Time-Resolved (Ultrafast) Electron Microscopy

Femto-UP School 2020-2021
March 29th, 2021
Funding sources

erc  FNS  SNF  Google
Outline

• Background
  – Transmission electron microscopy (TEM)
  – Motivation behind improving the temporal resolution of in-situ TEM
  – Brief history of the development of the Ultrafast TEM (UTEM)

• UTEM Technique
  – fs-Stroboscopic pump-probe approach
  – Space charge effects
  – Practical consideration when doing UTEM experiments

• UTEM experimental examples
  – Definitions of spatial and temporal coherence (coherent control)
  – Charge density waves, Phonons and Skyrmions
  – Light-electron interactions
Electron wave-particle duality and the birth of the electron microscope

L. de Broglie (1923) postulates that \( \rho = \frac{h}{\lambda} \) is valid for all particles \( \lambda \), wavelength of electrons (Nobel prize 1929)

1986: E. Ruska Nobel Prize

The resolution limit of the light microscope due to the length of the light wave which had been recognized 50 years before by Ernst Abbe and others could, because of lack of light, not be important at such magnifications. Knoll and I simply hoped for extremely low dimensions of the electrons. As engineers we did not know yet the thesis of the “material wave” of the French physicist de Broglie that had been put forward several years earlier (1925). Even physicists only reluctantly accepted this new thesis. When I first heard of it in summer 1931, I was very much disappointed that now even at the electron microscope the resolution should be limited again by a wavelength (of the “Materiestrahlung”). I was immediately heartened, though, when with the aid of the de Broglie equation I became satisfied that these waves must be around five orders of magnitude shorter in length than light waves. Thus, there was no reason to abandon the aim of electron microscopy surpassing the resolution of light microscopy.
Why do we use electrons as a probe?
High energy = short wavelengths = high spatial resolution

Electromagnetic radiation: \( E = \frac{hc}{\lambda} \) so if \( \lambda < 5 \text{ nm} \), \( E > 1 \text{ keV} \)

Electron wavelength according to de Broglie equation: \( \lambda = \frac{\hbar}{p} \)
with \( p = m_o v = (2m_o eV)^{1/2} \)

non relativistic (<50keV):
\[
\lambda = \frac{h}{(2m_e eV)^{1/2}}
\]

Relativistic correction (>50keV or >1% speed):
\[
\lambda = \frac{h}{\left[2m_e eV \left(1 + \frac{eV}{2m_e c^2}\right)\right]^{1/2}}
\]

<table>
<thead>
<tr>
<th>voltage [KV]</th>
<th>Nonrelativistic ( \lambda ) [pm]</th>
<th>Relativistic ( \lambda ) [pm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>38.79</td>
<td>0.03878</td>
</tr>
<tr>
<td>10</td>
<td>12.27</td>
<td>0.01221</td>
</tr>
<tr>
<td>80</td>
<td>4.34</td>
<td>0.00418</td>
</tr>
<tr>
<td>200</td>
<td>2.74</td>
<td>0.00251</td>
</tr>
<tr>
<td>300</td>
<td>0.00224</td>
<td>0.00197</td>
</tr>
<tr>
<td>1000</td>
<td>0.00123</td>
<td>0.00087</td>
</tr>
</tbody>
</table>
TEMs have sets of electromagnetic and electrostatic lenses and deflector that allow to flexibly change between many different imaging modes.

- **Diffraction mode**
  The intermediate and projector lens magnify and project the **back focal plane (first diffraction pattern formed in the microscope)** of the objective lens to the detector system.

- **Imaging mode**
  The intermediate and projector lens magnify and project the **image plane** of the objective lens to the detector system.
Electron-matter interactions in a thin sample

- Backscattered electrons (BSE)
- Characteristic X-rays
- Incdent beam
- Secondary electrons (SE)
- Visible light
- Electron-hole pairs
- Absorbed electrons
- Specimen
- ELS (EELS)
- Elastically scattered electrons
- Direct beam
- Inelastically scattered electrons
- Bremsstrahlung X-rays

1-100 nm
Transmission Electron Diffraction

Bragg Diffraction Powder Pattern

Convergent Beam Electron Diffraction

Single Crystal Spot Pattern

R = g = k + k' = 1/d_{hkl}
Imaging using Diffraction Contrast

Bright-field

Dark-field
Phase Contrast Imaging: High Resolution Transmission Electron Microscopy (HRTEM)

Phase contrast for crystalline specimen

Parallel beam

Electron wave

Crystal structure properly oriented

Objective lens

Projected image

Atomic resolved HRTEM image of Silicon

\[ \Psi_i = \Psi_o \exp(2\pi i \mathbf{r}) \]

Plane wave

\[ n = \frac{\lambda}{\lambda_m} \]

Exit wave

\[ \Psi_{EW}(\mathbf{r}) = a(\mathbf{r}) e^{i\varphi_s \mathbf{r}} \Rightarrow \Psi_{obj}(\sigma_E, V_{proj}(\mathbf{r})) \]

Exit wave function in terms of weak phase object approximation (WPOA)
The advent of aberration correctors have pushed the boundaries of spatial resolution (spatial coherence).

Uncorrected, resolution is ~100 X diffraction limit.

Cs Correction, resolution is ~15 x diffraction limit.
Electron Energy Loss Spectroscopy (EELS)

Energy filtered TEM with core loss electrons

Zero loss
Inner-shell loss of atom A
Inner-shell loss of atom B
Energy loss (eV)
Plasmon
E=0
E_F
Conduction bands
Valence bands
Potential
E_A
E_B
Atom A
Atom B
Atom A
Atom B

Parallel illumination (TEM)
Sample
Energy-selecting slit inserted
Image mode
Core-loss image projected onto CCD detector
Each EFTEM image is placed in the stack building up the spectrum image plane by plane.
EELS technology has made great advancements in the past decade that local phonon modes can be probe on the sub-nanometer scale.

Modern TEM technology can combine high energy EELS and sub-nm STEM probes for nano-vibrational spectroscopy.

Development of ultrafast techniques are being pursued across many fields, e.g., physics, biology, materials science, chemistry, engineering...

- **Diffract before Destroy**
  - Protein Structural Analysis

- **Bunch/Burst Modes at**
  - Synchrotron Sources

- **Free Electron (X-Ray) Lasers**

- **Ultrafast Electron Microscopy**

- **High Time Resolution** allows you to observe unknown structures, transient states in matter, dynamics, and physics that cannot be captured with conventional techniques.

- **Ultrafast Laser Spectroscopy**

---

thomas.lagrange@epfl.ch • lumes.epfl.ch • +41 (0)21 6935861

Laboratory of Ultrafast Microscopy and Electron Scattering
Ultrafast TEM grew out of developments in time-resolved electron microscopes that date back to the 1960's.
There are growing number of UTEM installations worldwide
There are 2 laser based instrumentation approaches to Ultrafast Microscopy: Single-Shot and Stroboscopic.

**Single-Shot Method:** one pump excitation to the sample and one electron pulse producing one image

- Probes irreversible processes
- Main challenges: limitations in resolution

**Stroboscopic Method:** integration of millions of pump excitations to the sample and electron pulses producing one image

- Can only probe highly reversible processes
- High spatial resolution
- Limited methods for sample excitation
fs-Ultrafast Electron Microscopy: Stroboscopic Technique

Stroboscopic → *train of weak pulses*

UV laser → photocathode

IR laser → pump

Object

Multiple electron bunches → probe

Camera/detector

Many pulses produce the image

➢ for reversible transitions

Ultrafast Transmission Electron Microscopy

Delay between IR and UV photons

Δt

UV pulse

IR pulse
The Stroboscopic UTEM installed at IPCMS CNRS Strasbourg (Florian Banhart) achieves atomic resolution with a few ps electron pulses

**Resolution in thermionic mode**
Ta disc cathode, heated
Sample: gold nanoparticles
Laser repetition rate: 2 MHz
UV output power: 15 mW

**Resolution in photoelectron mode**
Laser repetition rate: 2 MHz
UV output power: 15 mW

Lattice image resolution ~0.23nm

PINEM spectrum
Laser repetition rate: 500 kHz
UV output power: 8 µW

IR photons $h\nu = 1.2$ eV

Photon-induced near-field electron microscopy energy gain/loss by interaction with photon field

Photon induced near-field electron microscopy (PINEM) can be used to study coherent photo-induced processes, such as plasmons.

PINEM provides a means for determining “time zero” and temporal resolution of the electron bunch.


The major challenge for Ultrafast Electron Microscopy is using fermions as a probe!

Coulomb interactions and replusion (electron-electron scattering) limit the temporal and spatial resolution of Ultrafast Electron Microscopy techniques!

Electron-Electron scatter events scramble the high-resolution information of the specimen encoded on the electron waves.
With high electron currents, electron-electron interactions limit brightness and spatial resolution.

**Lateral Space Charge**

- Virtual Lens crossover

**Defocusing:**
- To first order, it's just a diverging lens
- Correction via readjustment of microscope alignment
- Higher orders create spherical aberration
High Current electron pulses act as a “moving”, variable lens.

Lateral Space Charge effects can be overcome easily in the TEM (every lens in the microscope has a tunable focal length) but these effects require that laser be stable and we work at Space Charge Limit.
With high electron currents, electron-electron interactions limit brightness and spatial resolution.

**Longitudinal Space Charge**

Child's Law:
- Field from recent electrons retards emission
- Fundamentally limits current density ($j$):

$$j \propto V^{3/2} / d^2$$
Electron yield saturates as the cathode laser energy increases.

The number of electrons per pulse is a function of the gun design. High spatial coherence (small emitting area) gives low signal. Larger signals are generated from large areas (low spatial coherence).
With high electron currents, electron-electron interactions limit brightness and spatial resolution.

**Longitudinal Inhomogeneous Boersch Effect:**
- Pulse viewed in rest frame
- Most velocity is lateral
- Equilibration increases $\Delta E$
- Can create 1 kV energy spreads in high charge beams
- Cannot be corrected, only minimized
Coulomb interactions increase bunch length and energy spread, that induce aberrations and greatly reduce temporal and spatial resolution.

- **Boersch effects** reduce energy resolution.
- **Inhomogeneous and homogenous space-charge effects** decrease spatial and temporal resolution.
- **Integration time and signal** are traded-off with temporal resolution and coherence.


Unpublished data acquired on Strasbourg UTEM:

- **Stroboscopic mode, 15 mW UV**
  - 95 eV

- **Stroboscopic mode, 1 mW UV**
  - 0.8 eV
Does FEG tip geometry provide better coherence?

“Flat” Thermionic TEM gun cathode

a

cathode (-) -- Δα -- Δx

Δα

anode (+)

e−

effective source size 10 - 20 μm

“Sharp” Field Emission Gun cathode

b

supressor (-) -- Δα -- Δx

Δα

extractor (+)

focus anode (+)

e−

effective source size 10 - 100 nm

Armin Feist, Nora Bach, Nara Rubiano da Silva, Thomas Danz, Marcel Möller, Katharina E. Priebe, Till Domröse, J. Gregor Gatzmann, Stefan Rost, Jakob Schauss, Stefanie Strauch, Reiner Bormann, Murat Sivis, Sascha Schäfer, and Claus Ropers, Ultramicroscopy 176, 63 (2017).
Flat (thermionic gun) vs. Sharp tip (FEG photogun) designs: Which is better?

Each design trade-offs parameters:
- Coherence and Temporal Resolution Vs. Signal and Average Brightness

Space Charge Controls Brightness!
Both designs have similar photoelectron brightness
Flat photocathode temporal resolution characteristics

Space charge at the cathode
Boersch effect in the gun cross-over
- XY-spread: loss of coherence
- Z-spread: increase of pulse duration
- High charge: increase energy spread
- High bias: increases pulse duration and signal, but decreases in energy spread

Guard Ring Cathodes limit the photoemission area (no shank electrons) providing high temporal resolution and moderate signal

Improves coherence at high charge regime when operating at moderate bias voltages

LaB₆ Guard Ring Cathode

Standard Truncated LaB₆ filament

High-Fluence, femtosecond optical pump-probe are challenging

**Practical Limitations of laser pumps in the UTEM approach**

1. High power fs-lasers (>10 fs) have limited repetition rates, <1 MHz
2. Samples need to sufficiently cool between pump pulses

- High fluence laser conditions
- High-repetition rate can lead to melting and material modifications

Thin foil or nanostructures on TEM carbon support grid

Nano-puddle on TEM carbon support grid
Resolution in photoelectron mode with Plate Cathode

- Laser repetition rate: 2 MHz
- Exposure time: 10s
- UV output power: 15 mW
- **Electrons per pulse (@source): ~500**
- **Total number of electrons: >10^9**
- Throughput: ~1%
- Pulse duration at specimen: ~10ps

**JEMS simulated CTF**
Energy spread: 1.5 eV, sample drift: 0.2 nm/min, Stability, Voltage: 2ppm/min, OL current: 1ppm/min, Field noise: <20 nT
JEOL 2100 HR pole piece: Cc 1.4mm and Cs 1.0mm

![Au TEM Image](image_url)

Product of envelopes: Spatial, Temporal, Noise
High resolution imaging is challenging with single electron per pulse operation in FEG due to inherently long exposure times.

Hypothetical conditions for 5 e⁻/pulse operation of the FEG Sharp Tip
- FEG photogun
- Laser rep rate: 2 MHz
- ΔE: 1 eV
- Throughput: 10%
- 5 electrons per pulse @ source
- Total electrons: >1x10⁹
- Exposure time: 100 s
- 10 ps pulses

Is sub-nm phase contrast imaging possible with exposure times >1 min?

Low Average Brightness and microscope instabilities prevent high spatial resolution imaging.
End of Part 1
Part 2: UTEM experiments

Spatial coherence
Constant phase difference between $P_1$ and $P_2$

Temporal coherence
Constant phase at point $P$ between $\tau_1$ and $\tau_2$

Laser sources
- Temporal coherence given by the inverse of the linewidth
- Excellent spatial coherence (depends on cavity and transport optics)

Pulsed electron sources
- Temporally incoherent on the femtosecond timescale
- Spatial coherence depends on the size of the source
- Space-charge degrades coherence

Spatial and Temporal Incoherence
- $x_c \tau_c$

Temporal coherence
- $\tau_c = \infty$

Spatial coherence
- $x_c = \infty$

Temporal Incoherence
- $x_c = \infty$
Spatial and temporal coherence

**HRTEM phase contrast** (spatial resolution)

**Diffraction resolution**
Effect of transverse coherence

**Effect of space-charge on energy resolution in fs-EELS**

Source brightness is a fixed quantity

Small focused spot, small signal, but high coherence (900 nm)

Large spot, 5x higher signal, but lower coherence (450 nm)

Effect of Boersch effects in the gun

Product of envelopes
Spatial coherence

Temporal coherence
Noise

Effect of lens aberrations and source

thomas.lagrange@epfl.ch • lumes.epfl.ch • +41 (0)21 6935861

Laboratory of Ultrafast Microscopy and Electron Scattering
There is a finite Electron Brightness, and trade-offs are made between signal, time resolution, temporal and spatial coherence.

High temporal, spatial and energy resolution, as well as high signal are impossible to achieve all at once.

**Ultrafast EELS**
- central spot: small \( \Delta t \) and moderate \( \Delta E \)
- weak signal

**HR EELS**
- small \( \Delta E \)
- high signal

**HRTEM imaging**
- high bias
- good signal
- low \( \Delta E \)

**Imaging**
- small \( \Delta t \)
- high signal
- moderate \( \Delta E \) acceptable

Textured ground states rule the physics of novel materials

Examples of UTEM science and applications: charge density waves

Applications of UTEM: lattice dynamics

Using Convergent Beam Electron Diffraction (CBED) for probing phonon modes
Mapping the excess line shifts give the vibrational harmonics of the system for breathing and in-plane shear modes.
Skyrmions are nanometer sized topologically nontrivial defects

Dzyaloshinskii-Moriya interaction (DMI) favors canting of neighboring spins

- Bloch type
  MnSi, FeCoSi, FeGe, Cu2OSeO3, CoZnMn...

- Néel type
  Ferromagnet/heavy metal bilayers

The defined chirality of the spin configurations provides topological barrier (protection) that stabilizes the skyrmions
Ultrafast Cryo-Lorentz TEM TEAM in LUMES

Gabriele Berruto  Ivan Madan

Help with the experiments:
EPFL, ER-C Julich: Y. Murooka
EPFL: G. M. Vanacore

Sample preparation:
Uni Glasgow: R. Lamb, D. McGrouther
Uni Osaka: Y. Togawa

Funding:
We use time-resolved cryo-Lorentz-transmission electron microscopy to characterize Skyrmion dynamics.

Manipulation of the magnetization texture using ultrafast laser pulses, and monitoring with high real space (nm) and time (ps) resolution.
Writing and erasing of Skyrmions was studied using \( ns \)-stroboscopic pump-probe experiments in a cryo-Lorentz imaging mode.

- Large and regular SkL upon adiabatic field cooling
- Skyrmions can be created by a single laser pulse
- Skyrmions erasing by magnetic field excitation, or by laser pulses with much higher fluence

\[ T = RT \]
\[ \vec{B} = 0 \, G \]

\[ T = 200 \, K \]
\[ \vec{B} = 0 \, T \]

\[ T = 200 \, K \]
\[ \vec{B} \sim 750 \, G \]

Write Skyrmions using laser pulses \( \vec{B} \sim 750 \, G \)
It is possible to “photocreate” skyrmions by irradiating the sample (in the helical phase) with multiple fs pulses.

Skyrmions are created at the edges, gradually filling of the sample with skyrmions with multiple laser pulses.

At a fluence of 2.5X above the threshold, skyrmions are mostly erased, i.e., they are annihilated, but a few of them reform during the cooling.
Example: Coherent control of Light-Electron interactions

Transition radiation

How can light and $e^-$ interact?
Surface plasmons polaritons and plasma resonances

Surface plasmon:
Electrons-light coupled at an interface

Plasma resonance

Suggested reading: Dressel, “Electrodynamics of solids”
Energy filtered TEM mapping in UTEM PINEM experiments


Surface plasmons polaritons and plasma resonances

Most cited article in Nat Comm since 2015

Lummen et al., *Nat. Comm.* 7 13156 (2016)
Coherent control of plasma resonances

Holography of plasma resonance

- Attosecond mapping of plasmon propagation
- Direct measurement of group and phase velocity

News and Views article: Ropers, Nature 571, 331 (2019)
Coherent control of the transverse electron wavefunction

- Chiral SPP launched by elliptically polarized light

- Simulated Chiral SPP

- A chiral plasmonic field can impart a phase singularity onto the transverse component of an e⁻ wavefunction


New and Views article: Jun Yuan
*Nature Materials* 18, 533 (2019)

https://www.youtube.com/watch?v=TU-mlh2LSI&feature=emb_logo
Generation of electron vortex beams

Electron wavefunction microscopy in momentum space

High Dispersion Electron Diffraction (CL~80m)

Coherent control of vortex beams

Three pulses experiment: attosecond control of the topological charge of an $e^-$ wavefunction between $+1$ and $-1$
Generating vortex electron beams with forked phase plates

Phase plate side view

SEM image

Diameter: 5 μm
Pitch: 350 nm

Nanofabrication

Simulations

Experimental
EMCD is based on the analysis of ionization edges in Electron Energy Loss Spectroscopy (EELS), allowing to quantitatively determine the spin and the orbital magnetic moments with atomic spatial resolution depth sensitivity.

**XMCD from literature** (H. Shiozawa et al. Scientific Reports (2015))

Mapping of spin polarized $2p_j \rightarrow 3d$ transitions in Ni

- $L_2$ - edge: $2p_{1/2} \rightarrow 3d$
- $L_3$ - edge: $2p_{3/2} \rightarrow 3d$

**EMCD with phaseplates**

Veronica Lecesse
Laser free UTEM for magnetic imaging with ~10 ps resolution

Xuewen Fu et al. Sci Adv 2020
Thanks for your attention!