SCHOOL OF ENGINEERING

# Status report: Baseline materials for characterizing the MUED configuration, their role verifying daily alignment and in operation and implementation of a non-destructive real-time machine learning diagnostic for ensuring beam stability

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DOE's Established Program to Stimulate Competitive Research (EPSCoR).

**Status:** Funded.

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Los Alamos NATIONAL LABORATORY

Rensselaer

# Funding acknowledgements

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# Funding secured through DOE EPSCoR program

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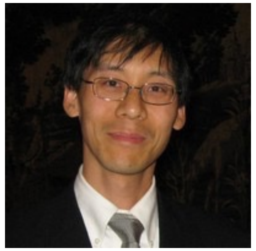
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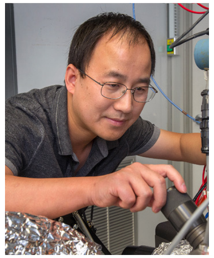
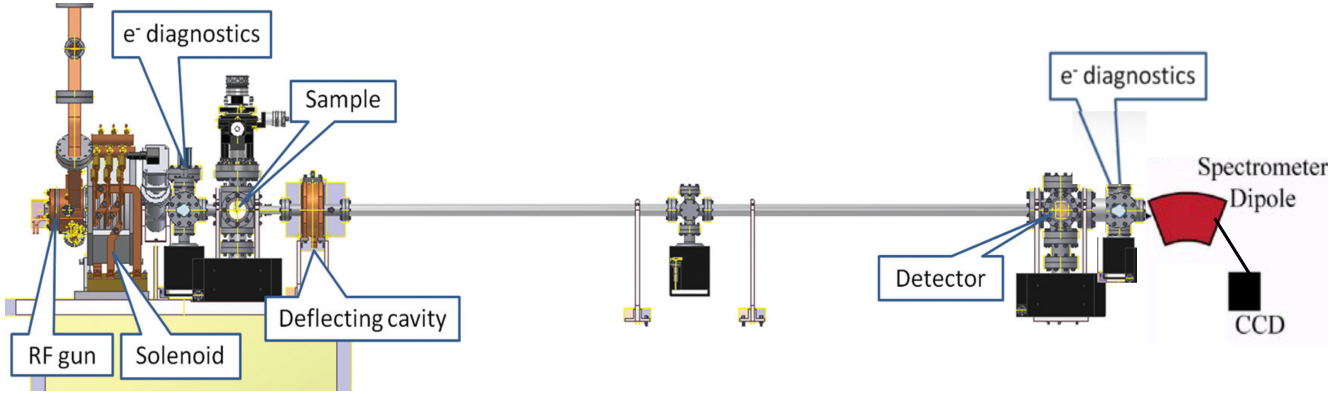


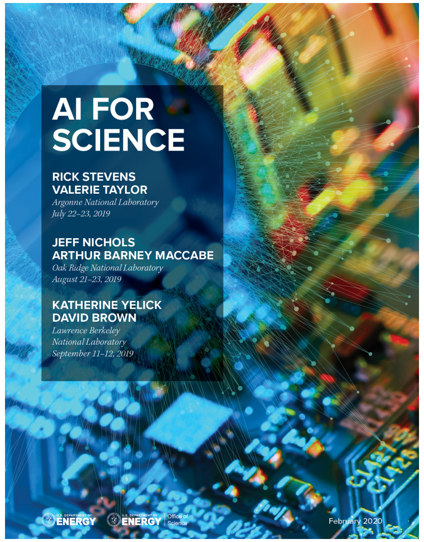

Mikhail Fedurin

# Two DOE facilities are involved: ATF and ALCF

Accelerator Test Facility (ATF)




Argonne Leadership Computing Facility (ALCF)


The combination of machine hardware, advanced computing for simulation, and data science for surrogate modelling, training of neural networks and data analysis is inspired by our past work and our participation on DOE meetings, workshops and reports such as AI for Science (*https://www.anl.gov/ai-for-science-report*).

# Special Equipment Requirements and Hazards

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## Special equipment:

- A ***second camera*** will be eventually required for the last step of the suite of experiments as is the ***associated controls, sample holder etc.*** Funding is secured for everything except for this last step.

## Hazards:

- Other potential hazards include the ***laser*** of the MUED instrument and the ***cryogenic system*** necessary to cool the samples to the desired temperatures. We will work with the BNL collaborators to exercise the necessary precautions.

# Experimental time request

CY2022 Time Request

| Capability | Setup Hours | Running Hours |
|---|---|---|
| UED Facility | 36 | 84 |

Time Estimate for Remaining Years of Experiment (including CY2021)

| Capability | Setup Hours | Running Hours |
|---|---|---|
| UED Facility | 72 | 168 |

# Overview of the proposal

We propose a suite of experiments that will span for a 3-year period aimed at characterizing baseline materials, optimizing their analysis and controlling alignment and stable operation of the instrument.

*We will characterize baseline materials, automate the analysis, apply artificial neural network based models and contribute to the development of a MUED database. Analysis of the diffraction patterns will allow determination of the relevant beam parameters and enable to maintain beam stability during the experiment.*

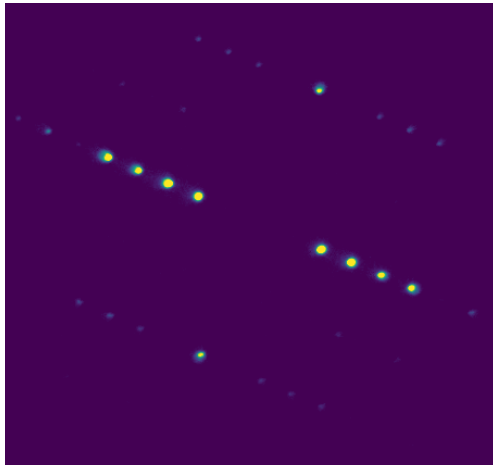
*At a later stage, a chosen calibration sample will be placed in an extension of the beamline and will interact with the undiffracted beam, providing real-time feedback to the instrument systems.*

**We expect the proposed experiments move the MUED instrument forward to make it turn-key, high stability, and high-throughput.**

# Characterization of baseline materials

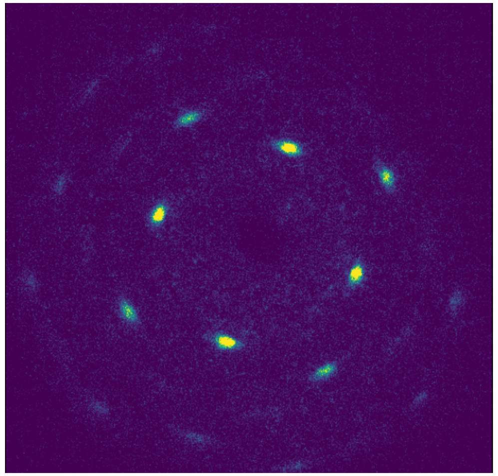
We measured 2 baseline materials this first year:

- $Ta_2NiS_5$



Samples courtesy of Junjie Li

- Au



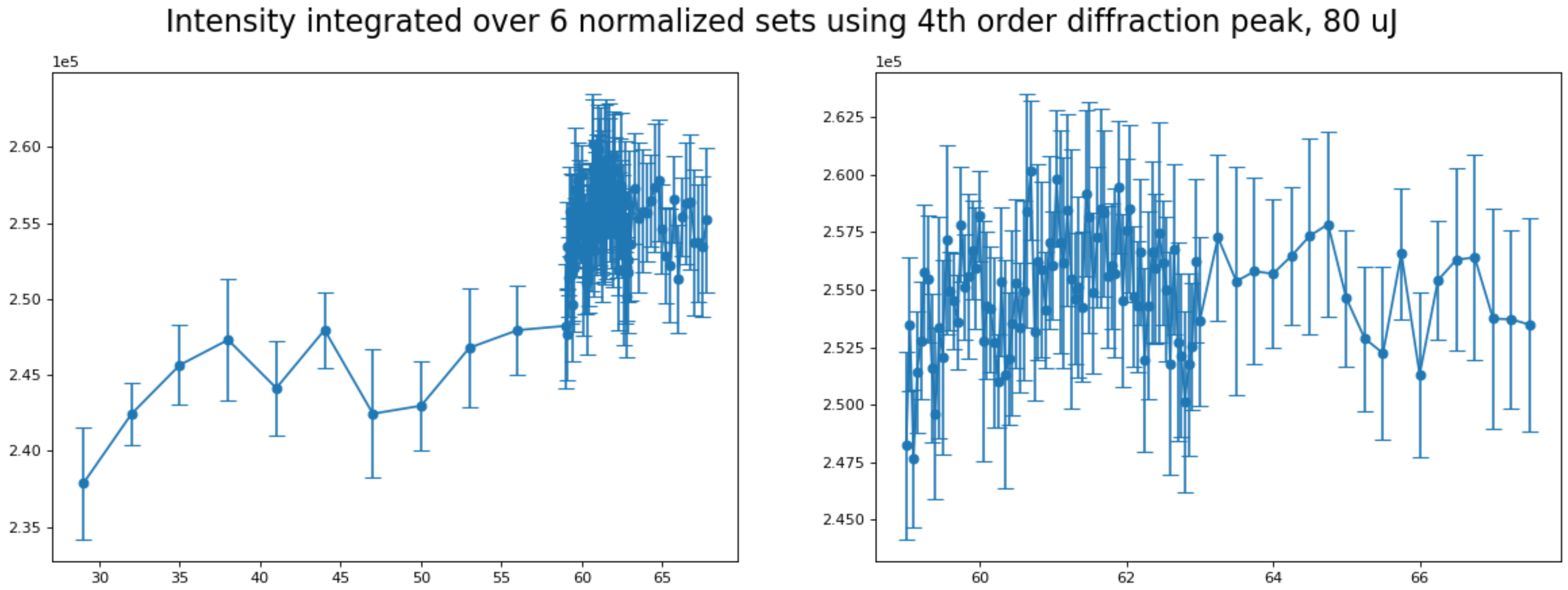
Commercial samples from Ted Pella (TEM standard)

✓ We collected more than 5000 single shot measurements.

✓ During our last beamtime, we conducted pump-probe measurements for both materials in single shot and integrated modes.

# Characterization of baseline materials

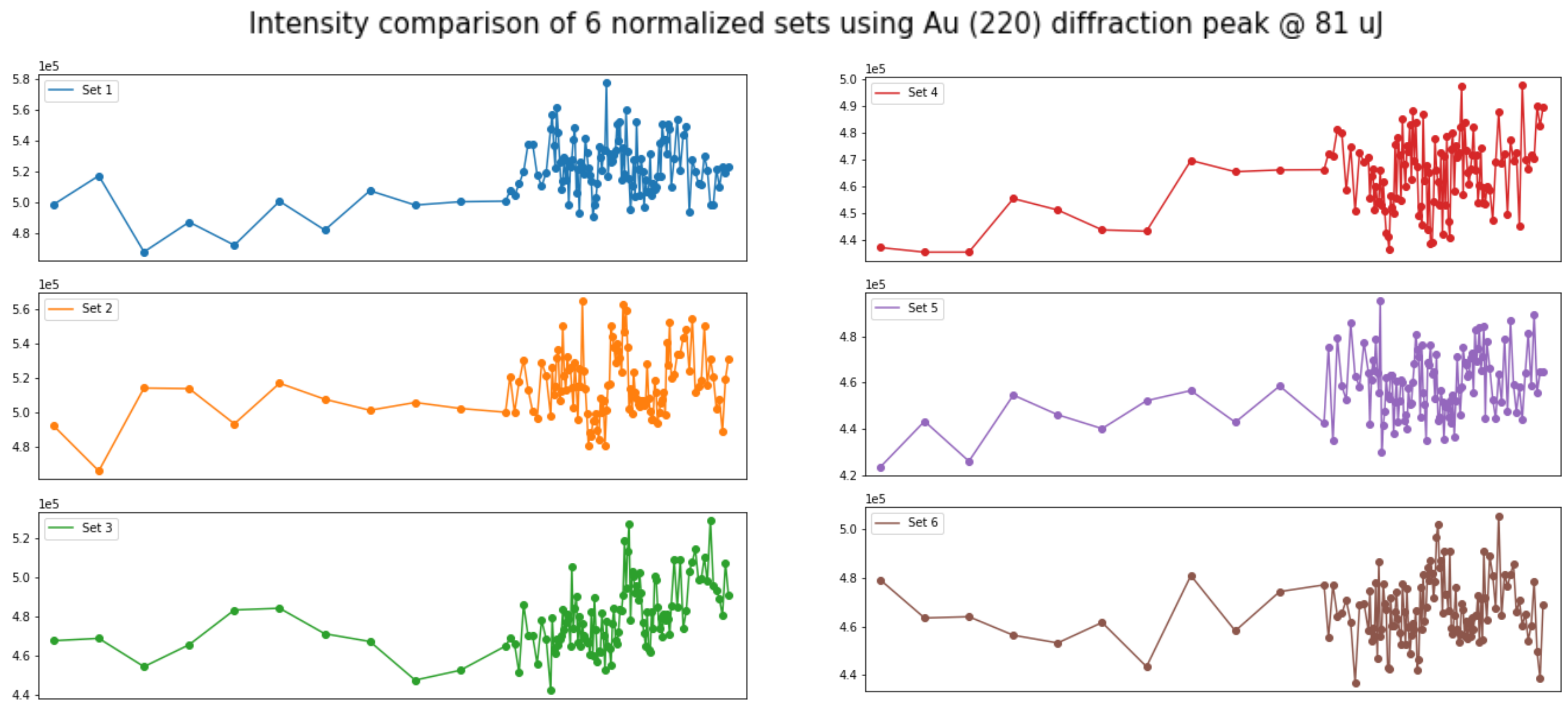
- During our Nov-Dec 2021 beamtime, we conducted pump-probe experiments.
- We were particularly interested in estimating the Debye-Waller factor of Au.
- Integrated measurements over 70 shots were employed.
- We were not able to estimate the time zero:



# Characterization of baseline materials

- There were some stability concerns regarding the beam with some frequency dependence, which were difficult to diagnose.

- Preliminary: presence of oscillations in the (assumed) negative time region:

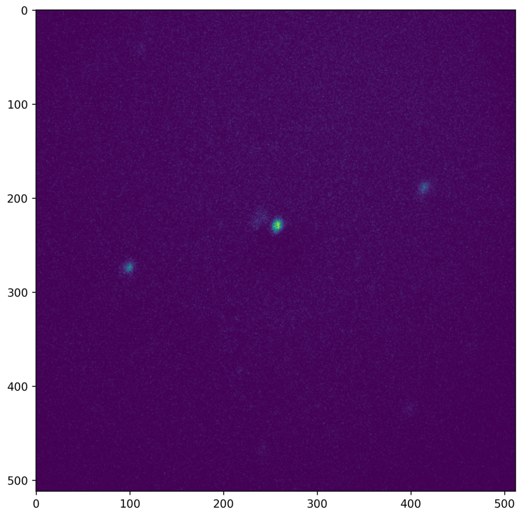


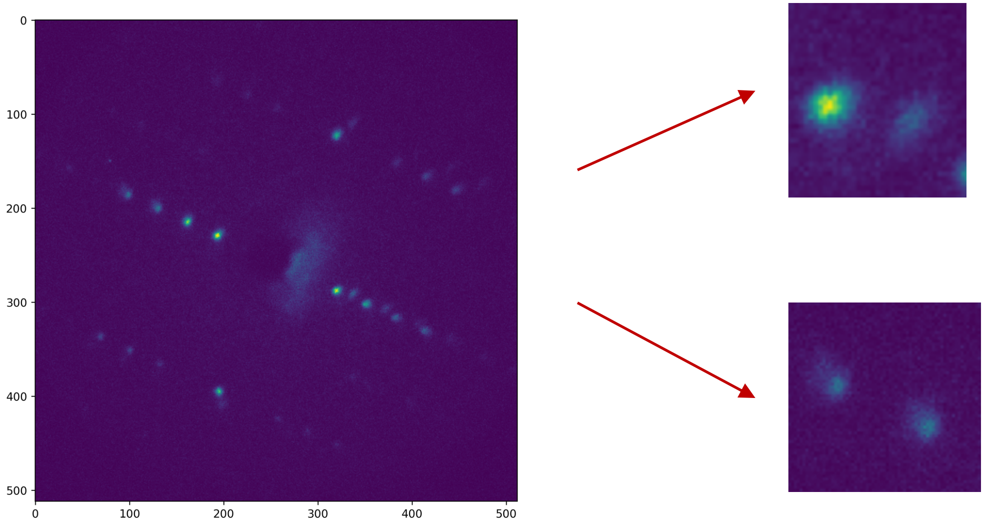
# Autonomous anomaly detection

- During single shot experiments, one is able to observe patterns that are **anomalous** due mostly to instabilities in the electron beam.

- These anomalies are integrated when accumulating several patterns (typically 70) and will be detrimental for the accuracy of the experiment.

- Some examples:




- The rate of anomalies is about 10% but can vary largely with experimental conditions (eg: 38% anomaly rate in a pump-probe experiment).

# Autonomous anomaly detection

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We want to be able to find anomalous patterns in the large datasets with no user input (autonomous)

- We have different types of anomalies and would like to also recognize unseen types.
- We will limit our analysis to $Ta_2NiS_5$ as it is single crystal.
- Given the low rate of anomalies, we can't employ a classification model.

  - We developed a convolutional autoencoder model to reconstruct the diffraction patterns.
  - Our model trains on all data (unsupervised).
  - An anomaly will have a large reconstruction error or different feature vector values.
  - We will test different strategies to detect anomalies, such as one-class support vector machines (unsupervised).

# Autonomous anomaly detection: preprocessing

- Input: images of 512 x 512 pixels, mostly filled with background.
- We split each image in 80 x 80 pixel tiles, using a sliding window with overlap.
- Some of the tiles are background, need to be filtered out if not model will train on mostly background.
- We devised a simple algorithm to decide if a tile contains background (= white noise):

For f(x) a discreet distribution of N samples that is normalized, we define the inverse participation ratio (IPR) as:

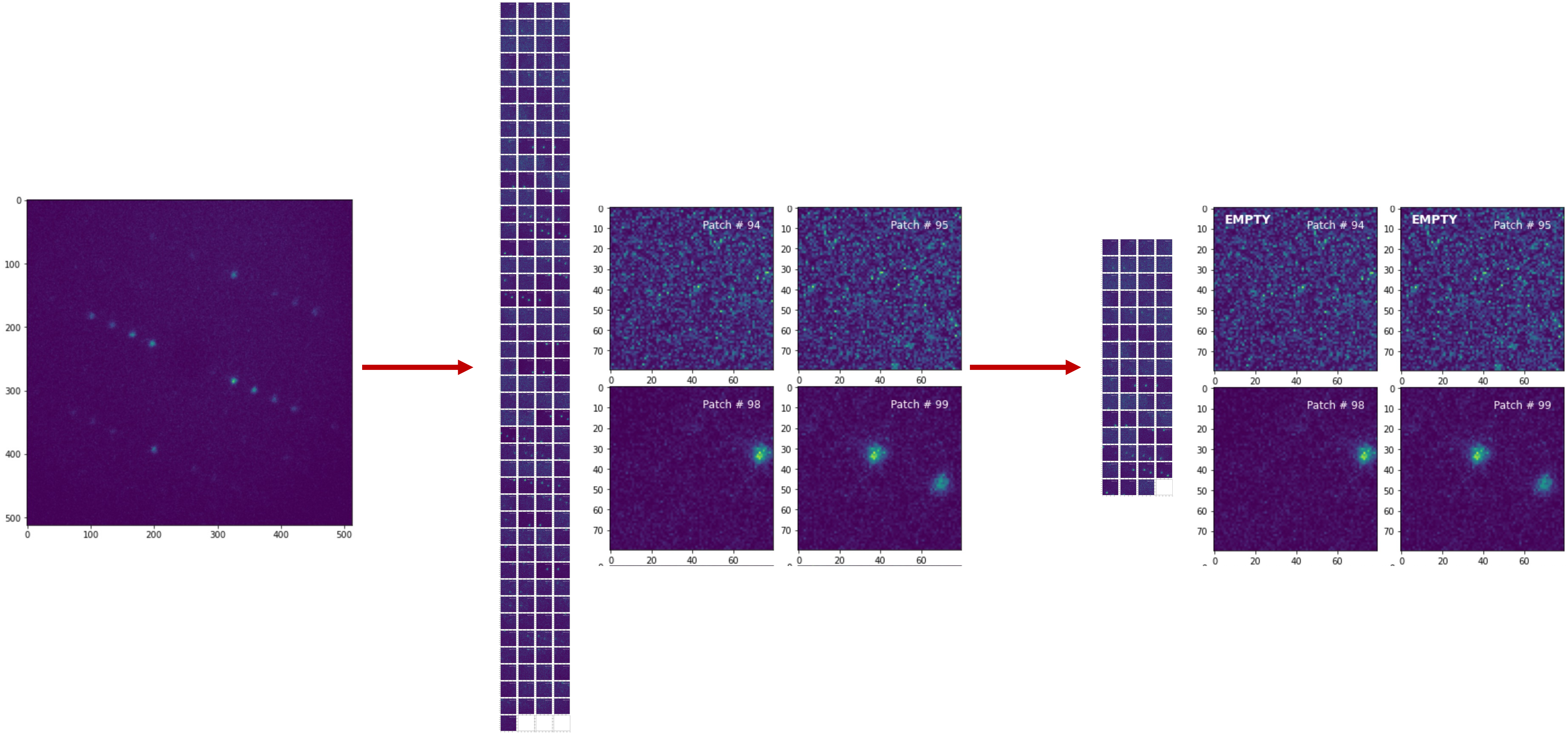
$$IPR = \sum_{i=1}^{N} f(x)^2$$

For white noise, all frequencies contribute equally so f(x) has the same value for all x then:

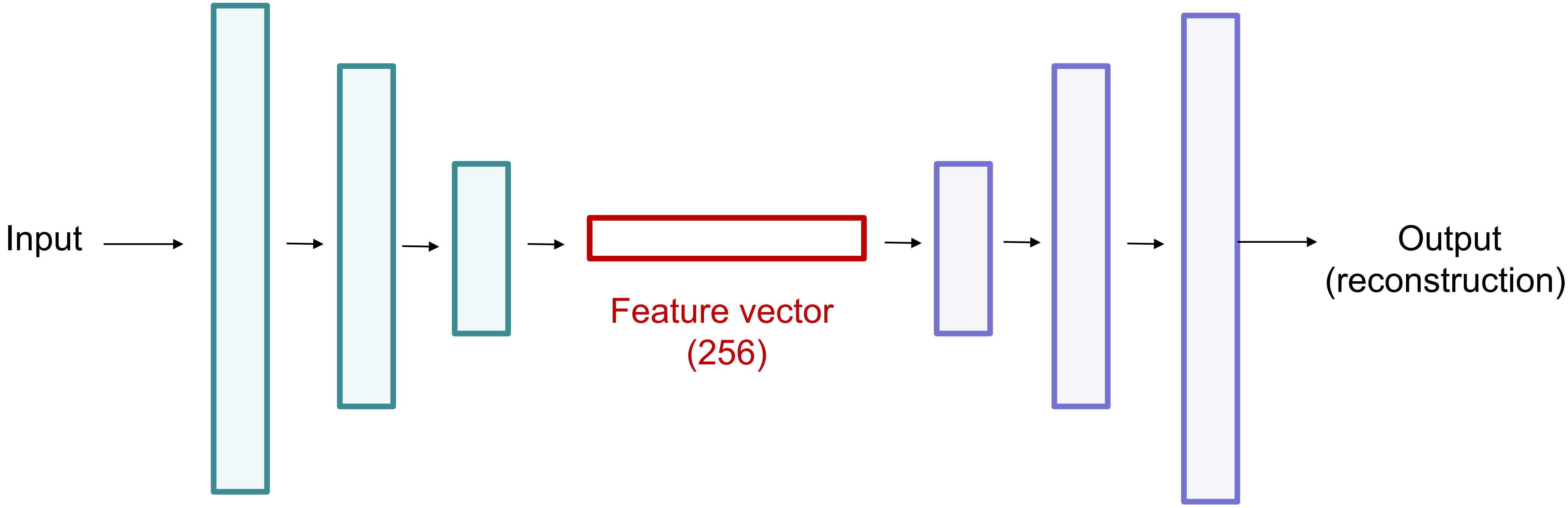
$$f_i(x) = 1/N \Rightarrow IPR = \sum_{i=1}^{N} 1/N^2 = 1/N$$

We do the FFT of the tile, calculate the IPR and if it is equal to 1/N the tile is not included in the dataset for the autoencoder.

# Autonomous anomaly detection: preprocessing



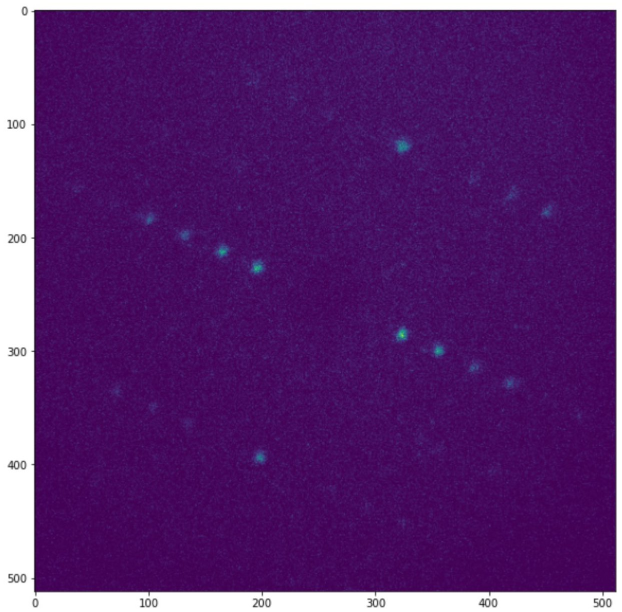
# Autonomous anomaly detection: autoencoder

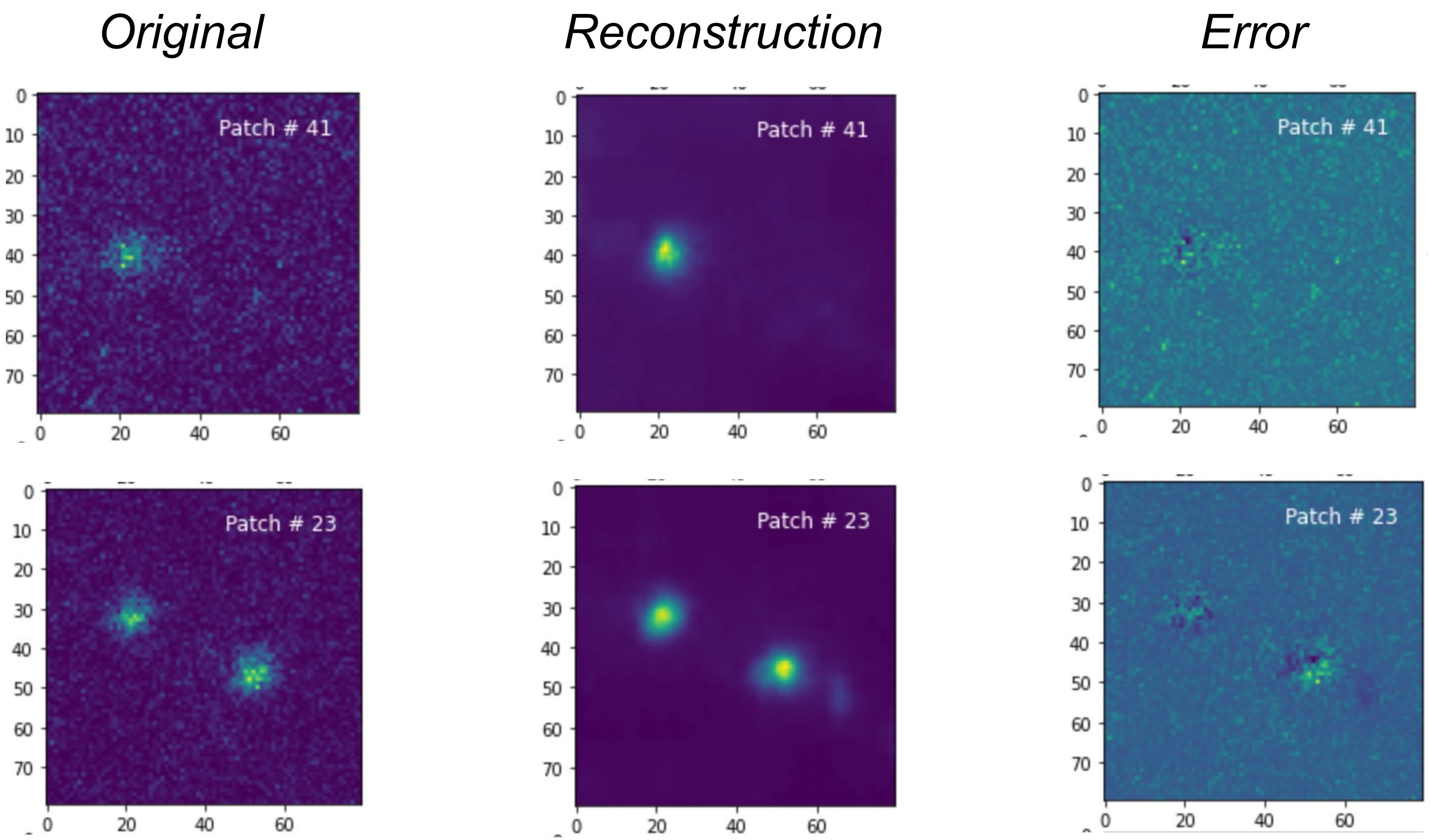



- Each layer of the encoder: Conv2d with relu activation followed by MaxPool.
- MSE loss is used, model trained for 100 epochs with 3789 diffraction patterns.
- Dataset is split 10-10-80 for test-validation-training.

# Autonomous anomaly detection: autoencoder

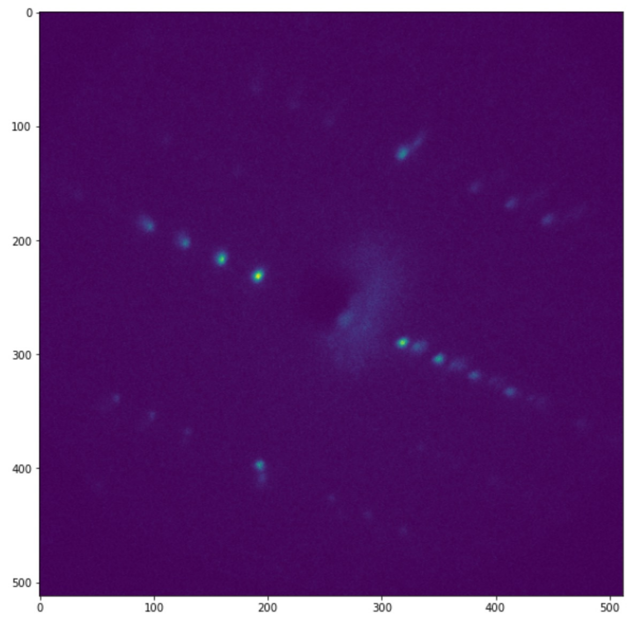
- The autoencoder performs very well and is trained in 100 epochs.
- It also served to denoised the images (which we plan to explore further)



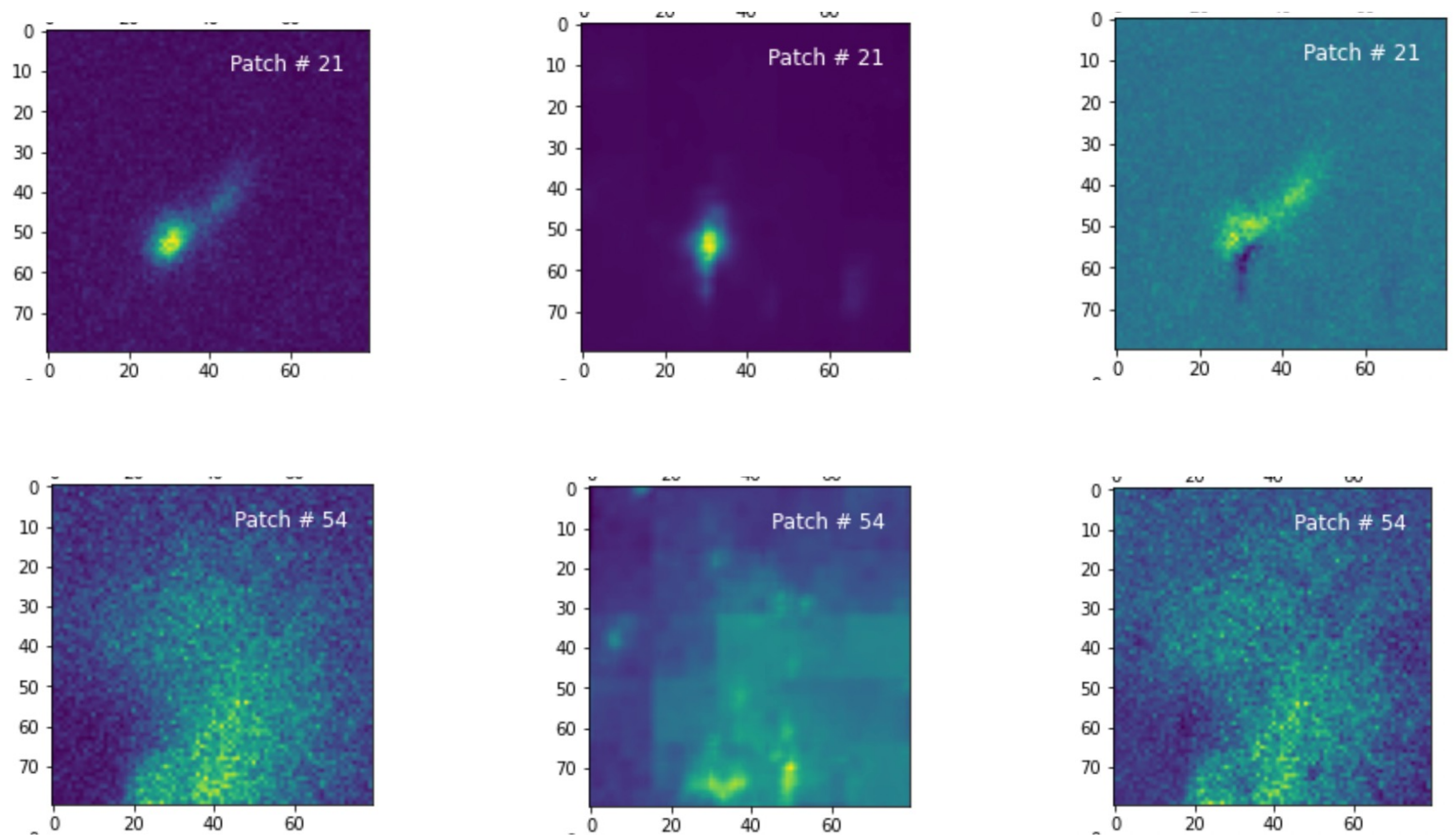


# Autonomous anomaly detection: autoencoder

- Recognizable features of anomalies are not well reconstructed:

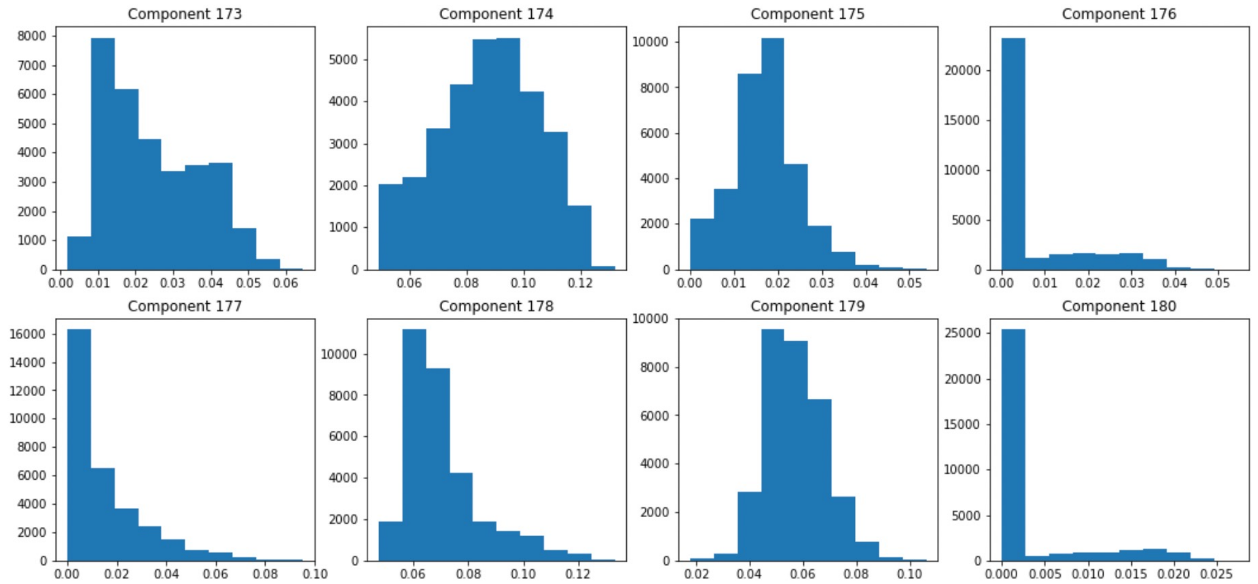


*Original* *Reconstruction* *Error*



# Autonomous anomaly detection: autoencoder

- We are exploring the best strategy for recognizing anomalies.
- Feature vector is not highly dimensional, but unknown distribution
- Reconstruction error is highly dimensional but probably multivariate Gaussian distribution.
- One- class SVM has the ability to learn new types of anomalies but likely requires dimensionality reduction.



# Progress on connection to ALCF

- We have allocation at Theta and ThetaGPU for this experiment.
- A computer will be installed in UED rack (thanks Bob Malone for the help!)
- Plan in place for synchronized measurements (more on this in UE117 update) (thanks Bob Malone for the help!)
- We have C-AD VPN access for facility (and once again, thanks Bob Malone for the help!)
- Tom Uram of ALCF helping on connection setup for data transfer.
- Model is being tested / trained in ThetaGPU with greatly reduced training time.
- We can visualize results and analyze data using Jupyter Notebook running on ALCF.

# Plans for second year of experiment

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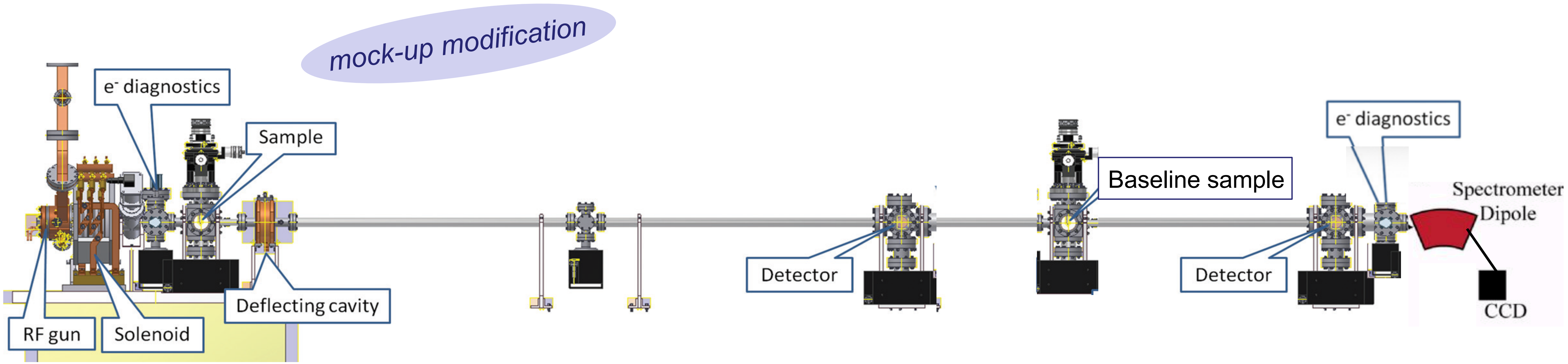
- **Collect synchronized single shot diffraction patterns.**
- Pattern analysis to be implemented in Python available for users to run (ideally on denoised images).
- Leverage existing Python packages such as scikit-ued for analysis.
- Test clustering methods for Au measurements during beam instabilities.
- Continue autonomous detection of anomalies.
- Test near real-time model training and inference using ALCF resources.

# Plans for second year of experiment: beamline extension






- Beamline extension plans are underway.
- Shot-to-shot control of the beam can be implemented using this diagnostic.
- This will provide a novel diagnostic technique and concurrent patterns can be used to ‘normalize’.

# Plans for second year of experiment: vanadium dioxide

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- **Samples in collaboration with Edwin Fohtung (Rensselaer Polytechnic Institute), also in collaboration with Robert Hull (RPI), John Gordon (LANL), and David Clark (LANL).**

- Study ultrafast dynamics resulting from an ultrafast disordering of the V-V dimers forming the M1 phase.

- Experiments will be conducted at low temperature (T ~ 100K) to probe the transient isostructural phase which mediates the transition between the structurally-distinct equilibrium phases.

- Challenges that could be addressed in this study:

  - Are there low energy states that mediate electronic switching in $VO_2$?
  - On what timescales do these metastable states exist?
  - How do such intermediate states determine the timescales and energy costs associated with MIT and switching?

# Non-destructive real-time diagnostic

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**Ultimate goal: achieve single-shot capabilities for MUED**

- We measured 2 baseline samples during our beamtime in integrated and single shot modes.
- We constructed an ML model for reconstruction / denoising of diffraction patterns.
- We are developing a procedure for autonomous anomaly detection.
- We started the connection for data transfer to ALCF.
- We will continue to develop ML/AI models for denoising.
- We have a plan in place for synchronized measurements of diffraction pattern – beam status.
- For our second year, we plan to advance with the beamline extension.
- We will also add a material with interesting physics $VO_2$ to our experiment.

# Summary of products delivered in 2021

**Conference paper:**

- M. Fazio, S. G. Biedron, M. Martinez-Ramon, D. Monk, S. I. Sosa, T. Talbott, M. Babzien, K. Brown, M. Fedurin, J. Li, M. Palmer, J. Tao, J. Chen, A. J. Hurd, N. Moody, R. Prasankumar, C. Sweeney, D. E. Martin, M. E. Papka, *Towards a Data Science Enabled MeV Ultrafast Electron Diffraction System*. In: Proceedings of the 2021 International Particle Accelerator Conference, www.JACoW.org, paper MOPAB286.

**Conference presentations:**

- M. Fazio, S. G. Biedron, M. Martinez-Ramon , S. I. Sosa, oral poster presentation: *MeV class electron beams for ultrafast diffraction*, PPC/SOFE 2021, Denver, Colorado, virtual conference.
- M. Fazio, S. G. Biedron, M. Martinez-Ramon, D. Monk, S. I. Sosa, T. Talbott, M. Babzien, K. Brown, M. Fedurin, J. Li, M. Palmer, J. Tao, J. Chen, A. J. Hurd, N. Moody, R. Prasankumar, C. Sweeney, D. E. Martin, M. E. Papka, oral poster presentation: *Towards a data science enabled MeV ultrafast electron diffraction system*, 2021 International Particle Accelerator Conference, May 24 - 28, Iguazu, Brazil, virtual conference.
- M. Fazio, S. Biedron, D. Monk, M. Martinez-Ramon, S. Sosa, D. Martin, M. Papka, M. Babzien, K. Brown, M. Palmer, J. Tao, A. Hurd, J. Chen, R. Prasankumar, C. Sweeney, oral presentation: *Progress towards a rapid-throughput MeV ultrafast electron diffraction system*, American Physical Society March Meeting, March 15 - 19, virtual conference.

# Thank you for your attention

We want to thank the ATF BNL team for their support during our beamtime.