



# Frontier nonequilibrium materials science enabled by ultrafast electron methods

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Most innovation in materials science and engineering resides in our ability to understand and control the intimate relationship between the structure of materials and their properties. Conventionally, the only route to discovering innovative new material properties has been to explore the structural and compositional phase space that is accessible at (or near) thermodynamic equilibrium. The characterization, manipulation and, ultimately, control of material properties far from equilibrium offers almost completely untapped possibilities for uncovering novel states and phases of materials. Investigations of materials far from equilibrium require the development of new techniques, which are capable of following dynamic processes in materials with extreme spatiotemporal resolution. Thus, ultrafast electron-based methods have become a major new frontier in materials science due to the capability of following dynamics on time scales as short as femtoseconds with the high spatial resolution and sensitivity afforded by electrons. The articles in this issue provide an exciting cross section of the novel research directions that have been enabled at this frontier. Imaging on ultrafast time scales carries critical information on phase transitions, fundamental processes involved in light-harvesting, magnetic, and plasmonic dynamics, the coupling of electronic and nuclear degrees of freedom in materials and has revealed previously hidden, nonequilibrium metastable states of matter that have no equilibrium analog.

## Introduction

An atomic-level description of structure is central to our understanding of the material world around us. As a result, enormous research effort has been devoted to determine the equilibrium structure of materials on a range of spatial scales. There are a broad range of experimental techniques capable of performing such measurements, and all areas of science have seen their influence.

Electron methods, including diffraction, imaging, and spectroscopy, have been a major tool of choice for structure determination, ranging from meso-dimension, to molecular and atomic scales, thanks to large scattering cross sections, high elastic scattering versus inelastic scattering ratio, and relative ease of focusing by electromagnetic lenses. State-of-the-art electron microscopes achieve unprecedented subatomic spatial resolution in images through aberration-corrected transmission electron microscopy.<sup>1</sup> To further advance electron scattering techniques that directly enable groundbreaking

science, instrumentation must advance beyond traditional two-dimensional (2D) imaging. Advances in three-dimensional spatial resolution using electron tomography,<sup>2</sup> *in situ* methods with novel gas/liquid/temperature/electrochemical environments,<sup>3</sup> new detectors that provide unprecedented elemental contrast, and advances in temporal resolution<sup>4</sup> all constitute new frontiers in electron microscopy. This issue of the *MRS Bulletin* focuses on the combination of high temporal resolution with *in situ* methods/environments to make visible the atomic-scale, dynamic processes that occur in materials “while they are working” (under nonequilibrium conditions). These dynamic electron methods now exist through the combination of ultrafast electron beams and short-pulse lasers, each of which is themselves relatively mature technologies with their own research communities. The rapid progress that has occurred in this field during the last decade is the direct result of an exciting ferment between ultrafast spectroscopists, electron microscopists, high-energy electron-beam physicists,

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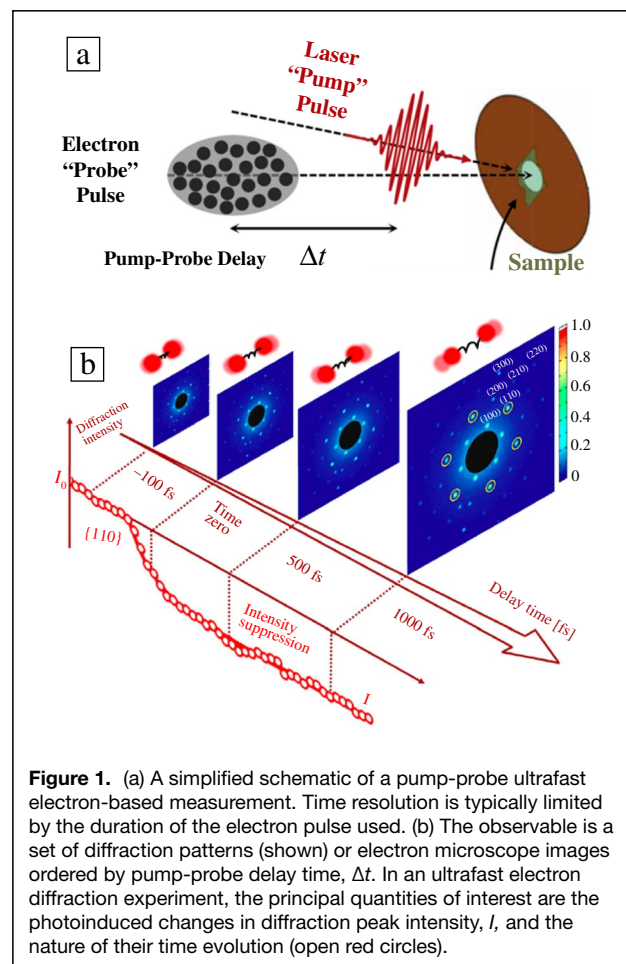
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and materials scientists. The complementary expertise of these communities has been essential to fostering the developments described in this issue. Ultrafast electron methods now provide a direct view of the most fundamental processes in materials as they occur, a capability that has ignited a revolution in research laboratories around the world.<sup>5–9</sup> This new view of the microscopic world is required to solve a range of real-world problems, as described by several US-DOE reports.<sup>10,11</sup>

### Ultrafast electron methods and technologies

Ultrafast electron methods use a pump-probe approach (Figure 1a) by which it is possible to capture a snapshot of the atomic,<sup>5,8</sup> nano- to microstructure<sup>6,7</sup> of materials as it evolves in time. The process of interest is initiated with an ultrashort pump laser pulse tunable over the range from UV to THz. The excitation wavelength is typically chosen to either selectively (in energy/momentum) “photo-dope” the material with carriers or drive specific vibrational modes (phonon pumping), although other excitations are possible. The intensity of the pump controls the carrier density or the amplitude of the driven phonon. At a well-controlled delay time after the “pump,” the sample is exposed to an ultrashort electron-probe pulse that produces either a diffraction/scattering pattern (Figure 1b) or transmission electron microscope image at the detector. These data, collected over a range of time delays, can be used to obtain a “molecular movie” of the photoinduced process and the structure of any metastable states that emerge along the relaxation (or transformation) pathway. In ultrafast electron instruments, the electron pulses themselves are produced via photoemission with part of the same femtosecond laser pulse train that is used to pump the samples (Figure 2). The duration of the photoelectron pulses at the sample effectively determines the minimum “exposure time” (or time resolution) and is now in the 100 fs regime for state-of-the-art instruments.<sup>12–15</sup> Recent work has demonstrated that many-electron pulses as short as 10 fs can be produced,<sup>16</sup> although there is still work left to be done before pump-probe experiments can be performed at this time resolution. It is important to appreciate that a snapshot of structure taken with a 10-fs pulse represents a truly instantaneous atomic configuration in most materials, since essentially all atomic motion is frozen on time scales shorter than the period of the highest frequency phonons. These are exciting times in the field of ultrafast electron methods.

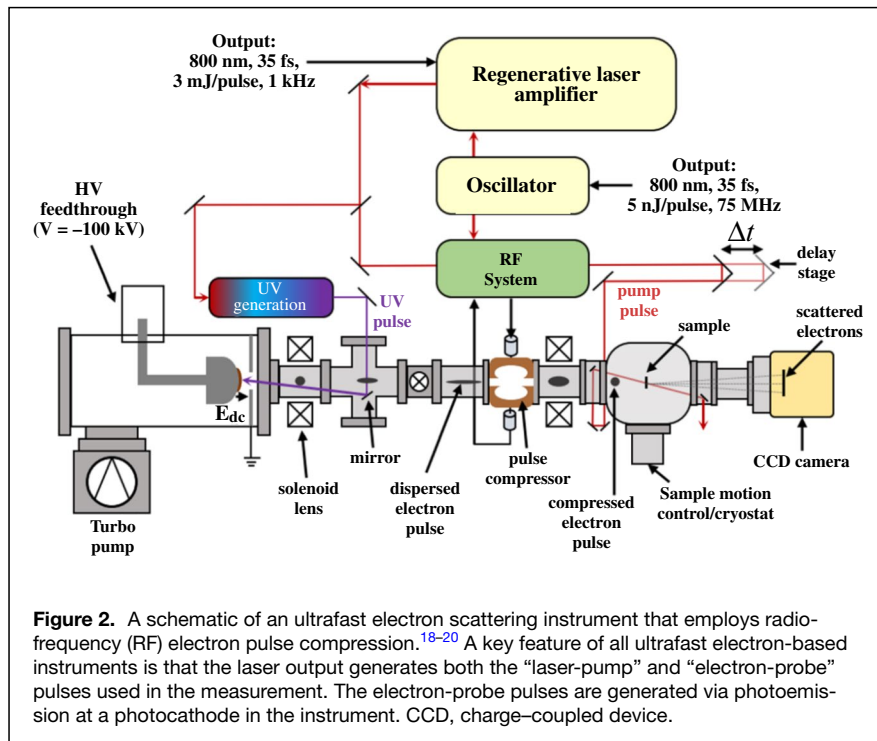
While electrons, x-rays, and neutrons all provide the ability to map the atomic composition and structure of molecules, materials, and devices, neutron sources are not suited to ultrafast experiments. Ultrafast electron scattering and ultrafast x-ray techniques have both flourished in recent years. These are both competitive and complementary approaches, in particular, due the high performance available from relatively inexpensive lab-scale ultrafast electron instrumentation when compared to x-ray free-electron lasers (XFELs) and other large x-ray user facilities. Electrons also



**Figure 1.** (a) A simplified schematic of a pump-probe ultrafast electron-based measurement. Time resolution is typically limited by the duration of the electron pulse used. (b) The observable is a set of diffraction patterns (shown) or electron microscope images ordered by pump-probe delay time,  $\Delta t$ . In an ultrafast electron diffraction experiment, the principal quantities of interest are the photoinduced changes in diffraction peak intensity,  $I$ , and the nature of their time evolution (open red circles).

have a significantly larger elastic scattering cross section than x-rays and a superior ratio of elastic to inelastic cross sections. This combination makes electrons particularly well suited for pump-probe experiments, since the laser absorption depth typically matches the single-electron scattering length within an order of magnitude. These properties pay real dividends when studying thin films, 2D materials, monolayers, surfaces, and radiation-sensitive samples. The high performance of electron lenses also provides for high-resolution real-space imaging as is well known from conventional electron microscopy. The small de Broglie wavelength of electrons and associated flat Ewald sphere allow electron scattering to easily cover a large scattering vector ( $Q$ ) range, enabling the efficient extraction of pair distribution functions (PDFs) for polycrystalline and gas-phase specimens. This large  $Q$ -range provides higher real-space resolution PDFs than achieved to date with ultrafast x-ray laser scattering.

The accurate measurement of subtle phenomena (that result in small photoinduced changes in electron scattering) and the study of irreversible physical processes (such as solid- to liquid-phase transitions, or nucleation and growth)



requires large numbers of electrons in a single pulse to produce diffraction patterns or images with sufficient signal-to-noise ratio. Packing as many electrons as possible into bunches with a duration 10–100 fs is the source of one of the primary technical challenges associated with ultrafast electron methods (i.e., space-charge-related [or Coulomb repulsion] effects in the electron beam).<sup>5</sup> Electron-beam brightness has historically been severely limited by space-charge-induced broadening effects, leading to a tradeoff between signal levels and time resolution. Such a tradeoff is obviously profoundly unfavorable and has been a significant barrier standing in the way of a broad application of ultrafast electron methods to complex structural problems. However, remarkable progress has been made addressing these challenges, improving both the temporal resolution and structural sensitivity of ultrafast electron methods. Several strategies are now being employed. First, simple ultracompact femtosecond electron sources have been developed<sup>13,17</sup> in which the time between photoemission and scattering is so short that space-charge broadening simply does not have time to occur provided the bunch charge does not get too high.<sup>5</sup> Second, radio-frequency (RF) cavities have been employed as “temporal lenses” to recompress space-charge-broadened electron pulses at the sample.<sup>18–20</sup> For several years, the performance of this approach was limited by laser-RF synchronization issues, but even this is now a solved problem.<sup>14</sup> Third, relativistic or MeV electron beams are now being used.<sup>12,21</sup> Transverse and longitudinal space-charge effects scale as  $1/\beta^2\gamma^3$  and  $1/\beta^2\gamma^5$ , where  $\beta$  and  $\gamma$  are the relativistic velocity and energy. Due to the favorable

longitudinal space-charge scaling, more MeV electrons can be packed into a single short bunch or, for a fixed number of MeV electrons, shorter bunch durations can be achieved. An example of single-shot images with an excellent signal-to-noise achieved at the SLAC MeV-UED facility are shown in Figure 1b. MeV beams can also bring further benefits. For example, the elastic scattering cross section for MeV electrons is also lower than it is for keV electrons, which has the potential to reduce the impact of multiple scattering, extending the range of samples that can be analyzed quantitatively within the single scattering (or kinematical) approximation. This can also allow for experiments to be performed on thicker samples, opening up a wide range of scientific opportunities involving liquids such as photocatalysis in solution.

There is now a diversity of ultrafast electron sources and instrumentation available, including home-built keV systems, modified commercial TEMs, and RF-photoguns/MeV electron-beam instrumentation that originally found their use filling the storage ring at synchrotron facilities or driving x-ray free-electron lasers (XFELs). These instruments cover a wide range of electron energy from 10 s of eV to MeV and have provided for ultrafast versions of many conventional electron-based scattering, imaging, and spectroscopy method beyond those previously described (e.g., low-energy electron diffraction (LEED),<sup>22</sup> reflection high-energy electron diffraction (RHEED),<sup>23</sup> electron energy-loss spectroscopy (EELS),<sup>24</sup> photoemission electron microscopy (PEEM),<sup>25</sup> and other techniques).

### In this issue

This issue is organized around three main themes that demonstrate the broad impacts that ultrafast electron methods are having on our understanding of fundamental processes and excitations in materials.

#### **Directly observing ultrafast structural dynamics in materials: Phase transitions and processes that underlie properties**

When trying to uncover the microscopic origins of many phenomena in materials science, equilibrium structure is only a starting point. For a thorough understanding of any process, the essential complement to equilibrium structural

information is dynamics. This dynamical information can be essentially hidden from view when using conventional techniques but can be revealed in full with ultrafast methods.

It is now possible to follow phase-transition dynamics in laser-driven solids, as described in the article by Mo et al.,<sup>26</sup> distinguishing heterogeneous and homogeneous nucleation processes involved in melting using ultrafast electron diffraction. Structural dynamics driven by photoexcitation in light-harvesting materials have also been probed by UED, and this is the subject of the article by Guzelturk and Lindenberg.<sup>27</sup> Real-space ultrafast electron imaging methods are also in development, and the article by Harvey et al.<sup>28</sup> demonstrates time-resolved Lorenz microscopy as a tool for studying magnetization dynamics in materials.

### **Properties on demand in quantum materials**

The exploration and control of nonequilibrium states of solids are an emerging area of research with the potential to discover exotic properties, combinations of properties, or ordered phases that are not present in materials at equilibrium. This work expands the methods of new materials discovery beyond the traditional approaches of materials synthesis that focus on equilibrium states and ground-state properties, and instead opens a new direction, selecting “properties on demand.”<sup>29</sup> This approach makes use of external perturbations to steer a material toward a competing ground, metastable, or transient state that has novel properties and behavior. Strongly correlated or quantum materials are particularly interesting in this regard because the interactions associated with lattice, charge, orbital, and spin degrees of freedom in these materials are commonly on par with the electronic kinetic energy. Any excitation that modifies the interplay between these degrees of freedom can result in a dramatic transformation in the material’s properties. The rather fragile balance between coexisting and competing ground states can be readily shifted via external stimuli, leading to many quantum phases and the possibility of initiating transitions between them. Many of these phases are completely inaccessible through simple changes in pressure or temperature.

The article by Zong et al.<sup>30</sup> provides a review of research in this direction that has been enabled by ultrafast electron methods, highlighting several examples of research from the recent literature.

### **Probing the nature of complex excitations and their mutual couplings directly**

Elementary excitations and their mutual couplings form the fundamental basis of our understanding of diverse phenomena in materials. For example, the interactions between collective excitations of the lattice system (phonons) and charge carriers, specifically, are known to lead to superconductivity, charge density waves, multiferroicity, and soft-mode phase transitions. These carrier–phonon interactions are also central to our understanding of electrical transport, heat transport, and

energy conversion processes in photovoltaics and thermoelectrics. Phonons can themselves be intimately mixed in to the very nature of more complex elementary excitations, as they are in polarons or polaritons, or intertwined with electronic, spin, or orbital degrees of freedom, as it now seems is the case for the emergent phases of many strongly correlated systems that exhibit complex phase diagrams such as high- $T_c$  superconductors. It has been a difficult experimental problem to fully characterize the nature of elementary excitations and to quantify the strength of their momentum-dependent interactions. This has been one of the primary barriers to our understanding of these phenomena, particularly in complex anisotropic materials.

Ultrafast pump-probe techniques provide an opportunity to study couplings between elementary excitations rather directly. Photoexcitation can prepare a nonequilibrium distribution of quasiparticles or other selected modes in which subsequent relaxation dynamics and coupling to other degrees of freedom can be followed in the time domain. Progress has been rapid over the last decade, both from the perspective of the selectivity of the initial excitation and the ability to probe the subsequent dynamics over a broad range of frequencies.

Ultrafast electron diffuse scattering (UEDS) methods, for example, have provided unprecedented, direct access to the momentum-dependent couplings between carrier and phonon systems, and within the phonon system itself. A review of this approach highlights from the recent literature that is the focus of the article by Dürr et al.<sup>31</sup>

Ultrafast photoemission electron microscopy (uPEEM) has recently provided remarkable access to plasmon dynamics at nano-femtospacial resolution,<sup>25</sup> and the article by Dai et al.<sup>32</sup> demonstrates an extension of these methods to imaging surface plasmon polaritons and coupled vectorial electromagnetic wave-charge density polarization fluctuations.

### **Outlook**

This issue of the *MRS Bulletin* gives a brief update on the exciting development of frontier nonequilibrium materials science enabled by ultrafast electron methods, providing a snapshot of current work to control materials and develop systems to capture, convert and store energy that will impact sustainability and advanced technologies. The establishment of ultrafast electron methods at user facilities (e.g., SLAC MeV-UED and ANL/CNM UEM) is helping to make these unique tools available to the broader material science communities by lowering the barrier to entry.

The exciting scientific opportunities presented in this issue have stimulated worldwide R&D effort to realize higher temporal resolution, better momentum resolution, and more versatile sample environments in ultrafast electron methods. By employing THz or double-bend achromat electron bunch compressor, time resolution on the order of 10 fs is expected.<sup>32,33</sup> Central to understanding how to synthesize and exploit functional materials is having the ability to apply external stimuli (such as heat, light, a reactive flux, an electrical bias) and to

observe the resulting dynamic process *in situ* or *operando*, and under the appropriate environment. The success of the first liquid-phase MeV-UED<sup>34</sup> opens the possibility of investigating electrochemical process with atomic view using ultrafast electron methods. A recent report on the universal phase dynamics in VO<sub>2</sub> switches demonstrates a first ultrafast electron *operando* diffraction measurement<sup>35</sup> and lays a foundation for exploring quantum material-based electronic device with electric and photon control as they operate. Ultrafast electron diffuse scattering (UEDS) has the great potential of becoming a tool of choice investigating energy flow and phonon dynamics in material science. To realize such potential, better temporal and momentum resolution and detectors with large dynamics range and single-electron sensitivity are needed. Direct electron detector (DED) developed for cryo-EM has been successfully tested for ultrafast MeV electrons.<sup>36</sup> With electron counting and shot-by-shot jitter correction, noise-free (almost) diffraction is expected in UEDS. Recent development of an ePix detector with high dynamic range and gain auto-ranging<sup>37</sup> could be a game changer for both UEDS and single-shot UED.

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## Conflict of interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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