# AI for Materials Geometric Representation Learning and High Tensor Order Property Predictions

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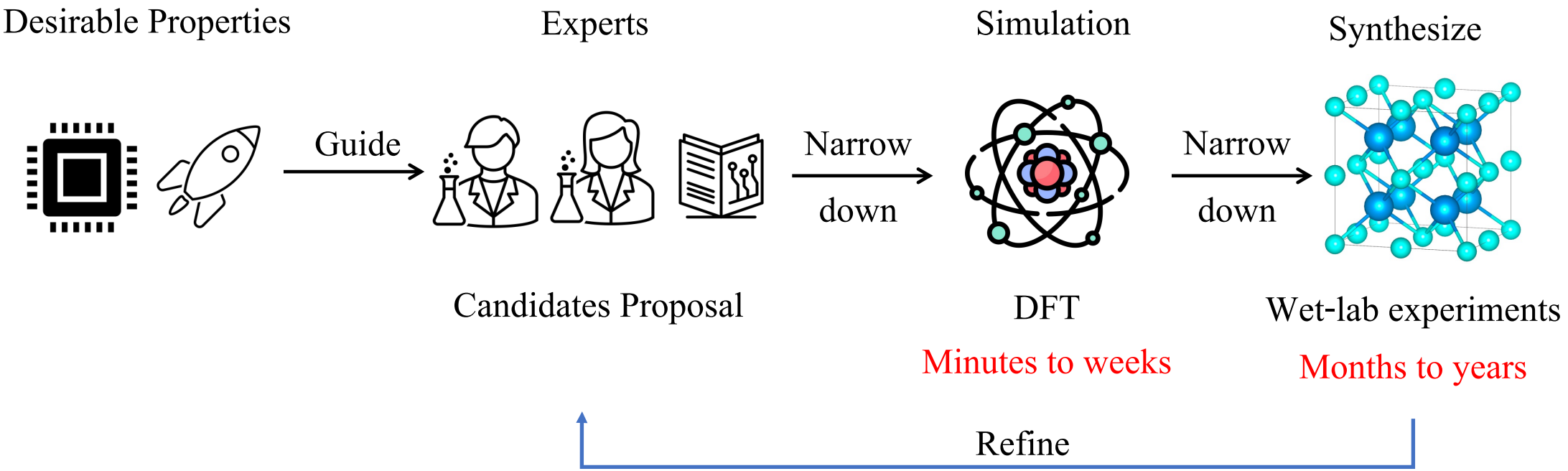
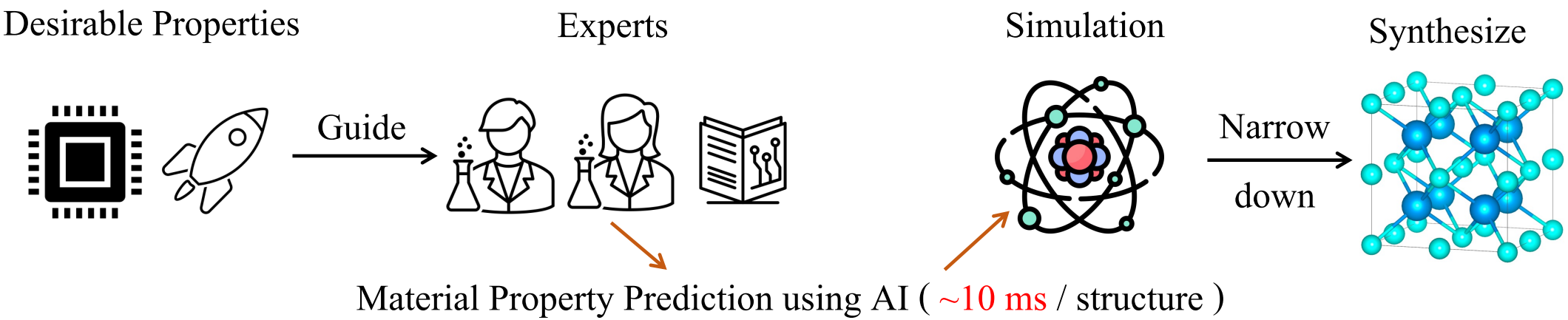
# Discovery of Novel Materials




- Experiments are expensive and time-consuming v.s. > **10^9** material search space
- Experts and DFT algorithms $O(n^3 \sim n^7)$ can narrow down the candidate list

*Figures designed using images from Flaticon.com*

# Speed Up Material Discovery using AI

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# Crystal Structures

- Crystal Structures
  - **Lattice**
    - $L \in R^{3\times3}$
    - How unit cell repeats itself in 3D space
  - **Atom matrix**
    - $A \in R^{n\times1}$
    - Atomic numbers of atoms in the unit cell
  - **Atom Position matrix**
    - $P \in R^{n\times3}$
    - 3D positions of atoms in the unit cell

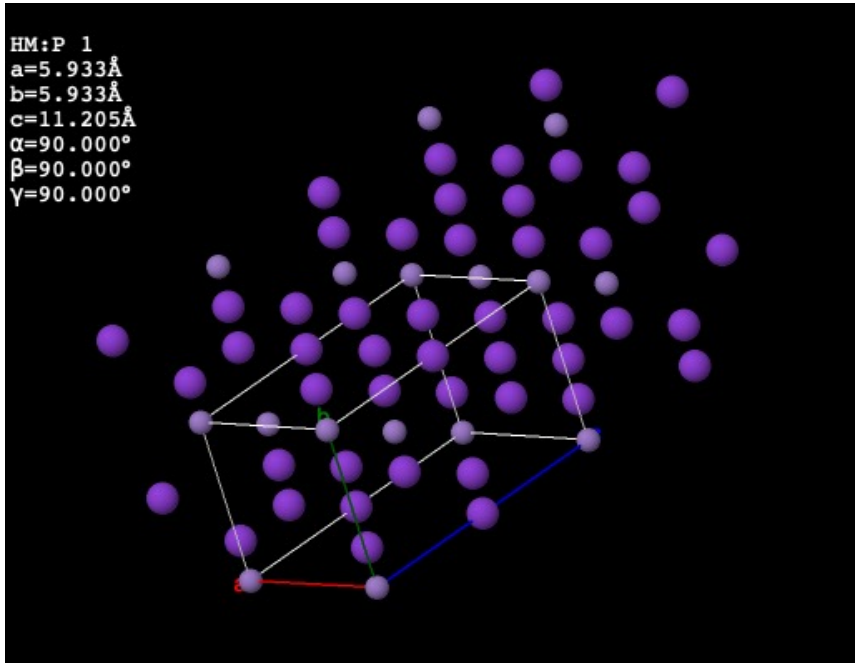
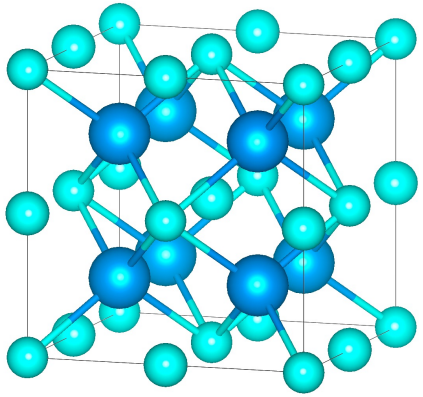
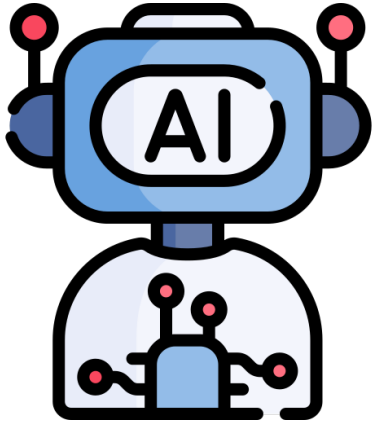



*Image from Material Project : https://materialsproject.org/batteries/mp-1184916_K/*

# Geometric Representation Learning

The bridge between real world atomic systems and AI






Crystal structures

Complex 3D structures

Near **infinite unexplored** structural space

Geometric representations

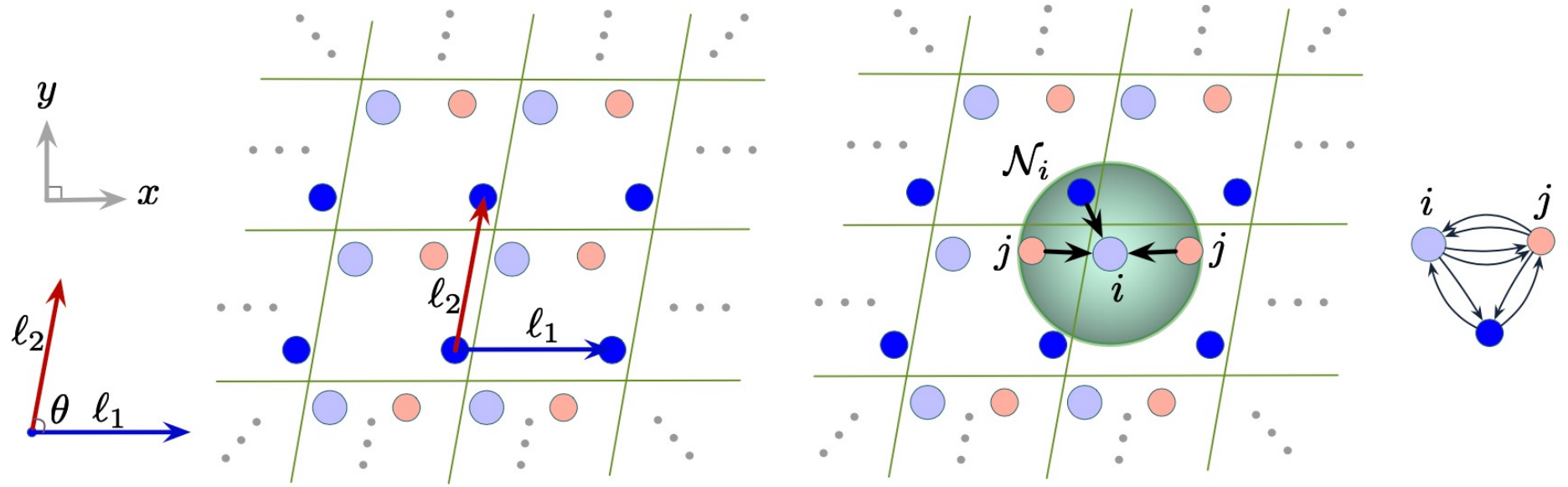
Converting structures into representations that **capture geometric information**

AI methods

Only consider information **contained in representations**!

*Figures designed using images from Flaticon.com*

# Previous Multi-edge Graphs




Only captures bond lengths and atom types within a radius

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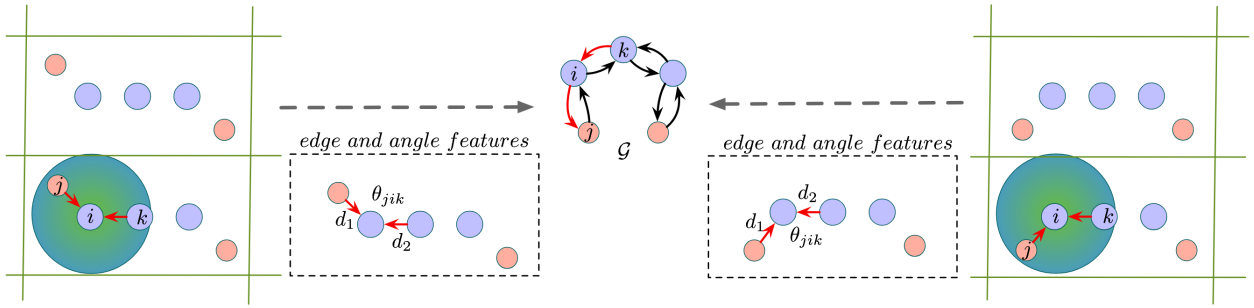
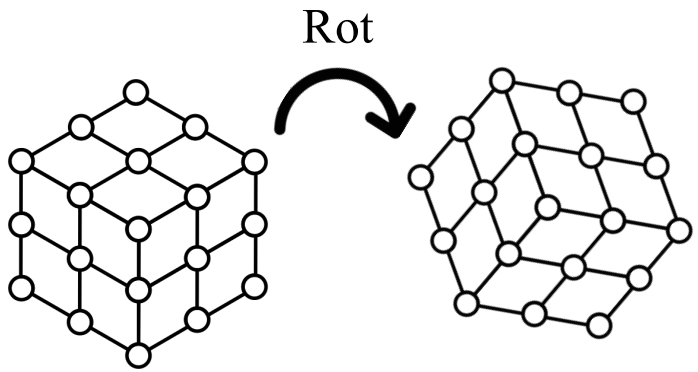
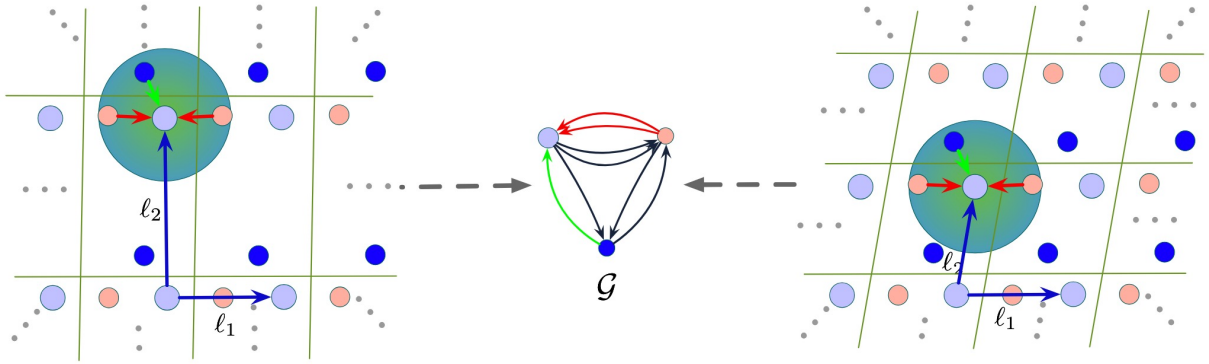
# Geometric Representation Learning

Instead, we want the geometric representations to

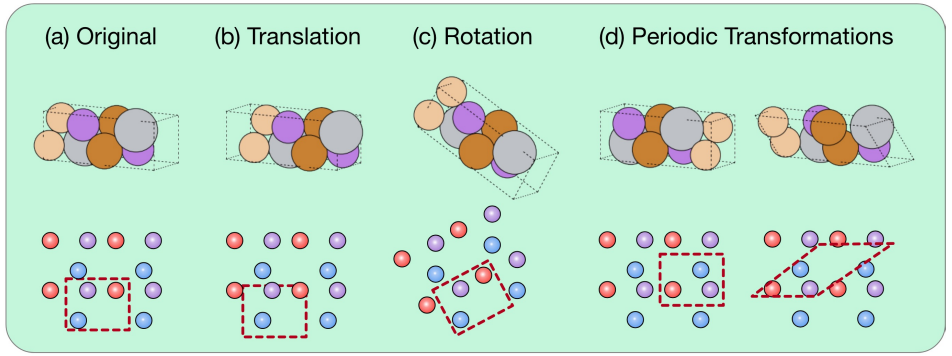



***Model different crystals differently***



***Model equivariant crystals equivariantly***

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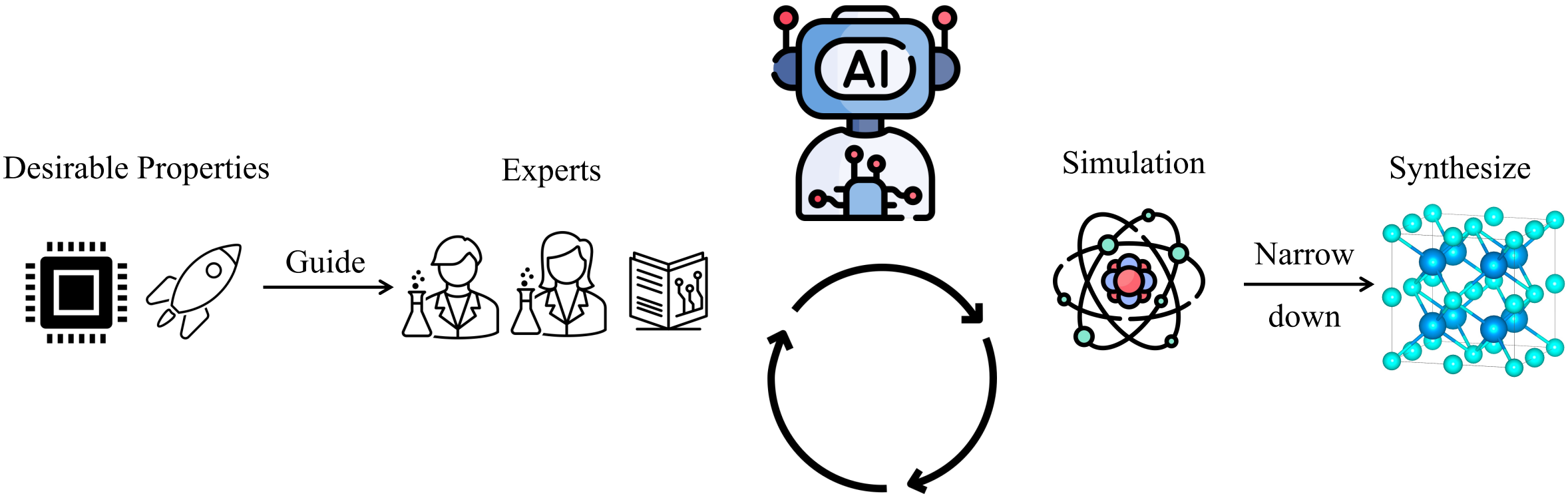
# Geometric Representation Learning + AI

Given robust and powerful geometric representations, designing AI methods that are

- Accurate, efficient, and robust
- Screening potential crystal structures $\sim 10^5$ times faster

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# Talk Overview

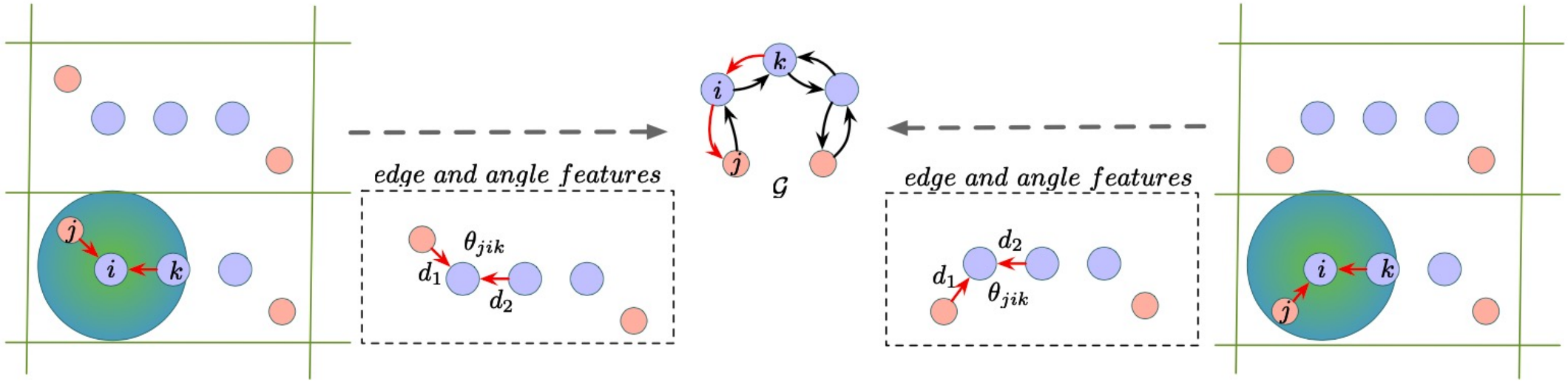
Empowering AI methods to

- distinguish any crystal structural differences - [ICLR 2024]
- predict crystal tensor properties of various order - [ICML 2024]

# Distinguish Any Structural Differences

Can previous methods (Matformer, ALIGNN, M3GNet) distinguish any **structural differences**?

- Distances are not enough (even with bond angles)



# Distinguish Any Structural Differences

Can we design a crystal representation that is complete?

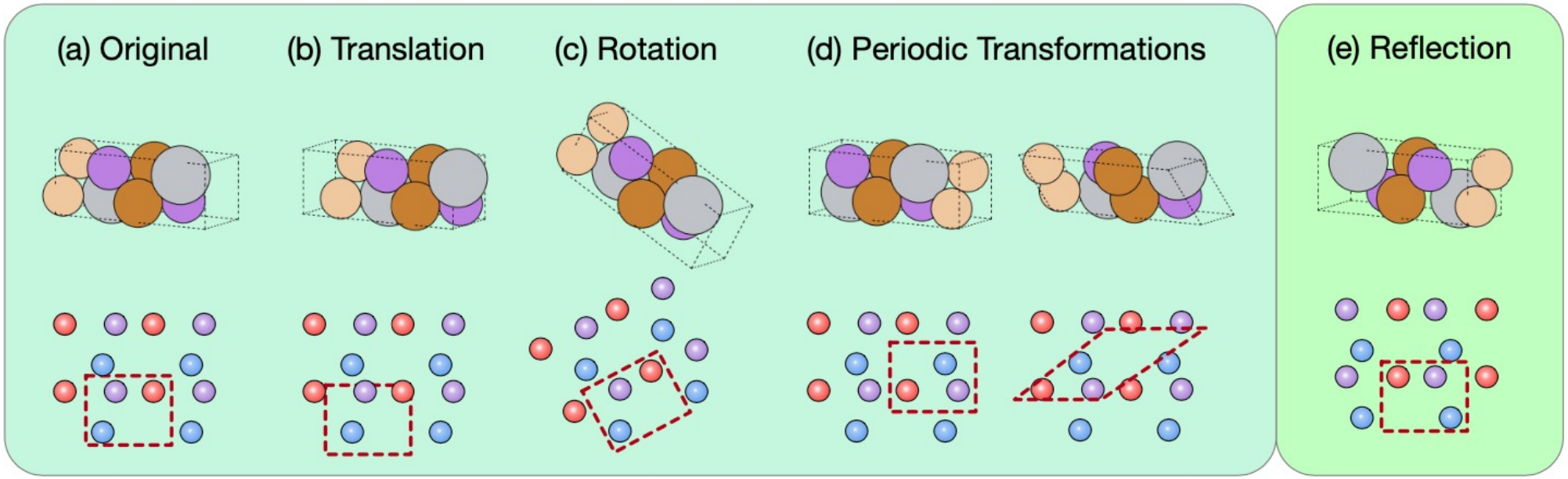
- Ability to capture any structural differences

**Definition 1** (Geometrically Complete Crystal Graph). *Following Widdowson & Kurlin (2022), a crystal graph $\mathcal{G}$ is geometrically complete if $\mathcal{G}_1 = \mathcal{G}_2 \rightarrow \mathbf{M}_1 \cong \mathbf{M}_2$, where $\cong$ denotes that two crystals are isometric as defined in Appendix A.3. It means if two crystal graphs $\mathcal{G}_1$ and $\mathcal{G}_2$ are the same, the infinite crystal structures represented by $\mathcal{G}_1$ and $\mathcal{G}_2$ are identical.*

Yan, Keqiang, et al. "Complete and Efficient Graph Transformers for Crystal Material Property Prediction." *ICLR* (2024).

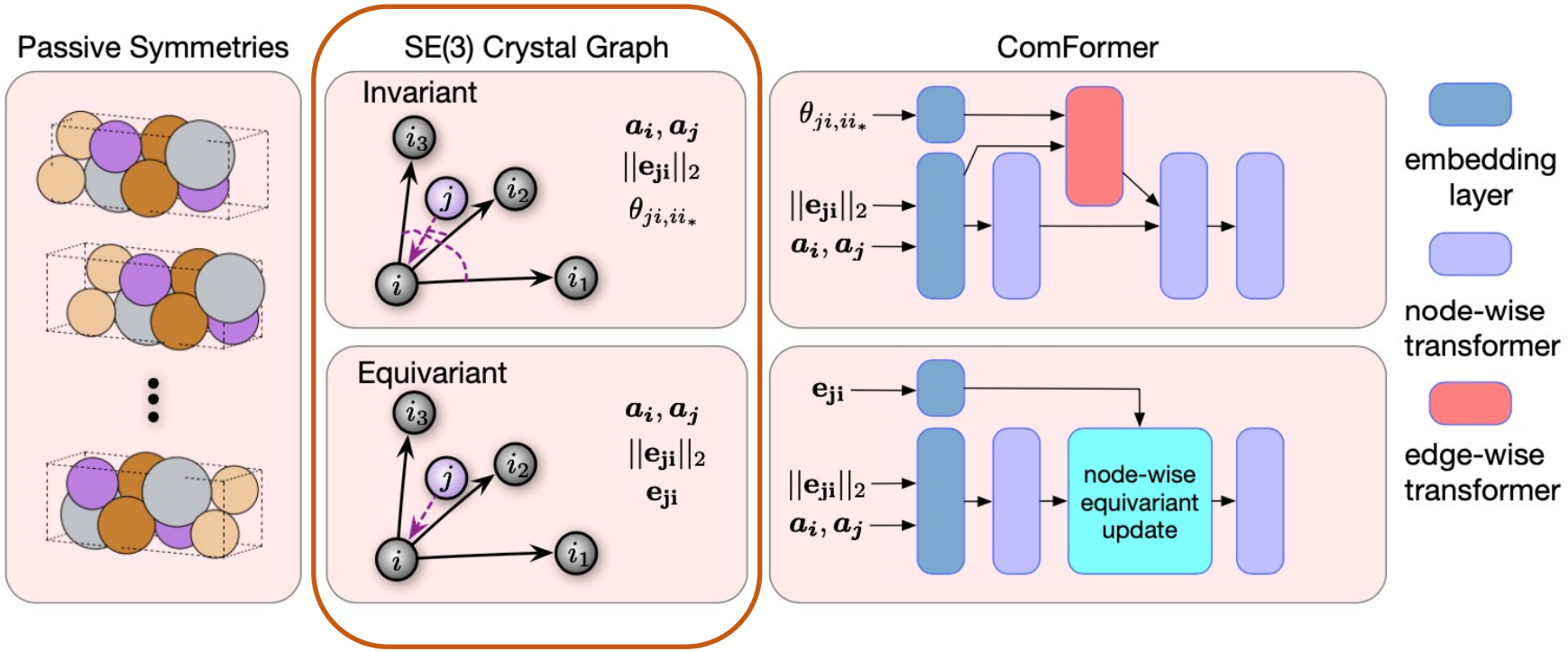
# Challenges

- A crystal can have ***dramatically different*** unit cells, but should have ***a unique*** representation
- Map equivariant crystals equivariantly, e.g., rotates when the structure is rotated
- Efficiency



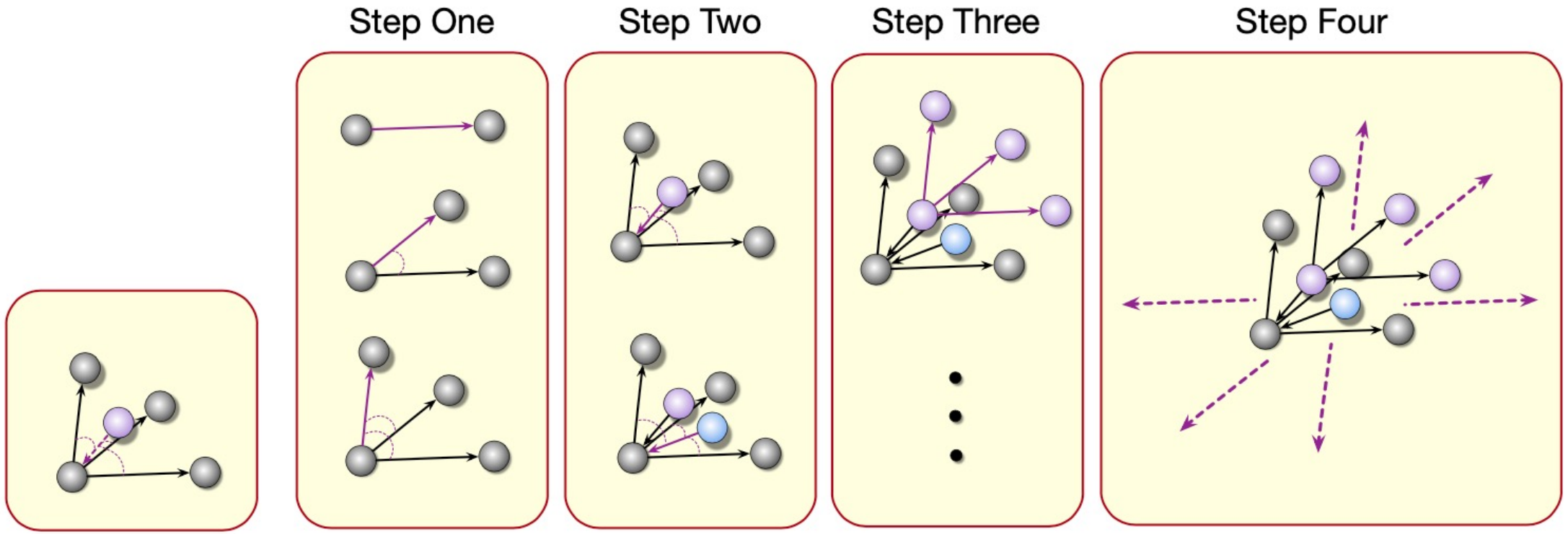
# The proposed ComFormer

- Insights: periodic patterns are the same for every atom in a crystal
  - First uniquely determine a unit cell
  - Use periodic patterns to form a ***lattice frame*** for every atom



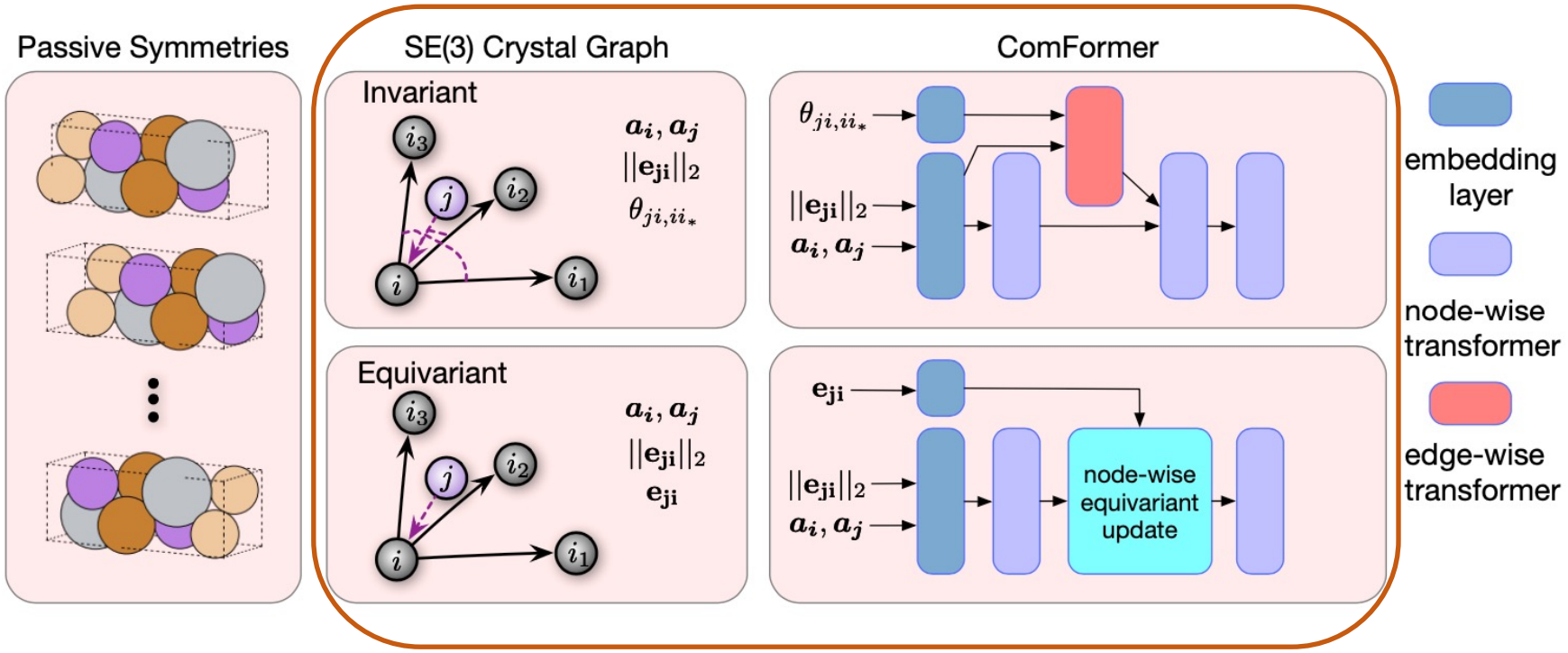
# The proposed ComFormer

- How to extend to global completeness?
  - Lattice frame alignment



# The proposed ComFormer

- Edge features: distances + three lattice angles, or $e_{ji}$
- Completeness: with rigorous mathematical proofs
- Efficiency: linear computational complexity with $O(nk)$



# Experimental Results

- SOTA prediction results for Matbench
  - **# 1 method** for the competitive formation energy task with **16.5** meV/atom MAE
  - # 2 method for jdft2d, with just 636 samples (#1 method is physics descriptor based)
- Even ***faster*** beyond ***in-complete*** methods

Table 4: Efficiency analysis.

| Method | Complexity | Num. Params. | Time/epoch |
|---|---|---|---|
| Matformer | $O(nk)$ | 2.9 M | 64 s |
| ALIGNN | $O(nk^2)$ | 4.0 M | 327 s |
| eComFormer | $O(nk)$ | 12.4 M | 115 s |
| eComFormer-half | $O(nk)$ | 5.6 M | 90 s |
| iComFormer(3) | $O(nk)$ | 4.1 M | 69 s |
| iComFormer(4) | $O(nk)$ | 5.0 M | 78 s |

# Takeaway

- Lattice frames can be used to achieve completeness with efficiency
- Complete geometric representations can boost the prediction accuracy
- ComFormer is complete, efficient with $O(nk)$, and robust across various material properties with ranging training data scales

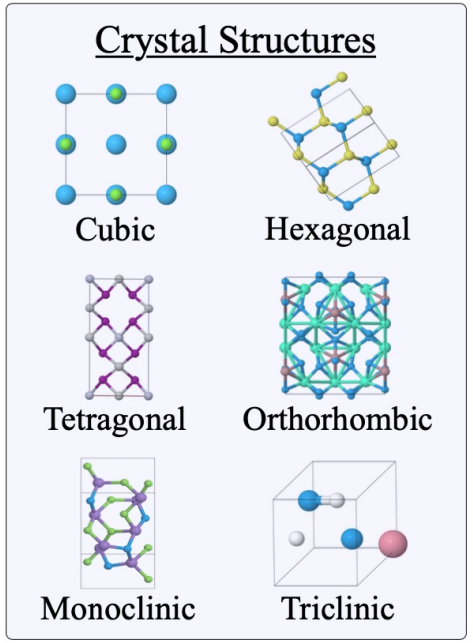
# Talk Overview

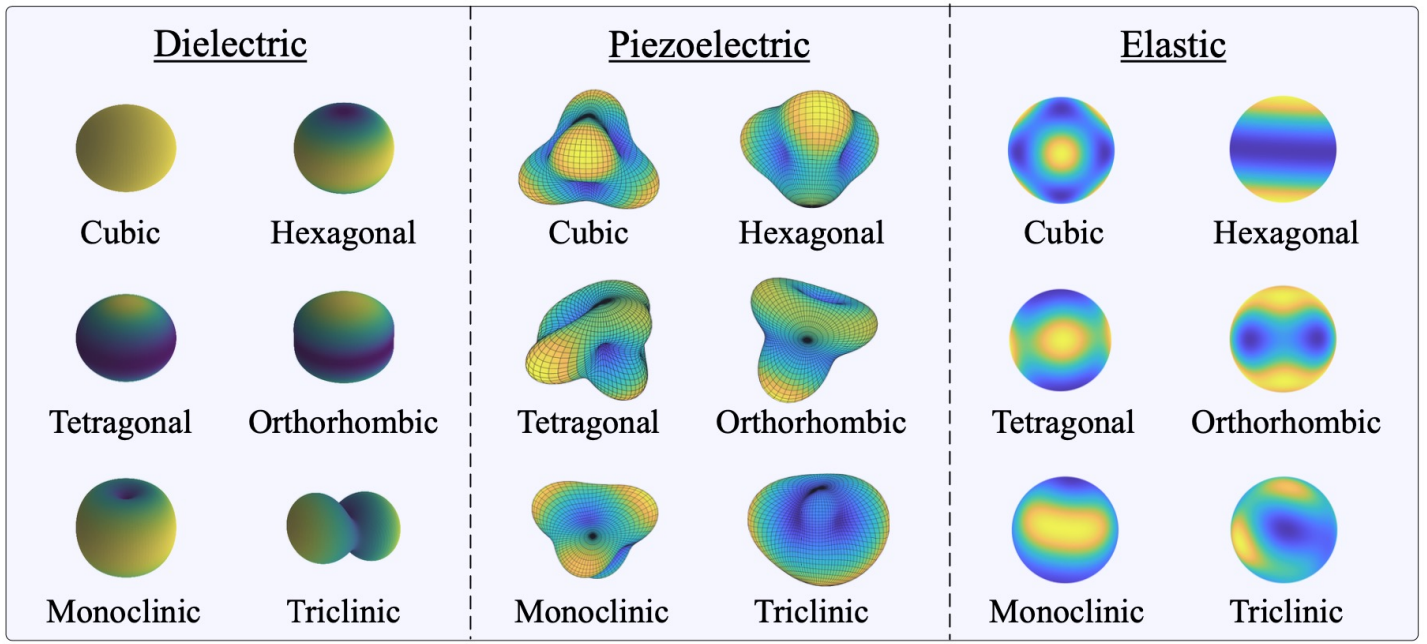
Empowering AI methods to

- distinguish any structural differences - [ICLR 2024]
- predict crystal tensor properties of various order - [ICML 2024]

# Predict General Materials Tensors

- Crystal tensor properties with different tensor orders
  - How to predict high tensor order properties?
  - How to guarantee underlying physics constraints, e.g., $O(3)$ and crystal space groups?




Yan, Keqiang, et al. "A Space Group Symmetry Informed Network for O (3) Equivariant Crystal Tensor Prediction." *Forty-first International Conference on Machine Learning*.

# Challenges

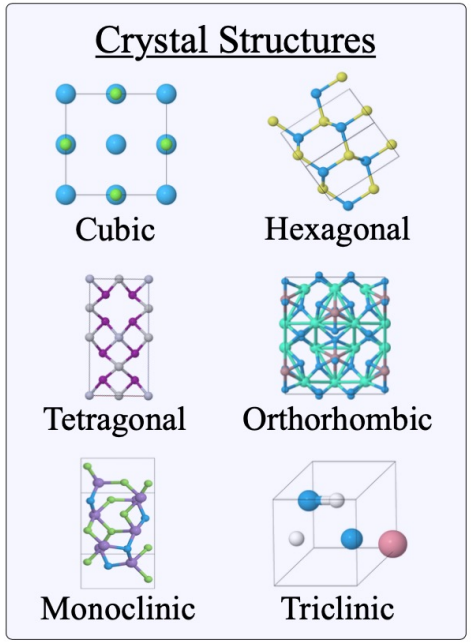
- Crystal tensor properties with tensor order ***n*** are of **matrix form** $\in \mathbb{R}^{3^n}$
  - Order 2 dielectric tensors $\in \mathbb{R}^{3\times3}$, order 3 piezoelectric tensors $\in \mathbb{R}^{3\times3\times3}$
  - *General ML models* developed cannot predict matrix output

- ***O*(3) tensor equivariance**
  - When the crystal is rotated, tensor properties will rotate in a complex way
  - Dielectric $\boldsymbol{\varepsilon} = \mathbf{R}_i \boldsymbol{\varepsilon} \mathbf{R}_i^T$, piezoelectric $\mathbf{e}'_{ijk} = \sum_{\ell mn} \mathbf{R}_{i\ell} \mathbf{R}_{jm} \mathbf{R}_{kn} \mathbf{e}_{\ell mn}$

- **Space group constraints**

# Space Group Constraints

- Space group of crystal structures
  - Space group transformations will map the crystal back to itself
  - e.g., rotate 90 degree, reflect along xy-plane, etc.




- Space group puts direct constraints to tensor matrix values

$$\begin{bmatrix} D_x \\ D_y \\ D_z \end{bmatrix} = \begin{bmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\ \varepsilon_{yx} & \varepsilon_{yy} & \varepsilon_{yz} \\ \varepsilon_{zx} & \varepsilon_{zy} & \varepsilon_{zz} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}, \quad \mathbf{R}_1 = \begin{bmatrix} 0 & -1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{bmatrix}.$$

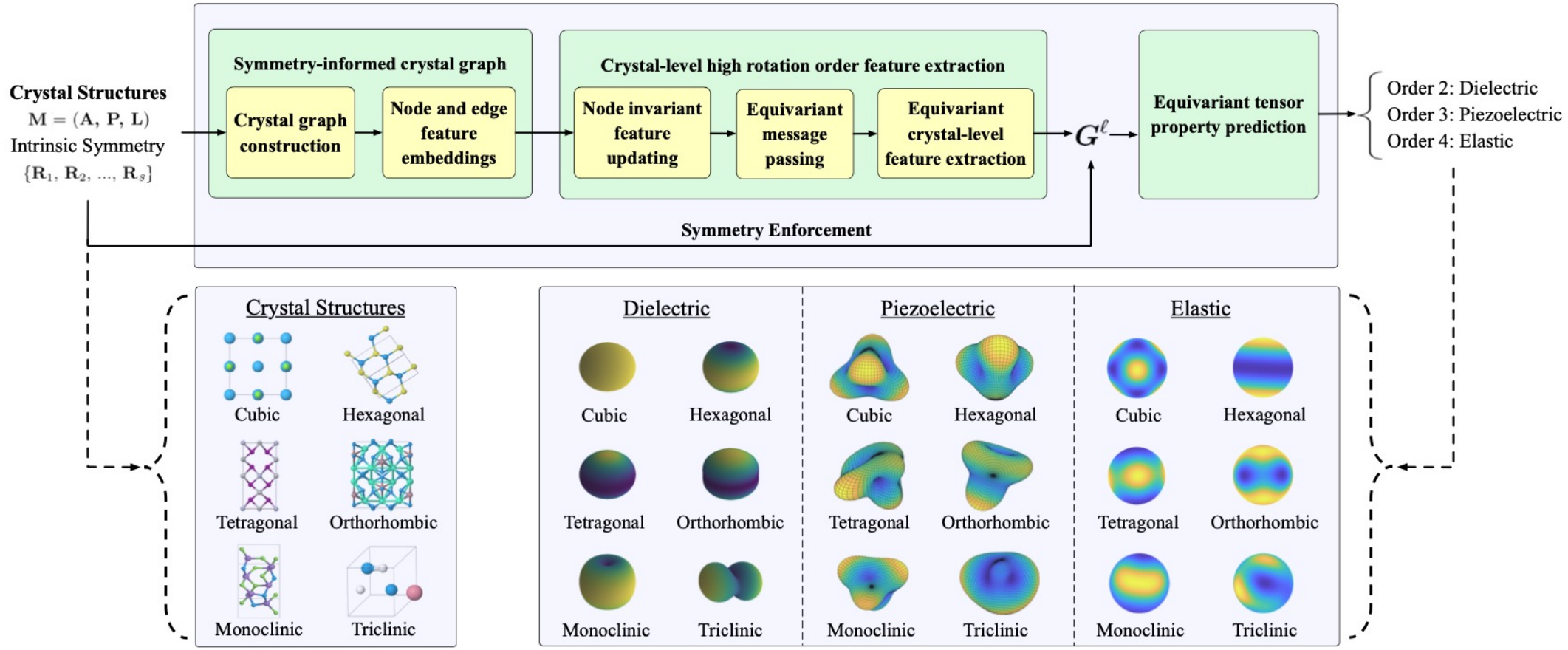
*Due to* $\boldsymbol{\varepsilon} = \mathbf{R}_i \boldsymbol{\varepsilon} \mathbf{R}_i^T$, *we can have*

$$\begin{bmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\ \varepsilon_{yx} & \varepsilon_{yy} & \varepsilon_{yz} \\ \varepsilon_{zx} & \varepsilon_{zy} & \varepsilon_{zz} \end{bmatrix} = \begin{bmatrix} \varepsilon_{yy} & -\varepsilon_{yx} & -\varepsilon_{yz} \\ -\varepsilon_{xy} & \varepsilon_{xx} & \varepsilon_{xz} \\ -\varepsilon_{zy} & \varepsilon_{zx} & \varepsilon_{zz} \end{bmatrix} \quad \boldsymbol{\varepsilon} = \begin{bmatrix} \varepsilon_{xx} & \varepsilon_{xy} & 0 \\ -\varepsilon_{xy} & \varepsilon_{xx} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{bmatrix}.$$

*Resulting in* ***strictly zero and equal entries in dielectric tensors***

# GMTNet

- GMTNet: General Materials Tensor Network
  - A carefully designed network ***satisfying all structural symmetry constraints for various tensors***



# GMTNet

- GMTNet: General Materials Tensor Network
  - ***Three components***
  - 1. Symmetry-informed crystal graphs
  - 2. Equivariant crystal high rotation order feature extraction
  - 3. Equivariant tensor property prediction

  - ***Satisfy crystal symmetries*** *at different levels, including* ***atom and crystal levels***

# Atom and Crystal Level Requirements

- Symmetry-informed crystal graphs
  - Requirements for atom-level features → satisfy crystal symmetries
    - *If two atoms can be mapped to each other by space group operation (R, τ), then they must have **identical invariant features and rotated equivariant features***

- Equivariant crystal high rotation order feature extraction
  - Requirements for crystal-level features
    - *According to Neumann's Principle, crystal features should retain the same spatial symmetry characteristics as the crystal → **invariant to space group ops***

# Atom and Crystal Level Requirements

By satisfying these requirements, we mathematically prove that the extracted crystal feature is invariant to (R, $\tau$) in corresponding space groups.

$$\boldsymbol{G}^{\ell} = \frac{1}{n} \sum_{1 \leq i \leq n} \boldsymbol{f}_{i}^{\ell} = \frac{1}{n} \sum_{1 \leq \theta_{\mathrm{re}}(i) \leq n} \boldsymbol{f}_{\theta_{\mathrm{re}}(i)}^{\ell} = \frac{1}{n} \sum_{1 \leq i' \leq n} \boldsymbol{f}_{i'}^{\ell} = \boldsymbol{G}_{\mathrm{rotated}}^{\ell},$$

$$\boldsymbol{G}_{\mathrm{rotated}}^{\ell} = \frac{1}{n} \sum_{1 \leq i \leq n} \boldsymbol{f}_{i,\mathrm{rotated}}^{\ell} = \frac{1}{n} \sum_{1 \leq i \leq n} \mathrm{WD}^{\ell}(\mathbf{R}) \circ \boldsymbol{f}_{i}^{\ell} = \mathrm{WD}^{\ell}(\mathbf{R}) \circ \boldsymbol{G}^{\ell}.$$

Global crystal feature is invariant to space group transformations (R, $\tau$)

# Equivariant Tensor Property Prediction

**Mimic** the actual **physical response** when external electric field / strain is applied

- General response theory inspired
  - Directly restricting the output to satisfy complex O(3) equivariance is hard
  - Predicting electrical displacement field (a 3D vector) is easy

***Dielectric:*** $\mathbf{D} = \sum (\boldsymbol{G}^{\ell_G} \otimes \mathbf{E}^{\ell_\mathbf{E}=1})^{\ell_\mathbf{D}=1}, \quad \boldsymbol{\varepsilon} = \frac{\partial \mathbf{D}}{\partial \mathbf{E}}.$

***Piezoelectric:*** $\mathbf{D} = \sum_{\ell_G} \sum_{\ell_\epsilon} (\boldsymbol{G}^{\ell_G} \otimes \boldsymbol{\epsilon}^{\ell_\epsilon})^{\ell_\mathbf{D}=1}, \text{ and } \mathbf{e} = \frac{\partial \mathbf{D}}{\partial \boldsymbol{\epsilon}}.$

***Elastic:*** $\boldsymbol{\sigma}^{\ell_\sigma} = \sum_{\ell_G} \sum_{\ell_\epsilon} (\boldsymbol{G}^{\ell_G} \otimes \boldsymbol{\epsilon}^{\ell_\epsilon})^{\ell_\sigma}, \text{ and } C = \frac{\partial \boldsymbol{\sigma}}{\partial \boldsymbol{\epsilon}}.$

# Curated Dataset

A dataset is curated specifically focusing on crystal tensor properties, including dielectric, piezoelectric, and elasticity tensors, sourced from the JARVIS-DFT database

| Dataset | # Samples | Fnorm Mean | Fnorm STD | # Elem. | Unit |
|---|---|---|---|---|---|
| Dielectric | 4713 | 14.7 | 18.2 | 87 | Unitless |
| Piezo | 4998 | 0.43 | 3.09 | 87 | $C/m^2$ |
| Elastic | 14220 | 327 | 249 | 87 | GPa |

# Experimental Results

**GMTNet satisfies all symmetry constraints**

Left / Right: success rate in predicting zero / equal entries in dielectric tensors

| Crystal System | MEGNET | ETGNN | GMTNet |
|---|---|---|---|
| Cubic | 0% | 13.5% | **100%** |
| Tetragonal | 0% | 1.3% | **100%** |
| Hexa-Trigonal | 0% | 2.3% | **100%** |
| Orthorhombic | 0% | 0% | **100%** |
| Monoclinic | 0% | 6.4% | **100%** |

| Crystal System | MEGNET | ETGNN | GMTNet |
|---|---|---|---|
| Cubic | 0% | 100% | **100%** |
| Tetragonal | 0% | 55.3% | **100%** |
| Hexagonal | 0% | 0% | **100%** |
| Trigonal | 0% | 0% | **100%** |

Accuracy comparison for dielectric tensors

| | MEGNET | ETGNN | GMTNet |
|---|---|---|---|
| Fnorm ↓ | 4.16 | 3.92 | **3.50** |
| EwT 25% ↑ | 74.9% | 81.3% | **84.5%** |
| EwT 10% ↑ | 38.9% | 41.6% | **57.1%** |
| EwT 5% ↑ | 19.1% | 23.8% | **27.8%** |

# Experimental Results

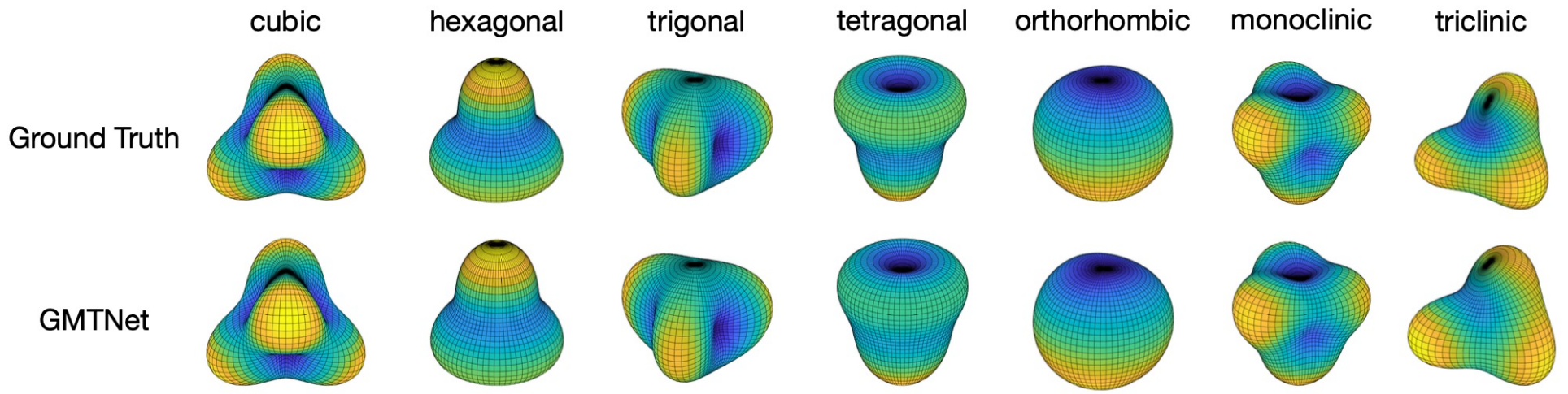
**GMTNet can generalize to higher tensor properties**

Accuracy for piezoelectric and elastic tensors

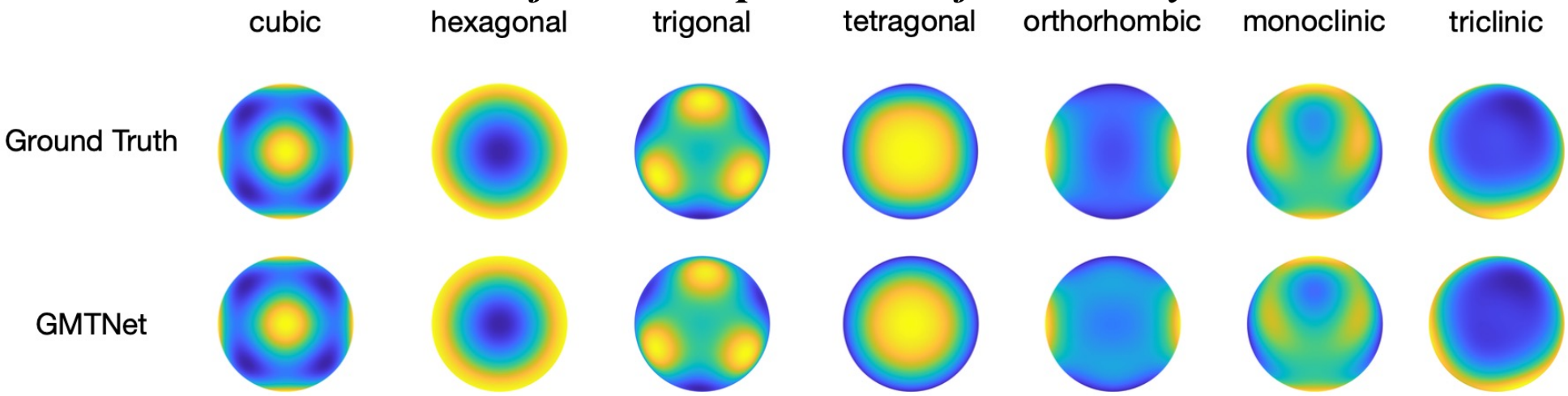
| | Piezo ($C/m^2$) | Elastic (GPa) |
|---|---|---|
| Data Fnorm (mean ± std) | 0.43 ± 3.09 | 326.9 ± 249.3 |
| Fnorm ↓ | **0.37** | **67.38** |
| EwT 25% ↑ | 49.1% | 66.1% |
| EwT 10% ↑ | 46.3% | 21.8% |
| EwT 5% ↑ | 45.7% | 7.7% |
| Symmetry-Zero ↑ | 100% | 100% |
| Symmetry-Equality ↑ | 100% | 100% |

# Visualization Comparison with Ground Truth

***Visualization of GMTNet predictions for piezoelectric tensors***




***Visualization of GMTNet predictions for elasticity tensors***



# Takeaway

- Crystal space group symmetry constraints can be decomposed into atom-level and crystal-level feature requirements

- The proposed GMTNet is a powerful, general, and robust machine learning tool to predict crystal tensor properties of various orders, satisfying all symmetry constraints

# Acknowledgement


- Everyone attending the talk!
- Advisors: Dr. Shuiwang Ji, Dr. Xiaofeng Qian, Dr. Xiaoning Qian
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