Revisiting material structure in the time of AI

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What I learned about materials from AI

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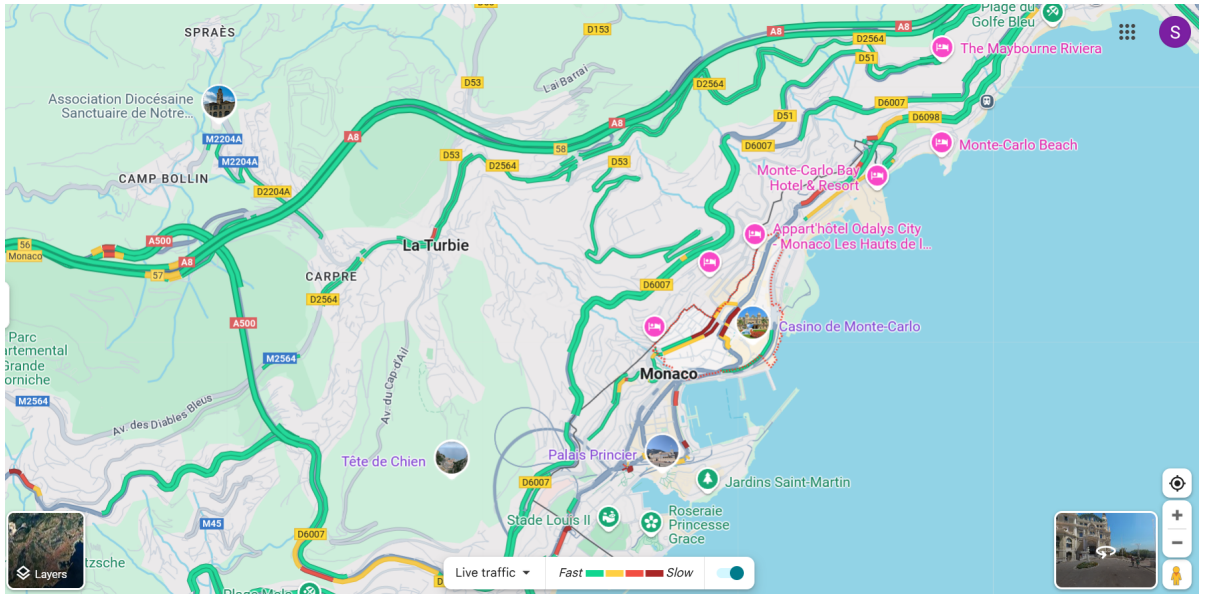
Let me recap a bit of my journey

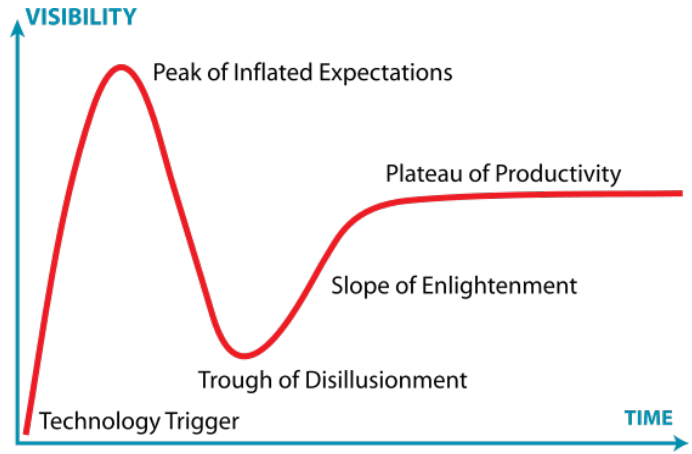


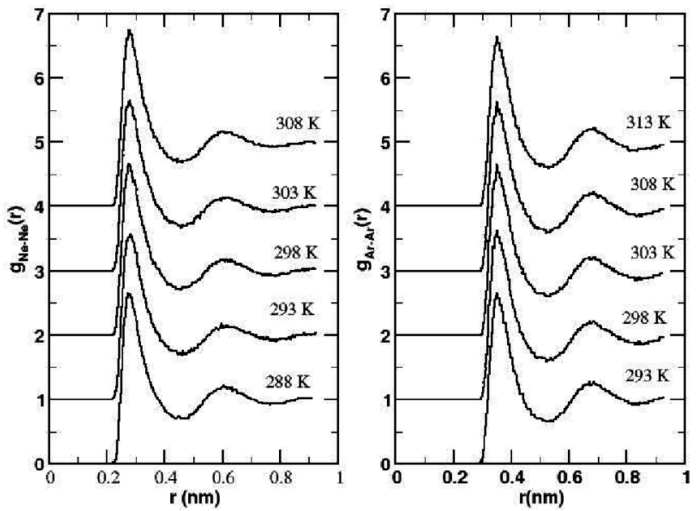
What I learned about materials from AI

It started with a classification problem

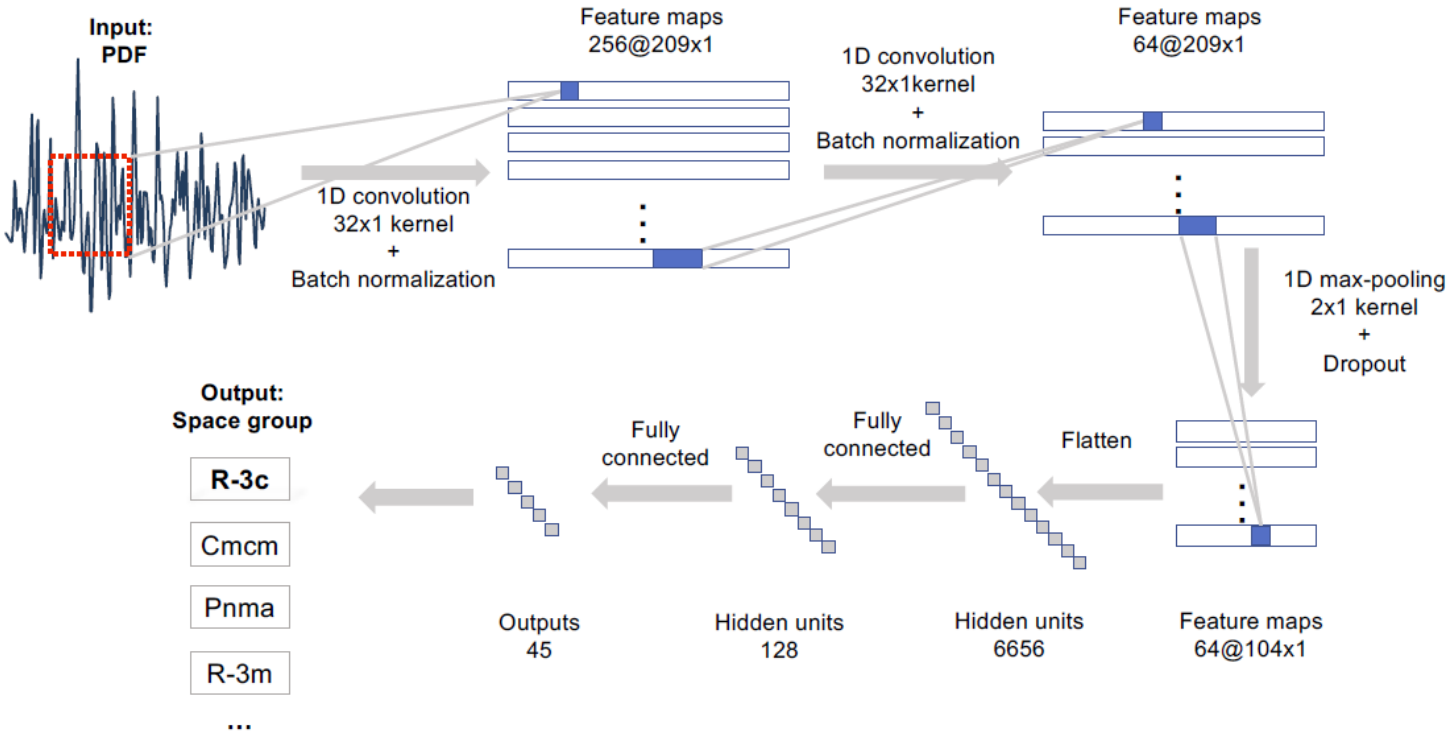
- We can't get space-group from PDF
- But PDF is just the FT of diffraction data
- We can get SG from diffraction data

So the information is there, can a ML model figure out a surrogate forward problem?





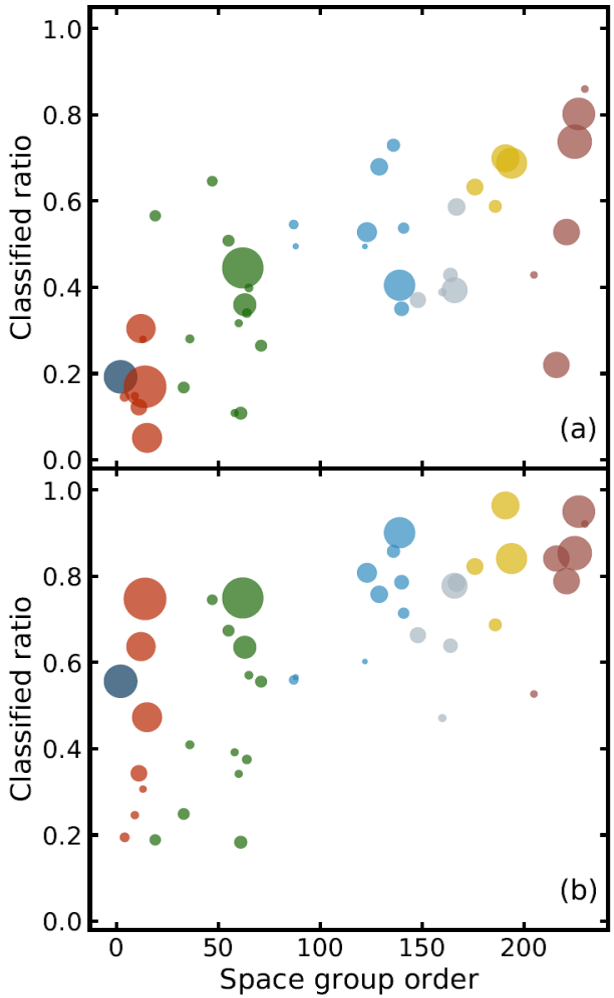
Given a PDF, what was the space-group of the material?



collaboration with group of Qiang Du (Columbia U)

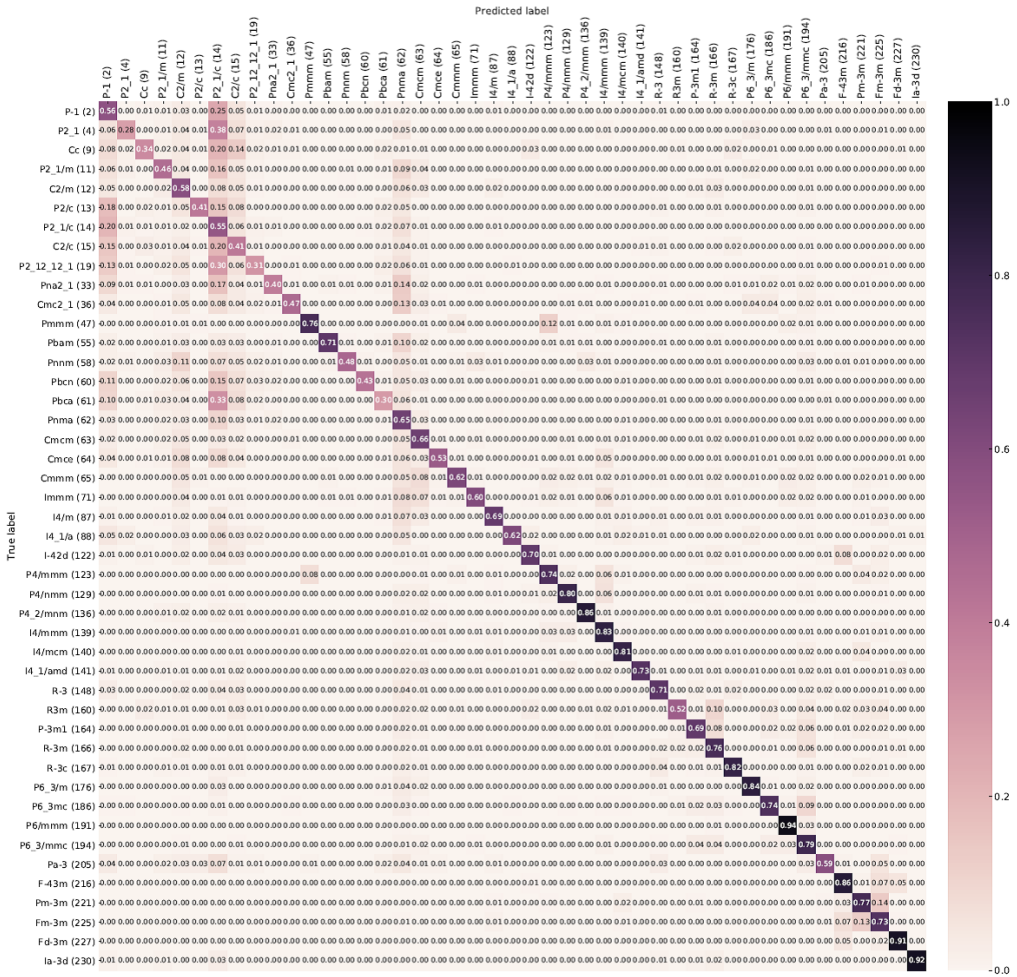
Chia-Hao Liu, SJLB et al., Acta Cryst. (2019), 10.1107/S2053273319005606

How well did it perform?



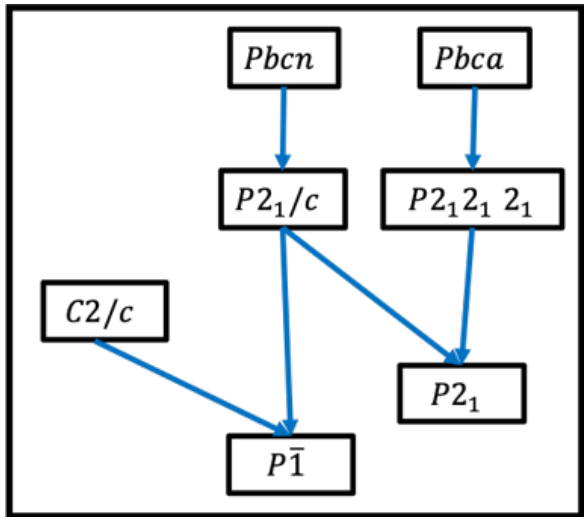
- Top six classification ratio
- (top) Logistic Regression baseline
- (bottom) CNN 91% overall
- Color indicates crystal system
- Size of spot indicates sample-size

We learned crystallography from the model



Matrix of confusion

- tear drops on space-groups C2/1, Pnma
- connected by group-sub-group relationships



Can we do structure solution with Generative AI?

The goal

- Input: the PDF or a powder diffraction pattern
- Output: the structure of the material

That would be cool!

What I learned about AI by applying it to structure solution

What I thought I knew

- Gen-AI is doing interpolation

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- For close-packed structures, I can interpolate from fcc to bcc by introducing stacking-fault defects.

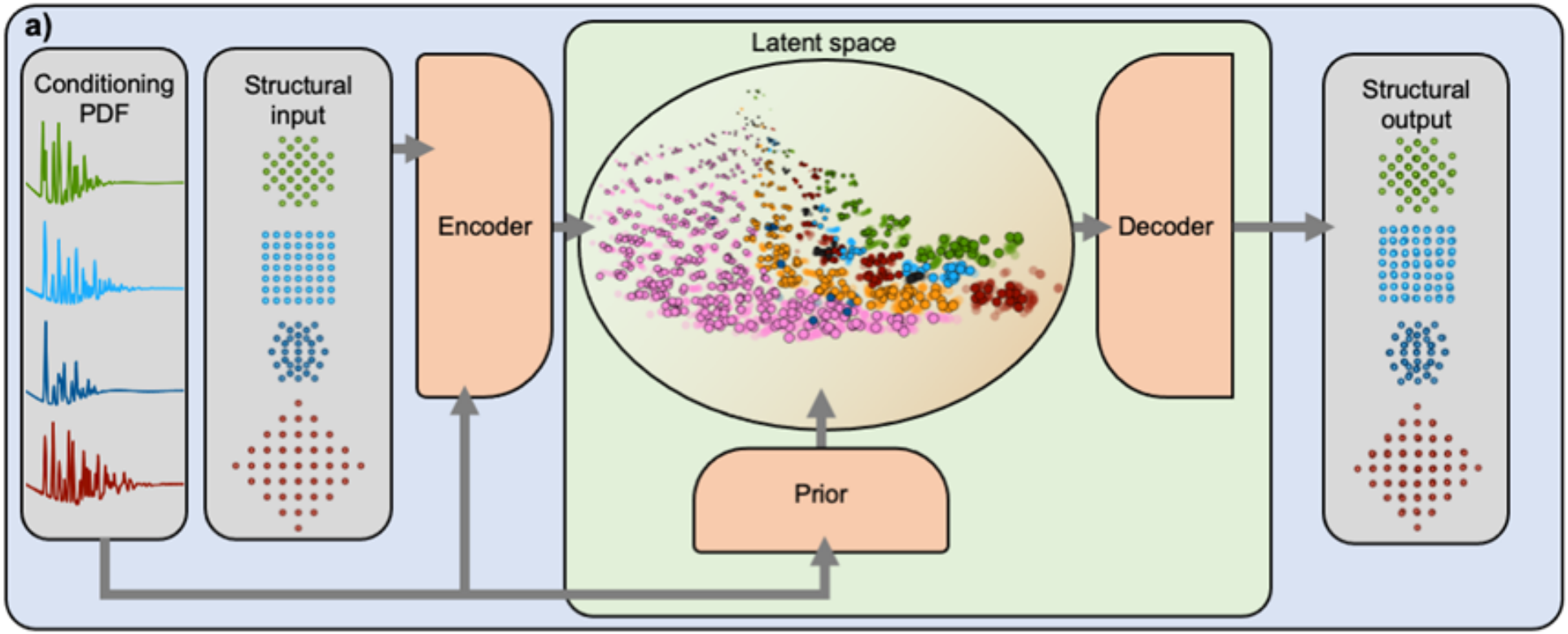
Let's do close-packed structures that interpolate

Case 1: close packed structures

- Graph Convolutional Variational Autoencoder (CVAE)
- Collaboration with Kirsten Jensen, Raghav Selvan, U. Copenhagen
- Work of students Emil Kjaer and Andy Anker
- Kjaer, Anker, et al., Digital Discovery (2023), 10.1039/D2DD00086E

Structure solution using Neural Nets

Deepstruc: close packed nanoparticles with a CVAE



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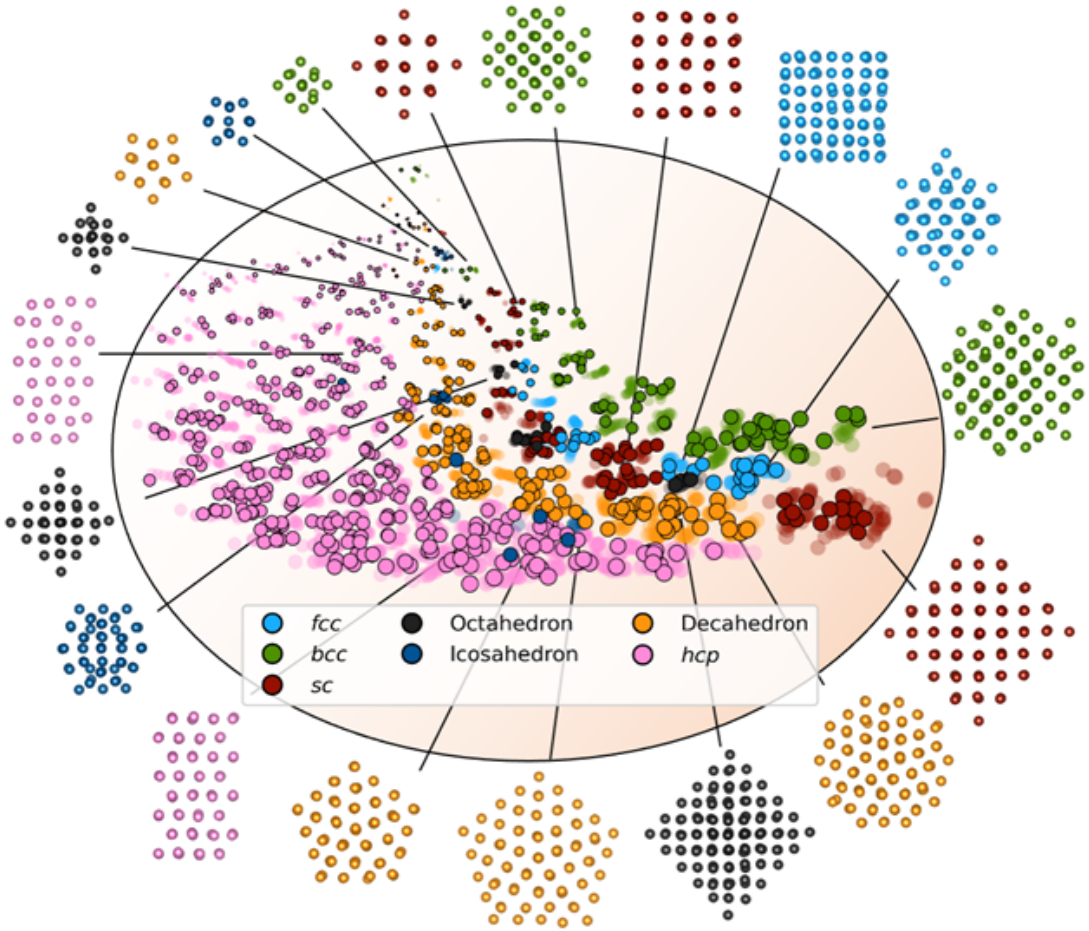

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- Gen-AI is doing interpolation
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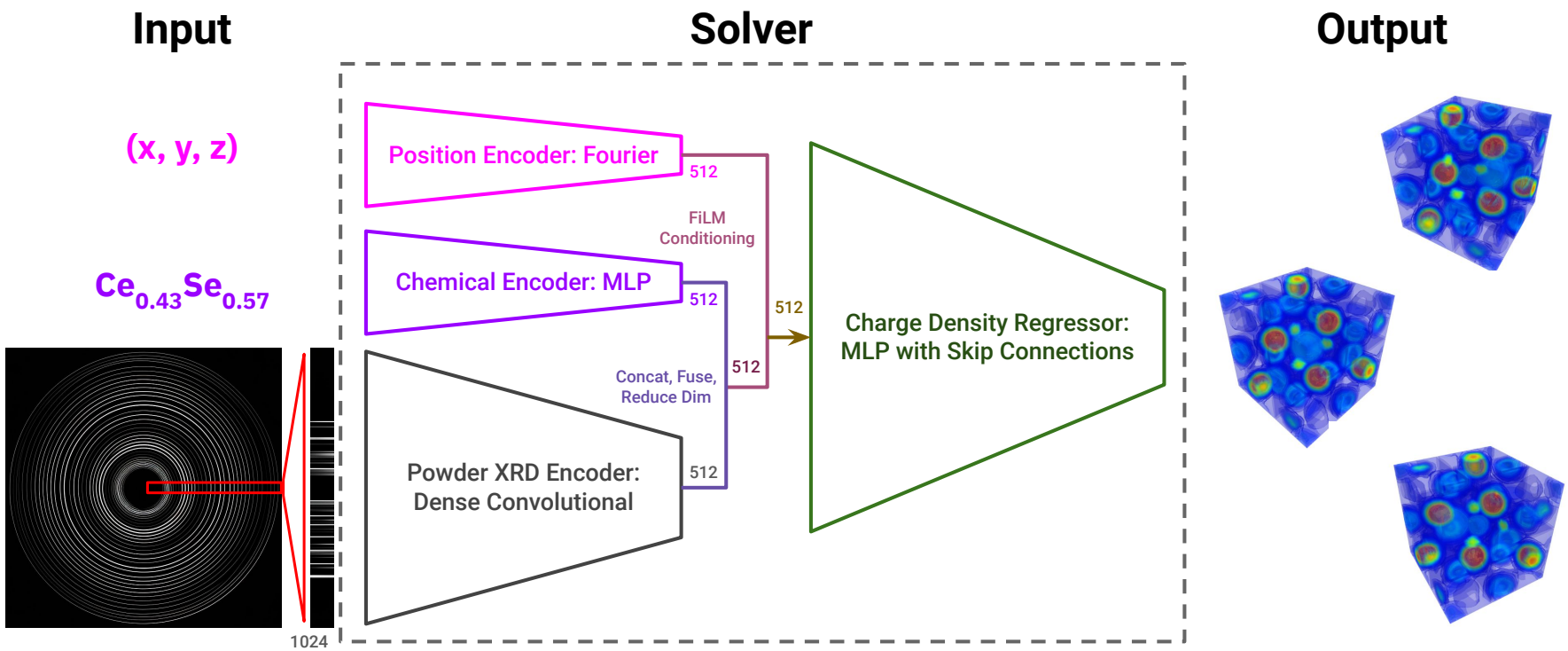
- For close-packed structures, I can interpolate from fcc to bcc by introducing stacking-fault defects.
- It worked!
- But in general? If I draw a line from NaCl to nickel, I don't find $CaCO_4$ in between

Let's change the problem to something continuous....electron-density!

Case 2: electron density determination

- Collaboration with Hod Lipson (Columbia University)
- Work of students Gabriel Guo and Ling Lan
- G. Guo et al., npj-Computational Materials, to be published
- Graph Convolutional Variational Autoencoder (CVAE)
- input: powder diffraction pattern and composition
- output: electron density distribution
- Trained on cubic and trigonal (non-orthogonal but high symmetry) crystal systems

CrystalNet: Electron density reconstruction with CVAE



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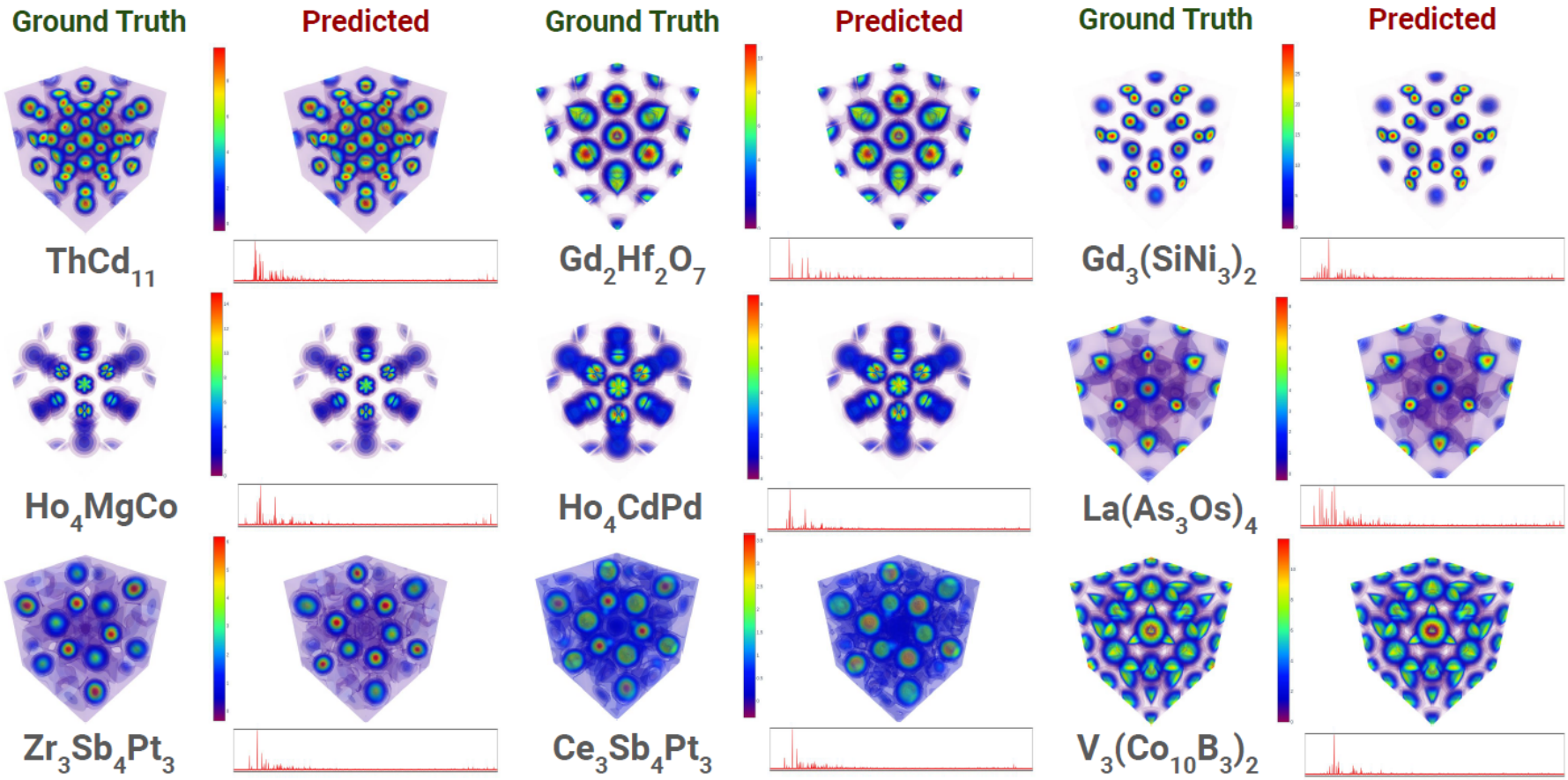

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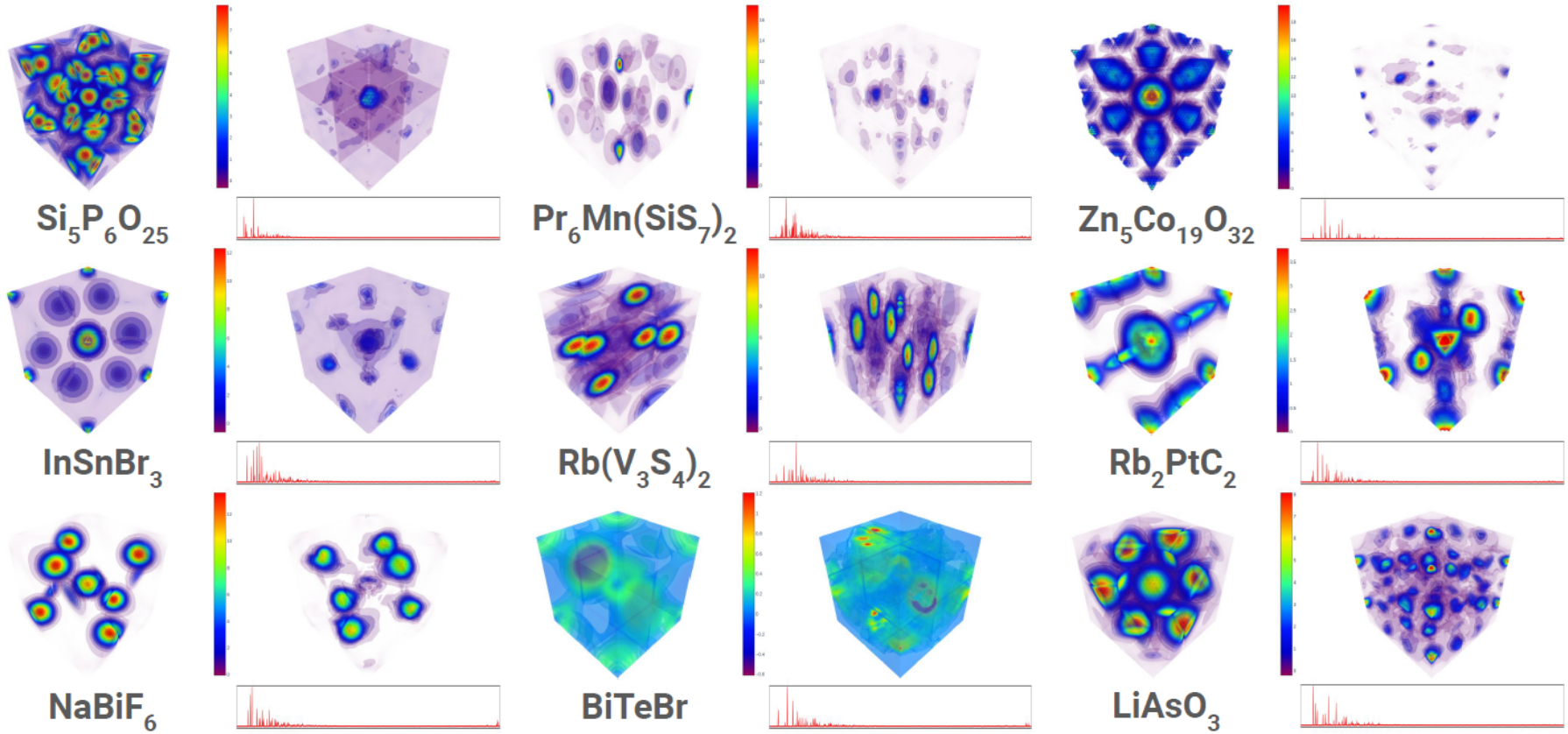

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The magic of diffusion models

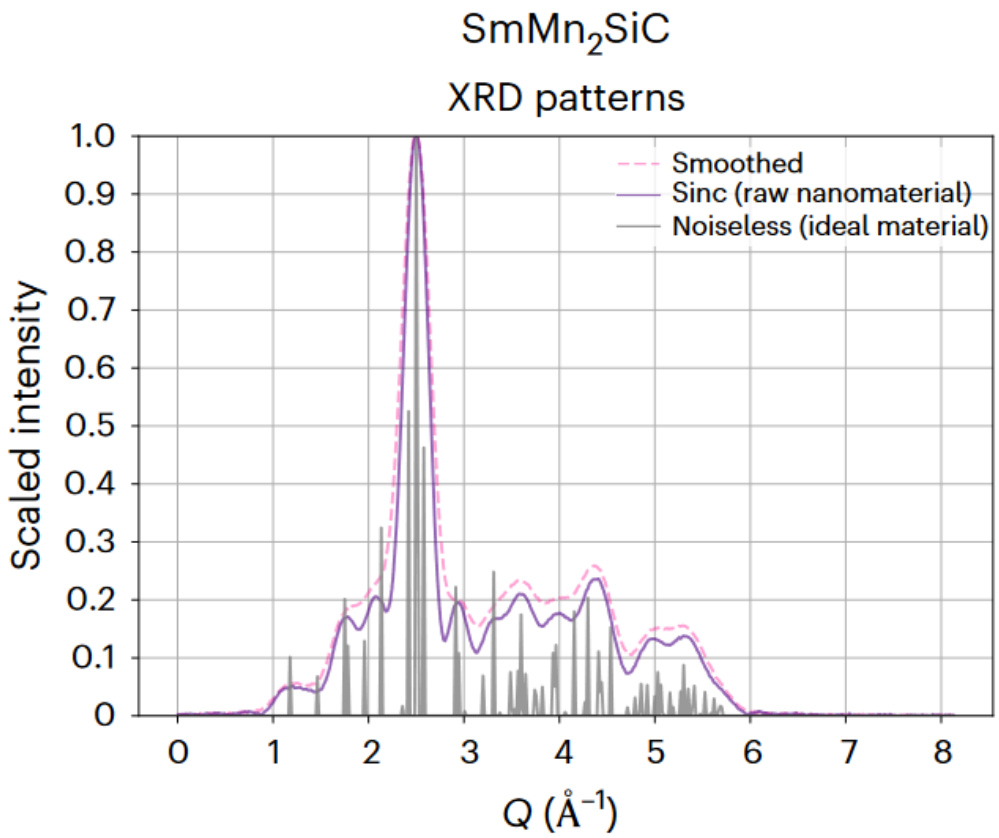
Case 3: structure solution of nanoparticles

- Collaboration with Hod Lipson (Columbia University)
- Work of students Gabriel Guo and Tristan Saidi
- G. Guo et al., Nature Materials, (2025) https://doi.org/10.1038/s41563-025-02220-y
- input: nanostructure powder diffraction pattern and composition
- output: "structure": unit cell, coordinates and coloring

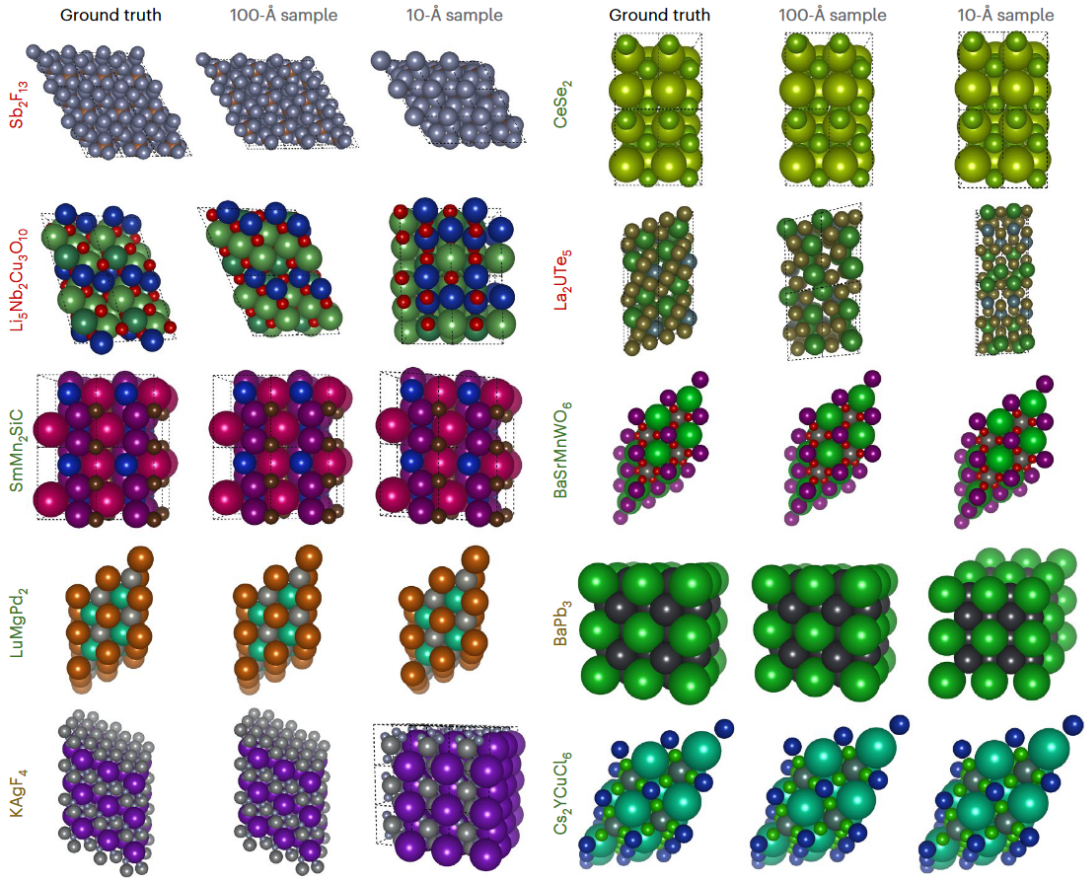
PXRDNet: structure solution with diffusion model

PXRDNet

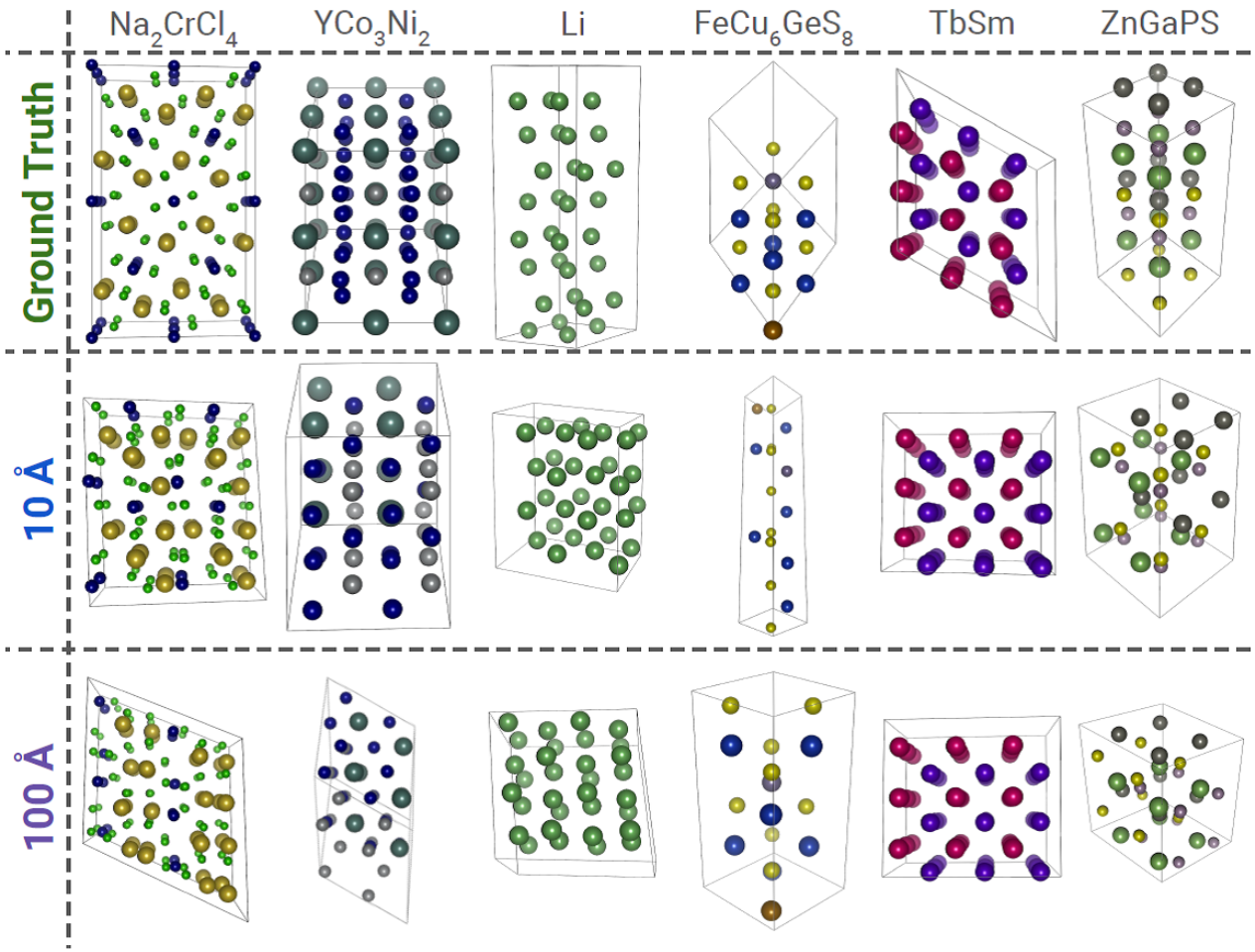
- 100 Å or 10 Å NP powder data. Low information content!
- Based on CDVAE (T. Xie, Proc. International Conference on Learning Representations (2022).
- SE(3) equivariant graph NN autoencoder for composition, lattice parameters and number of atoms.
- Denoising diffusion via noise-conditioned score networks for the atomic coordinates



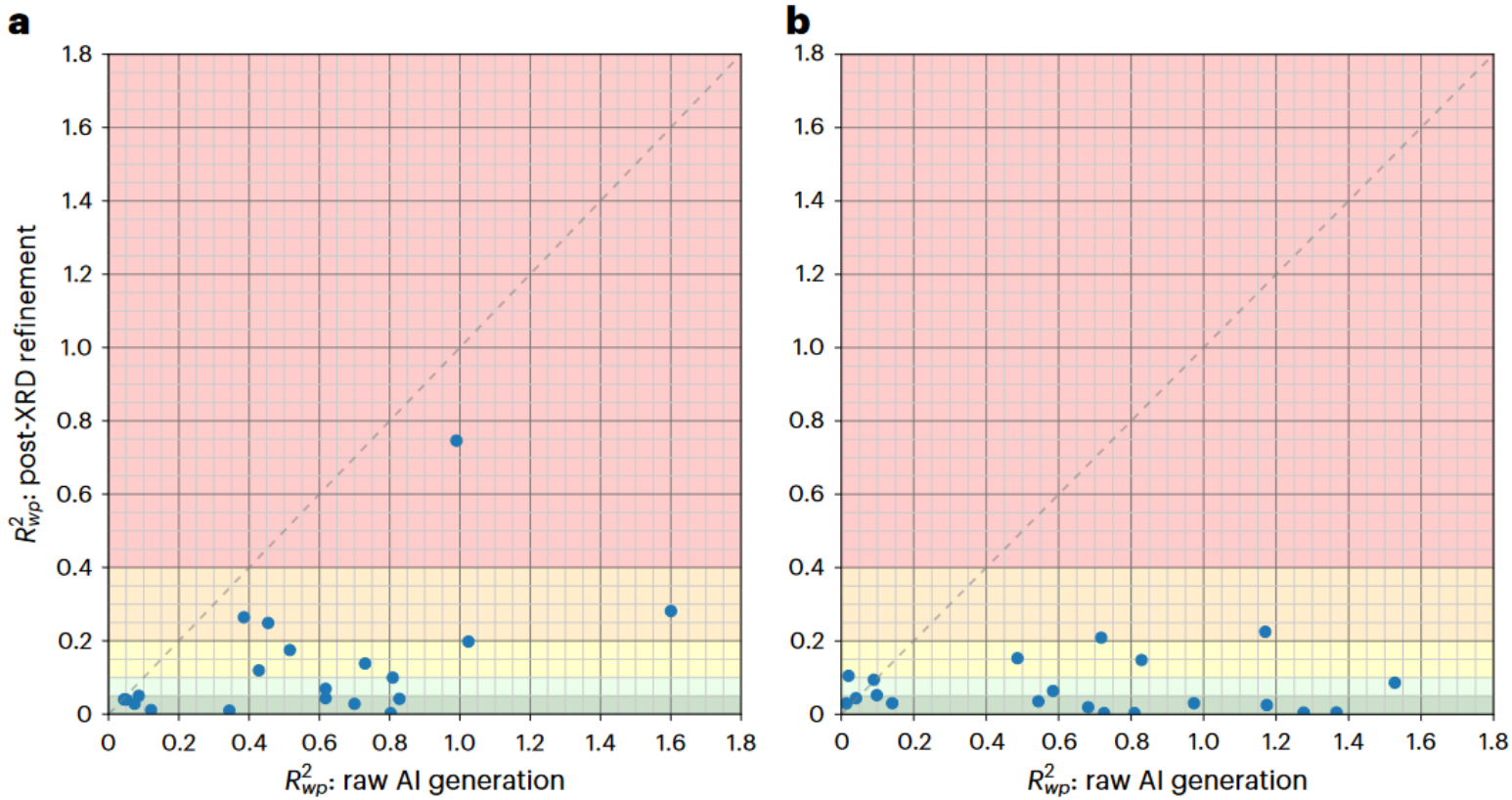
PXRDNet: structure solution with diffusion model



PXRDNet: structure solution with diffusion model



PXRDNet: structure solution with diffusion model



Diffusion models are the s$%t

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- They build latent spaces/embeddings that can interpolate non-interpolatable things
- They do it by starting with a large population of "good" things
- They then randomize them in a well defined way
- Any point in this space can be denoised
- A point in the space can be picked by a prompt, which could be a desired target

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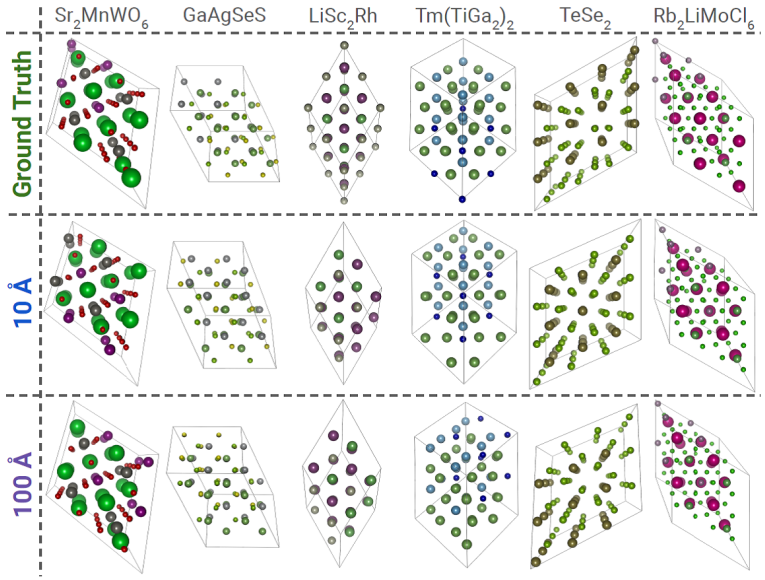
They work!

Example: PXRDnet

It works!

- After training the latent space and conditioning on (rather small amounts of) diffraction data, it makes a rather good guess of the structure solution
- Enforces SO(3) equivariance so only interested in rigid bodies

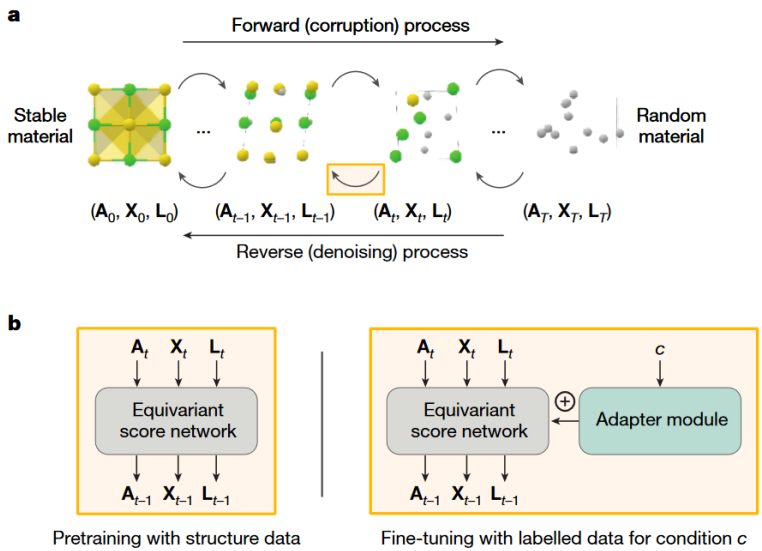


Example: matterGen

Let's throw some serious Microsoft money at the same problem

- Trained on a dataset that is $> 10\times$ larger than PXRDnet using serious Microsoft compute
- In most respects, in common with PXRDnet
- Can be conditioned on many different labels. Has to be trained, but the foundation model means that it can be trained for the new task on a small amount of data
- At the time of writing of the MS could only be conditioned on scalars. I'm sure that has changed.

It works!



What I learned about AI - Summary

- Diffusion models are the sh$%t
- Emerging foundation models built on large, diverse, datasets are adaptable for many task with modest retraining

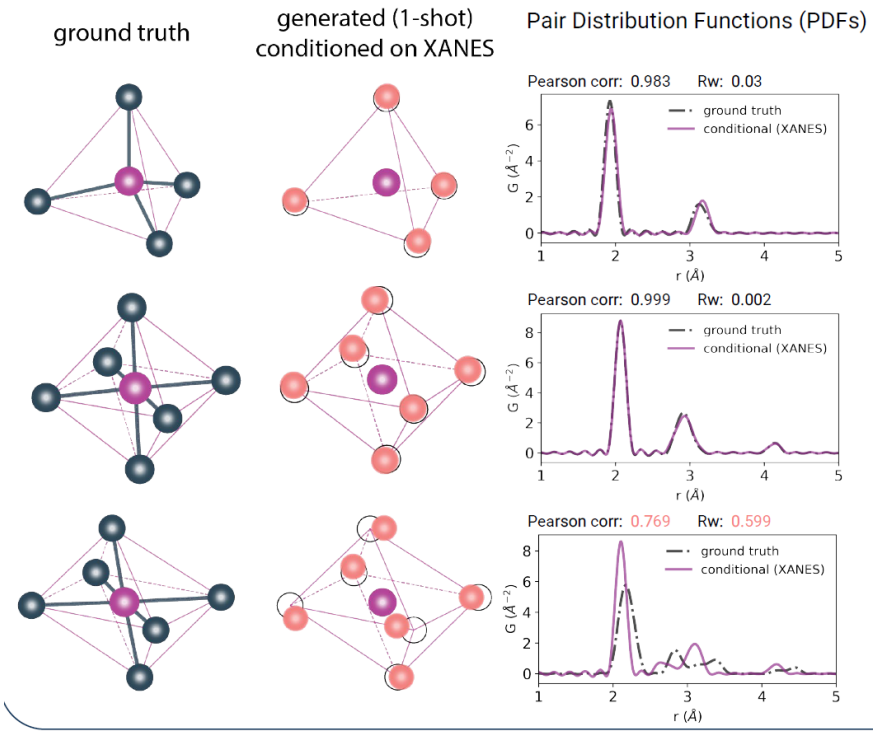
They work

Local structure from XANES with a diffusion model

Solving local structure from XANES (not XAFS!)

- Work of Aniv Ray and Tina Na Narong
- Collaboration with Steven Torrisi and Hod Lipson
- Use a diffusion model to extract local structure (with some surprises)

Please see Tina's poster



What I learned about materials by applying AI to structure solution

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Structure solution

Structure of the problem

- Input is data, a diffraction pattern or PDF
- Output is a structure
- To train the model we need to tell it when it got the structure right vs wrong
- We know the structure, but we need a categorical success/failure measure
- How do we do that?

Structure solution

How do we assess if a structure solution is right?

- Need a measure of similarity between structures
- Need a threshold when structures are sufficiently similar to determine them as being "the same" and so "the structure is solved"

Structure solution

How do we assess if a structure solution is right?

- Need a measure of similarity between structures
- Need a threshold when structures are sufficiently similar to determine them as being "the same" and so "the structure is solved"
- btw, this problem affects Materials Discovery. MatterGen talks about SUN materials, "stable", "unique", "new" (i.e., not the same as a known structure).

When are structures the same problem (WASTS problem)

But wait, there are many papers in the literature that do this without much discussion. How do they do it?

- They use a structure similarity metric in pymatgen "structureMatcher"
- But this appears not to have been heavily validated. We are taking on blind faith that is working as hoped
- Some chemists seem to disagree

When are structures the same problem (WASTS problem)

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The structure definition problem

What is a structure?

The structure definition problem

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Crystal structure

Actions

From Online Dictionary of Crystallography

Structure cristalline (*Fr*). Kristallstruktur (*Ge*). Struttura cristallina (*It*). 結晶構造 (*Ja*). Estructura cristalina (*Sp*).

A **crystal structure** is a crystal pattern consisting of atoms.

See also

- Point space
- Noncrystallographic symmetry
- *International Tables of Crystallography, Volumes A and A1*

Category: Fundamental crystallography

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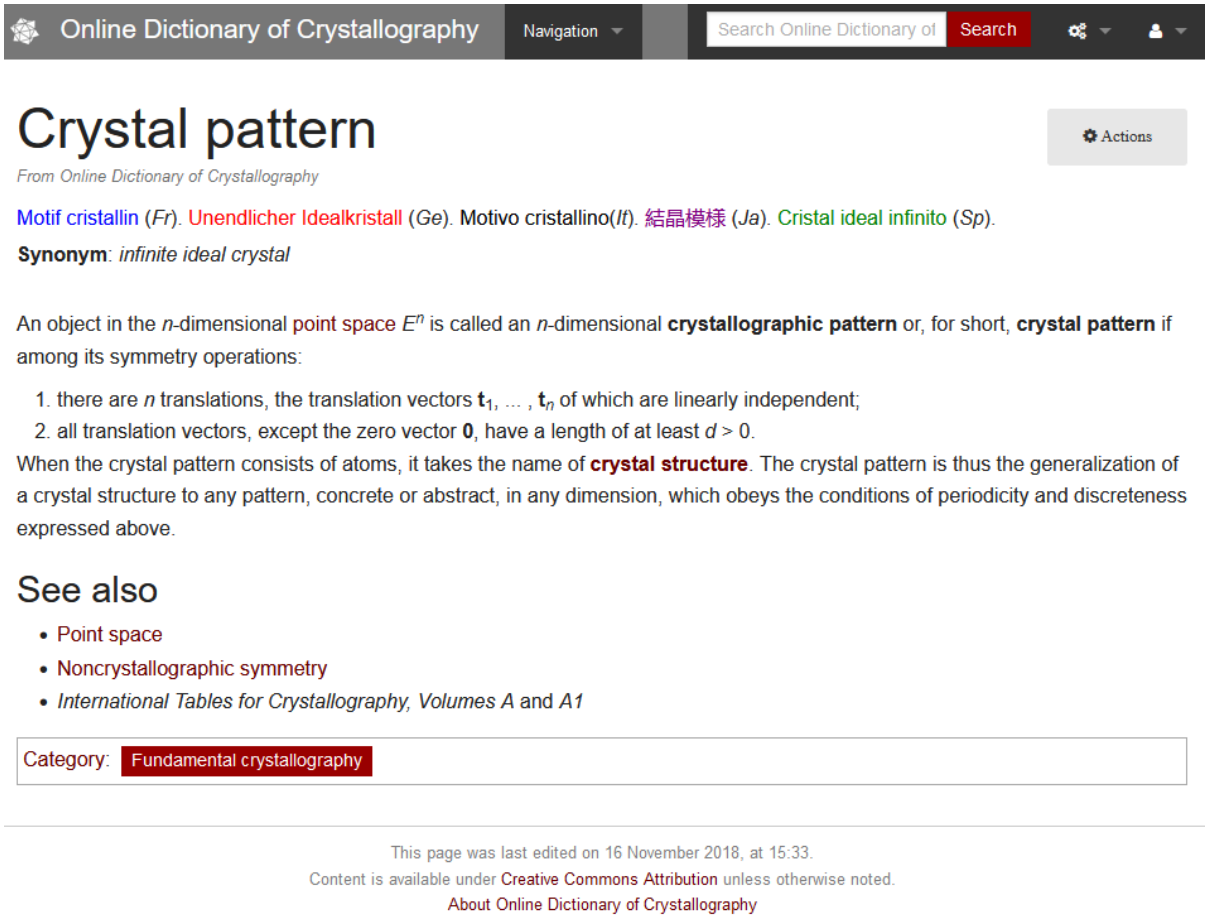
The structure definition problem



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Crystal pattern

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From Online Dictionary of Crystallography

Motif cristallin (*Fr*). Unendlicher Idealkristall (*Ge*). Motivo cristallino(*It*). 結晶模様 (*Ja*). Cristal ideal infinito (*Sp*).

Synonym: *infinite ideal crystal*

An object in the *n*-dimensional point space E^n is called an *n*-dimensional **crystallographic pattern** or, for short, **crystal pattern** if among its symmetry operations:

1. there are *n* translations, the translation vectors $\mathbf{t}_1, \ldots, \mathbf{t}_n$ of which are linearly independent;
2. all translation vectors, except the zero vector **0**, have a length of at least $d > 0$.

When the crystal pattern consists of atoms, it takes the name of **crystal structure**. The crystal pattern is thus the generalization of a crystal structure to any pattern, concrete or abstract, in any dimension, which obeys the conditions of periodicity and discreteness expressed above.

See also

- Point space
- Noncrystallographic symmetry
- *International Tables for Crystallography, Volumes A* and *A1*

Category: Fundamental crystallography

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About Online Dictionary of Crystallography

The structure definition problem

What is a structure, chemist's definition?

- Definition based on space-group classification
- Structures that change without breaking symmetry are the same structure

The structure definition problem

What is a structure, chemist's definition?

- Definition based on space-group classification
- Structures that change without breaking symmetry are the same structure
- In this world it is easy to say that two structures are the same (continuous crystallography)
- much harder to say whether two unequal structures are the same in the chemist's sense

The structure definition problem

Case 1: thermal expansion

- Take material
- Warm it up
- The unit cell expands (without breaking (point) symmetry)
- The structure hasn't changed (according to Chemists)
- The structure has changed (according to continuous crystallography measures like PDD)
- Actually, it has changed, all the bonds are different lengths which affects properties

The structure definition problem

Case 2: miniscule phase transition

- Take material
- Move one atom off a special position by a tiny amount
- The structure has changed (according to Chemists), the space group has changed
- The structure has changed (according to continuous crystallography measures like TPP), but by a miniscule amount

The structure definition problem

Case 2: miniscule phase transition

- Take material
- Move one atom off a special position by a tiny amount
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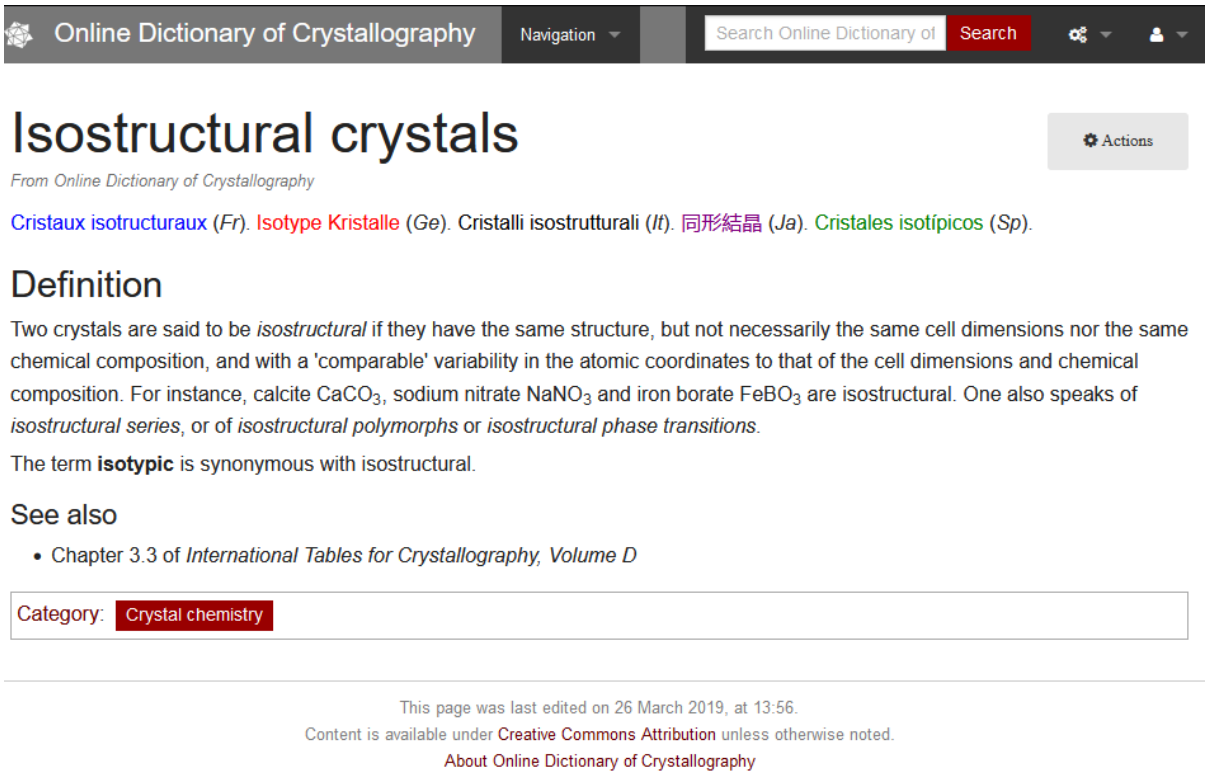
Case 2 the continuous crystallography thinks the materials are closer than Case 1, but chemists think otherwise

The structure definition problem



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Isostructural crystals

Actions

From Online Dictionary of Crystallography

Cristaux isotructuraux (*Fr*). Isotype Kristalle (*Ge*). Cristalli isostrutturali (*It*). 同形結晶 (*Ja*). Cristales isotípicos (*Sp*).

Definition

Two crystals are said to be *isostructural* if they have the same structure, but not necessarily the same cell dimensions nor the same chemical composition, and with a 'comparable' variability in the atomic coordinates to that of the cell dimensions and chemical composition. For instance, calcite $CaCO_3$, sodium nitrate $NaNO_3$ and iron borate $FeBO_3$ are isostructural. One also speaks of *isostructural series*, or of *isostructural polymorphs* or *isostructural phase transitions*.

The term **isotypic** is synonymous with isostructural.

See also

- Chapter 3.3 of *International Tables for Crystallography, Volume D*

Category: Crystal chemistry

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About Online Dictionary of Crystallography

The structure definition problem

Summary

As a community we need more practical definitions of what it means for two structures to be the same or similar

Ways to compare structures: structure metrics

How do crystallographers compare structures?

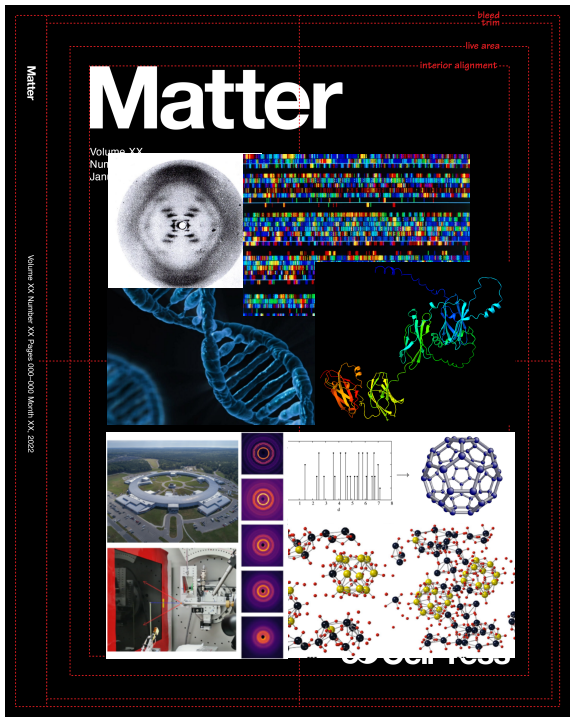
- Gold standard is Rw between measured and computed structure-factors or diffraction data coupled with crystallographic expertise
- CIFcheck (does some symmetry and chemistry common sense)

Ways to compare structures: continuous structure metrics

continuous structure representations

- PDF as a "materials genome", a 1D function that "codes for" 3D structure
- AMD (reduce the PDF to a scalar) - metric of Vitaliy Kurlin (U Liverpool)
- PDD (matrix of partial PDFs)
- smooth overlap of atomic positions (SOAP)
- atom-centered symmetry functions (ACSF)

AMD and PDD are not experimentally accessible but are valid metrics, and PDD is complete



Comparing StructureMatcher and PDF

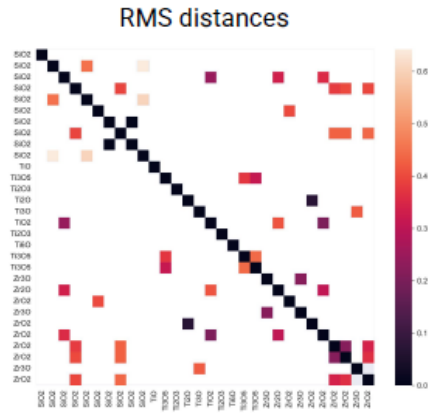
Baseline: pymatgen.StructureMatcher module

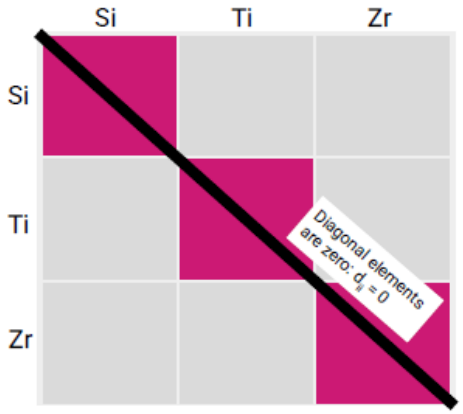
- Pymatgen: open-source Python library for materials analysis
- StructureMatcher module compares structures based off their atomic coordinates.

$$distance(\mathbf{x}, \mathbf{y}) = \sqrt{\sum_{i=1}^{n}(x_i - y_i)^2}$$

Distance measure:

- Root-mean-square distance between the atomic positions of two structures
- Ignores atomic species, only accounts for atomic sites and lattice parameters
- Permutes atomic sites to minimize RMS, account for rotations & symmetries





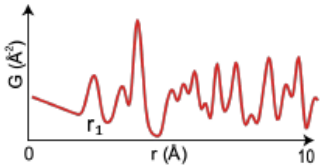
Pair Distribution Function (PDF):
"histogram" of bond lengths in the structure, experimentally measurable from diffraction data.

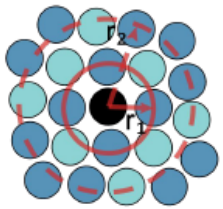
Distance measures:

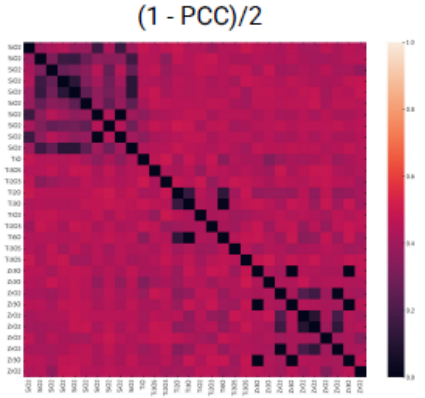
- Pearson correlation coefficient (PCC)
- Symmetrized R_w

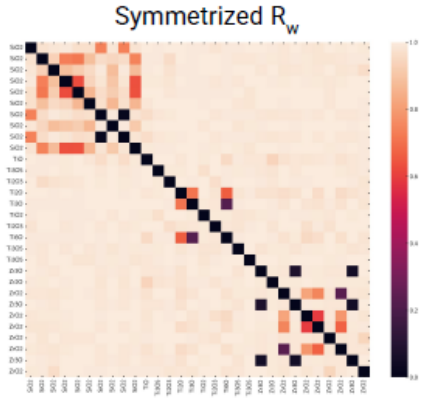
$$R_w = \frac{\sum_i (x_i - y_i)^2}{\sum_i (\vec{x}_i \vec{y}_i)}$$

**Pairs of PDFs are "morphed" before computing PCC and R_w. This accounts for differences in scale between structures, which affects R_w, PCC greatly.*









Work of Tina Na Narong, Zoe Zachko and Andrew Yang, Collaboration with Steven Torrisi

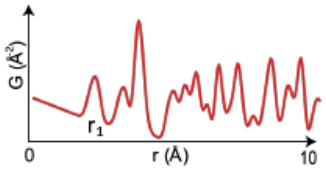
Comparing PDF and PDD/AMD

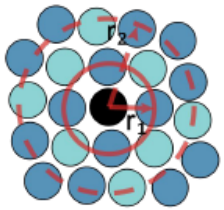
Pair Distribution Function (PDF):
"histogram" of bond lengths in the structure, experimentally measurable from diffraction data.

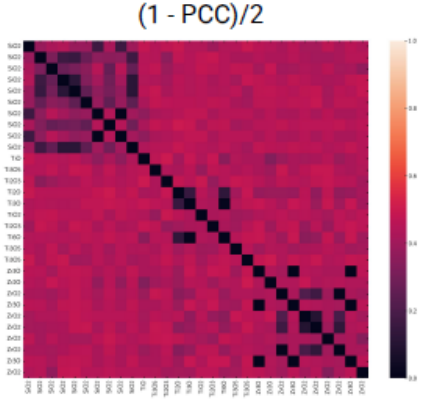
Distance measures:
- Pearson correlation coefficient (PCC)
- Symmetrized R_w

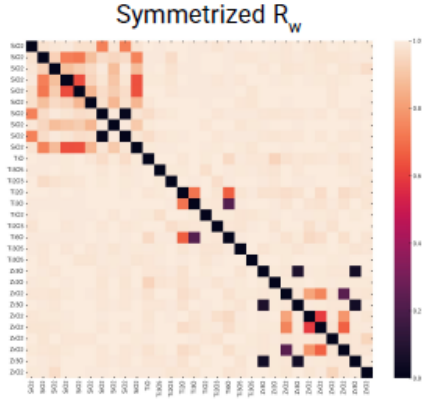
$$R = \frac{\sum_i (x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum_i (x_i - \bar{x})^2 \sum_i (y_i - \bar{y})^2}}$$

**Pairs of PDFs are "morphed" before computing PCC and R_w. This accounts for differences in scale between structures, which affects R_w PCC greatly.*



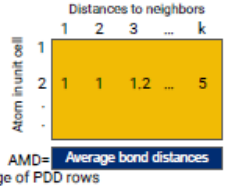






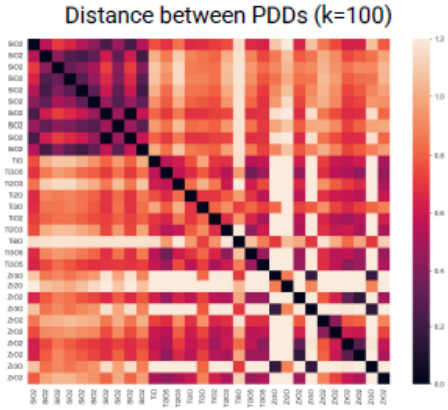
Pointwise Distance Distribution (PDD)
- Matrix of interatomic distances in the structure (isometry invariant)
- Each row of a PDD matrix lists *k* shortest distances (ascending order) from an atom in the unit cell to other atoms

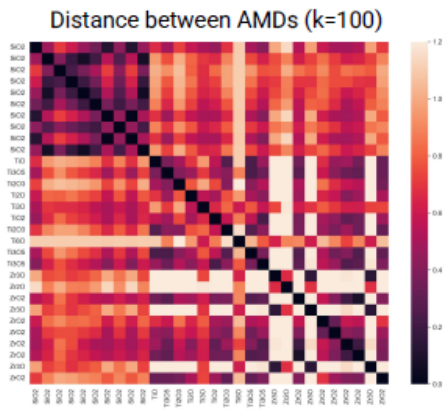
Distance measure: Earth's Mover Distance (EMD)
→ treat PDD matrix elements as "piles" of Earth
→ solve a linear programming problem to quantify the minimum "mass transport work" needed to make the two PDDs identical



Average Minimum Distance (AMD)
- AMD is an average of PDD: vector of *k* average shortest distances

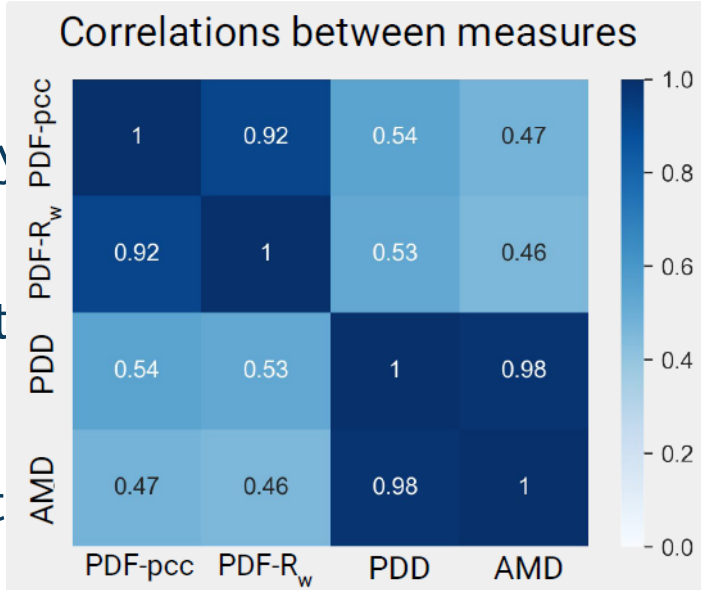
Distance measure: Chebyshev distance $D(x,y) = \max_i |x_i - y_i|$





Comparing structure metrics

- different similarity metrics differ in which structures they think are similar
- Which is the rightest
- We don't know
- How similar of result do they provide?



Correlations with StructureMatcher
(only distances below threshold included)

PDF-pcc	0.72
PDF-R$_w$	0.83
PDD	0.63
AMD	0.55

Summary

- Diffusion models are the s$%t
- They can be used to make really great foundation models
- As with LLMs, we will likely all be using these in the future
- But:

Summary

- Diffusion models are the s$%t
- They can be used to make really great foundation models
- As with LLMs, we will likely all be using these in the future
- But: be careful what you wish for...the answers are only as good as the question that was asked
- Bias in the data, bias in the network/noising protocol, choice of success metric
- These things are still being figured out

With GenAI, the problem is the problem, not the solution

Acknowledgements

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- Beamline and software teams
- Collaborators

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- The Facilities!