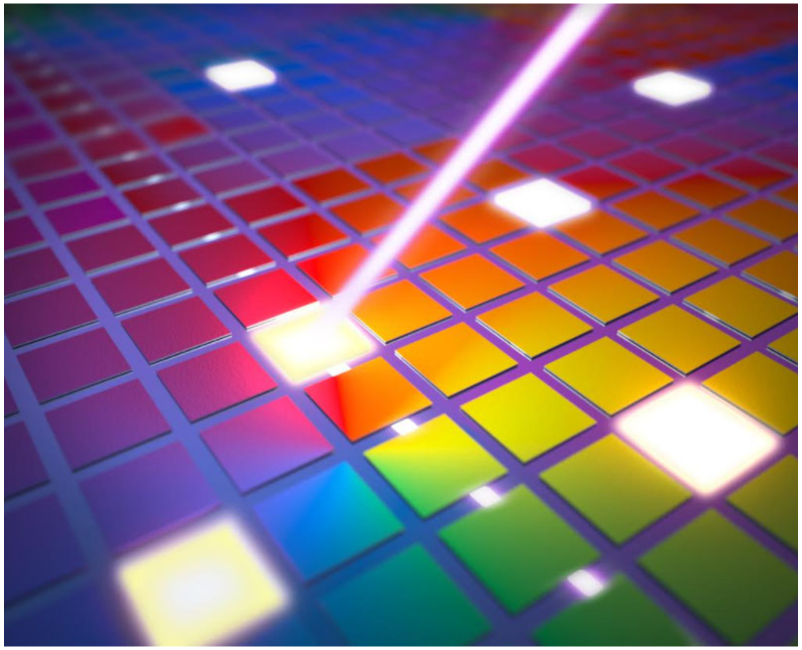
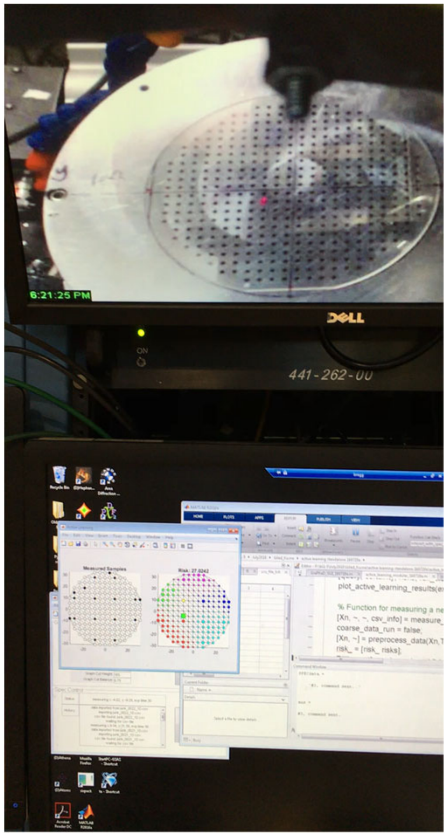
Real-time autonomous combinatorial experimentation: from atomic layer deposition to metal additive manufacturing



Ichiro Takeuchi
University of Maryland

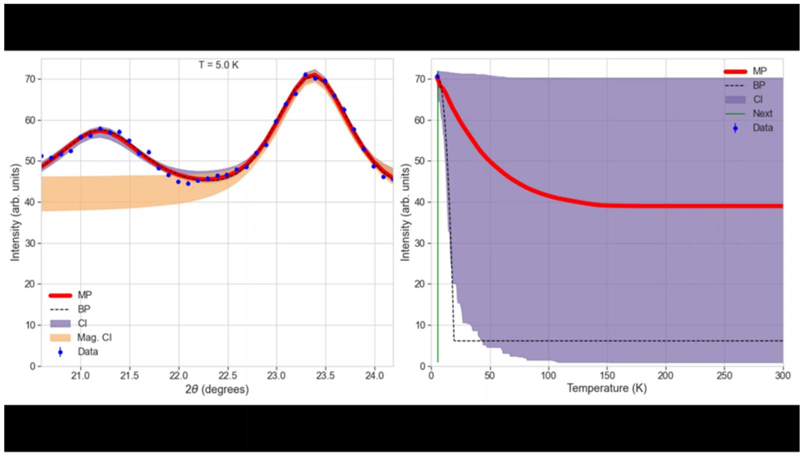
UNIVERSITY OF MARYLAND 18 56

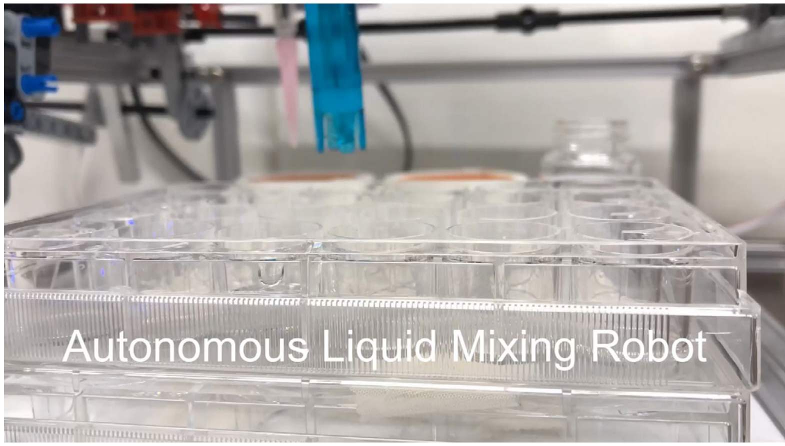
Live autonomous closed-loop materials science
w/ G. Kusne, A. McDannald (NIST)



CAMEO: Closed-loop autonomous materials exploration and optimization
Nature Communications 11, 5966 (2020)

LEGOLAS: LEGO based Low-cost Autonomous System for Education
MRS Bulletin **47**, 881 (2022)





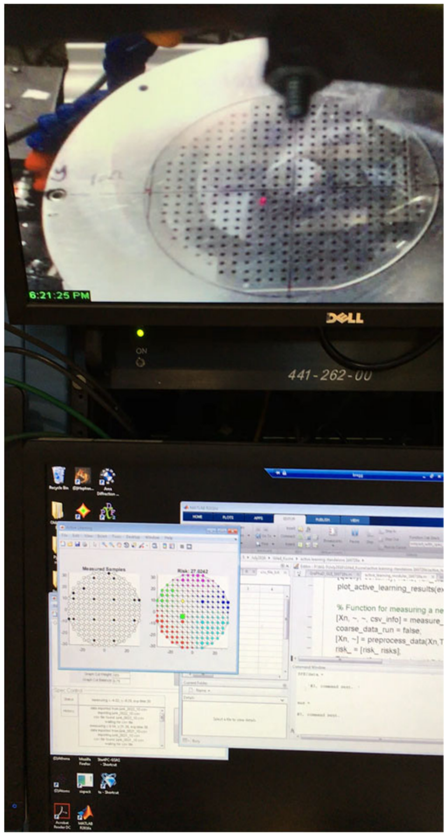
Autonomous neutron diffraction explorer (ANDiE)
Applied Physics Reviews 9, 021408 (2022)

SLAC

NIST
National Institute of Standards and Technology
U.S. Department of Commerce

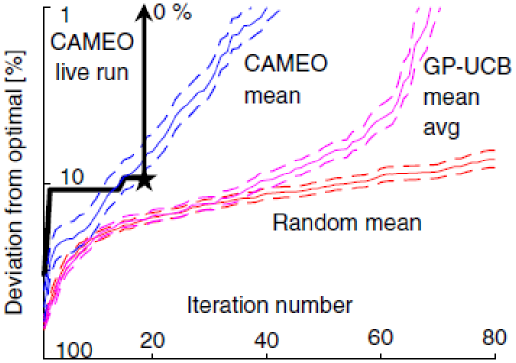
OAK RIDGE National Laboratory

Autonomous combinatorial experimentation: materials discovery via Bayesian active learning



Remote diffraction at SLAC

Nature Communications 11, 5966 (2020)



CAMEO: Closed-loop autonomous materials exploration and optimization

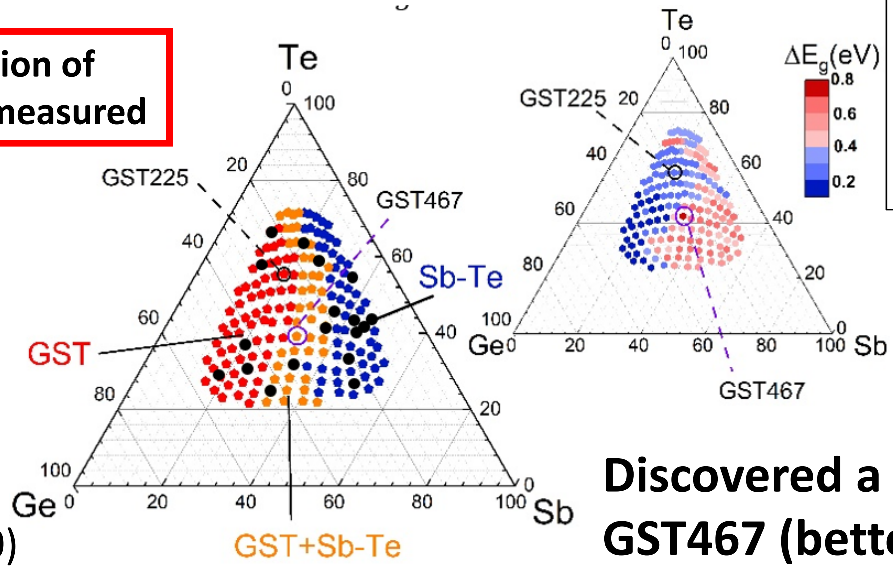
Performs phase map and property optimization together

Patent sponsored by SRC

Reproduced in multiple labs including industry

Wu et al., Nat. Commun. 15, 13 (2024)

Only a fraction of points are measured

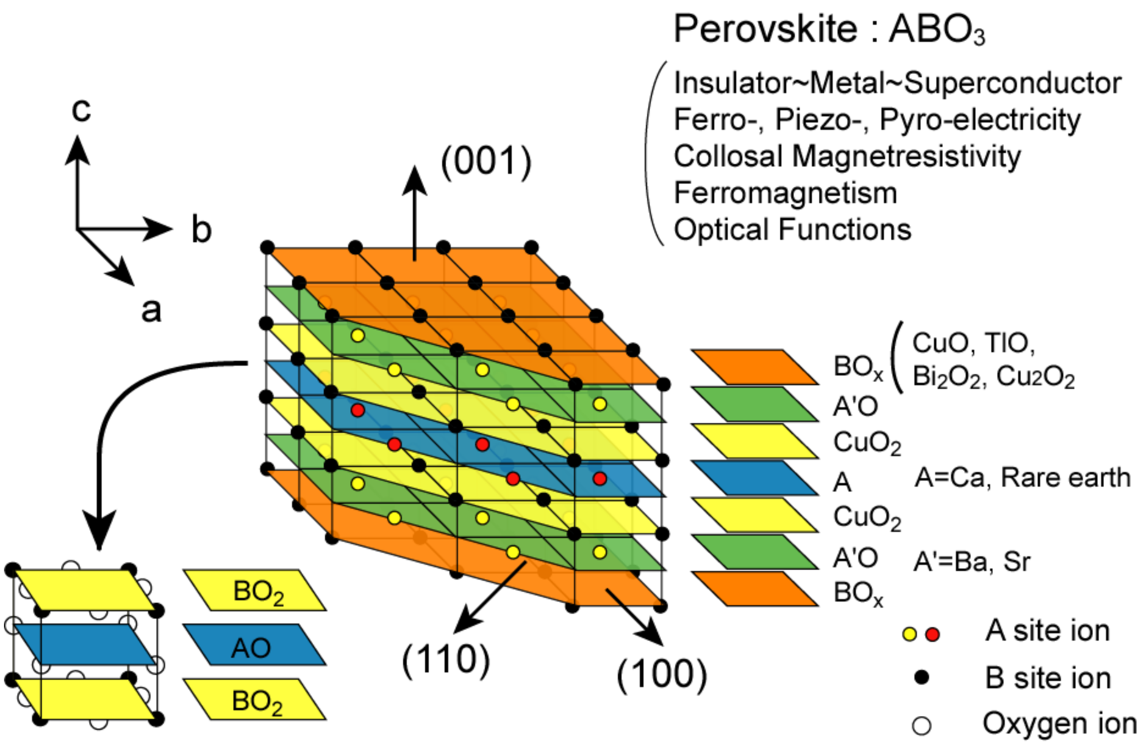


Discovered a new PCM material, GST467 (better than GST225)

Autonomous atomic-layer synthesis

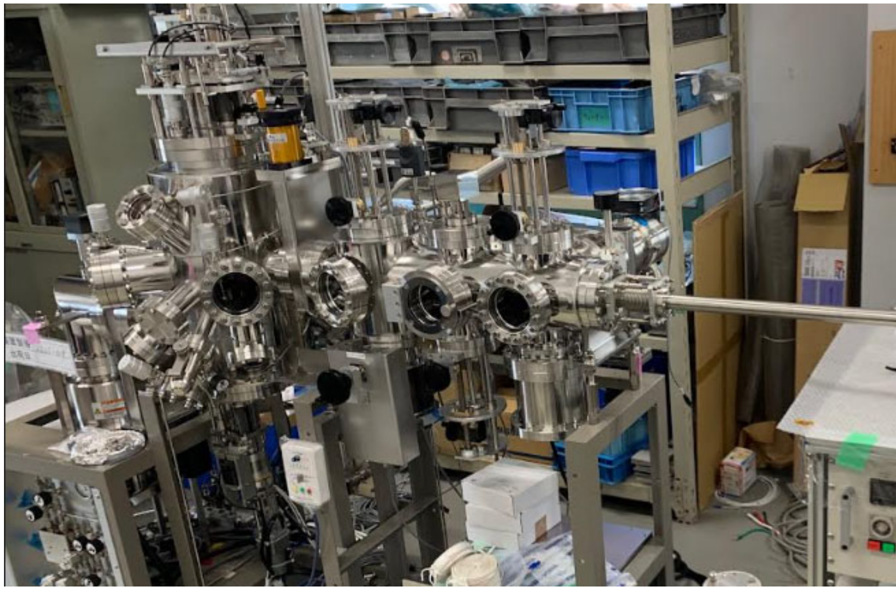
w/ Haotong Liang (UMD), Yunlong Sun (U. Tokyo), Mikk Lippmaa (U. Tokyo)

Pulsed laser deposition for creating new materials and tune properties by controlling atomic arrangement at unit-cell level



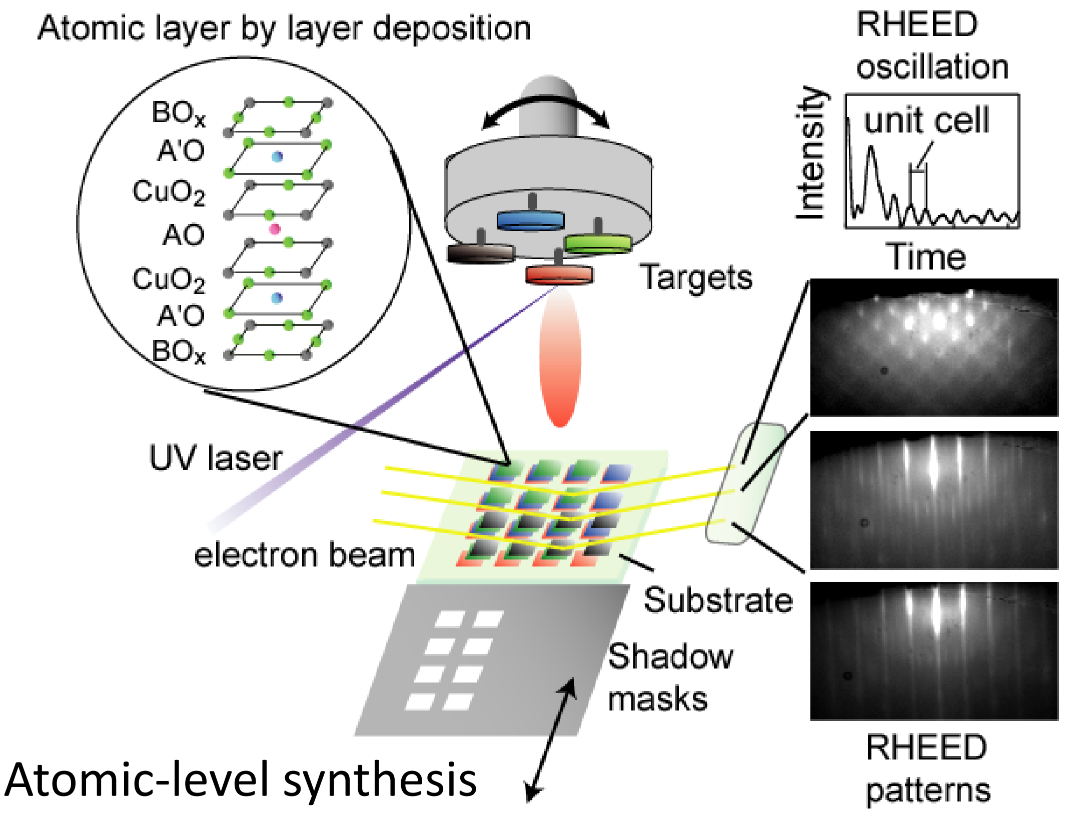
Perovskite : ABO_3

- Insulator~Metal~Superconductor
- Ferro-, Piezo-, Pyro-electricity
- Collosal Magnetresistivity
- Ferromagnetism
- Optical Functions



Autonomous atomic-layer synthesis

w/ Haotong Liang (UMD), Yunlong Sun (U. Tokyo), Mikk Lippmaa (U. Tokyo)



Atomic-level synthesis

Structure determination

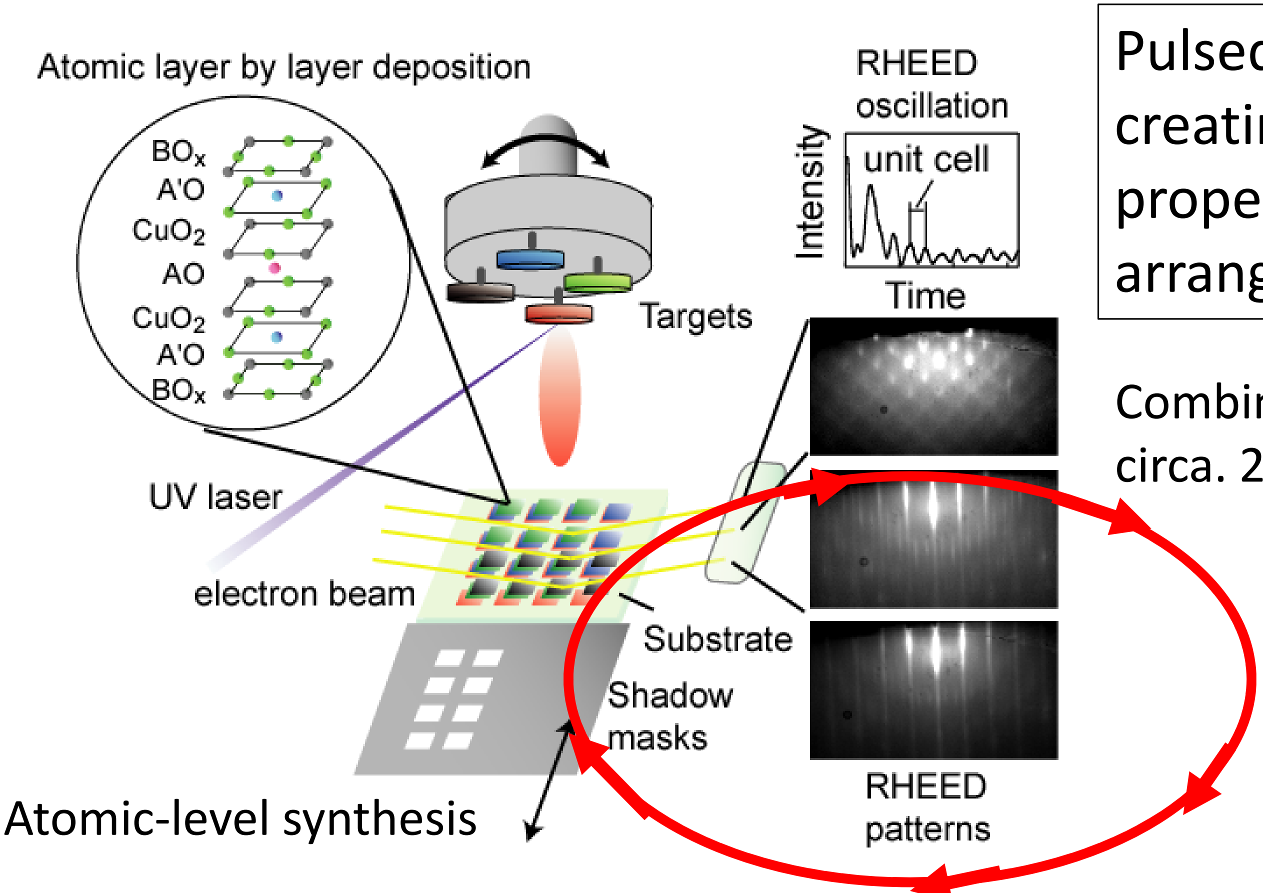
Pulsed laser deposition for creating new materials and tune properties by controlling atomic arrangement at unit-cell level

Combinatorial pulsed laser deposition circa. 2000

Autonomous atomic-layer synthesis

w/ Haotong Liang (UMD), Yunlong Sun (U. Tokyo), Mikk Lippmaa (U. Tokyo)

Pulsed laser deposition for creating new materials and tune properties by controlling atomic arrangement at unit-cell level



Atomic-level synthesis

Structure determination

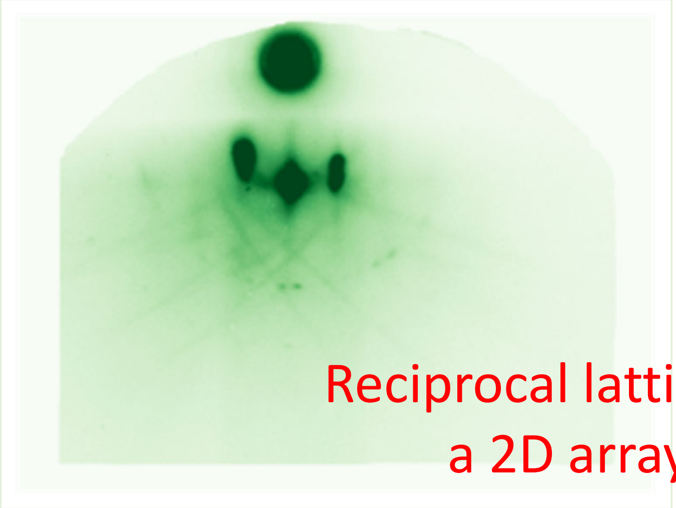
Combinatorial pulsed laser deposition circa. 2000

Live autonomous cycle (2023)

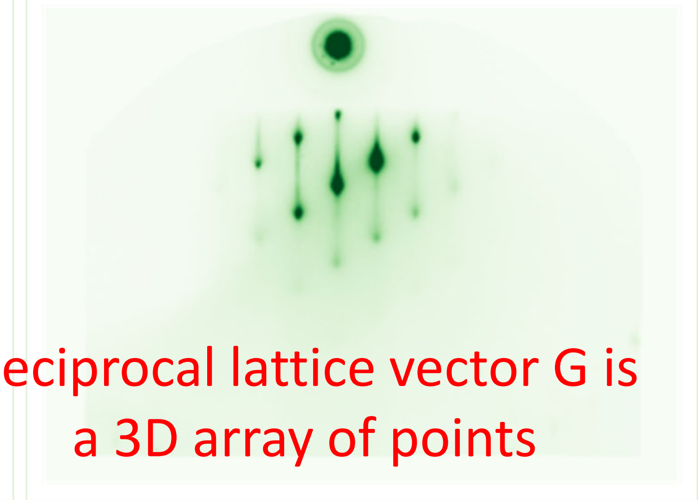
ML analysis of RHEED patterns

Bayesian optimization

RHEED is useful for understanding surface structures



Reciprocal lattice vector G is a 2D array of rods



Reciprocal lattice vector G is a 3D array of points

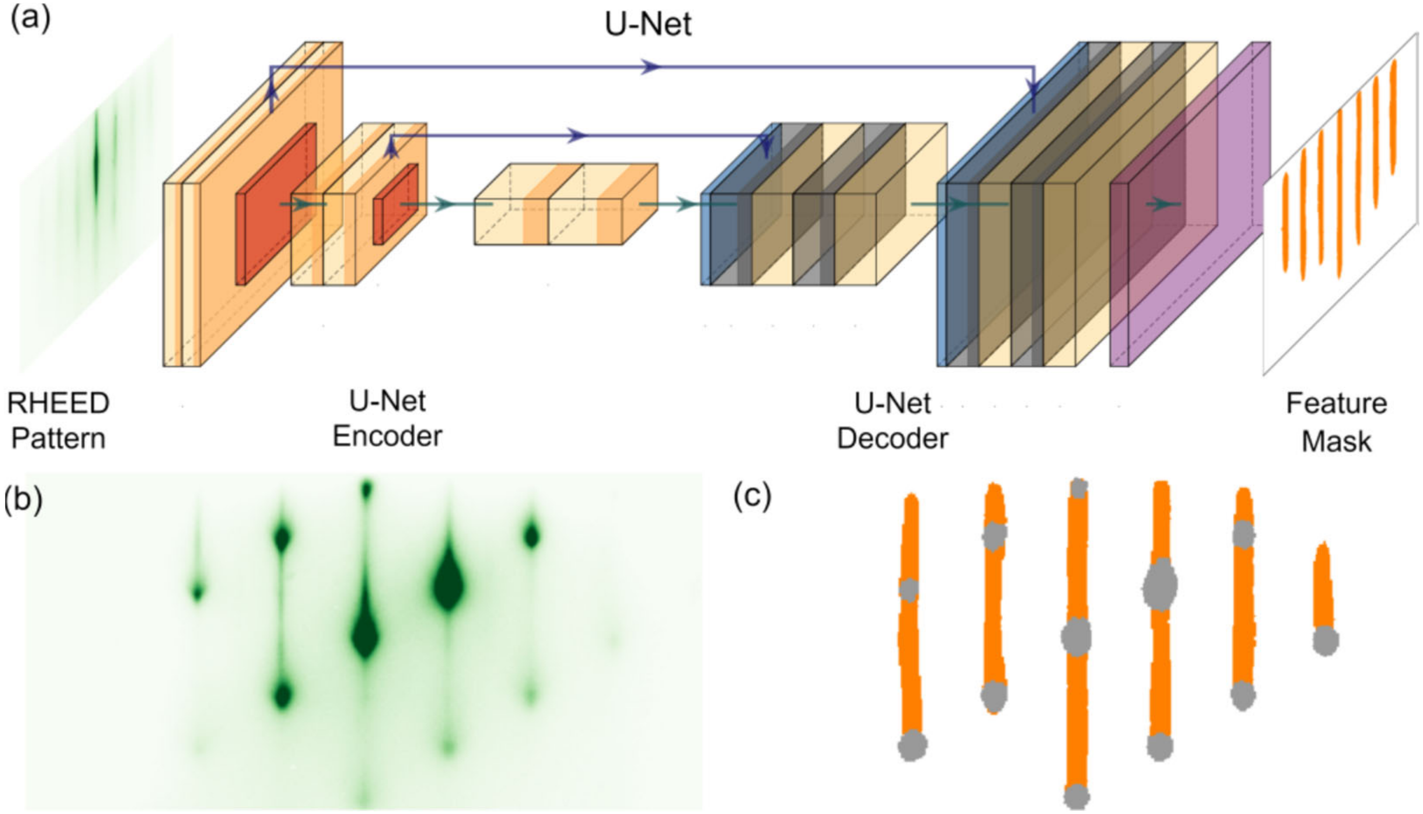
- Spotty pattern
- Kikuchi line (secondary scattering)
- Atomically **flat** surface

- Streaky pattern
- Common in MBE
- Disorder in the atomic-scale

- Streaky + spotty pattern: **3D structures**
- Transmission patterns: 3D structure mixed with 2D structure

……….but it's usually for qualitative characterization

Quantitative live analysis of RHEED patterns using ML

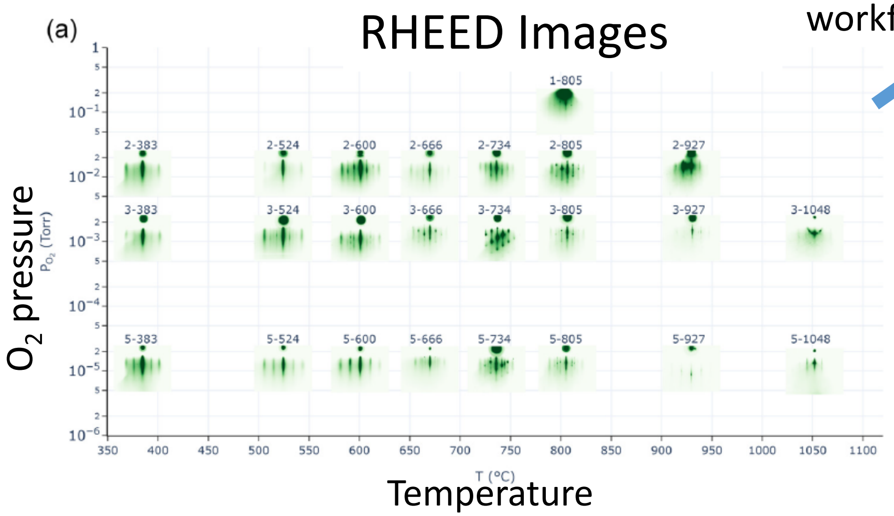


Liang et al., Phys. Rev. Mat. 6, 063805 (2022)

Other ML work on RHEED:
Vasudevan et al., ACS Nano **8**, 10899 (2014);
Provence et al., Phys. Rev. Materials **4**, 083807 (2020).

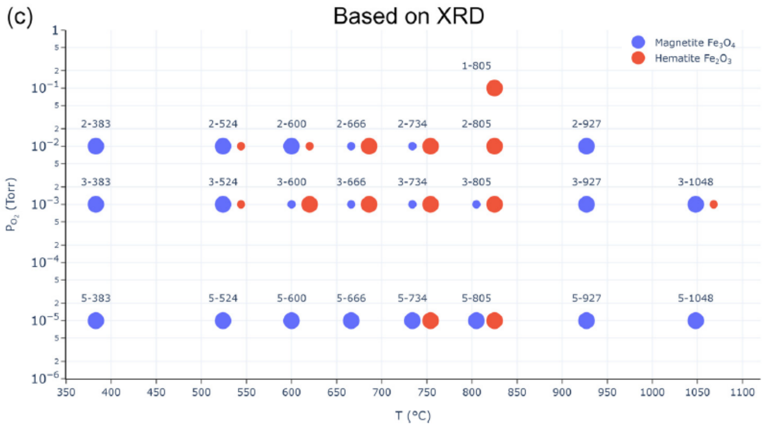
Quantitative analysis of RHEED patterns using ML

Growth phase map of FeO_x thin films:
Fe_2O_3 vs Fe_3O_4 vs… as functions of O_2 pressure and temperature during growth



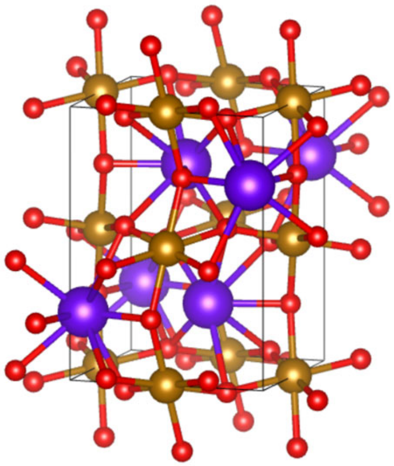
ML workflow

Good agreement with "post mortem" XRD



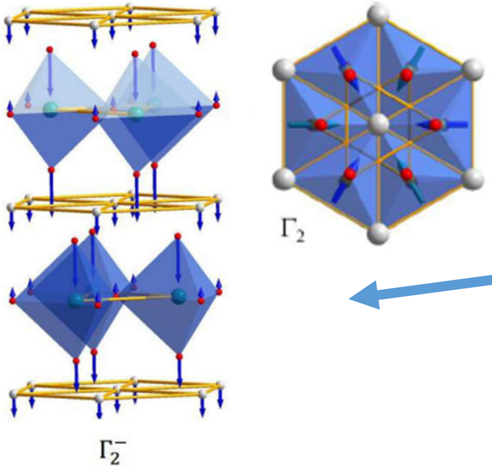
Liang et al., Phys. Rev. Mat. 6, 063805 (2022)

Target material: metastable hexagonal TbFeO_3



Pbnm Phase

Stable structure
colinear antiferromagnetic order



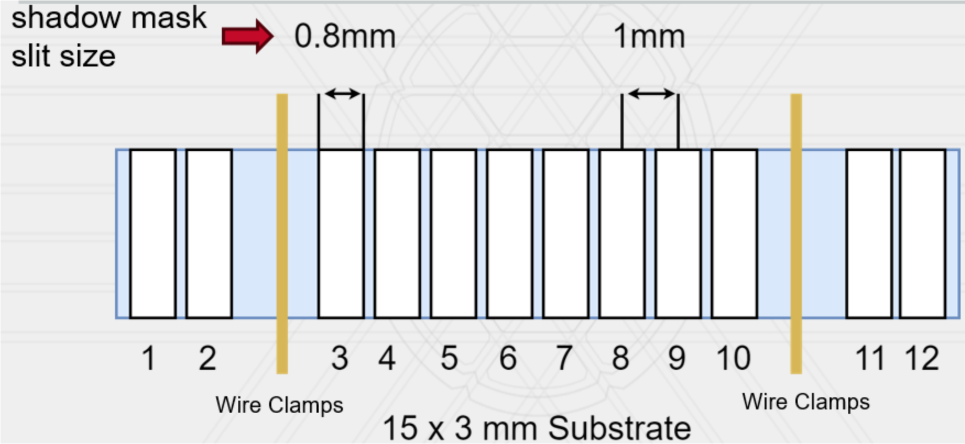
P63/mmc Phase

Metastable multiferroic, non-colinear antiferromagnetic order

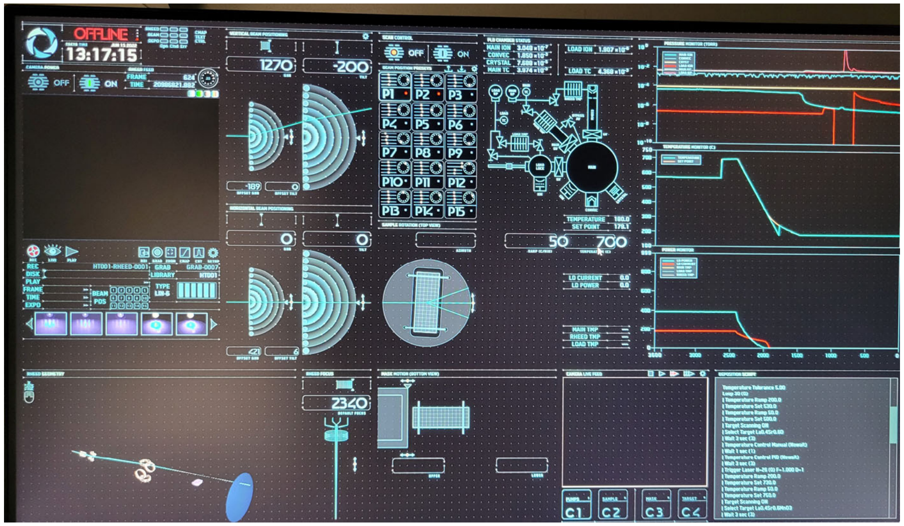
X. Xu and W. Wang, *Multiferroic Hexagonal Ferrites (h-RFeO3, R = Y, Dy-Lu): A Brief Experimental Review*, Mod. Phys. Lett. **28**, (2014).

- Stabilize metastable hexagonal P63/mmc
- Search for optimal growth condition for thin films

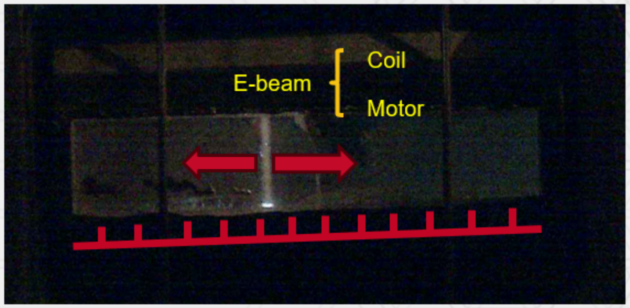
Autonomous atomic-layer synthesis setup 2023


shadow mask slit size

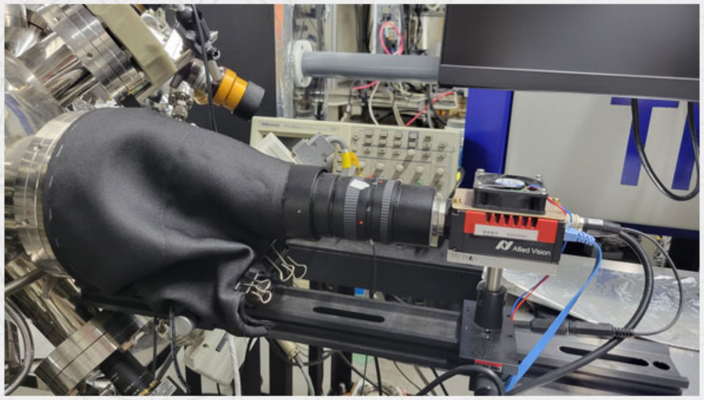
15 x 3 mm Substrate



Calibration

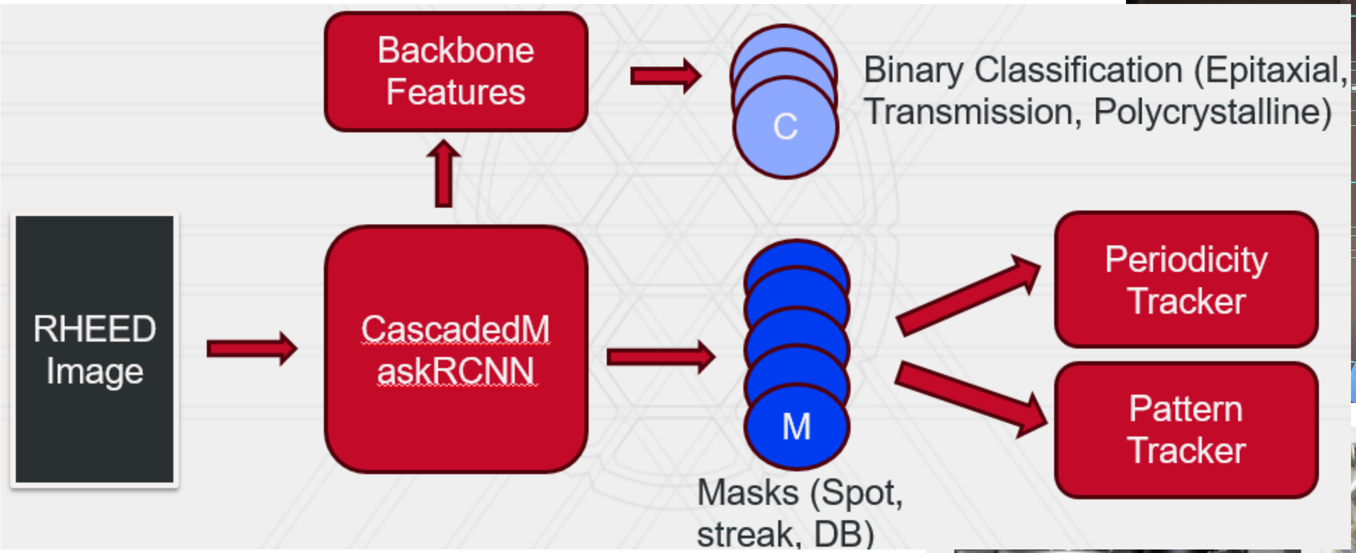



Low-light sensitive Camera (ELP)


High Speed Network Camera (Allied Vision)

Autonomous atomic-layer synthesis setup 2023

New RHEED analysis pipeline



RHEED Image

CascadedMaskRCNN

Backbone Features

C

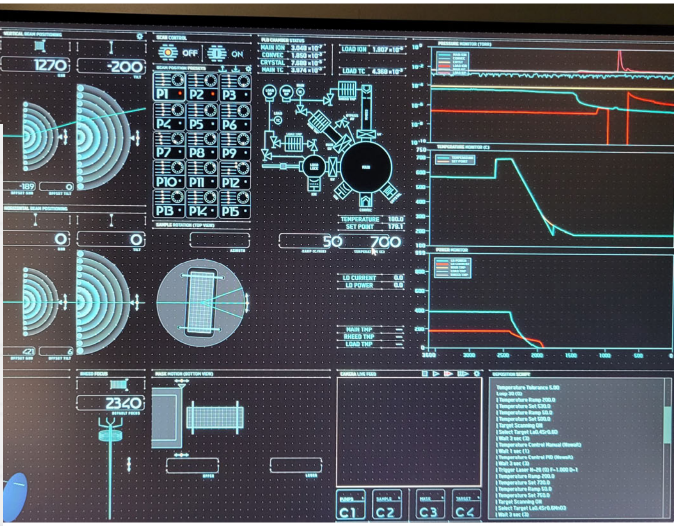
Binary Classification (Epitaxial, Transmission, Polycrystalline)

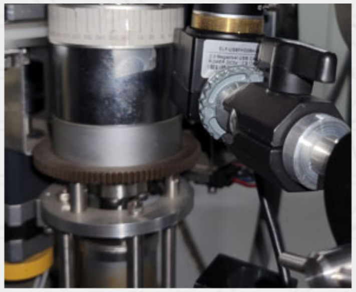
M

Masks (Spot, streak, DB)

Periodicity Tracker

Pattern Tracker





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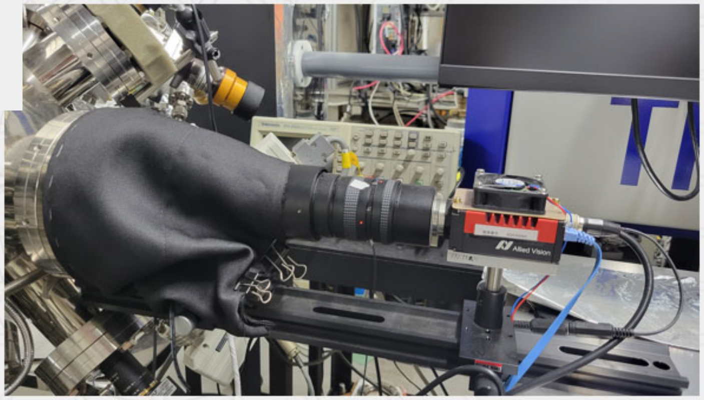
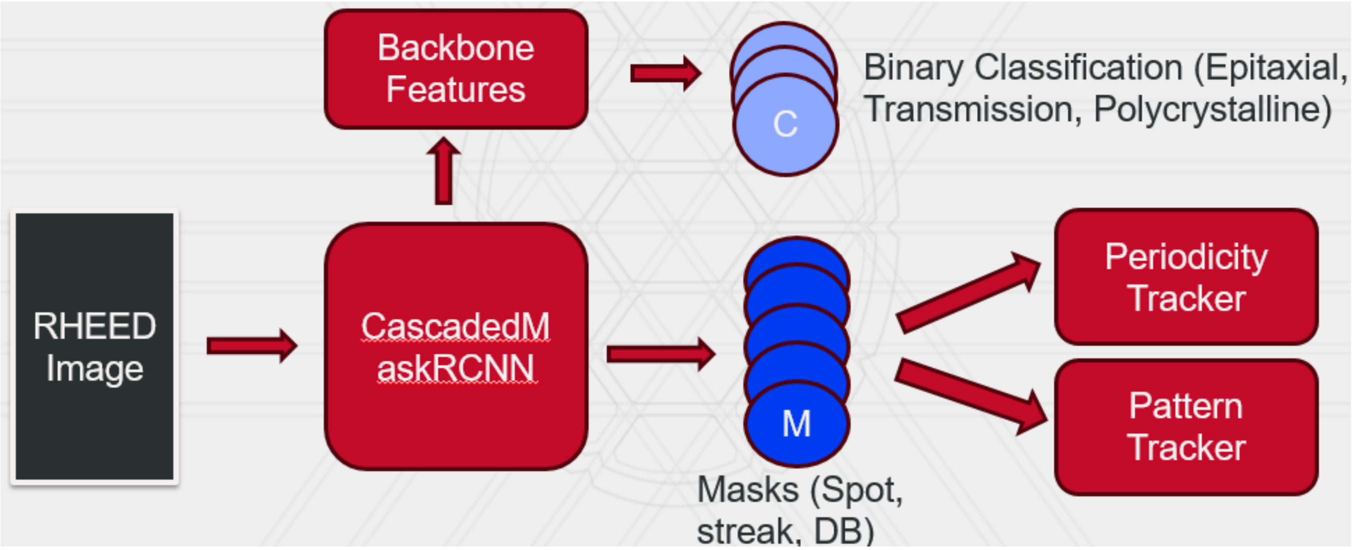


High Speed Network Camera (Allied Vision)

Autonomous atomic-layer synthesis setup 2023

New RHEED analysis pipeline



RHEED Image

CascadedMaskRCNN

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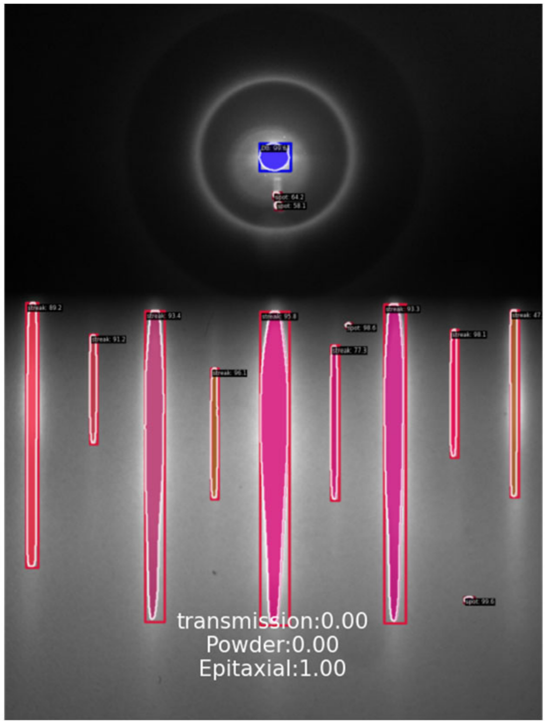
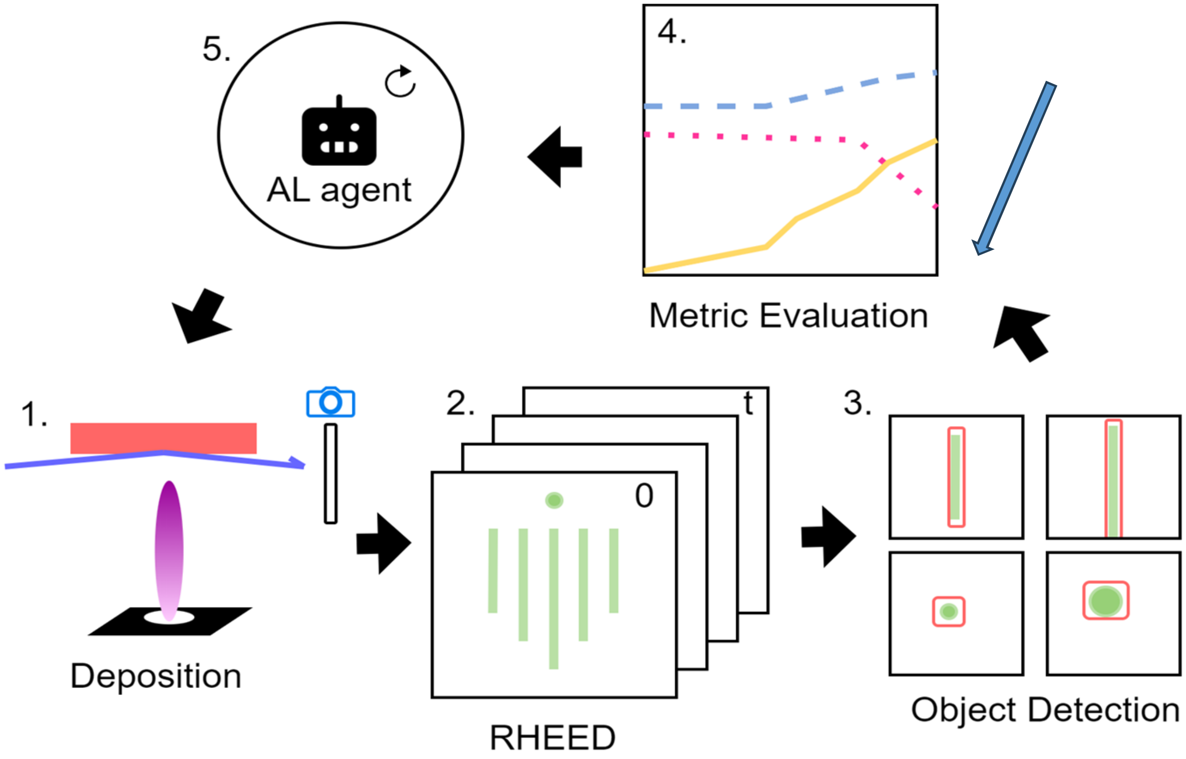


- Multiple features are identified: epitaxial vs islands vs ring (polycrystalline)
- Sharpness of the peak
- Periodicity gives us the lattice constant – can identify the phase

Autonomous live experimental loop for controlled growth of hexagonal $TbFeO_3$



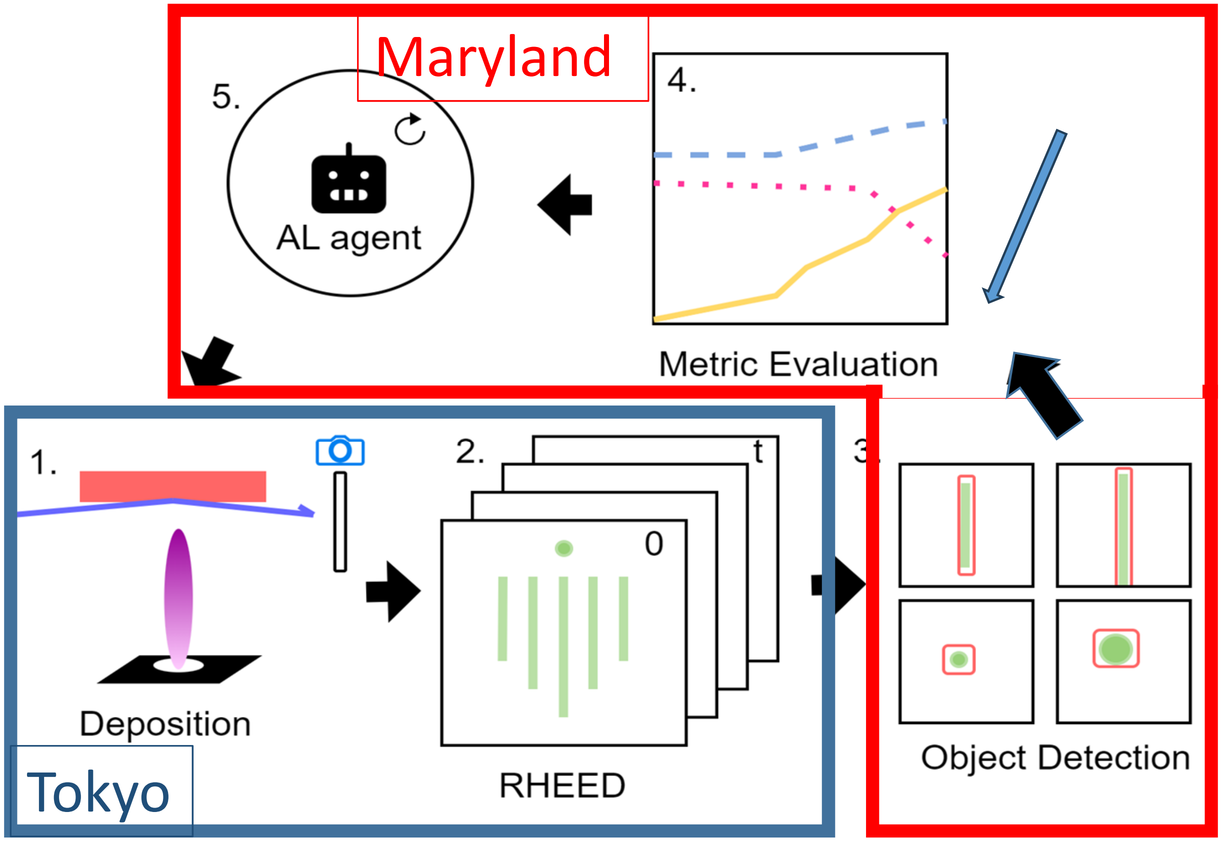
15 min per cycle

Parameters:

- O_2 pressure: 10^{-7}-10^{-4} Torr (10 increments)
- Temperature: 600 – 1000 ℃ (16 increments)
- Laser pulse rate: 0.5 – 20 Hz (7 increments)

Total grid: 1100 experiments

Autonomous live experimental loop for controlled growth of hexagonal $TbFeO_3$



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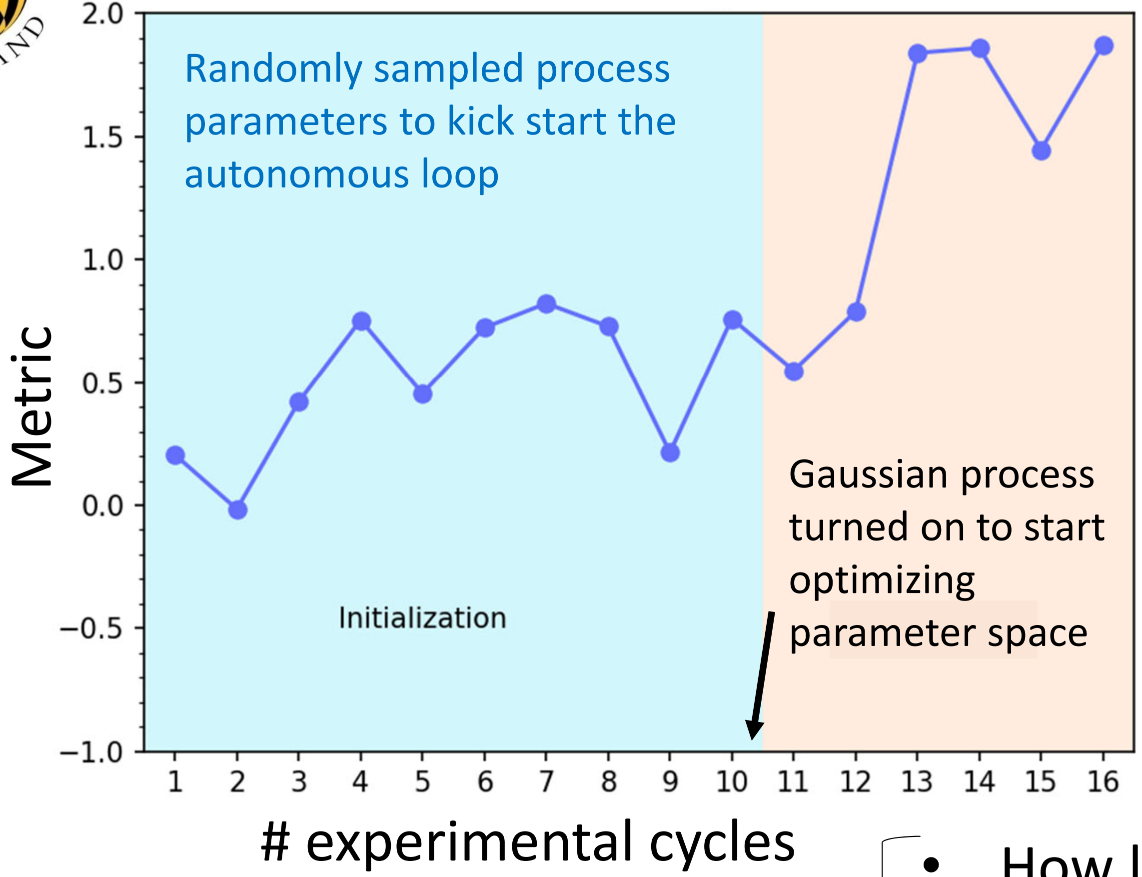
Total grid: 1100 experiments

Autonomous atomic-layer synthesis of $TbFeO_3$



Randomly sampled process parameters to kick start the autonomous loop

Initialization

Gaussian process turned on to start optimizing parameter space

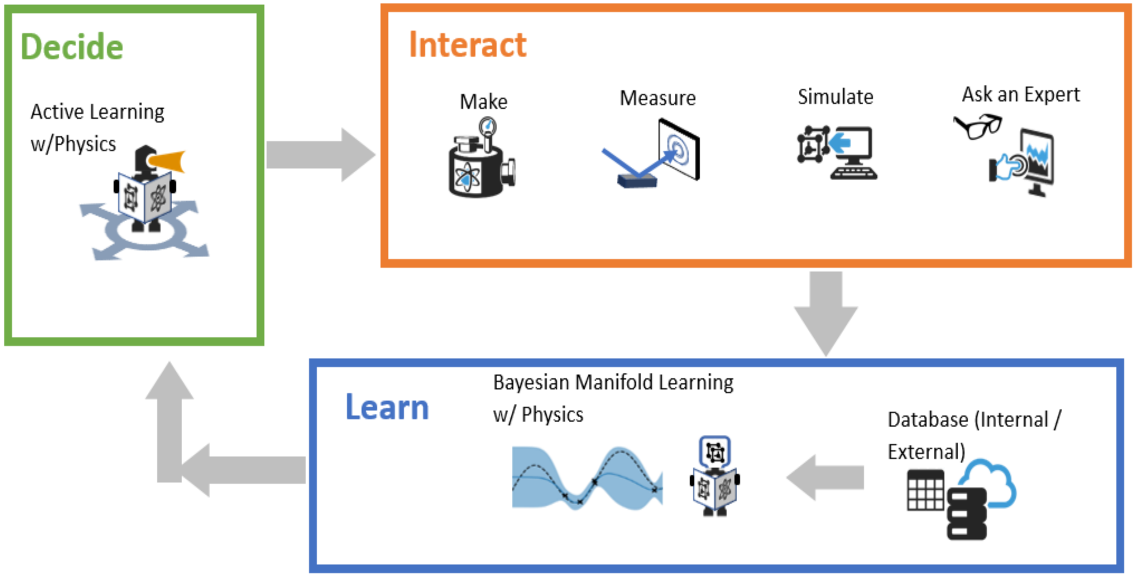
- 3 deposition parameters (pressure, temperature, dep rate: 1100 possibilities)
- Each cycle ~15 min
- Takes us to unexpected region of parameter space
- Always converges after only < 15 cycles of autonomous iteration

3 figures of merit from RHEED analysis combined into 1 metric (-1 < range < 2)

- How long does the phase stay stable?
- Sharpness of the peak
- How fast can we grow it? (Hz)

Autonomous additive manufacturing

Haotong Liang, J-C Zhao (MSE), Huapeng Huang (AAL)



Decide

Active Learning w/Physics

Interact

Make

Measure

Simulate

Ask an Expert

Learn

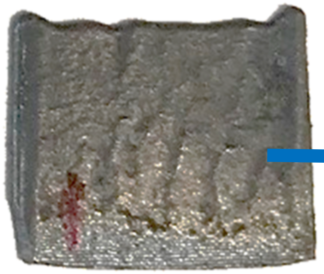
Bayesian Manifold Learning w/ Physics

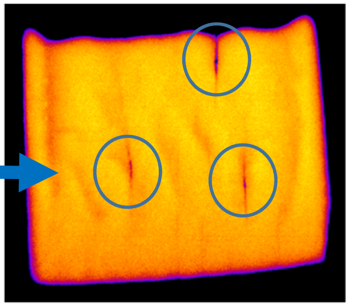
Database (Internal / External)

(LDED tool)

Compact X-ray source and detector mounted inside printer

Optical image of a printed superalloy





In-situ X-ray image detects defects not visible optically

Autonomous additive manufacturing

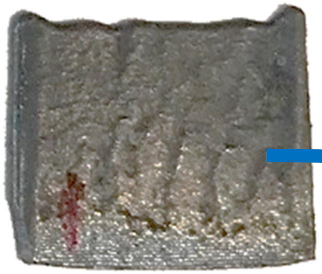
Haotong Liang, J-C Zhao (MSE), Huapeng Huang (AAL)

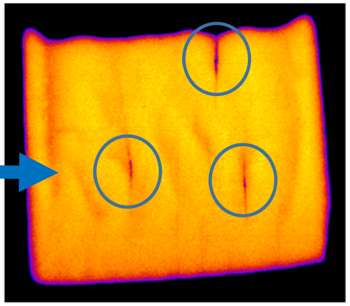
Goal: develop a live closed-loop metal AM tool, which automatically evaluates the quality of printed materials, figures out how to optimize, and proceeds to find the best process conditions with minimum # of iterations w/ no human intervention

(LDED tool)

Compact X-ray source and detector mounted inside printer

Optical image of a printed superalloy





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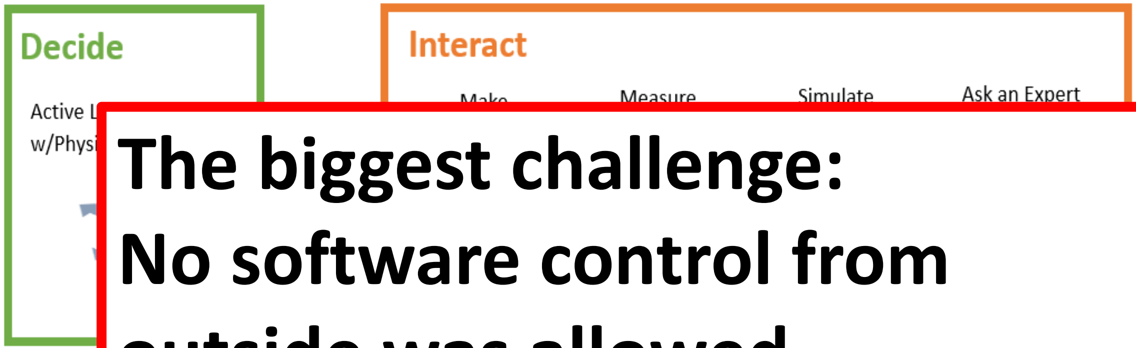


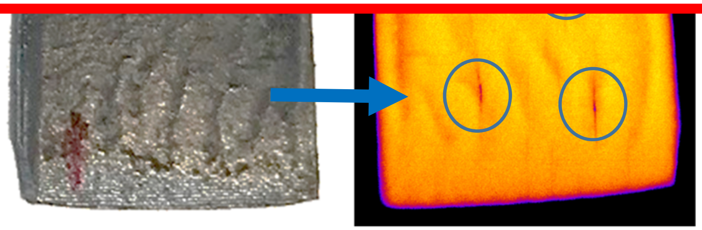
The biggest challenge:
No software control from outside was allowed

Solution: Hardware/software integration hack

(LDED tool)

Compact X-ray source and detector mounted inside printer

Optical image of a printed superalloy



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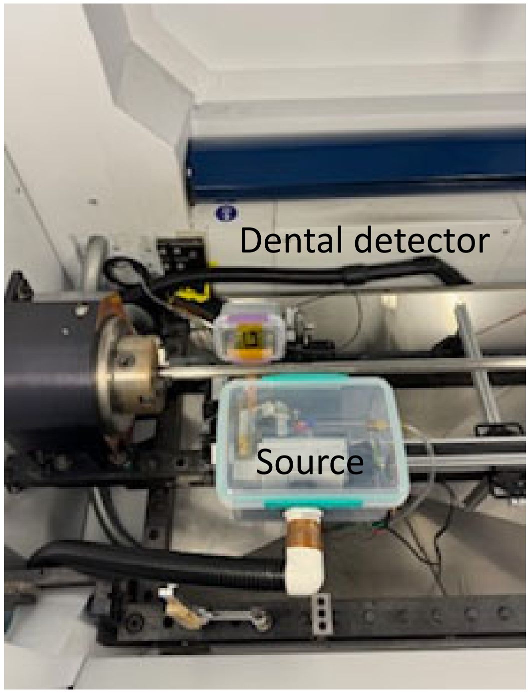
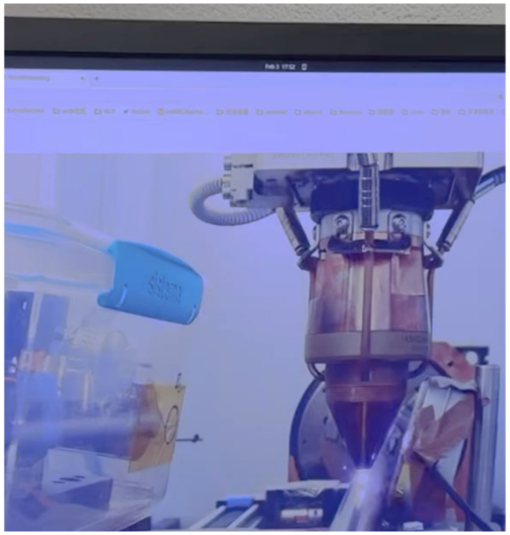
Haotong Liang, J-C Zhao (MSE), Huapeng Huang (AAL)

Hardware/software integration hack

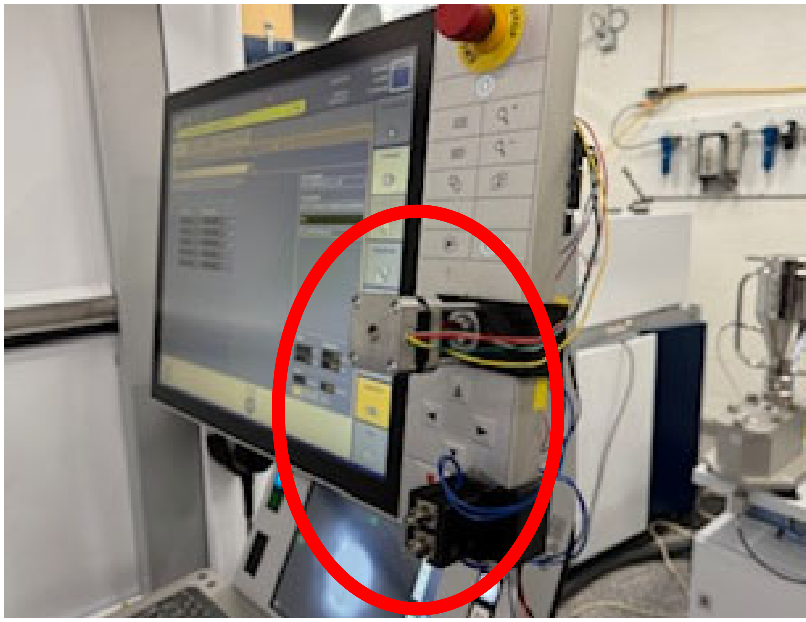
Compact X-ray source and detector



Camera for monitoring

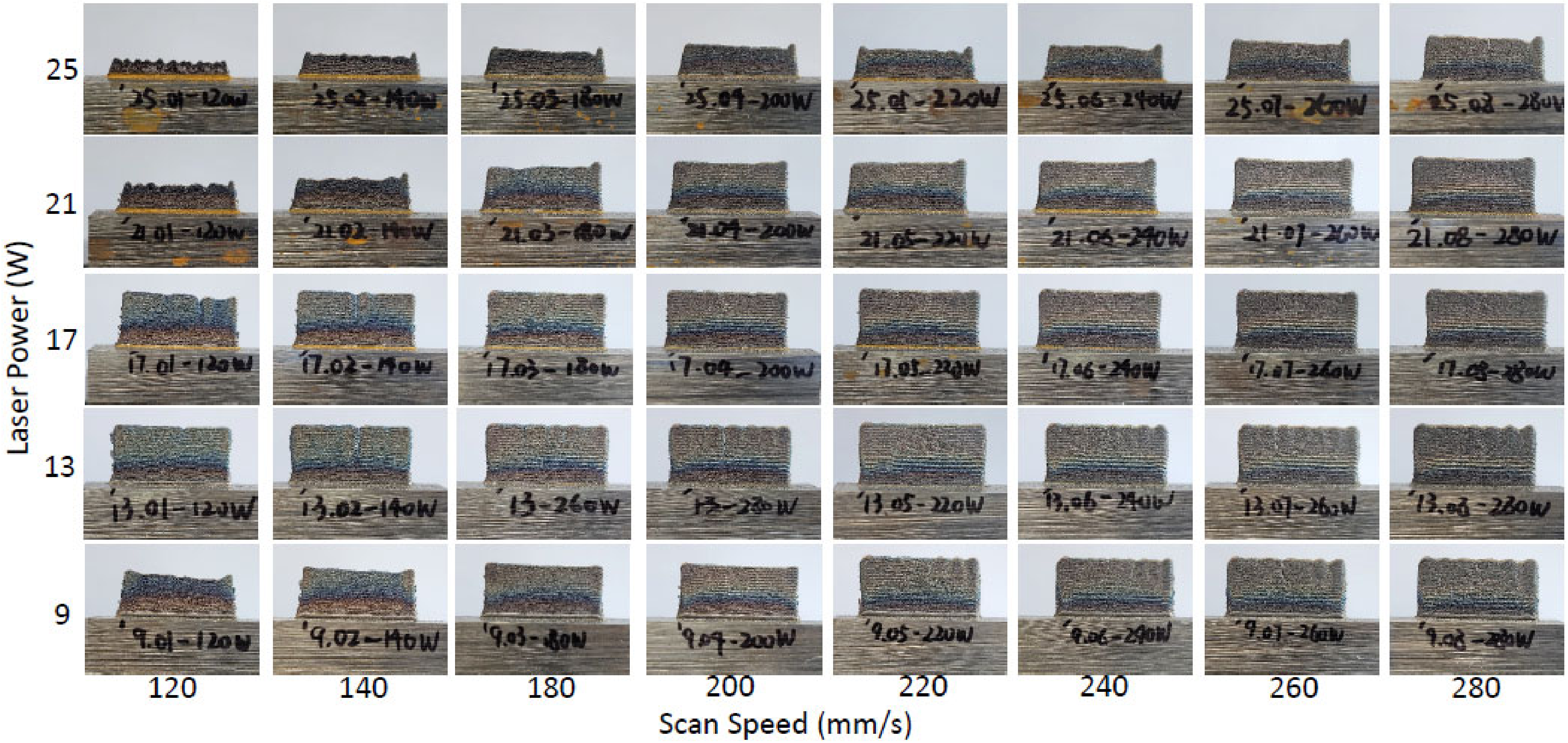


Motor and actuators to replace a knob and on/off button



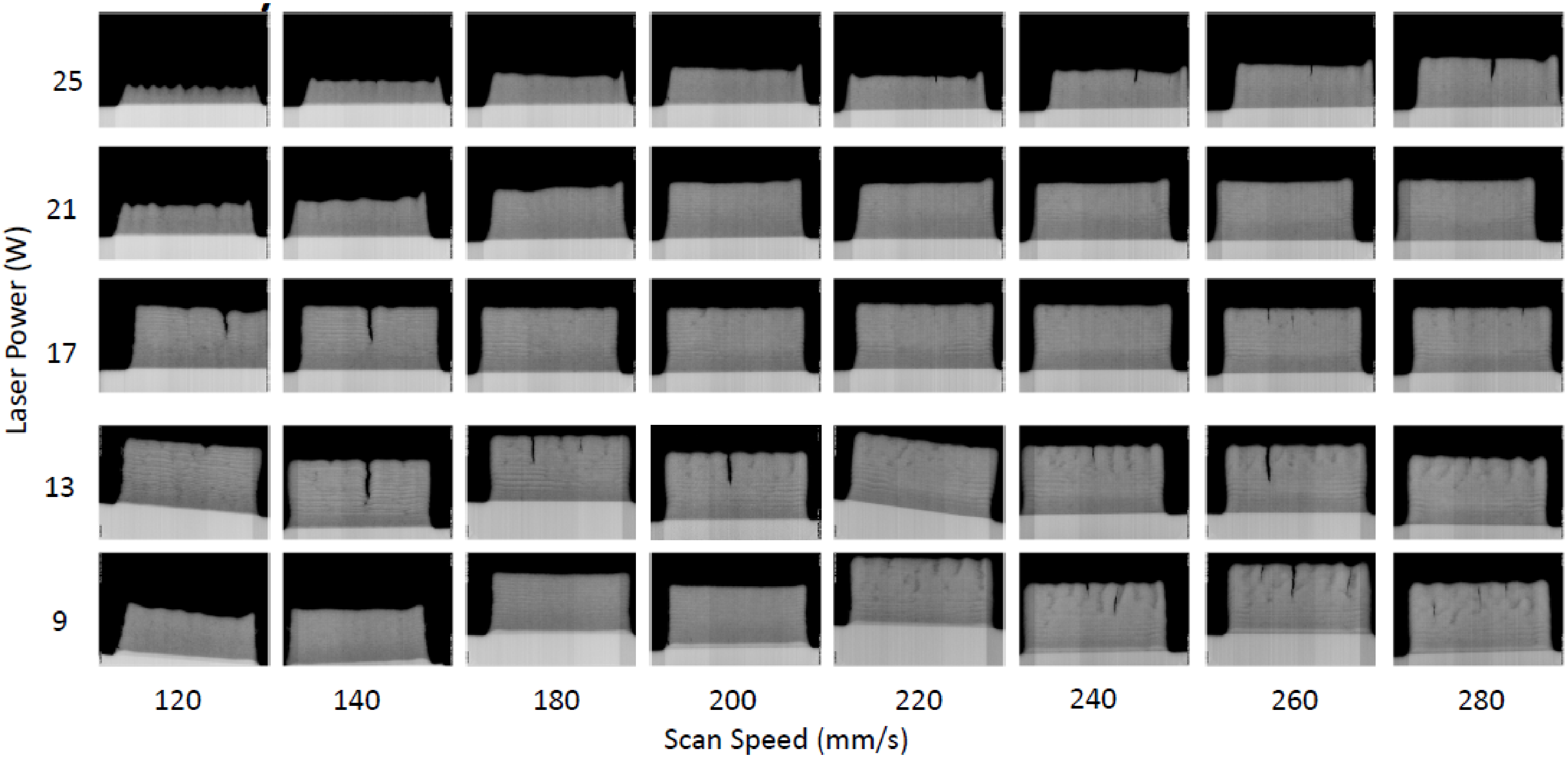
Superalloy (MAR-M247) printing: minimize crack formation by controlling laser power and scan speed

Reality check: grid measurements: optical images

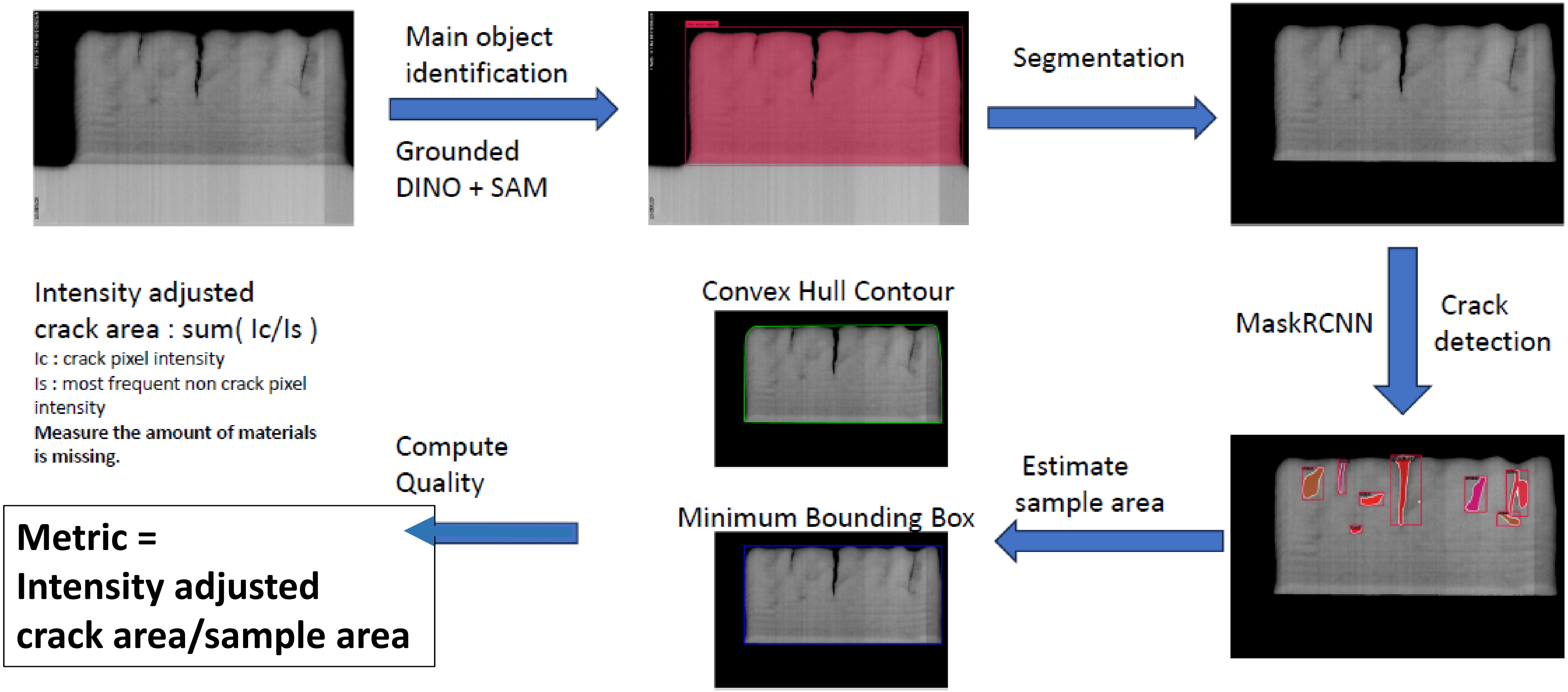


Superalloy (MAR-M247) printing:
minimize crack formation by controlling laser power and scan speed

Reality check: grid measurements: X-ray images



Computer vision workflow for identifying defects from X-ray images and quantify them



Main object identification

Grounded DINO + SAM

Segmentation

MaskRCNN

Crack detection

Estimate sample area

Convex Hull Contour

Minimum Bounding Box

Compute Quality

Intensity adjusted crack area : sum(Ic/Is)

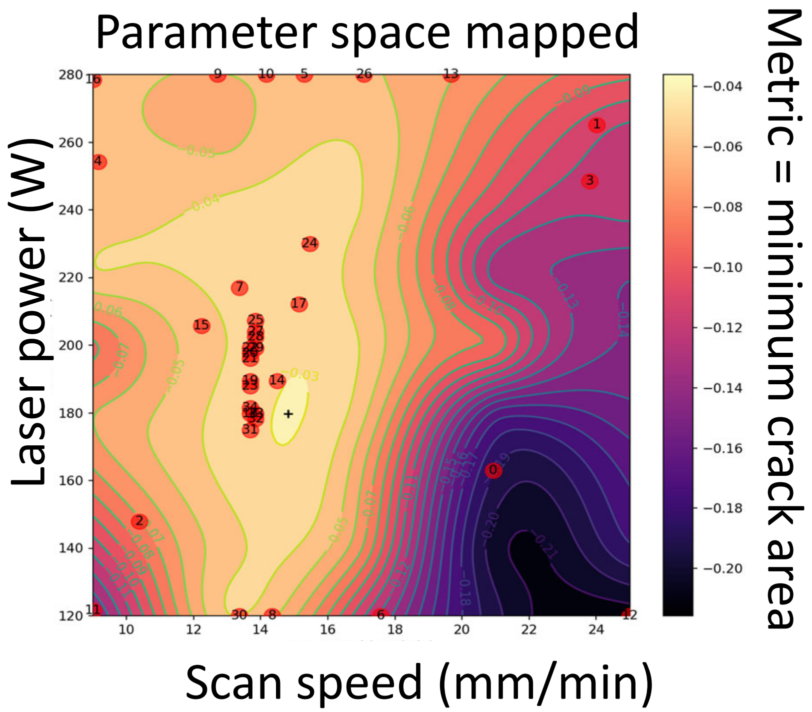
Ic : crack pixel intensity

Is : most frequent non crack pixel intensity

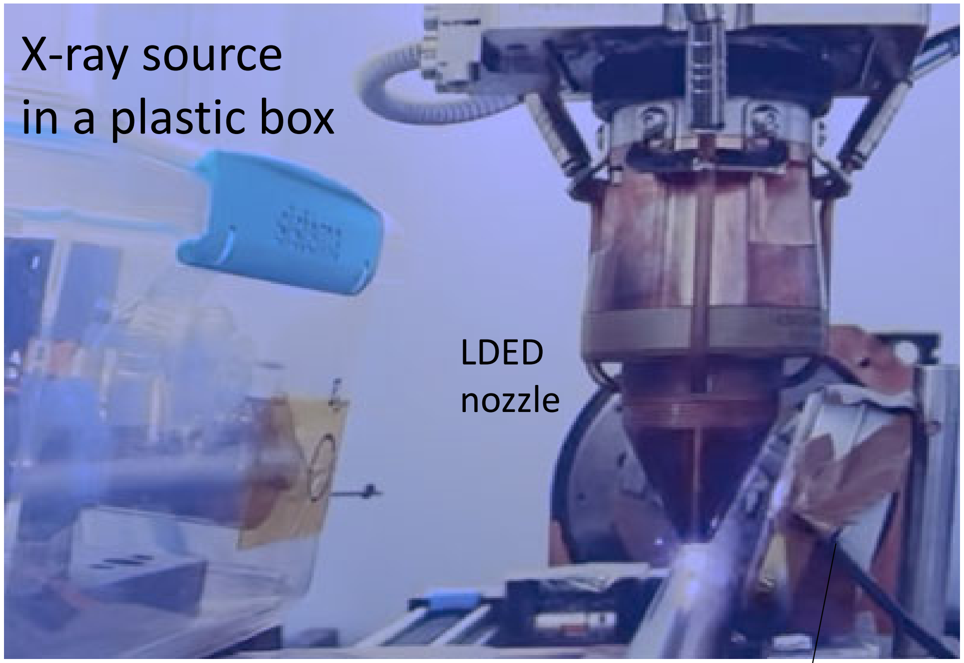
Measure the amount of materials is missing.

Metric = Intensity adjusted crack area/sample area

Demonstration of autonomous metal additive manufacturing


Parameter space mapped

After ~8 prints,
best conditions are identified



Dental X-ray imager in plastic

→ After 40 min:
the plastic had melted and imager was burned

Liang, Takeuchi et al., to be published (2024)

Summary

We are developing various nimble autonomous materials science platforms

We have demonstrated:

- autonomous control of oxide thin film growth by PLD
  (the basis for autonomous semiconductor manufacturing)

- autonomous control of metal additive manufacturing

Acknowledgement

University of Maryland	Advanced Analyzer	NIST	Univ. of Tokyo
H. Liang	H. Huang	A. G. Kusne	M. Lippmaa
J.-C. Zhao			Y. Sun

Supported by NIST, ONR, AFOSR, and SRC