Leveraging AI and robotics to accelerate inorganic materials synthesis

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Chemical Engineering & Materials Science
University of Minnesota

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AI and robotics are here…

Chatbots galore

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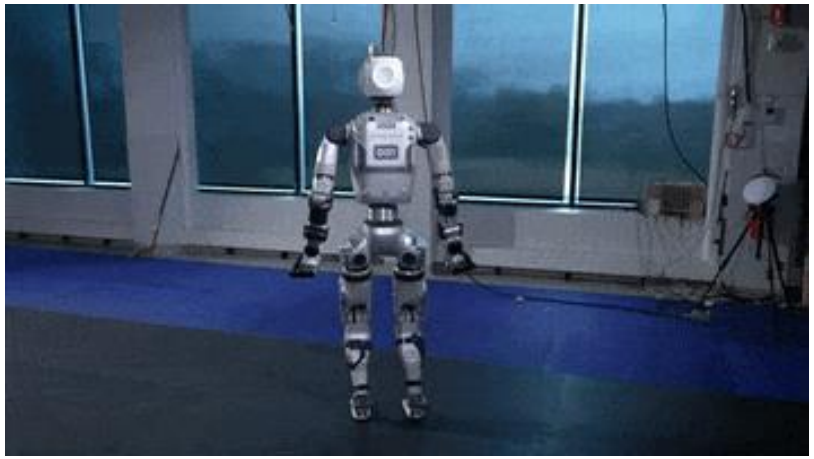
Chatbots galore



And cute (or creepy) robots

AI and robotics are here…

Chatbots galore

But can these tools be useful in science?

And cute (or creepy) robots

Early examples are promising!

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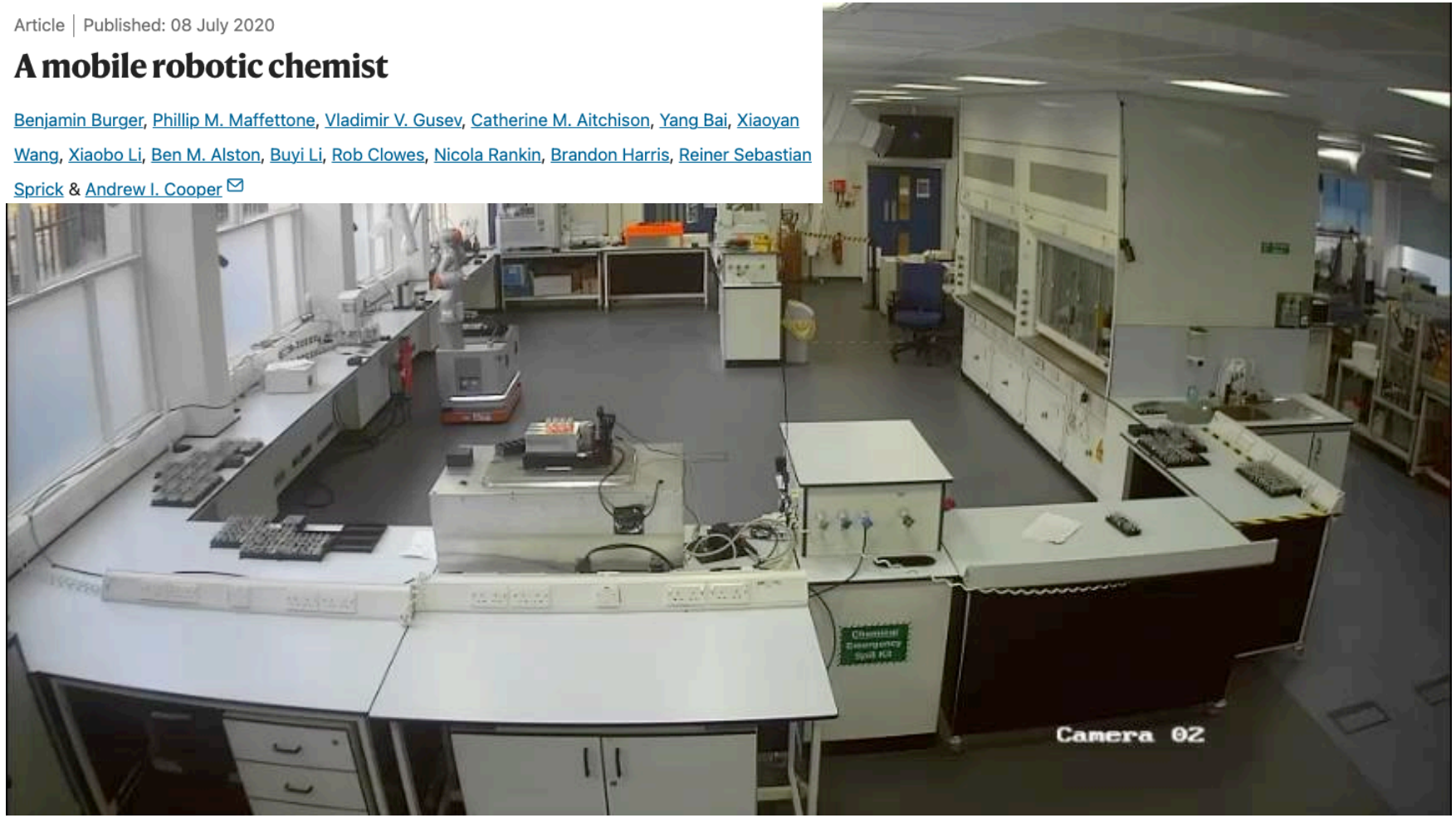
Chemical Engineering & Materials Science

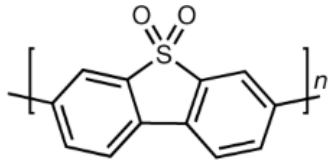
Article | Published: 08 July 2020

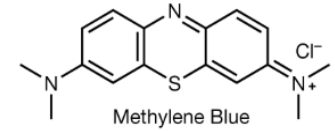
A mobile robotic chemist

Benjamin Burger, Phillip M. Maffettone, Vladimir V. Gusev, Catherine M. Aitchison, Yang Bai, Xiaoyan Wang, Xiaobo Li, Ben M. Alston, Buyi Li, Rob Clowes, Nicola Rankin, Brandon Harris, Reiner Sebastian Sprick & Andrew I. Cooper



Photocatalysts





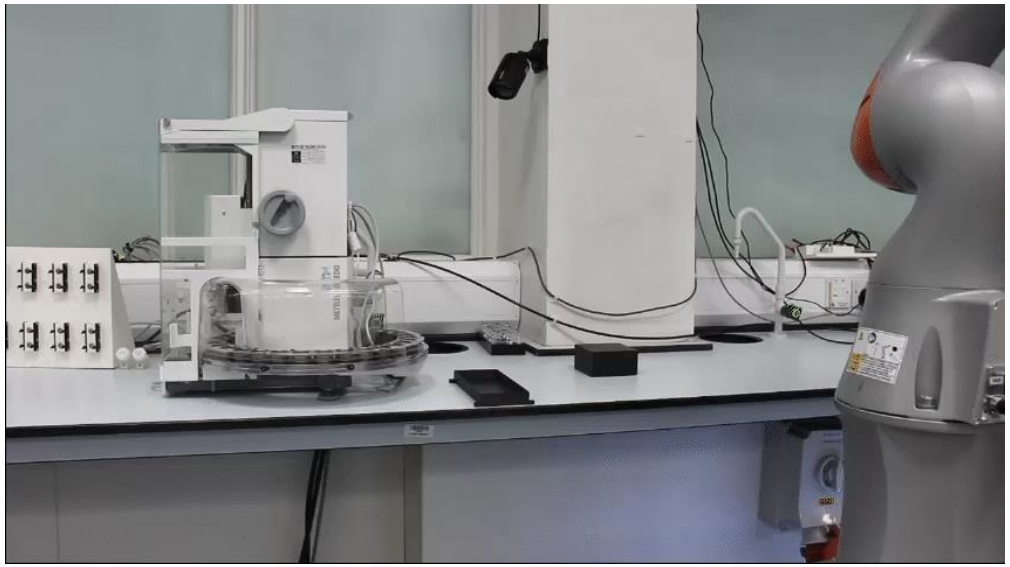
Solution synthesis

$2\ H_2O \rightarrow 2\ H_2 + O_2$

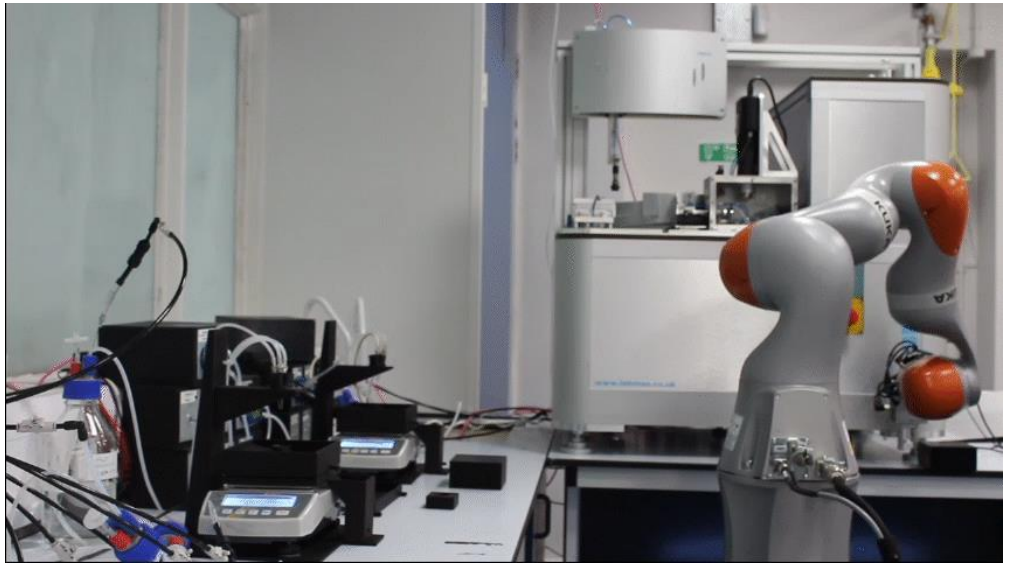
Water splitting

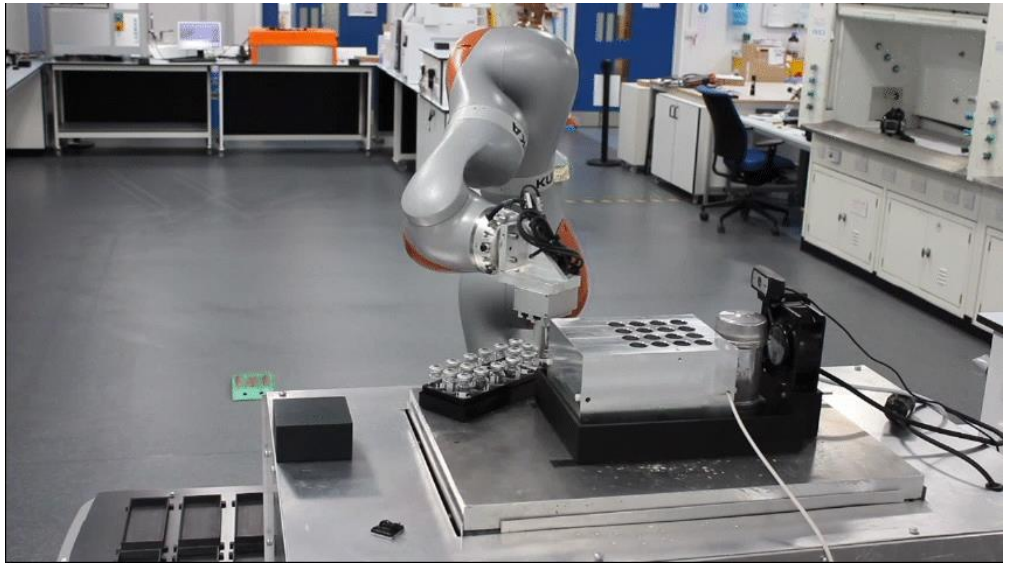
Early examples are promising!

The mobile chemist handles the vials and does sample transfer



Customized stations handle the finer tasks (weighing, mixing, *etc.*)





Early examples are promising!

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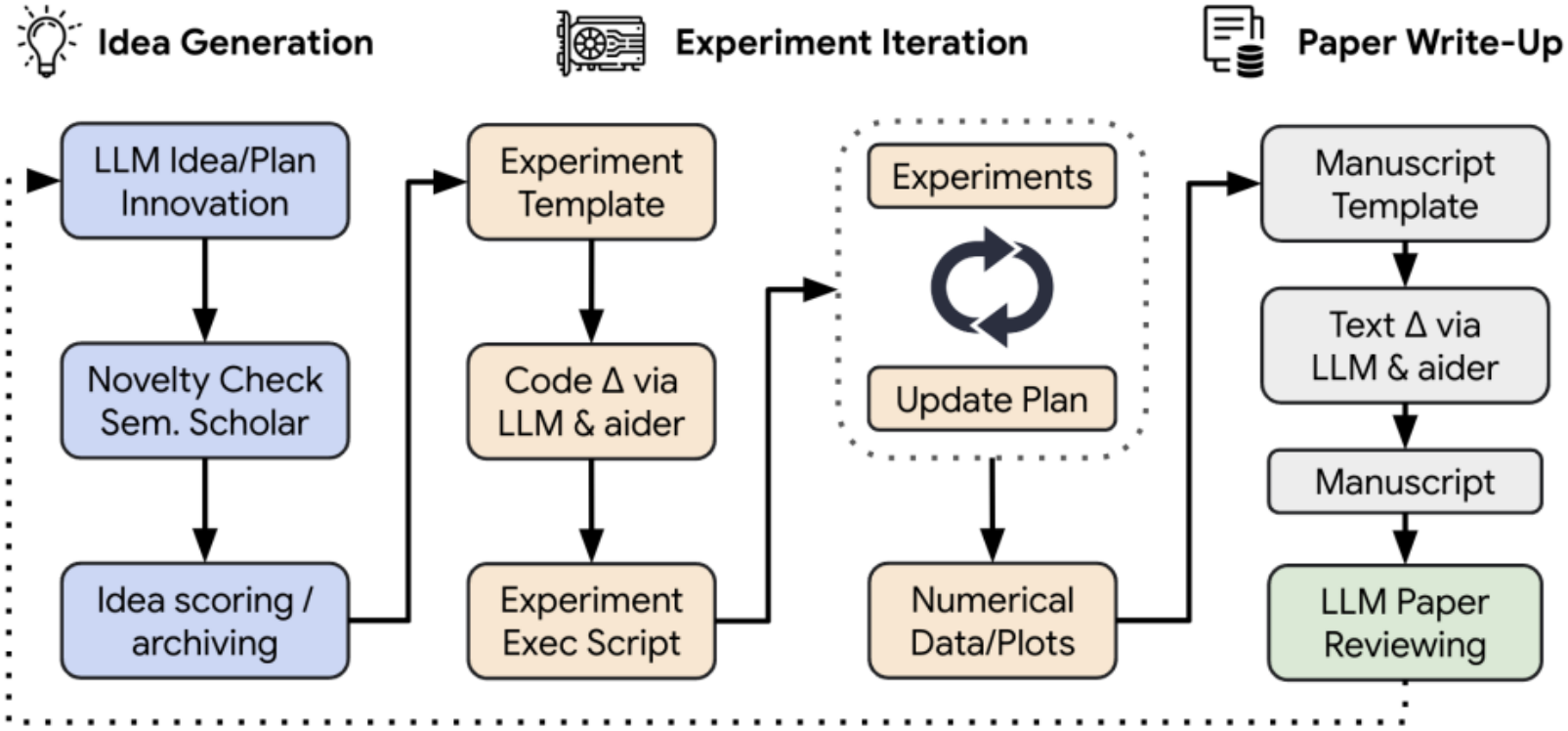
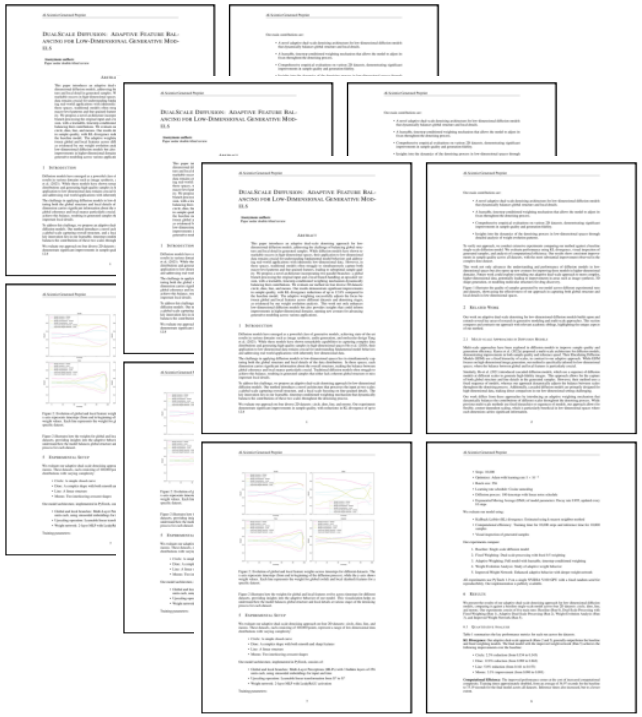
The AI Scientist: Towards Fully Automated Open-Ended Scientific Discovery

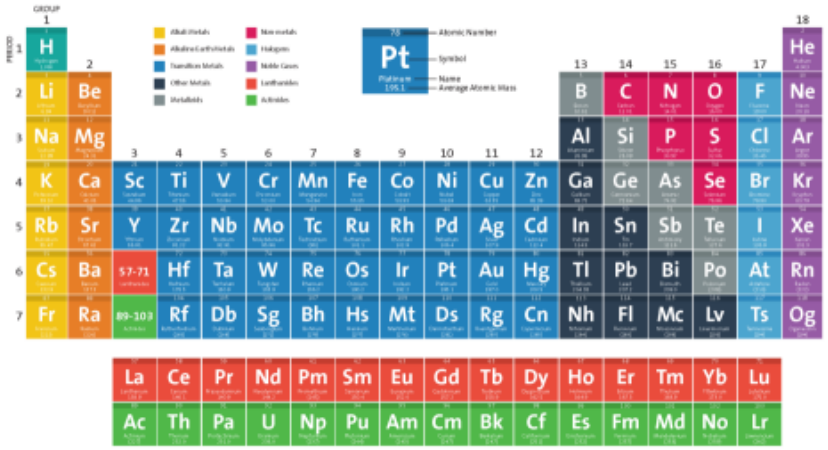
Chris Lu[1,2,*], **Cong Lu**[3,4,*], **Robert Tjarko Lange**[1,*], **Jakob Foerster**[2,†], **Jeff Clune**[3,4,5,†] **and David Ha**[1,†]
[*]Equal Contribution, [1]Sakana AI, [2]FLAIR, University of Oxford, [3]University of British Columbia, [4]Vector Institute, [5]Canada CIFAR AI Chair, [†]Equal Advising

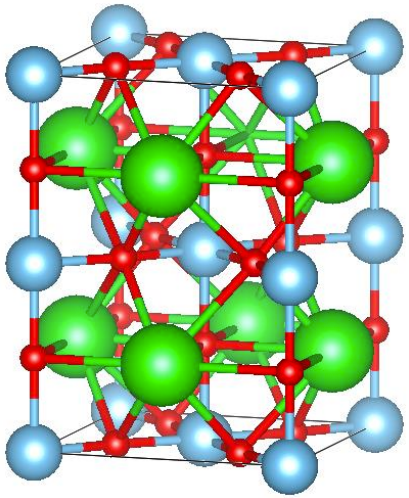
Here, the "experiments" involve writing code, training models, *etc.*





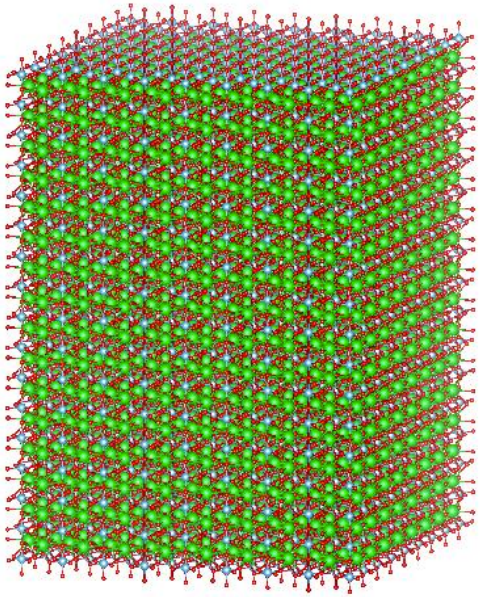
How about for (inorganic) materials science?





Building block

Repeated *many* times in 3-D to form a crystal



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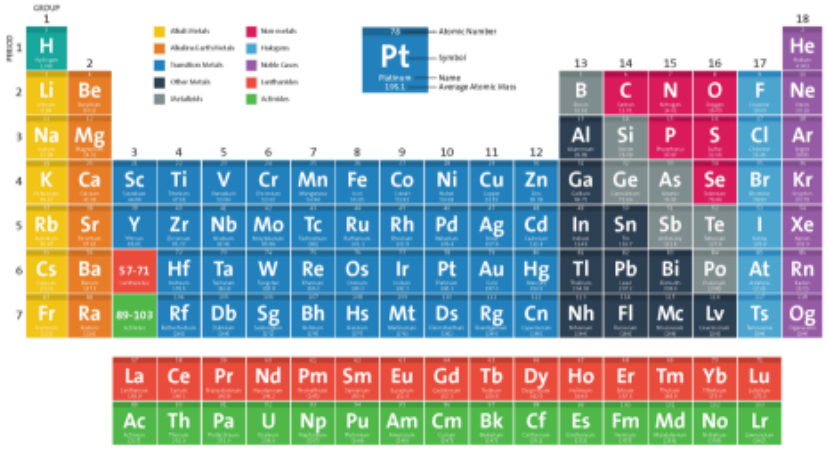

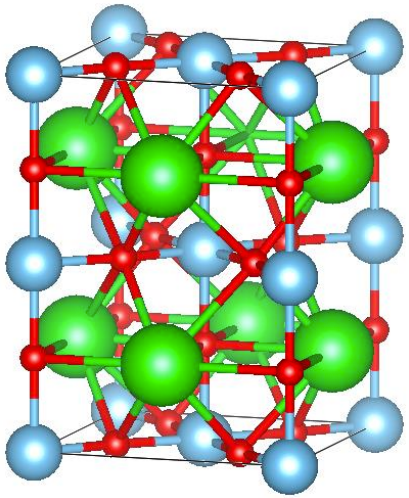

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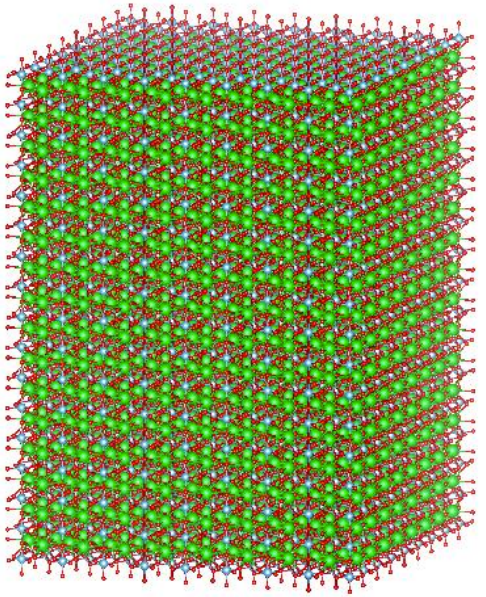
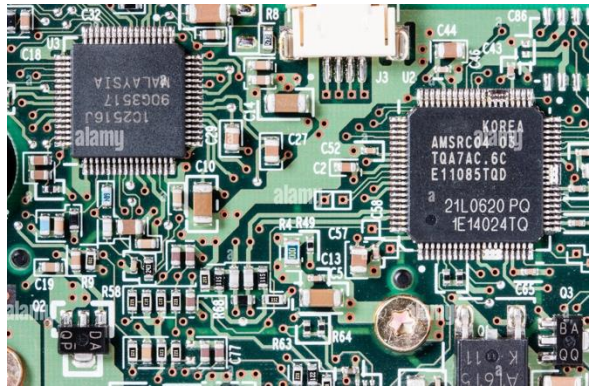
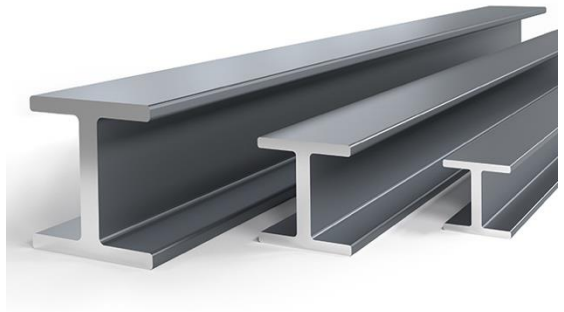
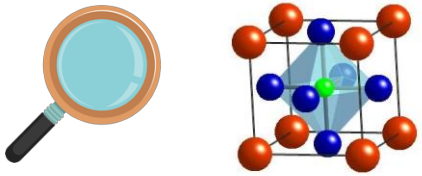


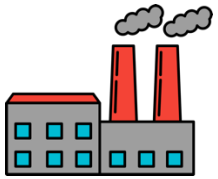




From discovery to commercialization still takes 10-20 years



Materials design



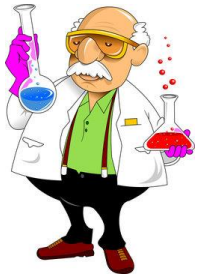
Testing, optimization, scale-up

Stage 1

Stage 2

Stage 3

Synthesis and characterization



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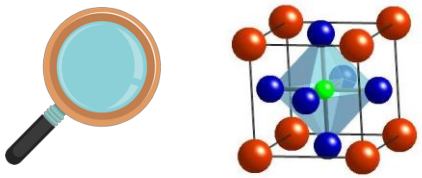
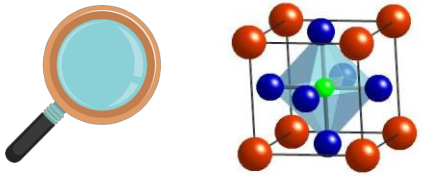


Materials design

Stage 1 → Stage 2 → Stage 3

Modification of a known material (composition, morphology, *etc.*)

From discovery to commercialization still takes 10-20 years



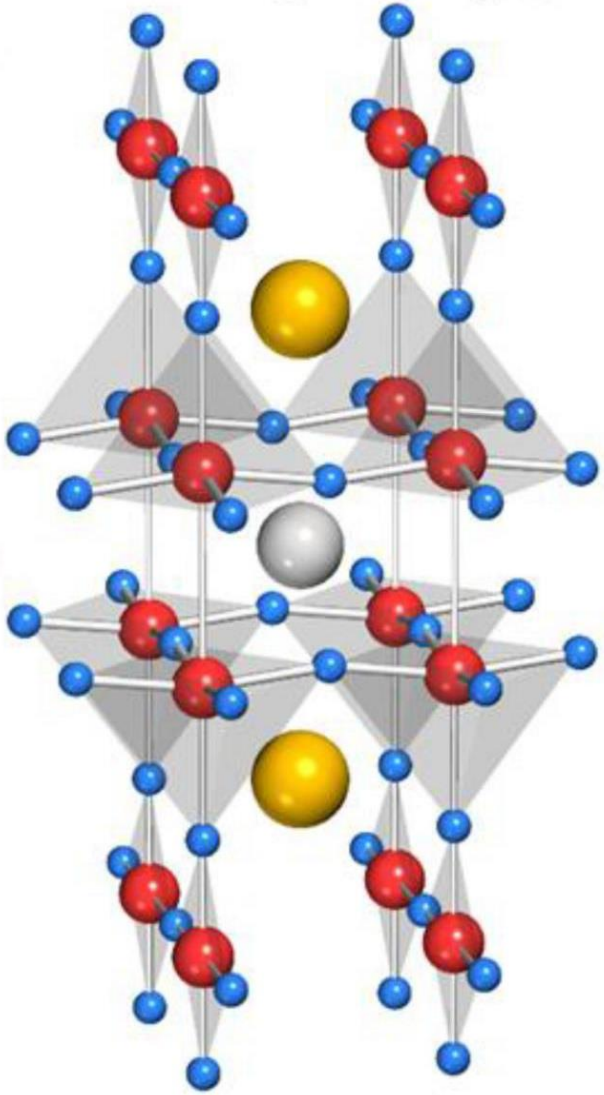
Materials design

Stage 1 | Stage 2 | Stage 3

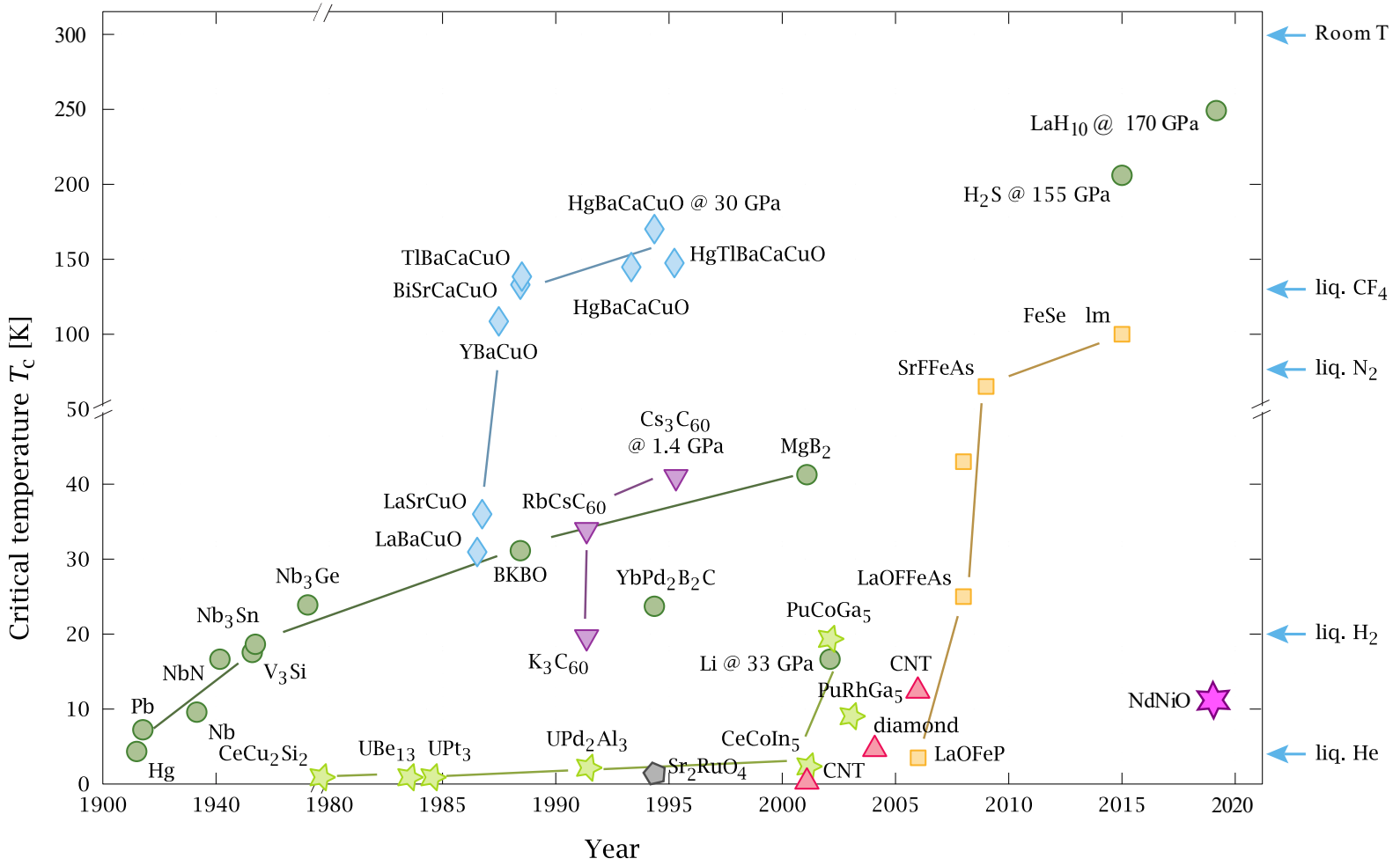
Modification of a known material (composition, morphology, *etc.*)

Discovery of a completely new material

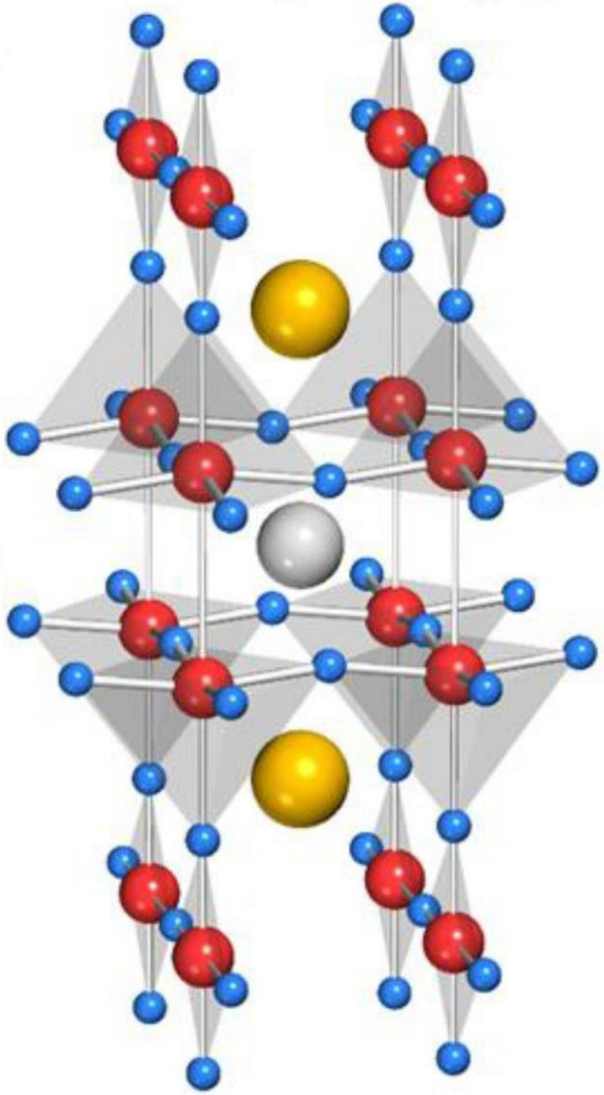
Success stories in materials discovery



Cuprate oxides led to **high-T_c superconductivity**



Success stories in materials discovery



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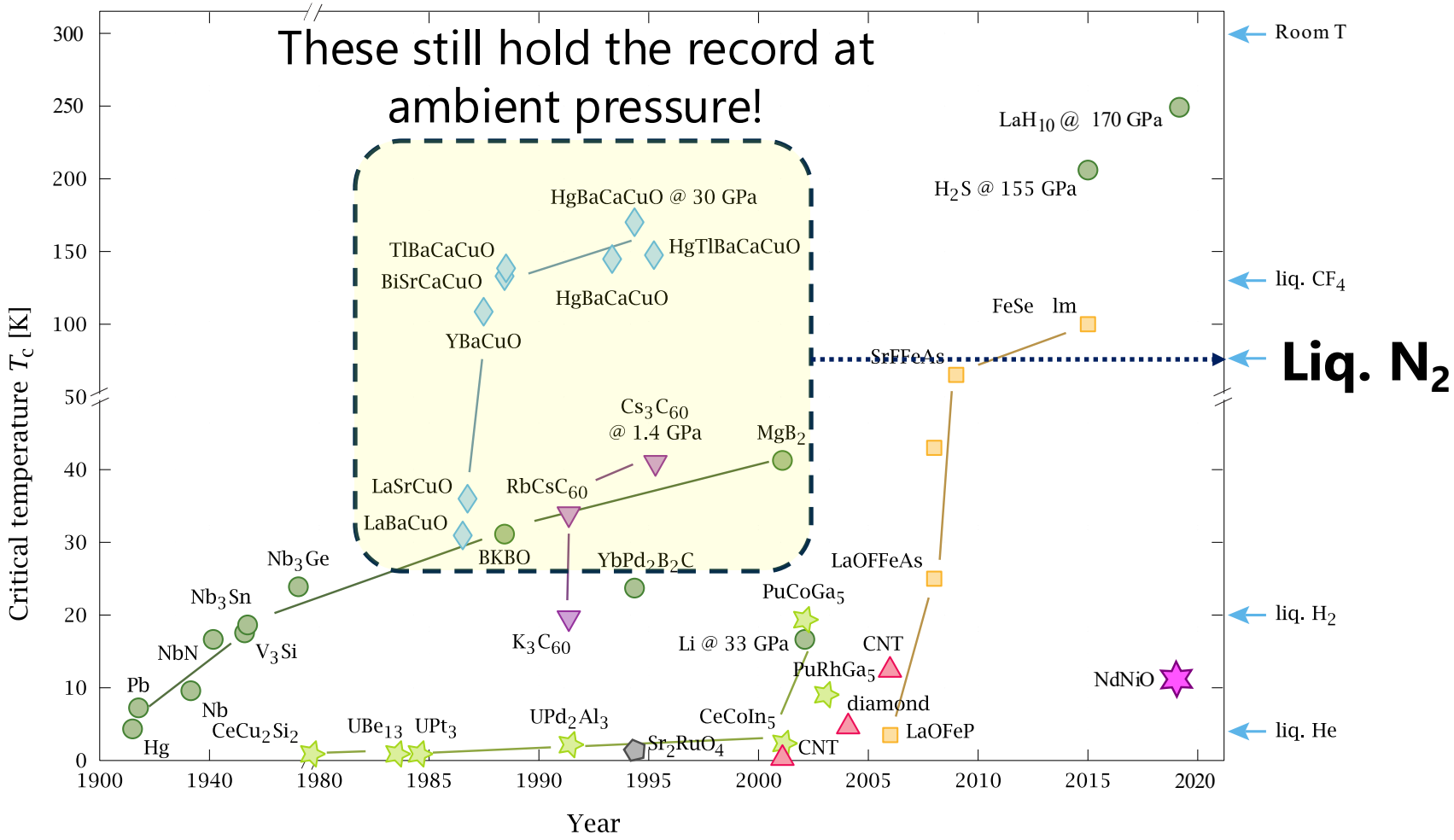

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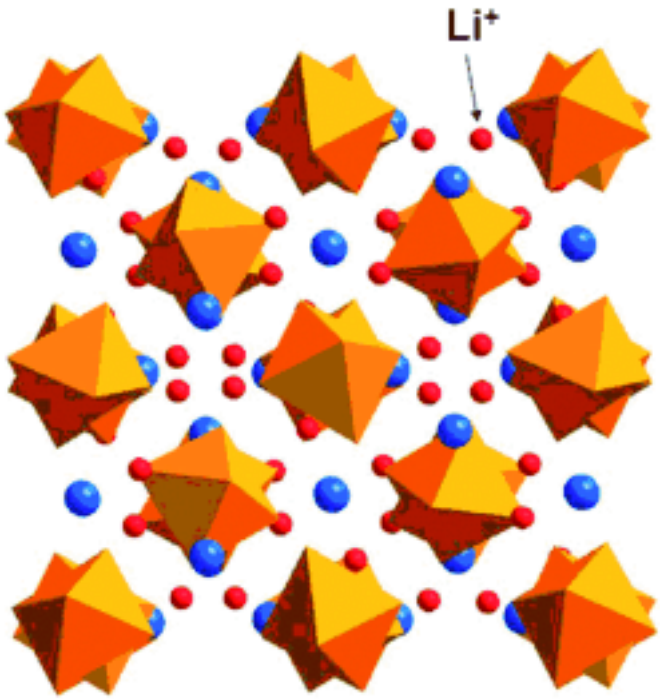
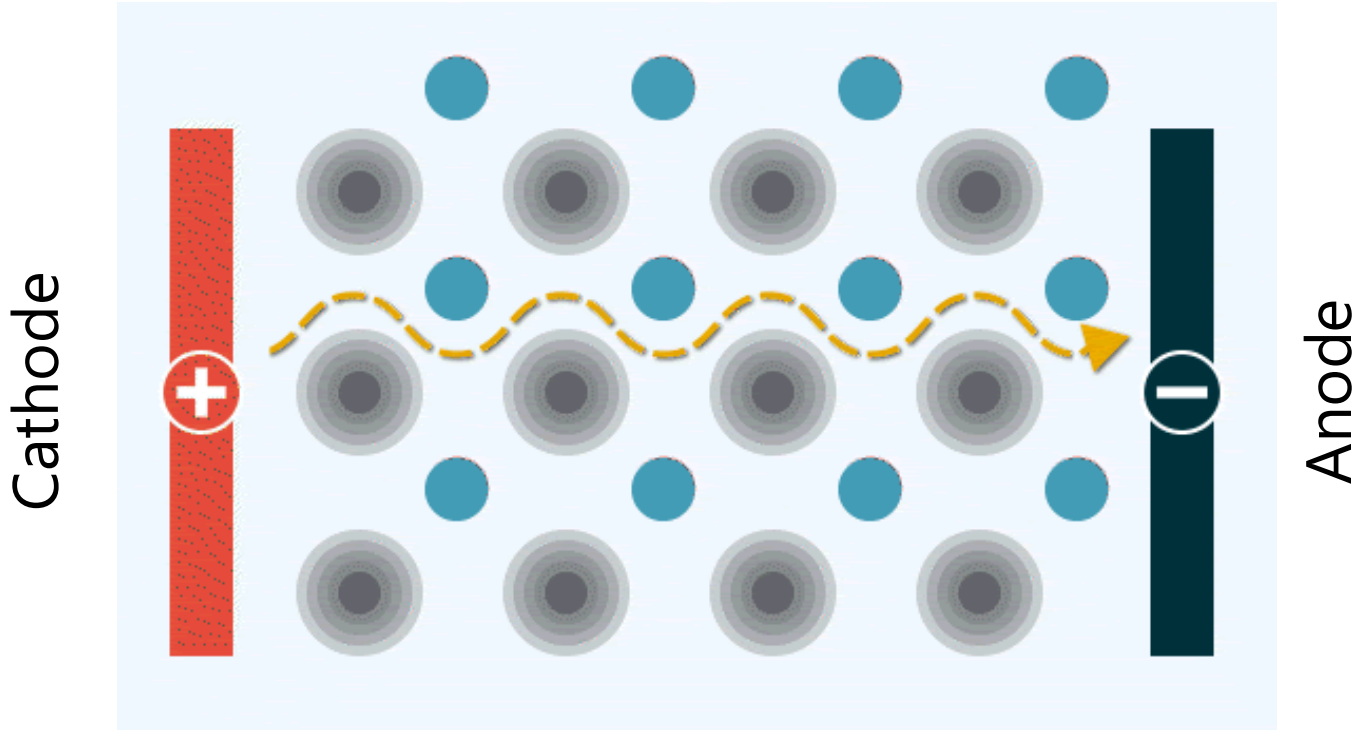


Discovered in 2007, not yet (successfully) commercialized…

Garnet-type $Li_7La_3Zr_2O_{12}$ for solid-state electrolytes



Computational materials design has come a long way!

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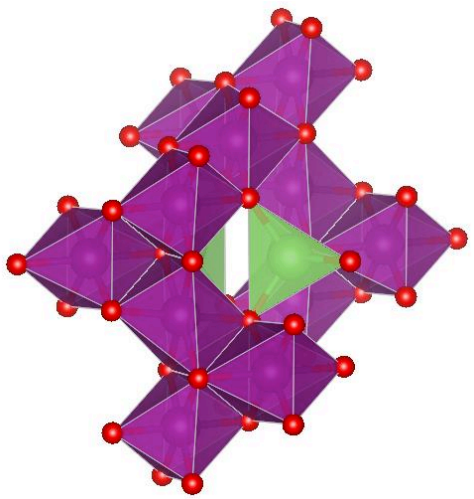
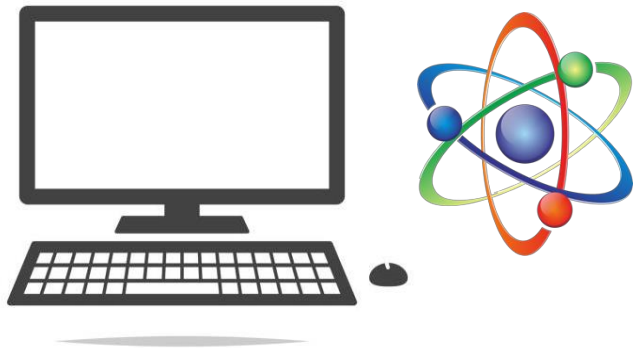
Density functional theory (DFT) can be used to screen for promising materials



Crystal structure



Minutes-to-days of computational time
required for each compound

Stability and
Properties

Computational materials design has come a long way!

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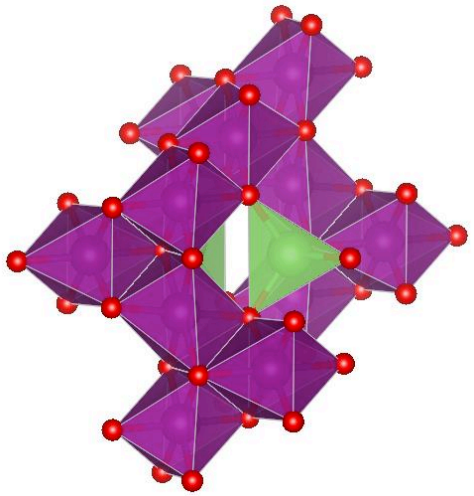
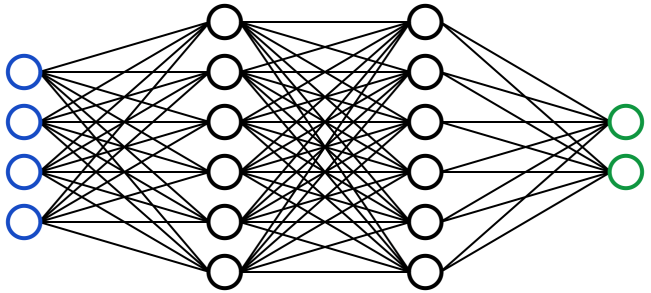
And more recently…**Machine learning Potentials (MLPs)** are coming online



Crystal structure

Seconds of compute time per compound



Stability and Properties

Computational materials design has come a long way!

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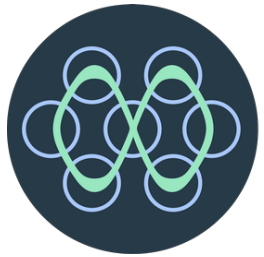
Result: more and more **databases of computed materials** are becoming available

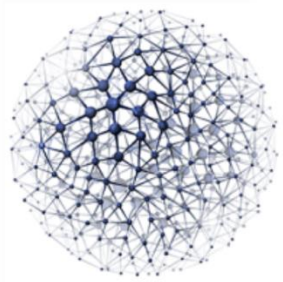


The Materials Project

155,000 materials

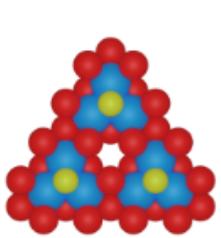
OQMD
The Open Quantum Materials Database

1.22 million materials



AFLOW
Automatic - FLOW for Materials Discovery

3.5 million materials



lexandria

2 million materials

Computational materials design has come a long way!

CEMS Chemical Engineering & Materials Science

Result: more and more **databases of computed materials** are becoming available

But there are *very* few examples of successfully commercialized materials originating from computations…

Why?

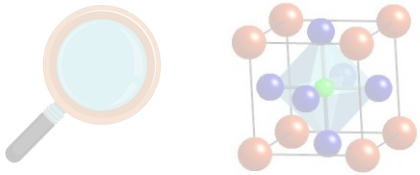
Automatic - FLOW for Materials Discovery

3.5 million materials

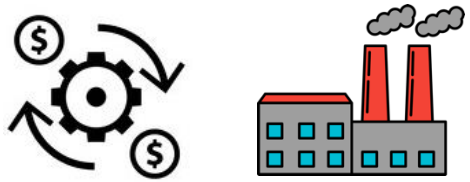
lexandria

2 million materials

Experimental synthesis and *optimization* are still bottlenecks



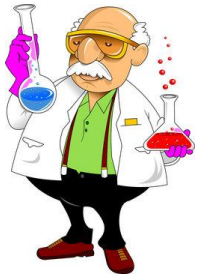
Materials design



Testing, optimization, scale-up

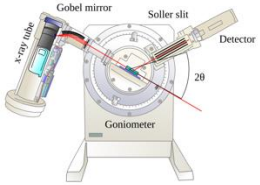
Stage 1 | Stage 2 | Stage 3

Synthesis and characterization



Experimental synthesis and *optimization* are still bottlenecks

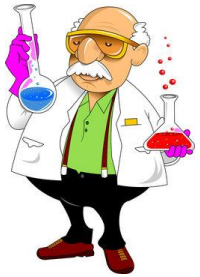
A grad student can **synthesize a "handful" of samples each day**



Even more time is needed to **characterize** them

Stage 1 | **Stage 2** | Stage 3

Synthesis and characterization



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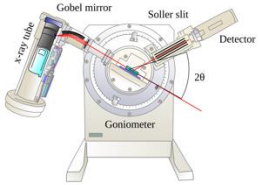


Even more time is needed to **characterize** them

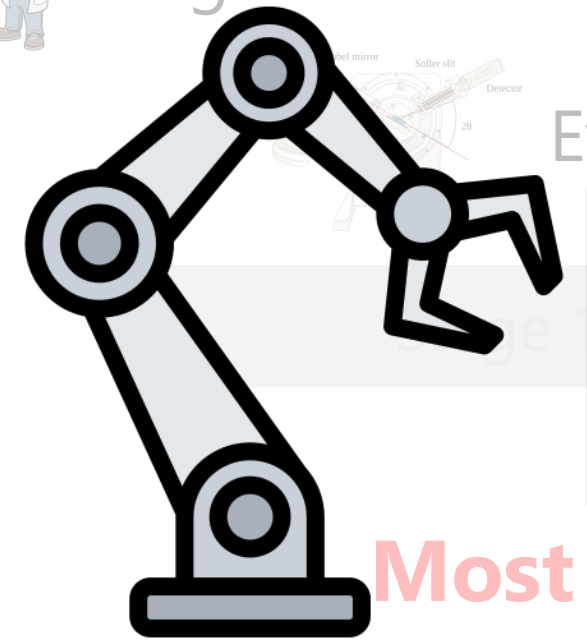
Stage 1 | **Stage 2** | Stage 3

Most initial synthesis attempts will fail!

Computations do not tell us how to make a material

Experimental synthesis and *optimization* are still bottlenecks

A grad student can **synthesize a "handful" of samples each day**



Even more time is needed to **characterize** them

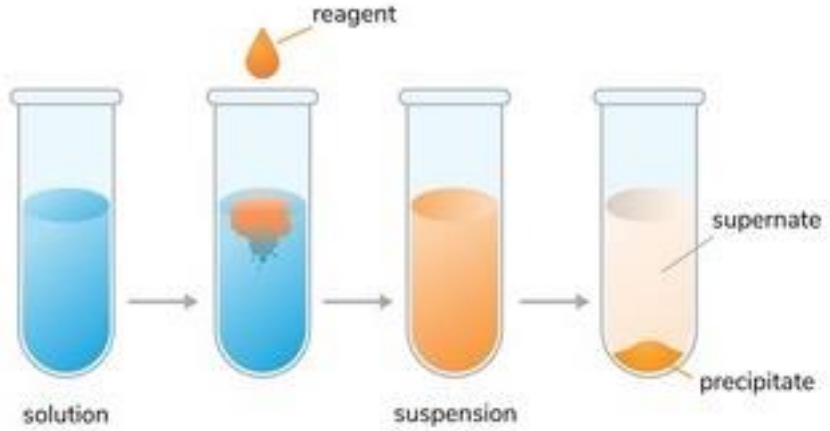
Can automation help alleviate this problem?

Most initial synthesis attempts will fail!

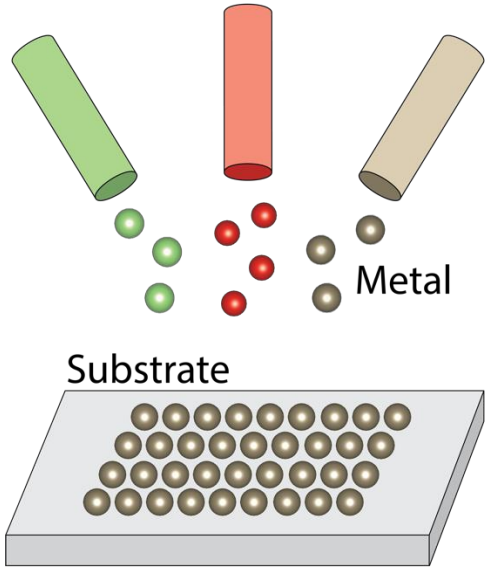
Computations do not tell us how to make a material

There are *many* approaches to make inorganic materials

High-temperature
(solid-state)



Low-temperature
(solution-based)



Deposition
(thin films)

There are *many* approaches to make inorganic materials

High-temperature (solid-state)

Long been the workhorse for inorganic materials synthesis…but remains stubbornly **difficult to automate in a versatile way**

The basics of solid-state synthesis

Target

$LiMnTiO_4$

Synthesis recipe

- ☑ 50 mg Li_2CO_3
- ☑ 80 mg MnO
- ☑ 20 mg TiO_2
- ☑ 800 °C (air)
- ☑ 24 hours

50 mg

20 mg

80 mg

Shake 'n bake

800 °C, 24 hours

Final product!

Even this “simple” technique has limitations

Target

$LiMnTiO_4$

Synthesis recipe

- ☑ 50 mg Li_2CO_3
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Shake ‘n bake

800 °C, 24 hours

Final product!

😢

- Starting precursors often react to give **unwanted byproducts/impurities**
- This can result in **zero target yield**, which is **difficult to optimize!**

Closing the loop for autonomous materials synthesis

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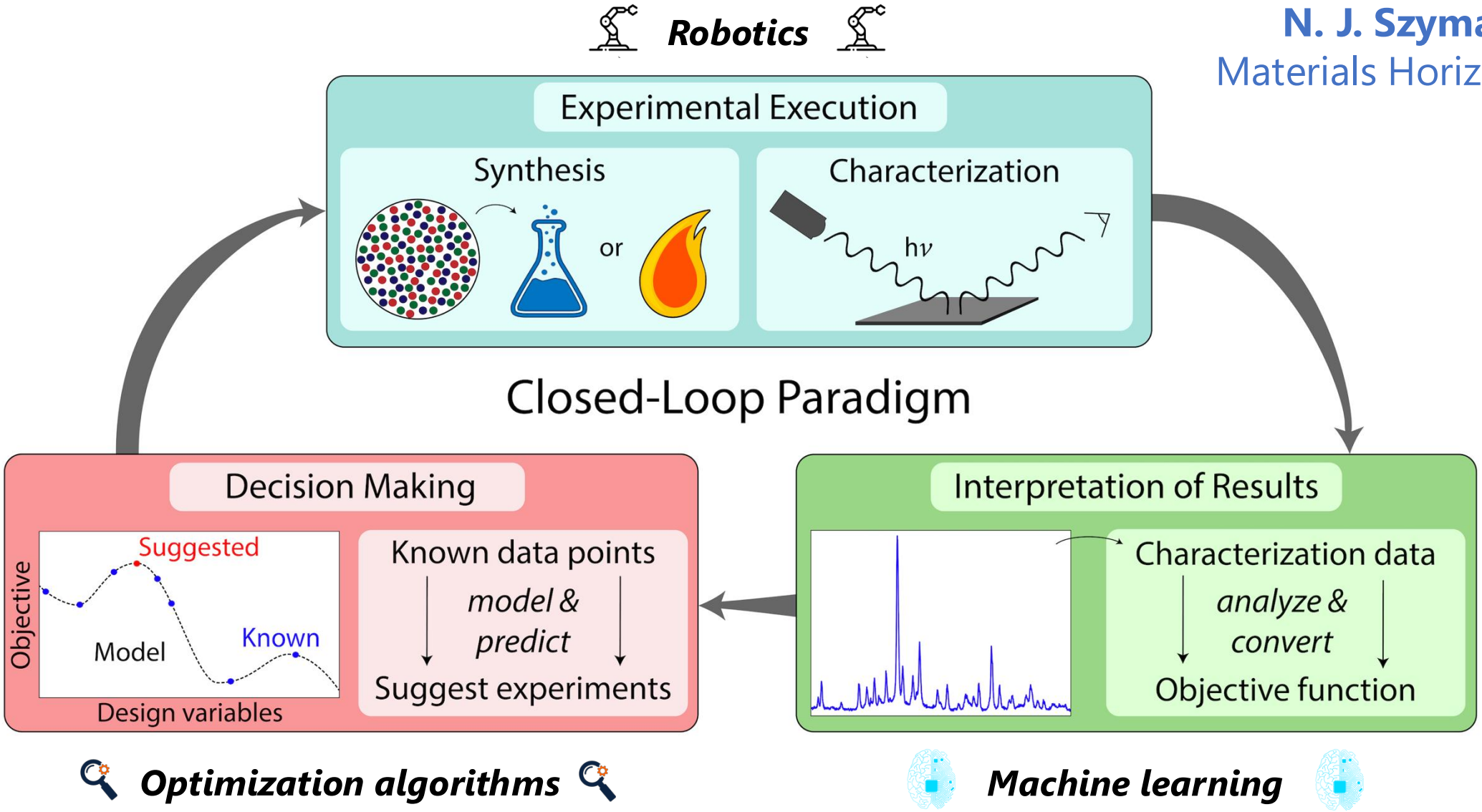
N. J. Szymanski *et al.*, Materials Horizons (2021).



Closing the loop for autonomous materials synthesis

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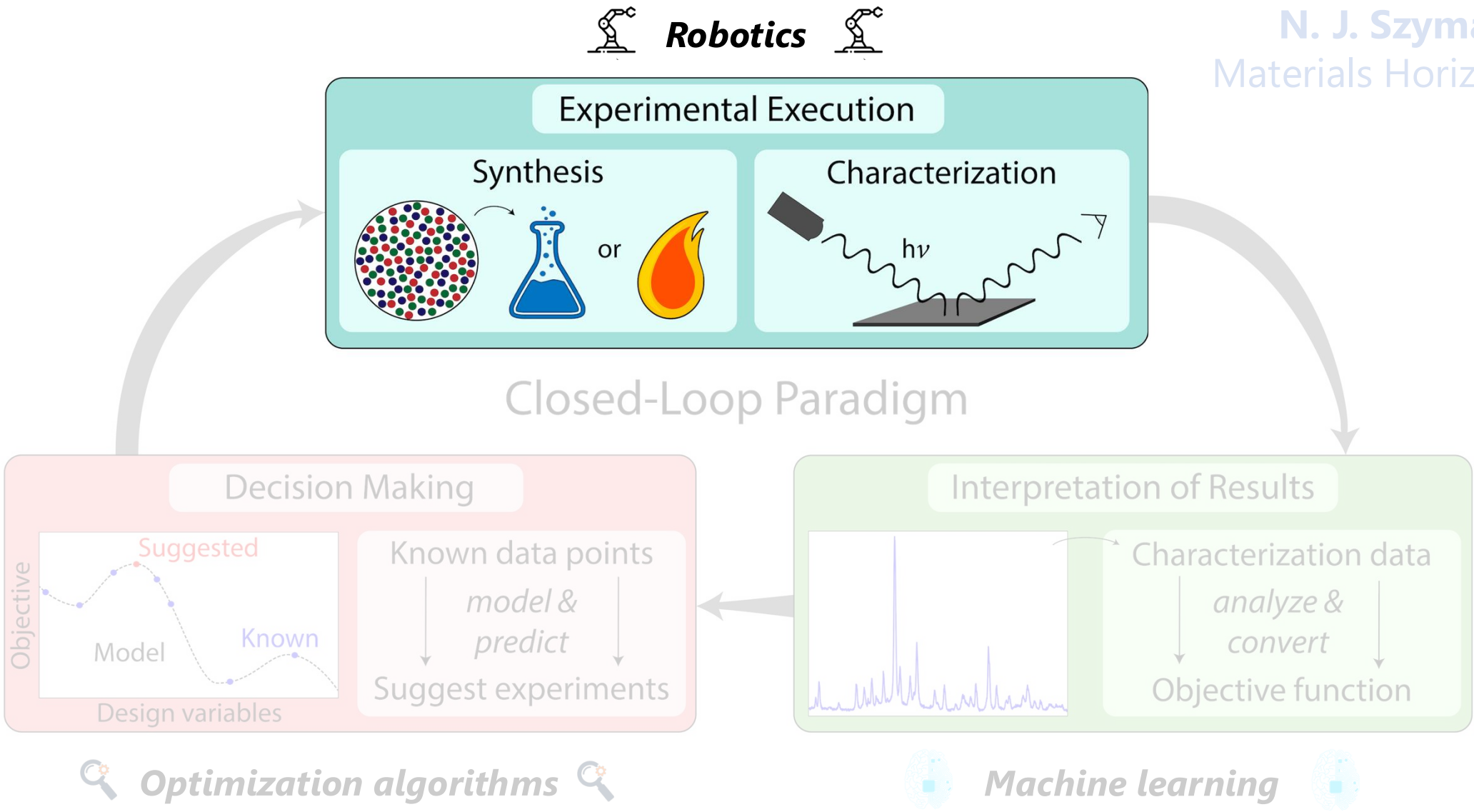

N. J. Szymanski *et al.*, Materials Horizons (2021).

The A-Lab: three robotic stations in cooperation

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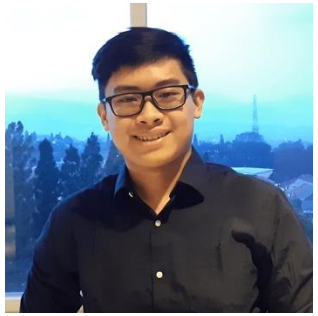
Precursor preparation

Heating station

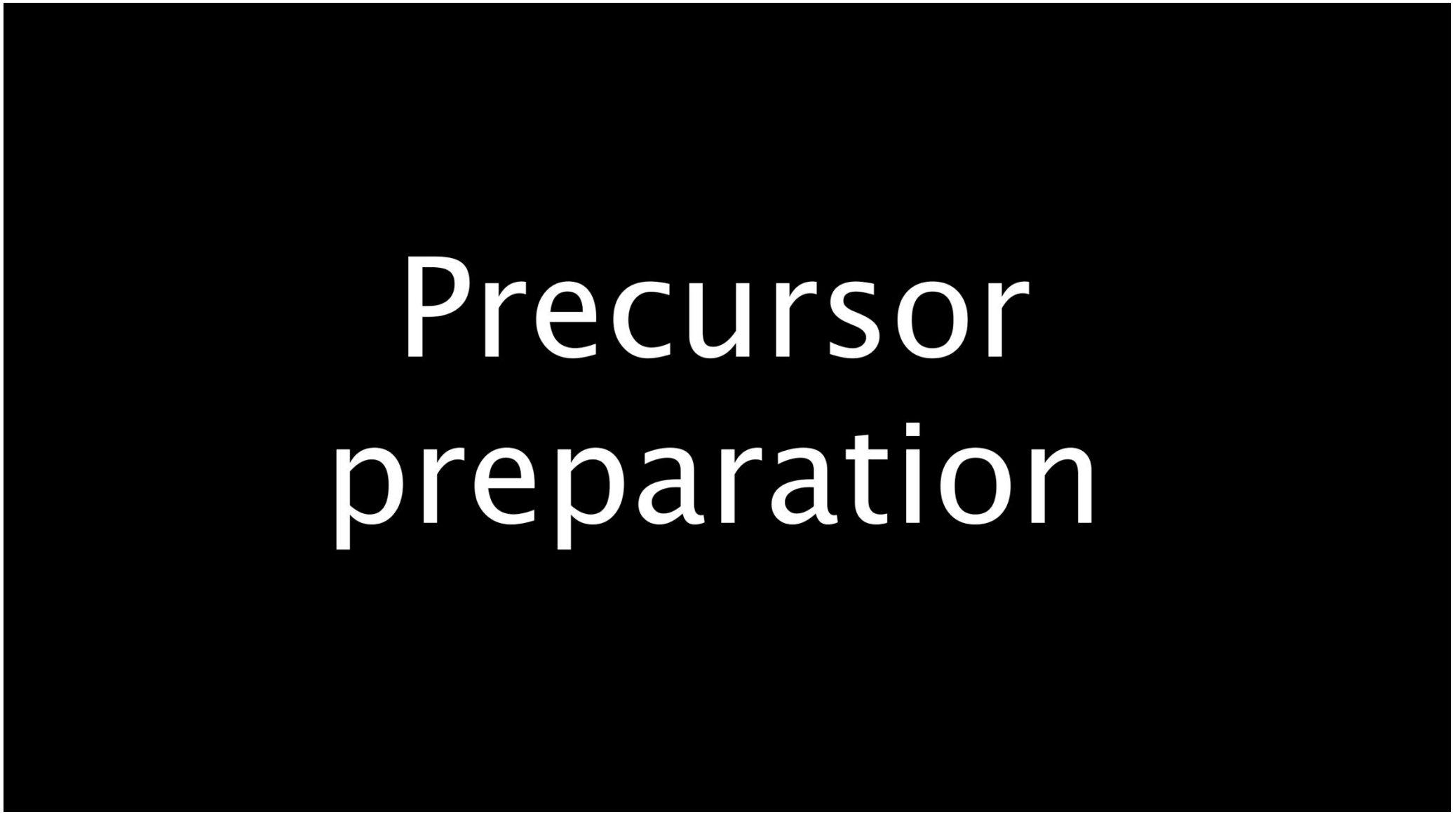
Characterization

The hardware team



N. J. Szymanski, B. Rendy, Y. Fei, *et al.*, Nature (2023).

The A-Lab: a video demo



How to interpret the XRD patterns that A-Lab produces?

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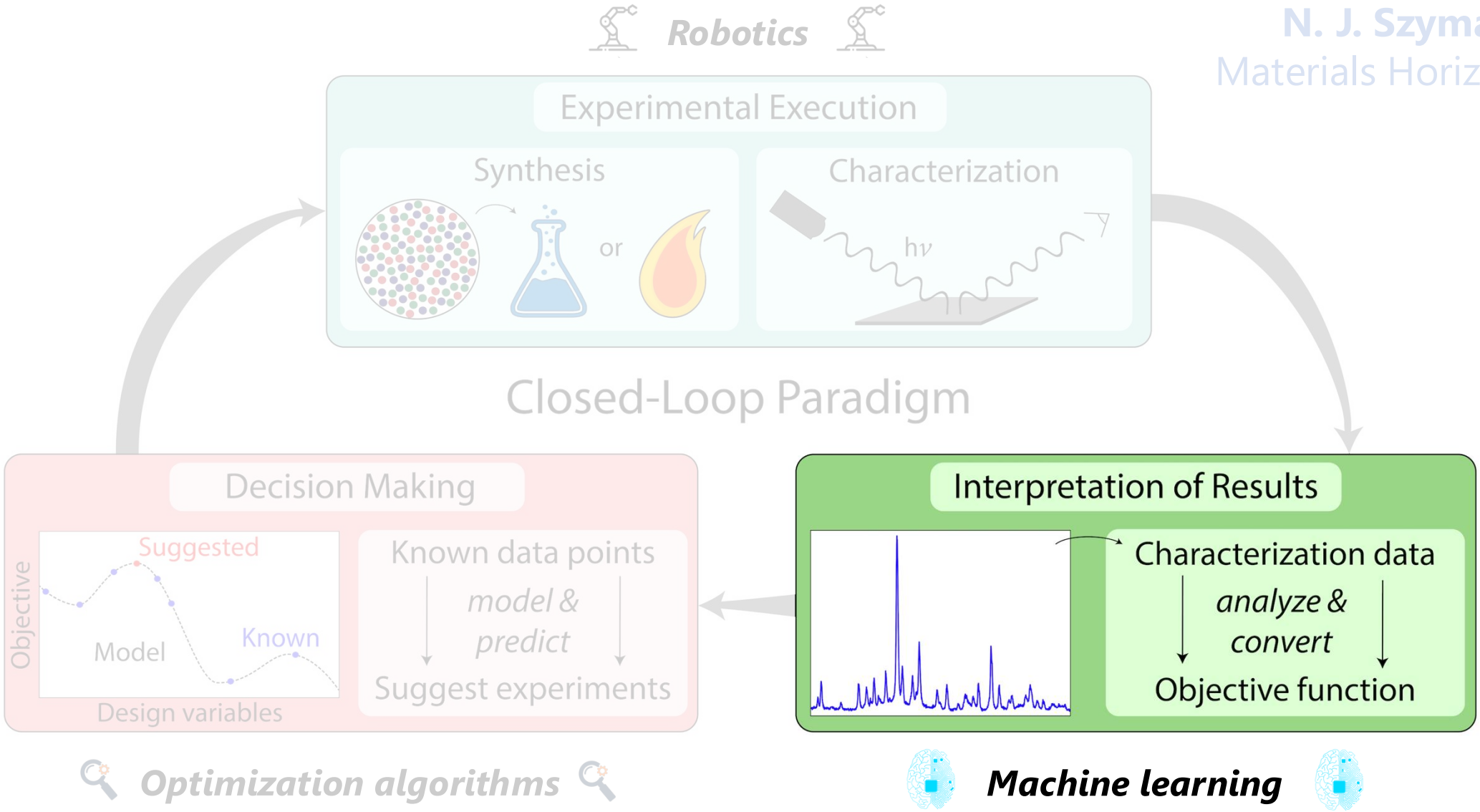
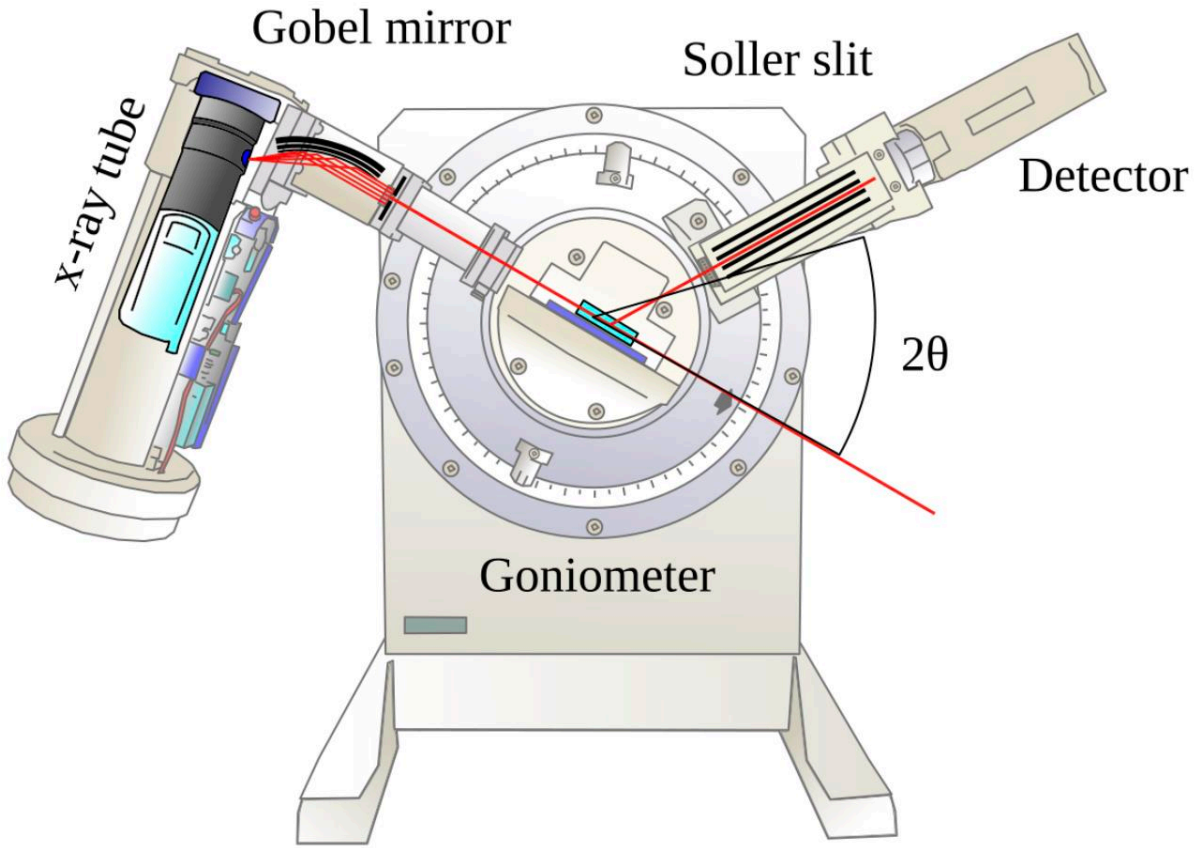
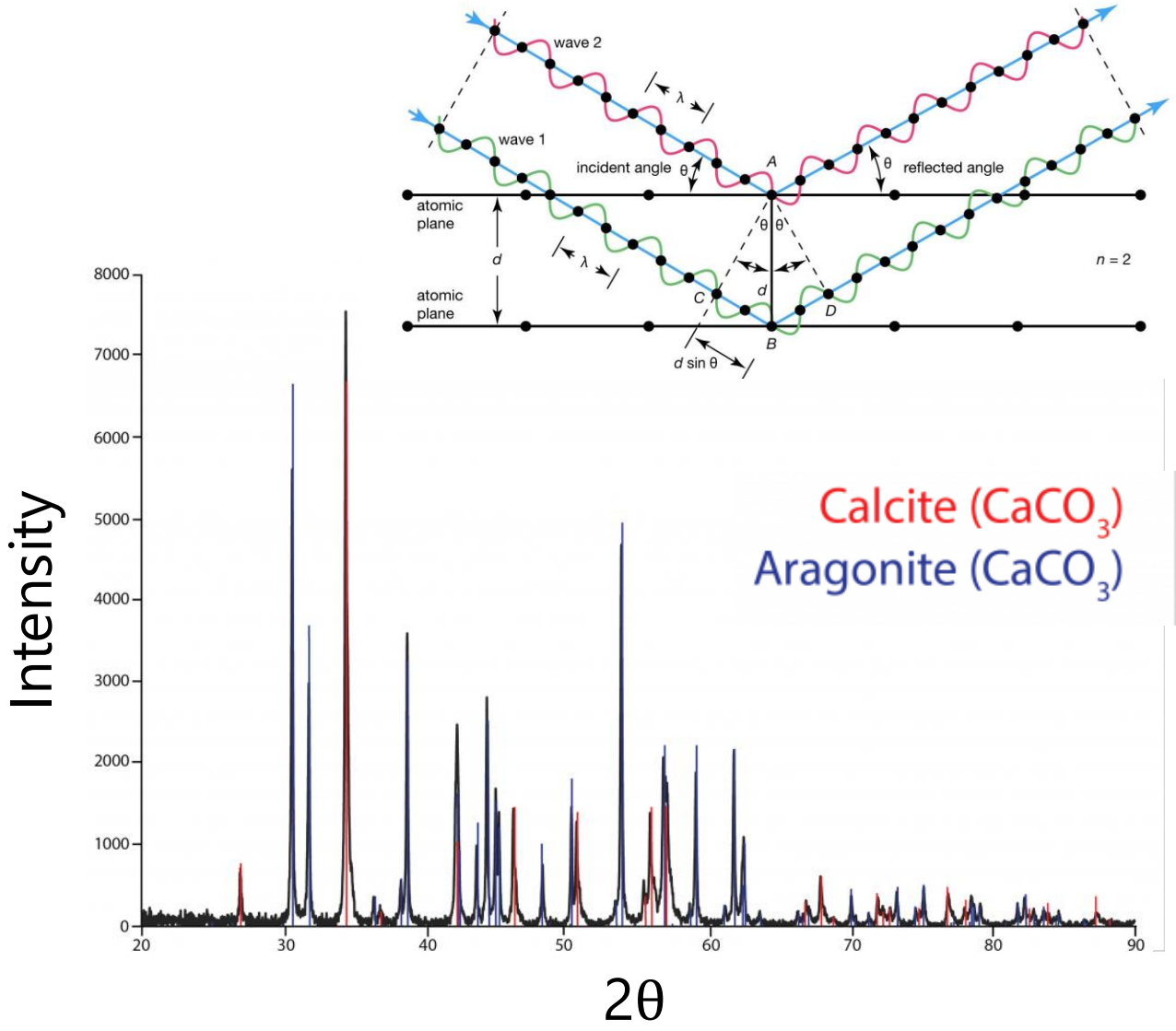
N. J. Szymanski *et al.*, Materials Horizons (2021).



A review of X-ray diffraction (XRD) for crystalline materials

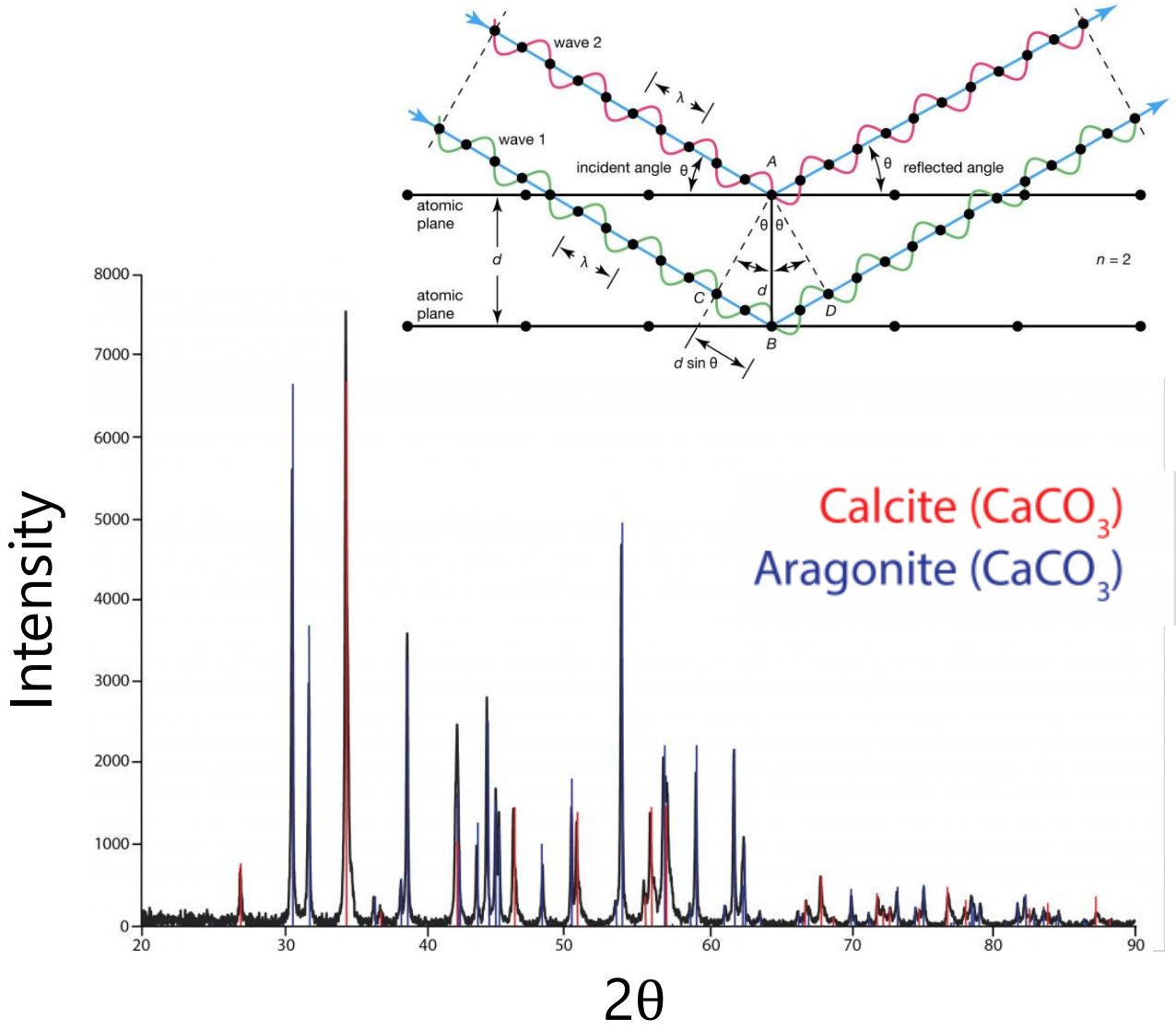




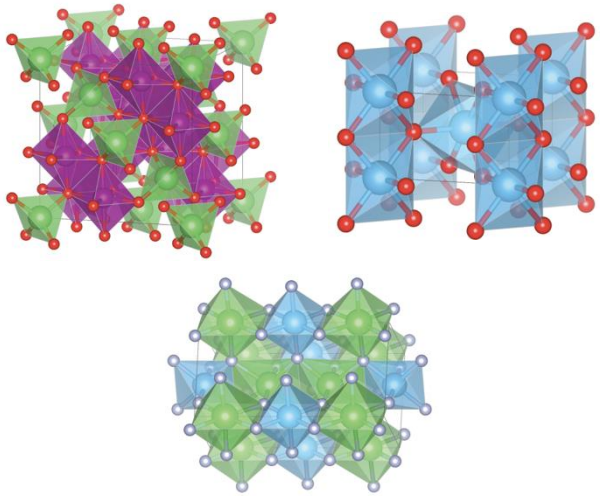
A review of X-ray diffraction (XRD) for crystalline materials

These patterns act as **“fingerprints”** for materials.

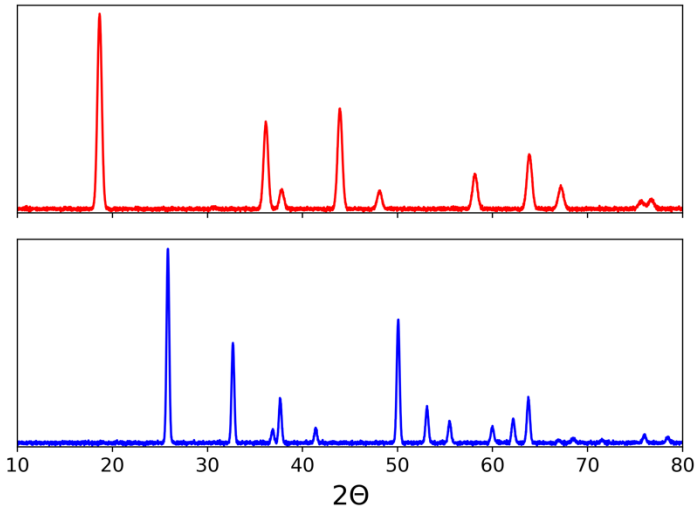
But **how do we know what they’ll look like?**



Forward problem is easy, but reverse problem is hard



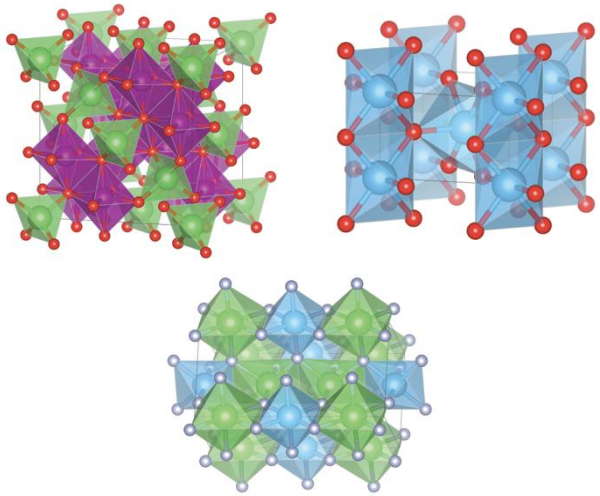
$$F_{hkl} = \sum_{j=1}^{N} f_j e^{-2\pi i(hx_j + ky_j + lz_j)}$$



How to go from XRD to structure?

- XRD pattern is **not unique**
- **Multi-phase mixtures** are common
- **Experimental artifacts** modify peaks

Forward problem is easy, but reverse problem is hard



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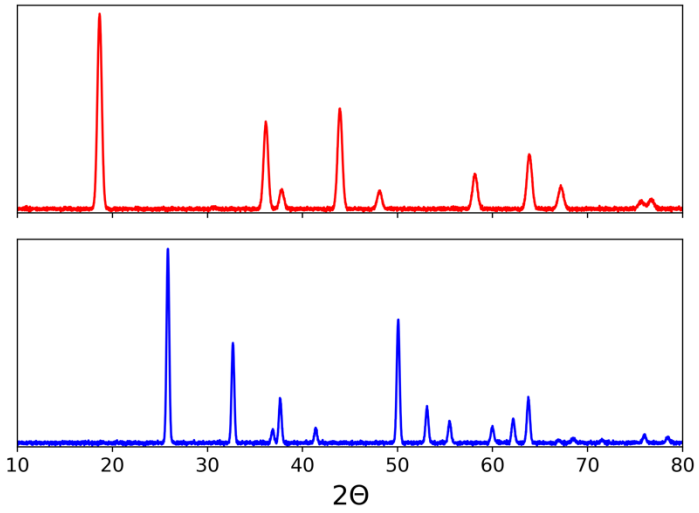


Train ML to solve reverse problem!

- XRD pattern is **not unique**
- **Multi-phase mixtures** are common
- **Experimental artifacts** modify peaks

These can all be ***simulated*** and fed to an ML model as training data

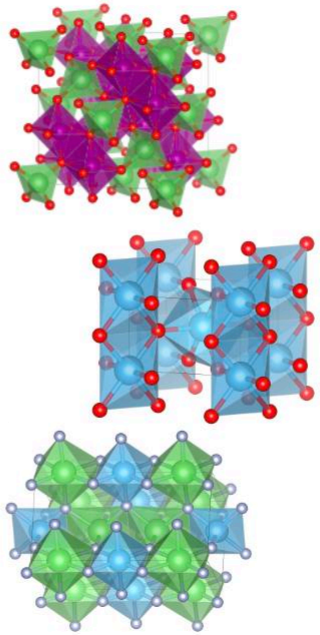
Neural networks + physics-informed data augmentation

Known structures

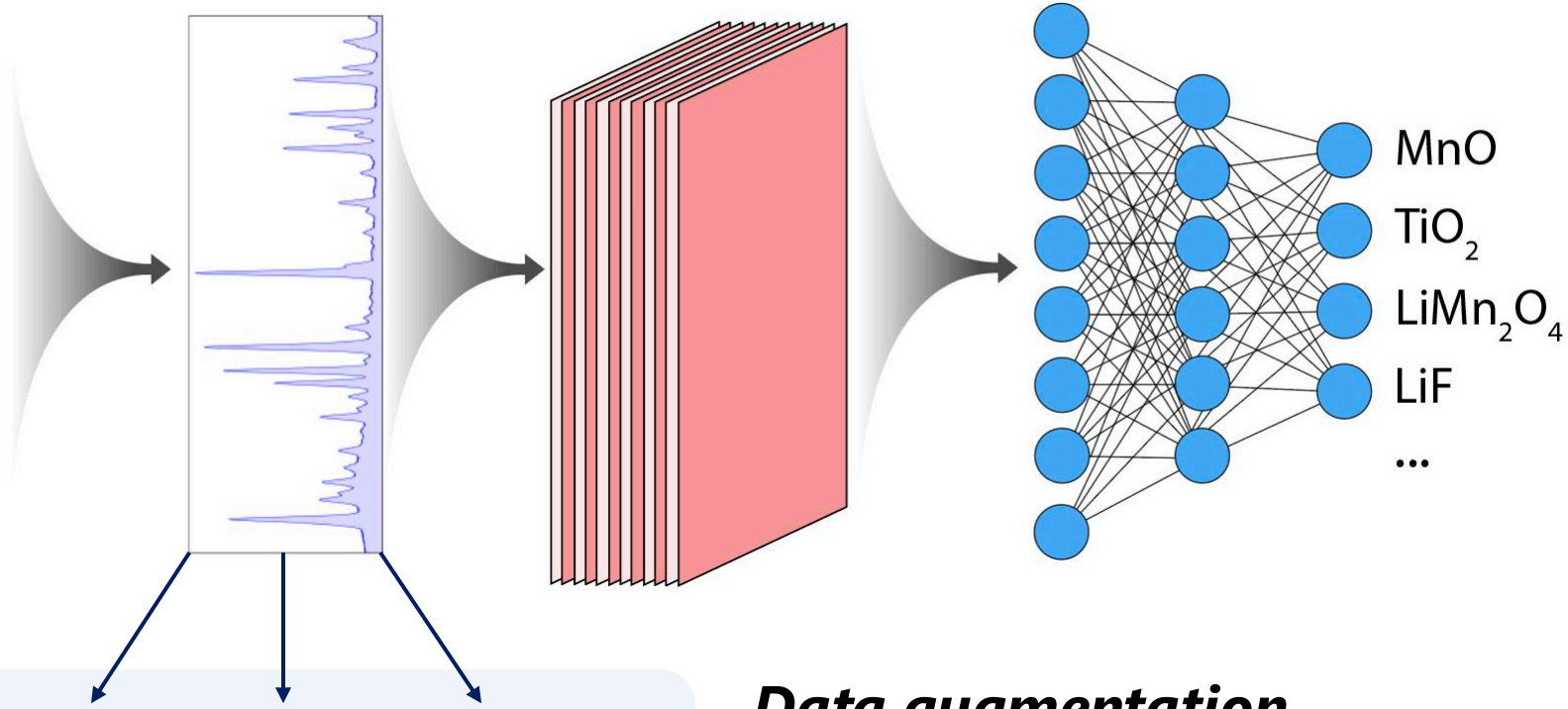
Simulated patterns

Convolutional neural networks

ICSD
FIZ Karlsruhe



Materials Project

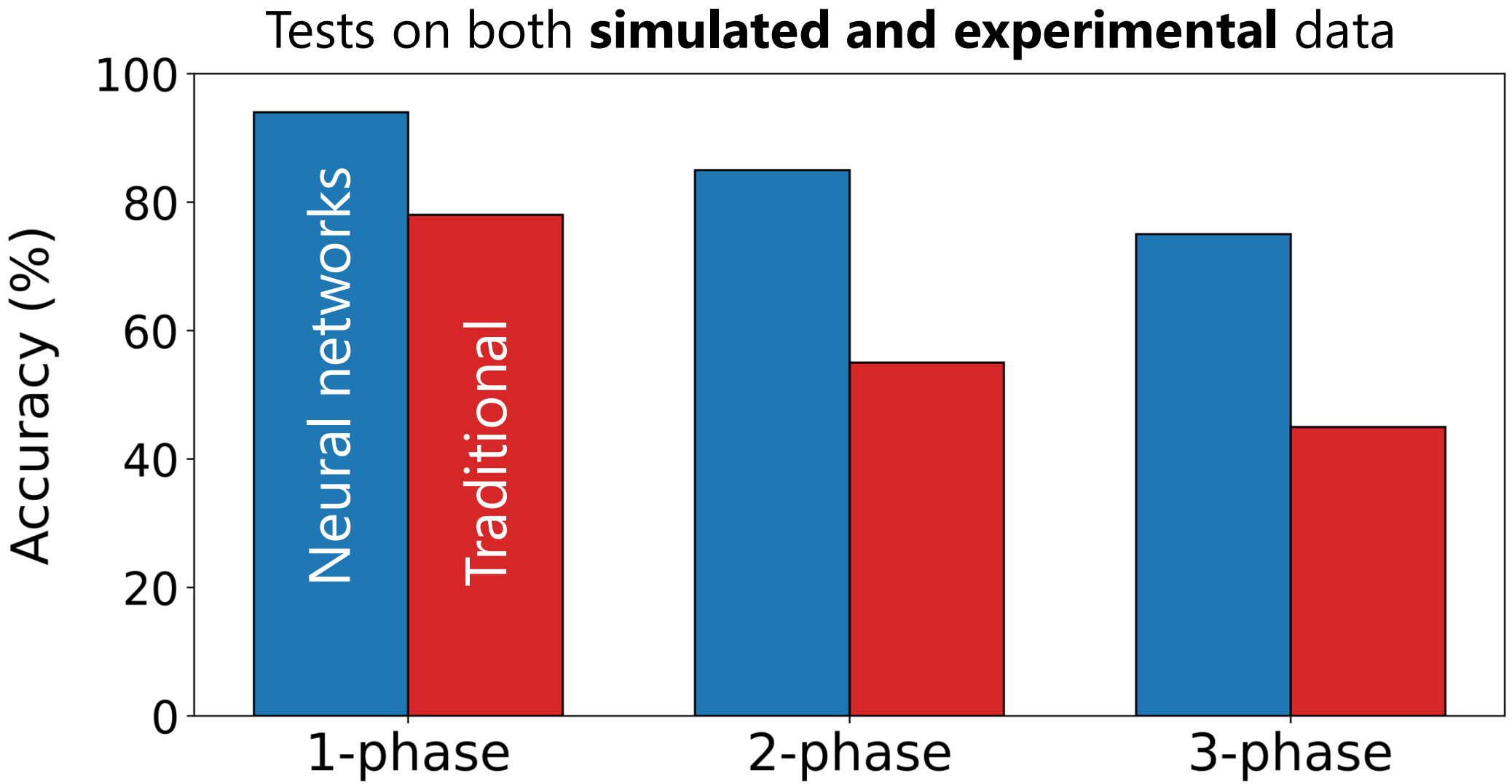


Shifts in 2θ | Intensity changes | Broad peaks

Data augmentation from strain, texture, and poor crystallinity

N. J. Szymanski *et al.*, Chem. Mater (2021).

Tests show that ML outperforms traditional methods



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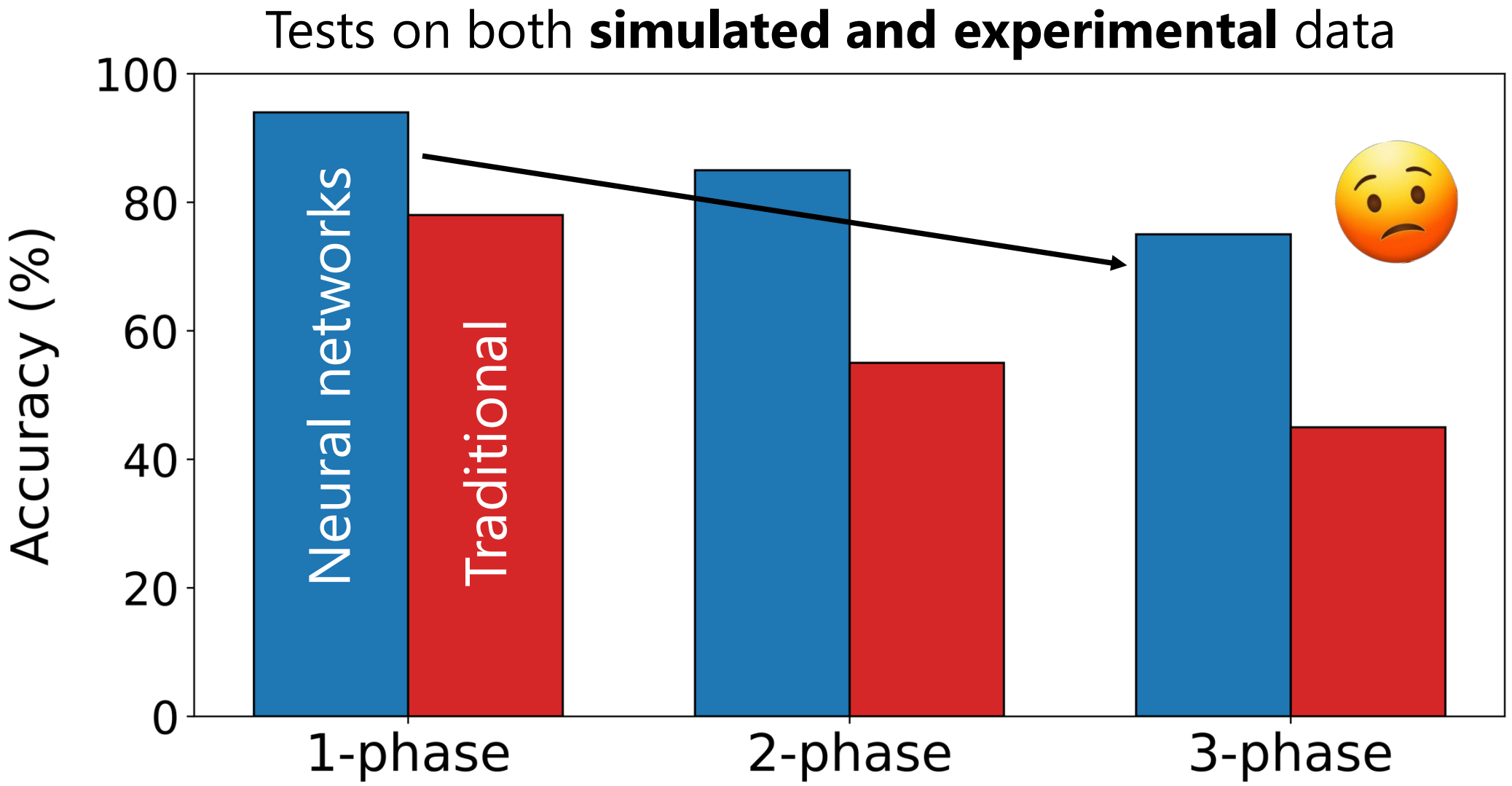

Mixtures are difficult to characterize reliably in XRD…

If the experiment failed, what should A-Lab do next?

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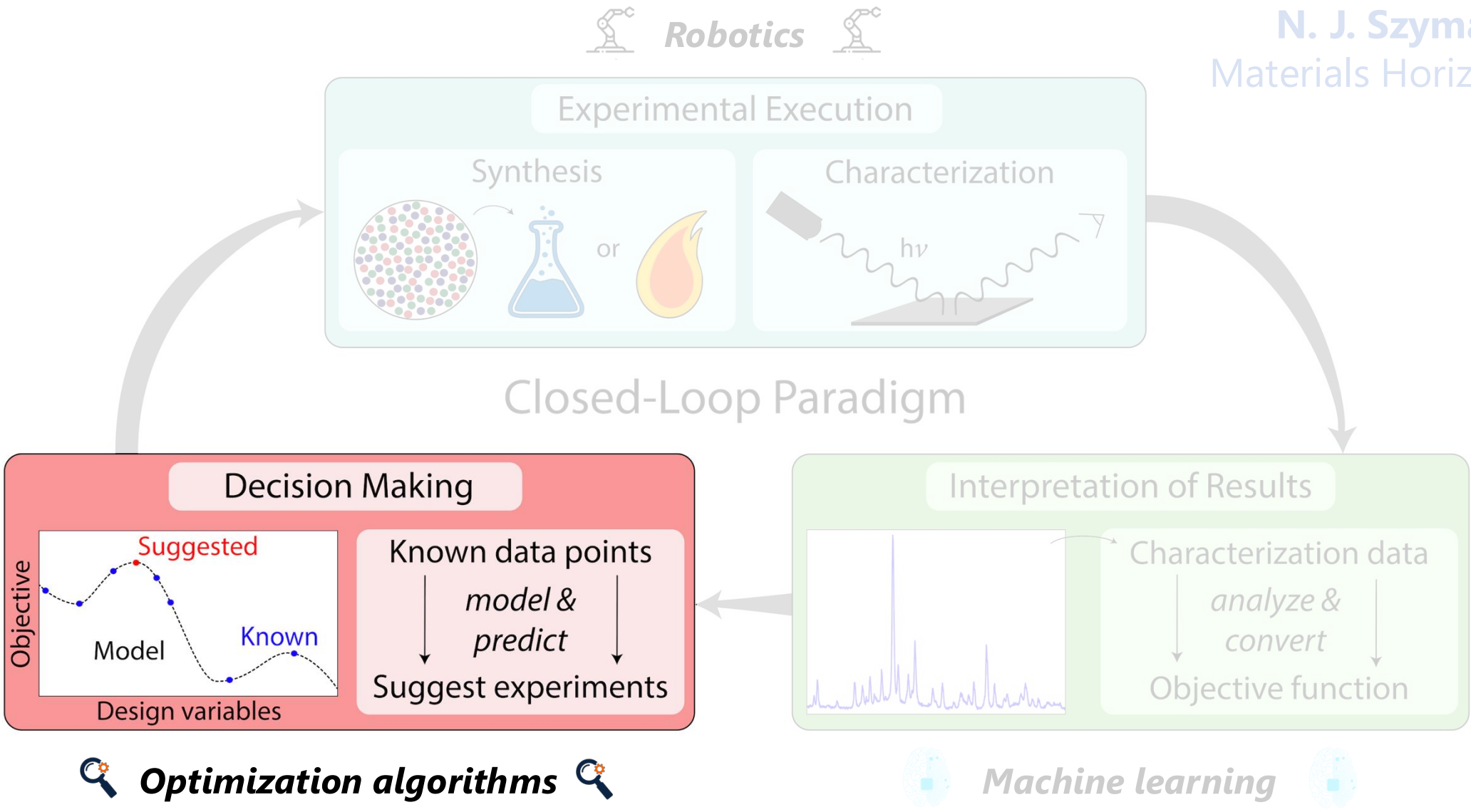


N. J. Szymanski *et al.*, Materials Horizons (2021).

Some background on synthesis science

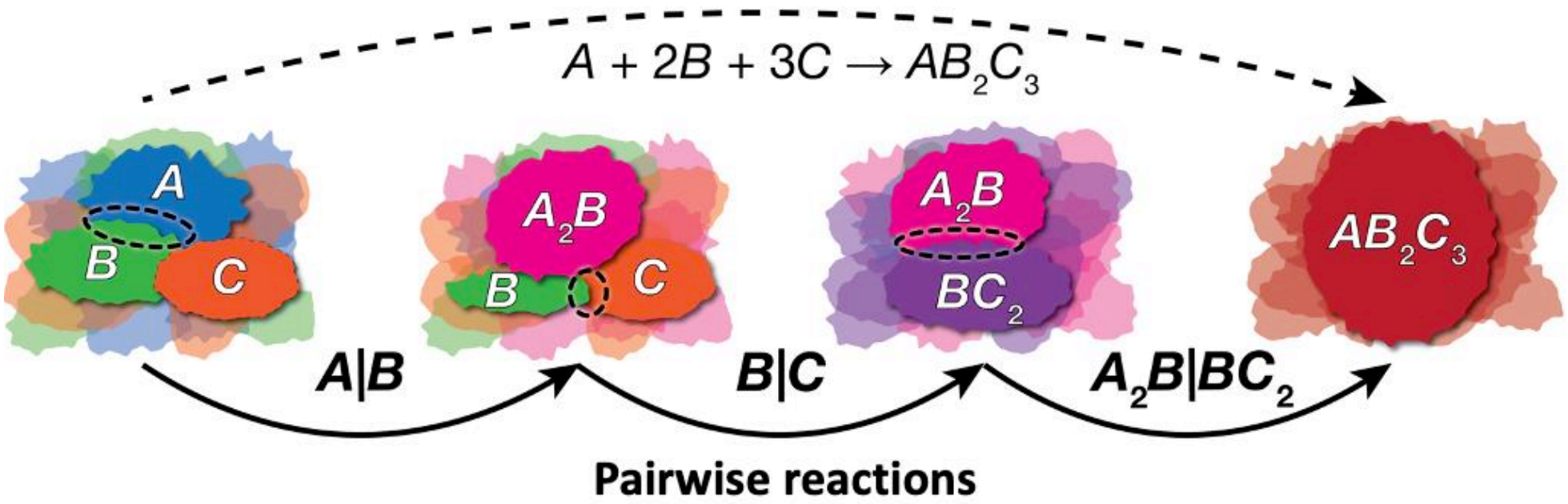
Precursors

What happens in between?

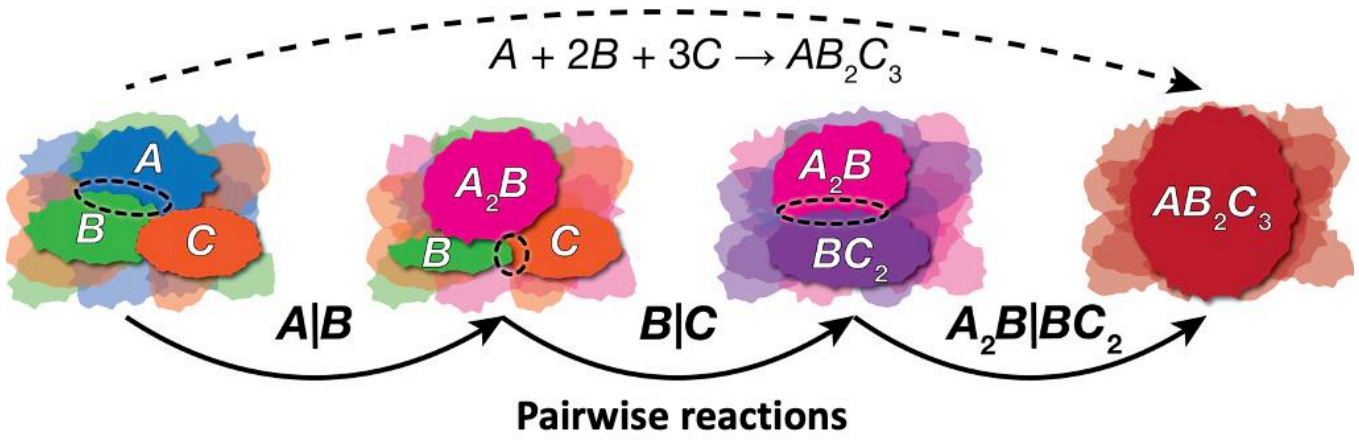
Heating

Product

Some background on synthesis science



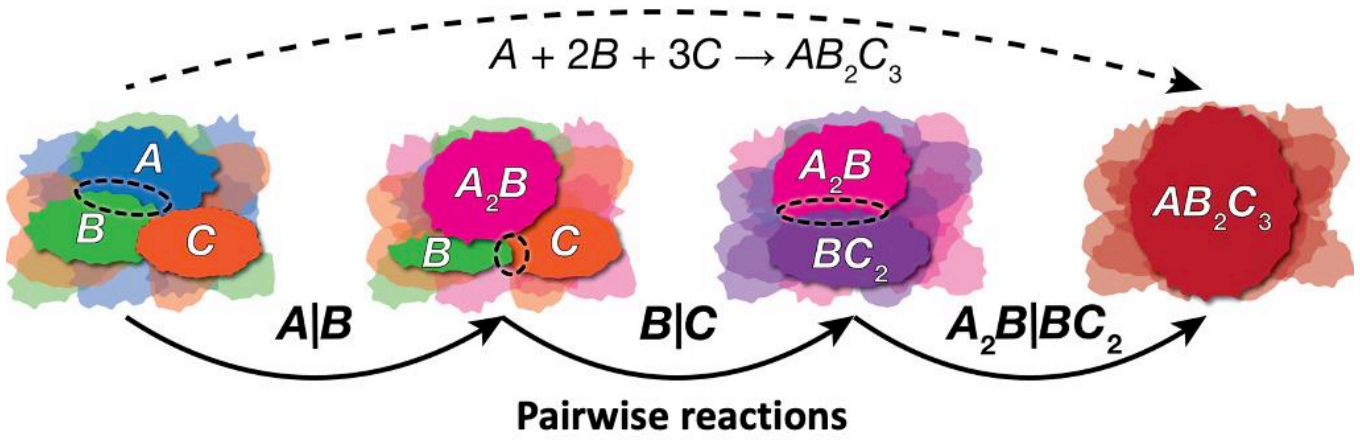
Some background on synthesis science



Change in Gibbs free energy provides a driving force for each reaction step:

$$\Delta G = \Delta H - T\Delta S$$

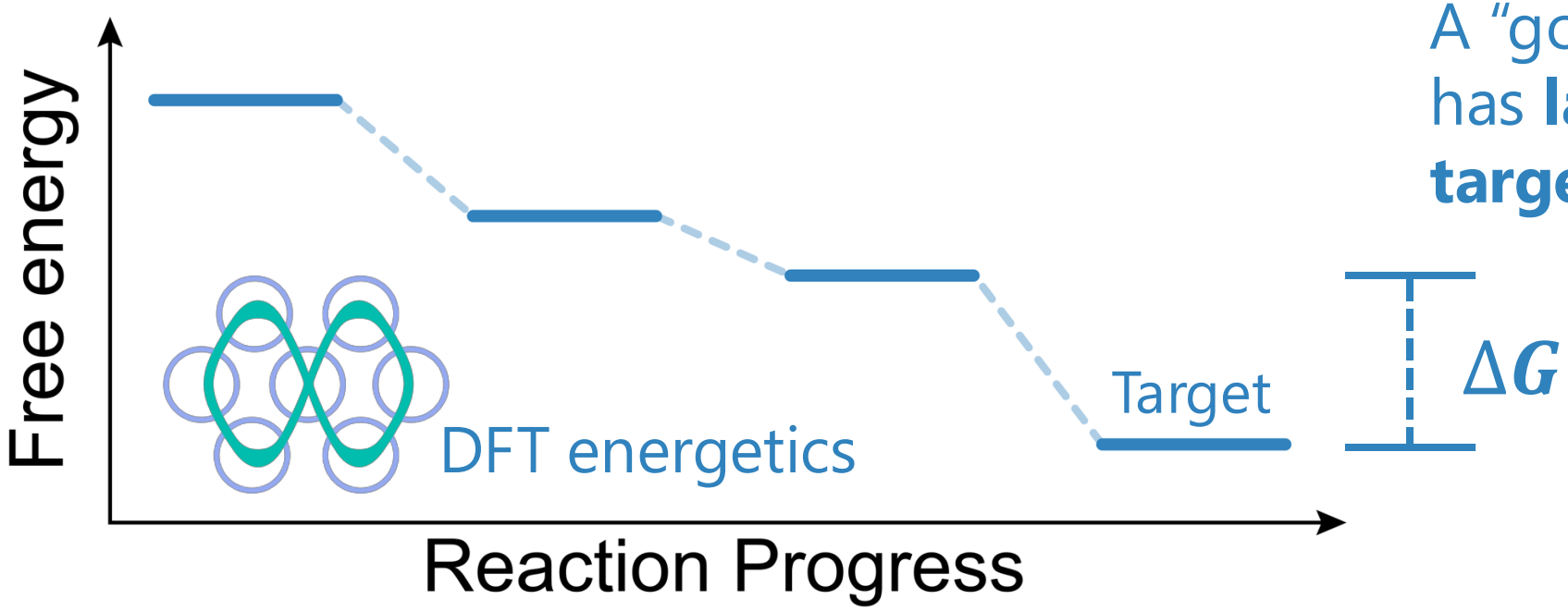
Some background on synthesis science



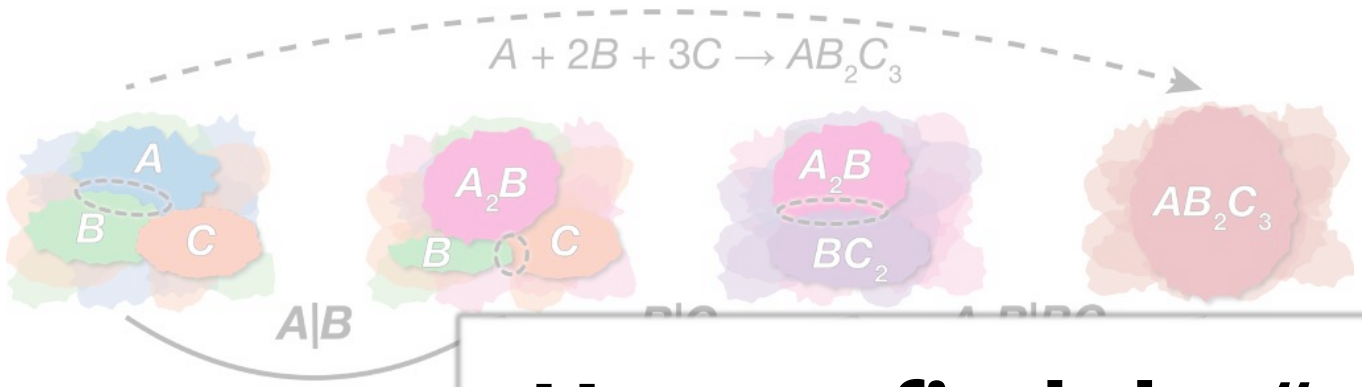
Change in Gibbs free energy provides a driving force for each reaction step:

$$\Delta G = \Delta H - T\Delta S$$

A "good" reaction pathway has **large** ΔG at the **target-forming step**



Some background on synthesis science

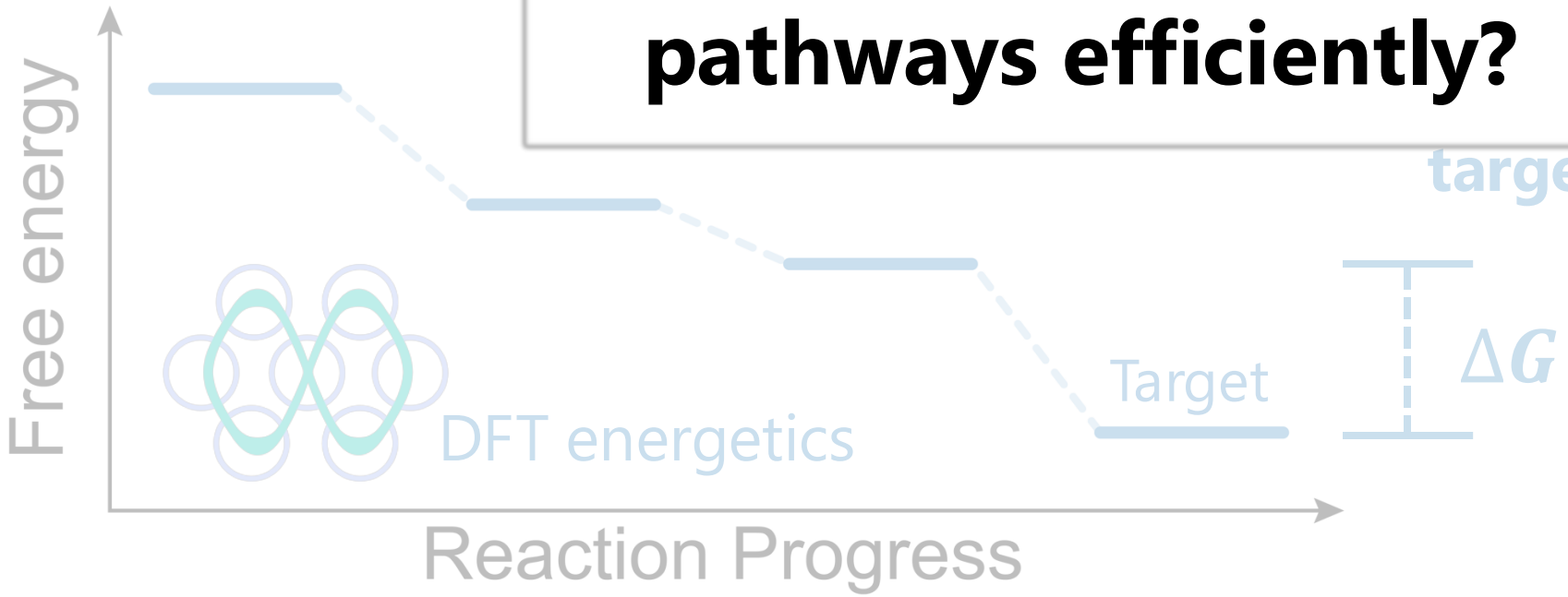


Change in Gibbs free energy provides a driving force for each reaction step:

$$\Delta G = \Delta H - T\Delta S$$

How to find the "good" pathways efficiently?

" reaction pathway
ge ΔG at the
target-forming step



ARROWS iteratively learns to prioritize max-ΔG pathways

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MATERIALS PROJECT

DFT-calculated energetics

Prioritize precursor sets with **large** ΔG

Precursors

ΔG

Target

ARROWS^3

Autonomous Reaction Route Optimization With Solid-State Synthesis

N. J. Szymanski*, P. Nevatia*, *et al.*, Nat. Commun. (2023).

ARROWS iteratively learns to prioritize max-ΔG pathways

CEMS Chemical Engineering & Materials Science

MATERIALS PROJECT

DFT-calculated energetics

Prioritize precursor sets with **large** ΔG

Precursors

ΔG

Target

ΔG is the **change in the Gibbs free energy** for the precursors to react and form the target

N. J. Szymanski*, P. Nevatia*, *et al.*, Nat. Commun. (2023).

ARROWS iteratively learns to prioritize max-ΔG pathways

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MATERIALS PROJECT

DFT-calculated energetics

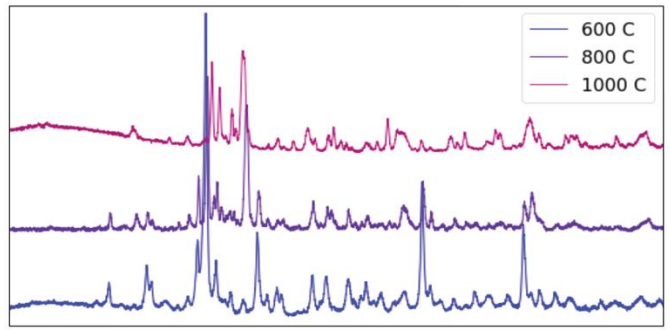
Prioritize precursor sets with **large** ΔG

Intermediates

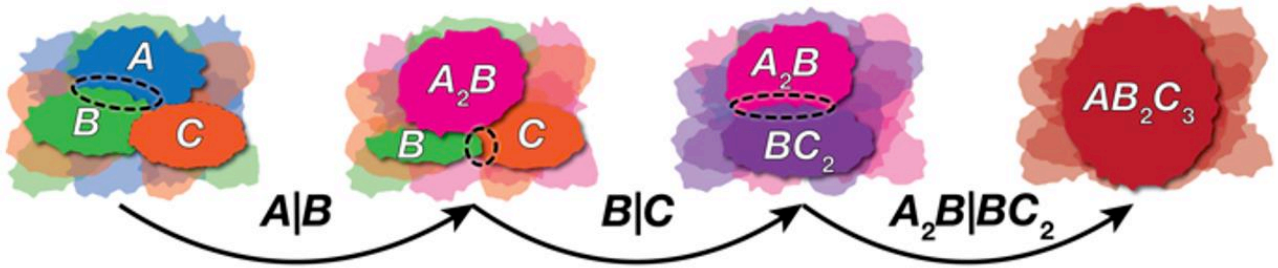
ΔG

Target

Perform experiments using suggested precursors



Learn which reactions occurred and **re-compute ΔG to form the target**



ARROWS iteratively learns to prioritize max-ΔG pathways

Prioritize precursor sets with **large** ΔG

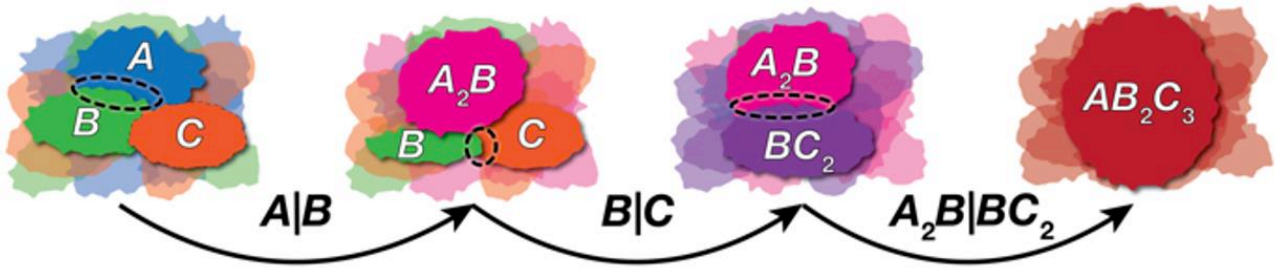
Intermediates

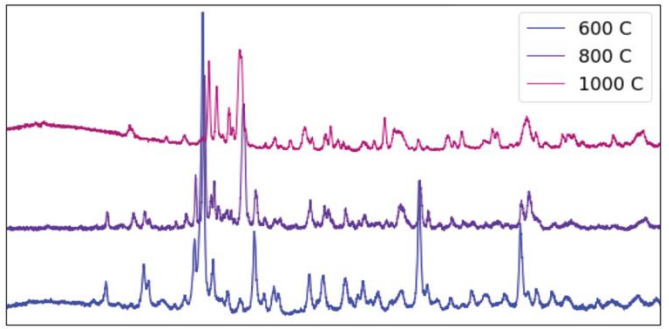
ΔG

Target

Perform experiments using suggested precursors

Predict reaction outcomes of new precursor sets





ARROWS iteratively learns to prioritize max-ΔG pathways

Avoid forming highly stable intermediates

Prioritize precursor sets that ***retain*** **large ΔG**

Intermediates

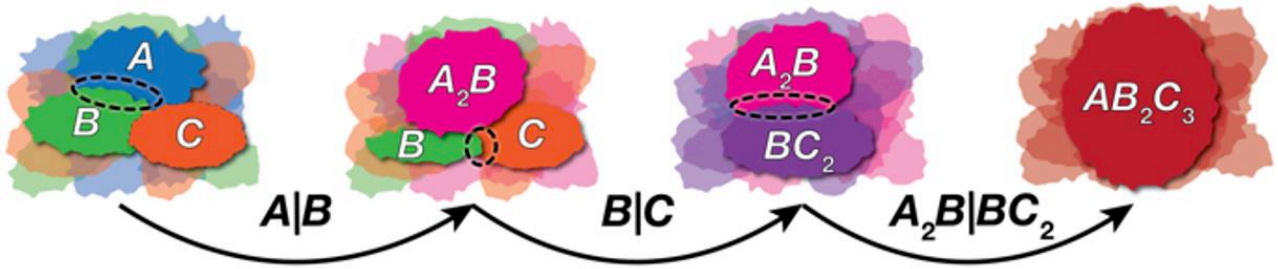
ΔG

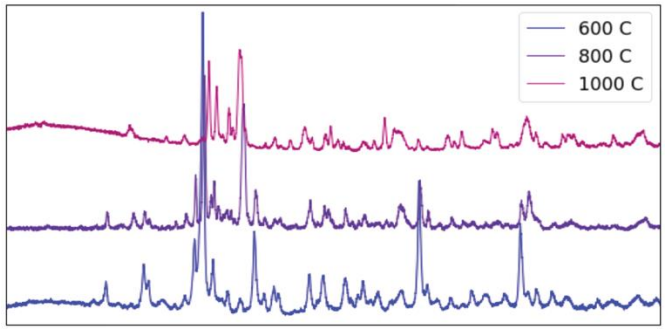
Target

Update ranking

Predict reaction outcomes of new precursor sets

Perform experiments using suggested precursors





Closing the loop…Does it work?

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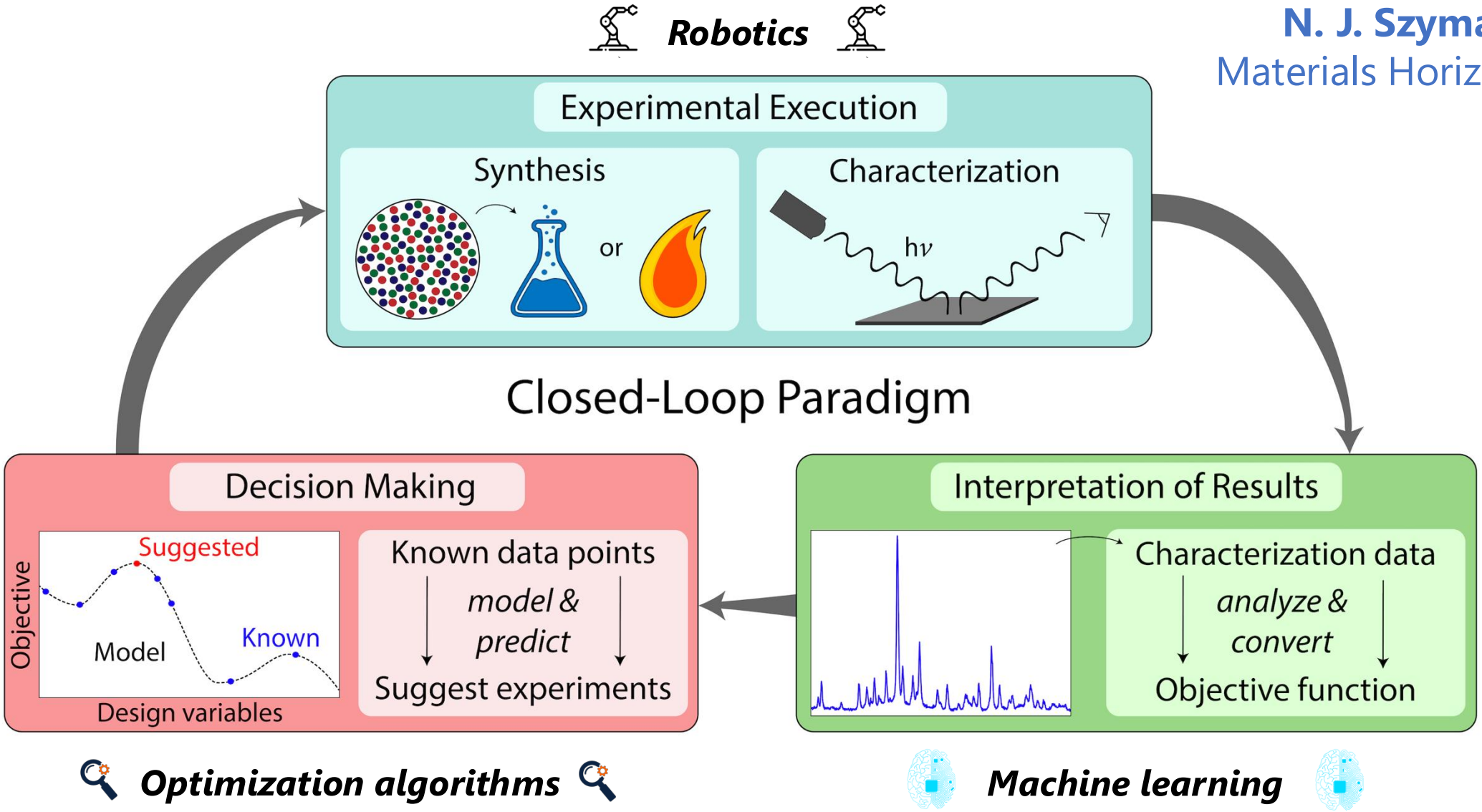
N. J. Szymanski *et al.*, Materials Horizons (2021).



Initial test case: synthesizing DFT-stable compounds

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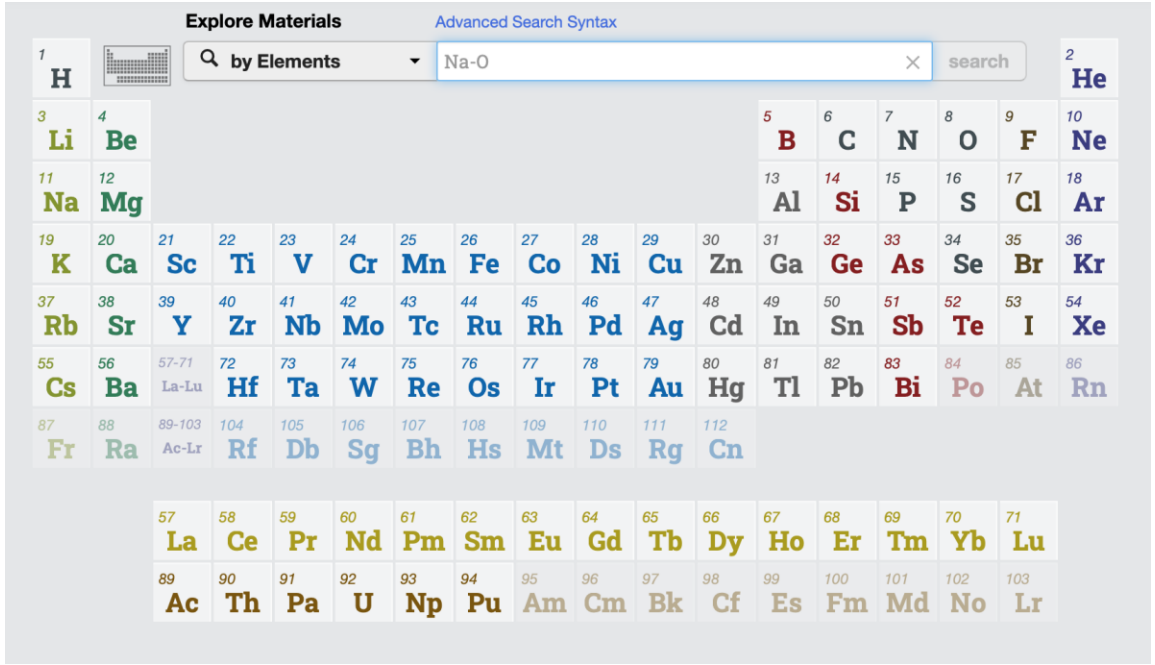
155,000 materials

We selected **58 compounds** that are stable in air

Ran **3 weeks** of targeted syntheses in the A-Lab

The Materials Project



N. J. Szymanski, B. Rendy, Y. Fei, *et al.*, Nature (2023).

Initial test case: synthesizing DFT-stable compounds

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71% success per target

37% success per recipe

A high success rate per target demonstrates the connection between DFT-calculated stability and synthesizability

A much lower success rate per recipe demonstrates how challenging synthesis can be… even for stable materials!

Initial test case: synthesizing DFT-stable compounds

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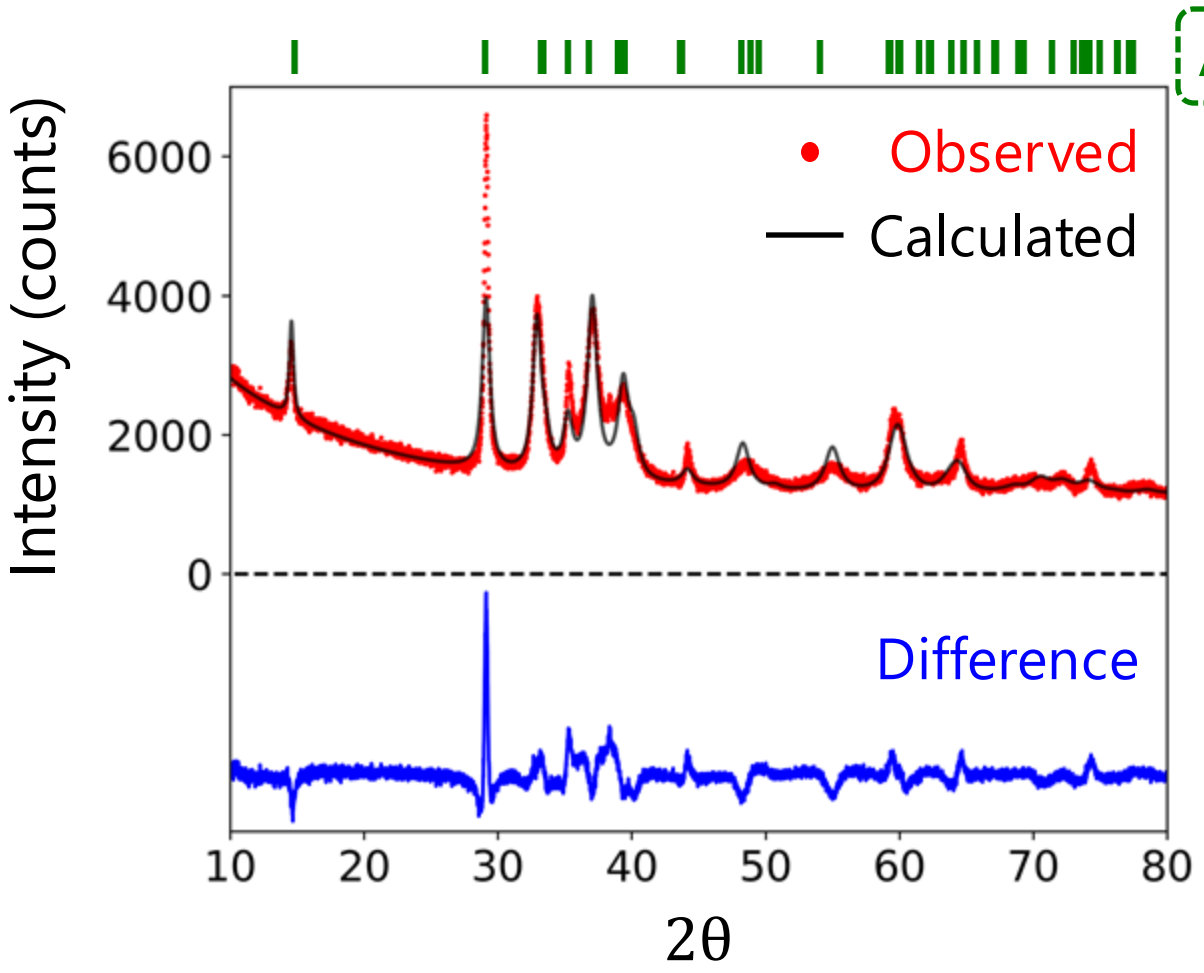
Again, does it work? Yes, but with limitations…

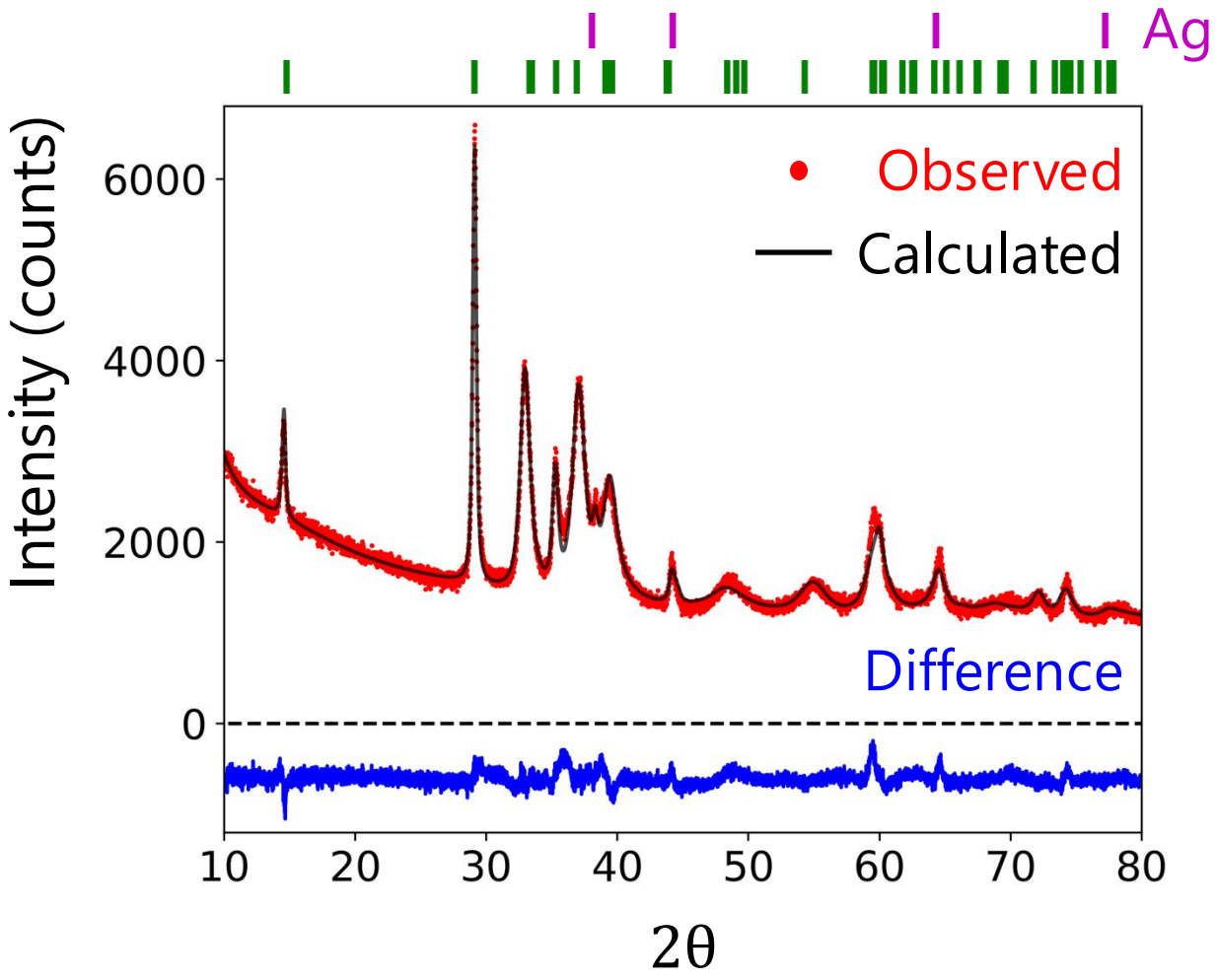


$AgMnO_2$ ⟷

Characterization remains challenging, and some of these materials contain impurities that are difficult to identify

Not a good fit!

Again, does it work? Yes, but with limitations…



Human experts to the rescue!

Manual analysis leads to a much better fit

Ag impurities are present…
More optimization needed!

In progress: incorporating additional characterization

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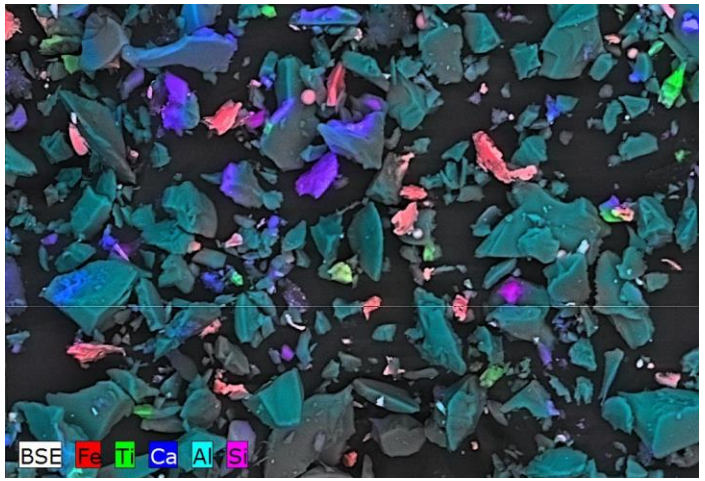
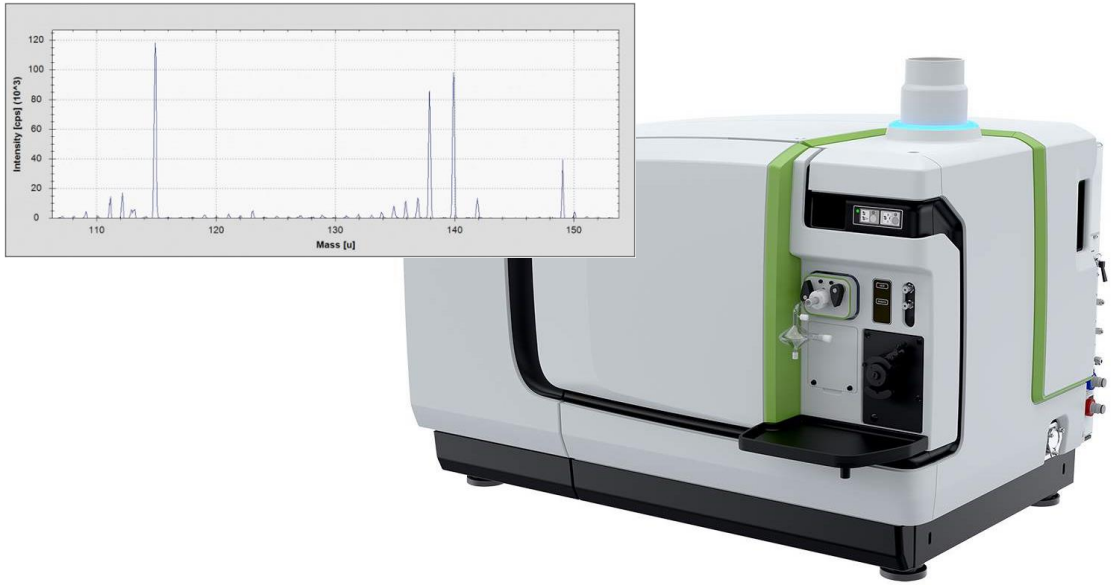
& Materials Science

In the loop: SEM/EDS

This can give information regarding **composition homogeneity**, but the results are limited in their precision

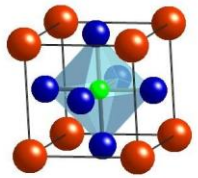




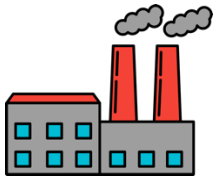
Out of the loop: ICP-MS

Provides more **precise composition,** but is also more difficult to automate

Many more opportunities for automation exist!

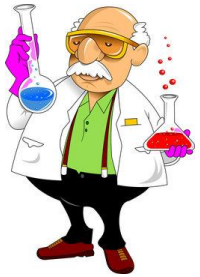
Materials design

Testing, optimization, scale-up

Stage 1 → Stage 2 → Stage 3

Synthesis and characterization



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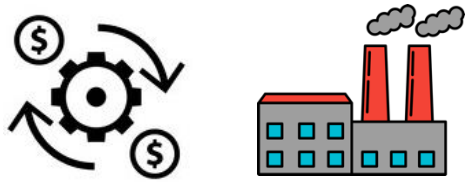


Testing, optimization, scale-up

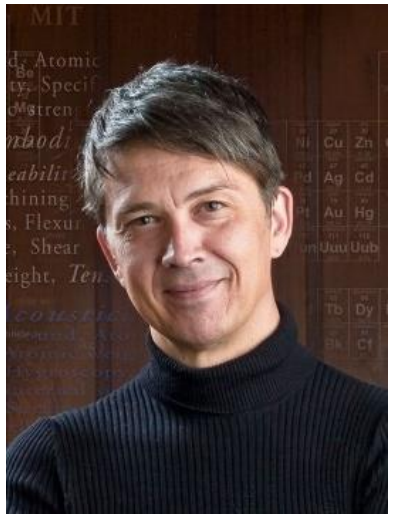
There is still quite a **disconnect between academia and industry** here

→ **Room for collaboration!** Educate us on what is important to you ☺

Acknowledgements

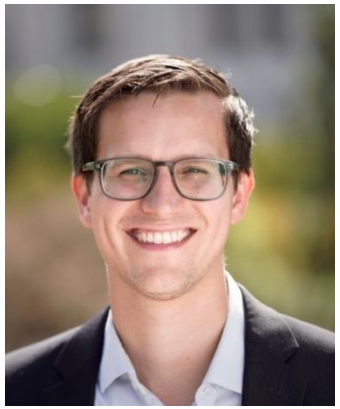
CEMS
Chemical Engineering
& Materials Science

A very large **team effort**! Some members of the "A-Team"






UCB, LBNL




Persson group, UCB

UMN


Google