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**Project Title:** Q-SORT. The Quantum Sorter: A New Measurement Paradigm in Electron Microscopy

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## Interdisciplinary Training Webinar (release 7) D3.11

Version 1.0

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**Authors:**

Raffaella Santucci (QED)  
Luca Marco Carlo Giberti (QED)  
Vincenzo Grillo (CNR)

**Reviewers:**

Enzo Rotunno (CNR)  
Stefano Frabboni (UM)

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Dissemination Level		
P	Public	X
C	Confidential, only for members of the consortium and the Commission Services	

## Revision History

Revision	Date	Author	Organisation	Description
0.1	16/01/2020	Raffaella Santucci Luca Marco Carlo Giberti	QED QED	Outline of the document
0.2	23/01/2020	Raffaella Santucci	QED	First draft
0.3	06/02/2020	Vincenzo Grillo	MPI	Comments
0.4	13/02/2020	Vincenzo Grillo Raffaella Santucci	CNR QED	First complete version
0.4.5	12/03/2020	Luca Marco Carlo Giberti	QED	Second complete version - overall cleanup
0.5	16/03/2020	Enzo Rotunno, Stefano Frabboni	CNR UMR	Review and comments
1.0	30/03/2020	Luca Marco Carlo Giberti Raffaella Santucci	QED QED	Final version integrating the comments of the peer reviewers and partners

### Statement of originality:

This deliverable contains original unpublished work except where clearly indicated otherwise. Acknowledgement of previously published material and of the work of others has been made through appropriate citation, quotation or both.

### Disclaimer

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## EXECUTIVE SUMMARY

This deliverable describes the “*Quantum aspects of the interaction between beam electrons and optical near fields*” webinar. This is the seventh in the planned series of 9 Interdisciplinary Training Webinars for the Q-SORT project.

The Interdisciplinary Training Webinars are part of Work Package 3 **Theory, optical testing, and TEM implementation of generalised Sorter** [Months: 1-41], Task 3.5 - Interdisciplinary Training Webinars (M6, M11, M12, M18, M24, M26, M30, M36, M41).

This document is comprised of four Chapters, including Introduction and Conclusions.

The Introduction describes what a webinar is.

Chapter 2 summarises what we mean by Interdisciplinary Training Webinars and what their function is within Q-SORT.

Chapter 3 illustrates context, aims, and content of the seventh Interdisciplinary Training Webinar of Q-SORT.

The conclusions outline future plans for the Interdisciplinary Training Webinars.

## 1 INTRODUCTION - WHAT IS A WEBINAR?

A webinar is a web-based seminar which can be streamed either live or on demand. It might be just a simple audio stream, or it might include visual aids, such as PowerPoint slides, recorded video clips, or live software demonstrations.

The experience of a webinar attempts to reproduce the benefits of attending a seminar. Audience members can ask questions and the speaker can survey or poll the audience and get feedback as he or she delivers the information.

Webinar technology providers need to support these interactive elements in addition to the basic delivery of the audio and video streams. Keyboard chat features and Q&A are usually subject to more control, whereby the presenters can see messages from the audience and choose whether to broadcast them to all participants, ignore them, or reply privately.

The Q-SORT Interdisciplinary Webinars make use of Facebook Live as a platform for live streaming. We chose this platform as it provides the best value for money and offers the added advantage of being very widespread. Users are able to pose questions by posting comments below the video. Answers can be provided by specialists during or after the webinar.

## 2 THE Q-SORT INTERDISCIPLINARY TRAINING WEBINARS

The Q-SORT project is intrinsically interdisciplinary, being born from the cross-fertilisation of three subject areas: light optics (UG, MPI), electron microscopy (CNR, FZJ, UMR, FEI), and biology (MU).

The silo-breaking nature of Q-SORT provides added value through the following cross-fertilisation dynamics: microscopists borrow techniques from light optics specialists and in turn explain the challenges of the new measurements; biologists explain to physicists the problems that usually arise when studying proteins and suggest alternative proteins for investigation; physicists develop the quantum mechanical approach to cryo-TEM; biologists communicate a priori knowledge to light optics theoreticians in order to streamline efforts in the project work-packages (WP3-WP5).

In order to facilitate mutual learning between physicists and biologists, as well as to foster possible further applications to biology and the life sciences, 9 Q-SORT Interdisciplinary Training Webinars have been planned during the lifetime of the project. Their main aim is to function as aids to facilitate the highly interdisciplinary work in WP3 and in the other research WPs. They will do so by providing opportunities for training and mutual learning mentored by senior scientists, for forging a common language, for exchanging ideas for further joint research.

However important their role within Q-SORT, these seminars are meant to be accessible to everyone, hence the choice to propose them as webinars, so as to reach as wide an audience as possible.

The Interdisciplinary Training Webinars introduce a sort of 'intermediate level' in the communication of science: i.e. their nature is intermediate between the specialist-to-specialist character of discipline-specific communication tools -such as colloquia, talks, papers- and the broad appeal of conventional public understanding of science.

This is why, besides their facilitating function within WP3 and the other research WPs, the Interdisciplinary Training Webinars also play an important role in the dissemination and engagement process. Together with the Website, they ensure that Q-SORT will reach a broad range of pertinent audiences across Europe and beyond.

In the following paragraph we describe the seventh of the 9 Interdisciplinary Training Webinars planned for Q-SORT.

### 3 THE Q-SORT SEVENTH INTERDISCIPLINARY TRAINING WEBINAR: QUANTUM ASPECTS OF THE INTERACTION BETWEEN BEAM ELECTRONS AND OPTICAL NEAR FIELDS

Javier García de Abajo, ICFO-Institut de Ciències Fotoniques (Spain)

03 July 2019, h 11:15-12:15 AM CET

Facebook Live: <https://www.facebook.com/quantumsorter/>

<http://www.qsort.eu/interdisciplinary-training-webinar-7/>

The “*Quantum aspects of the interaction between beam electrons and optical near fields*” *Interdisciplinary Training Webinar* was recorded and webcast on 03 July 2019, during the second [International Conference on Quantum Imaging and Beam Shaping](#) at Max Planck Institute for the Science of Light. The conference was organised by Q-SORT and the “[Science and Applications of Electron Wavefunctions Shaped and Manipulated by Engineered Nanoholograms](#)” DIP project (*Deutsch-Israelische Projekt cooperation grant*) with the support of the Max Planck Institute for the Science of Light and the [Nanoscience Institute of CNR \(CNR-NANO\)](#).

The webinar featured one of the conference [invited speakers](#), **Javier García de Abajo, ICFO-Institut de Ciències Fotoniques (Spain)**.

The webinar totaled 12K views and 212 engagements (post clicks, likes, comments, shares, etc.). Facebook posts related to this webinar reached over 39K people.

The webinar was recorded with two cameras -one for the speaker, the other for the screen and the laser pointer-, then stored on remote servers for subsequent on-demand viewing.

The content of the Webinar was agreed upon by the Project Coordinator, Dr. Vincenzo Grillo (CNR) with Prof. Garcia de Abajo, Dr. Luca Marco Carlo Giberti (QED) and Dr. Raffaella Santucci (QED).

The webinar can be re-watched on demand - interested parties can re-play it at their convenience by clicking on the following links:

on the Q-SORT website:

<http://www.qsort.eu/interdisciplinary-training-webinar-7/>

on Facebook:

<https://www.facebook.com/1223951817738187/videos/360860904622526/>

### 3.1 TECHNICAL DESCRIPTION

The seventh Q-SORT Interdisciplinary Training Webinar touches upon the following topics:

- Ultrafast TEM;
- PINEM;
- Beam shaping;
- Plasmons

#### **Quantum aspects of the interaction between beam electrons and optical near fields**

**F. Javier García de Abajo,<sup>1,2</sup> Valerio Di Giulio,<sup>1</sup>  
and Vahagn Mkhitarian<sup>1</sup>**

<sup>1</sup>*Institut de Ciències Fotoniques, The Barcelona Institute of Science and Technology, 08860 Castelldefels (Barcelona), Spain*

<sup>2</sup>*Institució Catalana de Recerca i Estudis Avançats, Passeig Lluís Companys, 23, 08010 Barcelona, Spain  
[javier.garciadeabajo@nanophotonics.es](mailto:javier.garciadeabajo@nanophotonics.es)*

Electron beams are ideal tools to controllably excite and probe plasmons and other nanoscale optical excitations with an unparalleled combination of space and energy resolutions. Spectroscopy performed through the analysis of electron energy loss and cathodoluminescence are widely used to obtain snapshots of these excitations. Additionally, access to the ultrafast sample dynamics is possible by recording photoelectrons excited with femtosecond light pulses, while several experiments demonstrate optical pumping followed by electron-beam probing with similar temporal resolution. In this talk, we will review recent advances in these techniques and present a unified theoretical description, along with several potential directions for improving the space-time-energy resolution and accessing quantum aspects of the samples.

As a first challenge, we will discuss fundamental limits to the coupling between electrons and optical excitations based on suitably tailored beam-electron wave functions, thus opening new directions for further increase in time resolution and the exploration of nonlinear phenomena with nanometer resolution.

Additionally, we will discuss recent theoretical results on fundamental aspects of the interaction of fast electrons with localized optical modes that are made possible by the noted advances. Using a quantum-optics description of the optical field, we predict that the resulting electron spectra strongly depend on the statistics of the sample excitations (bosonic or fermionic) and their population (Fock, coherent, or thermal), whose autocorrelation functions are directly retrieved from the ratios of electron gain intensities. We further explore feasible experimental scenarios to probe the quantum characteristics of the sampled excitations and their populations.

This webinar is aimed at:

- spreading awareness amongst physicists of the new possibilities afforded by the following new electron microscopy schemes:
  - time-resolved electron microscopy
  - pump-and-probe
  - beam-shaping,

with special emphasis placed on:

- electron-light interactions mediated by plasmons
- joint of control of the quantum states of electron and photon probes;
- exchanging ideas between scientists for furthering joint research.

### 3.2 THE SEVENTH INTERDISCIPLINARY TRAINING WEBINAR ON THE Q-SORT WEBSITE

A dedicated webpage for this webinar has been created on the Q-SORT website featuring:

1. the webinar;
2. a short description of the webinar thought for the general public;
3. a short bio of the speaker who offered the talk.

The link to the webpage is:

<http://www.qsort.eu/interdisciplinary-training-webinar-7/>

An overview and a screenshot of the webpage are provided below.

#### Quantum aspects of the interaction between beam electrons and optical near fields

Javier García de Abajo, ICFO-Institut de Ciències Fotoniques (Spain)

03 July 2019, h 11:15 -12:15 AM CET

[Facebook Live: https://www.facebook.com/quantumsorter/](https://www.facebook.com/quantumsorter/)

The seventh Q-SORT Interdisciplinary Training Webinar touches upon the following topics:

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### About the speaker



**Javier García de Abajo** received his PhD from the University of the Basque Country in 1993 and then visited Berkeley National Lab for three years. He was a Research Professor at the Spanish CSIC and in 2013 moved to ICFO-Institut de Ciències Fotoniques (Barcelona) as an ICREA Research Professor and Group Leader. He is Fellow of both the American Physical Society and the Optical Society of America. García de Abajo has co-authored 300+ articles cited 25,000+ times with a h index of 78 (March 2019 WoK data), including contributions on different aspects of surface science, nanophotonics, and electron microscope spectroscopies.

# INTERDISCIPLINARY TRAINING WEBINAR 7

Quantum aspects of the interaction between beam electrons and optical near fields

Javier García de Abajo, ICFO-Institut de Ciències Fòniques (Spain)

03 July 2019, h 11:15 -12:15 AM CET

Facebook Live: <https://www.facebook.com/quantumsorter/>

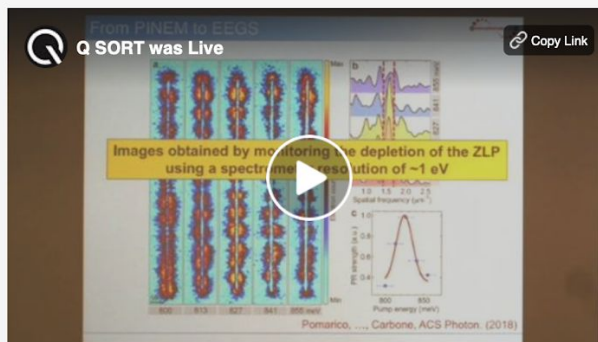


Figure 1 - Screenshot of the seventh interdisciplinary training webinar page on the Q-SORT website

## 4 CONCLUSIONS

This is the seventh Interdisciplinary Training Webinar produced for Q-SORT by [QED](#), with additional support from partner Max Planck Institute for the Science of Light.

In the past few years we have seen a consistent rise in the popularity and value of webinars as an essential training, communication, and engagement tool.

The video of the webinar also offers a unique opportunity for engagement. The project will continue to promote on-demand webinars so that their audiences will get larger and more engaged, and spend more time watching webinars, both live and on-demand. In planning the next webinars we will evaluate the effectiveness of the previous ones and use this information as a guideline to help the Q-SORT consortium create, promote, and deliver even more successful webinars.

**ANNEX 1: QUANTUM ASPECTS OF THE INTERACTION BETWEEN BEAM ELECTRONS AND OPTICAL NEAR FIELDS**

# Quantum aspects of the interaction between beam electrons and optical near fields

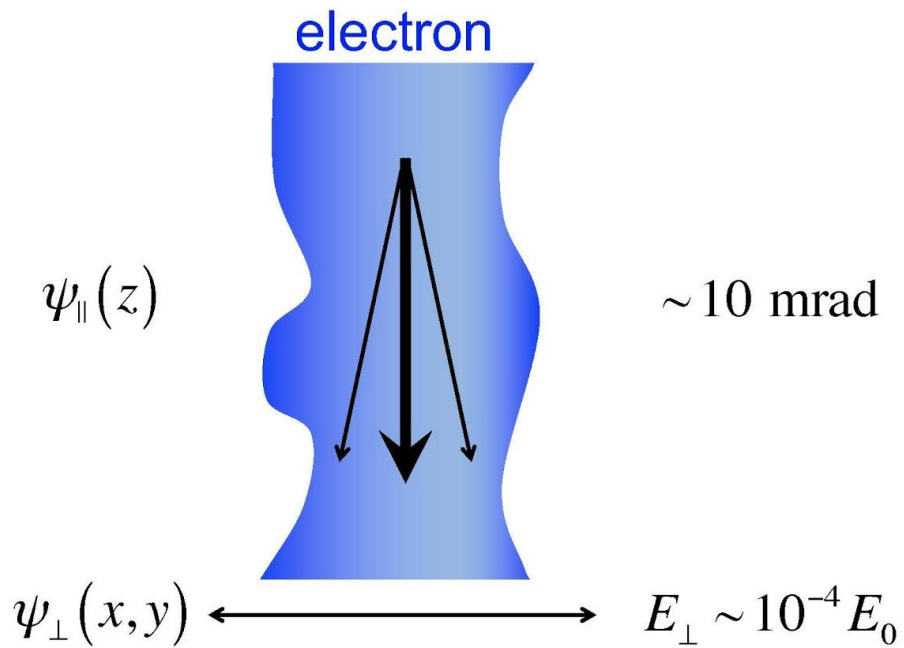
**Javier García de Abajo**

ICFO-The Institute of Photonic Sciences, Barcelona, Spain

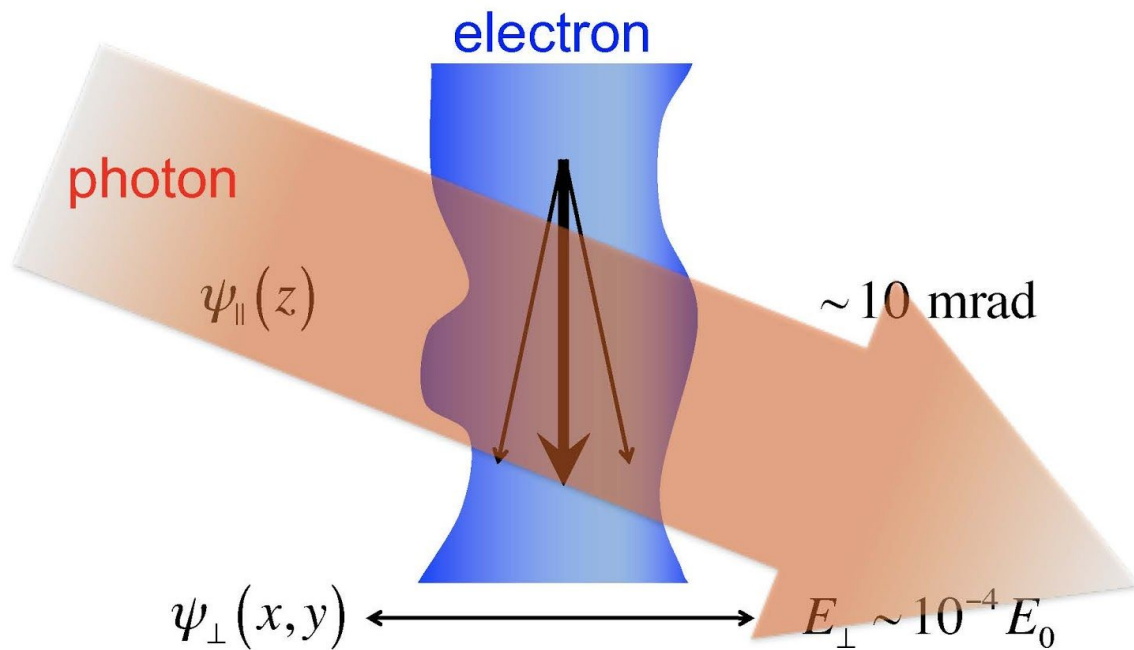


QSORT 2019 Conference – Erlangen  
July 3, 2019

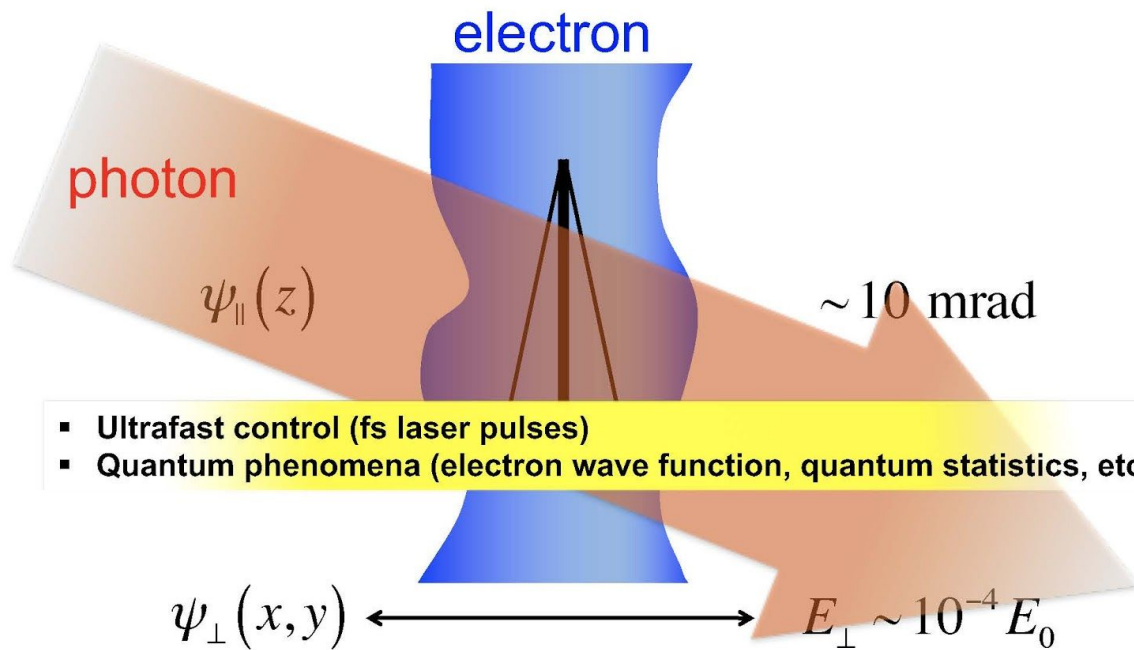
## Optical manipulation of electron beams



## Optical manipulation of electron beams



## Optical manipulation of electron beams



## Free-electron/photon interaction



photon

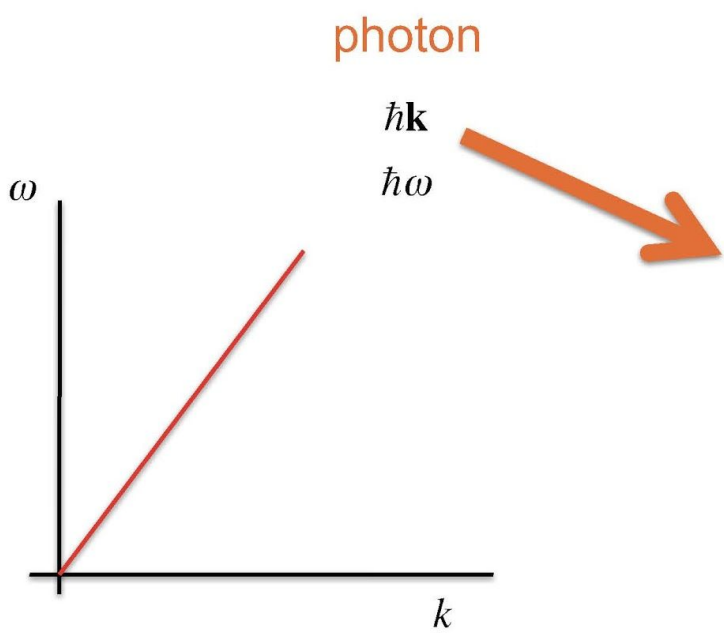
$$\hbar\mathbf{k}$$

$$\hbar\omega$$

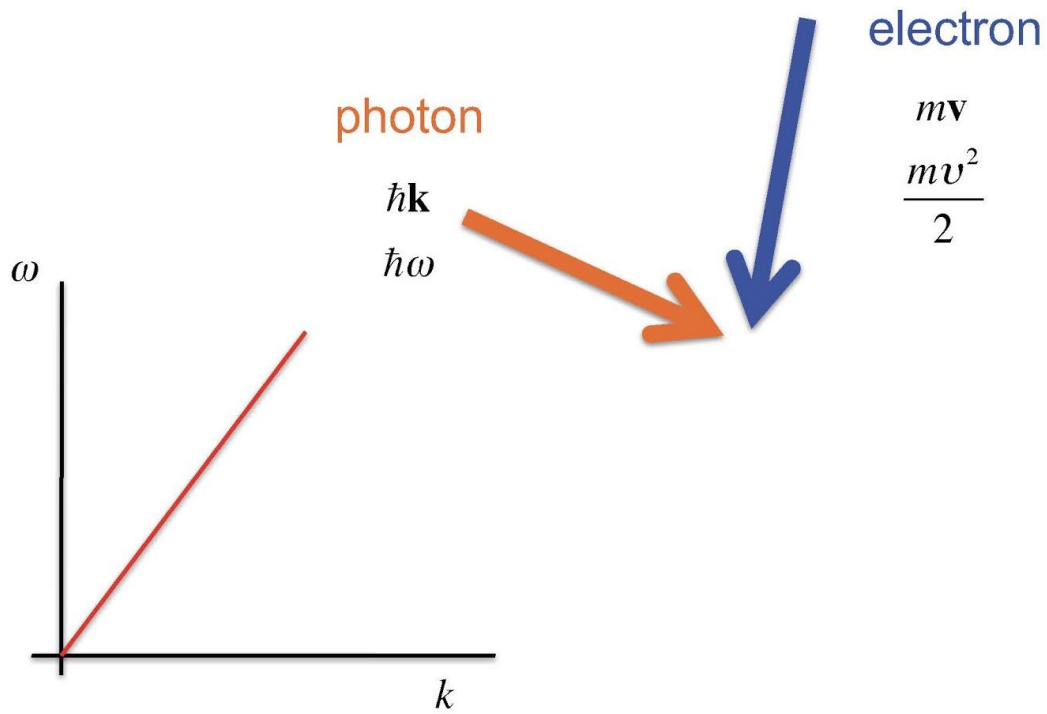




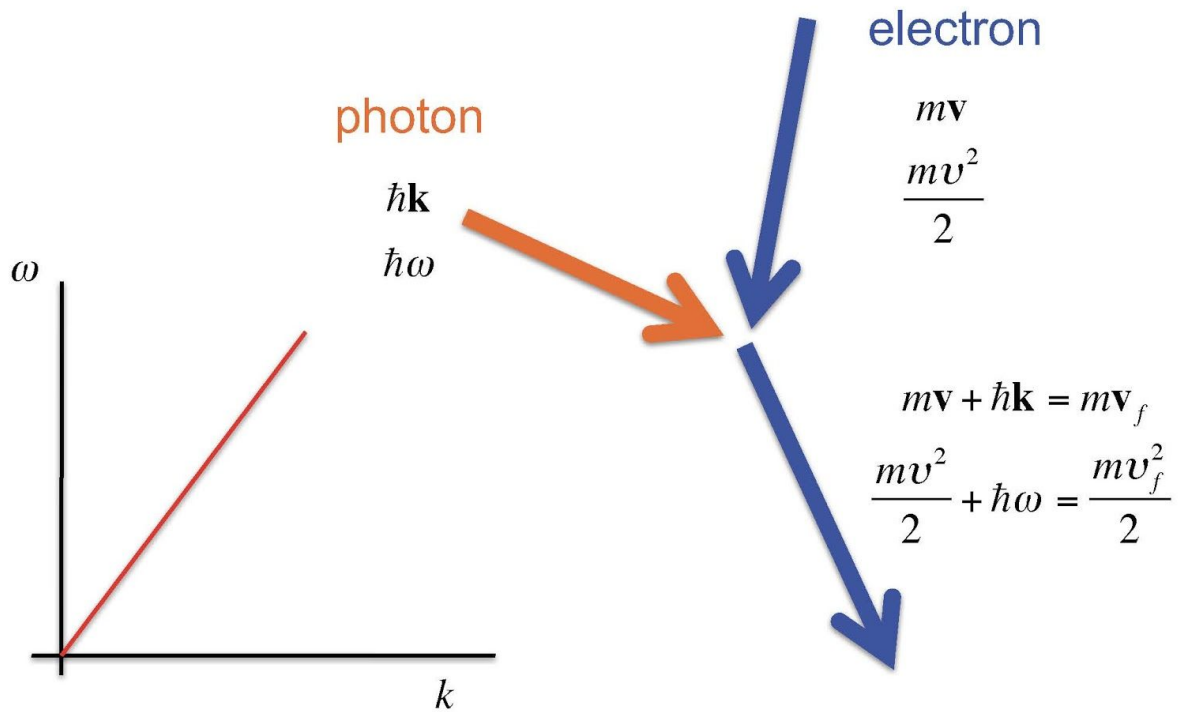
# Free-electron/photon interaction



## Free-electron/photon interaction



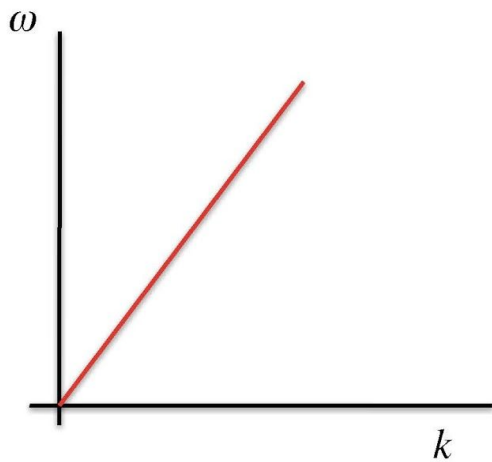
## Free-electron/photon interaction



## Free-electron/photon interaction



$$\omega = \frac{m}{2\hbar}(\mathbf{v}_f^2 - \mathbf{v}^2)$$



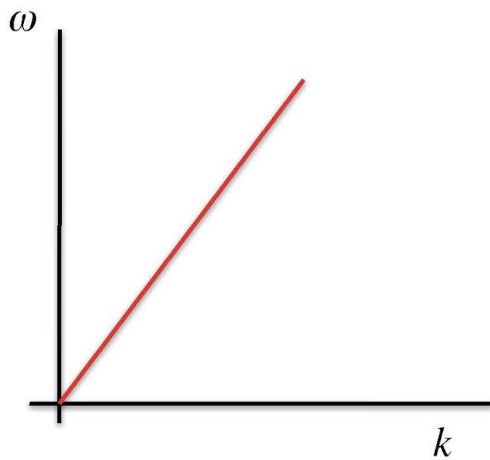
$$m\mathbf{v} + \hbar\mathbf{k} = m\mathbf{v}_f$$
$$\frac{mv^2}{2} + \hbar\omega = \frac{mv_f^2}{2}$$


## Free-electron/photon interaction



$$\omega = \frac{m}{2\hbar}(\mathbf{v}_f^2 - \mathbf{v}^2)$$

$$\omega = \mathbf{k} \cdot \mathbf{v} + \frac{\hbar k^2}{2m}$$




$$m\mathbf{v} + \hbar\mathbf{k} = m\mathbf{v}_f$$
$$\frac{m\mathbf{v}^2}{2} + \hbar\omega = \frac{m\mathbf{v}_f^2}{2}$$

## Free-electron/photon interaction

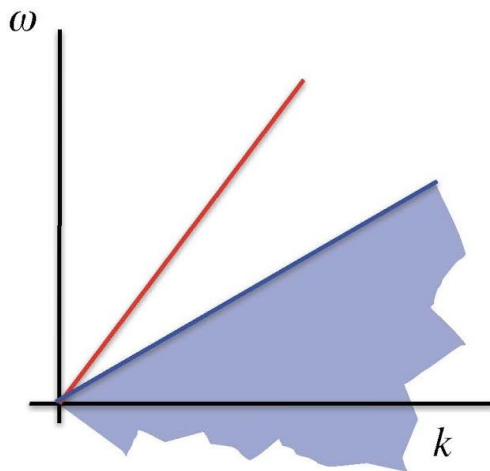


$$\omega = \frac{m}{2\hbar}(\mathbf{v}_f^2 - \mathbf{v}^2)$$

$$\omega = \mathbf{k} \cdot \mathbf{v} + \frac{\hbar k^2}{2m}$$

$$\omega \approx \mathbf{k} \cdot \mathbf{v}$$

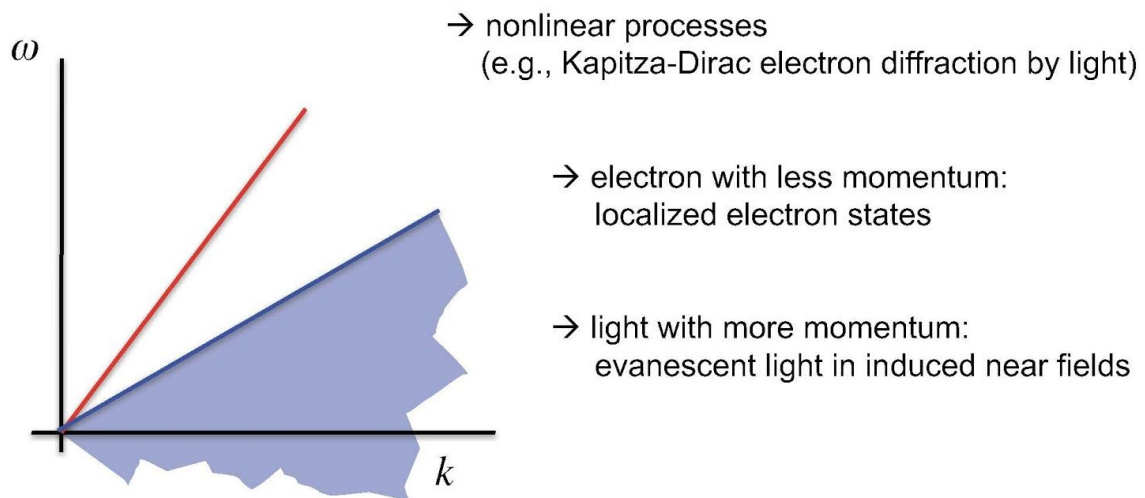
$$\hbar\omega \ll 1 \text{ MeV}$$



## Free-electron/photon interaction



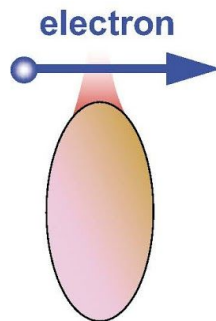
Free light and free electrons do not interact  
below gamma-ray energies unless ...



## Free-electron/optical-mode interaction



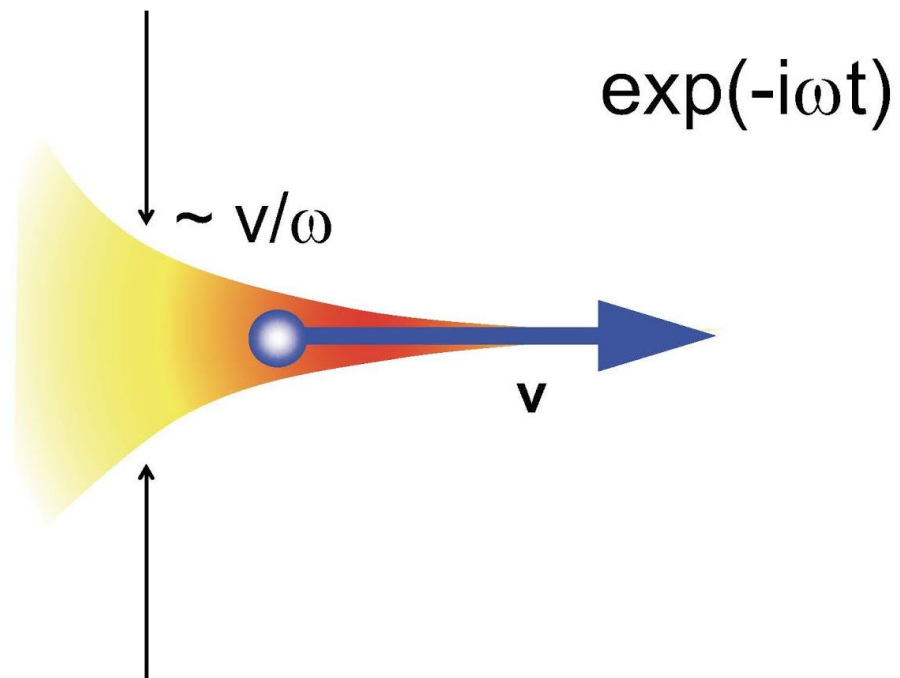
- **Cathodoluminescence (CL) – coherent light emission**



$$\mathbf{j}(\mathbf{r}, t) = -e \mathbf{v} \delta(\mathbf{r} - \mathbf{v}t)$$



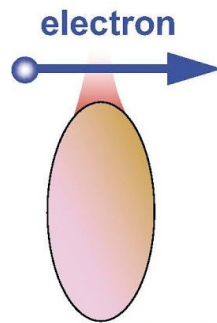
## EM field of a moving electron



## Free-electron/optical-mode interaction



- **Cathodoluminescence (CL) – coherent light emission**



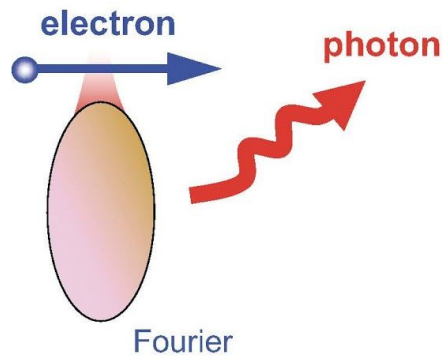
Fourier

$$\mathbf{j}(\mathbf{r}, t) = -e \mathbf{v} \delta(\mathbf{r} - \mathbf{v}t) \rightarrow \mathbf{j}(\mathbf{r}, \omega)$$

## Free-electron/optical-mode interaction



- Cathodoluminescence (CL) – coherent light emission



$$\mathbf{j}(\mathbf{r}, t) = -e \mathbf{v} \delta(\mathbf{r} - \mathbf{v}t) \rightarrow \mathbf{j}(\mathbf{r}, \omega)$$

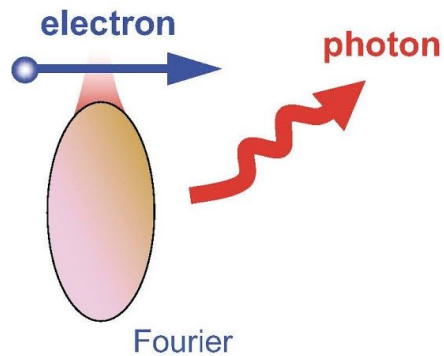
$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}, \omega) - (\omega/c)^2 \epsilon(\mathbf{r}, \omega) \mathbf{E}(\mathbf{r}, \omega) = \frac{4\pi i \omega}{c^2} \mathbf{j}(\mathbf{r}, \omega)$$

**Emission probability: integral of Poynting vector**

## Free-electron/optical-mode interaction



- Cathodoluminescence (CL) – coherent light emission

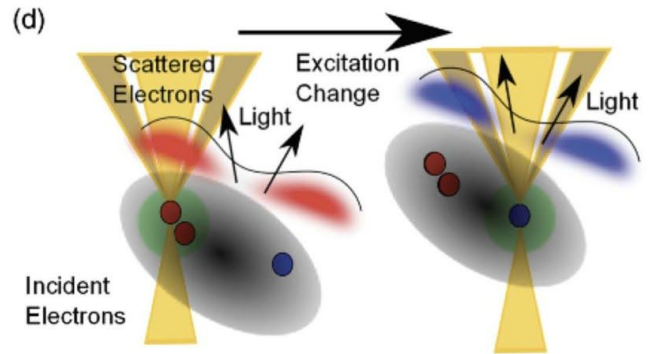
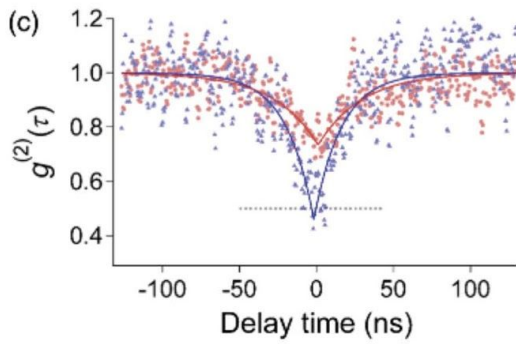
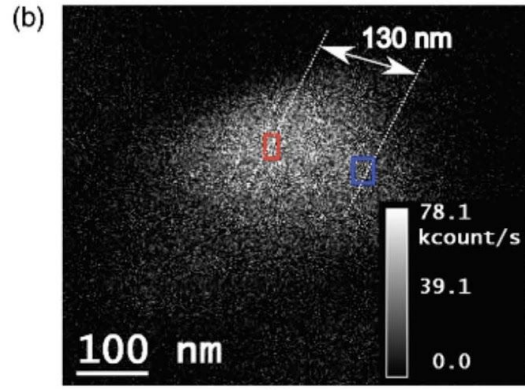
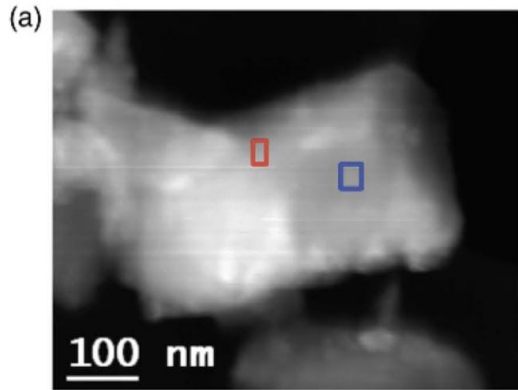


$$\mathbf{j}(\mathbf{r}, t) = -e \mathbf{v} \delta(\mathbf{r} - \mathbf{v}t) \rightarrow \mathbf{j}(\mathbf{r}, \omega)$$

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}, \omega) - (\omega/c)^2 \epsilon(\mathbf{r}, \omega) \mathbf{E}(\mathbf{r}, \omega) = \frac{4\pi i \omega}{c^2} \mathbf{j}(\mathbf{r}, \omega)$$

**Emission probability: integral of Poynting vector  
For a transparent sample: CL = EELS**

# Single-photon generation

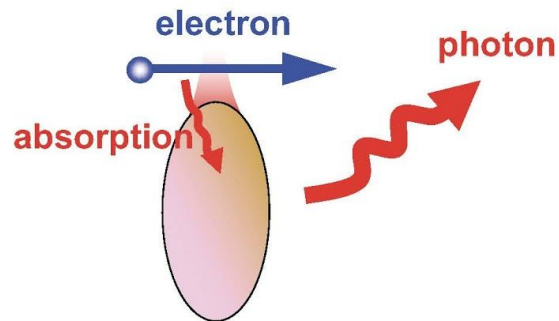


Tizei and Kociak, Phys. Rev. Lett. (2013)

## Free-electron/optical-mode interaction



- Cathodoluminescence (CL) – coherent light emission
- **Electron energy-loss spectroscopy (EELS)**



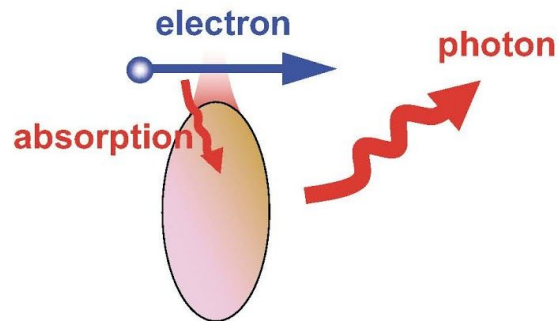
$$\text{EELS probability} \propto \int dt \operatorname{Re}\{e^{-i\omega t} \mathbf{v} \cdot \mathbf{E}(\mathbf{r}_t, \omega)\}$$

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}, \omega) - (\omega/c)^2 \varepsilon(\mathbf{r}, \omega) \mathbf{E}(\mathbf{r}, \omega) = \frac{4\pi i \omega}{c^2} \mathbf{j}(\mathbf{r}, \omega)$$

## Free-electron/optical-mode interaction



- Cathodoluminescence (CL) – coherent light emission
- **Electron energy-loss spectroscopy (EELS)**

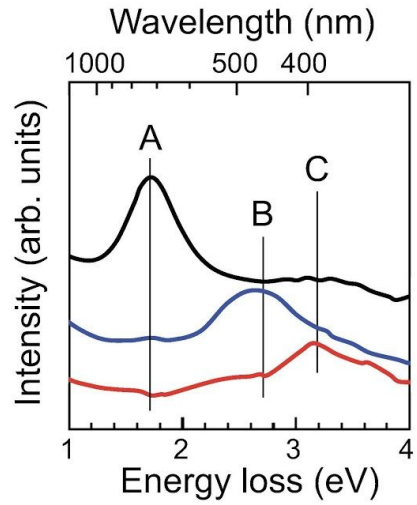
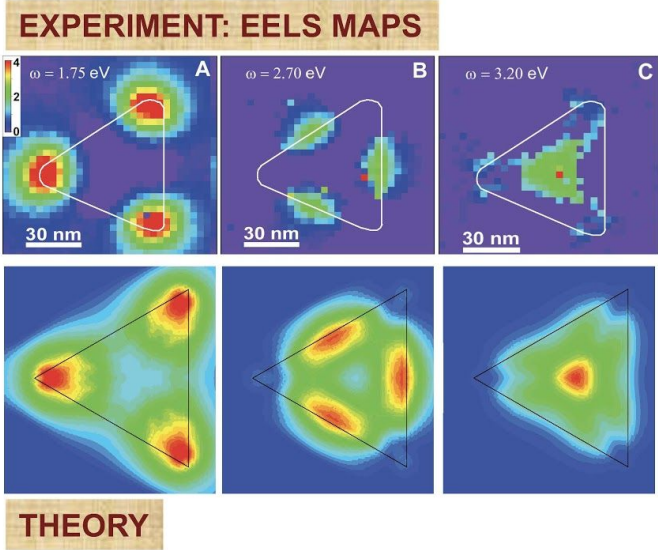


$$\text{EELS probability} \propto \int dt \operatorname{Re}\{e^{-i\omega t} \mathbf{v} \cdot \mathbf{E}(\mathbf{r}_t, \omega)\}$$

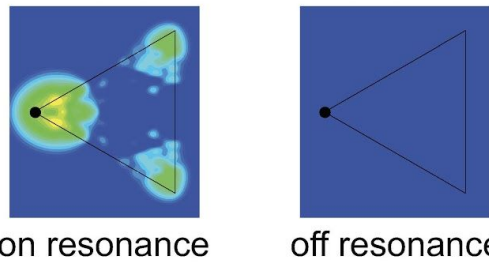
$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}, \omega) - (\omega/c)^2 \varepsilon(\mathbf{r}, \omega) \mathbf{E}(\mathbf{r}, \omega) = \frac{4\pi i \omega}{c^2} \mathbf{j}(\mathbf{r}, \omega)$$

The work done by the current  $\mathbf{j}(\mathbf{r}, \omega)$  on itself is  $-\hbar\omega$  times the EELS probability

# Plasmon near fields revealed by EELS



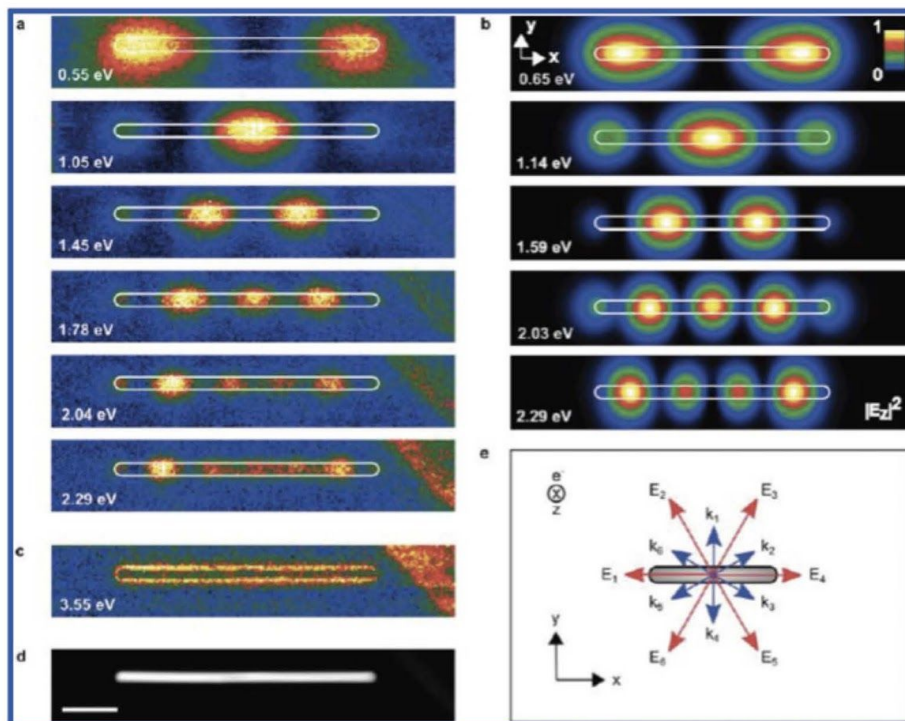
Near-field maps →



Nelayah *et al.*, Nature Physics (2007)



# Plasmon near fields revealed by EELS

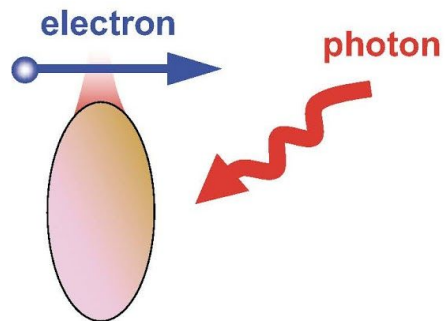


Rossouw et al., Nano Lett. (2011)

## Free-electron/optical-mode interaction



- Cathodoluminescence (CL) – coherent light emission
- Electron energy-loss spectroscopy (EELS)
- Electron energy-gain spectroscopy (EEGS)

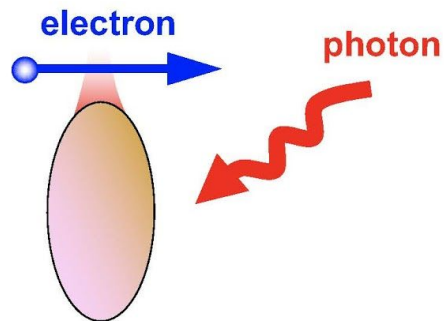


$$\mathbf{E}^{\text{ext}}(\mathbf{r}, \omega)$$

## Free-electron/optical-mode interaction



- Cathodoluminescence (CL) – coherent light emission
- Electron energy-loss spectroscopy (EELS)
- Electron energy-gain spectroscopy (EEGS)



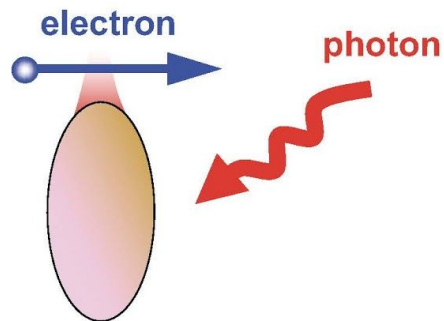
$$\text{EEGS probability} \propto \int dt \operatorname{Re}\{e^{-i\omega t} \mathbf{v} \cdot \mathbf{E}^{\text{ext}}(\mathbf{r}_t, \omega)\}$$

$$\mathbf{E}^{\text{ext}}(\mathbf{r}, \omega)$$

## Free-electron/optical-mode interaction



- Cathodoluminescence (CL) – coherent light emission
- Electron energy-loss spectroscopy (EELS)
- Electron energy-gain spectroscopy (EEGS)

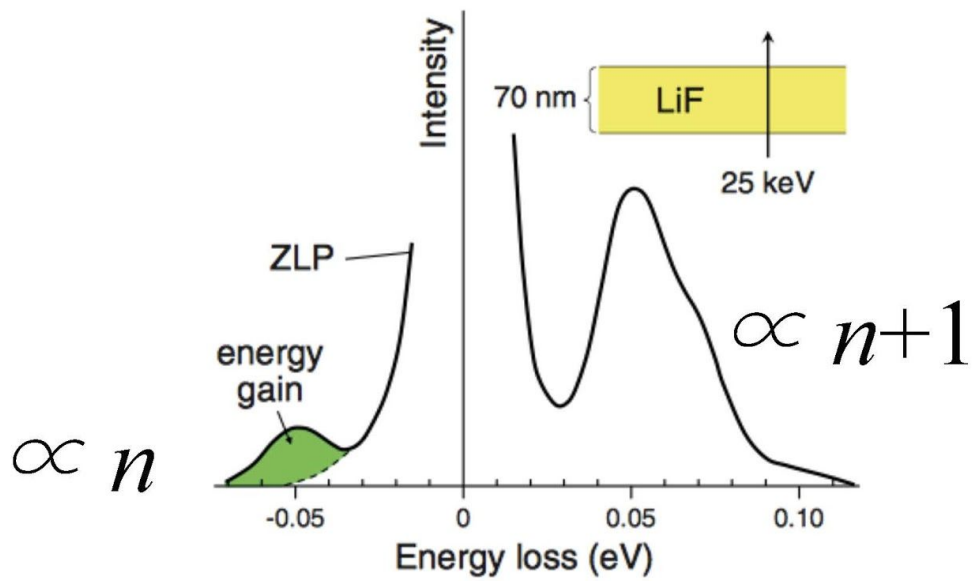


$$\text{EEGS probability} \propto \int dt \operatorname{Re}\{e^{-i\omega t} \mathbf{v} \cdot \mathbf{E}^{\text{ext}}(\mathbf{r}_t, \omega)\} \propto n_{\text{photon}}$$

$$n_{\text{photon}} \rightarrow n_{\text{photon}} - 1$$

Howie, Inst. Phys. Conf. Ser. (1999)  
García de Abajo and Kociak, New J. Phys. (2008)

# Thermal noise

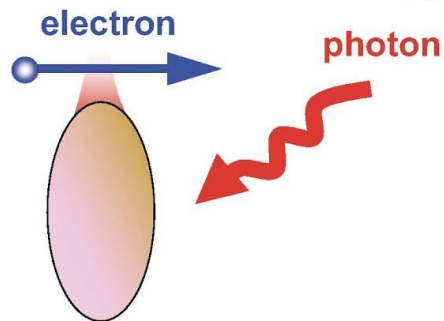


Boersch *et al.*, Phys. Rev. Lett. (1966)

## Free-electron/optical-mode interaction



- Cathodoluminescence (CL) – coherent light emission
- Electron energy-loss spectroscopy (EELS)
- Electron energy-gain spectroscopy (EEGS)
- **Photon-induced near-field electron microscopy (PINEM)**

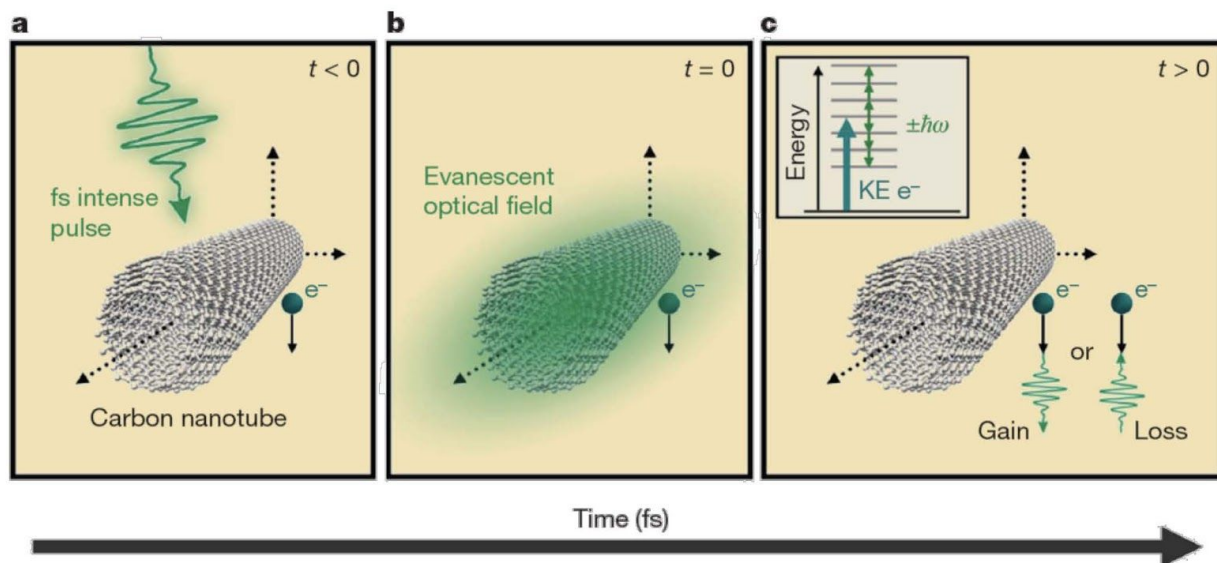


$$\text{EEGS probability} \propto \int dt \operatorname{Re}\{e^{-i\omega t} \mathbf{v} \cdot \mathbf{E}^{\text{ext}}(\mathbf{r}_t, \omega)\} \propto n_{\text{photon}}$$

$$n_{\text{photon}} \gg 1$$

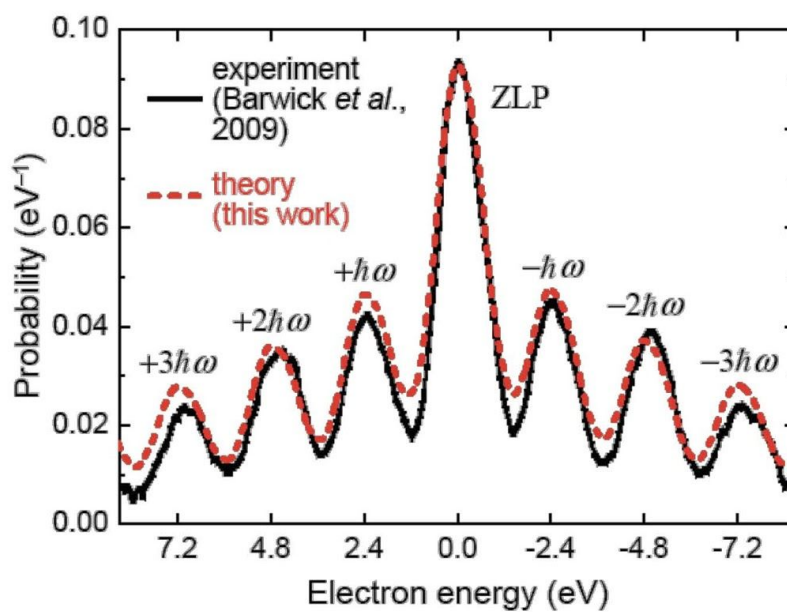
**EEGS probability = EELS probability via stimulated emission**

# PINEM



Barwick, Flannigan, and Zewail, Nature (2009)

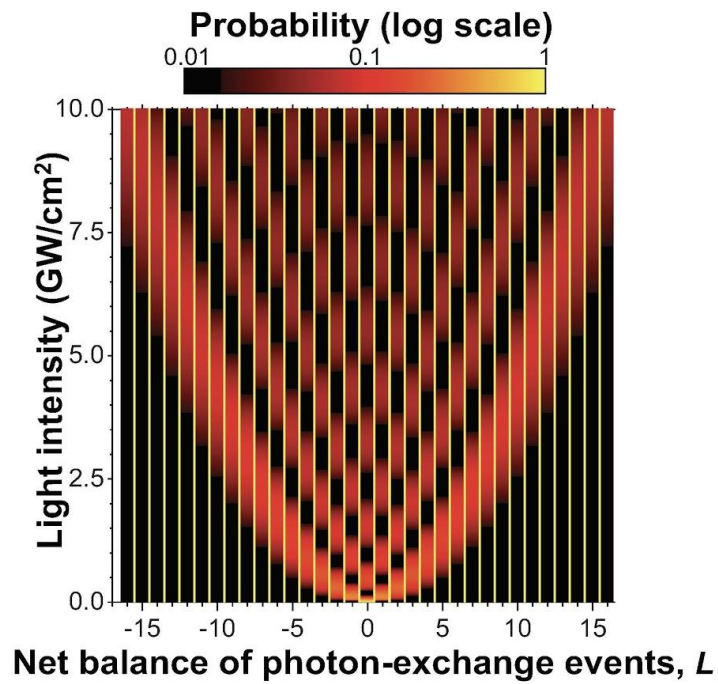
# PINEM



Experiment: Barwick, Flannigan, Zewail, Nature (2009)  
Theory: García de Abajo, Asenjo-Garcia, and Kociak, Nano Lett. (2010)

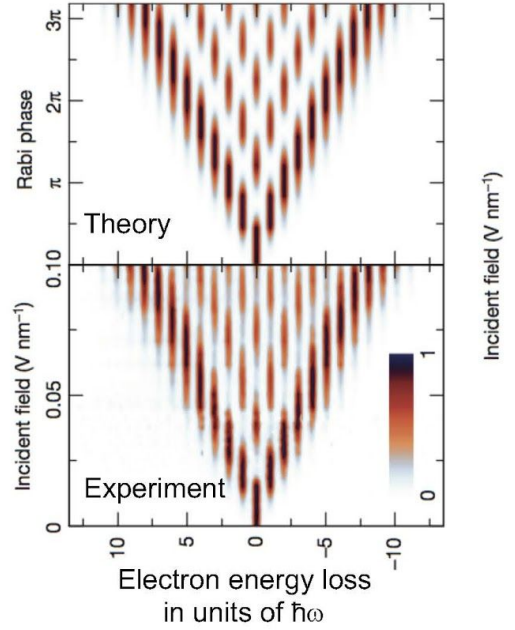
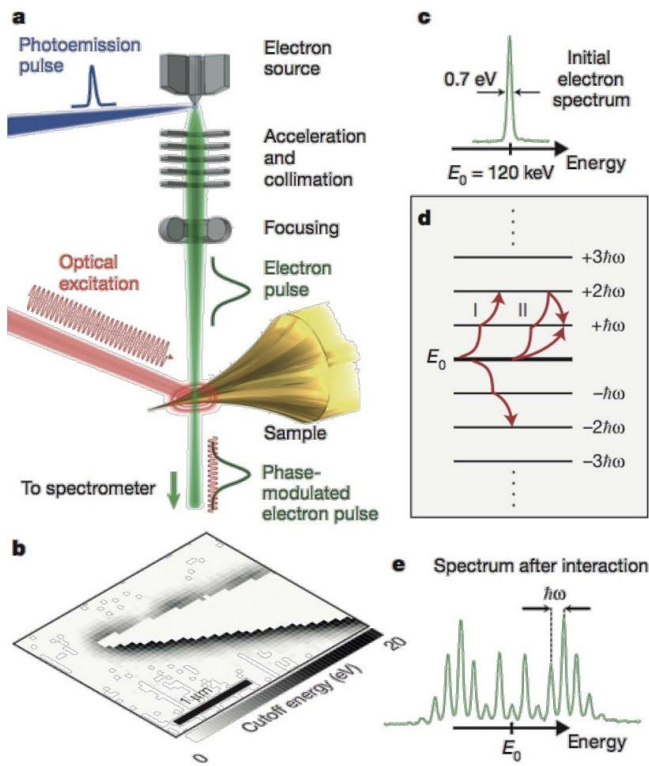


# PINEM



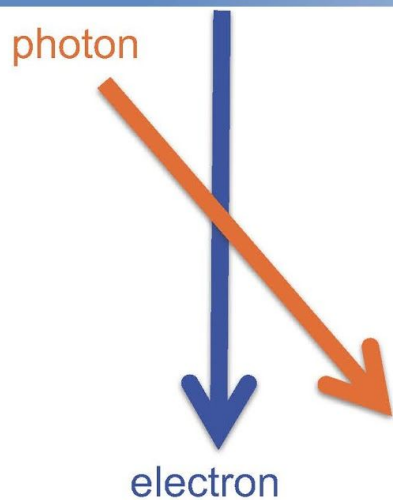
García de Abajo, Asenjo-Garcia, and Kociak, *Nano Lett.* (2010)

PINEM



Feist, ..., Ropers, Nature (2015)

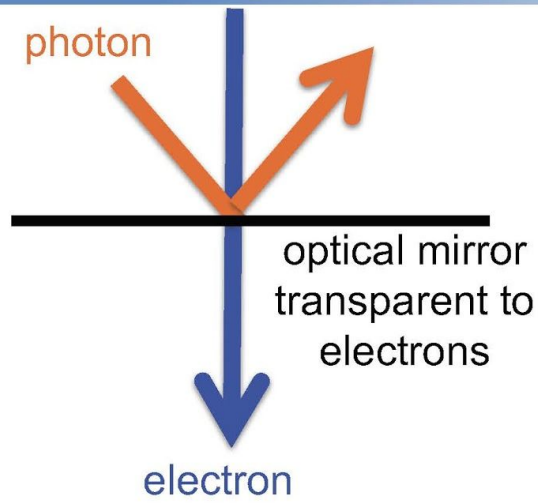
## Interaction with semi-infinite light plane waves



Vanacore, ..., Carbone, Nat. Commun. (2018)

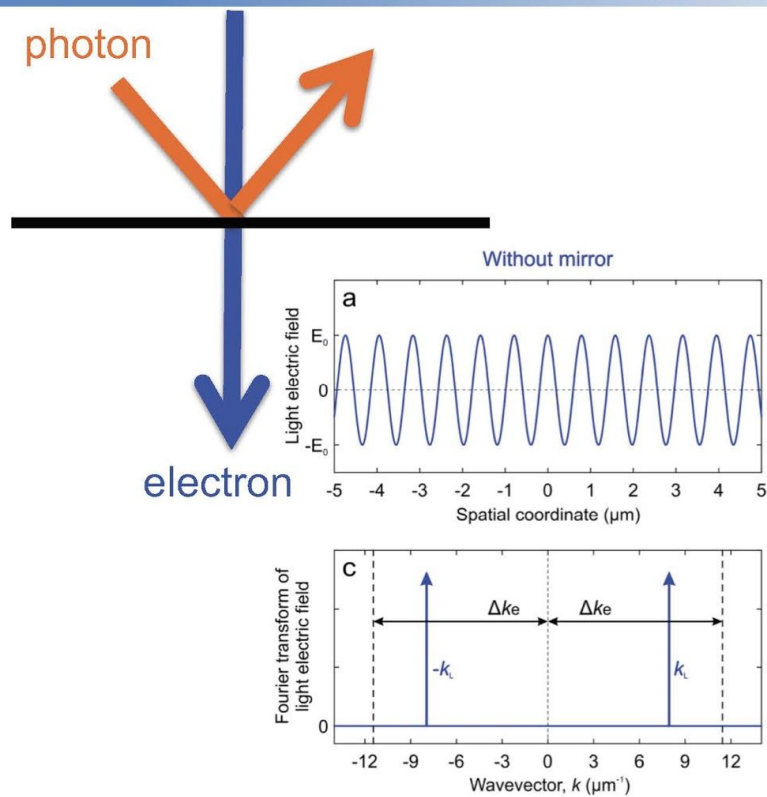
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## Interaction with semi-infinite light plane waves



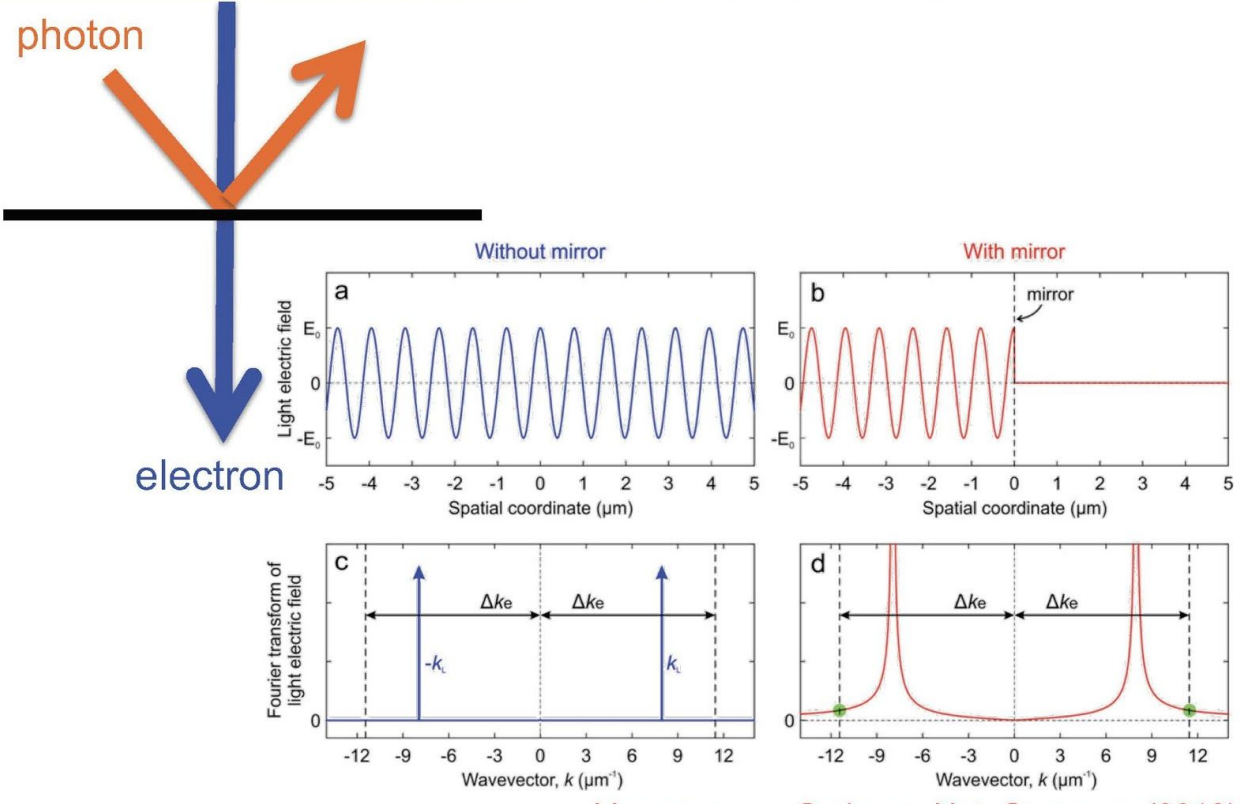
Vanacore, ..., Carbone, Nat. Commun. (2018)

## Interaction with semi-infinite light plane waves



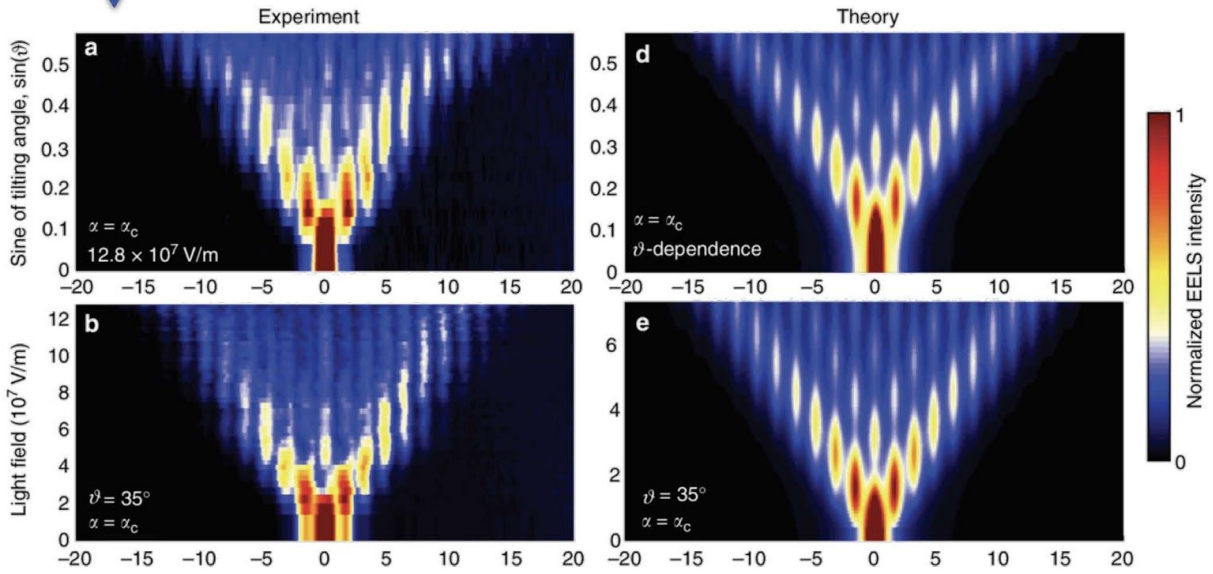
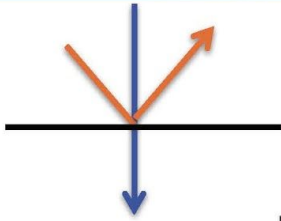
Vanacore, ..., Carbone, Nat. Commun. (2018)

Interaction with semi-infinite light plane waves



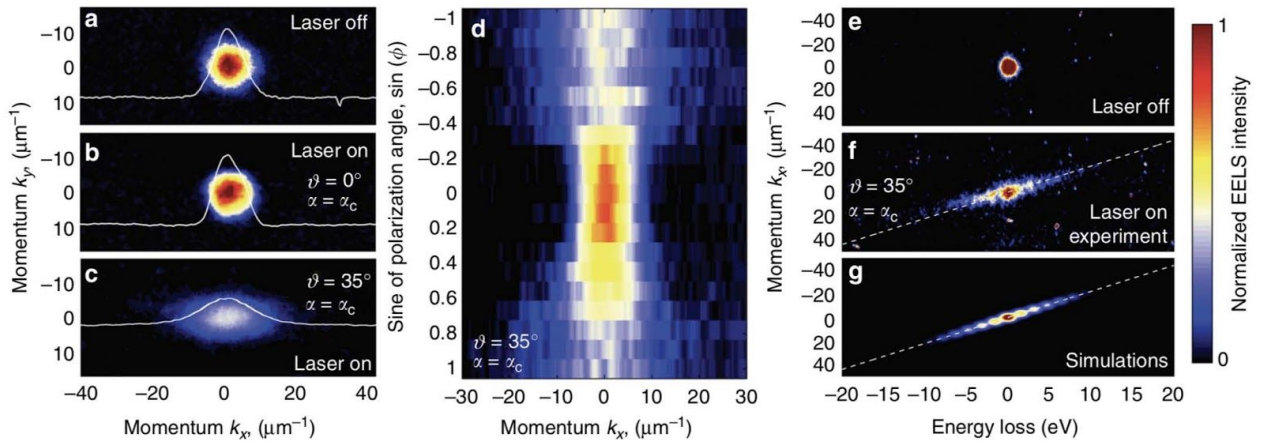
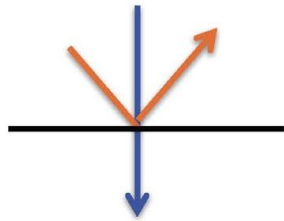
Vanacore, ..., Carbone, Nat. Commun. (2018)

# Interaction with semi-infinite light plane waves



Vanacore, ..., Carbone, Nat. Commun. (2018)

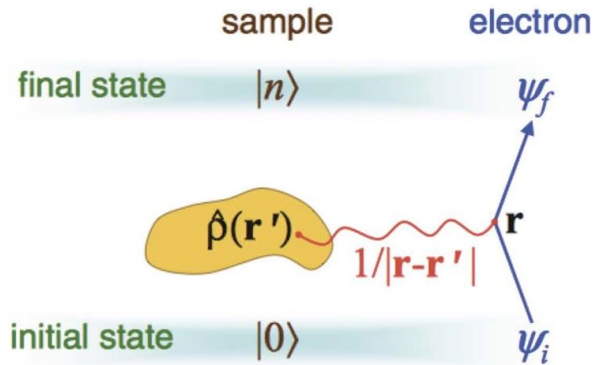
Interaction with semi-infinite light plane waves nanophotonics.es



Vanacore, ..., Carbone, Nat. Commun. (2018)



## Quantum treatment of EELS



$$\begin{aligned} \frac{d\Gamma(\omega)}{dt} &= \frac{8\pi\hbar e^2}{m_e^2} \sum_f \int d^3\mathbf{r} d^3\mathbf{r}' \psi_f(\mathbf{r}) \psi_f^*(\mathbf{r}') \\ &\times [\nabla \psi_i^*(\mathbf{r})] \cdot \text{Im}\{-G(\mathbf{r}, \mathbf{r}', \omega)\} \cdot [\nabla \psi_i(\mathbf{r}')] \\ &\times \delta(\epsilon_f - \epsilon_i + \omega) \end{aligned}$$

García de Abajo, Rev. Mod. Phys. (2010)

## Manipulation of transversal wave functions



$$\psi_{i,f}(\mathbf{r}) = \frac{1}{\sqrt{L}} e^{iqz} \psi_{i,f}(\mathbf{R})$$

$$\psi_i(\mathbf{R}) = \frac{1}{\sqrt{A}}$$

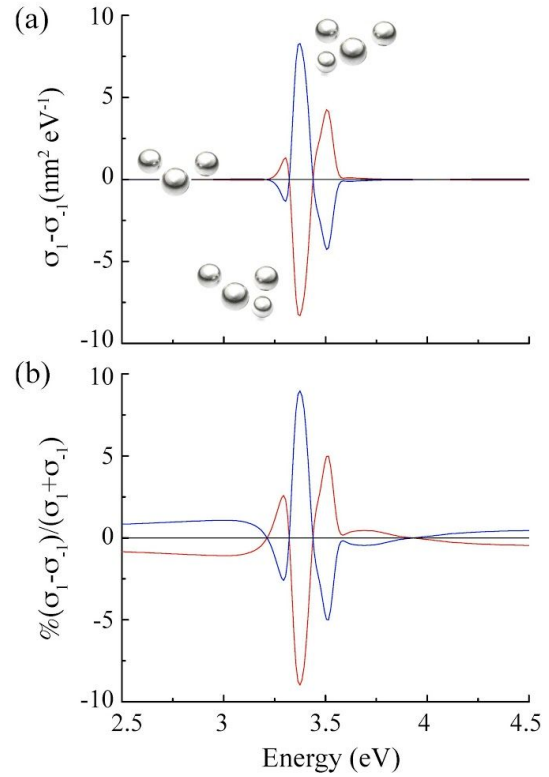
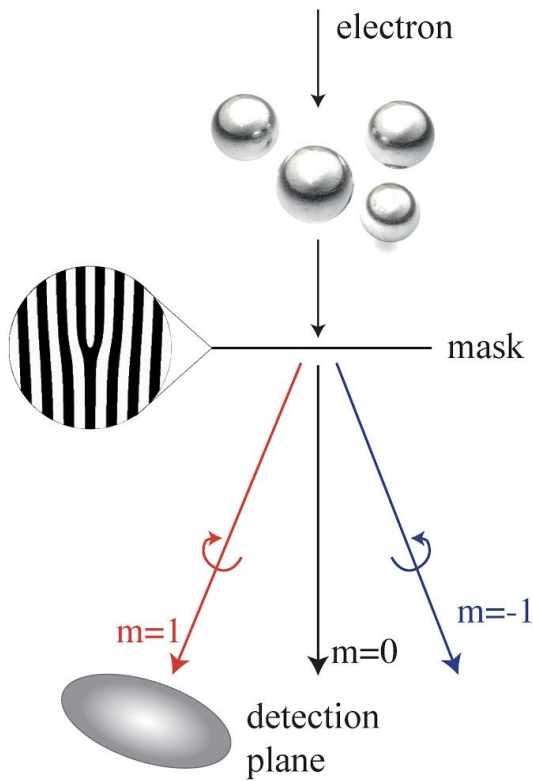
Plane  
wave

$$\psi_f(\mathbf{R}) = \frac{1}{\sqrt{A}} i^{m_f} e^{im_f\phi} J_{m_f}(Q_f R)$$

Vortex Beam

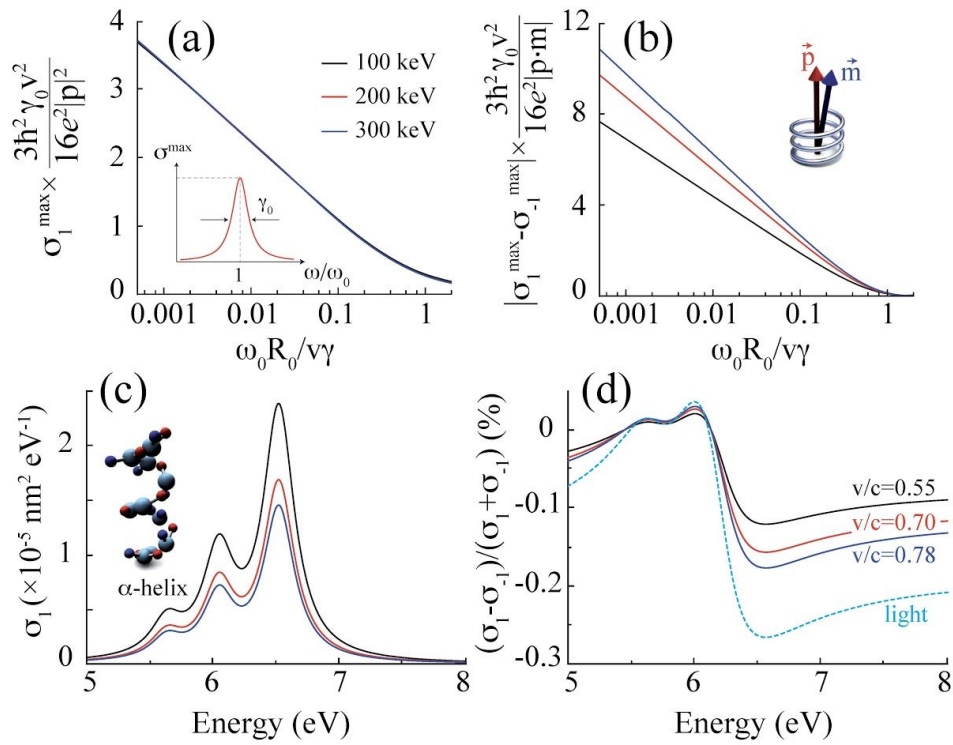
$$\begin{aligned} \frac{d\Gamma(\omega)}{dt} &= \frac{8\pi\hbar e^2}{m_e^2} \sum_f \int d^3\mathbf{r} d^3\mathbf{r}' \psi_f(\mathbf{r}) \psi_f^*(\mathbf{r}') \\ &\times [\nabla \psi_i^*(\mathbf{r})] \cdot \text{Im}\{-G(\mathbf{r}, \mathbf{r}', \omega)\} \cdot [\nabla \psi_i(\mathbf{r}')] \\ &\times \delta(\varepsilon_f - \varepsilon_i + \omega) \end{aligned}$$

Manipulation of transversal wave functions



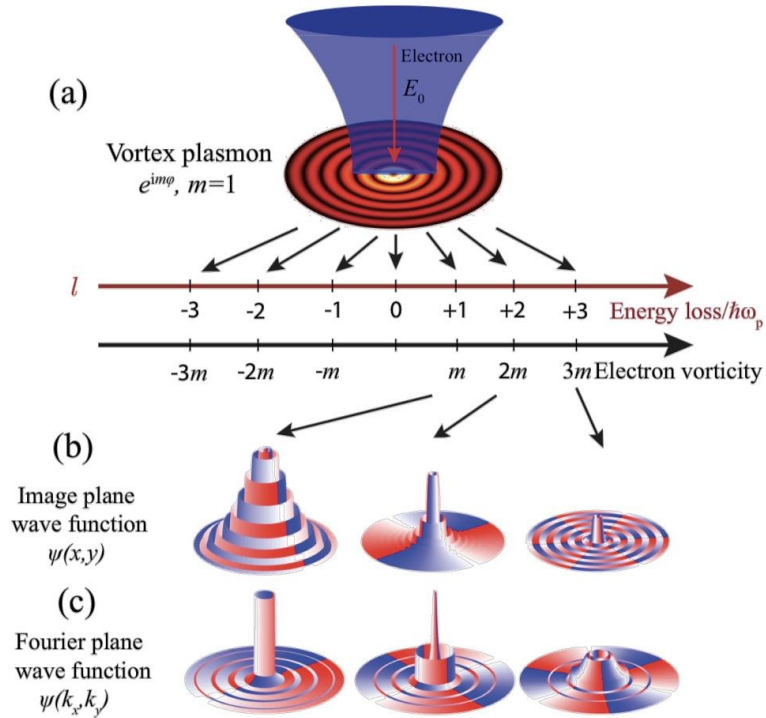
Asenjo and García de Abajo, PRL (2014)

Manipulation of transversal wave functions



Asenjo and García de Abajo, PRL (2014)

# Large photon-electron OAM transfer

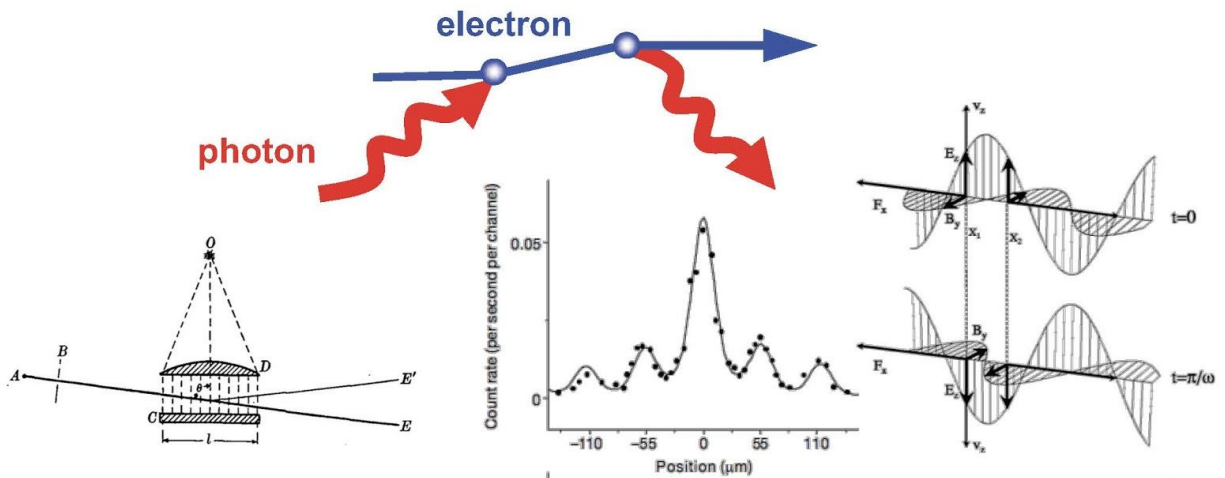


Cai et al., PRB 98, 045424 (2018)

## Free-electron/optical-mode interaction



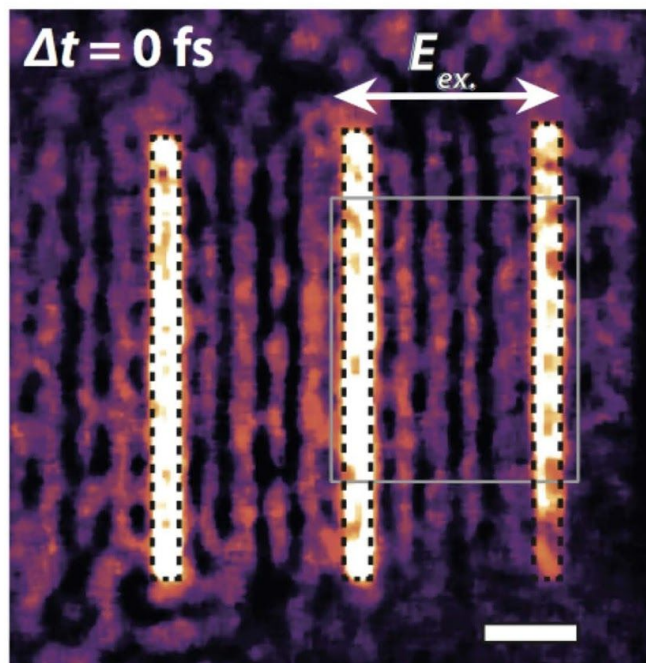
- Cathodoluminescence (CL) – coherent light emission
- Electron energy-loss spectroscopy (EELS)
- Electron energy-gain spectroscopy (EEGS)
- Photon-induced near-field electron microscopy (PINEM)
- **Kapitza-Dirac effect**



Kapitza and Dirac (1933)

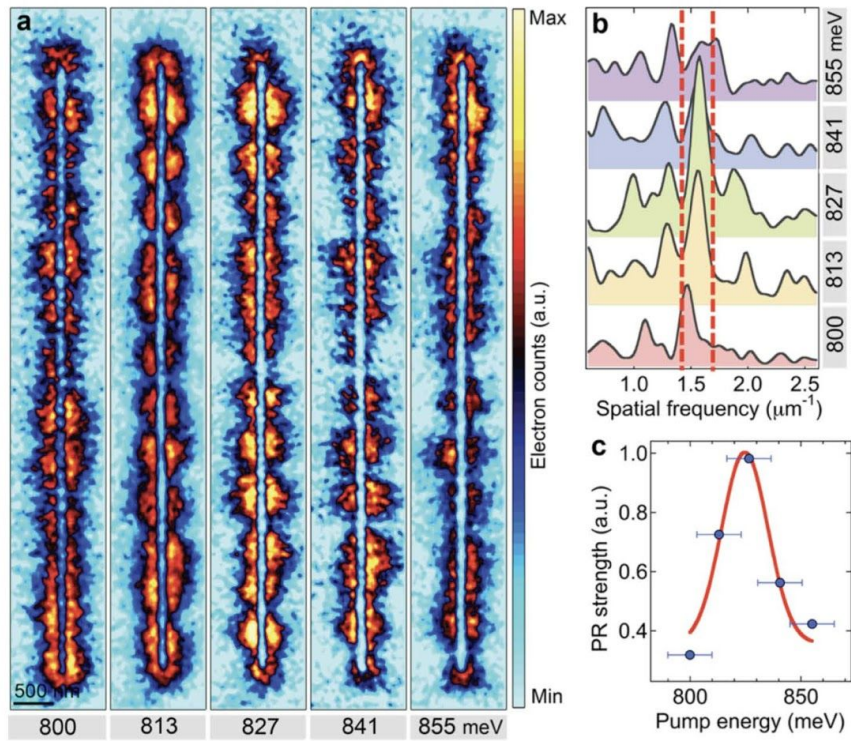
Freimund, Aflatooni, Batelaan, Nature (2001)  
 Batelaan, Rev. Mod. Phys. (2007)

## Plasmon Kapitza-Dirac effect – initial progress



Lummen, ..., Carbone, Nat. Commun. (2016)

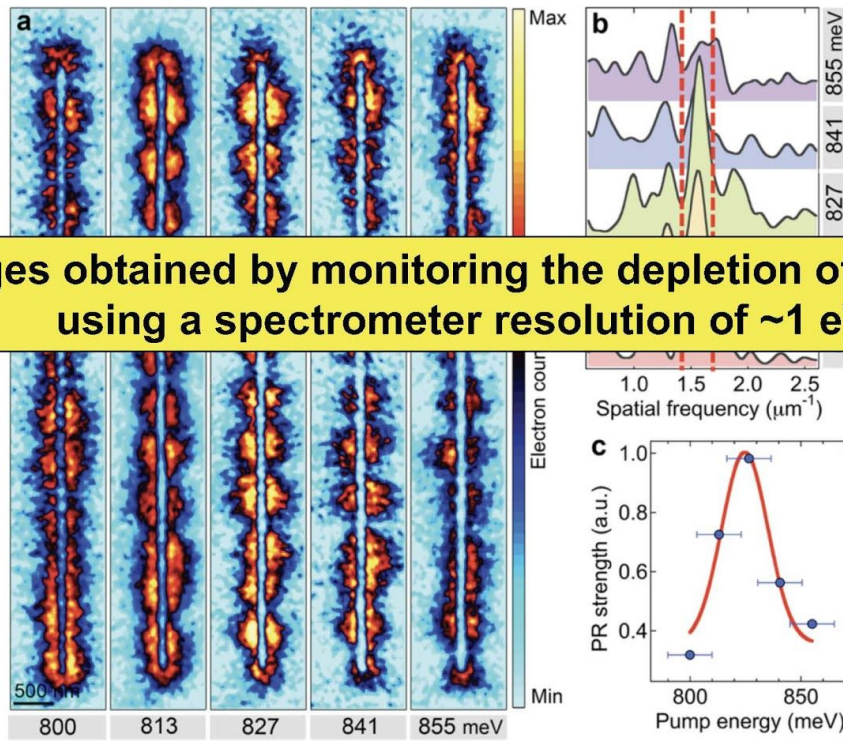
# From PINEM to EEGS



Pomarico, ..., Carbone, ACS Photon. (2018)



# From PINEM to EEGS



**Images obtained by monitoring the depletion of the ZLP using a spectrometer resolution of  $\sim 1$  eV**

Pomarico, ..., Carbone, ACS Photon. (2018)

## Electron-EM field interaction



$$\left[ m_e c^2 \beta + c \vec{\alpha} \cdot \left( \mathbf{p} + \frac{e}{c} \mathbf{A} \right) - e \varphi \right] \Psi = i \hbar \frac{\partial \Psi}{\partial t}$$

Di Giulio, Kociak, JGdeA,  
arXiv:19:05.06887

## Electron-EM field interaction



$$\left[ m_e c^2 \beta + c \vec{\alpha} \cdot (\mathbf{p} + \frac{e}{c} \mathbf{A}) - e \varphi \right] \Psi = i \hbar \frac{\partial \Psi}{\partial t}$$

$$\Psi = V^{-1/2} \sum_{\mathbf{k}} \psi_{\mathbf{k}} e^{i \mathbf{k} \cdot \mathbf{r} - i E_{\mathbf{k}} t / \hbar} \Psi_{\mathbf{k}}$$

$$\Psi_{\mathbf{k}} = \begin{bmatrix} A_{\mathbf{k}} \hat{\mathbf{s}} \\ B_{\mathbf{k}} \vec{\sigma} \cdot \mathbf{k} \hat{\mathbf{s}} \end{bmatrix}$$

$$(m_e c^2 \beta + c \vec{\alpha} \cdot \mathbf{p}) \Psi_{\mathbf{k}} = E_{\mathbf{k}} \Psi_{\mathbf{k}}$$

Di Giulio, Kociak, JGdeA,  
 arXiv:19:05.06887

## Electron-EM field interaction



$$\left[ m_e c^2 \beta + c \vec{\alpha} \cdot \left( \mathbf{p} + \frac{e}{c} \mathbf{A} \right) - e \varphi \right] \Psi = i \hbar \frac{\partial \Psi}{\partial t}$$

$$E_k \approx E_0 + (\hbar^2 c^2 / E_0) \mathbf{k}_0 \cdot (\mathbf{k} - \mathbf{k}_0)$$

$$\Psi = V^{-1/2} \sum_{\mathbf{k}} \psi_{\mathbf{k}} e^{i \mathbf{k} \cdot \mathbf{r} - i E_k t / \hbar} \Psi_{\mathbf{k}}$$

$$\Psi_{\mathbf{k}} = \begin{bmatrix} A_k \hat{\mathbf{s}} \\ B_k \vec{\sigma} \cdot \mathbf{k} \hat{\mathbf{s}} \end{bmatrix}$$

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Di Giulio, Kociak, JGdeA,  
 arXiv:19:05.06887

## Electron-EM field interaction



$$\left[ m_e c^2 \beta + c \vec{\alpha} \cdot \left( \mathbf{p} + \frac{e}{c} \mathbf{A} \right) - e \varphi \right] \Psi = i \hbar \frac{\partial \Psi}{\partial t}$$

$$E_k \approx E_0 + (\hbar^2 c^2 / E_0) \mathbf{k}_0 \cdot (\mathbf{k} - \mathbf{k}_0)$$

$$\mathbf{k} \rightarrow -i \nabla$$

$$\Psi = V^{-1/2} \sum_{\mathbf{k}} \psi_{\mathbf{k}} e^{i \mathbf{k} \cdot \mathbf{r} - i E_k t / \hbar} \Psi_{\mathbf{k}}$$

$$\Psi_{\mathbf{k}} = \begin{bmatrix} A_k \hat{\mathbf{s}} \\ B_k \vec{\sigma} \cdot \mathbf{k} \hat{\mathbf{s}} \end{bmatrix}$$

$$(m_e c^2 \beta + c \vec{\alpha} \cdot \mathbf{p}) \Psi_{\mathbf{k}} = E_k \Psi_{\mathbf{k}}$$

Di Giulio, Kociak, JGdeA,  
 arXiv:19:05.06887

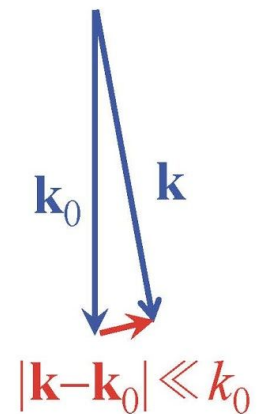
## Electron-EM field interaction



$$\left[ E_0 - (\hbar^2 c^2 / E_0) \mathbf{k}_0 \cdot (i\nabla + \mathbf{k}_0) + e\vec{\alpha} \cdot \mathbf{A} - e\varphi \right] \Psi = i\hbar \frac{\partial \Psi}{\partial t}$$

$$\Psi = V^{-1/2} \sum_{\mathbf{k}} \psi_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{r} - iE_{\mathbf{k}} t / \hbar} \Psi_{\mathbf{k}}$$

$$\Psi_{\mathbf{k}} = \begin{bmatrix} A_k \hat{\mathbf{s}} \\ B_k \vec{\sigma} \cdot \mathbf{k} \hat{\mathbf{s}} \end{bmatrix}$$



Di Giulio, Kociak, JGdeA,  
 arXiv:19:05.06887

## Electron-EM field interaction

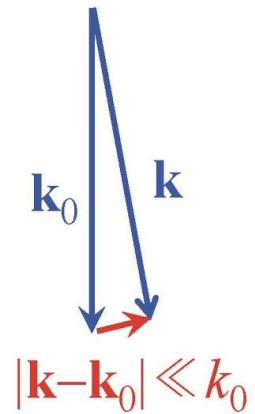


$$\left[ E_0 - (\hbar^2 c^2 / E_0) \mathbf{k}_0 \cdot (i\nabla + \mathbf{k}_0) + e\vec{\alpha} \cdot \mathbf{A} - e\varphi \right] \Psi = i\hbar \frac{\partial \Psi}{\partial t}$$

$$\Psi \approx \underbrace{\psi(\mathbf{r}, t)}_{\text{envelope}} \Psi_{\mathbf{k}_0}$$

$$\Psi = V^{-1/2} \sum_{\mathbf{k}} \psi_{\mathbf{k}} e^{i\mathbf{k} \cdot \mathbf{r} - iE_{\mathbf{k}} t / \hbar} \Psi_{\mathbf{k}_0}$$

$$\Psi_{\mathbf{k}} = \begin{bmatrix} A_k \hat{\mathbf{s}} \\ B_k \vec{\sigma} \cdot \mathbf{k} \hat{\mathbf{s}} \end{bmatrix}$$



Di Giulio, Kociak, JGdeA,  
 arXiv:19:05.06887

## Electron-EM field interaction



$$\left[ E_0 - (\hbar^2 c^2 / E_0) \mathbf{k}_0 \cdot (i\nabla + \mathbf{k}_0) + e\vec{\alpha} \cdot \mathbf{A} - e\varphi \right] \Psi = i\hbar \frac{\partial \Psi}{\partial t}$$

$$\Psi \approx \psi(\mathbf{r}, t) \Psi_{\mathbf{k}_0}$$

$$\Psi_{\mathbf{k}} = \begin{bmatrix} A_k \hat{\mathbf{s}} \\ B_k \vec{\sigma} \cdot \mathbf{k} \hat{\mathbf{s}} \end{bmatrix} \begin{cases} \Psi_{\mathbf{k}}^+ \Psi_{\mathbf{k}} = 1 \\ \Psi_{\mathbf{k}}^+ \vec{\alpha} \Psi_{\mathbf{k}} = (\hbar c / E_k) \mathbf{k} \end{cases}$$

Di Giulio, Kociak, JGdeA,  
arXiv:19:05.06887



## Electron-EM field interaction



$$\left[ E_0 - (\hbar^2 c^2 / E_0) \mathbf{k}_0 \cdot (i\nabla + \mathbf{k}_0) + e\vec{\alpha} \cdot \mathbf{A} - e\varphi \right] \Psi = i\hbar \frac{\partial \Psi}{\partial t}$$

$$\Psi \approx \psi(\mathbf{r}, t) \Psi_{\mathbf{k}_0} \quad \begin{aligned} \hbar \mathbf{k}_0 &= m_e \mathbf{v} \gamma \\ E_0 &= m_e c^2 \gamma \end{aligned}$$

$$\Psi_{\mathbf{k}} = \begin{bmatrix} A_k \hat{\mathbf{s}} \\ B_k \vec{\sigma} \cdot \mathbf{k} \hat{\mathbf{s}} \end{bmatrix} \begin{cases} \Psi_{\mathbf{k}}^+ \Psi_{\mathbf{k}} = 1 \\ \Psi_{\mathbf{k}}^+ \vec{\alpha} \Psi_{\mathbf{k}} = (\hbar c / E_k) \mathbf{k} \end{cases}$$

Di Giulio, Kociak, JGdeA,  
 arXiv:19:05.06887



## Electron-EM field interaction



$$[E_0 - \hbar \mathbf{v} \cdot (i\nabla + \mathbf{k}_0) + (e\mathbf{v}/c) \cdot \mathbf{A} - e\phi] \psi(\mathbf{r}, t) = i\hbar \frac{\partial \psi(\mathbf{r}, t)}{\partial t}$$

$$\hat{\mathbf{A}} = \sum_j (-ic/\omega_j) \vec{\mathcal{E}}_j(\mathbf{r}) e^{-i\omega_j t} \hat{a}_j + \text{h.c.}$$

Di Giulio, Kociak, JGdeA,  
arXiv:19:05.06887

## Electron-EM field interaction



$$[E_0 - \hbar \mathbf{v} \cdot (i\nabla + \mathbf{k}_0) + (e\mathbf{v}/c) \cdot \hat{\mathbf{A}} - e\phi] \psi(\mathbf{r}, t) = i\hbar \frac{\partial \psi(\mathbf{r}, t)}{\partial t}$$

$$\hat{\mathbf{A}} = \sum_j (-ic/\omega_j) \vec{\mathcal{E}}_j(\mathbf{r}) e^{-i\omega_j t} \hat{a}_j + \text{h.c.}$$

$$(\mathcal{H}_0 + \mathcal{H}_1) |\psi(\mathbf{r}, t)\rangle = i \frac{\partial |\psi(\mathbf{r}, t)\rangle}{\partial t}$$

$$\mathcal{H}_0 = \sum_j \hbar \omega_j \hat{a}_j^\dagger \hat{a}_j + E_0 - \hbar \mathbf{v} \cdot (i\nabla + \mathbf{k}_0)$$

$$\mathcal{H}_1 = (e\mathbf{v}/c) \cdot \hat{\mathbf{A}}$$

Di Giulio, Kociak, JGdeA,  
 arXiv:19:05.06887

## Electron-EM field interaction



$$\left[ m_e c^2 \beta + c \vec{\alpha} \cdot \left( \mathbf{p} + \frac{e}{c} \mathbf{A} \right) - e \varphi \right] \Psi = i \hbar \frac{\partial \Psi}{\partial t}$$

( ... )

$$(\mathcal{H}_0 + \mathcal{H}_1) |\psi(\mathbf{r}, t)\rangle = i \frac{\partial |\psi(\mathbf{r}, t)\rangle}{\partial t}$$

$$\mathcal{H}_0 = \hbar \omega_0 \hat{a}^\dagger \hat{a} + E_0 - \hbar \mathbf{v} \cdot (i \nabla + \mathbf{k}_0),$$

$$\mathcal{H}_1 = (-ie \mathbf{v} / c \omega_0) \left[ \vec{\mathcal{E}}(\mathbf{r}) e^{-i \omega_0 t} \hat{a} + \text{h.c.} \right]$$

Di Giulio, Kociak, JGdeA,  
 arXiv:19:05.06887

## Electron-EM field interaction



$$|\psi(\mathbf{r}, t)\rangle = \psi_0(\mathbf{r}, t) \sum_{\ell=-\infty}^{\infty} \sum_{n=0}^{\infty} e^{i\omega_0[\ell(z/v-t)-nt]} f_{\ell}^n(z) |n\rangle$$

$$\frac{df_{\ell}^n}{dz} = \sqrt{n} u^* f_{\ell+1}^{n-1} - \sqrt{n+1} u f_{\ell-1}^{n+1}$$

$$(\mathcal{H}_0 + \mathcal{H}_1)|\psi(\mathbf{r}, t)\rangle = i \frac{\partial |\psi(\mathbf{r}, t)\rangle}{\partial t}$$

$$\mathcal{H}_0 = \hbar\omega_0 \hat{a}^{\dagger} \hat{a} + E_0 - \hbar\mathbf{v} \cdot (i\nabla + \mathbf{k}_0),$$

$$\mathcal{H}_1 = (-iev/c\omega_0) \left[ \vec{\mathcal{E}}(\mathbf{r}) e^{-i\omega_0 t} \hat{a} + \text{h.c.} \right]$$

Di Giulio, Kociak, JGdeA,  
 arXiv:19:05.06887

## Electron-EM field interaction



$$|\psi(\mathbf{r}, t)\rangle = \psi_0(\mathbf{r}, t) \sum_{\ell=-\infty}^{\infty} \sum_{n=0}^{\infty} e^{i\omega_0[\ell(z/v-t)-nt]} f_{\ell}^n(z) |n\rangle$$

$$\frac{df_{\ell}^n}{dz} = \sqrt{n} u^* f_{\ell+1}^{n-1} - \sqrt{n+1} u f_{\ell-1}^{n+1}$$

$$f_{\ell}^n(\infty) = \sqrt{p_{n+\ell}} e^{i\chi} \sqrt{(n+\ell)!n!} e^{-|\beta_0|^2/2} (-\beta_0)^{\ell} \\ \times \sum_{n'} \frac{(-|\beta_0|^2)^{n'}}{n'!(\ell+n')!(n-n')!}$$

$$\beta = \frac{e}{\hbar\omega} \int dz \mathcal{E}_z(z) e^{-i\omega z/v}$$

Di Giulio, Kociak, JGdeA,  
 arXiv:19:05.06887

## From PINEM to EEGS



$$\beta = \frac{e}{\hbar\omega} \int dz \mathcal{E}_z(z) e^{-i\omega z/v}$$

$$\Gamma(\omega) \propto |\beta|^2 [\bar{n}\delta(\omega + \omega_0) + (\bar{n} + 1)\delta(\omega - \omega_0)]$$

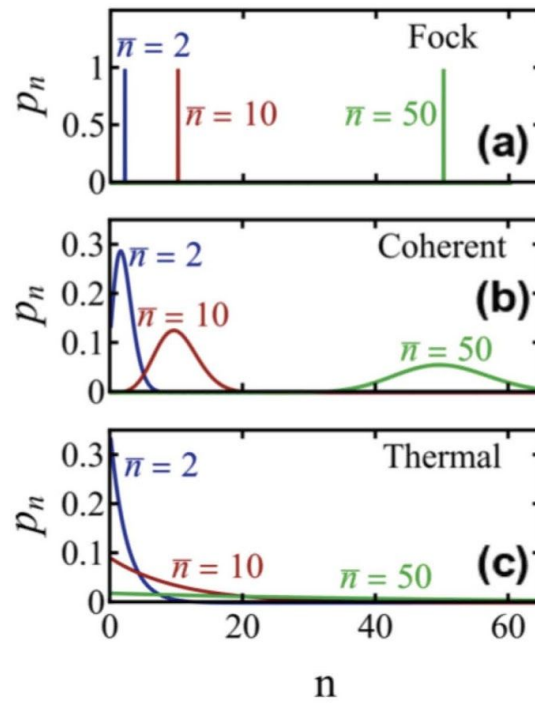
$$\vec{\mathcal{E}} = [(\omega^2/c^2)\mathbf{p} + (\mathbf{p} \cdot \nabla)\nabla] \frac{e^{i(\omega/c)r}}{r}$$

$$\Gamma_{\text{EELS,dipole}}(\omega) = \frac{1}{\hbar\pi} \left( \frac{2e\omega}{v^2\gamma} \right)^2 [K_1^2(\omega b/v\gamma) + K_0^2(\omega b/v\gamma)/\gamma^2] \text{Im}\{\alpha\}$$

Di Giulio, Kociak, JGdeA,  
arXiv:19:05.06887

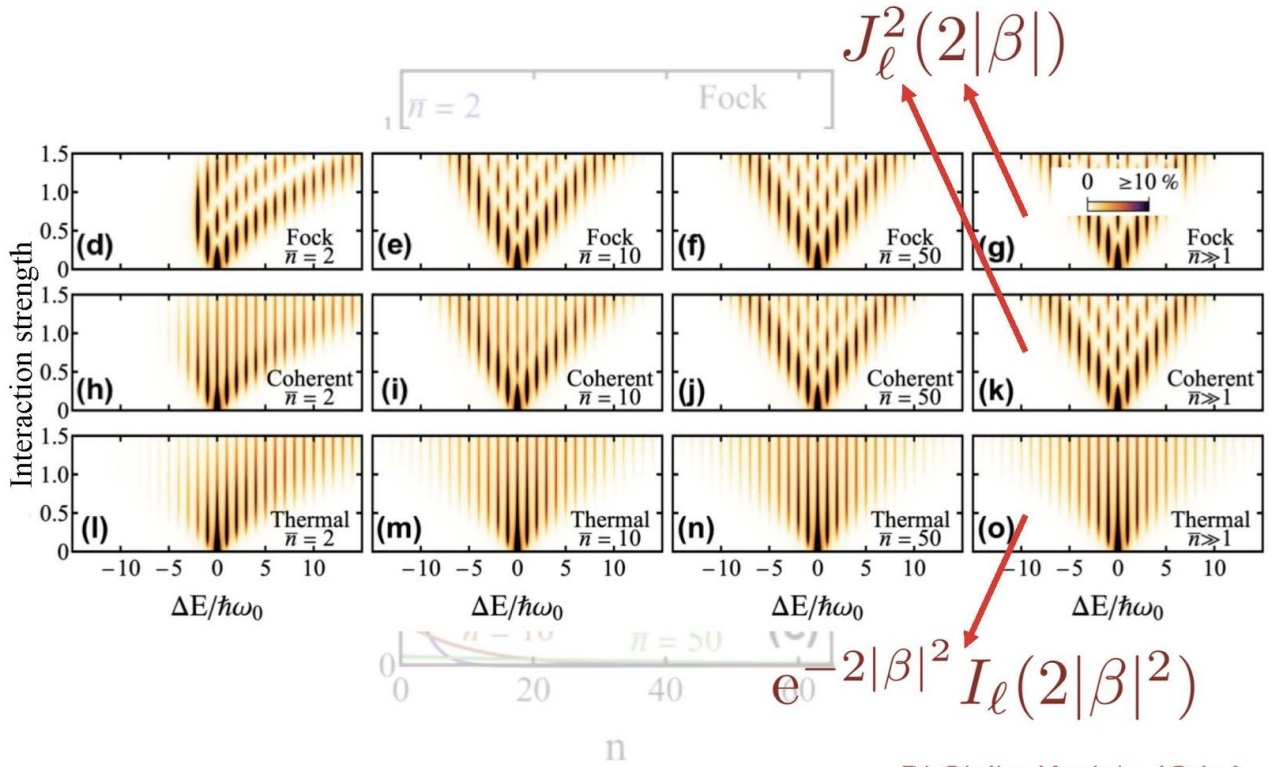


## Interaction with non-coherent light



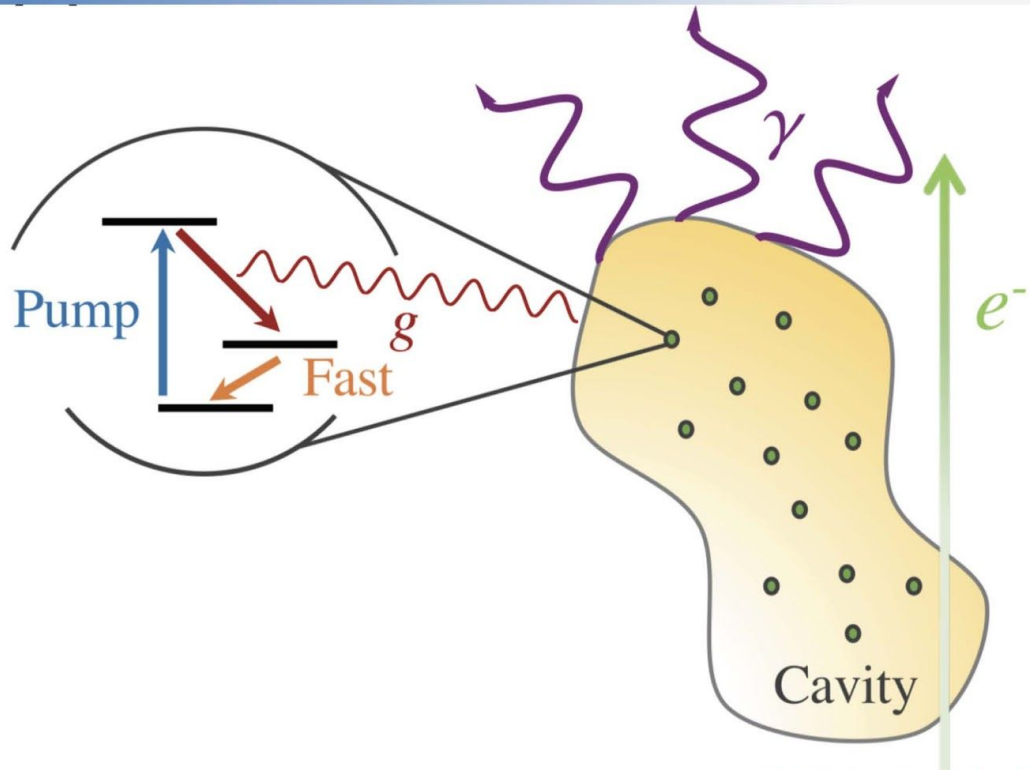
Di Giulio, Kociak, JGdeA,  
arXiv:19:05.06887

Interaction with non-coherent light



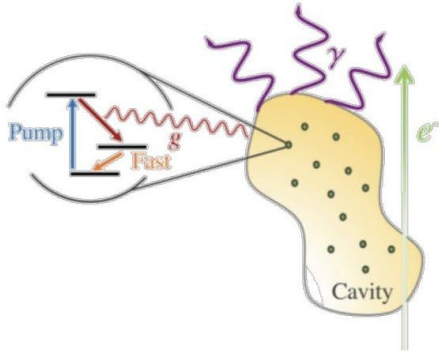
Di Giulio, Kociak, JGdeA,  
 arXiv:19:05.06887

## Interaction with non-coherent light

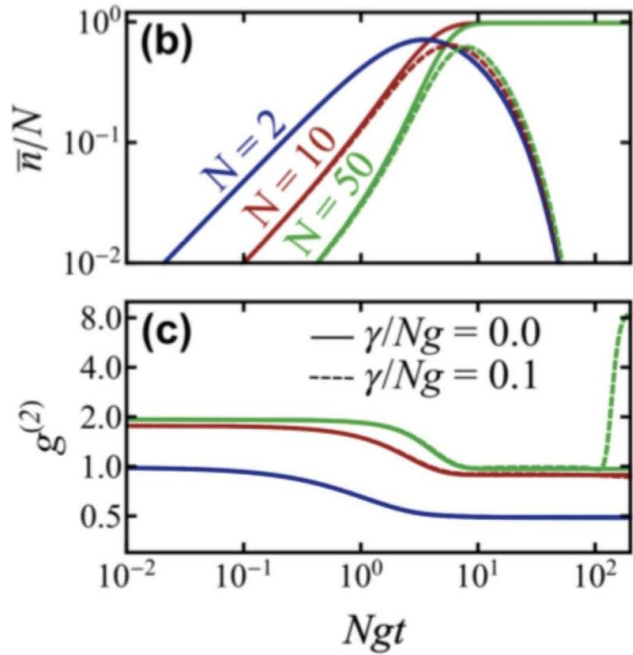


Di Giulio, Kociak, JGdeA,  
arXiv:19:05.06887

# Interaction with non-coherent light

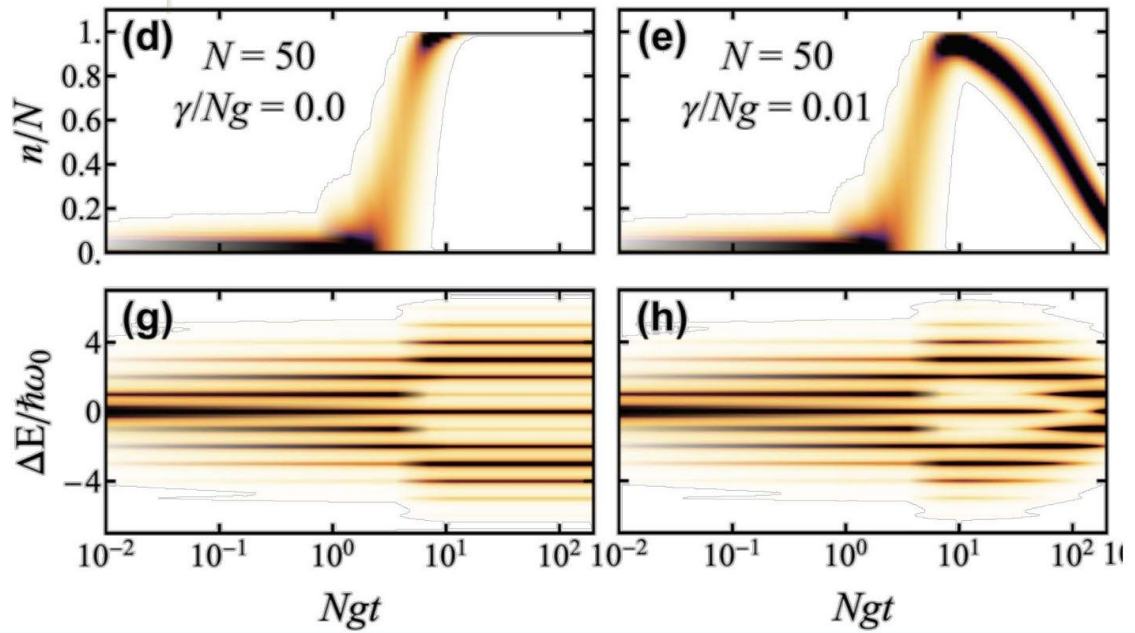
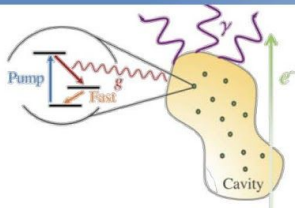


$$g^{(2)} = \frac{\langle n(n-1) \rangle}{\langle n \rangle^2}$$

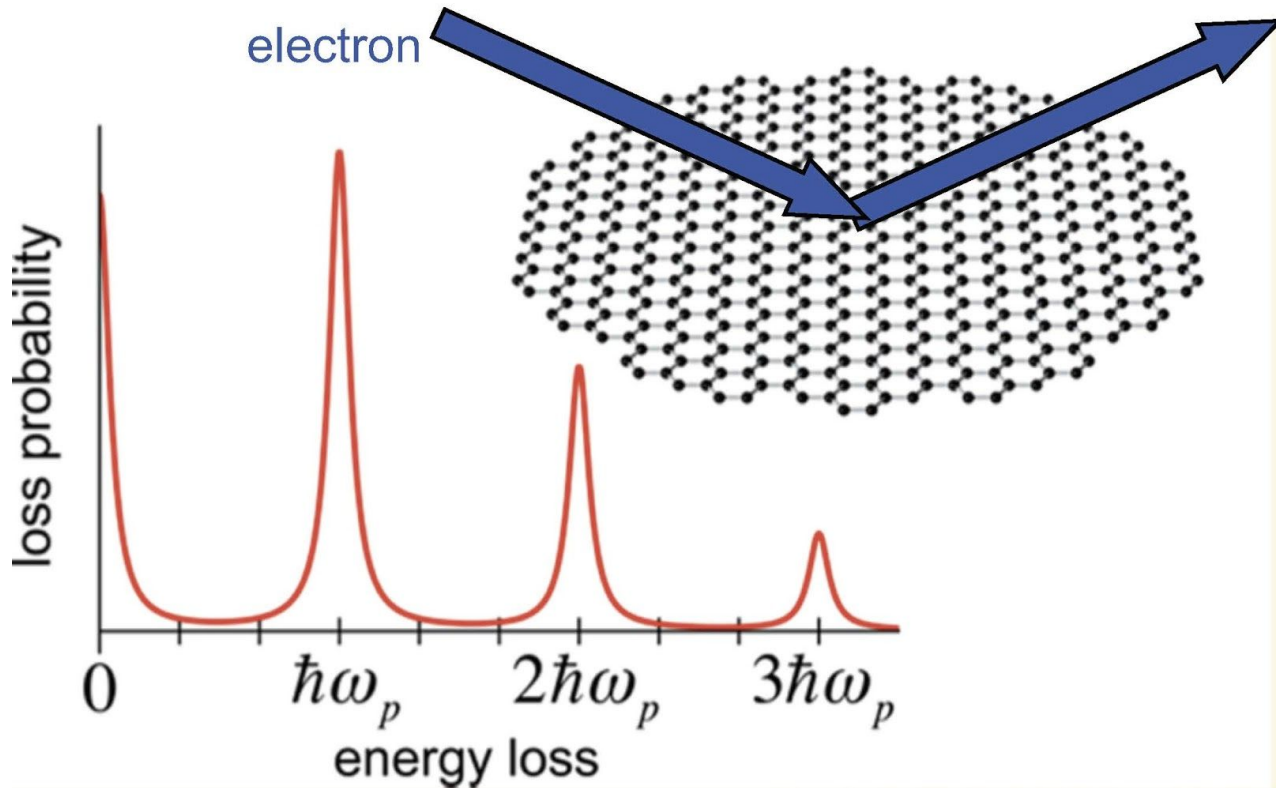


Di Giulio, Kociak, JGdeA,  
 arXiv:19:05.06887

## Interaction with non-coherent light



## Multiple excitations of the same mode



García de Abajo, ACS Nano (2013)

## Acknowledgments



Nanophotonics -  
Theory Group  
@ ICFO



## ANNEX 2: ABBREVIATIONS

### SHORT NAME OF PARTICIPANTS

Partner	Country	Short Name
National Research Council (Project scientific coordinator)	Italy	<b>CNR</b>
Forschungszentrum Jülich, Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons	Germany	<b>FZJ</b>
FEI - Thermo Fisher Scientific	The Netherlands	<b>FEI</b>
The Max Planck Institute for the Science of Light	Germany	<b>MPI</b>
University of Glasgow, Department of Physics and Astronomy	United Kingdom	<b>UG</b>
QED Film & Stage Productions Ltd.	United Kingdom	<b>QED</b>
University of Modena and Reggio Emilia, Department of Physics, Informatics, and Mathematics	Italy	<b>UMR</b>
Maastricht University, Maastricht MultiModal Molecular Imaging Institute	The Netherlands	<b>MU</b>

### LIST OF ABBREVIATIONS

Consortium Agreement	<b>CA</b>
Description of Action	<b>DoA</b>
Description of Work	<b>DoW</b>
European Commission	<b>EC</b>
Grant Agreement	<b>GA</b>
Kick-off Meeting	<b>KoM</b>
Project Management Board	<b>PMB</b>
Work Package	<b>WP</b>
Work Package Leader	<b>WPL</b>