

EPFL

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Time-Resolved (Ultrafast) Electron Microscopy



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



Fabrizio Carbone

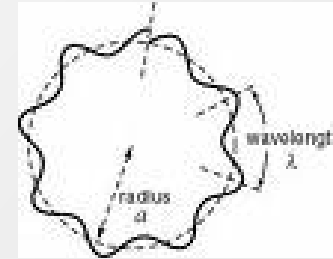
Funding sources



- 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

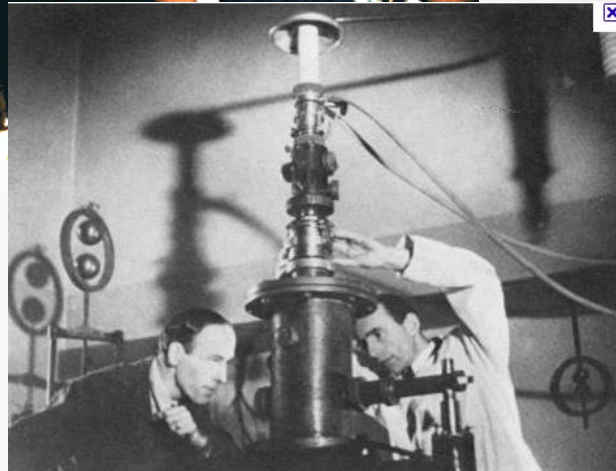
L. de Broglie (1923) postulates that $p=h/\lambda$ is valid for all particles λ wavelength of electrons (Nobel prize 1929)



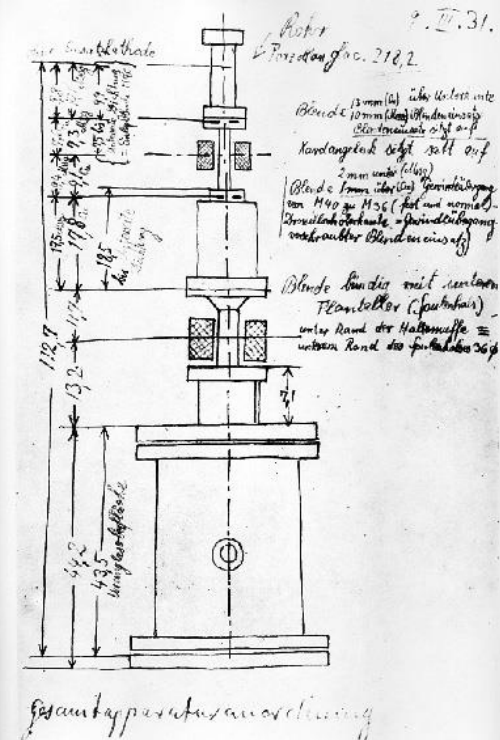
1986: E. Ruska Nobel Prize



E. Ruska: 1931



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: $E = hc/\lambda$ so if $\lambda < 5 \text{ nm}$, $E > 1 \text{ keV}$

Electron wavelength according to de Broglie equation: $\lambda = h/p$

with $p = m_0 v = (2m_0 eV)^{1/2}$

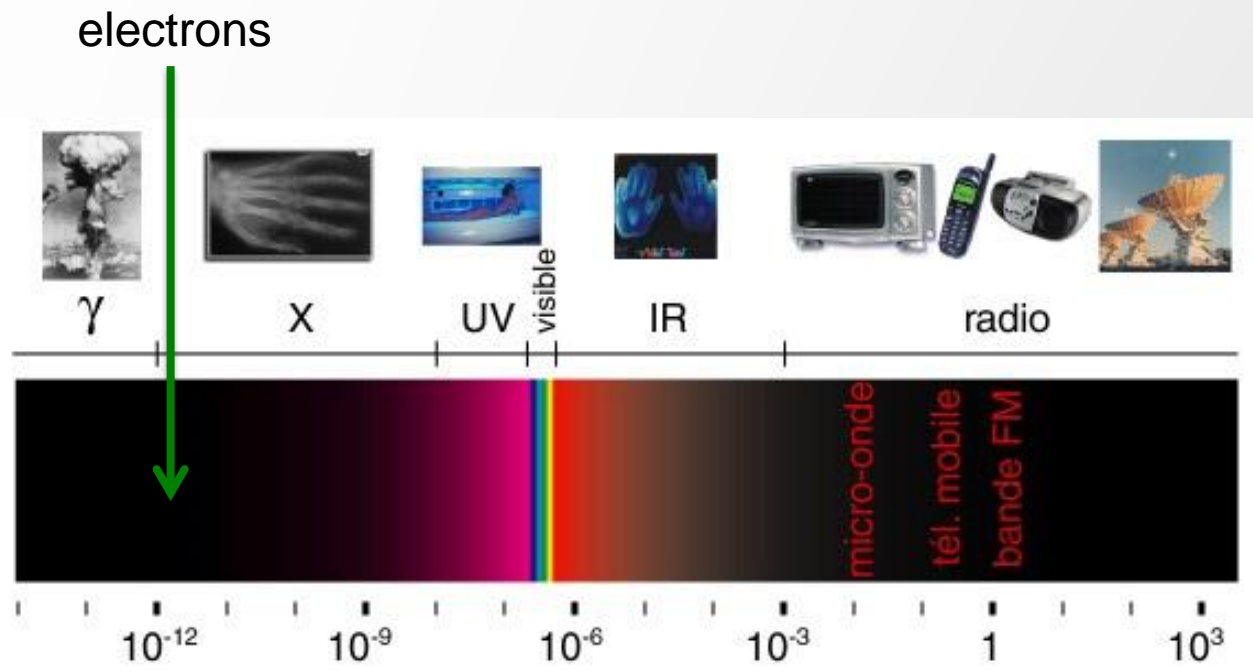
non relativistic (<50keV):

$$\lambda = \frac{h}{(2m_0 eV)^{1/2}}$$

Relativistic correction

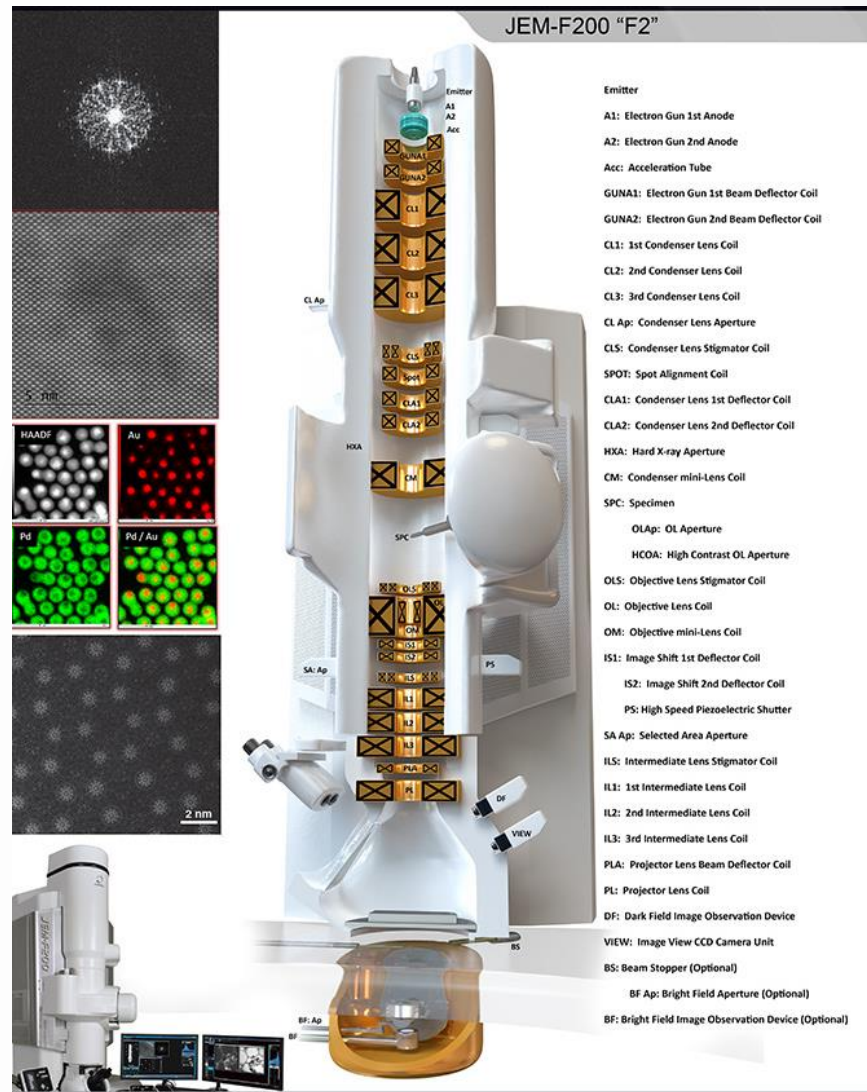
(>50keV or >1% speed) :

$$\lambda = \frac{h}{\left[2m_0 eV \left(1 + \frac{eV}{2m_0 c^2}\right)\right]^{1/2}}$$



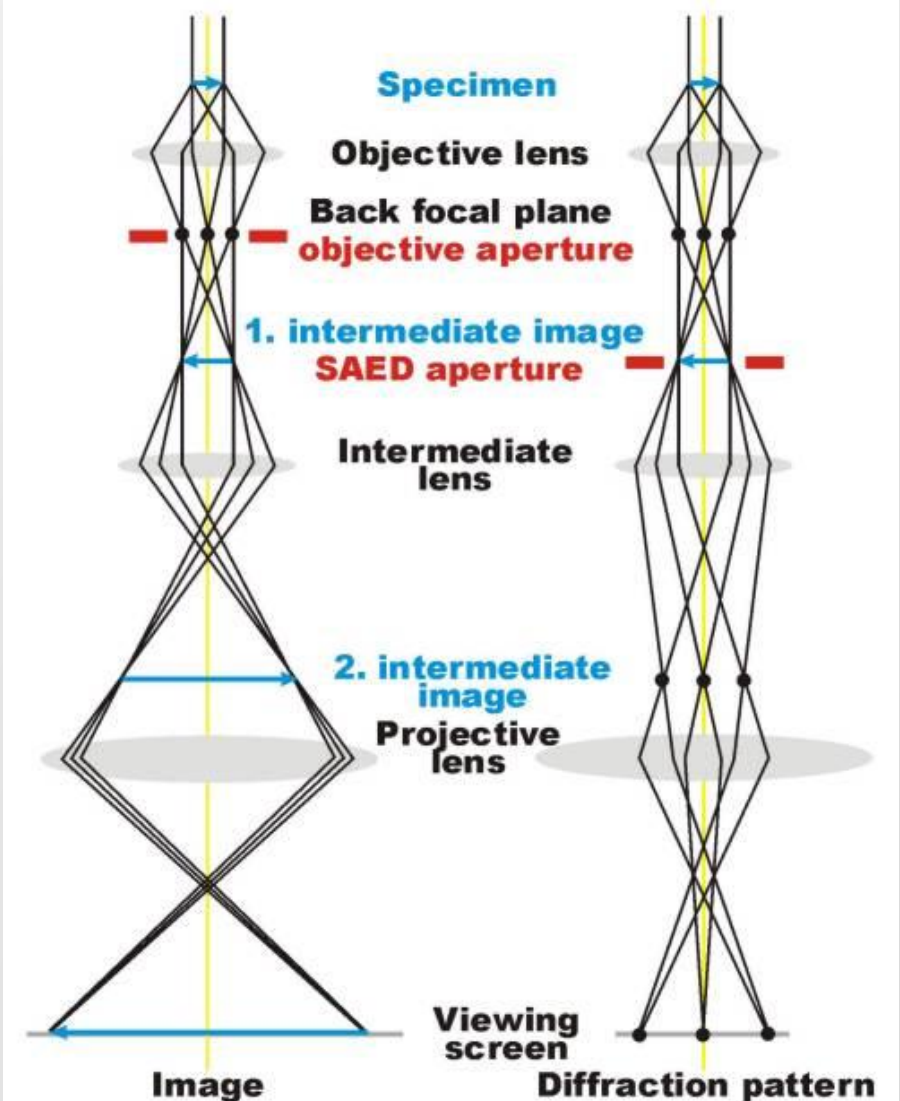
voltage [KV]	Nonrelativistic λ [pm]	Relativistic λ [pm]
1	38.79	0.03878
10	12.27	0.01221
80	4.34	0.00418
200	2.74	0.00251
300	0.00224	0.00197
1000	0.00123	0.00087

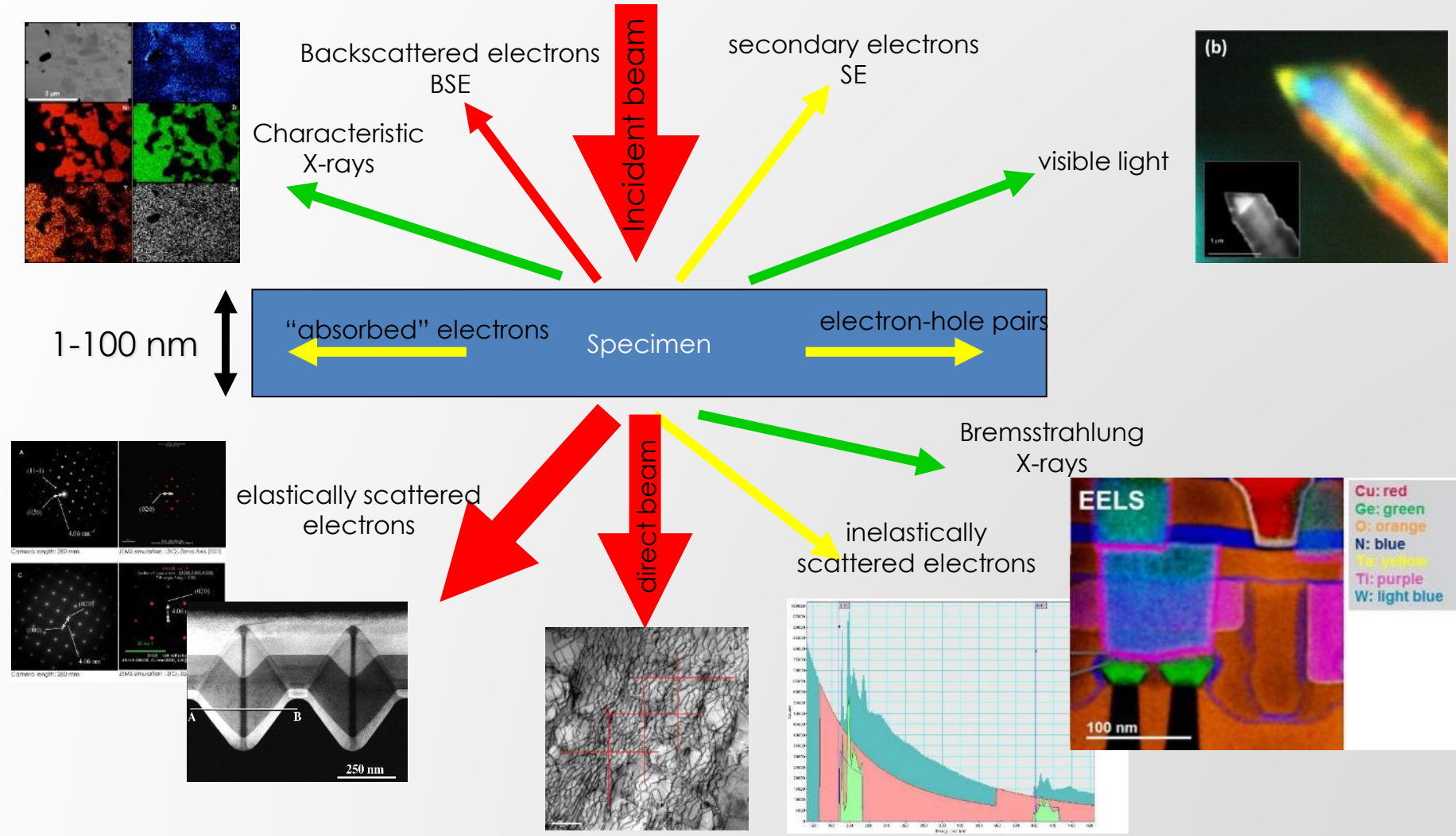
TEMs have sets of electromagnetic and electrostatic lenses and deflector that allow to flexibly change between many different imaging modes

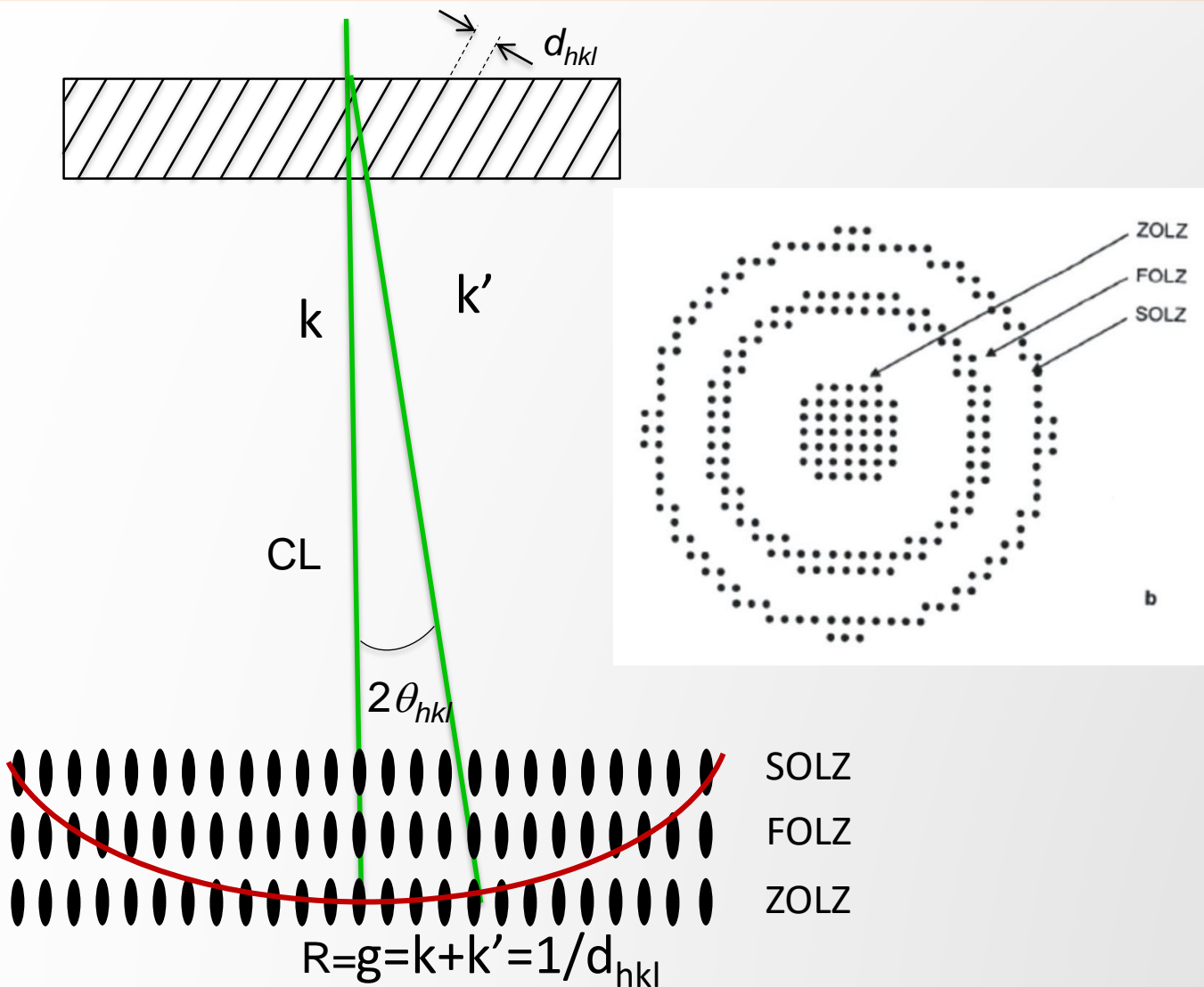


❑ Diffraction mode
The intermediate and projector lens magnify and project the **back focal plane (first diffraction pattern formed in the microscope)** of the objective lens to the detector system

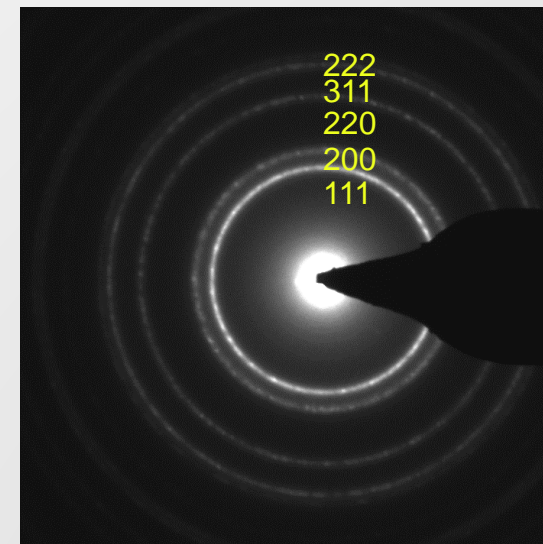
❑ Imaging mode
The intermediate and projector lens magnify and project **the image plane** of the objective lens to the detector system







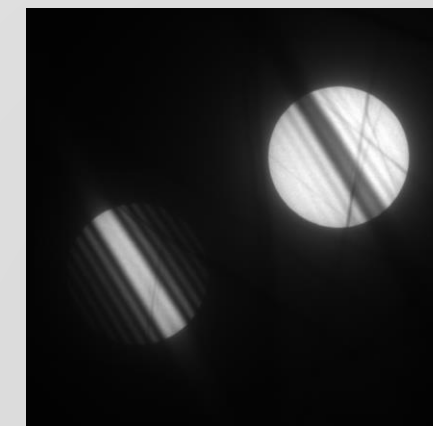
Bragg Diffraction Powder Pattern



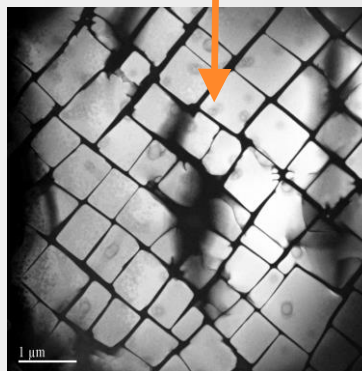
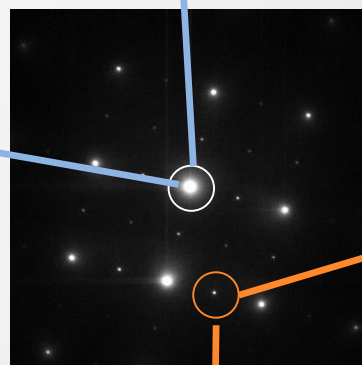
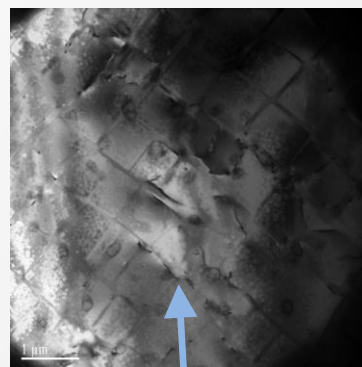
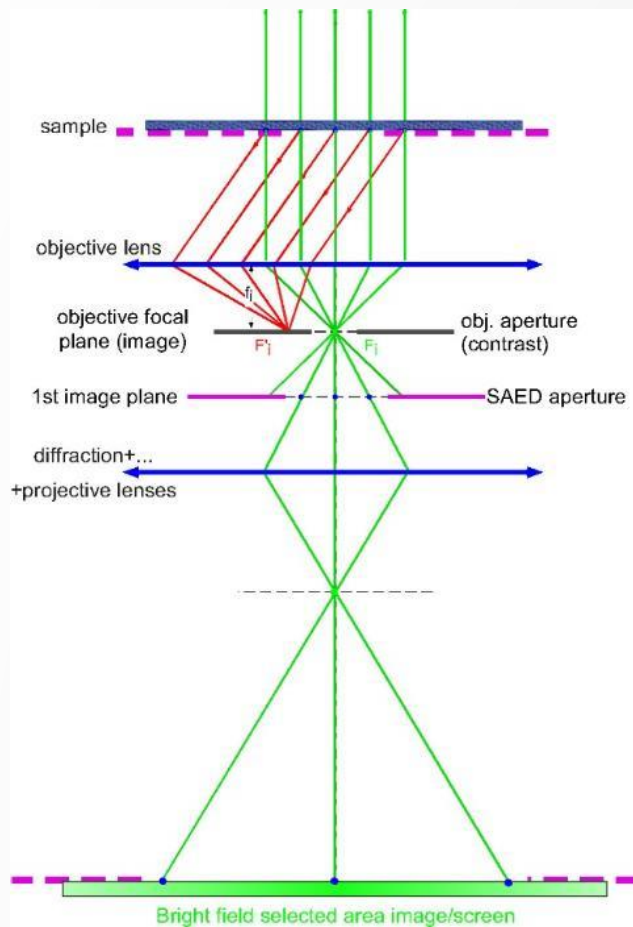
Single Crystal Spot Pattern



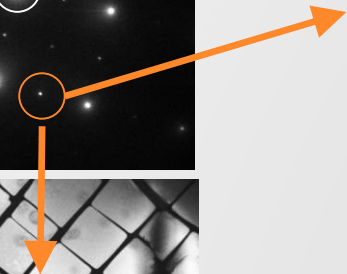
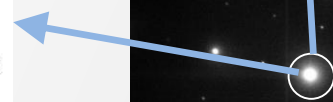
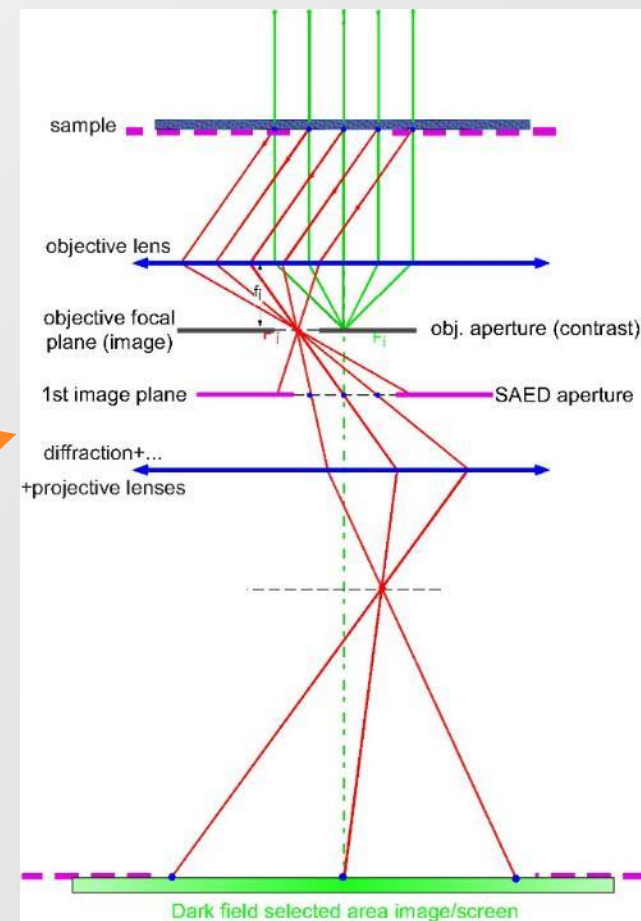
Convergent Beam Electron Diffraction



Bright-field

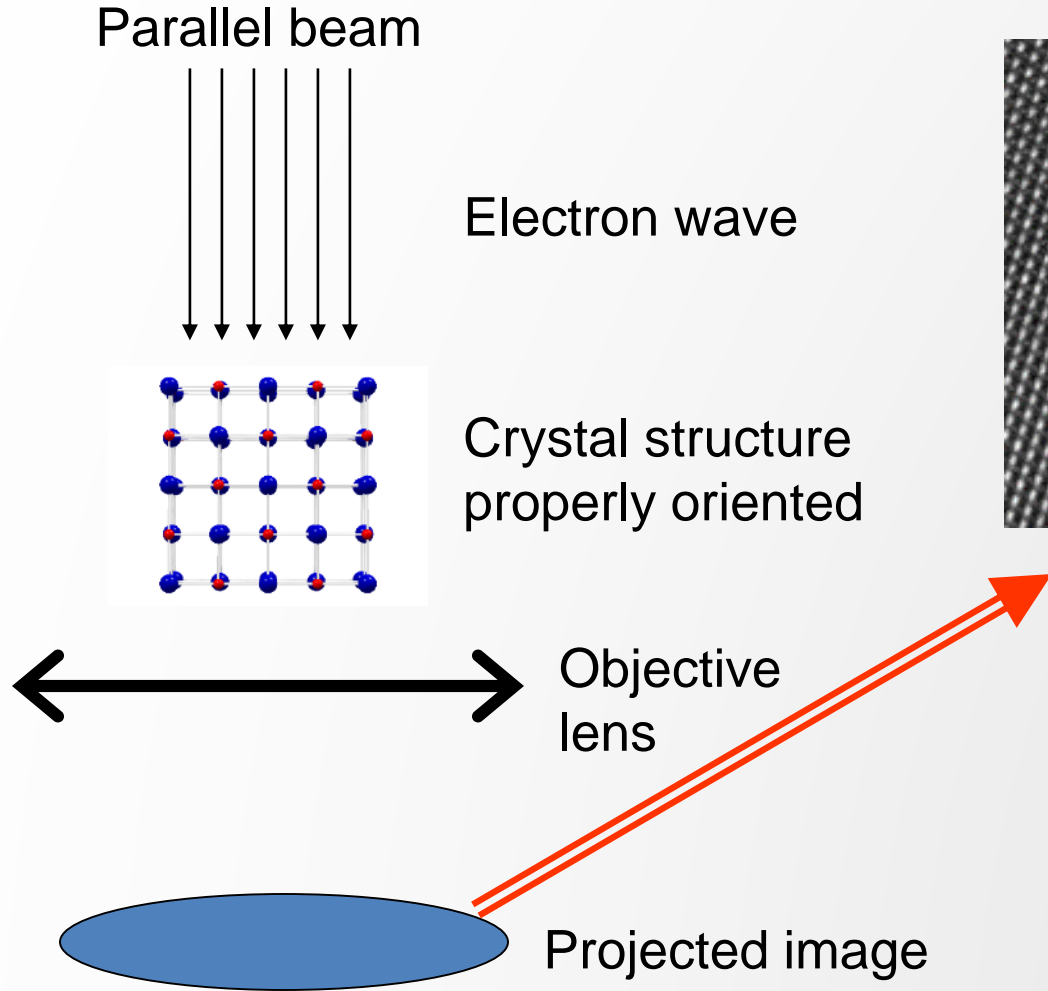


Dark-field

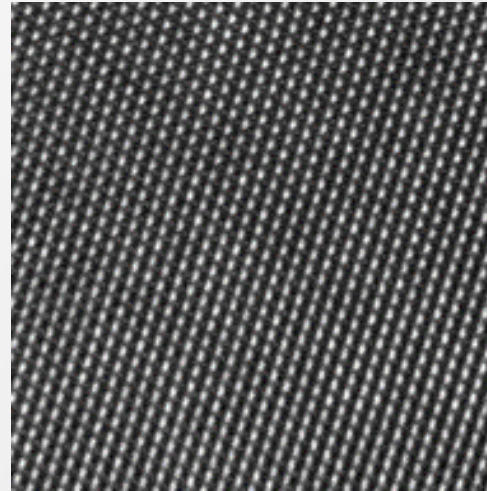


Phase Contrast Imaging: High Resolution Transmission Electron Microscopy (HRTEM)

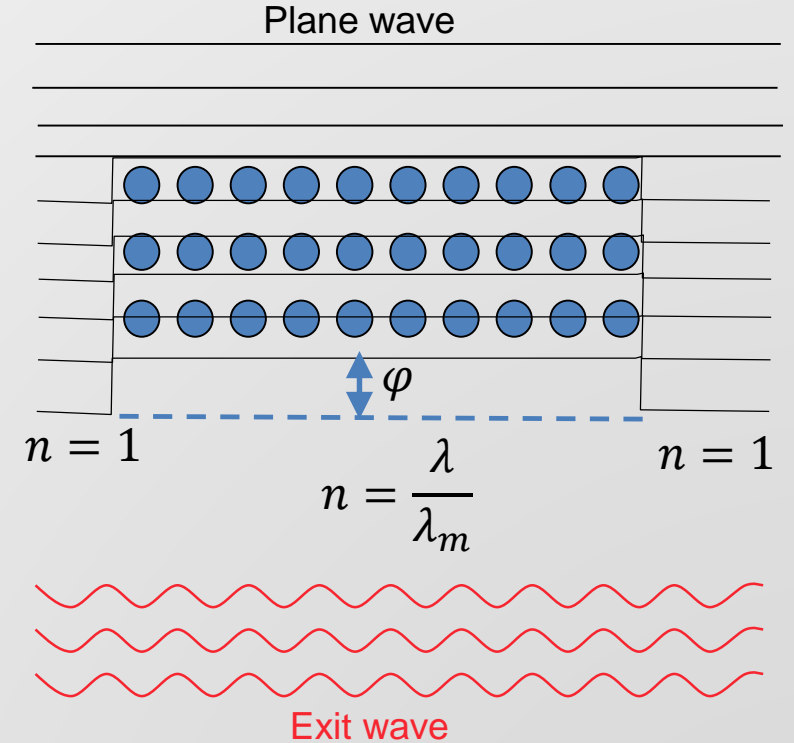
Phase contrast for crystalline specimen



Atomic resolved HRTEM image of Silicon



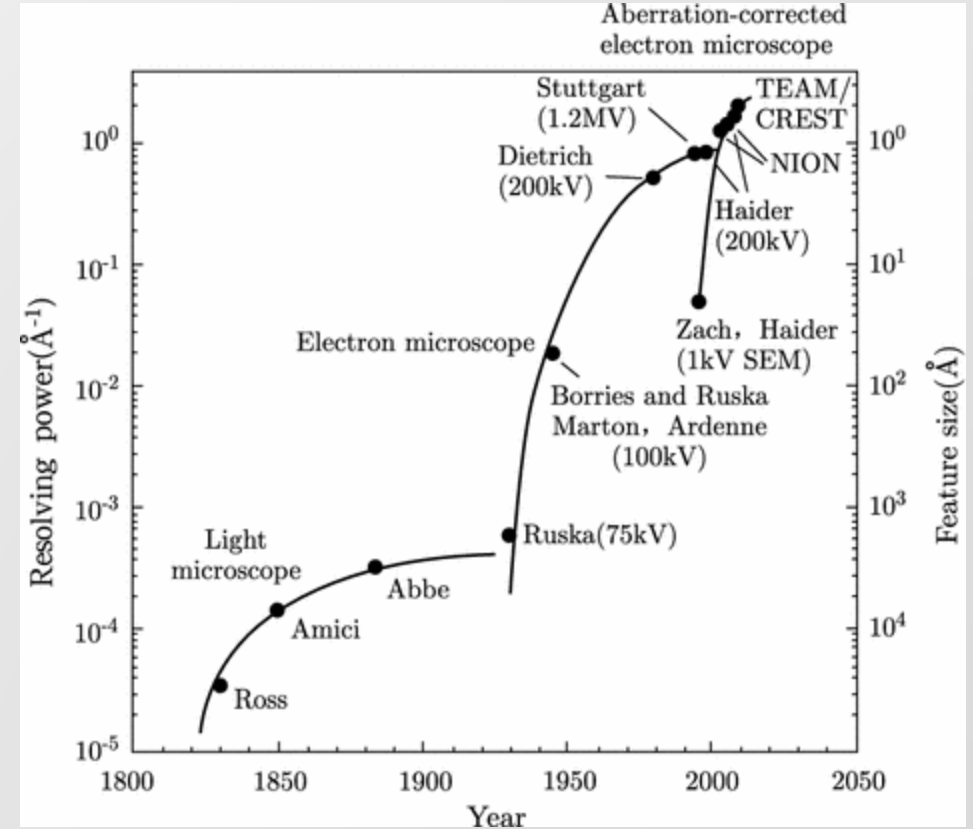
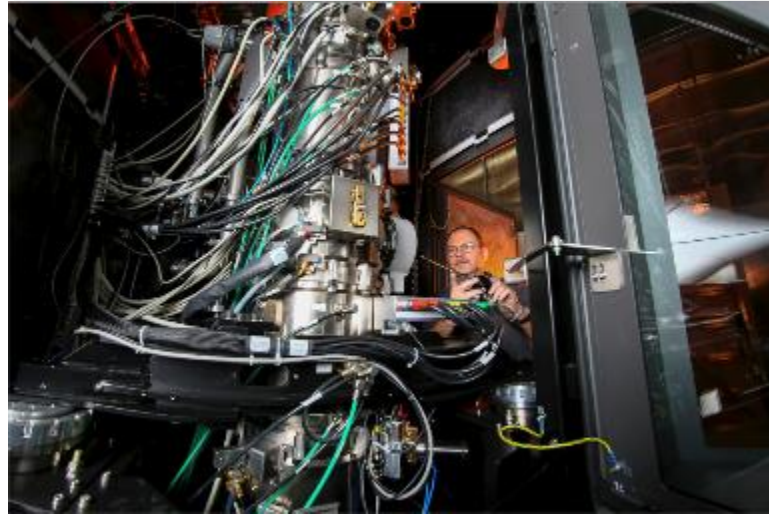
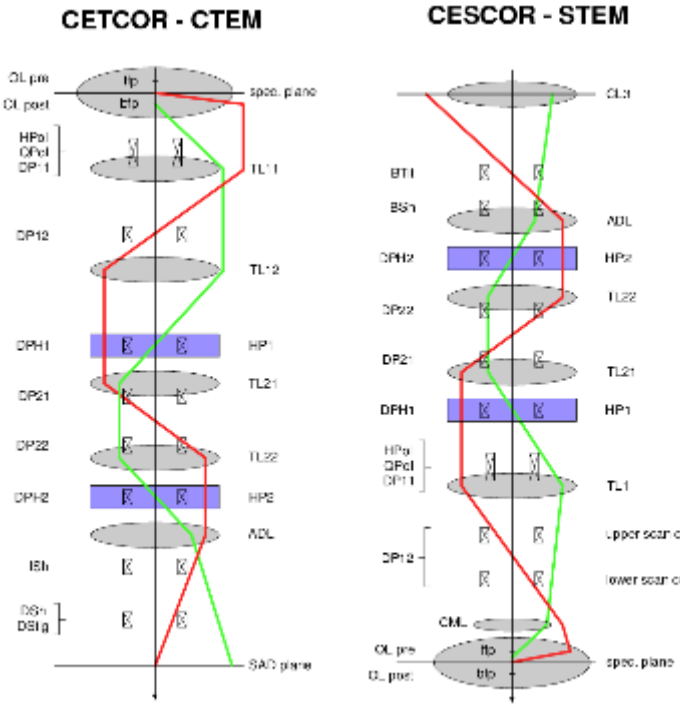
$$\Psi_i = \Psi_o \exp(2\pi i \vec{r})$$



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

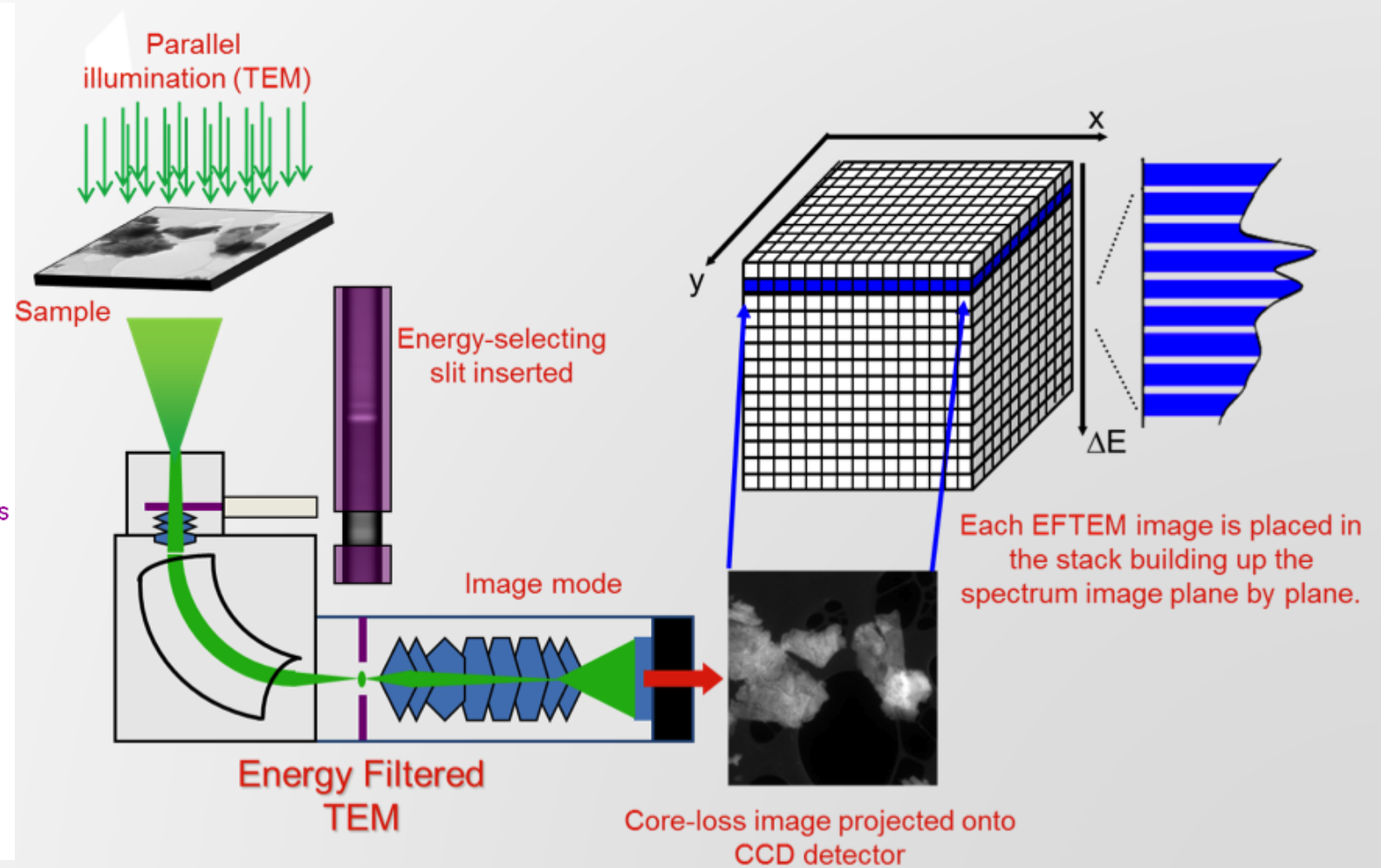
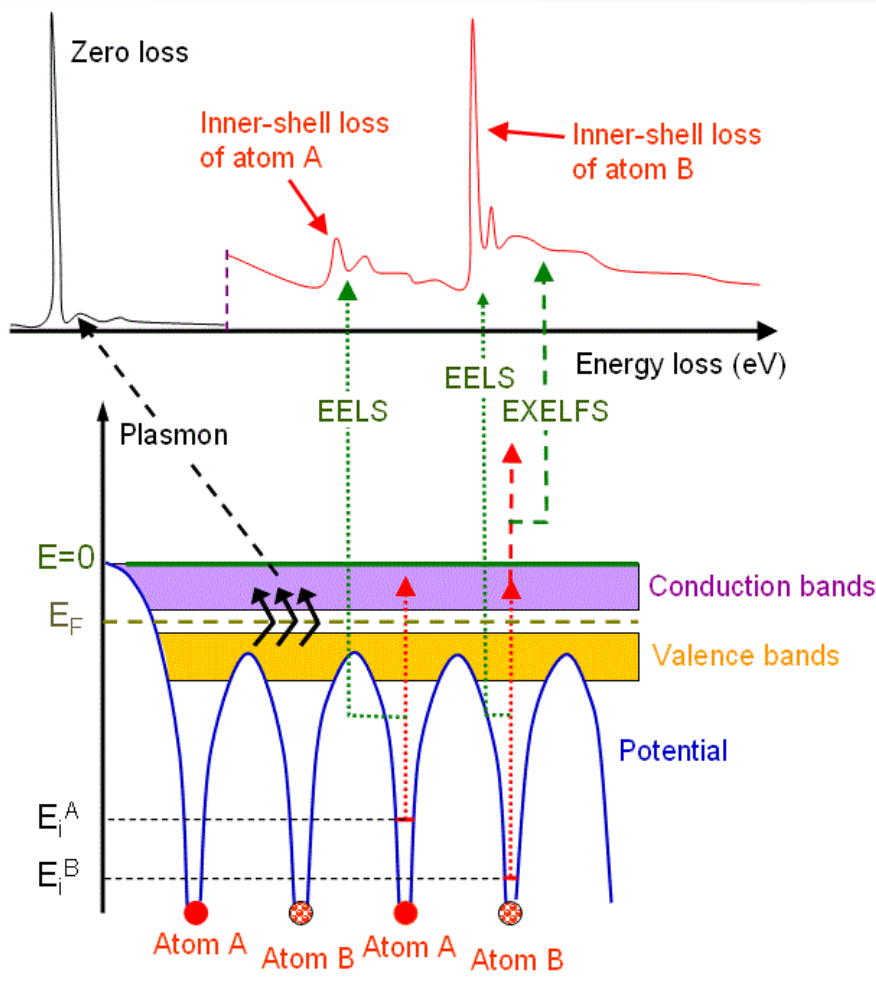
Exit wave function in terms of weak phase object approximation (WPOA)

The advent Aberration correctors have pushed the boundaries of spatial resolution (spatial coherence)

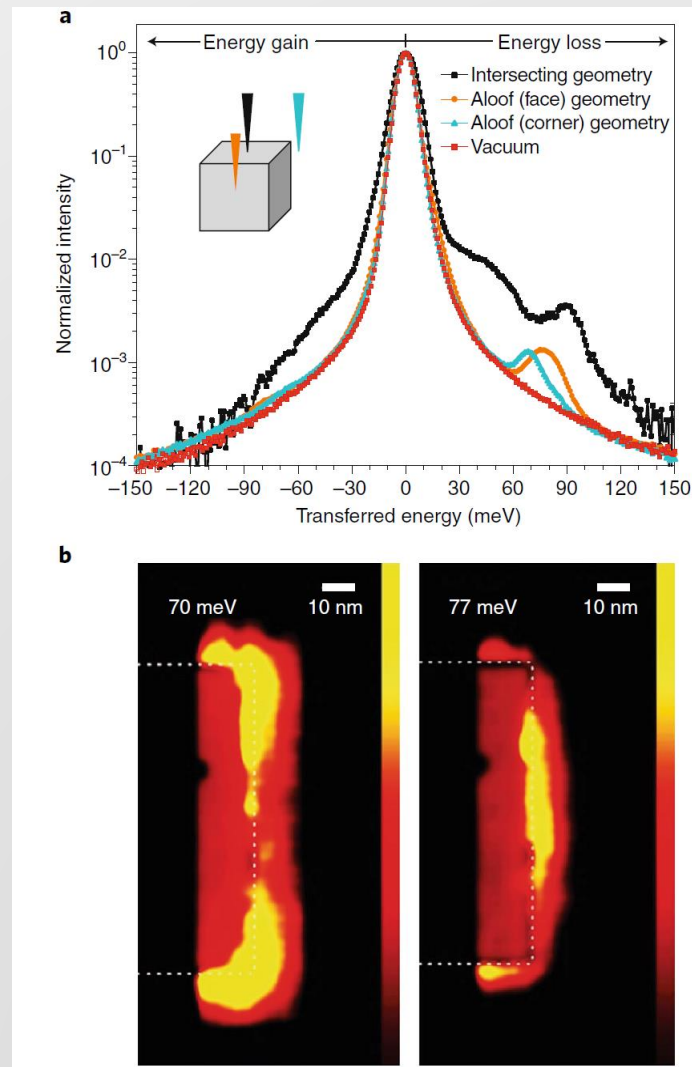
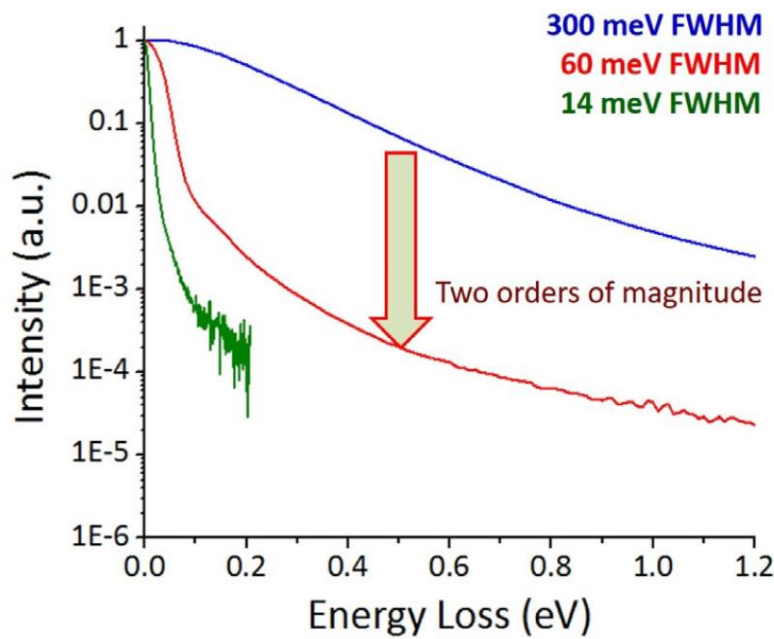
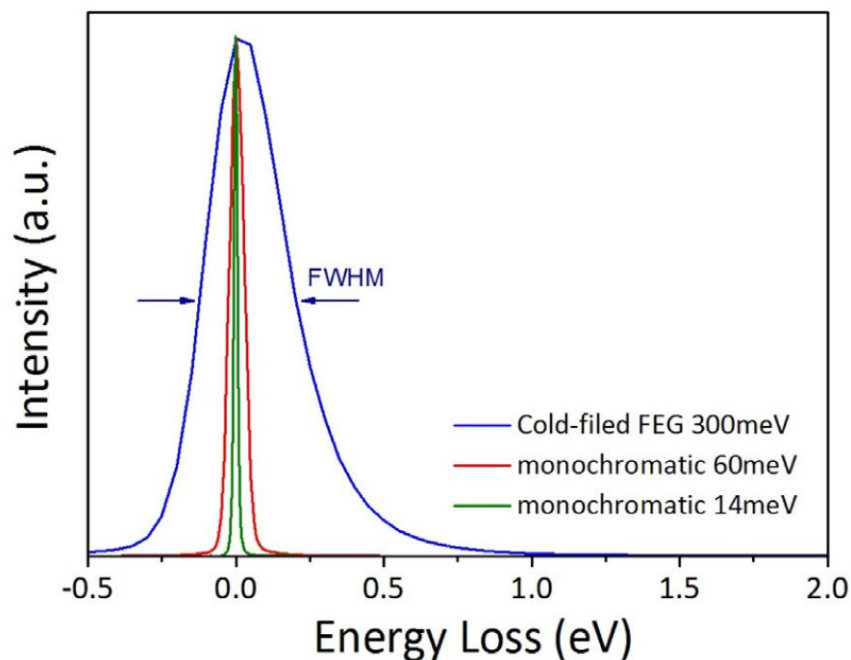


Uncorrected, resolution is $\sim 100 \times$ diffraction limit
 Cs Correction, resolution is $\sim 15 \times$ diffraction limit

Energy filtered TEM with core loss electrons



EELS technology has made great advancements in the past decade that local phonon modes can be probed on the sub-nanometer scale.



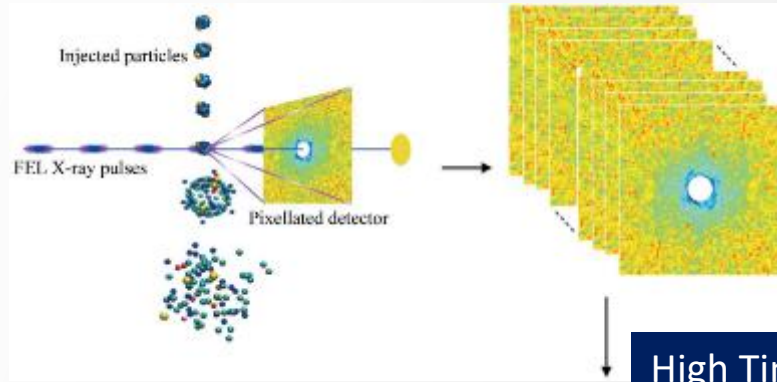
Maureen J. Lagos, Andreas Trügler, Ulrich Hohenester, and Philip E. Batson, *Nature* **543** (7646), 529 (2017).

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

Lagos, M. J., Trügler, A., Hohenester, U. & Batson, P. E. Mapping vibrational surface and bulk modes in a single nanocube. *Nature* **543**, 529–532 (2017).

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

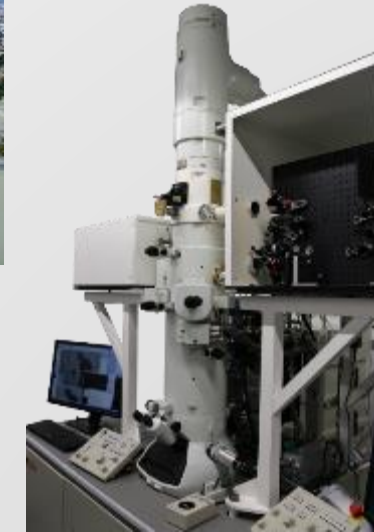
Diffract before Destroy Protein Structural Analysis



Bunch/Burst Modes at Synchrotron Sources



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

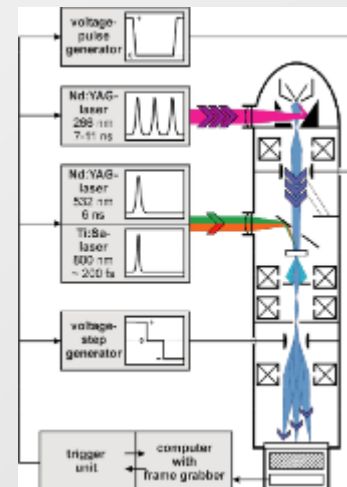
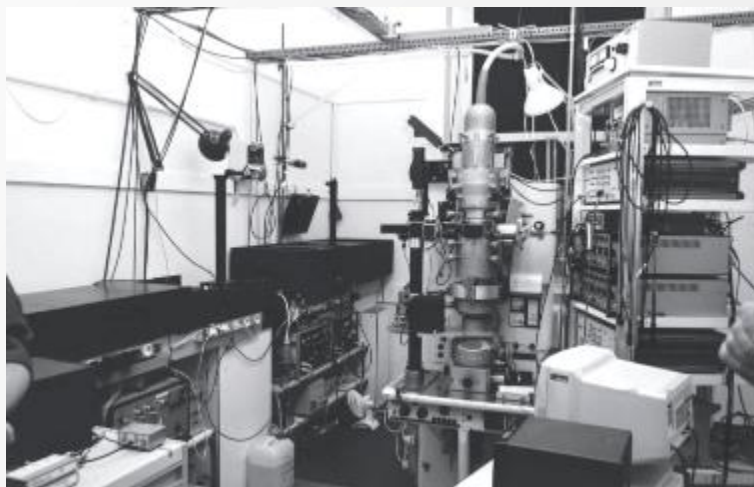
Free Electron (X-Ray) Lasers



Ultrafast Laser Spectroscopy

Ultrafast TEM grew out of developments in time-resolved electron microscopes that date back to the 1960's

Technical University of Berlin

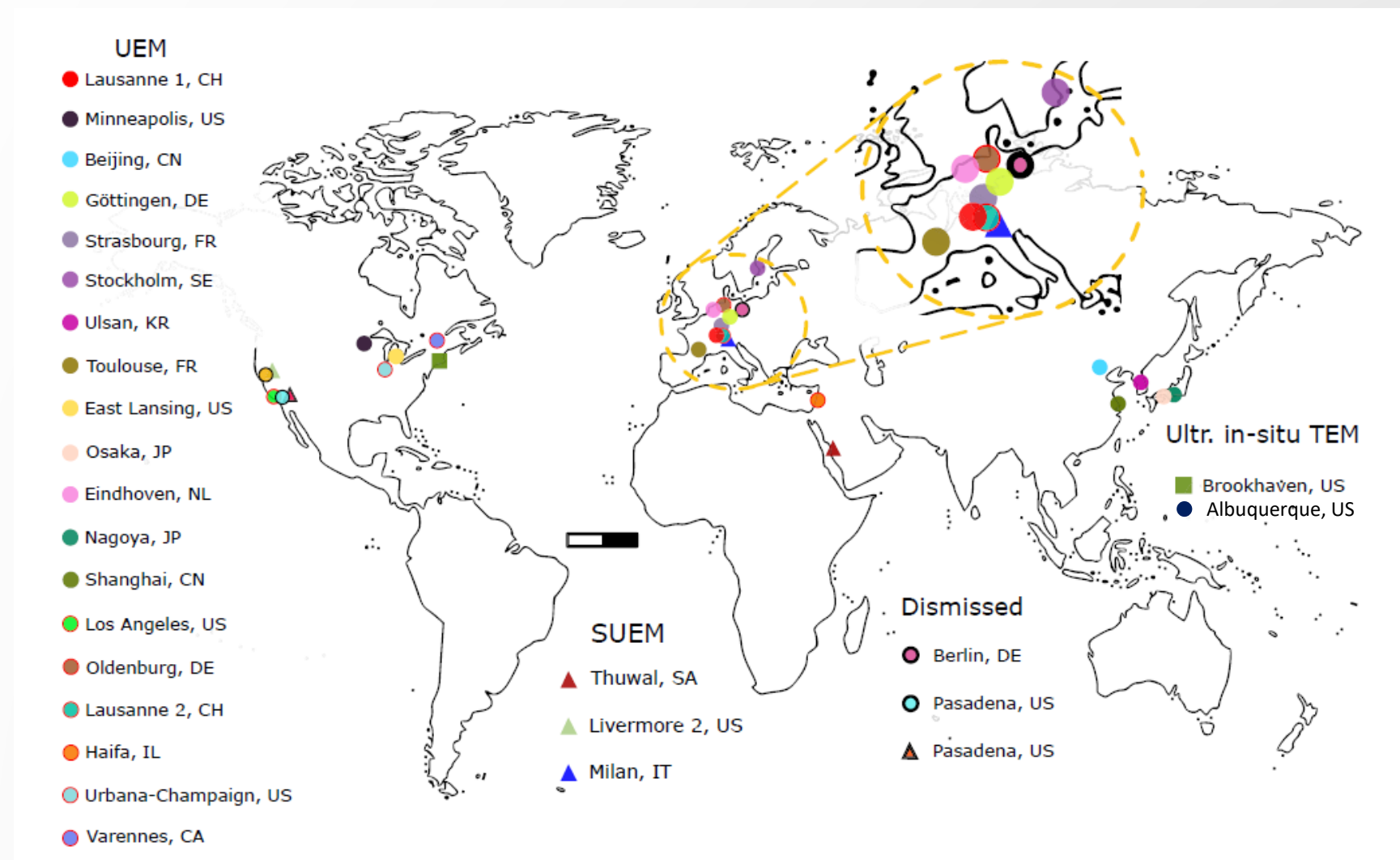


Lawrence Livermore National Lab



Caltech

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



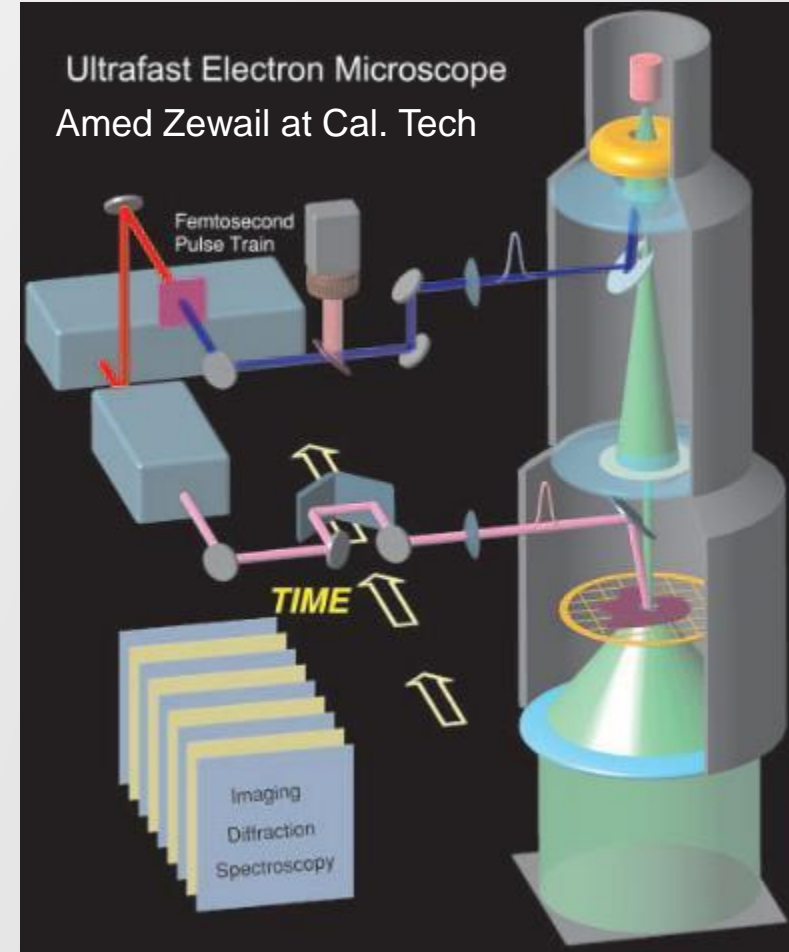
Single-Shot method

- Probes irreversible processes
- Main challenges: limitations in resolution

Stroboscopic method

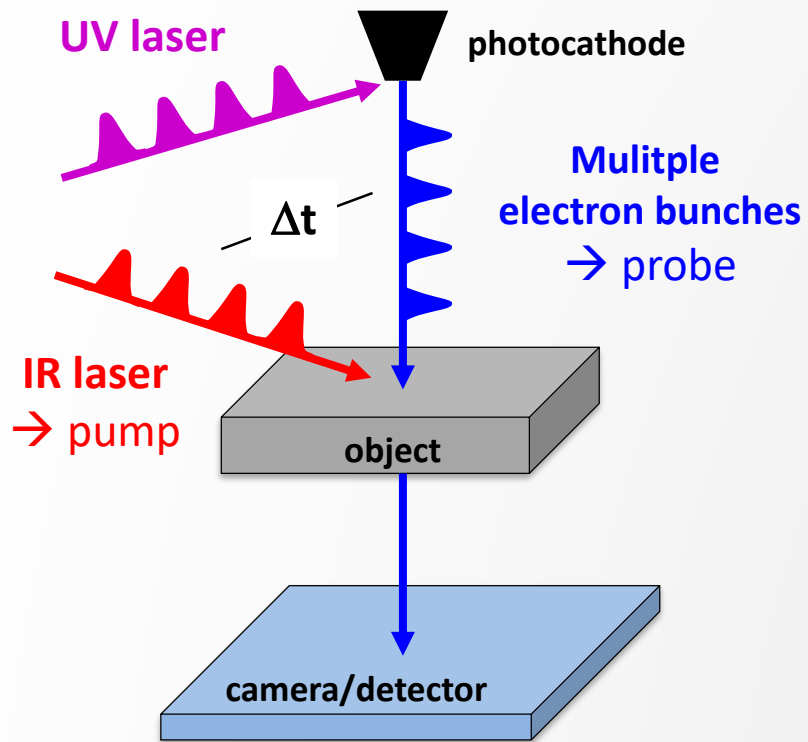
- Can only probe highly reversible processes
- High spatial resolution
- Limited methods for sample excitation

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



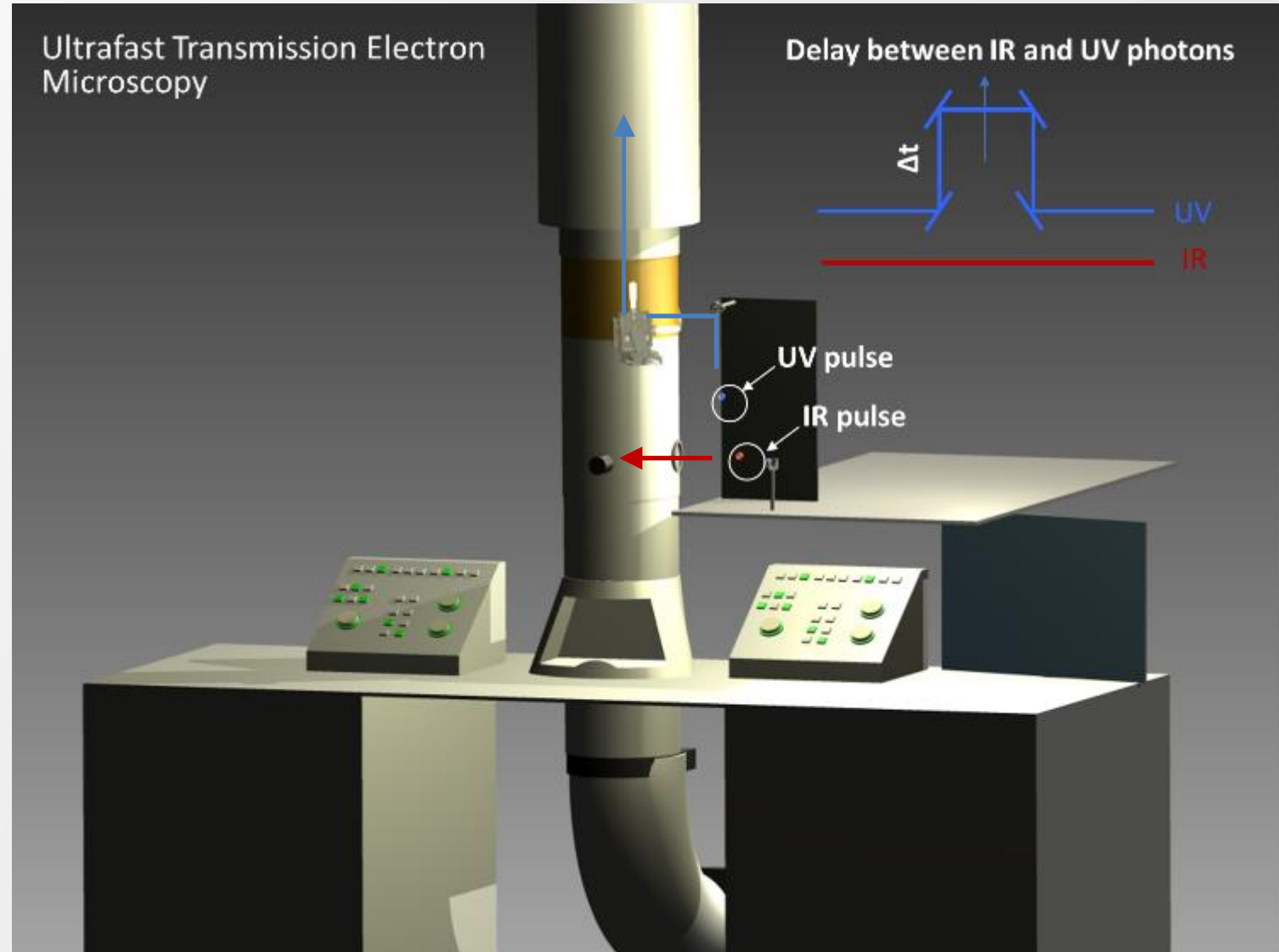
Stroboscopic

→ *train of weak pulses*



many pulses produce the image

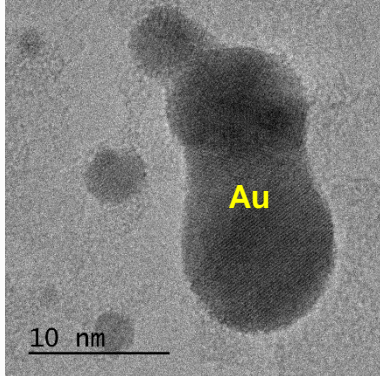
➤ for reversible transitions



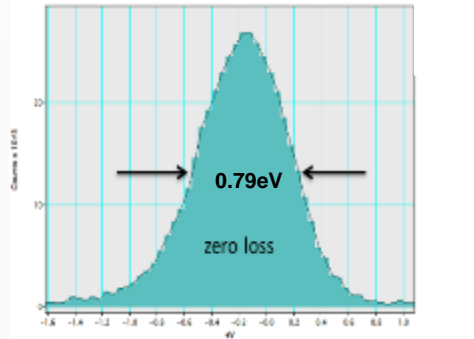
The Stroboscopic UTEM installed at IPCMS CNRS Strasbourg (Florian Banhart) achieves atomic resolution with a few ps electron pulses

Resolution in thermionic mode

Ta disc cathode, heated
Sample: gold nanoparticles



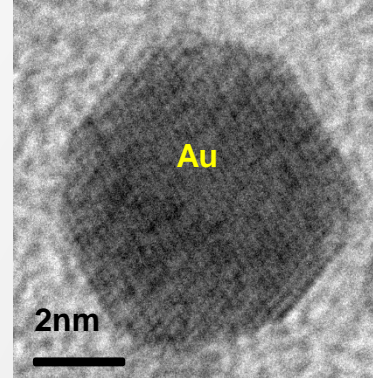
Laser repetition rate: 2 MHz
UV output power: 1 mW



Spread of zero loss
resolution ~ 0.79 eV

Resolution in photoelectron mode

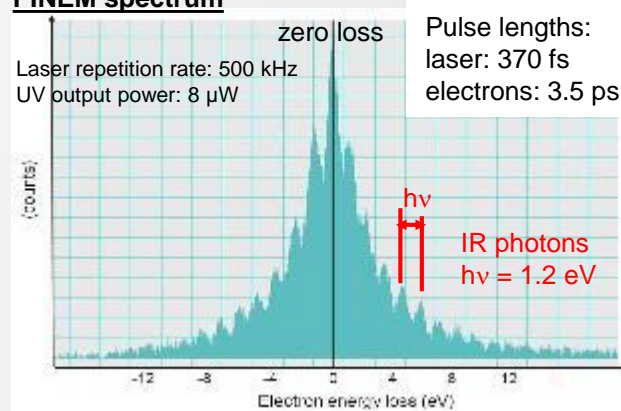
Laser repetition rate: 2 MHz
UV output power: 15 mW



lattice image
resolution
~0.23nm

PINEM spectrum

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

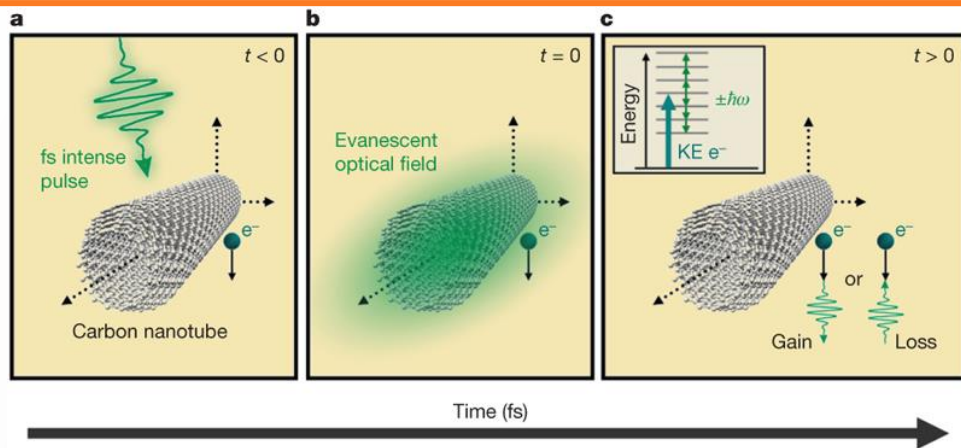


Photon-induced near-field electron microscopy
energy gain/loss by interaction with photon field



K. Bucker, M. Picher, O. Crégut, T. LaGrange, B.W. Reed, S.T. Park, D.J. Masiel, F. Banhart, "Electron beam dynamics in an ultrafast transmission electron microscope with Wehnelt electrode, *Ultramicroscopy, Volume 171 (2016) pp.8-18*

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

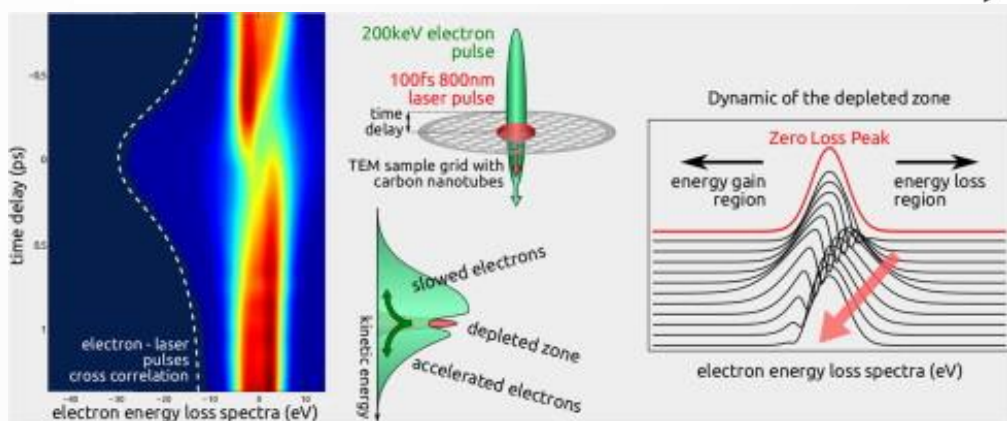


Inelastically accelerated electrons

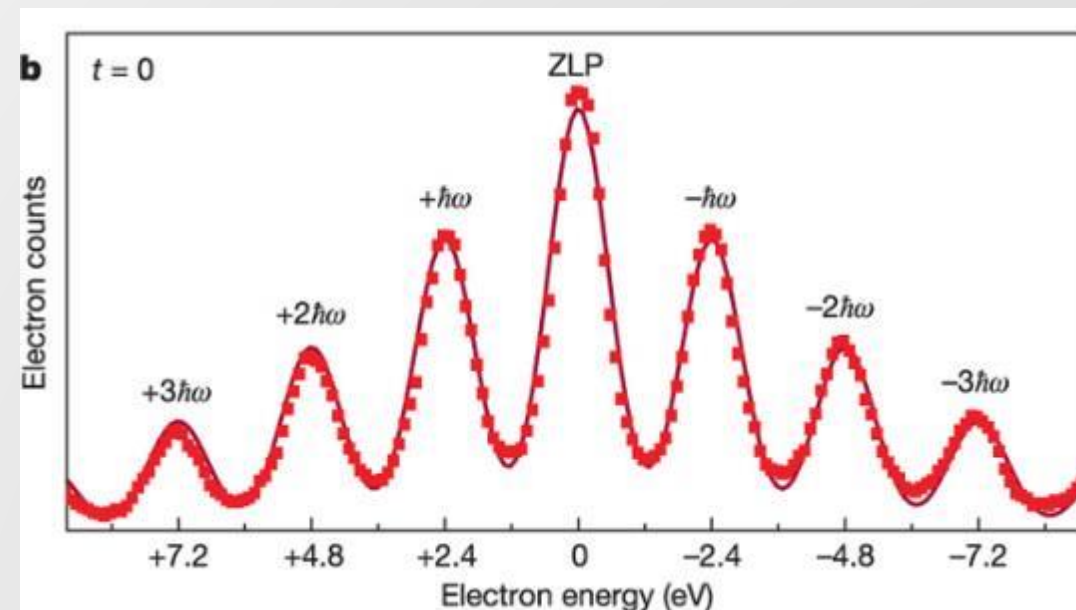
$$200 \text{ KeV} + n \cdot \hbar\omega$$

Inelastically decelerated electrons

$$200 \text{ KeV} - n \cdot \hbar\omega$$



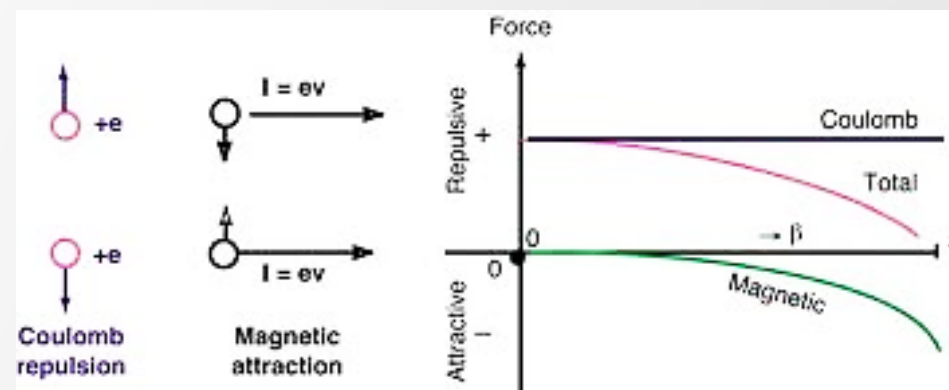
L. Piazza et al., *Chem Phys* **423**, 79 (2013).



Brett Barwick, David J. Flannigan & Ahmed H. Zewail *Nature* **462**, 902-906 (2009)

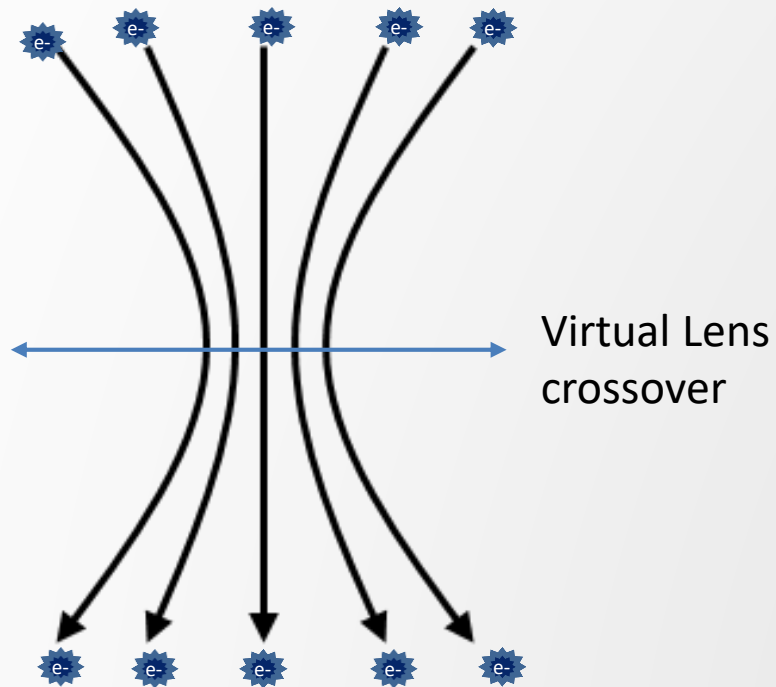
PINEM provides a means for determining “time zero” and temporal resolution of the electron bunch

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

Lateral Space Charge

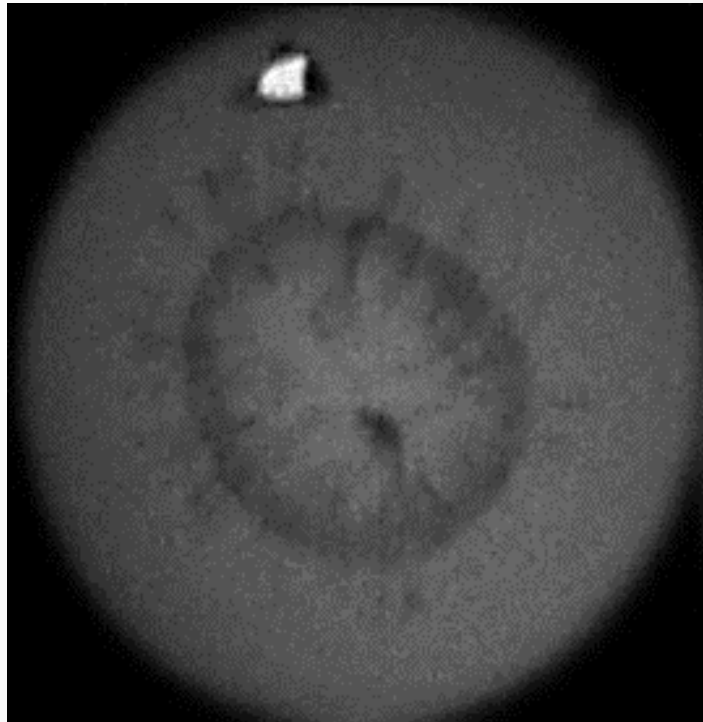


Defocusing:

- 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.

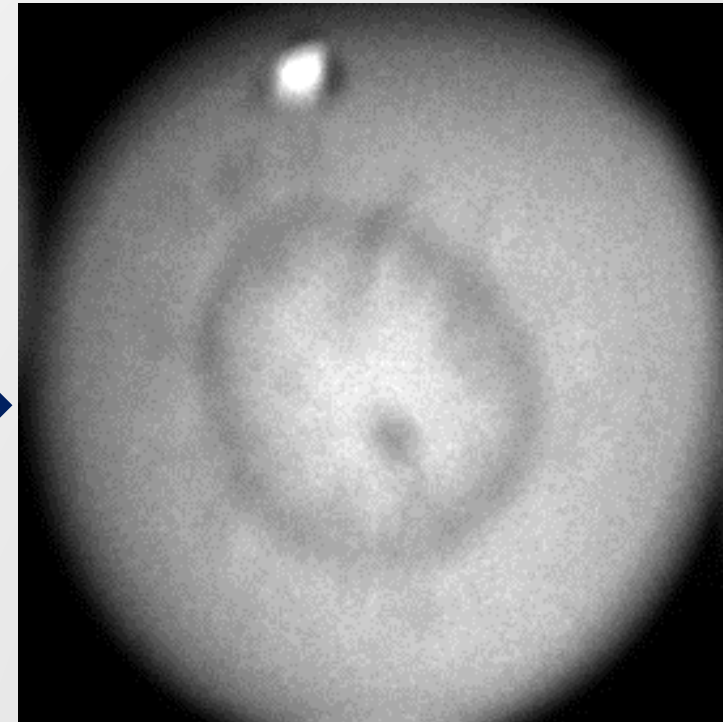
Focused image



5X more electrons

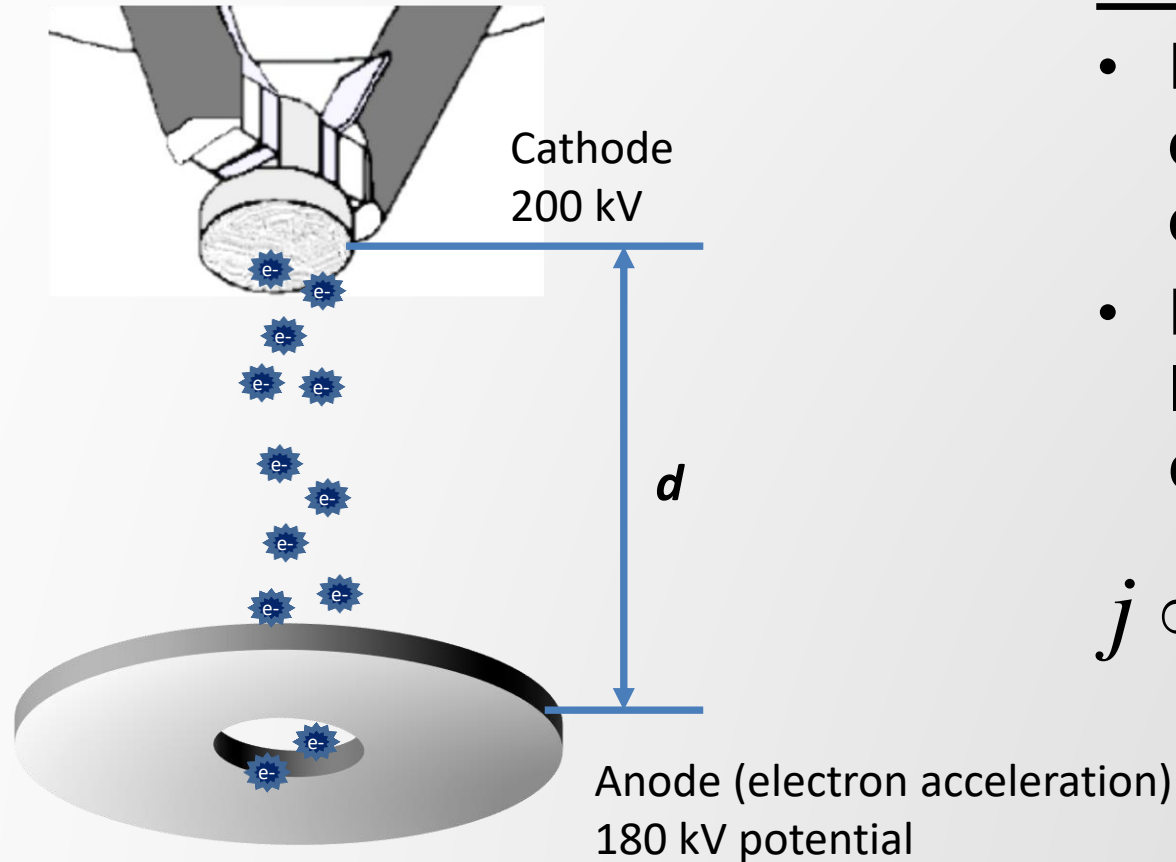


Defocused due to Lateral Space Effects



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

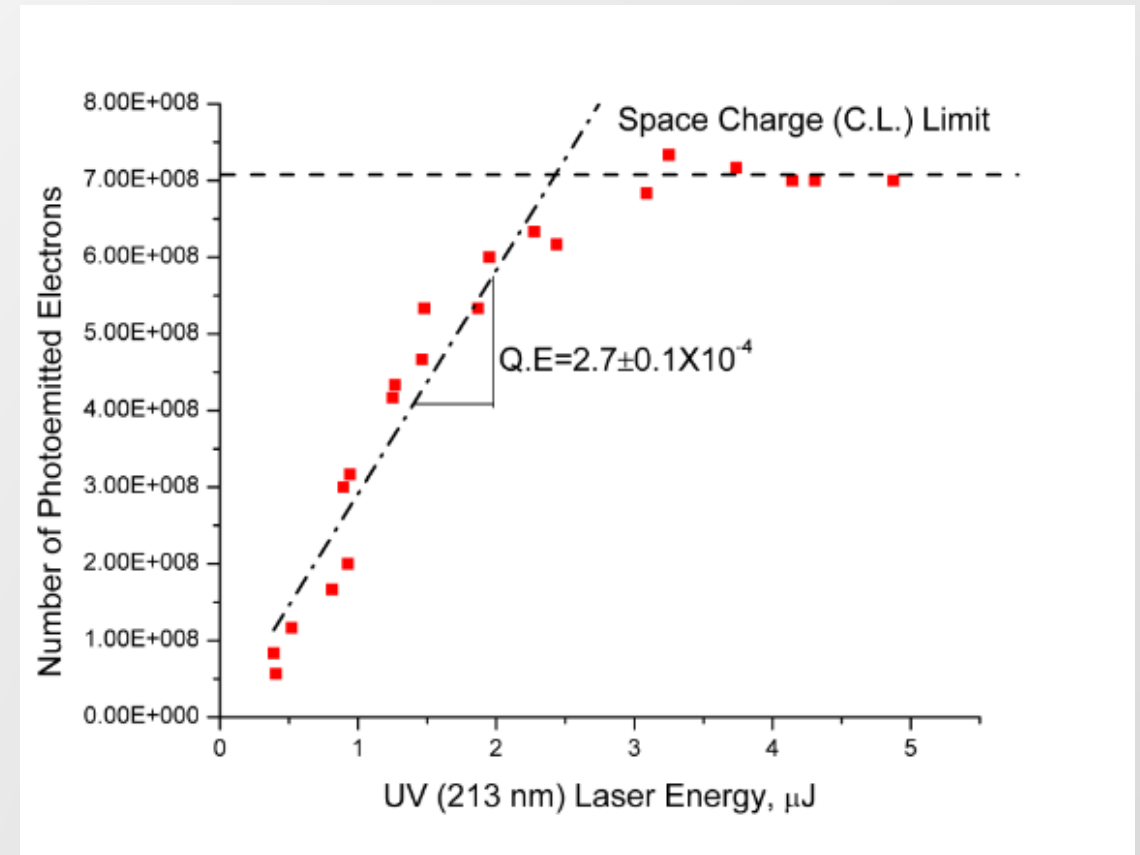
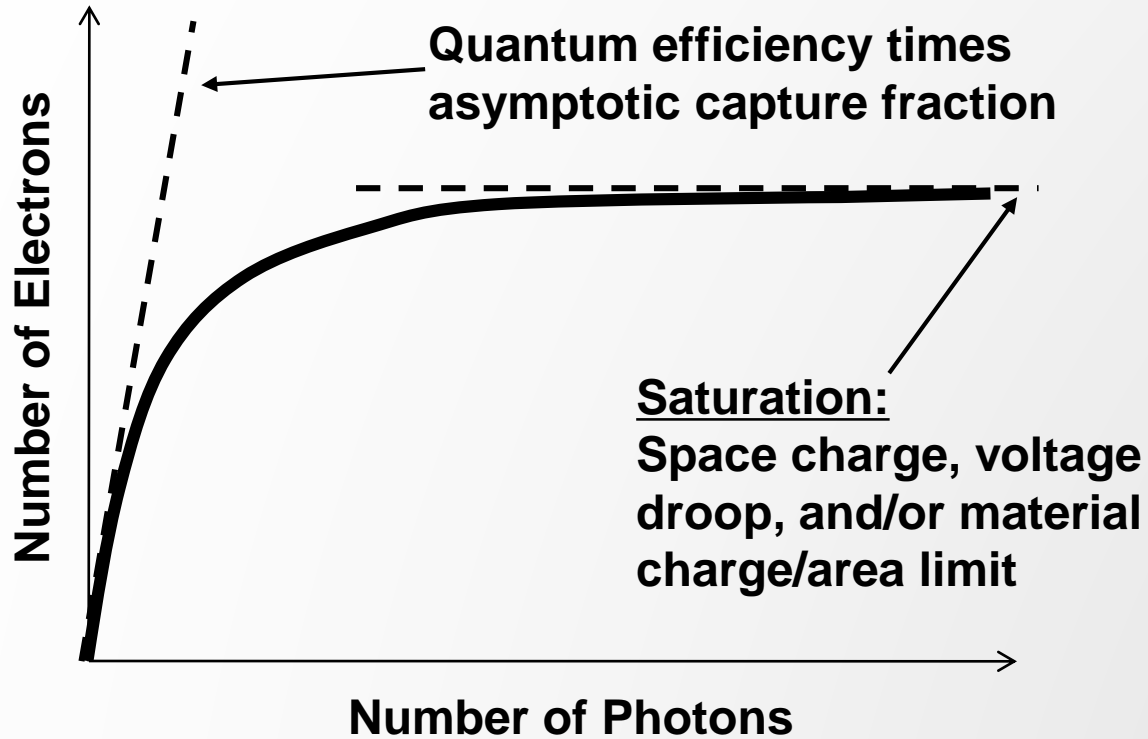
Longitudinal Space Charge



Child's Law:

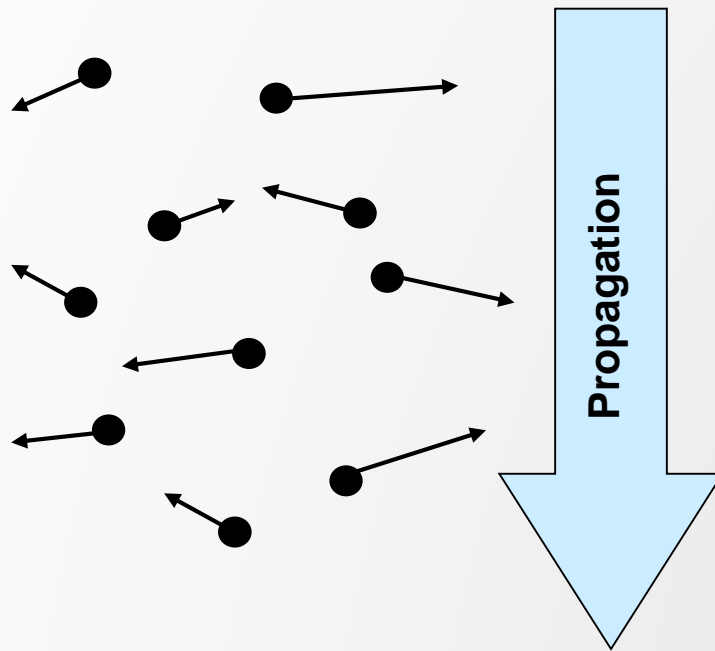
- Field from recent electrons retards emission
- Fundamentally limits current density (j):

$$j \propto V^{3/2} / d^2$$



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).

Longitudinal Inhomogeneous



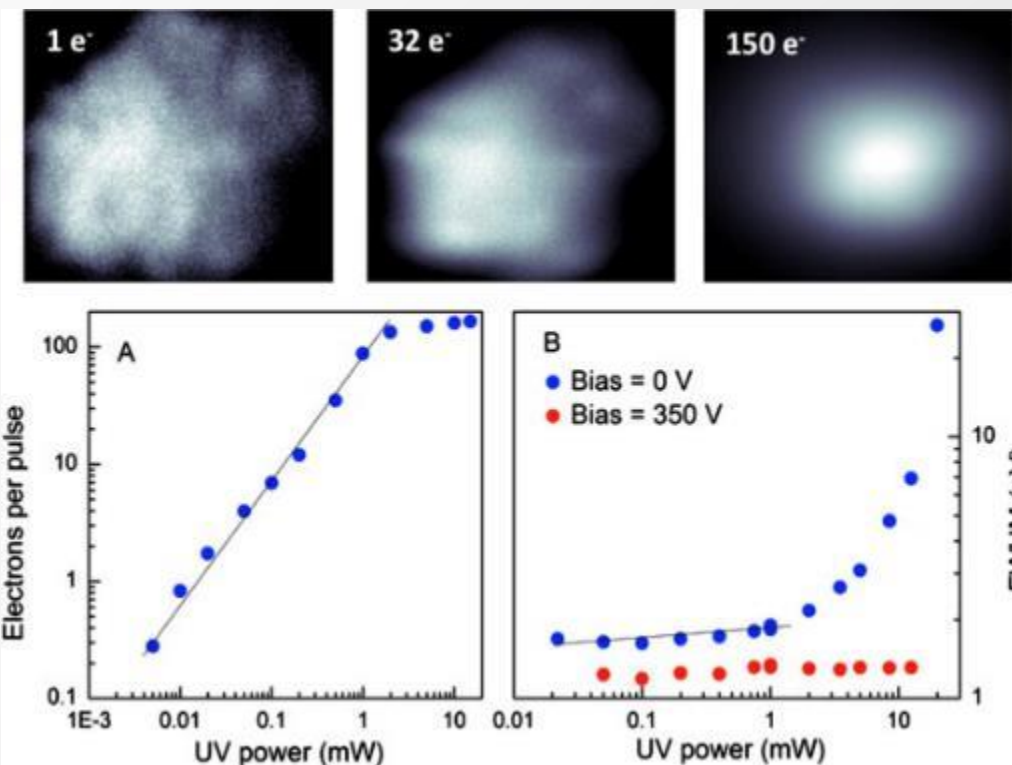
Boersch Effect:

- Pulse viewed in rest frame
- Most velocity is lateral
- Equilibration increases ΔE
- 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

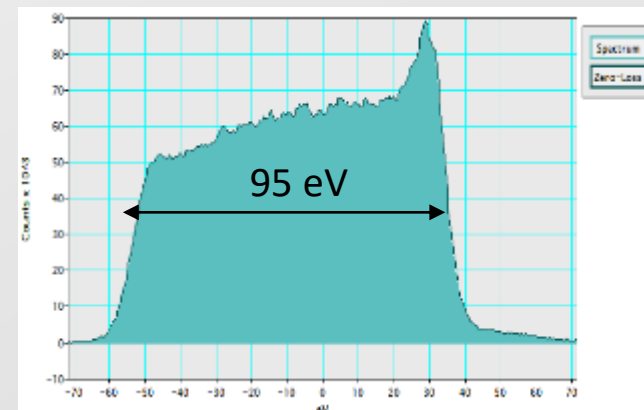
Operating with 10s of electrons per pulse causes

- **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

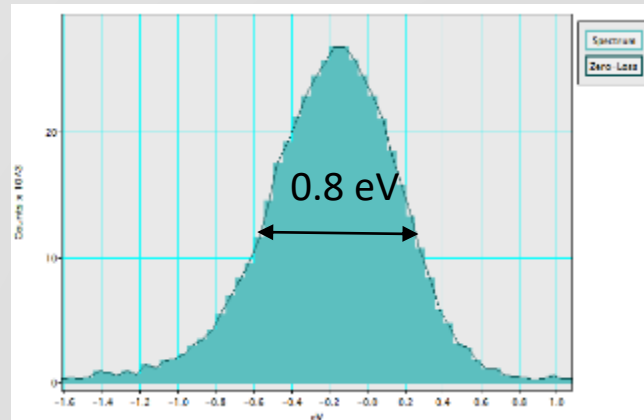


L. Piazza et al, *Chem Phys* **423**, 79 (2013).

Stroboscopic mode, 15 mW UV

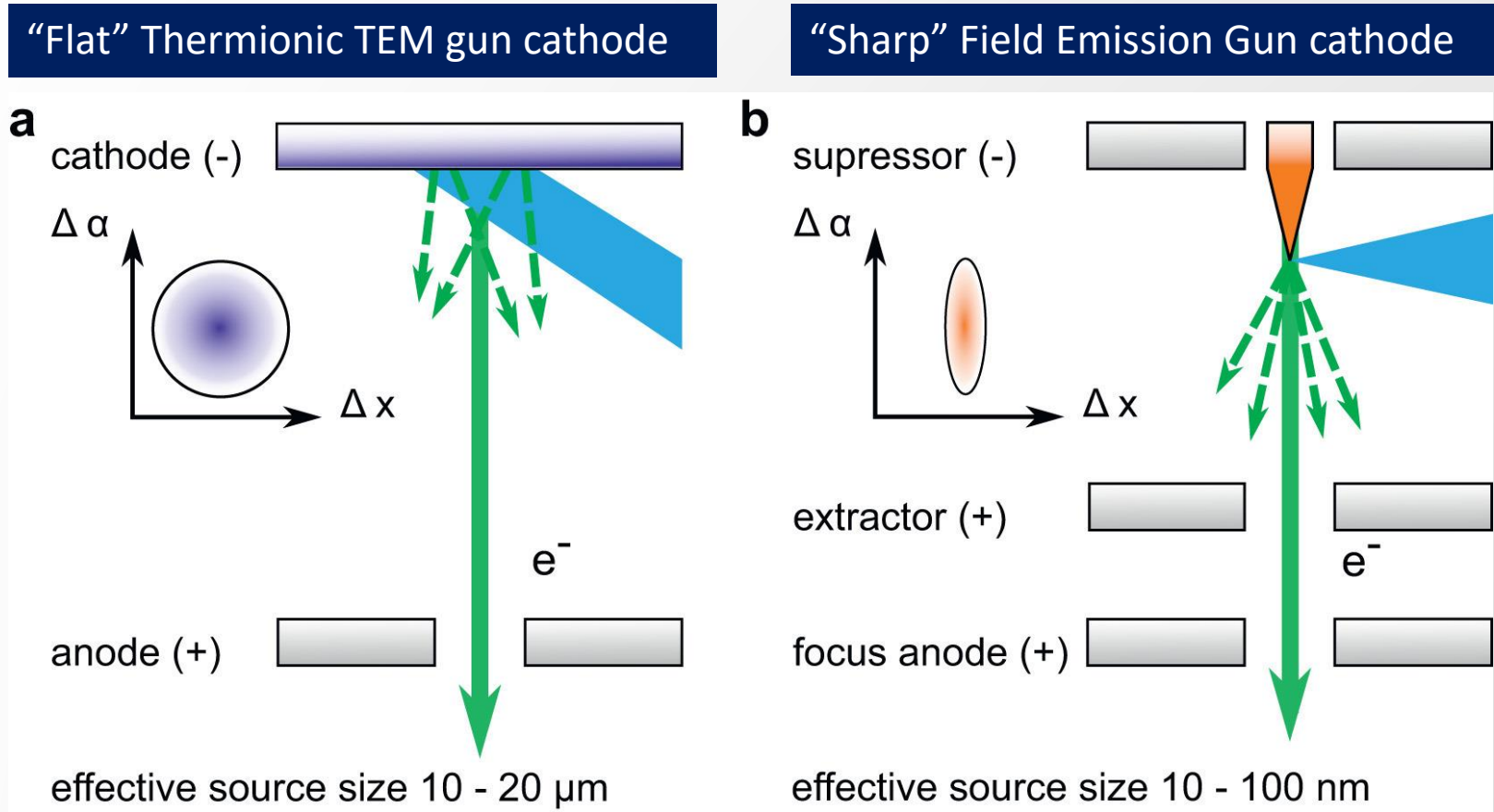


Stroboscopic mode, 1 mW UV



Unpublished data acquired on Strasbourg UTEM

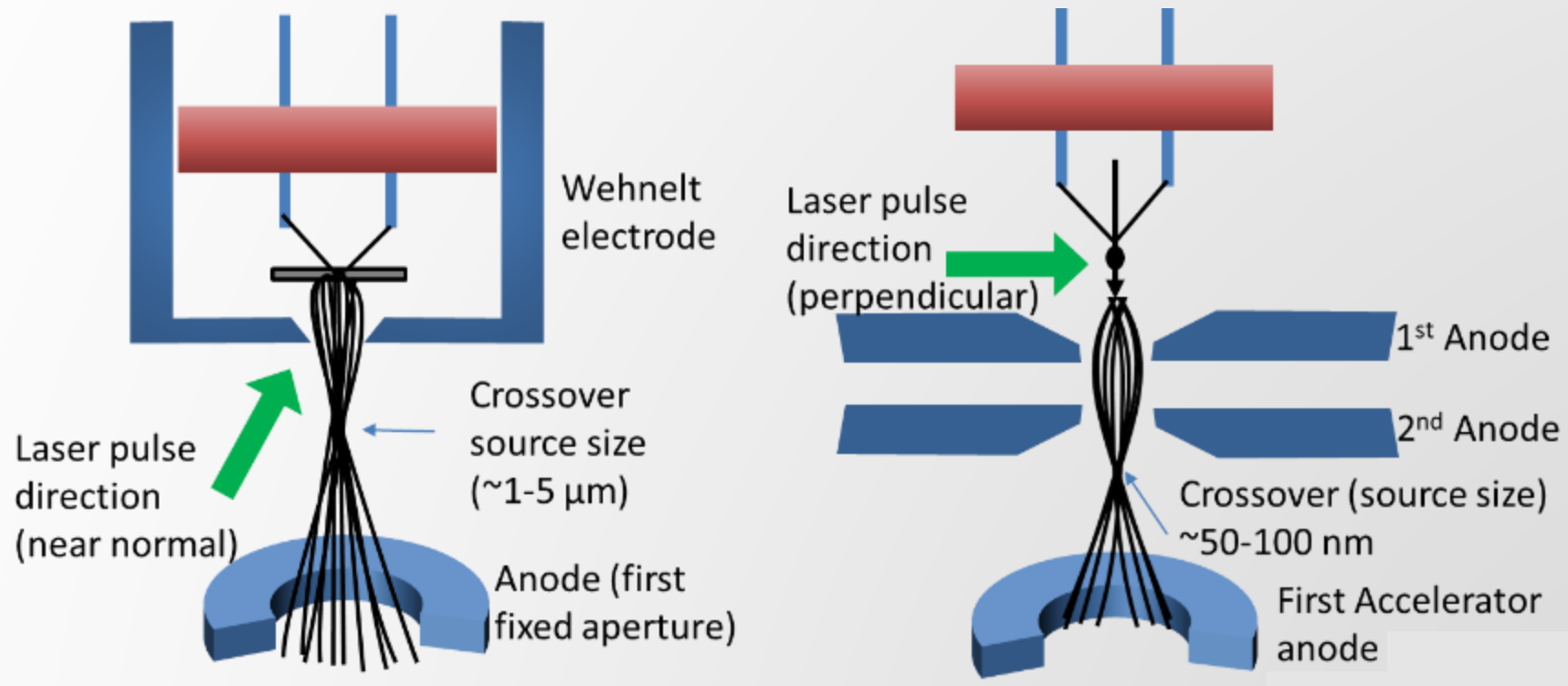
Does FEG tip geometry provide better coherence?



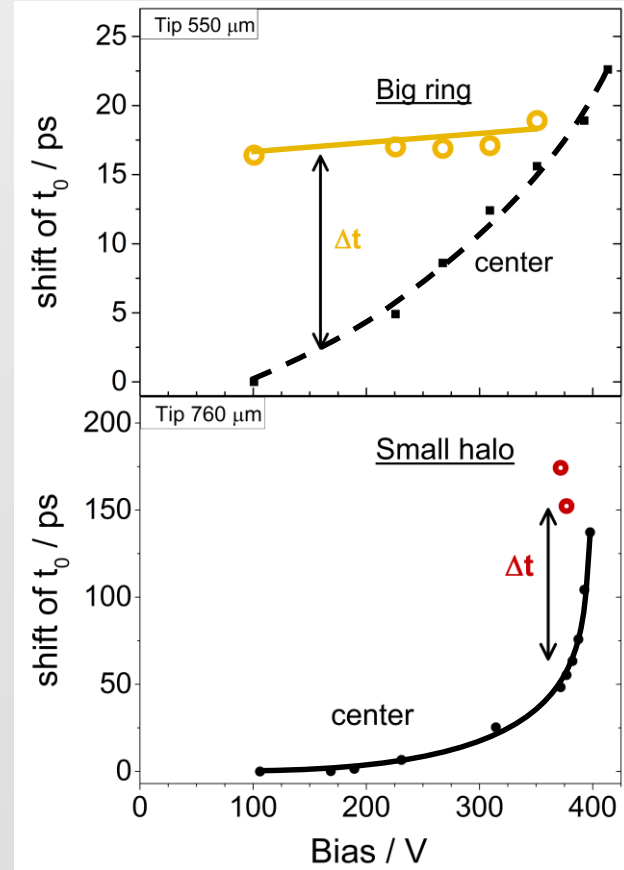
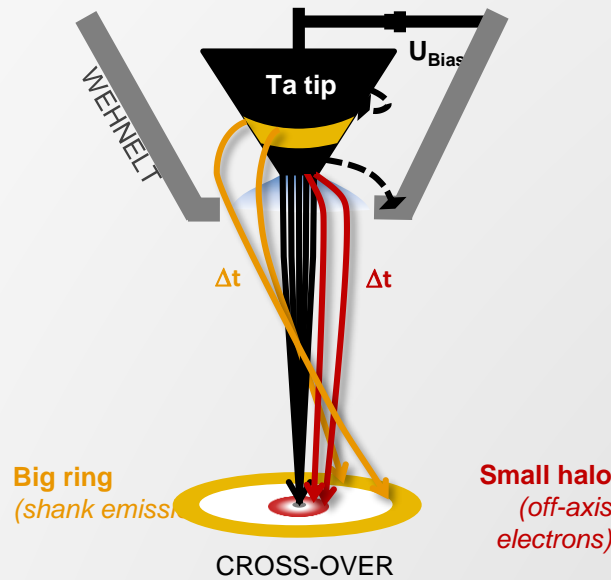
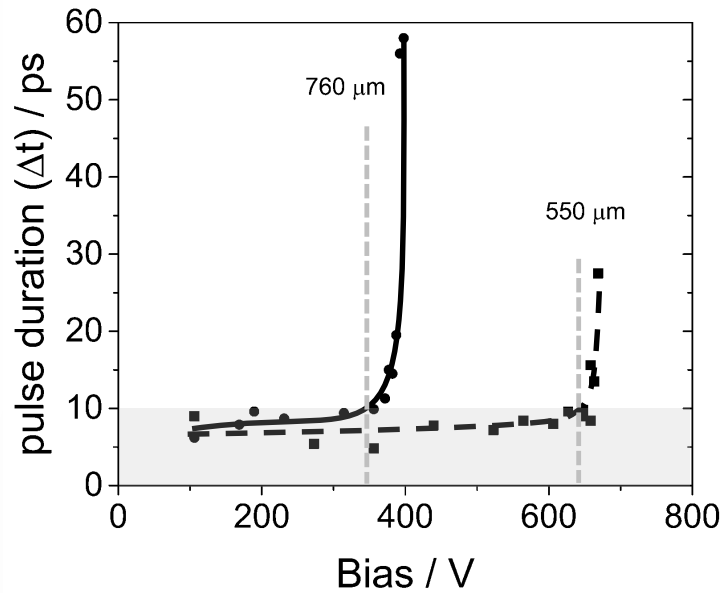
Armin Feist, Nora Bach, Nara Rubiano da Silva, Thomas Danz, Marcel Möller, Katharina E. Priebe, Till Domröse, J. Gregor Gatzmann, Stefan Rost, Jakob Schauss, Stefanie Strauch, Reiner Bormann, Murat Sivis, Sascha Schäfer, and Claus Ropers, *Ultramicroscopy* **176**, 63 (2017).

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



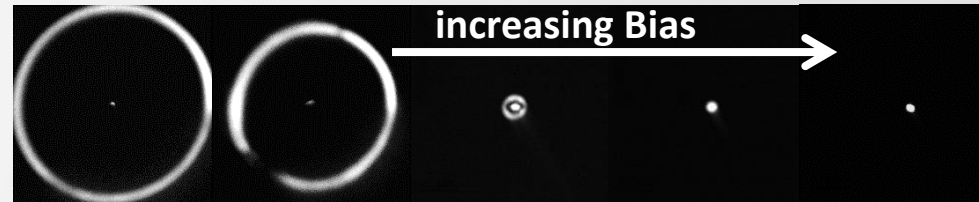
Space Charge Controls Brightness!
Both designs have similar photoelectron brightness



Space charge at the cathode

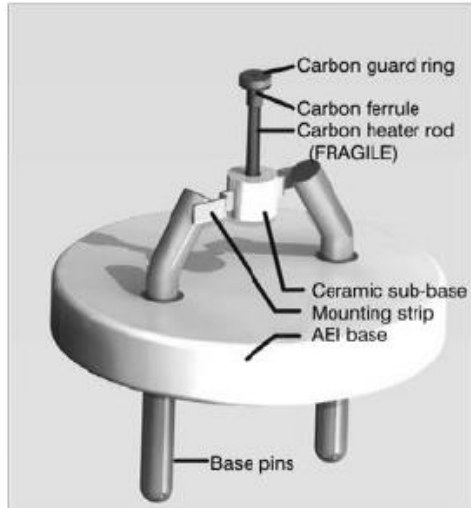
Boersch effect in the gun cross-over

- 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**



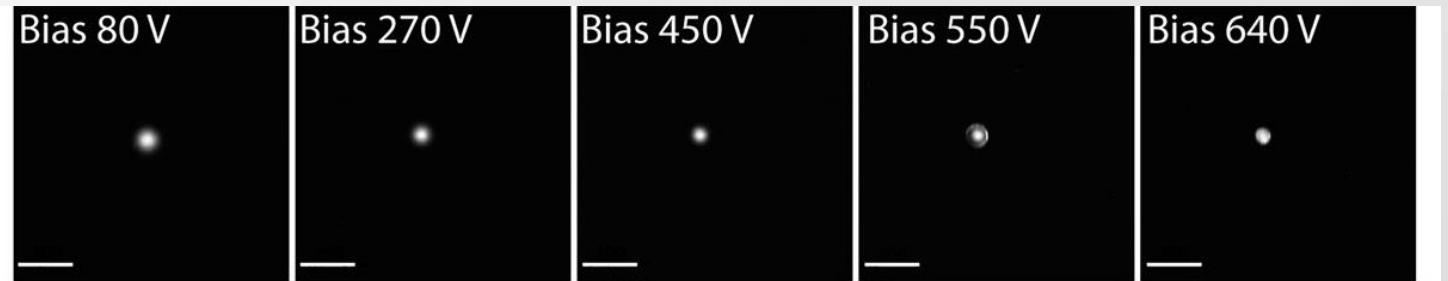
K. Bückler, M. Picher, O. Crégut, T. LaGrange, B.W. Reed, S.T. Park, D.J. Masiel, F. Banhart, "Electron beam dynamics in an ultrafast transmission electron microscope with Wehnelt electrode, *Ultramicroscopy*, Volume 171 (2016) pp.8-18

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

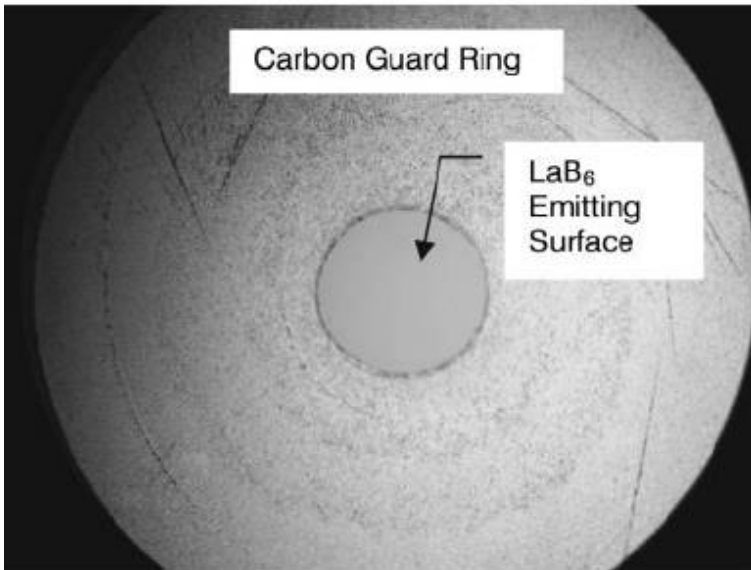


Improves coherence at high charge regime
when operating at moderate bias voltages

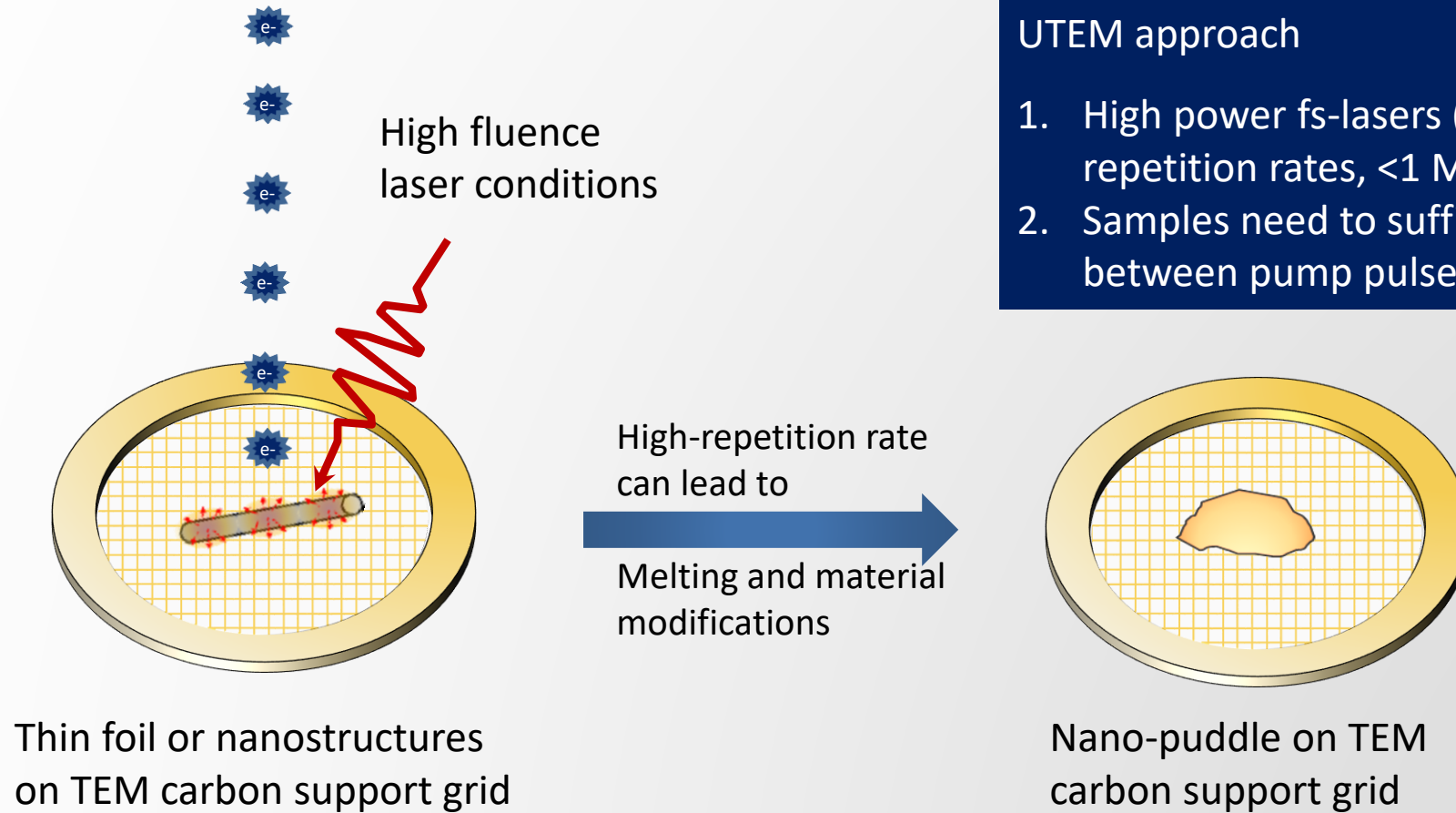
LaB₆ Guard Ring Cathode



Standard Truncated LaB₆ filament



J. Shaozheng et al., "Influence of cathode geometry on electron dynamics in an ultrafast electron microscope, Structural Dynamics 4, 054303 (2017)



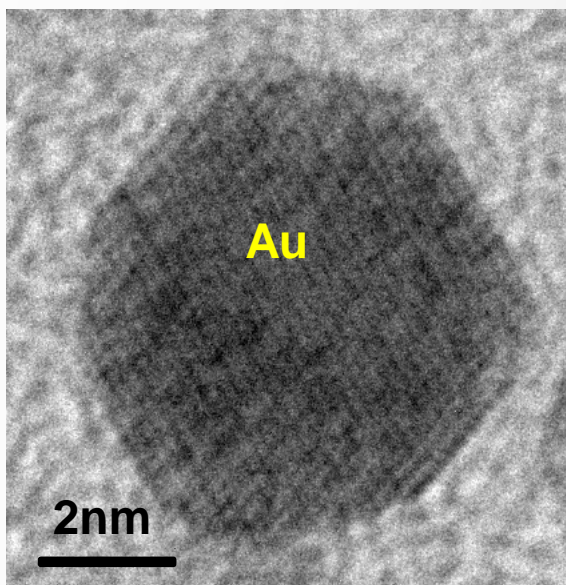
Practical Limitations of laser pumps in the UTEM approach

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

In practice, imaging requires high signal, i.e., >1 e⁻/pulse, and thus Boersch effects are the limiting factor in the spatial resolution

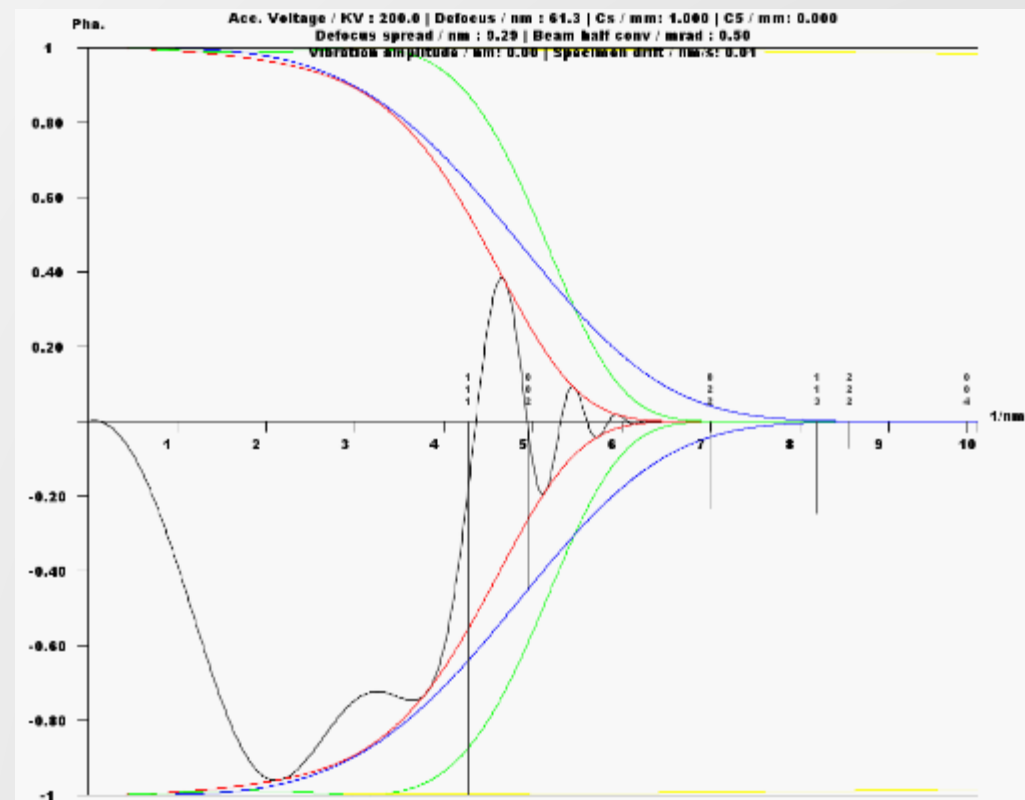
Resolution in photoelectron mode with Plate Cathode

- 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



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?

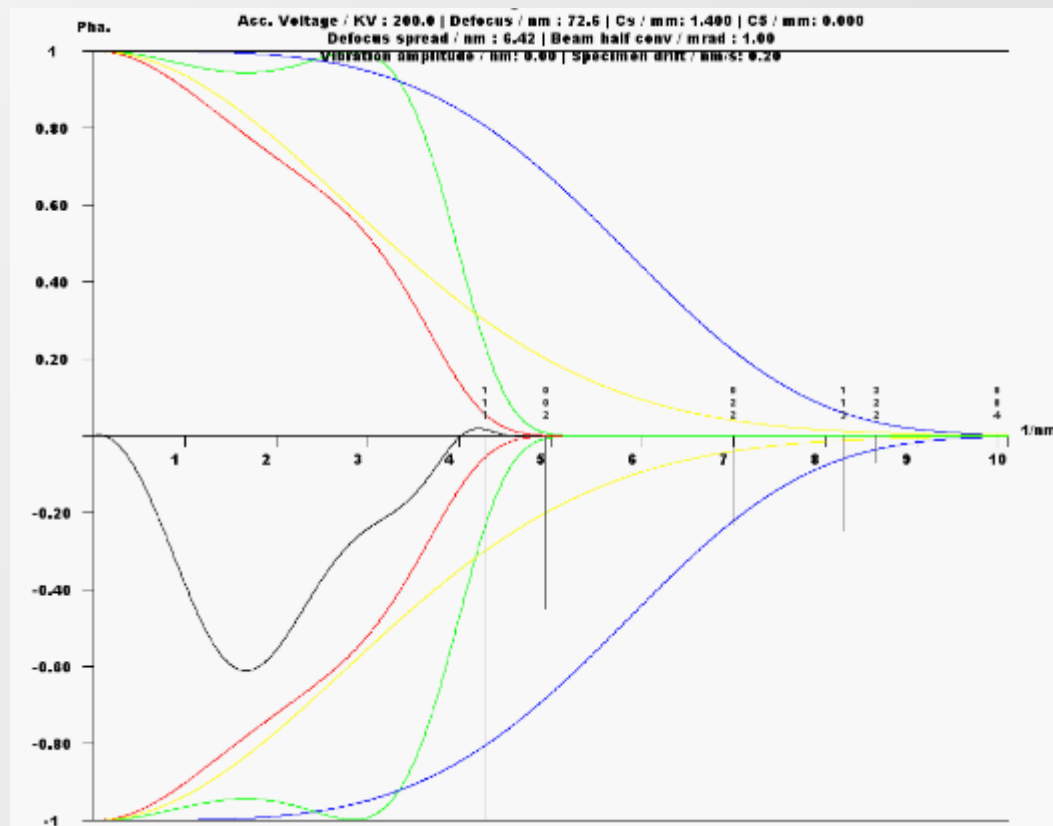
Low Average Brightness and microscope instabilities prevent high spatial resolution imaging

JEMS simulated CTF for long exposure times, 100s

JEOL 2100FHR pole piece: Cc 1.4mm and Cs 1.0mm

Energy spread: 1 eV, sample drift: **0.2 nm/min!!**

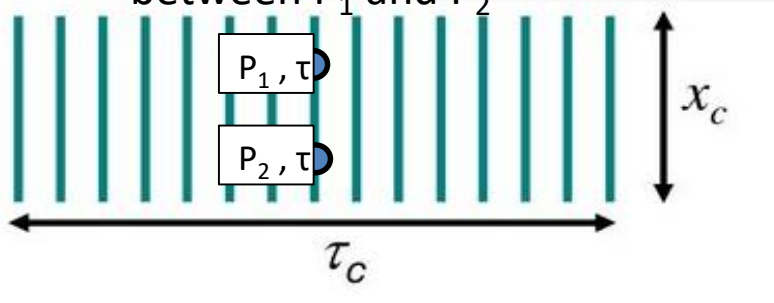
Stability Voltage: <1ppm/min & OL current: 1ppm/min, noise: <10 nT



Product of envelopes: **Spatial** **Temporal** **Noise**

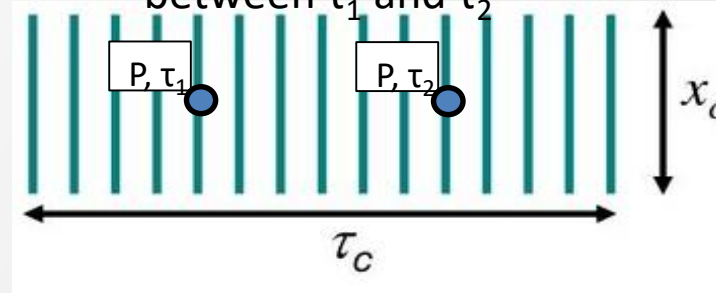
Spatial coherence

Constant phase difference
between P_1 and P_2



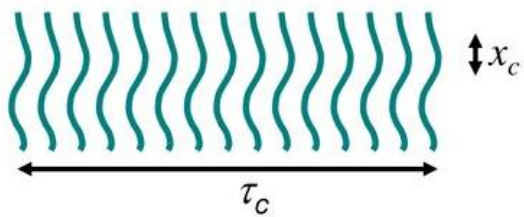
Temporal coherence

Constant phase at point P
between τ_1 and τ_2

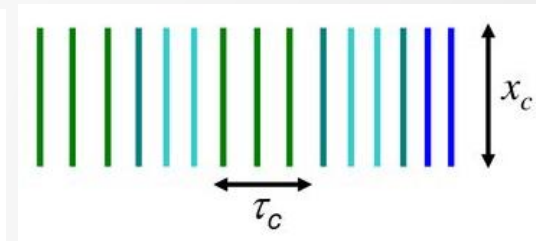


Laser sources

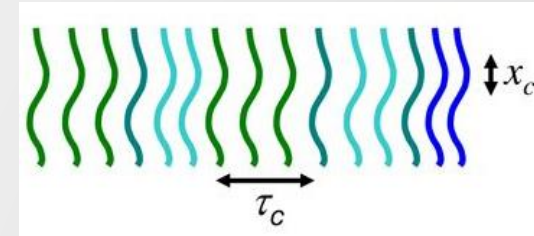
- Temporal coherence given by the inverse of the linewidth
- Excellent spatial coherence (depends on cavity and transport optics)



Temporal coherence
Spatial **In**coherence
 $x_c = \text{coherence length}$
 $\tau_c = \infty$



Temporal **In**coherence
Spatial coherence
 $\tau_c = \text{coherence time}$
 $x_c = \infty$

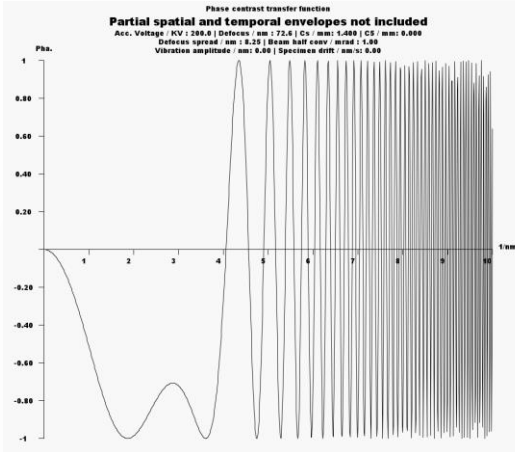


Spatial **and** Temporal
Incoherence
 x_c, τ_c

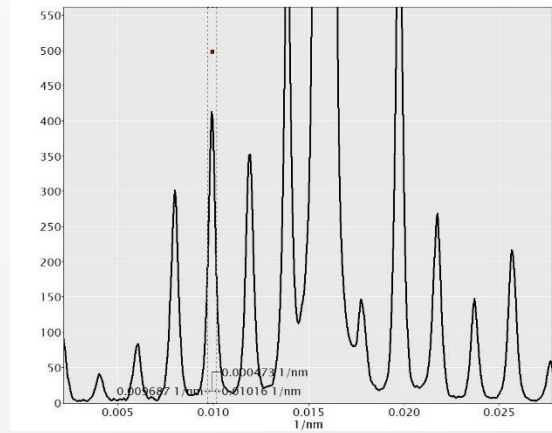
Pulsed electron sources

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

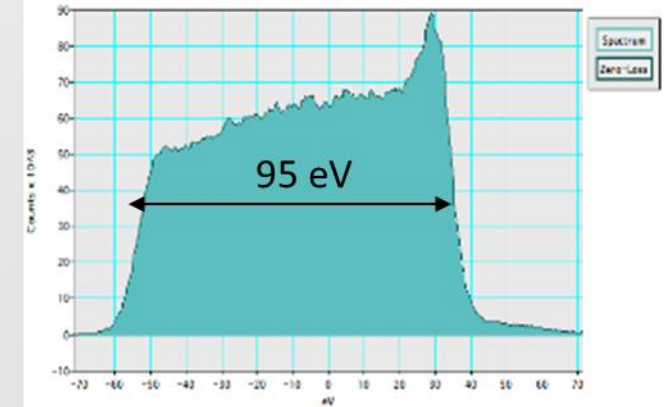
HRTEM phase contrast (spatial resolution)



Diffraction resolution Effect of transverse coherence



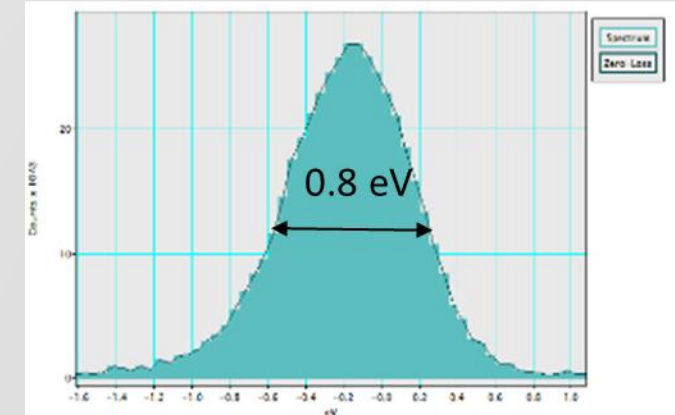
Effect of space-charge on energy resolution in fs-EELS



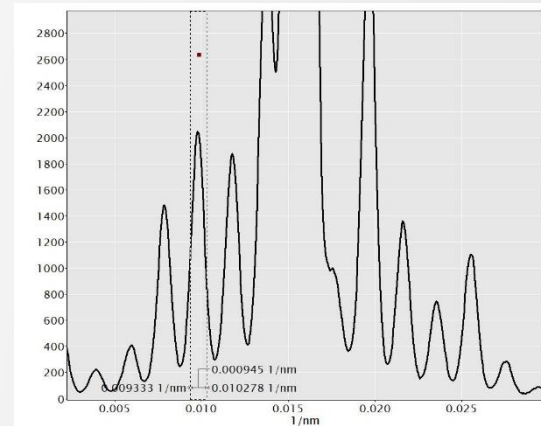
Source brightness is a fixed quantity

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

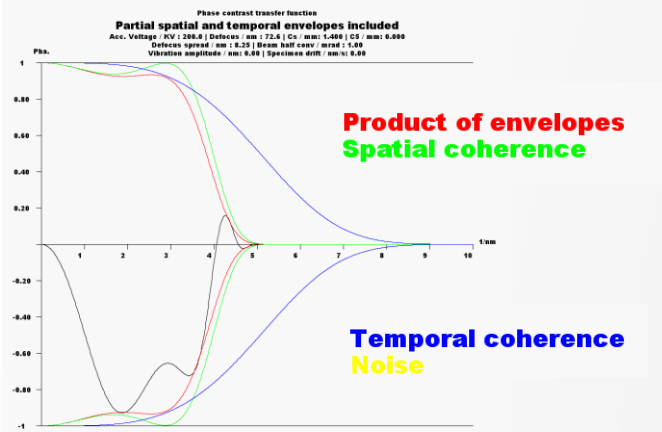
Effect of Boersch effects in the gun



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

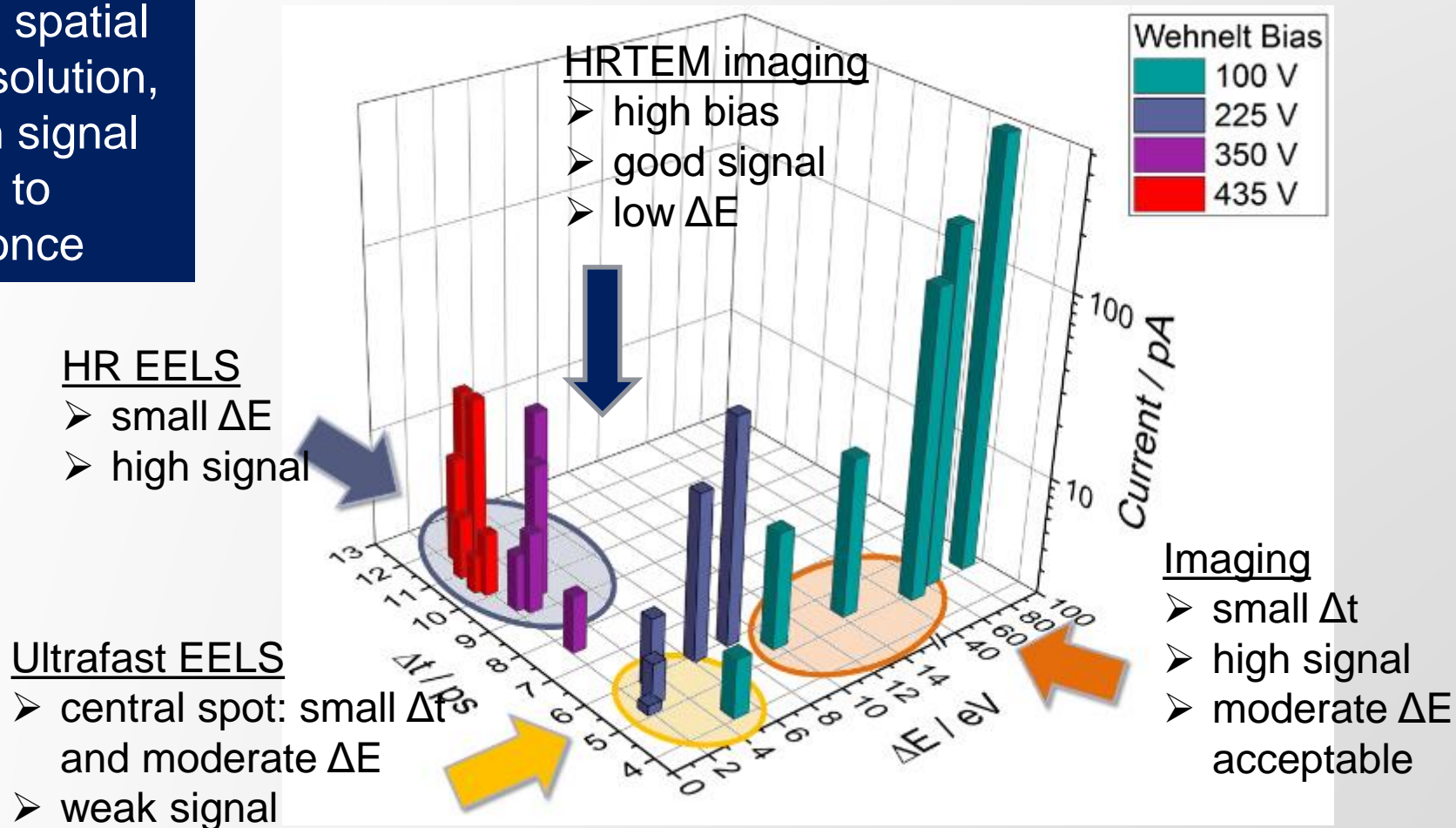


Effect of lens aberrations and source



There is a finite Electron Brightness, and trade-offs are made between signal, time resolution, temporal and spatial coherence

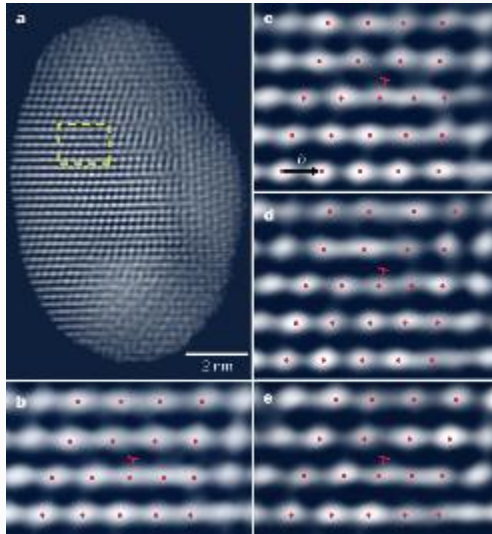
High temporal, spatial and energy resolution, as well as high signal are impossible to achieve all at once



K. Bückler et al, *Ultramicroscopy*, Volume 171 (2016) pp.8-18

Applications of Ultrafast Transmission Electron microscopy in Condensed Matter Physics

Observing structure (lattice)



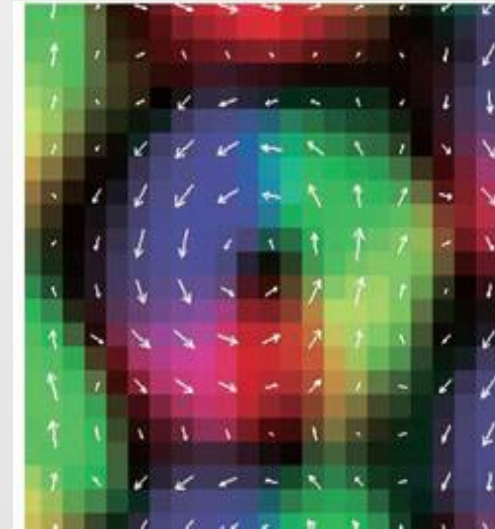
Nature 496 74 (2013)

Observing charges/orbitals



Tokunga et al., Nat. Mater. 5 (2006)

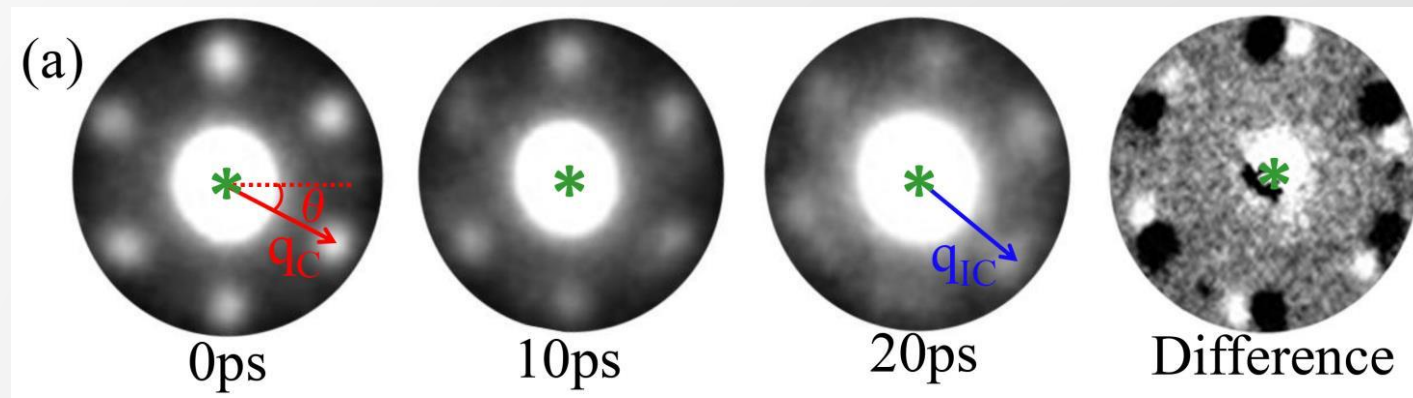
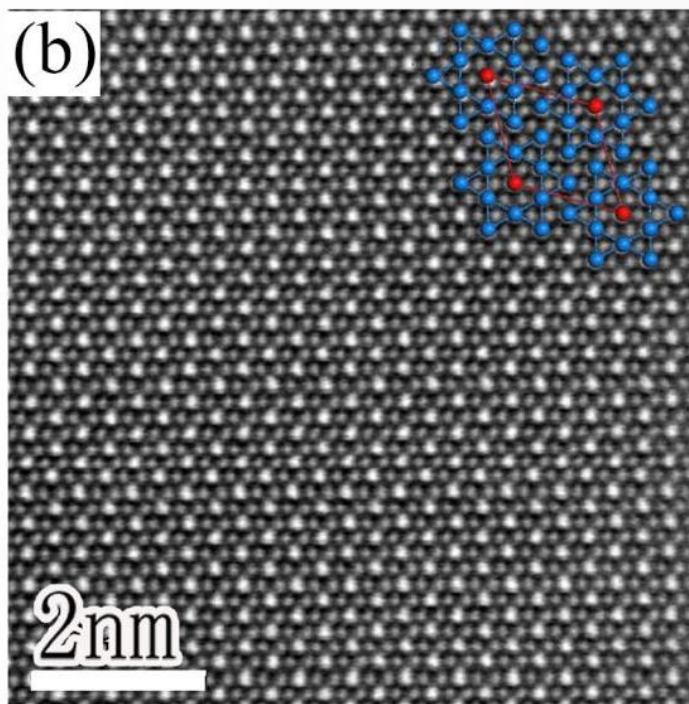
Observing spins



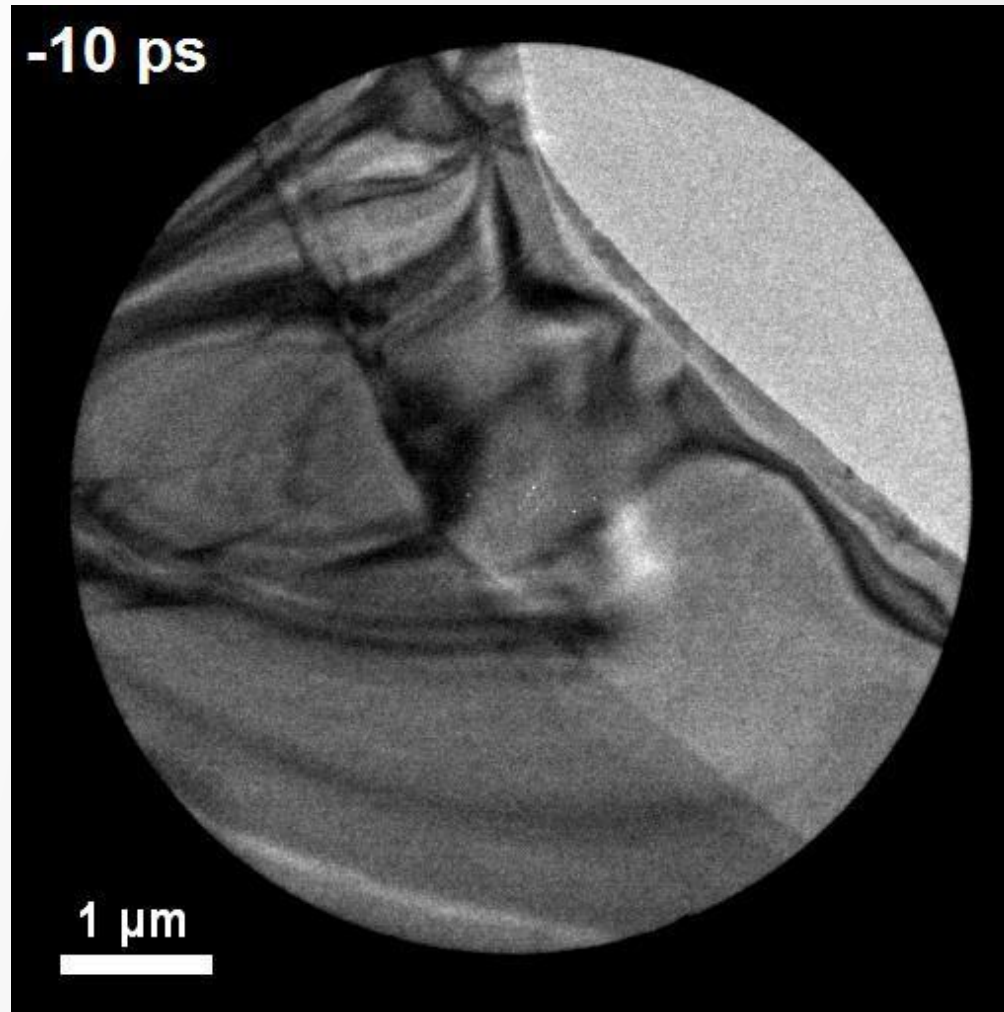
Seki et al, Science 336 (2012)

Textured ground states rule the physics of novel materials

W. Witczak--Krempa, et al., "Correlated Quantum Phenomena in the Strong Spin--Orbit Regime." *Annual Review of Condensed Matter Physics*, 5 57 (2014).

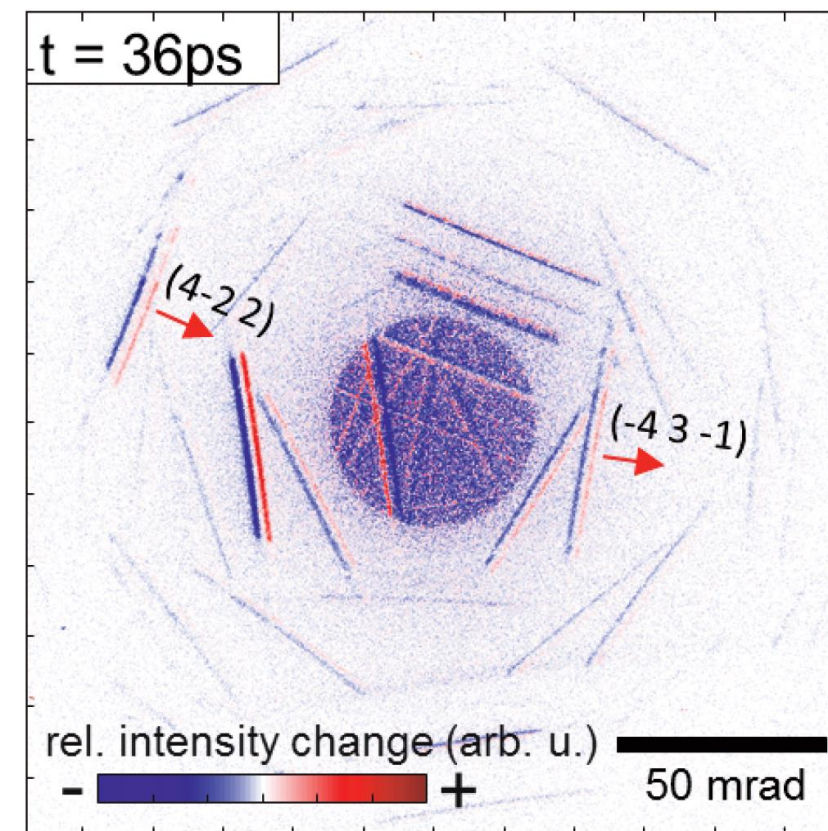
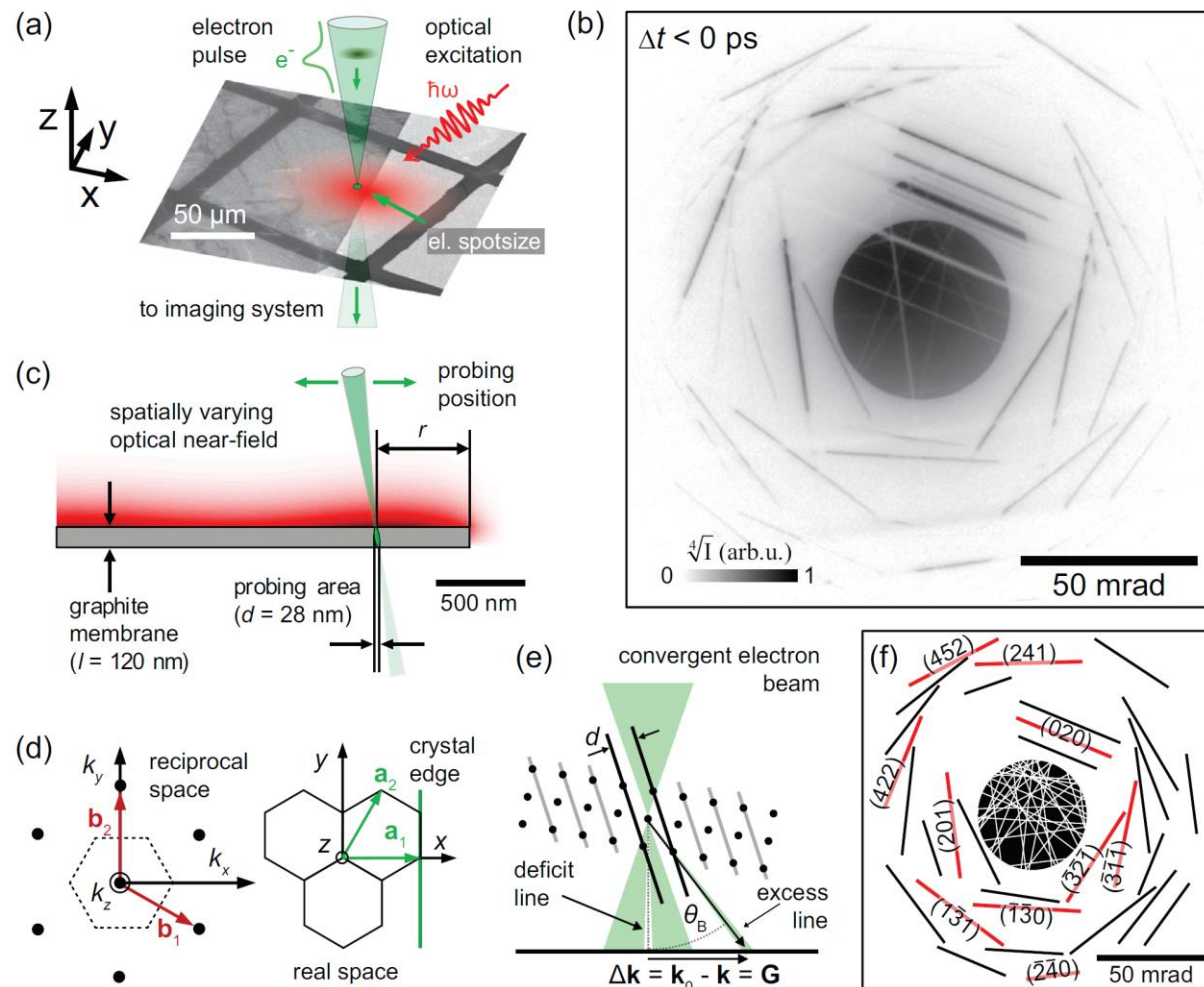


Sun et al., Phys. Rev. B 92 224303 (2015)

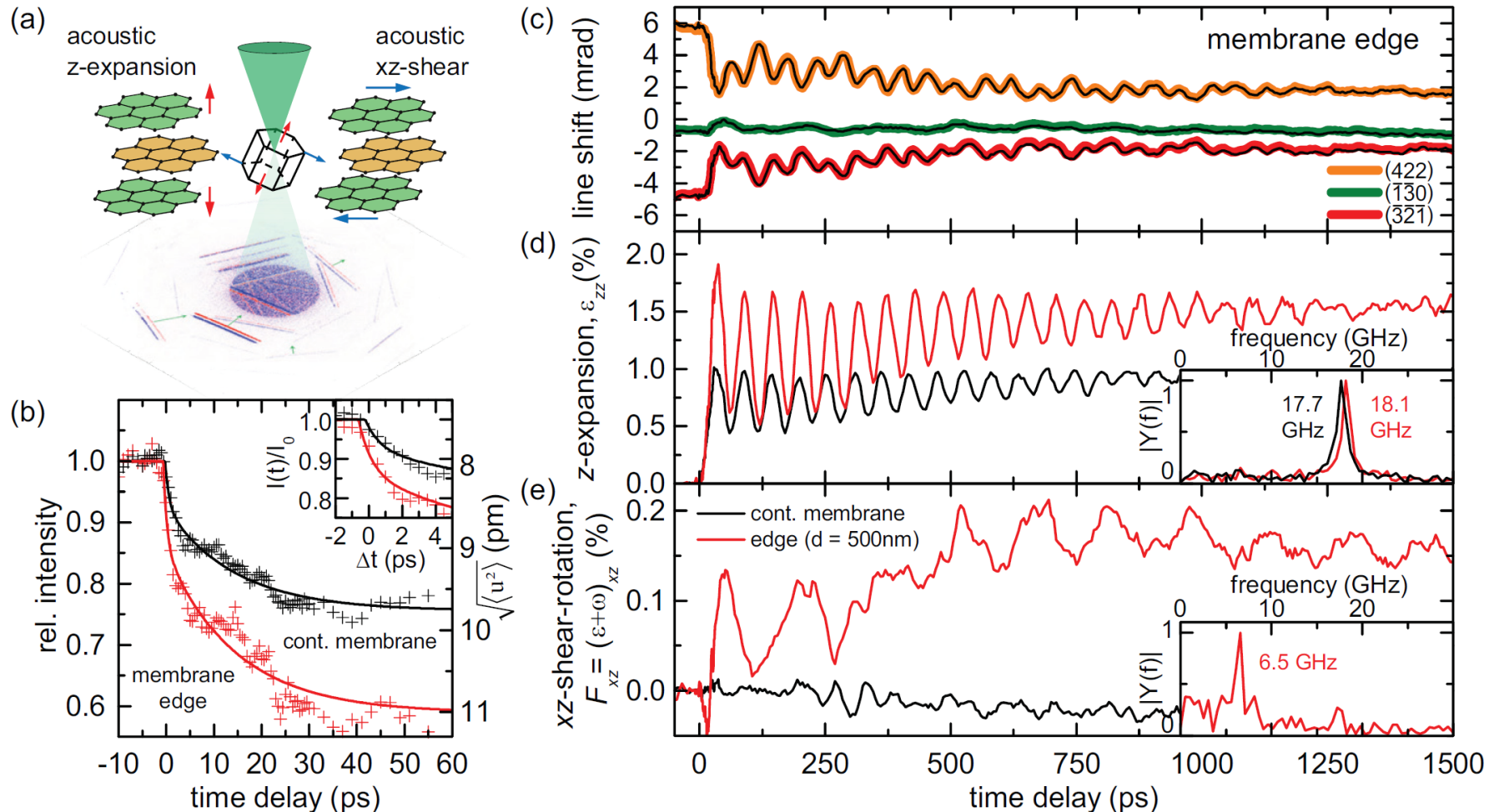


Cremons et al, Nat. Comm. 7, 11230 (2016)

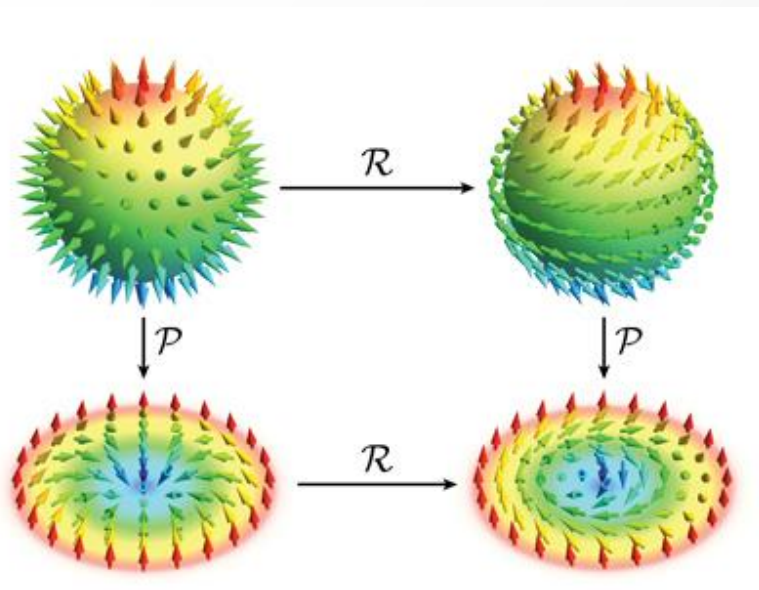
Using Convergent Beam Electron Diffraction (CBED) for probing phonon modes



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



Néel skyrmion

Bloch skyrmion

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

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

Néel type
Ferromagnet/heavy metal bilayers

Racetrack memory:

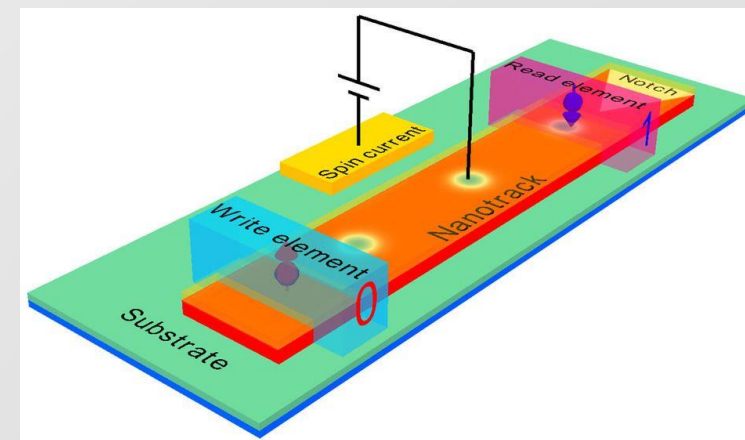


Image from Zhang et al. Sci Rep. 5 7643 (2015)

The defined chirality of the spin configurations provides topological barrier (protection) that stabilizes the skyrmions



Gabriele Berruto



Ivan Madan

Help with the experiments:

EPFL, ER-C Julich: Y. Murooka

EPFL: G. M. Vanacore

Sample preparation:

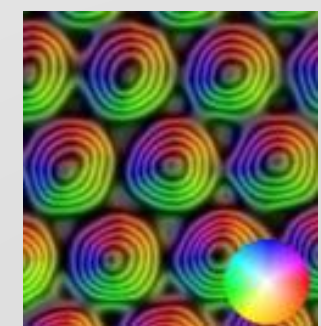
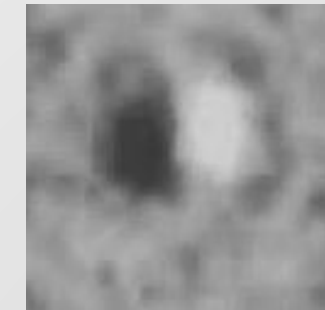
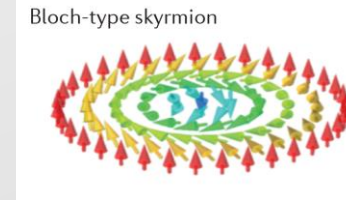
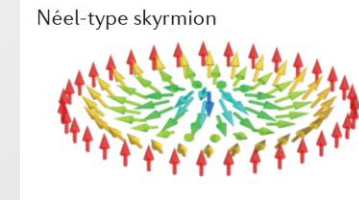
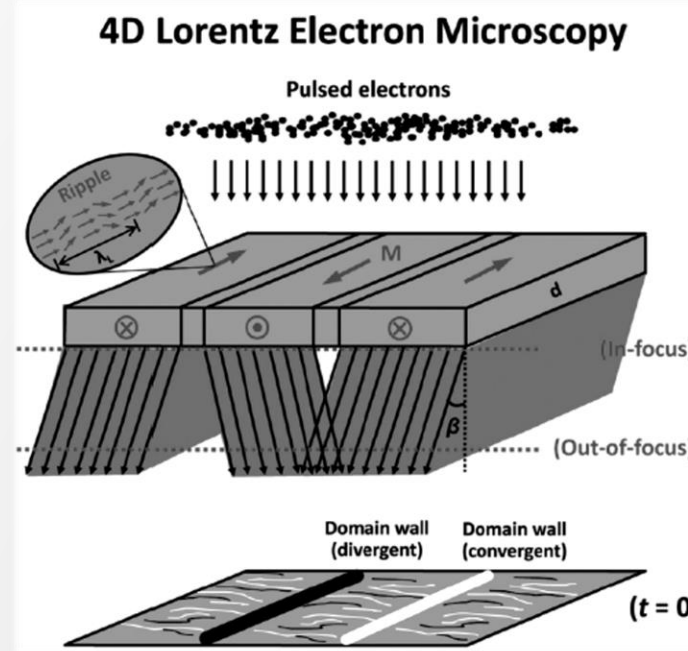
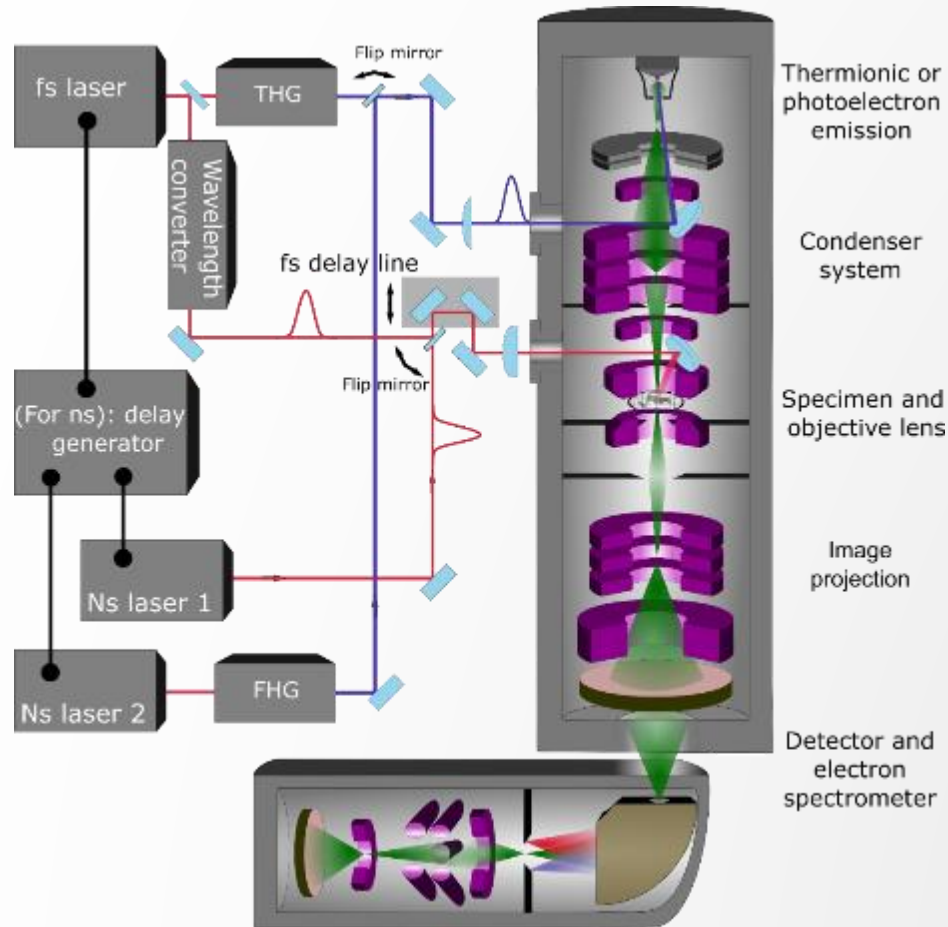
Uni Glasgow: R. Lamb, D. McGrouther

Uni Osaka: Y. Togawa

Funding:

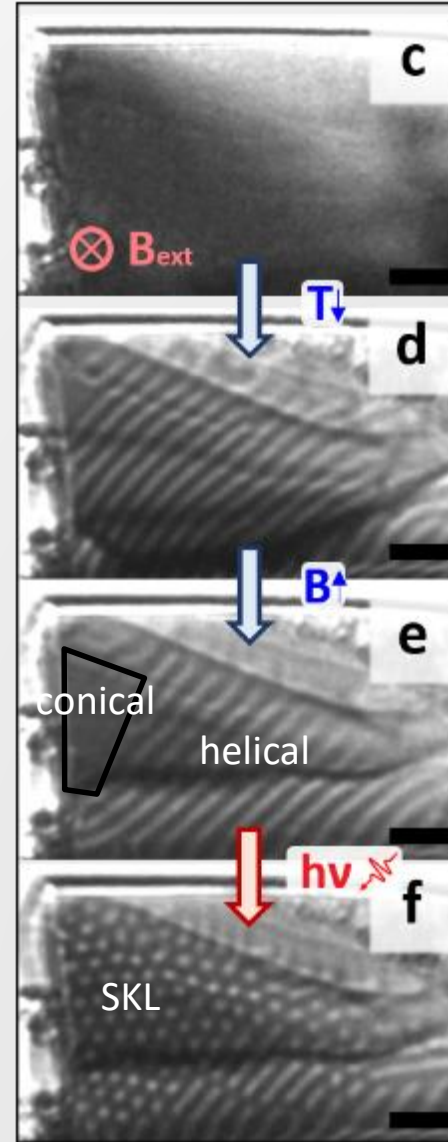
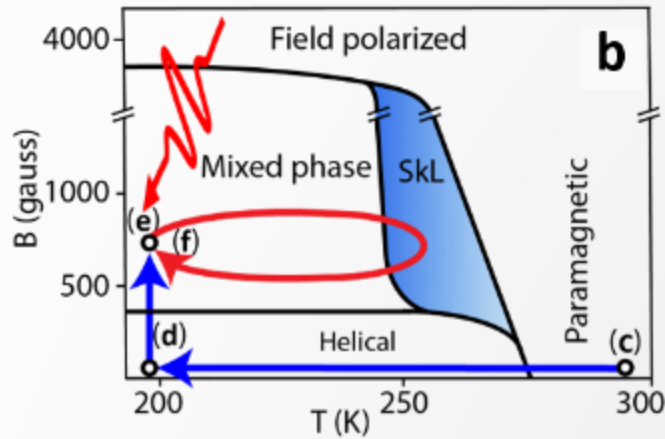


We use time-resolved *cryo*-Lorentz-transmission electron microscopy to characterize Skyrmion dynamics



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



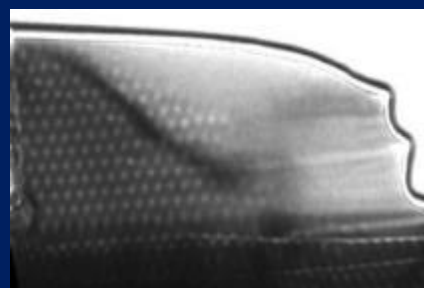
T = RT
 $\vec{B} = 0$ G

T = 200 K
 $\vec{B} = 0$ T

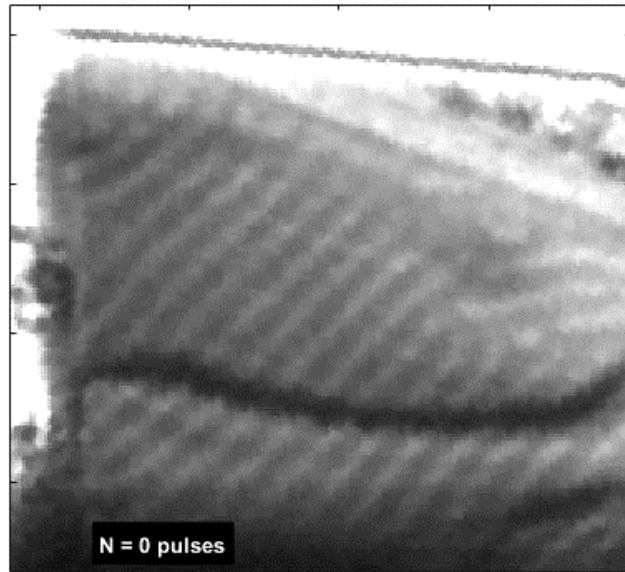
T = 200 K
 $\vec{B} \sim 750$ G

Write Skyrmions
 using laser pulses
 $\vec{B} \sim 750$ G

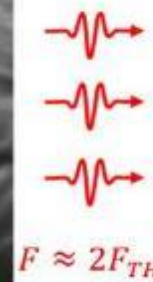
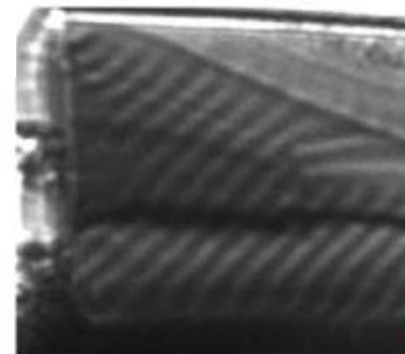
- Large and regular SkL upon adiabatic field cooling
- Skyrmions can be created by a single laser pulse
- Skyrmions erasing by magnetic field excitation, or by laser pulses with much higher fluence



It is possible to “photocreate” skyrmions by irradiating the sample (in the helical phase) with multiple fs pulses



“Mixed” phase regime



Photocreated SkL

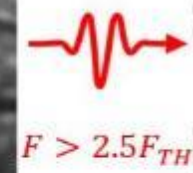
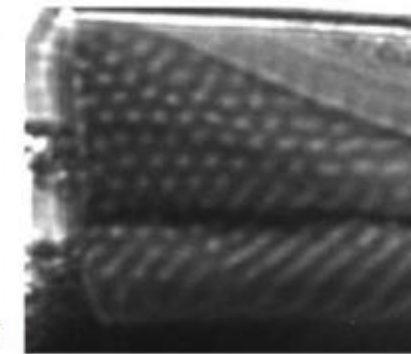
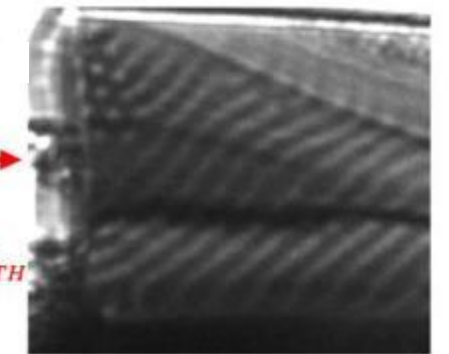


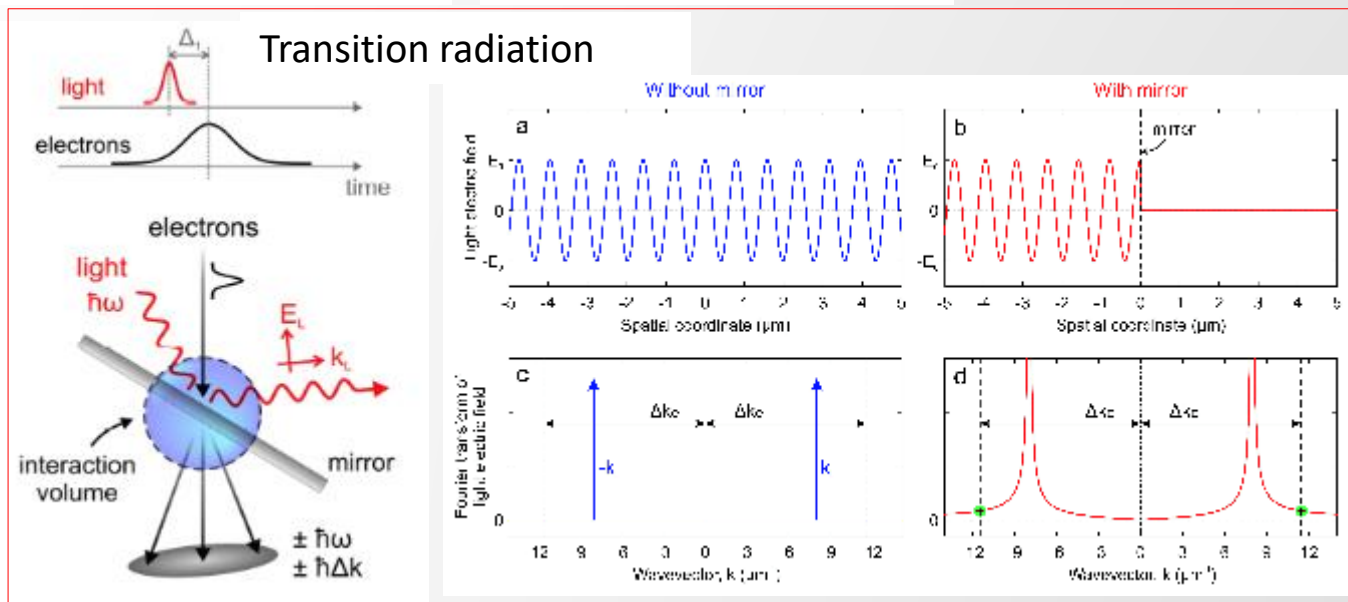
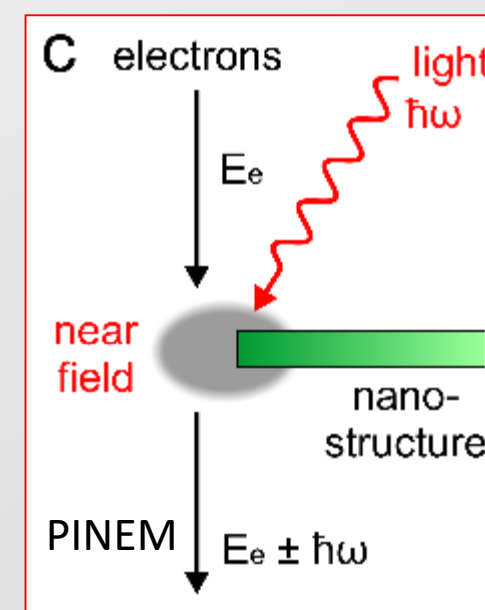
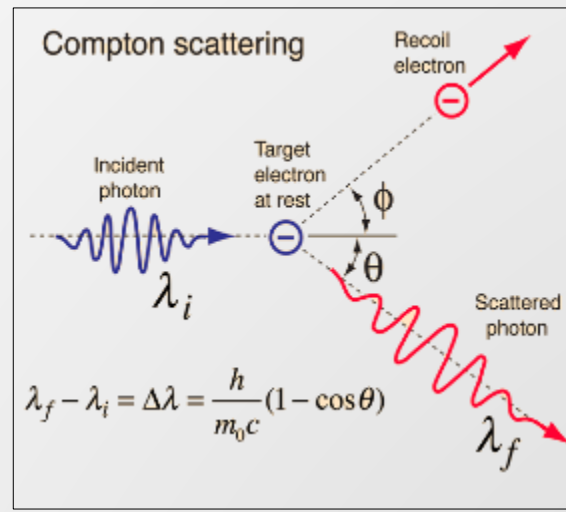
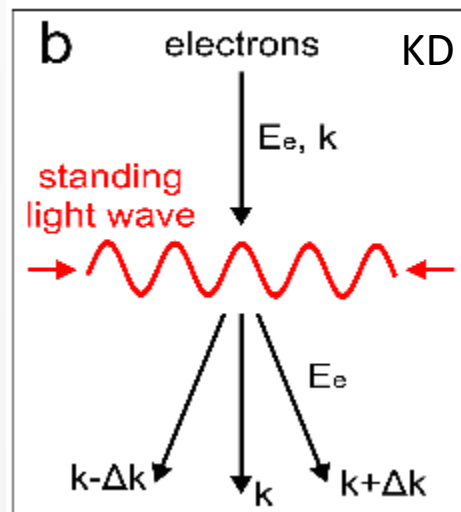
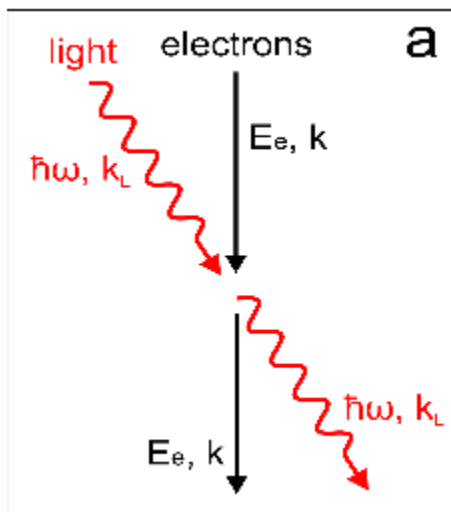
Photo-Erased SkL



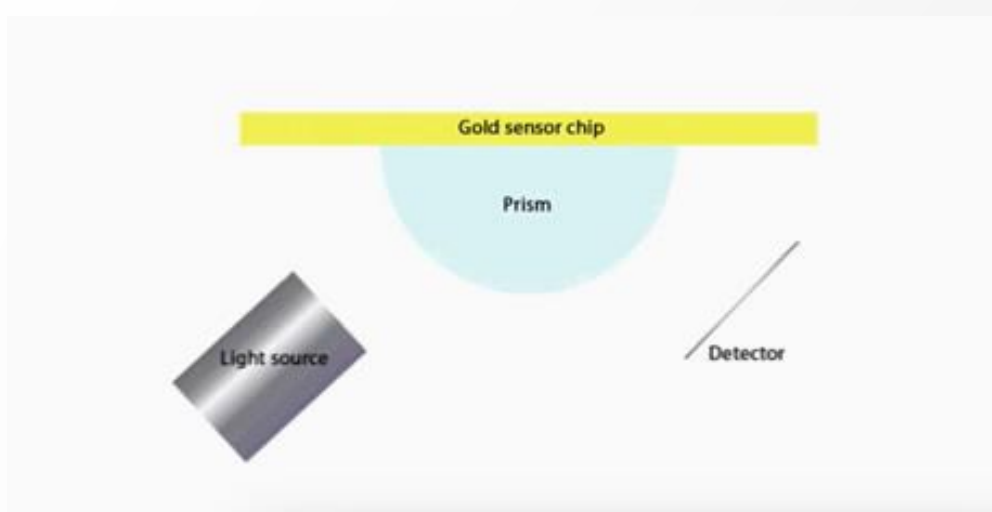
Skyrmions are created at the edges, gradually filling of the sample with skyrmions with multiple laser pulses

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

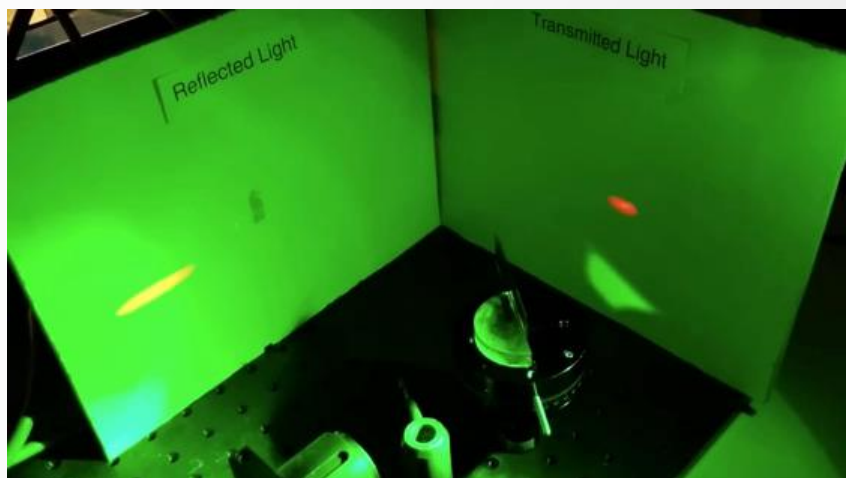
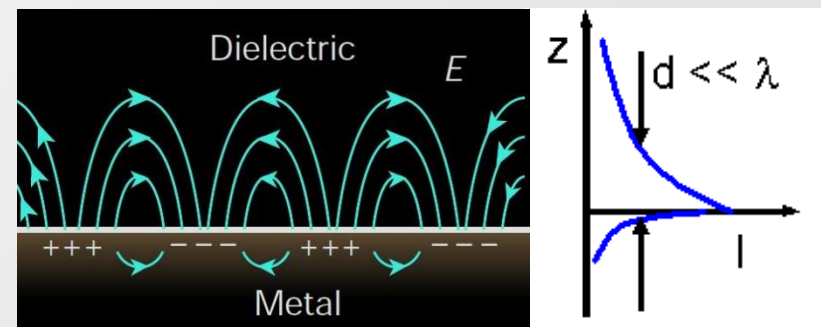
Example: Coherent control of Light-Electron interactions



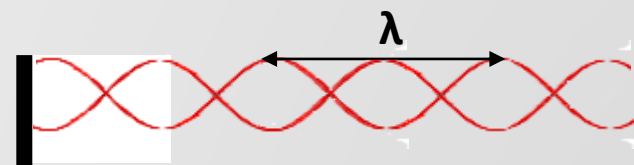
How can light and e^- interact?



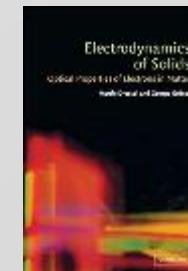
Surface plasmon:
Electrons-light coupled at an interface



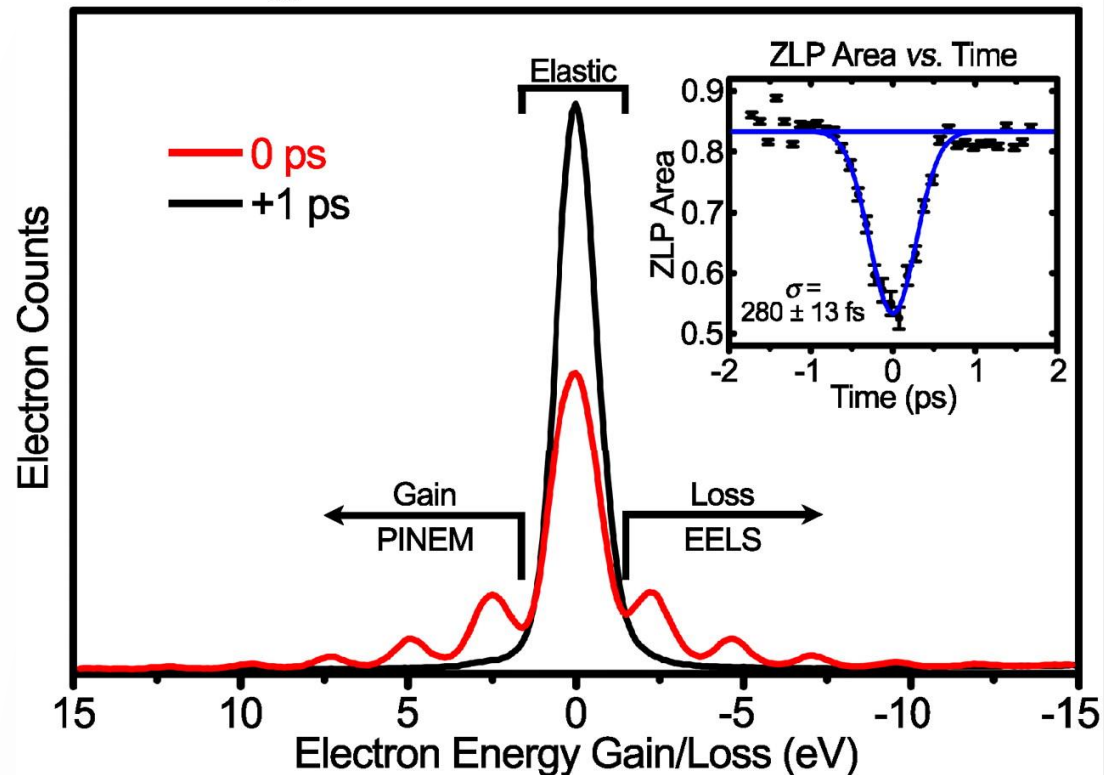
Plasma resonance



Suggested reading: Dressel, "Electrodynamics of solids"

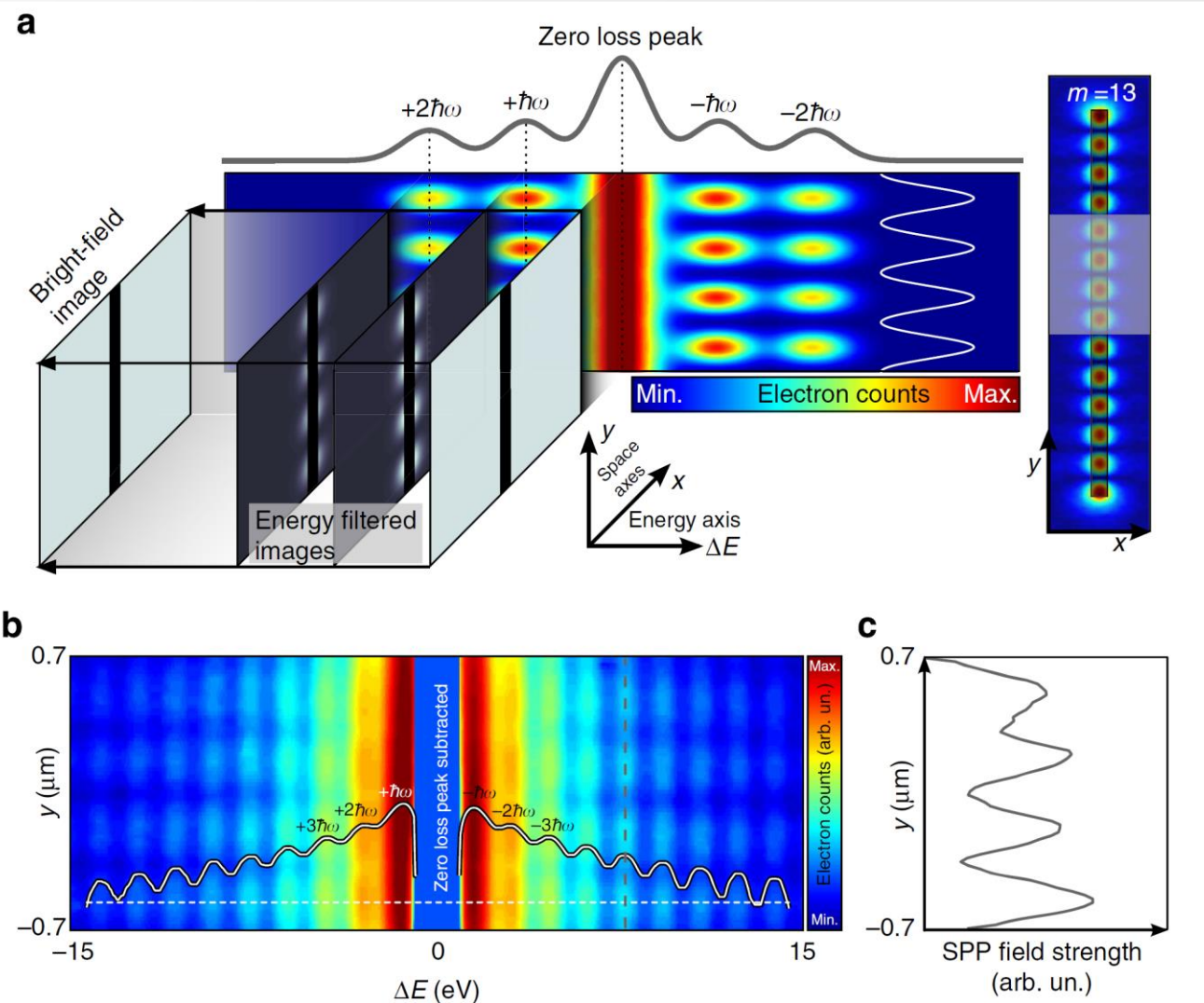


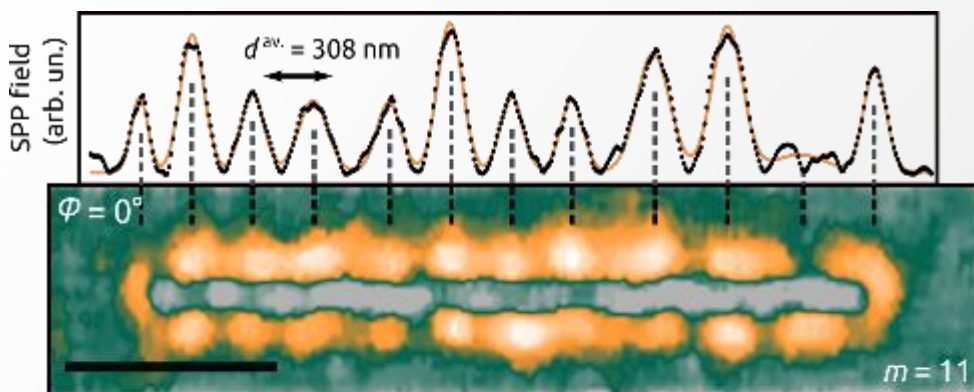
UEM: Energy-filtered fs electrons



David J. Flannigan, Brett Barwick, and Ahmed H. Zewail, Proceedings of the National Academy of Sciences **107** (22), 9933 (2010).

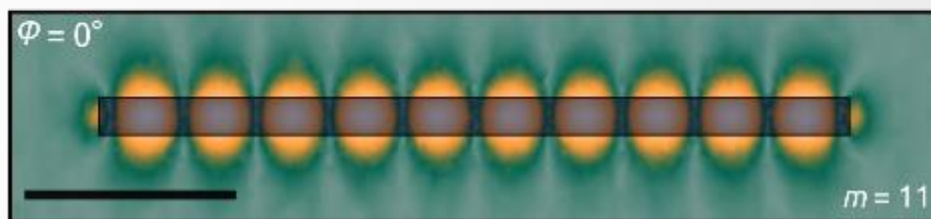
L. Piazza, T. T. A. Lummen, E. Quiñonez, Y. Murooka, B. W. Reed, B. Barwick, and F. Carbone, Nature Communications **6** (1), 6407 (2015).



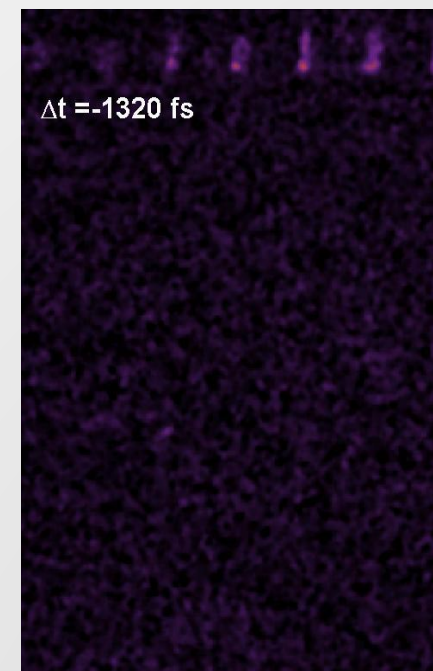


Experiment

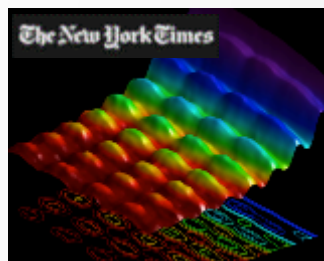
Polarization \longleftrightarrow



Simulation

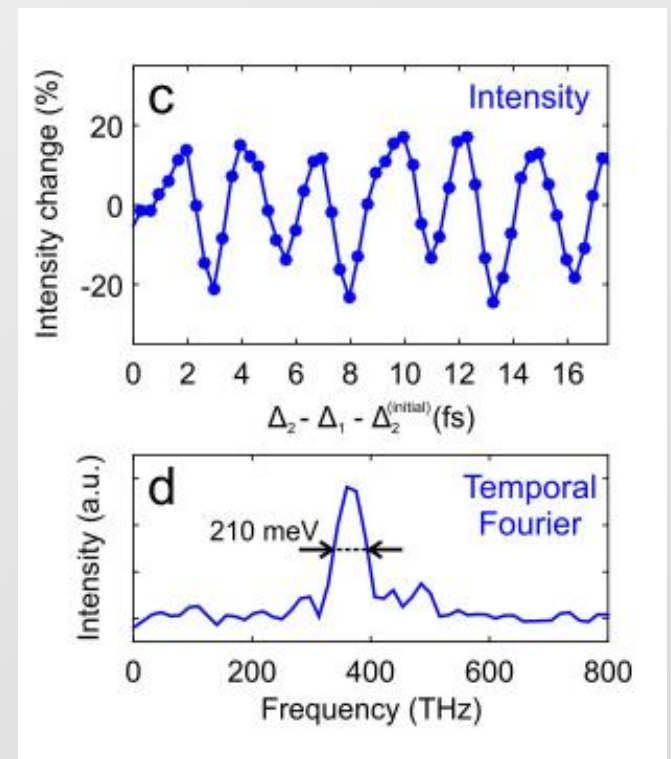
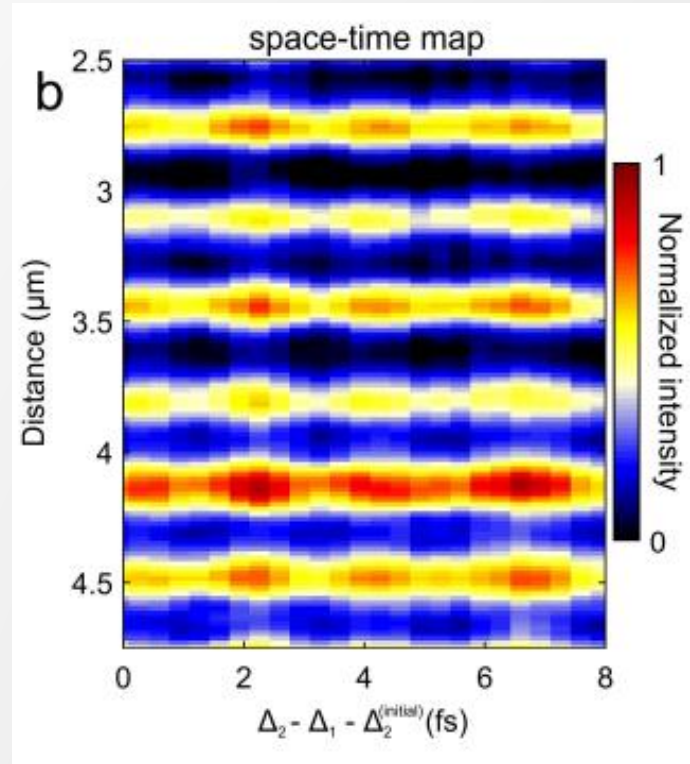
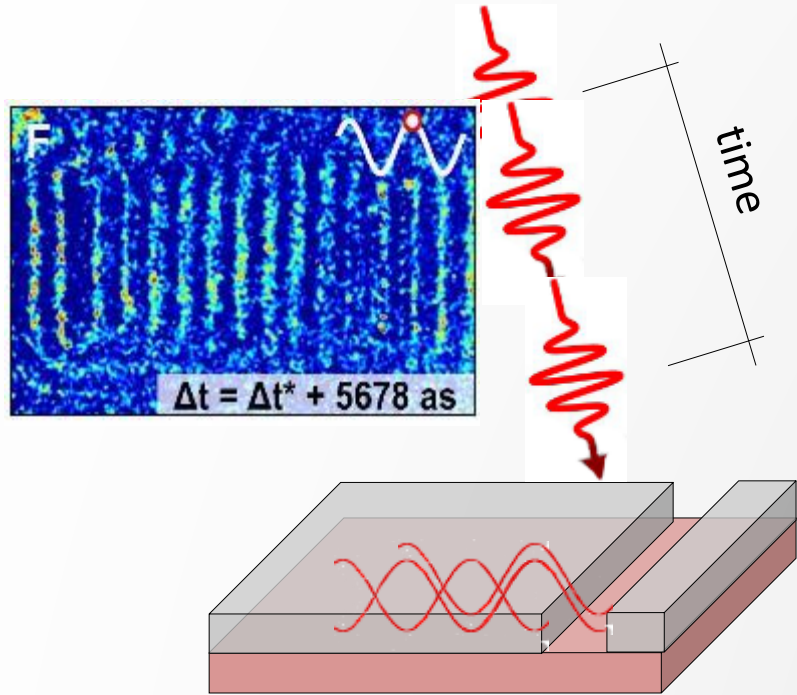


Lummen et al., *Nat. Comm.* 7 13156 (2016)



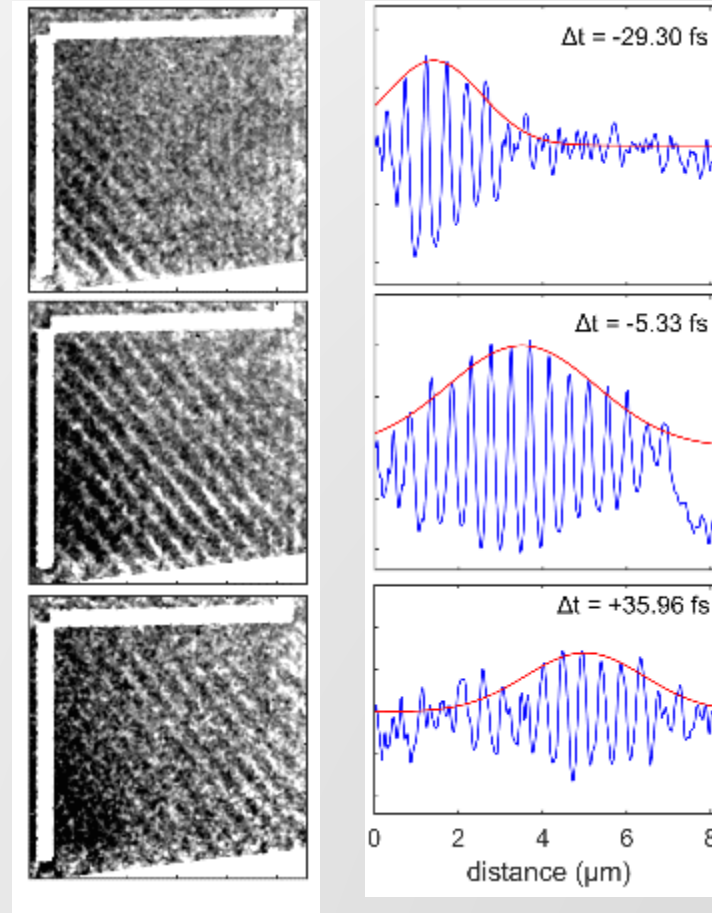
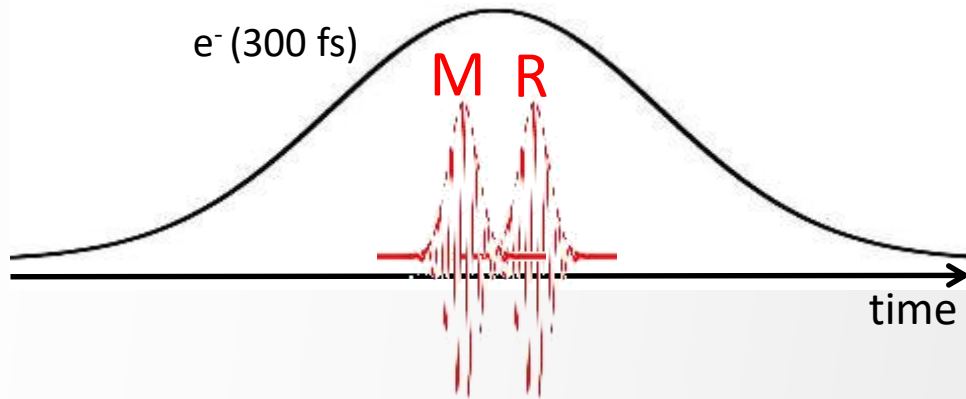
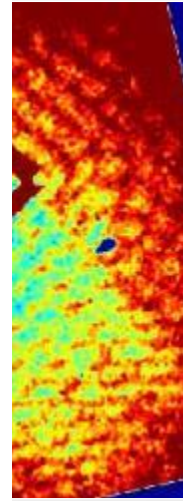
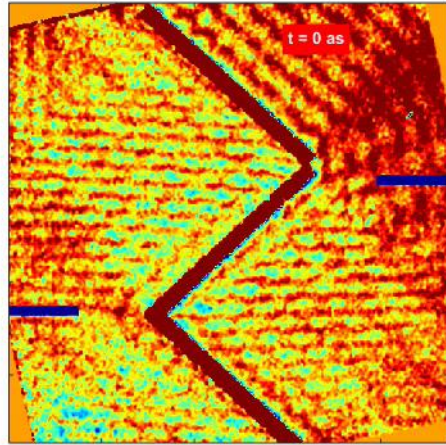
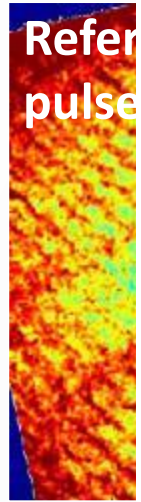
Piazza et al., *Nat. Comm.* 6 6407 (2015)

Most cited article in Nat Comm since 2015



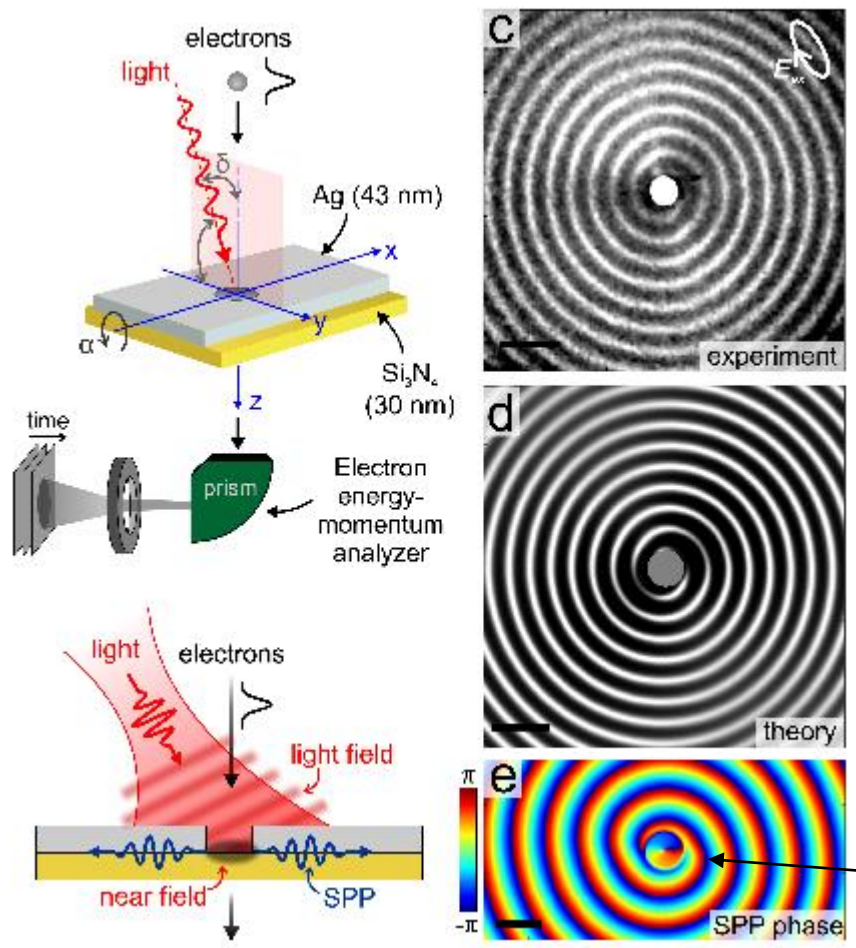
Vanacore, et al. *Nat. Comm* 9 2694 (2018)



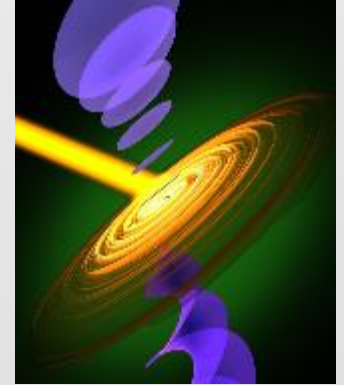


Madan et al, *Science Advances* 6 eaav8358 (2019)
News and Views article:
Ropers, *Nature* 571, 331 (2019)

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



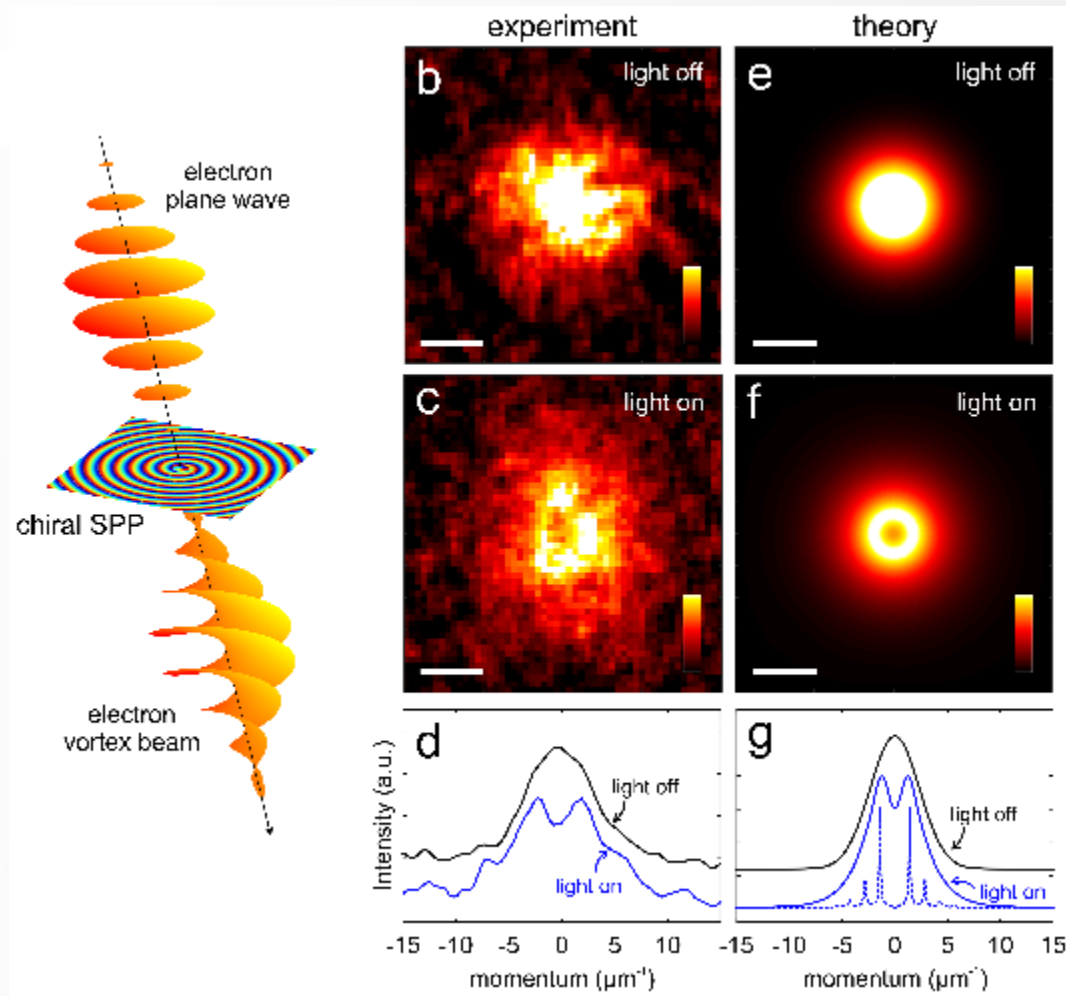
- Chiral SPP launched by elliptically polarized light
- Simulated Chiral SPP
- A chiral plasmonic field can impart a phase singularity onto the transverse component of an e^- wavefunction



Vanacore et al., *Nature Materials* 18, 573 (2019)

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

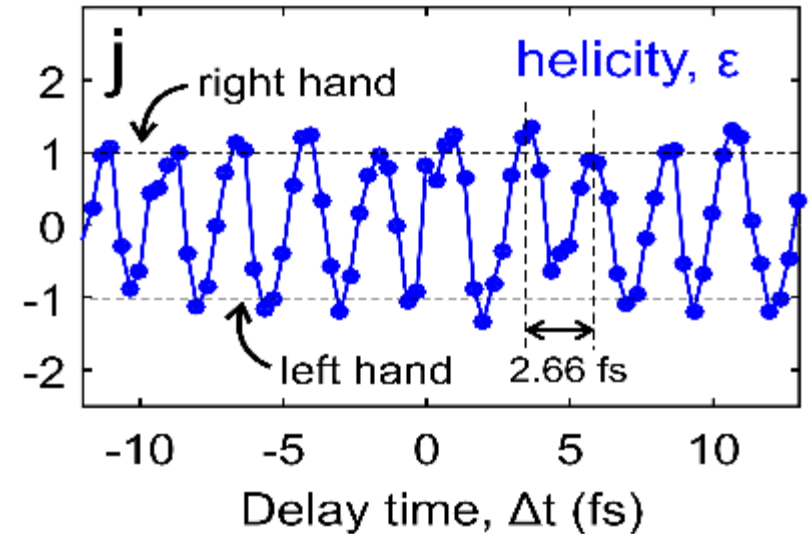
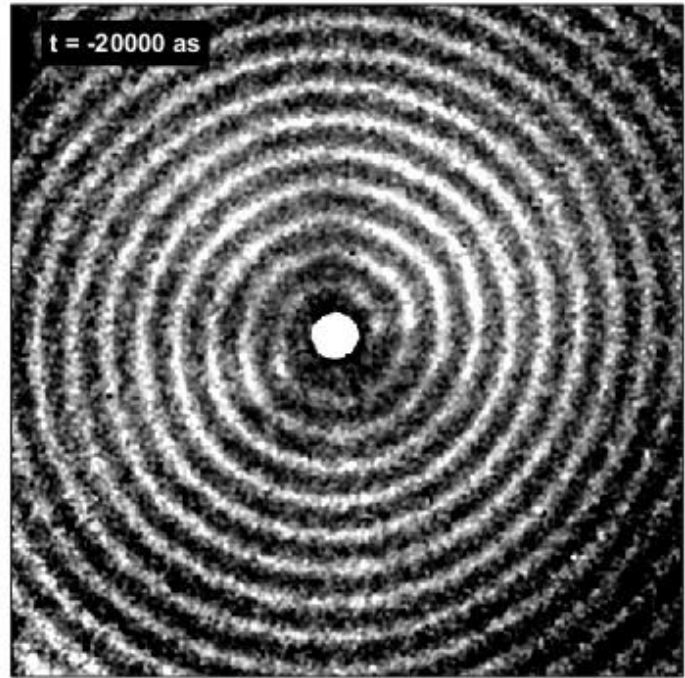
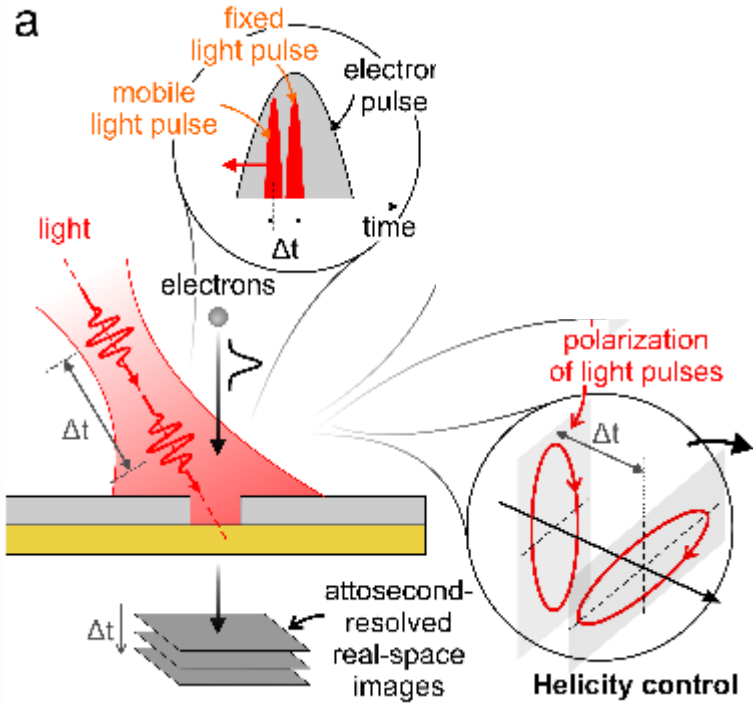
https://www.youtube.com/watch?time_continue=5&v=TIJ-mlh2LSI&feature=emb_logo



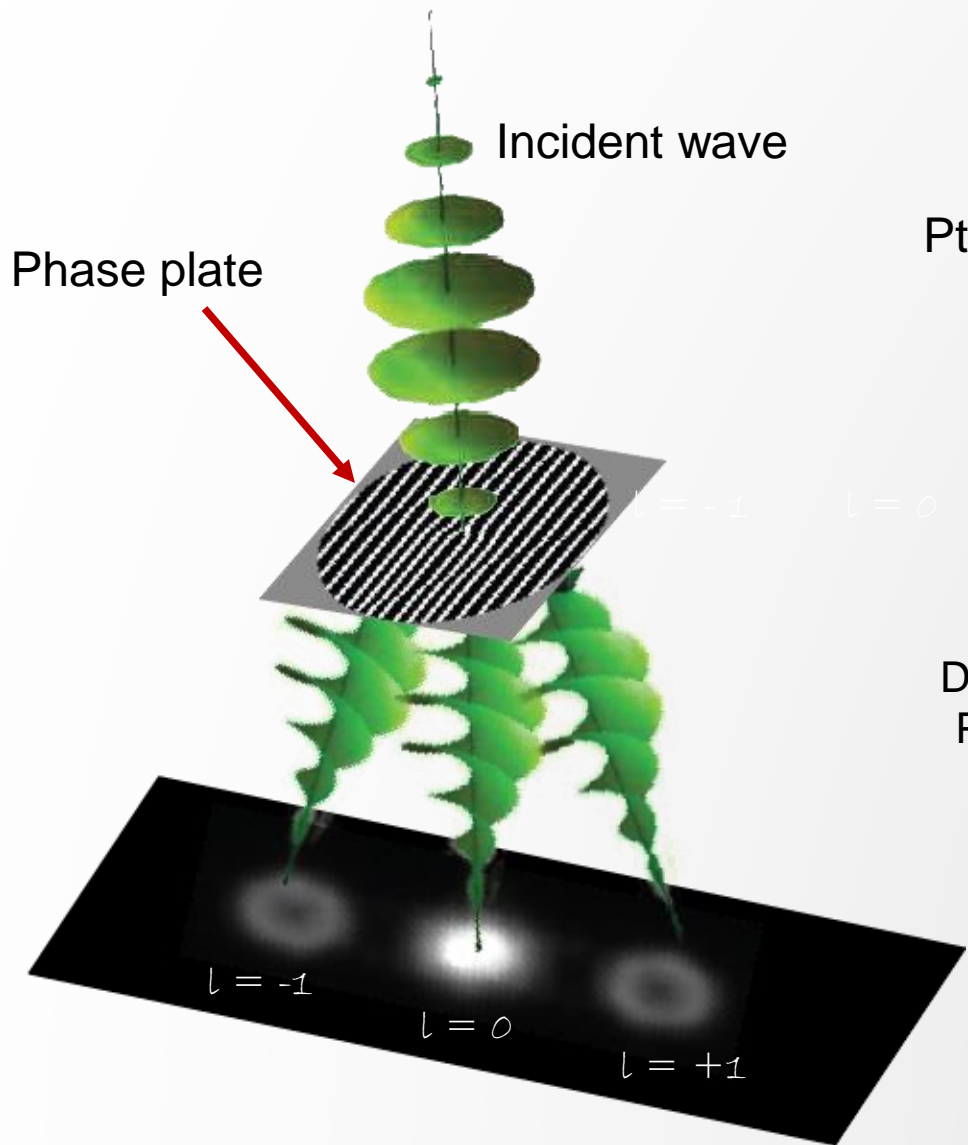
Electron wavefunction microscopy
in momentum space

High Dispersion Electron Diffraction (CL~80m)

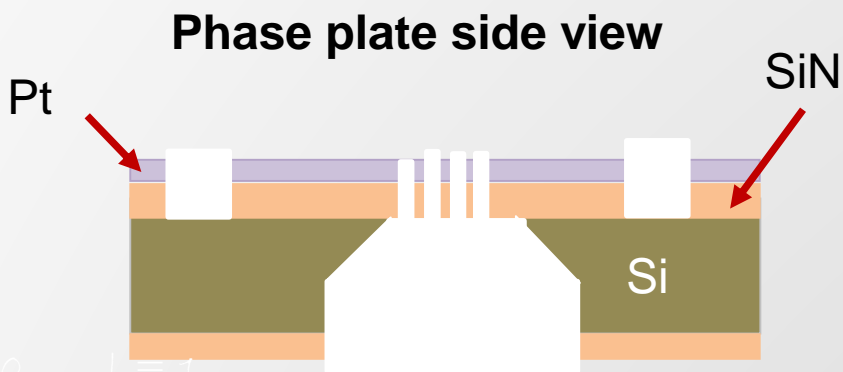
Vanacore et al., *Nature Materials* 18, 573 (2019)



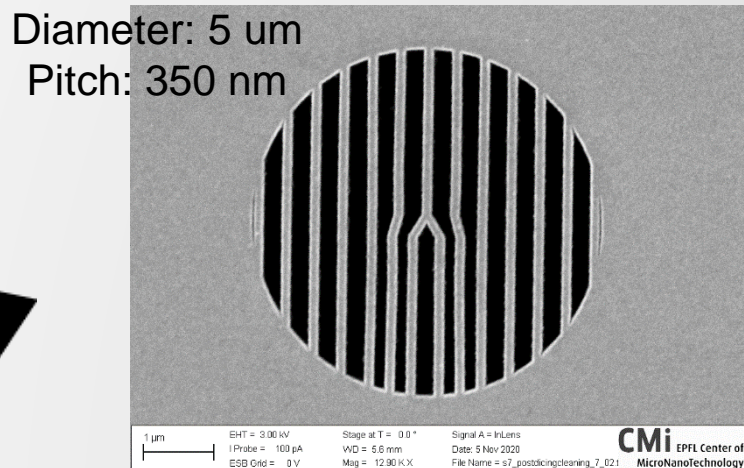
Three pulses experiment: attosecond control of the topological charge of an e^- wavefunction between +1 and -1



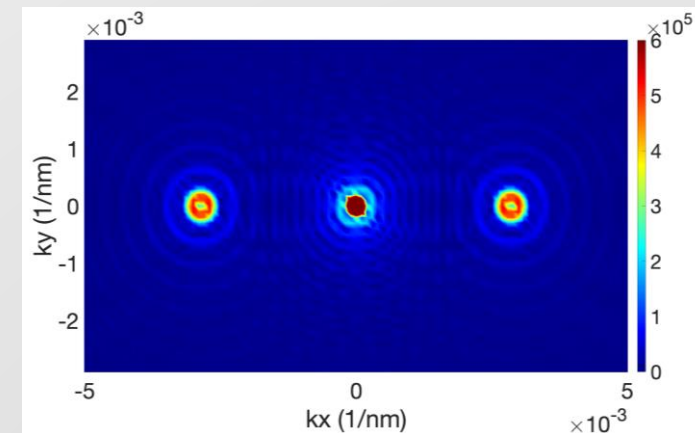
Nanofabrication



SEM image



Simulations



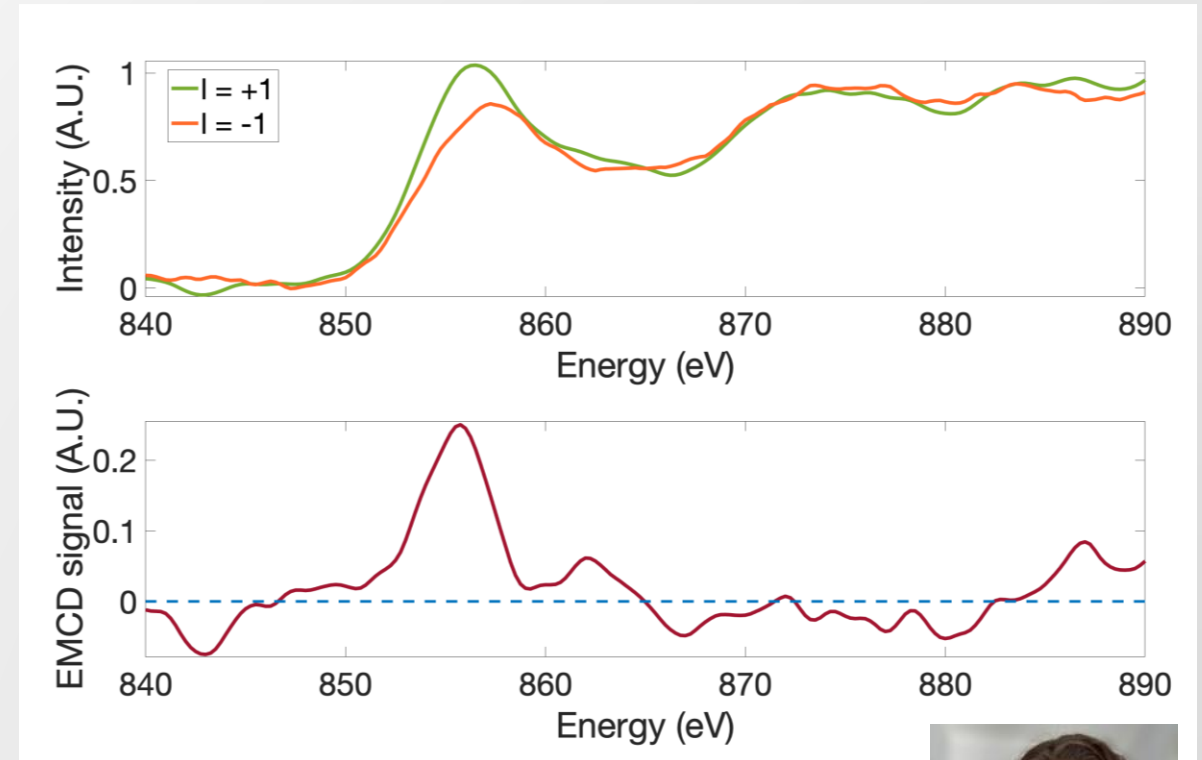
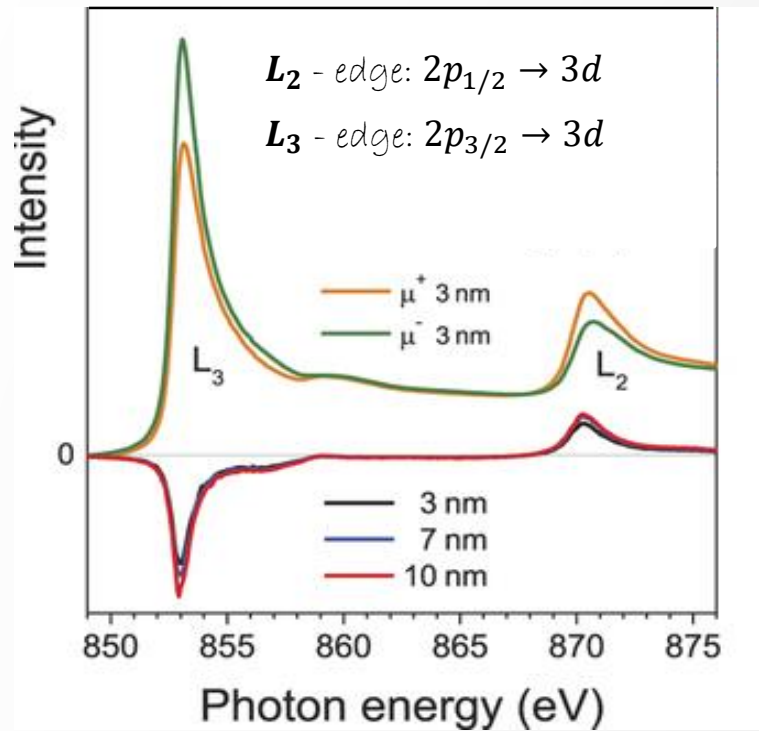
Experimental



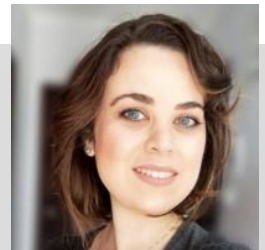
XMCD from literature (H. Shiozawa et al. Scientific Reports (2015))

EMCD with phaseplates

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

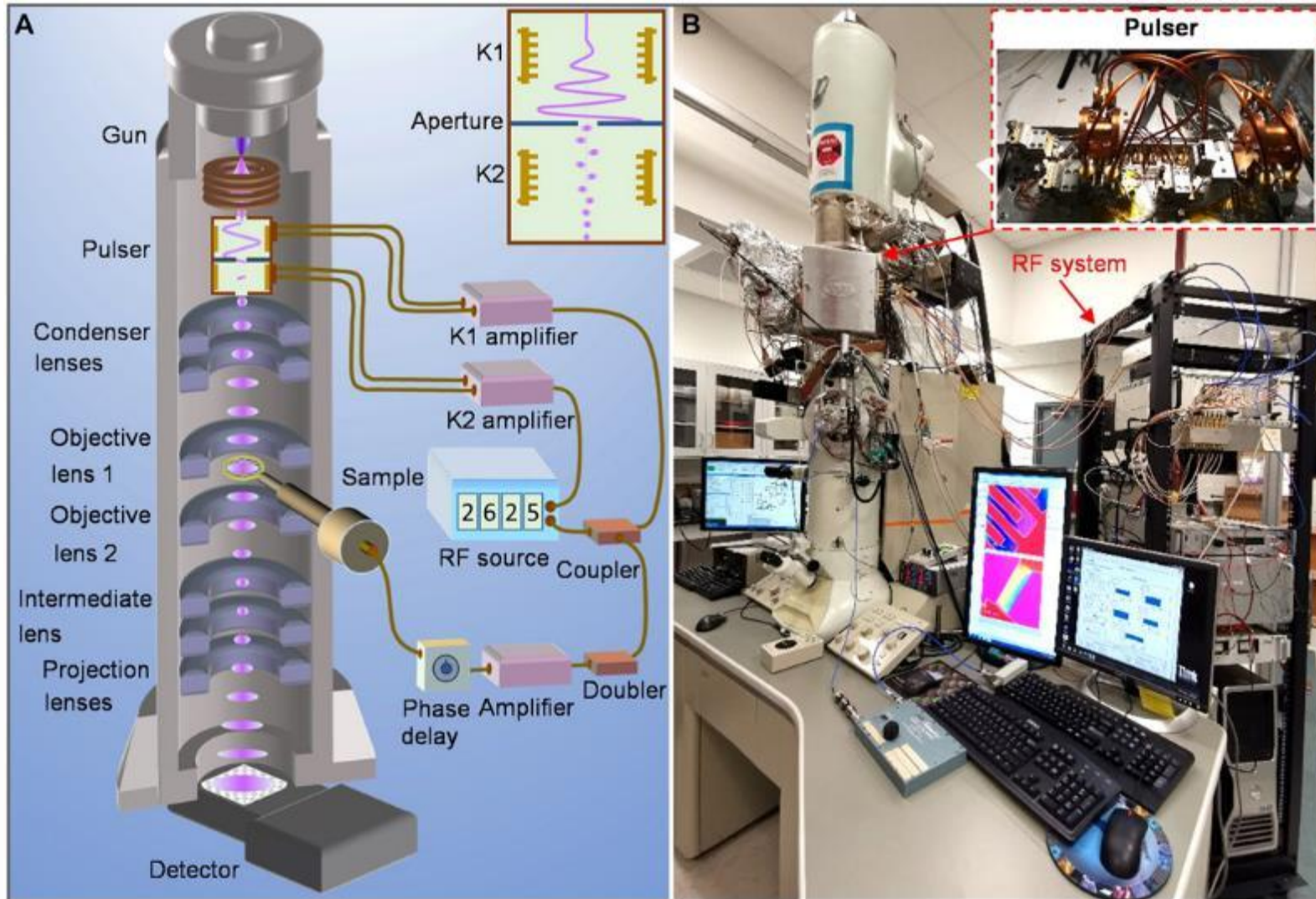


EMCD is based on the analysis of ionization edges in Electron Energy Loss Spectroscopy (EELS), allowing to quantitatively determine the spin and the orbital magnetic moments with atomic spatial resolution depth sensitivity

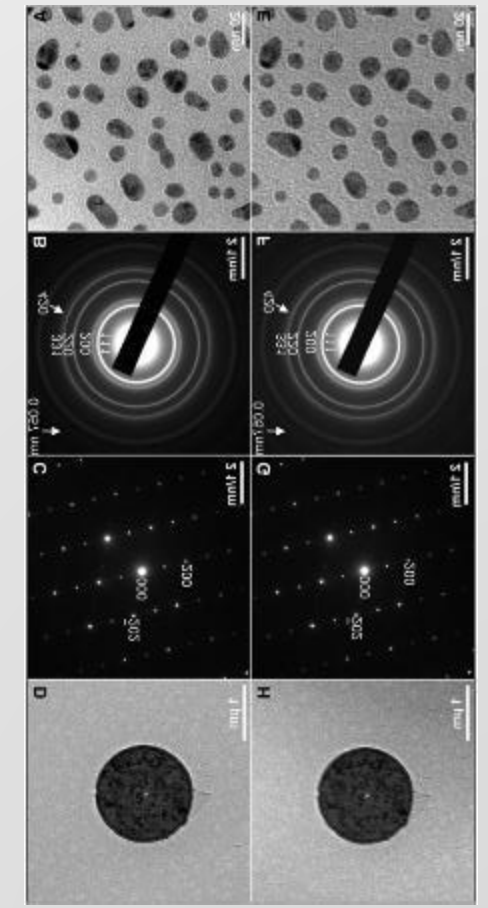


Veronica Lecesse

Laser free UTEM for magnetic imaging with ~ 10 ps resolution



Conventional mode RF cavity mode



Xuwen Fu et al. Sci Adv 2020

