# Program :

## *Thursday July the 18th :*

- 12.30: Welcome and overview (Fabrizio Carbone)
- 13.00: Jom Luiten *«Ultracold and ultrafast electron source»*
- 13.35: Fulvio Parmigiani *«Science driven requirements for seeded free electron lasers»*
- 14.10: Tom Lagrange *« Movie Mode Dynamic Transmission Electron Microscopy (DTEM): Multiple Frame Movies of Transient States in Materials with Nanosecond to Microsecond Temporal Resolution»*
- 14.45: Peter Baum *«Towards an atomic-scale recording of electronic motion in 4D»*
- 15.20: Arnaud Arbouet *« Development of a high-brightness ultrafast TEM : the FemTOTEM project »*

Coffee break

- 16.15: Sasha Schafer *«Laser-triggered needle emitters in ultrafast transmission electron microscopy»*
- 16.50: Florian Banhart *«In-situ transmission electron microscopy at conventional time scales»*
- 17.25: Brett Barwick *«Imaging at the nm and fs scales with ultrafast electron microscopy (UEM)»*
- 18.00: Yoshie Murooka *« Ultrafast Relativistic-electron Diffraction in transmission with 100fs temporal resolution for structural dynamics »*

Conference dinner

## *Friday July the 19th :*

- 9.00 : UED lab tour
- 10.00: UEM lab tour

Early lunch

- 13.00: Jure Demsar *«Dynamics of Charge Density Wave Order in dichalcogenides probed by Ultrafast Electron Diffraction»*
- 13.35: Yimei Zhu *«Decoupling Electron and Phonon Contributions in Charge/orbital Ordered Systems using MeV Ultrafast Electrons»*
- 14.10: Oh Hoon Kwon *«Single-Particle Structural Dynamics in 4D Electron Microscopy»*
- 14.45: Hyun Soon Park *«4D Lorentz Electron Microscopy»*
- 15.20: Jonas Weissenrieder, *« Ultrafast Electron Microscopy Studies of Phase Transitions in Vanadium dioxide »*

Coffee break

- 15.55: Nigel Browning, *« Quantifying In-situ Reactions in the STEM/DTEM »*
- 16.30: David Flannigan *« Considerations for Environmental and Specimen Stability in Ultrafast Electron Microscopy Experiments »*
- 17.05: Ahmed Zewail

End of the workshop.

**Dr Jom Luiten**
**University of Eindhoven, The Netherlands**

# Ultracold and ultrafast electron source

We have developed a new class of electron source by using near-threshold, pulsed photoionization of laser-cooled, trapped rubidium atoms which allows us to produce intense electron pulses with large intrinsic coherence length. Several properties of electron pulses extracted from such an ultracold source have been investigated, showing in particular that temperatures in the ten Kelvin range can be achieved with tens of thousands electrons in a picosecond pulse. The ultralow source temperature – more than three orders of magnitude lower than conventional photoemission sources – seems to be in contradiction with the spread in photon energy associated with femtosecond ionization laser pulses. We explain this surprising result by a detailed study of the electron trajectories during photoionization. In addition, we show how laser polarization effects arise and that these can be used to further control the electron beam properties. Recently we have produced the first diffraction patterns from graphene-like samples with the ultracold and ultrafast electron beam. Our goal is to demonstrate single-shot electron diffraction of macromolecules and ultimately enable dynamical studies of biomolecules in an ambient environment.

# Dr Fulvio Parmigiani

**Physics Department, University of Trieste**
**and Elettra-Sincrotrone Trieste (Italy)**


## Science driven requirements for seeded free elecron lasers

Recent theoretical and experimental progress in the physics and technology of electron beams has shown the possibility of producing fully coherent, variable polarization and tunable X-ray pulses. This development opens new possibilities to design and build free-electron lasers (FELs), to produce high intensity, femtosecond and fully coherent soft X-ray pulses.

Here we will review the experiments and the ideas that represent the science frontier in time-resolved spectroscopy, coherent imaging and scattering studies.

A particular attention is given to the new free-electron laser (FEL) source FERMI@Elettra seeded at 260 nanometers with an external laser and operating in the single cascade high gain high harmonic (HGHG) mode and in the double cascade mode. This marks the first successful operation of FERMI@Elettra in its planned configuration, i.e., as a next-generation seeded free-electron laser source.

# Dr Thomas LaGrange

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# Movie Mode Dynamic Transmission Electron Microscopy (DTEM): Multiple Frame Movies of Transient States in Materials with Nanosecond to Microsecond Temporal Resolution

Most processes in materials naturally occur in conditions far from equilibrium on short time scales, ranging from the femtosecond-scale processes occurring from non-equilibrated electron states to microsecond transient events of phase transformations and deformation processes. Typically, we are confined in the laboratory to conduct experiments near equilibrium or observe material post process due to the resolution limitations of conventional analytical techniques. Though insight about material's behavior gained from these observations, much of coupled and convoluted events of complex processes not well understood that limit and require technique that both high spatial and temporal resolution to observe nanoscale microstructural features evolving on short timescales. In effort to meet the need for studying fast dynamics and transient states in material processes, we have constructed a nanosecond dynamic transmission electron microscope (DTEM) at Lawrence Livermore National Laboratory to improve the temporal resolution of in-situ TEM observations.

Prior DTEM hardware only allowed single-pump/single-probe operation, building up a process's typical time history by repeating an experiment with varying time delays at different sample locations. Movie Mode DTEM upgrade now enables single-pump/multi-probe operation. The two core components of the Movie Mode technology are the arbitrary waveform generator (AWG) cathode laser system and a high-speed electrostatic deflector array. The AWG cathode drive laser produces a series of laser pulses with user-defined pulse durations and delays that stimulates a defined photoemitted electron pulse train. Each pulse captures an image of the sample at a specific time. The fast-switching electrostatic deflector array located below the sample directs each pulse (image) to a separate patch on a large, high-resolution CCD camera. At the end of the experiment, the entire CCD image is read out and segmented into a time-ordered series of images, i.e., a movie. These technical improvements provide the ability to track the creation, motion, and interaction of individual defects, phase fronts, and chemical reaction fronts, providing invaluable information of the chemical, microstructural and atomic level features that influence the dynamics and kinetics of rapid material processes. For example, the potency of a nucleation site is governed by many factors related to defects, local chemistry, etc. While a single pump-probe snapshot provides statistical data about these factors, a multi-frame movie of a unique event allows all of the factors to be identified and the progress of nucleation and growth processes can be explored in detail. It provides unprecedented insight into the physics of rapid material processes from their early stages (e.g. nucleation) to completion, giving direct, unambiguous information regarding the dynamics of complex processes. This presentation will discuss the technical aspects of the Movie Mode DTEM technology in the context of recent material science studies using the novel in situ TEM capability.


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# Dr. Peter Baum

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## Towards an atomic-scale recording of electronic motion in 4D

Time-resolved microscopy and diffraction now allow to record atomic motion in space and time. For example, the insulator metal phase transformation in VO2 [1] and the interlayer dynamics of graphite [2] had revealed sequential mechanisms of coherent and incoherent atomic displacements, resolved on picometer and femtosecond scales. But what about electron densities? These can move within attoseconds. Our aim is to advance ultrafast electron diffraction and microscopy towards that novel regime. Our concepts include single-electron pulses [3], compression in microwave fields [4], development of 'isochronic' magnetic lens systems [5], increase of coherence into biomolecular regimes [6], and pulse characterization by streaking in laser fields. We will discuss our progress and what discoveries we may expect to see, in the regime of 4D electron dynamics [7-8].


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# Dr Arnaud Arbouet

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# Development of a high-brightness ultrafast TEM : the FemTOTEM project


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The recent development of ultrafast TEMs with temporal resolution in the subpicosecond range bears exciting promises. However, the ultrafast TEMs currently available exploit photocathodes which have a limited brightness as the electrons are emitted from an illuminated area having a size of the order of several tens of microns. From a general point of view, the *brightness* of the electron source, defined as the current per unit area and unit solid angle, is the key figure of merit in order to get the highest spatial coherence and highest spatial resolution mandatory for the most demanding TEM applications. Indeed, the development of cold field-emission guns at the end of the 1960s has opened a new era. These sources in which electrons are tunneling out of the apex of a metallic nanotip under a strong DC bias voltage have a brightness at least a hundred times larger than all other electron sources. Following the initial suggestion of Gabor in 1948, new coherent electron microscopy techniques such as off-axis electron holography have relied on these new sources to produce electron interferograms from which small changes in the phase of the electron quantum-mechanical wavefunction are analyzed. These phase changes are so sensitive to the electrostatic field, strain field or magnetic field inside nano-objects that electron holography allows their quantitative mapping with nanometer resolution. Recently, several studies have evidenced the emission of ultrashort electron pulses from field-emission tips triggered by femtosecond low energy laser pulses[1]. Beyond fundamental issues, these investigations raise great expectations as they provide the ideal candidate of high brightness ultrafast electron source for time-resolved electron microscopy and holography experiments. Our project proposes to build on these studies and develop an alternative approach based on the implementation of a laser-driven field emission source inside a TEM[2].


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# Laser-triggered needle emitters in ultrafast transmission electron microscopy

Ultrafast transmission electron microscopy enables the exploration of dynamical processes with combined ultrafast temporal resolution and nanoscale spatial resolution. However, the ability to focus a sufficiently high number of electrons into a nanometric volume relies on the emission characteristics of the pulsed electron source. Specifically, small effective source size and low beam divergence are required to obtain low emittance electron beams. Laser-triggered needle-shaped photoemitters allow the reduction of the effective source size far below the diffraction-limited focal size of the driving laser and are thus a promising alternative to the presently employed flat photocathodes. In this contribution, we present experimental results on the performance of laser-triggered needle emitters and compare these to particle trajectory simulation. An emphasis is placed on the beam emittance and temporal pulse widths which can be achieved in the electron optics environment of a transmission electron microscope. Furthermore, the feasibility of laser-triggered needle emitters for time-resolved electron diffraction and imaging is demonstrated in an ultrafast low-energy electron diffraction setup.

# Dr Florian Banhart

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## In-situ transmission electron microscopy at conventional time scales

The talk will present an overview of recent in-situ experiments in transmission electron microscopy at a temporal resolution on the "conventional" scale of seconds. The focus will be on nanomaterials based on carbon such as graphene, carbon nanotubes, and metal particles [1]. Dynamic phenomena are getting accessible to direct observation such as the nucleation and growth of graphene or carbon nanotubes [2], the diffusion of metal atoms on graphene [3], the formation of chains of carbon atoms, or the interaction between energetic electrons and graphitic nanomaterials. The formation of lattice defects by electron irradiation is another important subject of in-situ TEM as it allows the observation of lattice reconstructions after ballistic atom displacements [4]. Examples for defect formation in graphene-based materials will be shown [5]. Irradiation effects can be used to tailor the structure and properties of specific nanomaterials and for joining different species as will be shown for the example of metal-carbon junctions [6]. Different techniques for in-situ experimentation were used such as heating, irradiation, or electrical probing of nano-objects under observation.
The necessity of an improved temporal resolution, needing observations at timescales between milliseconds and femtoseconds, will be discussed, and the plans to set up an ultrafast TEM in Strasbourg will be presented.


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# Dr Brett Barwick

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## Imaging at the nm and fs scales with ultrafast electron microscopy (UEM)


Investigating ultrafast phenomena with femtosecond (10-15 s) and attosecond (10-18 s) temporal resolution is pivotal to understanding the dynamic processes that atomic, molecular and condensed matter systems undergo. The time scale for dynamics, at the atomic length scale, ranges from picoseconds to attoseconds for processes such as the heating of a thin metallic crystal and the motion of plasmons in metals. In this talk I will describe ultrafast imaging using "single-electron" packets as applied to several different nanoscale ultrafast processes. In particular, I will describe a new imaging method that exploits the fact that "free" electrons (when near a third body) can absorb and emit multiple photons. The physics describing the absorption and emission of photon quanta by free electrons is well known in AMO physics as a "free-free" transition and is manifested in the laser assisted photoemission effect. We form images by using only electrons that have absorbed photons; allowing us to observe the evanescent electric field created by plasmons that have been excited by an intense ultrafast optical pulse. In describing this imaging technique dubbed, photon-induced near-field electron microscopy (PINEM), I will also discuss future plans to extend the temporal resolution to tens of femtoseconds and possibly even the attosecond regime.

**Dr Y. Murooka**
**Advanced Science Institute, RIKEN, Saitama, Japan**

# Ultrafast Relativistic-electron Diffraction in transmission with 100fs temporal resolution for structural dynamics

With relativistic electron pulses, we have developed a compact ultrafast electron diffractometer in transmission mode with ~100fs time resolution [1]. This is to directly reveal ultrafast structural dynamics triggered by short laser pulses in solids.

The diffractometer has three characteristic features. First is the homemade rf photocathode [2] specifically designed for electron diffraction to generate 3 MeV electron beams with the pulse width of ~100 fs and the brightness of more than 106 e/pulse. Second is the three electromagnetic lenses arranged identical to those for a transmission electron microscope. Because of the flexibility and richness of the lens operation, the entire system becomes compact. The last is the homemade electron detection system for 3MeV electrons, using a CsI:Tl scintillater with a CCD camera. Its sensitivity is high enough so as to allow both the pump-probe experiments for reversible processes and the single-shot experiments for irreversible processes. The laser-induced transient crystalline deformation of Si single crystals will be shown. The mechanism of generating the ultrafast transient distortion is discussed.
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**Dr Jure Demsar**
**University of Kostanz, Germany**

# Dynamics of Charge Density Wave Order in dichalcogenides probed by Ultrafast Electron Diffraction

Reduced dimensionality seems to be an intrinsic property governing phenomena like high temperature superconductivity and charge density wave (CDW) formation. The latter are typical for quasi one- or two-dimensional metals at reduced temperatures. According to the standard Peierls Fermi surface nesting scenario the appearance of CDW phase at low temperatures is driven by the divergent static electronic susceptibility at a wave vector q=2k¬F. Recently, this classical picture has been challenged [1]. It was argued that the appearance of CDWs, in particular in transition-metal dichalcogenides, is due to strong (q-dependent) electron-phonon coupling [1].

Here I will review recent time-resolved experiments on transition-metal dichalcogenides, where the evolution of the order parameter (atomic displacement) following excitation with a femtosecond optical pulse has been measured directly using femtosecond X-ray [2] end electron diffraction [3,4] techniques. The results on 1T-TaS2 [3] and 4Hb-TaSe2 [4] suggest that strong electron-phonon coupling is indeed crucial for stabilization of CDWs. Moreover, comparison of CDW build-up dynamics in 4Hb-TaSe2 [4], where the coupling between CDW carrying octahedral (1T) layers is strongly reduced, to that of 1T-TiSe2 [2] and 1T-TaS2 [3] reveals the importance of three-dimensionality for the existence of CDWs in these quasi-2D systems.

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# Decoupling Electron and Phonon Contributions in Charge/orbital Ordered Systems using MeV Ultrafast Electrons

Ultrafast electron diffraction, imaging and spectroscopy represents a major challenge for modern electron microscopy, but also provides a unique opportunity for understanding the interactions between electron, orbital, spin and lattice and their competing degree of freedoms in complex materials that exhibit fascinating properties. In this presentation I will give a brief overview on our recent development of the 2.8MeV-120fs electron diffraction apparatus at Brookhaven National Laboratory. The principle and design of pump-probe electron diffraction approach and their applications in strongly correlated electron systems will be discussed. A particular example will be given on the photoinduced structural dynamics and the evolution of the charge-density wave (CDW) state in 2H-TaSe2. The observed three time-constants for the superlattice peaks, Bragg reflections and diffuse scattering of the compound after photo-excitation allow us to separate the contribution of individual dynamic transient processes in the corresponding subsystems and may shed new insight into the mechanism of CDW formation.

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# Single-Particle Structural Dynamics in 4D Electron Microscopy

With advances in spatial resolution reaching the atomic scale, 2 and 3 dimensional (D) imaging in transmission electron microscopy (TEM) has become an essential methodology in various fields of research providing static structural information. Now, it has become possible to integrate the ultrahigh temporal resolution (fourth dimension) to the 3D spatial resolution of TEM for the studies of structural dynamics of fine objects (1). Here, presented is the recent achievement of single-particle sensitivity in 4D electron microscopy (2). The examples shown include the studies of light-induced phase transitions in spin-crossover metal organic frameworks and vanadium dioxide.

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# Dr Hyun Soon Park

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# 4D Lorentz Electron Microscopy

In four-dimensional (4D) ultrafast electron microscopy (UEM),[1] timed-pulse electron imaging is used to study structural dynamics with space- and time-resolutions that allow direct observation of transformations affecting the fundamental properties of materials. Only recently, the UEM studies have begun to reveal a variety of dynamic responses of nanoscale specimens to material excitation, on ultrafast time scales and up to microseconds. In this talk, I present 4D imaging of domain walls (DWs) by the out-of-focus Fresnel method in Lorentz ultrafast electron microscopy,[2] with in situ spatial and temporal resolutions. The temporal change in magnetization, as revealed by changes in image contrast, is clocked using an impulsive optical field to produce structural deformation of the specimen, thus modulating magnetic field components in the specimen plane. Directly visualized are DW nucleation and subsequent annihilation and oscillatory reappearance (periods of 32 and 45 ns) in nickel films on two different substrates. For the case of Ni films on a Ti/Si3N4 substrate, under conditions of minimum residual external magnetic field, the oscillation is associated with a unique traveling wave train of periodic magnetization reversal. The velocity of DW propagation in this wave train is measured to be 172 m/s with a wavelength of 7.8 μm. The success of this study demonstrates the promise of Lorentz UEM for real-space imaging of spin switching, ferromagnetic resonance, laser induced demagnetization in ferromagnetic nanostructures and possible applications to spintronics.


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# Ultrafast Electron Microscopy Studies of Phase Transitions in Vanadium dioxide

In the presentation a review of the first time resolved results using the first generation ultrafast electron microscope (UEM) at Caltech will be given. Experimental difficulties together with some tried and proposed strategies and solutions will be presented. The first system of study in the microscope was the prototypical phase transition in the strongly correlated material vanadium dioxide ($VO_2$). $VO_2$ exhibit a metal to insulator transition that can be optically driven at ultrafast timescales. Using four-dimensional imaging the transition was initiated by a femtosecond near-infrared laser pulse and probed by electron pulses (at 120 keV) in the microscope. Real-space imaging and diffraction could follow the transition from the monoclinic to tetragonal phase, but also points to the presence of a metastable structure whose nature is determined by electronic, carrier-induced, structural changes. Because of the selectivity of excitation from the 3d|-band, and the relatively low fluence used, these results show the critical role of carriers in weakening the V4+□V4+ bonding in the monoclinic phase and the origin of the nonequilibrium phase. In the presentation some time will be devoted to discussing the possibilities to extend the UEM technique into near ambient studies of catalysts, sensors, and solarcells.

# Dr N.D. Browning

# Quantifying In-situ Reactions in the STEM/DTEM

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The goal of the Dynamic Transmission Electron Microscope (DTEM) is to be able to directly observe transient phenomena in materials/biological systems with both high spatial (~1nm or better) and high temporal (~1 □s or faster) resolution. The main issue in the design of experiments in the DTEM is that for many of the key scientific challenges involving structure-property relationships in materials science and structure-function relationships in biology, the transient processes involve significant re-arrangement of the atoms/molecules in the structure at specific locations – making dynamic observations essentially irreversible in nature. This requirement for "single shot" imaging effectively limits the temporal resolution that can be obtained, and defines the types of reactions that can be studied in the microscope. A key first step in the use of DTEM to study transient processes is to control the environment around the sample. An in-situ gas stage has been developed in collaboration with Fischione Instruments that allows atmospheric pressure in a range of reactive gases to be maintained around the sample while atomic resolution images can be obtained in a high resolution microscope. By utilizing a novel laser heating source, temperatures up to 2000$^{o}$C can also be obtained in small areas of the sample. Such capabilities allow for direct imaging of oxidation and reduction processes in metals, ceramics and catalytic systems. In addition, an in-situ liquid stage has also been developed in collaboration with Hummingbird that allows atomic scale images and electron energy loss spectra to be obtained from samples suspended in solution. This has a wide range of applications from corrosion in materials science to live biological systems. The use of these in-situ stages to quantify in-situ reactions will be discussed using results obtained from aberration corrected scanning transmission electron microscopes (STEM) with particular emphasis on the control of the electron dose in determining the reaction rate and the compatibility of the results with ex-situ chemical reactions. In addition, the progress in incorporating the high spatial resolution of an aberration corrected microscope into the DTEM configuration will also be discussed along with design optimizations that are underway that may extend the DTEM into the ultrafast regime.

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# Considerations for Environmental and Specimen Stability in Ultrafast Electron Microscopy Experiments

The general approach in ultrafast electron microscopy (UEM) experiments is to combine a properly-modified transmission electron microscope (TEM) with a laser system capable of producing femtosecond pulses. In this way, the millisecond temporal resolution of current TEM detection systems is circumvented such that sub-picosecond dynamics can be observed while in principle preserving spatial (1 Å) and energy (1 eV) resolutions. Conceptually, UEM can be thought of as consisting of combined ultrafast spectroscopy and electron microscopy laboratories having all the relevant apparatus and support equipment. To date, much attention has been focused on the photoelectron pulse properties (e.g., coherence, temporal profile, compression schemes, etc.) in UEM and ultrafast/femtosecond electron diffraction setups. In contrast, little emphasis has been placed on outlining and quantifying limitations imposed by environmental and specimen stability on spatial and temporal resolutions. Here, I will discuss effects of ambient conditions well-known to both the ultrafast spectroscopy and electron microscopy communities on various experimental parameters. Further, those effects will be addressed within the context of UEM experiments, and the potential impact on observables (e.g., intensity, position, energy, etc.) will be illustrated. In short, the combination of sophisticated experimental techniques, desired high energy-space-time resolutions, and sensitivities to picometer/milli-electron volt changes requires a detailed, quantitative understanding of specific environmental and specimen factors on femtosecond electron-based techniques. The magnitudes of these factors are non-universal and will vary greatly across lab spaces, thus necessitating individualized efforts in identifying and deconvoluting inevitable artifacts from intrinsic dynamics.