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


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


Davidovic and Sainz, comment in EPN (2013)
Delayed choice experiments

Motivation Quantum mechanics

Test: detected in the interferometer
Corroborative: tells if it behaved as wave or particle before any information could be exchanged via hidden variables

Polarization entangled photons

Hidden variable

Corroborative photon apparatus
Test photon apparatus

Entanglement source

$\frac{1}{\sqrt{2}}(c_H^\dagger t_H^\dagger + c_V^\dagger t_V^\dagger)|\text{vac}\rangle$

Forward light cone

Corroborative photon detected

Pair generation

Position (m)

Time (ns)

230 ns

250 ns

3 m

-17 m

20 m

$\theta$

BS$_1$

PBS$_1$

PBS$_2$

PDBS

D$_a''$

D$_b''$

D$_a$

D$_b$

$\phi$

Corroborative photon apparatus

Test photon apparatus

Hidden variable
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 than 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

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
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 $>> \lambda$
- Inherent limitations:
  - spatial resolution (~10nm)
  - point-by-point probing
  - Time-resolution

Alternative method?

Quantum properties of plasma waves


Plasmons entanglement

Two-plasmons quantum interference
Wave-particle duality of single surface plasmons
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 than 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:

• energy-filtering using GIF

• Image using only ‘inelastic’ electrons

**PINEM**

- Energy-filtering using GIF
- Image using only ‘inelastic’ electrons

**Carbon nanotubes**


**E. Coli**


**Protein Vesicles**

PINEM vs Spectral imaging

A (PRL 110, 066801 (2013))

Free space wavelength (μm)

Energy loss (eV)

B (PRL 110, 066801 (2013))

C (NanoLett 1499 (2012)).
PINEM spectroscopy mode: Time-resolved Electron Energy Loss Spectroscopy (EELS)

Energy exchange of over 30 photon quanta
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Energy exchange of over 30 photon quanta

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

Energy exchange indeed yields Planck’s constant

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

Energy exchange of over 30 photon quanta
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 imaging mode: energy-filtered imaging

- energy-filtering using GIF
- Image using only ‘inelastic’ electrons

Time-resolved visualization of plasmonic field

Silver nanowire, hanging off a TEM grid
PINEM imaging mode: energy-filtered imaging

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

Single Ag wire, suspended on graphene sheet
Laser driven plasmons

(NanoLett 1499 (20122).)

EELS

plasmonic resonances frequency

drive laser frequency

PINEM

ZLP

+1\omega_3  +2\omega_3  +3\omega_3  +4\omega_3  ...

electron gain energy
‘Hybrid’-PINEM

PINEM modes of acquisition:

Hybrid PINEM mode: space-energy information ??

Energy filtered) imaging
- GIF: energy axis projected out
- Visualizes wave aspect

Electron energy spectroscopy
- GIF: spatial axes projected out
- Visualizes particle aspect

interaction @ t = 0

energy distribution

spatial distribution
‘Hybrid’-PINEM

Hybrid PINEM mode: space-energy information

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

- Standard TEM image
- Raw EELS acquisition
- Unfocused EELS acquisition

only ZLP electrons

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**

**Energy-space image**

quanta: particle aspect

interference: wave aspect
‘Hybrid’-PINEM

Hybrid PINEM mode: space-energy information

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

Energy-space image

Energy-filtered images
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 than 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 simultaneously 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!

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Interaction

**Semi-classical treatment:**

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

- **KE**
- **Field interaction**
- **Ponderomotive potential negligible**
- *(\rho_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 \( h \)**

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
    
  
Approach & Instrumentation

- **Effect on static TEM**

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

Results

Polarization dependence: numerical simulation

Continuous evolution of evanescent field with polarization

Numerical Simulation
Silver wire, suspended in air

COMSOL Multiphysics® simulation