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TOWARDS A DATA SCIENCE ENABLED MeV ULTRAFAST ELECTRON DIFFRACTION SYSTEM

M. A. Fazio*, S. G. Biedron[1,2], M. Martínez-Ramón, D. Monk, S. I. Sosa
Department of Electrical and Computer Engineering, University of New Mexico, Albuquerque, NM 87131, USA
T. Talbott
Department of Mechanical Engineering, University of New Mexico, Albuquerque, NM 87131, USA
M. Babzien, K. Brown, M. Fedurin, J. Li, M. Palmer, J. Tao
Brookhaven National Laboratory, Upton, NY 11973, USA
J. Chen, A. J. Hurd, N. Moody, R. Prasankumar, C. Sweeney
Los Alamos National Laboratory, Los Alamos, NM 87545, USA
D. E. Martin, M. E. Papka[3]
Argonne National Laboratory, Lemont, IL 60439, USA
[1]also at Department of Mechanical Engineering, University of New Mexico, Albuquerque, NM 87131, USA
[2]also at Element Aero, Chicago, IL 60643, USA
[3]also at Department of Computer Science, Northern Illinois University, DeKalb, IL 60115, USA

Abstract

A MeV ultrafast electron diffraction (MUED) instrument is a unique characterization technique to study ultrafast processes in materials by a pump-probe method. This relatively young technology can be advanced further into a turn-key instrument by using data science and artificial intelligence (AI) mechanisms in conjunctions with high-performance computing. This can facilitate automated operation, data acquisition and real-time or near-real-time processing. AI-based system controls can provide real-time feedback on the electron beam which is currently not possible due to the use of destructive diagnostics. 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. We will discuss the progress made on the MUED instrument in the Accelerator Test Facility of Brookhaven National Laboratory.

INTRODUCTION

MeV ultrafast electron diffraction (MUED) is a pump-probe characterization technique for studying ultrafast processes in materials. The use of relativistic beams leads to decreased space-charge effects compared to typical ultrafast electron diffraction experiments employing energies in the keV range [1, 2]. Compared to other ultrafast probes such as X-ray free electron lasers, MUED has a higher scattering cross section with material samples and 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 which can lower the effective spatial and temporal resolution.

In the past 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. In particular, 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 acquired data. In this later case, convolutional neural network architectures such as autoencoder 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. By supplying a paired training dataset of noisy and "clean" data, these ML models can denoise measurements quite effectively [3, 4]. 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 [5]. 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

* mfazio@unm.edu

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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 [6]. ML has yet to be applied to the MUED technique, where it can certainly enable advances that can further our understanding of ultrafast material processes in a variety of systems.

EXPERIMENTAL

The MUED instrument is located in the Accelerator Test Facility at Brookhaven National Laboratory. A schematic representation of the experimental setup is presented in Fig. 1. The femtosecond electron beams are generated using a frequency-tripled Ti:Sapphire laser that illuminates a copper photocathode generating a high brightness beam. The electrons are then accelerated and compressed in a 1.6-cell RF cavity achieving energies up to 5 MeV. Current parameters of the electron beam source optimized for stability are presented in Table 1. The sample 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. Next to the chamber a RF deflecting cavity is located and 4 m downstream the detector system is placed 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) and a CCD Andor camera of 512 pixels x 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 of the probed material.

Table 1: MUED Source Parameters for Typical Operation

Beam energy:	3 MeV
Number of electrons per pulse:	1.25×10^6
Temporal resolution:	180 fs
Beam diameter:	100 - 300 µm
Repetition rate:	5 - 48 Hz
Number of electrons per sec per µm^2:	88 - 880

FUTURE PLANS

Due to the impact of the COVID pandemic on research facilities, no experiments have yet been carried out the MUED instrument. When beamtime becomes available for the authors, measurements on the following material systems are planned: black phosphorus, graphite and polycrystalline gold films. These material systems have been previously investigated using keV-ultrafast electron diffraction under the same photoexcitation employed in the MUED instrument [7–10]. The lattice dynamics and electron-phonon coupling effects on these systems are of itself significant phenomena to study using MUED. However, our focus will be on implementing ML methods to increase the resolution of single shot measurements. Currently, diffraction patterns are obtained by averaging over multiple shots to minimize the effect of instabilities such as fluctuations in the electron beam energy. We propose to apply ML methods such as autoencoders, with or without a GAN model, to estimate the noise distribution in order to denoise single shot diffraction patterns. We will produce our input training datasets from single shot measurements of the different materials listed above. Paired datasets can be constructed by averaging over several shots or utilizing simulations for constructing the diffraction patterns. A GAN model will be implemented to attempt blind denoising following the work of Chen *et al.* [5]. We will also explore active learning implementation of the denoising algorithms to provide users with near-real-time single shot diffraction patterns that could be analysed on the fly. As a large amount of data is expected to be generated, we will also incorporate existing visualization and data management tools, such as Cinema:Bandit [11] for prompt display after initial processing.

Accessing single shot capabilities of the MUED instrument can not only provide significant insights into material processes and higher spatial resolution in the measurements, but they can also be employed to estimate electron beam parameters. Yang *et al.* demonstrated that by analysing the Bragg peaks the shot-to-shot energy fluctuation and the spatial-pointing jitter can be measured, thus determining the electron beam energy spread for each pattern [12]. The features in the diffraction pattern could be extracted effectively by a convolutional neural network, which when implemented after denoising the images can serve as a nondestructive probe of the electron beam. The convolutional neural network will take denoised single shot diffraction patterns as input and output electron beam characteristics, such as energy spread. Single shot diffraction patterns can then be filtered according to beam energy leading to increased resolution in the technique as the effects of beam fluctuations are minimized.

Our work will also focus on the analysis of diffraction patterns. Deep learning algorithms can be implemented to attempt structural reconstruction based on the work by Laanait *et al.* [6] to understand subtle lattice changes induced by the laser pumping. In addition, search and match protocols can be put in place leveraging existing databases of density functional calculations. Bridging computing resources between user facilities will be a priority to facilitate supercomputing and cloud computing, enabling near-real-time analysis that can steer experiments and provide relevant analysis during limited time in the MUED instrument.

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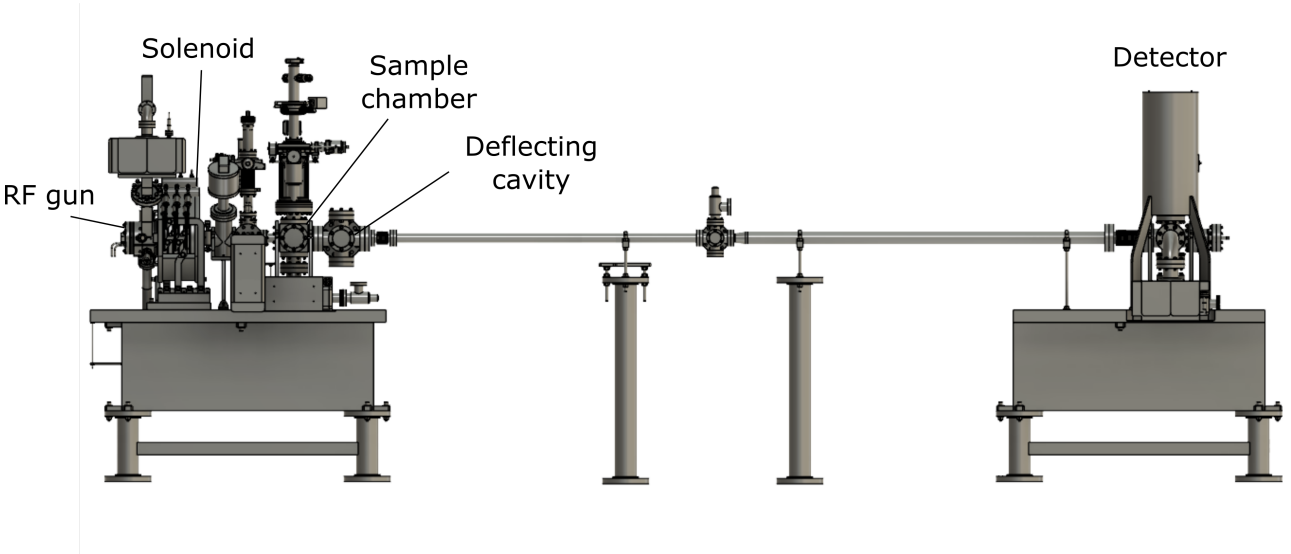

Figure 1: Schematic representation of the experimental setup for the MUED instrument located in the Accelerator Test Facility at Brookhaven National Laboratory.

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