Adaptively driven XRD measurements for autonomous phase identification

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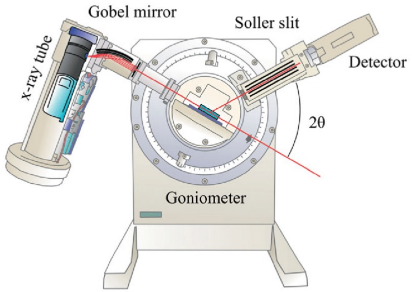
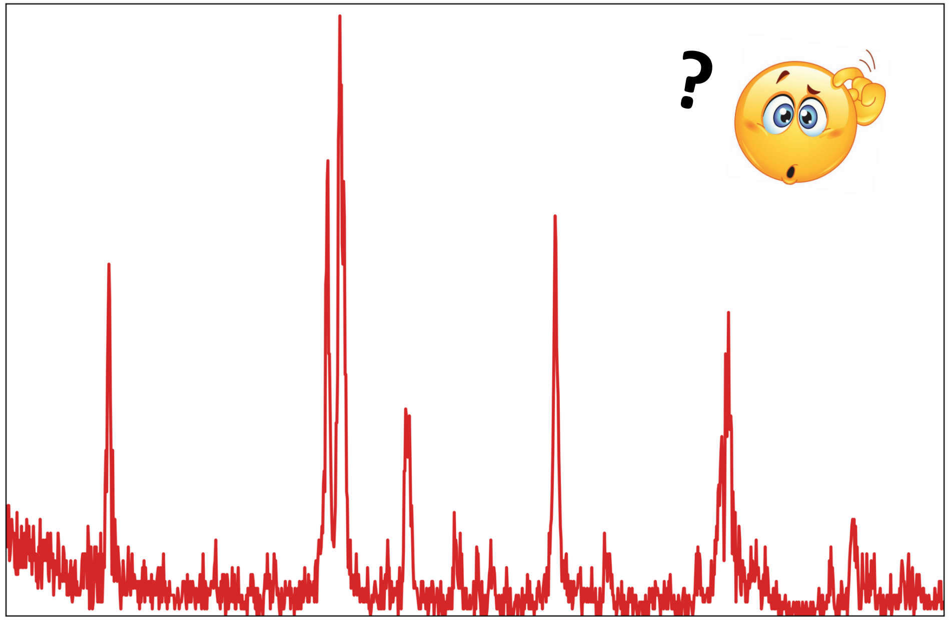
Automating XRD for *fast* and accurate phase analysis

Measurement speed tradeoff

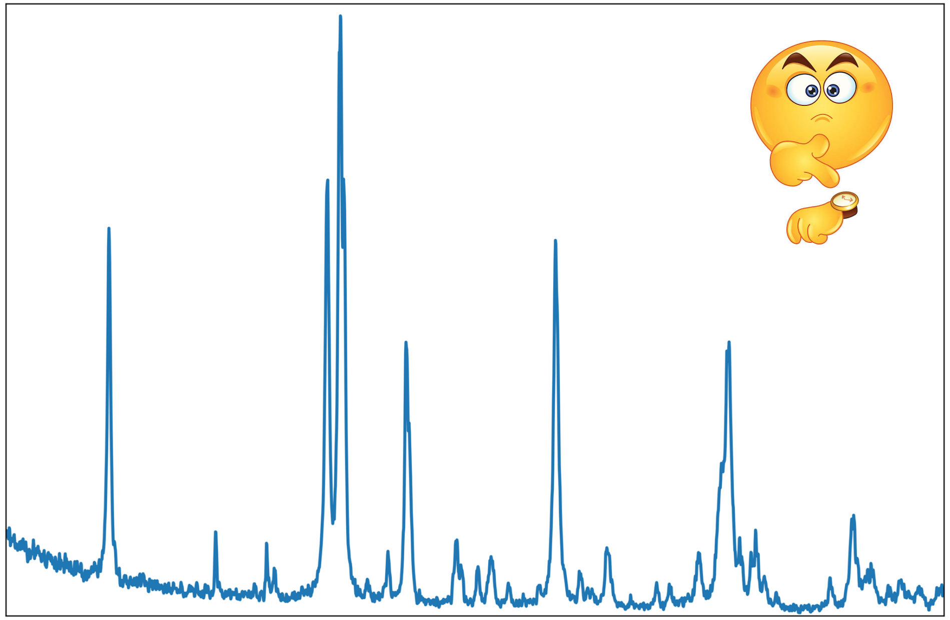
Fast XRD scan
Low resolution

Slow XRD scan
High resolution



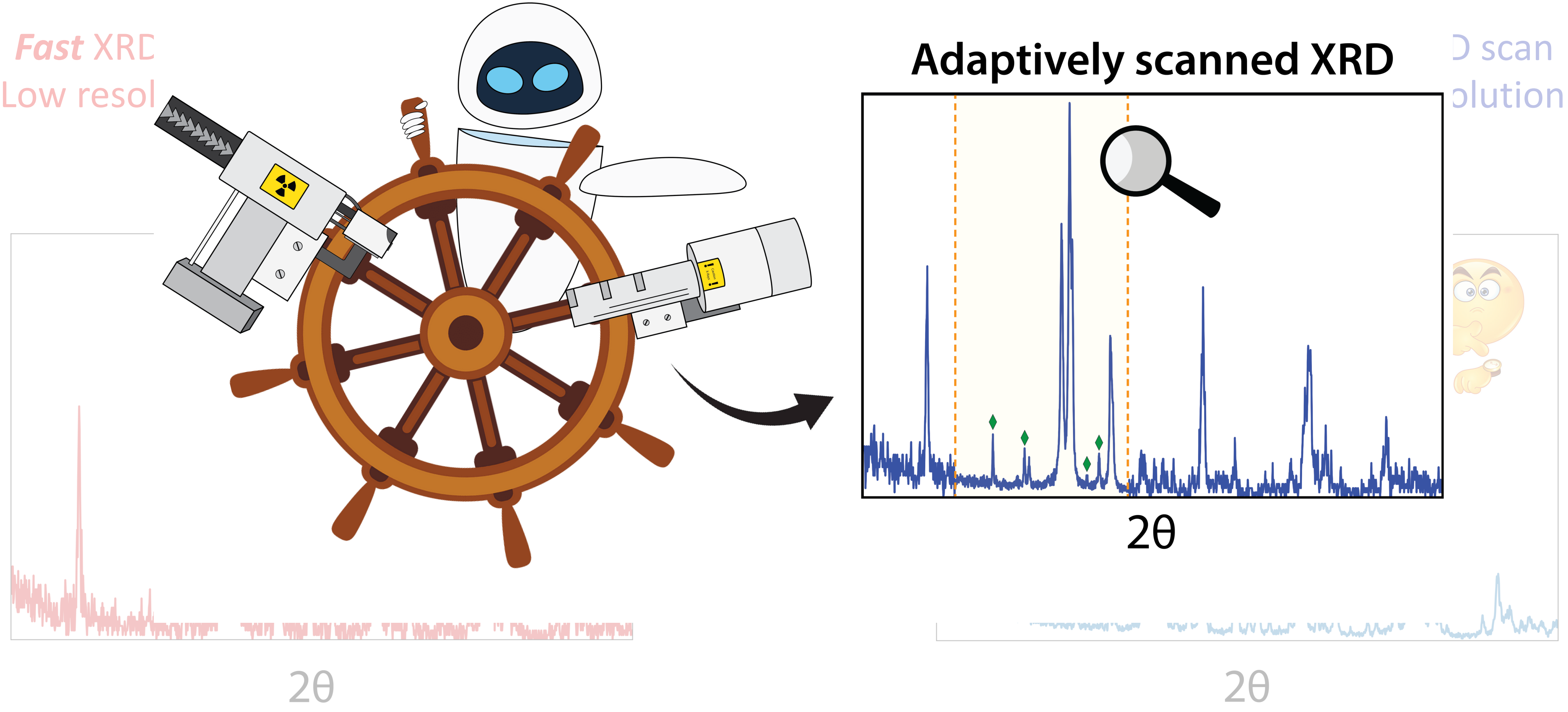


2θ



2θ

Automating XRD for *fast* and accurate phase analysis



Three components required for adaptive XRD

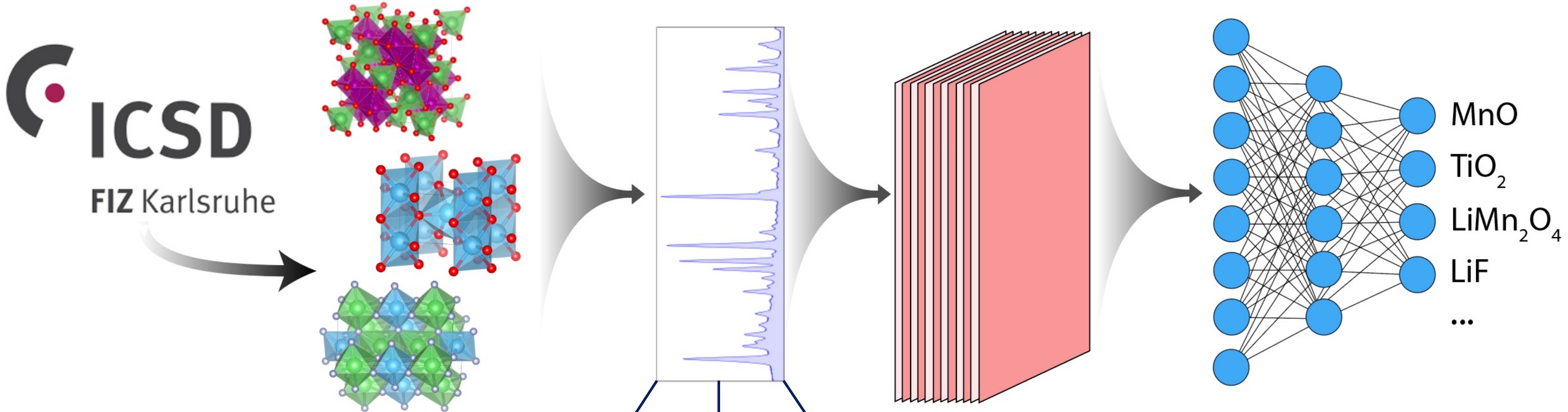
Model to perform phase identification → ***What*** phases may be present

Uncertainty quantification → ***When*** should you scan more

Feature importance analysis → ***Where*** should you scan

Component #1: phase ID with neural networks

Experimental structures | Simulated patterns | Convolutional neural network

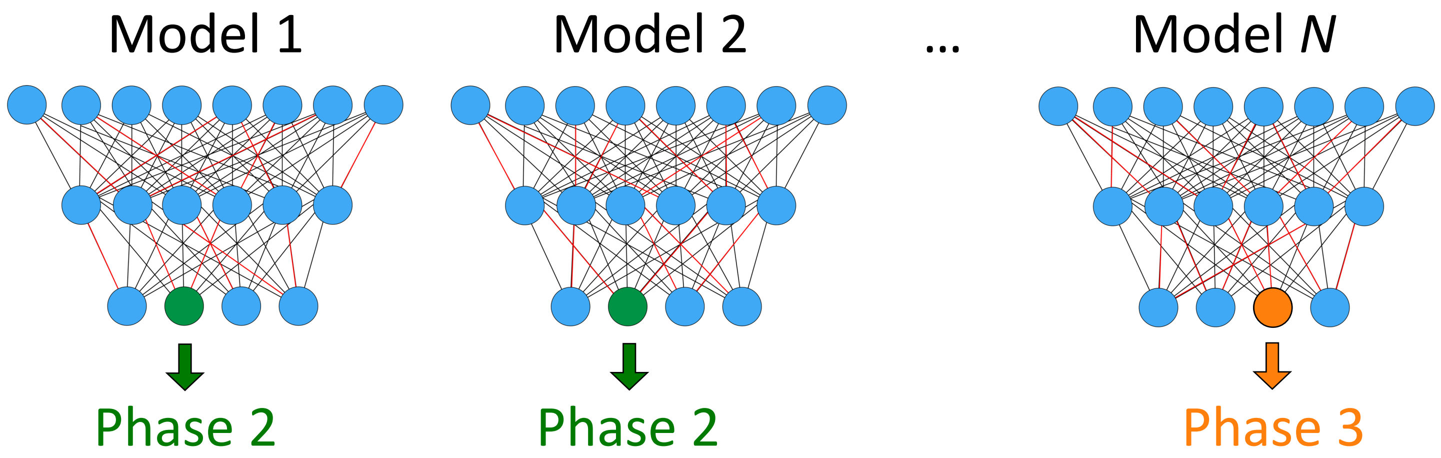


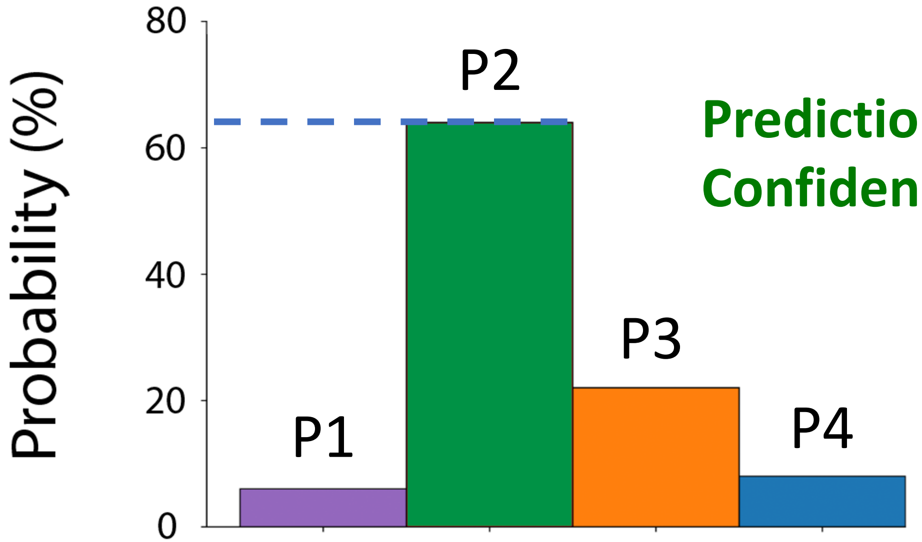
Broad peaks | Intensity changes | Shifts in 2θ

Data augmentation
from strain, texture, and poor crystallinity

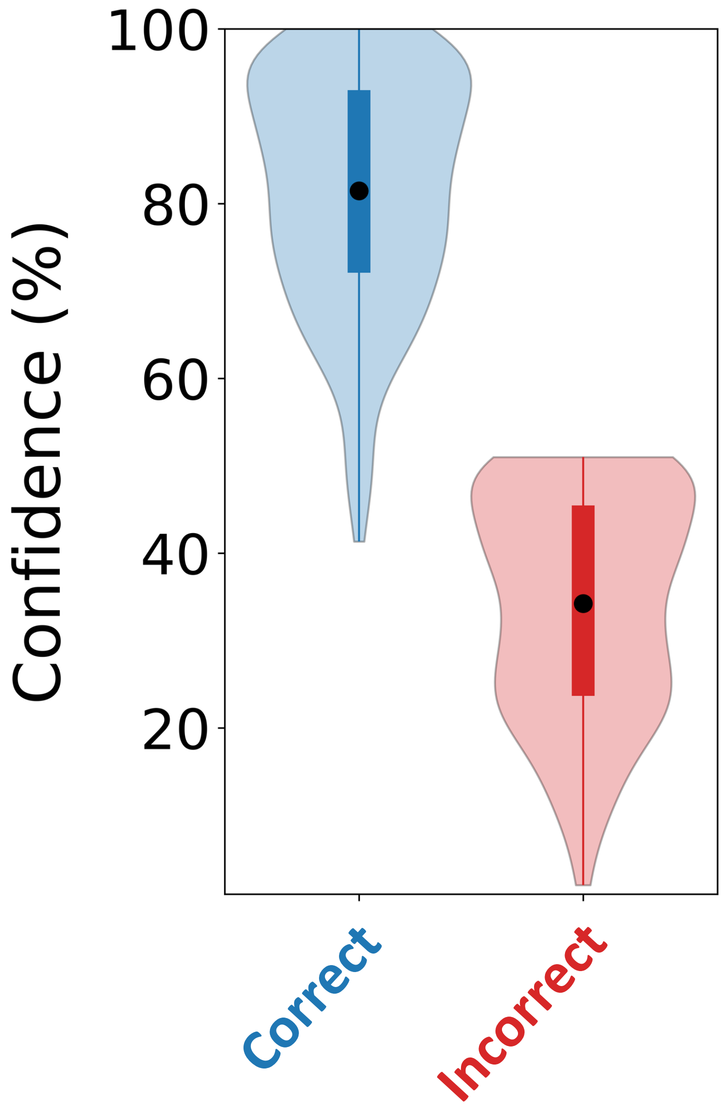
Component #2: uncertainty assessment *via* ensemble

Prediction confidence estimated using an **ensemble** approach





Prediction: Phase 2
Confidence: 65%

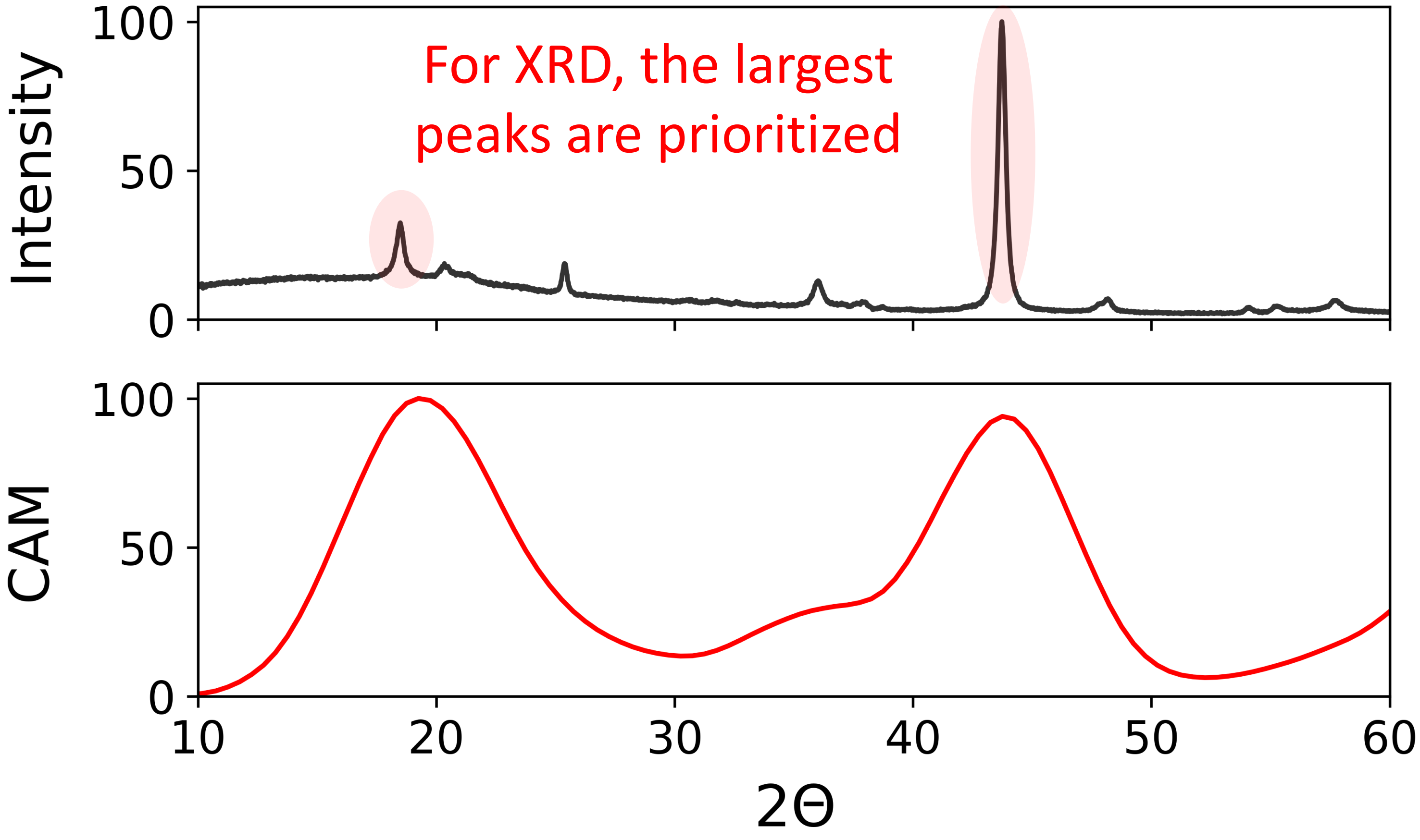


Component #3: feature analysis with CAMs

CNN predicts **dog**

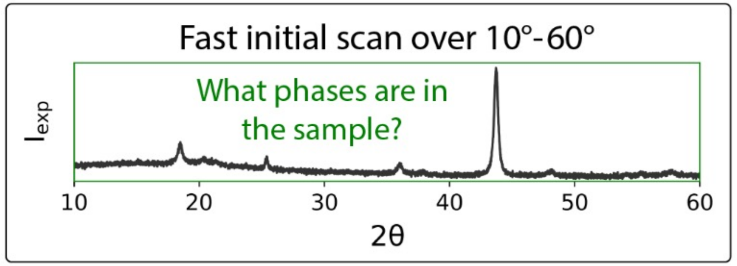
CNN predicts **cat**

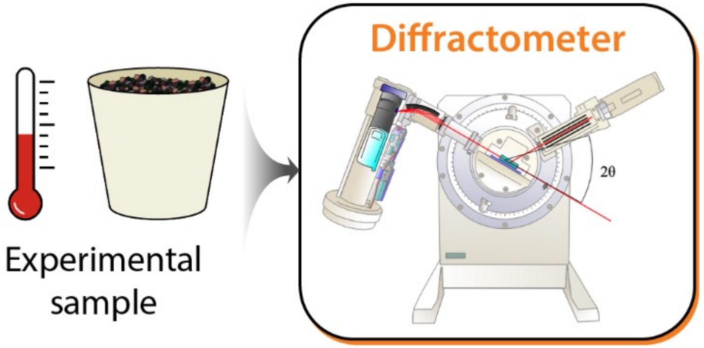
Class activation map (CAM) highlights the features that contribute most to the model's prediction



Example from Keras tutorials

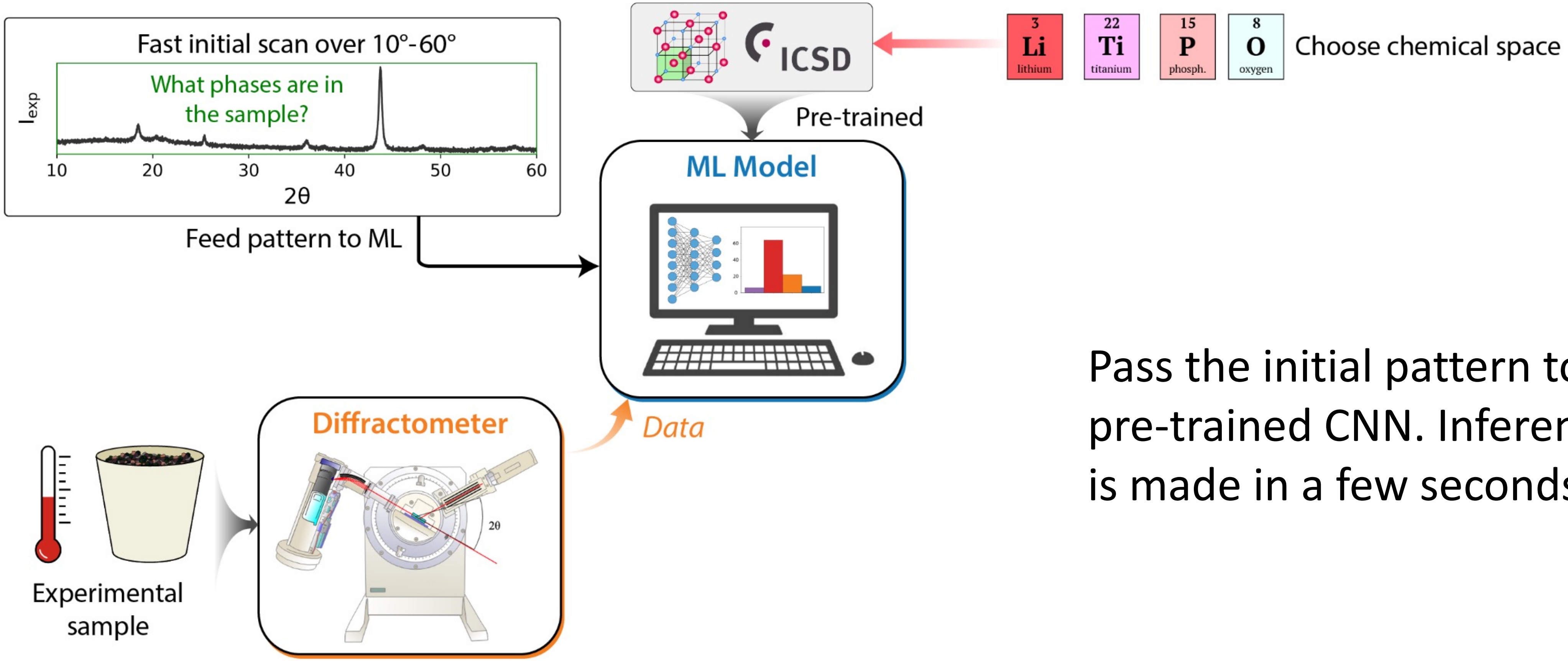
Adaptively driven XRD to maximize prediction confidence





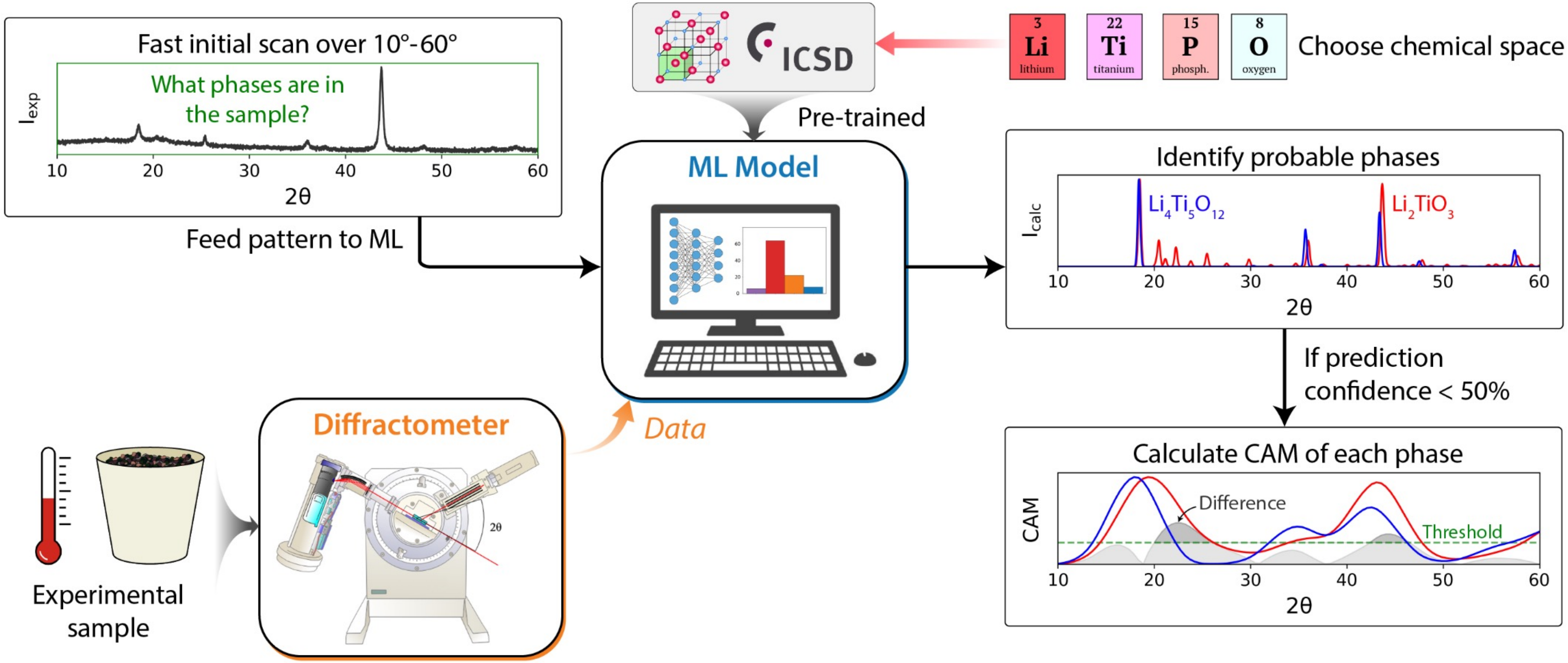
Given an unknown sample, first perform a very fast initial scan – 2 min is usually enough to make a preliminary prediction

Adaptively driven XRD to maximize prediction confidence



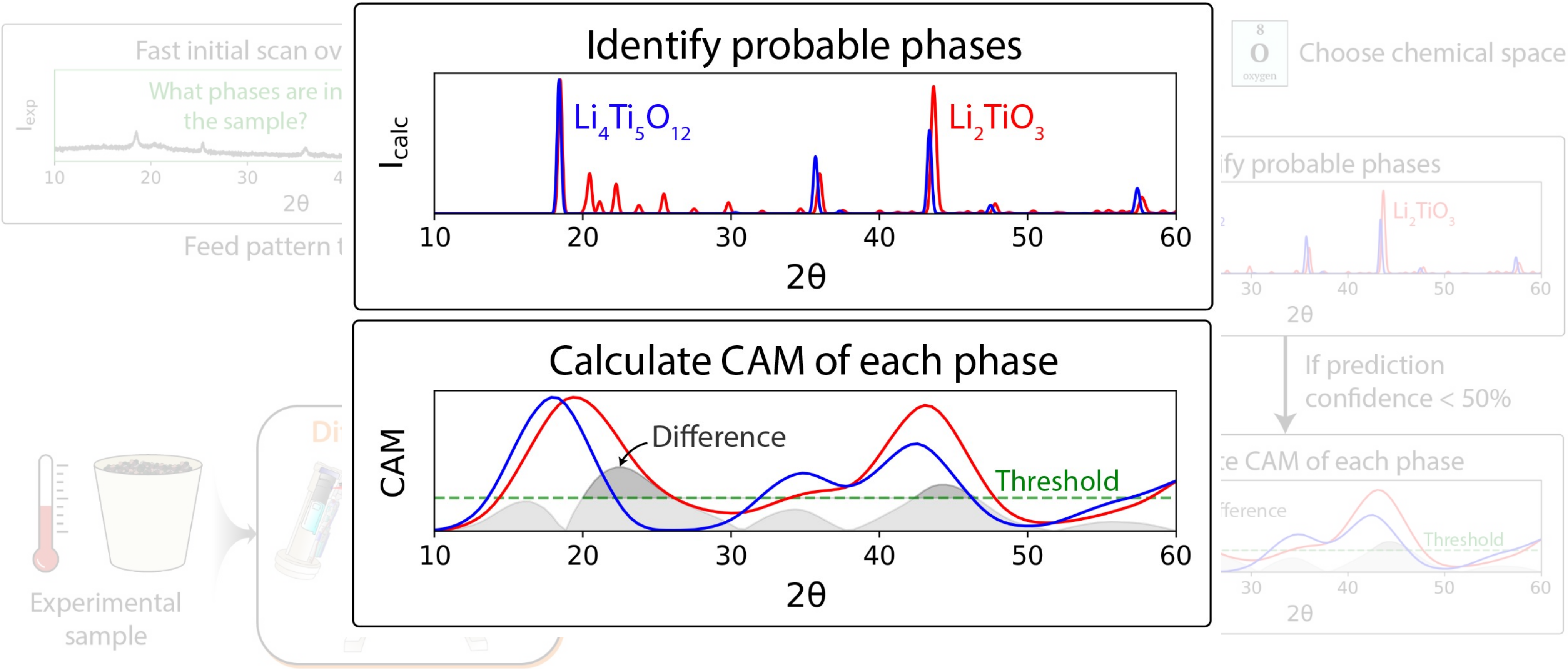
Pass the initial pattern to a pre-trained CNN. Inference is made in a few seconds.

Adaptively driven XRD to maximize prediction confidence



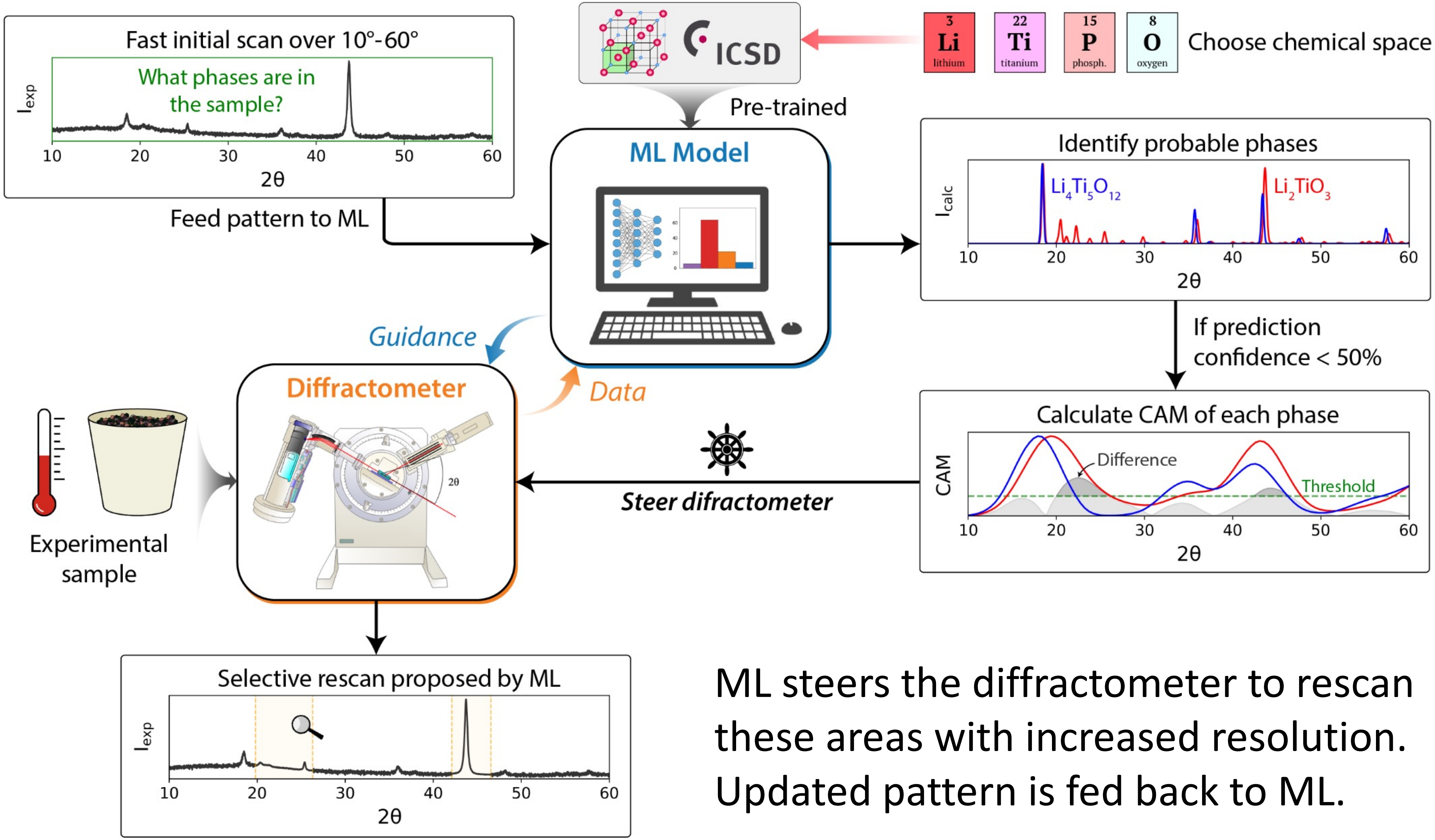
Make initial predictions and check if the model is confident.
If not, calculate the CAMs for all suspected phases.

Adaptively driven XRD to maximize prediction confidence



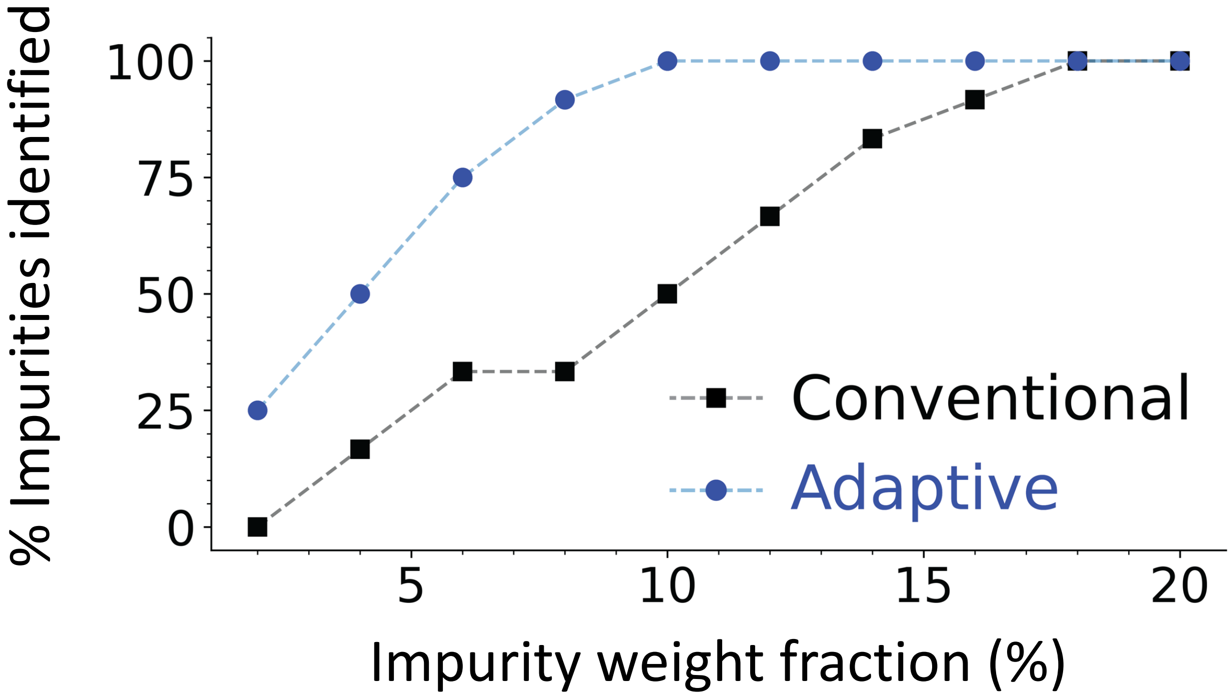
The areas with large *differences* in CAMs contain the most valuable information to distinguish these two phases.

Adaptively driven XRD to maximize prediction confidence



ML steers the diffractometer to rescan these areas with increased resolution. Updated pattern is fed back to ML.

Adaptive XRD on 240 two-phase mixtures



Adaptive XRD gives **a higher detection rate** while requiring **less scan time**

All possible combinations of:

- Li_2CO_3, LiOH, $La(OH)_3$, ZrO_2
- TiO_2, Li_2TiO_3, and Li_3PO_4

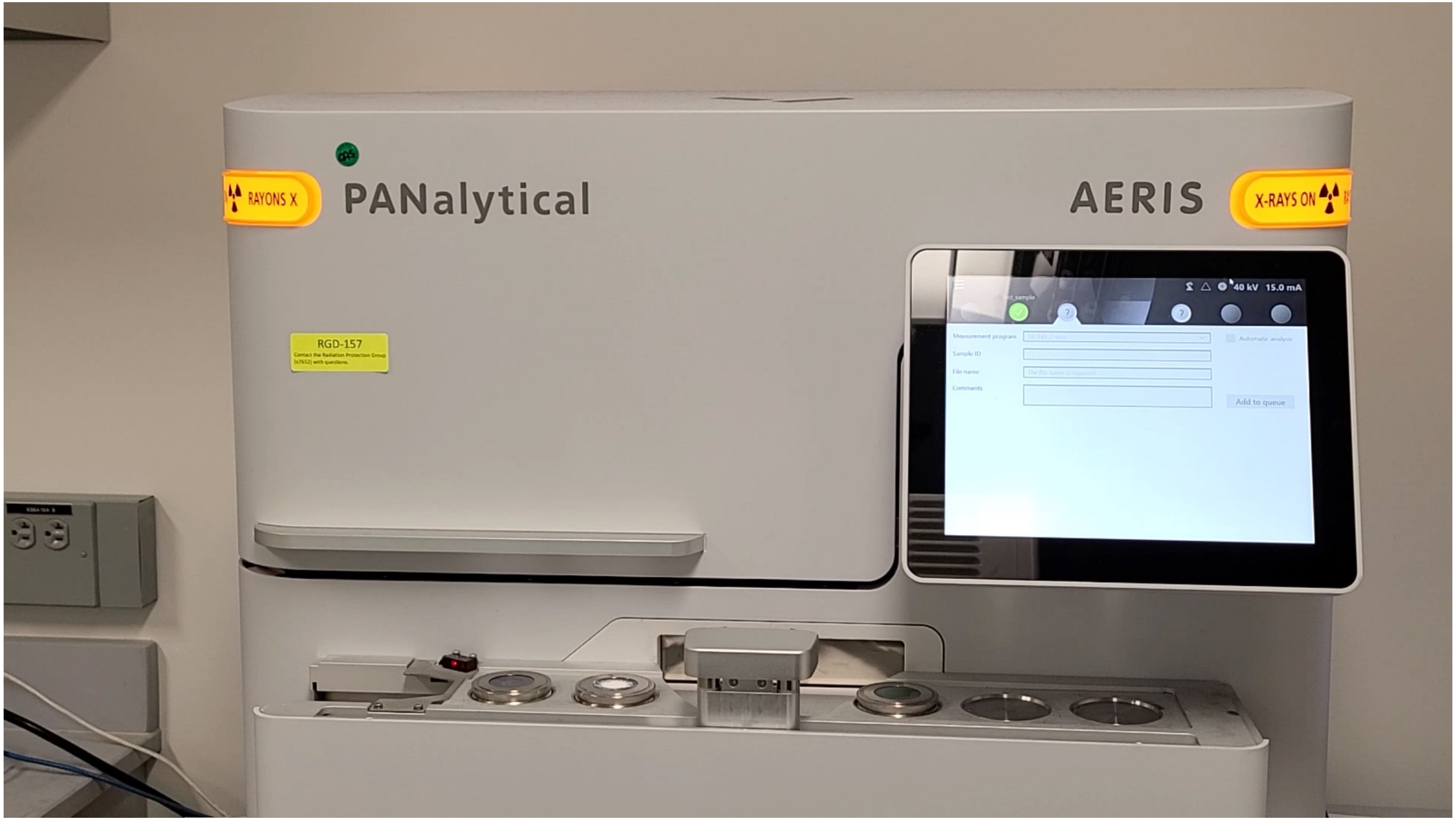
With different weight fractions:

- 2%, 4%, 6%, …, 18%, 20%

Two types of XRD measurements:

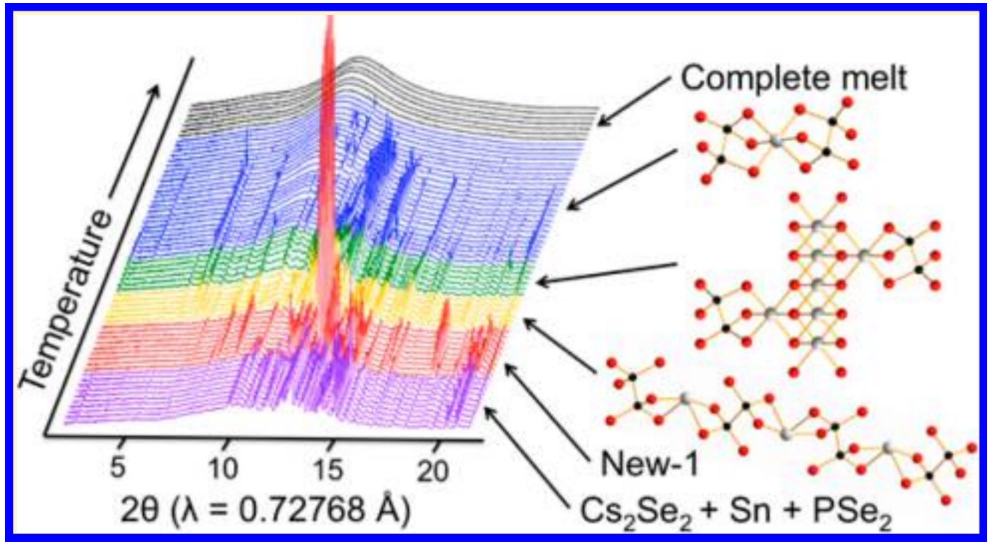
- Conventional **10 min** scan
- Adaptive scan, **6 min** on avg

Adaptive XRD demo for impurity detection



In situ XRD: where scan speed is critical

When XRD is used to monitor chemical reactions, it *must be fast* as to not miss short-lived intermediates



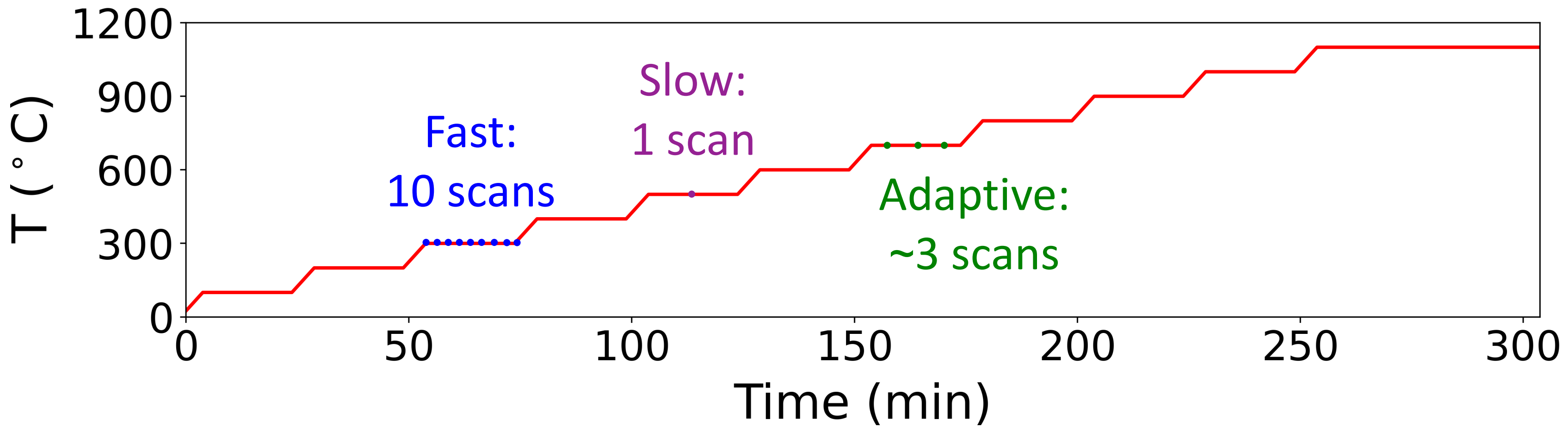
Haynes *et al.*, JACS 139, 10814-10821 (2018).

In situ XRD: where scan speed is critical

Our test case: $Li_7La_3Zr_2O_{12}$ (LLZO) synthesis

Precursors: $La(OH)_3$, Li_2CO_3, ZrO_2

Heating profile: up to 1100 ℃ with 10-minute hold every 100 ℃



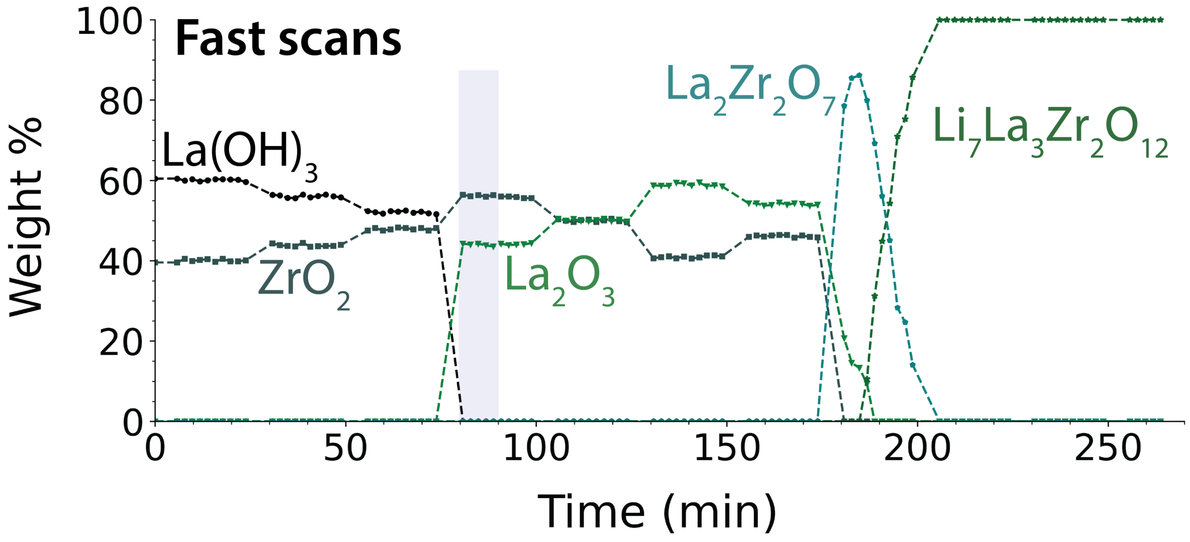
Bruker D8 Advance

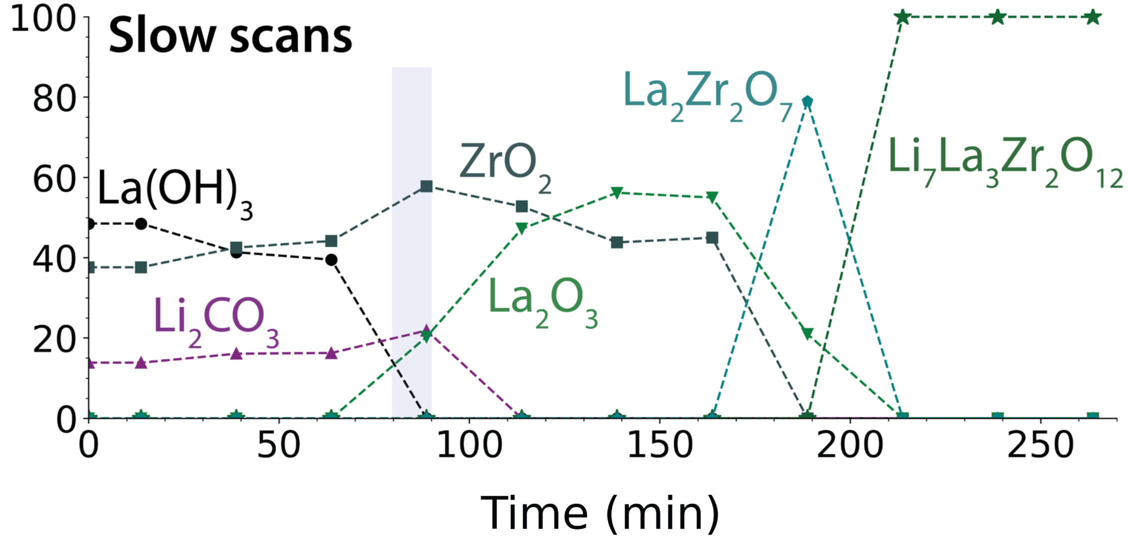
Three different procedures were tested:

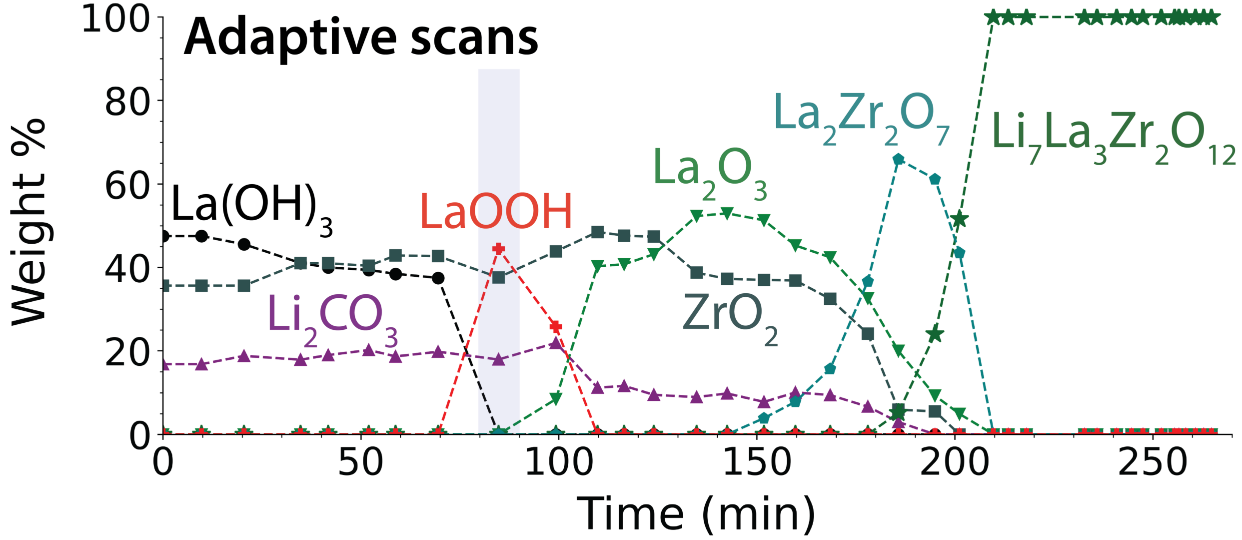
Fast scans (1 min), Slow scans (10 min), Adaptive scans (1-10 min)

Non-Adaptive

Adaptive scans enable rapid *in situ* phase detection

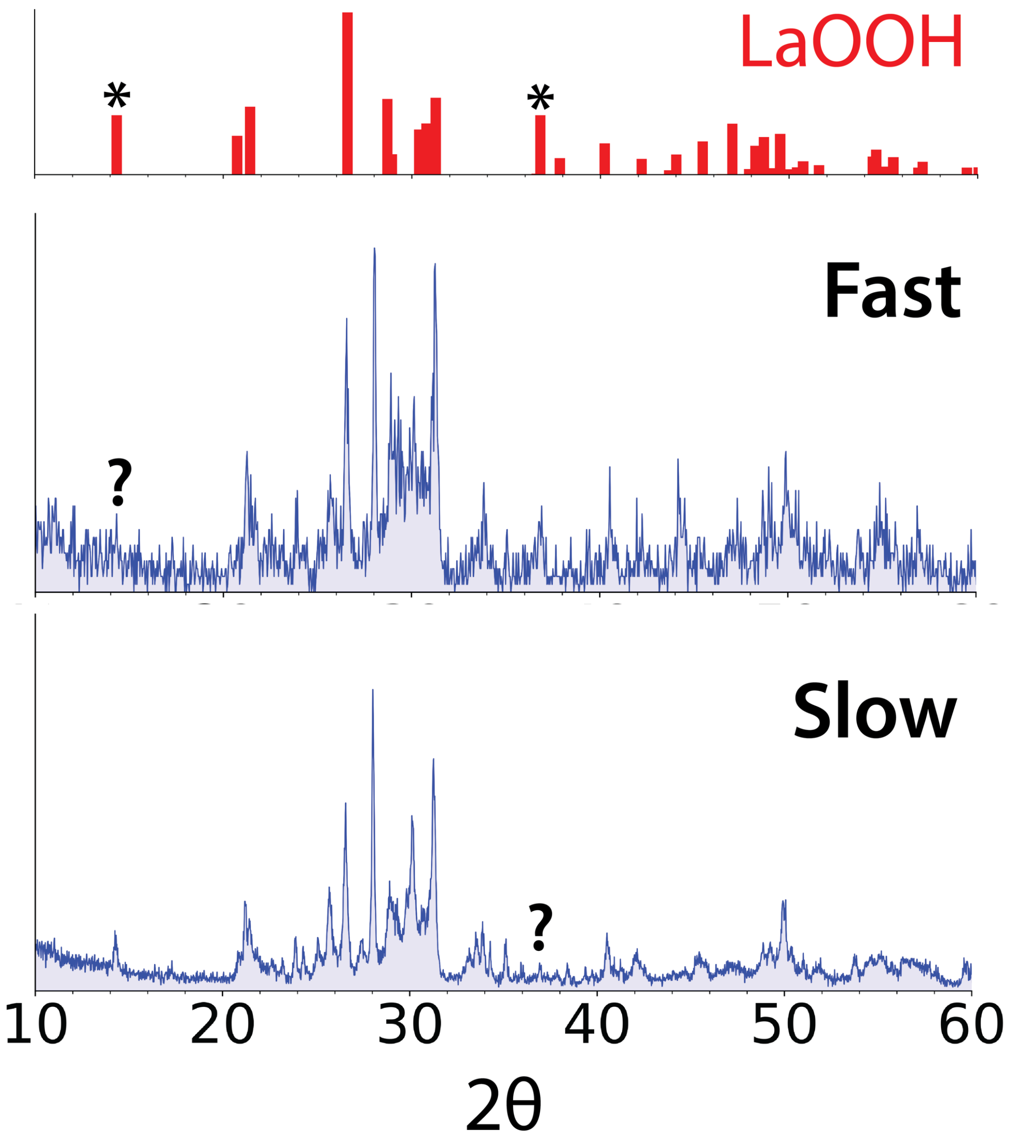






LaOOH is detected only by adaptive XRD scans

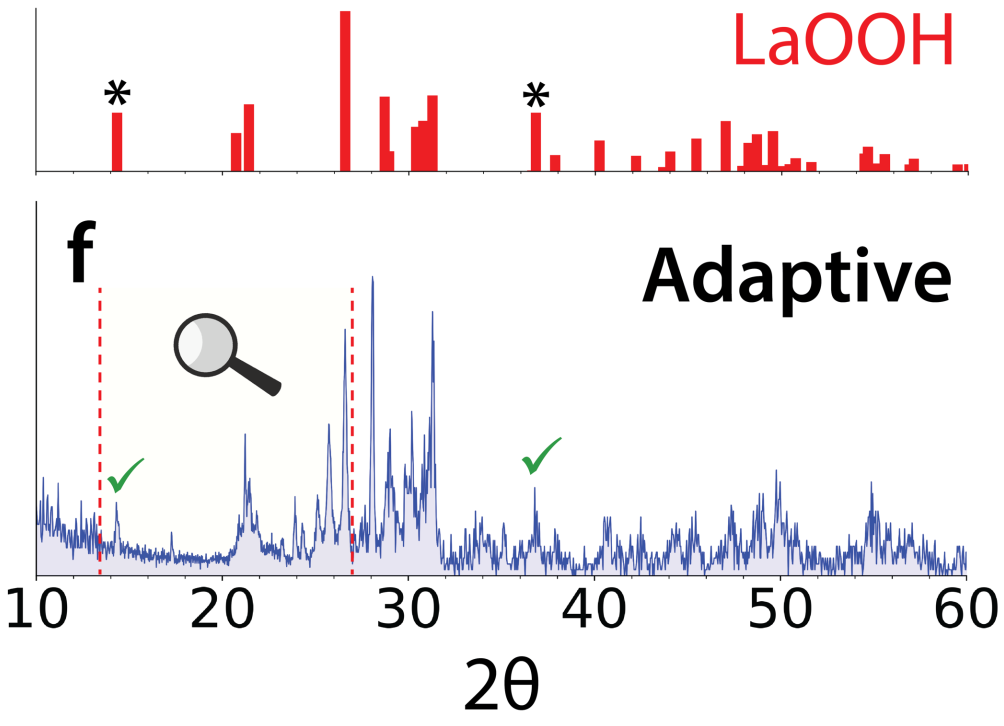
Adaptive scans enable rapid *in situ* phase detection



When the scan is too **fast**, measurement **noise** clouds the detection of smaller peaks

When the scan is too **slow**, phases transform during the measurement → **peaks disappear**

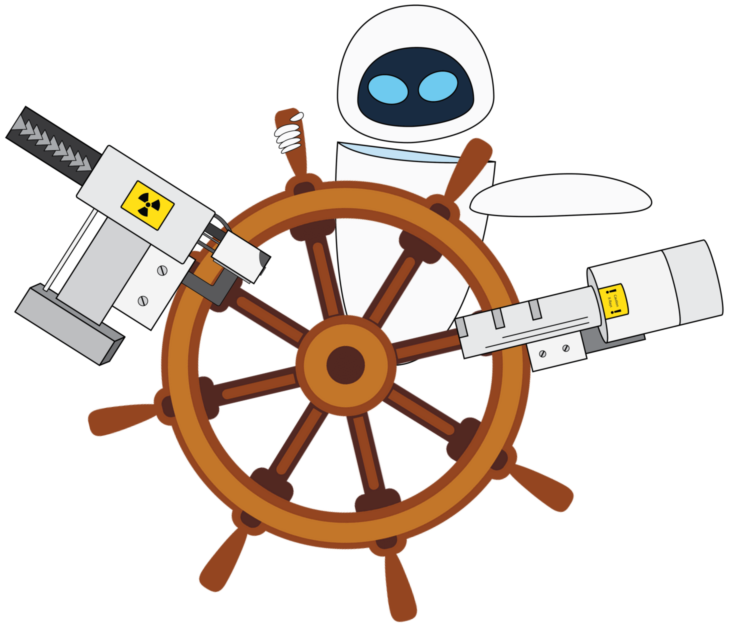
Adaptive scans enable rapid *in situ* phase detection



Adaptive XRD reduces noise where needed while maintaining fast scan time

→ All peaks are detected

Conclusion



- Because ML is fast, it can be integrated directly with experiments for on-the-fly analysis and feedback → **autonomous steering**

- Adaptive XRD proves this concept as it reduces the necessary scan time, enabling *in situ* XRD on a standard in-lab diffractometer

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Adaptively driven X-ray diffraction guided by machine learning for autonomous phase identification

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Details in publication

Code is available for use @ https://github.com/njszym/AdaptiveXRD

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