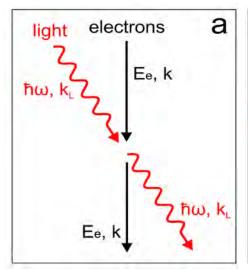
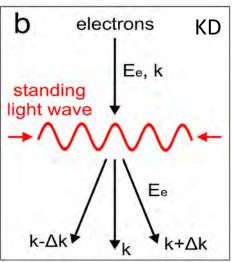
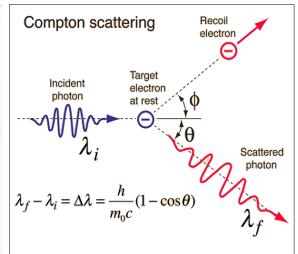
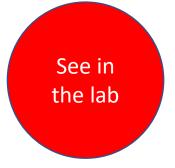
## Coherent control of light electrons interaction

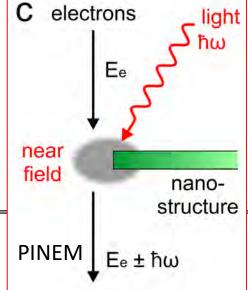
How can light and e-interact?

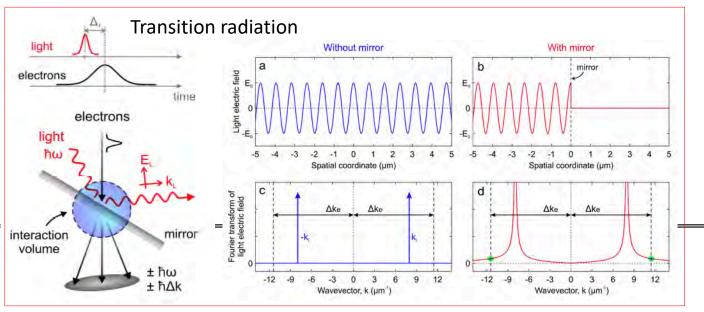




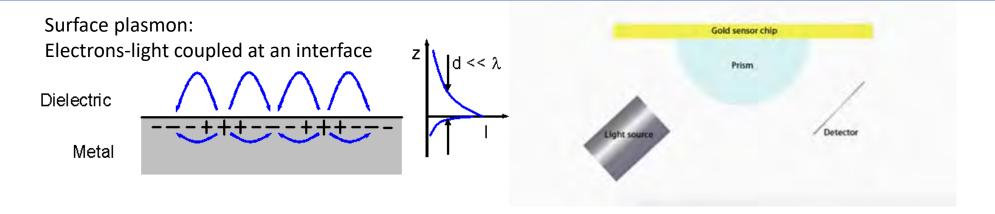




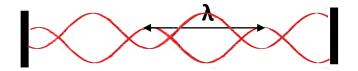




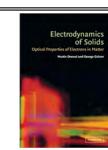
## Surface plasmons polaritons and plasma resonances

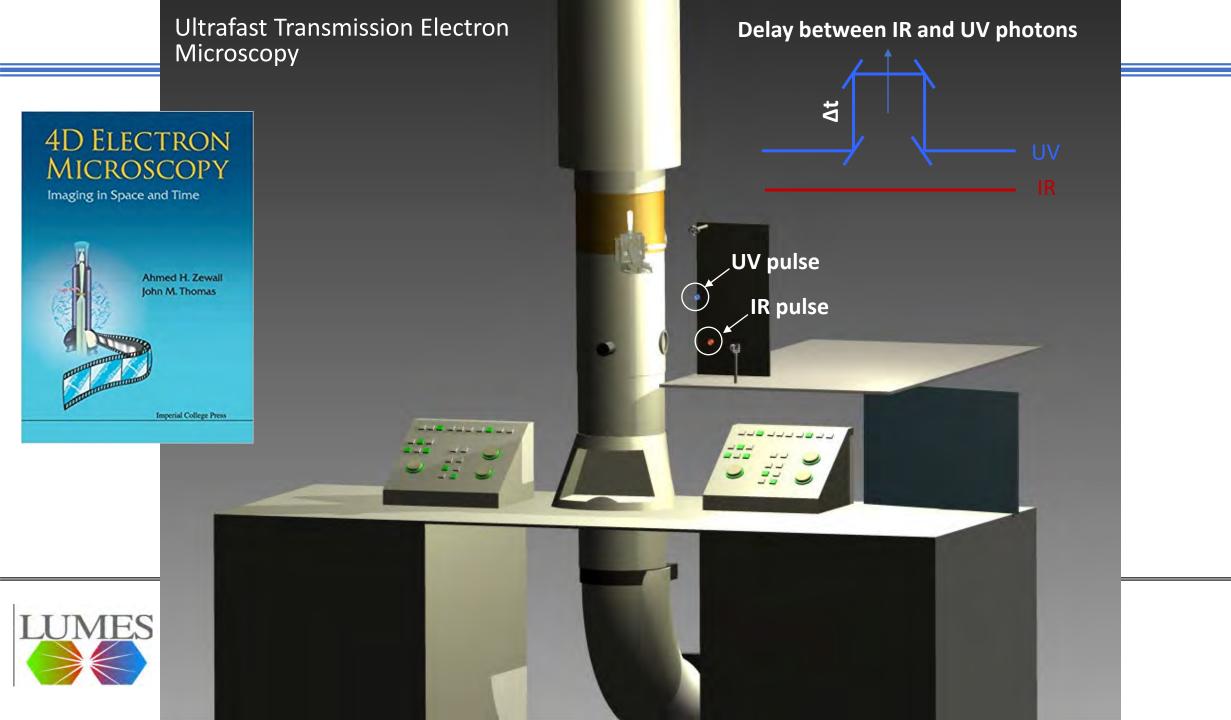


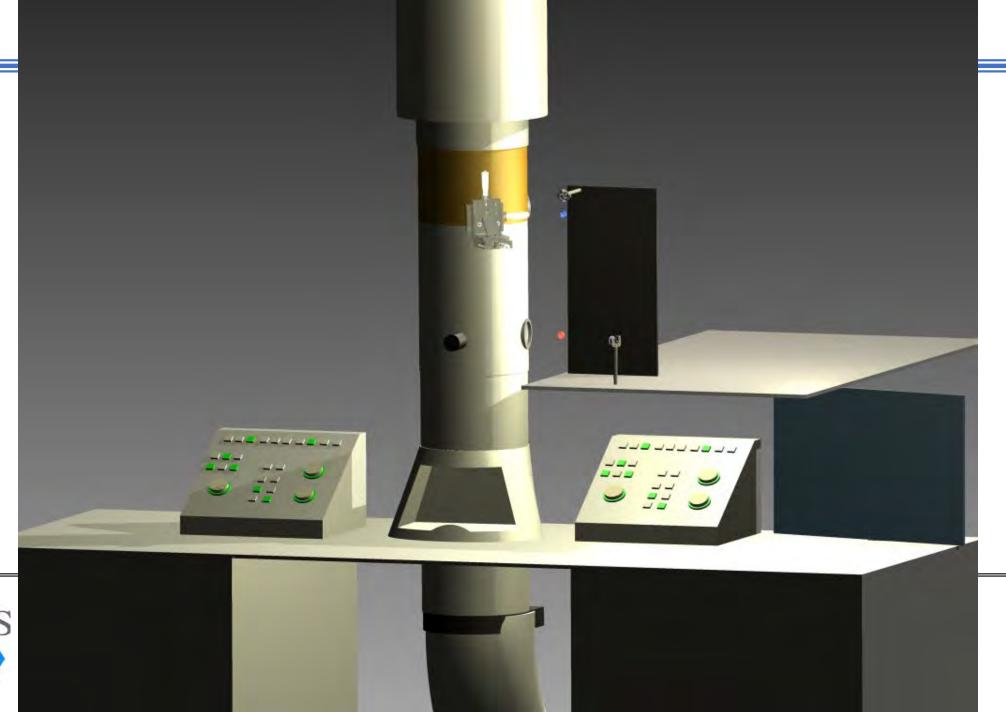
## Plasma resonance



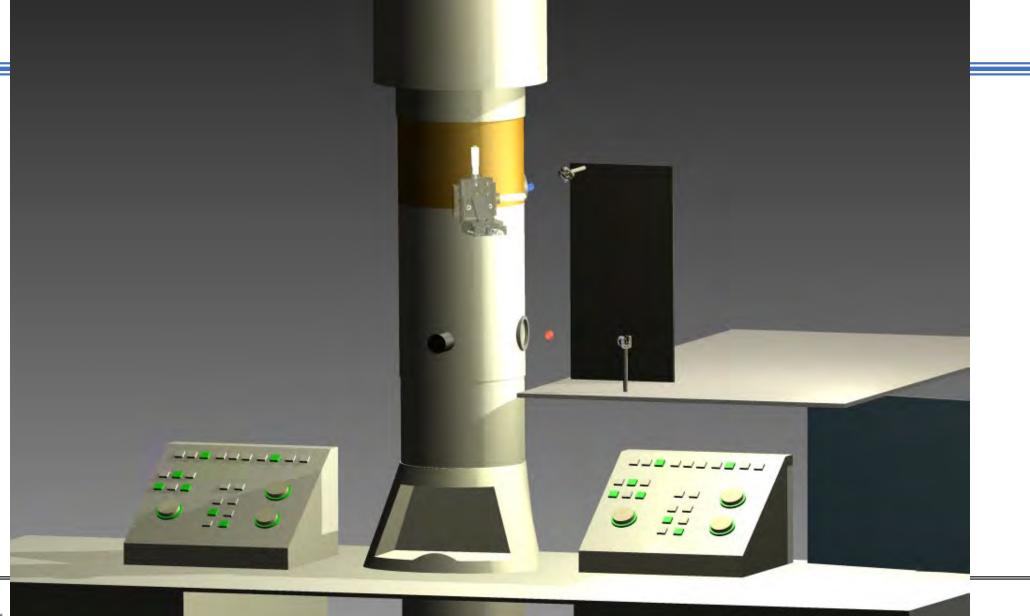




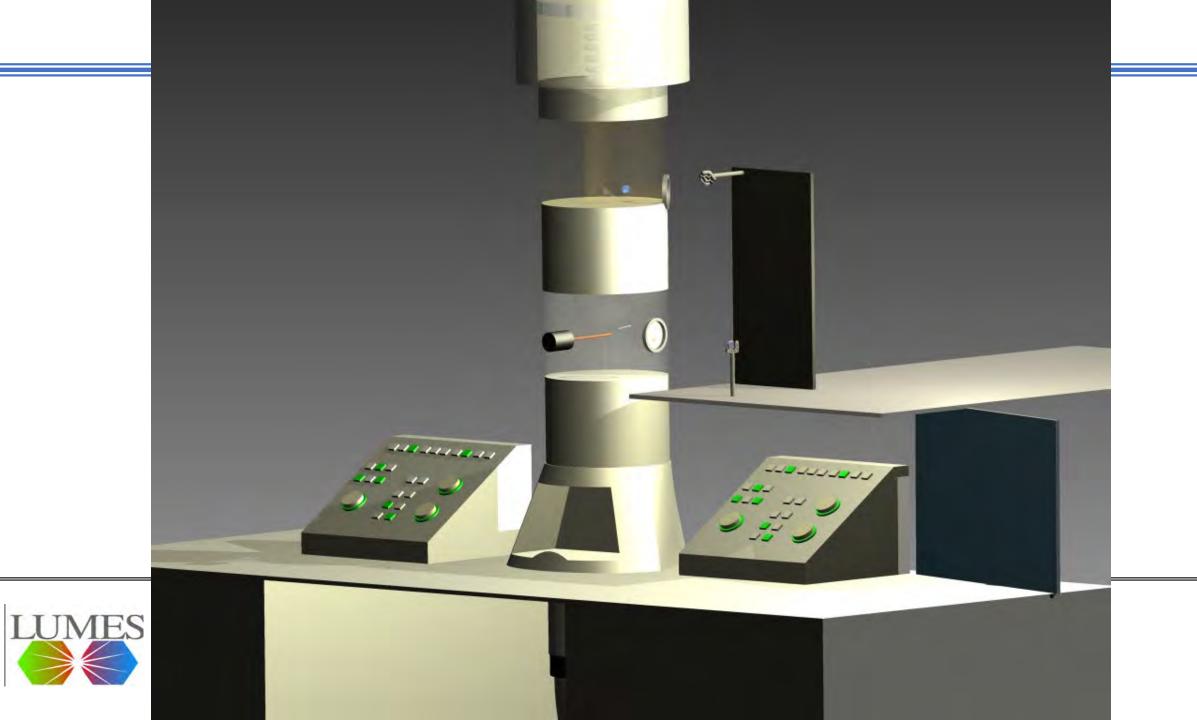


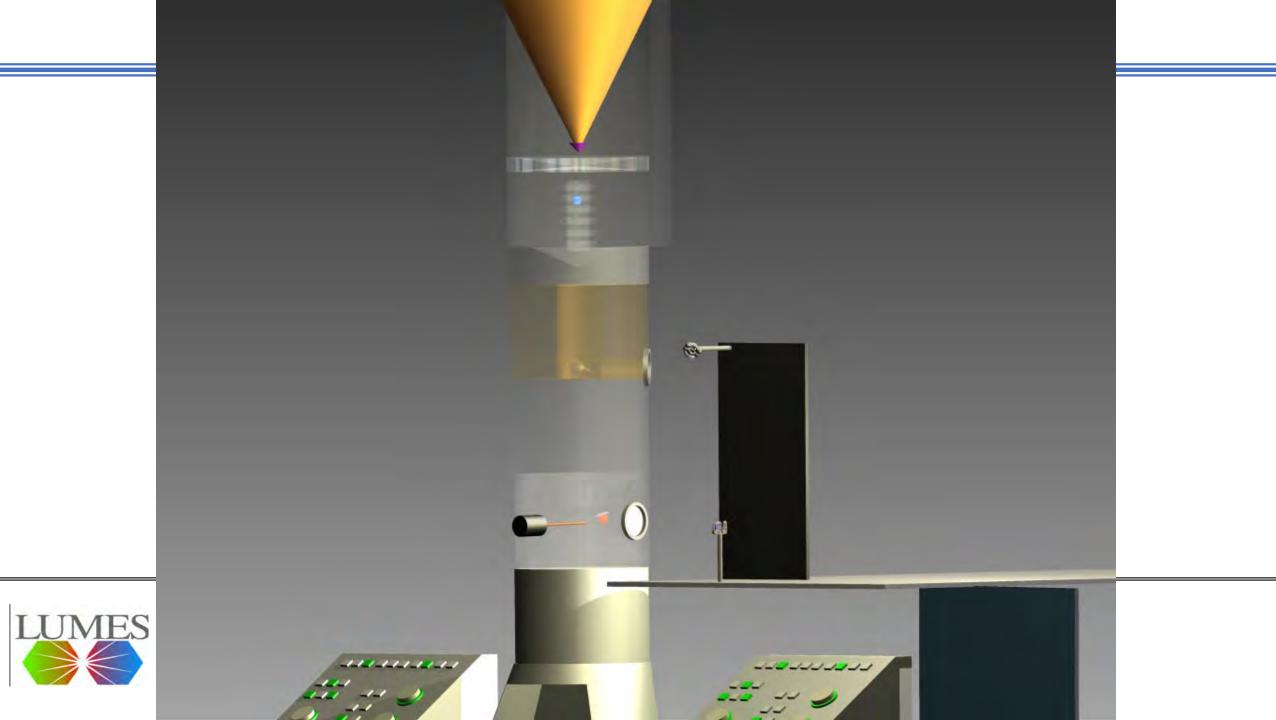


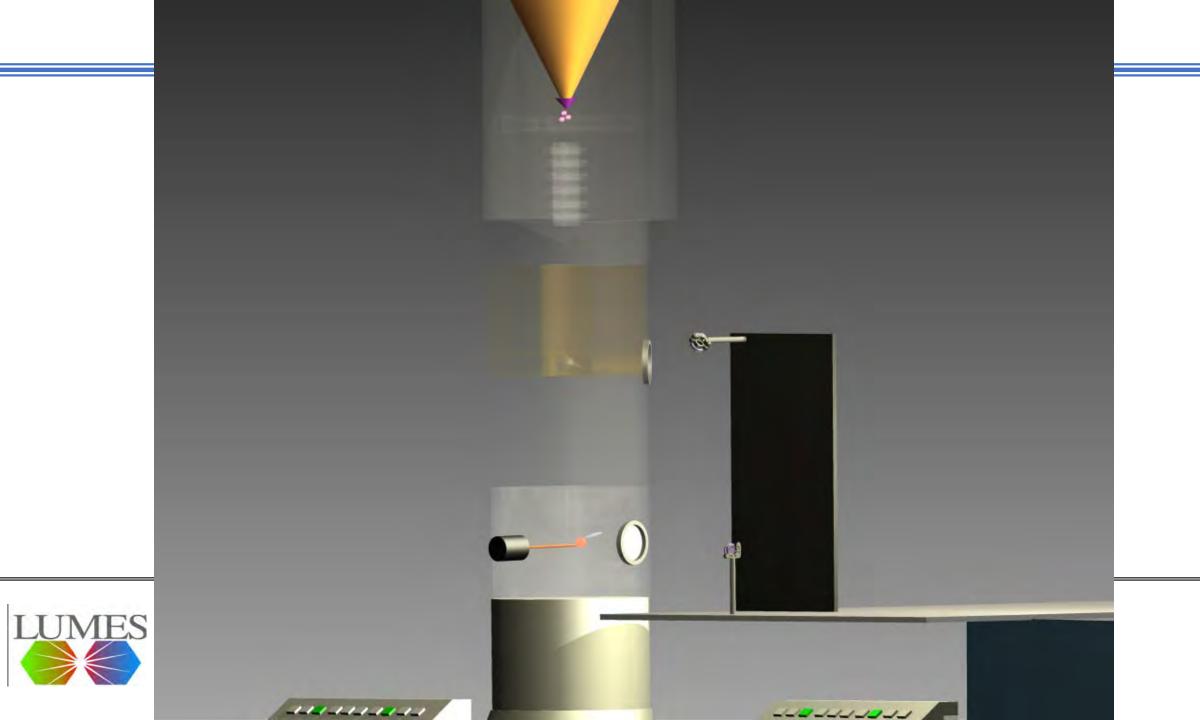


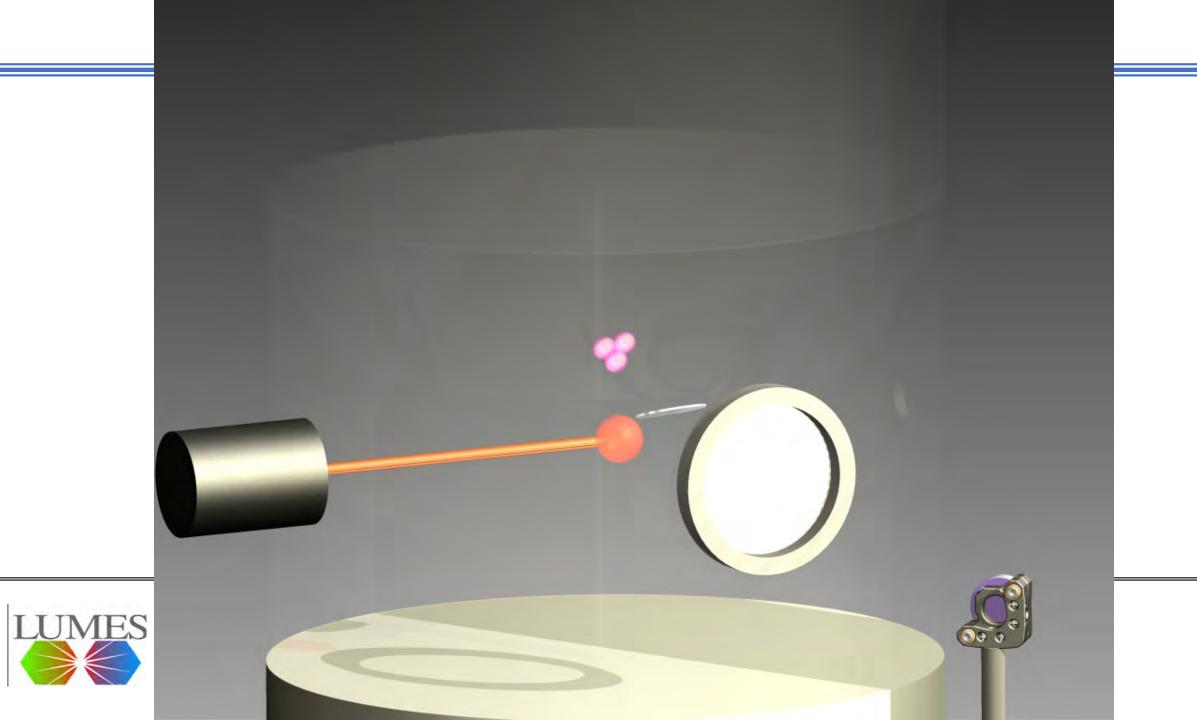


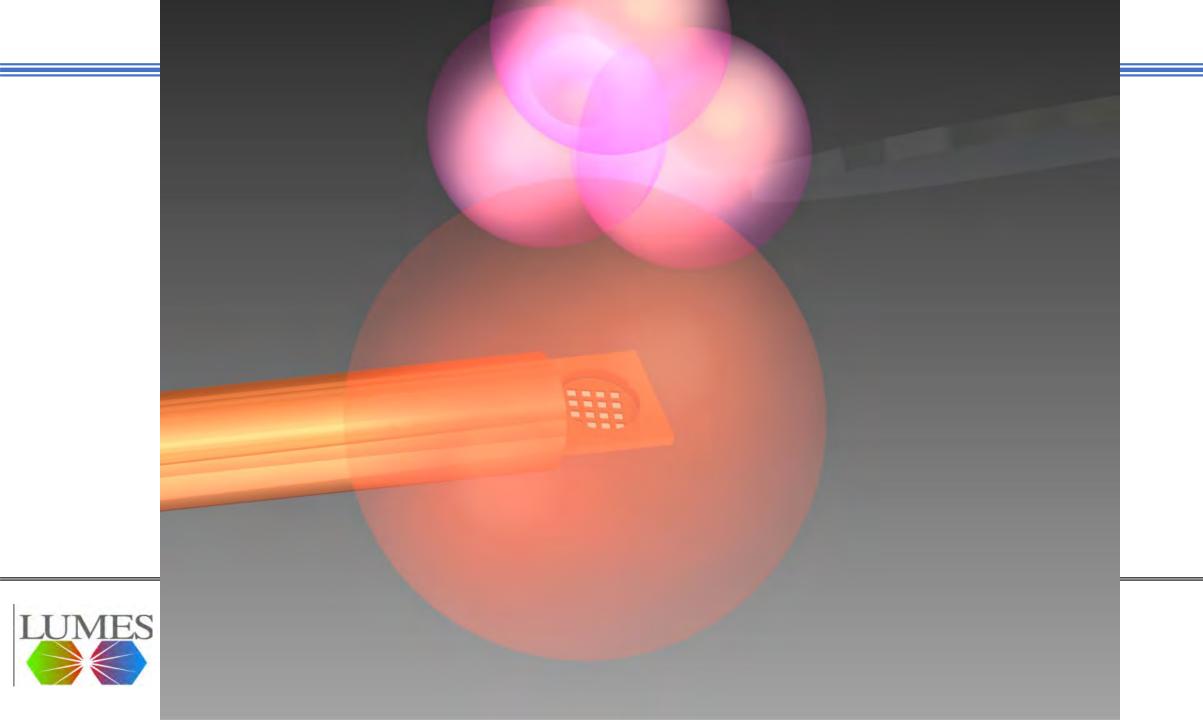


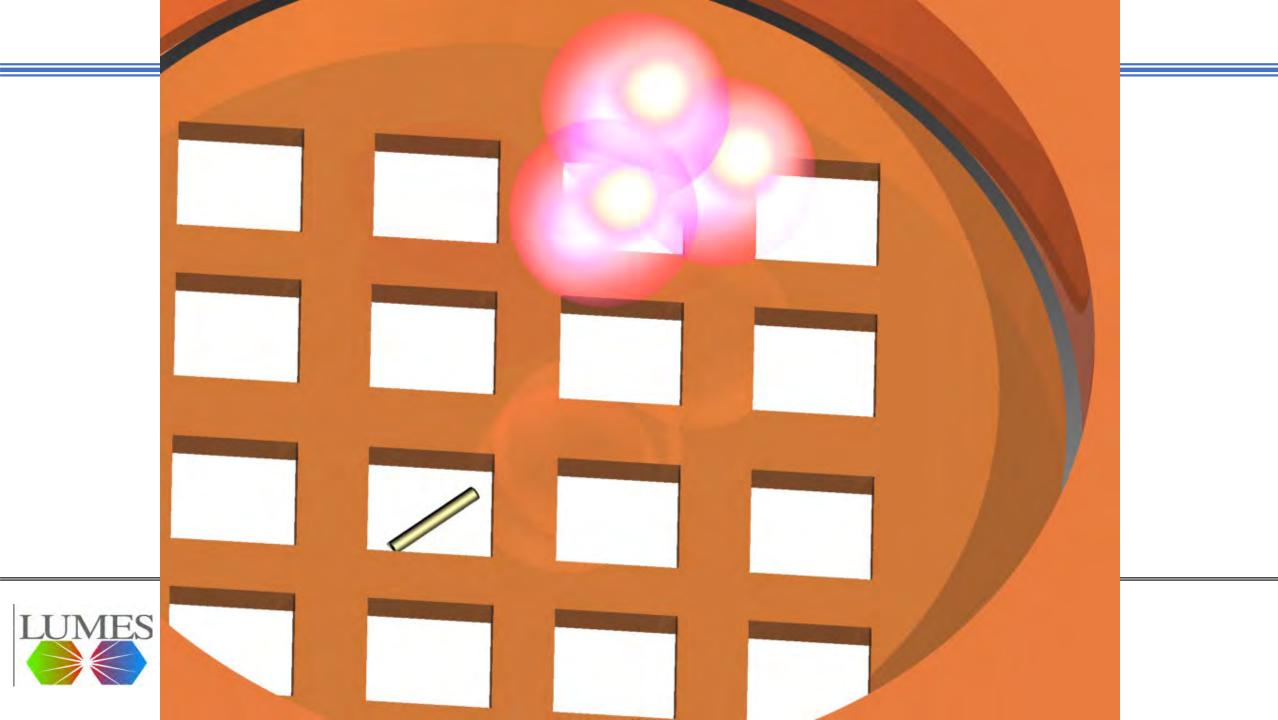


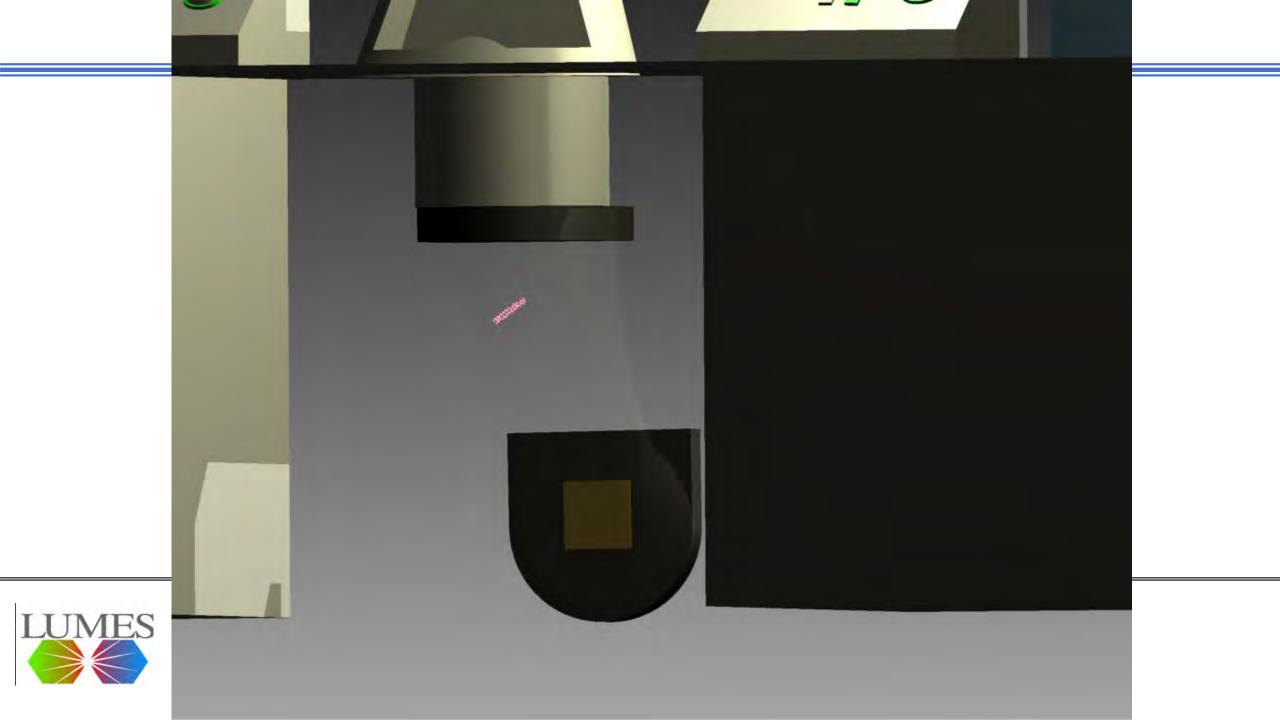


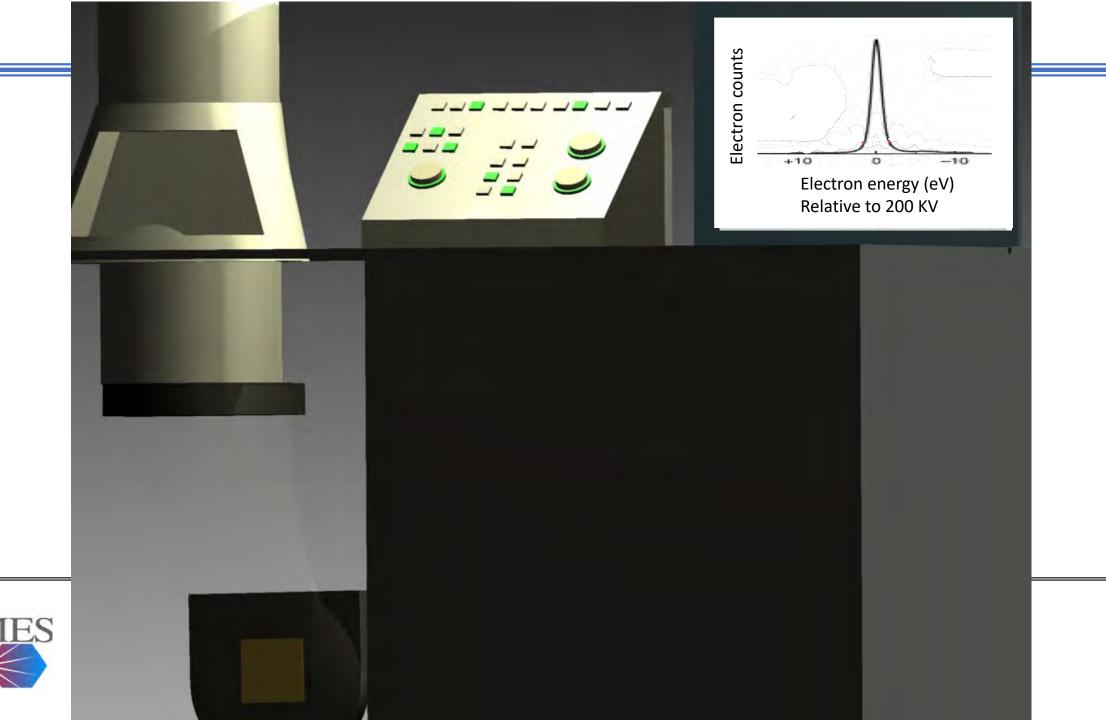


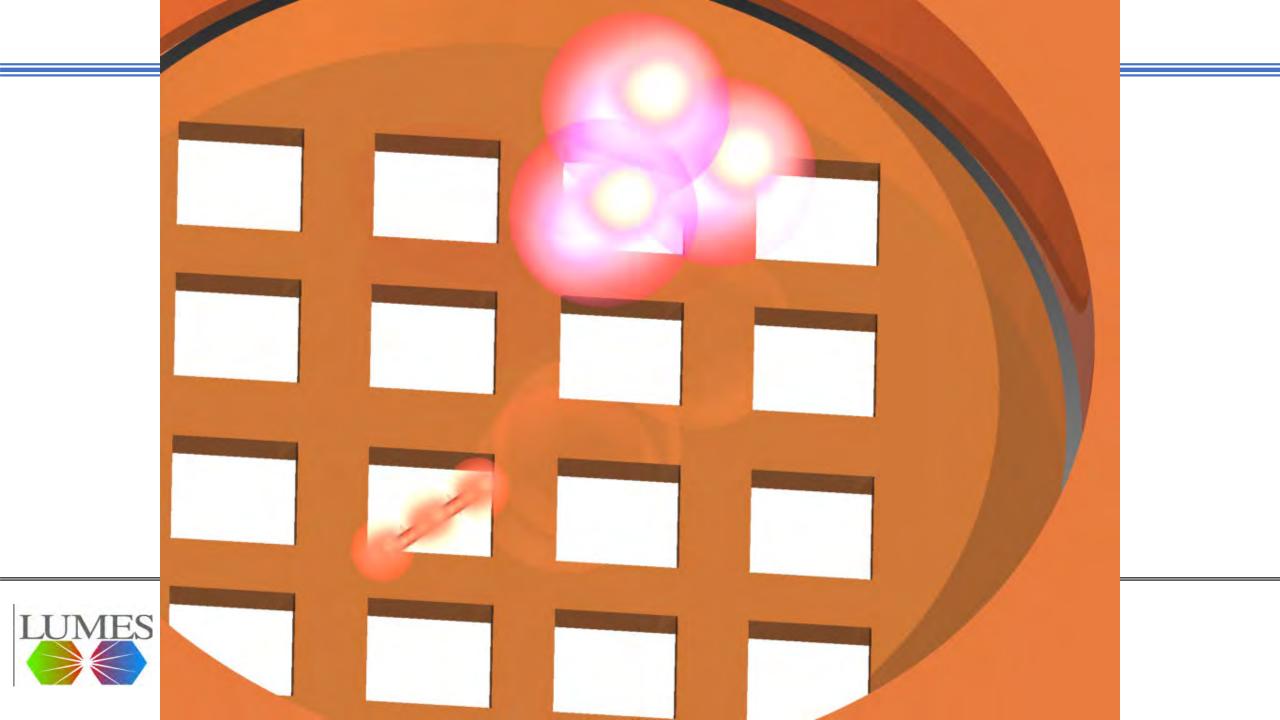


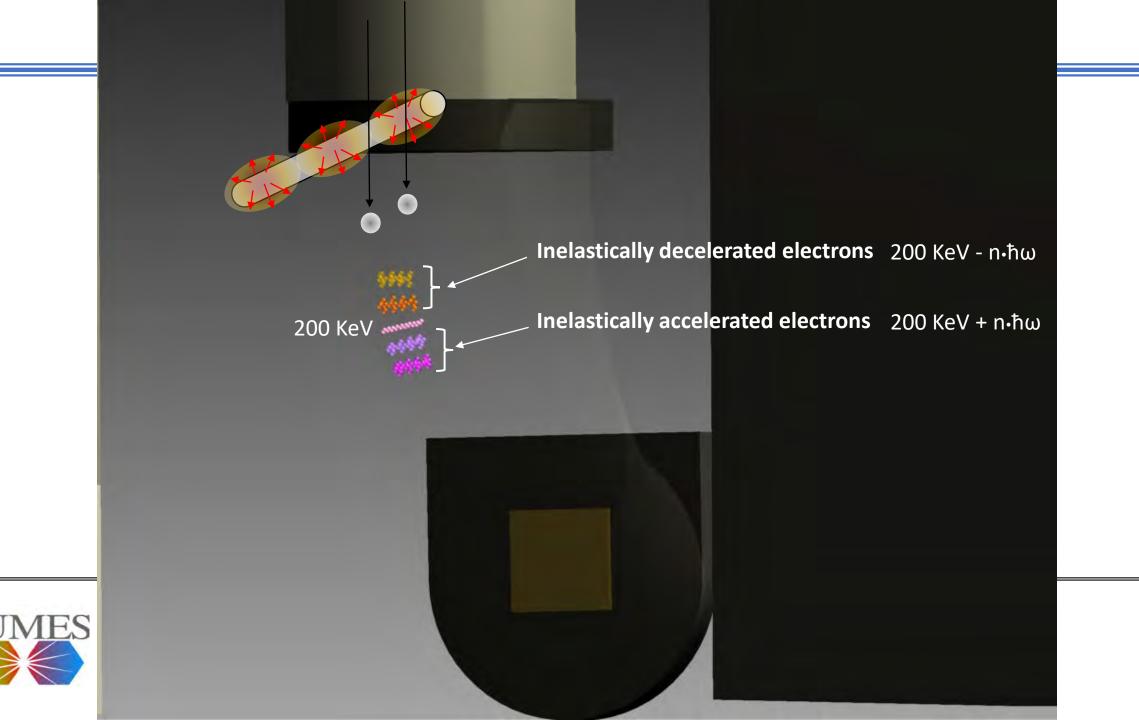


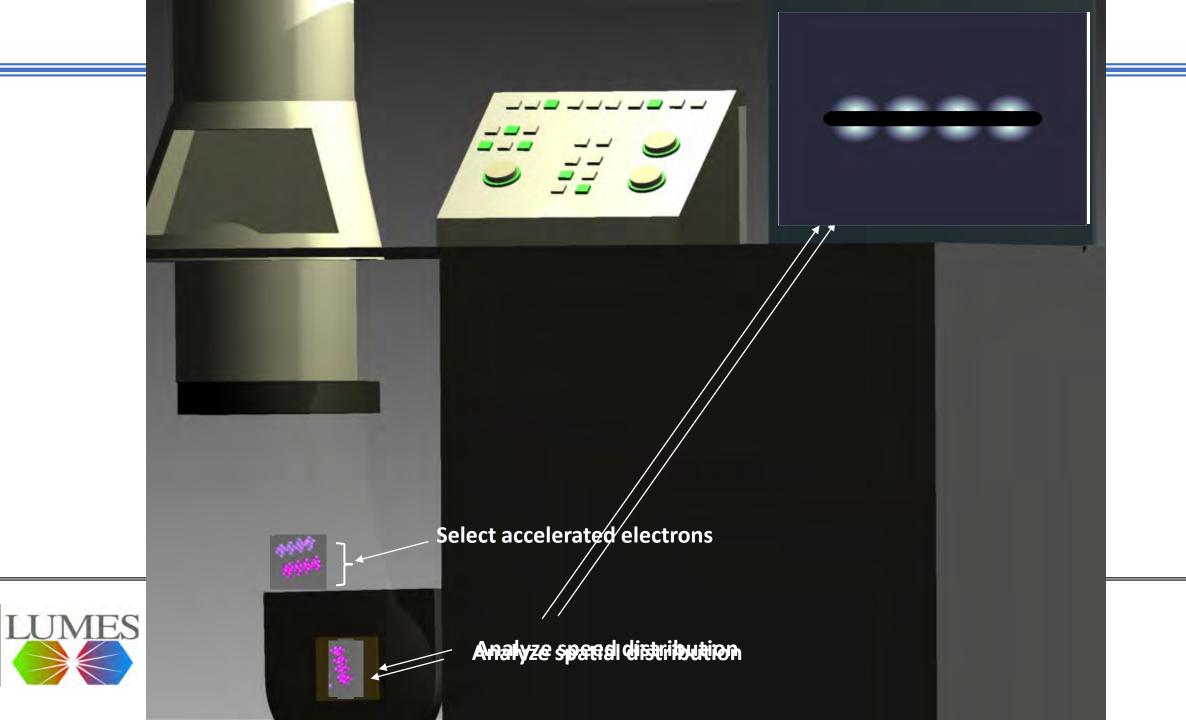




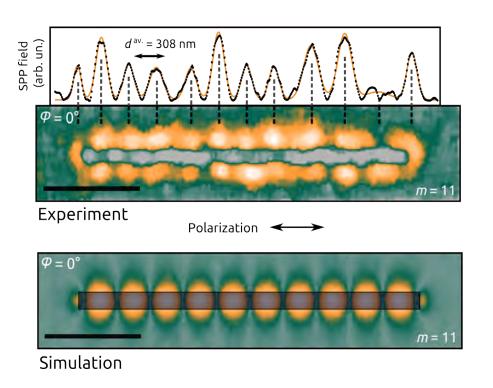


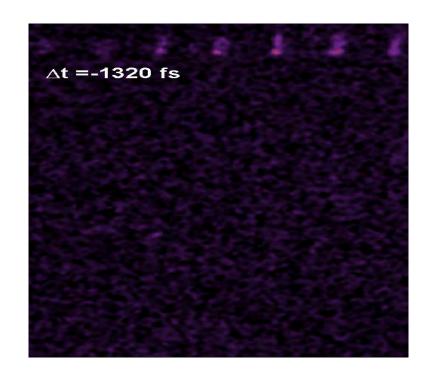






## Filming plasma resonances











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

Most cited article in Nat Comm since 2015

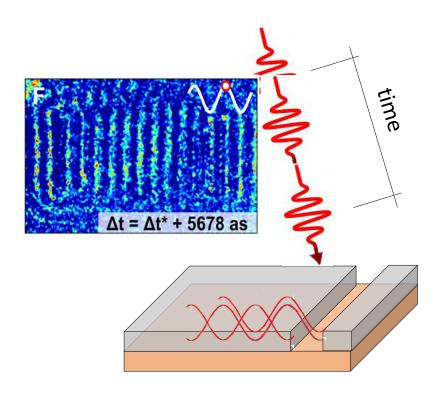
Now product manager at Dectris, Zurich

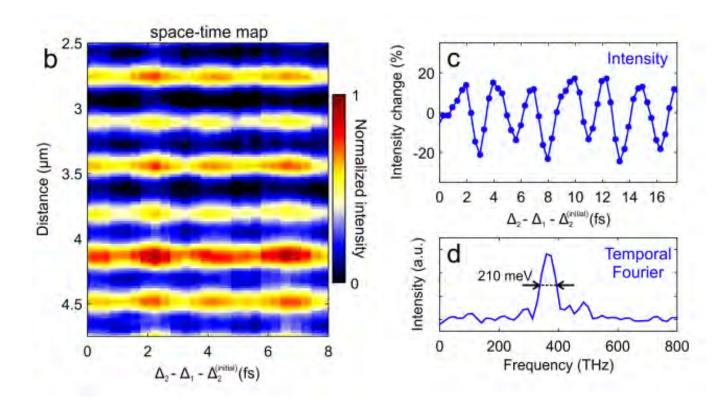


**Lummen** et al., **Nat. Comm**. 7 13156 (2016)

Now senior scientist at ETHZ

## Coherent control of plasma resonances





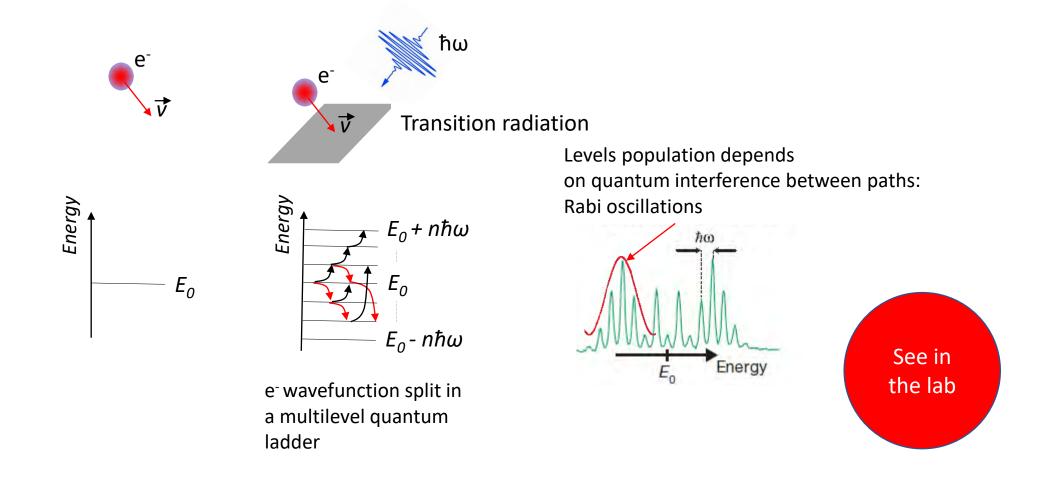




Vanacore, et al. Nat. Comm 9 2694 (2018)

Now tenure track professor at the University of Milan Bicocca

## What about the e- after these interactions?

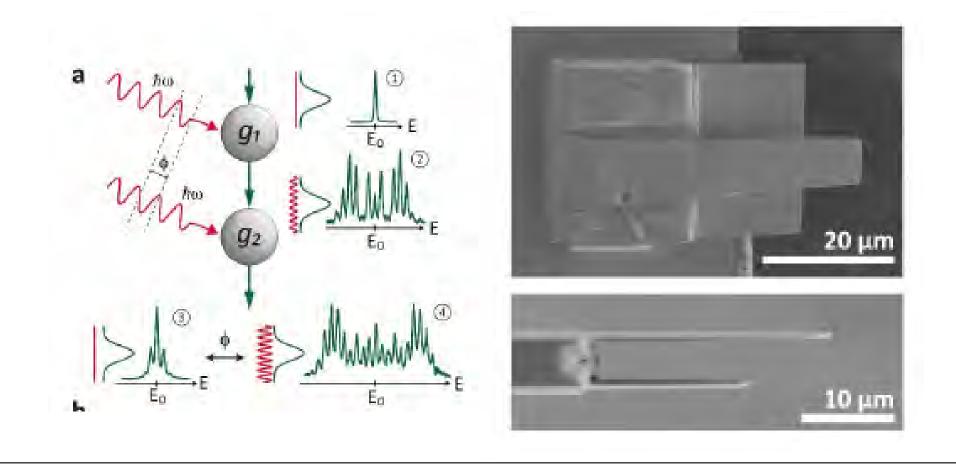




Park *et al.*, **New J. Phys.** 12, 123028 (2010) García de Abajo *et al.*, **Nano Lett.** 10, 1859 (2010)

Feist et al., Nature 521, 14463 (2015)

## Coherent control of the longitudinal electron wavefunction by plasma resonances





## What about the e- after these interactions?

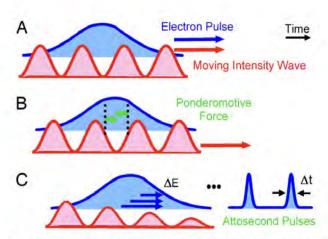
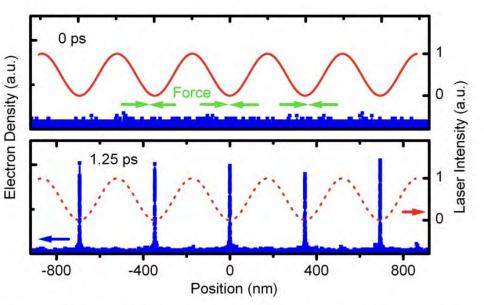
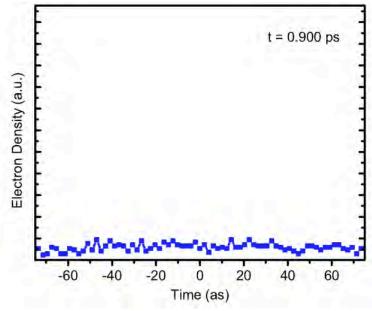


Fig. 2. Concept of optical compression in a moving intensity grating. (A) An electron packet (blue) copropagates at a velocity of less than the speed of light with the maxima and minima of a moving intensity wave (red), synthesized from two counterpropagating laser pulses of different frequencies (see text). (B) The ponderomotive forces (green arrows) push electrons out of regions with high intensity. Within every optical cycle, the parts of the electron packets that are located at a falling slope are accelerated (left green arrow) and the parts located at a rising slope are retarded (right green arrow). (C) The resultant momentum and velocity distribution  $\Delta p$  leads into self-compression to the final attosecond duration ( $\Delta t$ ) within every optical cycle (see text).



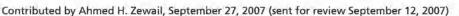
**Fig. 4.** Trajectory calculations showing the compression. (*Upper*) A long electron packet (blue) is overlapped with a moving intensity wave (red wave). Over time, the ponderomotive force (green arrows) accumulates and initiates self-compression. (*Lower*) At later times, when the intensity wave has faded (red, dotted wave), the electron density shows marked peaks of attosecond duration, which are located at the minima of the intensity wave. Note that the position scale is relative to the electron packet center.



# Attosecond electron pulses for 4D diffraction and microscopy

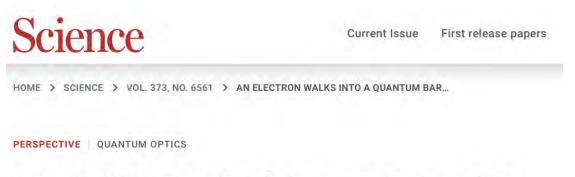
Peter Baum and Ahmed H. Zewail\*

Physical Biology Center for Ultrafast Science and Technology, Arthur Amos Noyes Laboratory of Chemical Physics, California Institute of Technology, Pasadena, CA 91125





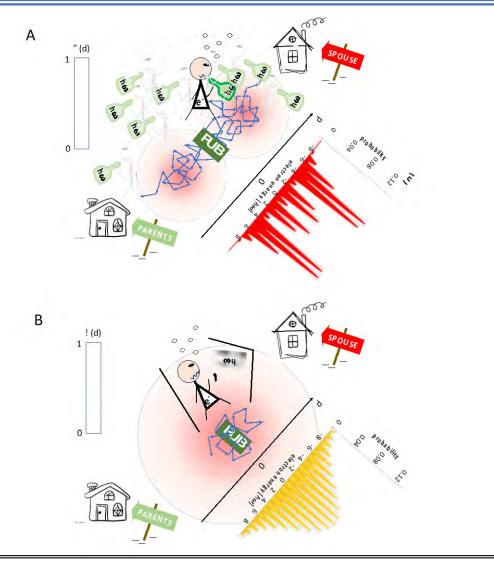
- Coherent light can be easily described as a wave (in the high intensity limit)
- Incoherent light needs a quantum description of its noise distribution



## An electron walks into a quantum bar...

ABRIZIO CARBONE

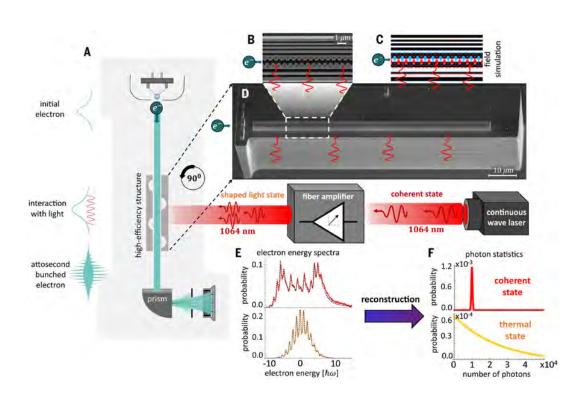
SCIENCE • 16 Sep 2021 • Vol 373, Issue 6561 • pp. 1309-1310 • DOI: 10.1126/science.abl6366

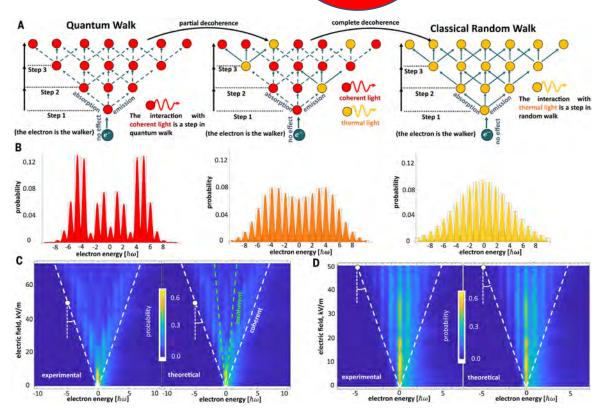




R. Dahan et al., *Science* **373**, eabj7128 (2021).

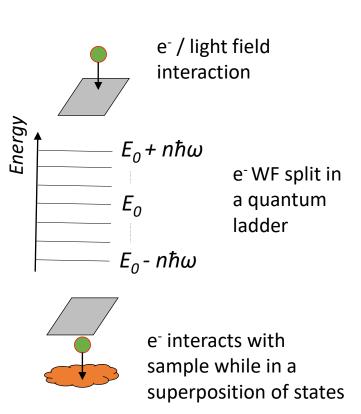


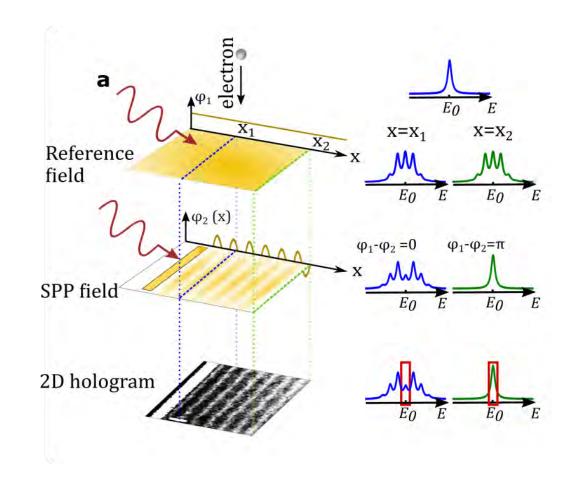






## Quantum holography





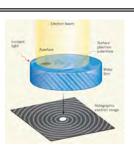
Vary dt between the two interactions



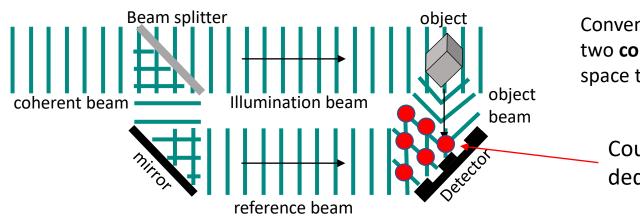


Madan et al, Science Advances 6 eaav8358 (2019) News and Views article: Ropers, Nature 571, 331 (2019)

https://actu.epfl.ch/news/new-holographic-technique-opens-the-way-for-quantu/https://www.youtube.com/watch?v=s2iiBbTuZn4&feature=emb\_logo



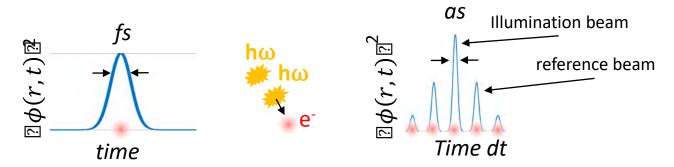
Madan et al., Sci. Adv. 2019; **5**: eaav8358 3 May 2019



Conventional holography: two **coherent** beams are separated in space to provide signal and reference

Count interferences deduce depth information

Quantum holography: two beams (made of one e-) are separated in time to provide as-resolved signal and references





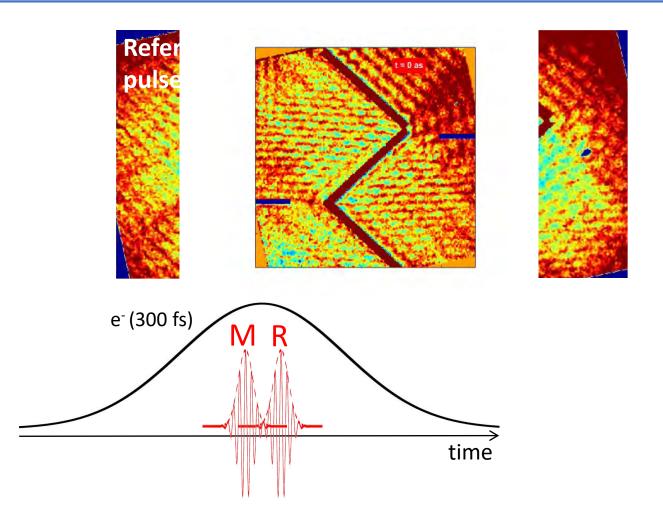
Light scatters electrons to make holograms

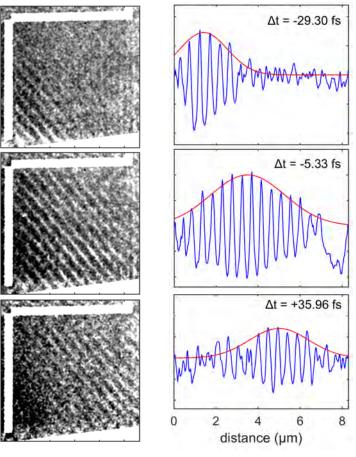


as-nm microscopic **quantum holography** of guided light in a nanostructure **Image the sample with WF-manipulated e**-



## Holography of plasma resonance





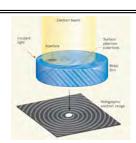
- Attosecond mapping of plasmon propagation
- Direct measurement of group and phase velocity





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

https://actu.epfl.ch/news/new-holographic-technique-opens-the-way-for-quantu/https://www.youtube.com/watch?v=s2iiBbTuZn4&feature=emb\_logo



#### Article

## Attosecond electron microscopy of sub-cycle optical dynamics

https://doi.org/10.1038/s41586-023-06074-9

David Nabben<sup>1,2</sup>, Joel Kuttruff<sup>1,2</sup>, Levin Stolz<sup>1</sup>, Andrey Ryabov<sup>1™</sup> & Peter Baum<sup>1™</sup>

Received: 12 October 2022

Accepted: 12 April 2023

Published online: 31 May 2023



The primary step of almost any interaction between light and materials is the electrodynamic response of the electrons to the optical cycles of the impinging light wave on sub-wavelength and sub-cycle dimensions<sup>1</sup>. Understanding and controlling the electromagnetic responses of a material<sup>2-11</sup> is therefore essential for modern optics and nanophotonics<sup>12–19</sup>. Although the small de Broglie wavelength of electron beams should allow access to attosecond and angström dimensions<sup>20</sup>, the time resolution of ultrafast electron microscopy<sup>21</sup> and diffraction<sup>22</sup> has so far been limited to the femtosecond domain<sup>16–18</sup>, which is insufficient for recording fundamental material responses on the scale of the cycles of light 1,2,10. Here we advance transmission electron microscopy to attosecond time resolution of optical responses within one cycle of excitation light<sup>23</sup>. We apply a continuous-wave laser<sup>24</sup> to modulate the electron wave function into a rapid sequence of electron pulses, and use an energy filter to resolve electromagnetic near-fields in and around a material as a movie in space and time. Experiments on nanostructured needle tips, dielectric resonators and metamaterial antennas reveal a directional launch of chiral surface waves, a delay between dipole and quadrupole dynamics, a subluminal buried waveguide field and a symmetry-broken multi-antenna response. These results signify the value of combining electron microscopy and attosecond laser science to understand lightmatter interactions in terms of their fundamental dimensions in space and time.



Ived with nanometre and atto second precision. Black arrows and white line indicate the space–time propagation. Scale bar, 500 nm.  $\Delta$ /is electron current change. See Supplementary Video I for full data.



https://doi.org/10.1038/s41567-017-0007-6



# Diffraction and microscopy with attosecond electron pulse trains

Yuya Morimoto<sup>1,2</sup> and Peter Baum<sup>1,2</sup>\*

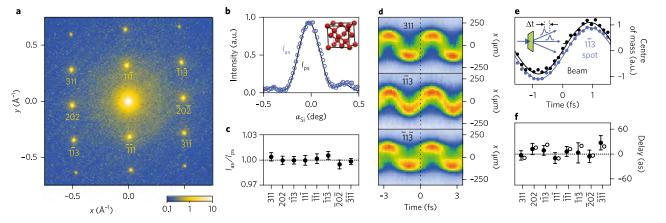
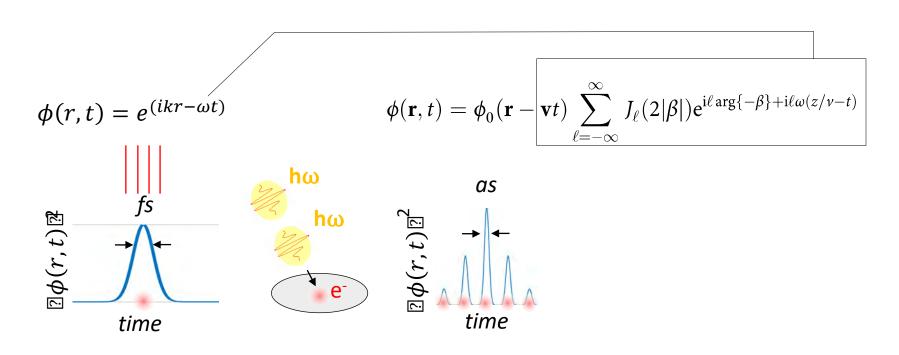


Fig. 3 | Atomic diffraction with attosecond electron pulses. a, Diffraction pattern of single-crystalline silicon, taken with the attosecond electron pulse train. The non-labelled Bragg spots are forbidden but visible via multiple scattering. b, Rocking curve of the 173 spot for picosecond pulses in comparison with the attosecond electron pulse train. Inset, crystal structure of silicon. c, Absolute intensity of all Bragg spots, measured with the attosecond pulses and normalized to the picosecond case. The error bars denote the shot noise of the measurement (-140 s integration). d, Streaking deflectograms of three example Bragg spots; all of them behave similarly. e, Centre-of-mass of the deflectogram of the 173 spot versus the direct beam for delay analysis. Inset, illustration of delays in Bragg spot emission. f, Measured attosecond-level delay of different Bragg spot emissions with respect to the direct beam. The filled and open circles are the results of centre-of-mass analysis and deflectogram fitting, respectively. The error bars represent standard deviation. The average delay is (4.6 ± 5.4) as.

# Attosecond coherent control of free-electron wave functions using semi-infinite light fields



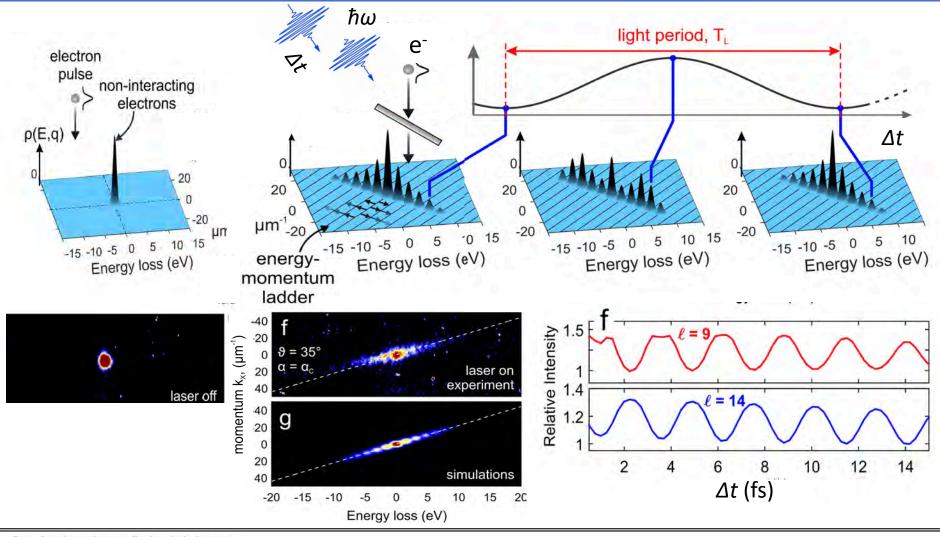
Vanacore et al, 9 2694 (2018)



*Temporal* Tomography of the e<sup>-</sup> wavefunction Image electrons



## Coherent control of the longitudinal electron wavefunction by transition radiation



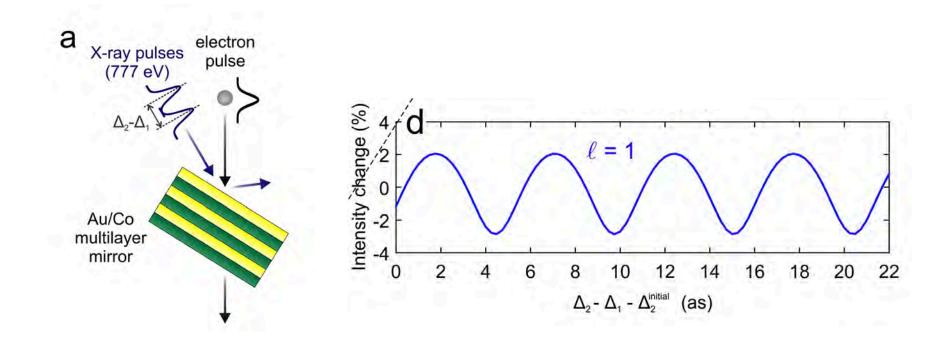




Vanacore, et al Nat. Comm 9 2694 (2018)

https://phys.org/news/2018-07-ultrashort-electron-harvest-nuclear-energy.html

## Zeptoseconds (10<sup>-21</sup>) electron pulses



## Zeptosecond physics:

- Nuclear processes (electron capture, core-level photoemission)
- Intramolecular processes (charge transfer)

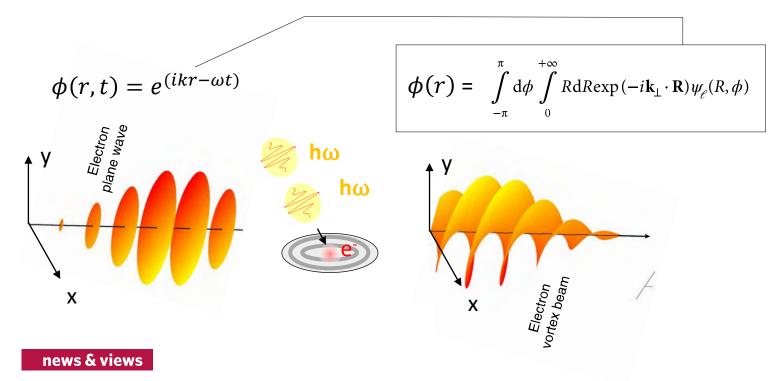


# Ultrafast generation and control of an electron vortex beam via chiral plasmonic near fields



Cited by 116

Vanacore et al., 18 573 (2019)



#### **PHOTONICS**

## Vorticity induced by chiral plasmonic fields

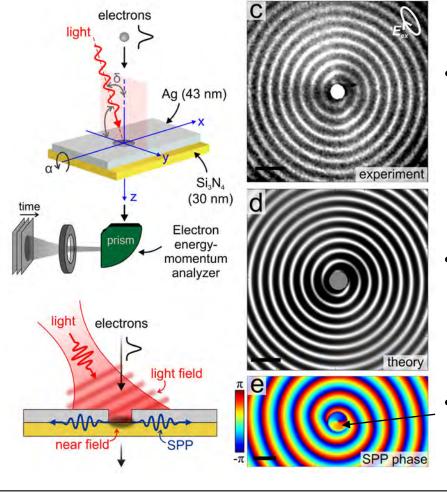
Coherent shaping of matter waves in temporal and spatial domains by photon-induced near fields opens up new possibilities for the quantum control of matter.

Jun Yuan

**Spatial** Tomography of the e<sup>-</sup> wavefunction **Image the electrons** 



## Coherent control of the transverse electron wavefunction



• Chiral SPP launched by elliptically polarized light

Simulated Chiral SPP

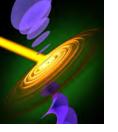
 A chiral plasmonic field can impart a phase singularity onto the transverse component of an e<sup>-</sup> wavefunction



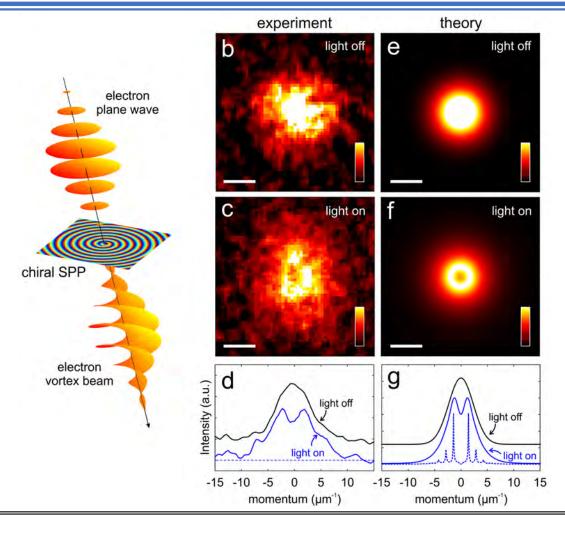
Vanacore et al., Nature Materials 18, 573 (2019)

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

https://actu.epfl.ch/news/twisting-whirlpools-of-electrons/
https://www.youtube.com/watch?time\_continue=5&v=TIJ-mlh2LSI&feature=emb\_logo



## Generation of electron vortex beams

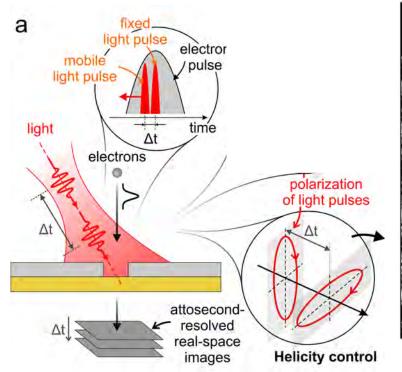


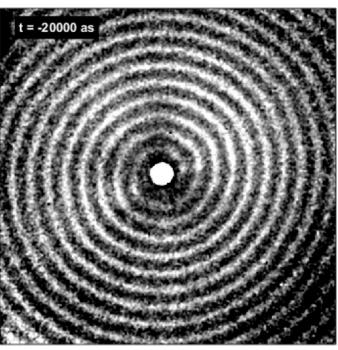
Electron wavefunction microscopy in momentum space

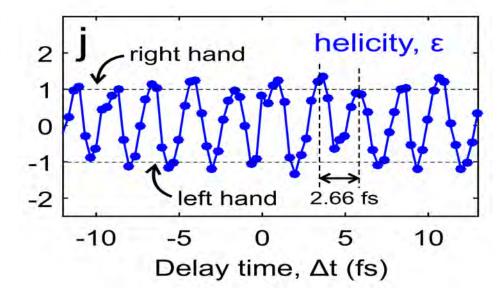


Vanacore et al., Nature Materials 18, 573 (2019)

## Coherent control of vortex beams





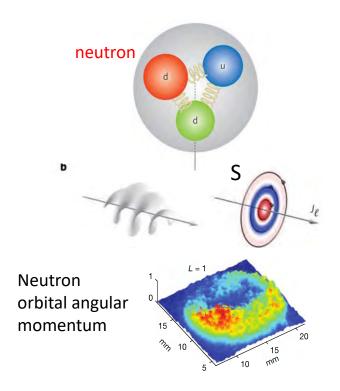


Three pulses experiment: attosecond control of the topological charge of an  $e^-$  wavefunction between + 1 and - 1



## Application of vortex beams

#### Twisted neutrons



Make a particle with a fine structure spiral reveals its inner structure

- Can we twist protons as well?
- Can we twist atoms and ions?
- Exploit the different radiation/matter interaction for selective tissue ablation (ongoing project with ADAM and CERN)

#### Vortex beams, magnetic sensing

- Spin-polarized TEM does not exist yet
- Left handed/right handed vortex beams are sensitive to magnetization orientation
- Transfer topological charge from e<sup>-</sup> vortex
   beam to spins in materials



Clark et al., Nature 525, 504 (2015)

Laroque et al., Nat. Phys. 14 1 (2018)

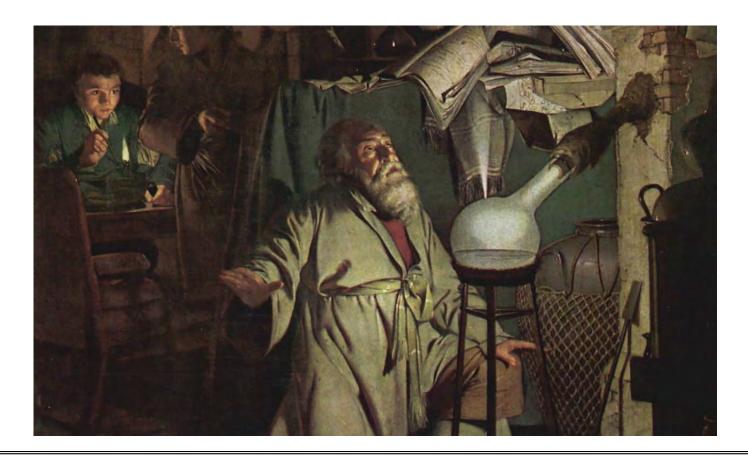
## Coherent control of nuclear reactions

## **Manipulating the element transmutation**

Dreamt of: 1600

Observed and used: 1950s to now

Controlled: not yet







## Orbital electron capture

Ionized atom captures a free e<sup>-</sup>
Instead of Auger recombination with gamma ray emission the atomic nucleus is excited

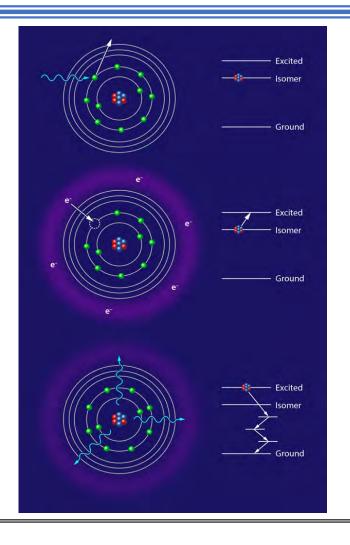
#### **Nuclear isomer:**

metastable atomic nucleus made of one or more of its nucleons (protons or neutrons) in an excited state.

Half-lives 100 to 1000 times longer (ns) than other excited nuclear states (ps or less)

Create metastable isomers: high energy process
The metastable state is not necessarily the most convenient for EC

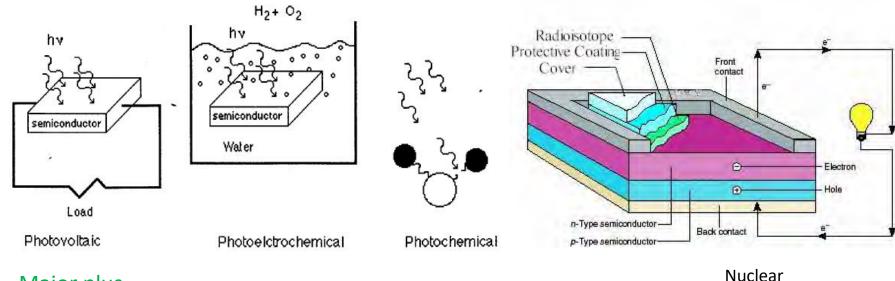
Sample short-lived states: impossible until now Expand parameter space from which to study NEEC





Chiara et al., Nature 554, 216 (2018)

## **Nuclear batteries**



## Major plus

- 4-5 orders of magnitude higher energy density
- Long-lasting (even 100s of years, use in satellites)

### Major minus

- Low power density (slow energy release process)
- Dangerous/poisonous materials (not eco-friendly)
- Low nuclear to electric energy conversion efficiency

#### **Solutions:**

- Control decay-rates: control power delivery
- Transiently induce radioactivity in stable elements: reduce environmental hazard, cost, danger
- Electrons and light technology can be integrated. Common nuclear technology cannot





https://physicsworld.com/a/nuclear-excitation-by-electron-capture-seen-at-long-last/

## Coherent control of nuclei

Can we use out of equilibrium methods to control the release of nuclear energy?

- Speed up the decay of nuclear waste
- Engineer long-lasting/high power nuclear batteries
- Induce radioactivity on demand in stable atoms



## Nuclear Excitation by Electron Capture

216 | NATURE | VOL 554 | 8 FEBRUARY 2018

## ≠ electronic configurations ≠ channels available

Nuclear Excitation by Electron Capture in Excited Ions

Simone Gargiulo, Ivan Madan, and Fabrizio Carbone Phys. Rev. Lett. **128**, 212502 – Published 27 May 2022

## $\neq$ orbitals $\neq$ cross section (10<sup>6</sup> higher!)

Dynamical Control of Nuclear Isomer Depletion via Electron Vortex Beams

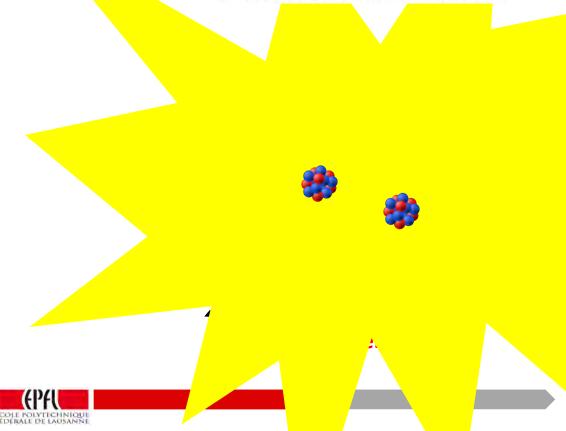
Yuanbin Wu, Simone Gargiulo, Fabrizio Carbone, Christoph H. Keitel, and Adriana Pálffy Phys. Rev. Lett. **128**, 162501 – Published 22 April 2022

## ≠ particles ≠ effect (induced fission)

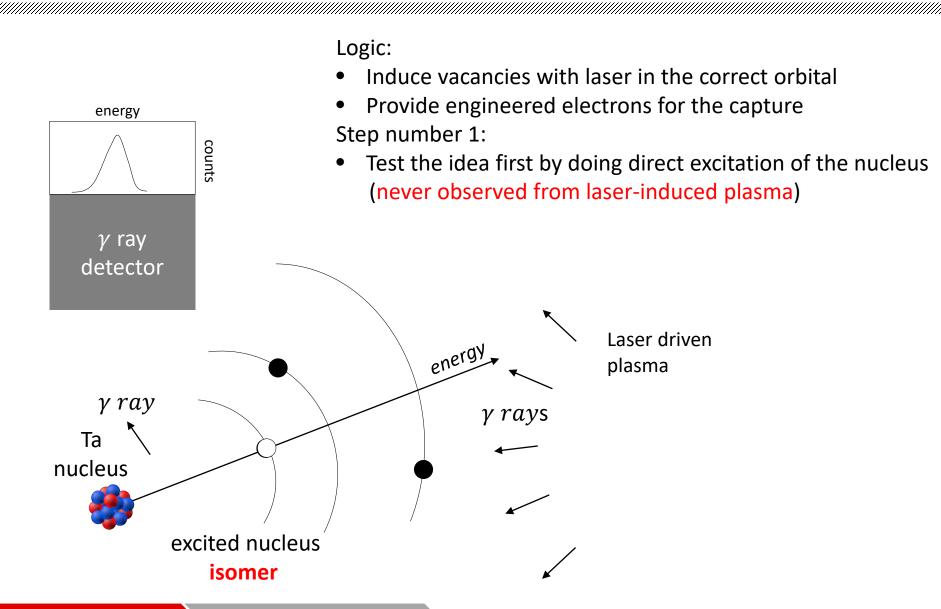
### Nuclear Excitation by Free Muon Capture

Simone Gargiulo, Ming Feng Gu, Fabrizio Carbone, and Ivan Madan Phys. Rev. Lett. **129**, 142501 – Published 27 September 2022

The modifications of the FAC code made it the only freely-available online tool that can solve the Dirac equations for muonic atoms



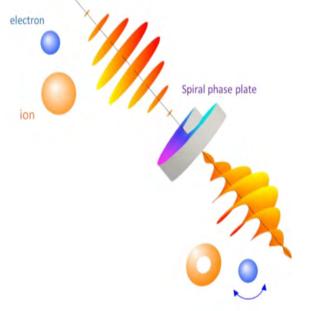
## **Experimental Implementation**



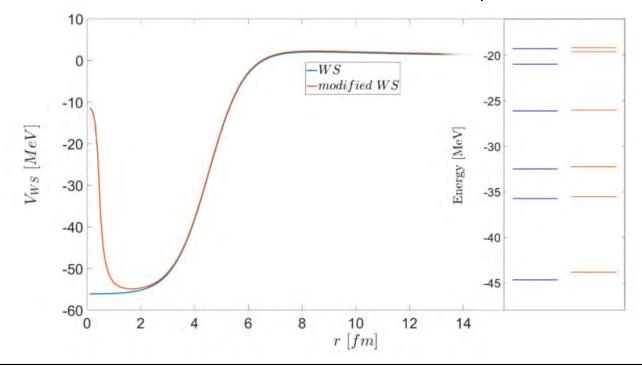


## Nuclear physics with vortex beams

- Twisting ions WF modifies nuclear excitations spectrum
- Vortex e<sup>-</sup> can be captured in higher l-shells, created ad hoc
- NEEC  $\sigma$  up to 6 orders of magnitude (article in preparation) ENGINEERED NUCLEAR DECAY!!

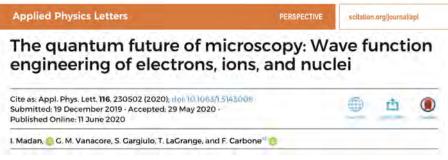


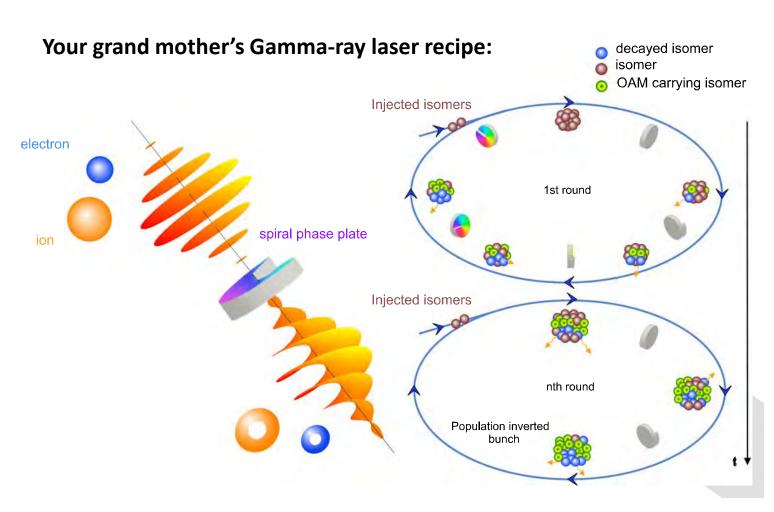
## Nuclear excitations and nuclear Wood-Saxon potential







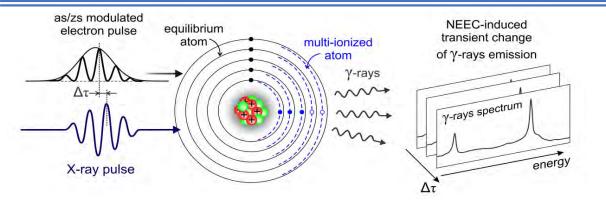


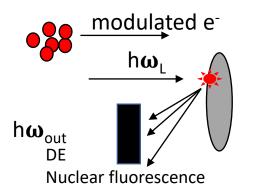




Thank you!

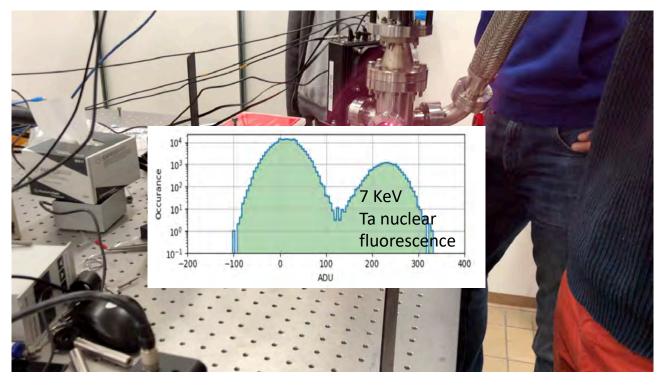
## Experimental concept and implementation





New medipix detector: Energy-resolution + ns time resolution

Same energy! Different time......



We don't have an FEL: Use light induced plasma to generate the needed X-rays for direct photoexcitation



Rudek et al., Nat. Phot. 6 858 (2012)

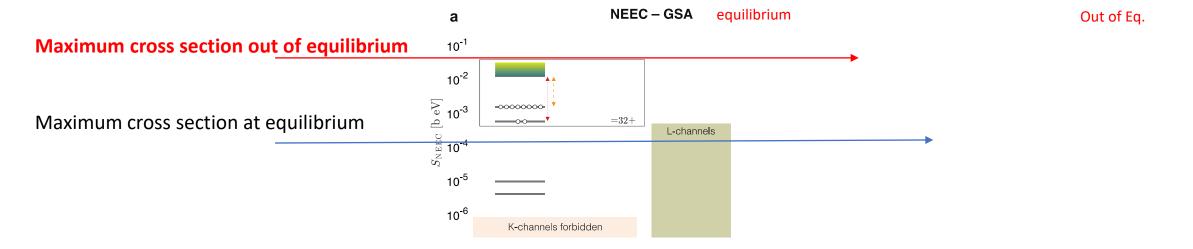
Gargiulo et al (submitted to PRL, <a href="http://arxiv.org/abs/2102.05718">http://arxiv.org/abs/2102.05718</a>)

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

## Out of equilibrium nuclear physics:

Observed NEEC  $\sigma$  vs theoretical NEEC  $\sigma$ :  $10^{11}$ 

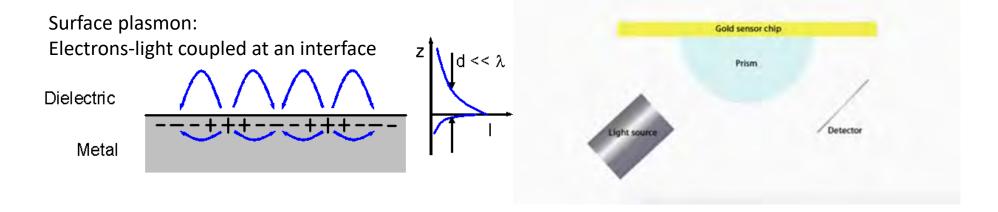
Recalculating  $\sigma$  in out of equilibrium ions: 3 orders of magnitude gained, 8 to go





Gargiulo et al (submitted to PRL, <a href="http://arxiv.org/abs/2102.05718">http://arxiv.org/abs/2102.05718</a>)

## Surface plasmons polaritons and plasma resonances





Plasma resonance

