Imaging plasmons in an ultrafast Transmission electron microscope

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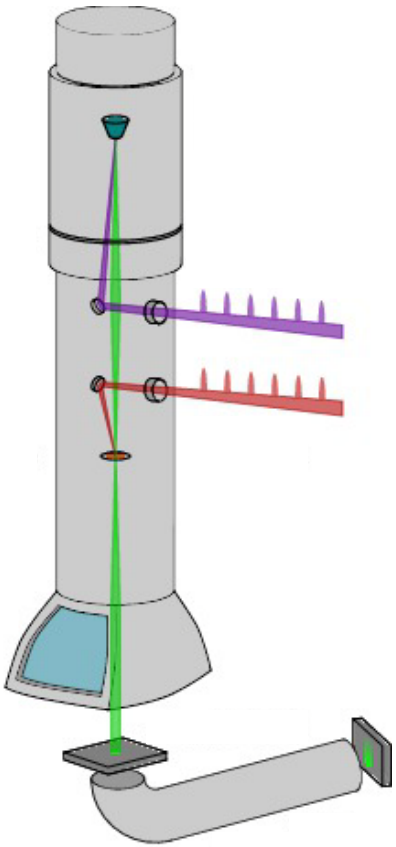
Outline

- Motivation
  - ✓ Visualizing photonic near-fields
  - ✓ Testing fundamental aspects of quantum mechanics
- Instrumentation and Approach
  - ✓ Time-resolved electron microscopy
  - ✓ Setup characteristics and specifications
- Photon-Induced Near-Field Electron Microscopy
  - ✓ The PINEM effect
  - ✓ PINEM spectroscopy: photon quanta
  - ✓ PINEM imaging: plasmonic fields
- Multi-dimensional imaging
  - ✓ 'Hybrid' PINEM: simultaneous energy-space imaging



Testing the wave/particle duality

Black-body radiation theory 1901

(wave and particle aspects both necessary)

Photoelectric effect 1905

(curiosity: quantized field not necessary)

Lamb&Scully, "The photoelectric effect without photons", 1968

Particle-wave duality for electrons

demonstration, Tonomura *Am. J. Phys.* **57**, 117–120 (1989).

Particle-wave duality for photons

Single photons sources needed

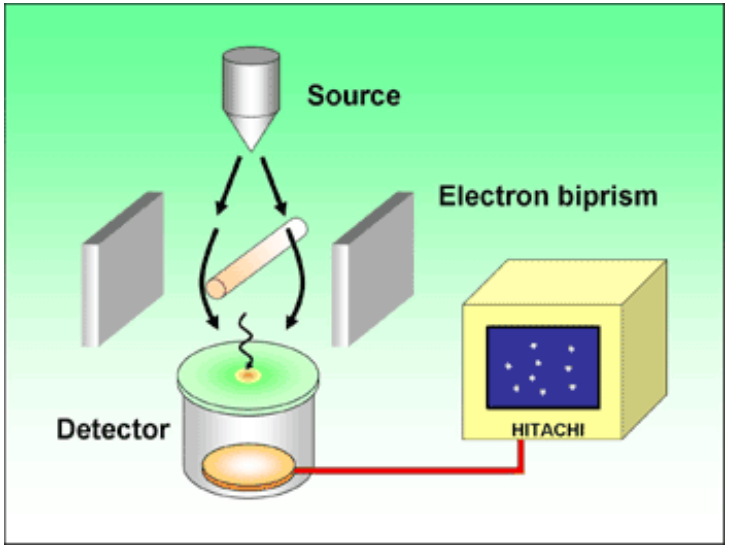
1988-now Aspect and others, delayed choice experiments.

Simultaneous observation of wave/particle duality?

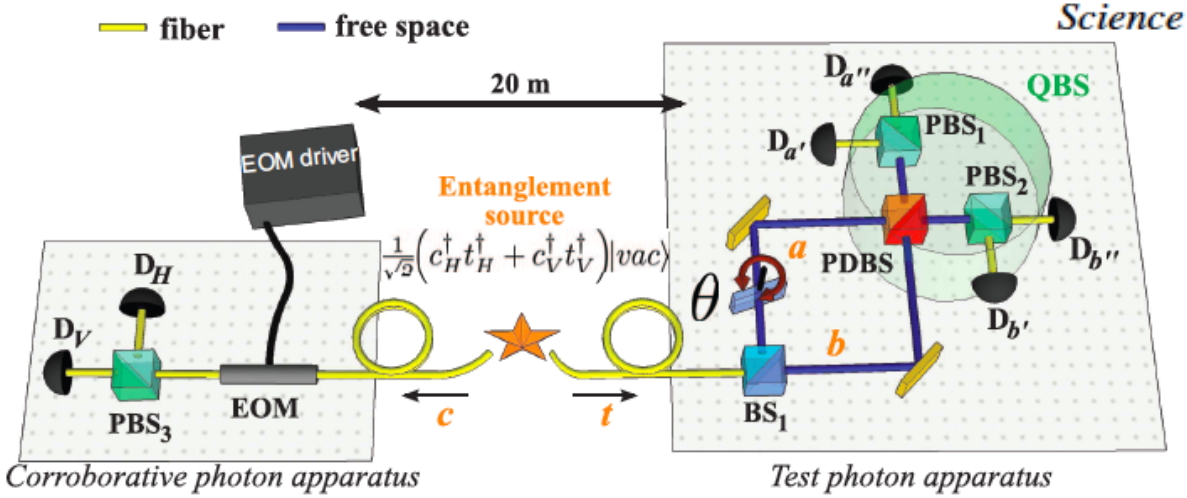
Low flux, double slit experiment

"Observing the Average Trajectories of Single Photons in a Two-Slit Interferometer". S. Kocsis et al., Science 332, 1170 (2011).

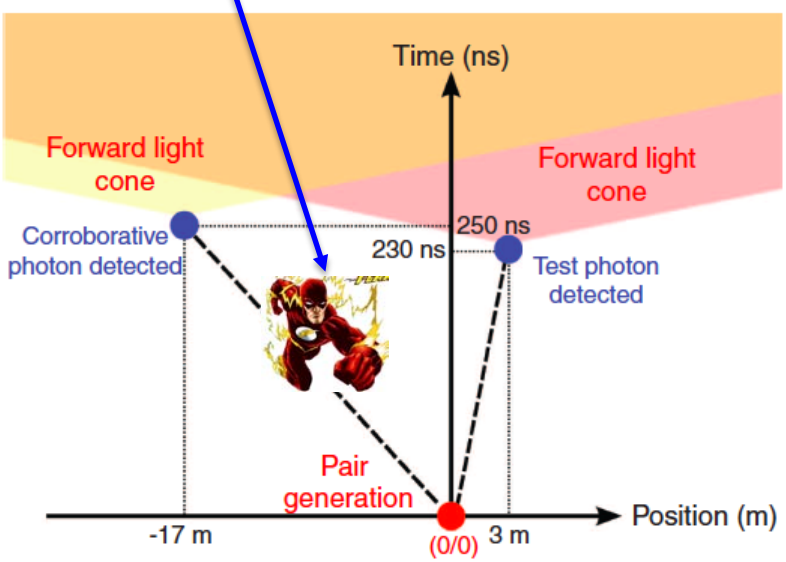
Davidovic and Sainz, comment in EPN (2013)



Delayed choice experiments



Hidden variable



Polarization entangled photons

Test: detected in the interferometer

Corroborative: tells if it behaved as wave or particle before any information could be exchanged via hidden variables

Hidden variable and Bell's inequality

Hidden variables:

Can a 007 particle "sense" the experiment and tell the photon what to do?

- What if hidden variables travel faster then the speed of light?
- In a delayed choice experiment particle and wave behavior are not recorded simultaneously

Bell's theorem (or inequality):

$p(a,c) - p(b,a) - p(b,c) \leq 1$ deals with correlation of particles that interacted and have been separated

X

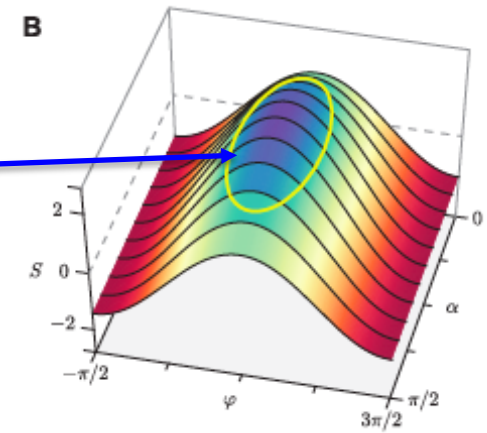
Y

Z

3 coin flips X,Y,Z highly correlated:

if X,Y coincide 99% of times
if Y, Z coincide 99% of times
X, Z have to give the same answer 98% of times

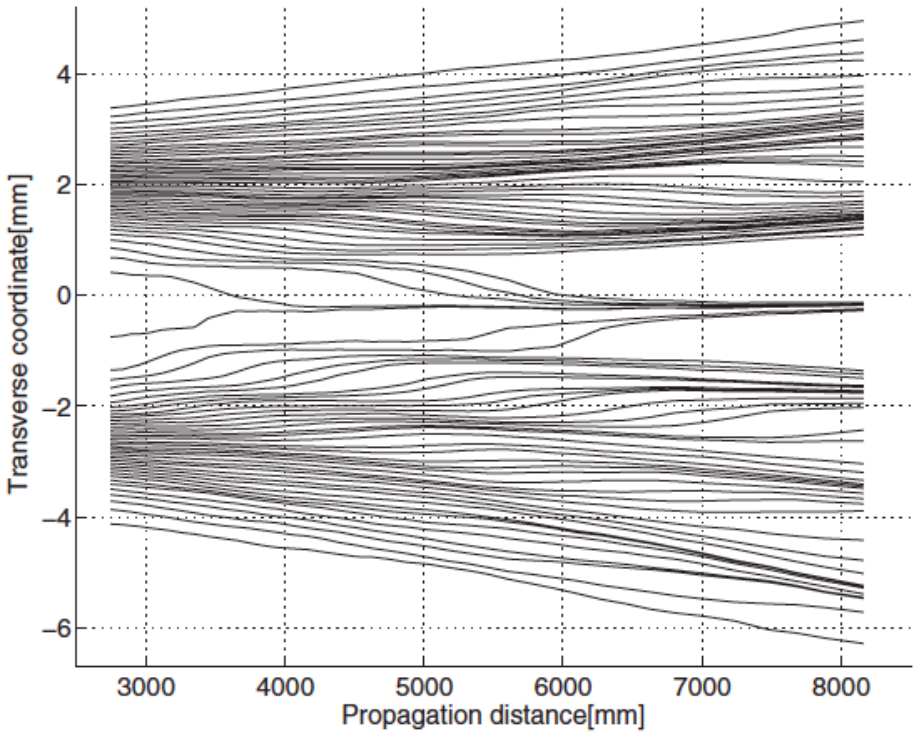
Particle-wave duality causes a violation of Bell's inequality in the measurement of correlated photons



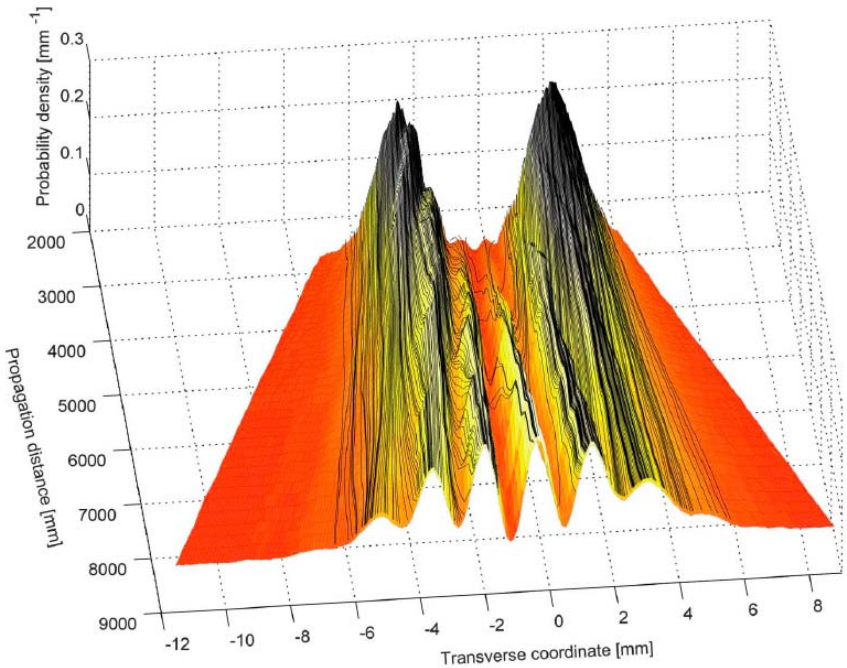
We want to have a visual and simultaneous demonstration of the two aspects, delayed choice experiments cannot do that

Low-flux double slit experiment

Observing the Average Trajectories of Single Photons in a Two-Slit Interferometer
Sacha Kocsis *et al.*
Science **332**, 1170 (2011);
DOI: 10.1126/science.1202218



Single particles trajectories



Interference pattern

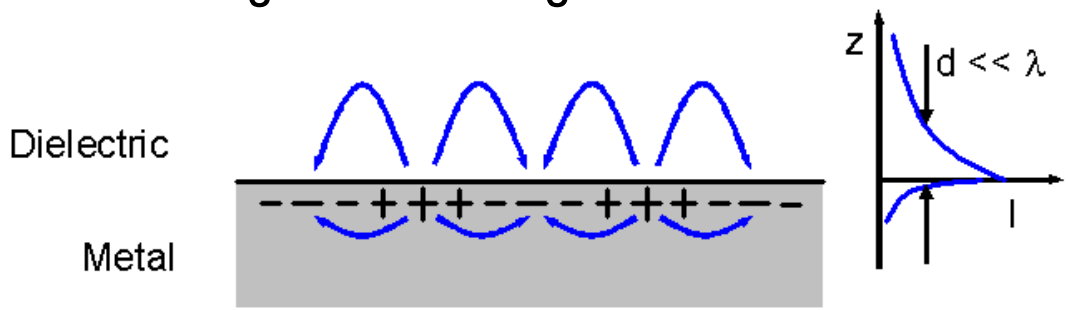
Is it the final word?

In that sense, even though the trajectories reconstructed from the experiment cannot be associated with the paths followed by individual photons, but with electromagnetic energy streamlines, the experiment constitutes an important milestone in modern physics.

Visualizing photonic near-fields

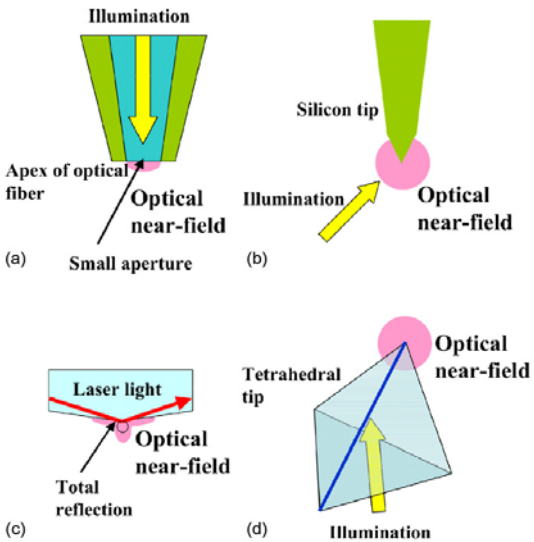
Nanoscale demands the use of sub-wavelength confined light:

- Evanescent optical fields
- Surface plasmon polaritons
- No coupling to the far-field



Scanning Near-field Optical Microscopy (SNOM/NSOM)

- Probe proximity to sample >> λ
- Inherent limitations:
  - spatial resolution (~10nm)
  - point-by-point probing
  - Time-resolution



Alternative method?

Y. Oshikane *et al.*, *Sci. and Technol. Adv. Mat.* **8**, 181 (2007)

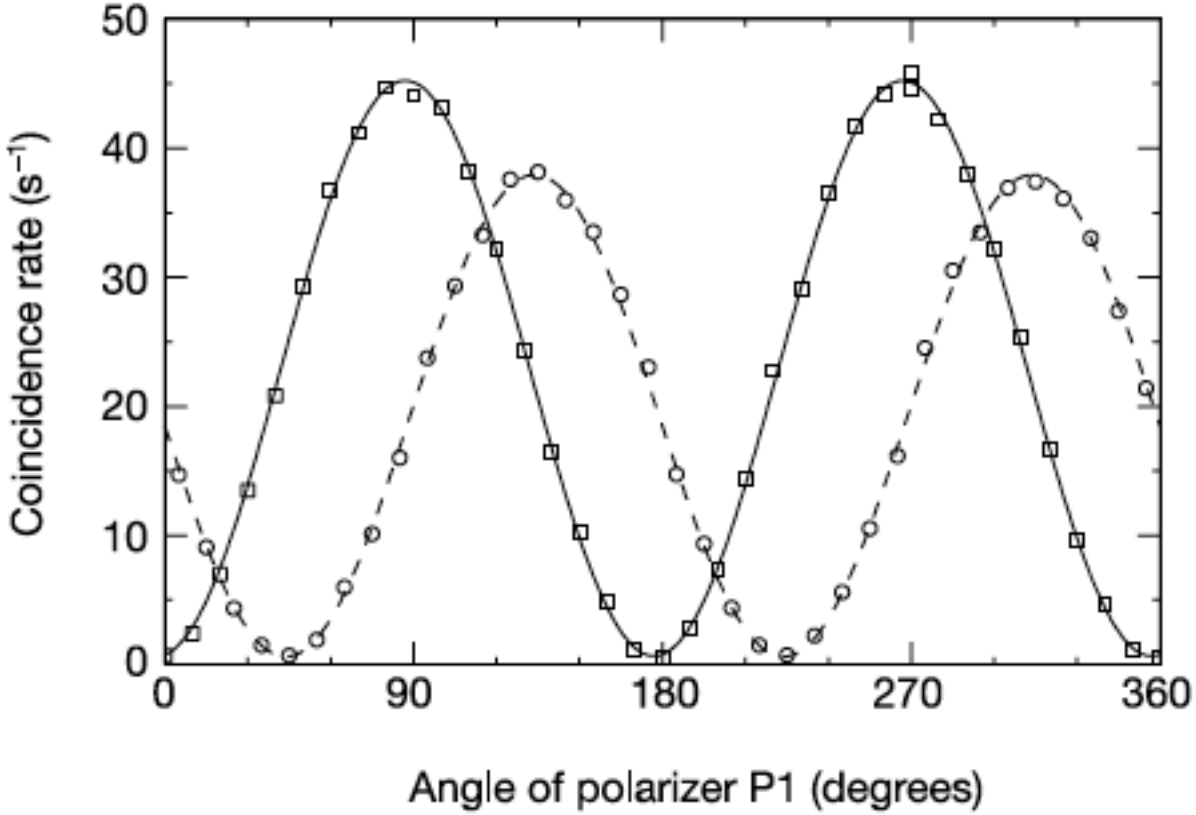
Quantum properties of plasma waves

Di Martino, G. *et al.* Quantum statistics of surface plasmon polaritons in metallic stripe waveguides. *Nano Lett.* **12,** 2504–2508 (2012).

Akimov, A. V. *et al.* Generation of single optical plasmons in metallic nanowires coupled to quantum dots. *Nature* **450,** 402–406 (2007).

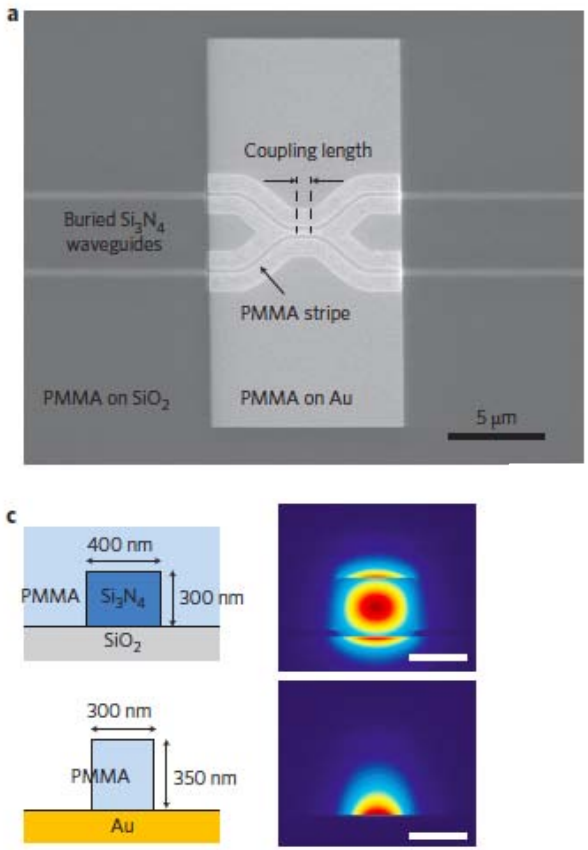
Huck, A. *et al.* Demonstration of quadrature-squeezed surface plasmons in a gold waveguide. *Phys. Rev. Lett.* **102,** 246802 (2009).

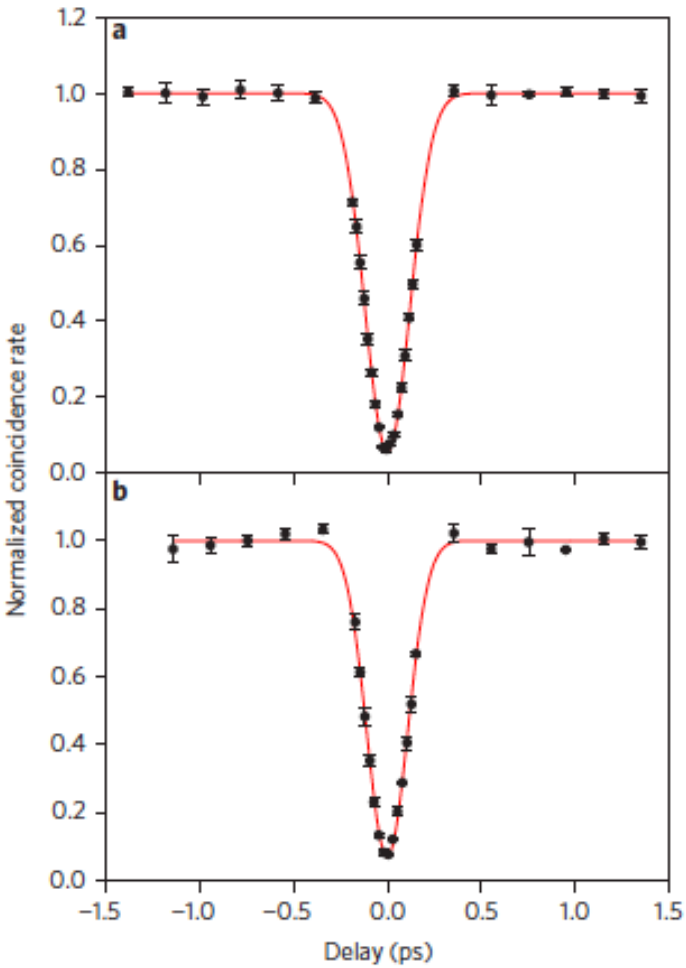
Plasmons entanglement



Altewischer, E., van Exter, M. P. & Woerdman, J. P. Plasmon-assisted transmission of entangled photons. *Nature* **418**, 304–306 (2002).

Two-plasmons quantum interference





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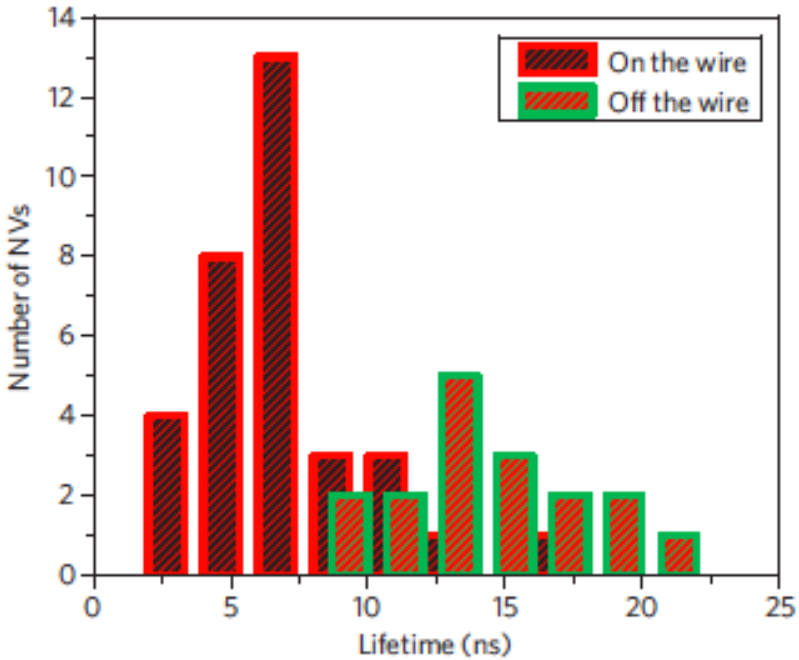
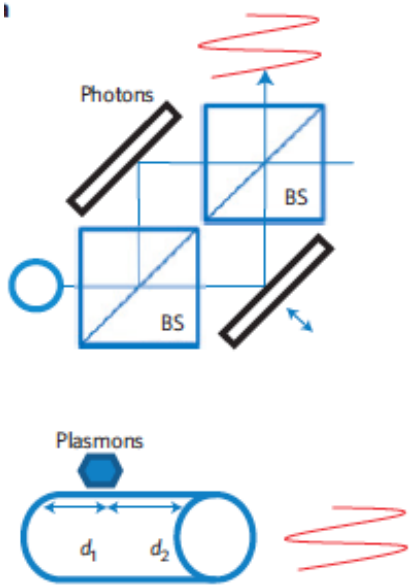
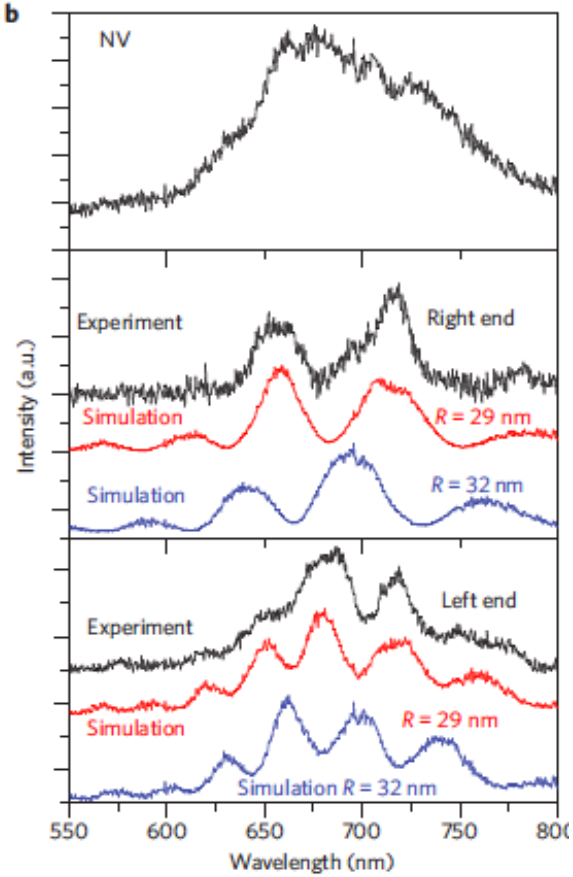
Wave-particle duality of single surface plasmons







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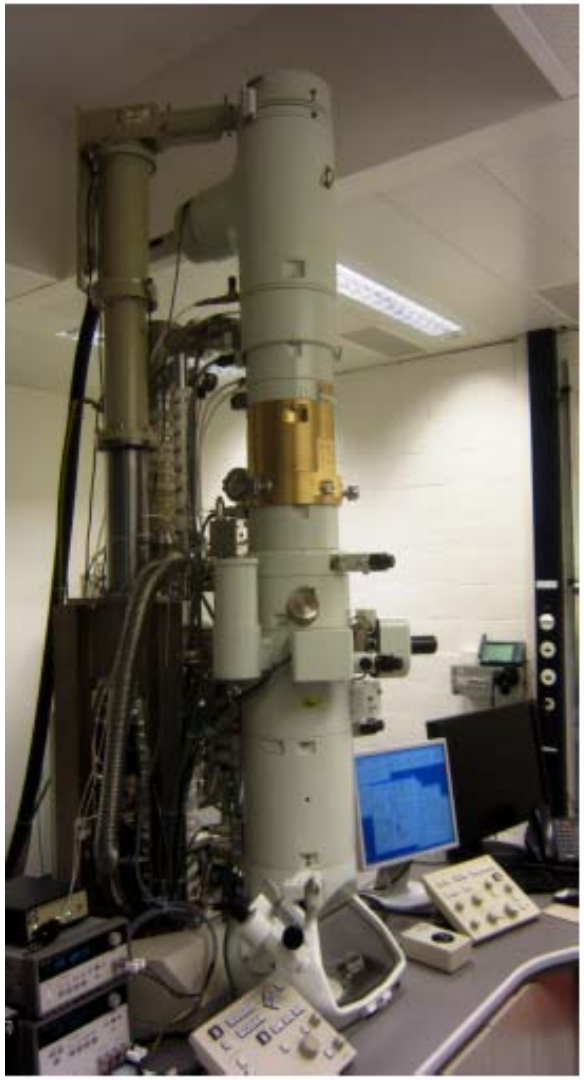
Instrumentation

Use electrons to probe the near-field

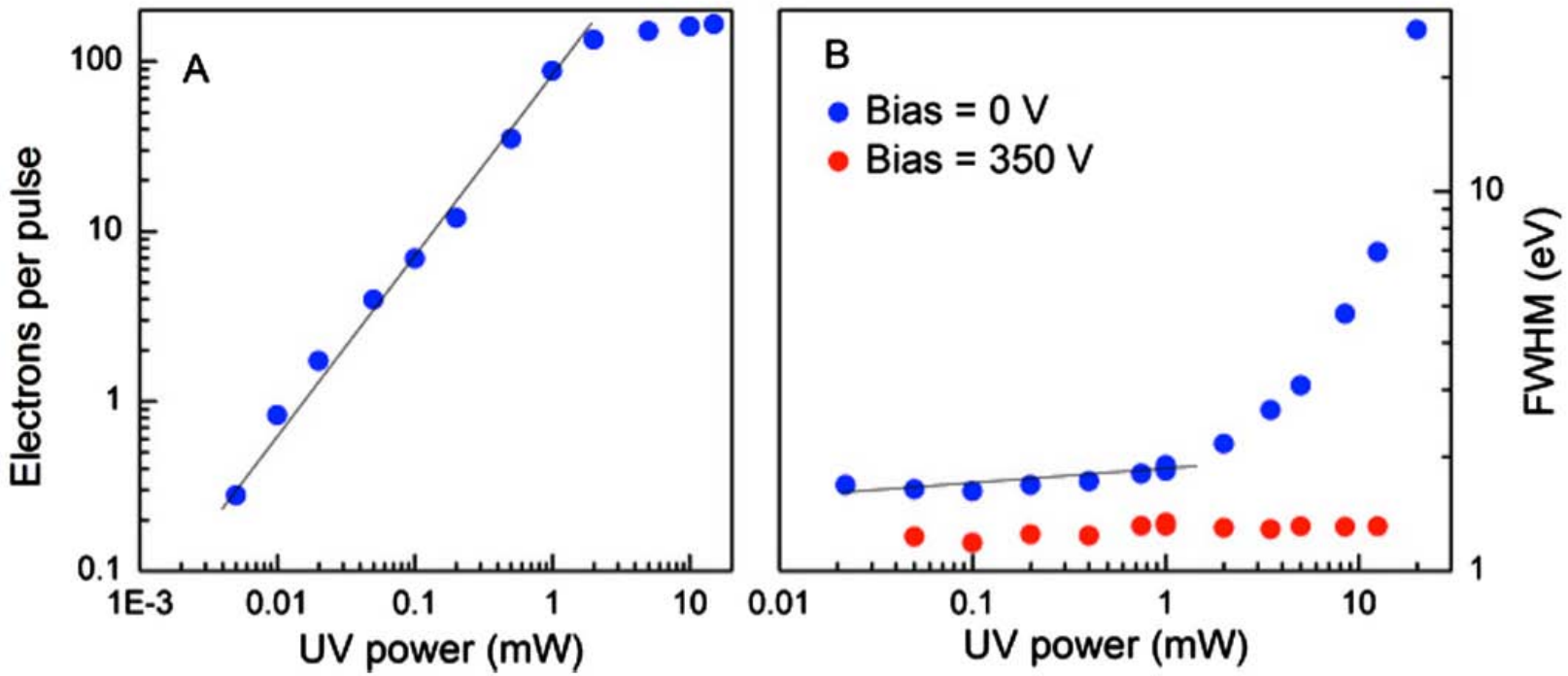
time-resolved Transmission Electron Microscope

- Modified 200 keV Jeol JEM-2100
- Optical port for specimen excitation
- Optical port for photo-electron generation
- Extra 'C0' electrostatic lens
- Electron Energy Filter (GATAN Quantum GIF) for energy-filtered spectroscopy



L. Piazza *et al.*, *Chemical Physics* **423**, 79 (2013)

Single electron microscopy

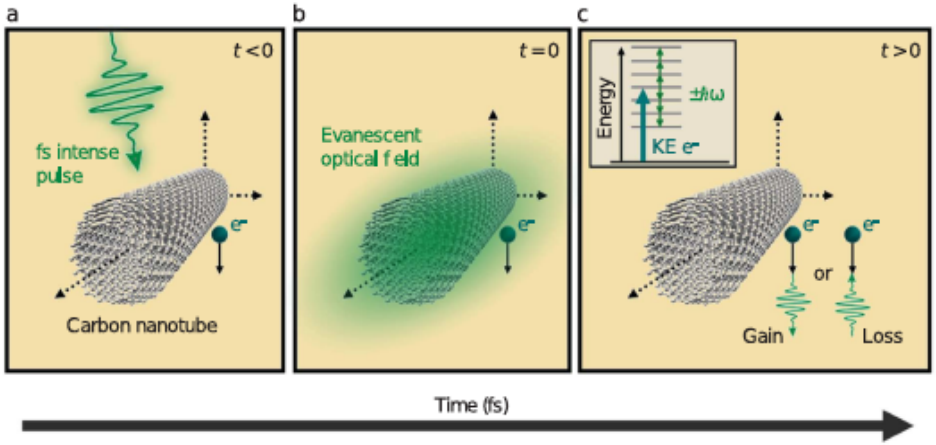


Statistically less then one electron per pulse are needed for optimal time and energy resolution

Photon-Induced Near-field Electron Microscopy (PINEM)

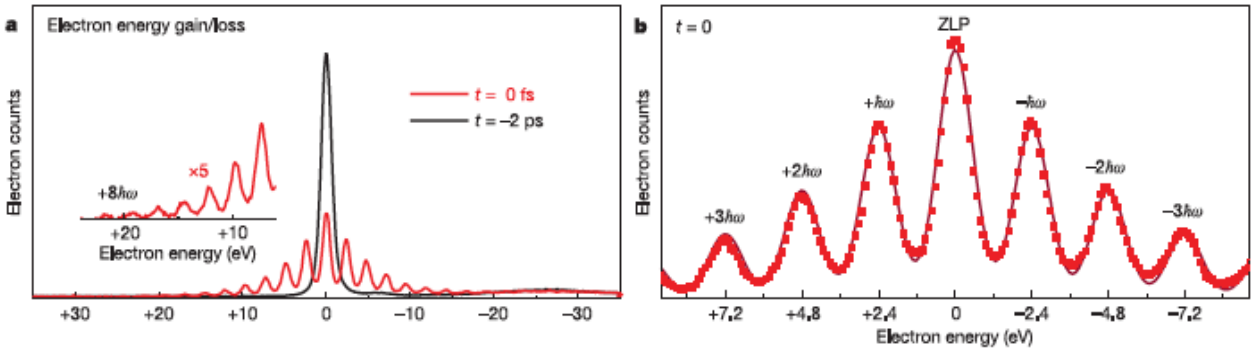
Pump-probe TEM technique

- fs optical pump
- fs electron probe



Energy exchange

- electrons gain/lose photon quanta
- structure-mediated

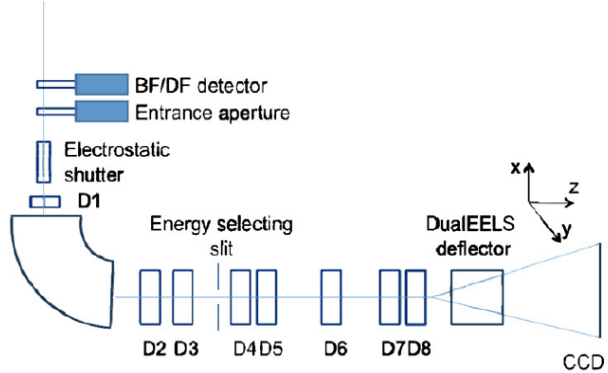


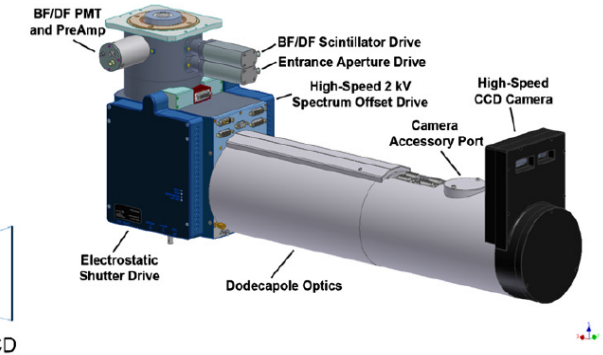
B. Barwick *et al.*, *Nature* **462**, 902 (2009)

PINEM

PINEM:

- energy-filtering using GIF

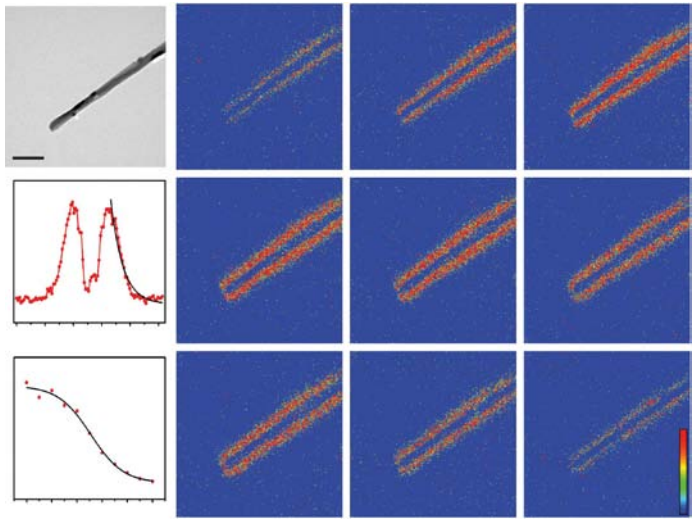




A. Gubbens *et al.*, *Ultramicroscopy* **110**, 962 (2010)

- Image using only 'inelastic' electrons

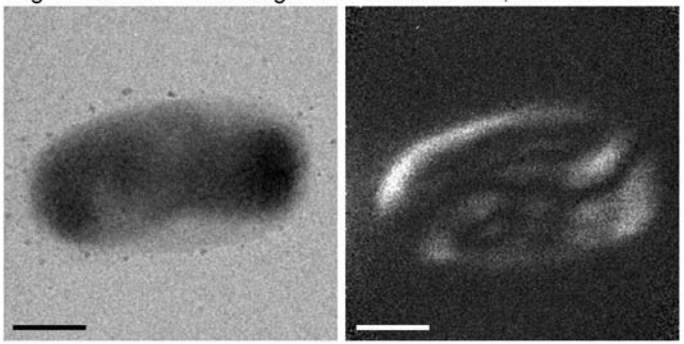
Carbon nanotubes



B. Barwick *et al.*, *Nature* **462**, 902 (2009)

E. Coli

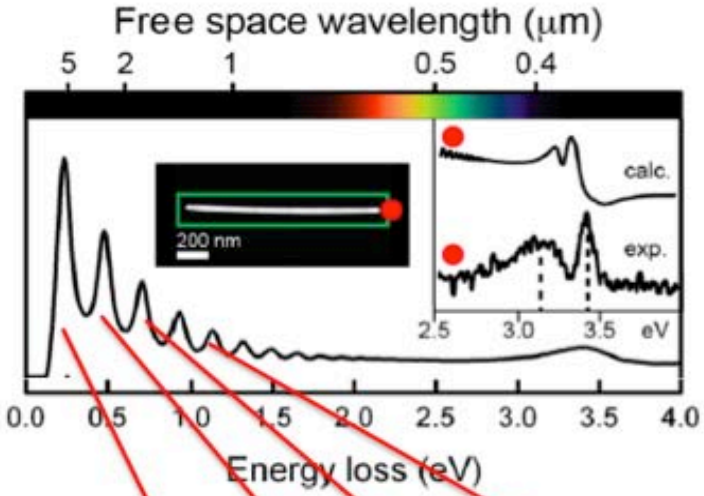
Bright-field and PINEM images of whole unstained, unfixed *E. coli*



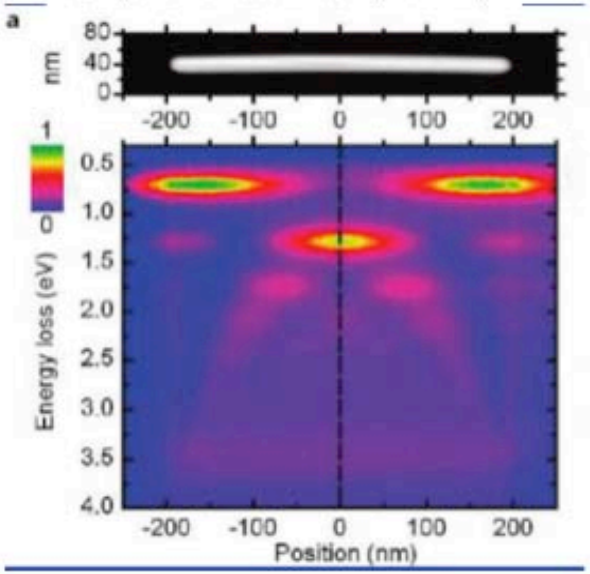
D. Flannigan *et al.*, *PNAS* **107**, 9933 (2010)

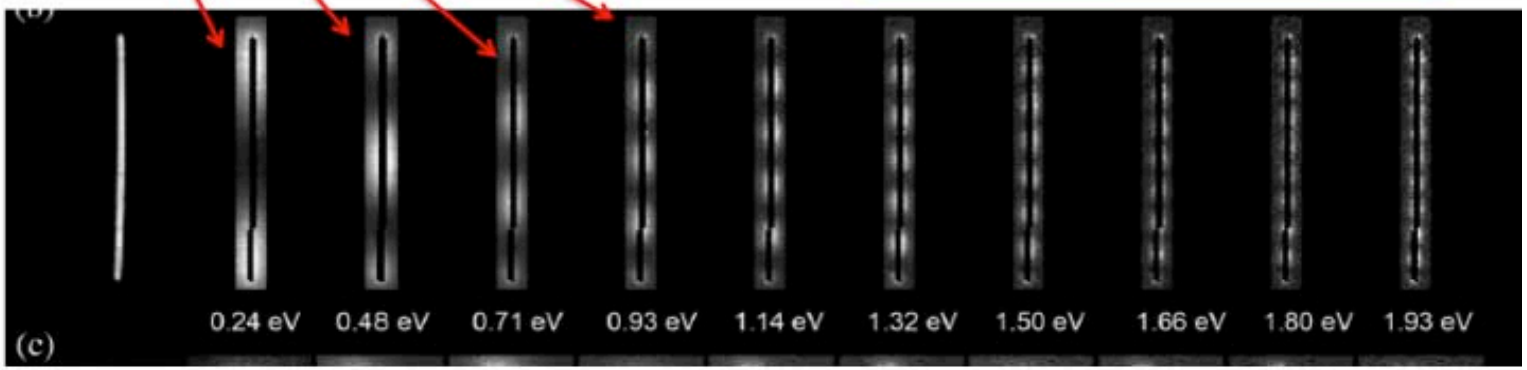
PINEM vs Spectral imaging

A (PRL 110, 066801 (2013))



C (NanoLett 1499 (20122).

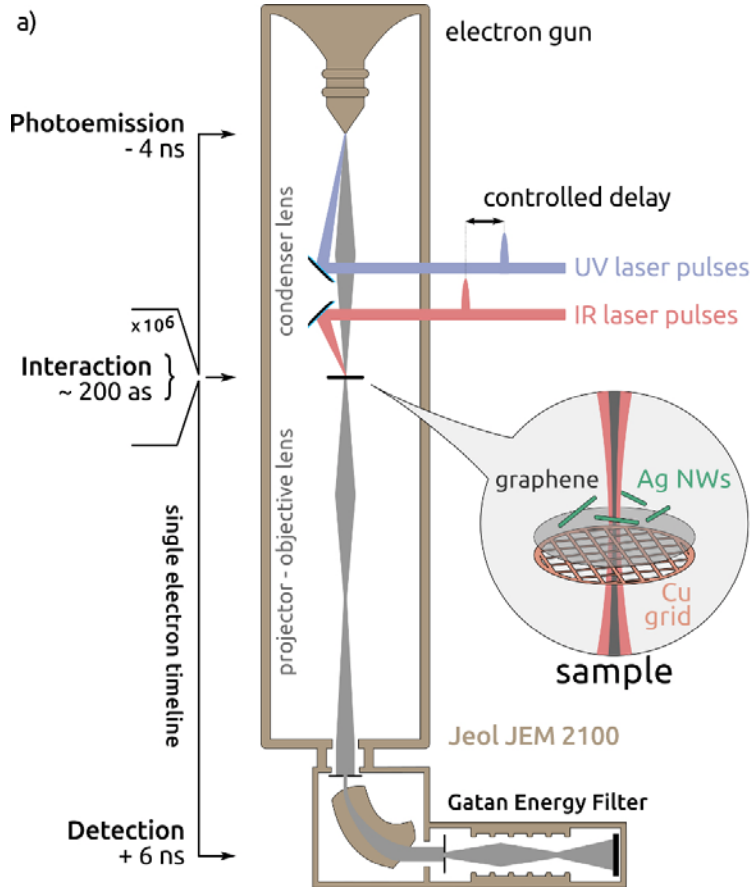


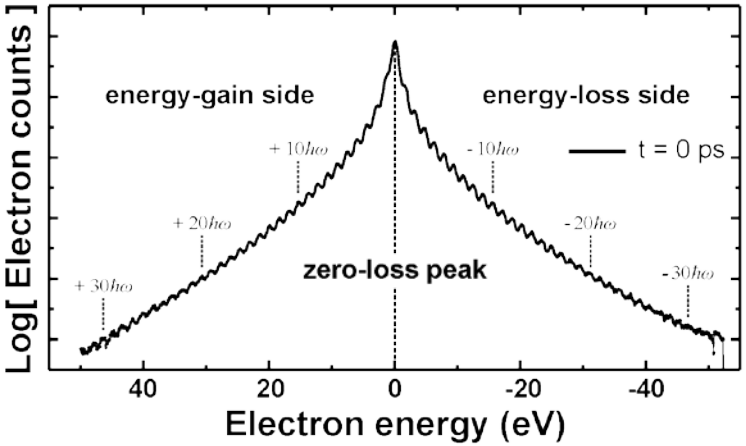


B (PRL 110, 066801 (2013))

PINEM

PINEM spectroscopy mode: Time-resolved Electron Energy Loss Spectroscopy (EELS)

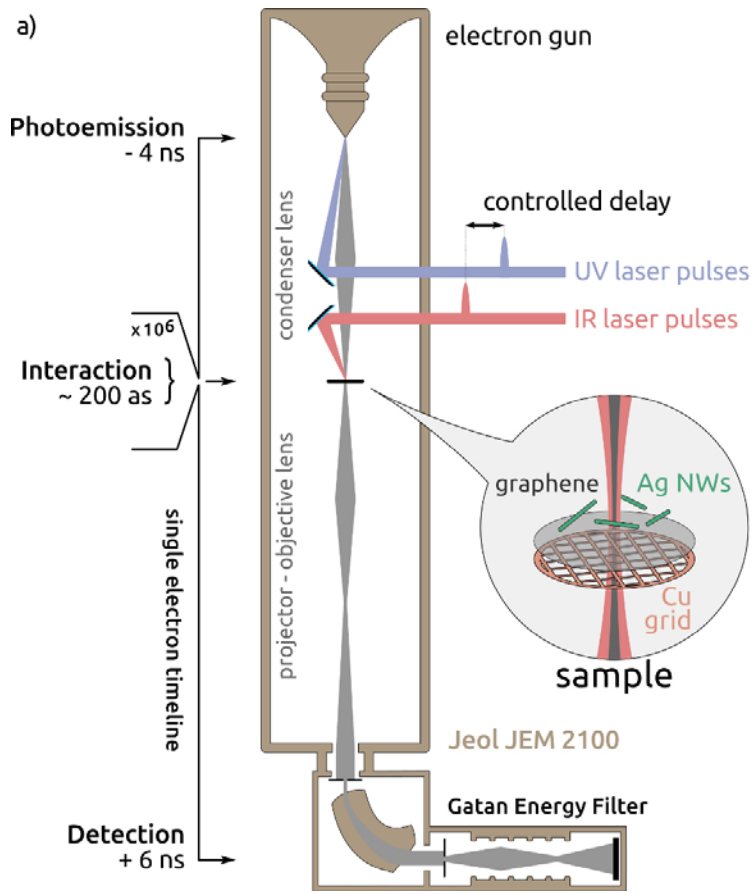


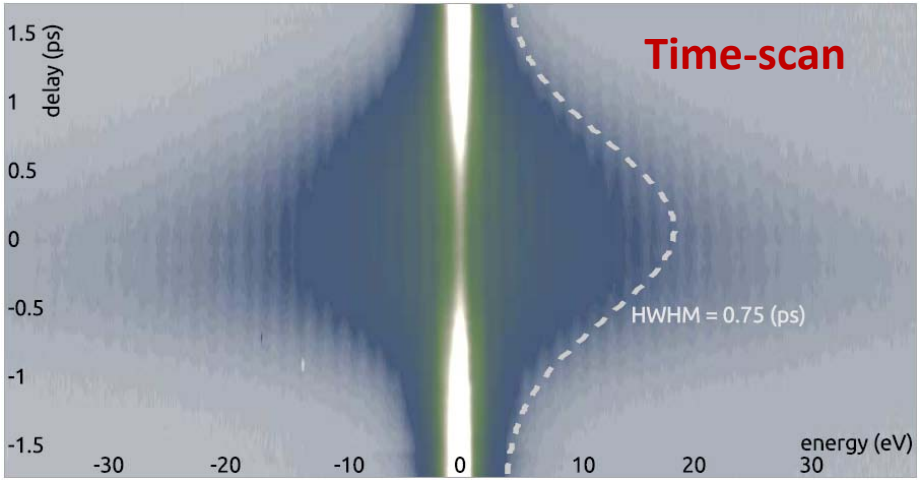


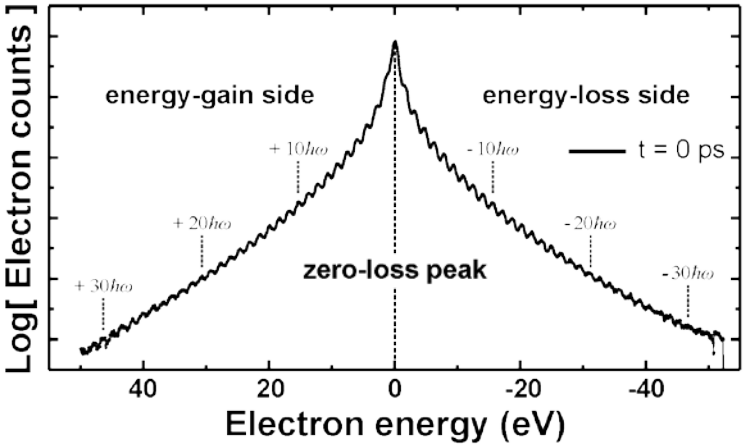
Energy exchange of over 30 photon quanta

PINEM

PINEM spectroscopy mode: Time-resolved Electron Energy Loss Spectroscopy (EELS)







Energy exchange of over 30 photon quanta

PINEM

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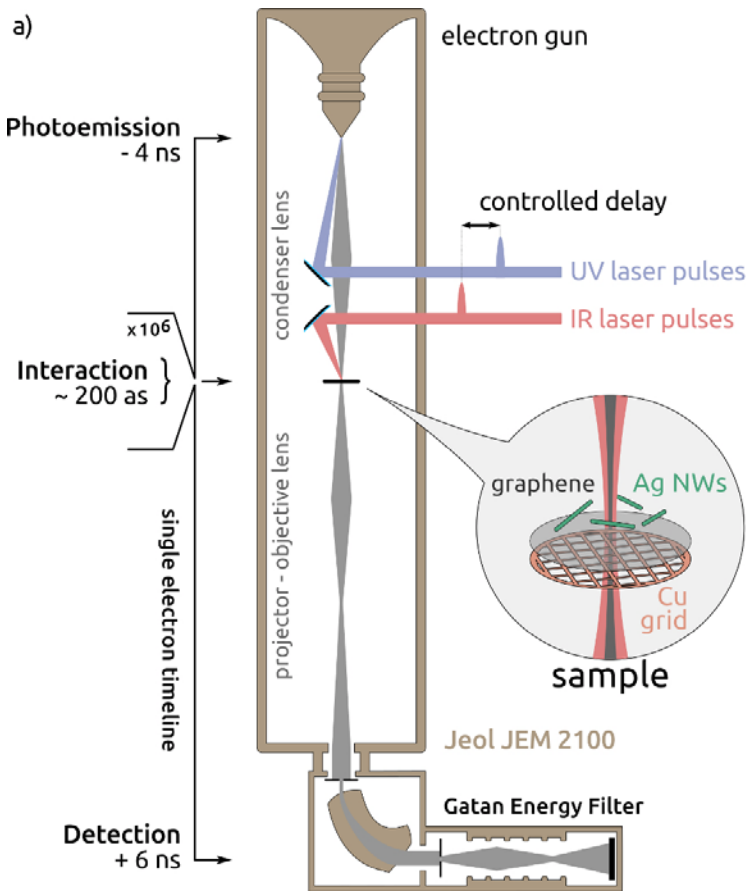
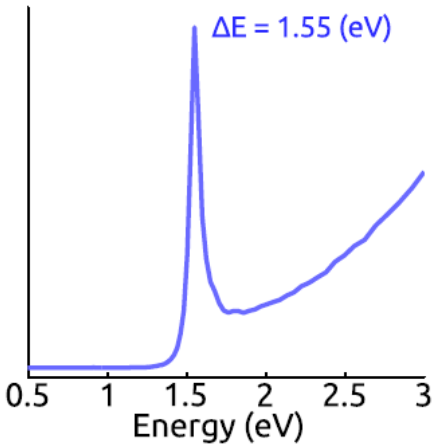
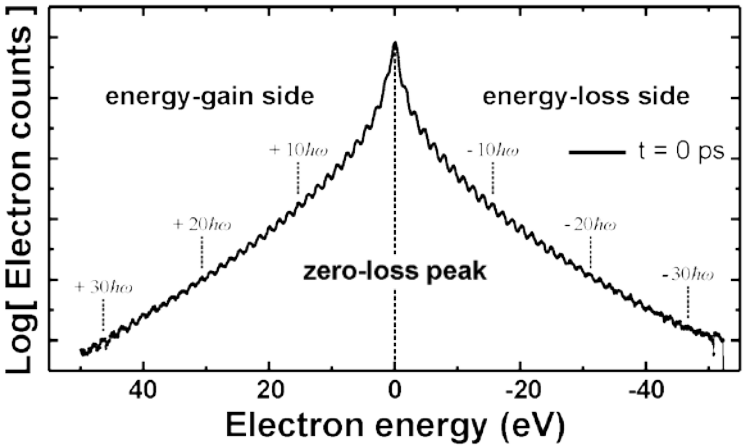




Fourier transform



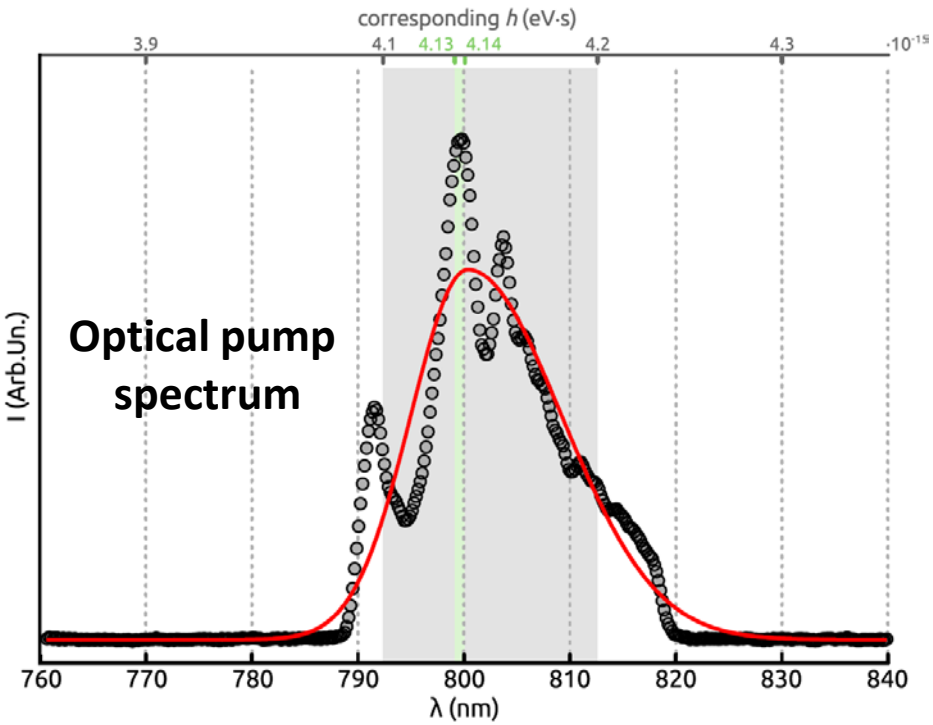
Energy exchange of over 30 photon quanta

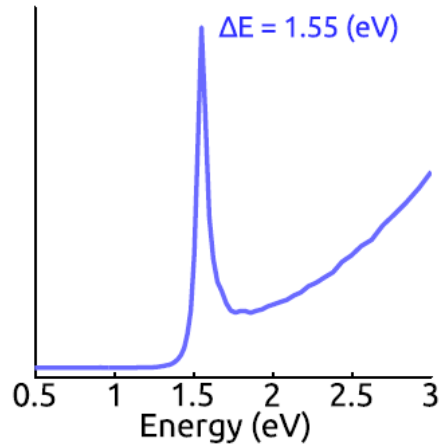
PINEM

PINEM spectroscopy mode: Time-resolved Electron Energy Loss Spectroscopy (EELS)

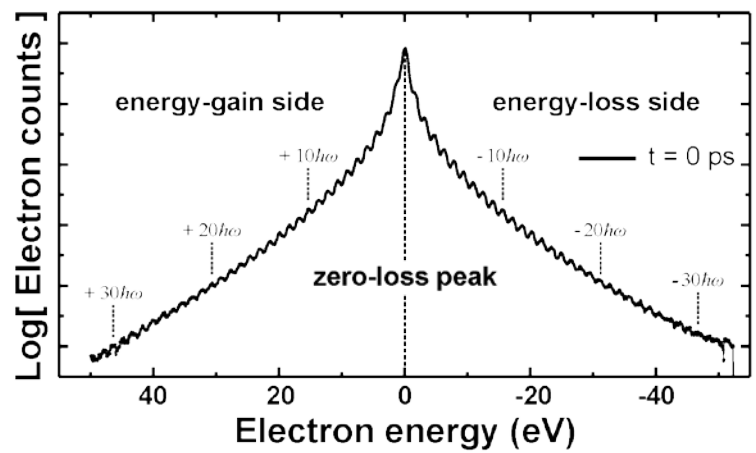
Energy exchange indeed yields Planck's constant

NIST reference value: $h = 4.135667516 \cdot 10^{-15}$ eV · s





Fourier transform

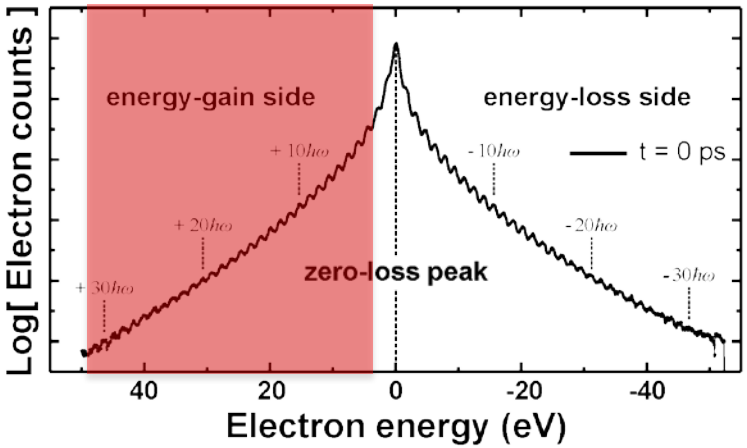


Energy exchange of over 30 photon quanta

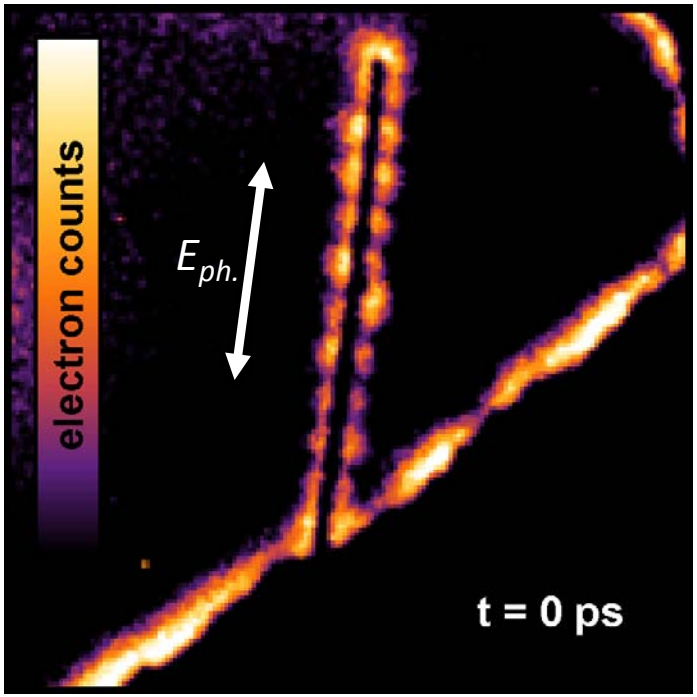
PINEM

PINEM imaging mode: energy-filtered imaging

- energy-filtering using GIF
- Image using only 'inelastic' electrons



Silver nanowire, hanging off a TEM grid

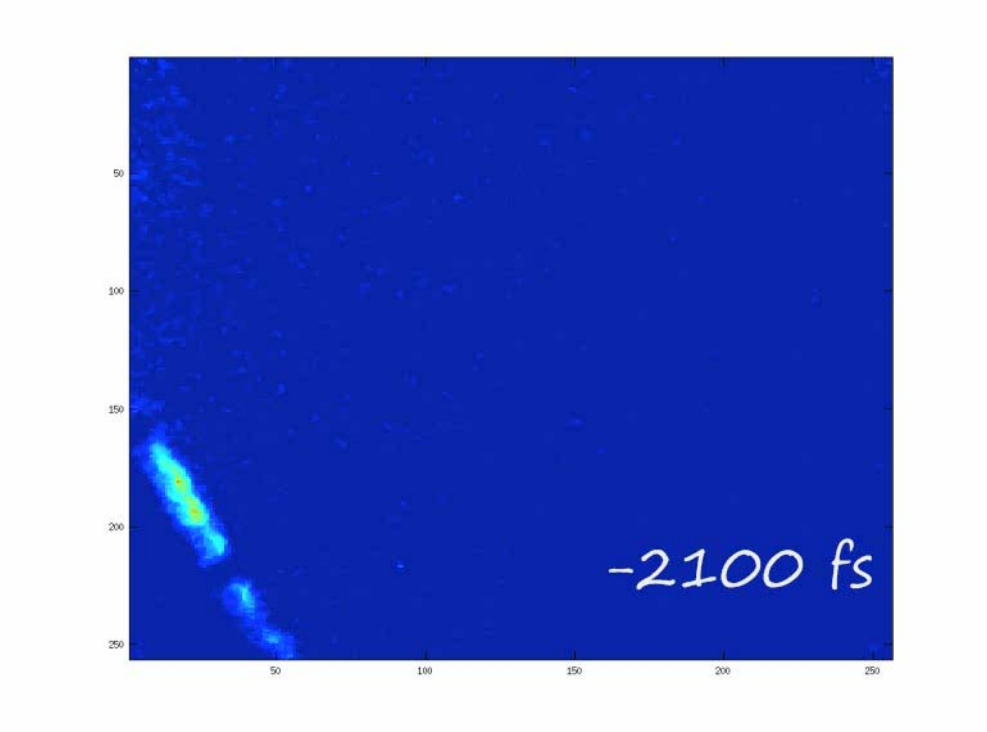


Time-resolved visualization of plasmonic field

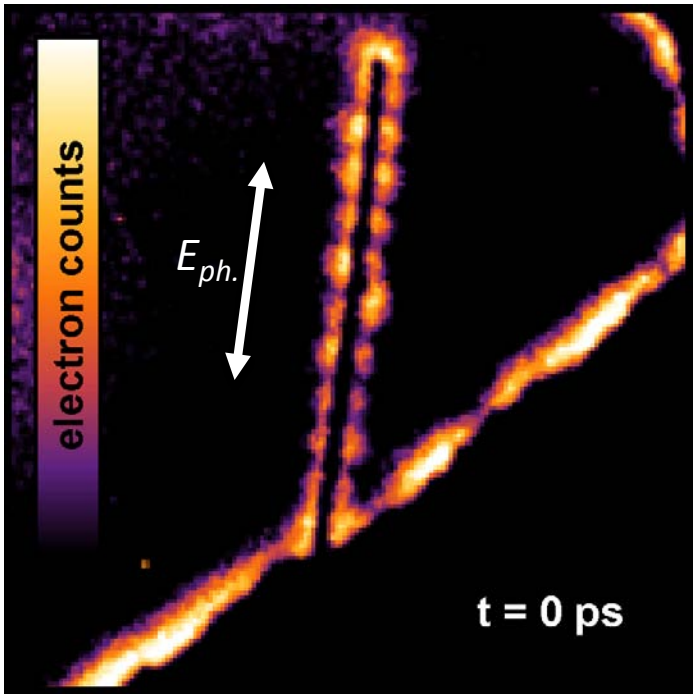
PINEM

PINEM imaging mode: energy-filtered imaging

- energy-filtering using GIF
- Image using only 'inelastic' electrons



Silver nanowire, hanging off a TEM grid



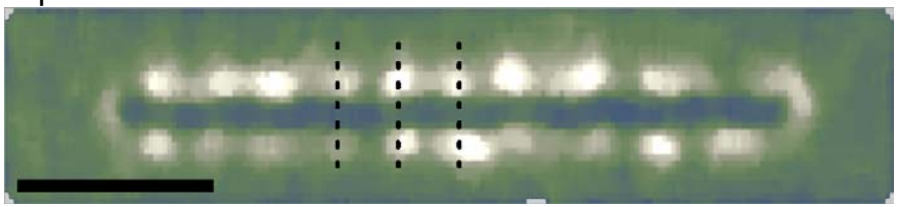
Time-resolved visualization of plasmonic field

PINEM

PINEM imaging mode: energy-filtered imaging

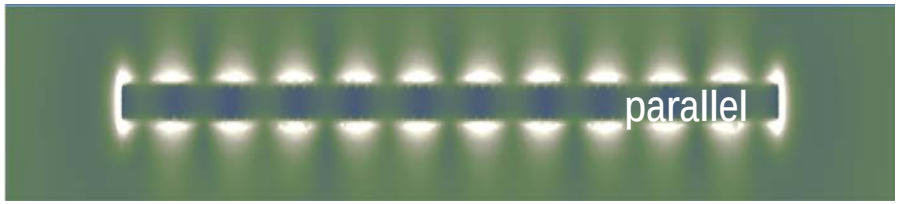
- energy-filtering using GIF
- Image using only ‘inelastic’ electrons
- Control plasmonic field through polarization

Experiment

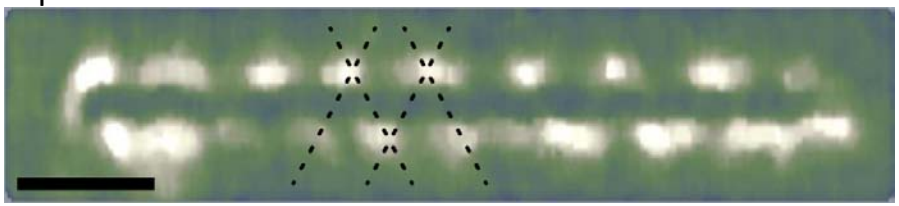


$E_{ph.}$ ⟷

Simulation

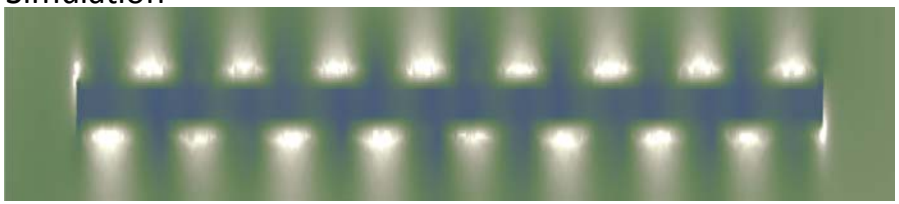


Experiment



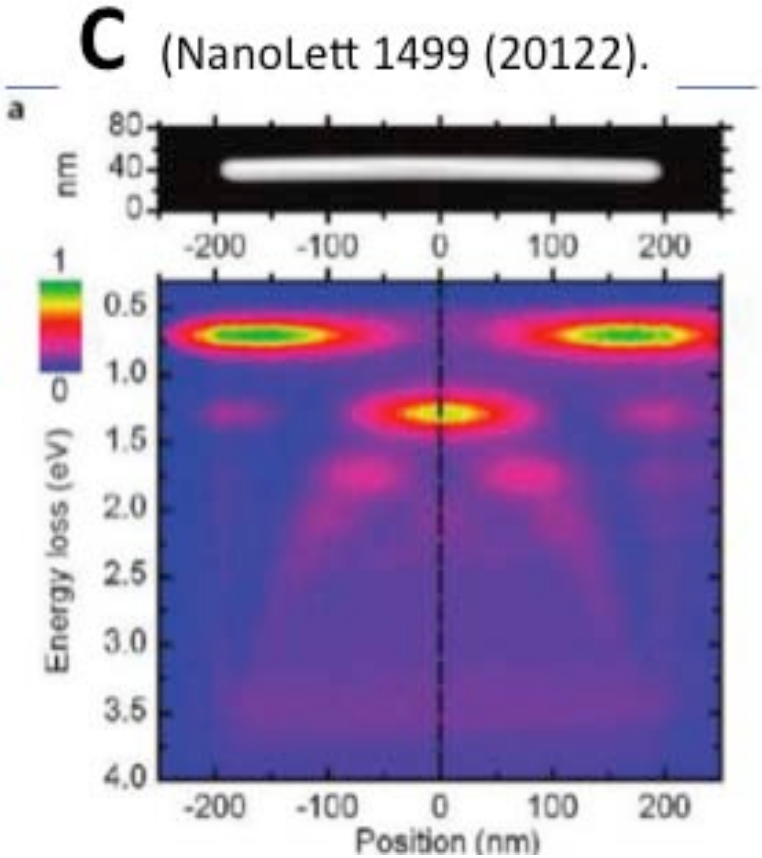
$E_{ph.}$ ↗

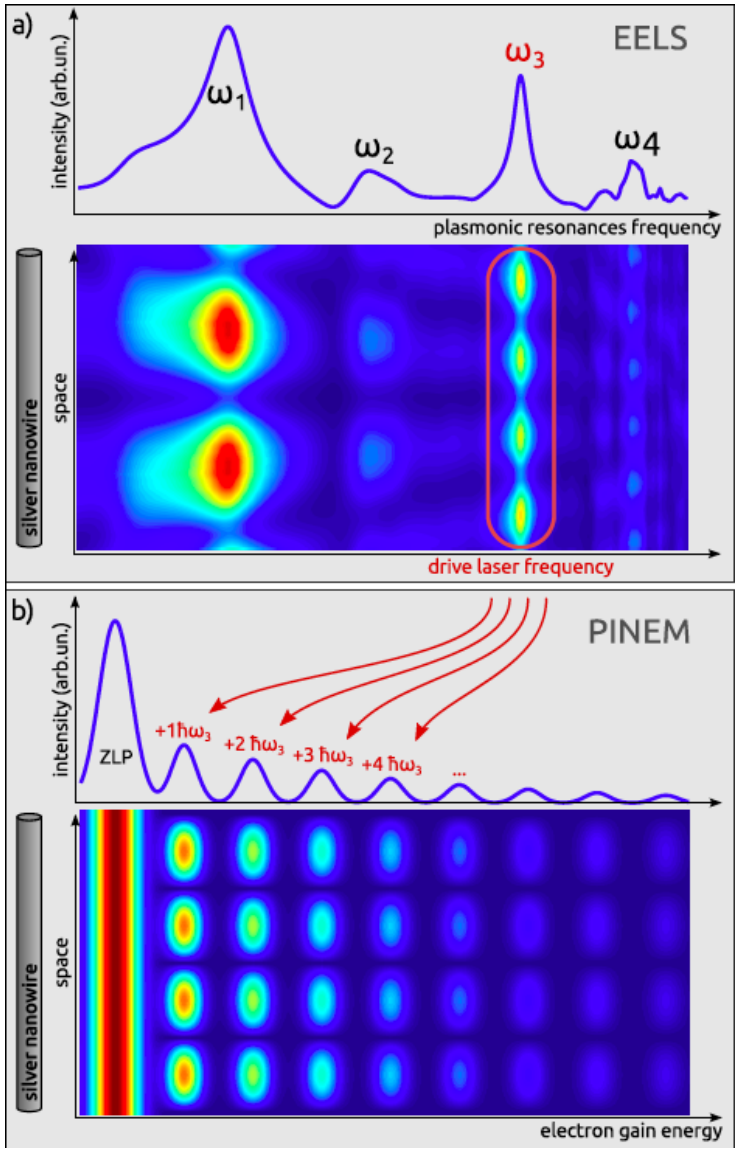
Simulation



Single Ag wire, suspended on graphene sheet

Laser driven plasmons

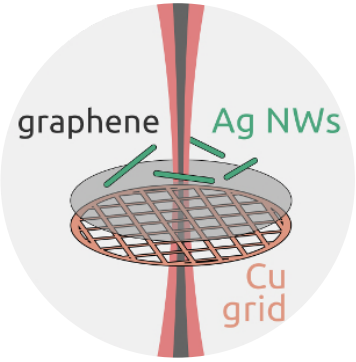




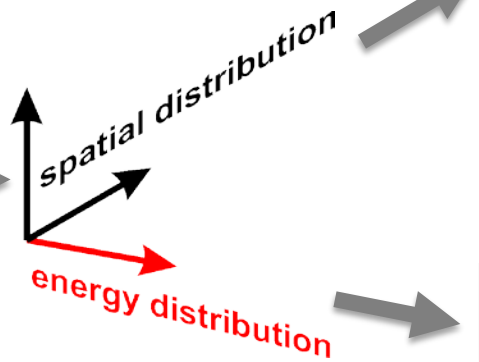
‘Hybrid’-PINEM

PINEM modes of acquisition:

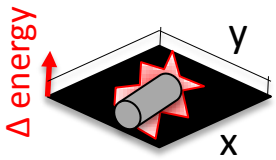
interaction @ t = 0



information

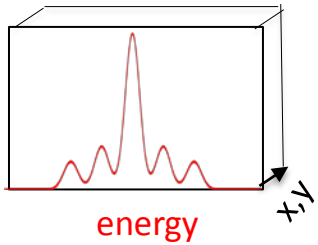


detection



(Energy filtered) imaging

- GIF: energy axis projected out
- Visualizes wave aspect



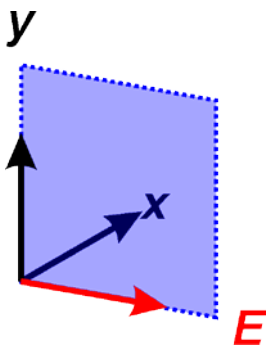
Electron energy spectroscopy

- GIF: spatial axes projected out
- Visualizes particle aspect

Hybrid PINEM mode: space-energy information ??

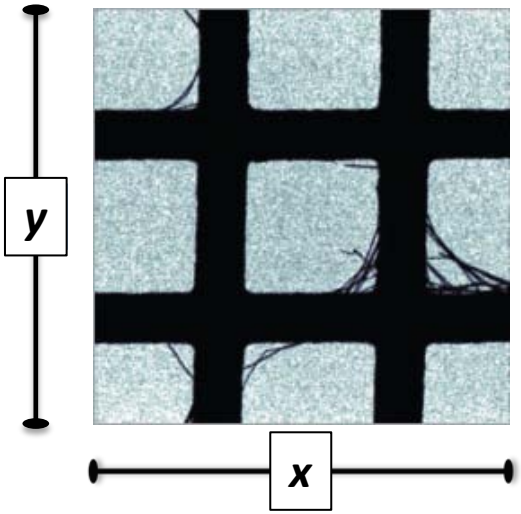
‘Hybrid’-PINEM

Hybrid PINEM mode: space-energy information



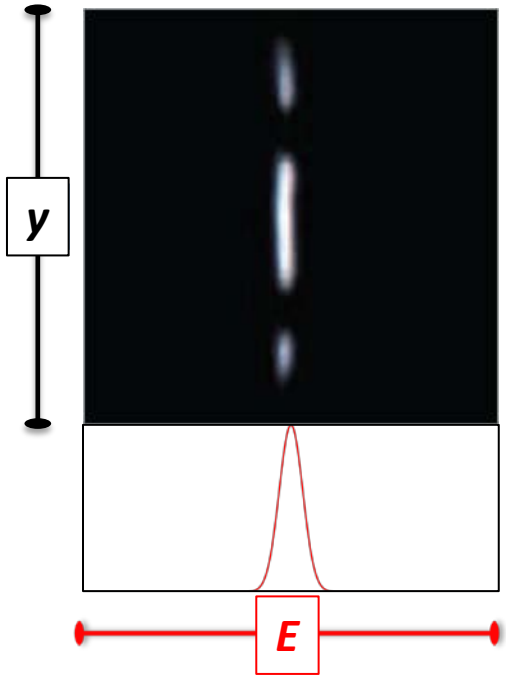
- 2D detection: GIF → project out only one spatial axis
- Proof of principle:

standard TEM image

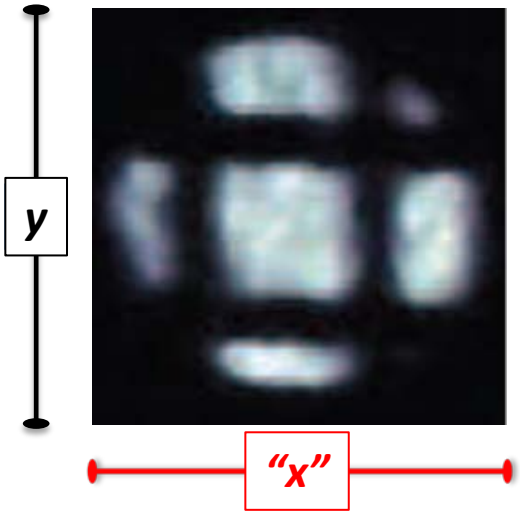


only ZLP electrons

raw EELS acquisition



unfocused EELS acquisition

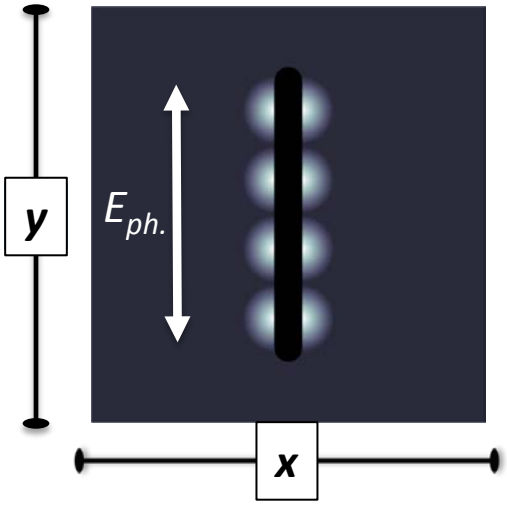


x-axis not projected out well

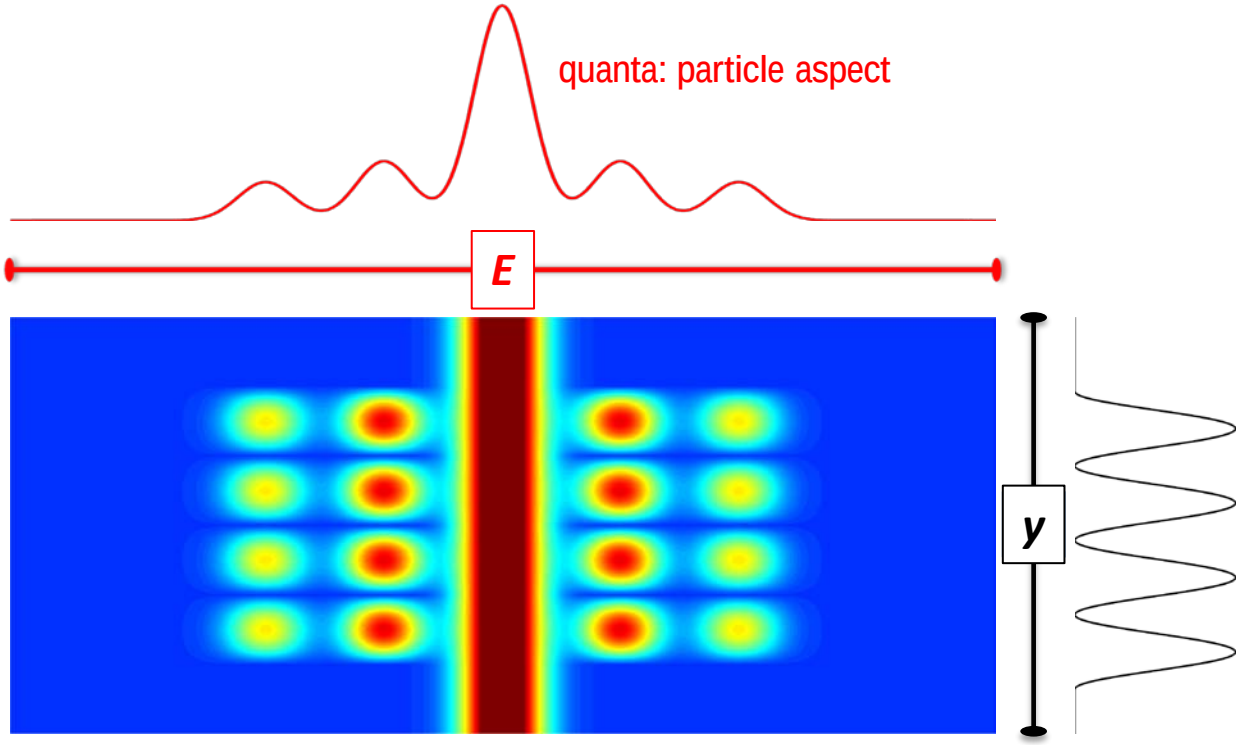
‘Hybrid’-PINEM

Hybrid PINEM mode: space-energy information

- PINEM image symmetric along x
- Idealized case @ t = 0



PINEM image



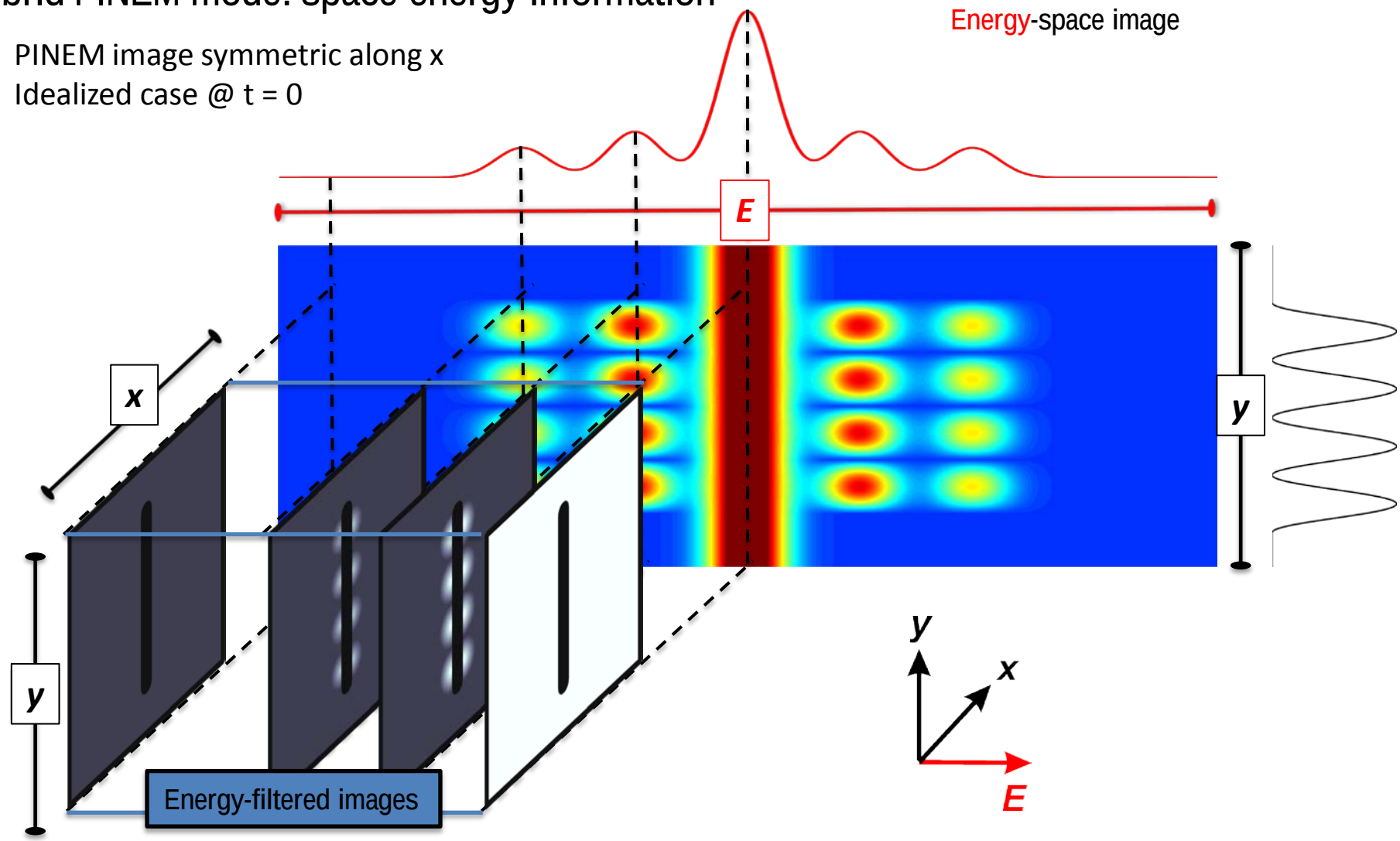
interference: wave aspect

Energy-space image

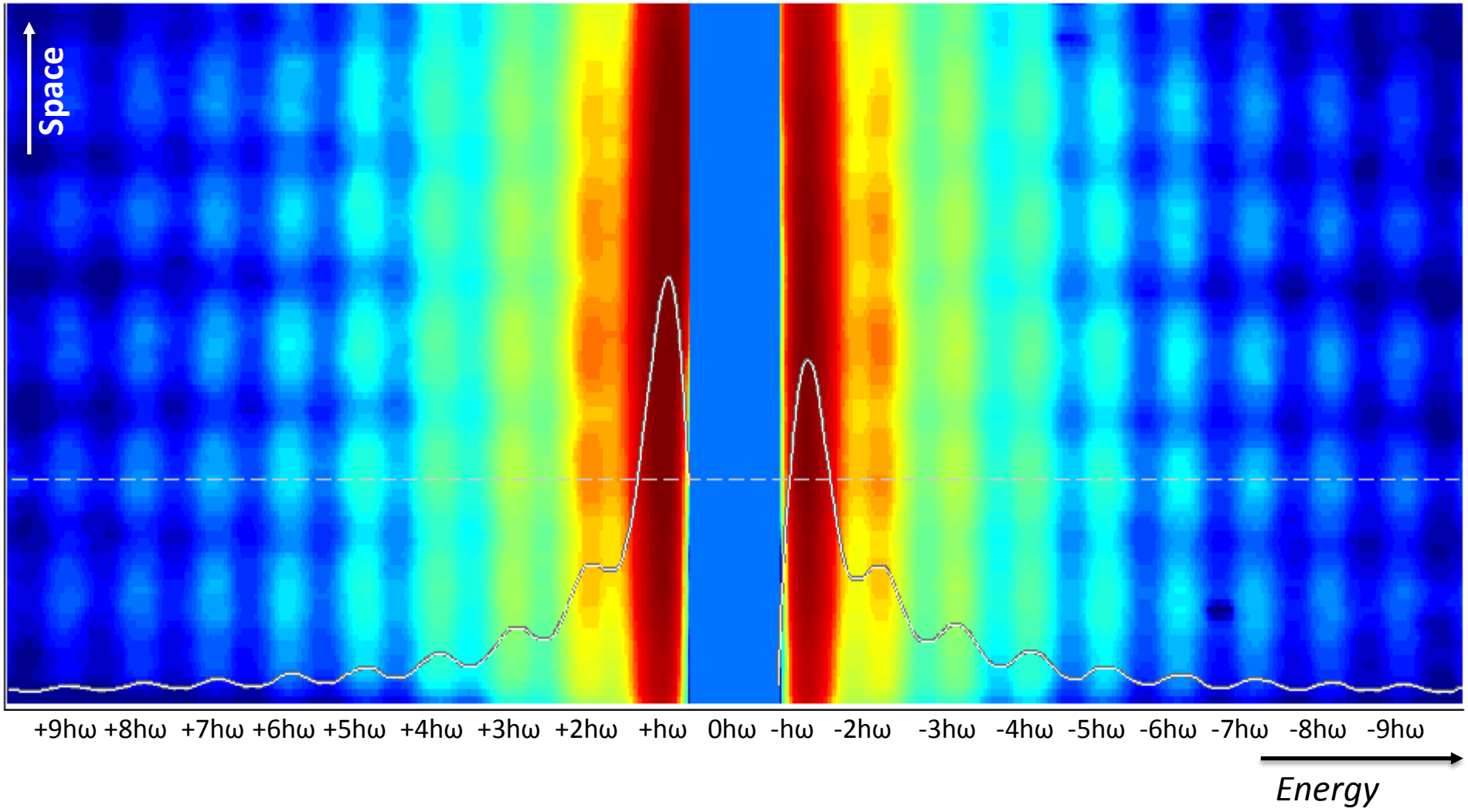
‘Hybrid’-PINEM

Hybrid PINEM mode: space-energy information

- PINEM image symmetric along x
- Idealized case @ t = 0



Simultaneous visualization of quantization and interference



Is THIS the end of the story?

Advantages:

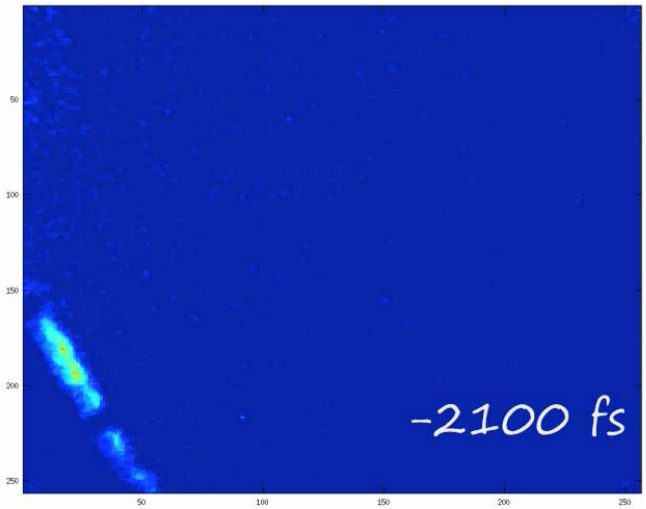
- The particle whose duality is tested does not “perform” the experiment. This completely defies the hidden variables.
- We use electrons in their particle limit (associated wavelength is ways smaller then any distance in the experiment) to image the EM field.
- We accumulate repeatedly the interaction between 1 electron and a discrete and numerable number of photons

Disadvantages:

- We observe simultanously the duality of a plasmonic field, i.e. a field sustained by a charge distribution (vacuum EM field vs the field of an antenna).
  Solution: photonic crystal for plasmons supporting intense interference between plasmonic fields.
- Is the quantization due to the EM field or the electrons sustaining the plasmon?
  Answer: in a semiclassical treatment, *h* has to be plugged in artificially. In a QED treatment of the interaction the field needs to be quantized.

Take-home messages

- Time-resolved electron microscopy for imaging plasmons
- Successful observation of a plasmonic standing wave on a single, isolated nanowire
- Demonstration of polarization control of the plasmonic field
- Demonstration of energy-space plasmon imaging
- Simultaneous observation of both quantization and interference of the plasmonic field

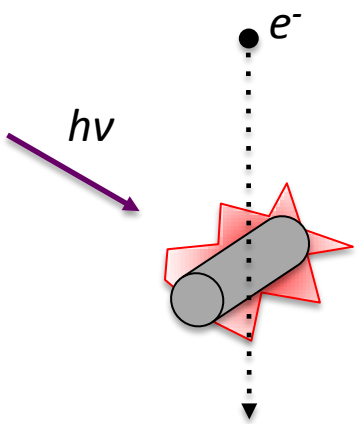


Thank you!

Contact:

Prof. Fabrizio Carbone
EPFL – LUMES
Fabrizio.carbone@epfl.ch

Interaction



Semi-classical treatment:

$$H = \frac{\hat{p}_e^2}{2m_e} + \left(\frac{e}{m_e}\right)\vec{A}\cdot\hat{p}_e + \frac{e^2A^2}{2m_e} \approx -\frac{\hbar^2\nabla^2}{2m_e} - i\frac{e\hbar}{m_e}\frac{1}{2}\left(\frac{\tilde{E}}{+i\omega_p} + \frac{\tilde{E}^*}{-i\omega_p}\right)\cdot\vec{\nabla}$$

KE *Field interaction* *Ponderomotive potential negligible (p_e) very large in PINEM*

Plasmonic near field has z component which accelerates and decelerates electrons.
Electron wavefunction:

$$\Psi(z, t \to +\infty) = g(z - v_e t, -\infty) \sum_{n=-\infty}^{\infty} \xi_n(z - v_e t) \exp\left[i\left(k_e + n\frac{\omega_p}{v_e}\right)z - i(\omega_e + n\omega_p)t\right],$$

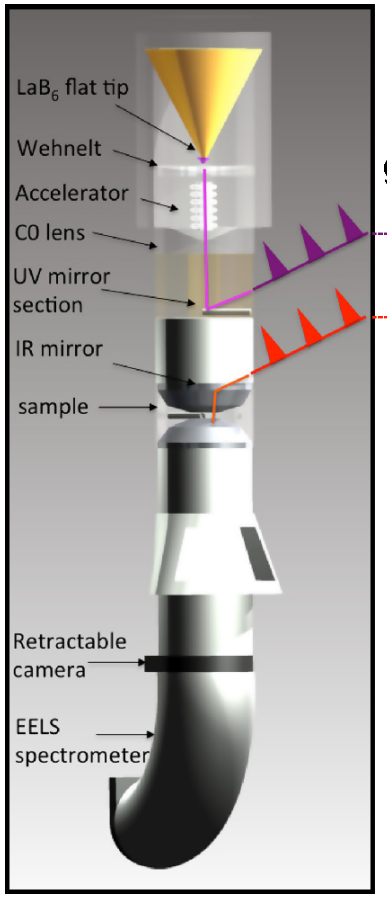
- Sum of electron wavelets at equidistant momenta.
- This originates from the decomposition of the near-field in its harmonics

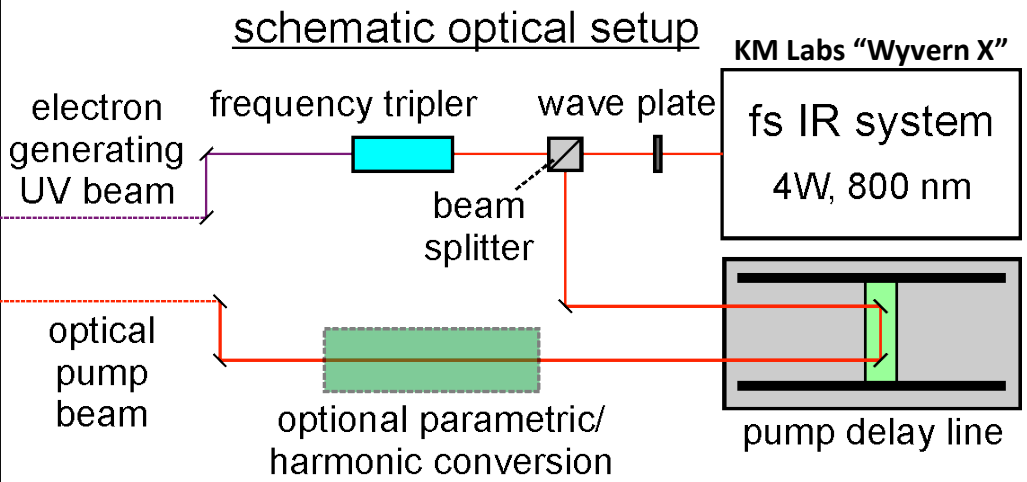
→ To obtain the energy spectrum one has to multiply by **Planck's constant *h***

S.T. Park *et al.*, *New J. Phys.* **12**, 123028 (2010)

Approach & Instrumentation

- Modified 200 keV Jeol JEM-2100
- Amplified Ti:sapphire laser system





Modified elements:

- extra 'C0' electrostatic lens
- UV mirror section
- Pump mirror
- GATAN GIF spectrometer

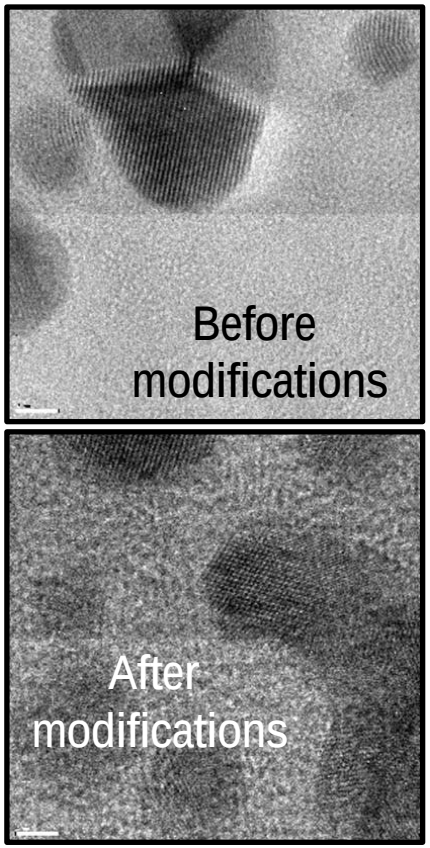
L. Piazza *et al.*, *Chemical Physics* **423**, 79 (2013)

Approach & Instrumentation

- Effect on static TEM

L. Piazza *et al.*, *Chemical Physics* **423**, 79 (2013)

Spatial Resolution



Gold nanoparticles

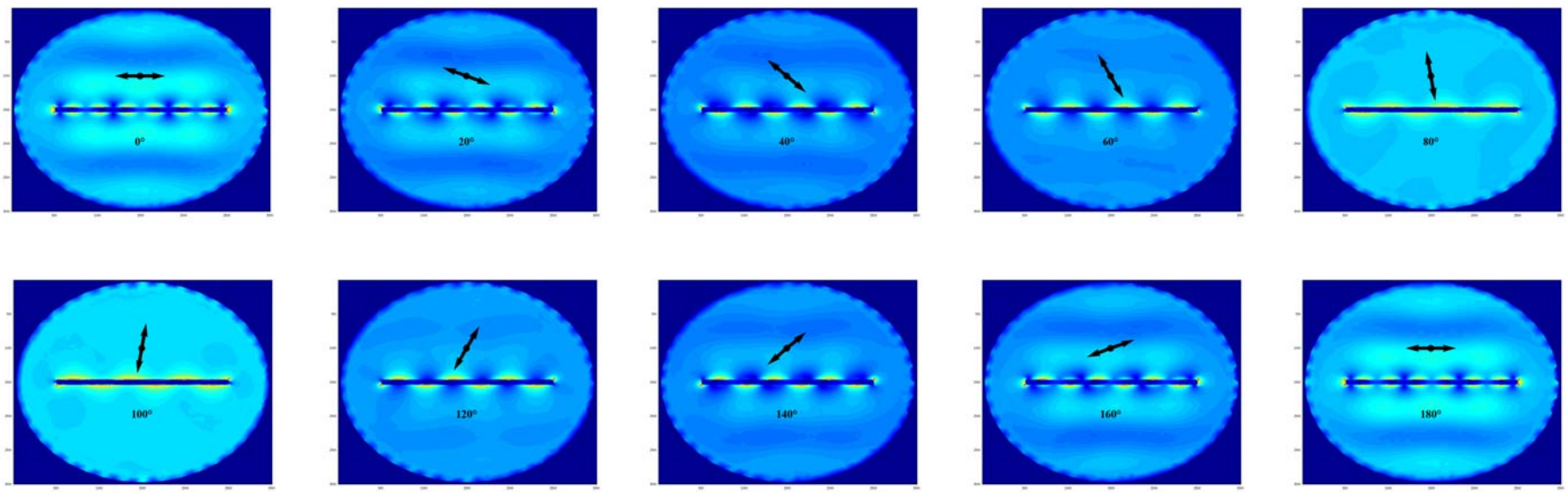
- Slight blurring at image edge
- Lattice fringes observed
- Reasonable atomic resolution even after modifications

Results

Polarization dependence: numerical simulation

Numerical Simulation
Silver wire, suspended in air

COMSOL Multiphysics® simulation



Continuous evolution of evanescent field with polarization