

# Time-resolved nano-optics in electron microscopy

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Dôme de La Grave, Toulouse

### Acoustic whispering gallery mode



St Paul's Cathedral, London (Wikipedia)



The Whispering Gallery







Matsuda & Tsuchiya, "Watching whispering-gallery waves », Laboratory of Applied Solid-State Physics, Hokkaido University

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# Optical whispering gallery mode

Glass micro-sphere







Source: OIST

Optical whispering gallery



Foreman *et al.,* Adv. Opt. Photon. **7**, 168-240 (2015)

Optical Whispering-gallery modes (WGM)

Length scale : µm Time scale : f**s-ps** 



# Optical whispering gallery mode

Optical Whispering-gallery modes (WGM)

Length scale : µm Time scale : f**s-ps** 



1.0 0.8 0.6 0.4 0.2 0.0 0.4 0.1 0.2 0.0 0.4 0.2 0.0 0.4 0.2 0.0 1 0.2 0.4 0.5 time (ps)

- Period of the round trip : fs
- Typical time of decay: **ps**



### What do we mean by being time-resolved?

### Hummingbird wings beating $\sim$ 70 Hz



### High speed camera $\sim$ 500 Hz



Movie by Anand Varna<sup>1</sup>

### The time scales of (nano-)optics



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# Outline of the course (if $t \rightarrow \infty$ )







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# 1. Chronophotography -Fifty shades of imaging

(no, actually just two)

Bullet cutting a card, Harold Edgerton, MIT *https://edgerton-digital-collections.org* 

# A brief history of Chronophotography



Eadweard Muybridge (1830-1904) The Horse in Motion (1878)





## A brief history of Chronophotography



 $\Delta$  The process needs to be **reversible** 

### Dynamic vs. Ultrafast transmission electron microscopy



*Principle of Streak imaging (single shot)* 

Irreversible processes



### Dynamic Transmission Electron Microscope\* (**DTEM**)

• O. Bostanjoglo, TU Berlin (1980s) • G.H. Campbell, Lawrence Livermore National Laboratory

Dömer and Bostanjoglo, Review of Scientific Instruments 74, 4369 (2003) Kim et al., Science **321**, 1472-1475 (2008) Campbell et al., Applied Physics Review 1, 041101 (2014) ✦ Santala et al., Applied Physics Letter 102, 174105 (2013)

Phase transition in GeTe (ns)

### *Principle of Pump-Probe imaging (stroboscopic)*



**Reversible processes** 



### A. Zewail, Caltech (2010s)

Danz et al., Science **371**, 371-374 (2021)



# Femto-photography: Light-in-flight recording



See the work of Philipp Haslinger: http://seecphotography.com/



2. A quick introduction to ultrafast transmission electron microscopy ZrO<sub>2</sub>- Reservoir

100µm

Schottky-emitter electron source of an Electron microscope Source: https://en.wikipedia.org/wiki/Field\_emission\_gun

CNIS

W{100}

### How does it look like?

• High-Speed TEM: The modified Siemens Elmiskop 1A at TU Berlin (1990s')





Oleg Bostanjoglo, Advances in Imaging and Electron Physics Volume **121**, 1-51 (2002)

# How does it look like?

• Schottky-FEG UTEM:

The modified JEOL 2100F at the university of Göttingen (2020)

# The modified Hitachi 3300 in CEMES Toulouse (2024)

• Cold-FEG UTEM:



Danz et al., Science **371**, 371-374 (2021)

### Ultrafast photo-emission



precise trigger time, therefore arrival time on the sample.

# Different type of electron photo-emitters





z (nm)





Strong-field PE



Armin Feist, PhD thesis (2018)

0

f(E)

### Different type of electron photo-emitters



A. Feist et al, Ultramicroscopy 176, 63-73 (2017)

• Electron energy width

$$\Delta E = N\hbar\omega - \phi_W + \Delta E_{\rm kin} + \Delta E_{\rm thermal} + \Delta E_{\rm laser}$$

- Ideal case:
- 1 photon photo-emission
- Cold source
- Close to the work function

### What are the properties of this ultrashort electron pulse?

• Effective source size (as reduced by apertures)

Typical TEM figures of merit

O Source brightness

o Emission stability.

o Source energy spread





UTEM adds more ! o The electron pulse duration o Number of electrons per pulse o The repetition rate o Impact of Coulomb repulsion for high density electron pulses. o The electron and laser chirp o ....

### A short bibliography on tip photo-emitters 🌌

Ultrafast Electron Pulses from a Tungsten Tip Triggered by Low-Power Femtosecond Laser Pulses P. Hommelhoff *et al.*, Phys. Rev. Lett. **97** (2006)

Attosecond control of electrons emitted from a nanoscale metal tip M. Krüger *et al.*, Nature **475**, 78–81 (2011)

Localized Multiphoton Emission of Femtosecond Electron Pulses from Metal Nanotips C. Ropers *et al.*, Phys. Rev. Lett. **98** (2007)

Laser-induced ultrafast electron emission from a field emission tip B. Barwick *et al.*, New J. Phys. **9** (2007)

Tip-Enhanced Strong-Field Photoemission R. Bormann *et al.*, Phys. Rev. Lett. **105** (2010)

**Field-driven photoemission from nanostructures quenches the quiver motion** G. Herink *et al.*, Nature **483** (2012)

**Highly coherent electron beam from a laser-triggered tungsten needle tip.** D. Ehberger *et al. Phys. Rev. Lett.* **114** (2015).

An ultrafast electron microscope gun driven by two-photon photoemission from a nanotip cathode R. Bormann *et al.*, J. Appl. Phys. **118** (2015)

### Longitudinal properties of ultrashort electron pulses



A strong temporal coherence does <u>not</u> equal a short pulse duration!

On the physics of ultrashort single-electron pulses for time-resolved microscopy and diffraction Peter Baum, Chemical Physics **423**, 55-61 (2013)

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![](_page_21_Figure_1.jpeg)

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![](_page_22_Figure_1.jpeg)

Danz et al., Science 371, 371-374 (2021)

![](_page_23_Figure_1.jpeg)

### What is the temporal resolution?

![](_page_24_Figure_1.jpeg)

### Quantum Improvement of Time Transfer between Remote Clocks

B. Lamine, et al., Physical Review Letters 101 (12) (2008)

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### Other time-resolved methods not based on photo-emission

![](_page_25_Figure_1.jpeg)

• Fast Electron beam blankers

Y. Auad et al., Nature Communications 14, 4442 (2023)
P. Das et al., Ultramicroscopy 203, 44-51 (2019)
Liu et al., ACS Photonics 6 (10), 2499–2508 (2019)
Collette et al., J. Chem. Phys. 153, 044711 (2020)
Collette et al., Scientific Reports 10, 12537 (2020)
Garfinkel et al., ACS Appl. Nano Mater. 5 (2) 1798–1807 (2022)

### • Microwave cavity choppers and camera streaking

![](_page_25_Figure_5.jpeg)

W. Verhoeven et al., Ultramicroscopy, 188, 85–89 (2018) S. A. Reisbick et al., Ultramicroscopy **235**, 113497 (2022)

# Summary: Ultrafast electron transmission microscopy (UTEM)

![](_page_26_Figure_1.jpeg)

### From the Schwarz-Hora effect to PINEM

![](_page_27_Picture_1.jpeg)

![](_page_27_Picture_2.jpeg)

Helmut Schwarz

Heinrich Hora

![](_page_27_Picture_5.jpeg)

Brookhaven National Laboratory Accelerator Test Facility (ATF)

1.0 0.8 0.6 0.6 MODEL (2 mm DIA. EXIT HOLE) 0.4 0.2 0.0 -2 0 0.4 ENERGY SHIFT (MeV) W. D. Kimura *et al.*, Phys. Rev. Lett. 74, 546 (1995)

H. Schwarz & H. Hora, Appl. Phys.

Lett. 15, 349–351 (1969)

![](_page_27_Picture_8.jpeg)

![](_page_27_Picture_9.jpeg)

![](_page_27_Picture_10.jpeg)

Brett Barwick

David J. Flannigan

Ahmed H. Zewail

![](_page_27_Figure_14.jpeg)

Barwick et al., Nature 462, 902–906 (2009)

![](_page_28_Picture_0.jpeg)

Picture of the optical setup of the Göttingen UTEM, the green laser (515 nm) is used for the photo-emission while the red one (640 nm) pumps the sample.

![](_page_29_Figure_1.jpeg)

![](_page_30_Figure_1.jpeg)

![](_page_31_Figure_1.jpeg)

![](_page_32_Figure_1.jpeg)

![](_page_33_Figure_1.jpeg)

![](_page_34_Figure_1.jpeg)

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![](_page_35_Figure_1.jpeg)
## Photon-induced near-field electron microscopy (PINEM)







g-map

0.8

0



Map of the laser-induced optical near-field



Kfir et al., Nature 582, 46–49 (2020)



## Where is the time resolution?



Kfir et al., Nature 582, 46–49 (2020)



## Where is the time resolution?





Christen and Bimberg, Oyo Buturi 57, 69-77 (1988)

300

## 3b. Coincidence measurements

Cathodoluminescence of an ensemble of  $NV^0$  centers of a nano-diamond ACS Photonics 5 (2), 324-328 (2018)

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## Nitrogen-Vacancy (NV) centers in nano-diamonds



## Nitrogen-Vacancy (NV) centers in nano-diamonds









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## Correlation electronics







## Decay kinetics can be more complex

• Simple exponential



Meuret *et al.*, Appl. Phys. Lett. **119**, 062106 (2021) Kim and Kwon, ACS Nano **15** (12), 19480–19489 (2021) Liu, et al. Appl. Phys. Lett. 109, 042101 (2016) Finot, et al. Appl. Phys. Lett. 117, 221105 (2020)

#### • Stretched exponential (Kohlrausch function)



Meuret *et al.*, Ultramicroscopy **197**, 28-38 (2019) Benny Lee *et al*, Biophysical Journal 81 (3), 1265-1274 (2001) Martins *et al.*, Phys. Chem. C **126** (48) 20480–20490 (2022)

#### • Multiple exponentials



Corfdir et al, J. Appl. Phys. 107, 043524 (2010)



## Measurement of the emitted photon statistics



## Measurement of the emitted photon statistics



## The Hanbury-Brown and Twiss (HBT) interferometer







Figure 2. Picture of the two telescopes used in the HBT experiments. The figure was extracted from Ref.[1].



### • Each photon comes one by one





Anti-bunched emission

**Bunched** emission

 $\dot{ au}$ 

# of events

Mark Fox, Quantum Optics: An Introduction, Oxford University Press (2006)

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## The Hanbury-Brown and Twiss (HBT) interferometer

HBT accesses:O The excitation lifetimeO The emission statistics

Second order correlation function of light  $g^{(2)}(\tau) = \frac{\langle n_1(t)n_2(t+\tau)\rangle_t}{\langle n_1(t)\rangle_t \langle n_2(t)\rangle_t}$ 

Glauber, Phys. Rev. **130**, 2529 (1963)







Anti-bunched emission

• Photons come in packets



#### Bunched emission





- S. Meuret et al., Nano Lett. 18 (4) 2288–2293 (2018) S. Finot et al., Appl. Phys. Lett. **117**, 221105 (2020)
- S. Finot et al., ACS Photonics **9**, 173–178 (2022)

## Bunching in HBT-CL



• Each electron can emit multiple photons



## Anti-bunching in HBT-CL



• Each electron can emit only one photon



## Bunching in HBT-CL – dependence on electron current



S. Meuret *et al.*, Phys. Rev. Lett. **114**, 197401 (2015)
M. Feldman *et al.*, Phys. Rev. B **97**, 081404 (2018)
M. Solà-Garcia *et al.*, ACS Photonics **8** (3) 916–925 (2021)

## N=2 centers: Competition between bunching and anti-bunching



W. Borst & L. Liu, Review of Scientific Instruments 70, 41 (1999)
S. Meuret *et al.*, Phys. Rev. Lett. 114, 197401 (2015)
M. Feldman *et al.*, Phys. Rev. B 97, 081404 (2018)
M. Solà-Garcia *et al.*, ACS Photonics 8 (3) 916–925 (2021)



S. Fielder et al., Nanophotonics 12 (12), 2231-2237 (2023)

## Photon emission pathways



Several excitations and decay pathways lead to the emission of the same photon

## Cathodoluminescence excitation (CLE) spectroscopy



Y. Auad, Ultramicroscopy **239**, 113539 (2022) Y. Auad, Phd Thesis, U Paris-Saclay (2022)

## Cathodoluminescence excitation (CLE) spectroscopy



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## Cathodoluminescence excitation (CLE) spectroscopy



Subscription Statistics depends on the electron final state





# 3c. Electron pulsecompression –attosecond bunching

Free-Electron Homodyne Detection on a gold nanotriangle. Picture courtesy from Murat Sivis

## Conventional PINEM



## Double PINEM interaction - toward attosecond/zeptosecond resolution



Lin et al., arXiv:2504.17770 (2025)














## Propagation of modulated electrons: Temporal lensing



#### • Experimental realizations

K. Priebe et al., Nature Photonics 11, 793–79 (2017)
A. Ryabov and P. Baum, Science 353, 6297, 374-377 (2016)
G. Vanacore et al., Nature Communications 9, 2694 (2018)
M. Kozák et al., Nature Physics 14, 121–125 (2018)
Y. Morimoto & P. Baum, Nature Physics 14, 252–256 (2018)
Y. Morimoto & P. Baum, Phys. Rev. A 97, 033815 (2018)
M. Kozák et al., Phys. Rev. Lett. 120, 103203 (2018)





655 attoseconds

Improving the pulse compression
 Multiple interaction stages
 S. Yalunin et al., Phys. Rev. Research 3, L032036 (2021)
 Large Talbot revival
 M. V. Tsarev et al., Physical Review Research 3 (4), 043033 (2021)
 Use quantum states of light
 V. Di Giulio & F. Javier García de Abajo, Optica 7 (12), 1820-1830 (2020)

#### • Application in coherent control

D. Rätzel *et al.*, Phys. Rev. Research 3, 023247 (2021) Kfir et al., Science Advances 7 (18), eabf6380 (2021)



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## Experimental examples

Surface plasmon Polariton along a gold tip



J. Gaida *et al.,* Nature Communication, **14**, 6545 **(2023)** 

# Surface plasmon Polariton along a gold tip



100 nm

J. Gaida *et al.*, Nature Photonics **18**, 509–515 (2024)

Dielectric nano-slit



63–67 (2023)

See also

T. Bucher *et al.*, Science Advances **9**, 51 (2023) D. Hui *et al.*, Science Advances **10**, 34 (2024)

### Further reading

- Reviews:
  - o J. García de Abajo, Rev. Mod. Phys. **82**, 209 (2010)
  - M. Kociak & Zagonel, Ultramicroscopy 176, 112-131 (2017)
  - S. Meuret et al., Ultramicroscopy 197, 28-38 (2019)
  - A. Polman, M. Kociak & J. García de Abajo, Nature Materials **18**, 1158–1171 (2019)
  - P. Dombi *et al.*, Rev. Mod. Phys. 92, 025003 (2020)
  - J. García de Abajo & V. Di Giulio, ACS Photonics 8 (4) 945–974 (2021)

- Books:
  - P. Hommelhoff & M. Kling, Attosecond Nanophysics: From Basic Science to Applications, Wiley – VCH (2015)

# Thank you for your attention !

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