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DATA ANALYSIS AND CONTROL OF AN MeV ULTRAFAST ELECTRON DIFFRACTION SYSTEM USING MACHINE LEARNING *

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Abstract

An MeV ultrafast electron diffraction (MUED) instrument system is a unique characterization technique used to study ultrafast processes in a variety of materials systems by a pump-probe method. This relatively young technology can be advanced further into a turnkey instrument by using data science and artificial intelligence (AI) mechanisms in conjunction with high-performance computing. This can facilitate automated operation, data acquisition, and real-time or near-real-time processing. The AI-based system controls can provide real-time feedback on the electron beam, or provide virtual diagnostics of the beam. Deep learning can be applied to the MUED diffraction patterns to recover valuable information on subtle lattice variations that can lead to a greater understanding of a wide range of material systems. A data-science-enabled MUED facility will also facilitate the application of this technique, expand its user base, and provide a fully automated state-of-the-art instrument. Updates on research and development efforts for the MUED instrument in the Accelerator Test Facility of Brookhaven National Laboratory are presented.

INTRODUCTION

MeV ultrafast electron diffraction (MUED) system is a pump-probe characterization technique for studying ultrafast processes in materials. The use of relativistic electron beams leads to decreased space-charge effects compared to typical ultrafast electron diffraction experiments employing energies in the keV range [1-3]. MUED has a higher scattering cross section with material samples as compared to other probes such as X-ray free electron lasers, and as such allows access to higher-order reflections in the diffraction patterns due to the short electron wavelengths.

However, this is a relatively young technology, and several factors contribute to making it challenging to utilize, such as beam instabilities that can lower the effective spatial and temporal resolution. In recent years, machine learning (ML) approaches to materials and characterization techniques have provided a new path towards unlocking new physics by improving existing probes and increasing the user's ability to interpret data. Ideally, anomalous contribution detection and removal should not require *a priori* knowledge of what those contributions would be or how they would present themselves in the data. Particularly, with proper preprocessing, ML methods can be employed to control characterization probes in near-real time, acting as virtual diagnostics, or ML can be deployed to extract features and effectively denoise data. With respect to denoising, convolutional neural network architectures, such as auto encoder models, are an attractive and more powerful alternative to conventional denoising techniques. The autoencoder models provide a method of unsupervised learning of latent space representation of data that can help reduce the noise in the data. It should be noted that noise and anomalies aren't necessarily the same thing, as systematic stochastic noise issues may be present. In principle, AI/ML can facilitate distinguishing both.

By supplying a paired training dataset of "noisy" and "clean" data, these ML models can denoise measurements quite effectively [4, 5]. This method relies on the existence of an ideal dataset with no noise, which can be obtained by simulation or by averaging existing noisy datasets. However, in some cases these are not accessible or practical to use. Generative adversarial networks (GANs) are a more suitable option when no "clean" data are available and have been proven to perform well for blind image denoising [6]. They can be trained to estimate and generate the noise distribution, thus producing paired training datasets that can be fed to an autoencoder model. These approaches can lead to increased resolution if employed to denoise, for example, diffraction patterns. In addition, deep convolutional neural network architectures can be used for data analysis. Laanait *et al*. measured diffraction patterns of different oxide perovskites using scanning transmission electron microscopy and, by applying a custom ML algorithm, were able to invert the materials structure and recover 3-dimensional atomic distortions [7]. ML has yet to be applied to the MUED technique, where it can certainly enable advances that can further understanding of ultrafast material processes in a variety of systems.

EXPERIMENTAL

The MUED instrument is located at the Accelerator Test Facility at Brookhaven National Laboratory. A schematic of the experimental setup is presented in Fig. 1. The details of data collection are very briefly described here. The femtosecond electron beam is generated using a frequency-tripled Ti:Sapphire laser that illuminates a copper photocathode, generating a high brightness beam. The electrons are bunched in a 1.6-cell rf cavity and accelerated to 5 MeV. Current parameters of the electron beam source optimized for stability are presented in Table 1. The sample

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chamber is located downstream from the source with a motorized holder for up to nine samples with cryogenic cooling capabilities and a window to allow laser pumping of the material. The detector system is placed 4 m downstream of the photocathode to collect the diffraction patterns. The detector consists of a phosphor screen followed by a copper mirror (with a hole for non-diffracted electrons to pass through) and a CCD Andor camera of 512 pixels × 512 pixels with a large aperture lens. Suitable material systems for MUED require careful preparation with typical lateral sizes of 100-300 µm and roughly < 100 nm thickness to assure electron transparency. Laser fluency is adjusted to avoid radiation-induced damage to the sample.

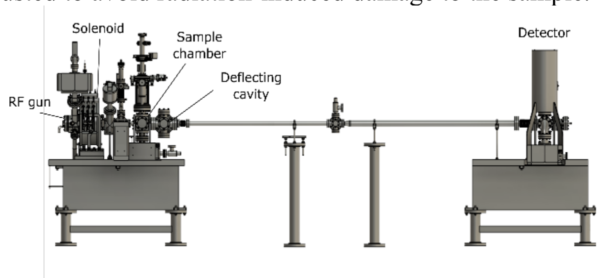

Figure 1: MUED beamline schematic.

Table 1: MUED Source Parameters for Typical Operation

Beam Energy	**3 MeV**
Electrons per pulse	1.25×10^6
Temporal resolution	180 fs
Beam diameter	100-300 µm
Repetition rate	5-48 Hz
Electron fluence	88-880 $s^{-1}\mu m^{-2}$

A schematic of the data pre-processing for ML application for noise detection and removal is presented in Fig. 2. A given image (dataset) is divided into an array of tiles in Fig. 2(a). Noting that for N samples with white noise all frequencies contribute equally to a function, these tiles are examined for those having an inverse participation ratio (IPR) value of 1/N. The IPR is a measure of the contribution of each frequency (in this case spatial). These tiles are ignored. The resulting image is shown in Fig. 2c.

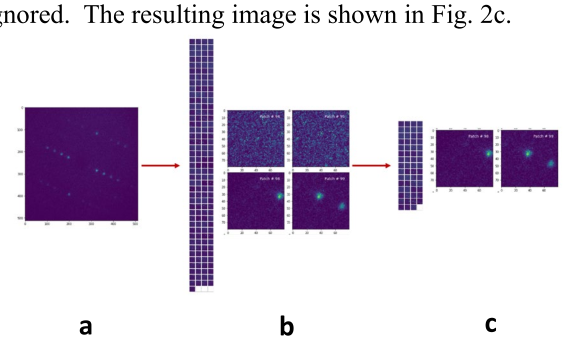

Figure 2: ML schematic for data denoising.

CONCLUSIONS AND FUTURE PLANS

MeV ultrafast electron diffraction (MUED) is a pump-probe system to measure dynamic material structure evolution in the time range from femtoseconds to nanoseconds. A convolutional autoencoder model was developed to reconstruct large sets of diffraction patterns. The model trained on all data (unsupervised). An anomaly was found to produce a large reconstruction error or different feature vector values. Different strategies to detect anomalies were also tested. Anomaly detection is ongoing, and multiple approaches are being considered. The large datasets expected from the ATF are well suited for data analysis on a high-performance computing system, such as at the Argonne Leadership Computing Facility, located at Argonne National Laboratory. There is an existing account at THETA and THETAGPU for this work.

COVID restrictions have had a significant impact on in-person testing and experiments and access to resources. With the restrictions now lifted, beam time at the facility has resumed, and future visits are in the planning process. There have been three talks resulting from this work [8-11]. A manuscript is in preparation on unsupervised anomaly detection for MeV ultrafast electron diffraction. Applications of ML combined with MeV ultrafast electron diffraction at facilities such as the ATF are expected to encompass not only materials science; interest has been expressed in global security challenges such as pandemics.

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