Developments in **Ultrafast Electron Microscopy**

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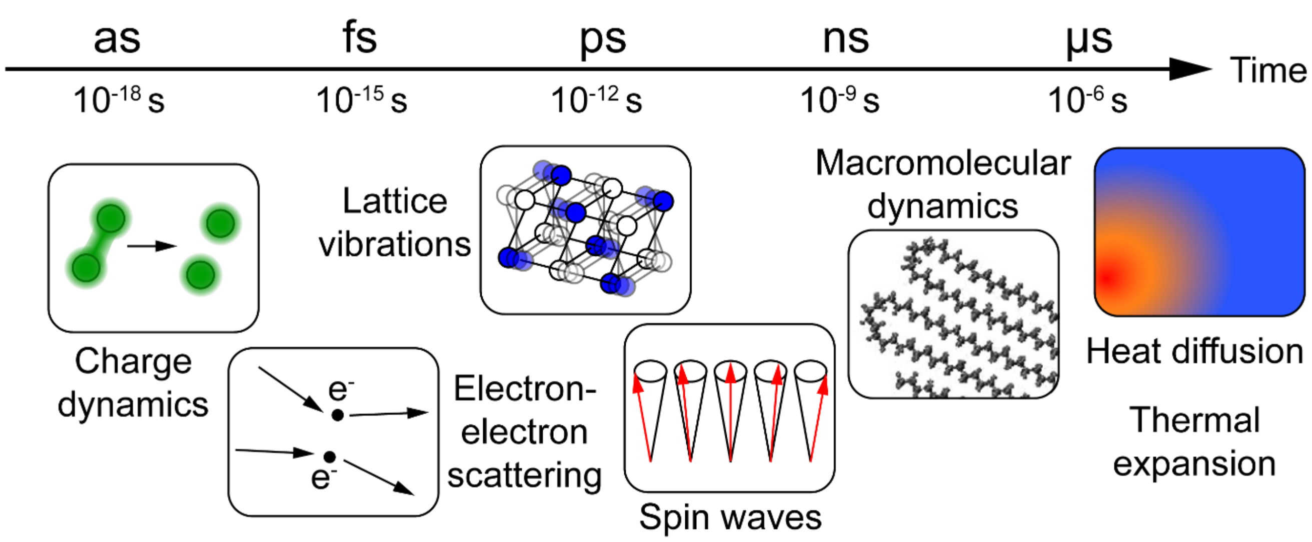
FOR MULTIDISCIPLINARY SCIENCES

Solids and Nanostructures: Selected Challenges

- Emergence and control of correlated states of matter
- Energy transfer and redistribution among different degrees of freedom
- Pathways in structural and magnetic transformations



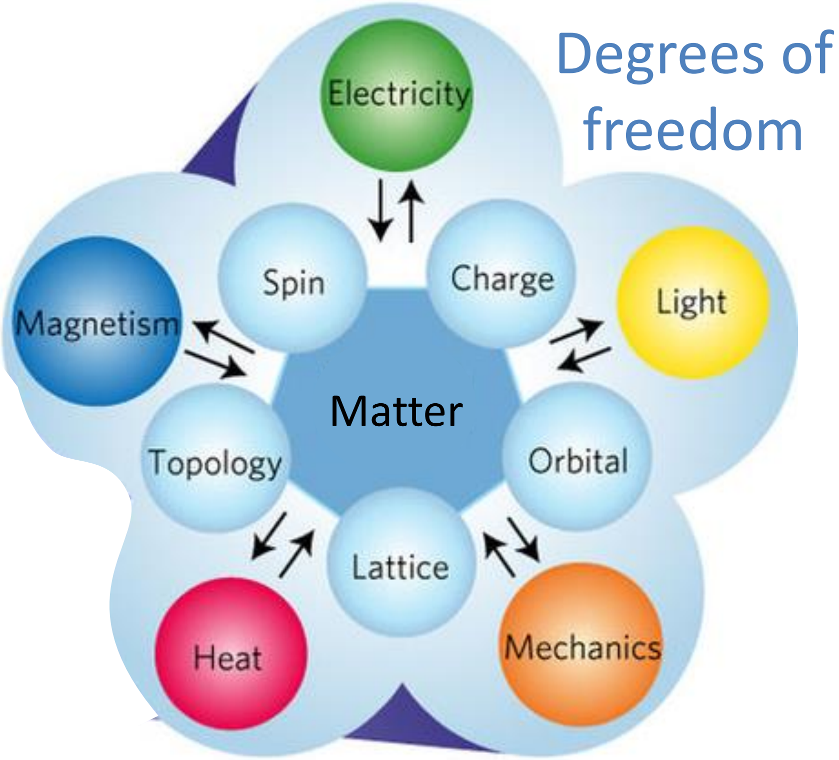
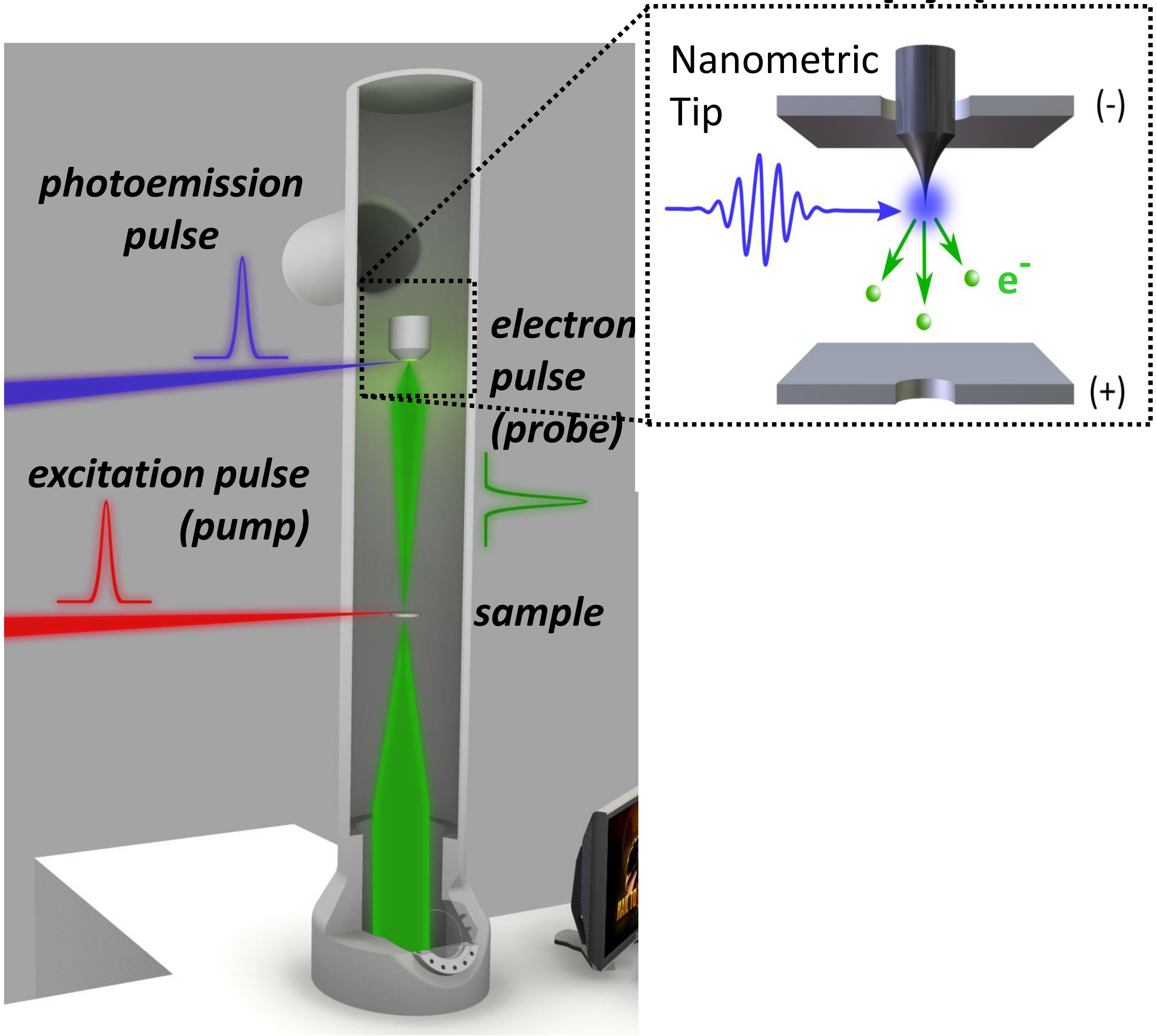


Fig. adapted from: Tokura et al., Nat. Phys. (2017)

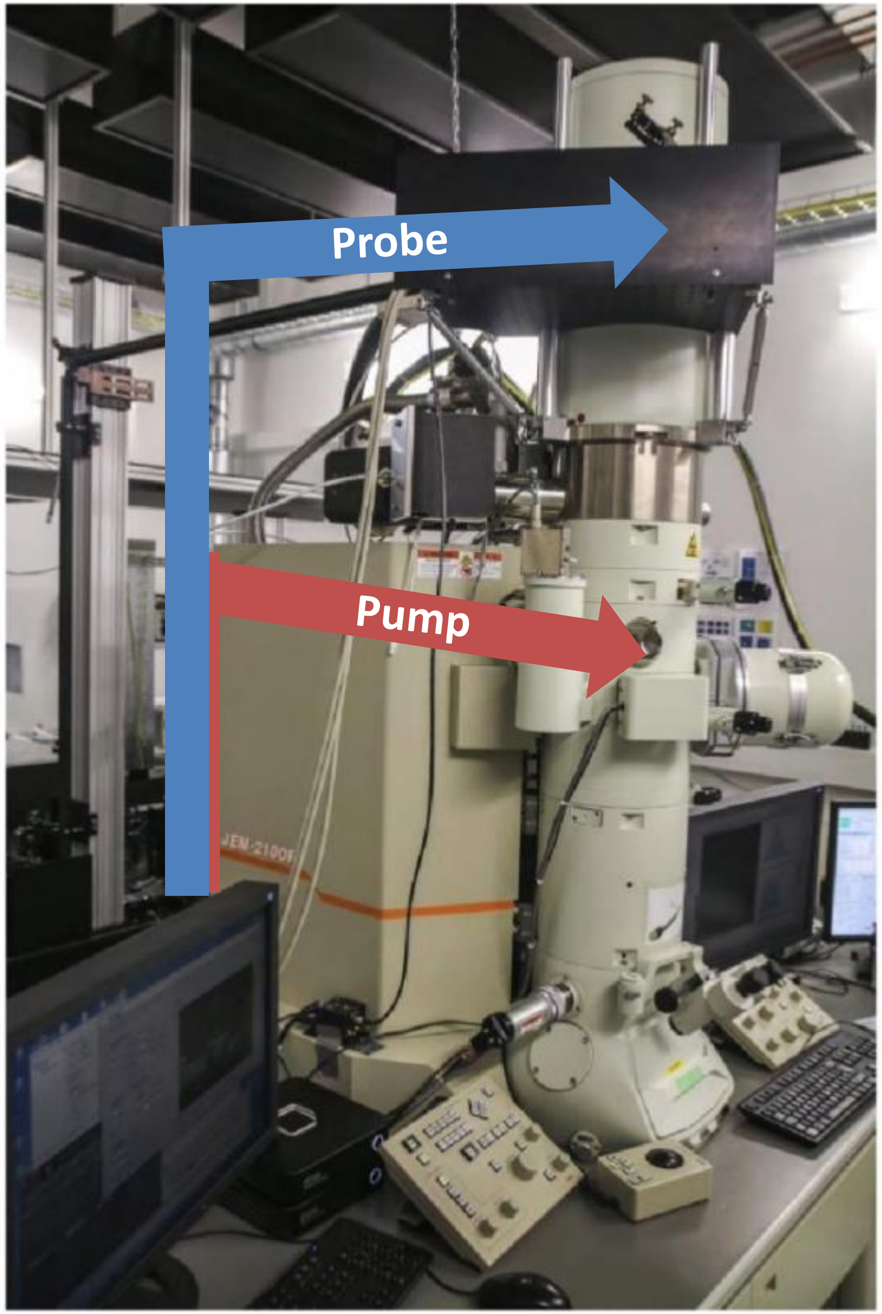
Ultrafast transmission electron microscopy (UTEM)

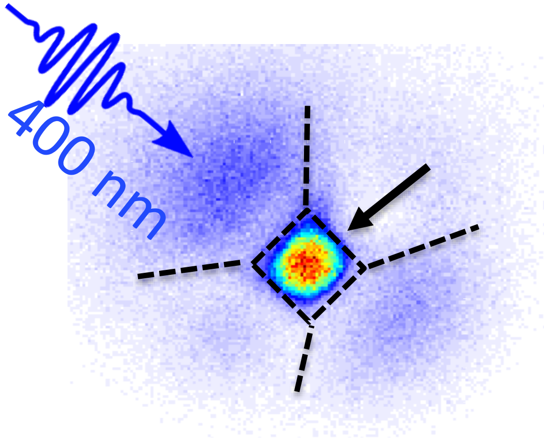


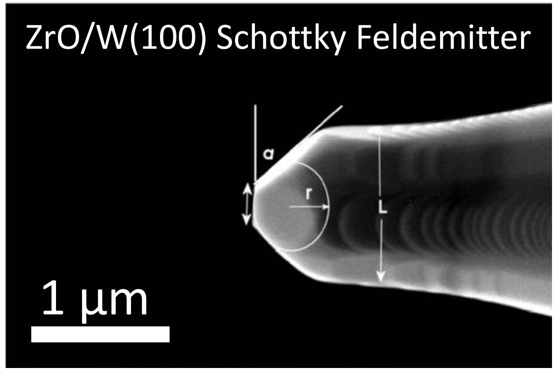
Nanocathodes:

- **High coherence**
- **Large extraction fields**
- **Short electron pulses**

Ultrafast transmission electron microscope (UTEM)

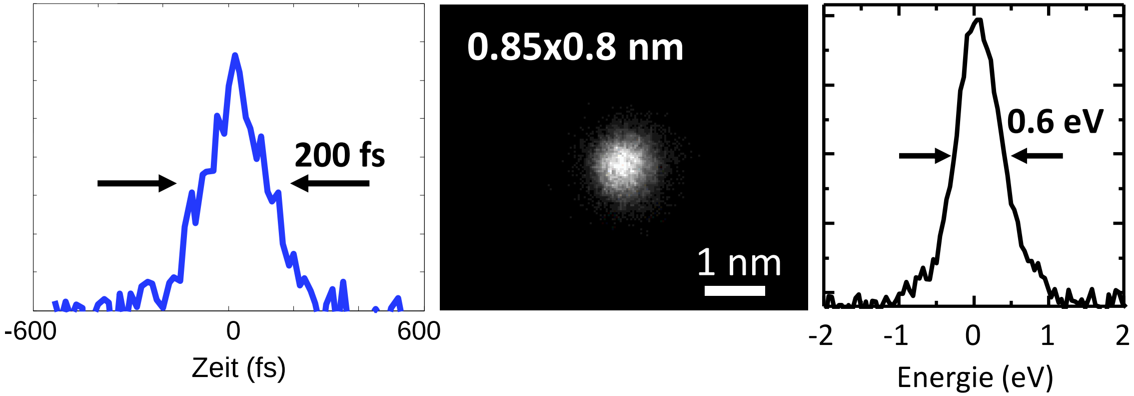






Liu et al., J. Vac. Sci Tech. (2010).

Temporal/spatial/spectral electron pulse properties



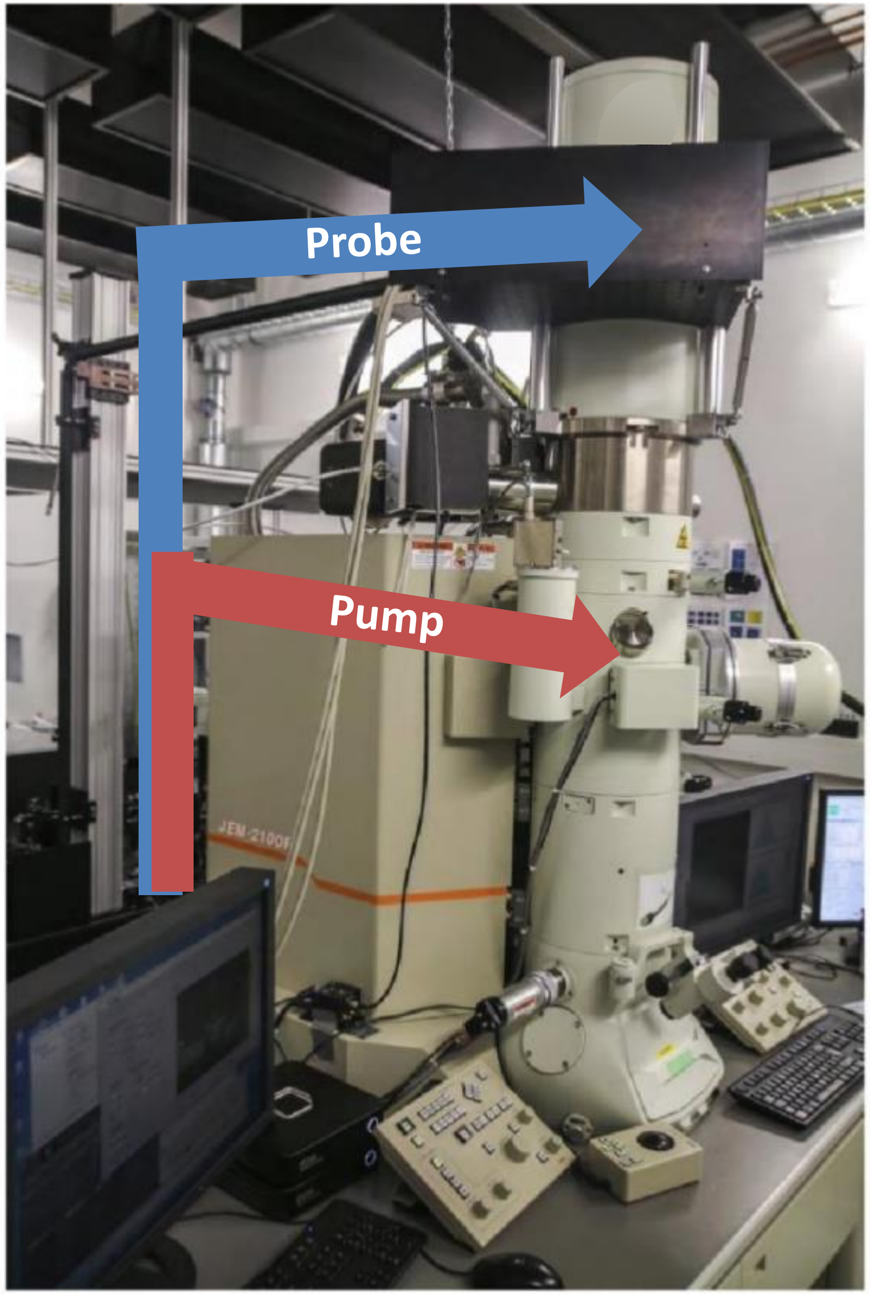
→ rms-Emittance: $\boldsymbol{\varepsilon = 2\, pm \cdot rad}$ (m. Apertur)

→ Peak Brillance: $1.75{\cdot}10^{13}$ A/m^{2}sr

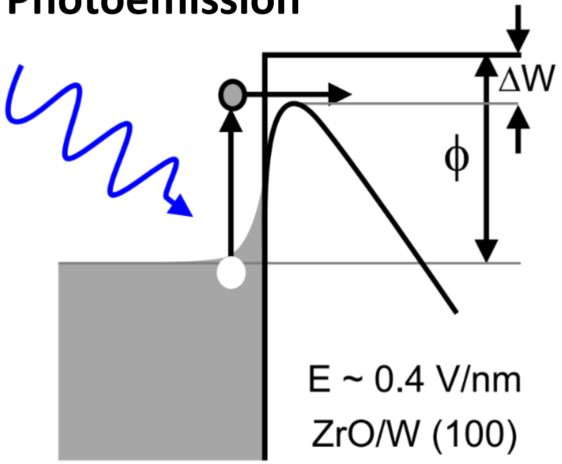
A. Feist *et al.*, Nature 521, 200 (2015)

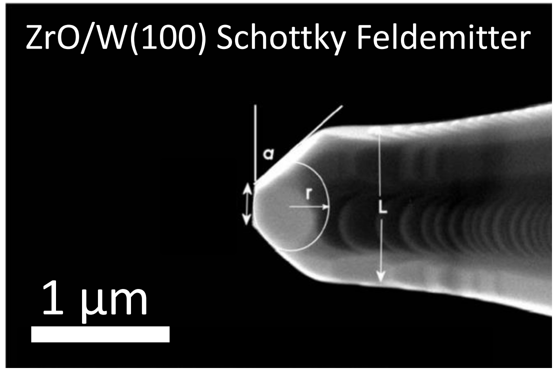
A. Feist *et al.*, Ultramicroscopy (2017)

Ultrafast transmission electron microscopy (UTEM)



Photoemission





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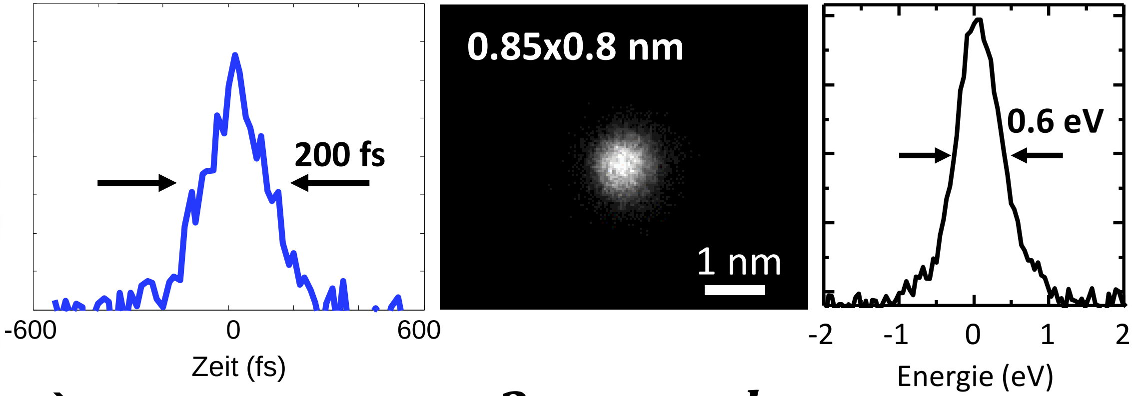
Temporal/spatial/spectral electron pulse properties

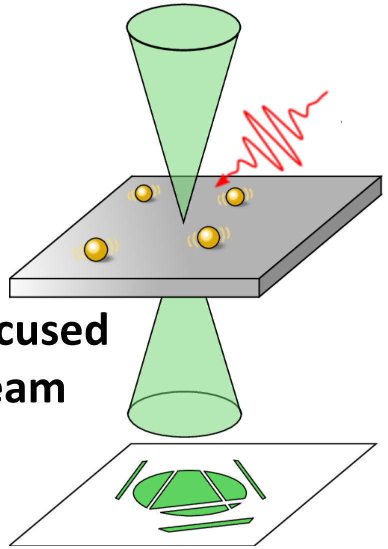


→ rms-Emittance: $\varepsilon = 2\ pm \cdot rad$ (apertured)

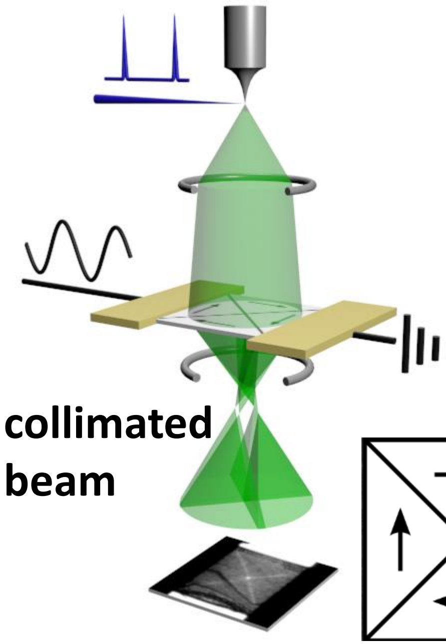
→ Peak brightness: $1.75 \cdot 10^{13}$ A/m^2sr

B. Cook *et al.* Ultramicroscopy **109**, pp. 403-412 (2009)
A. Feist *et al.*, Ultramicroscopy (2017)

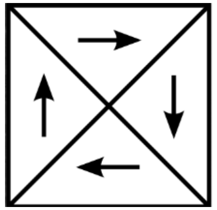
Ultrafast transmission electron microscopy (UTEM)

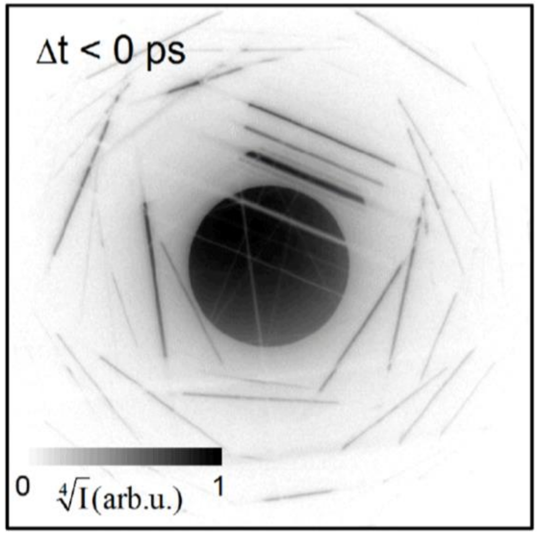


focused
beam



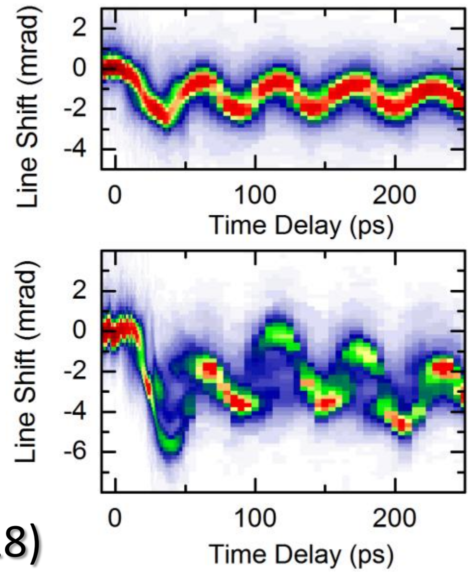
collimated
beam



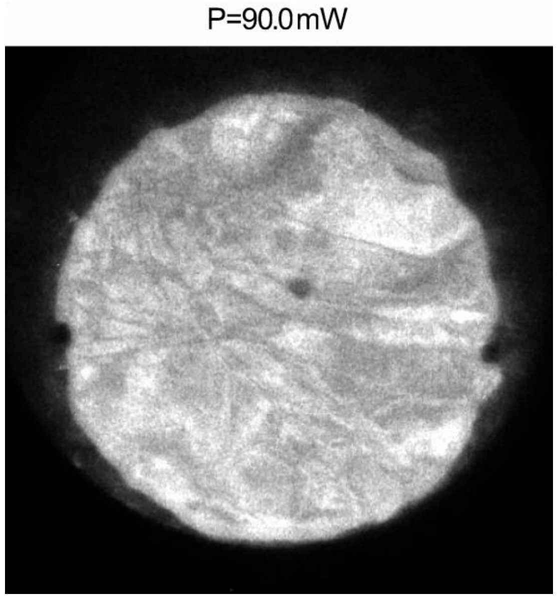


A. Feist et al., Struct. Dyn. (2018)

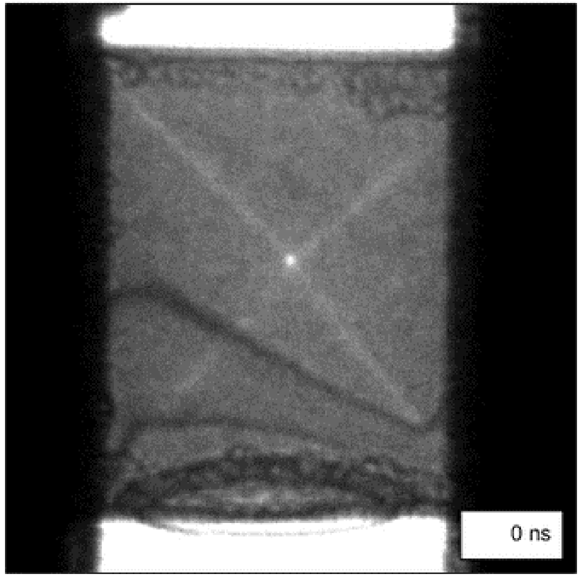
Structural dynamics



Phase transitions



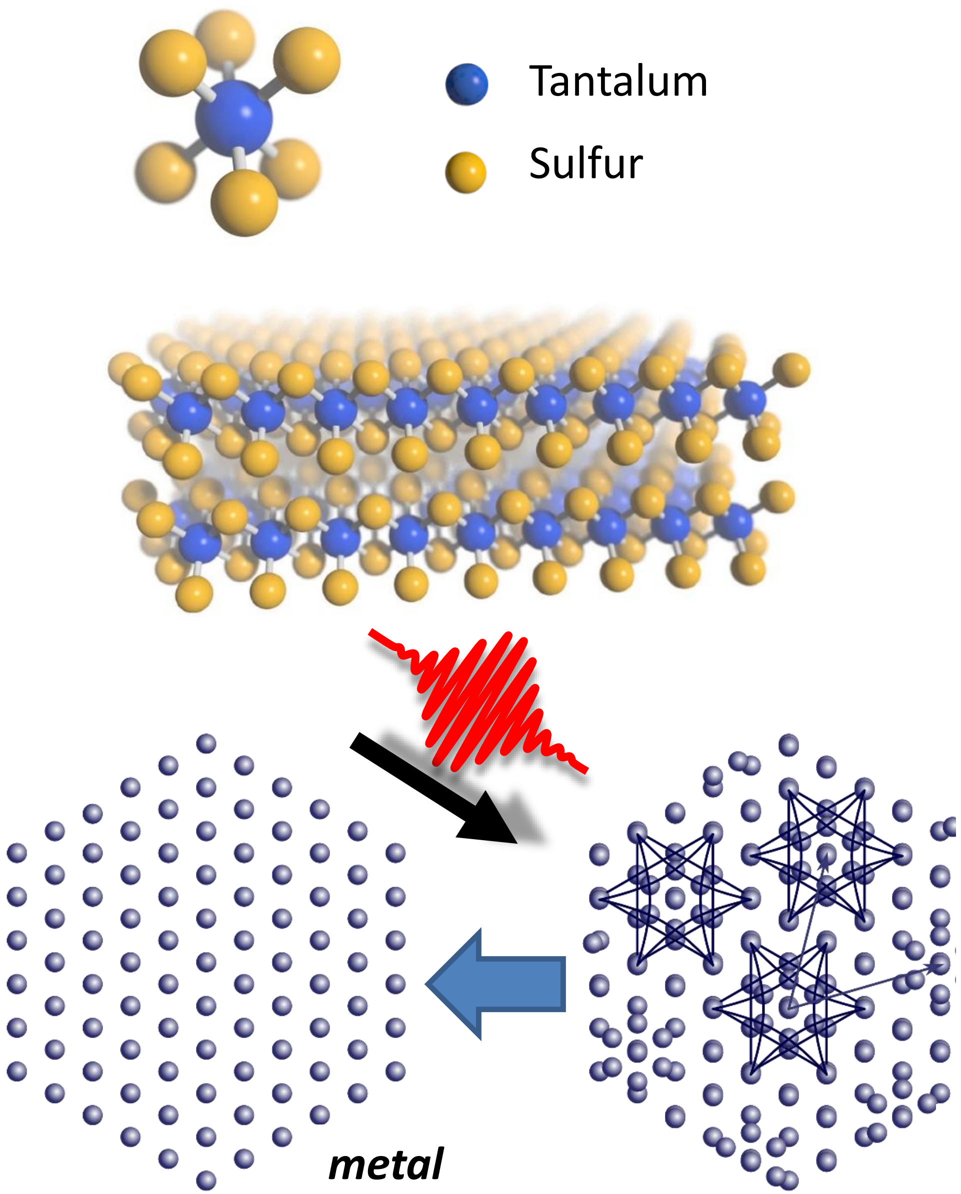
Th. Danz et al.,
Science (2021)

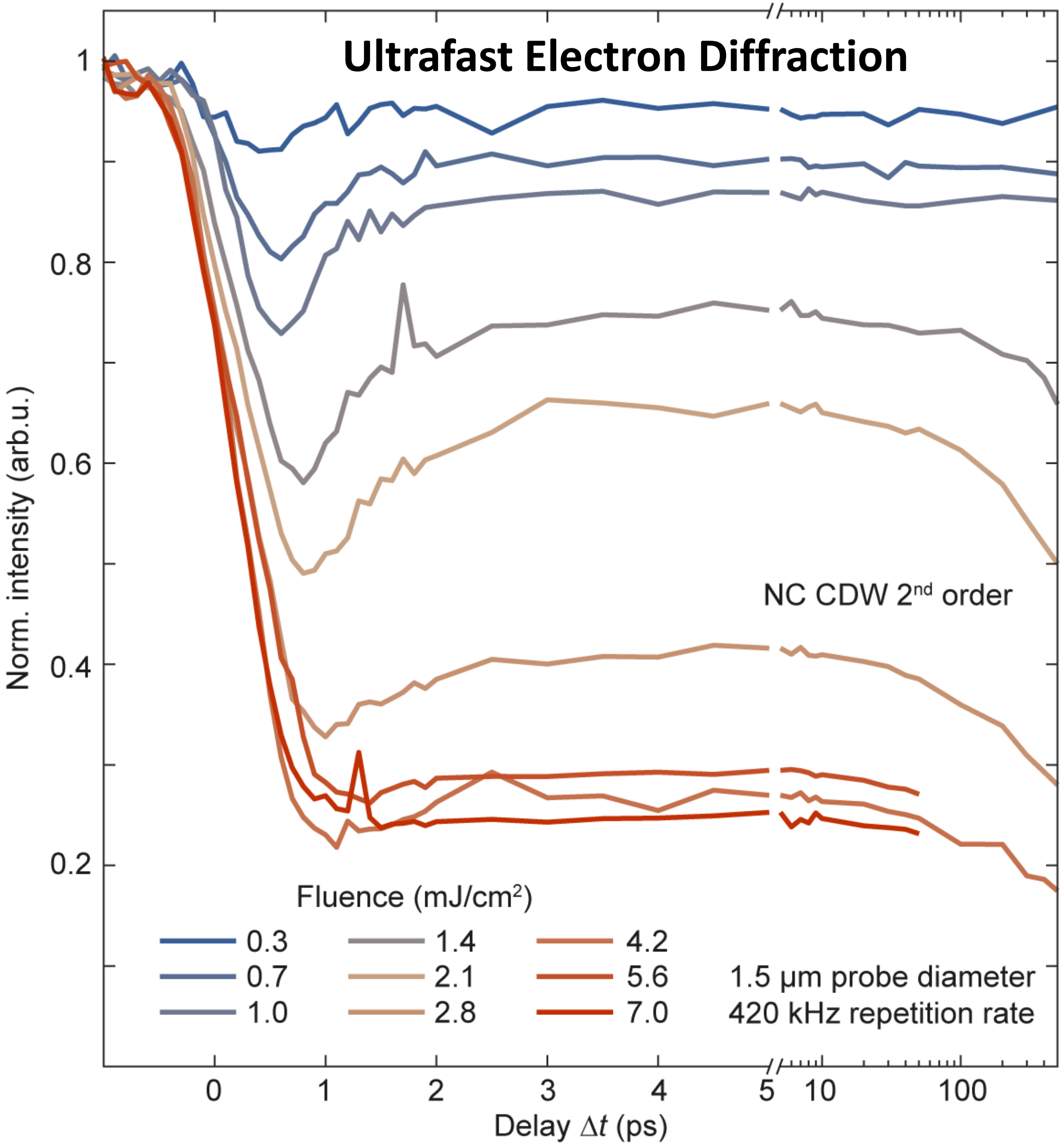


Magnetization Dynamics

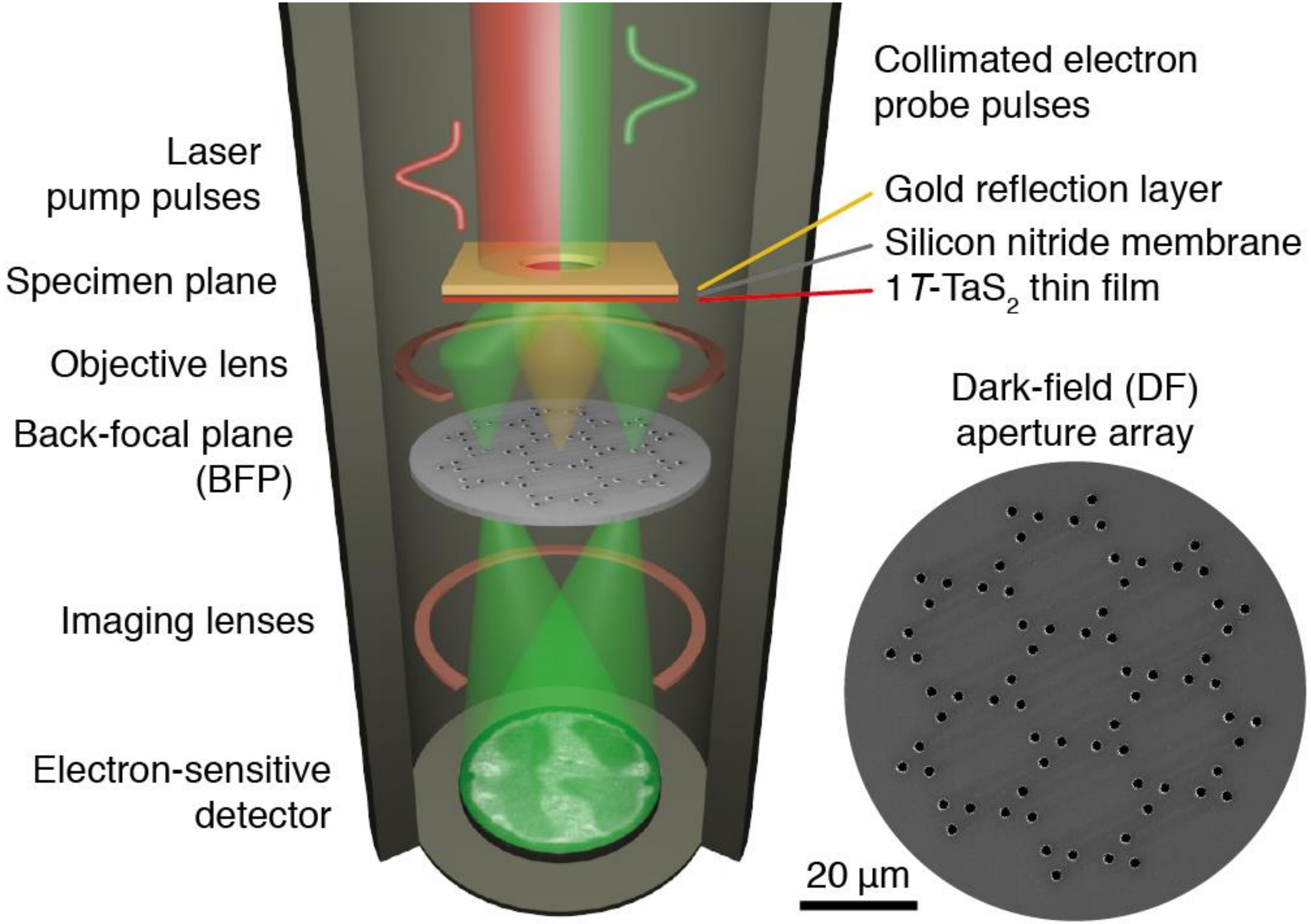
M. Möller *et al.*,
Comms. Phys. (2019)
Phys. Rev. Res. (2022)

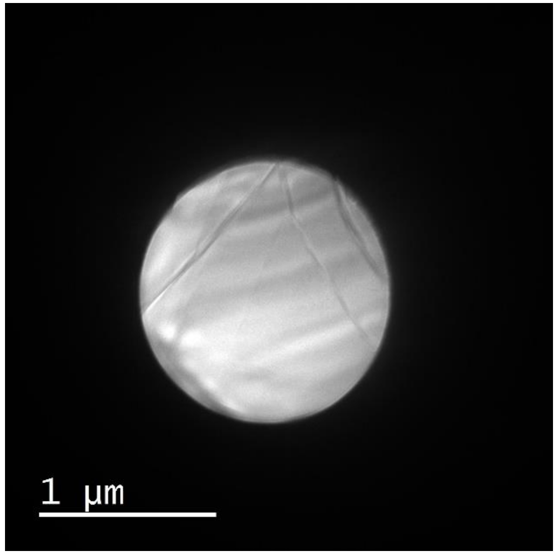
Ulrafast Imaging of a Metal-Insulator Transition

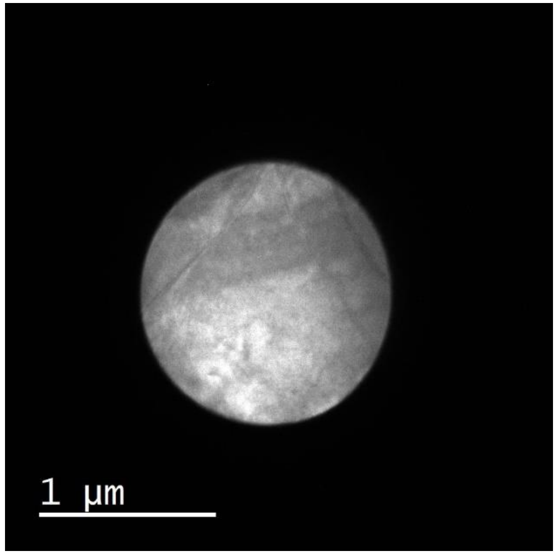




Ultrafast dark-field imaging

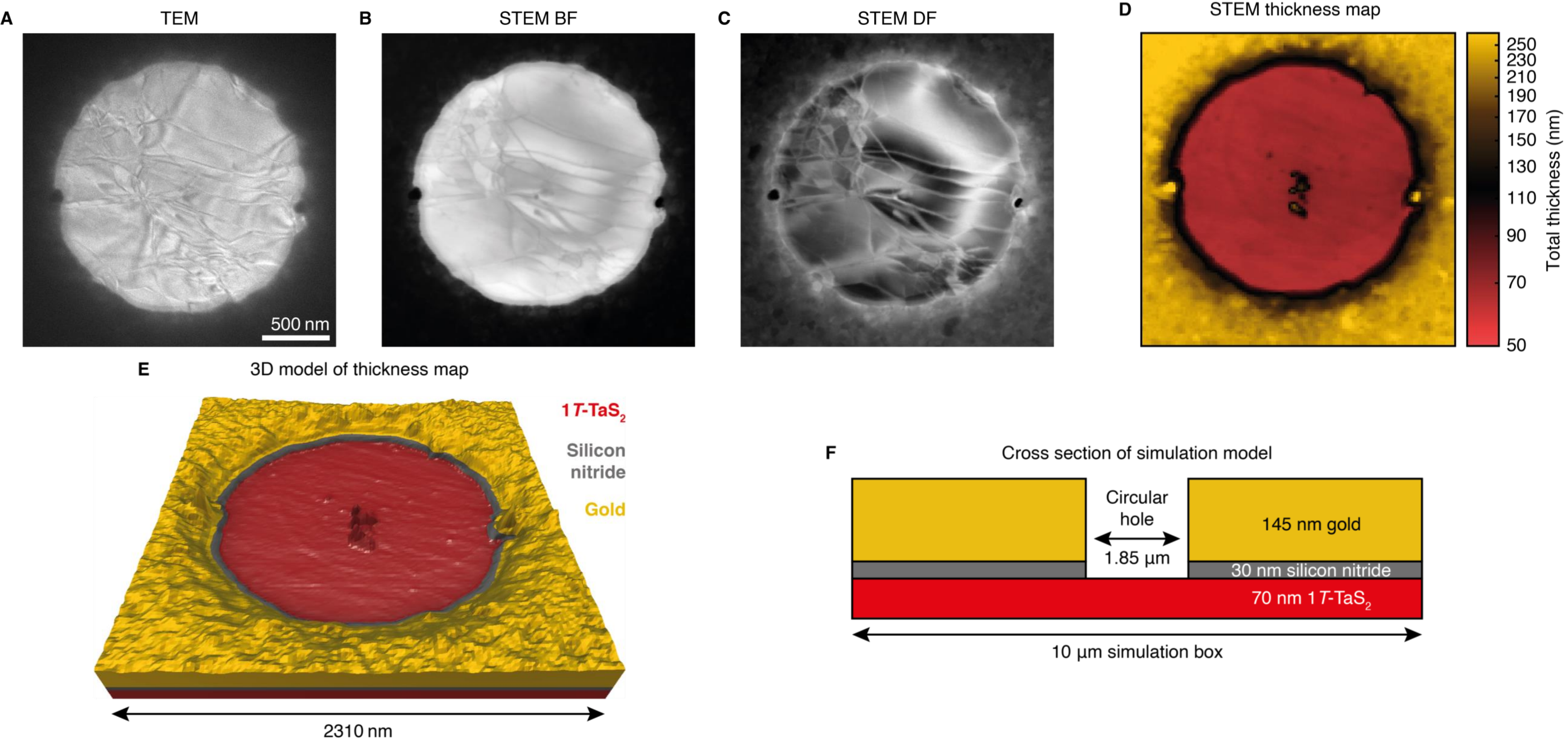






Th. Danz, T. Domröse, C. Ropers, Science (2021)

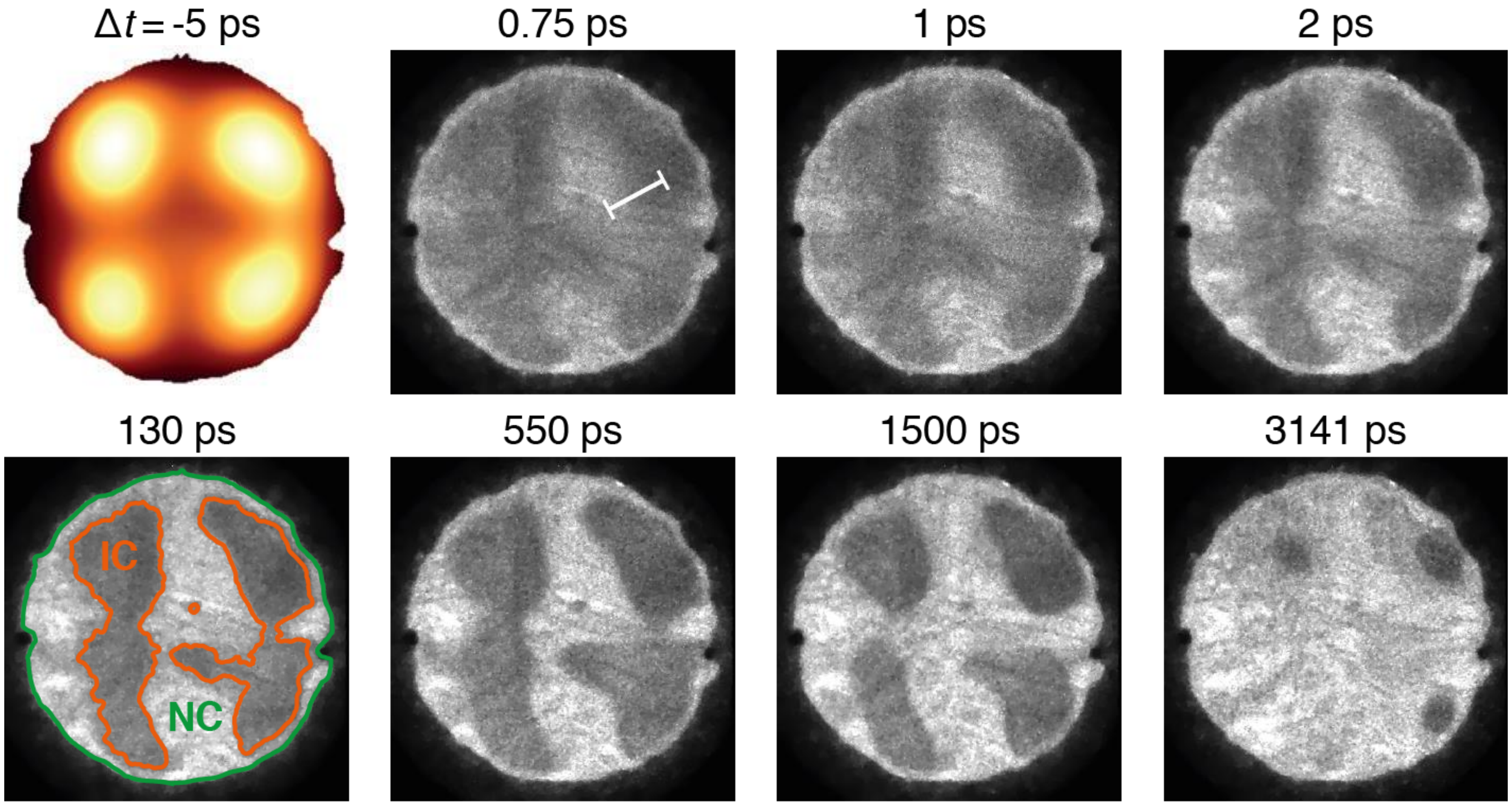
1*T*-TaS_2 sample design



Th. Danz, T. Domröse, C. Ropers, *Science* **371.6527** (2021): 371-374.

Sample: K. Rossnagel (Kiel)

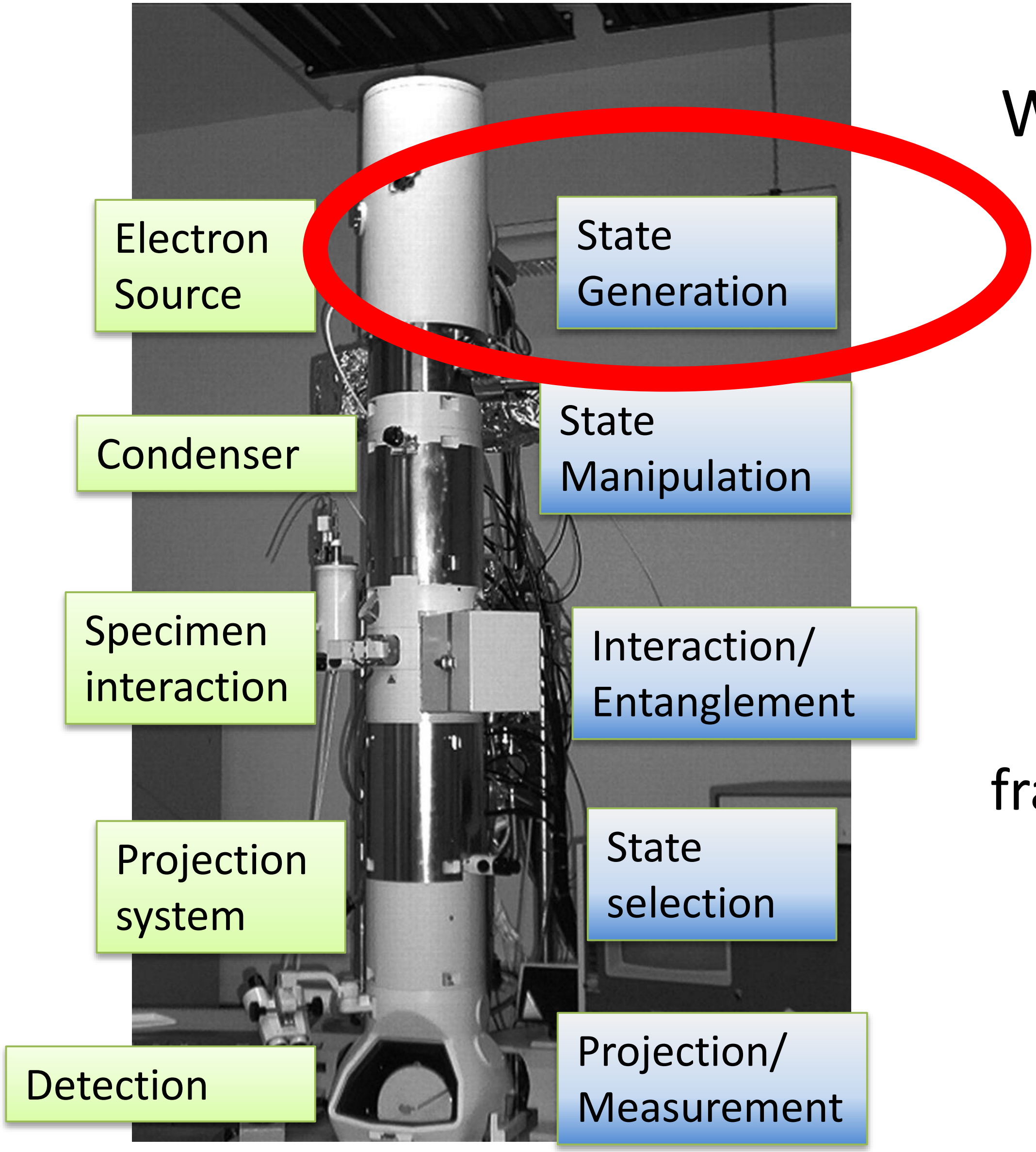
Ultrafast charge-density wave dynamics in 1*T*-TaS_2



- Rapid out-of-plane carrier transport
- Formation of phase boundaries
- Relaxation governed by thermal diffusion
- Study of phase transition at hundreds of kHz

Th. Danz, T. Domröse, C. Ropers, Science (2021)

Quantum probing in electron microscopy



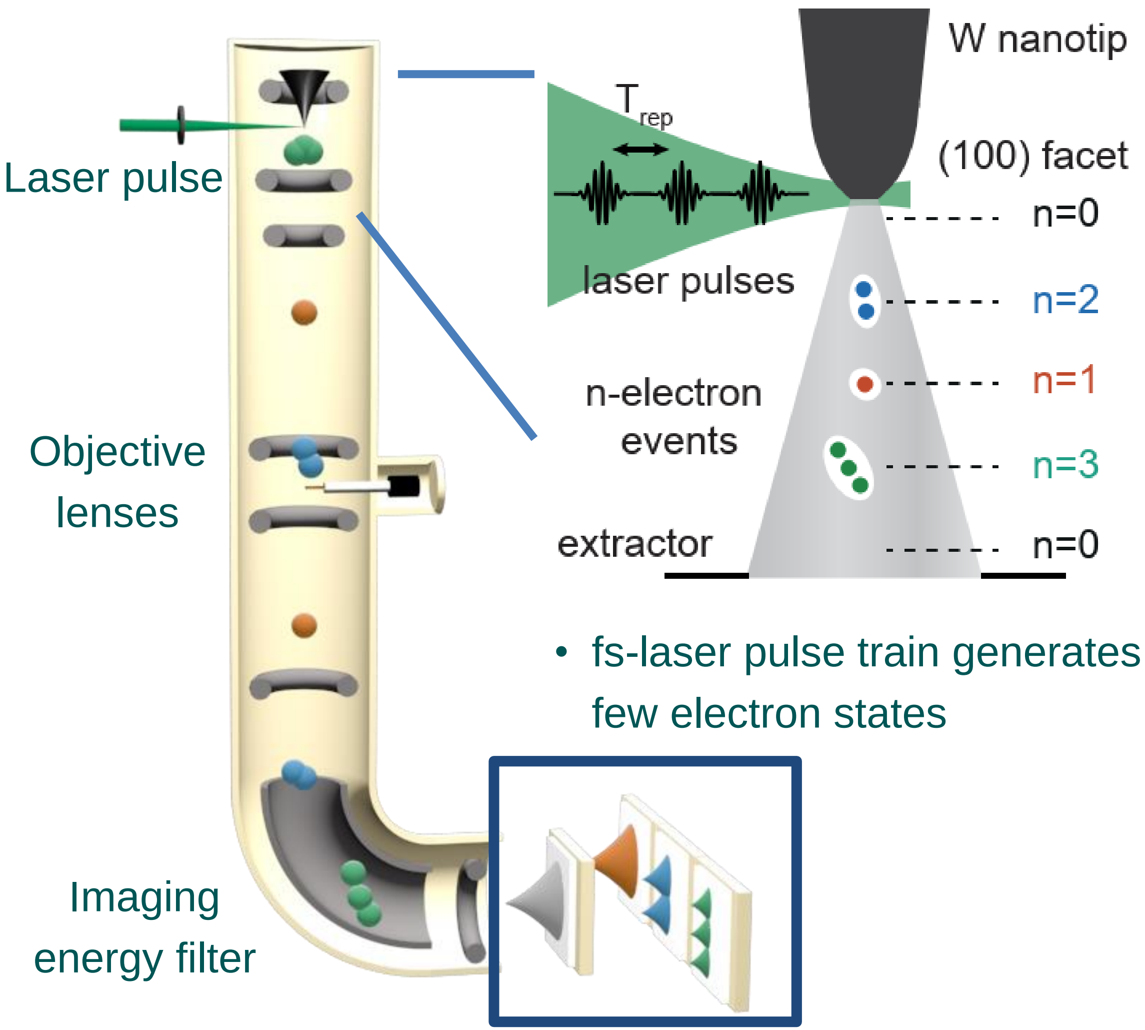
Want to develop strategies to **prepare, manipulate** and **characterize** the **quantum state** of electron pulses for novel measurement schemes

->Develop a „Quantum Optics“ framework for electron microscopy

Image: adapted from M. Haider

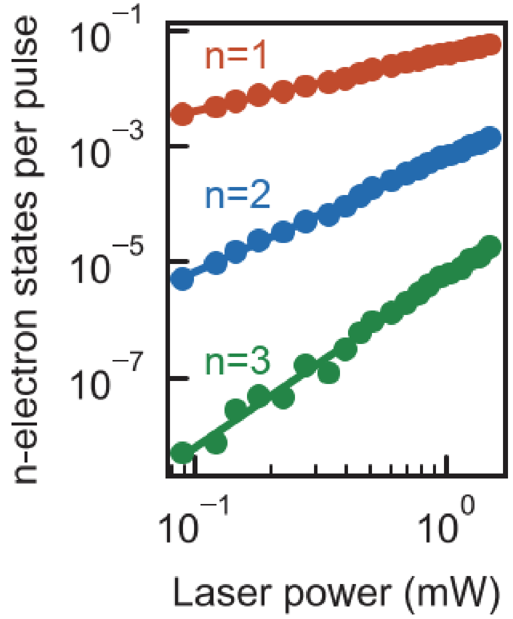
Counting electrons for each pulse

n-event classification



- fs-laser pulse train generates few electron states

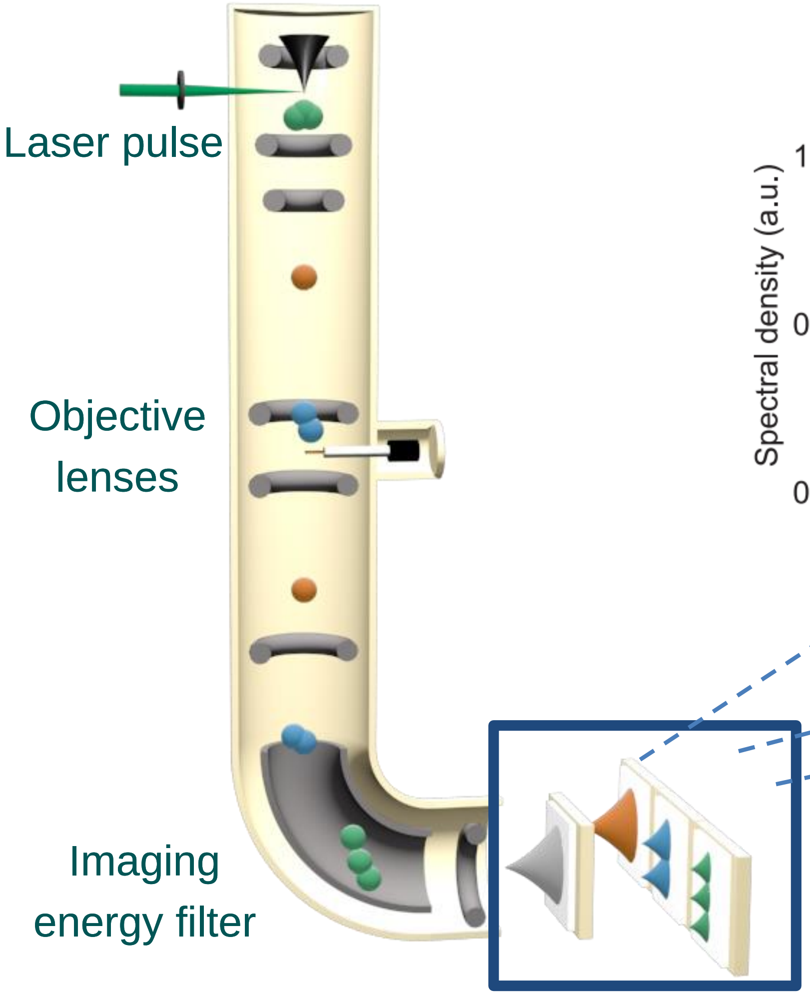
R. Haindl et al., arXiv:2209.12300 (2022)

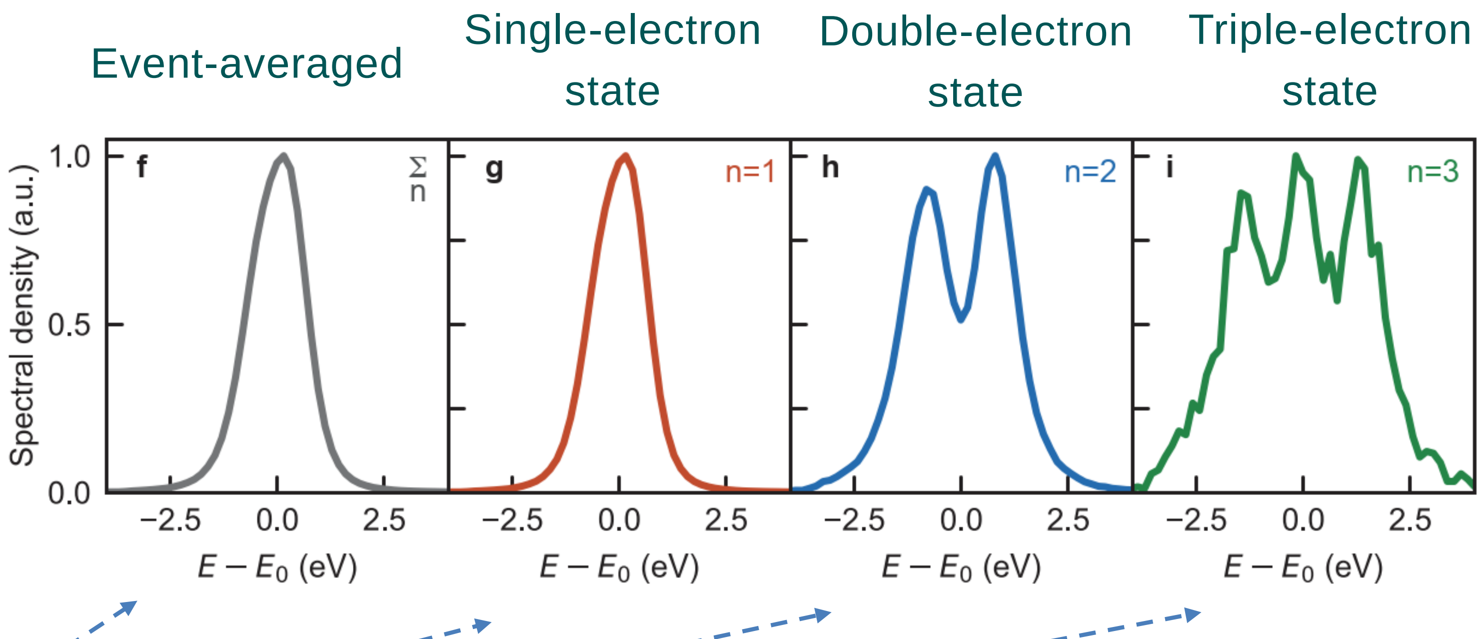


- We observe considerable antibunching.
- Poisson statistics: $P_n = r_n \frac{P_1^n}{n!}$ with $r_n = 1$
- In the measurement: $r_{n=2} = 0.85$, $r_{n=3} = 0.57$

see also: S. Keramati et al., Phys. Rev. Lett. 127, 180602 (2021)

Electron number-state spectra



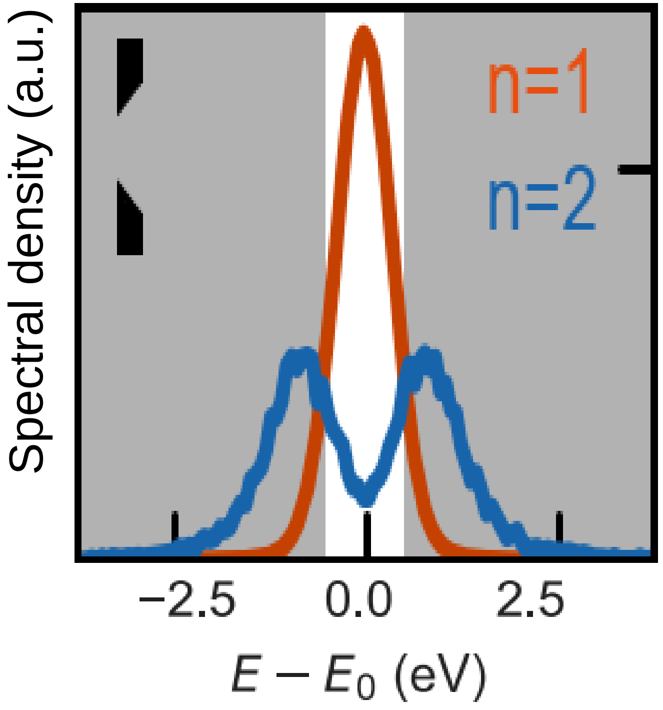


Characteristic single/double/triple lobe structure

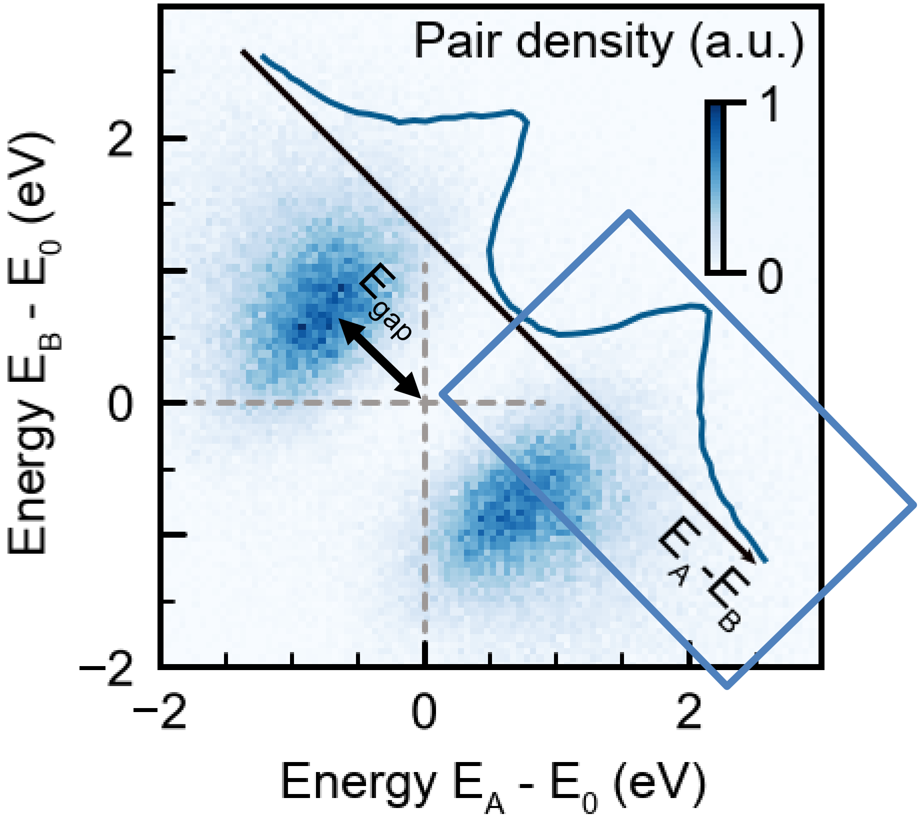
R. Haindl et al., arXiv:2209.12300 (2022)

Two-electron energy correlation

n=2 Spectrum

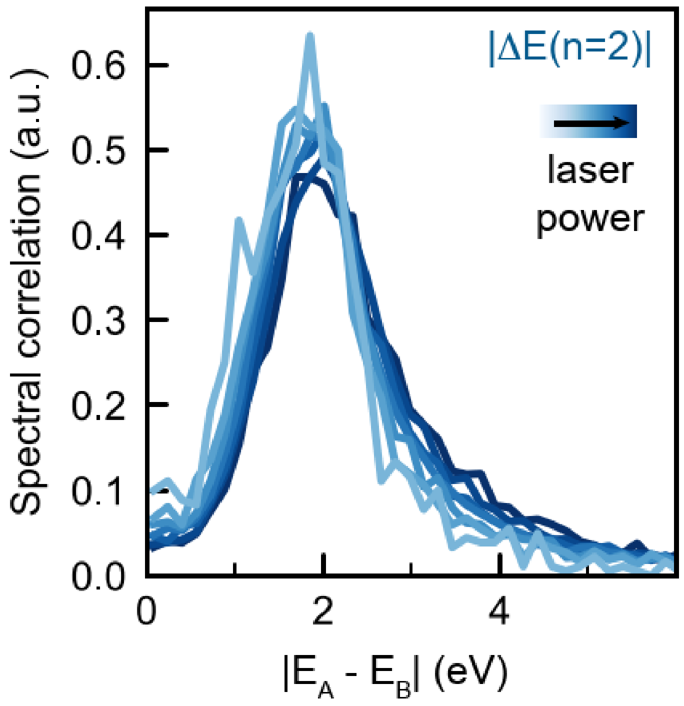


Electron pair distribution



- Correlation gap of 1.7 eV
- Energy antibunching

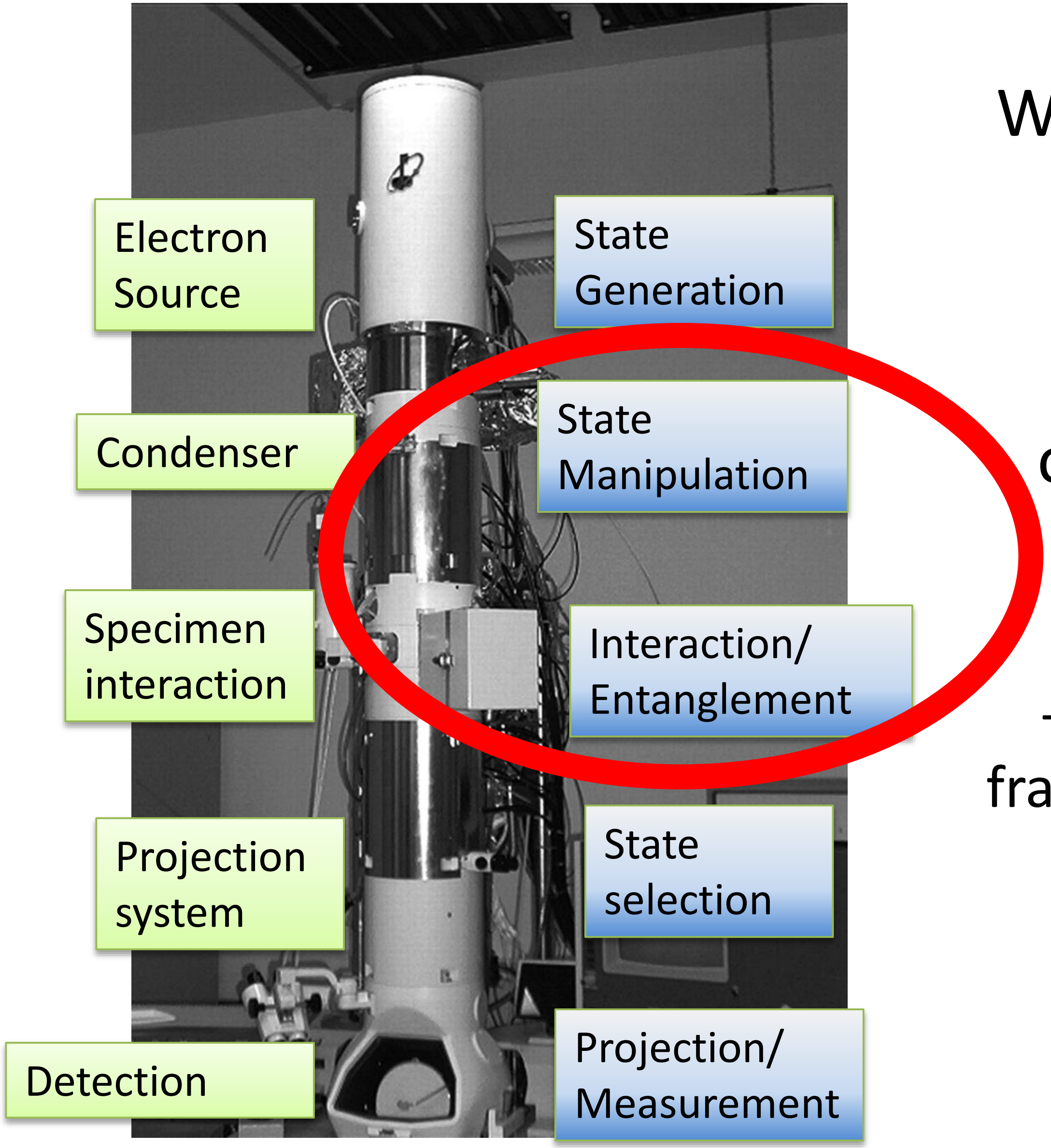
Two-electron correlation function



- Independent of laser power
- **Strongly Coulomb-correlated two-electron states**

R. Haindl et al., arXiv:2209.12300 (2022)

Quantum probing in electron microscopy



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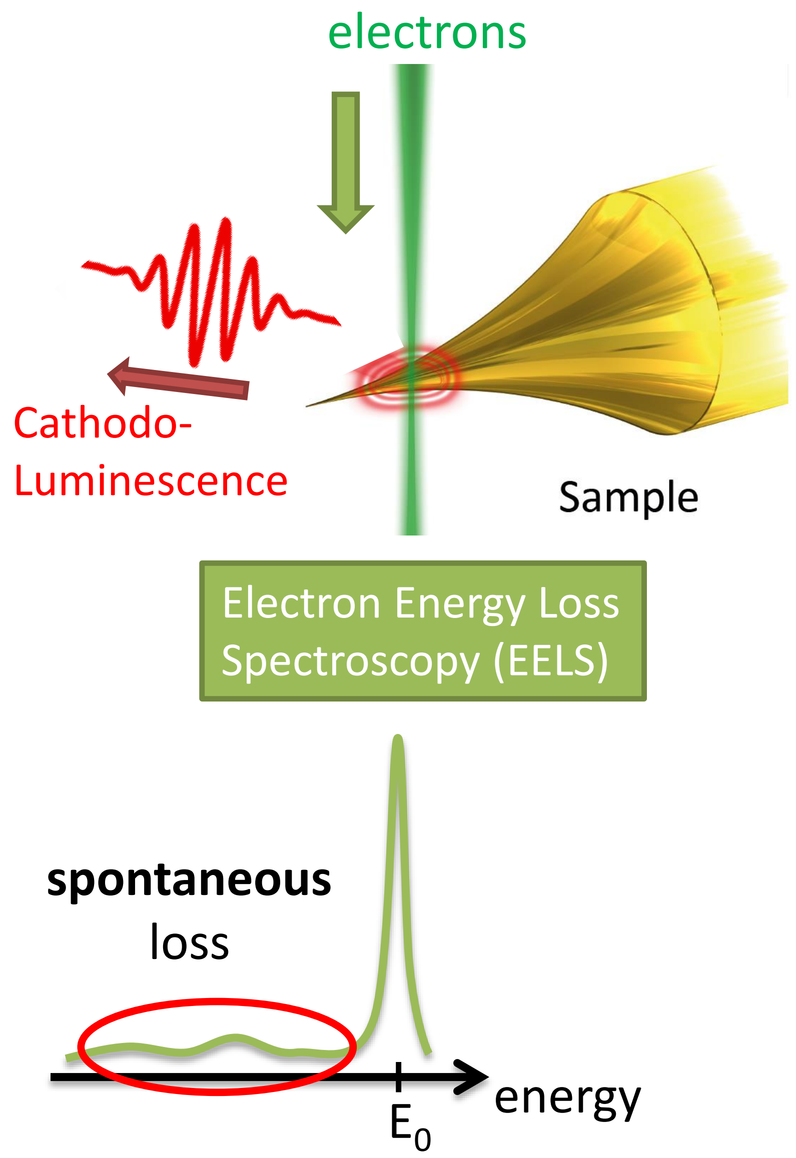
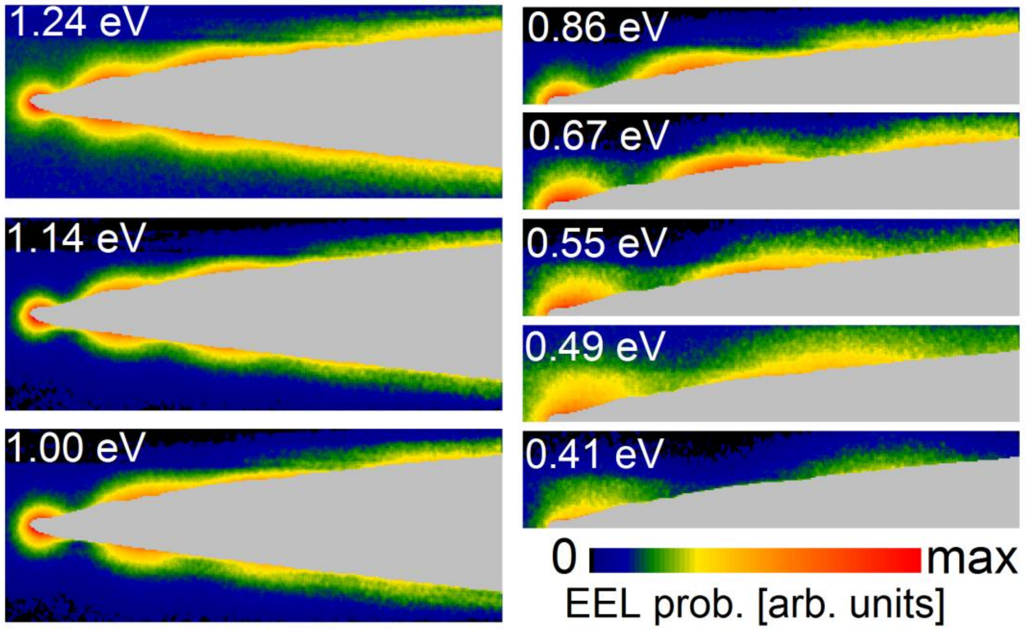
Interaction of fast electrons with optical near fields

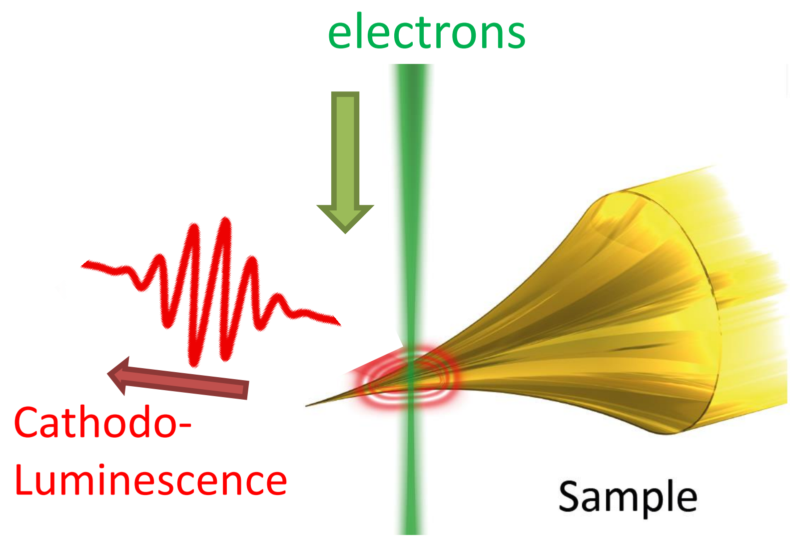




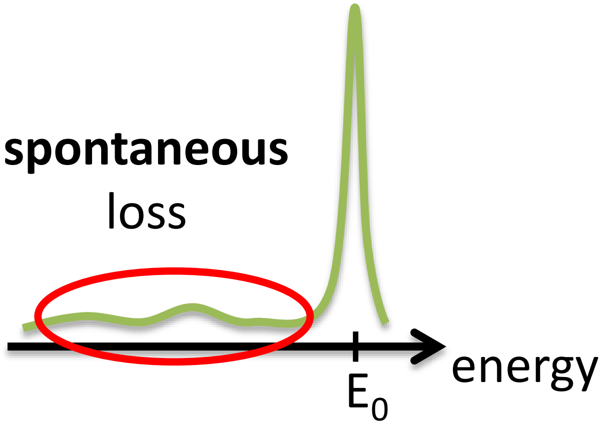
Real-space imaging of nanotip plasmons
B. Schröder *et al.*, Phys. Rev. B (2015)

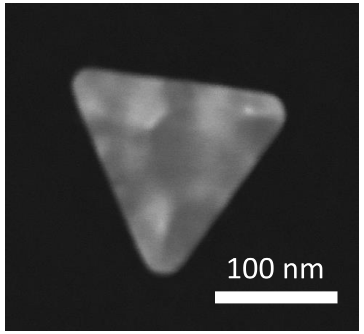
Literature:
Feist et al., Nature (2015)
Barwick et al., Nature (2009)
A. Howie, Inst. Phys. Conf. Ser. 161, 311 (1999)
F.J. García de Abajo *et al.*, Nano Lett. (2010)
S.T. Park *et al.*, NJP (2010)

Interaction of fast electrons with optical near fields

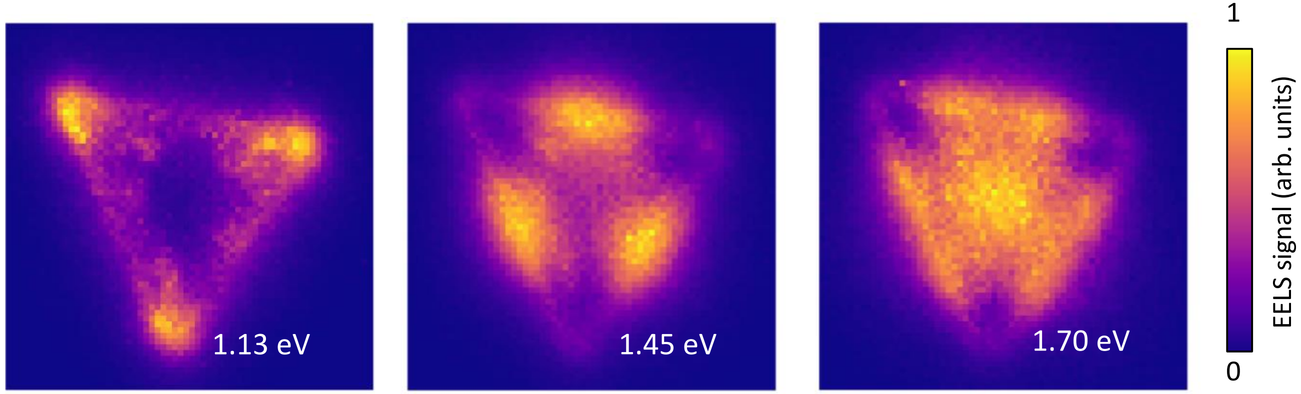


Electron Energy Loss Spectroscopy (EELS)





Energy-filtered EELS maps

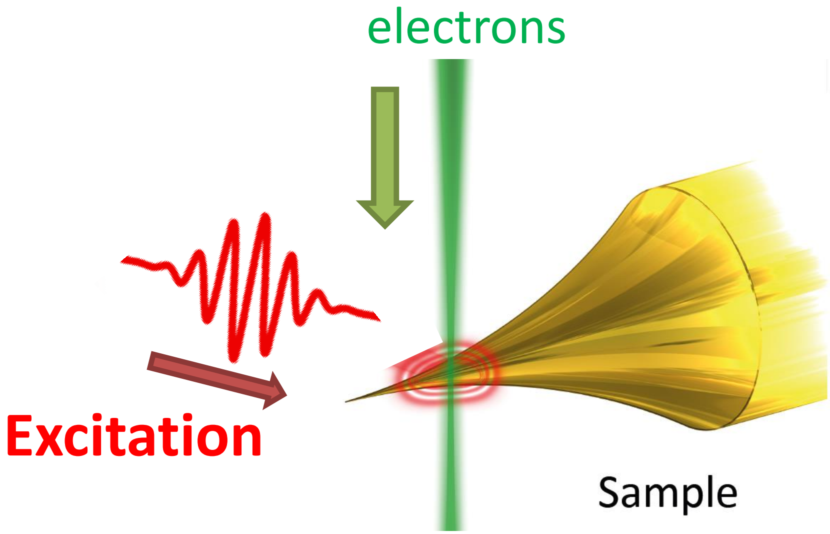


$$\Gamma_{EELS}(\omega) = \frac{e}{\pi\hbar\omega}\int dt\, Re\{e^{-i\omega t} v \cdot E^{ind}[r_e(t),\omega]\}$$

loss probability — self-induced E-field

See e.g.: F. J. Garcia de Abajo, Rev. Mod. Phys. 82, 209 (2010)

Interaction of fast electrons with optical near fields



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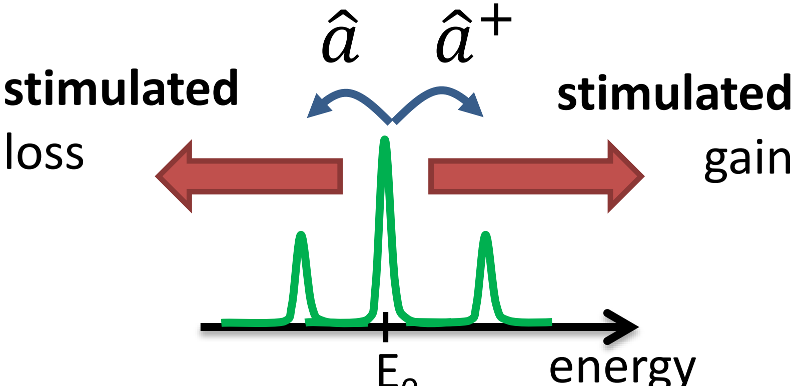

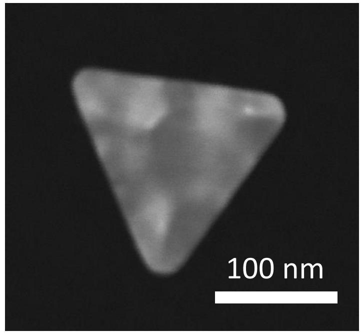

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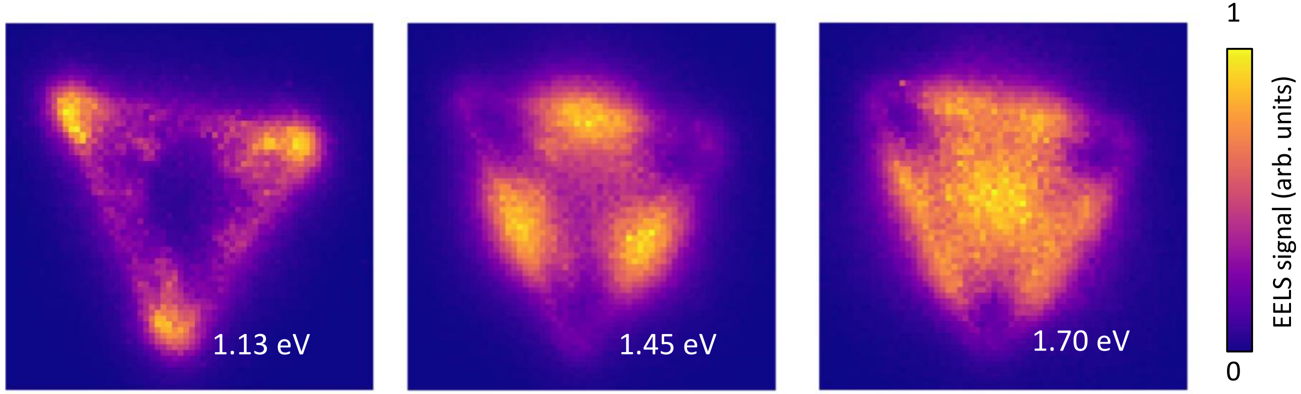

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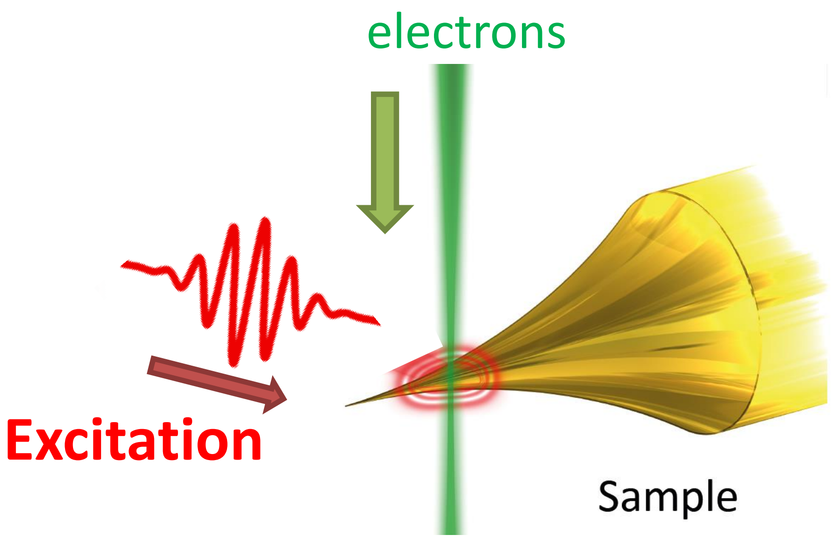

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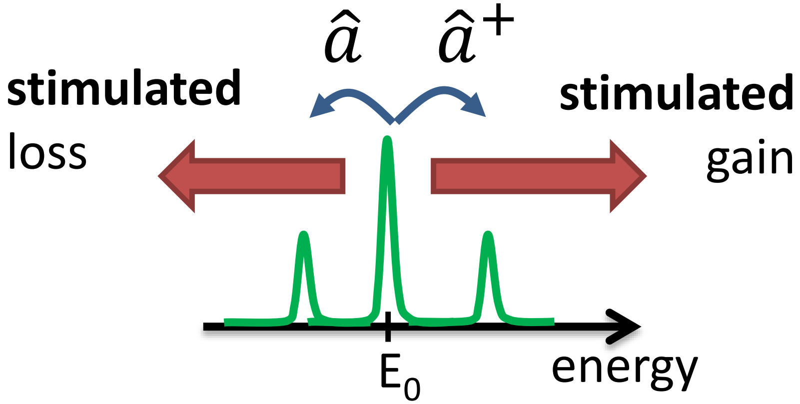
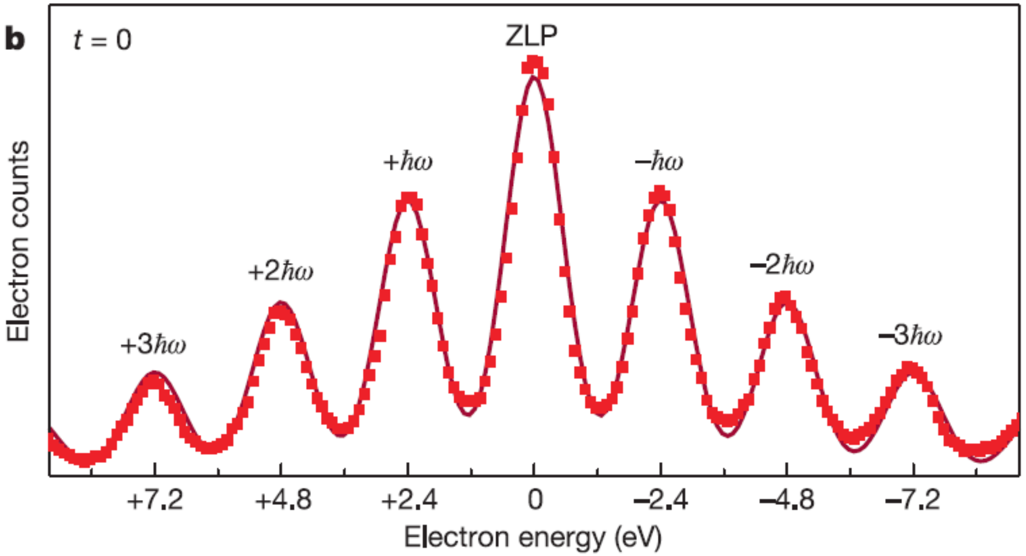




B. Barwick *et al.*, Nature (2009)

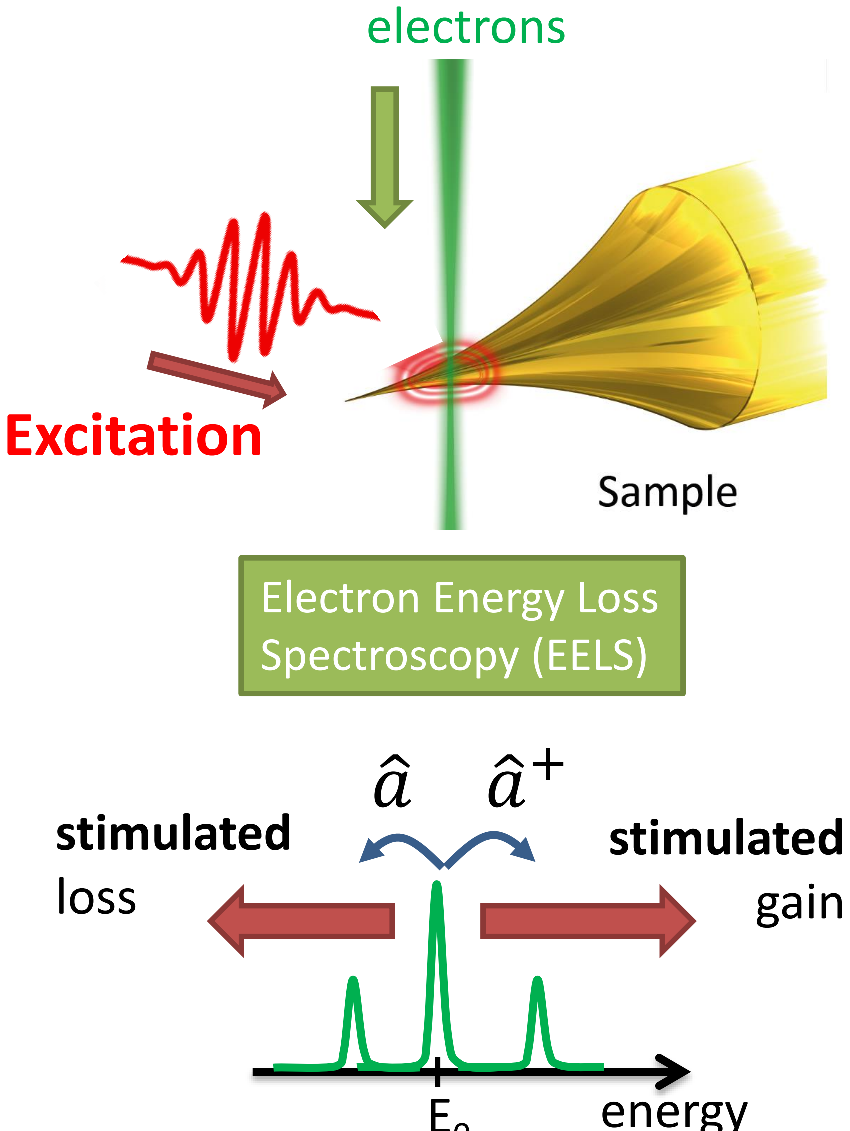
Photon-induced near-field electron microscopy (PINEM)

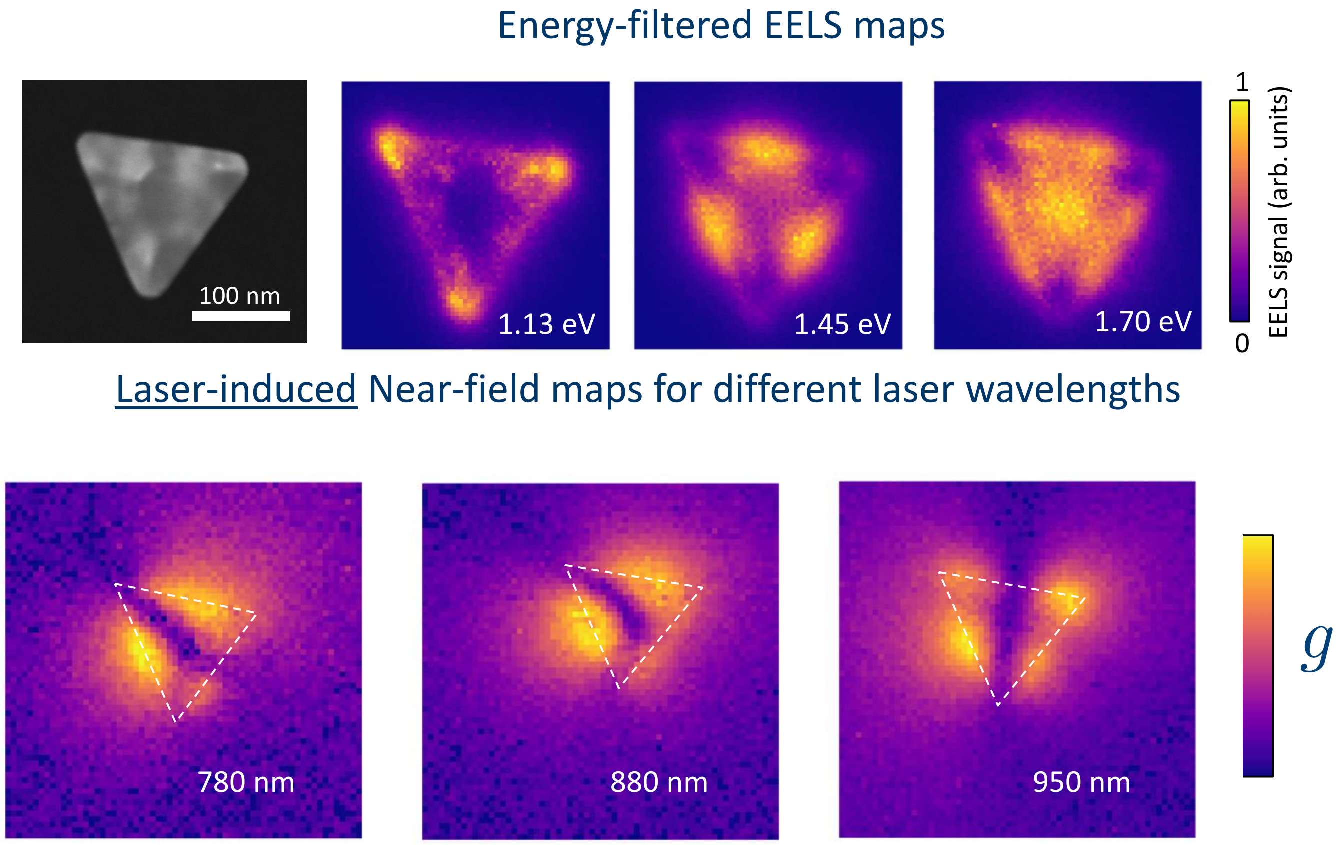
$$\Gamma(\omega) = \frac{e}{\pi\hbar\omega}\int dt\; Re\{e^{-i\omega t} v \cdot E^{ext}[r_e(t),\omega]\}$$

Loss/gain probability — external E-field

See e.g.: F. J. Garcia de Abajo, Rev. Mod. Phys. 82, 209 (2010)

Interaction of fast electrons with optical near fields

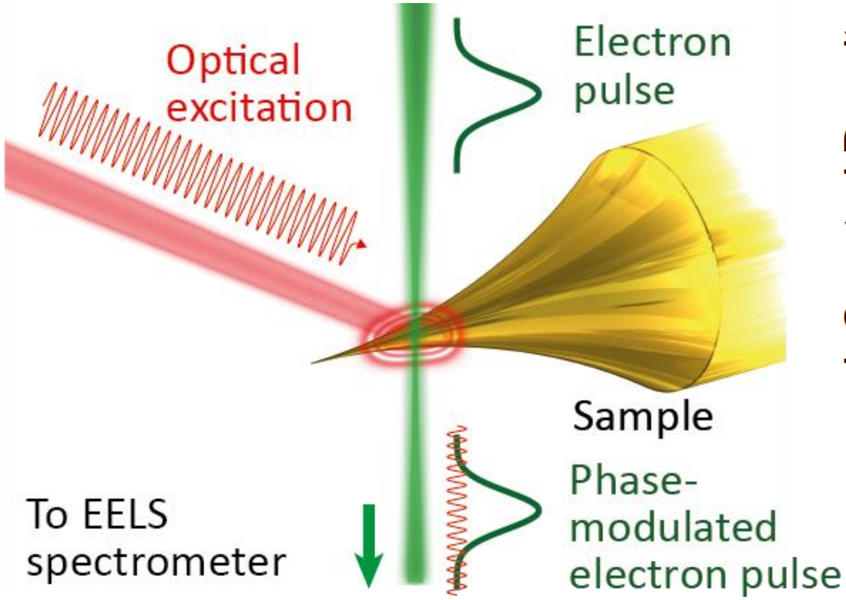


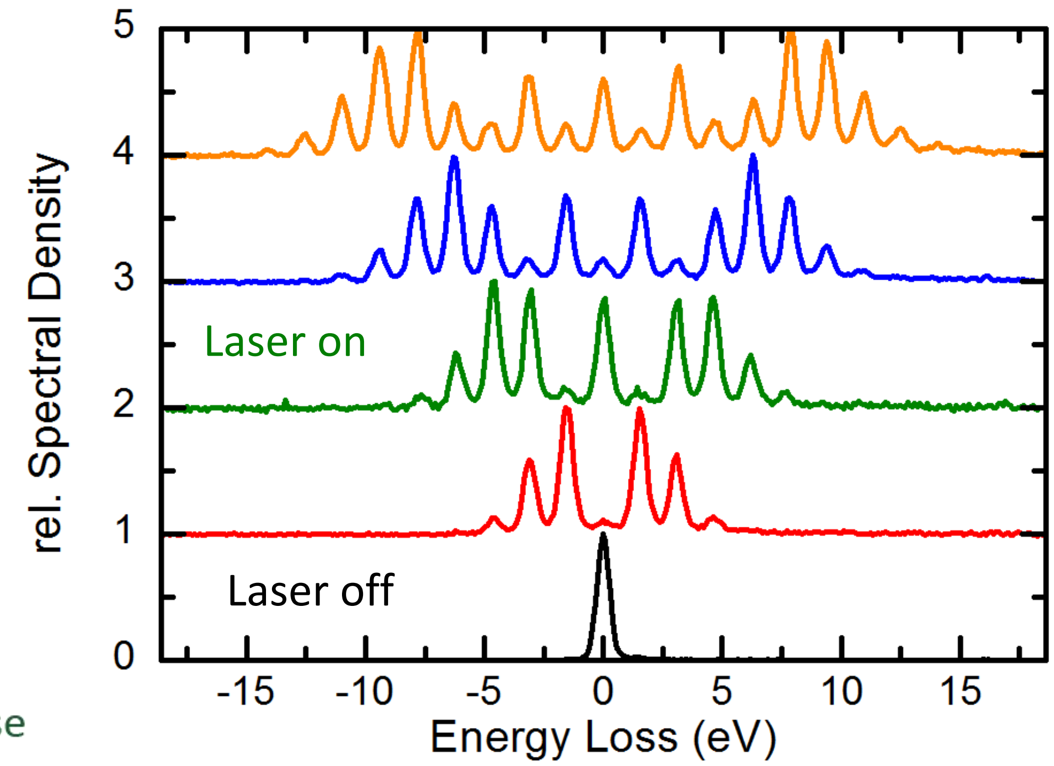


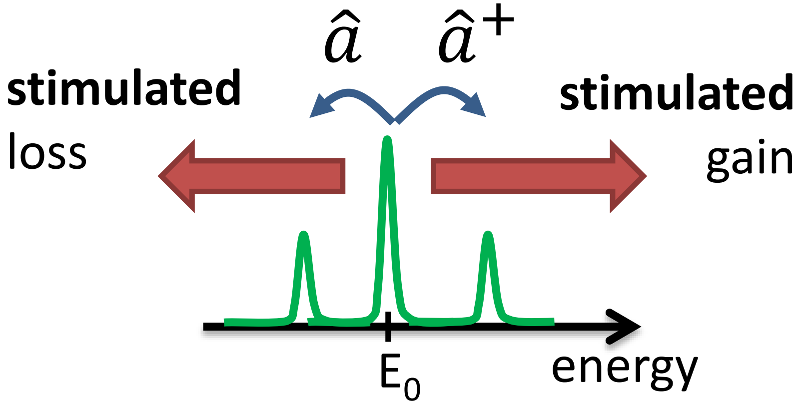
PINEM probes specific modes for incident frequency and polarization

H. Lourenco-Martins (unpublished, 2022)

Interaction of fast electrons with optical near fields





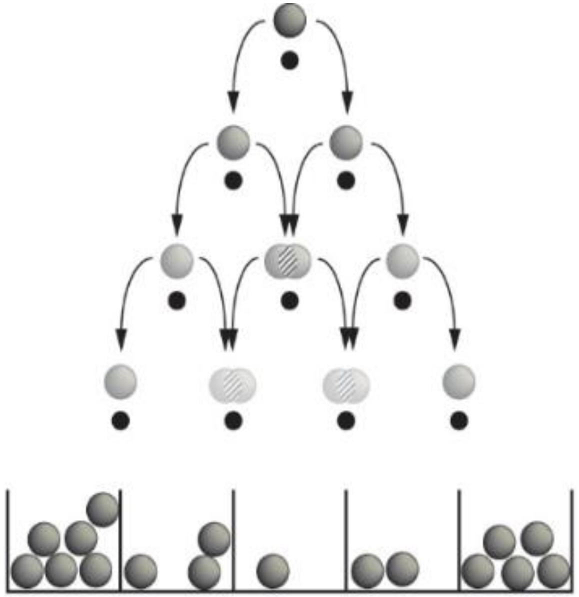


A. Feist *et al.*, Nature **521**, 200-203 (2015)

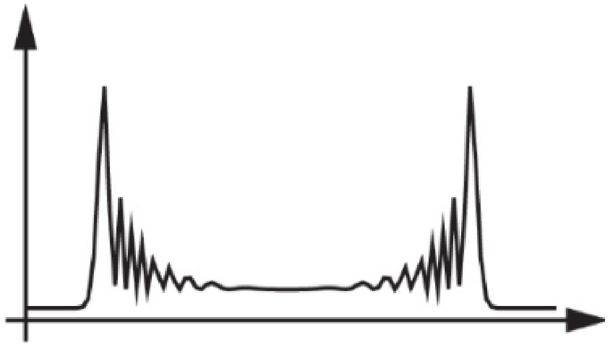
Literature:

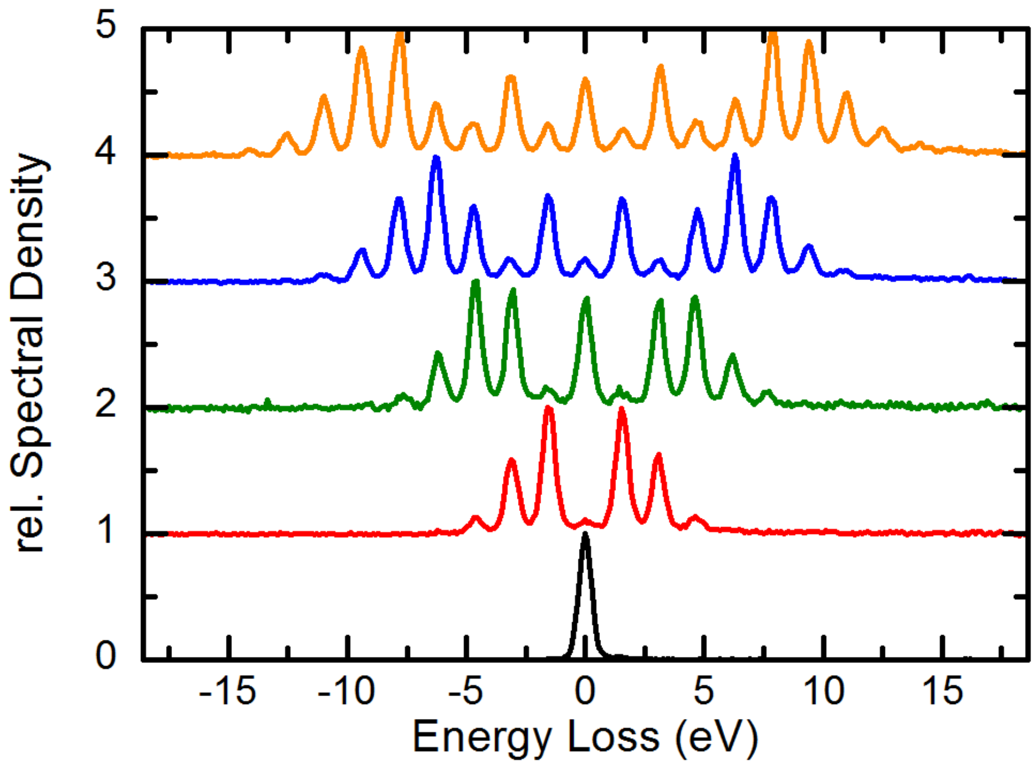
Feist et al., Nature (2015)
B. Barwick *et al.*, Nature (2009)
A. Howie, Inst. Phys. Conf. Ser. 161, 311 (1999)
F.J. García de Abajo *et al.*, Nano Lett. (2010)
S.T. Park *et al.*, NJP (2010)

Equivalent to a continuous-time quantum walk



$$\hat{S} = \exp\left(g_0 \hat{a}^{+} \hat{b} - h.c.\right)$$





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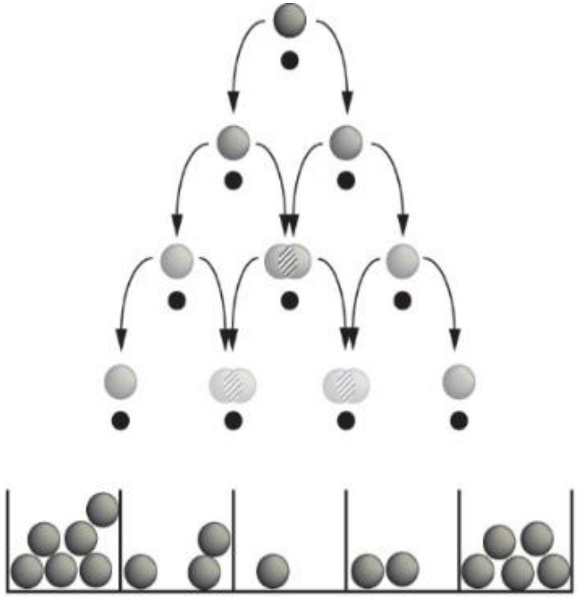
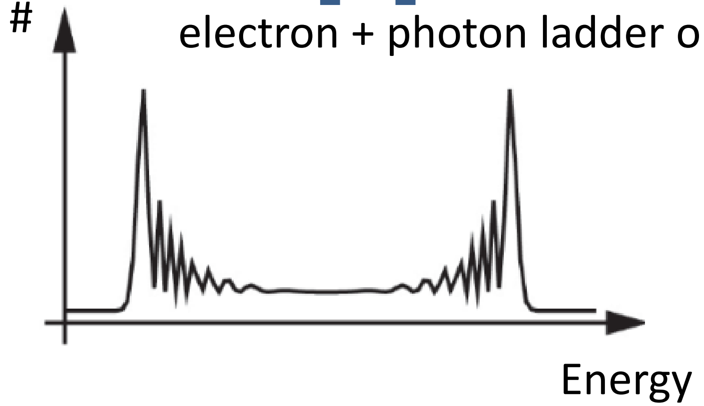
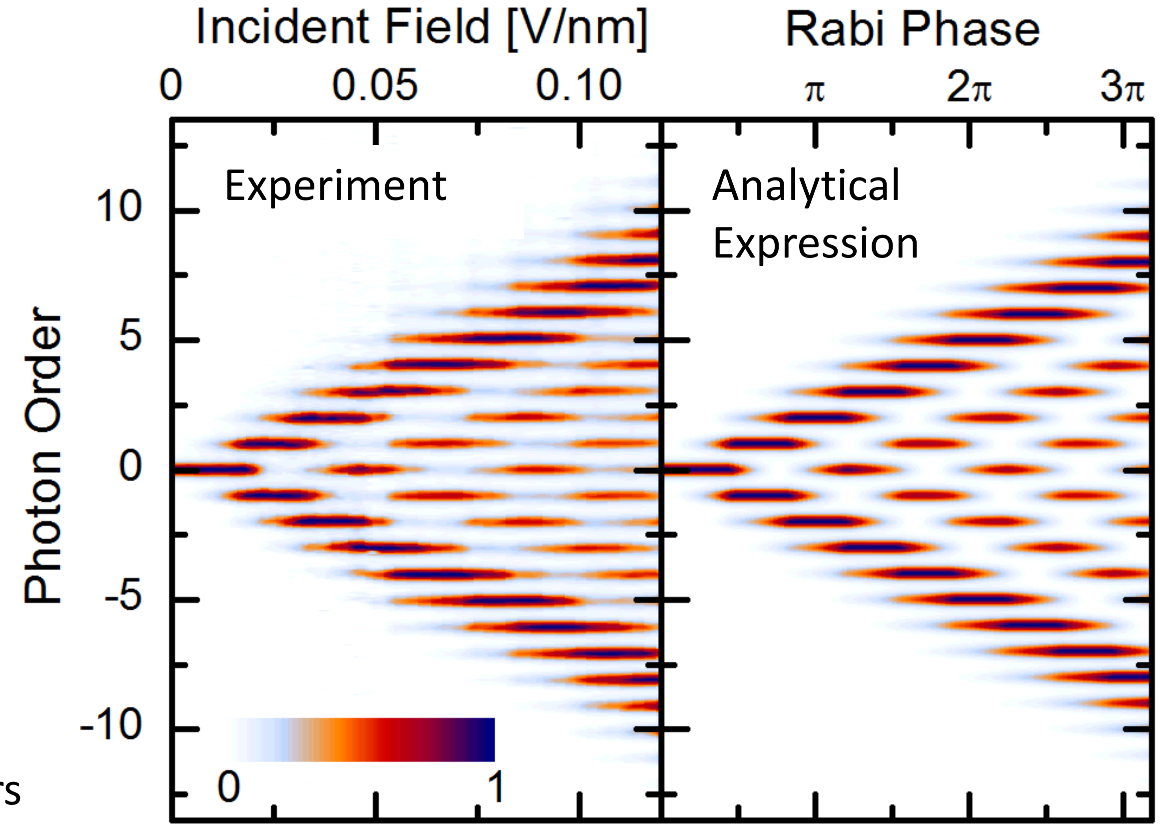


$$\hat{S} = \exp\left(g_0 \hat{a}^+ \hat{b} - h.c.\right)$$

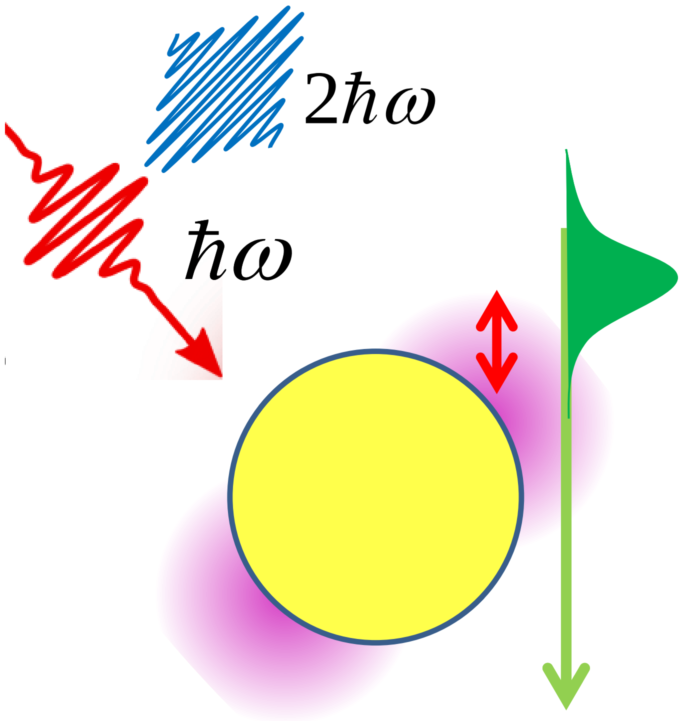
electron + photon ladder operators

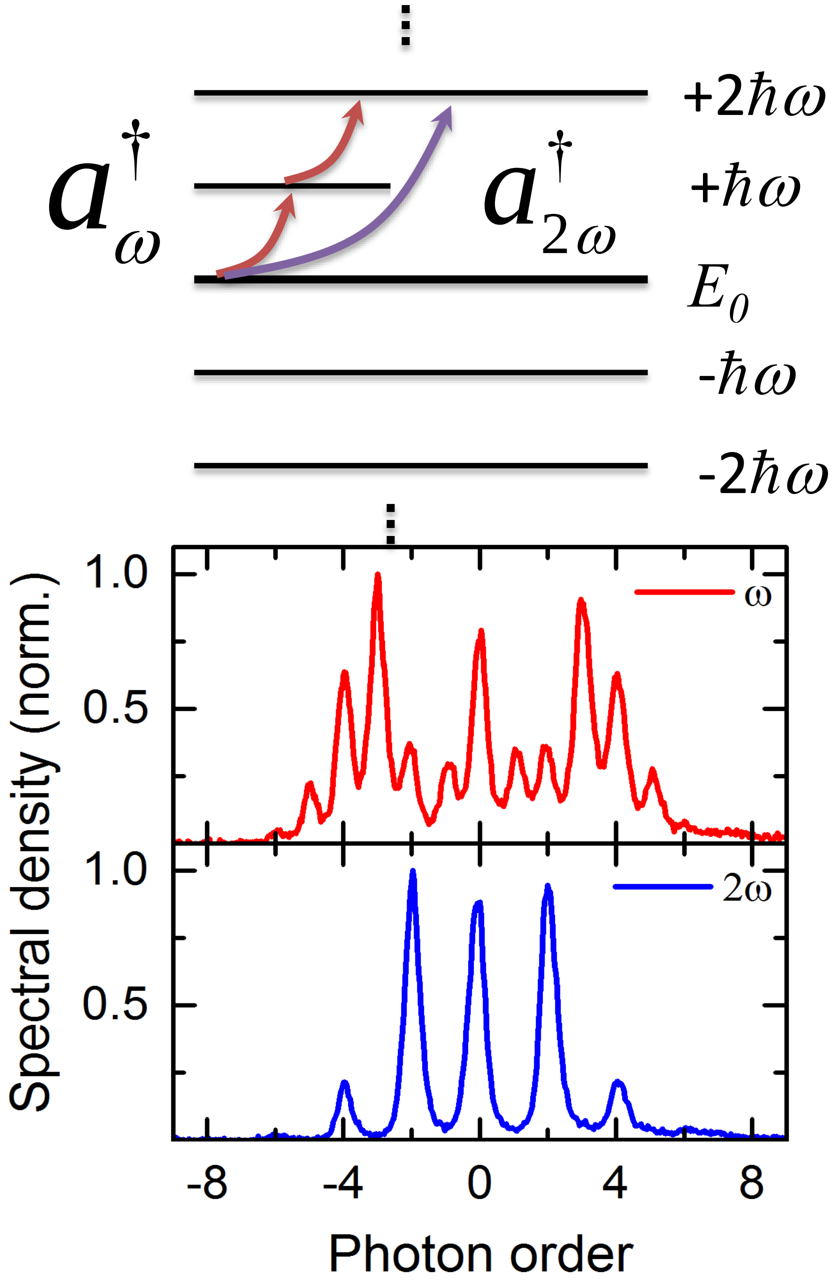




A. Feist *et al.*, Nature **521**, 200-203 (2015)

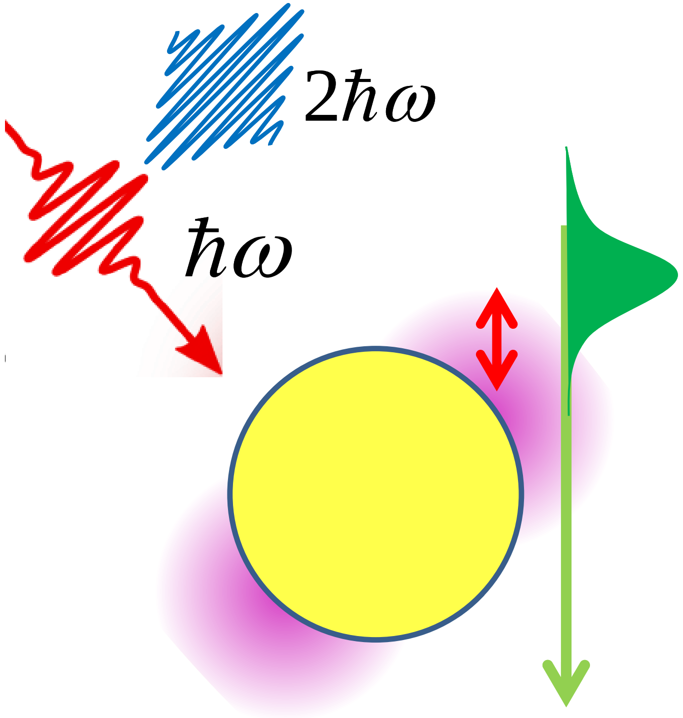
Two-colour coherent state control

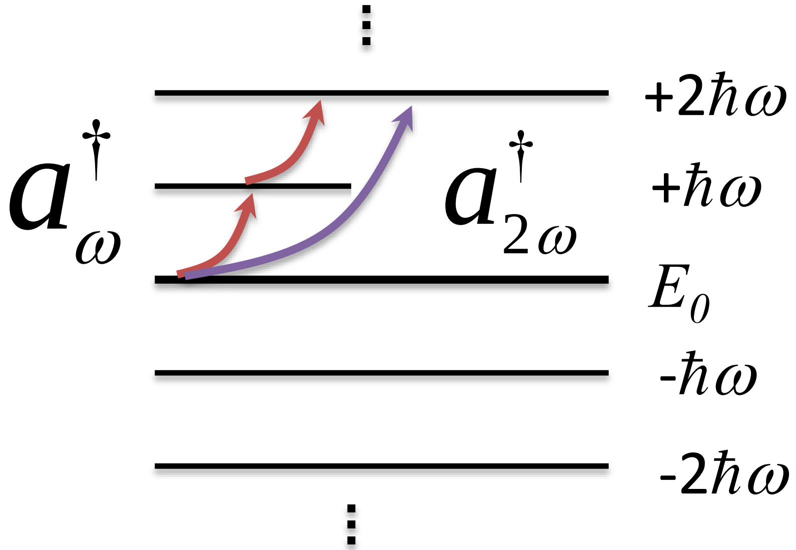


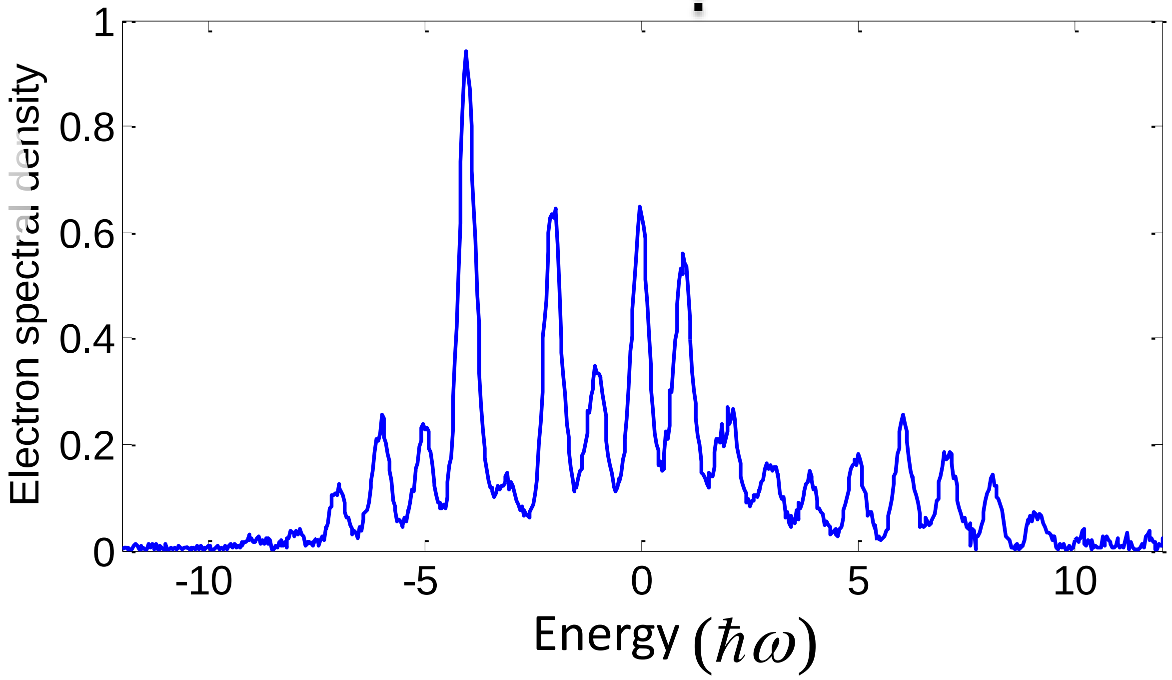


K. Priebe *et al.* Nat. Phot. 11, 793 (2017)

Two-colour coherent state control

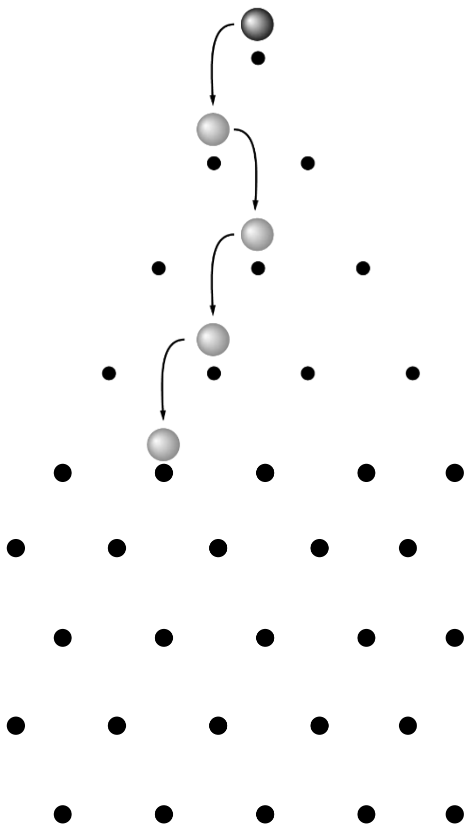




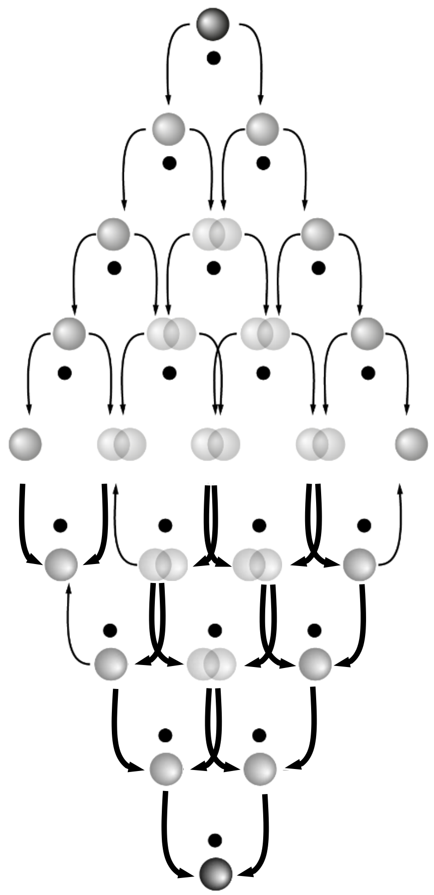


K. Priebe *et al.* Nat. Phot. 11, 793 (2017)

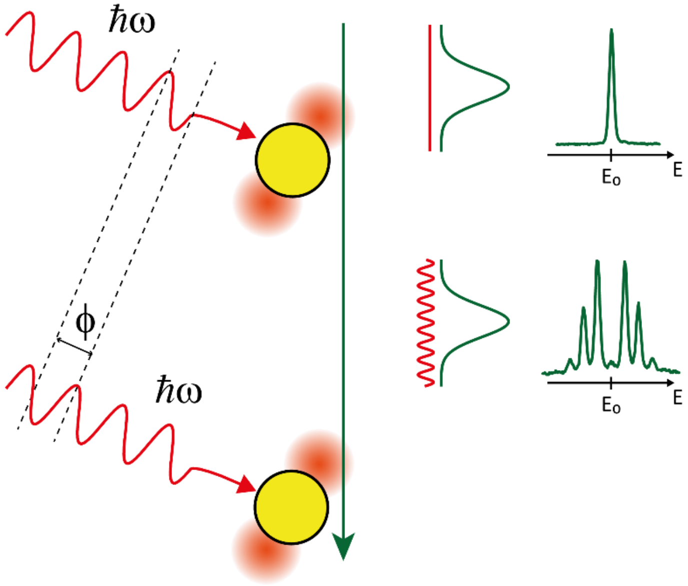
Two sequential Galton boards

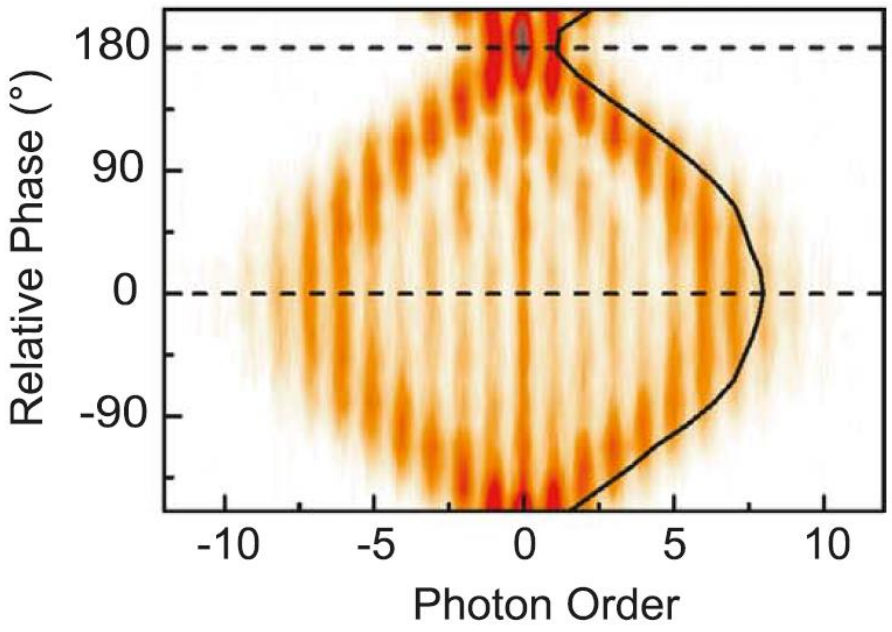


?



Ramsey-type double interaction

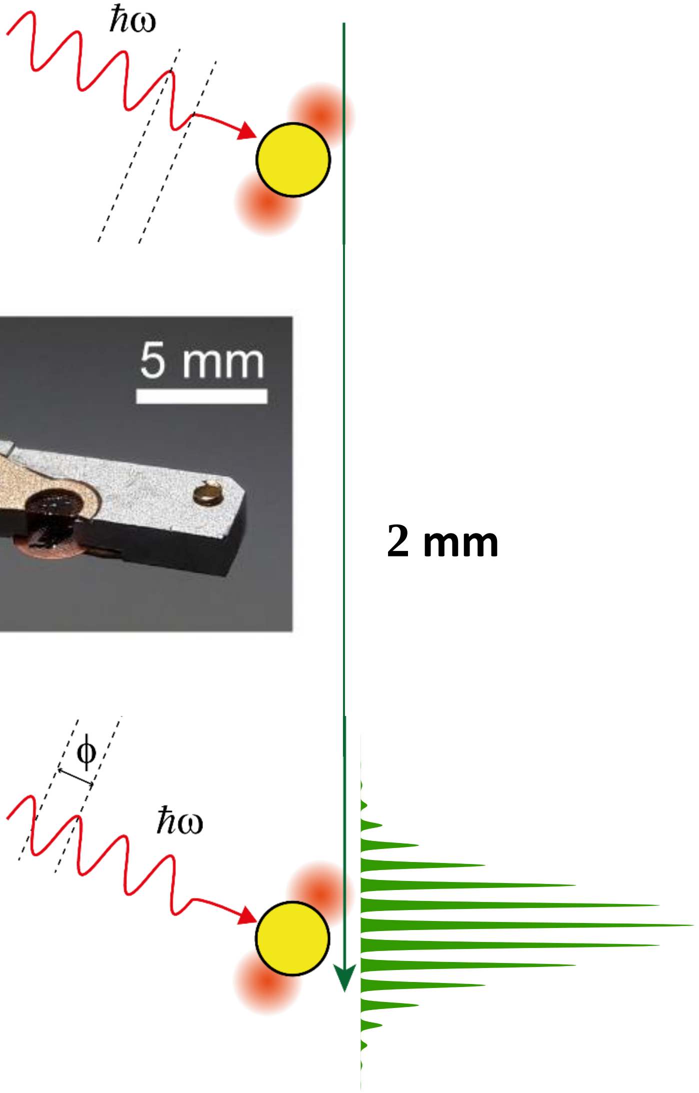


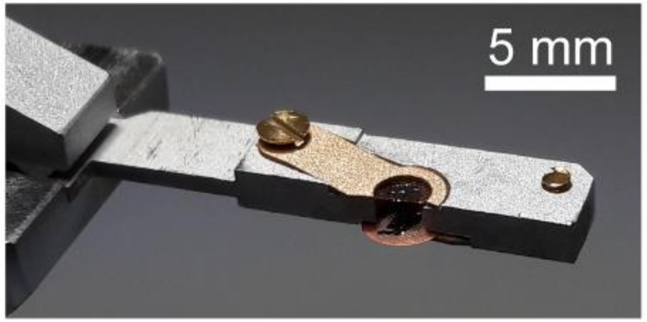


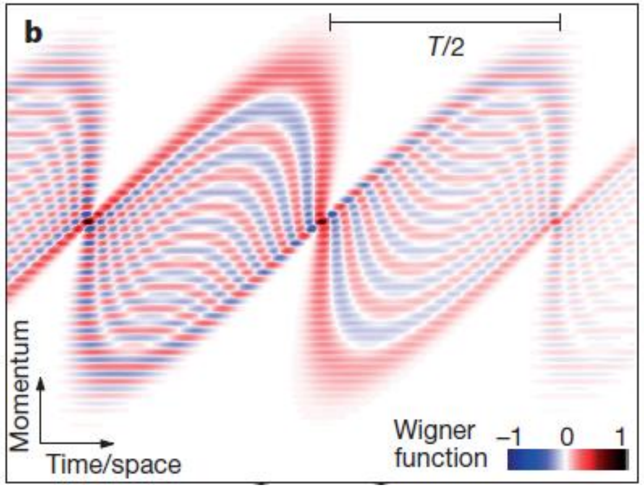
- Single interaction: phase modulation along the pulse
- quantum coherent control of free electron momentum states

K. E. Echternkamp *et al.*, Nature Phys. (2016)

Attosecond bunching

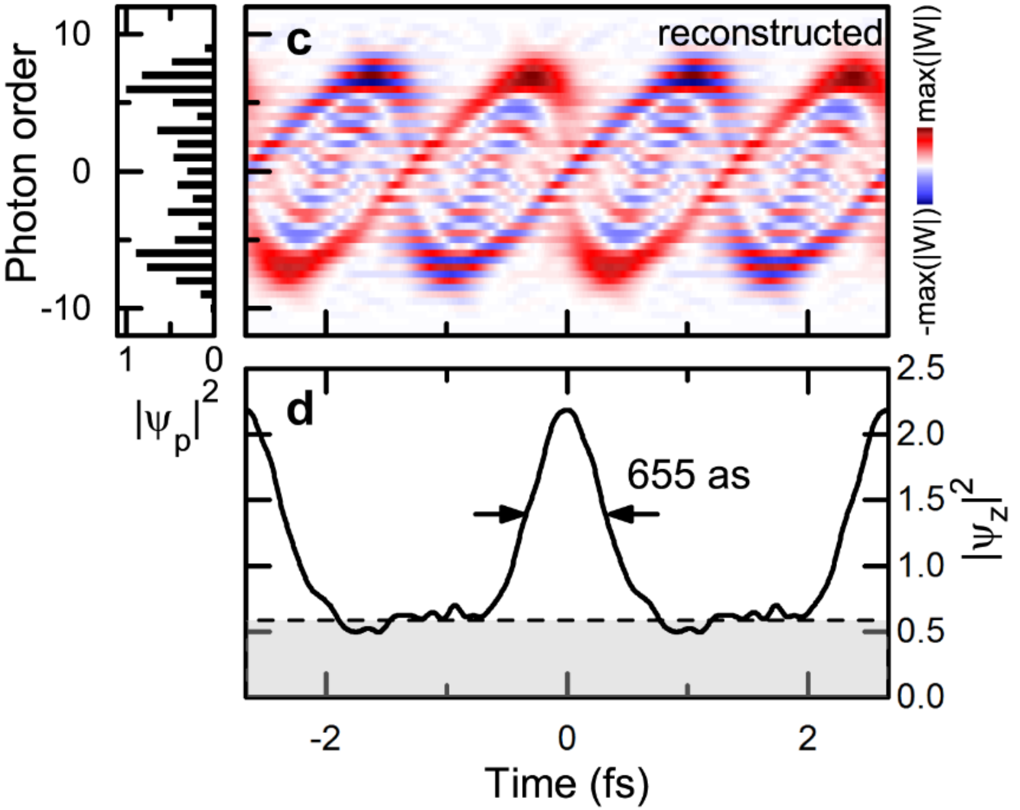






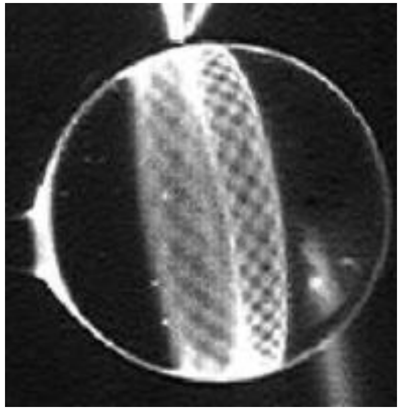
simulated

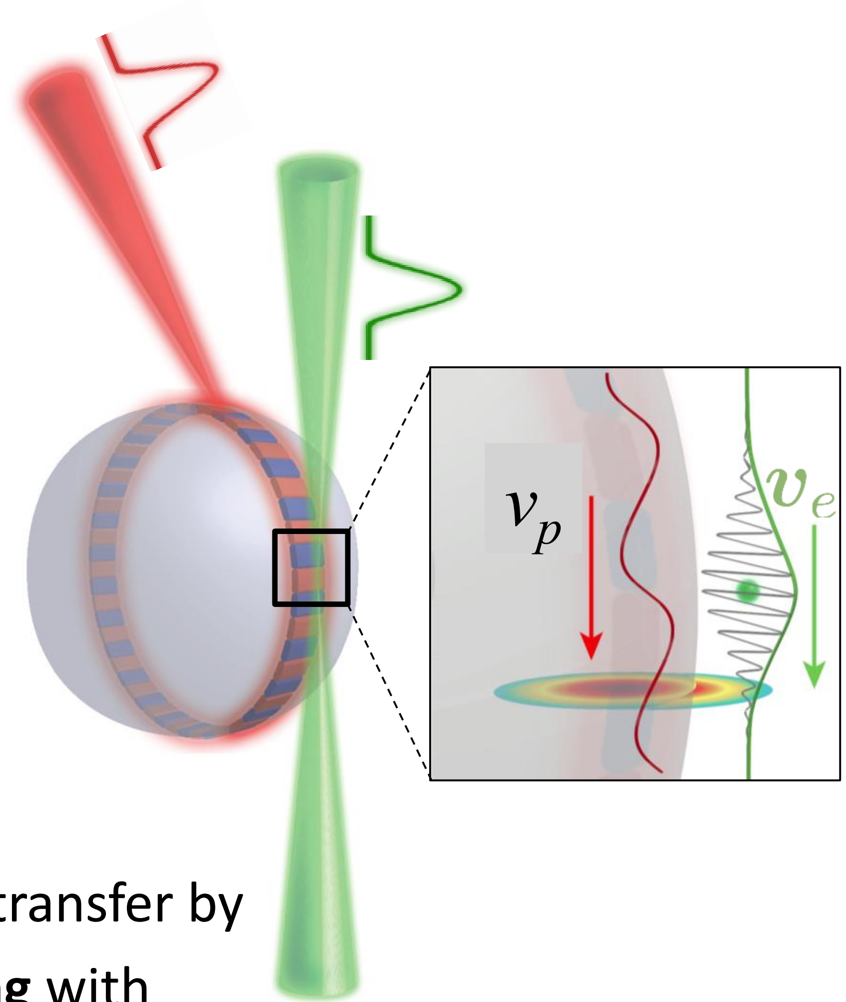
Wigner function reconstruction



K. E. Priebe *et al.*, Nat. Phot. (2017)

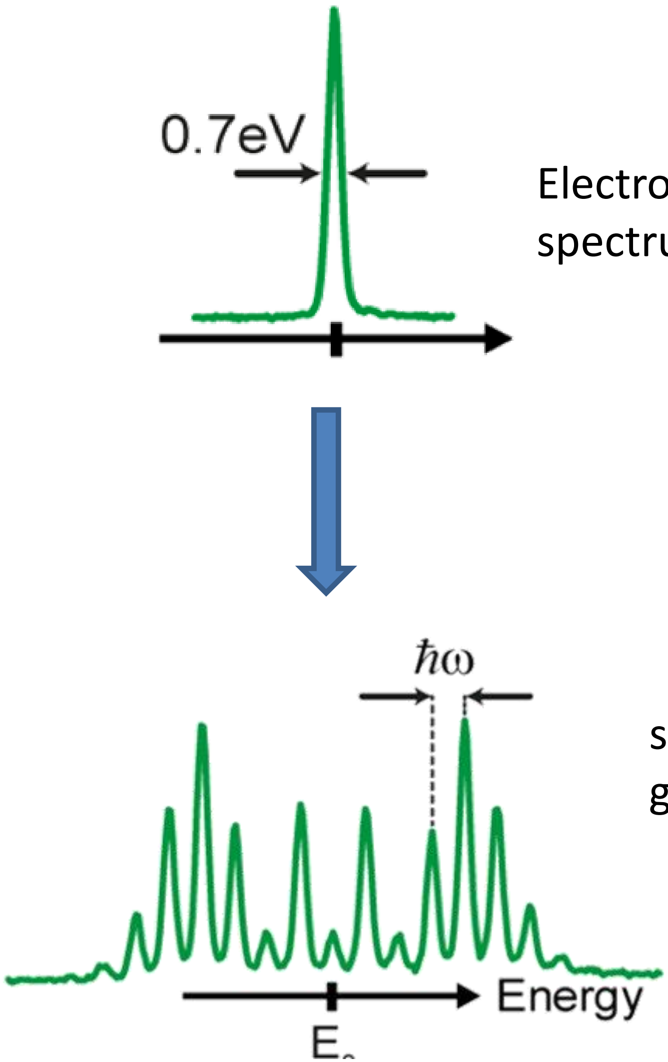
Phase-matched interaction in whispering gallery mode resonators





efficient energy transfer by **velocity matching** with **whispering-gallery modes (WGM)**

O. Kfir *et al.*, *Nature* (2020).

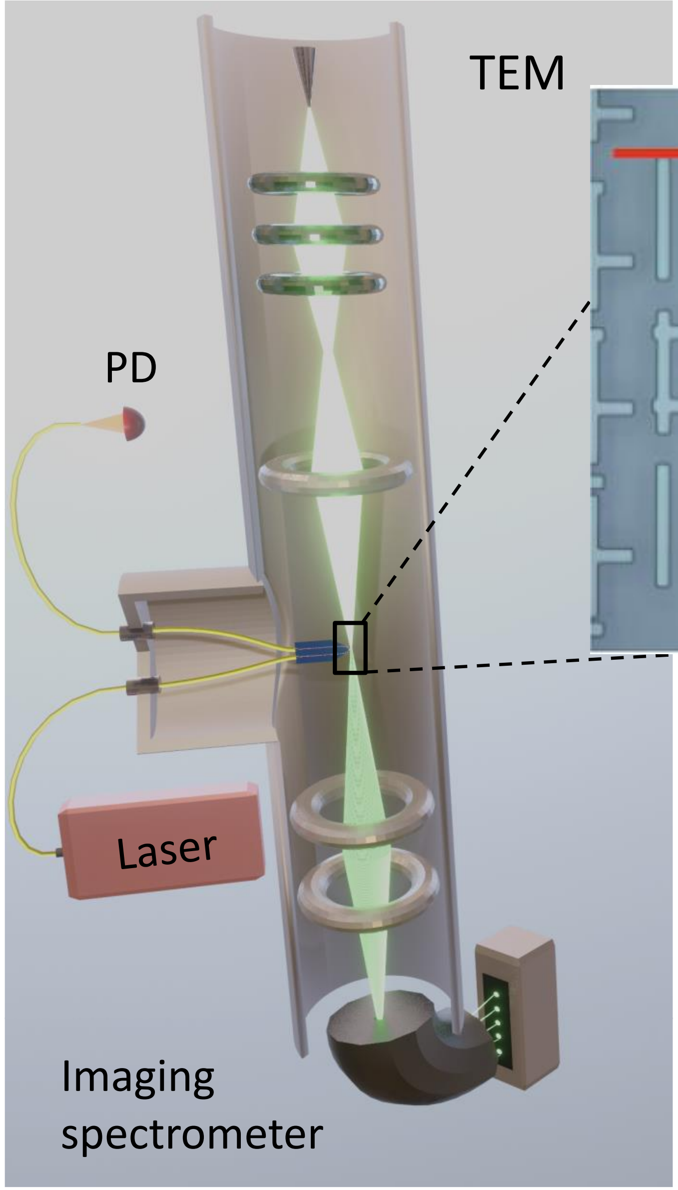


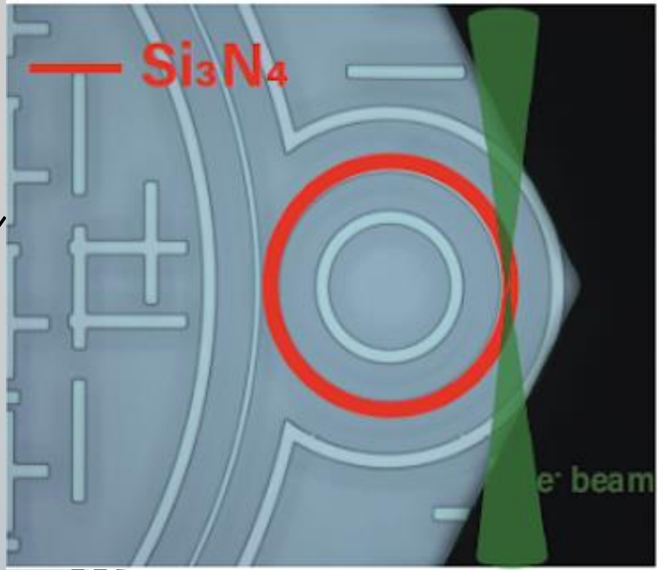
Electron spectrum

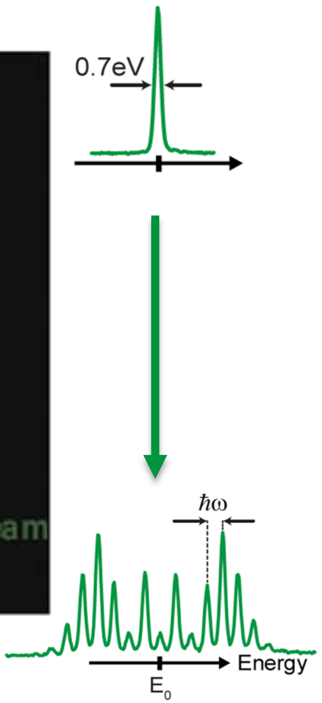
sideband generation

- Temporal structuring
- Ultrafast beam modulation

Electron-light interaction at high-Q resonators

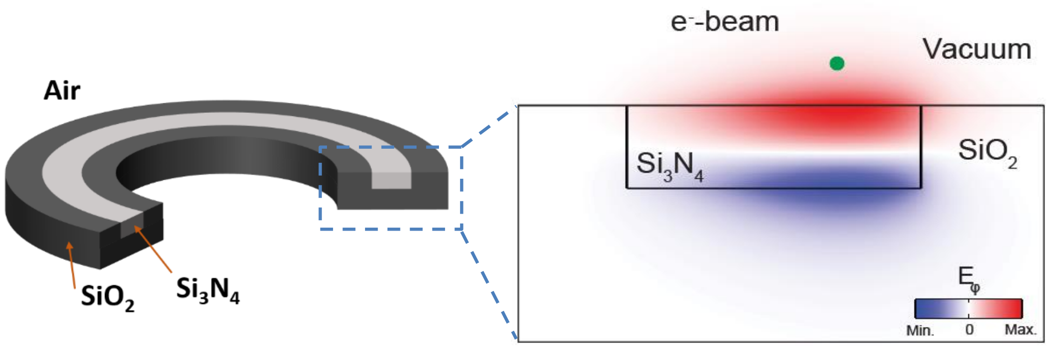






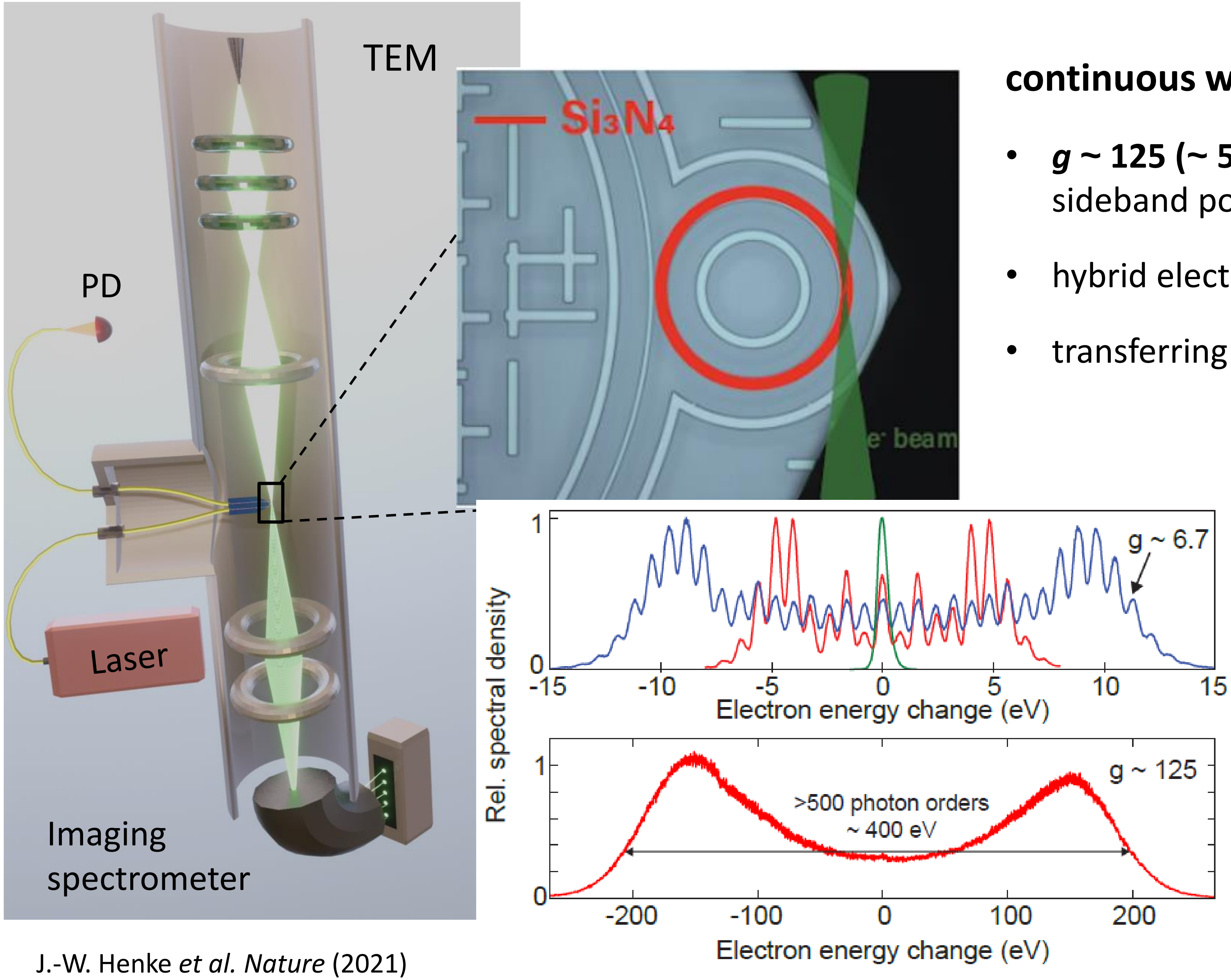
Chip-based high-Q Microresonators

- Resonators designed & fabricated by Kippenberg group at EPFL
- Air-cladded Si_3N_4 microresonator in SiO_2
- Fiber coupled
- high Q-factor > 6 * 10^5



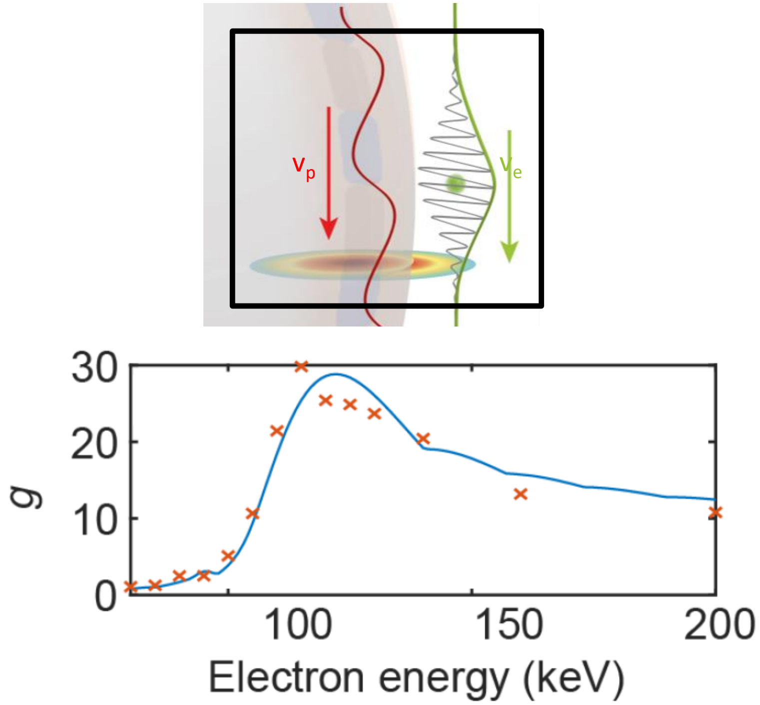
J.-W. Henke *et al.* *Nature* **600,** 653–658 (2021)

Electron-light interaction at high-Q resonators



continuous wave laser tuned to resonance

- ***g* ~ 125 (~ 500 photon sidebands)** for P_{bus} = 38 mW sideband population:
- hybrid electron-photon quantum technology
- transferring concepts to state-of-the-art TEMs



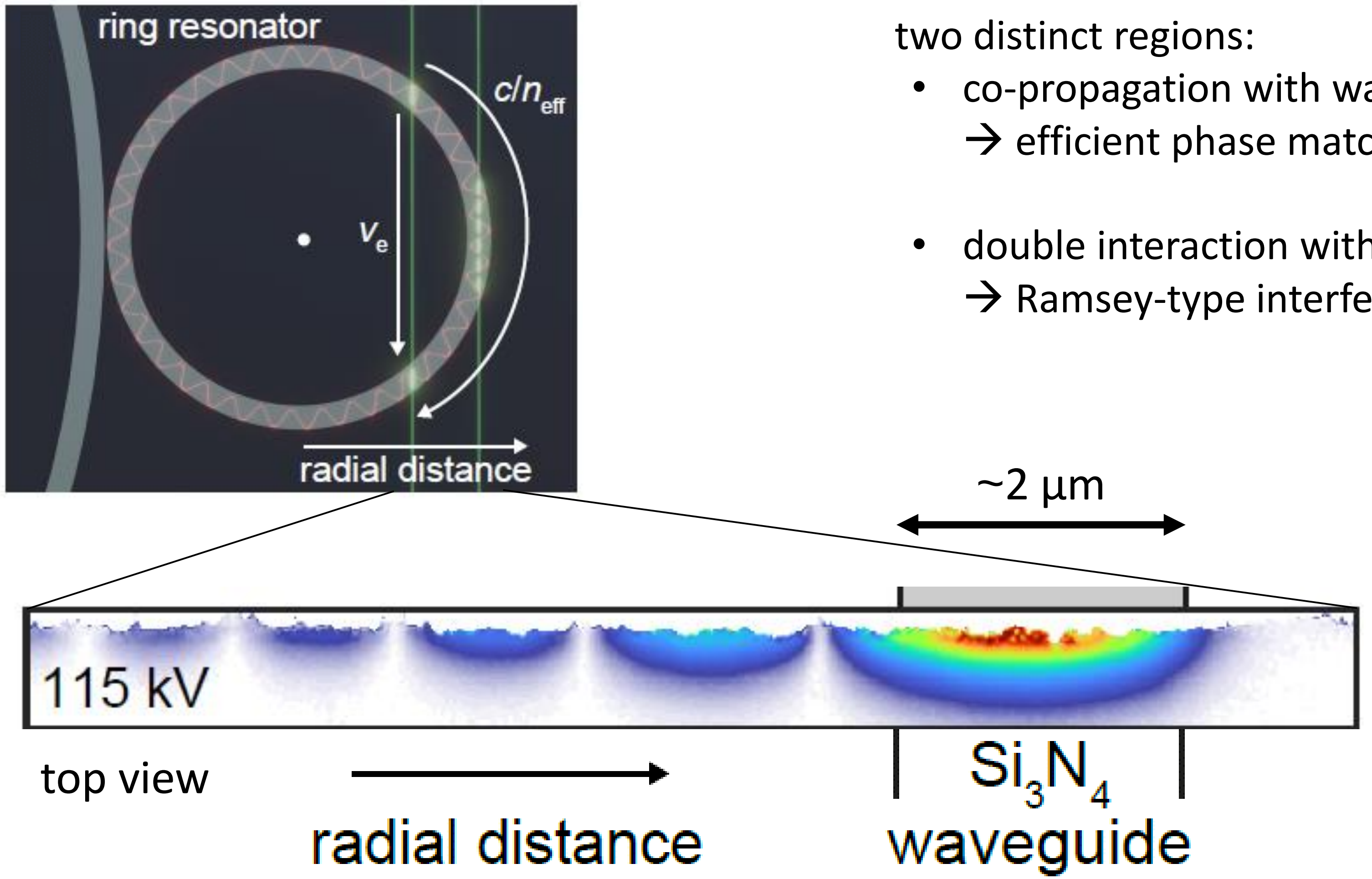
J.-W. Henke *et al. Nature* (2021)

quantitative PINEM of larger area

two distinct regions:

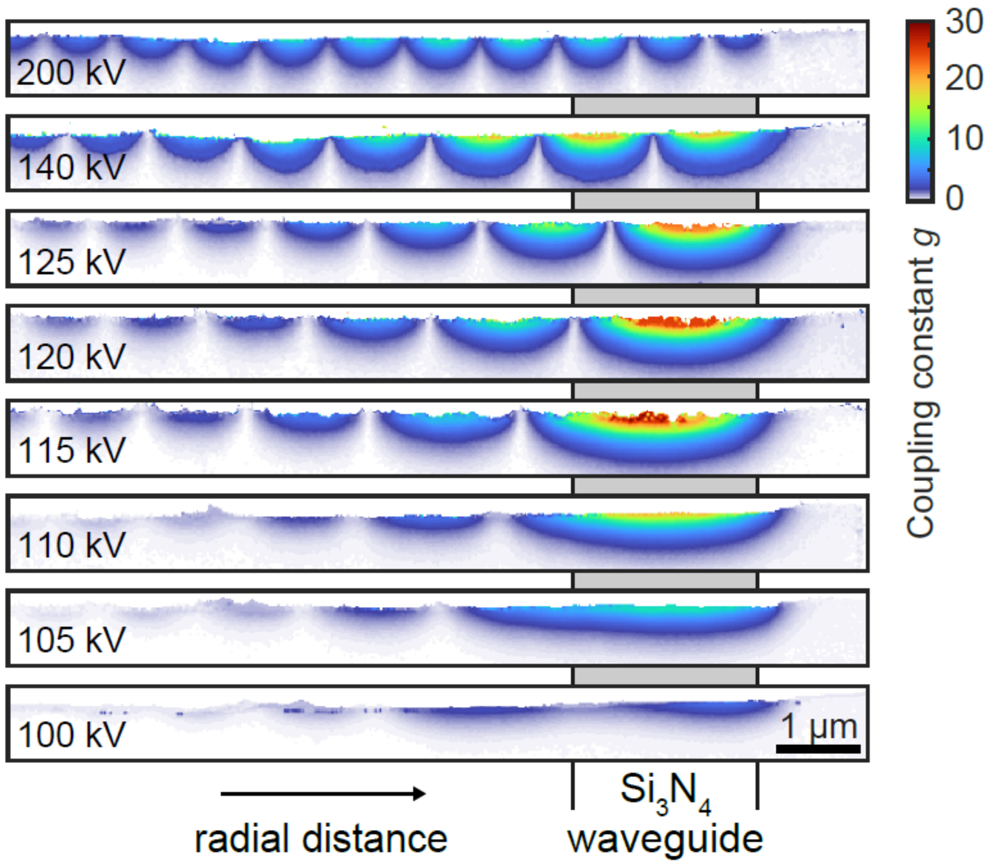
- co-propagation with waveguide
  → efficient phase matching
- double interaction with resonator mode
  → Ramsey-type interference & spatial interference pattern

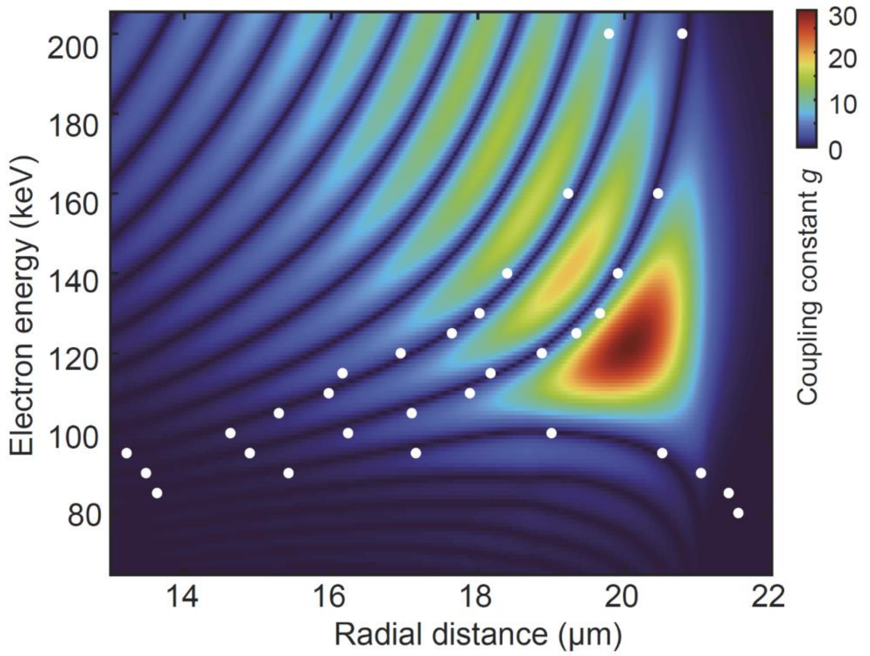
side view

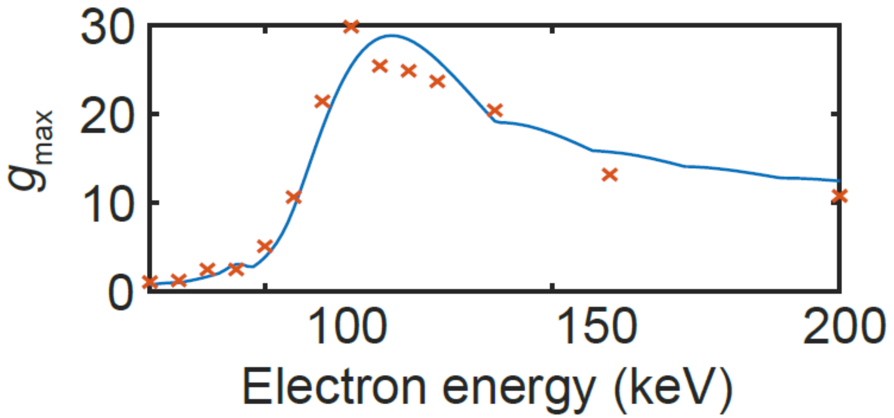


J.-W. Henke, AF *et al., Nature* **600**, 653–658 (2021).

Electron energy dependent phase matching



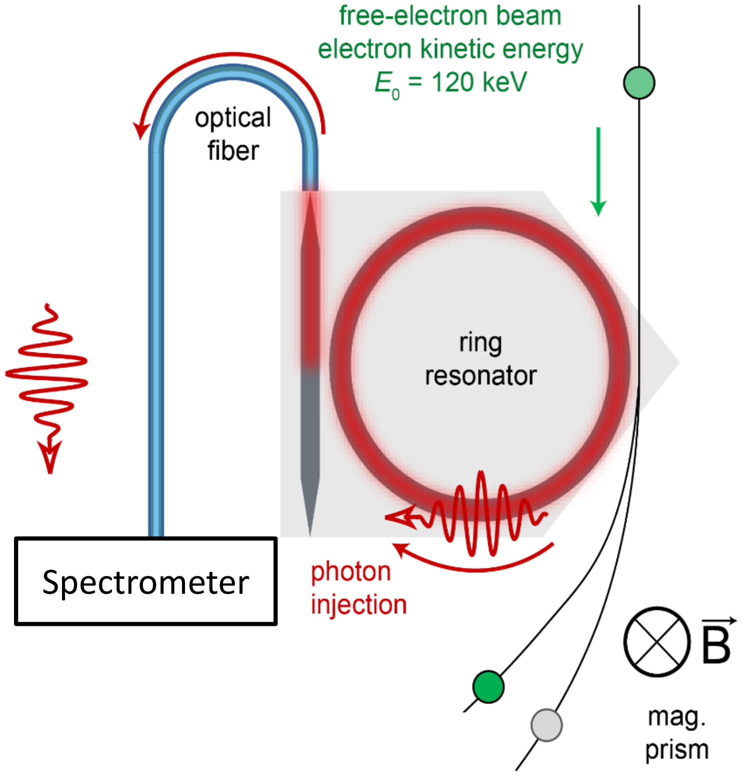


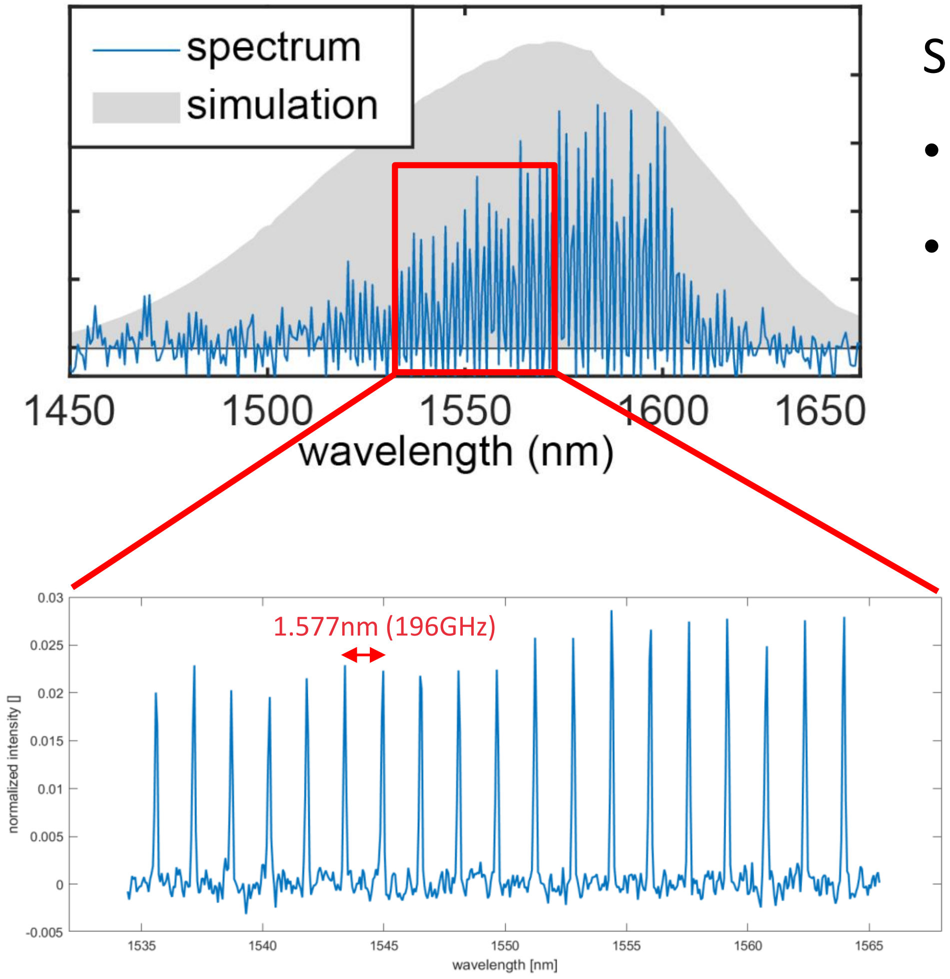


- PINEM for varying the electron energy between 80-200 keV
- visualization of velocity phase matching
- good agreement with numerical simulations

J.-W. Henke, AF *et al., Nature* **600**, 653–658 (2021).

Coupling to an empty cavity: Spontaneous photon generation





Free spectral range: 194 GHz (design)

Spectrum limited by

- detector
- coupling of resonator

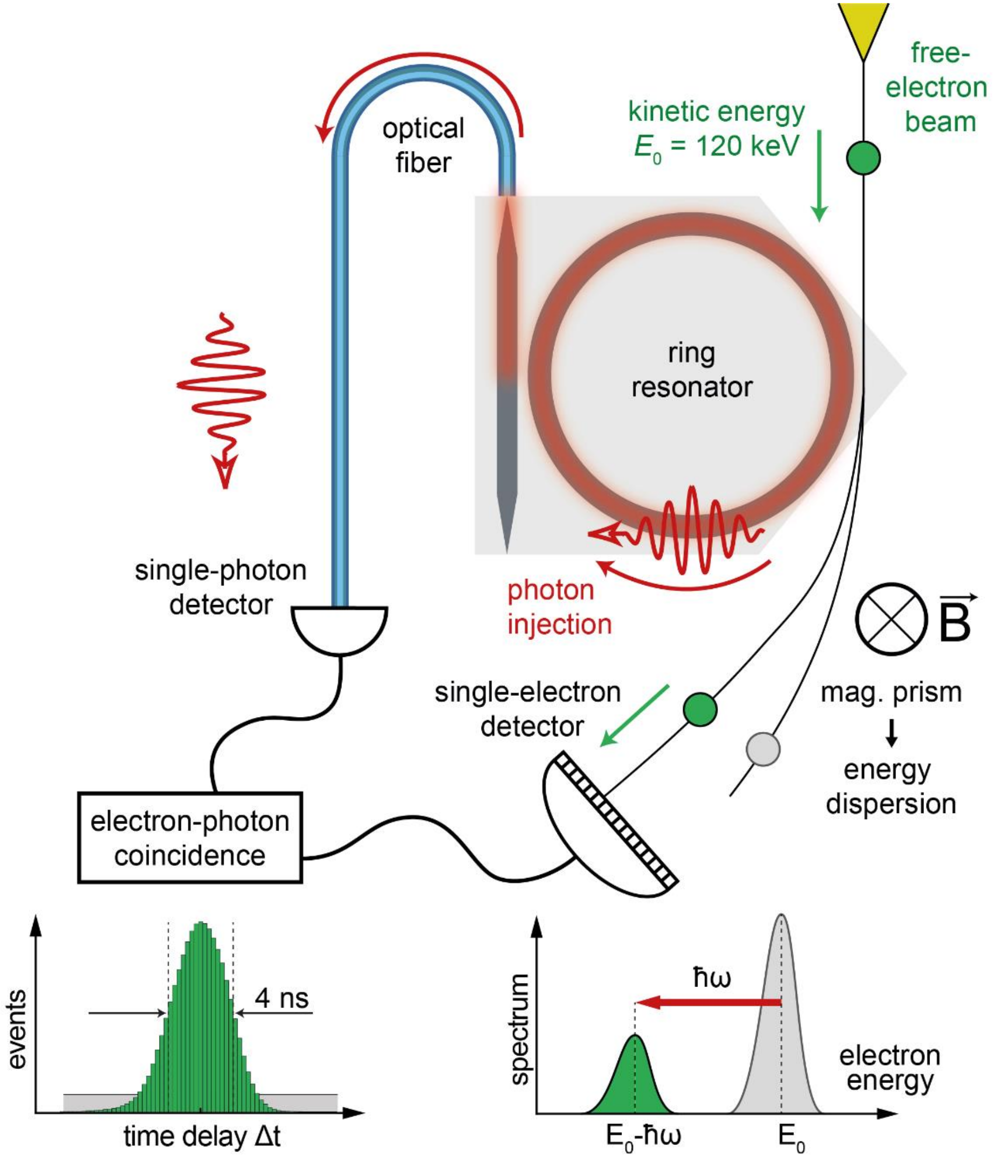
A. Feist *et al.*, Science (2022)

Coincidence measurements:

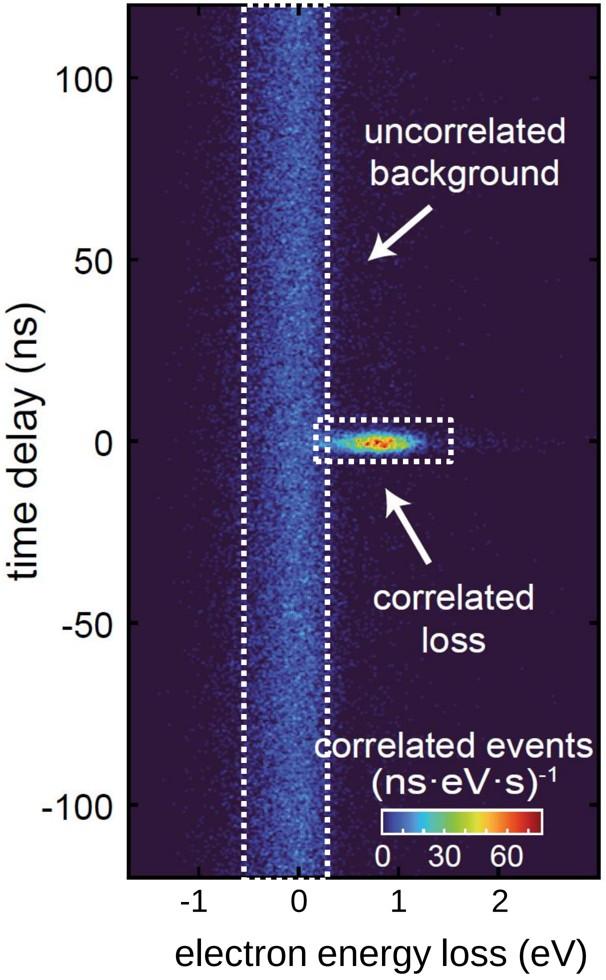
see also D. Jannis et al. , Appl. Sci. (2021)

D. Varkentina et al., Sci Adv. (2022)

Electron-photon pair state



A. Feist *et al.*, Science (2022)



two distinct features

- uncorrelated background

  $|E_0, 0\rangle$ Initial state

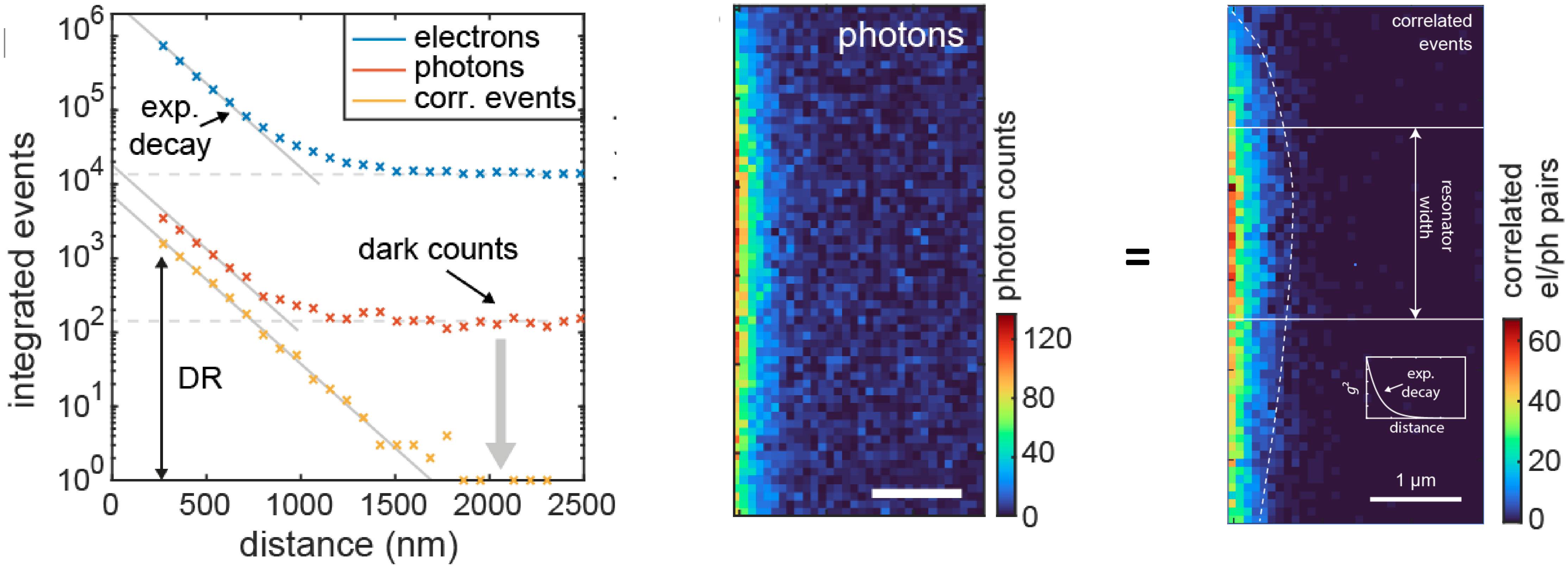
- photon correlated electron energy loss peak

  $|E_0 - \hbar\omega, 1\rangle$ Single photon generation

→ **unique identification of correlated electron-photon pairs**

- **Correlation-enhanced imaging and spectroscopy**
- **Heralded Single Photon Generation**
- **Strong indication for entanglement**

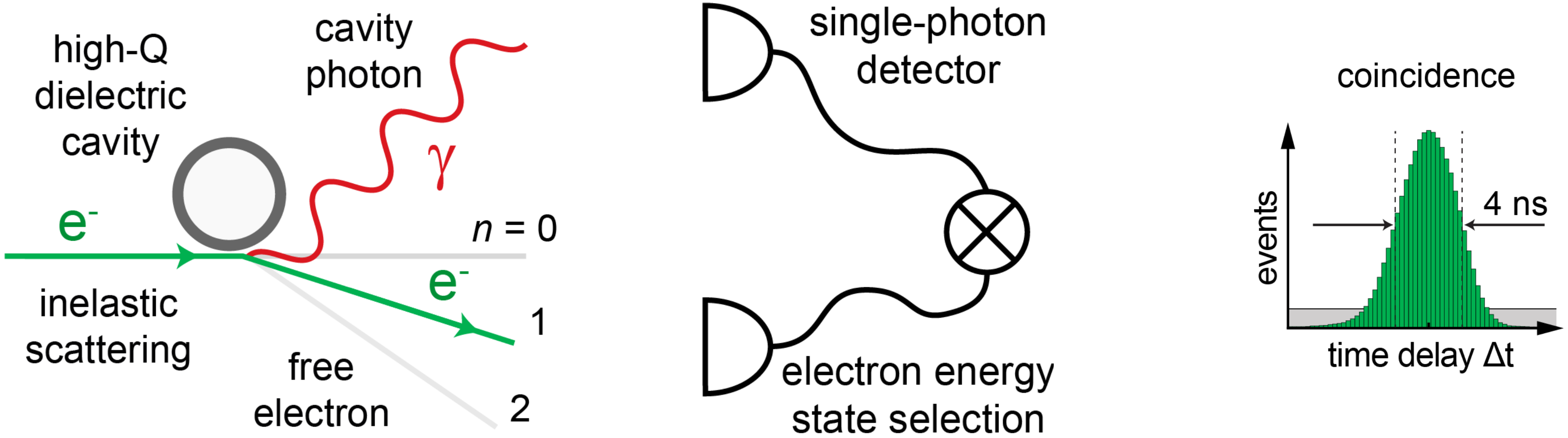
Contrast enhancement by coincidence gating



- imaging the resonator mode with loss scattered electrons, generated photons and correlated events

→ **two orders of magnitude contrast enhancement by coincidence gating**

Electron-photon pair-state preparation



initial state	entangled state	projection	pair state
$\vert E_0\rangle \vert 0\rangle_\omega$	$\sum_n c_n \vert E_0 - n\hbar\omega\rangle \vert n\rangle_\omega$	$\vert E\rangle\langle E\vert$	$\vert E_0 - \hbar\omega\rangle \vert 1\rangle_\omega$

A. Feist *et al.*, Science (2022)

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DFG Deutsche Forschungsgemeinschaft

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Photonic Resonators: T. Kippenberg (EPFL)

Thank you for your attention!