# CNM Workshop 4: Probing Fast and Ultrafast Dynamics with Time-resolved Electron Microscopy Spanning from Microseconds to Femtoseconds

**Wednesday, May 7, Morning**

**Session I: Ultrafast Electron Microscopy**

8:00 – 8:10 Ilke Arslan (Physical Sciences and Engineering, Argonne National Laboratory)
*Opening Remarks*

8:10 – 8:35 David Flannigan (Department of Chemical Engineering and Materials Science, University of Minnesota)
*UEM Imaging of Coherent-phonon and Order-parameter Dynamics*

8:35 – 9:00 Suman Kumari (Argonne National Laboratory)
*Unfolding the Dynamics of Materials via Single-shot Electron Microscopy*

9:00 – 9:20 Haihua Liu (Center for Nanoscale Materials, Argonne National Laboratory)
*Nanoscale Imaging of Surface Plasmons by Ultrafast Electron Microscopy*

9:20 – 9:40 Burak Guzelturk (X-ray Science, Argonne National Laboratory)
*Ultrafast Electron Diffraction on Nanocrystals: From Phonons to Polarons and Symmetry Transformations*

9:40 – 10:00 Bolin Liao (Department of Mechanical Engineering, University of California-Santa Barbara)
*Spatial-temporal Imaging of Photocarrier Dynamics Using Scanning Ultrafast Electron Microscopy*

10:00 – 10:30 Break

**Session II: Ultrafast Science**

10:30 – 10:55 Haidan Wen (Materials Science, Argonne National Laboratory)
*Ultrafast Spin-shear Coupling in van der Waals Antiferromagnets*

10:55 – 11:20 Paul Evans (Materials Science and Engineering, University of Wisconsin-Madison)
*Ultrafast Magnetic and Ferroelectric Dynamics in Functional Complex Oxides*

11:20 – 12:00 EM Tour – Building 216/212

12:00 – 1:30 Lunch Break

**Wednesday, May 7, Afternoon**

**Session III: *In-situ* Electron Microscopy**

1:30 – 1:55 Judith Yang (Center for Functional Nanomaterials, Brookhaven National Laboratory)
*Dynamics of the Early Stages of Metal and Alloy Oxidation*

1:55 – 2:20 Qian Chen (Department of Materials Science and Engineering, University of Illinois Urbana-Champaign)
*Liquid-phase Electron Microscopy and its Automation for Colloidal Nanoparticles*

2:20 – 2:40 Yuzi Liu (Center for Nanoscale Materials, Argonne National Laboratory)
*Multiscale* In-situ *Microscopy for Energy Materials*

2:40 – 3:00 Paul Voyles (Department of Materials Science and Engineering, University of Wisconsin-Madison)
*Heterogeneous Dynamics and Crystallization of Supercooled Metallic Liquids from* In-situ *5D STEM*

3:00 – 3:30 Break

**Session IV: Electrically Driven Dynamics**

3:30 – 3:55 Spencer Reisbick (Condensed Matter Physics and Materials Science, Brookhaven National Laboratory)
*Elucidation of Strongly Correlated Dynamics Using Electrically Driven Ultrafast Electron Microscopy*

3:55 – 4:20 Chris Regan (Department of Physics and Astronomy, University of California-Los Angeles)
*Comprehensive, High-resolution Mapping of Ferroelectric Fields in Hafnium Zirconium Oxide*

4:20 – 4:40 Daniel B. Durham (Center for Nanoscale Materials, Argonne National Laboratory)
*Nanosecond Electron Microscopy of Electrically Triggered Material Dynamics*

4:40 – 5:00 Workshop Organizers
*Closing Discussion*

5:00 Adjourn

# UEM Imaging of Coherent-phonon and Order-parameter Dynamics

David J. Flannigan[1], Jialiang Chen[1], Yichao Zhang[1,2], Alfred Zong[3], and Michael Zuerch[4]

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Ultrafast photoexcitation of coherent phonons is driven by an impulsive, collective displacement of constituent atoms from their average equilibrium lattice positions [1]. Models describing the generation of coherent acoustic modes typically invoke the creation of an anisotropic strain profile arising from the relatively instantaneous absorption of an ultrafast laser pulse [2]. If the skin depth is shallow relative to the specimen thickness, a steep tensile strain gradient perpendicular to the surface $\left(\frac{\partial \varepsilon}{\partial z}\right)$ results. Initial relaxation occurs via rapid contraction of the surface layers followed by subsequent coherent oscillations of the lattice and launch of a train of coherent elastic strain waves [i.e., coherent acoustic phonons (CAPs)]. Macroscopically, responses are generally well-described by constitutive relations as gleaned from data gathered using ultrasonic methods or ultrafast spectroscopies. At the atomic to nanoscale level, individual lattice discontinuities and their impact on CAP behaviors can be modeled using multiscale methods [3,4]. Further, average unit-cell level responses on ultrafast timescales can be probed using femtosecond electron and x-ray scattering [5,6].

Here we discuss our development and use of 4D ultrafast electron microscopy (UEM) [7-9] for the study of CAP dynamics in a variety of material types and structures. One of the main objectives has been to explore and establish the spatiotemporal limits of UEM bright-field imaging in a thermionic-based TEM [10]. In particular, emphasis has been placed on probing CAP dynamics – including generation, launch, scattering, and decay behaviors – at individual lattice defects. Select examples will be presented here and will include the direct imaging of (i) sub-picosecond nucleation and launch of CAPs from individual crystal surface steps in layered materials [11], (ii) propagation and time-domain dispersion behaviors of hypersonic CAPs in diamond cubic materials [12,13], (iii) distinct precursor thermalization prior to CAP launch in iron pnictides and metal nanoparticles [14,15], (iv) localized hot-spot generation at nanoparticle-nanoparticle interfaces [15], and (v) distinct CAP decoherence and softening in the vicinity of individual few-layer crystal step edges [16-18]. Following this, we will discuss our study of domain dynamics of polar textures in $PbTiO_3/SrTiO_3$ superlattices using UEM imaging. Specifically, we will discuss our findings of preferential domain melting and subsequent growth via boundary propagation by textures that persist following fs photoexcitation.

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# Unfolding the Dynamics of Materials via Single-shot Electron Microscopy

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The development of nanosecond and faster photoemission electron sources offers the chance to move the high spatial resolution world of electron microscopy into the ultrafast world of materials dynamics. The interaction of the materials subjected to either a pulsed or a continuous laser beam generates extremes of temperature and pressure in the material. Therefore, the incorporation of high temporal probing capabilities and lasers to the TEMs has been a critical development required to discover new knowledge about the dynamic evolution of materials on varying time and length scales. In particular, the single-shot approach enables the capture of various stages of irreversible processes with high temporal resolution, providing critical understanding of how dynamic processes unfold in materials. In this presentation, examples of single-shot electron microscopy studies will be presented for selected systems. Short lived transient processes involved in dynamic processes in materials will be discussed to obtain new insights into their evolution.

# Nanoscale Imaging of Surface Plasmons by Ultrafast Electron Microscopy

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Ultrafast Electron Microscopy (UEM) is one powerful tool to study the ultrafast dynamics of light-matter interactions in physics, chemistry, and materials science with temporal resolution of femtoseconds, which is 10 orders better than that of conventional electron microscopy limited by the camera read-out rate [1]. We established one state-of-art UEM user facility at the Center for Nanoscale Materials (CNM), Argonne National Laboratory. Besides the capabilities of imaging and diffraction, the UEM at the CNM was equipped with one GIF spectrometer, which enables it to work in energy filtering mode and characterize the near field of surface plasmon resonance [2]. Surface plasmons, collective oscillating charge at the surface of metals or interface of metals/dielectrics under light excitation, are among the most attractive candidates for next-generation information revolution due to their ability to extremely confine electromagnetic field and empower strong coupling of light and matter beyond the diffraction limit [3]. Replacing electronic signals with light pulses as information carriers is a prime motivation behind research on plasmonics and nanophotonics. Therefore, direct imaging of the plasmonic near field and its coupling between photonics circuit components or plasmonic nanostructures at the nanoscale is critical for nanophotonics applications, while scientists in this field mainly rely on theoretical simulation of the plasmonic field by FDTD or other programs. Here, we investigated the excitation and manipulation of Localized Surface Plasmon Resonances (LSPR) at surface of metal nanoparticles and propagating surface plasmon polaritons (SPPs) at the interface of metal and dielectric by UEM and their dependence on laser wavelength, polarization, fluence at nanometer scale and sub-picosecond time scale. The nanoscale near field detection of surface plasmons is critical for the biological sensing and SERS methods, plasmonics, and nanophotonics applications, quantum information processing.

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# Ultrafast Electron Diffraction on Nanocrystals: From Phonons to Polarons and Symmetry Transformations

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Ultrafast electron diffraction (UED) has emerged as a powerful tool for capturing structural dynamics in nanocrystalline materials, revealing fundamental processes such as electron-phonon coupling, polaron formation, and excited-state symmetry transformations [1-5]. In this talk, I will discuss how UED provides critical insights into these ultrafast phenomena, offering a unique perspective beyond conventional optical characterization techniques.

First, I will describe how Debye-Scherrer diffraction ring analysis enables tracking of unit cell dynamics upon photoexcitation. By analyzing shifts in diffraction ring intensity and position—indicative of bond length variations and mean-squared atomic displacements—we can quantitatively extract electron-phonon coupling strength [1-4]. This coupling is key to understanding hot-carrier properties of metallic and semiconductor nanocrystals, essential for applications in photocatalysis and photochemistry.

Next, I will highlight how atomic pair distribution function analysis reveals ultrafast, short-range structural distortions in photoexcited nanocrystals. I will show evidence of local lattice deformations in core/shell nanocrystals driven by small polaron formation [2]. This finding sheds light on the microscopic mechanism behind surface charge trapping in nanocrystals, a key factor influencing device performance in light-emitting diodes, photodetectors and related optoelectronic technologies.

Finally, I will demonstrate how Debye scattering equation modeling, combined with UED, allows for tracking structural symmetry in nanocrystals on picosecond timescale. I will show that PbS quantum dots undergo symmetry transformations driven by excited-state electronic carriers [5]. This discovery opens new avenues for controlling electronic and optical properties through photoinduced symmetry switching.

Overall, these studies illustrate the power of UED in capturing non-equilibrium structural dynamics in nanocrystals, deepening our understanding of their functional properties and expanding their potential applications.

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# Spatial-temporal Imaging of Photocarrier Dynamics Using Scanning Ultrafast Electron Microscopy

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The spatial-temporal dynamics of photoexcited charge carriers in materials are crucial for a wide range of applications including photovoltaics, photocatalysis, and optoelectronics. To fully resolve the microscopic details of photoexcited carrier dynamics, a combination of high spatial and temporal resolution is required. Scanning ultrafast electron microscopy (SUEM) is a promising photon-pump-electron-probe technique that is highly sensitive to surface photocarrier dynamics with simultaneously high spatial and temporal resolutions. In this talk, I will discuss the recent development of SUEM at UCSB and showcase its capabilities by describing some recent applications of SUEM to image photocarrier dynamics in a variety of technologically relevant materials, including semiconductors and Mott insulators.

*The initial development of SUEM at UCSB was supported by the DOE Early Career Faculty Program under the award number DE-SC0019244. Current applications of SUEM are supported by ARO under the award number W911NF2310188 and AFOSR under the award number FA9550-22-1-0468.*

# Ultrafast Spin-shear Coupling in van der Waals Antiferromagnets

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Van der Waals magnets possess unique structural and spin anisotropies that lead to pronounced magnetoelastic interactions. By employing ultrafast electron diffraction and microscopy, we discover a strong coupling between interlayer shear and magnetic order in vdW antiferromagnets. On the picosecond timescale, a seesaw-like motion of the reciprocal lattice is observed in $FePS_3$, exhibiting a thirty-fold amplification when the sample is cooled below the Néel temperature [1]. This rotation in reciprocal space results from an unusually large interlayer shear in real space, where individual micro-patches of the film behave as synchronized shear oscillators along the same in-plane axis. Ultrafast electron microscopy further reveals shear acoustic harmonics up to the fourth order arising from structural heterogeneities [2]. On nanosecond timescales probed by time-resolved x-ray scattering, the recovery of lattice shear and magnetic order concurrently slow down at the Néel temperature, sharing identical critical behaviors [3]. Time-dependent Ginzburg-Landau theory indicates that this concurrent critical slowing originates from a linear coupling between interlayer shear and magnetic order, dictated by the broken mirror symmetry intrinsic to the monoclinic stacking. Our work provides the first microscopic view of spin-mediated mechanical motion in an antiferromagnet and identifies a novel route towards realizing high-frequency resonators operating up to the millimeter-wave band.

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# Ultrafast Magnetic and Ferroelectric Dynamics in Functional Complex Oxides

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The nanoscale structure of ferroic complex oxide ferroelectric and magnetic thin films is characterized by heterogeneity in the ferroelectricity and magnetism in the form of domains, domain boundaries, and interfaces. These features have profound impacts on dynamical phenomena exhibited at timescales as short as picoseconds. In the particular case of the ferroelectric nanodomain pattern of ferroelectric/dielectric superlattices unique dynamics emerge after femtosecond optical excitation, these dynamics correspond to an oscillation of the polarization and have a timescale set by the spatial period of the superlattice [1]. Similar oscillatory dynamics arise in superlattice systems exhibiting Skyrmion polarization configurations. The development of highly intense x-ray nanoprobes combined with pulsed laser excitation raises the potential to probe similar effects at domain boundaries in ferroelectric and magnetic systems.

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Dynamics of the Early Stages of Metal and Alloy Oxidation

Judith C. Yang[1], Meng Li[1], Brian Lee[1], Dmitri Zakharov[1], Guangwen Zhou[2], Matthew T. Curnan[3], Stephen D. House[4], and Wissam A. Saidi[5]

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Oxidation reactions at the nanoscale are of tremendous importance to energy, corrosion, catalysis, and nano-oxide synthesis for a variety of applications in electronic, magnetic, and optical devices. Furthermore, as dimensions of materials systems approach nanoscale, it is critical to fundamentally understand their interactions with the environment at this length scale for their long-term durability. Limiting oxidation is critical for environmental stability and controlled oxidation is used for nano-oxide processing. Yet, the fundamental understanding of the atomic mechanisms of surface oxidation processes that occur at the nanoscale and below is currently very limited. The recent availability of *in-situ* tools, especially electron microscopy, has enabled the ability to directly observe the structural changes under environmental conditions, leading to an atomic- and nanoscale understanding of the fundamental nature of metal and alloy oxidation. Here, we present our *in-situ* high resolution transmission electron microscopy (TEM) that the formation of epitaxial $Cu_2O$ islands during the transient oxidation of Cu single crystal thin films bear a striking resemblance to heteroepitaxy, where the initial stages of growth are dominated by oxygen surface diffusion and strain impacts the evolution of the oxide morphologies. *To deepen our understanding of the atomic-scale dynamic processes of $Cu_2O$ island formation on Cu during oxidation in situ,* we are *using correlated in-situ environmental high-resolution TEM (ETEM) and atomistic simulations to explain the observed fascinating phenomena on how metals rust.*

# Liquid-phase Electron Microscopy and its Automation for Colloidal Nanoparticles

Qian Chen[1]

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I will present our group's recent progress on establishing and utilizing liquid-phase electron microscopy to image, understand, and engineer synthetic and natural nanoparticle systems, in space and time at a nanometer resolution. This involves systems that underpin the fundamentals of structure–functional relationship for a wide range of phenomena and applications. In this talk, we will discuss in detail two types of such systems. The first focuses on metallic nanoparticles assembling into various complex lattices such as Maxwell lattice, a chiral pinwheel lattice, a colloidal moiré pattern, and nanoparticle swarms as promising optical and mechanical metamaterials. The second is on our efforts to couple liquid-phase TEM with electron tomography and machine learning, to visualize the dynamic reactions of nanoparticles in 3D. I will end the talk by discussing the prospects of autonomous electron videography for understanding and discovery of dynamic multifunctional nanoparticle systems in liquid and at operation at the otherwise inaccessible spatiotemporal precision.

# Multiscale *In-situ* Microscopy for Energy Materials

Yuzi Liu[1], Tao Zhou[1], Zhong Hou Cai[1], and Martin Holt[1]

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Integrated correlative electron and x-ray microscopy offers insights through complementary multi-modal approaches across a broad range of length scales, enabling the study of functional materials like catalysts and their operational mechanisms. Data from one imaging platform can pinpoint intriguing sample regions and scientific questions for further exploration on another platform. These complementary techniques help validate findings by cross-checking the underlying science, which can sometimes be skewed by experimental artifacts, instrumentation issues, or beam-induced damage—common challenges with beam-sensitive materials. Here, lithium dendrite plating on Ta-doped Li7La3Zr2O12 (LLZTO) and the structural evolution of cathode materials were examined using *in-situ* transmission electron microscopy (TEM), scanning electron microscopy (FIB-SEM), and hard x-ray nanoprobe microscopy. Together, these methods provide a detailed view of structural and compositional changes across multiple scales.

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# Heterogeneous Dynamics and Crystallization of Supercooled Metallic Liquids from *In-situ* 5D STEM

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Fast readout direct electron cameras enable *in-situ* five-dimensional scanning transmission electron microscopy (5D STEM) with acquisition time per diffraction pattern of tens of microseconds and acquisition time for an entire scan of tenths of a second. This rate is fast enough to capture dynamics of highly viscous supercooled liquids (SCLs). This talk will summarize two examples. The first example is studies of liquid state dynamics directly using electron correlation microscopy (ECM), the electron scattering equivalent of x-ray photon correlation spectroscopy. ECM captures nanometer-length scale variations in the structural relaxation time of SCLs and careful consideration of the scattering angle dependence can be used to separate the dynamics by partial structure factor, proving composition-dependence. The second example is studies of the emergence of crystalline order from the SCL state. For at least one alloy SCL, crystallization occurs in a two-step process involving a disordered precursor state before the emergence of a fully ordered, growing crystal.

# Elucidation of Strongly Correlated Dynamics Using Electrically Driven Ultrafast Electron Microscopy

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At Brookhaven National Laboratory, we have developed a completely electrically driven ultrafast electron microscope (UEM) with the goal of pursuing *operando* time-resolved responses of devices functioning in standard operating conditions for the development of new technology [1,2]. We have expanded our broadly tunable UEM system to cover frequency ranges spanning from 1 Hz to 12 GHz with picosecond temporal resolution. I will discuss the capabilities of our UEM system and the technological advancements made towards nanoscale materials imaging at high spatiotemporal scales. I will also discuss two case studies which have successfully provided direct visualization of strongly correlated systems containing complicated magnetic and crystallographic structure. First, I will discuss the initiation and propagation of spin-waves away from magnetic anti-vortex cores [3]. Here, microwave excitation through an RF antenna is used to disturb the magnetic domain boundaries by inducing an oscillating out-of-plane magnetic field through a soft magnet, creating spin-localized wave fronts. Secondly, I will demonstrate the growth of a switchable domain in materials common for neuromorphic computing [4]. Specifically, our UEM experiments identified that devices in operation undergo structural metastability which remain in a transitionary state during specific experimental parameters (i.e., frequency, voltage, etc.). Once the device receives enough power, a steady state is observed, effectively locking the material out of the switchable state, and preventing high frequency computing capabilities [5].

*The development of the microwave-driven pulsed electron microscopy and part of the microscopy analysis efforts at BNL were supported by DOE-BES, Materials Science and Engineering Division, under Contract No. DE-SC0012704. The materials synthesis and microscopy of frequency-dependent study of VO2 were supported as part of the Quantum Materials for Energy Efficient Neuromorphic Computing (Q-MEEN-C), an Energy Frontier Research Center funded by the U.S. DOE-BES, under Award #DE-SC0019273. S.R. acknowledges AFOSR grant FA9550-23-1-0215 for support pertaining to discussions on RF interactions. This research used nanofabrication facility of the CFN, which is a U.S. Department of Energy Office of Science User Facility at BNL and supported under Contract No. DE-SC0012704.*

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# Comprehensive, High-resolution Mapping of Ferroelectric Fields in Hafnium Zirconium Oxide

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Ferroelectric hafnium zirconium oxide (HZO) could form the basis for the ultimate electronic memory technology, where ones and zeroes are stored by driving individual atoms back and forth inside their unit cells. What stands between the present day and the realization of this idealized vision is the defective, polymorphic nature of the real material. How HZO's ferroelectric phase wakes up, stabilizes, and eventually fatigues is, as yet, poorly understood. Electron beam-induced current (EBIC) imaging in a scanning transmission electron microscope (STEM) quantitatively maps both the remanent and the coercive fields in an operating HZO device with excellent spatial resolution and contrast [1]. STEM EBIC imaging thus provides a picture of HZO's key ferroelectric properties that is unprecedented in its completeness. This picture can be correlated with the physical structure determined via standard STEM, which should, with some more work, allow us to understand how HZO must be managed in a practical memory technology.

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# Nanosecond Electron Microscopy of Electrically Triggered Material Dynamics

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Materials with fast electrical responses (e.g., piezoelectric, ferroelectric, phase transitions, electrochemical) are critical for modern and next-generation electronics. These responses can be dictated by complex nanoscopic dynamics. Visualizing them is important for understanding the rate-limiting steps and for improving the dynamic properties.

I will introduce our new voltage-triggered ultrafast electron microscopy capability at CNM, which enables imaging electrically driven dynamics at both nanometer length scales and nanosecond time resolution [1]. I will present a first demonstration in which we determine the electrical pulse switching mechanism of room temperature charge density waves in 1T-$TaS_2$ [2]. I will then discuss ongoing developments to further improve the accessible length and time scales of electrically driven phenomena, highlighting opportunities for future user collaborations. We anticipate that these capabilities will provide key insights into the mechanisms underlying the dynamic properties of a broad range of electronic materials and nanostructures.

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