Sources d’électrons rapides: quelles nouvelles performances en résolutions spatiale et énergétique pour quelles applications?

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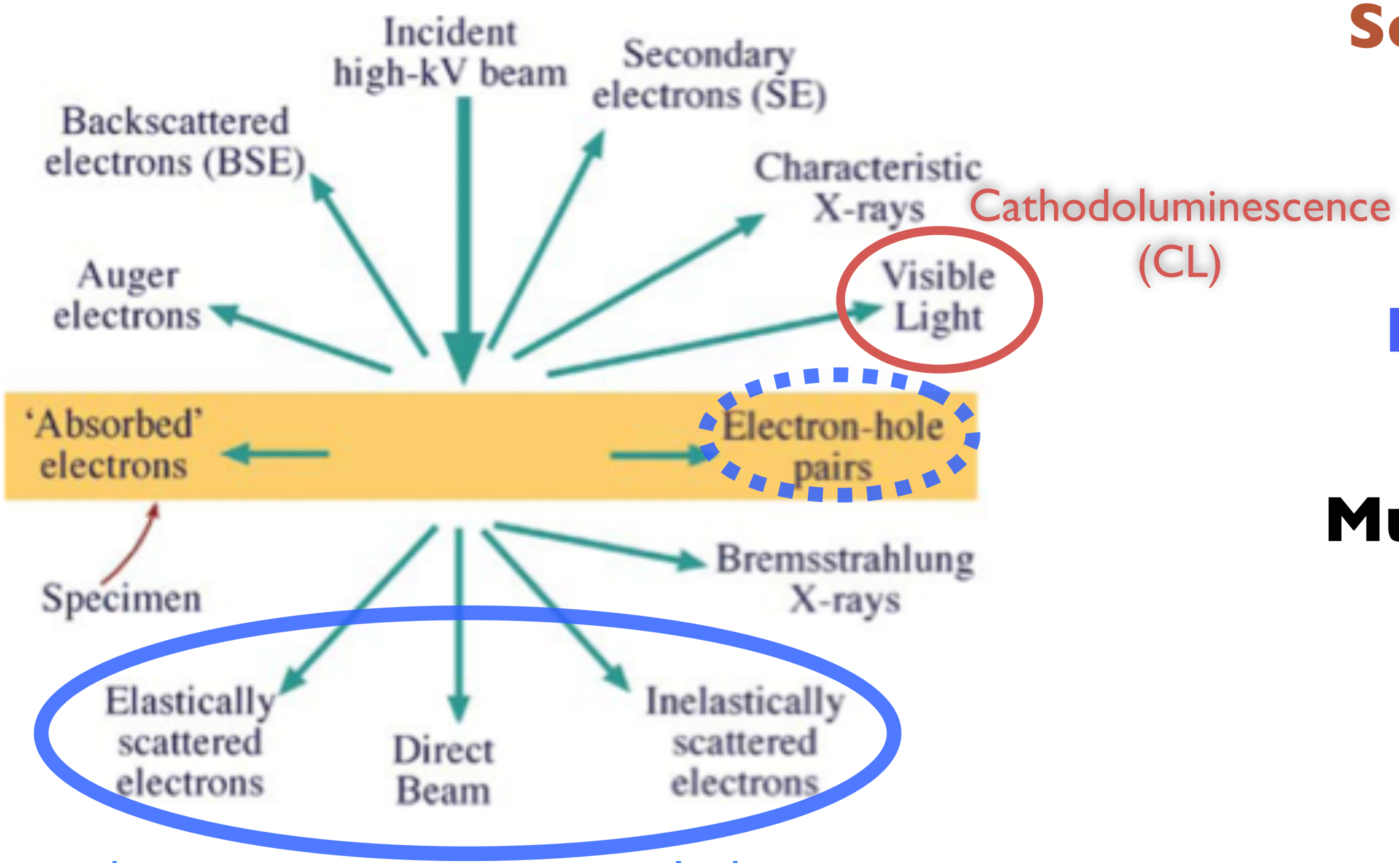
Equipement d’excellence TEMPOS

Transmission Electron Microscopy at Palaiseau Orsay Saclay

Electron matter interaction

a huge variety of measureable signals in SEM and (S)TEM



Coherent or incoherent
elastic imaging

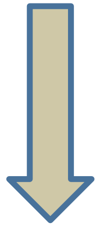
Inelastic imaging
EELS = Electron Energy-Loss Spectroscopy

Secondary Event

vs.

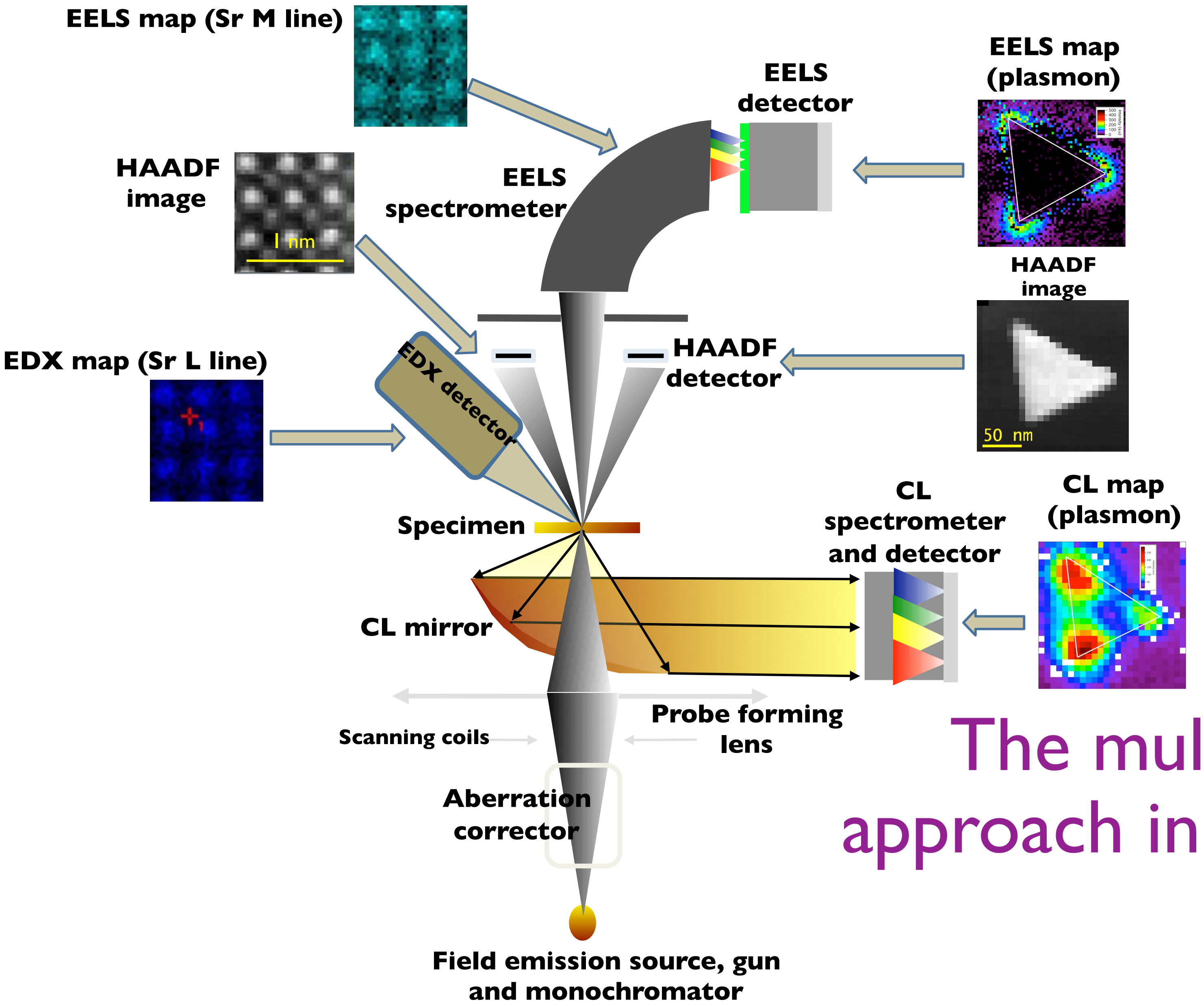
Primary Event

Multi-signal



Multi-information

Exploiting new physical events/signals combinations



The multisignal approach in the STEM

- Pushing the limits of the existing spectroscopic techniques
- Inventing new spectroscopies

Quelles performances actuelles en microscopie électronique en transmission?

- Résolution spatiale (correcteurs d'aberrations de la lentille objectif)
- Résolution énergétique (monochromateurs)
- Convergence photons/électrons

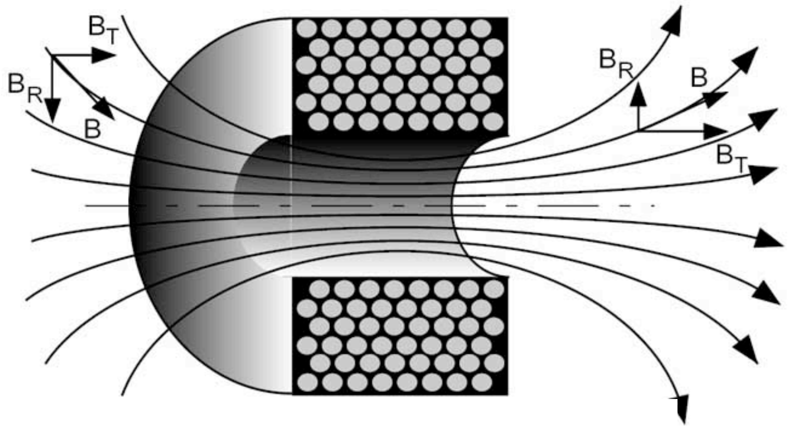
Pour quelles applications?

Recent advances in spatial resolution

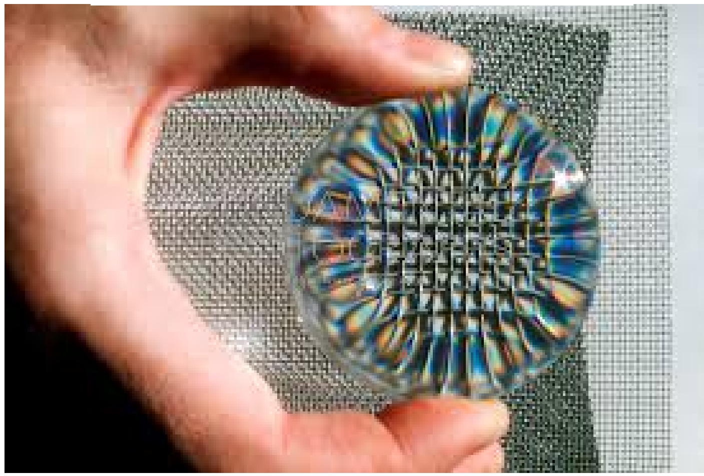
- Electron aberration correctors
- New possibilities in imaging

Imaging down to the atomic scale

λ (100 keV) = h/p = 3 pm

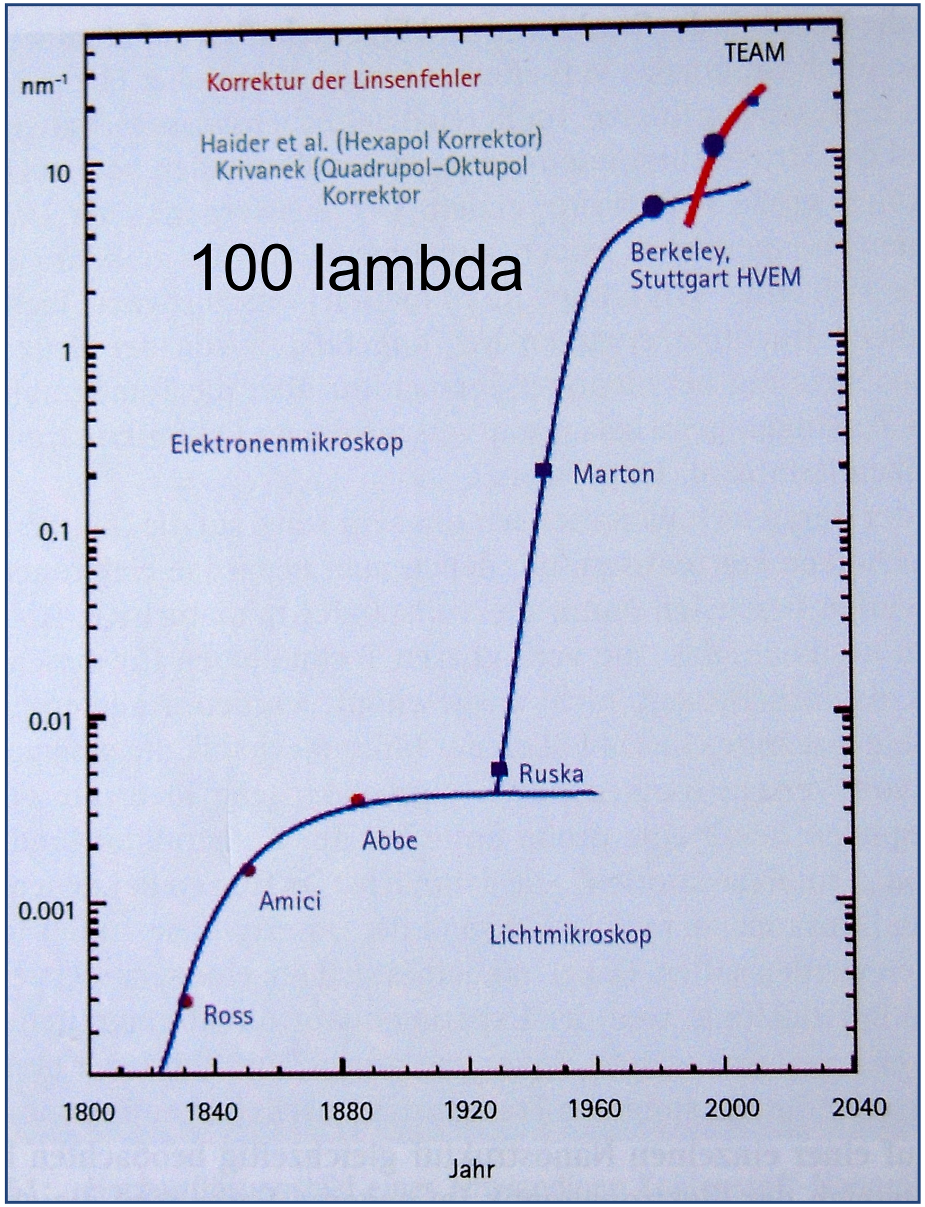


$$d_o = 0.61\,\lambda/\alpha_o$$



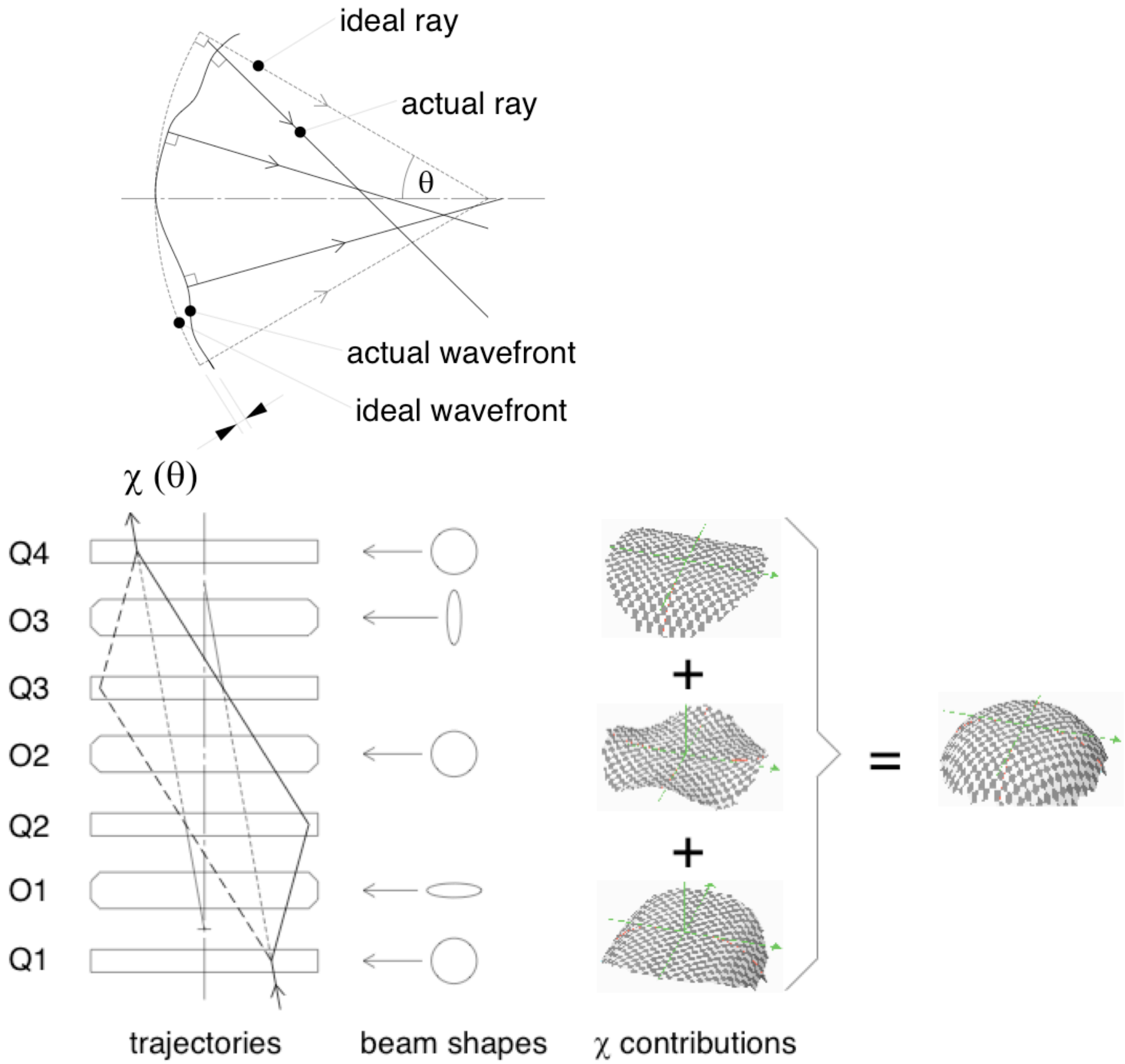
d~1,5 Å =50 λ

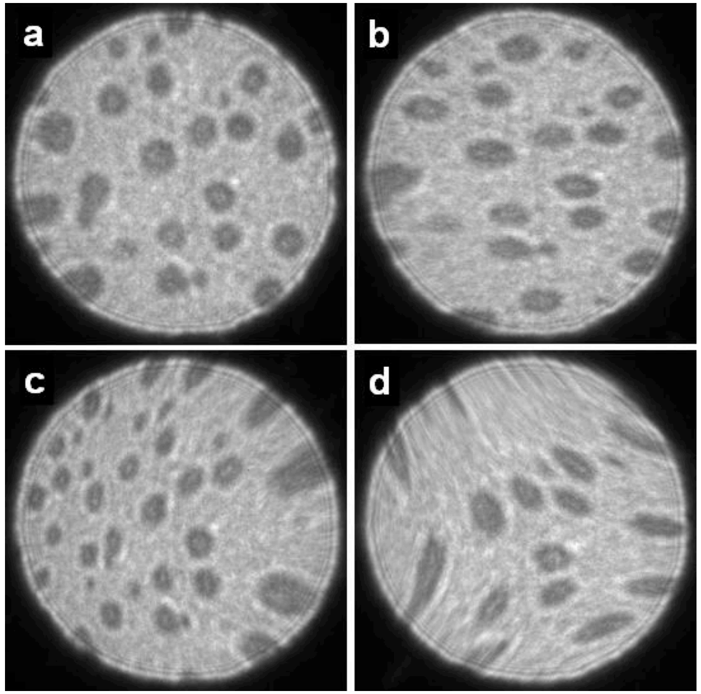
20 lambda = 60 pm



Harald Rose (2008)

Electron aberration correctors



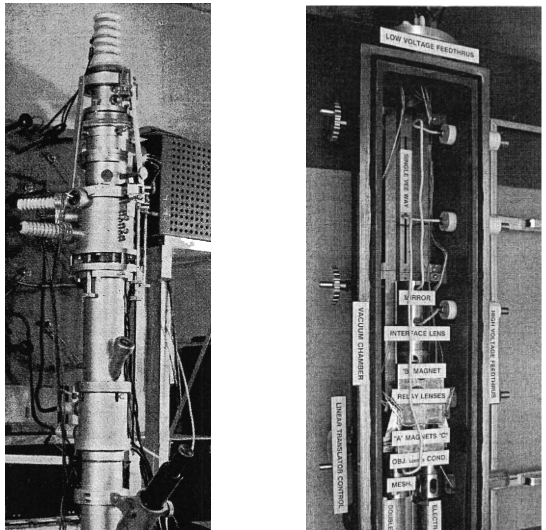


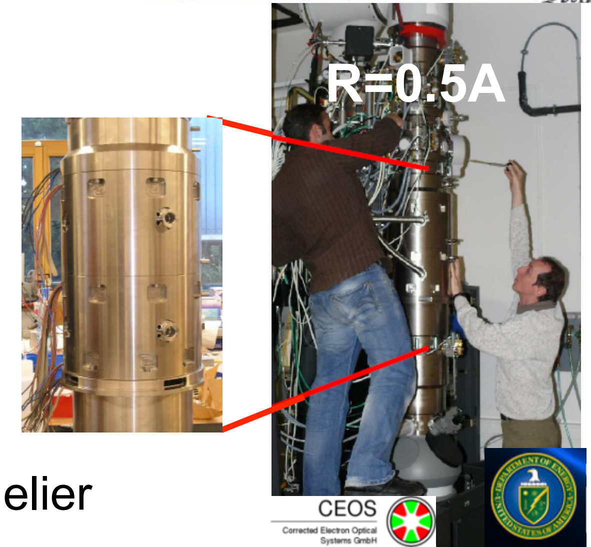
O. Krivanek (2007)

Augmenter la résolution d'un microscope: deux solutions

λ 1 MeV

corriger les aberrations





30 keV

courtesy: F. Houdelier

(Incoherent) elastic imaging

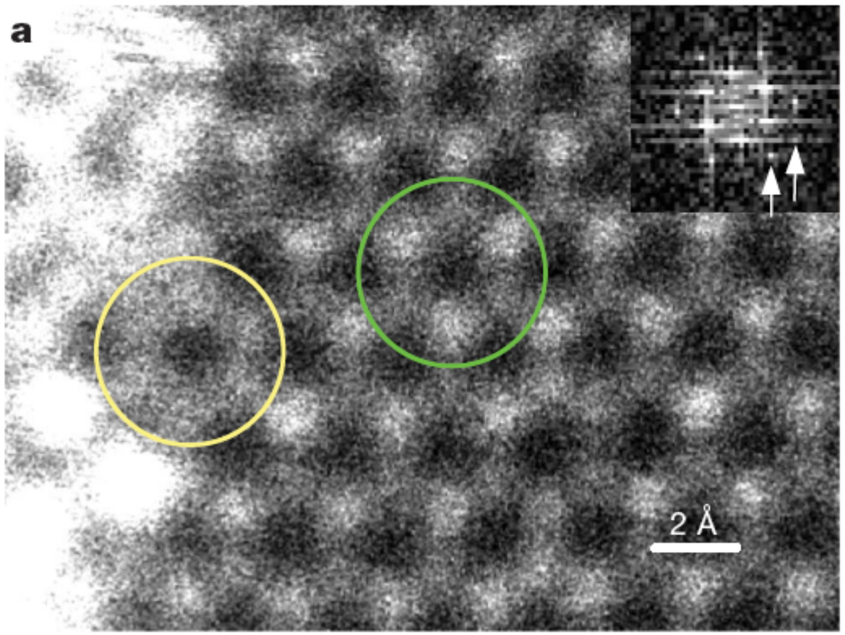
Z-contrast on h-BN monolayer

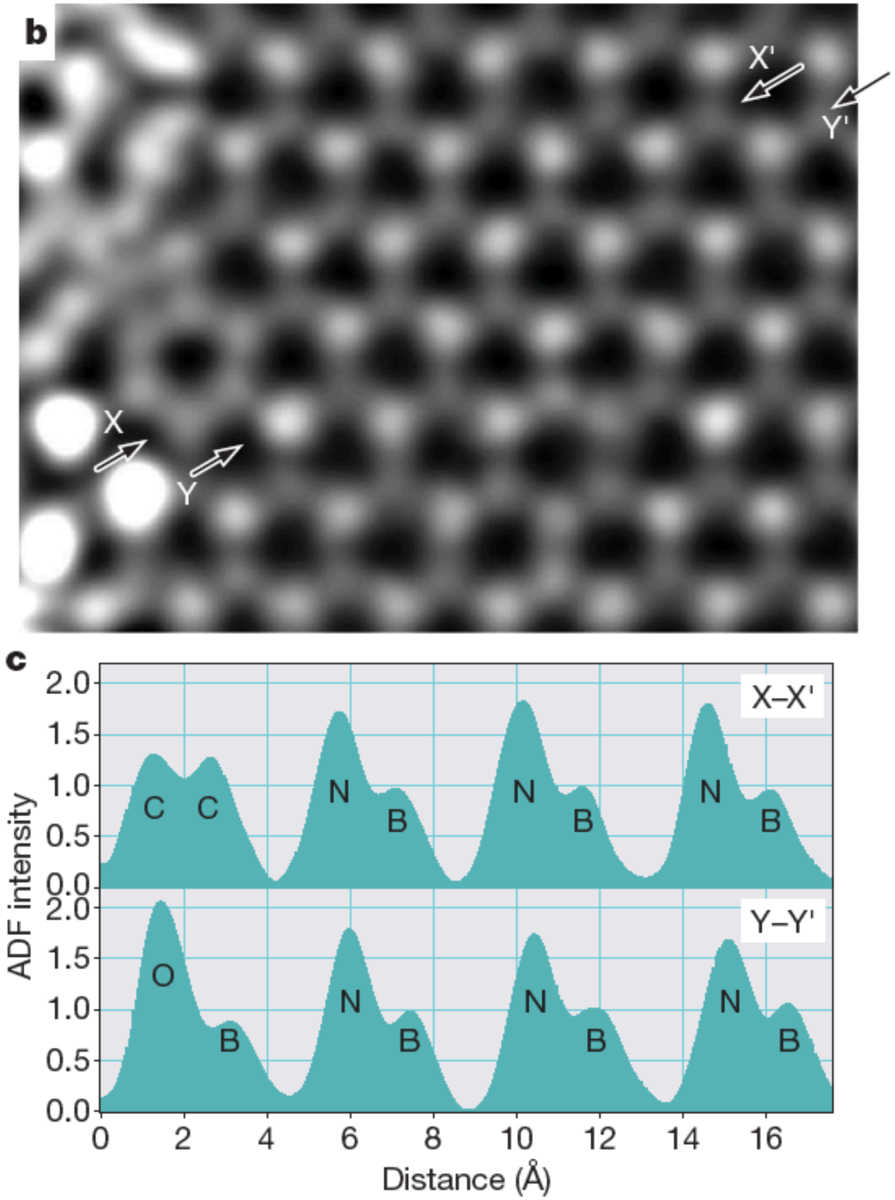
Vol 464 | 25 March 2010 | doi:10.1038/nature08879 nature

LETTERS

Atom-by-atom structural and chemical analysis by annular dark-field electron microscopy

Ondrej L. Krivanek[1], Matthew F. Chisholm[2], Valeria Nicolosi[3], Timothy J. Pennycook[2,4], George J. Corbin[1], Niklas Dellby[1], Matthew F. Murfitt[1], Christopher S. Own[1], Zoltan S. Szilagyi[1], Mark P. Oxley[2,4], Sokrates T. Pantelides[2,4] & Stephen J. Pennycook[2,4]

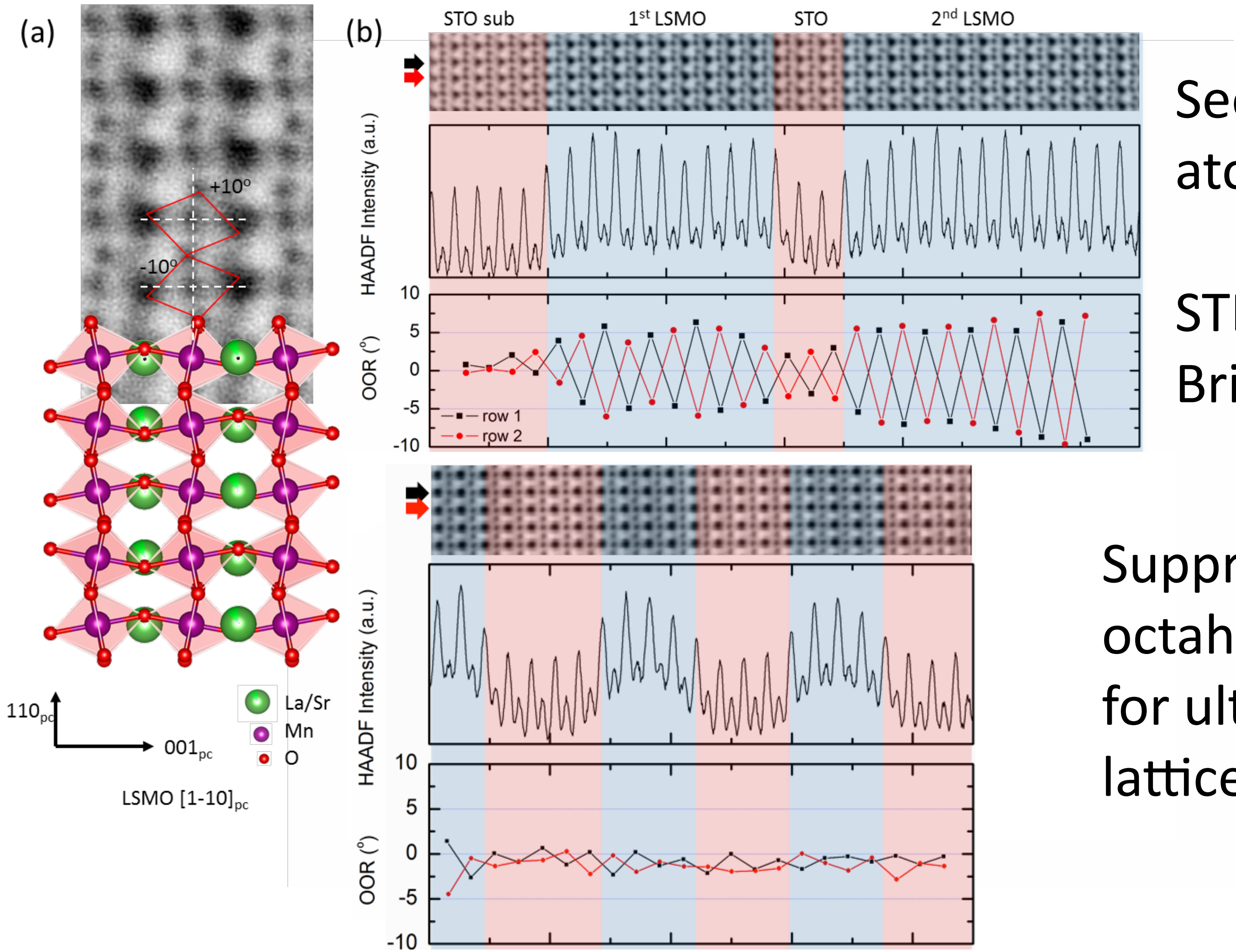




Nature, Mars 2010

(Coherent) elastic imaging

Impact of interfacial coupling of oxygen octahedra on ferromagnetic order in $La_{0.7}Sr_{0.3}MnO_3/SrTiO_3$ superlattices



Seeing oxygen atoms in real space

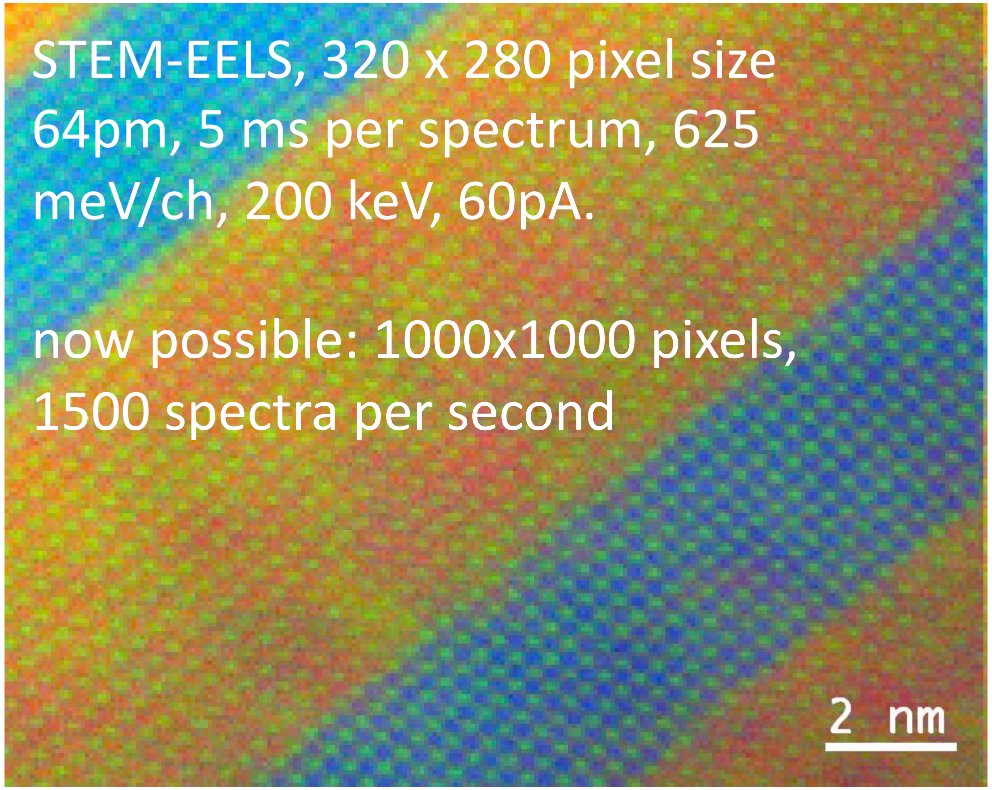
STEM-ABF (annular Brigth field)

Suppression of octahedra rotation for ultra thin super lattices

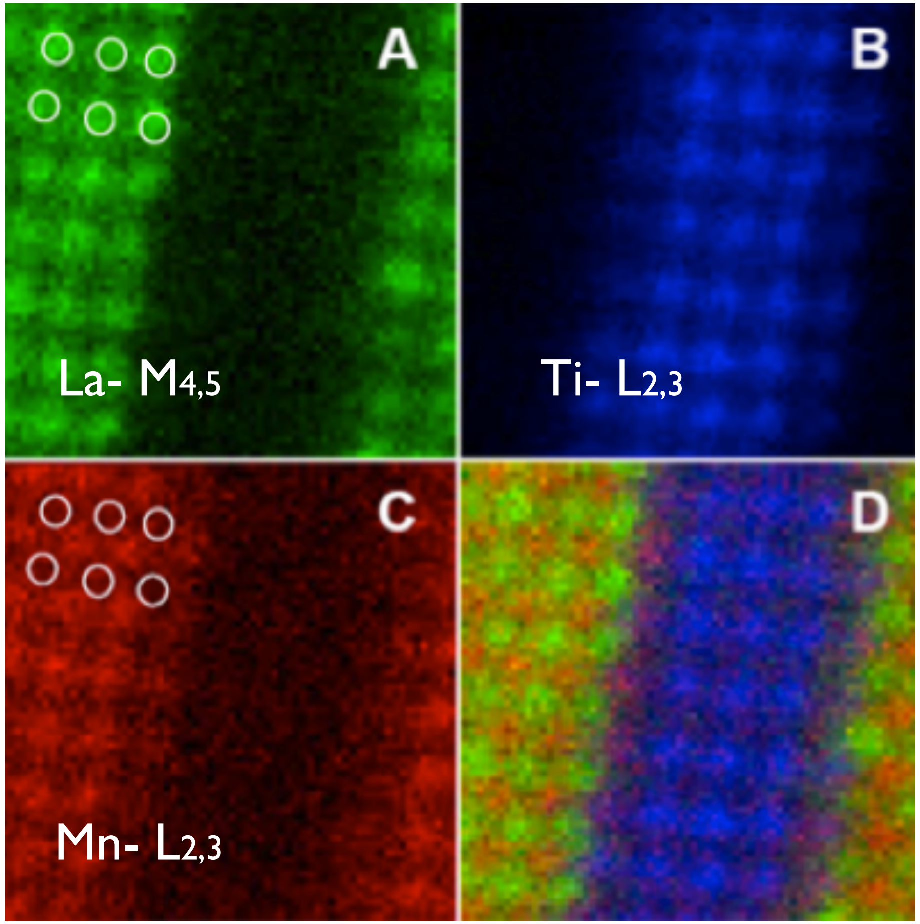
Xiaoyan Li, Ionela Lindfors-Vrejoiu, Michael Ziese, Alexandre Gloter and Peter A. van Aken

Inelastic imaging

atomic scale chemical imaging of composition by EELS



Data NION USTEM200 (Orsay)



D. Muller et al. Science 319 (2008) 1073)

D. Muller et al Science (2008)

Recent advances in energy (spectral) resolution

- New generation monochromators/ energy resolution and stability

- New application range

Electron Monochromators: two strategies

- Monochromator working with **high energy electron (100keV)** (native dispersion is weak so a lot of optical element/quad to increase the dispersion; ground, no space charge, analyzer can be quite symmetric with mono), Krivanek et al.

- Monochromator working with **low energy electron (1keV)** (higher dispersion, low aberration, integrated in the gun), For example Wien filter from Mook and Kruit, UM (2000) 129-139.

Drawbacks: « Boersch » effect/low intensity, selected energy dependent from HT

Nion high energy-resolution monochromator

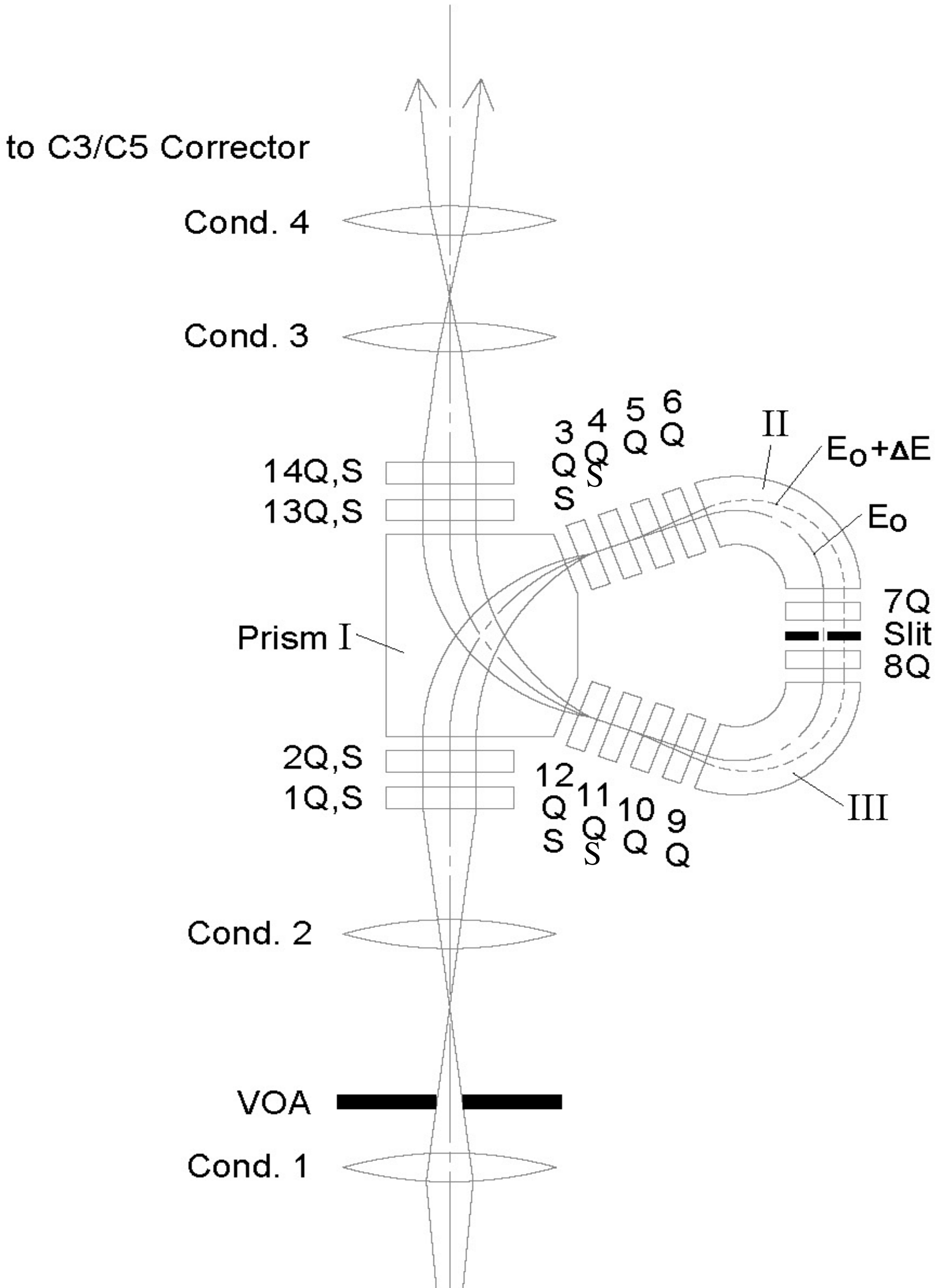


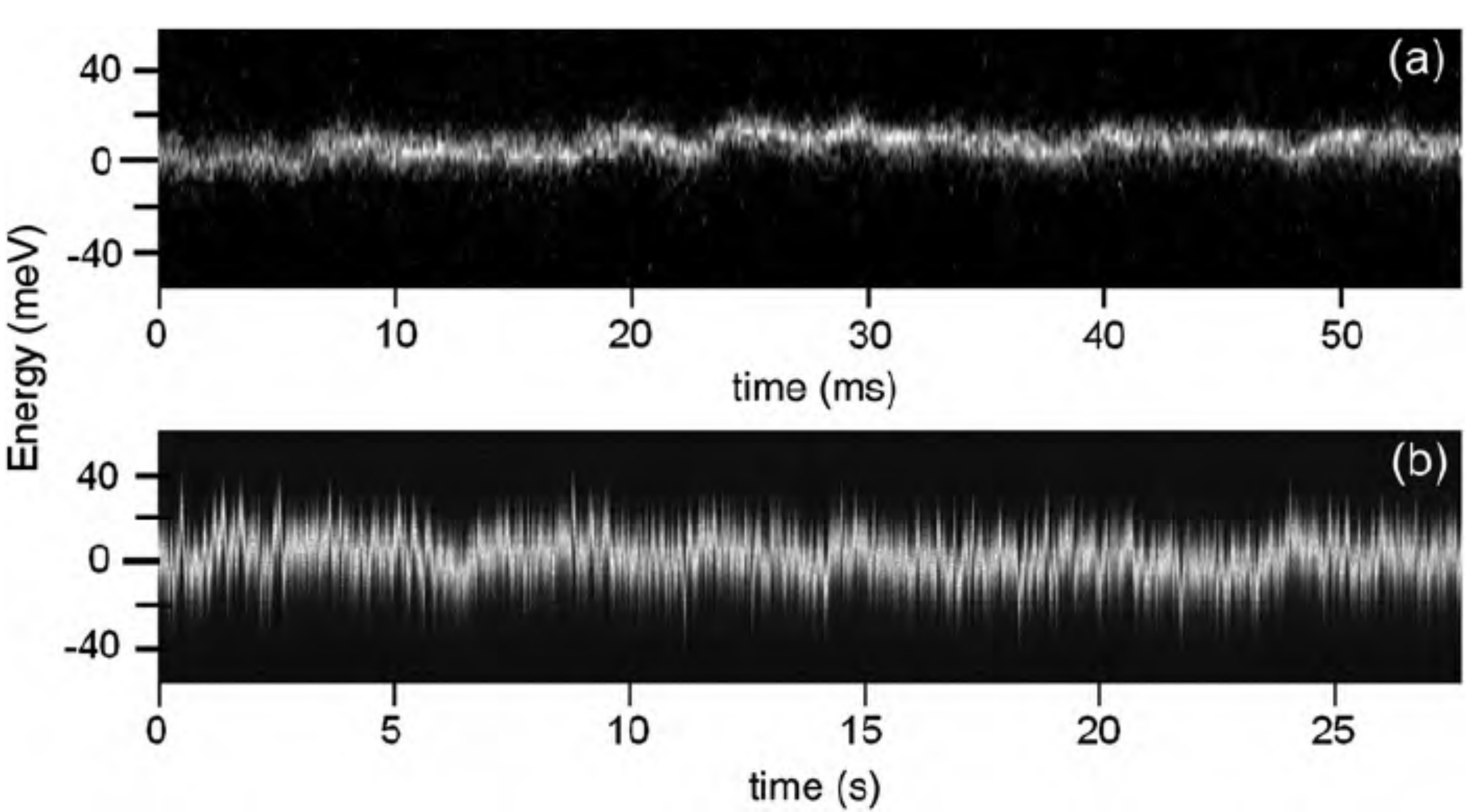
Figure shows Nion's design of a dispersing-undispersing monochromator operating on 20-200 kV electrons.

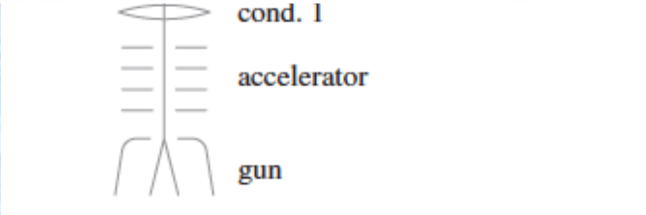
Spectrum is magnified at the slit. The spectrum is demagnified and the dispersion is canceled by the second half of the monochromator.

~ 30 meV energy resolution should be possible soon, better in the long term.

Now being built at Nion, for Arizona State U. and Rutgers U.

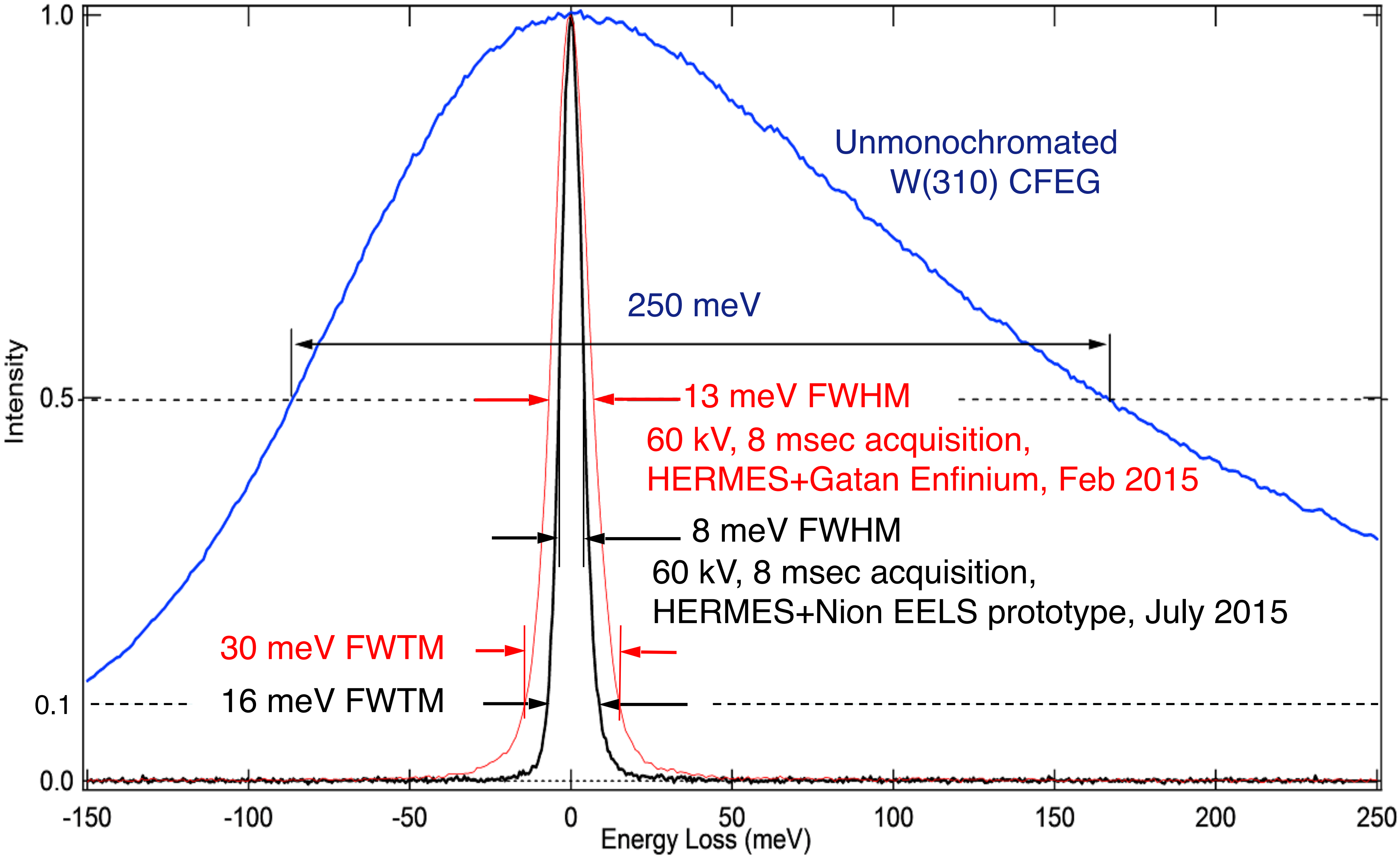
Nion high energy-resolution monochromator





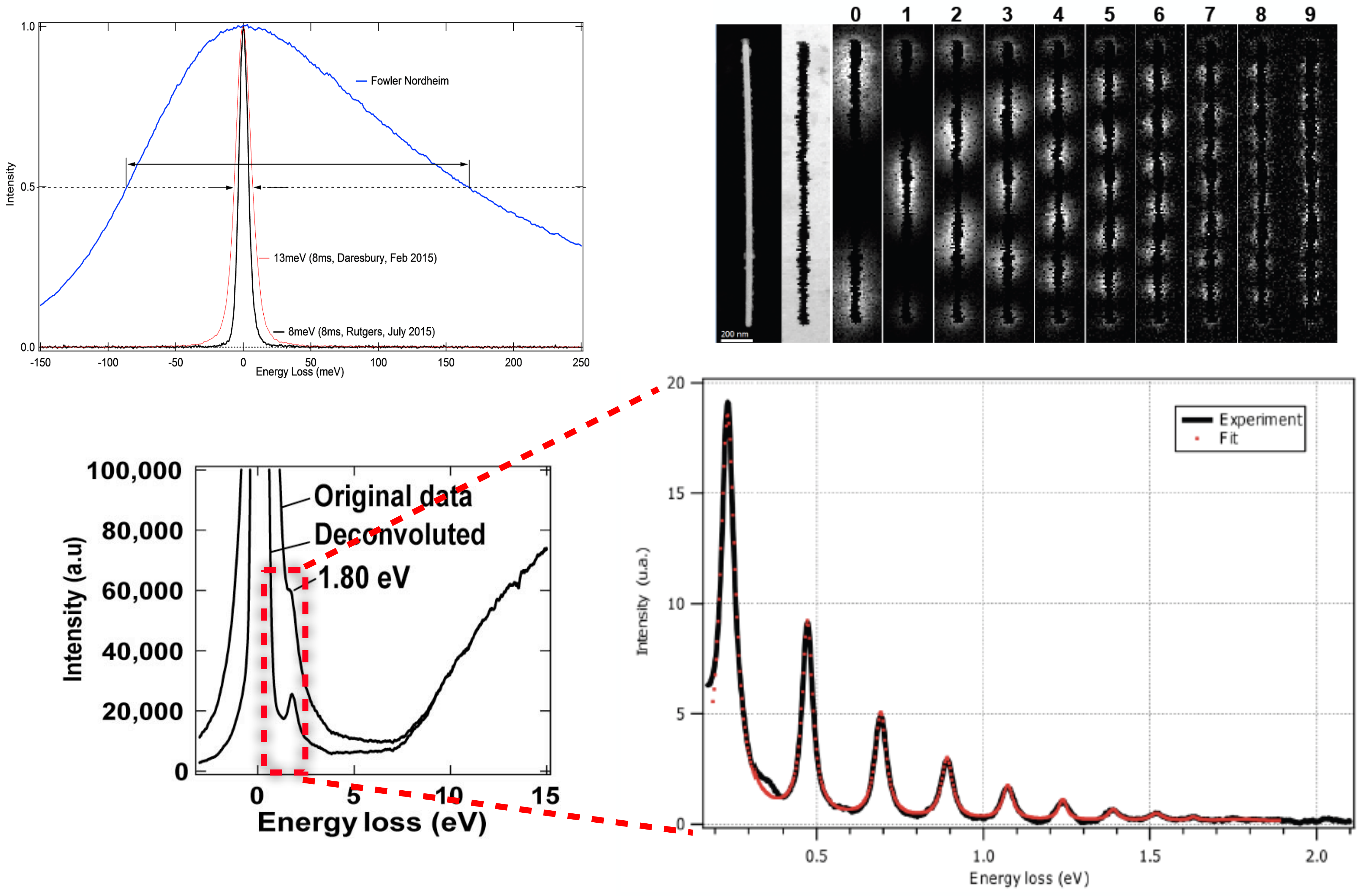
12 meV p-p (3.5 meV rms) ~10^{-7}

HERMES – Progress in Resolution and ZLP Tails

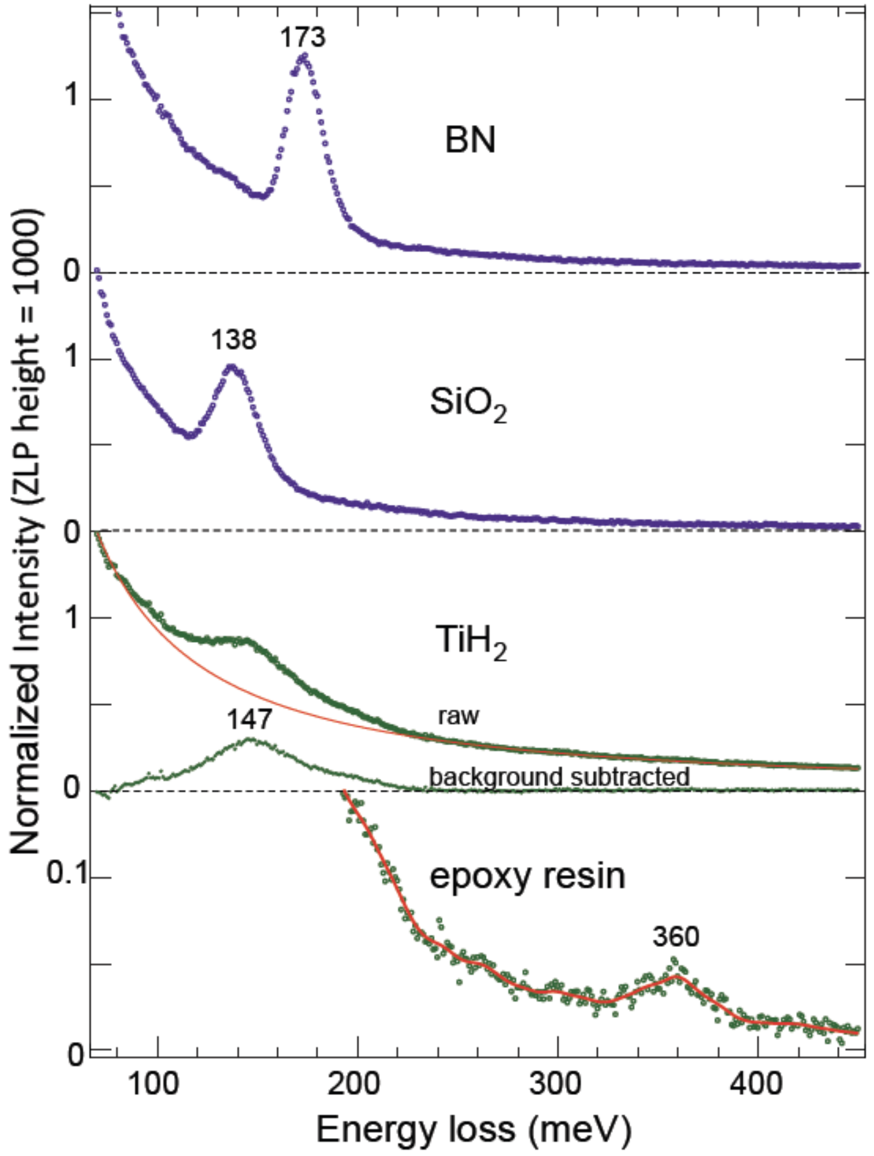


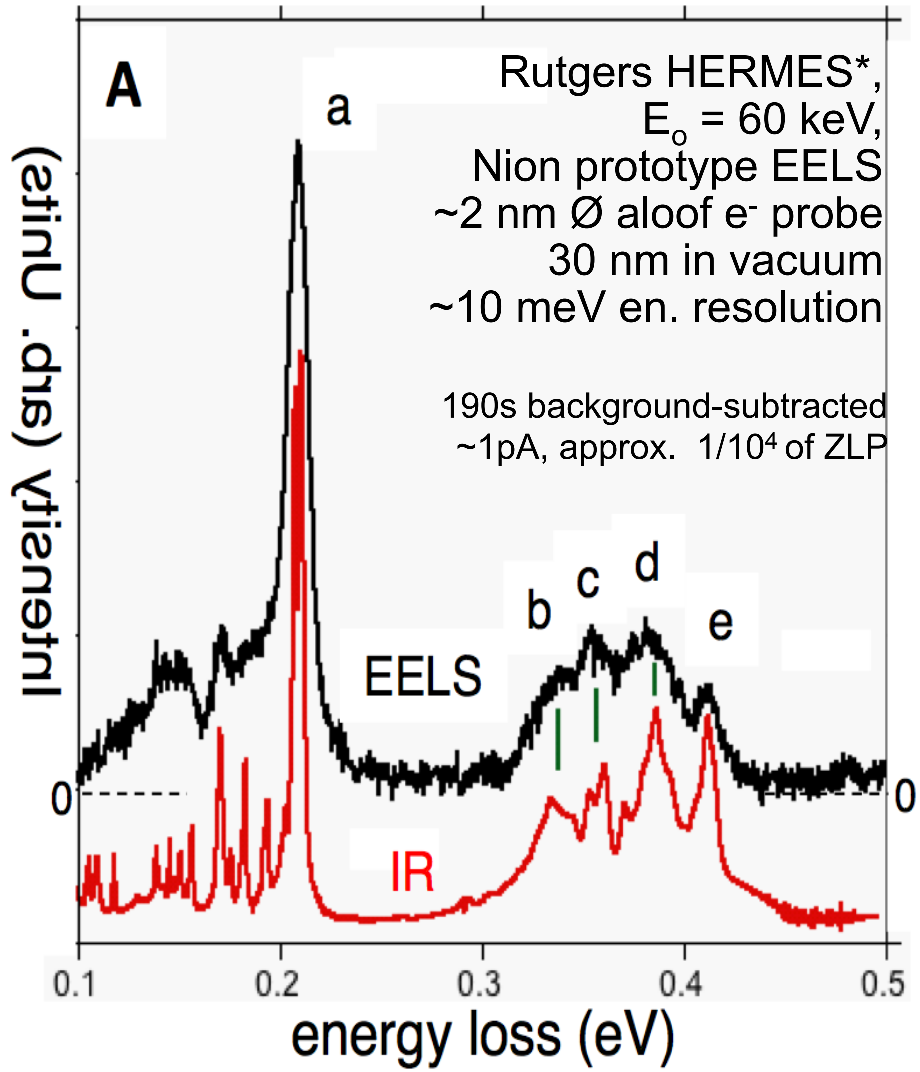
Courtesy O. Krivanek

Accessing new excitations/spectral ranges
IR plasmonic modes



Accessing new excitations/spectral ranges (vibrational and phonon modes)





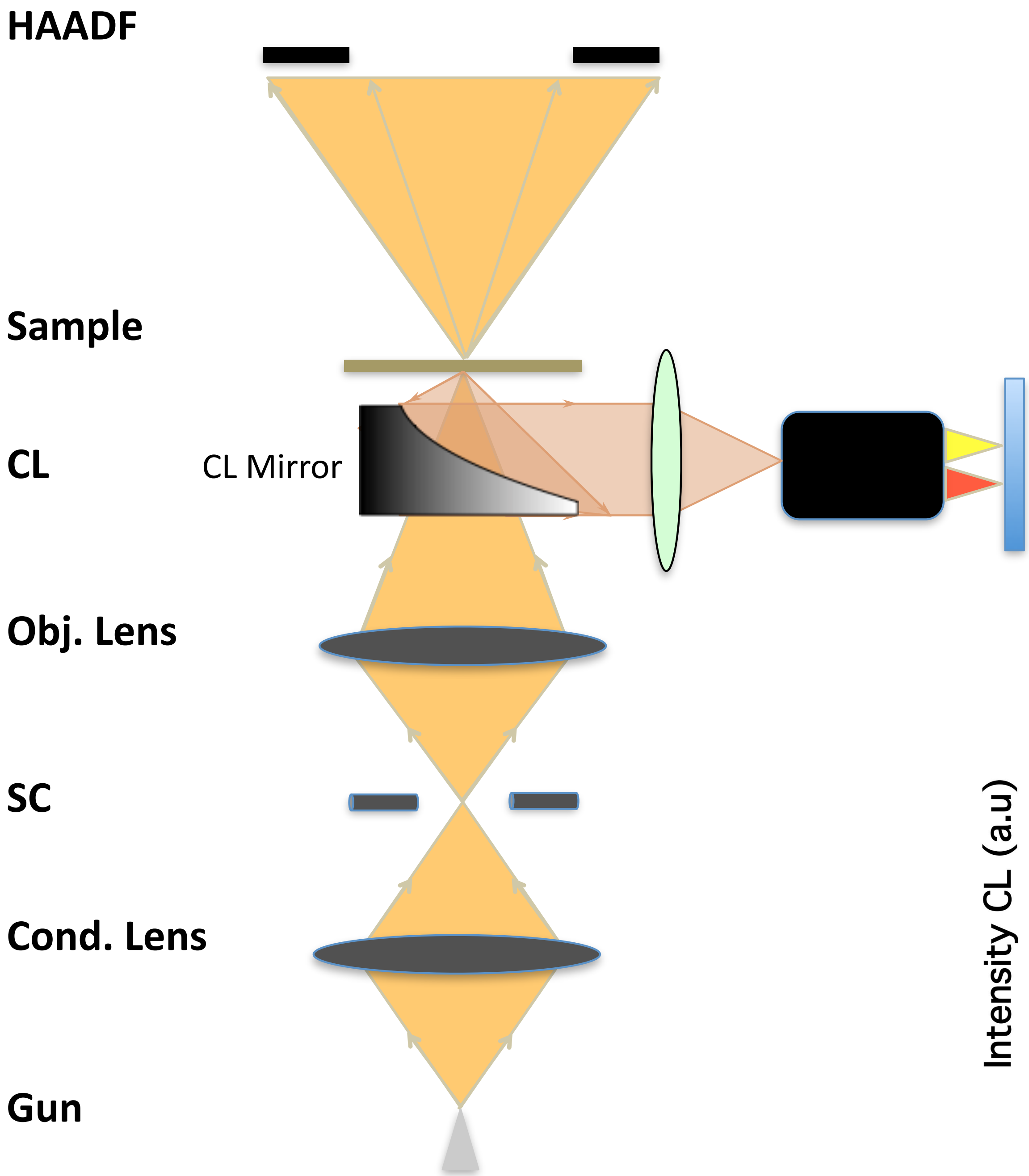
aloof EELS-FTIR comparison

Courtesy O. Krivanek

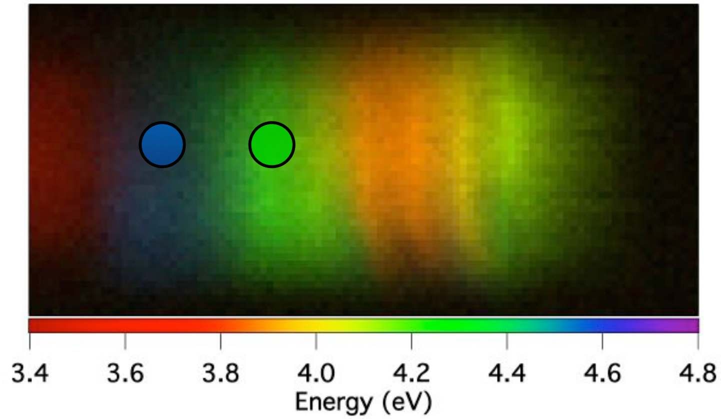
When electrons meet photons

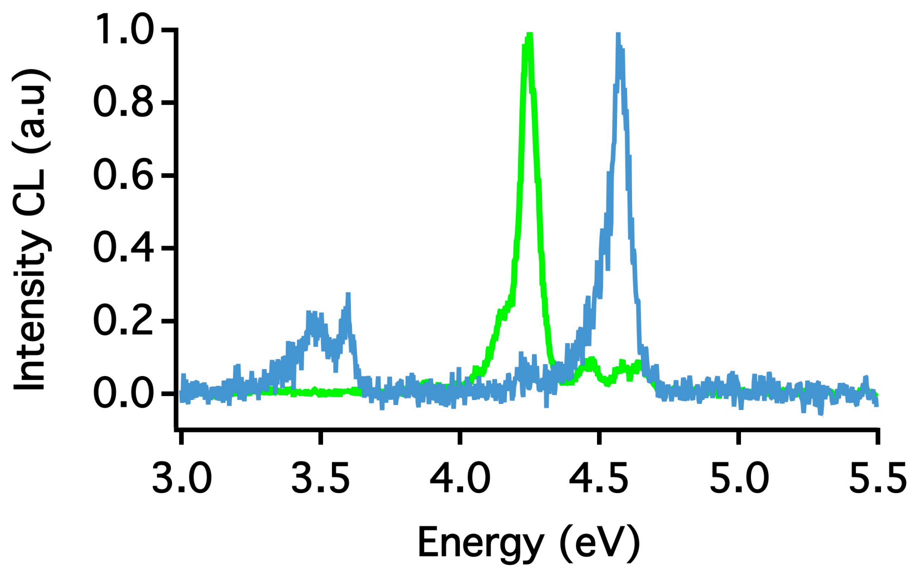
- Benefit from high spatial resolution from electrons and temporal resolution from photons
- Compare the physics of excitation
- Inventing new spectroscopy approaches

Cathodoluminescence Spectrum imaging



$10^4 \ldots 10^6$ pixels



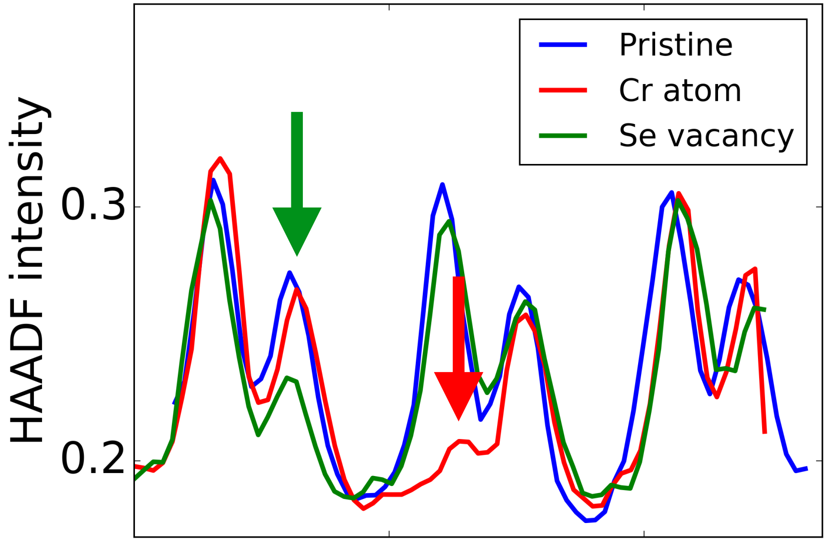


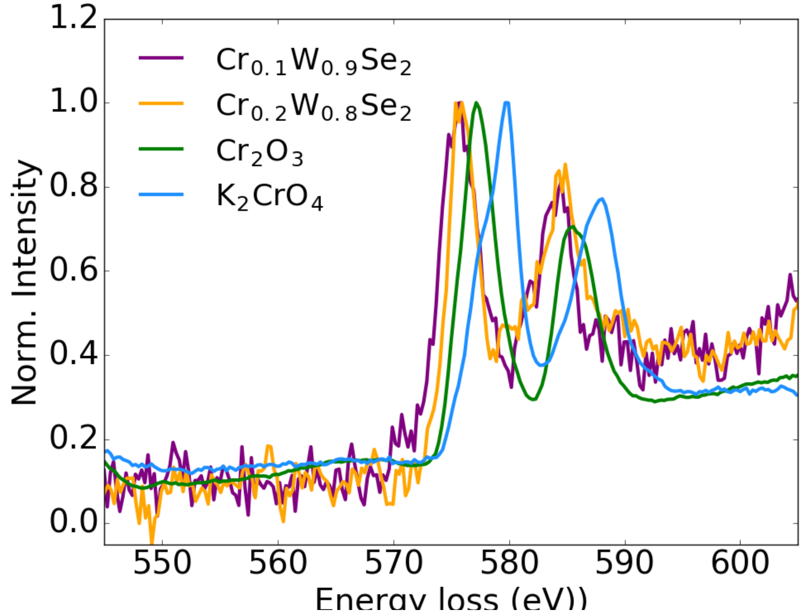
60 keV, 100 pA

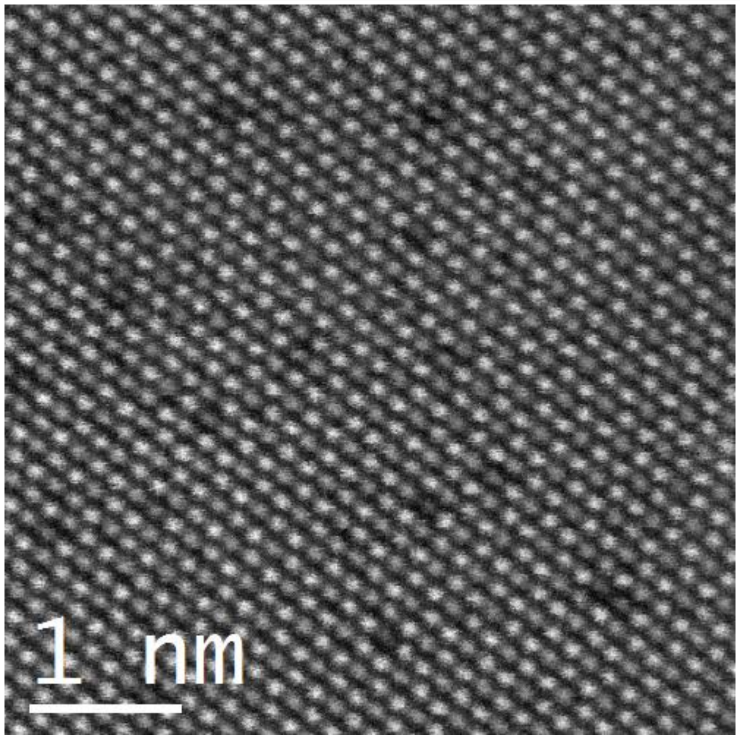
$10^{-2} \ldots 10$ ms dwell time (AlInGaN)

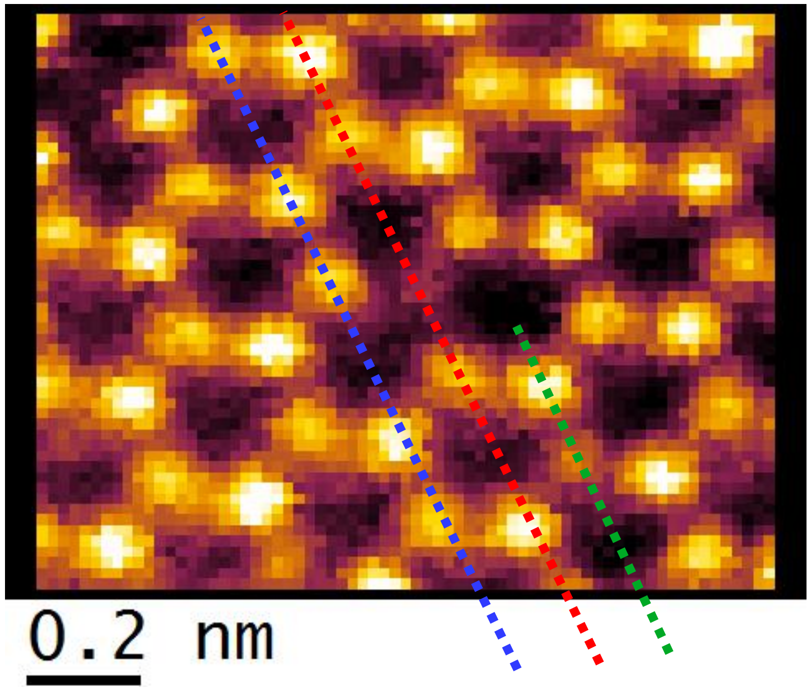
Some more examples in EELS and CL spectroscopy

Cr^{3+} doping in WSe_2

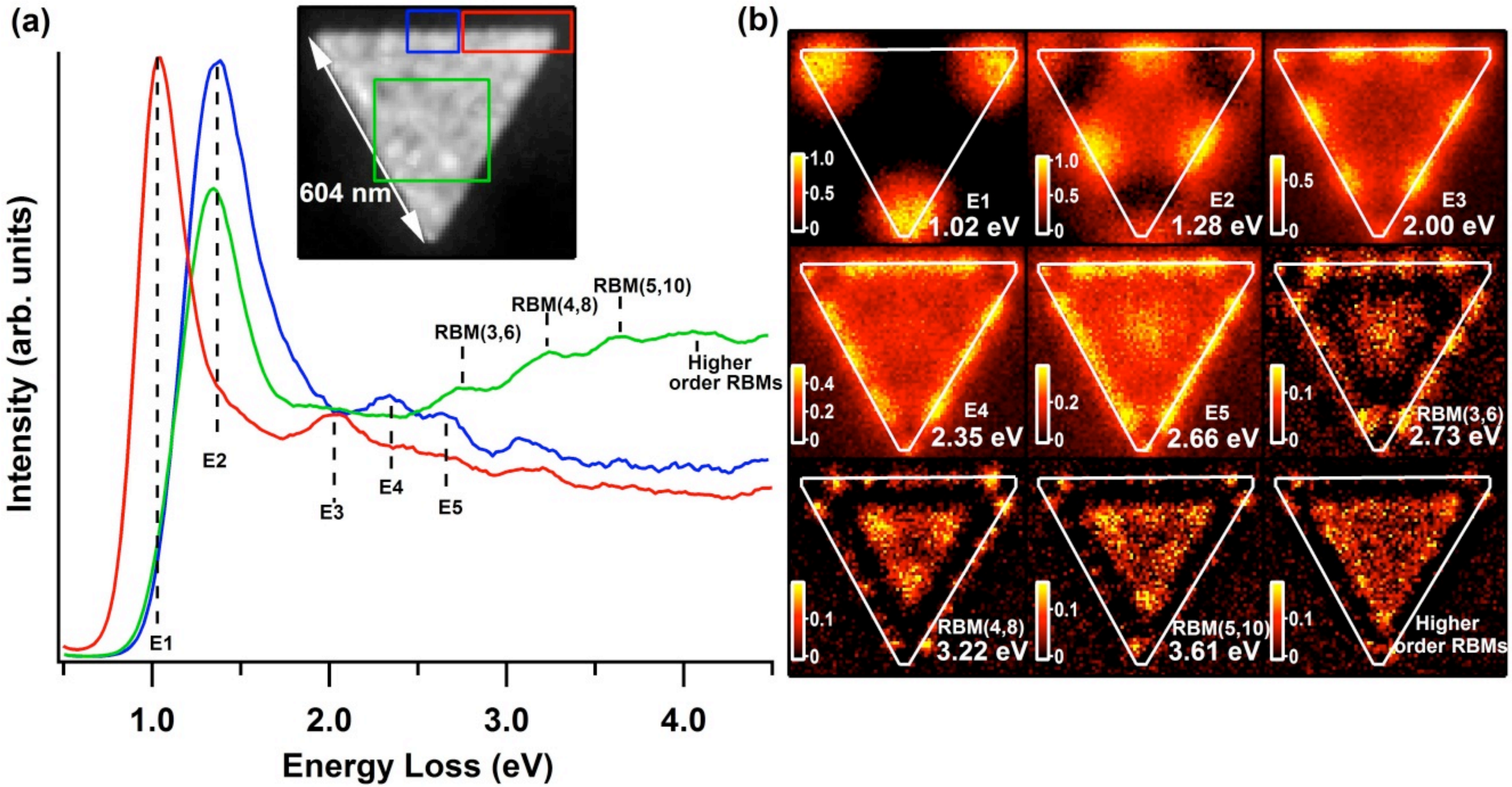




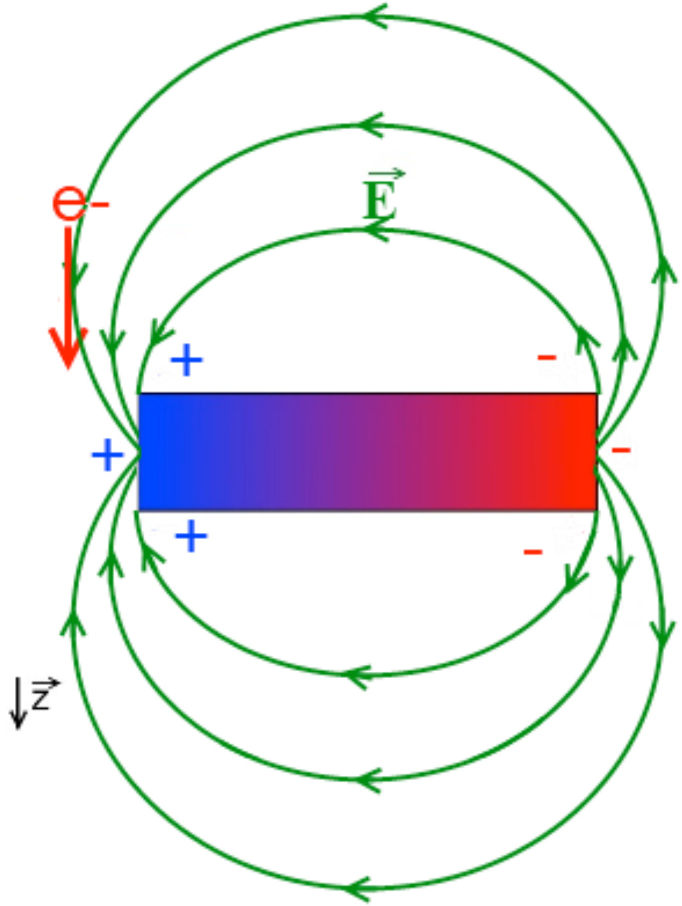




Mapping the plasmonic response (eigen modes) in individual nanoparticles

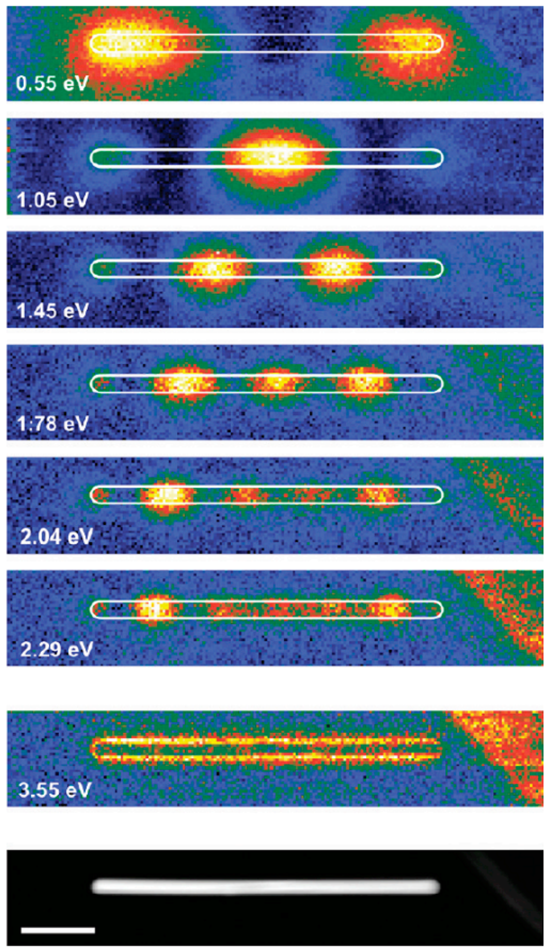


EELS for mapping EM fields
Probing a projected EMLDOS



One measure the **induced evanescent field**

The electron is loosing energy **only** when **the induced field on the antenna is parallel to its trajectory**

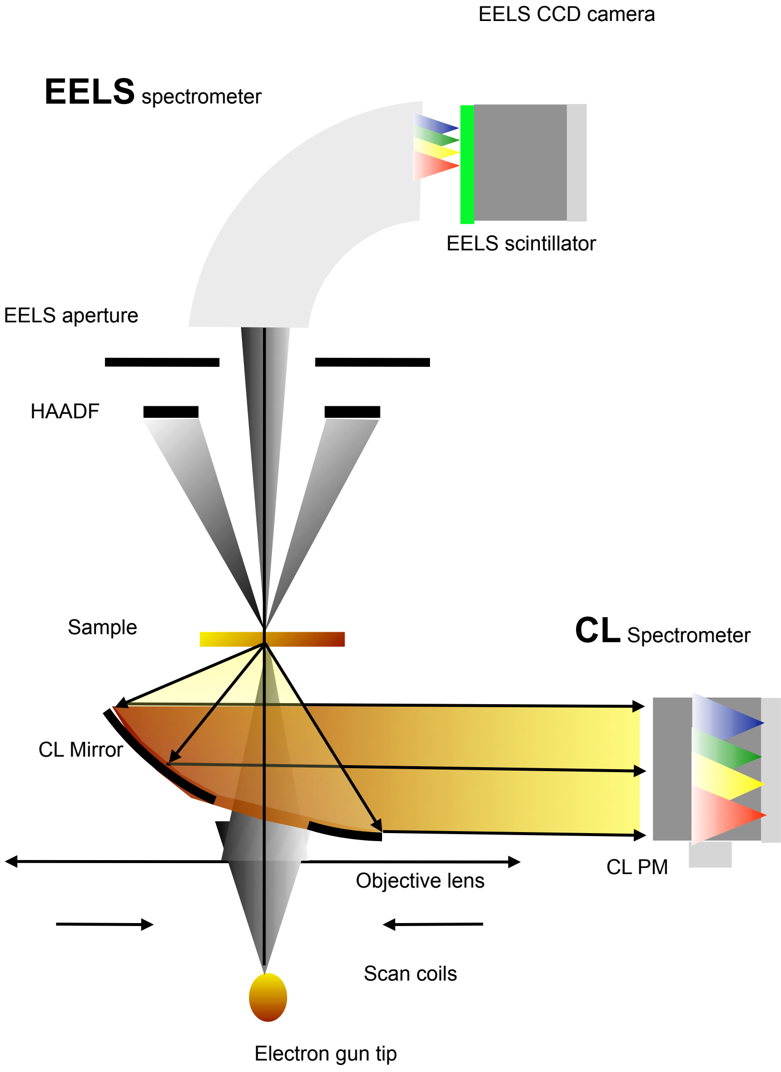


D. Rossouw et.al, Nanoletters (2011)

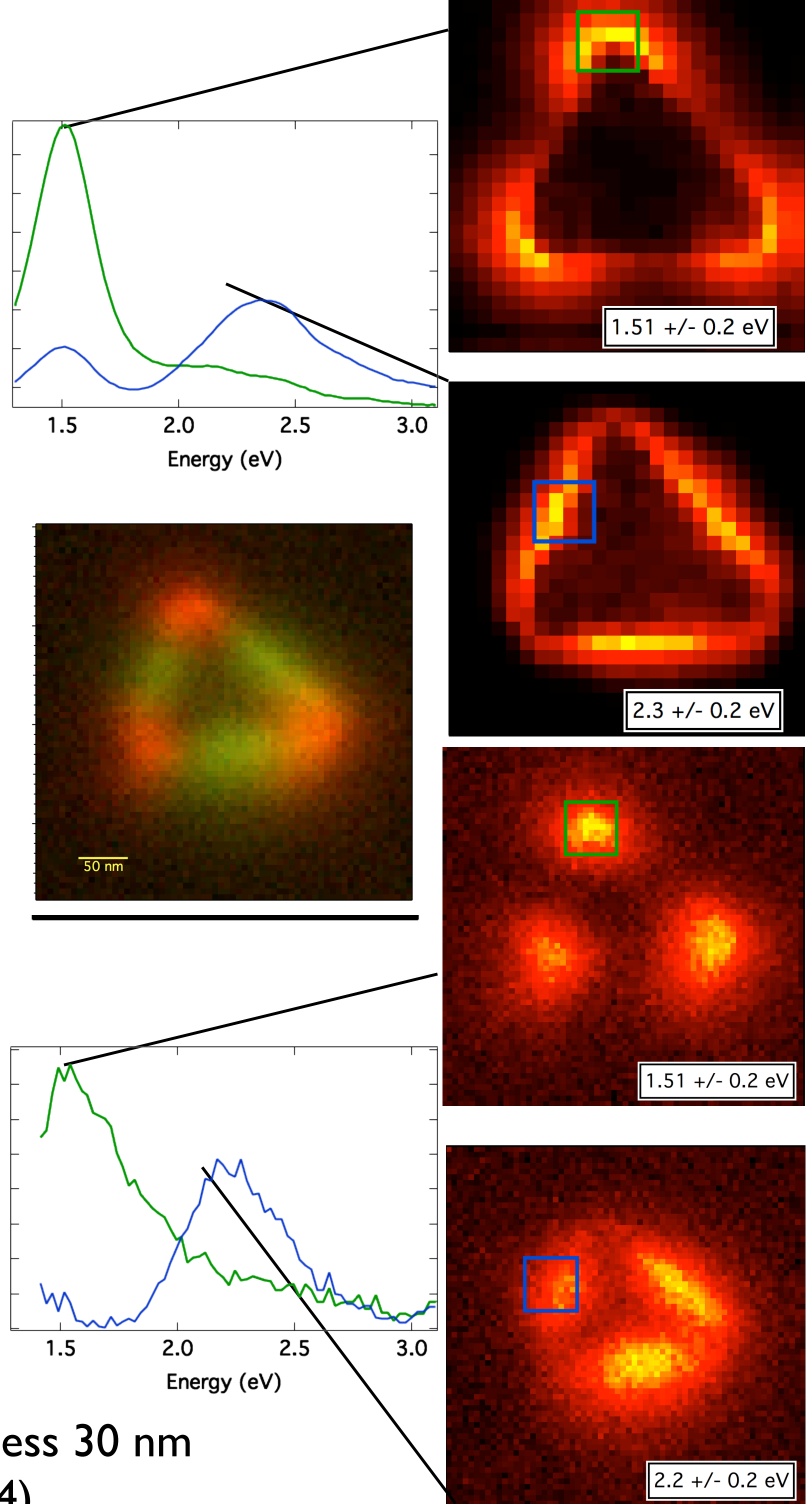
(1) spectrum(ω) ~ polarisability(ω)

(2) one measures the z-projected E.M. LDOS

Spectral Imaging with electrons



ΔE=300 meV
0.6<E<2000 eV
40-100 keV
ca 1 nm spatial resolution



40 X 40 pixels
Total SI acquisition time: < 80s
30 pA, 100 keV

64 X 64 pixels, Total SI acquisition time: < 4s
3 nA, 100 keV

Kociak, M. & Stéphan, O. *Mapping plasmons at the nanometer scale in an electron microscope*. Chem. Soc. Rev. 43, 3865–3883 (2014).

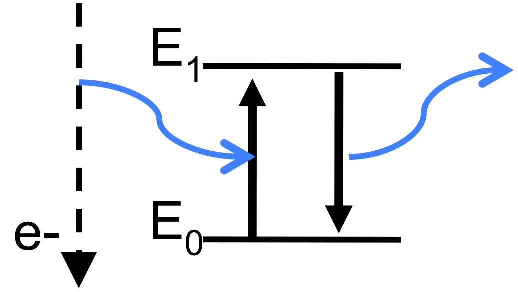
Silver nanoprism, 200 nm long thickness 30 nm
M. Kociak, Unpublished (2014)
coll. F. Schmidt, F. Hofer, J. Krenn, Graz Univ

Towards QUANTUM NANO-optics?

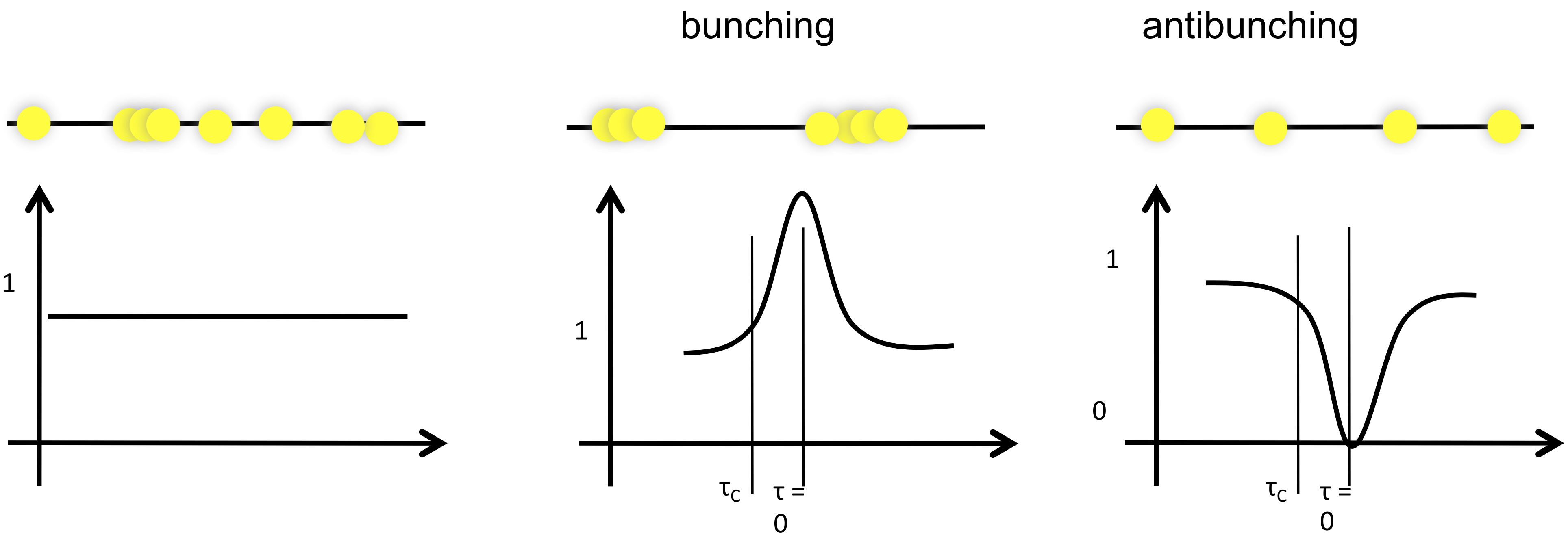
measuring the statistics of the emitted photons

Time auto-correlation (measuring photon statistics)

$$g^{(2)}(\tau) = \frac{\langle I(t)I(t+\tau)\rangle}{\langle I(t)\rangle\langle I(t+\tau)\rangle}$$



Quantum regime

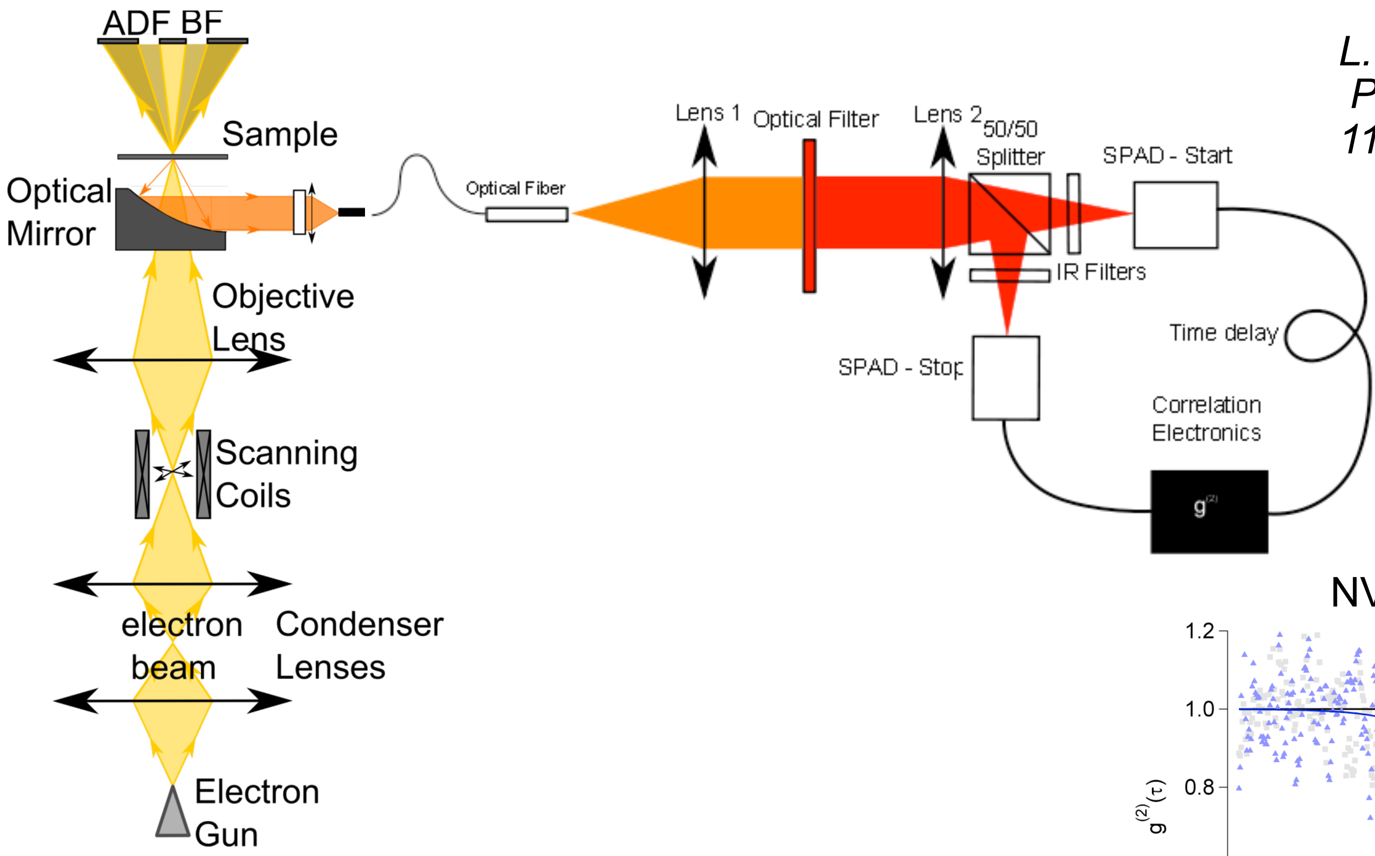


Poisonian (laser)

overpoisonian
(chaotic light)

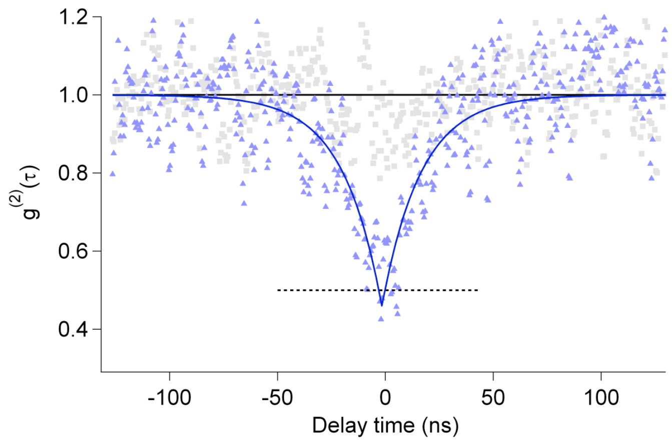
subpoisonian
(single photon emitters)

Quantum nanooptics in a STEM: light intensity interferometry (HBT)



L. H. G. Tizei and M. Kociak, Phys. Rev. Lett. 110, 153604 (2013).

NV centers in diamond

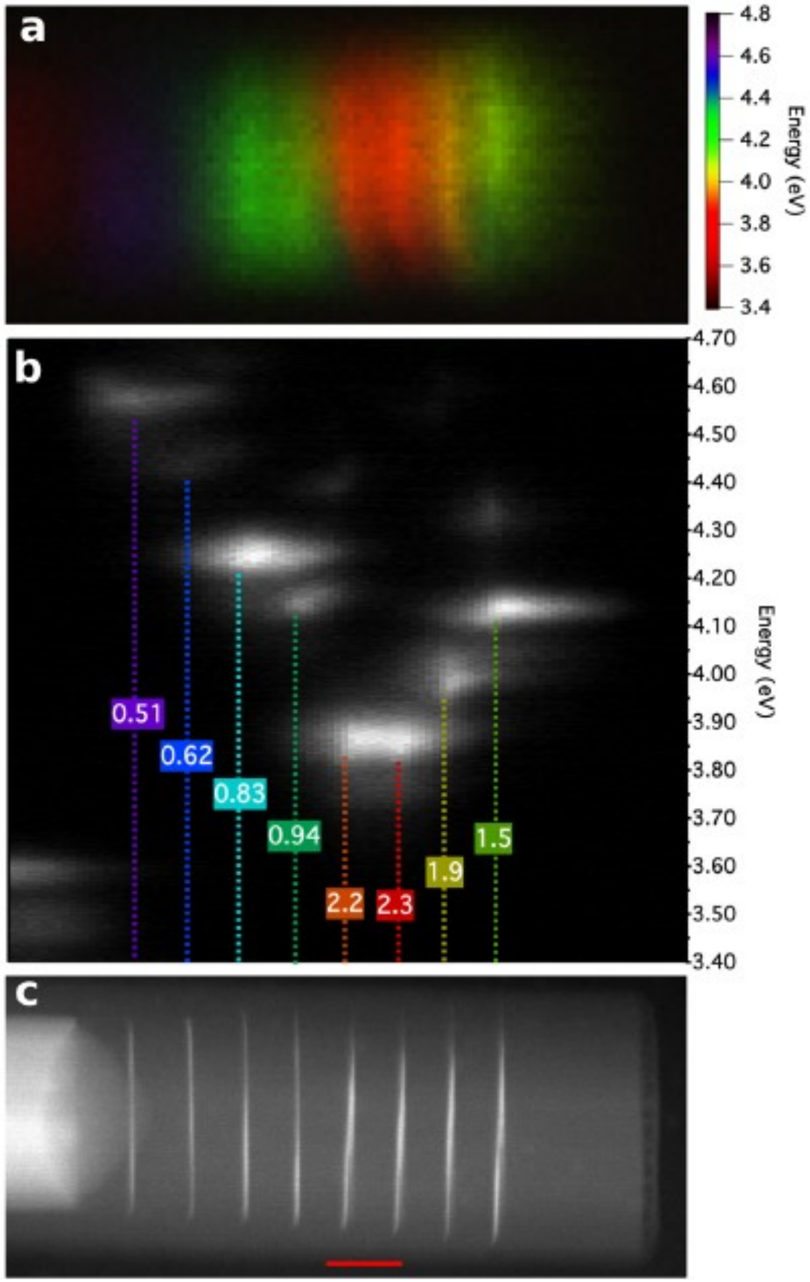


Exposure time 250s - 300 s

Typical count rate:
30 kcounts/s

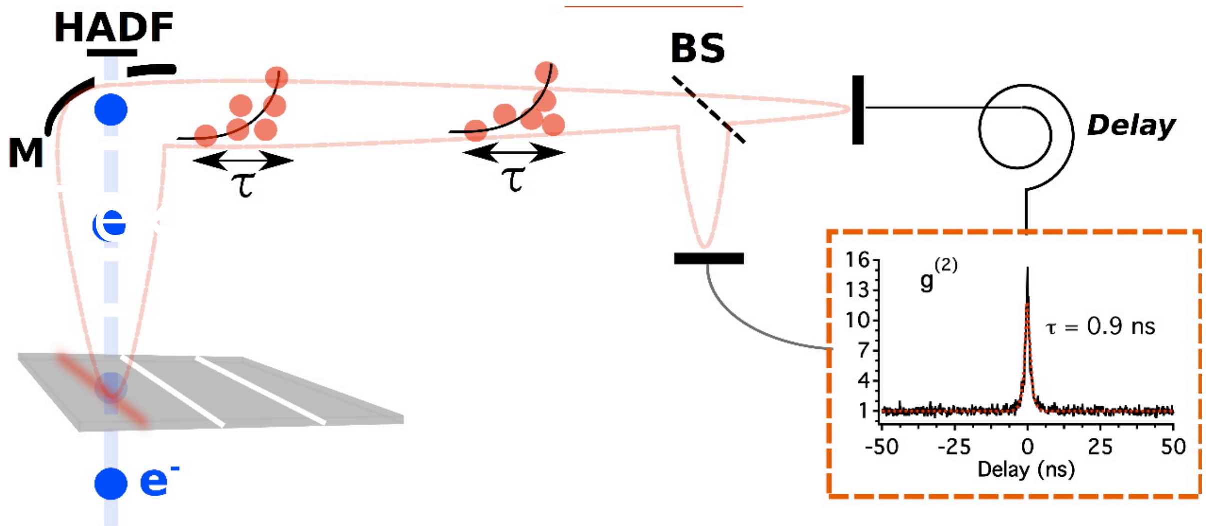
- Optimized CL system with **low excitation** needed, not a single photon can be lost!
- Stealing Quantum Optics knowledge (A. Beveratos et al., Quantum Communication, Computing, and Measurement 3 (2000))

Spatially resolved lifetime measurements in III-N heterostructures



Scale bar 20 mn

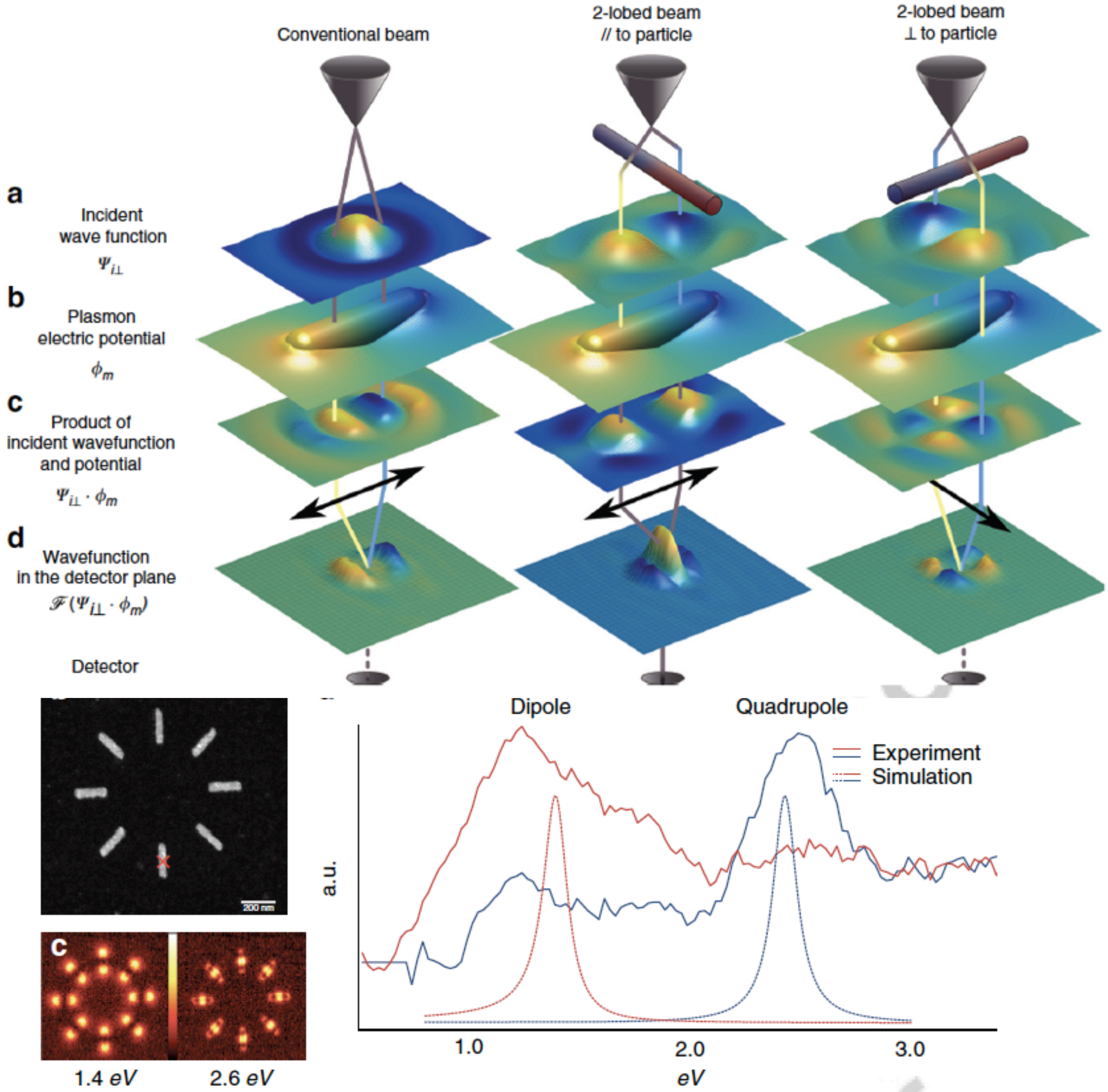
HBT experiments = a nice alternative to time-resolved PL (and CL)



Sub 15 nm ns lifetime measurements
In addition to the spatial resolution, it is fast: acquisition time tip. 30x faster than PL!

Meuret et al., ACS Photonics (2016)

Manipulating the phase of electron beams



Analogy with optical transitions in atomic physics

Selectively and directionally detect dipolar resonances in a plasmonic nanorod, analogously to what is currently possible with linearly polarized light

Some instrumental perspectives

1) Probe (in absorption and/or emission) localised plasmons, plasmon polaritons, excitons...

Routine simultaneous acquisition of EELS and CL signals

2) Push downward in the IR regime: map phonons, magnons, ...

Next generation of monochromators, how far can we go in terms of the probed energies?

3) Combine photons and electrons at the nanometer to perform « pump probe » experiments:
Non linear EELS, Electron Energy-Gain Spectroscopy, time-resolved experiments

Pulsed guns, synchronized e-/photon excitation/detection

4) Perform spatially-resolved NRIXS experiments:
probe non-dipolar transitions (d-d and others), tackle modern issues in Condensed Matter Physics

Explore the reciproqual space below 1 eV

Go to (very) low (controlled) temperatures

The STEM group at Orsay

J.M. Triscone and coll. Univ. Genève

C. Berger, W. de Heer, Giorgia Tech

A. Tejeda, A. Taleb-Ibrahimi, LPS, SOLEIL

A. Benito and W. Maser, ICB-CSIC, Zaragoza

L. Liz-Marzan and coll. San Sebastian

J. Garcia de Abajo, CSIC Madrid

M. Tchernicheva, F. Julien and coll. IEF, Univ. Paris-Sud

J.Plain and coll., UT Troyes

F. Schmidt, F. Hofer, J. Krenn, Graz Univ