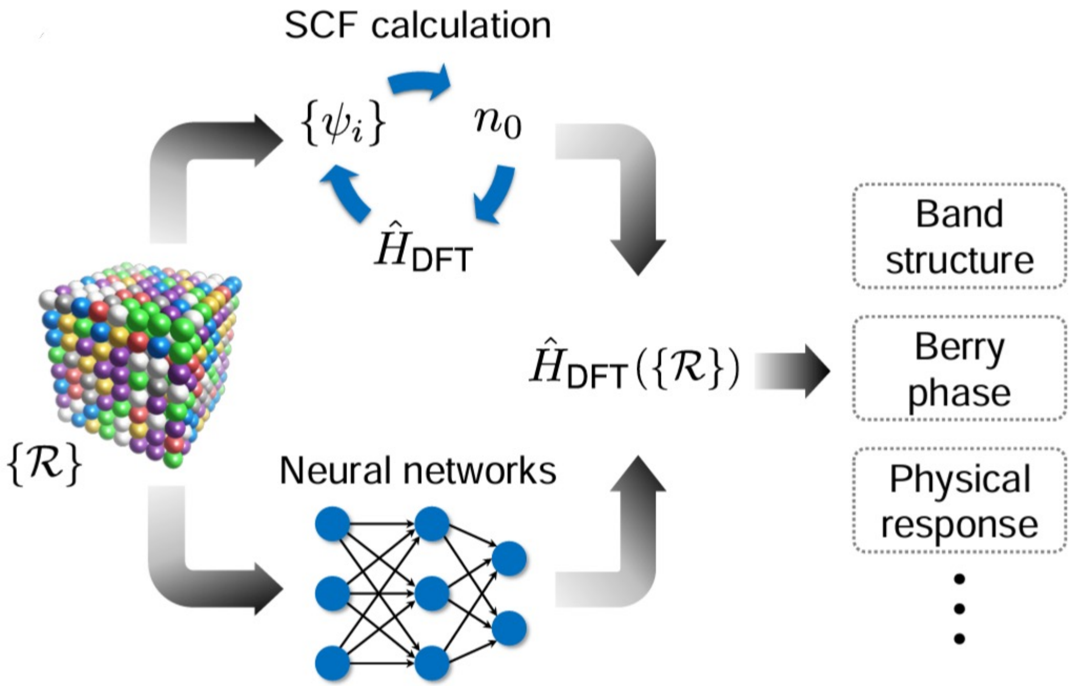
Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

Modeling Structure-$\widehat{H}_{\mathrm{DFT}}$ Relationship

||

Modeling Structure-Property Relationship



Deep Learning Approaches: DeepH

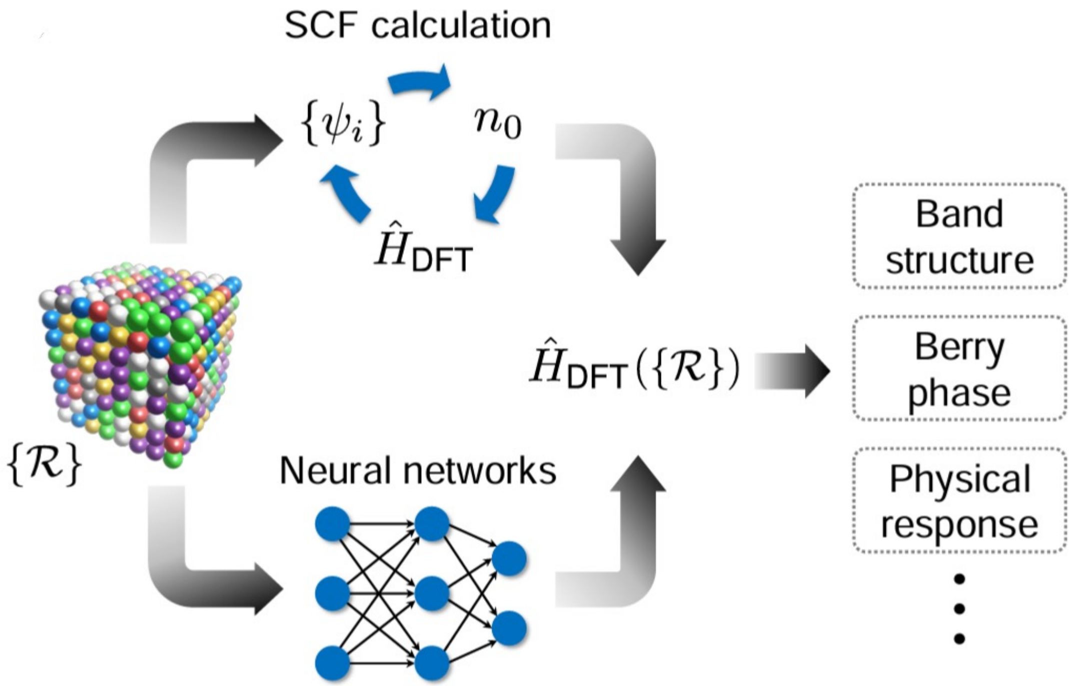
Deep Learning DFT Hamiltonian (DeepH):

Fundamental Facts:

- DFT does support high-accuracy calculation with acceptable expenses for **small systems**
- DFT is NOT suitable for calculating **large systems**

Key Problem:

- How to learn from small system DFT data and **expand/generalize** to large systems?

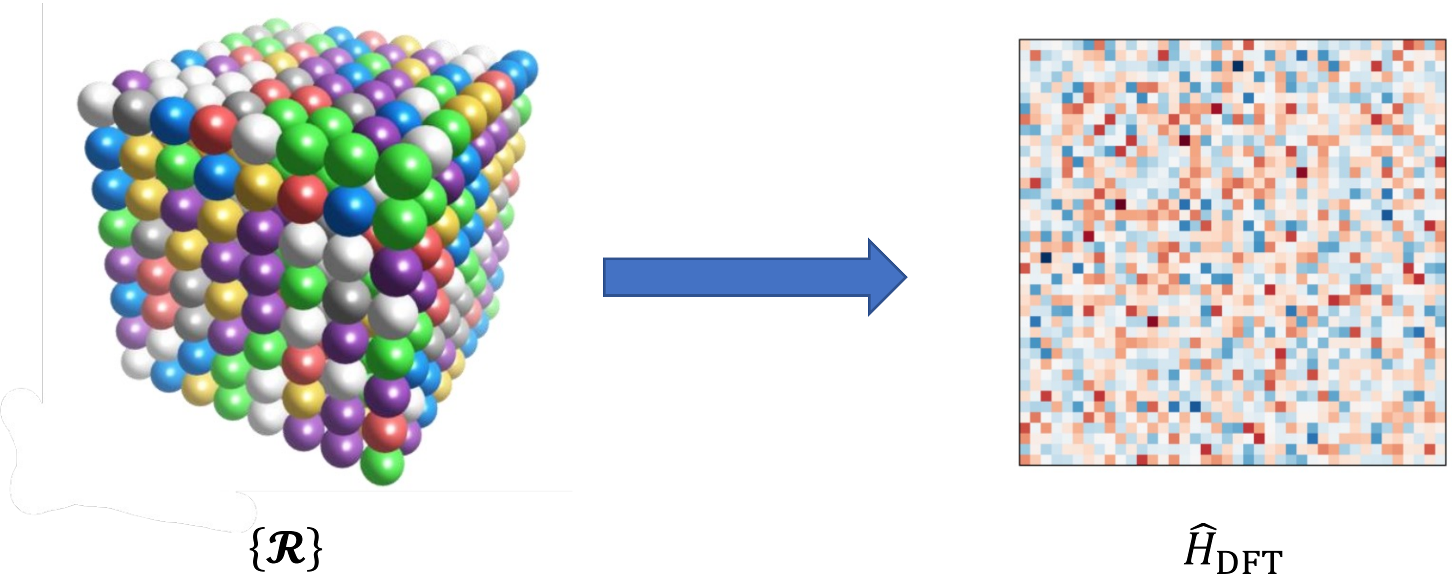


Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

Challenges:

1. Infinite dimension of $\widehat{H}_{\mathrm{DFT}}$ in the extended systems
2. SO(3) Rotation Equivariance of $\widehat{H}_{\mathrm{DFT}}$

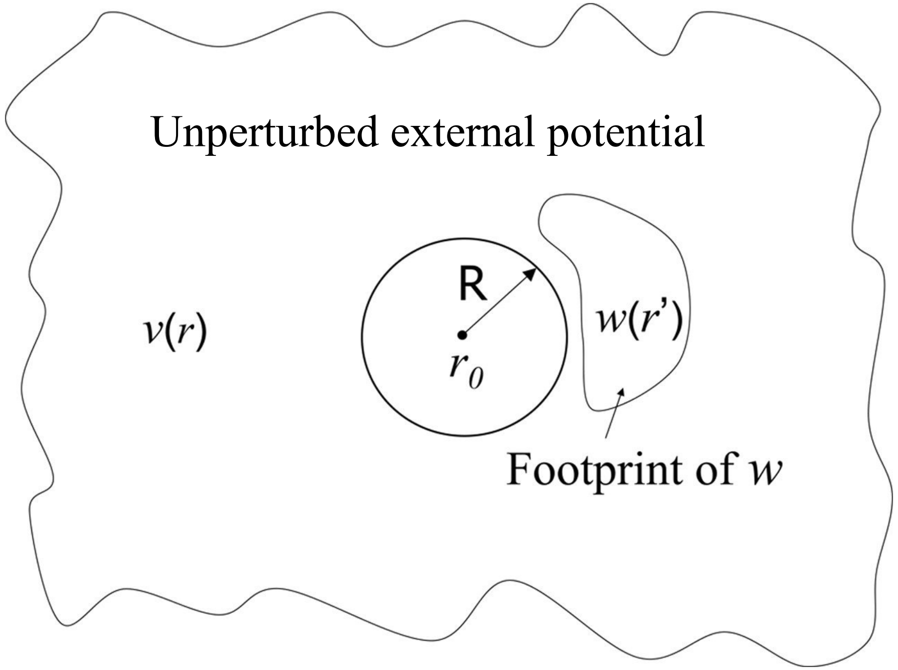


Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

Important Prior Physics Knowledge: Nearsightedness Principle [1]

- In the many-body system, **local electron properties** at r_0 **does not** response to the distant, local **perturbing** potential $w(r')$ outside a sphere of radius R



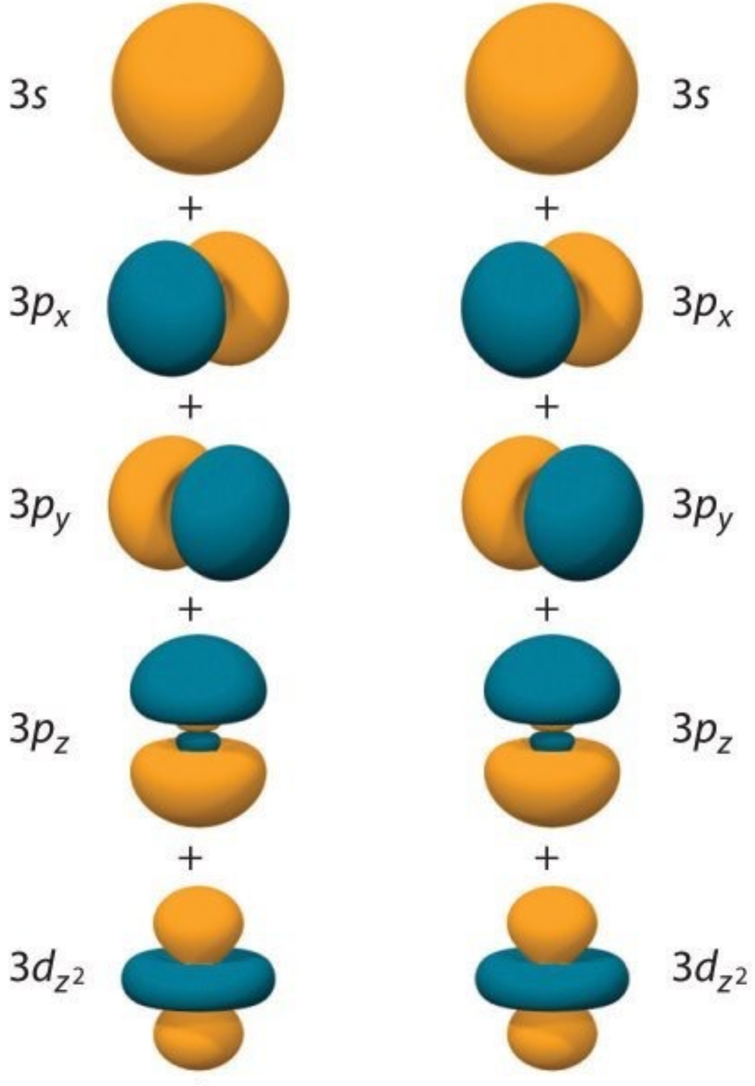
[1] Prodan, Emil, and Walter Kohn. "Nearsightedness of electronic matter." *Proceedings of the National Academy of Sciences* 102.33 (2005): 11635-11638.

Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

Localized Basis:

- Atomic orbitals
- Interact only when overlapped
- Well-defined rotation transformations



Deep Learning Approaches: DeepH

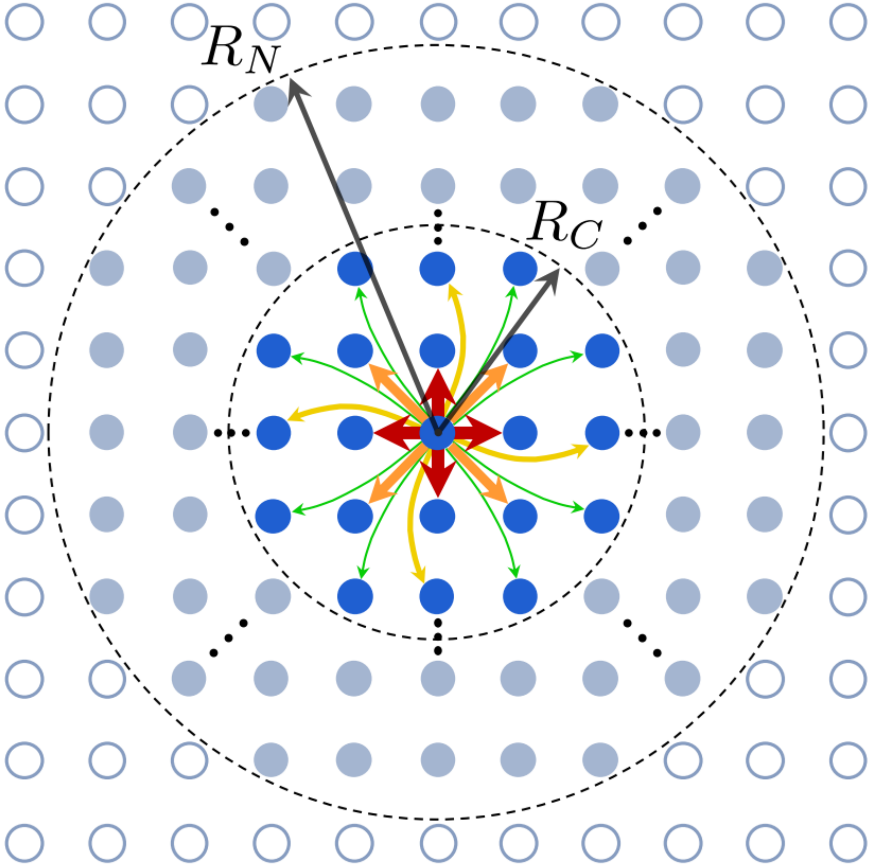
Deep Learning DFT Hamiltonian (DeepH):

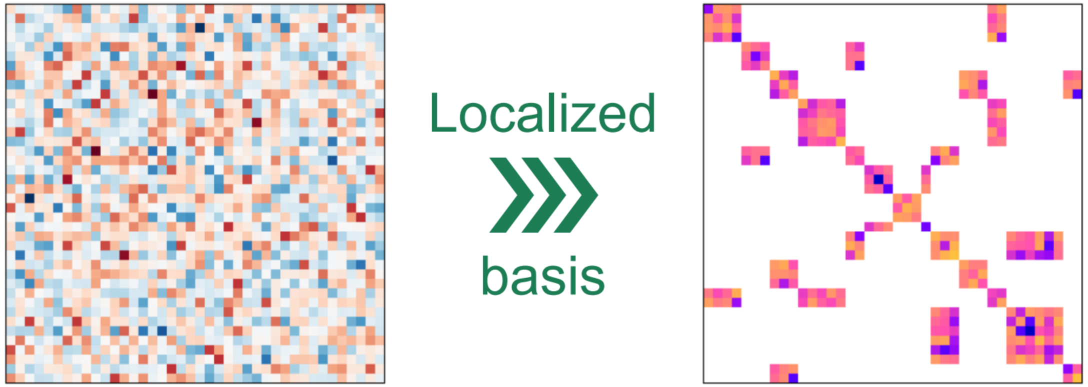
Localized Basis: Sparseness

- Only H_{ij} between neighboring atoms (within R_C, determined by the spread of orbitals, few Å) are nonzero

Nearsightedness Principle:

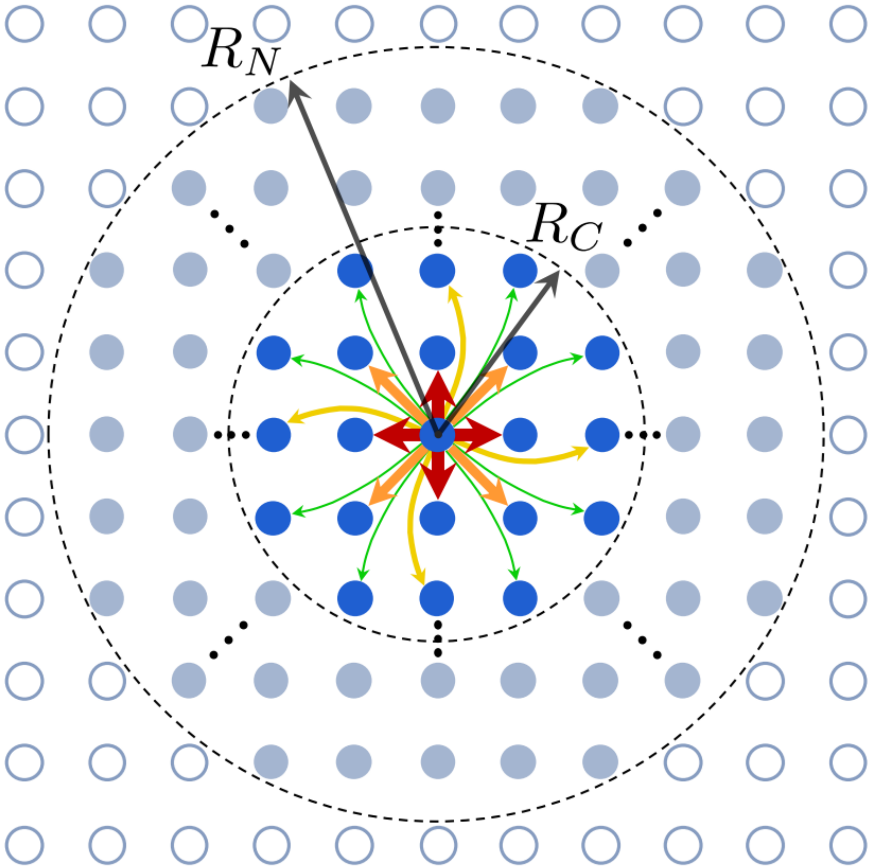
- Only information of neighborhood (within R_N) should be considered





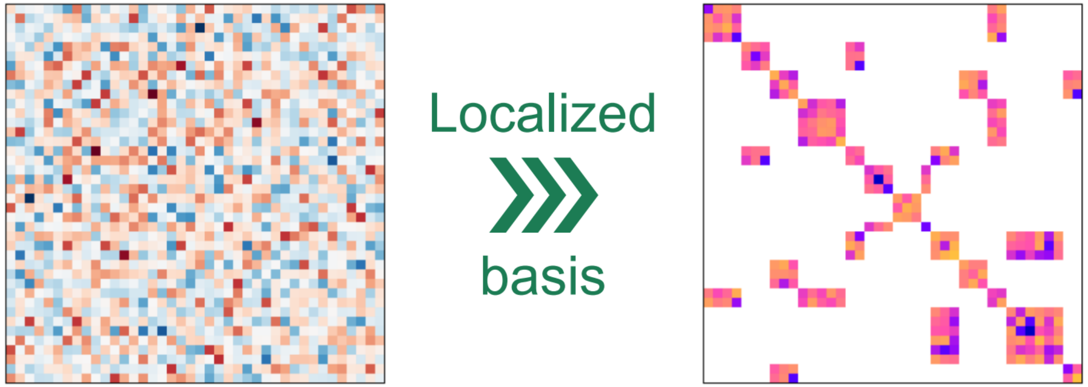
Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):



Localized Basis:

- Modeling Hamiltonian blocks from **local interactions**
- Does NOT respond to long-range configurations



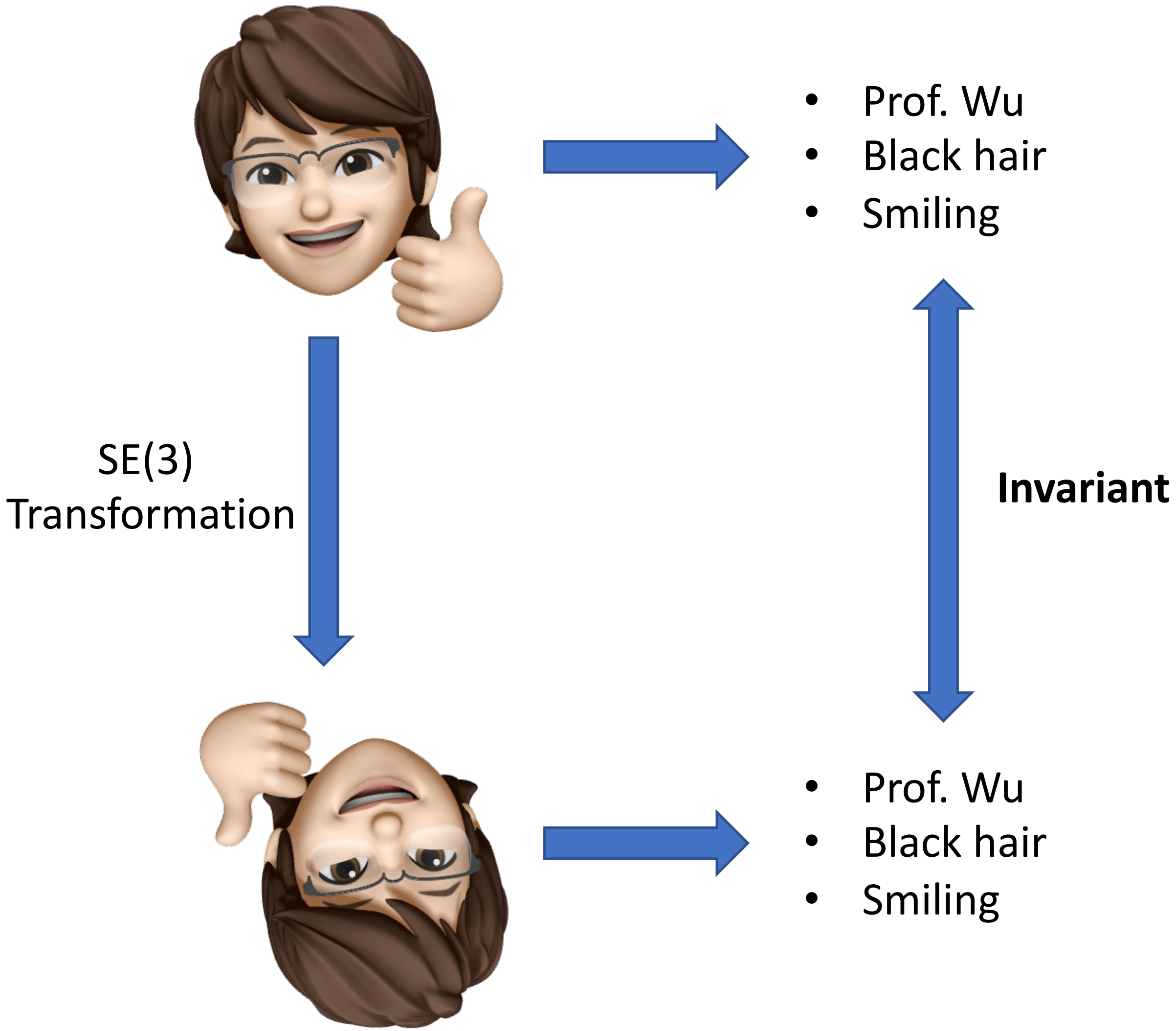
Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

SE(3) Equivariance vs. Invariance:

- SE(3) (Special Euclidean group in 3D) includes 3 translation transformation and 3 rotation transformation

- SE(3) Invariance:
  - The output does NOT change with the transformation of the input structure



- Prof. Wu
- Black hair
- Smiling

SE(3) Transformation

Invariant

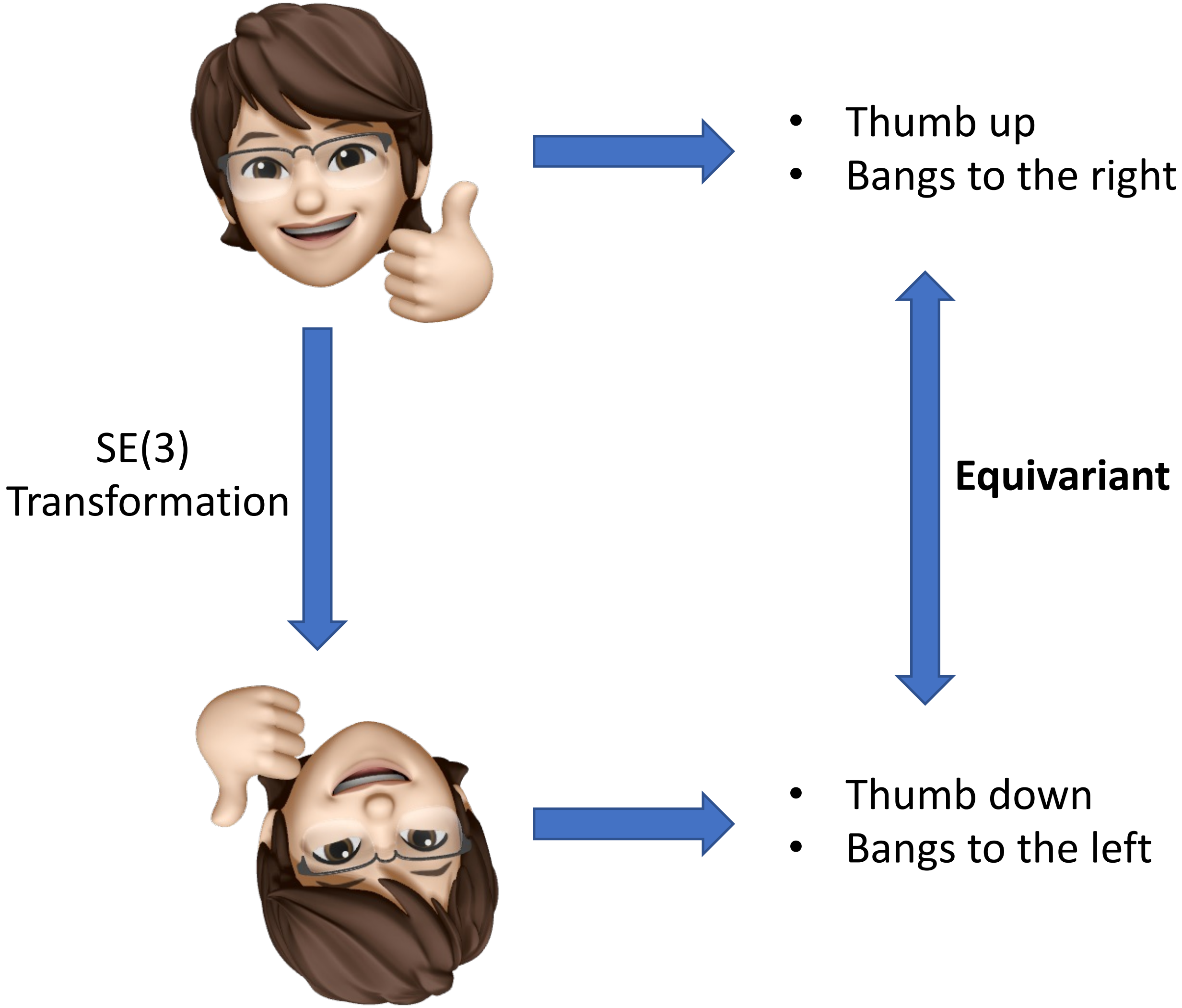
- Prof. Wu
- Black hair
- Smiling

Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

SE(3) Equivariance vs. Invariance:

- SE(3) (Special Euclidean group in 3D) includes 3 translation transformation and 3 rotation transformation
- SE(3) Equivariance:
  - The output changes together with the transformation of the input structure



- Thumb up
- Bangs to the right

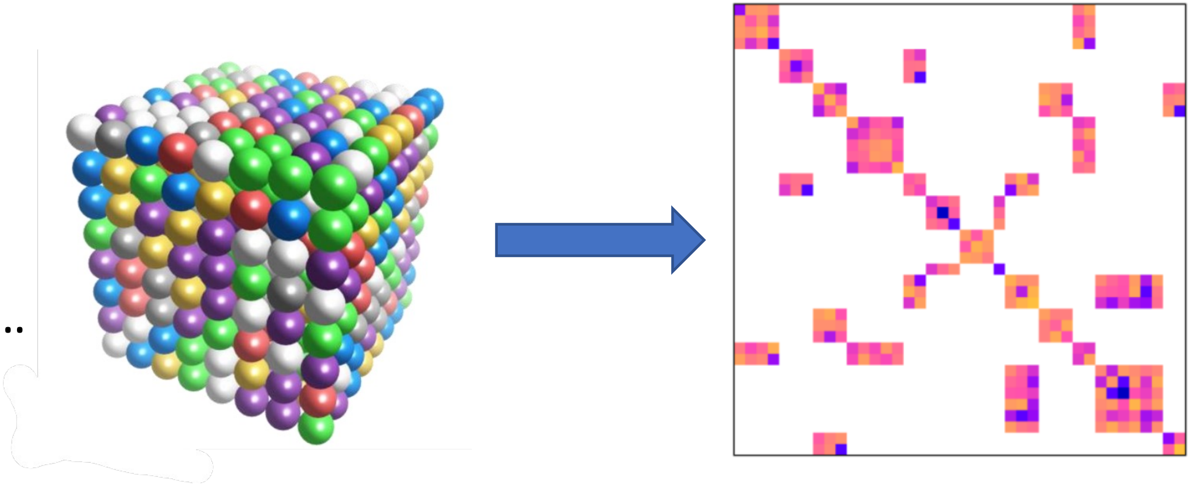
SE(3) Transformation

Equivariant

- Thumb down
- Bangs to the left

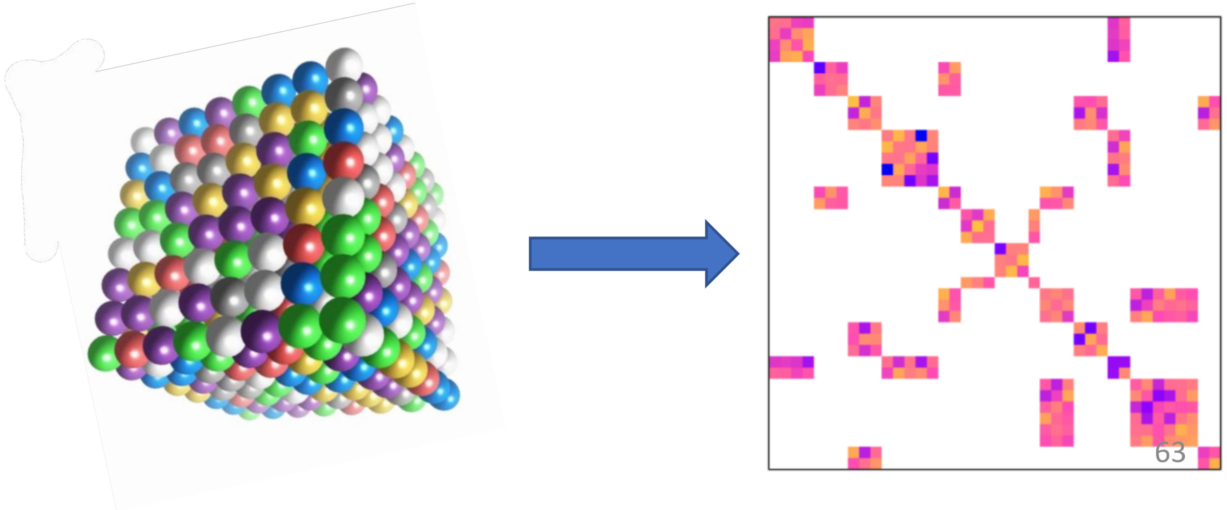
Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):



SE(3) Equivariance vs. Invariance:

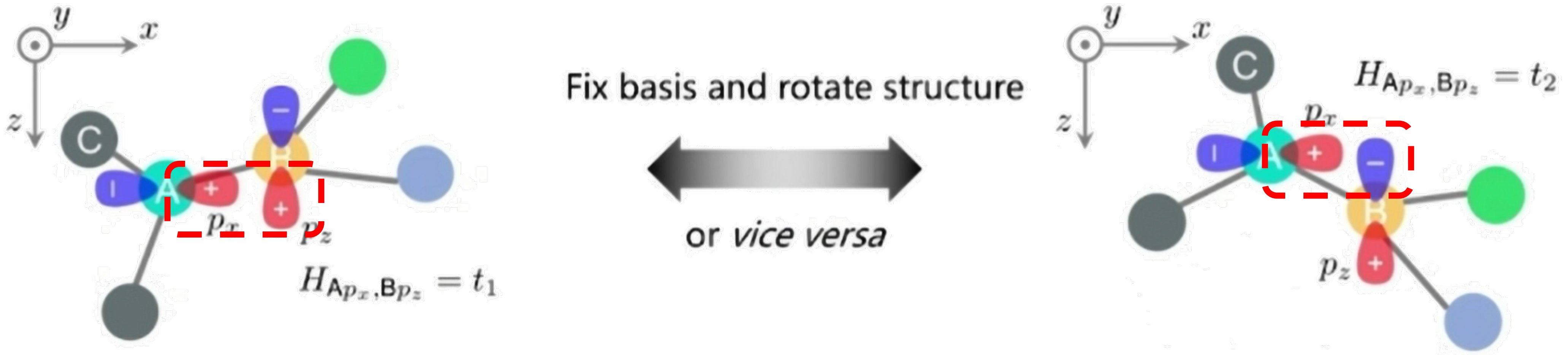
- **Invariant** quantity: total energy, band gap, …
- **Equivariant** quantity: $\widehat{H}_{\mathrm{DFT}}$, force field, …



Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

Global coordinates: rotation transformation

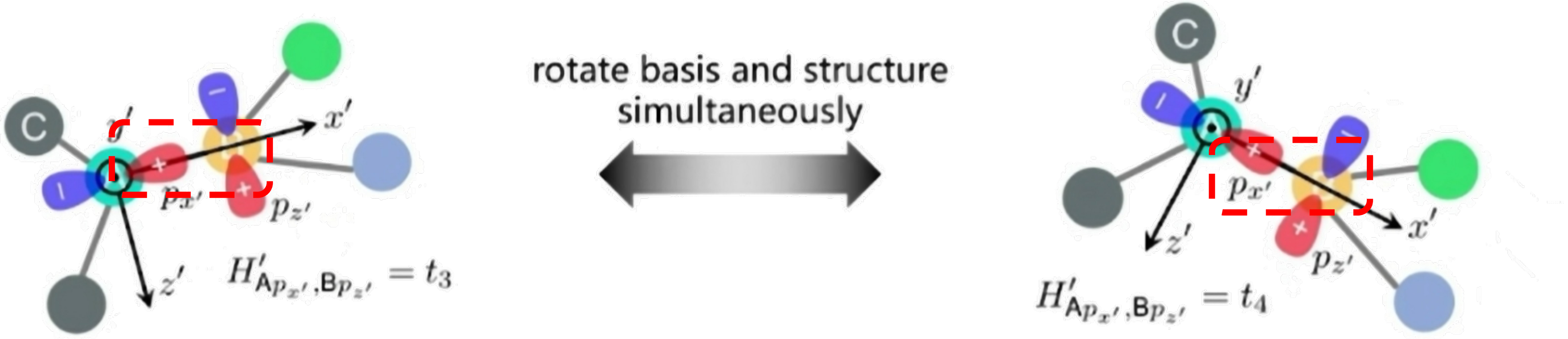


The overlaps between orbitals are **misoriented**

Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

Local coordinates: rotation transformation



The overlaps between orbitals remain **unchanged**

Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

Solution to $\widehat{H}_{\mathrm{DFT}}$ equivariance requirement:

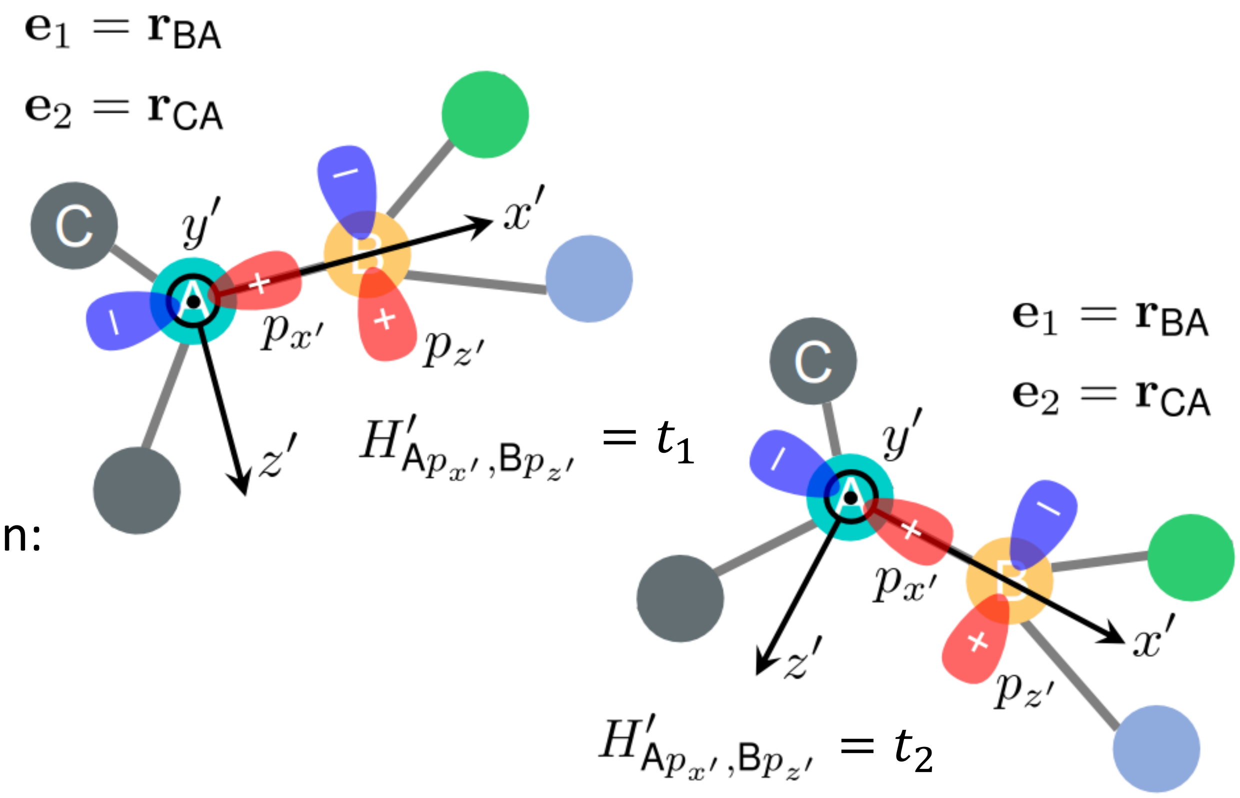
- Similar to DPMD
- Local coordinates $\{\hat{x}', \hat{y}', \hat{z}'\}$ for bond AB:

$$\hat{x}' = \frac{\boldsymbol{e}_1}{\boldsymbol{e}_1}, \hat{y}' = \frac{\boldsymbol{e}_1 \times \boldsymbol{e}_2}{|\boldsymbol{e}_1 \times \boldsymbol{e}_2|}, \hat{z}' = \hat{x}' \times \hat{y}'$$

- $\boldsymbol{e}_1 = \mathbf{r}_{\mathrm{BA}}$
- $\boldsymbol{e}_2$: second nearest $\mathbf{r}_{\mathrm{kA}}$ non-parallel to $\boldsymbol{e}_1$
- Ensure the $\widehat{H}_{\mathrm{DFT}}$ element invariant to rotation:

$$t_1 = t_2$$

- $p_{x'}, p_{z'}$: p orbitals of different angular momentum quantum numbers



Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

Solution to $\widehat{H}_{\mathrm{DFT}}$ equivariance requirement:

- The rotation transformation from DFT global coordinates to local coordinates of bond AB:

$$R^{AB} = (\hat{x}', \hat{y}', \hat{z}')$$

- with $\hat{x}', \hat{y}', \hat{z}'$ being column vectors
- And $\widehat{H}'_{\mathrm{DFT}}$ under **local coordinates** can be obtained for training:

$$H'_{A\alpha,B\beta} = \sum_{a,b} D^{l_\alpha}_{\alpha,a}\,(R^{AB}) H_{A\alpha,B\beta} D^{l_\beta}_{b,\beta}\big((R^{AB})^{-1}\big)$$

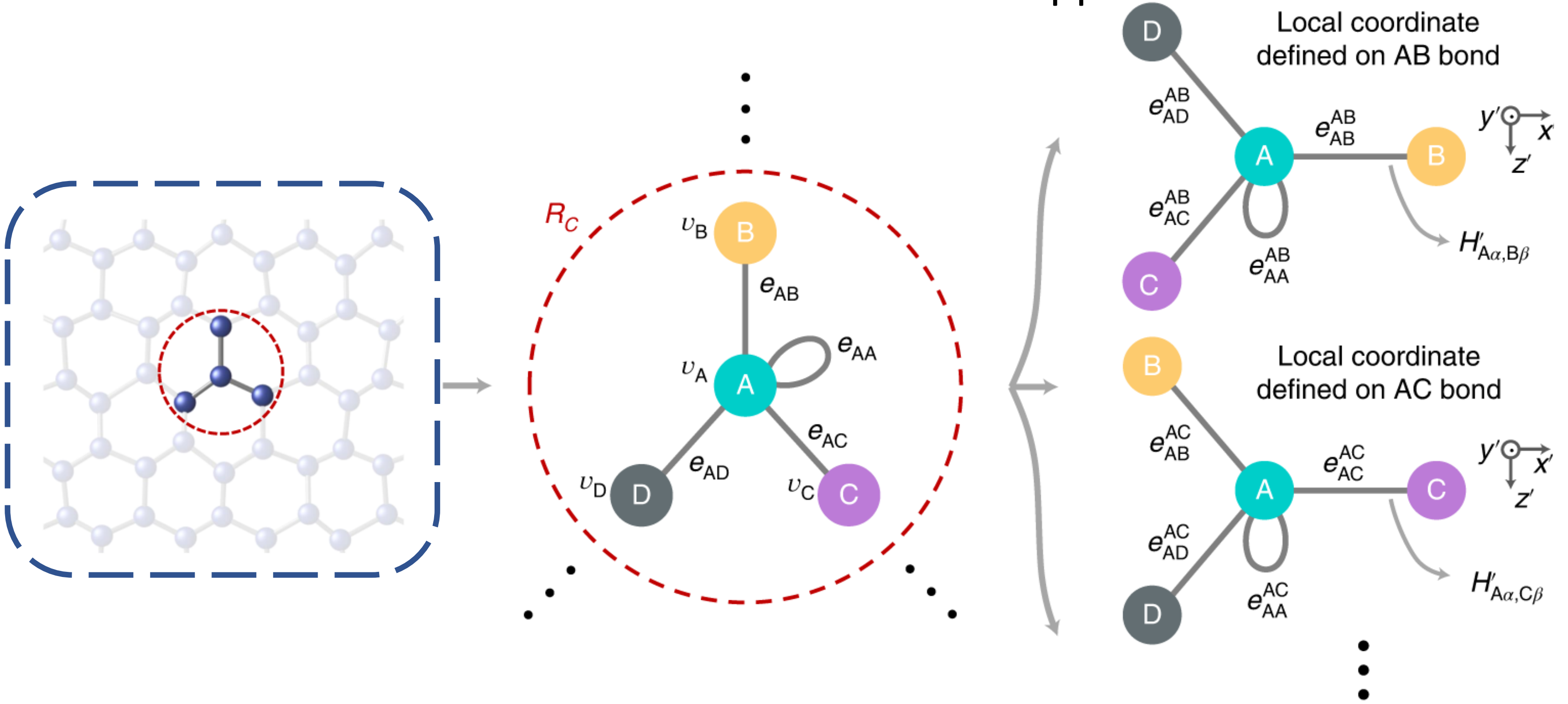
- where $D^{(l)}$ is the Wigner matrix, and l_α is angular momentum quantum number of the orbital α
- The predicted $\widehat{H}'_{\mathrm{DFT}}$ is transformed back to **global coordinates** to preserve **equivariance**:

$$H_{A\alpha,B\beta} = \sum_{a,b} D^{l_\alpha}_{\alpha,a}\,\big((R^{AB})^{-1}\big) H'_{A\alpha,B\beta} D^{l_\beta}_{b,\beta}(R^{AB})$$

Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

Crystal graph: consists of vertices v_i and edges e_{ij} within cutoff radius R_C
An edge is added between two atoms if the orbitals are overlapped



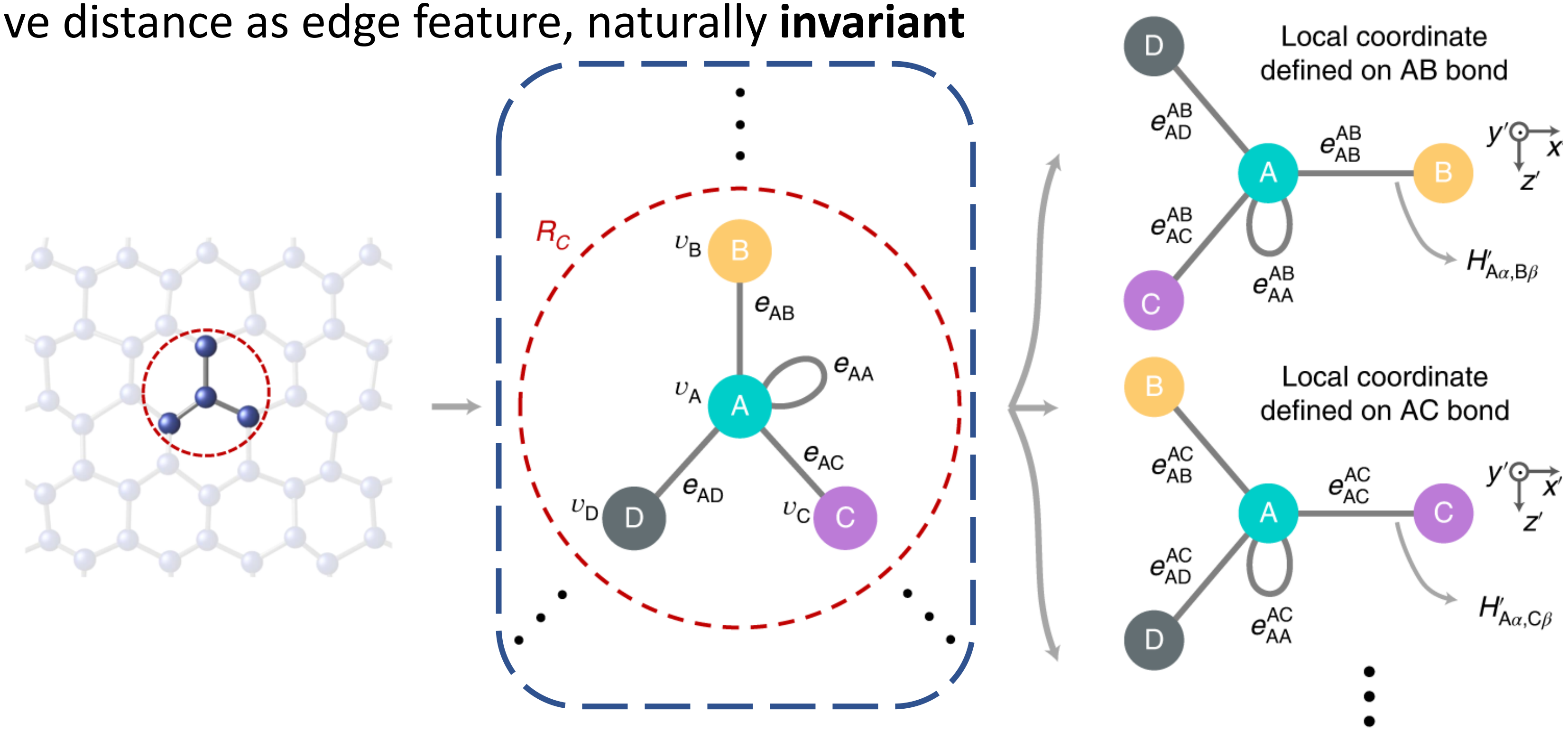
Message Passing
(MP)

Local Coordinates MP
(LCMP)

Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

MP Layer: node and edge representations are updated according to the local topology
Takes relative distance as edge feature, naturally **invariant**



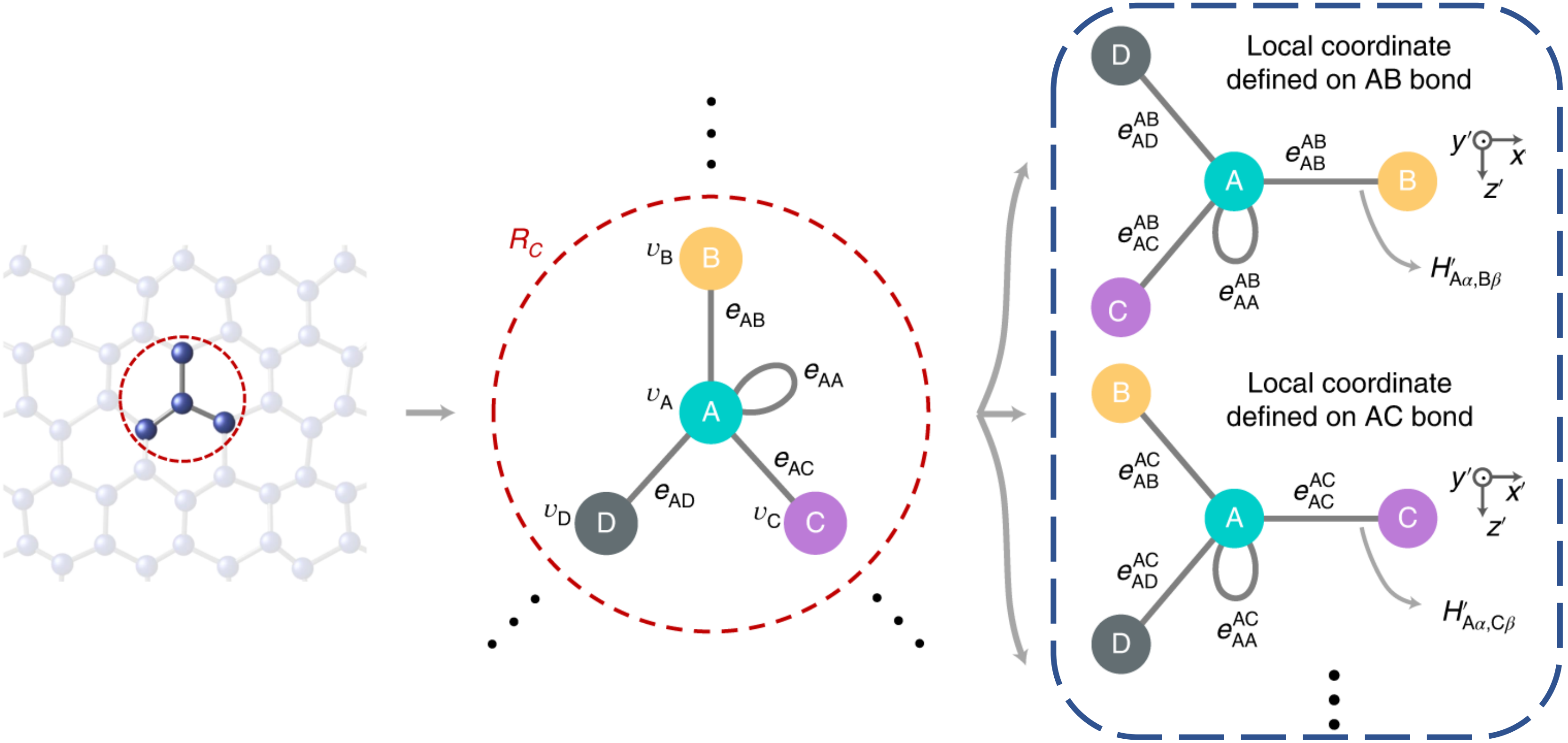
Message Passing (MP)

Local Coordinates MP (LCMP)

Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

LCMP Layer: relative orientation information $\hat{\mathbf{r}}_{ik}^{pq}$ of bond ik under local coordinate defined for edge pq is added into the initial edge features, and predict Hamiltonian element $\widehat{H}_{i\alpha,j\beta}$



$$\hat{\mathbf{r}}_{ik}^{pq} = R^{pq} \cdot \boldsymbol{r}_{ik}$$

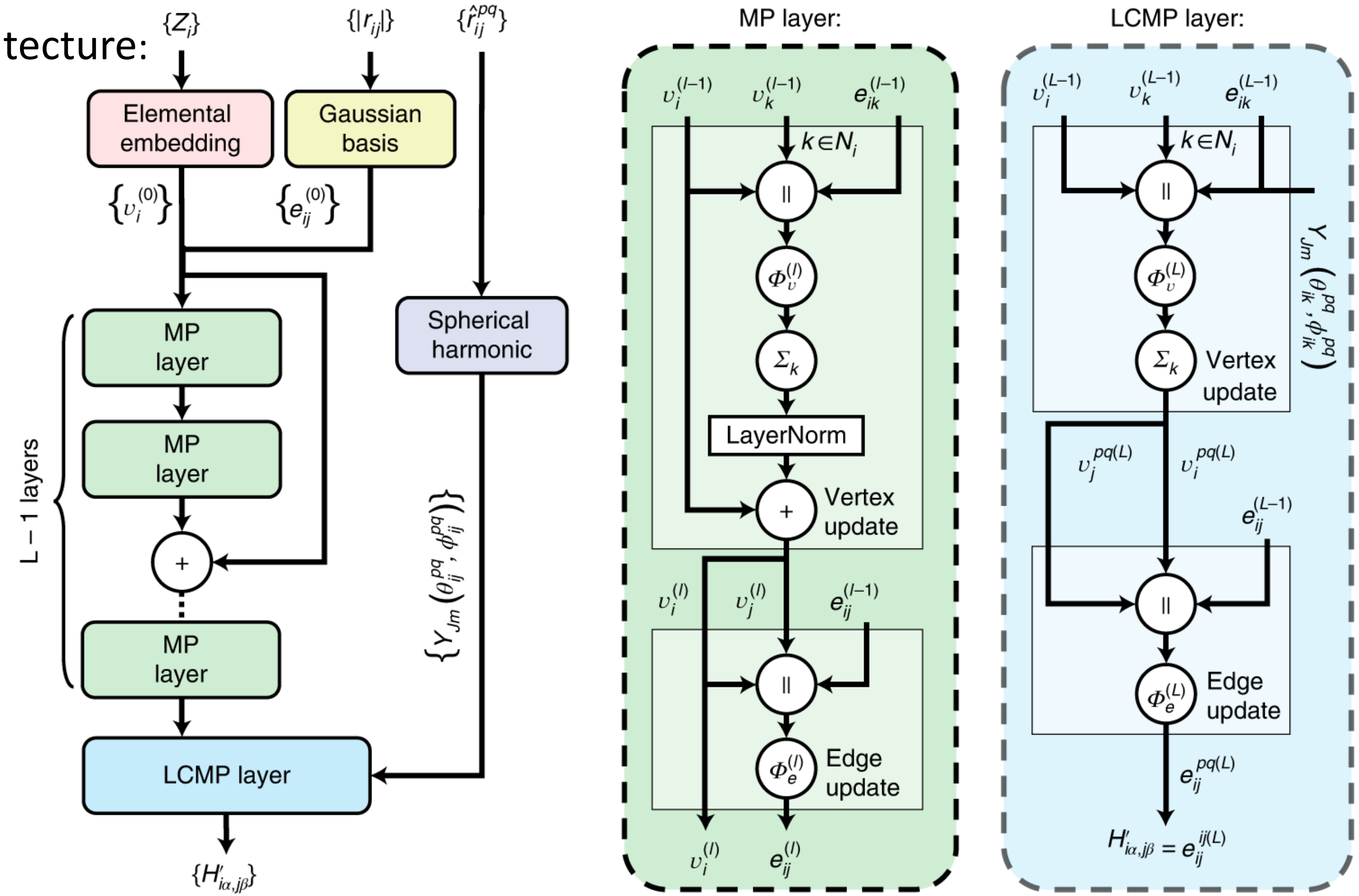
Message Passing (MP)

Local Coordinates MP (LCMP)

Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

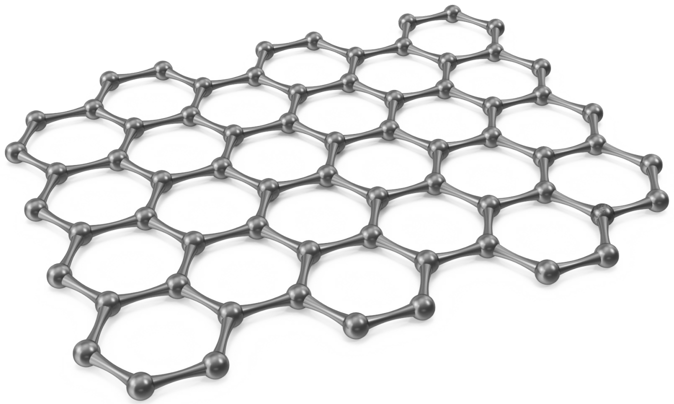
Network architecture:

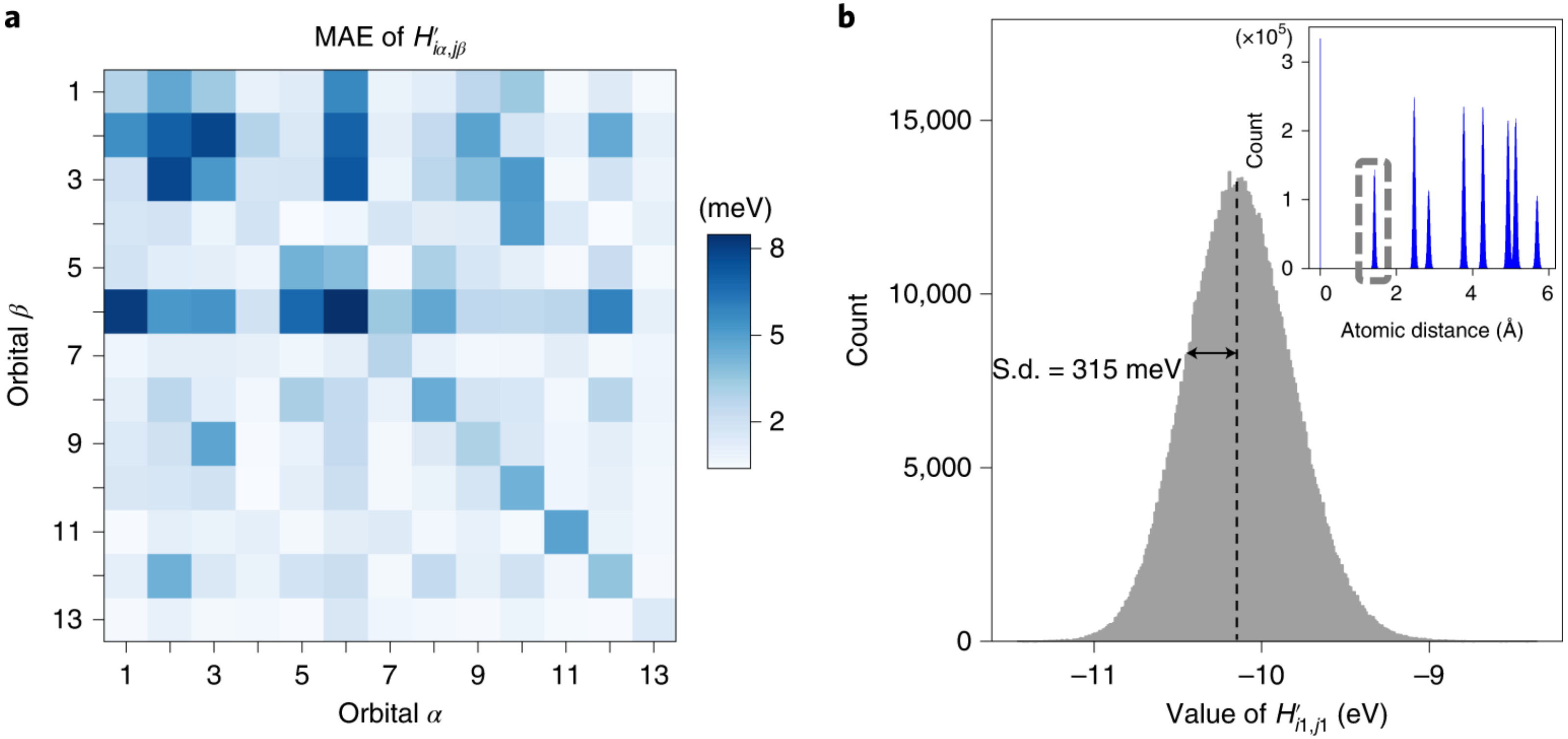


Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

Performance: trained and tested on graphene 6x6 supercell MD data (containing a variety of configurations)





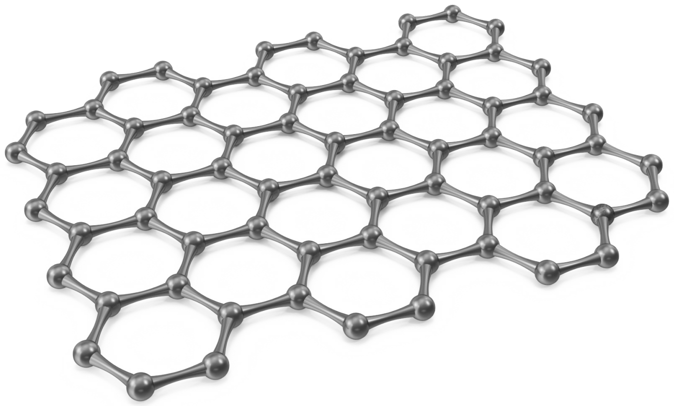
meV level error of $\widehat{H}_{i\alpha,j\beta}$

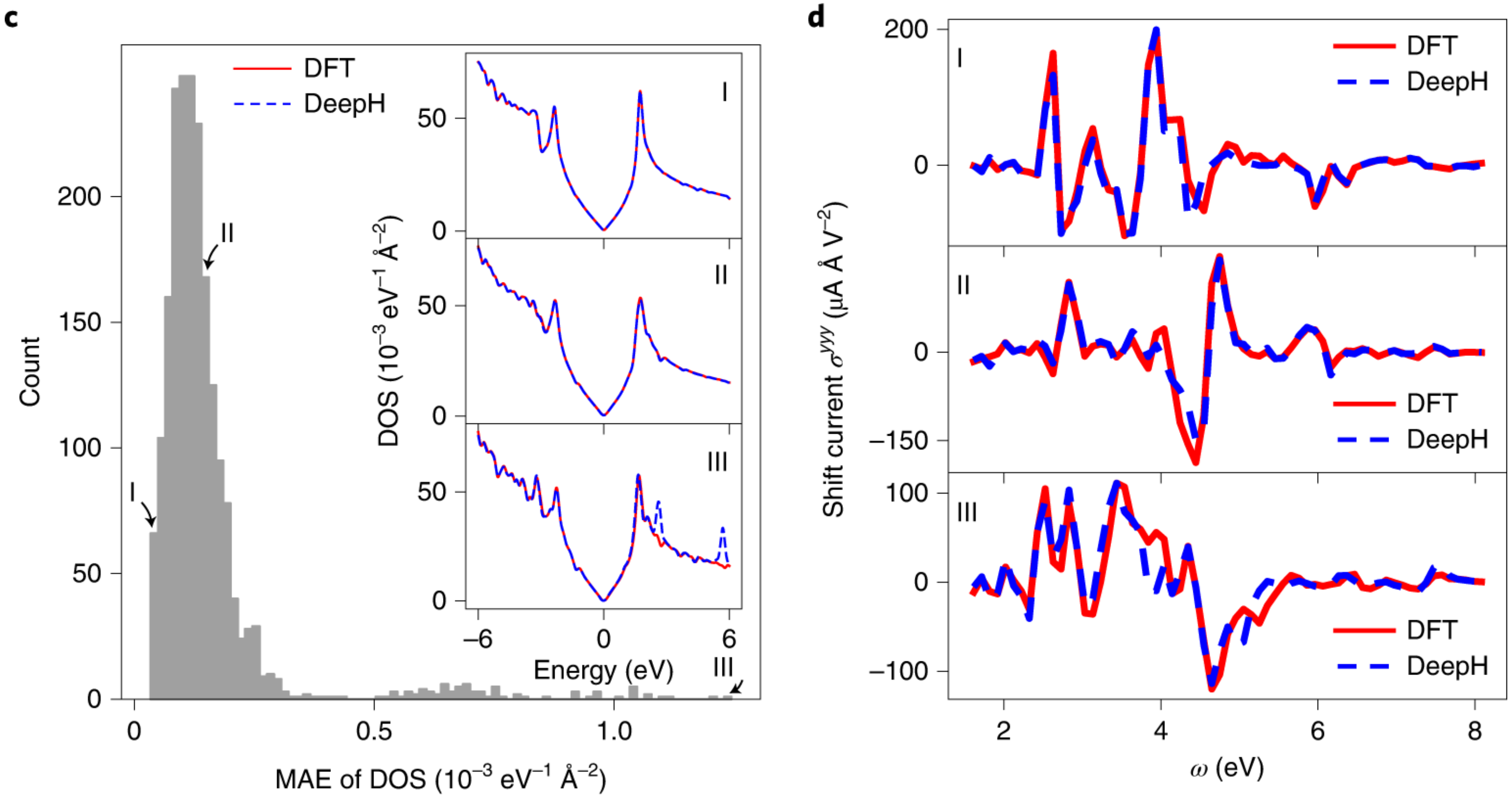
Narrow distribution of $\widehat{H}_{i1,j1}$ between identical orbital pairs

Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

Performance: generalization for 2,000 **unseen** graphene configurations



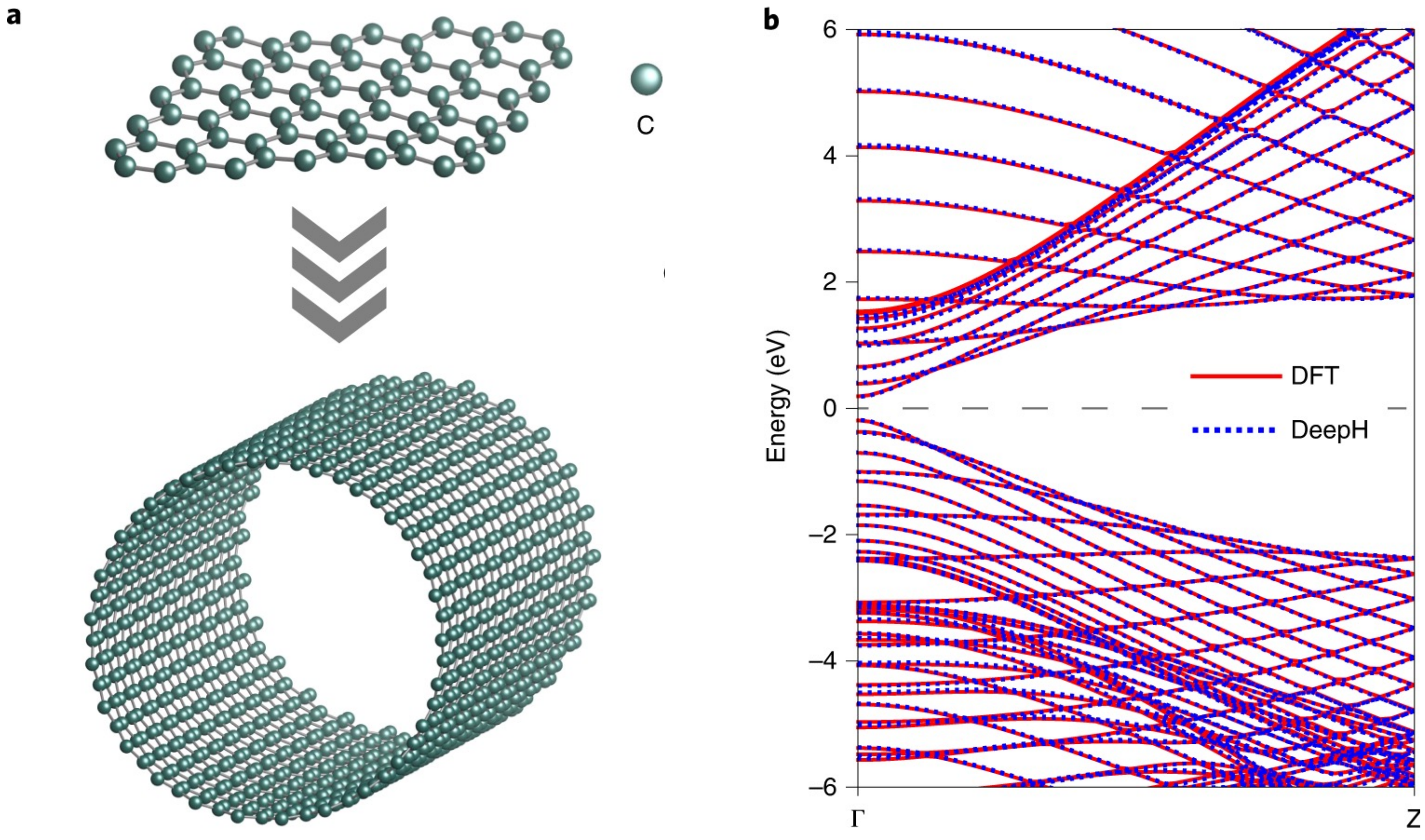


Density of State (DOS): distribution of electrons of different energy states
Shift current conductivity σ^{yyy}: conductivity of electrons excited by photons of different energy ω

Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

Generalization Performance: trained on **flat sheet** graphene, tested on **curved** carbon nanotubes

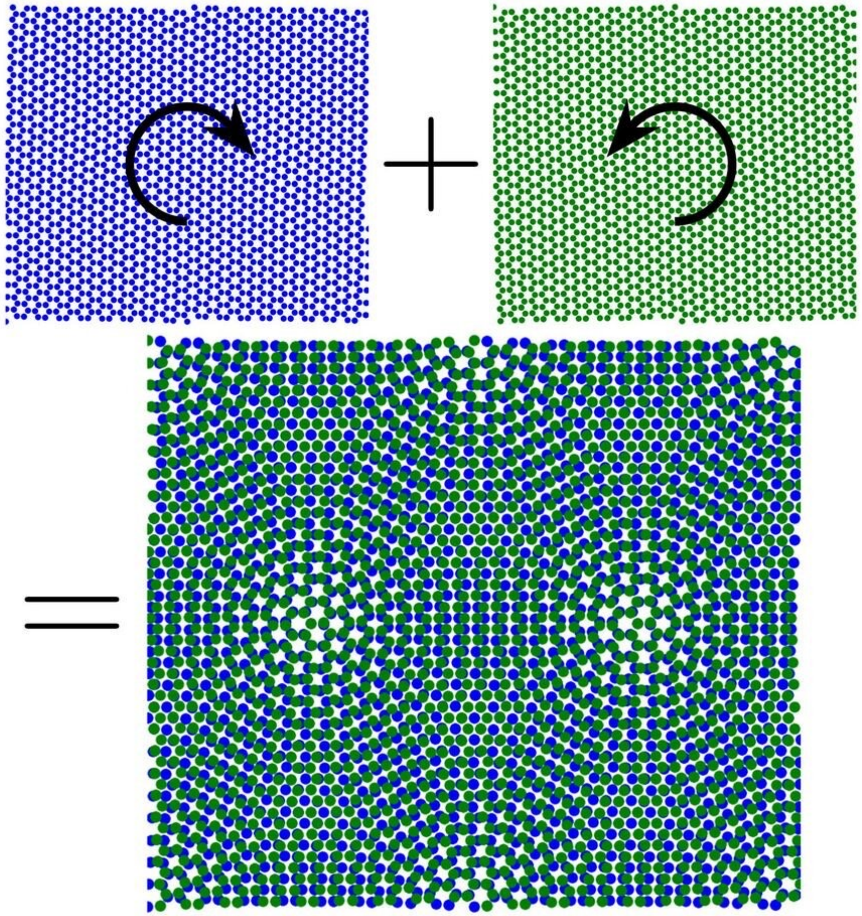


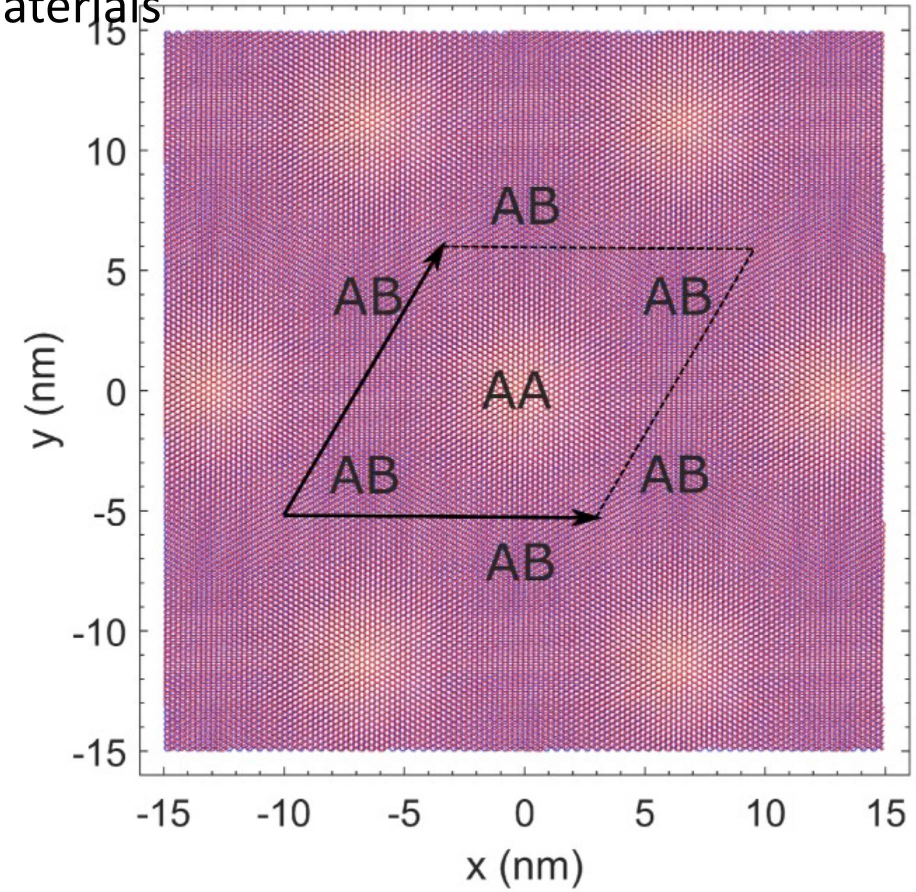
Band structures by DFT and DeepH

Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

Generalization Performance: moiré-twisted bilayer materials



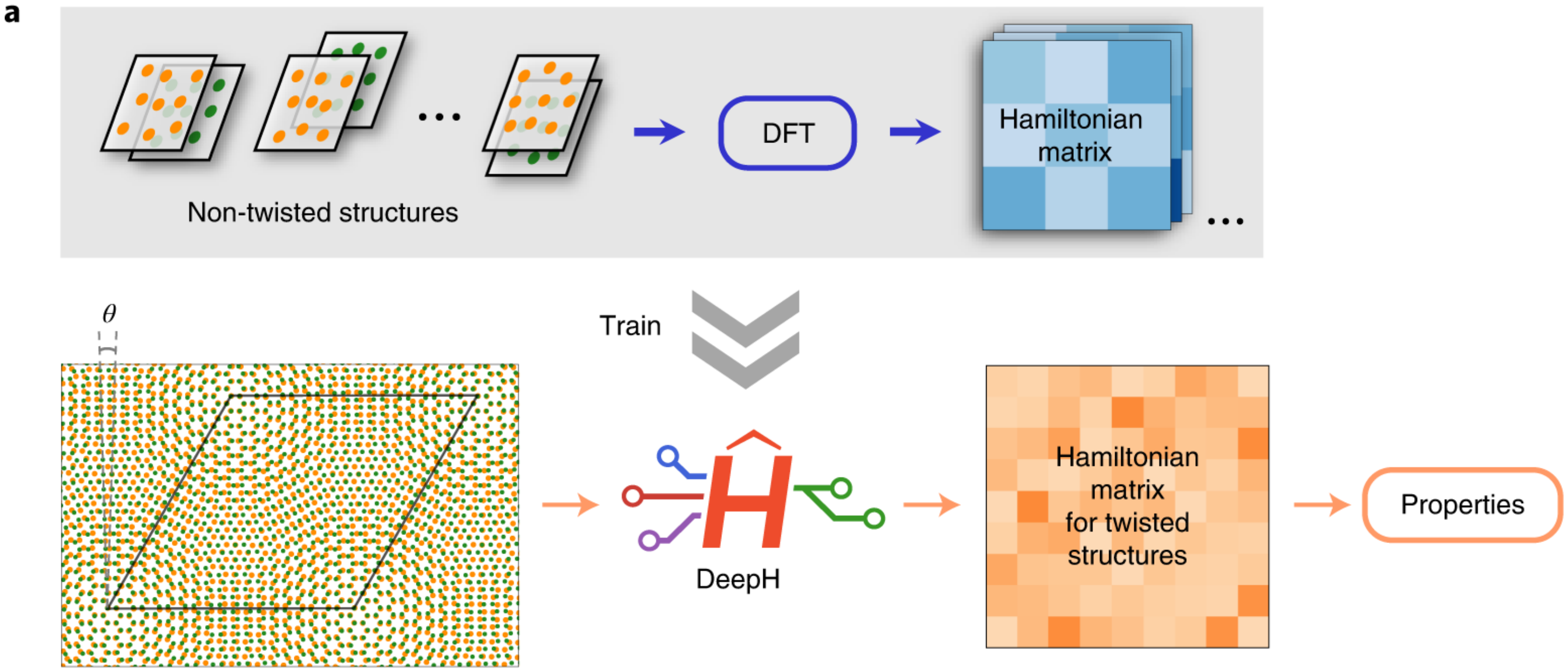


1.08 ° magic angle graphene: **11,164** carbon atoms per unit cell

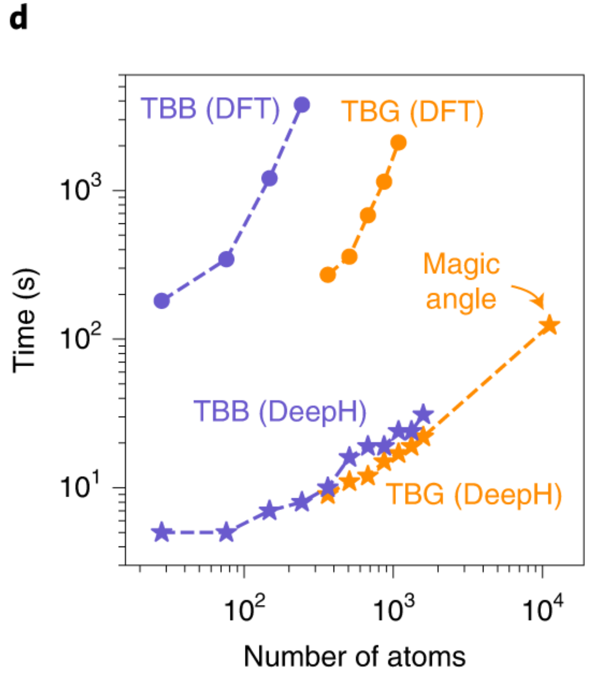
Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

Generalization Performance: moiré-twisted bilayer materials



trained on **non-twisted small** bilayer structures
tested on **twisted large** structures

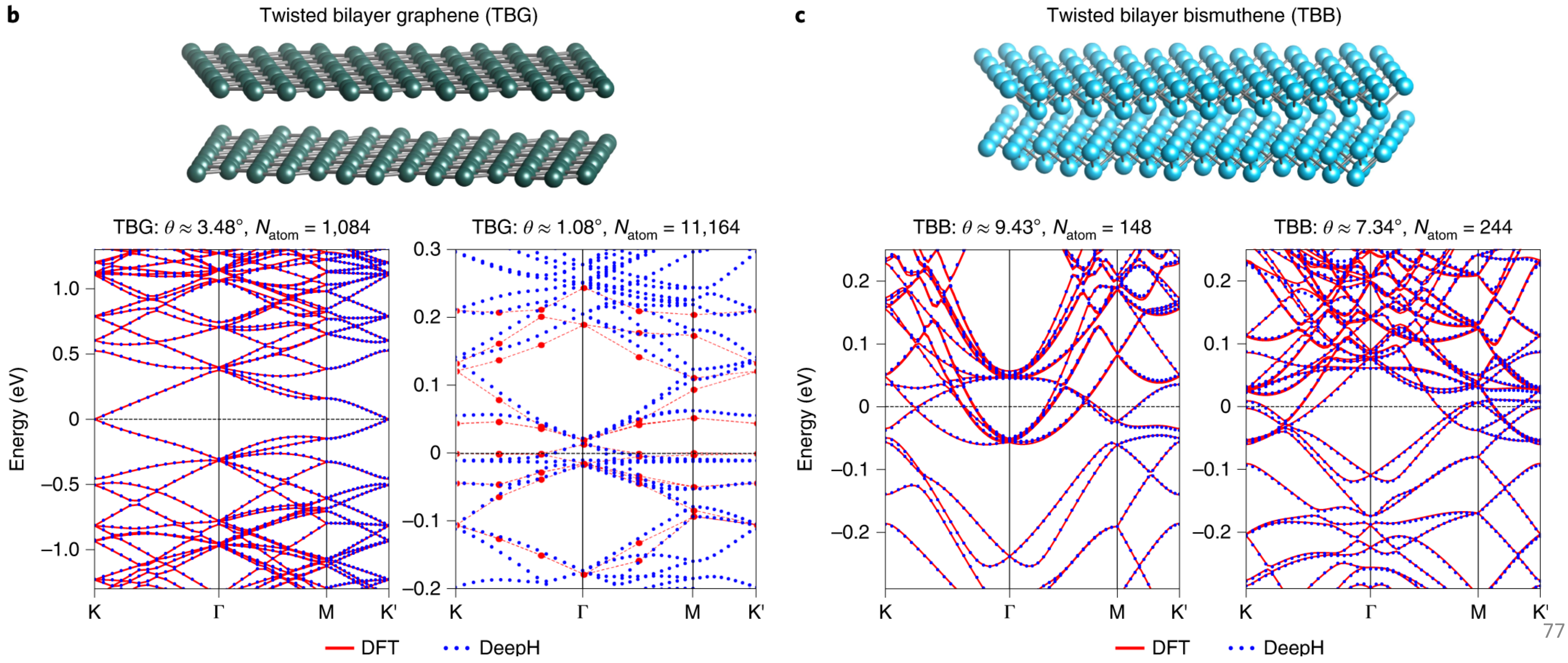


Linear complexity for large systems

Deep Learning Approaches: DeepH

Deep Learning DFT Hamiltonian (DeepH):

Generalization Performance: trained on non-twisted small bilayer structures, tested on twisted large



AI in Materials Science

Directly Bridging

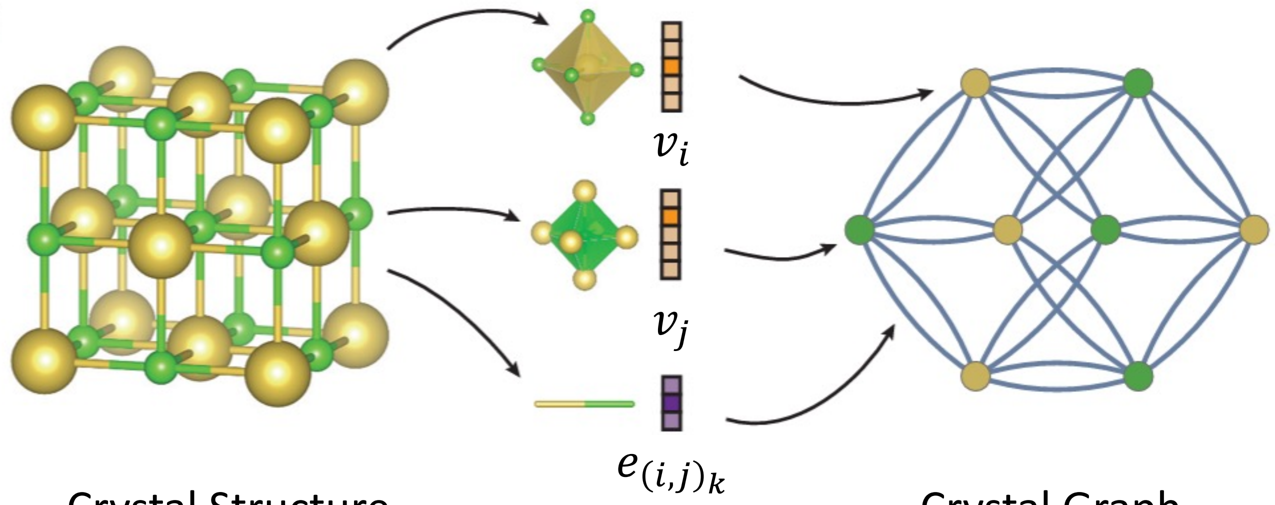
Deep Learning Approaches: CGCNN

Crystal Graph Convolutional Neural Networks (CGCNN) [1]:

Directly bridging the gap between crystal structures and material properties via GNN

Construction of crystal graph:

- Node v_i representing physical properties of atom i
- Edge $e_{(i,j)_k}$ representing the distance between atom i and its k-th nearest neighbor j



Crystal Structure

Crystal Graph

Preserving **invariant** chemical environment of atoms

[1] Xie, Tian, and Jeffrey C. Grossman. "Crystal graph convolutional neural networks for an accurate and interpretable prediction of material properties." *Physical review letters* 120.14 (2018): 145301.

Deep Learning Approaches: CGCNN

Crystal Graph Convolutional Neural Networks (CGCNN):

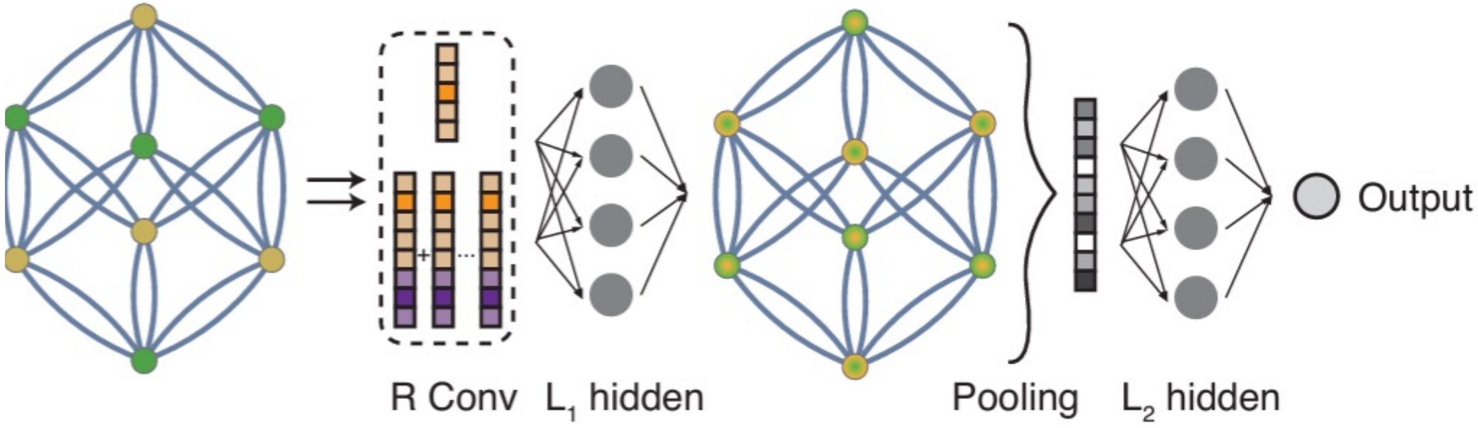
The node feature is updated following:

$$v_i^{(t+1)} = v_i^{(t)} + \sum_{j,k} \sigma\left(z_{(i,j)_k}^{(t)} W_f^{(t)} + b_f^{(t)}\right) \odot g\left(z_{(i,j)_k}^{(t)} W_s^{(t)} + b_s^{(t)}\right)$$

Where

$$z_{(i,j)_k}^{(t)} = v_i^{(t)} \oplus v_j^{(t)} \oplus e_{(i,j)_k}$$

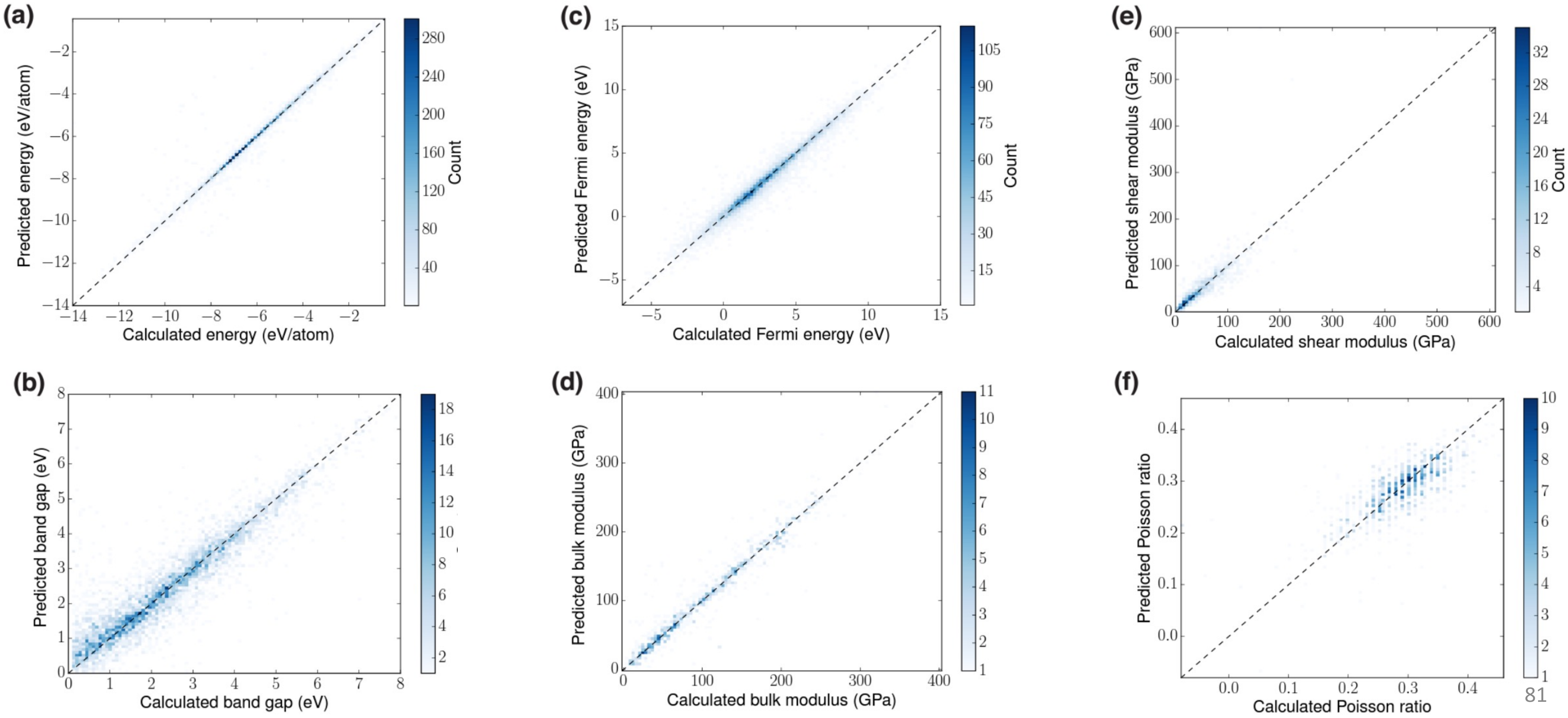
And node features are pooled for global properties prediction



$\odot$: element-wise production, $\oplus$: concatenation

Deep Learning Approaches: CGCNN

Crystal Graph Convolutional Neural Networks (CGCNN):

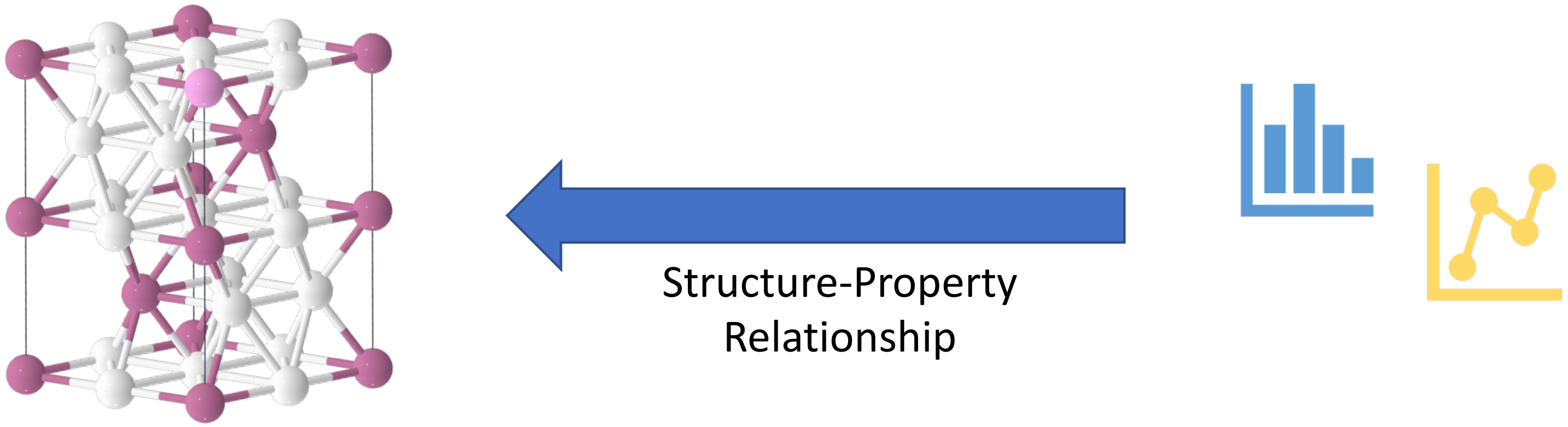


AI in Materials Science

Inverse Problem

Next Level: Inverse Problem

Can we start with the desired properties?



Structure-Property Relationship

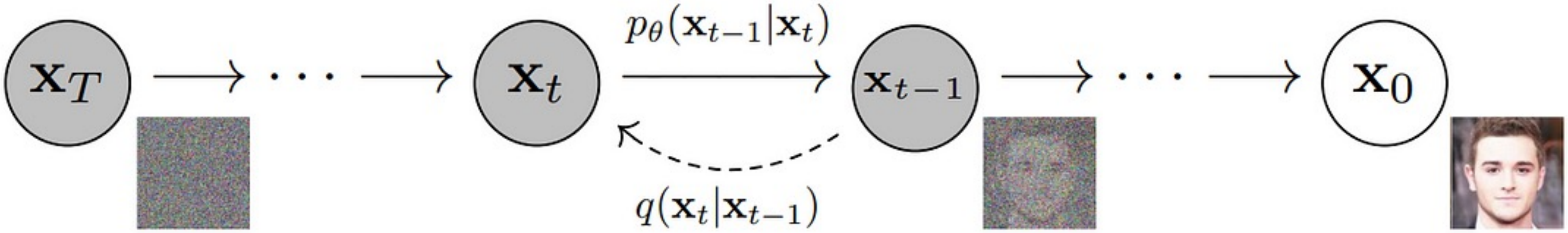
Structure

Properties

Next Level: Inverse Problem

Denoising Diffusion Probabilistic Models (DDPM) (Lecture 3):

- Mapping unknown data distribution to a known prior distribution (e.g. standard Gaussian)
- Making **effective sampling** from the original data distribution feasible

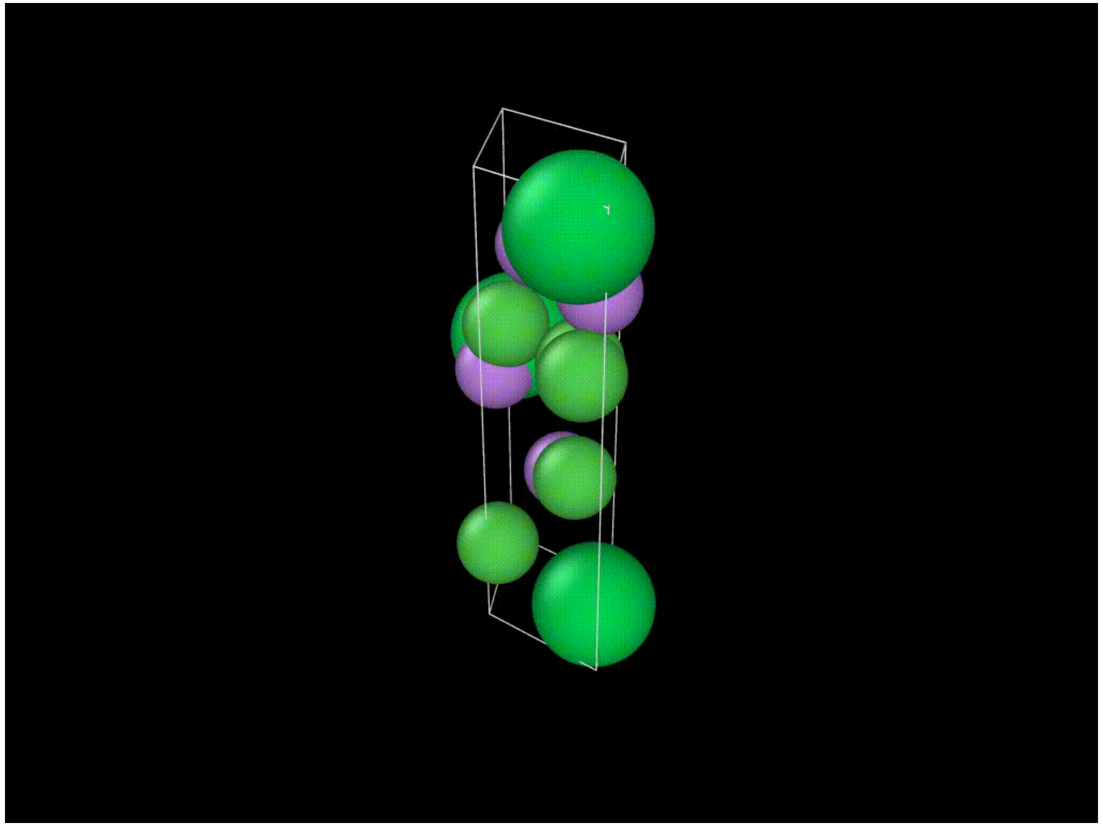


Gaussian distribution

data distribution

Inverse Problem: CDVAE

Crystal Diffusion Variational AutoEncoder (CDVAE) [1]:



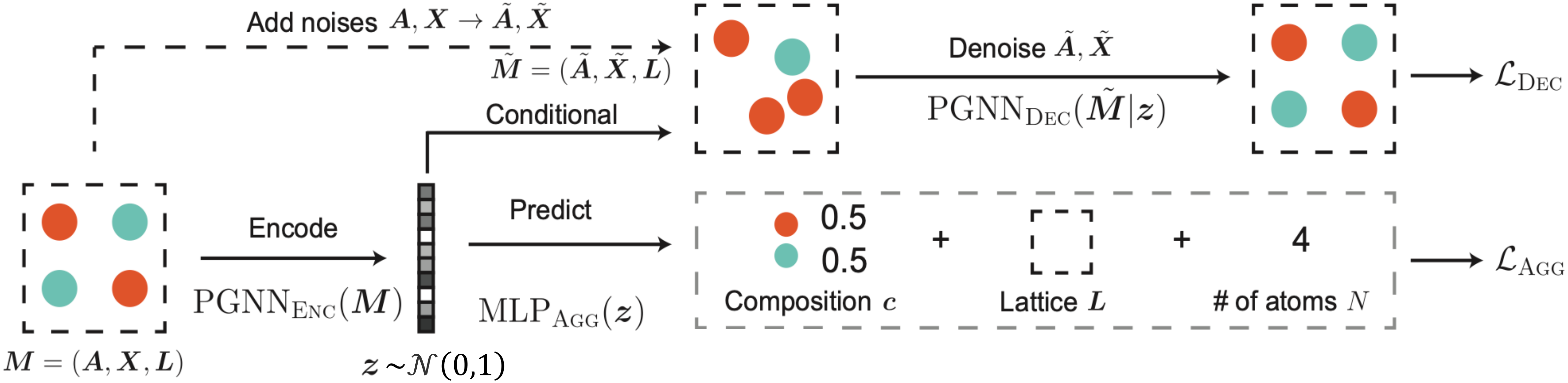
[1] Xie, Tian, et al. "Crystal diffusion variational autoencoder for periodic material generation." *arXiv preprint arXiv:2110.06197* (2021).

Inverse Problem: CDVAE

Crystal Diffusion Variational AutoEncoder (CDVAE):

Training:

- Encode crystal structure M into latent representation $\boldsymbol{z}$ with a periodic graph neural network $\mathrm{PGNN_{ENC}}$
- Decode crystal aggregation properties (c, L, N) through $\mathrm{MLP_{AGG}}$
- Denoise corrupted structure $\widetilde{M} = \left(\tilde{A}, \tilde{X}, L\right)$ conditioned on $\boldsymbol{z}$ through $\mathrm{PGNN_{DEC}}$

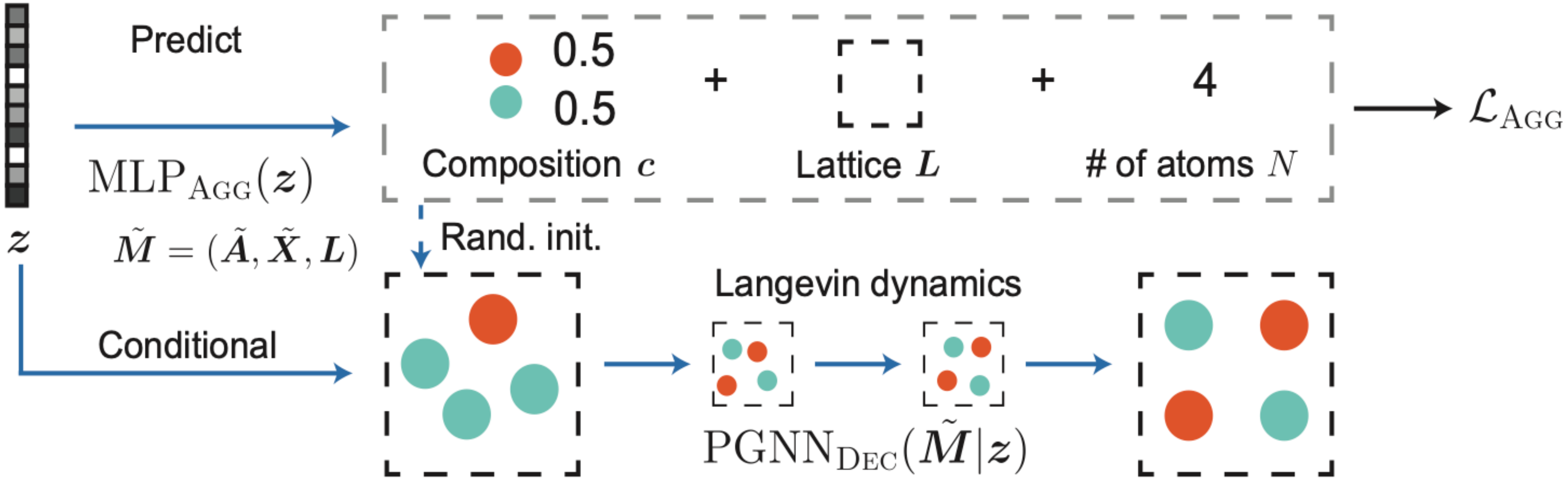


Inverse Problem: CDVAE

Crystal Diffusion Variational AutoEncoder (CDVAE):

Generation:

- Sample a latent representation $\boldsymbol{z} \sim \mathcal{N}(0,1)$
- Decode crystal aggregation properties (c, L, N) through $\mathrm{MLP}_{\mathrm{AGG}}$
- Randomly initialize a disordered crystal structure $\widetilde{M}$ according to (c, L, N)
- Denoise corrupted structure $\widetilde{M} = (\tilde{A}, \tilde{X}, L)$ conditioned on $\boldsymbol{z}$ through $\mathrm{PGNN}_{\mathrm{DEC}}$

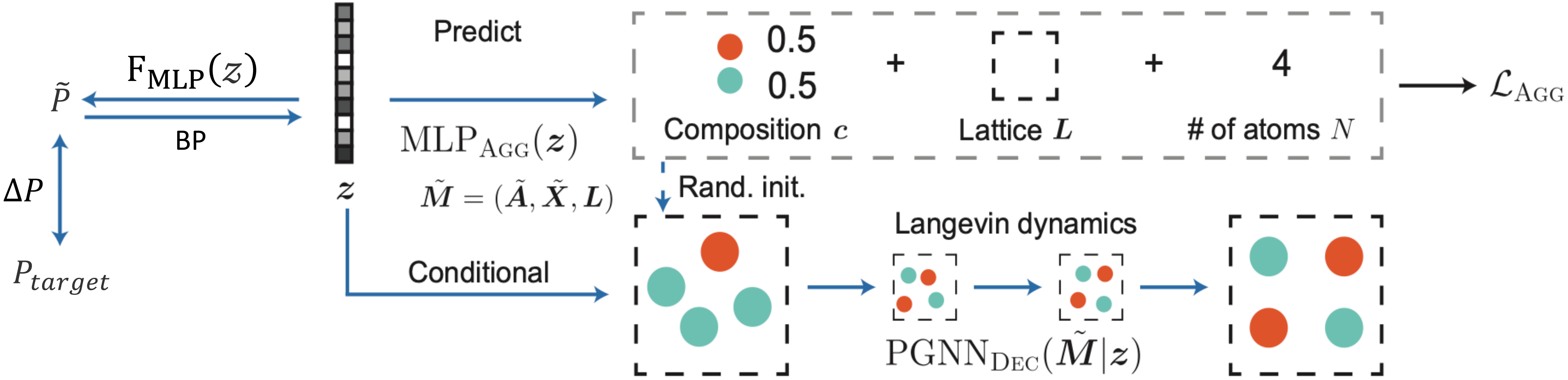


Inverse Problem: CDVAE

Crystal Diffusion Variational AutoEncoder (CDVAE):

Property optimization:

- Jointly trained property predictor: $\tilde{P} = \mathrm{F}_{\mathrm{MLP}}(z)$
- Optimize latent using back propagation (BP) for 5,000 steps
- Decode 10 crystal structures every 500 steps from the latent trajectory
- Select one best structure with closest $\tilde{P}$ predicted by an individual predictor



Inverse Problem: MatterGen

MatterGen [1]:
Tailored diffusion process for crystalline materials:

$$q(\boldsymbol{A}_{t+1}, \boldsymbol{X}_{t+1}, \boldsymbol{L}_{t+1} | \boldsymbol{A}_t, \boldsymbol{X}_t, \boldsymbol{L}_t) = q(\boldsymbol{A}_{t+1} | \boldsymbol{A}_t)\, q(\boldsymbol{X}_{t+1} | \boldsymbol{X}_t)\, q(\boldsymbol{L}_{t+1} | \boldsymbol{L}_t)$$

- Atom types: $\boldsymbol{A} = (\boldsymbol{a}_1, \boldsymbol{a}_2, \dots, \boldsymbol{a}_N) \in \mathbb{A}^N$
- Atom fractional coordinates: $\boldsymbol{X} = (\boldsymbol{x}_1, \boldsymbol{x}_2, \dots, \boldsymbol{x}_N) \in [0,1)^{N \times 3}$
- Lattice: $\boldsymbol{L} = (\boldsymbol{l}_1, \boldsymbol{l}_2, \boldsymbol{l}_3) \in \mathbb{R}^{3 \times 3}$

[1] Zeni, Claudio, et al. "Mattergen: a generative model for inorganic materials design." *arXiv preprint arXiv:2312.03687* (2023).

Inverse Problem: MatterGen

MatterGen:

Tailored diffusion process for crystalline materials:

- Atom type $\boldsymbol{A}$:

$$q(\boldsymbol{a}_t|\boldsymbol{a}_{t-1}) = \mathrm{Cat}(\boldsymbol{a}_t; \boldsymbol{p} = \boldsymbol{a}_{t-1}\boldsymbol{Q}_t)$$

- $\mathrm{Cat}(\boldsymbol{a}; \boldsymbol{p})$: categorical distribution over 1-hot vectors whose probabilities are given by the row vector $\boldsymbol{p}$
- $[\boldsymbol{Q}_t]_{ij} = q(a_t = j|a_{t-1} = i)$: Markov transition matrix at time step t

$$[\boldsymbol{Q}_t]_{ij} = \begin{cases} 1 & i = j = m \\ 1 - \beta_t & i = j \neq m \\ \beta_t & j = m \neq i \\ 0 & m \neq i \neq j \neq m \end{cases}$$

- β_t: probability of transiting to a **MASK** state
- $1 - \beta_t$: probability of staying unchanged

Inverse Problem: MatterGen

MatterGen:
Tailored diffusion process for crystalline materials:

- Fractional coordinates $\boldsymbol{X}$:

$$q(\boldsymbol{x}_t|\boldsymbol{x}_0) = \mathcal{N}_W(\boldsymbol{x}_t; \boldsymbol{x}_0, \sigma_t^2 \boldsymbol{I})$$

- $\mathcal{N}_W$: wrapped normal distribution preserving periodic boundary condition (PBC)

$$\mathcal{N}_W(\boldsymbol{x}_t; \boldsymbol{x}_0, \sigma_t^2 \boldsymbol{I}) = \sum_{k \in \mathbb{Z}^3} \mathcal{N}(\boldsymbol{x}_t; \boldsymbol{x}_0 - \boldsymbol{k}, \sigma_t^2 \boldsymbol{I})$$

Inverse Problem: MatterGen

MatterGen:

Tailored diffusion process for crystalline materials:

- Lattice $\boldsymbol{L}$:
- Naïve diffusion process:

$$q(\boldsymbol{L}_t|\boldsymbol{L}_0) = \mathcal{N}(\sqrt{\bar{\alpha}_t}\boldsymbol{L}_0, (1-\bar{\alpha}_t)\boldsymbol{I})$$

- Leads to extremely narrow and small lattices
- Custom limiting mean & variance:

$$q(\boldsymbol{L}_t|\boldsymbol{L}_0) = \mathcal{N}(\sqrt{\bar{\alpha}_t}\boldsymbol{L}_0 + (1-\sqrt{\bar{\alpha}_t})\mu(n)\boldsymbol{I}, (1-\bar{\alpha}_t)\sigma_t^2(n)\boldsymbol{I})$$

- which yields the limit distribution for $T \to \infty$:

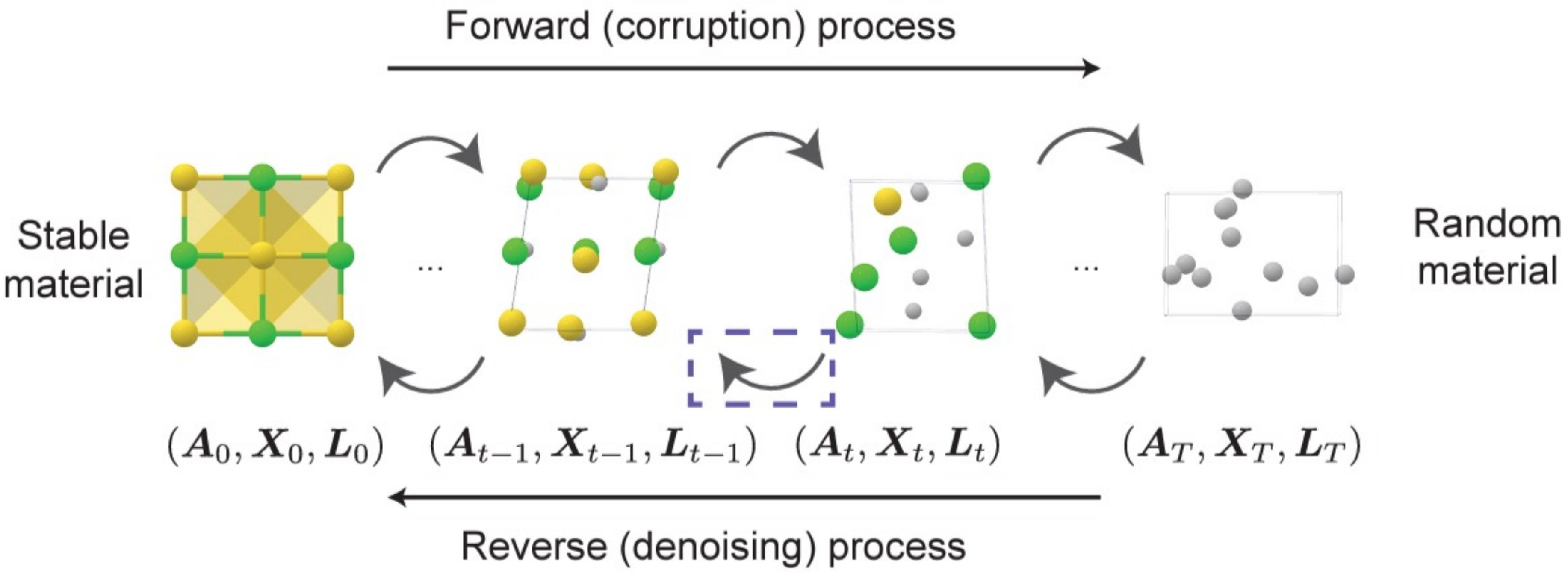
$$q(\boldsymbol{L}_T) = \mathcal{N}(\mu(n)\boldsymbol{I}, \sigma_T^2(n)\boldsymbol{I})$$

- where $\mu(n) = \sqrt[3]{nc}, \sigma_T^2(n) = \sqrt[3]{nv}$
- c: inverse average atomic density of the dataset
- v: average unit cell volume of the dataset
- The signal-to-noise-ratio at T is therefore independent to the number of atoms n:

$$SNR = \lim_{T\to\infty}\frac{|\mu(n)|}{\sigma(n)} = \sqrt[3]{\frac{c}{v}}$$

Inverse Problem: MatterGen

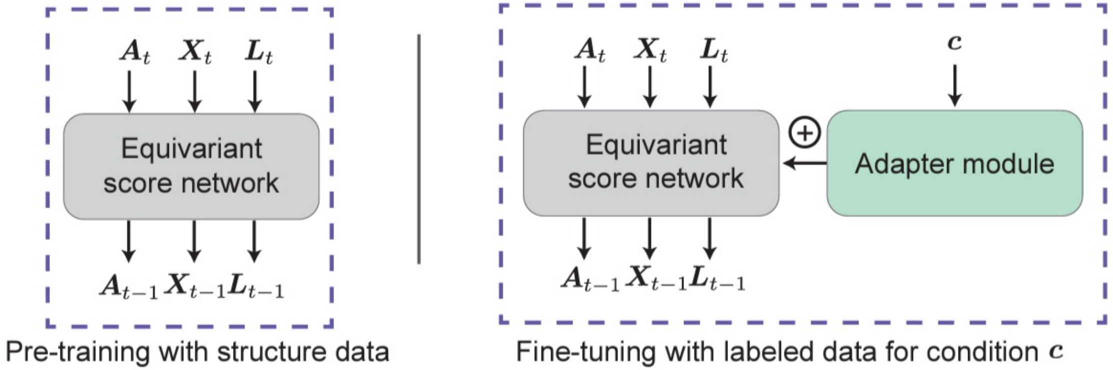
MatterGen:
Tailored diffusion process for crystalline materials:



Inverse Problem: MatterGen

MatterGen:
Conditional generation of materials:

"ControlNet, Zhang, et al. 2023"

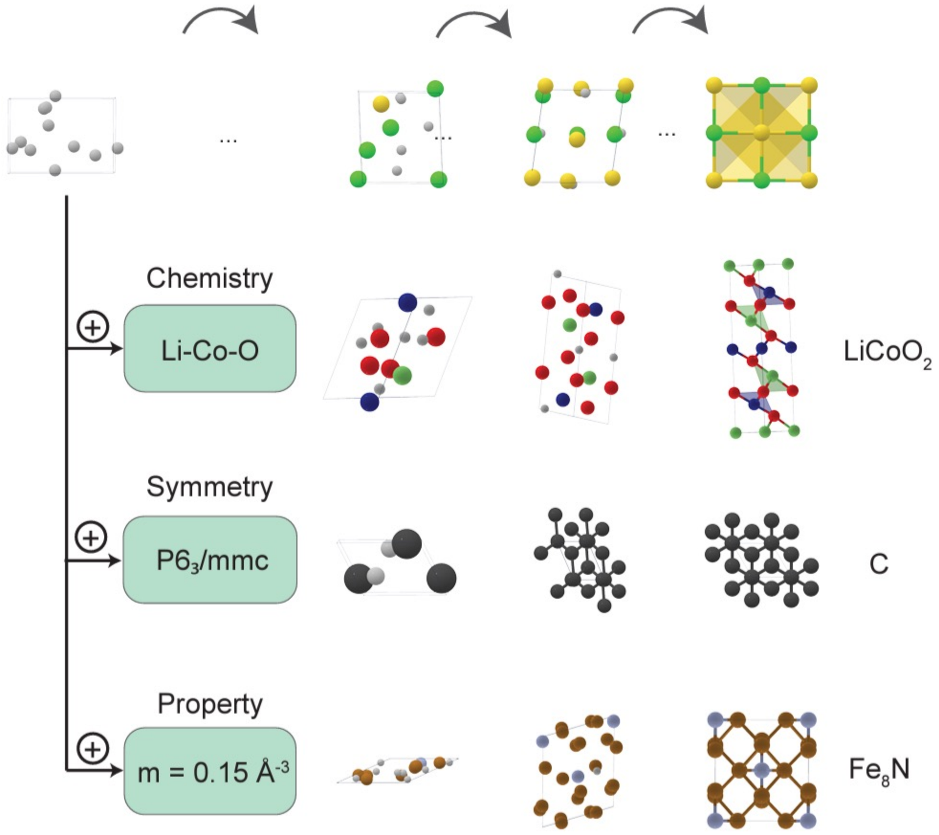


Pre-training with structure data

Fine-tuning with labeled data for condition c

Unconditional pre-training of the base model

Fine-tuning with labeled data for condition c



Fine-tuned for different tasks with a **frozen base model**

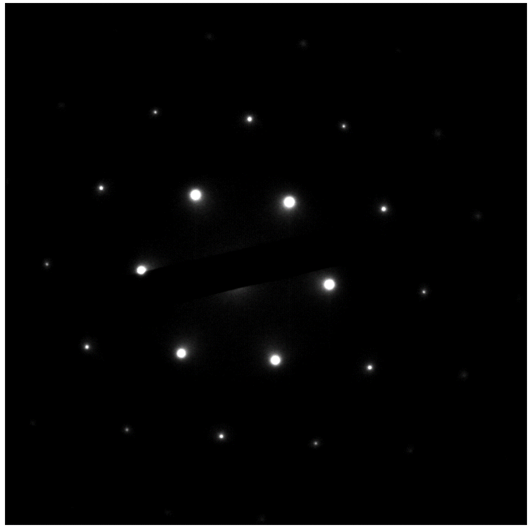
AI in Materials Science

AI as a Powerful Assistance

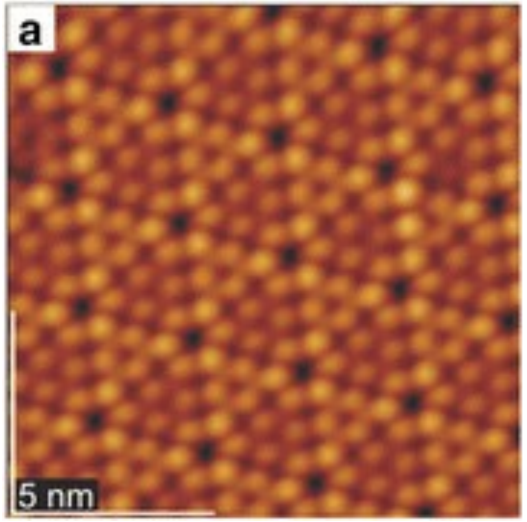
AI as a Powerful Assistance:

Single crystal Si

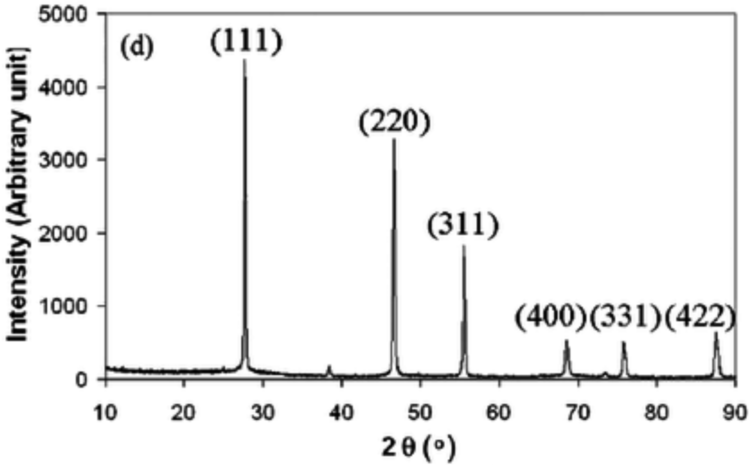
Characterization



Transmission Electron Microscopy (**TEM**)

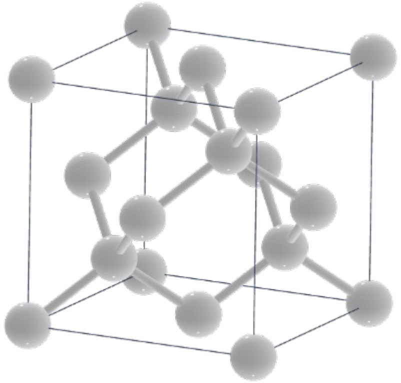


Scanning Tunneling Microscopy (**STM**)



X-Ray Diffraction (**XRD**)

Analysis & Interpretation

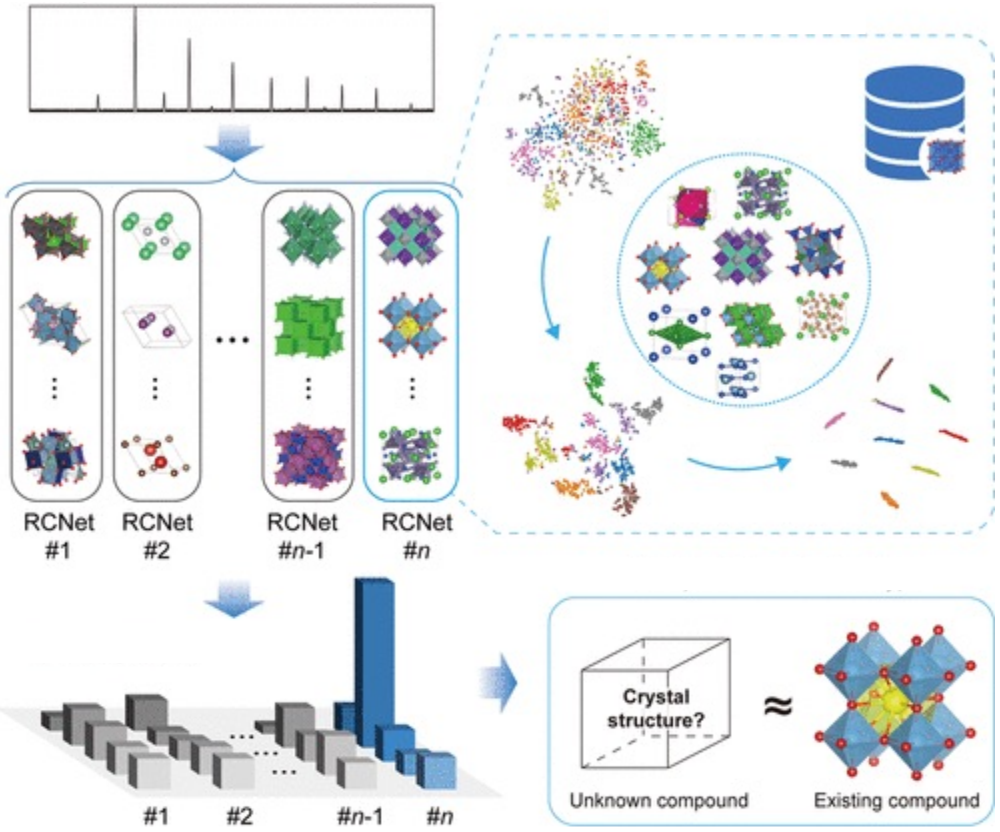


Crystal structure

AI as a Powerful Assistance: CrySTINe

Crystal Structure-Type Identification Network (CrySTINet) [1]:

Input the XRD pattern of an unknown compound



ResNet Confidence Networks (RCNet)

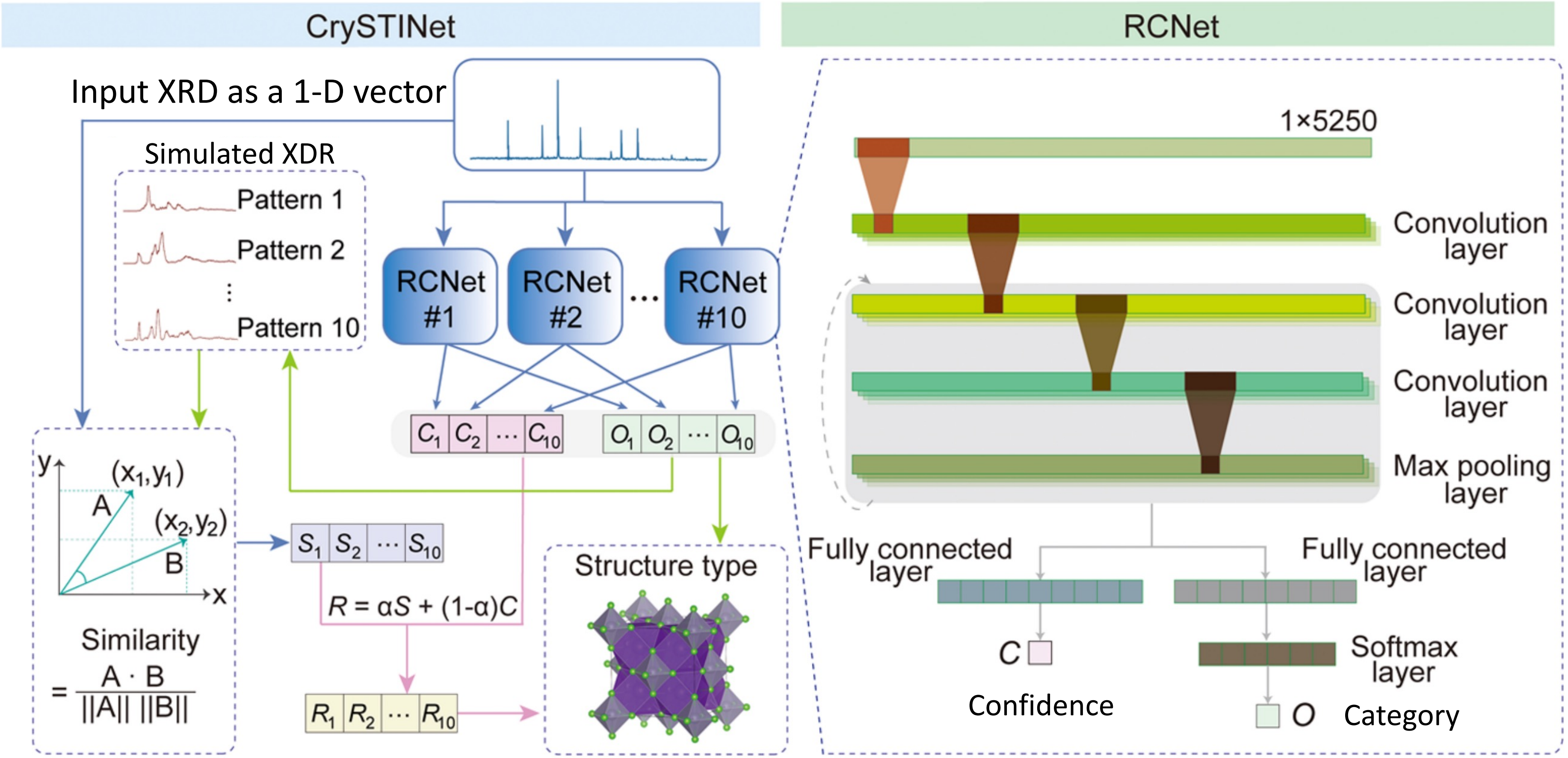
Predict both categories O and reliabilities R

Choose the **most probable structure type** as the final result

[1] Chen, Litao, et al. "Crystal Structure Assignment for Unknown Compounds from X-ray Diffraction Patterns with Deep Learning." *Journal of the American Chemical Society* 146.12 (2024): 8098-8109.

AI as a Powerful Assistance: CrySTINe

Crystal Structure-Type Identification Network (CrySTINet):

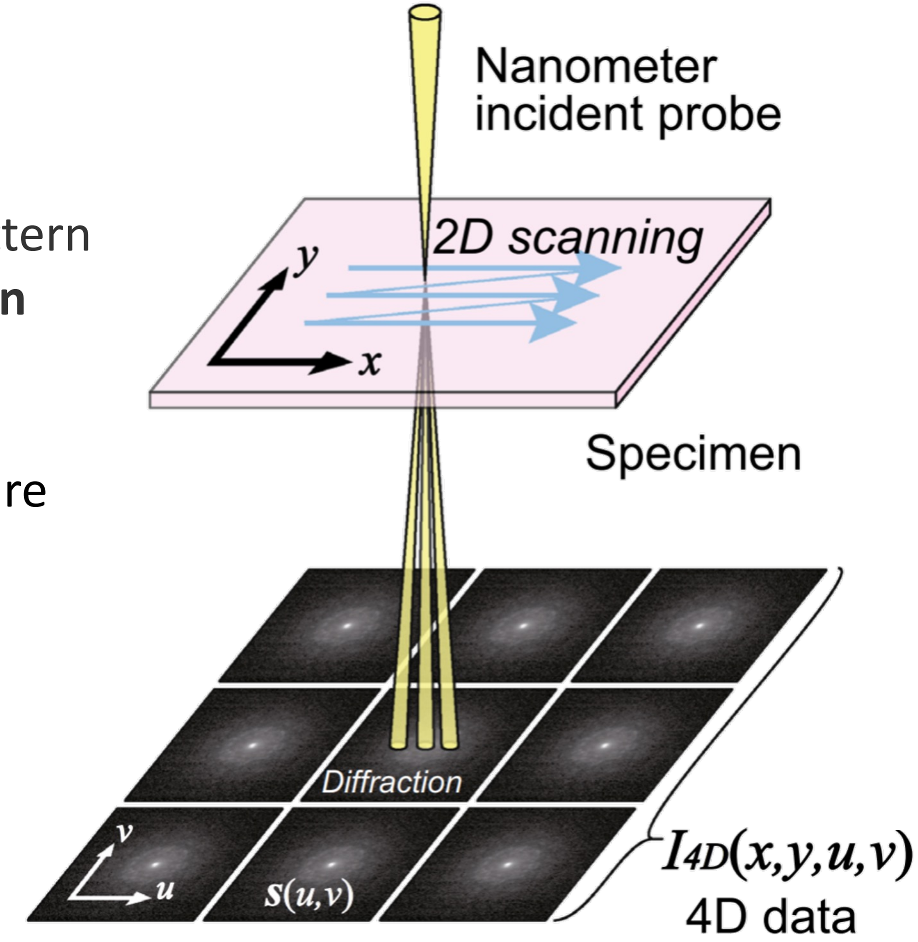


Cosine similarity as an extra criterion

AI as a Powerful Assistance: 4D-STEM

4-D Scanning Transmission Electron Microscopy (4D-STEM) :

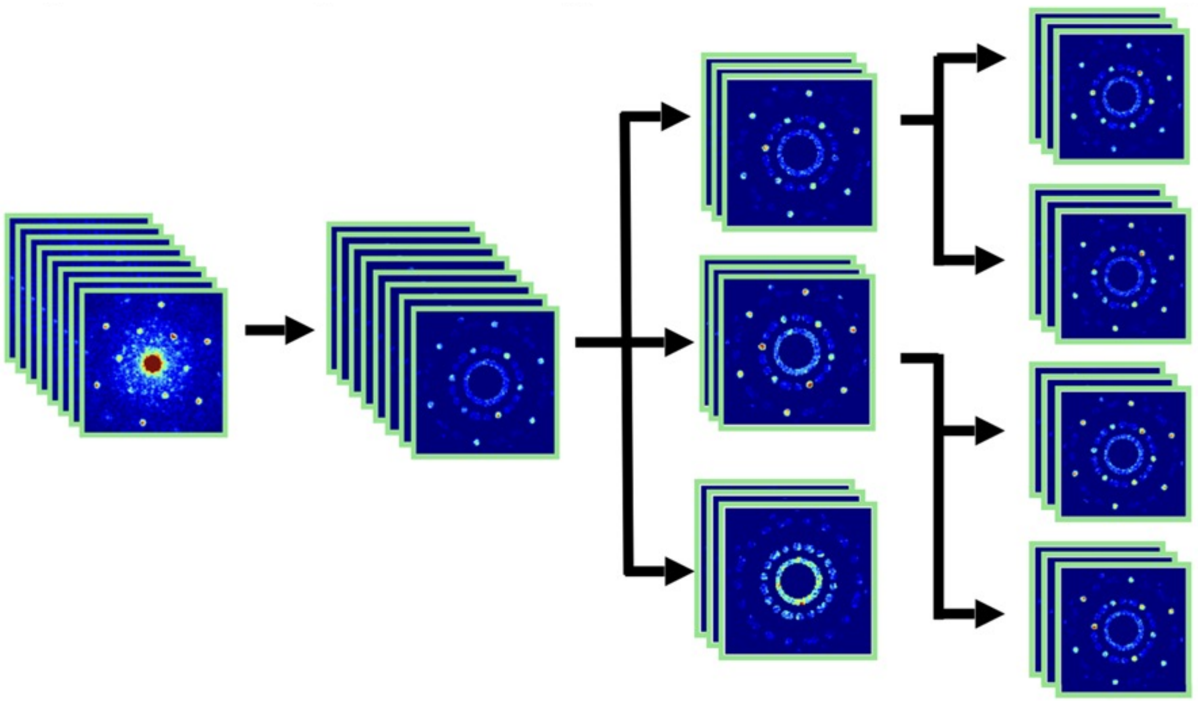
- Each pixel in real-space corresponds to a 2-D diffraction pattern
- Each diffraction pattern contains **local structure information** within the nm range
- 4D-STEM data can easily reach GBs
- Statistical or computational approaches for interpretation are indispensable



>3k diffraction patterns

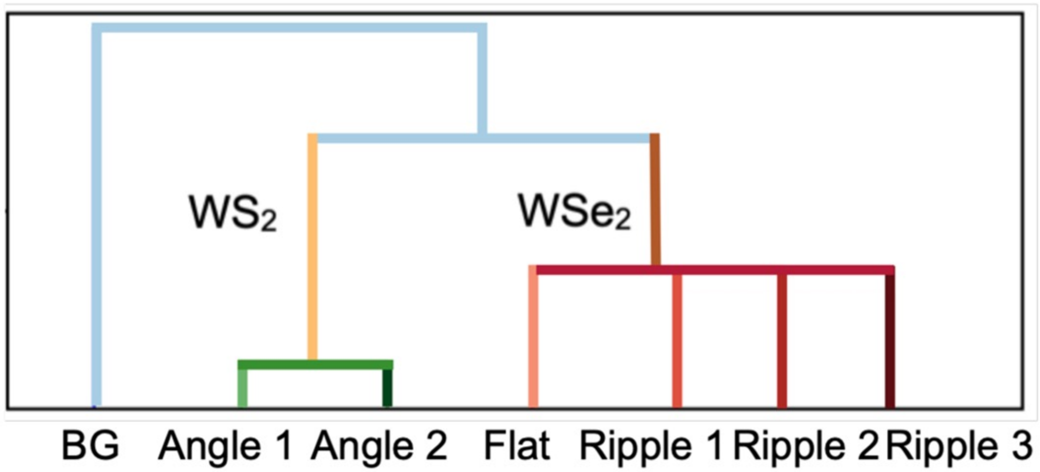
AI as a Powerful Assistance: 4D-STEM

4D-STEM Data Analysis [1]:



Input **Alignment** **Unsupervised Clustering**

Hierarchical k-means clustering

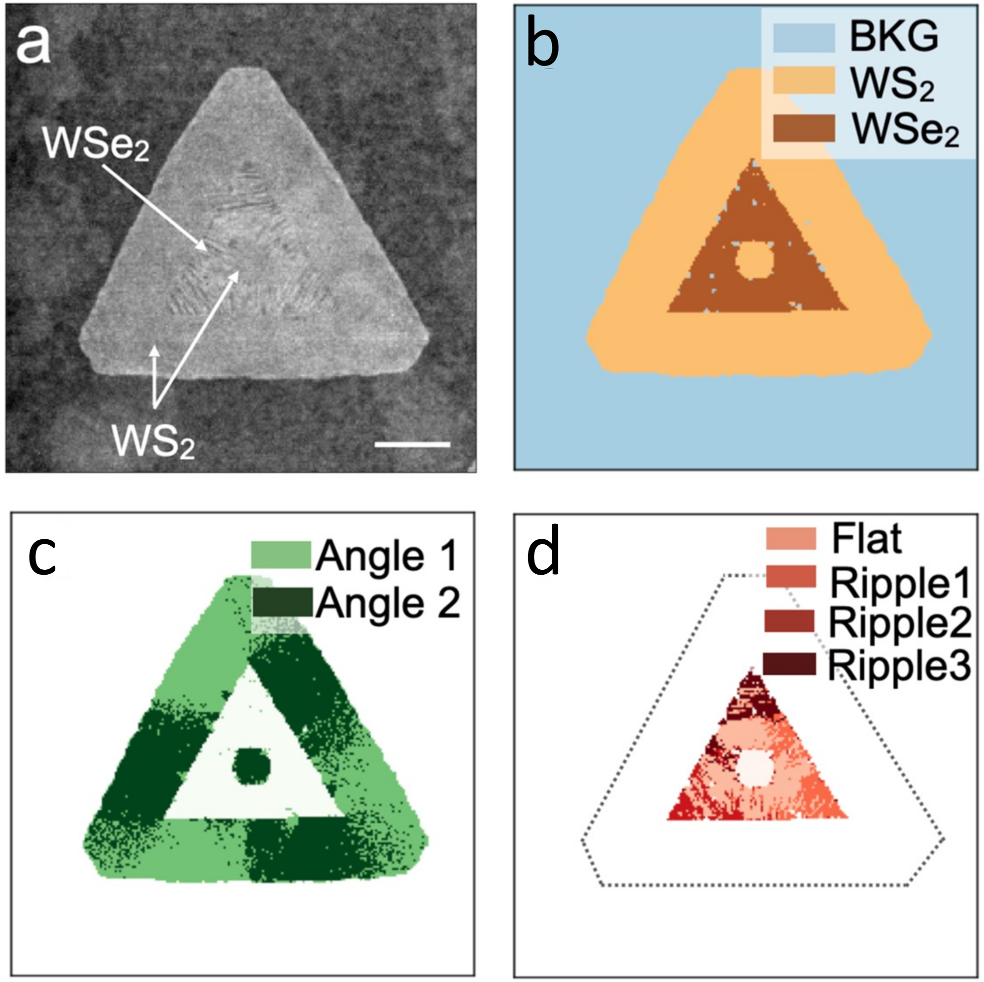


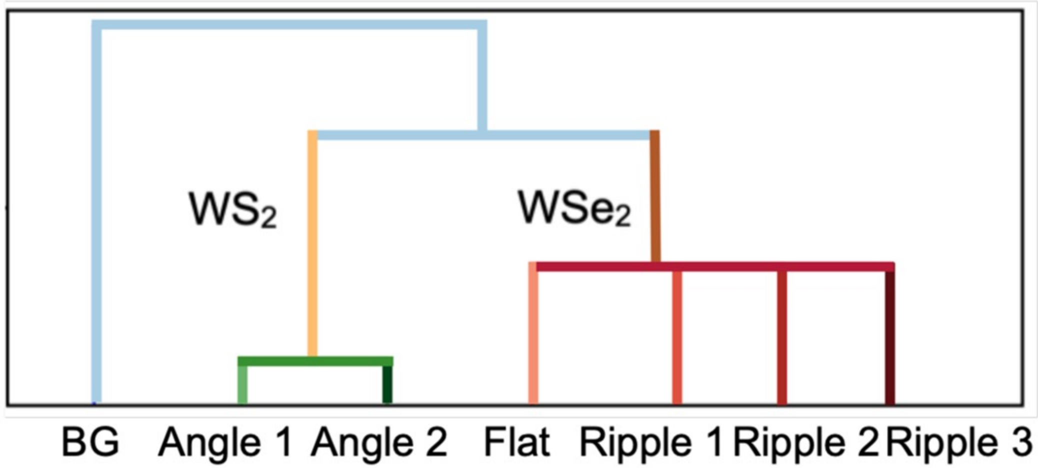
Features of different scales are **clustered sequentially**

[1] Kimoto, Koji, et al. "Unsupervised machine learning combined with 4D scanning transmission electron microscopy for bimodal nanostructural analysis." *Scientific Reports* 14.1 (2024): 2901.

AI as a Powerful Assistance: 4D-STEM

4D-STEM Analysis:

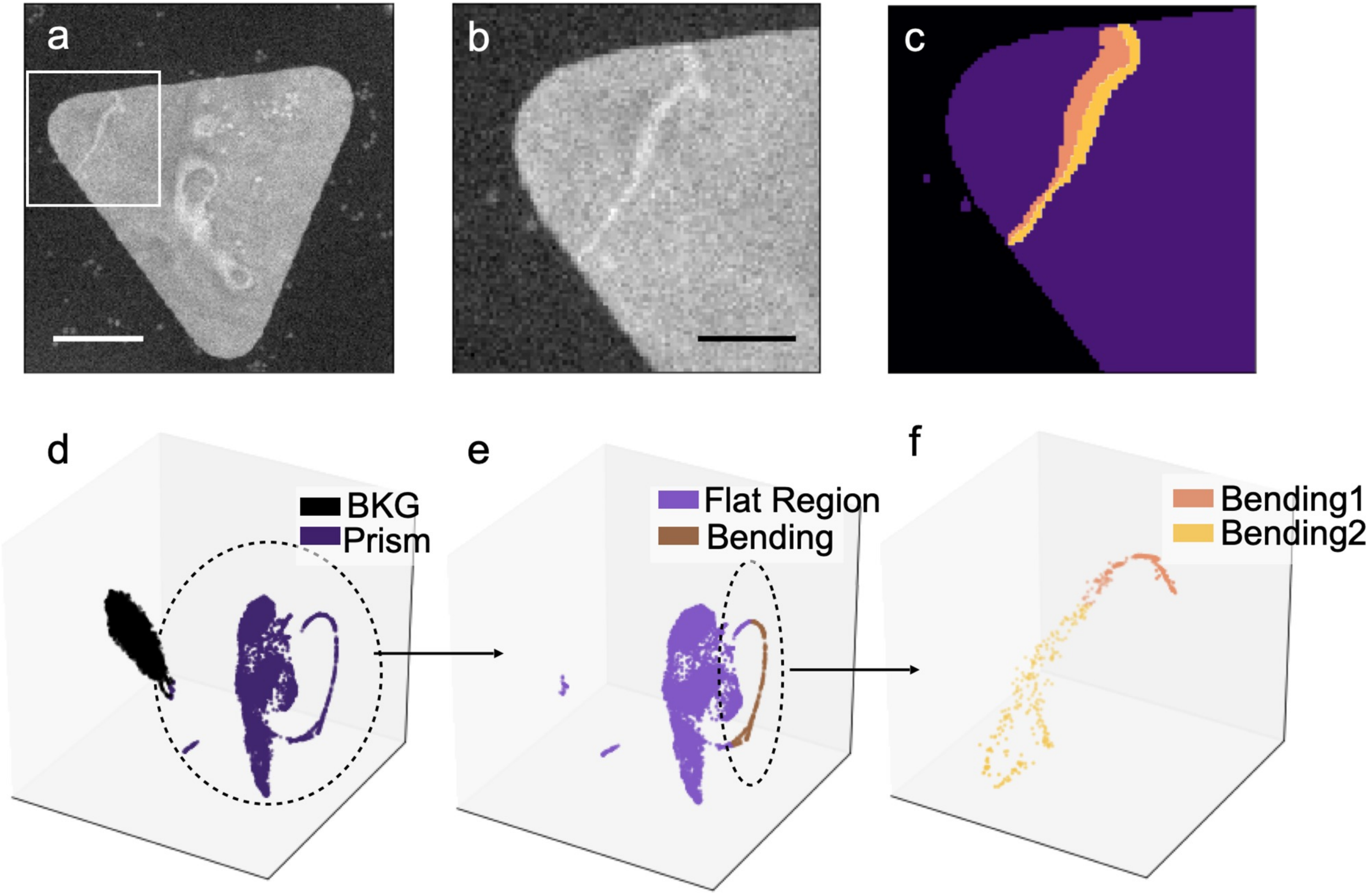




Features of different scales are **clustered sequentially**

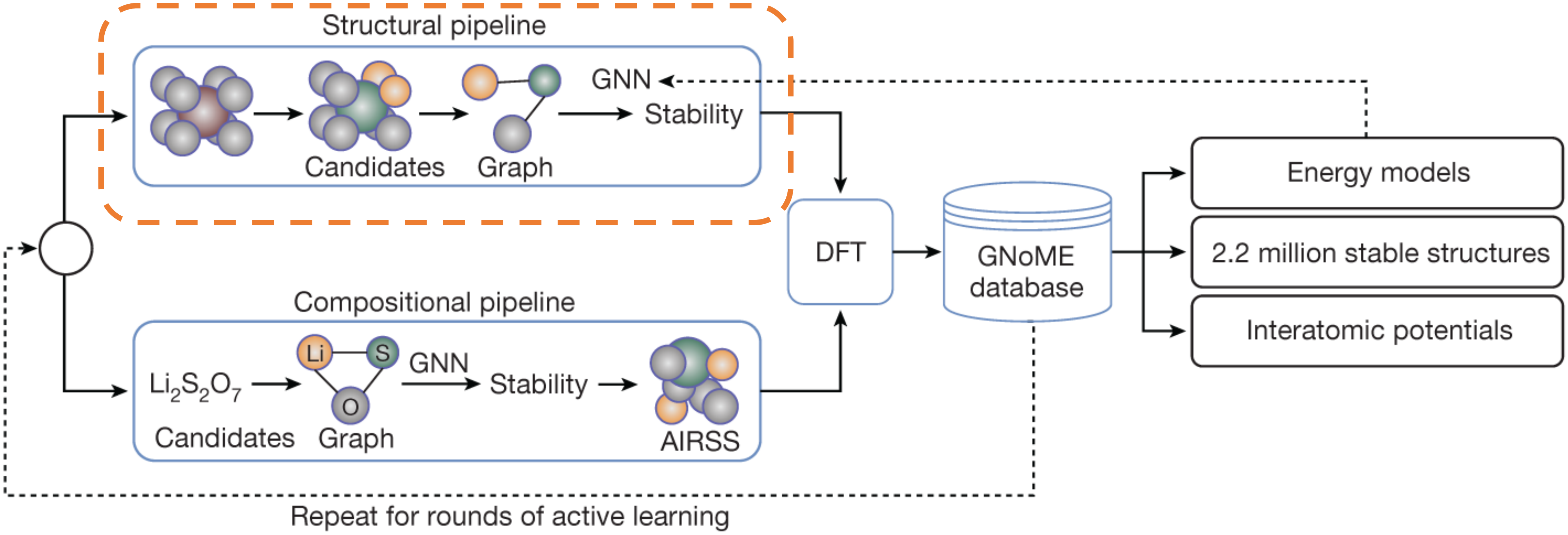
AI as a Powerful Assistance: 4D-STEM

4D-STEM Analysis:



AI as a Powerful Assistance: Material Discovery

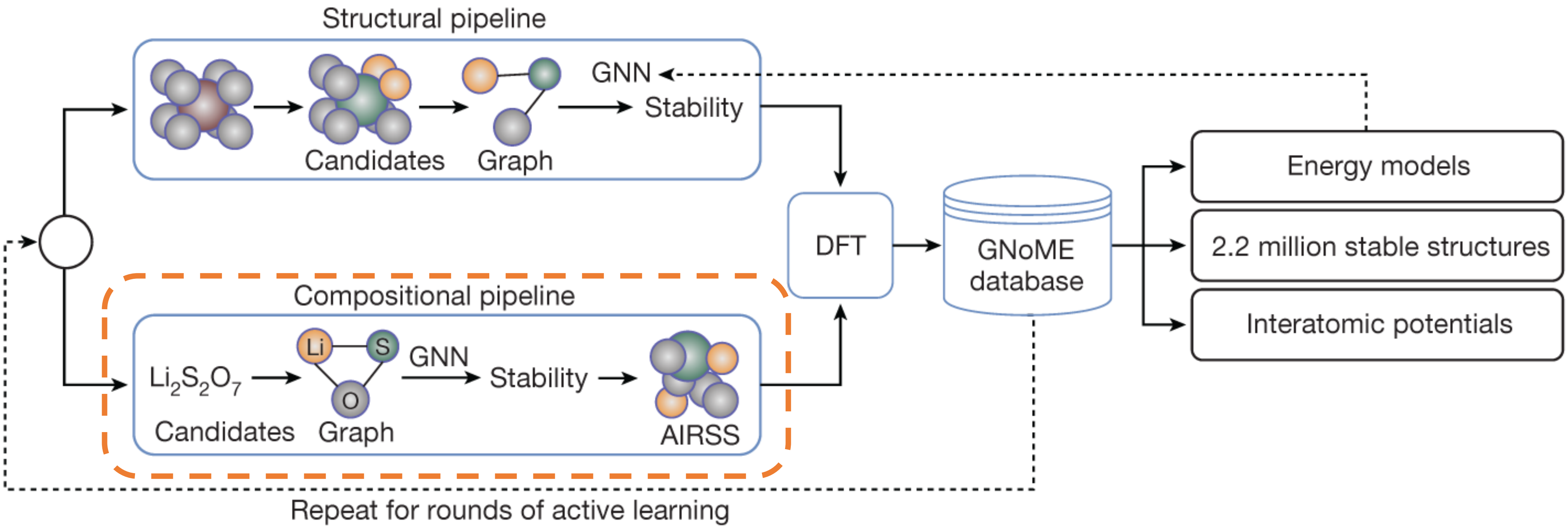
GNoME [1]:



[1] Merchant, Amil, et al. "Scaling deep learning for materials discovery." *Nature* 624.7990 (2023): 80-85.

AI as a Powerful Assistance: Material Discovery

GNoME:



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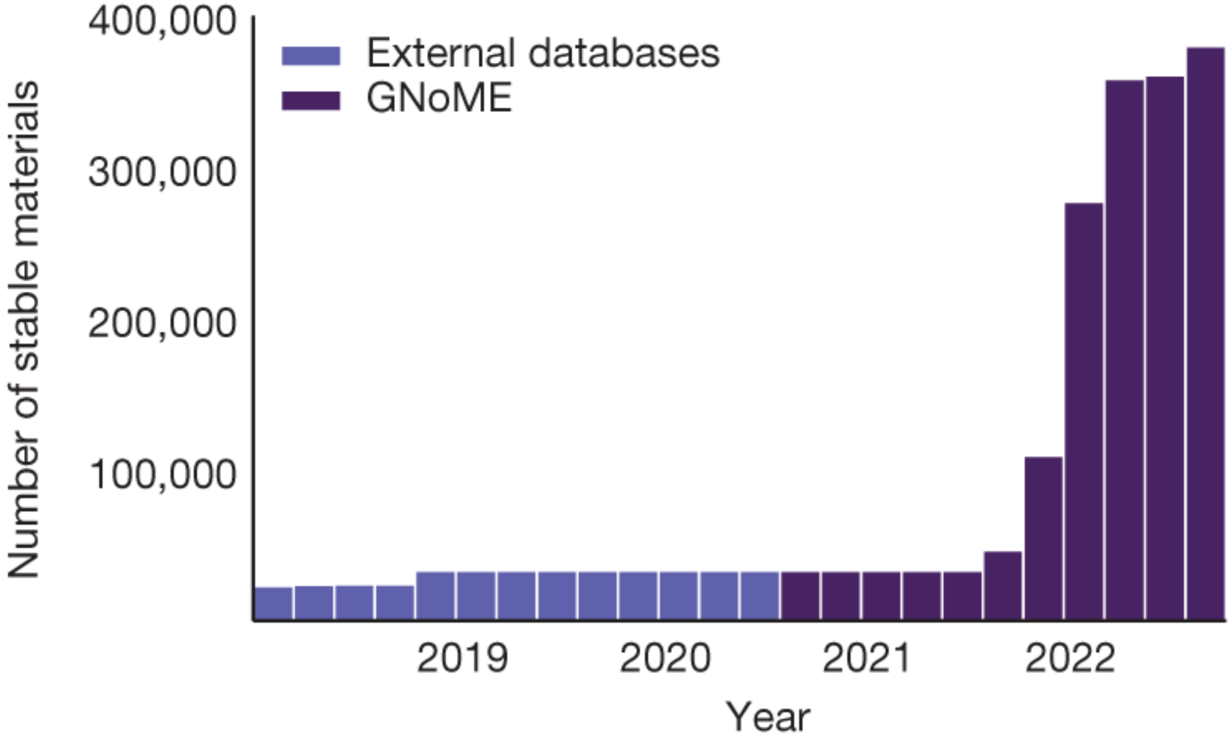
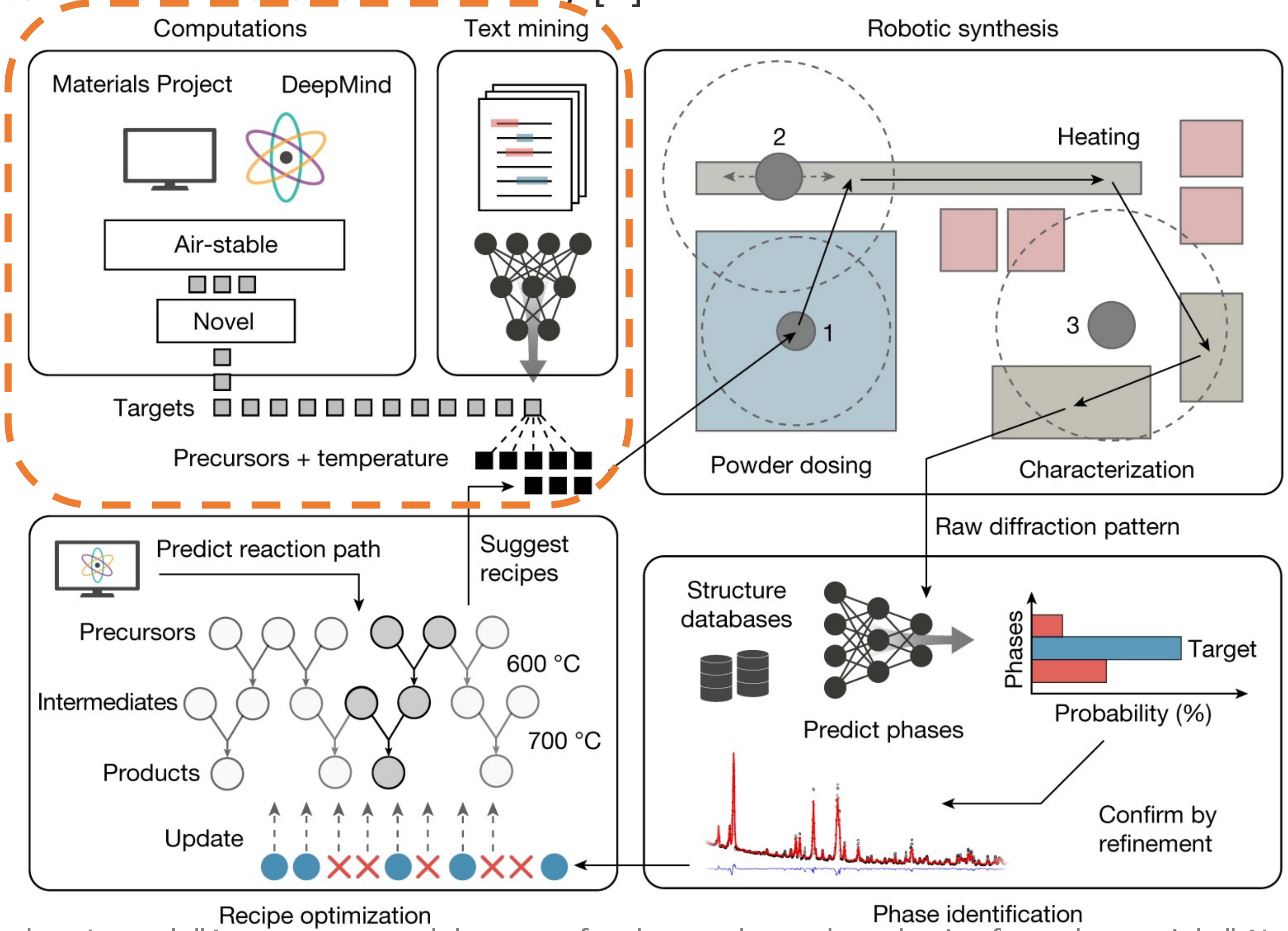


Discovered up to **380,000** computationally stable structures never found before

AI as a Powerful Assistance: Material Discovery

Autonomous materials discovery [1]:



1

2

3

[1] Szymanski, Nathan J., et al. "An autonomous laboratory for the accelerated synthesis of novel materials." *Nature* 624.7990 (2023): 86-91.

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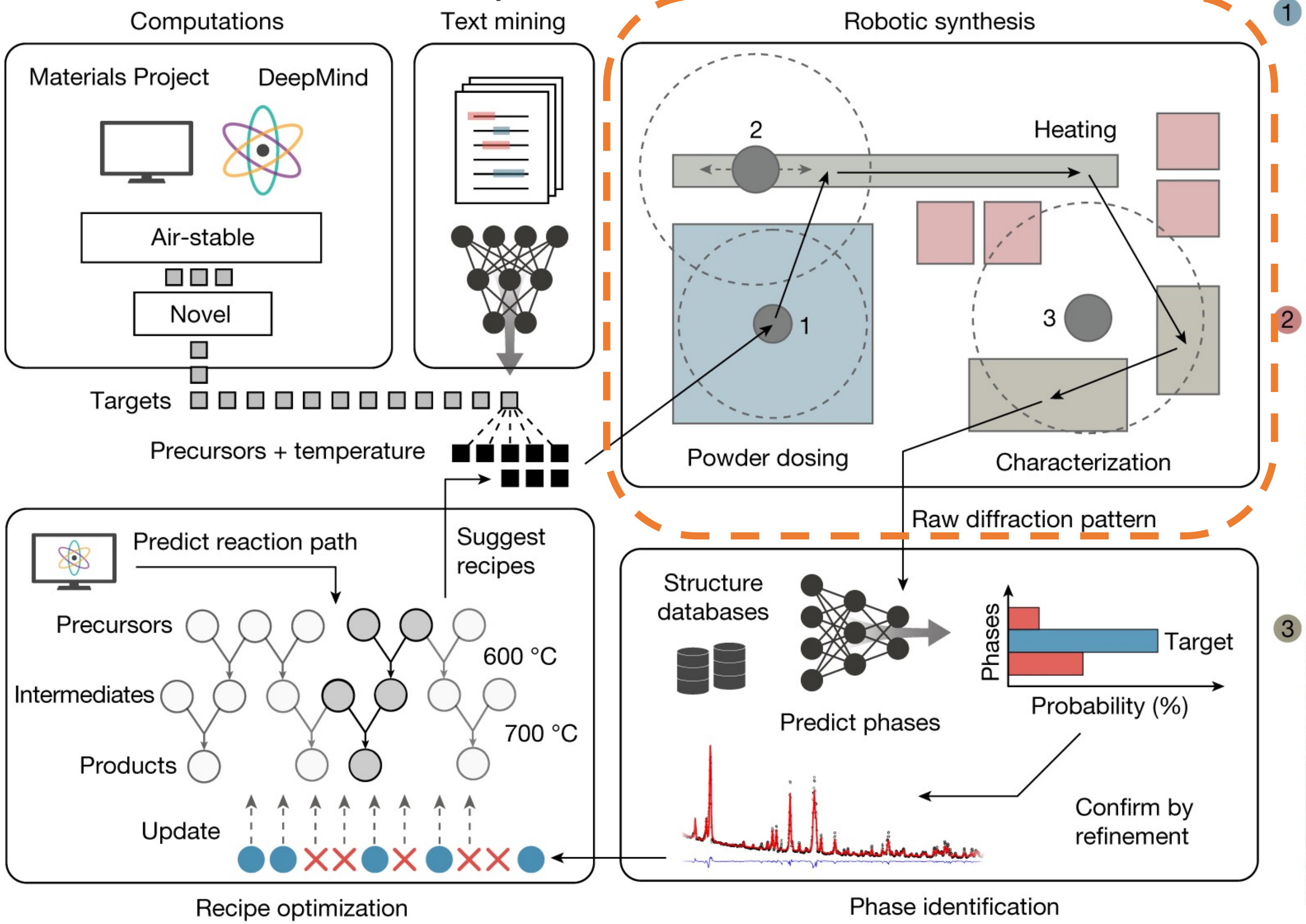
Autonomous materials discovery:



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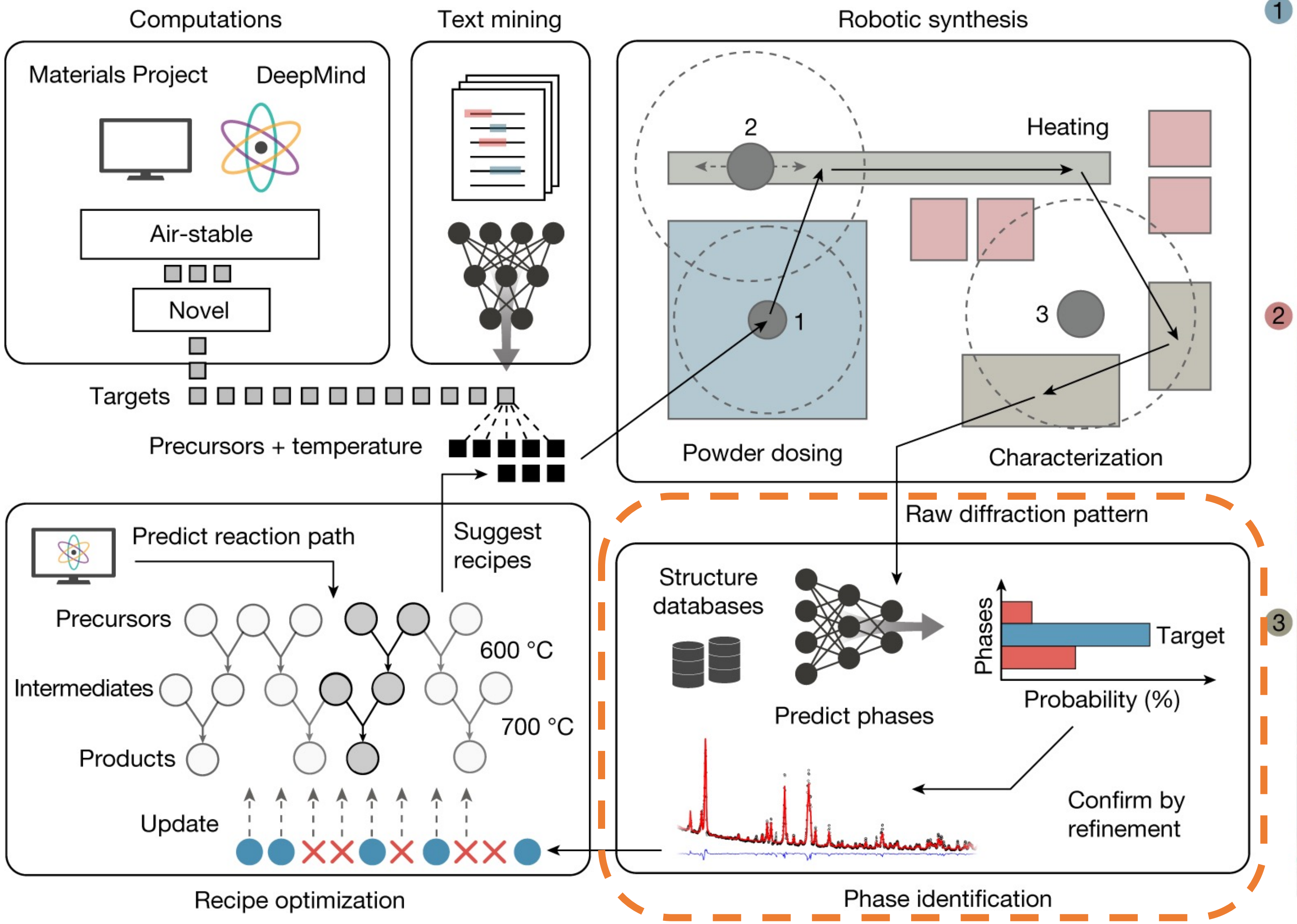
Autonomous materials discovery:



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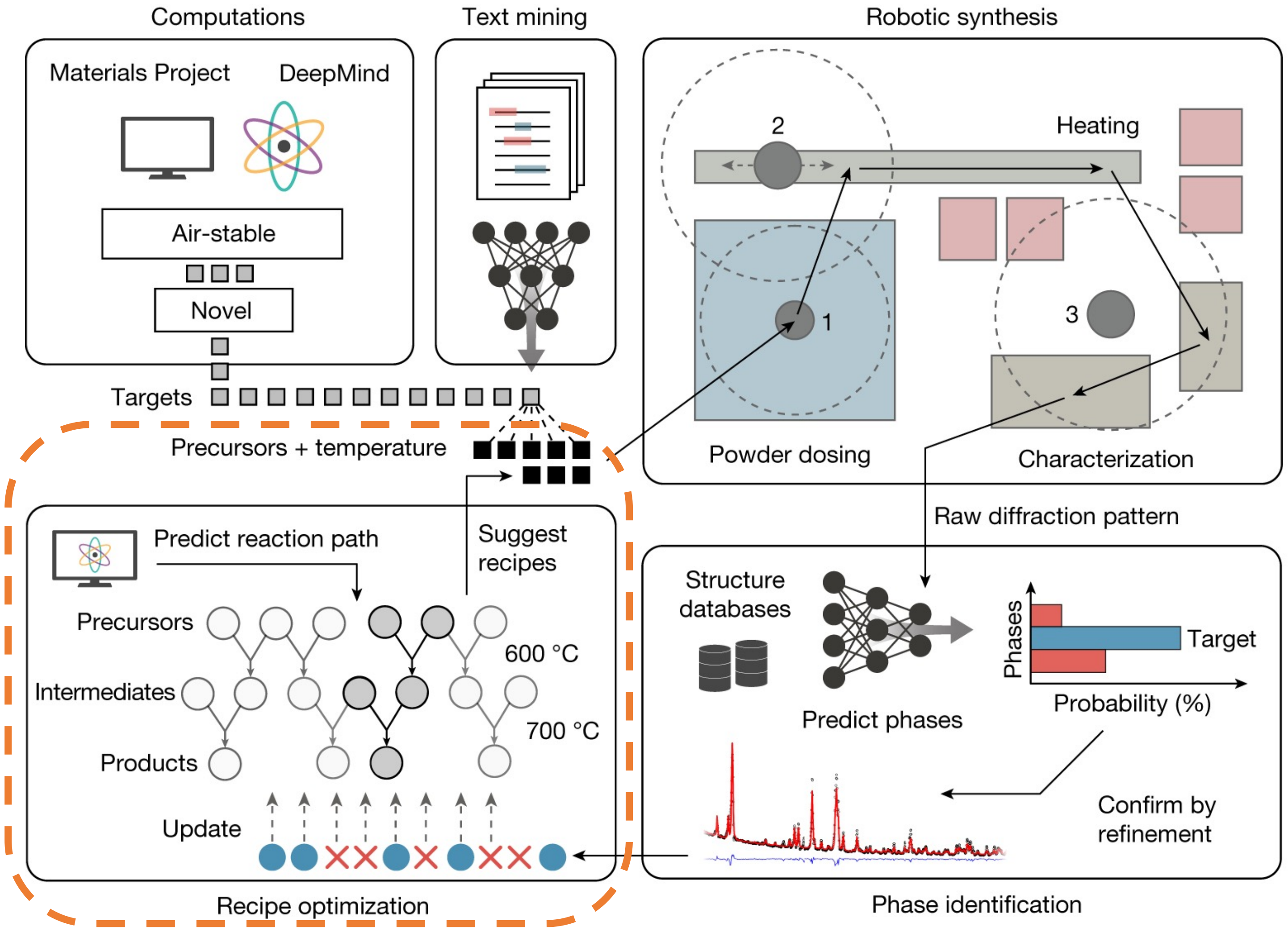
Autonomous materials discovery:



1

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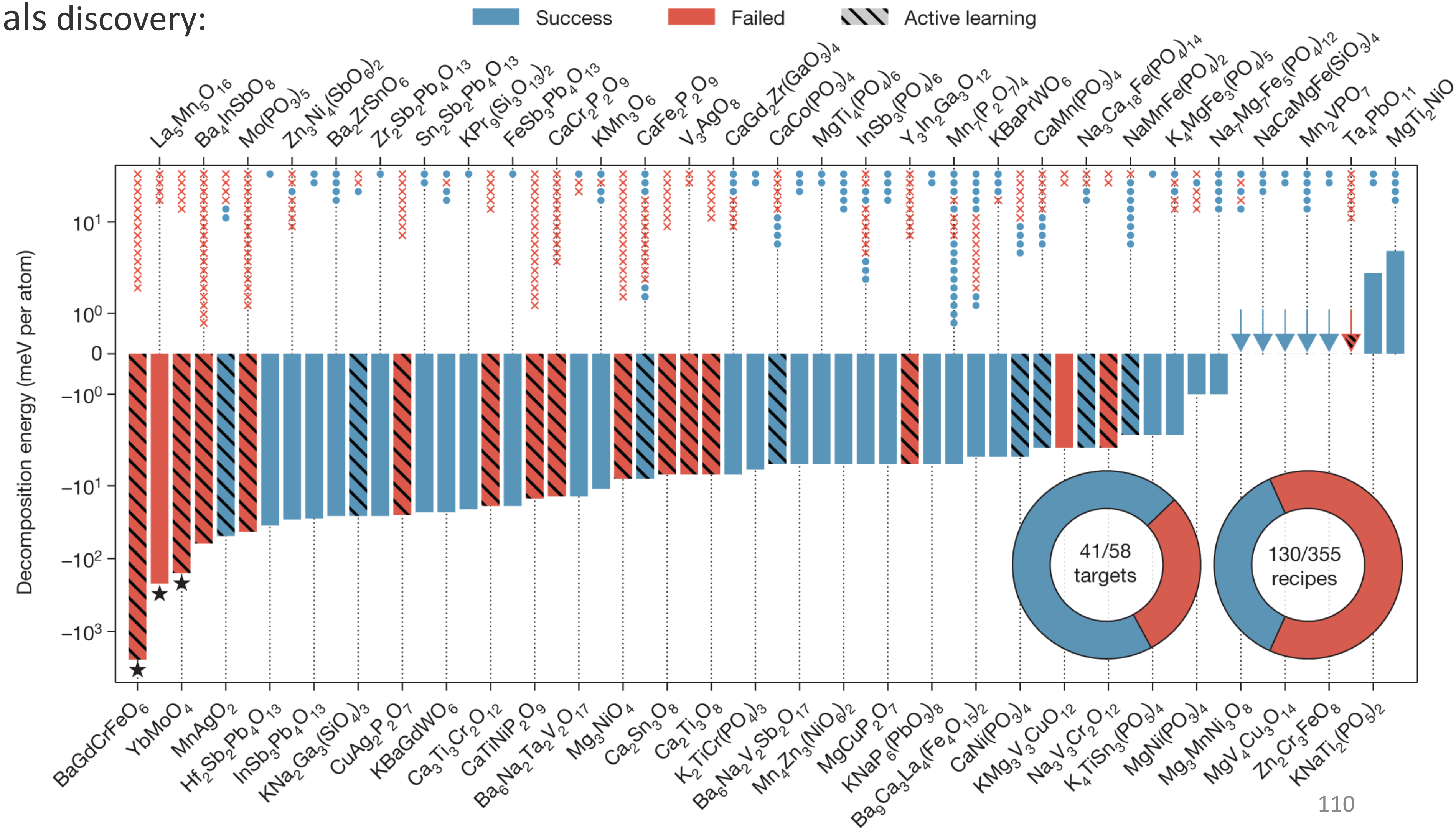
AI as a Powerful Assistance :

Autonomous materials discovery:

17 days:

- 41 new materials
- 9 w/ active learning
- 17 failed



AI in Materials Science

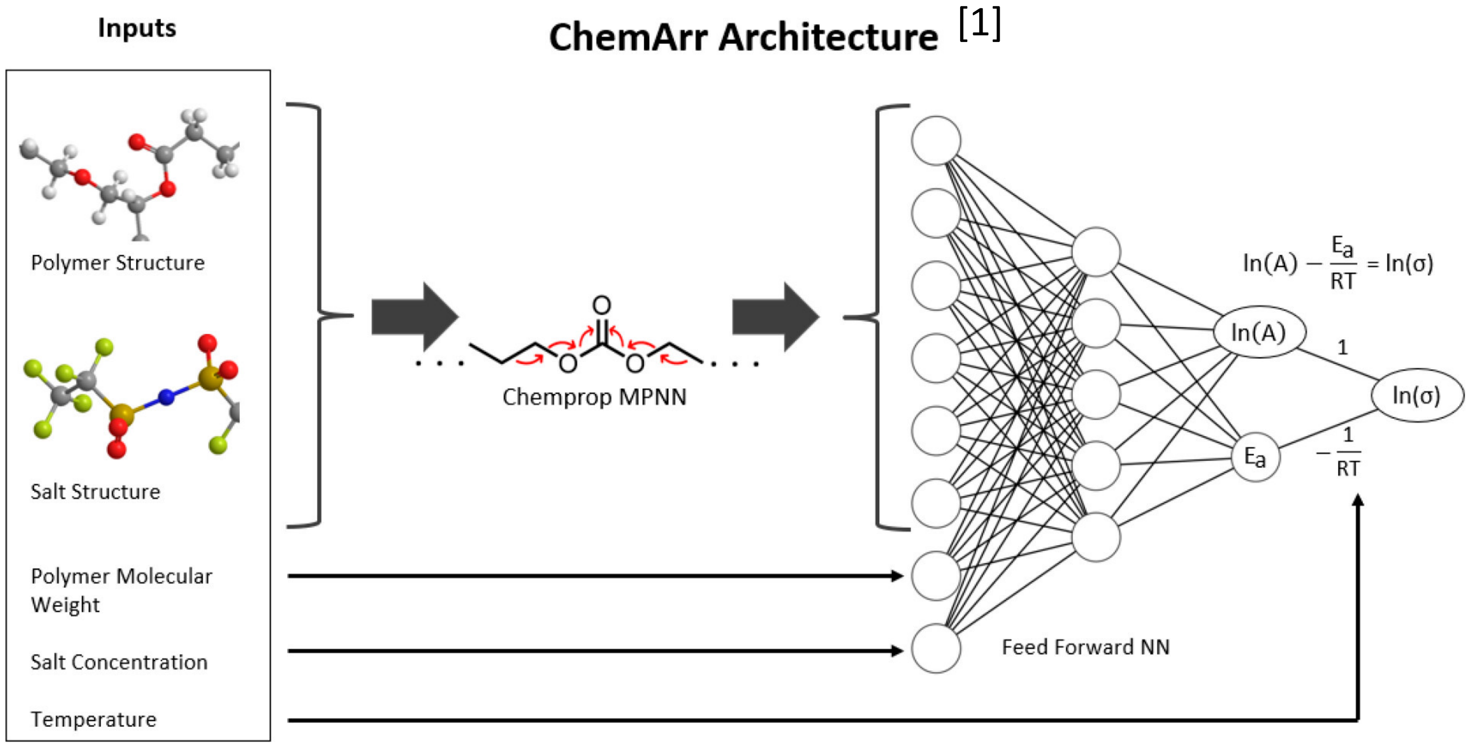
Outside and Beyond Crystals

Outside of the Crystals: Amorphous Materials

AI in Polymer Science:

- Input polymer electrolyte structure and formula
- Predicating ionic conductivities σ by predicting pre-exponential factor A and activation energy E_a in the Arrhenius equation:

$$\ln(\sigma) = \ln(A) - \frac{E_a}{RT}$$

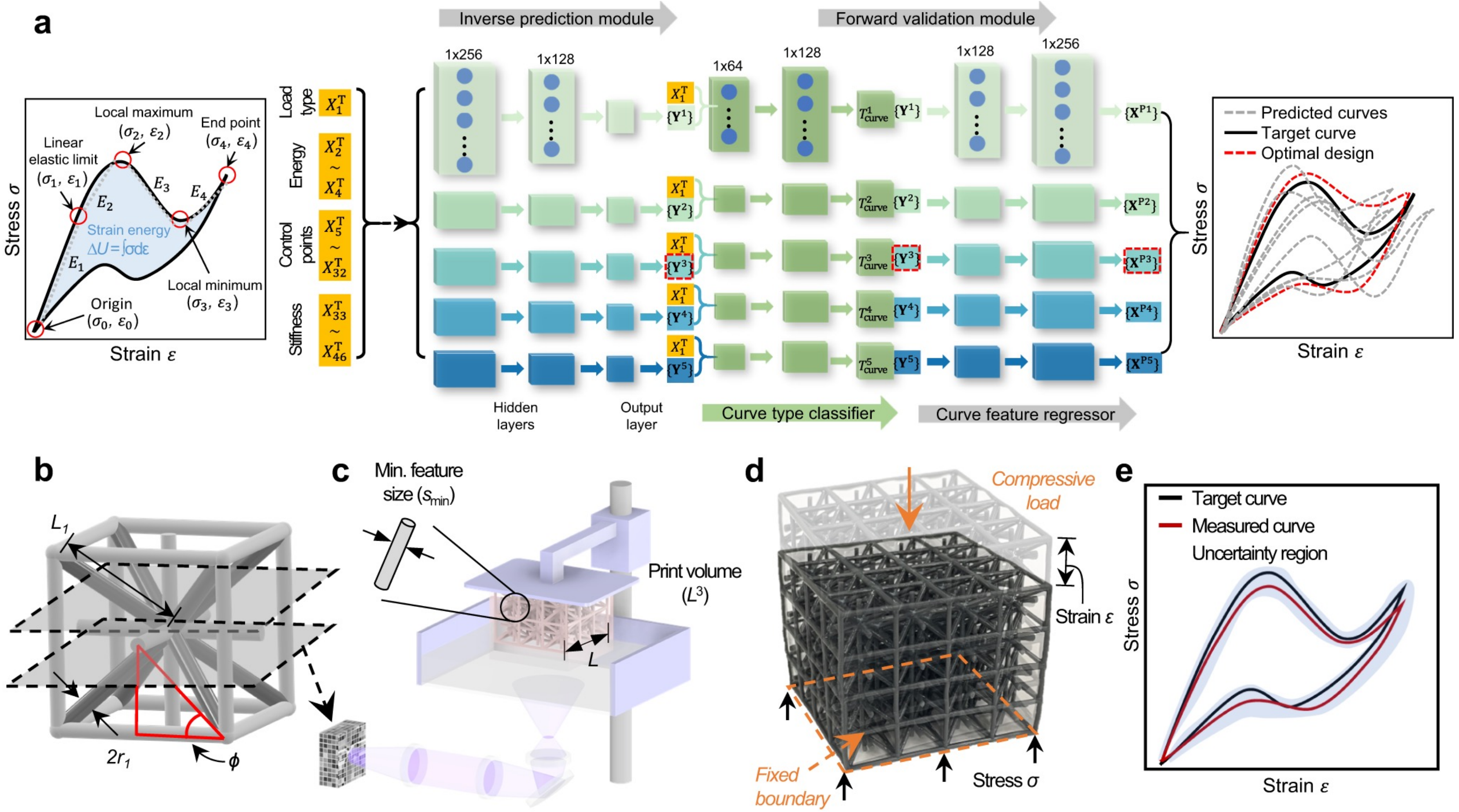


[1] Bradford, Gabriel, et al. "Chemistry-informed machine learning for polymer electrolyte discovery." *ACS Central Science* 9.2 (2023): 206-216.

Beyond Crystals: MetaMaterials

Metamaterials based on prescribed mechanical behavior [1]

- Mechanical properties determined by mm scale topology (10 mm unit cell)
- Inverse design of topology structure with the target compressive behavior in the form of curve features



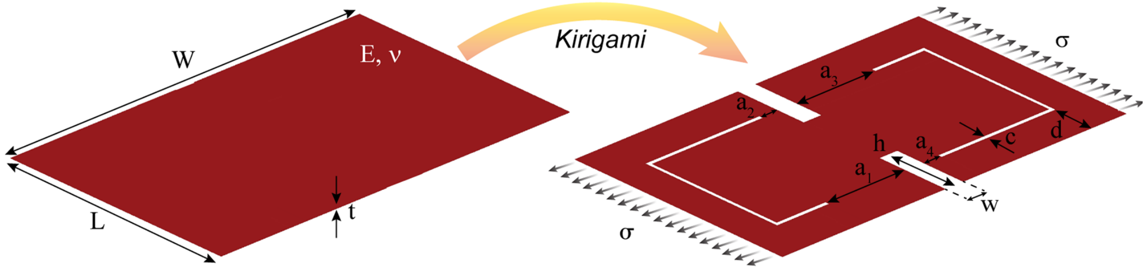
[1] Ha, Chan Soo, et al. "Rapid inverse design of metamaterials based on prescribed mechanical behavior through machine learning." Nature Communications 14.1 (2023): 5765.

Beyond Crystals: MetaMaterials

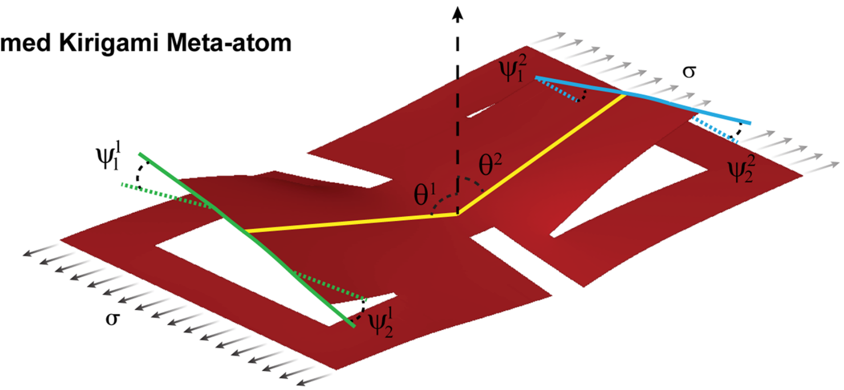
Shape-programmable 3D kirigami metamaterials [1]

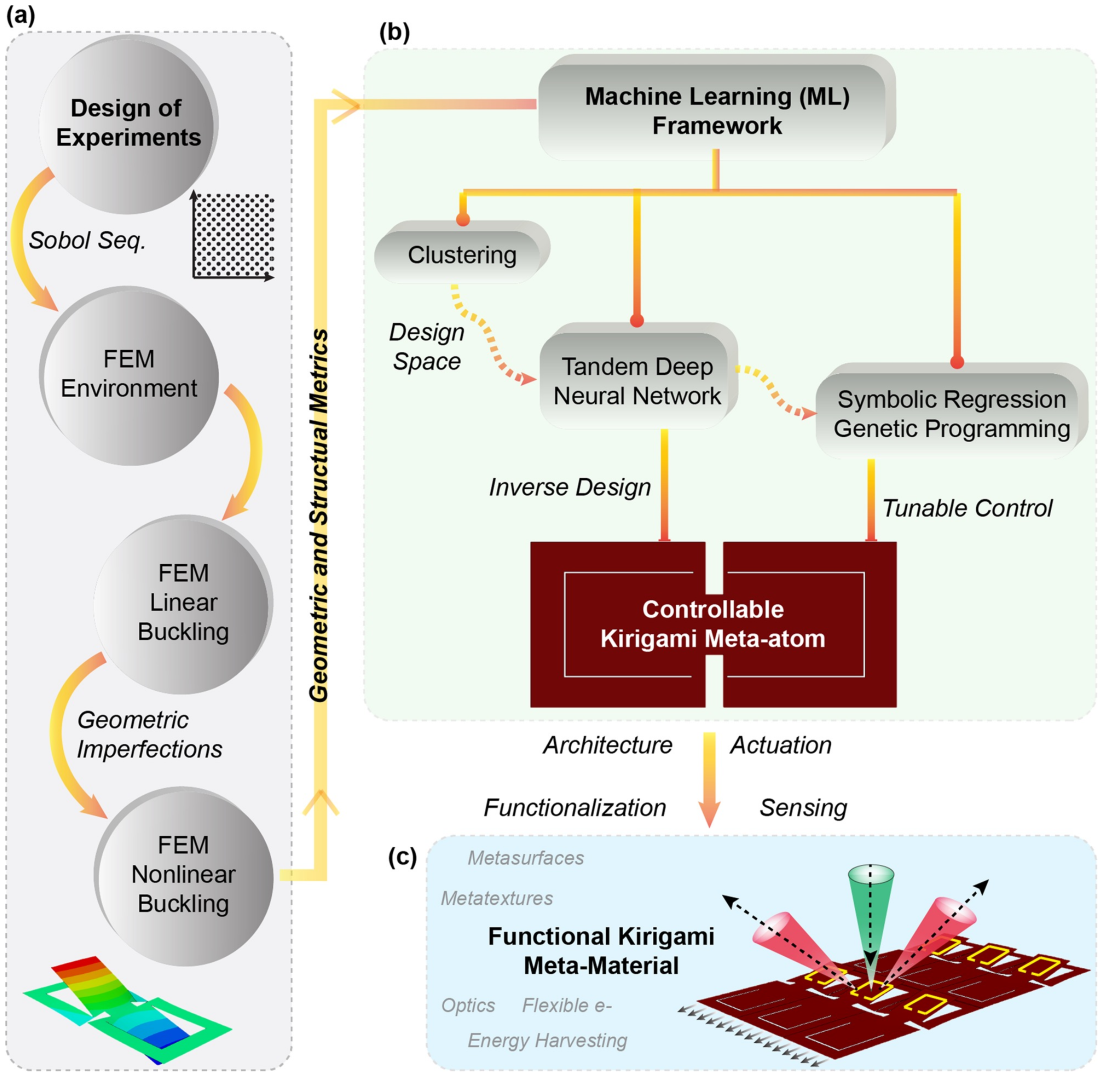
- Inverse design of cut layout with the desired deformation

(a) Kirigami Meta-atom



(b) Deformed Kirigami Meta-atom





[1] Alderete, Nicolas A., Nibir Pathak, and Horacio D. Espinosa. "Machine learning assisted design of shape-programmable 3D kirigami metamaterials." npj Computational Materials 8.1 (2022): 191.

Useful Resources:

- A Tutorial on Density Functional Theory: https://www.researchgate.net/publication/226474665_A_Tutorial_on_Density_Functional_Theory
- Material Project: https://next-gen.materialsproject.org/
- MP Seminar – Inverse Design: Why Aren't We There Yet?: https://youtu.be/0lO7QNAexRc?si=EaNuCcuwwDlDfBxM
- MP Seminar – MatterGen: https://youtu.be/Smz1go6_Spo?si=xnU8kXWNFVIYf-Zf

Additional materials

Physical properties derived from the DFT Hamiltonian

- Band structure and DOS:
  - The eigenvalues $\varepsilon_{n\boldsymbol{k}}$ and eigenstates $v_{n\boldsymbol{k}}$ of the Hamiltonian $\widehat{H}$ at band n and wavevector $\boldsymbol{k}$ can be obtained by solving the generalized eigenvalue problem:
  $$H(\boldsymbol{k})v_{n\boldsymbol{k}} = \varepsilon_{n\boldsymbol{k}}S(\boldsymbol{k})v_{n\boldsymbol{k}}$$
  - where the overlap matrix S is obtained by the inner product of the basis at very low computational cost
  - $\varepsilon_{n\boldsymbol{k}}$ of at band n and wavevector $\boldsymbol{k}$ construct the band structure
  - DOS is obtained by integrating the number of electronic in momentum space in the band structure over each value of energy:
  $$D(E) = \int \delta\big(E - E(\boldsymbol{k})\big)d^3\boldsymbol{k}$$

Physical properties derived from the DFT Hamiltonian

- Shift current conductivity σ^{abc}:

$$\sigma^{abc}(\omega) = \frac{\pi e^3}{\hbar^2}\int \frac{d^3\boldsymbol{k}}{(2\pi)^3} \times \sum_{n,m} f_{nm}\mathrm{Im}\left(r_{mn}^{b} r_{nm}^{c,a} + r_{mn}^{c} r_{nm}^{b,a}\right)\delta(\omega_{mn}(\boldsymbol{k}) - \omega)$$

- $\omega_{mn}(\boldsymbol{k}) = \frac{E_{n\boldsymbol{k}} - E_{m\boldsymbol{k}}}{\hbar}$: difference of energy eigenvalues
- $f_{nm} = f_n(\boldsymbol{k}) - f_m(\boldsymbol{k})$: Fermi–Dirac occupations of bands n and m at wavevector $\boldsymbol{k}$
- r_{mn}^{a} and $r_{nm}^{b,a} = \frac{\partial r_{nm}^{b}}{\partial k^a} - i(r_{nn}^{a} - r_{mm}^{a})r_{nm}^{b}$ are Berry connection and its general derivative, calculated with the DFT Hamiltonian using the method developed in ref [1]

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[1] Wang, Chong, et al. "First-principles calculation of optical responses based on nonorthogonal localized orbitals." *New Journal of Physics* 21.9 (2019): 093001.