Photo-Induced Near-field Electron Microscopy (PINEM)

Thomas LaGrange

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Doctoral Course PHYS-637

Spring 2024

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Outline:

Photo-Induced Near-field Electron Microscopy (PINEM) was first discovered in the group Ahmed Zewail at Caltech during pump-probe femtosecond EELS experiments in a unique Ultrafast TEM. Femtosecond laser excitation of nanostructures induces evanescent surface optical modes (surface plasmon polaritons). These optical modes can modulate the longitudinal phase of the electron beam into quantized energy loss and gain multiples of the exciting laser photon frequency. Experiments have explored the quantum nature of PINEM and have shown coherent control of the phase, PINEM-based holography, and the ability to shape the orbital angular momentum of the electron beam.

1) Photo-induced Near Field Electron Microscopy
   A. Can light and electrons interact?
   B. Ultrafast TEM and first Observations
2) Coherent phase modulation
   A. Rabi Oscillations- quantum walk
   B. Electron-photon coupling constant - $|g|$
3) PINEM experiments
   A. SPP velocity
   B. Coherent control and attoseconds
   C. Electron beam shaping
4) CWPINEM

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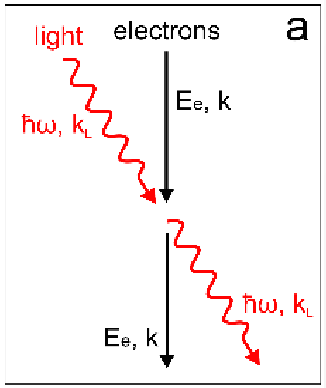
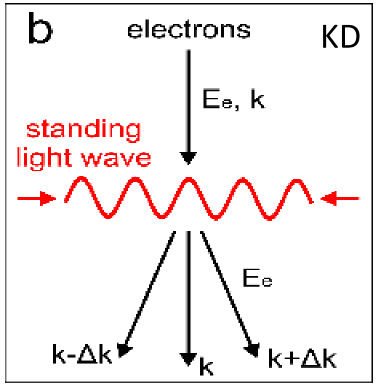
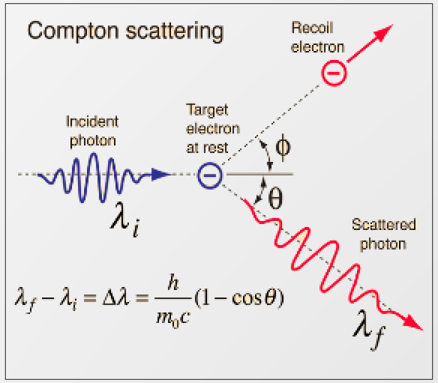
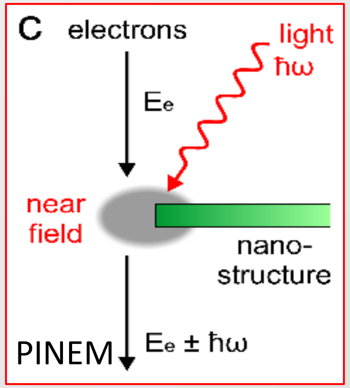
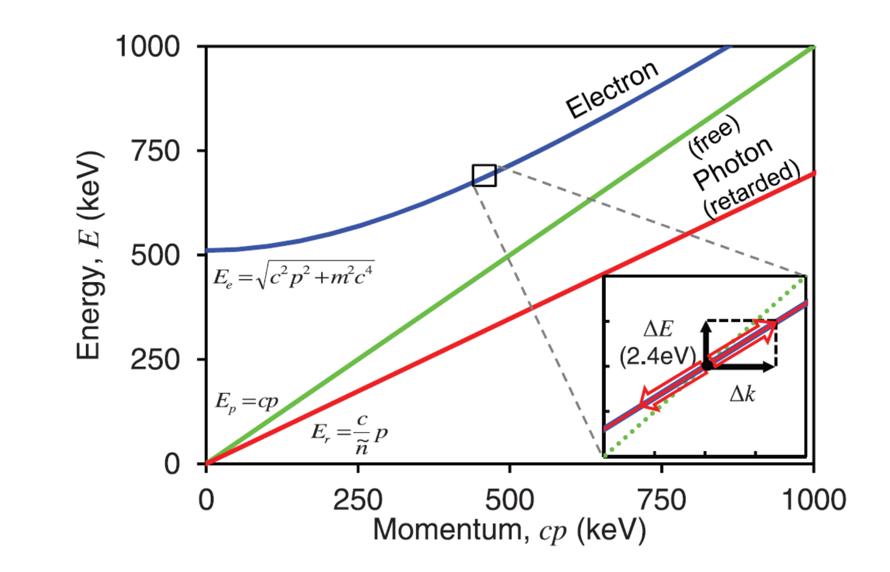
How can fast electrons and photons interact? It should not be possible due to the momentum mismatch!!

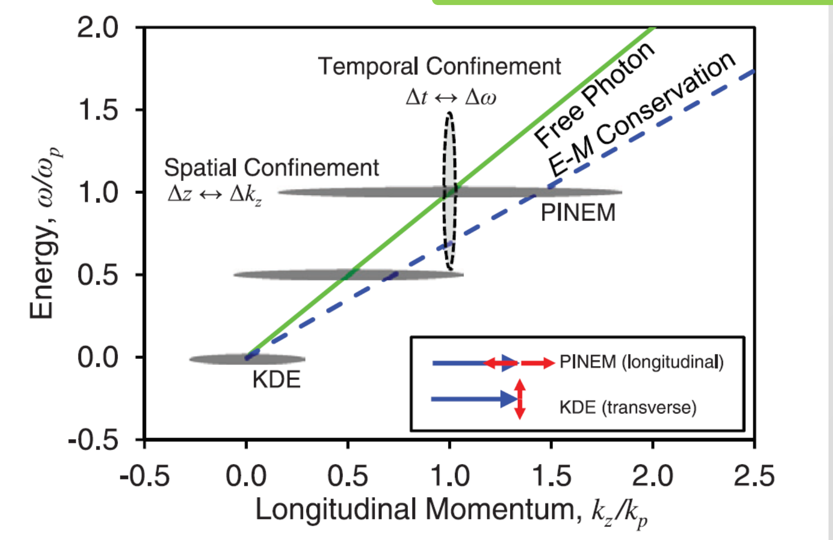












In PINEM, interaction with the spatially confined light induces a longitudinal momentum change large enough to allow for electrons-photon to exchange energy, i.e., acceleration and deceleration of the electrons

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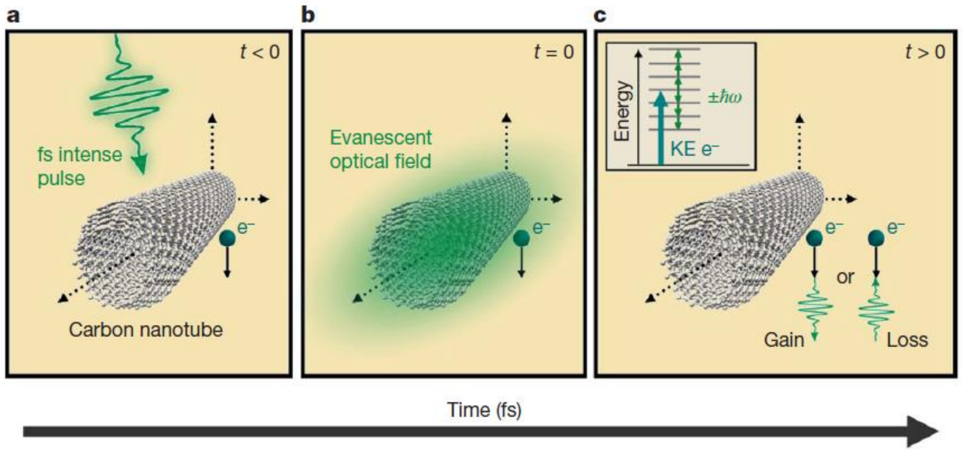
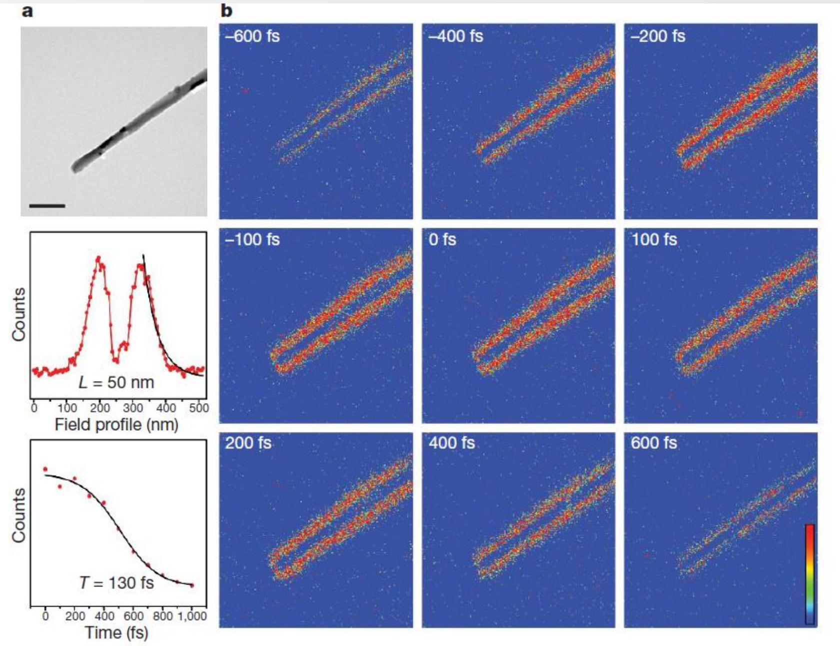
First observations of Photo-induced Near-Field effects

LETTERS

Photon-induced near-field electron microscopy

Brett Barwick[1], David J. Flannigan[1] & Ahmed H. Zewail[1]





The intense fs light pulse couples into the carbon nanotube and generates electromagnetic modes (plasma resonances) that decay within a few ps

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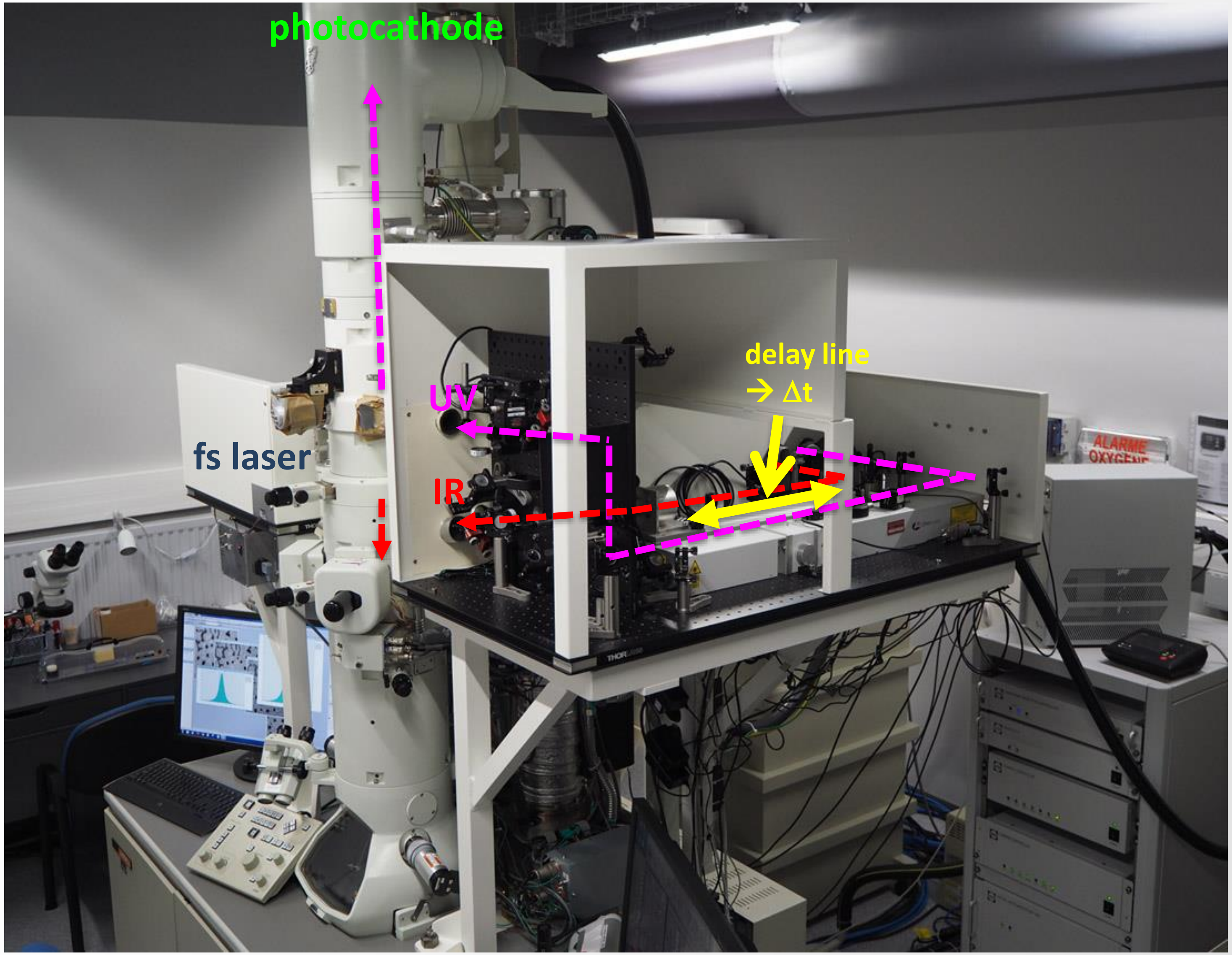
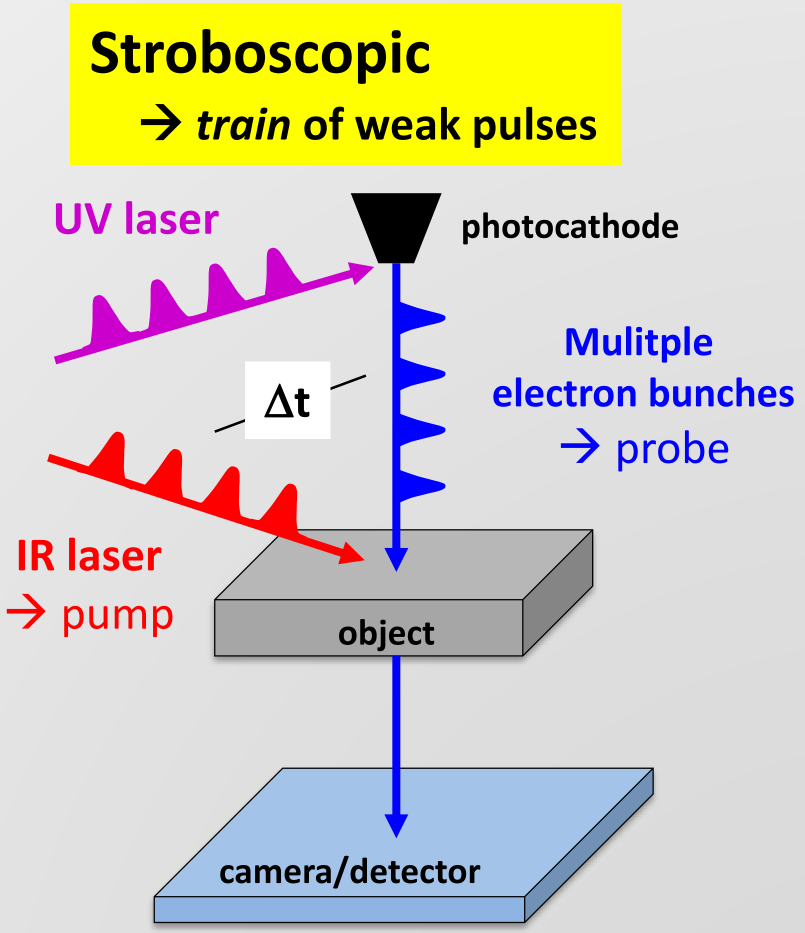
Femtosecond Stroboscopic UTEM setup





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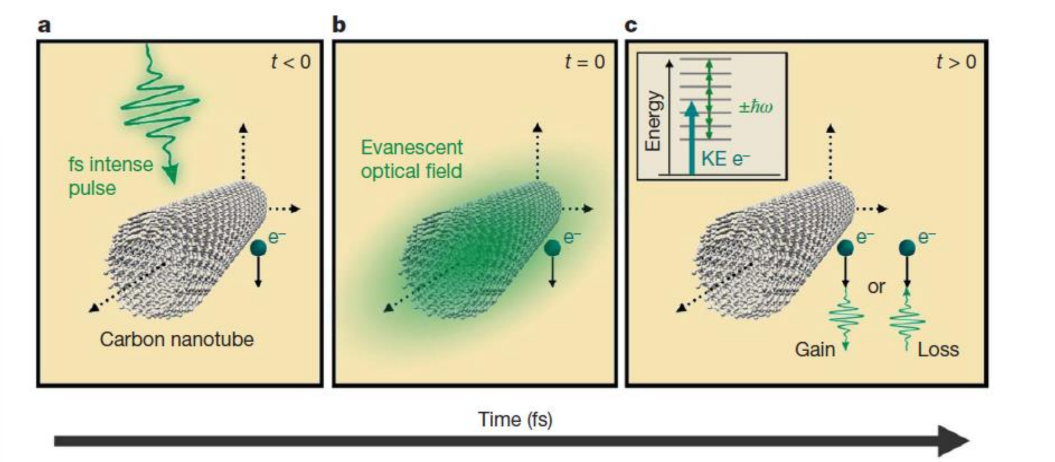

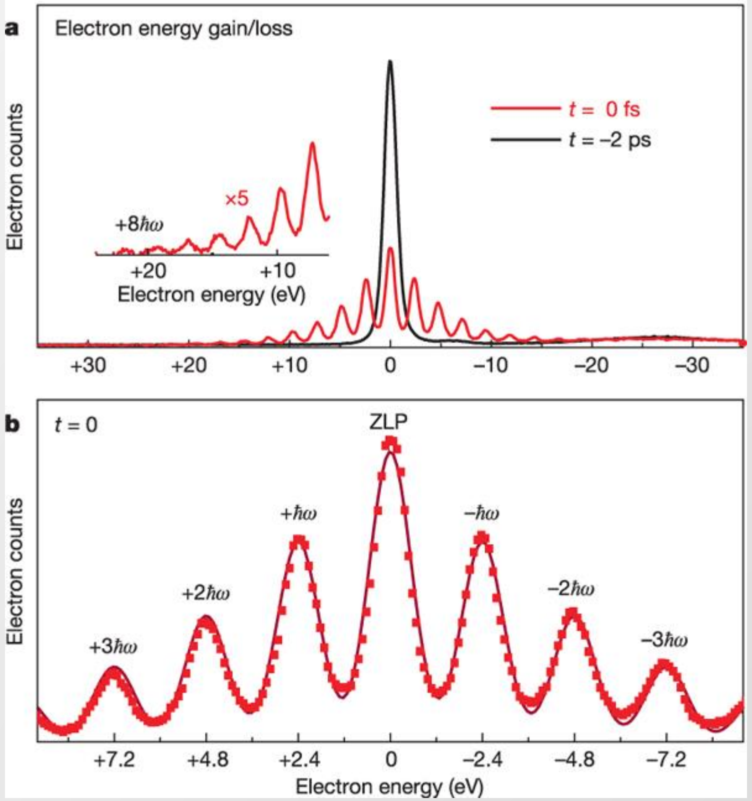

The phase velocity of light is slowed in the media, allowing momentum matching with electrons and for the exchange energy at the quantized photon frequency of the exciting light

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Electrons scatter off the confined, evanescent optical field causing a modulation of their longitudinal phase (quantized energy)

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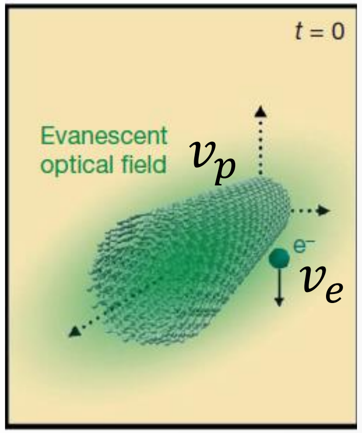
Initial envelope

Bessel function of the first kind of order k

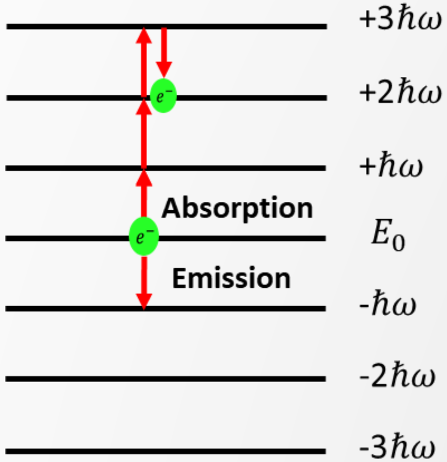
Change in the energy and momentum distribution

$$\Psi_e(z,t) = \phi(z - v_e t, t \to -\infty) \sum_{k=-\infty}^{+\infty} J_k(2|g|) e^{ik\arg(g)} e^{i\left[\left(k_e + k\frac{\omega_p}{v_e}\right)z - (\omega_e + k\omega_p)t\right]}$$

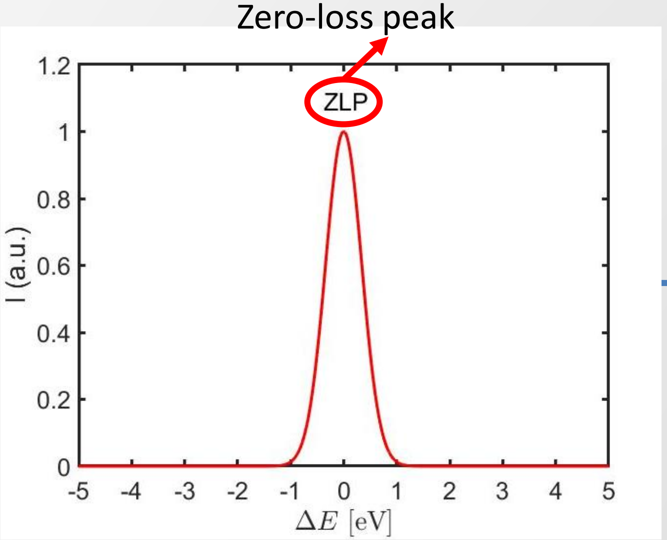
Final electron state

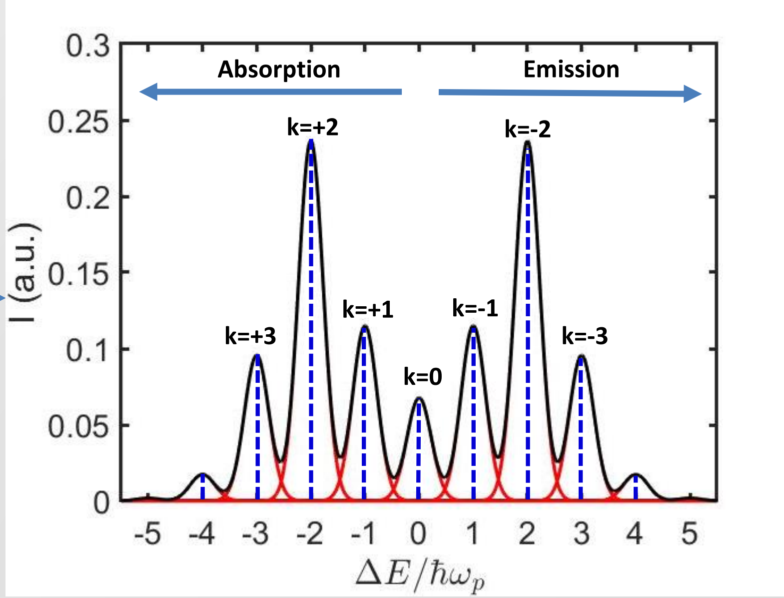


Phase matched electrons & photons



Rabi Oscillations
Quantum walk





$$g(x,y) = \frac{q_e}{2\hbar\omega_p} \int_{-\infty}^{+\infty} \tilde{\mathcal{E}}_z(x,y,z) e^{-i\frac{\omega_p}{v_e}z} dz$$

$$P_k = J_k^2(2|g|)$$

Electron-photon coupling constant

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LUMES

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

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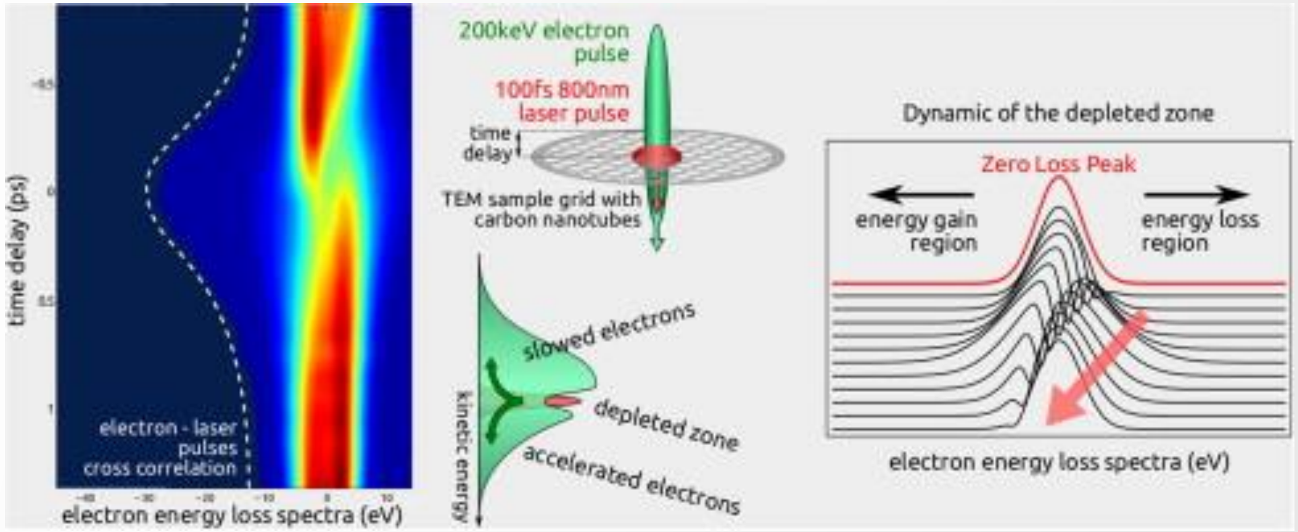
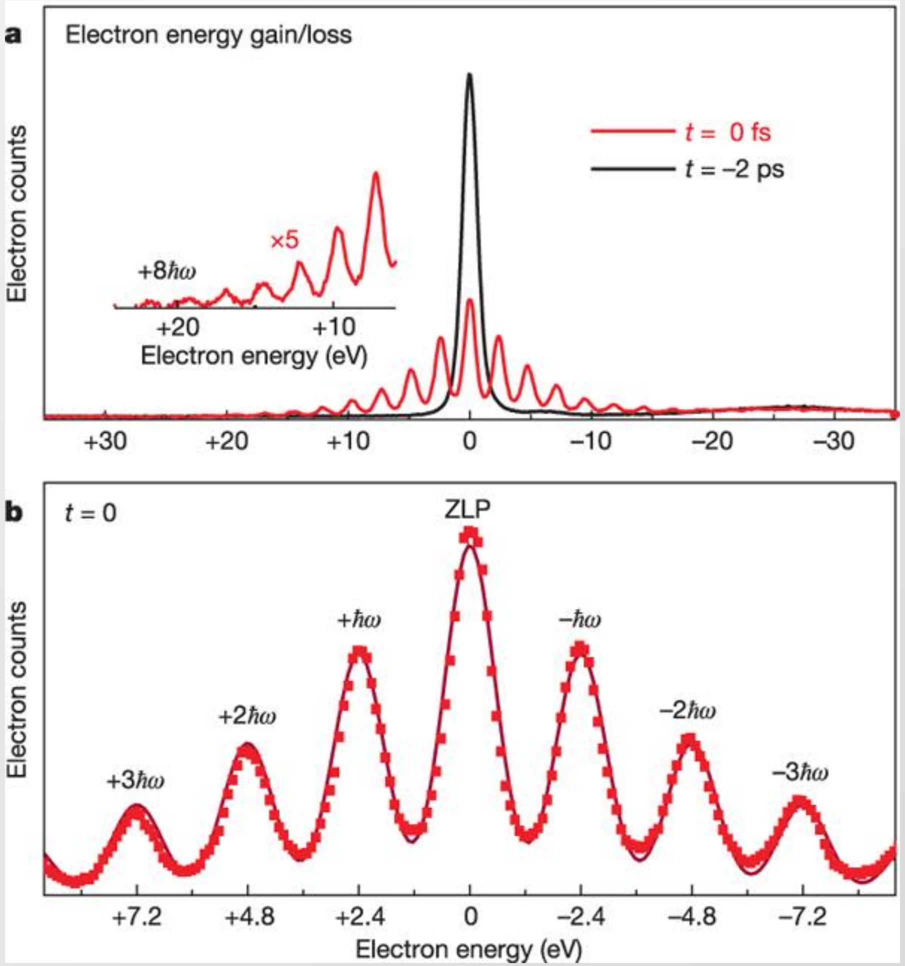
Fs optical excitation of nanostructures produces evanescent optical fields that interact with “aloof” electrons, giving them either an energy gain or loss equivalent to the quantized photons



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

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



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

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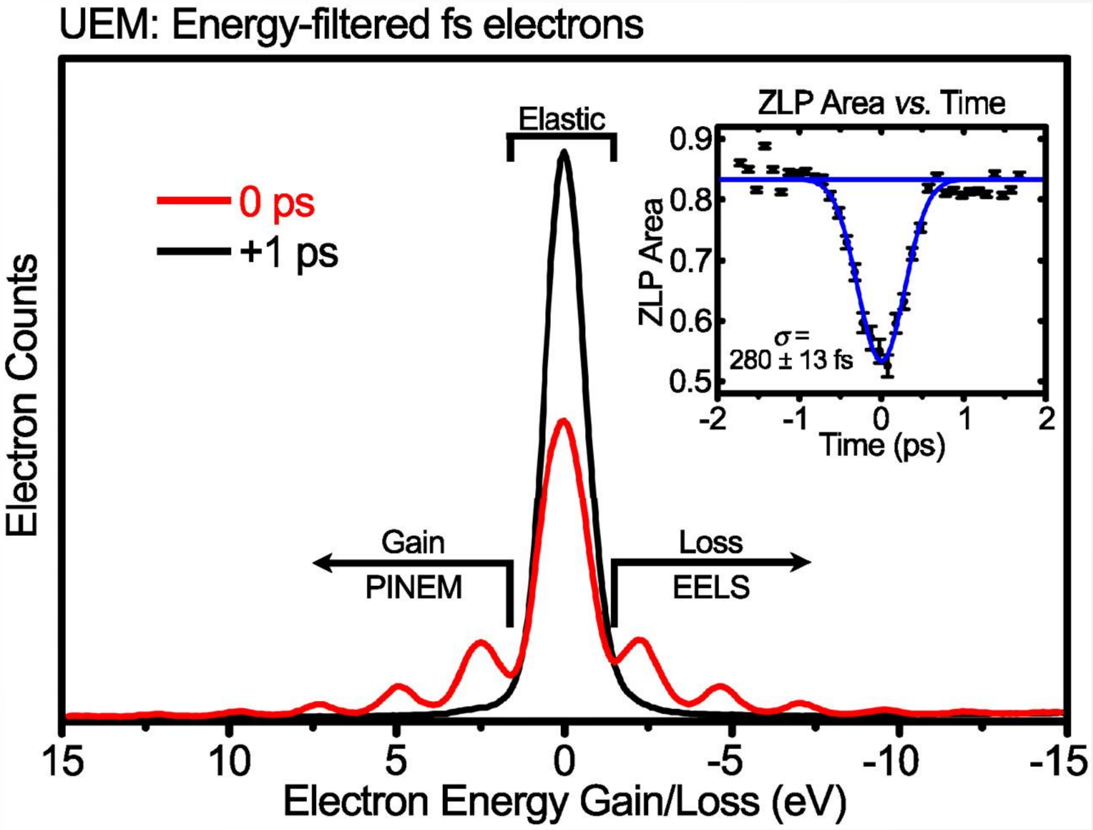
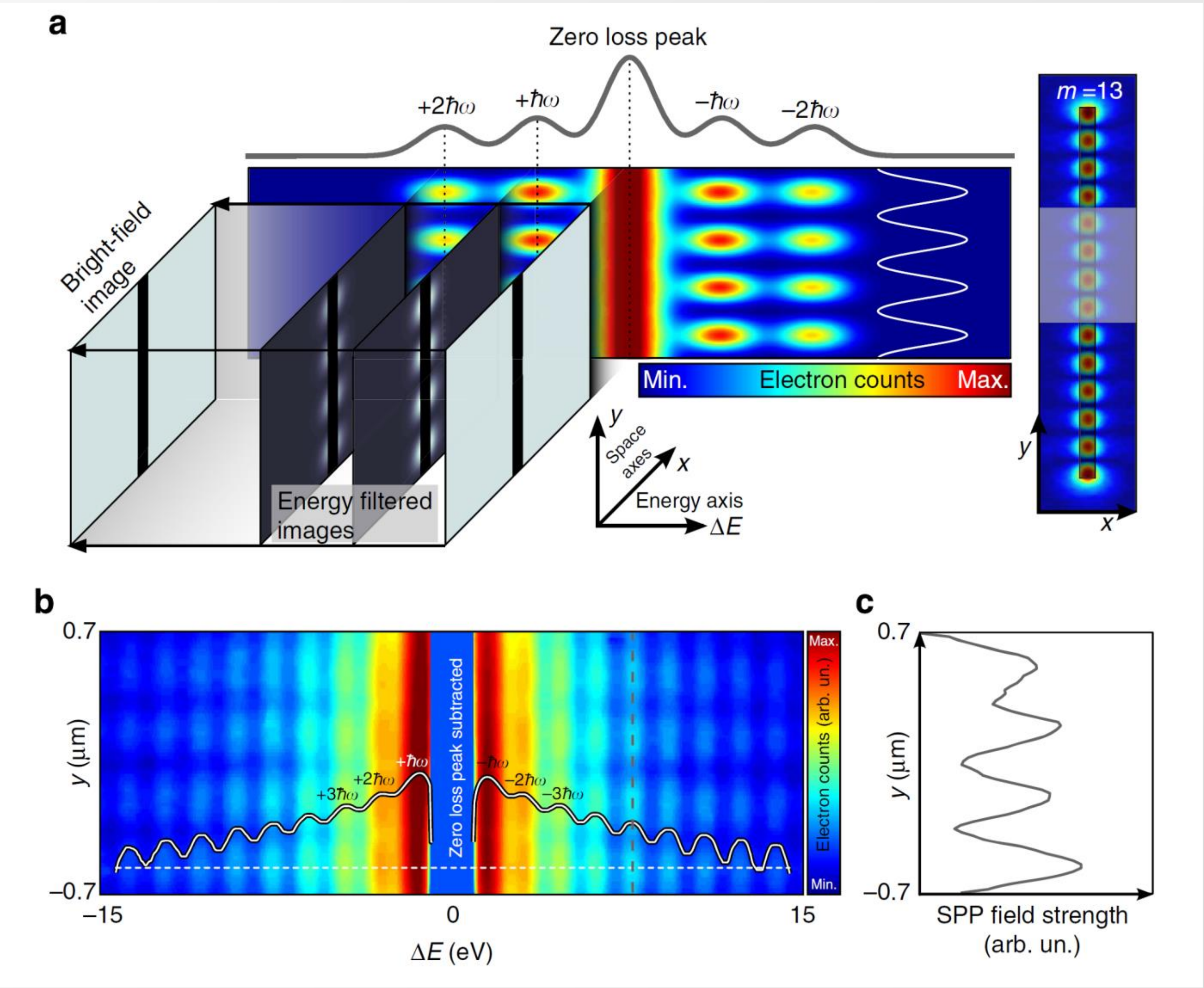
LUMES

Energy filtered TEM mapping in UTEM PINEM experiments

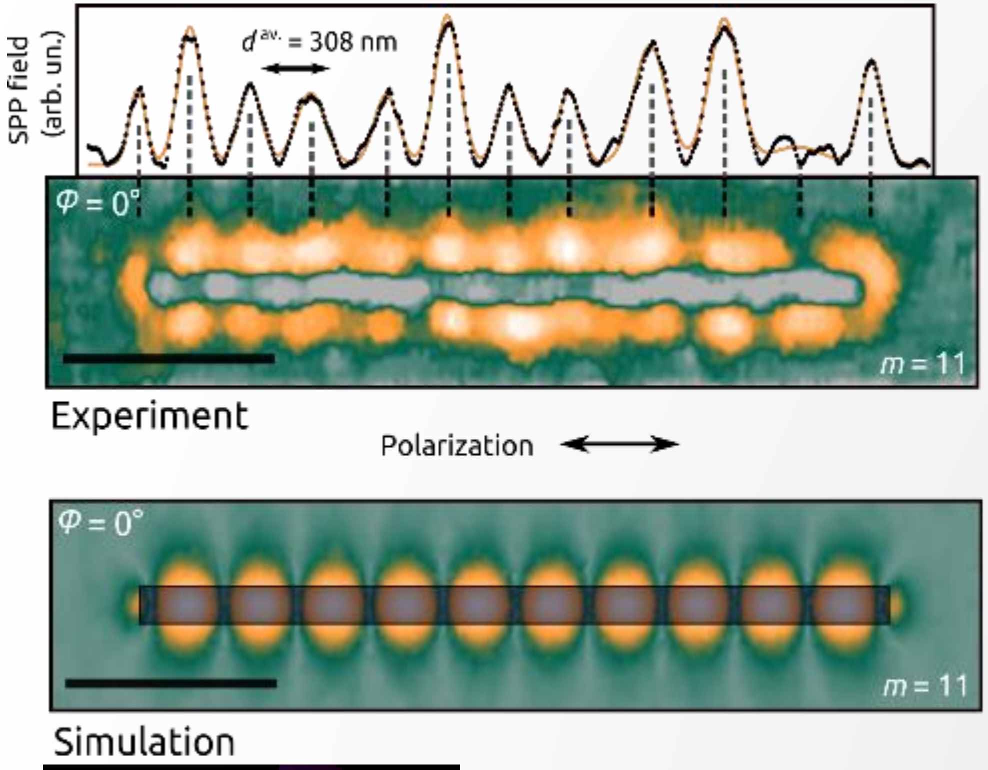


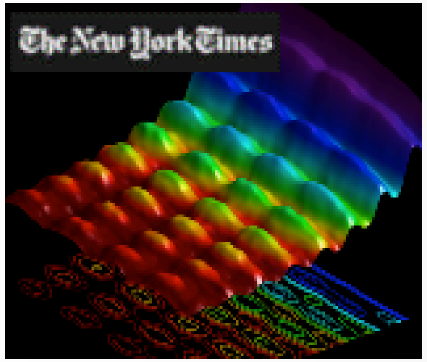


David J. Flannigan, Brett Barwick, and Ahmed H. Zewail, Proceedings of the National Academy of Sciences **107** (22), 9933 (2010).

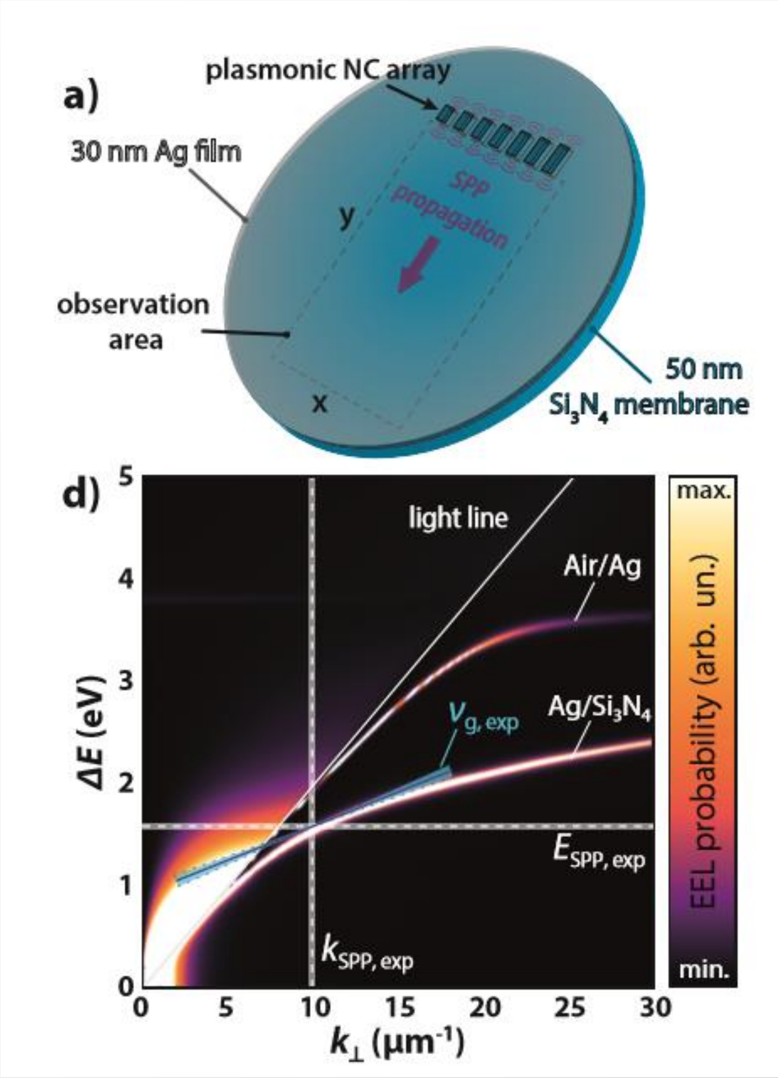
L. Piazza, T. T. A. Lummen, E. Quiñonez, Y. Murooka, B. W. Reed, B. Barwick, and F. Carbone, Nature Communications **6** (1), 6407 (2015).

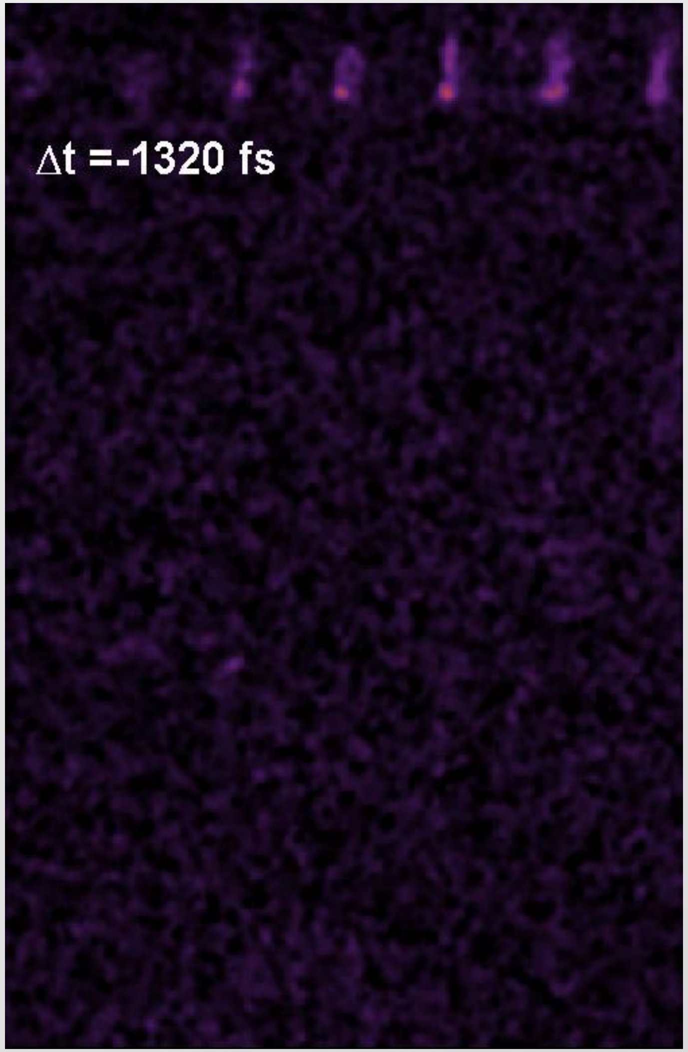
Surface plasmons polaritons and plasma resonances





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





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

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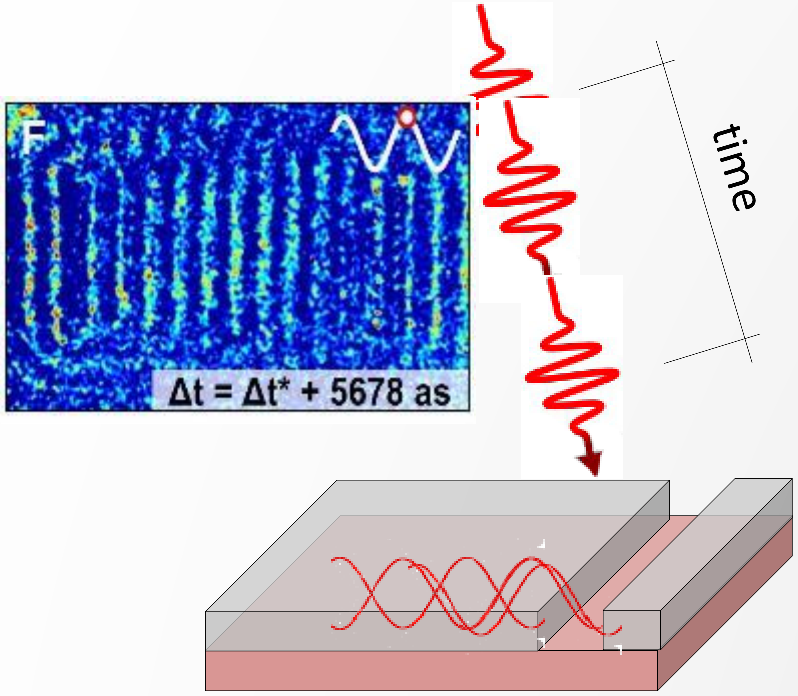
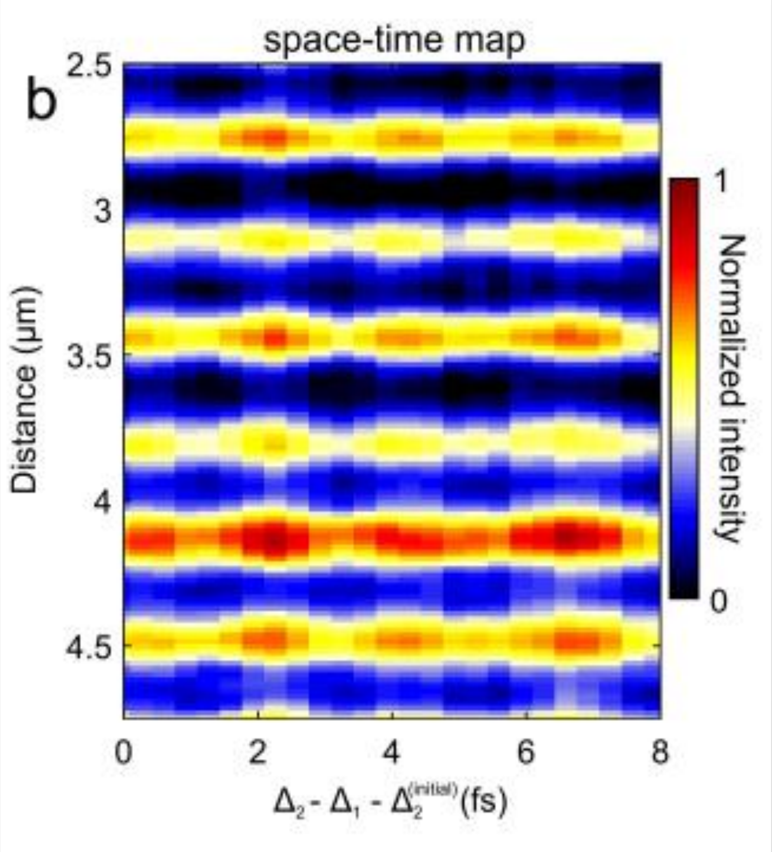
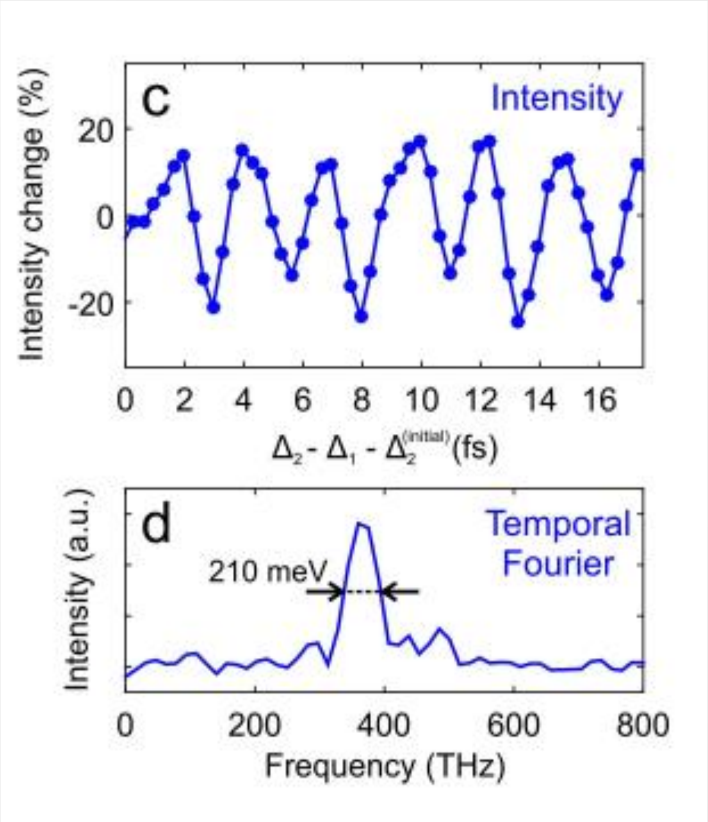
Coherent control of plasma resonances







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

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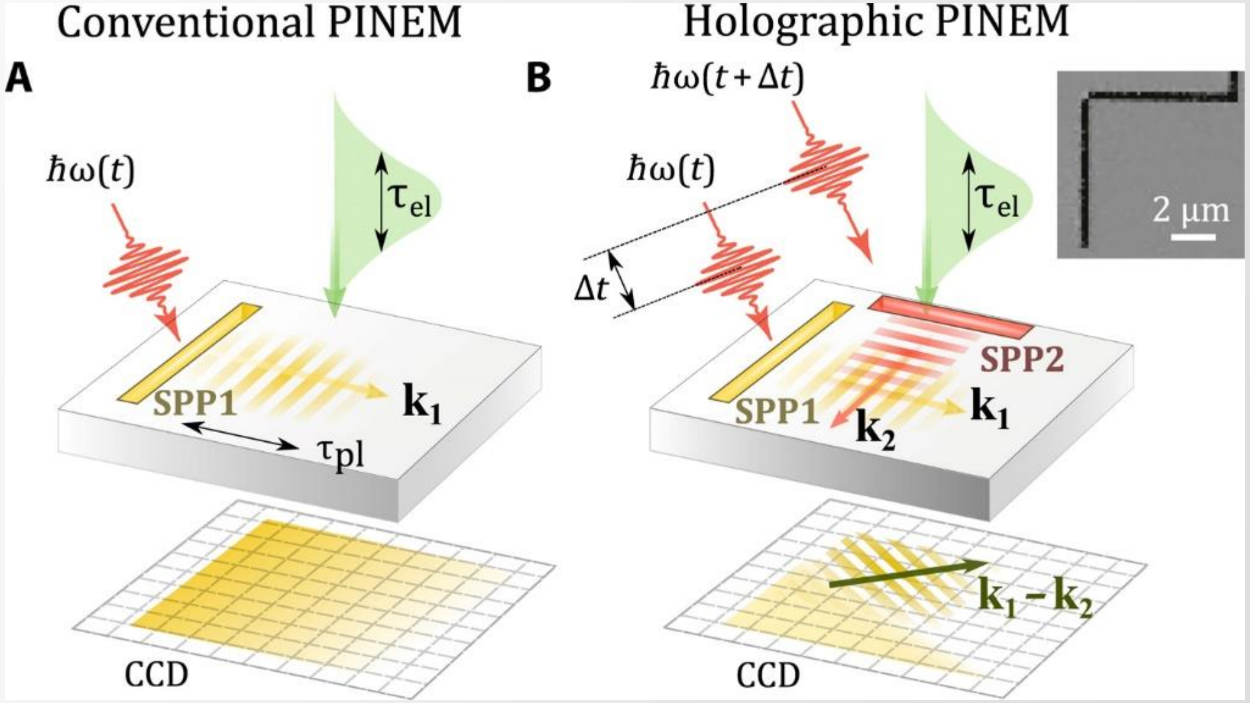
Holographic PINEM using gratings and optical gating

Holographic imaging of electromagnetic fields via electron-light quantum interference

I. Madan[1]*, G. M. Vanacore[1]*, E. Pomarico[1], G. Berruto[1], R. J. Lamb[2], D. McGrouther[2], T. T. A. Lummen[1†], T. Latychevskaia[1], F. J. García de Abajo[3,4], F. Carbone[1‡]



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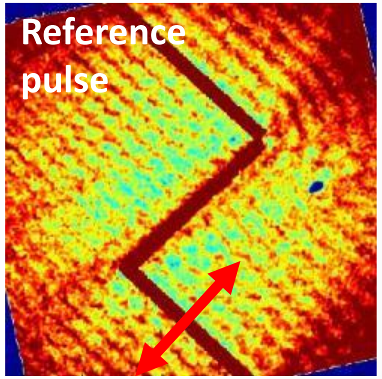
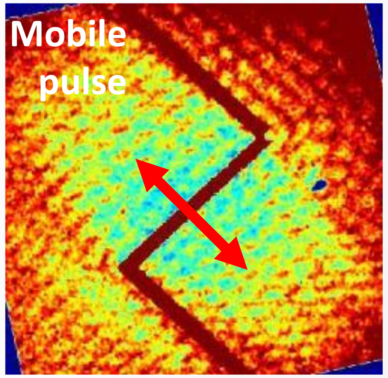
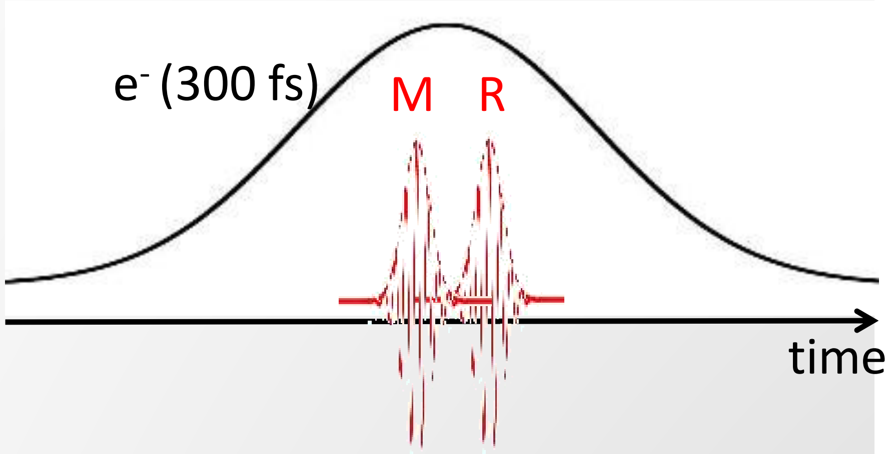
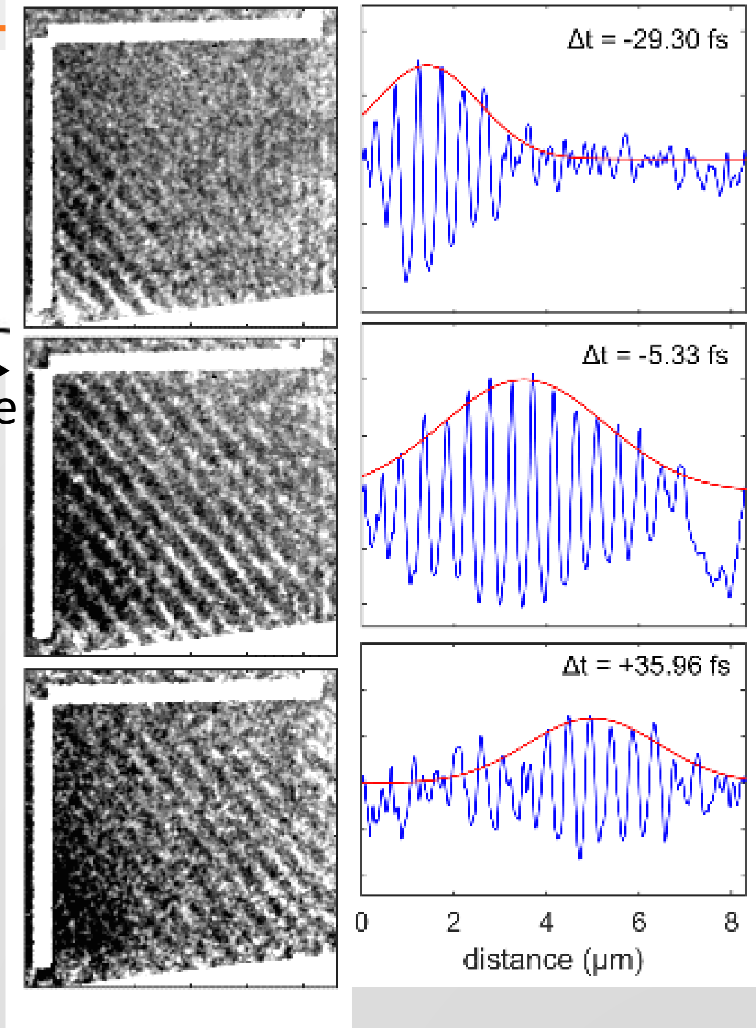
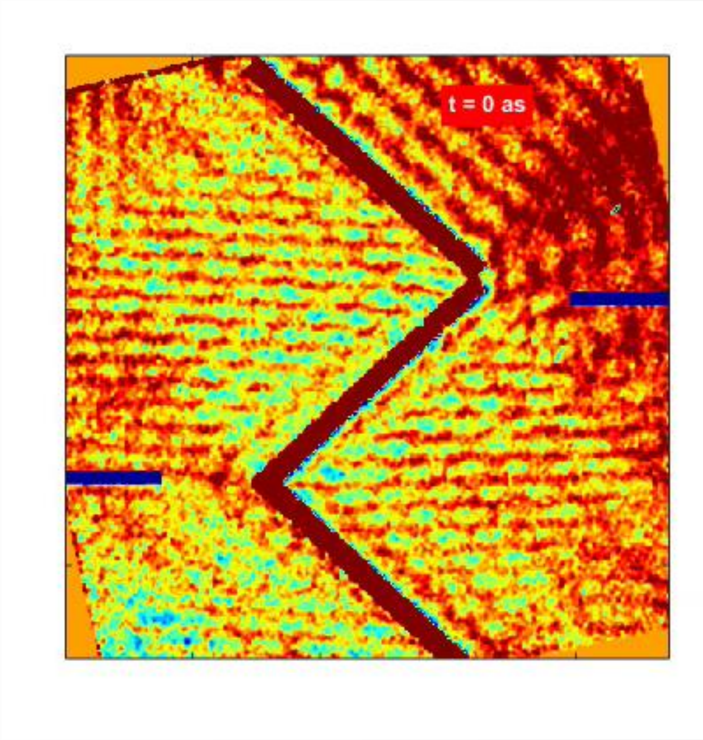
Holography of plasma resonance











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

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Time-resolved Vortex Electron beams generated by PINEM

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nature materials

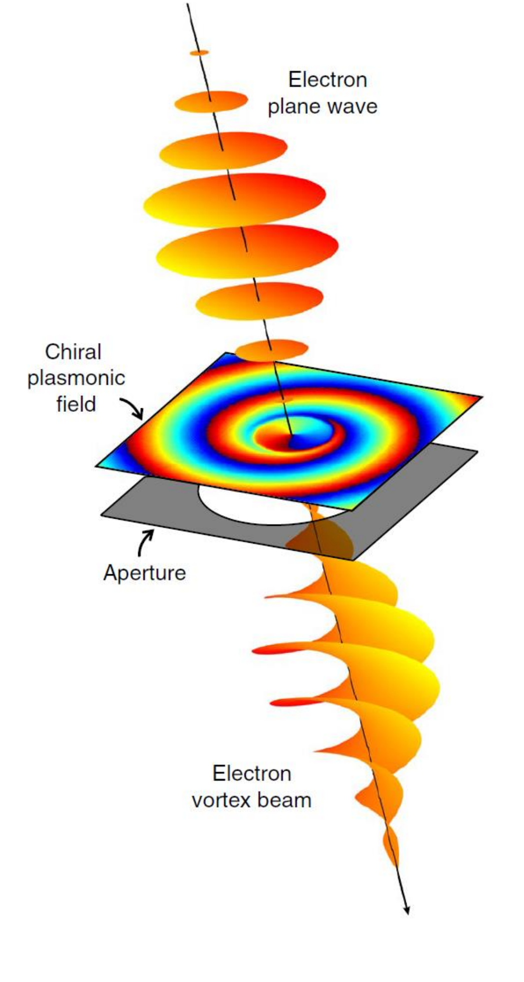
ARTICLES

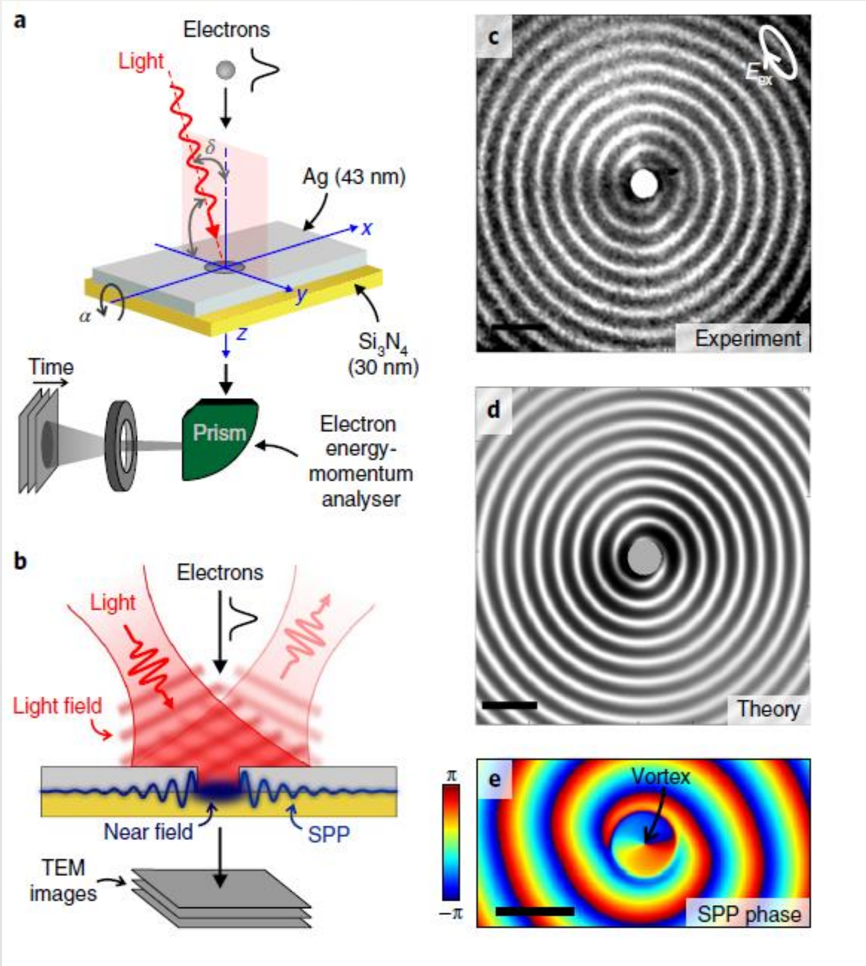
https://doi.org/10.1038/s41563-019-0336-1

Ultrafast generation and control of an electron vortex beam via chiral plasmonic near fields

G. M. Vanacore[1,10]*, G. Berruto[1,10], I. Madan[1], E. Pomarico[1], P. Biagioni[2], R. J. Lamb[3], D. McGrouther[3], O. Reinhardt[4], I. Kaminer[4], B. Barwick[5], H. Larocque[6], V. Grillo[7], E. Karimi[6], F. J. García de Abajo[8,9] and F. Carbone[1]

Using the holographic interference of light from a circular hole in the Ag film, a chiral plasmonic field develops that couples to the electron plane wave via the PINEM effect, creating an electron vortex beam carrying orbital angular momentum.





PINEM can be used to tailor the electron wave function!

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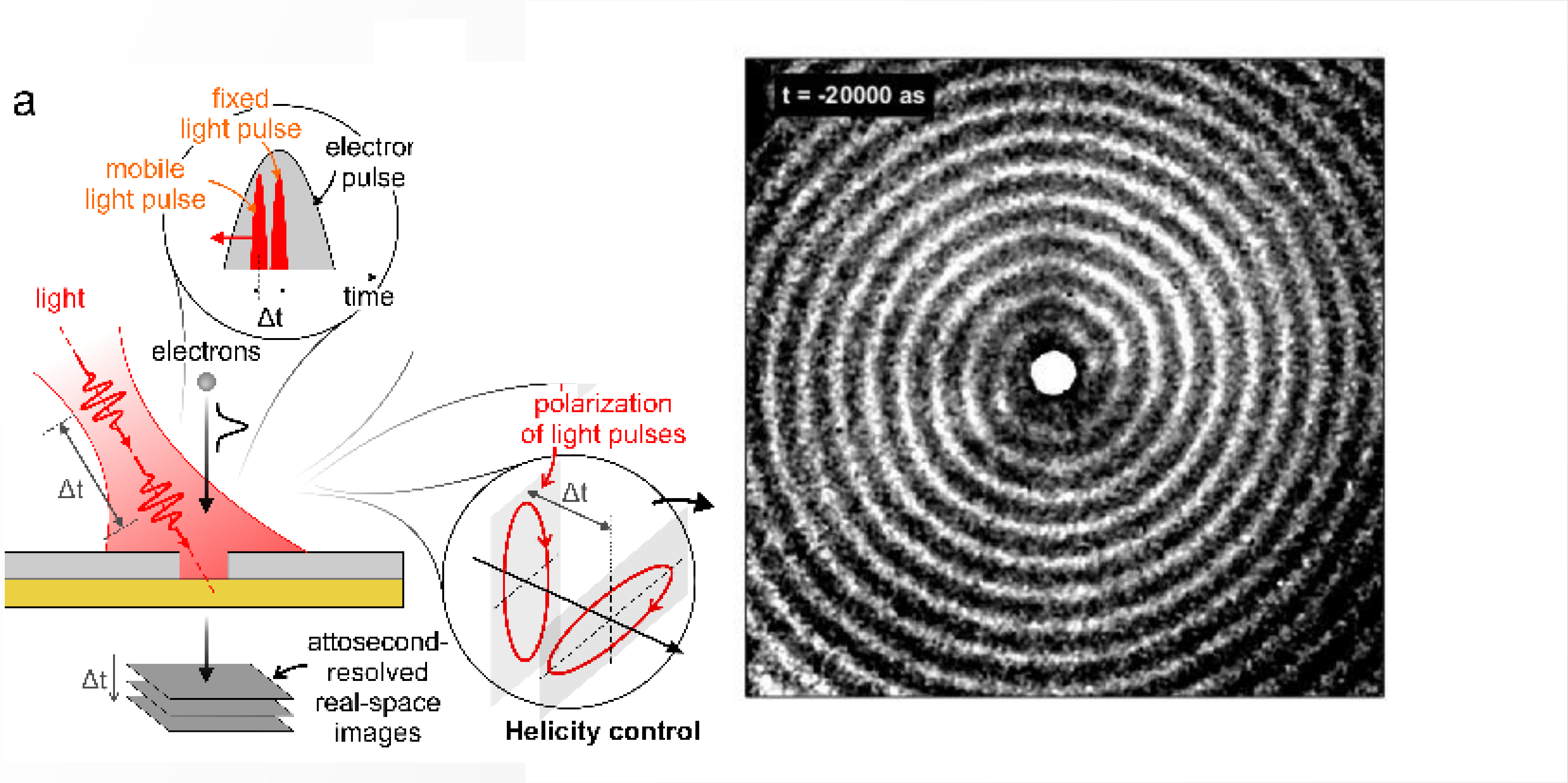
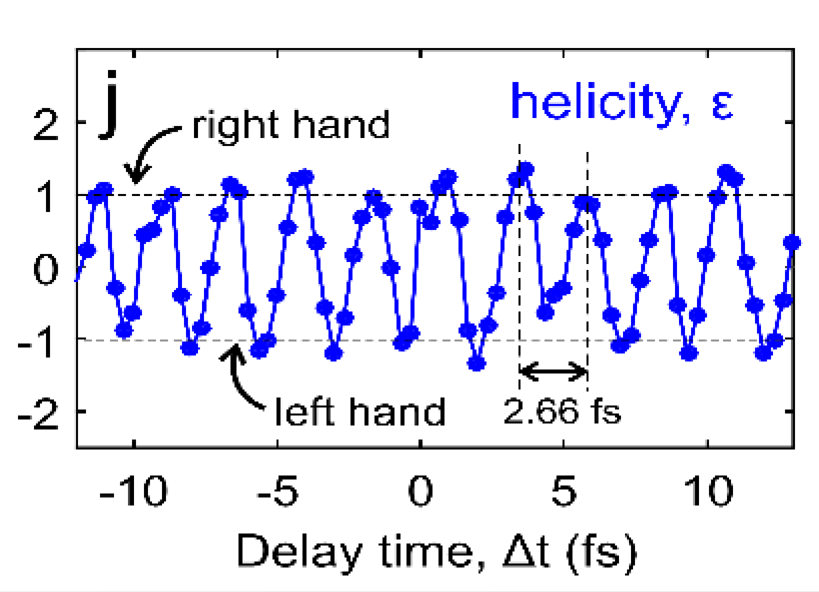
LUMES

Coherent control of vortex beams





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

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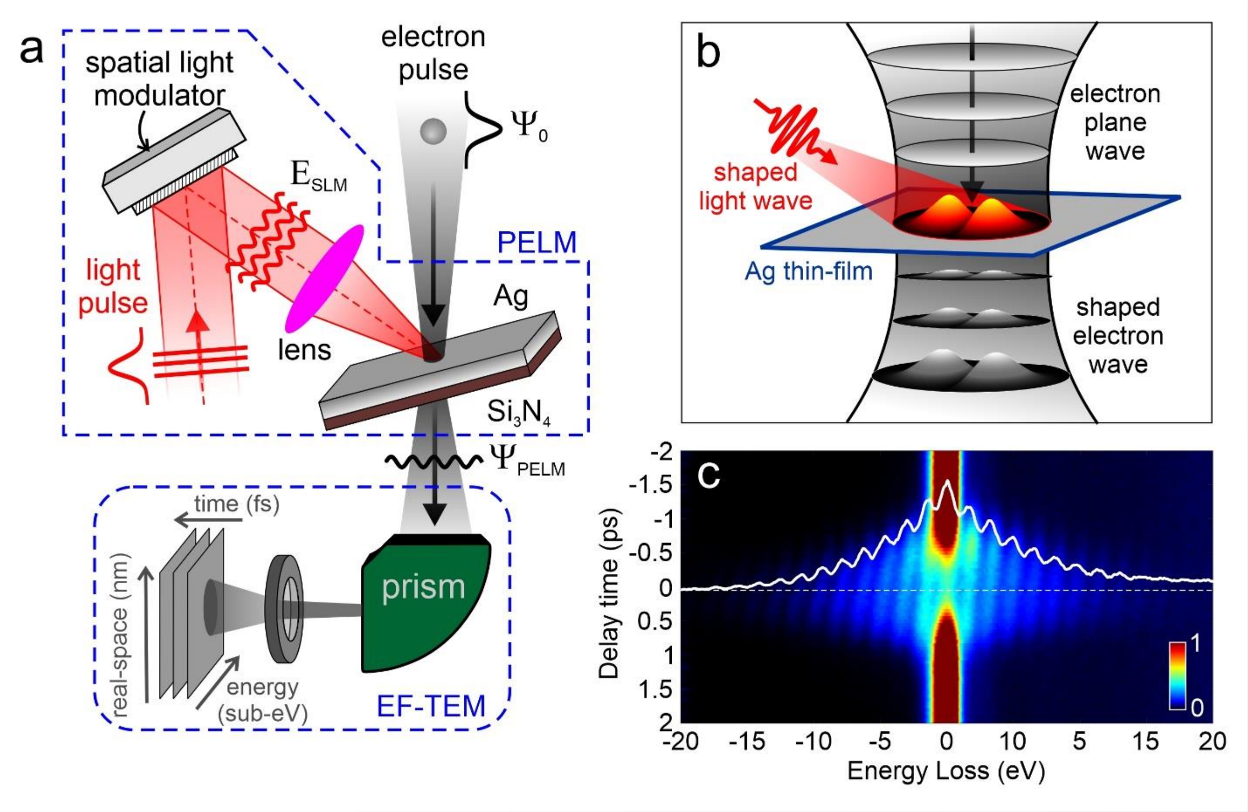
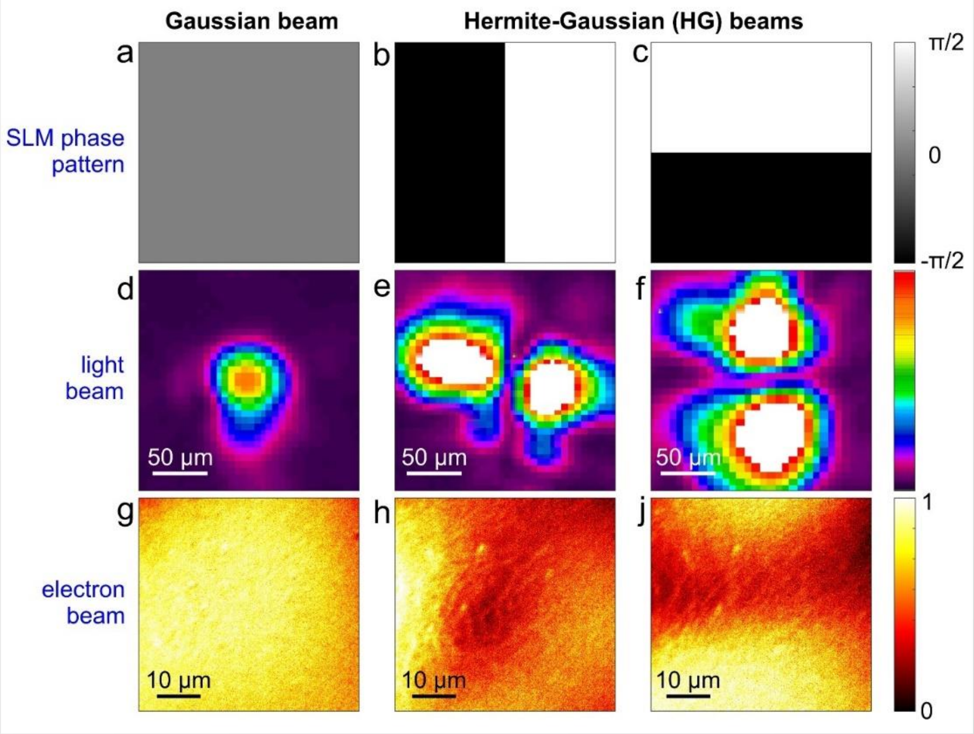
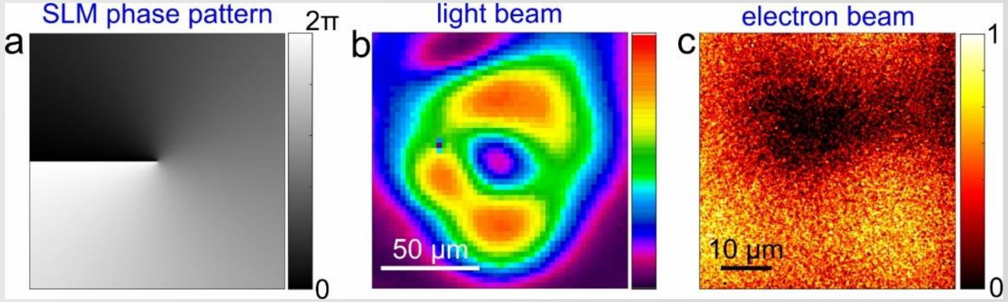
Patterned femtosecond light can be used to shape electron pulses, e.g, generate femtosecond vortex electron beams







Ultrafast Transverse Modulation of Free Electrons by Interaction with Shaped Optical Fields, I. Madan et. al, ACS Photonics 2022, 9, 10, 3215–3224

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Continuous Wave –PINEM in conventional microscopes using quasi-phase-matched silicon-photonic nanostructures

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RESEARCH ARTICLE SUMMARY

QUANTUM OPTICS

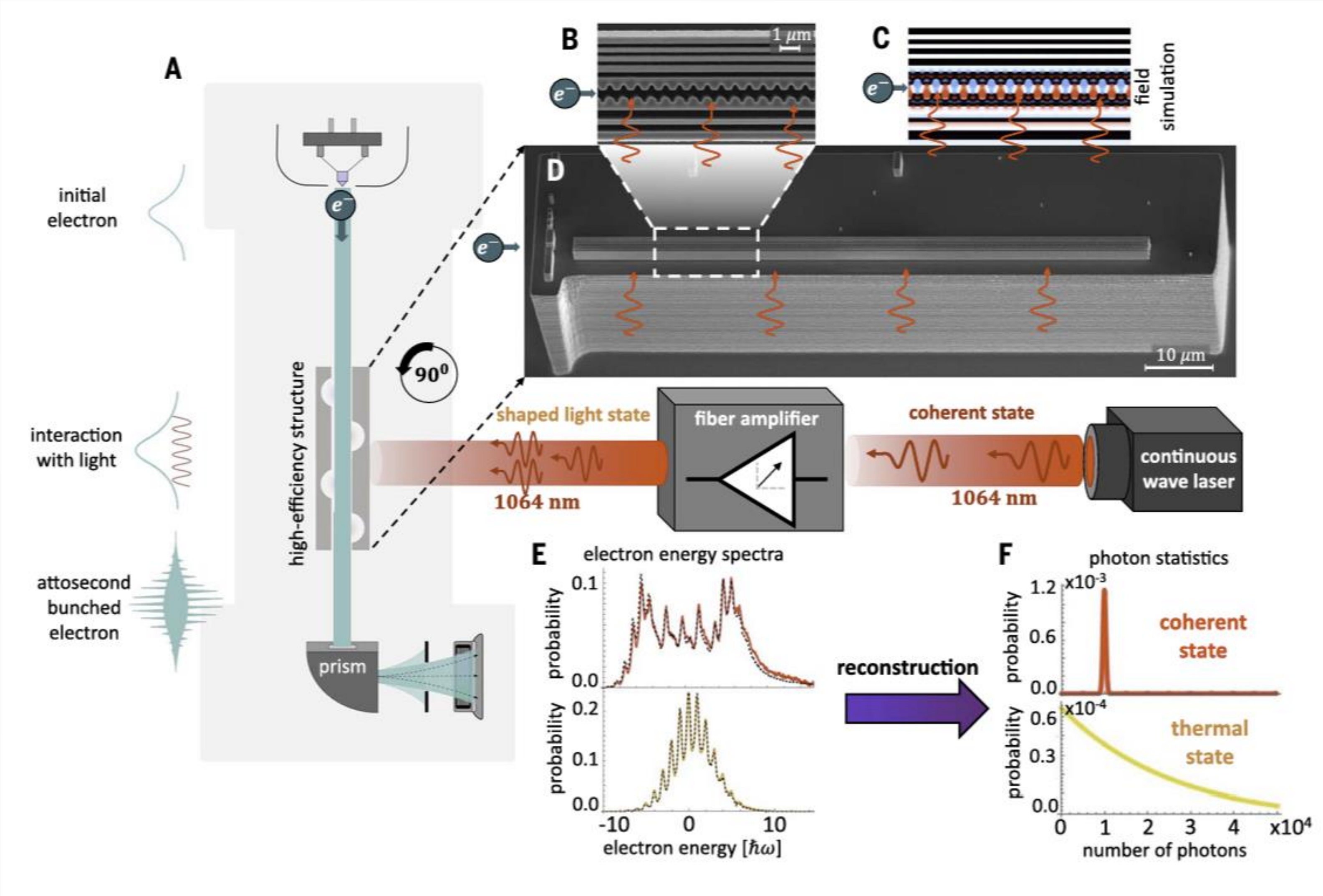
Imprinting the quantum statistics of photons on free electrons

Raphael Dahan†, Alexey Gorlach†, Urs Haeusler†, Aviv Karnieli†, Ori Eyal, Peyman Yousefi, Mordechai Segev, Ady Arie, Gadi Eisenstein, Peter Hommelhoff, Ido Kaminer*

Dahan et al., Science 373, 1324 (2021)

They could observe quantum statistics effects of photons on free-electron–light interactions and the transitions from quantum walk to classical random walk on the free-electron energy ladder.

- Attosecond metrology
- Quantum optics applications
- Quantum tomography of light
- Ramsey Holography



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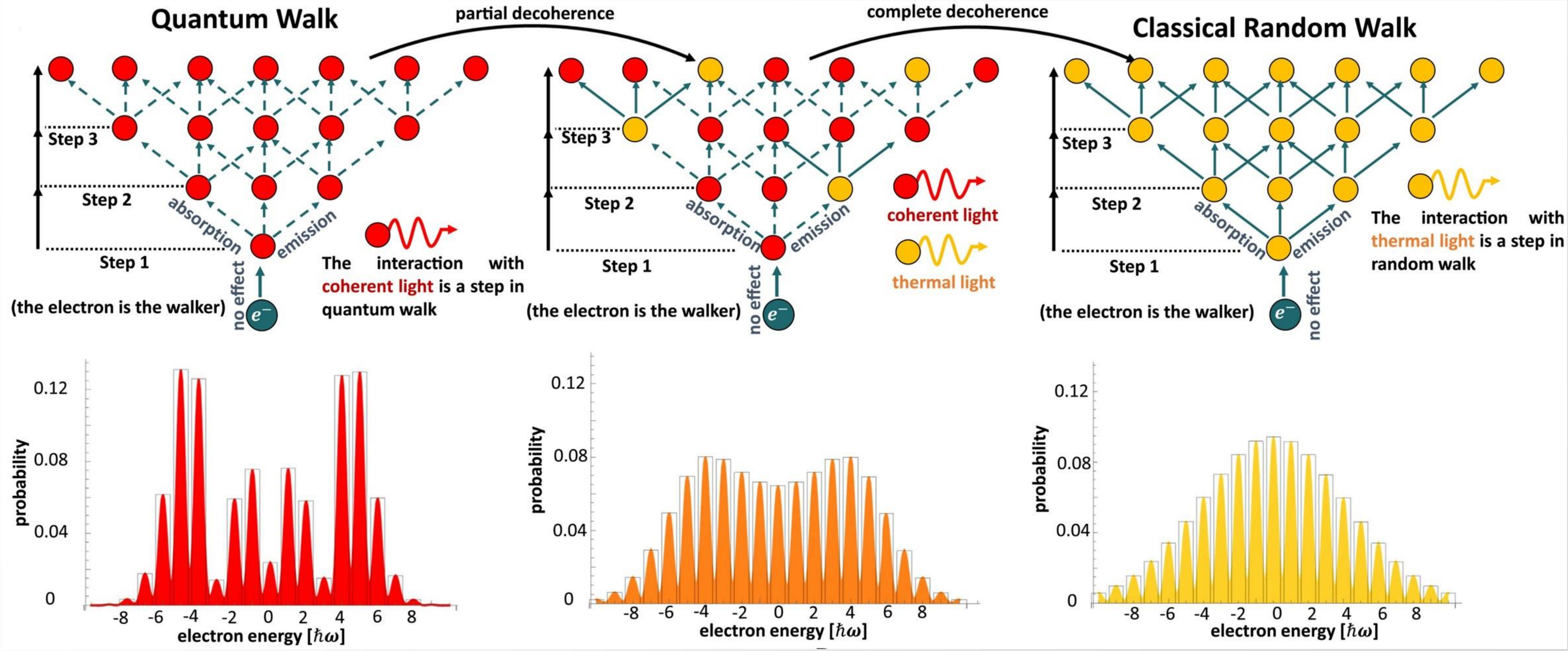
LUMES

Quantum walk and coherent phase modulation of free electrons by light



R. Dahan et. al., Imprinting the quantum statistics of photons on free electrons. Science 373 (6561), 2021

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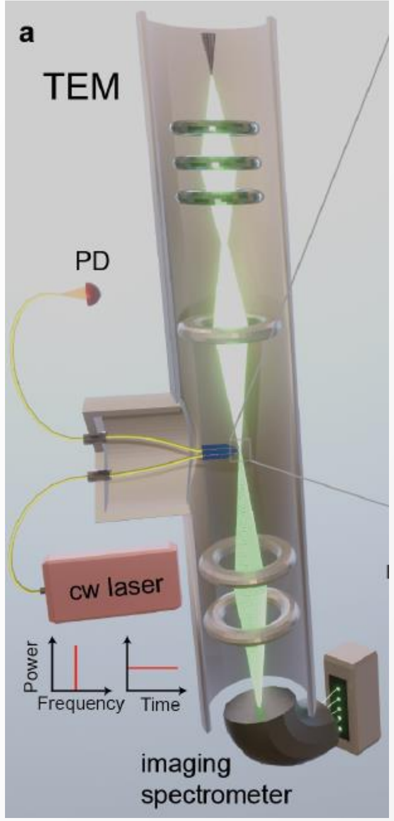
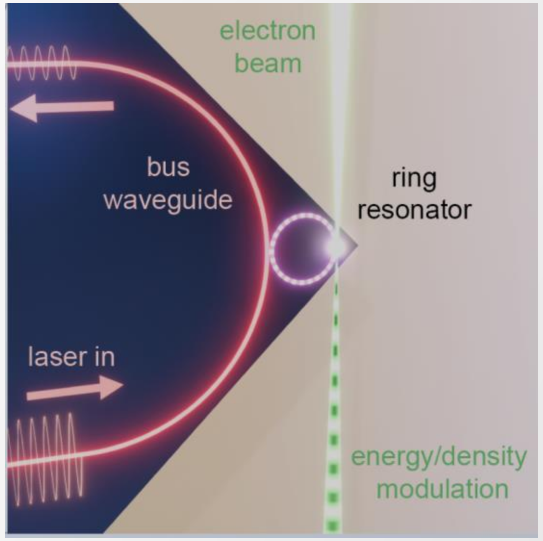
High-Q Microring Resonators provide strong coupling of photons with electron and enable CW-PINEM

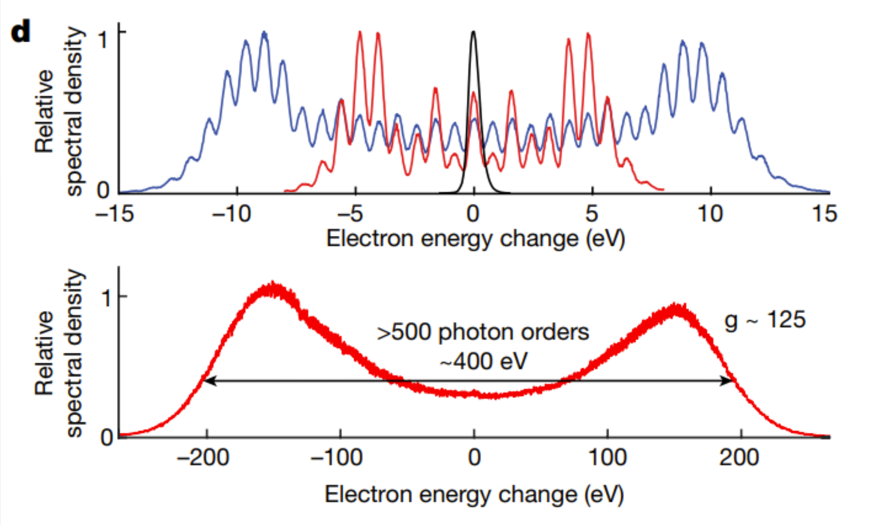


Integrated photonics enables continuous-beam electron phase modulation

https://doi.org/10.1038/s41586-021-04197-5
Received: 7 May 2021
Accepted: 1 November 2021

Jan-Wilke Henke[1,2,5], Arslan Sajid Raja[3,4,5], Armin Feist[1,2], Guanhao Huang[3,4], Germaine Arend[1,2], Yujia Yang[3,4], F. Jasmin Kappert[1,2], Rui Ning Wang[3,4], Marcel Möller[1,2], Jiahe Pan[3,4], Junqiu Liu[3,4], Ofer Kfir[1,2], Claus Ropers[1,2]✉ & Tobias J. Kippenberg[3,4]✉





- Phase matching between 120-keV electrons and the modes of a Si3N4 microresonator -> greatly enhanced interaction distance
- Allows for performing the PINEM experiments in CW mode of a TEM (normal TEMs)
- Prospective for electron beam shaping, single electron detector technology, etc.

Electron Spectrometer Calibration

ELSEVIER

Ultramicroscopy 55 (1994) 43–54

ultramicroscopy

Characterization of an analytical electron microscope with a NiO test specimen

R.F. Egerton, S.C. Cheng

Physics Department, University of Alberta, Edmonton, Canada T6G 2J1

Received 20 October 1993; in final form 31 March 1994

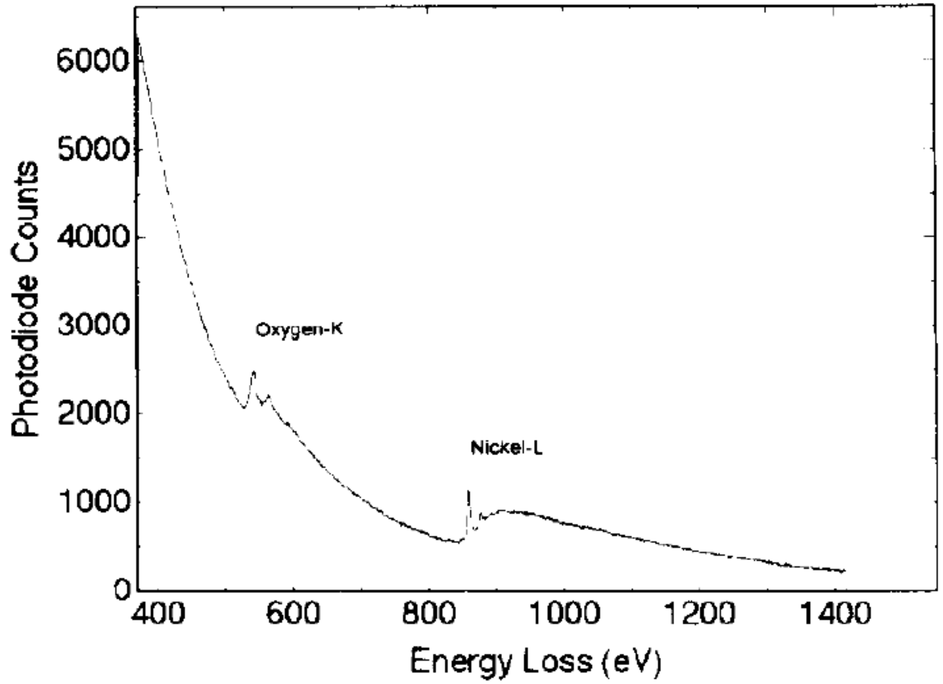
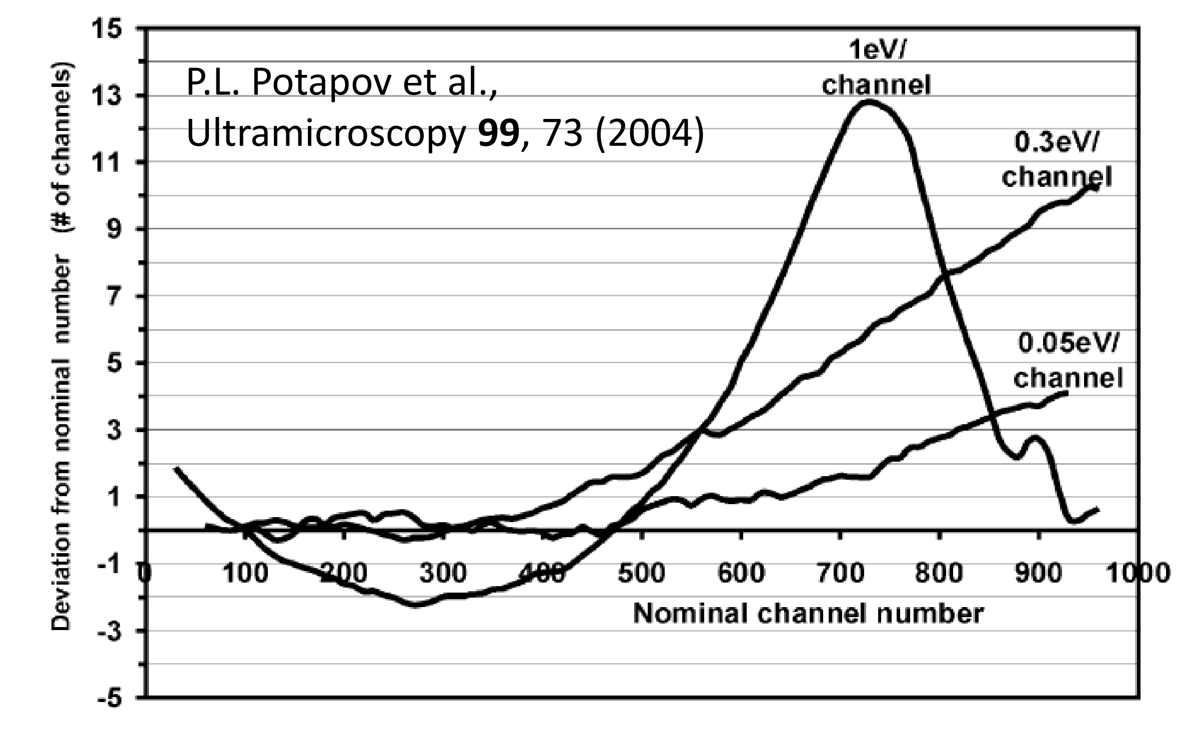


Fig. 2. Core-loss region of the electron energy-loss spectrum of a 47 nm NiO specimen, recorded using the Gatan 666 parallel-recording spectrometer with 200 keV electrons and 14 mrad collection semi-angle.

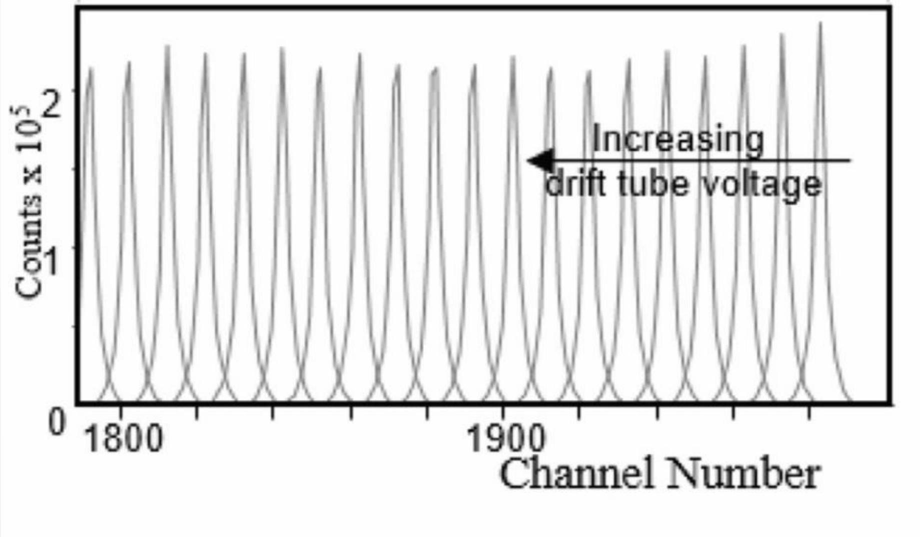
Manufacturers verify the dispersion calibration using white lines Ni L-edge and O K-edge from known NiO sample composition

Current calibration methods fall short…

NiO standard and commercial techniques
- Calibration precision is realistically ~1%
- No way to measure nonlinearity of spectrometer



Cumulative errors can be hundreds meVs to serval eVs!



Drift Tube Scan of the ZLP across spectrometer detector
- Relative calibration precision of 0.03%
- Measures nonlinearities
- Vulnerable to instrument instabilities
- No real-time drift correction
- Time consuming

R.W.H. Webster et al., Ultramicroscopy **217**, 113069 (2020)

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Electron Spectrometer Calibration device

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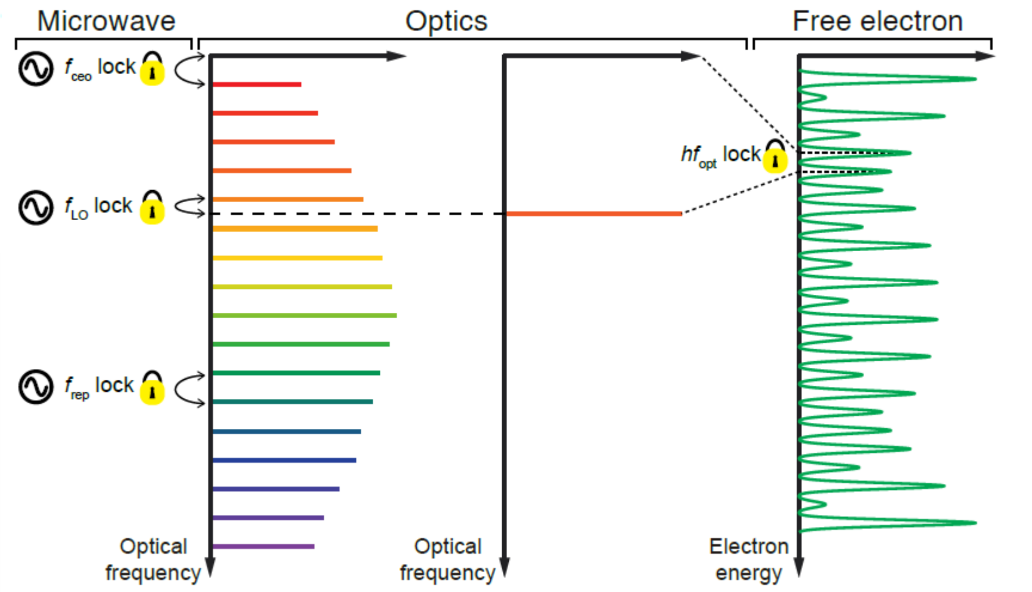
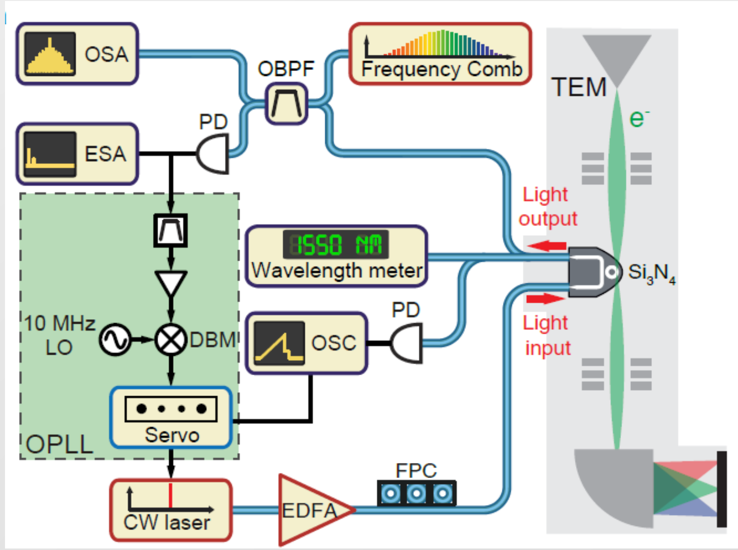
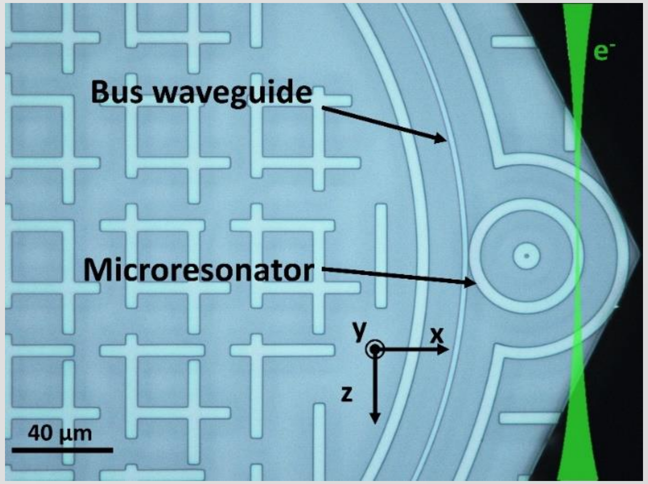
CWPINEM is useful tool for frequency based metrology with electrons

"High-Q"-10^6 microring resonators can modulate the longitudinal phase of free electrons in conventional TEM **(CW-PINEM) that act like an energy ruler**





By making a coherent frequency link between optical and CWPINEM, one can calibrate the dispersion of electron spectrometers with **~10 µeV/ch precision**



This method can also be used to **correct nonlinearities and drift with µeV/ch precision**

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Summary

- Light and electrons can interact under special conditions in which their phase velocities are similar (phase matching)
- Under "phase-matched" conditions, electrons can absorb and emit photons modulating the electron energy with quantized photon energy- PINEM spectra with sidebands spaced by the photon energy.
- Using the TEM, PINEM can achieve high spatial (nanometer) and temporal (attosecond) resolution, allowing the study of the interaction between light and matter with high precision.
  - Applications for Plasmonics devices: For observing surface plasmon resonances and understanding the behavior of metallic nanostructures
  - Application Photocatalysis: Examination of photocatalytic processes at the nanoscale for developing efficient catalytic materials.
  - Applications in Nanophotonics: For studying of optical antennas that can enhance light emission and absorption at the nanoscale.
  - Applications in Low Dose Imaging: Ramsey Holography, Ghost imaging, shaped electron beams–chiral electron beams

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QUESTIONS?

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LUMES