In-situ Ultrafast Transmission Electron Microscopy: Advancing Ultrafast Dynamics Research under Multi-Field Coupling at the Nanoscale

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In recent years, the development of ultrafast transmission electron microscopy (UTEM) has created new opportunities for studying dynamic processes at the nanoscale with unprecedented temporal resolution.[1–3] The significant advances in femtosecond and even attosecond temporal resolution are achieved through the integration of the pump-probe principle with transmission electron microscopy (TEM). In this technique, a femtosecond laser pulse is directed at the sample to initiate a transient dynamic event. After a variable time delay, an ultrashort electron pulse captures the sample's transient state. This ultrashort electron pulse is typically generated by photoemission from the cathode of the electron gun, which is illuminated by the femtosecond laser pulse. In UTEM, dynamics are typically triggered by a femtosecond laser pulse, revealing the dynamic response of multiple degrees of freedom in materials and demonstrating the coupling between orbitals, charge, spin, and lattice systems during relaxation processes. The majority of prior studies have focused on the intrinsic dynamic processes of samples in a vacuum under ultrashort laser pulse stimuli. However, in real applications, materials often operate in complex environments or under multifield conditions. Understanding dynamic responses under such conditions and exploring the possibility of manipulating these processes through external fields are crucial for improving material functionality and linking structure-dynamics-function relationships.

Simulating multi-field conditions in TEM has become increasingly feasible due to significant advancements in *in-situ* TEM techniques. Researchers have introduced external fields, such as electric, magnetic, stress, heating/cooling, and various gas or liquid environmental conditions, at the sample holder position to characterize material properties and dynamic responses with nanoscale or even atomic resolution.[4] Substantial progress in *in-situ* TEM research has enabled the visualization of numerous fundamental processes with high spatial resolution.[5–8] From a timescale perspective, structural responses to external fields can be captured with microsecond resolution, typically constrained by the camera frame rate.[9] However, for ultrafast processes occurring faster than nanoseconds, conventional camera speeds are insufficient to capture dynamic processes. The integration of UTEM with *in-situ* TEM techniques enables the investigation of ultrafast dynamics in complex environments under multi-field conditions. This combined approach facilitates the study of external field influences on relaxation processes following photoexcitation. Additionally, dynamic responses triggered by other impulsive excitations, such as electric pulses, ultrasonic waves, and microwaves, can be examined. These investigations are particularly relevant for applications in electronic devices.[10] Understanding dynamic processes in multi-field coupling and specific environmental conditions is essential for practical applications, including catalysts, solar cells, photodetection materials, and life sciences. This perspective aims to provide an in-depth exploration of current UTEM applications in multi-field dynamics research and to discuss future directions for advancing this cutting-edge technology.

In-situ Liquid/Gas Environments. The ability to observe nanoscale processes in real-time within liquid or gas environments has opened new avenues for research across various fields. In biology, nearly all biological processes occur in aqueous environments, and the function of biomolecules can be significantly influenced by these conditions.[11,12] Studying the structural dynamics of proteins in liquid environments can further enhance our understanding of protein function in living organisms.[11] In catalysis research, catalysts operate in gas or liquid environments. To characterize charge transfer processes or structural evolution during catalytic reactions, it is essential to study catalysts under real working conditions.[5] Numerous studies have demonstrated that coupling nuclear and carrier dynamics is crucial for understanding the catalytic properties of materials. Using first-principles dynamics simulations, You *et al.* have shown the correlated electron-nuclear dynamics of photoinduced water dissociation on rutile TiO_2.[13] UTEM may enable direct observa-

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tion of photo-induced nuclear dynamics at the nanoscale in water using an *in-situ* liquid holder, offering insights into reaction mechanisms, particularly the coupling between lattice vibrations and carrier dynamics. Fu *et al.* have made significant progress by integrating a liquid environment into UTEM to study the active Brownian dynamics of nanoparticles in water triggered by laser pulses.[14–16] Using this liquid-phase UTEM, a series of studies have demonstrated the complex dynamic behaviors of nanoparticles in liquid, highlighting the potential for ultrafast dynamic studies in liquid environments with high spatiotemporal resolution.

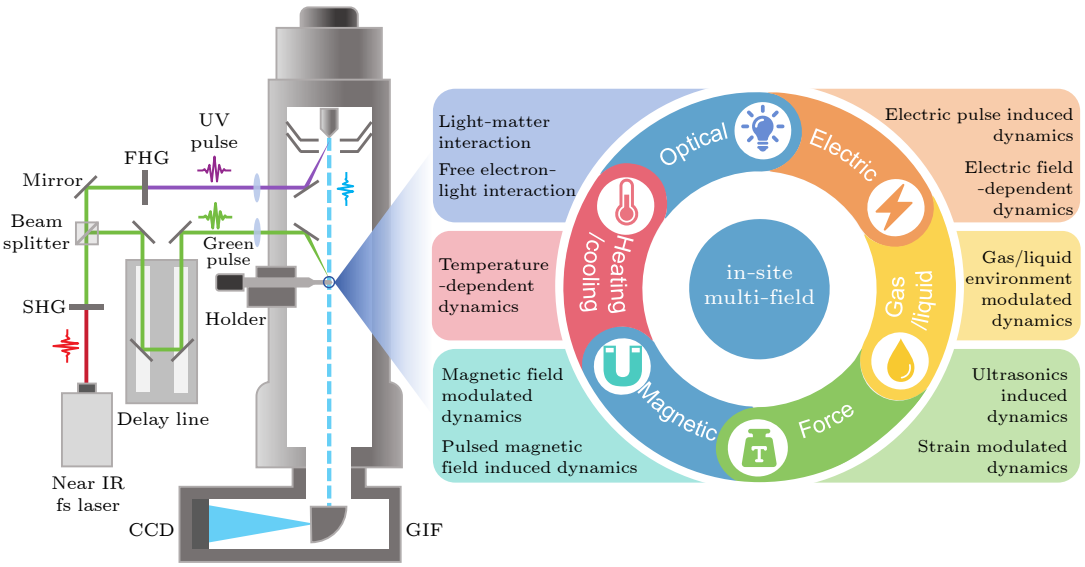

Fig. 1. Schematic illustrating the integration of *in-situ* multi-field techniques with UTEM for studying ultrafast dynamics in complex environments.

In-situ Electric Fields. By applying electric fields to samples within the TEM, researchers can observe field-induced phenomena such as electrochemical reactions,[17] polarization changes,[18] magnetic evolution,[19] and electromechanical responses.[20] This has significant implications for the development of next-generation electronic devices, energy storage systems, and sensors with enhanced performance and functionality. UTEM uniquely enables the visualization of atomic responses and carrier dynamics under electric fields, making it a powerful tool for studying electronic and structural dynamics in nanoscale devices. Applying a pulsed electric field allows for the study of material responses to electric stimuli. This is particularly important for research on data storage devices, such as resistive storage devices, ferroelectric storage devices, phase-change materials, and magnetic materials, which use electric pulses to switch material states. The dynamics of this switching process, which determine data recording and reading speeds, present an intriguing research topic for UTEM. The key challenge is synchronizing the electric pulse with the electron pulse. Mattes *et al.*[10] demonstrated that an ultrafast electron beam probe with terahertz-compressed electron pulses can directly sense local electromagnetic fields in electronic devices, achieving femtosecond, micrometer, and millivolt resolution under normal operating conditions. Fu *et al.*[21] developed a laser-free UTEM based on an RF-driven pulser and successfully visualized gigahertz electromagnetic wave propagation dynamics in an interdigitated comb structure. This revealed the transient oscillating electromagnetic field around the tines of the comb, with time-resolved polarization, amplitude, and local field enhancement at nanometer and picosecond scales.

In-situ Magnetic Fields. The combination of UTEM with magnetic field manipulation techniques has enabled the study of laser-induced magnetic domain dynamics, spin textures, and magnetic phase transitions in nanomaterials.[22–24] By applying external magnetic fields to samples within the TEM, researchers can investigate the magnetic properties of materials under varying conditions, leading to a deeper understanding of magnetic phenomena at the nanoscale. Li *et al.*[25] demonstrated the manipulation of the magnetic phase transition in an $Fe_{2.84\pm0.05}GaTe_2$ sample using a femtosecond laser pulse at various magnetic fields. This has implications for the development of novel magnetic materials, spintronic devices, and magnetic storage technologies with enhanced performance and functionality. Additionally, it can be used to study the magnetic-phonon coupling effect. For example, Tauchert *et al.*[26] demonstrated that applying a magnetic field to laser-demagnetized nickel reveals an almost instantaneous, long-lasting, non-equilibrium population of anisotropic phonons that oscillate predominantly in a plane perpendicular to the direction of the initial magnetization.

In-situ Strain Fields. The application of controlled strain fields to samples within the TEM has enabled the study of mechanical properties, deformation behaviors, and other aspects of strain engineering in materials at the nanoscale with high spatial resolution.[27,28] By visualizing

the response of materials to strain, UTEM offers valuable insights into the mechanisms governing mechanical behavior, deformation processes, and structural changes in nanomaterials. This has significant implications for designing advanced materials with tailored mechanical properties and enhanced performance across various applications. Photo-induced strain wave dynamics have been observed in two-dimensional (2D) materials using UTEM.[29,30] Ji *et al.* also reported the influence of local strain fields on photo-induced phase transitions in WTe_2.[31] However, the active application of strain fields to investigate their influence on structural dynamics remains relatively unexplored. This could provide valuable insights, particularly into the polar structural dynamics of ferroelectric materials. Another area of interest involves applying impulsive strain or shock waves to samples. For instance, O. Bostanjoglo and Th. Rosin conducted pioneering work revealing ultrasonic wave-induced magnetic reversals using stroboscopic electron microscopy.[32]

In summary, combining *in-situ* TEM techniques with UTEM capabilities allows researchers to visualize ultrafast dynamic processes and investigate complex phenomena in multifaceted environments. UTEM serves as a powerful tool for studying ultrafast nanoscale dynamics, providing unparalleled insights into material behavior under diverse conditions. Efforts are underway to enhance UTEM's brightness, spatial resolution, temporal resolution, and energy resolution. One promising approach involves leveraging advanced detector technologies, such as direct electron detectors, which enable high-resolution imaging and spectroscopy at lower electron doses. This approach significantly mitigates space charge effects, reducing temporal elongation and energy dispersion broadening. Additionally, techniques like RF/THz compressors are being explored to further improve temporal resolution of UTEM experiments. Advancements in spatial resolution, brightness, spectroscopic capabilities, and automation are expected to expand UTEM's potential, driving transformative discoveries in multifield dynamics. These innovations will pave the way for groundbreaking progress in fields such as materials science, condensed matter physics, chemistry, and biology.

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