EPFL

Electron-matter interactions: Plasmonics & nano-optics

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"Depending on the situation, surface plasmons must be regarded as many-body electron states or as electromagnetic waves, thus requiring either a quantum or a classical treatment with the possible inclusion of relativistic effects. This variety is a major obstacle to the establishment of a general theoretical treatment.

Even more crucially, surface plasmons suffer from huge ohmic losses in the metal which lead to very short lifetimes (typically a few femtoseconds). Theoretically, this presence of dissipation dramatically hardens the theoretical description of surface plasmons."

Hugo Lourenço-Martins, PhD thesis

Contents

- Kramers-Kronig analysis and band gap measurements in visible light regime
- An evanescent probe
- Introduction to localised surface plasmon resonances (LSPRs) in EELS
- Theoretical approaches and numerical methods for EELS LSPR analysis
  Dielectric in optical regime
- Coupling of plasmonic nanoparticles: EELS vs planewave illumination; imaging of hybridisation and bonding modes
- Plasmon damping; scattering and absorption
- Dielectrics and bound-states-in-the-continuum

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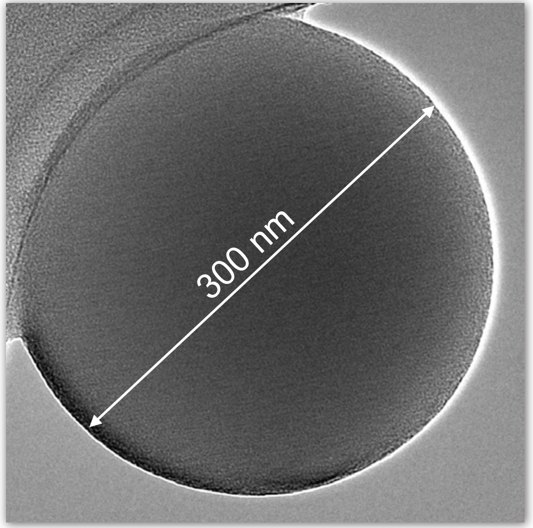
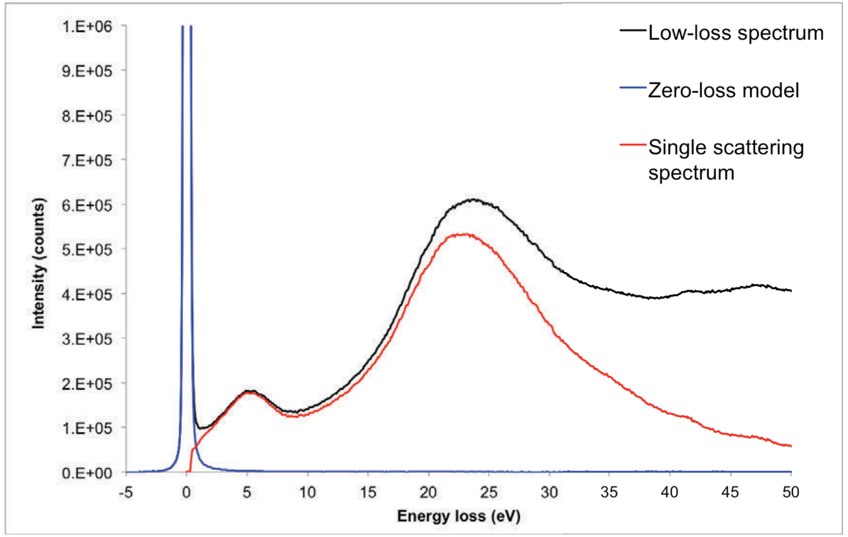
EELS ⟺ Optical response

- Deep relationship between EELS and light-optical response
- Example: determination of optical properties of atmospheric amorphous carbon spheres using Kramers-Kronig analysis of SSD EEL spectrum via:

$$\Gamma_{\text{bulk}}^{\text{NR}}(\omega) = \frac{2e^2 L}{\pi \hbar v^2} \mathrm{Im}\left\{-\frac{1}{\epsilon}\right\} \ln\left(\frac{q_c v}{\omega}\right)$$

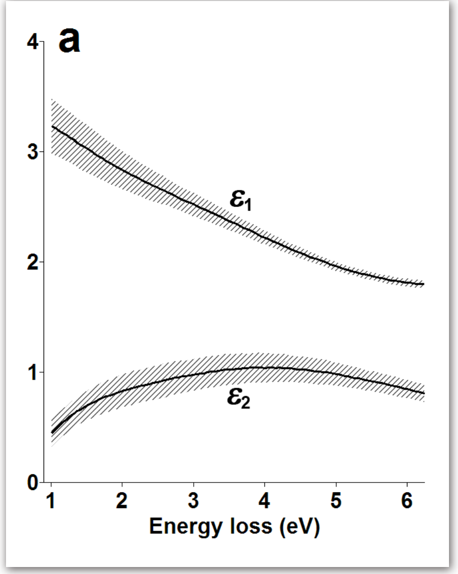


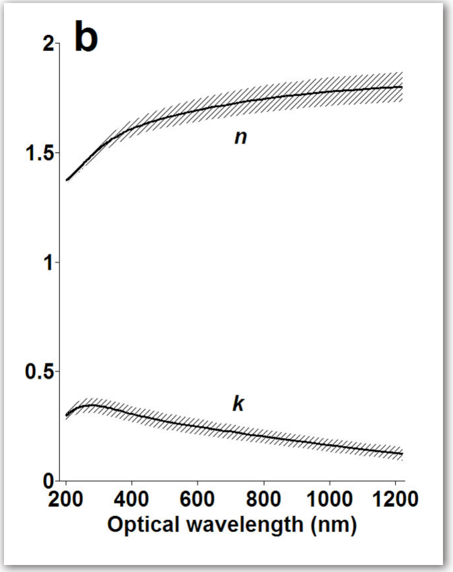


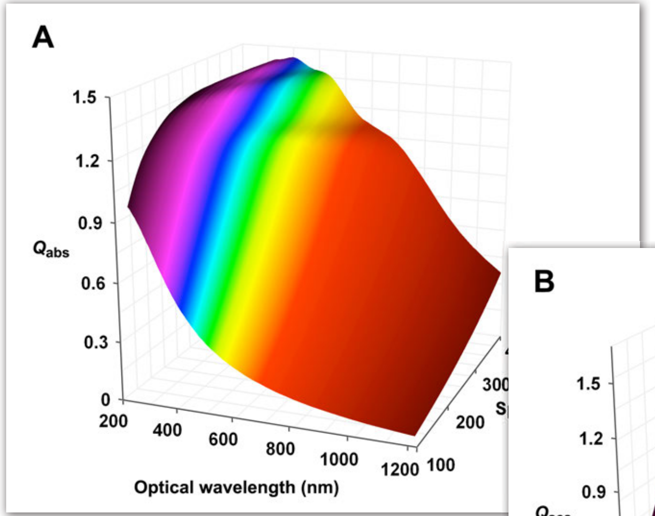
Alexander et al. *Science* **321** (2008) 833–836

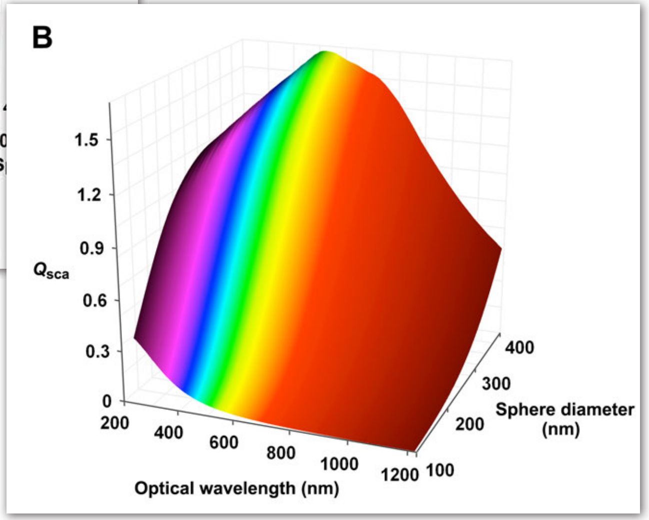
EELS ⟺ Optical response

- Kramers-Kronig normalisation using thickness of spheres









Property	Carbon sphere	Soot carbon
$N = n - ik$	$1.27 - 0.27i$	$1.95 - 0.79i$
Average spherule diameter (nm)	230	32
Number of spherules	1	55
σ_{abs} ($\times 10^{-14}$ m^2)	4.0	0.8
σ_{sca} ($\times 10^{-14}$ m^2)	3.2	0.2
ω_0	0.44	0.23
ρ (g cm^{-3})	~1.4–1.6	1.8–2.0
MAE (m^2 g^{-1})	~3.6–4.1	4.3–4.8
MSE (m^2 g^{-1})	~3.2–3.7	1.3–1.4
$Å_{abs}$	~1.5	~1

Alexander et al. *Science* **321** (2008) 833–836

Mie theory: Bohren and Huffman
"Absorption and scattering of light by small particles"

Band gap measurements

- In band gap $\epsilon'' \rightarrow 0$ therefore we should have $\Gamma_{\text{bulk}} \rightarrow 0$
- Measurements fraught with difficulties (Cherenkov radiation, surface damage/oxidation, surface plasmons, removal of ZLP…). See papers by M. Stöger-Pollach
- Example of good study: Bowman et al. *Ultramicroscopy* **167** (2016) 5–10:

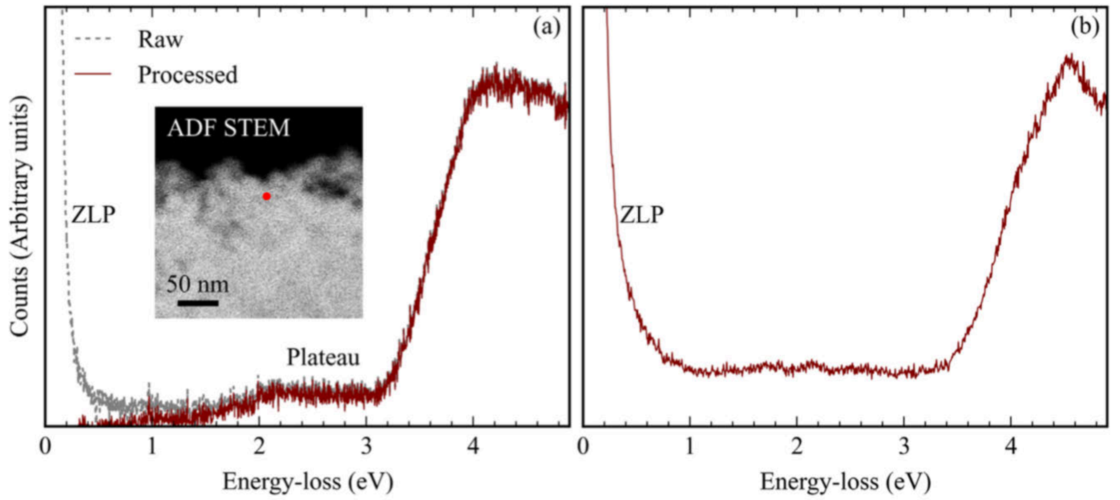


Fig. 2. (a) Typical valence-loss spectra from (a) oxidized PCO—showing ZLP, inter-band plateau feature and conduction band onset, and (b) fragment of bulk CeO_2. The beam position during PCO spectra acquisition is indicated in the monochromated ADF STEM image inset in (a).

- What is the spatial resolution of such a measurement?

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An evanescent probe

- Point charge moving in vacuum: evanescent source of radiation
  $\Longrightarrow$ can explore regions of momentum-space that lie outside the light cone

- Electric field of swift charged particle moving in homogeneous medium (distance r, frequency ω, time t):

$$\vec{E}(\vec{r},t) = \int \frac{d\omega}{2\pi} \vec{E}(\vec{r},\omega) e^{-i\omega t}$$

- In spectral domain, direct solution of Maxwell's equations yields:

$$\vec{E}(\vec{r},\omega) = \frac{ie}{\pi} \int d\vec{q} \, \frac{\vec{q}/\epsilon - k\vec{v}/c}{q^2 - k^2\epsilon} e^{i\vec{q}\cdot\vec{r}} \delta(\omega - \vec{q}\cdot\vec{v})$$

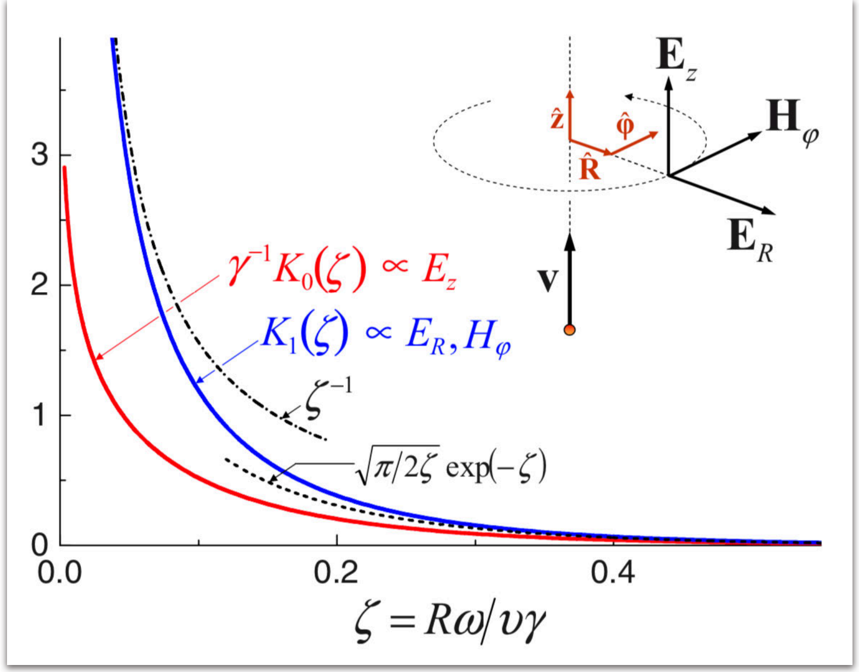
for wave vector $\vec{q}$ where $k = \omega/c$ is the light wave number in free space and ϵ is the dielectric function of the homogeneous medium

F.J. García de Abajo "Optical excitations in electron microscopy," Rev. Mod. Phys. **82** (2010) 209–275

An evanescent probe

- Applying cartesian coordinates & with $v < c$ integral yields:

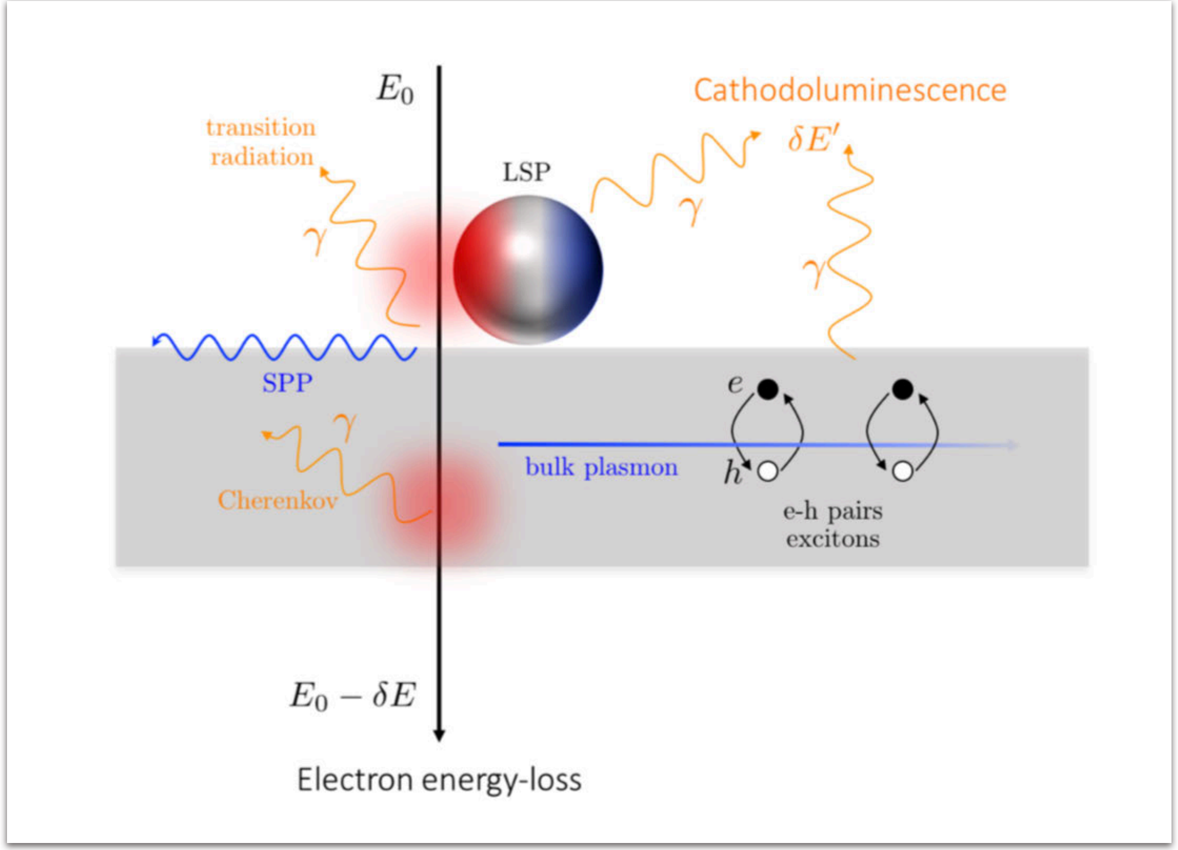
$$\vec{E}(r,\omega) = \frac{2e\omega}{v^2\gamma\epsilon} e^{i\omega z/v} \left[\frac{i}{\gamma} K_0\left(\frac{\omega R}{v\gamma}\right)\hat{z} - K_1\left(\frac{\omega R}{v\gamma}\right)\hat{\boldsymbol{R}}_\perp \right]$$



- Non vanishing components decay exponentially $\Longrightarrow$ response delocalisation
  Greater $v \Longrightarrow$ field extends further

F.J. García de Abajo "Optical excitations in electron microscopy," Rev. Mod. Phys. **82** (2010) 209–275

Probe–sample interactions



- e⁻-beam does not need to penetrate sample to stimulate surface excitation
  ⟹ measurements in "aloof" geometry

Figure from: Hugo Lourenço-Martins, PhD thesis

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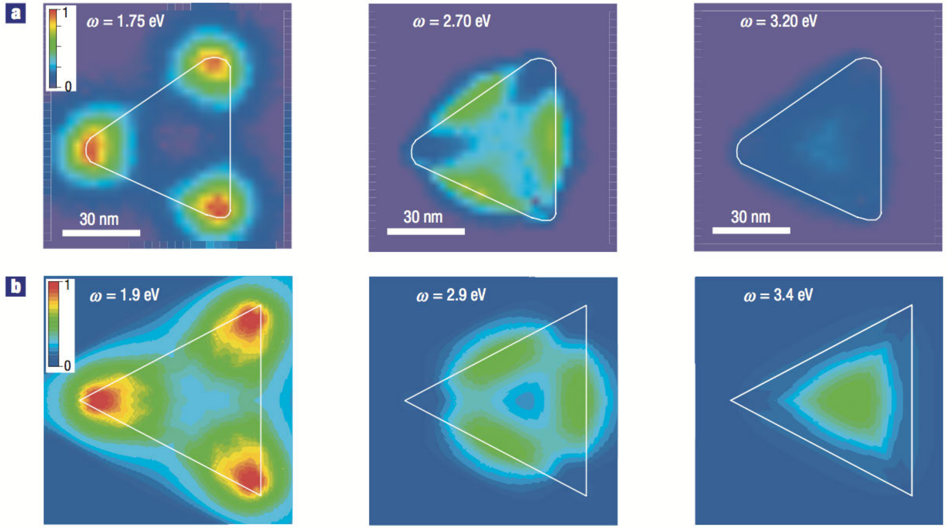
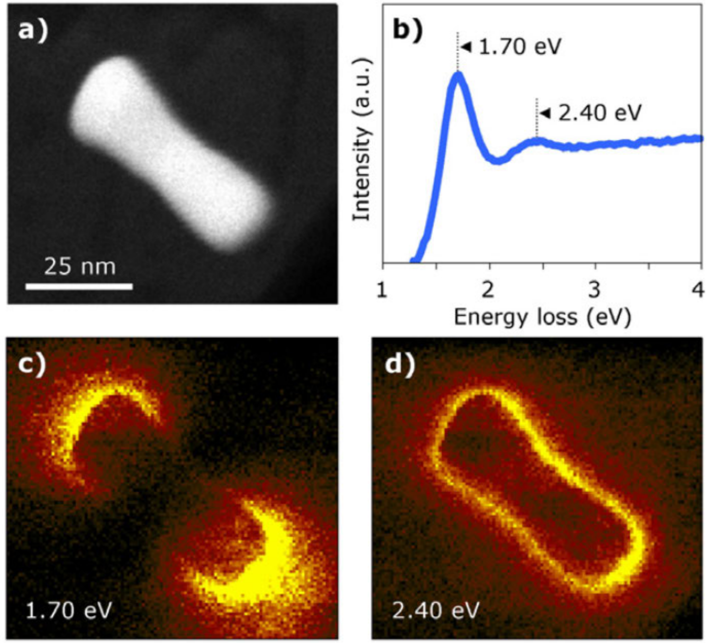
First EELS mapping of LSPRs

- EELS measurement of localised surface plasmon resonance (LSPR) eigenmodes



Nelayah et al. "Mapping surface plasmons on a single metallic nanoparticle," Nature Phys. **3** (2007) 348–353



Bosman et al. "Mapping surface plasmons at the nanometre scale with an electron beam," Nanotechnology **18** (2007) 165505

- Looks impressive: spatial resolution ~λ/(30–40) – *but what are we actually measuring?*

Surface plasmons(-polaritons)

- SPs: collective charge density waves of e⁻s propagating at interface associated to strongly confined EM fields
- Consider interface of 2 semi-infinite media with dielectric constants ϵ_m and ϵ_d
- Solution of Helmholtz wave eq. and continuity relations are surface plasmons (SPs):

$$\vec{\boldsymbol{E}}_j = \begin{pmatrix} E_{x,j} \\ 0 \\ E_{z,j} \end{pmatrix} e^{i(k_x x - \omega t)} e^{ik_{z,j} z}$$

- Dispersion relation: $k_{\mathrm{SP}} = \dfrac{\omega}{c}\sqrt{\dfrac{\epsilon_m \epsilon_d}{\epsilon_m + \epsilon_d}}$

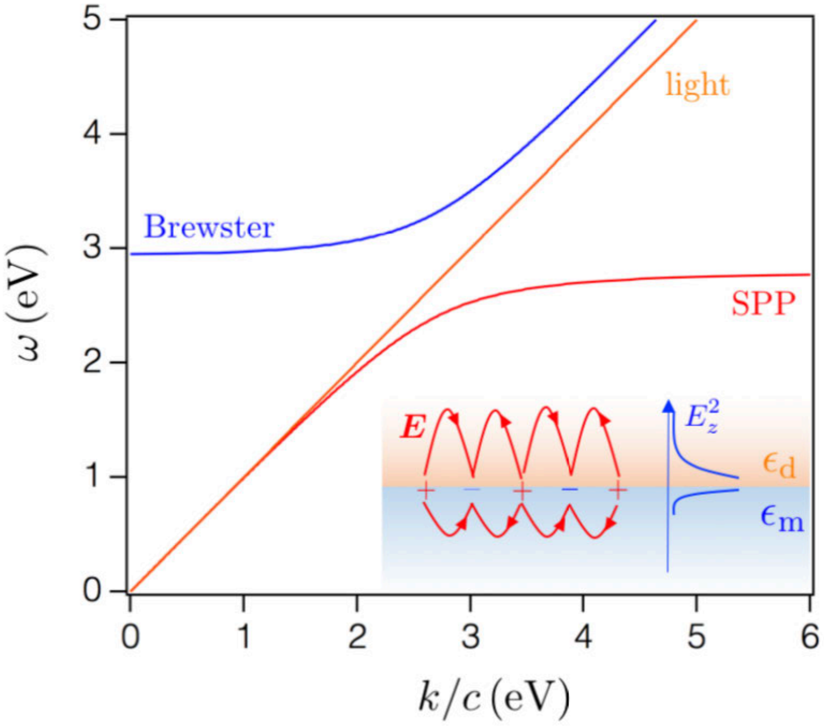
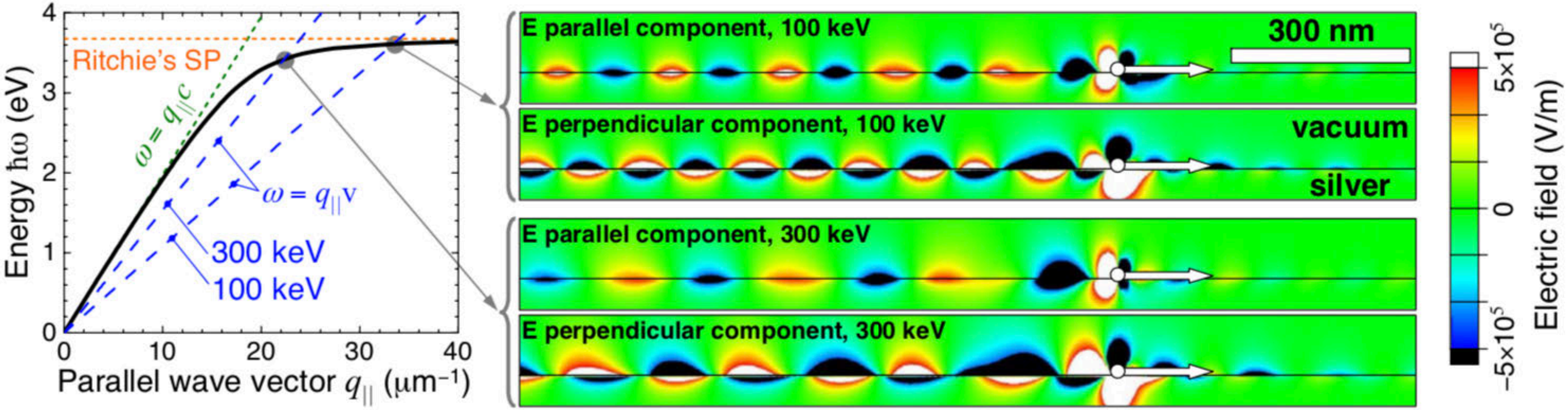


Figure for model Au/vac. from: Hugo Lourenço-Martins, PhD thesis

Surface plasmons(-polaritons)

- Semi-infinite interface: SPs are traveling waves with evanescent field in z direction: surface plasmon-polaritons (SPPs)
- SPPs: TM modes; cannot couple directly with light
- Modelling of excitation by fast e^-s moving parallel to Ag/vacuum interface at impact parameter of 10 nm:



F.J. García de Abajo "Optical excitations in electron microscopy," Rev. Mod. Phys. **82** (2010) 209–275

Localised surface plasmon resonances

- Confine SP on nanoparticle: set up a stationary standing wave
- Example: modelling of 400 x 30 nm Ag rod

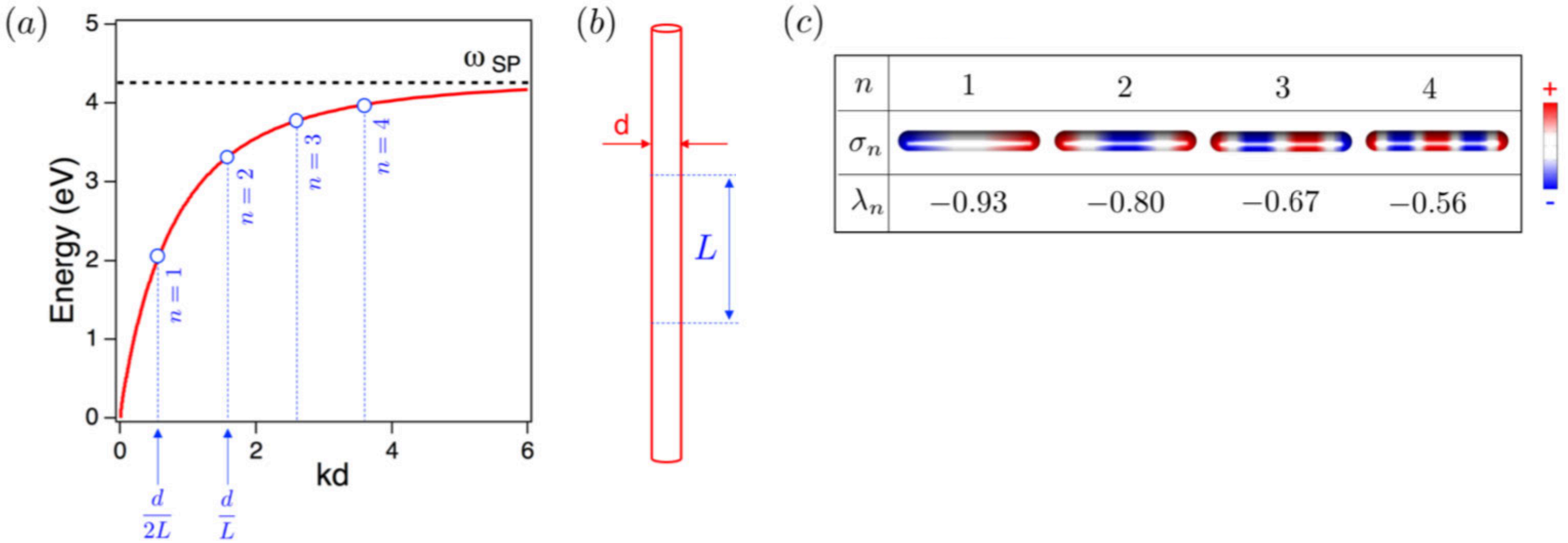


Figure from: Hugo Lourenço-Martins, PhD thesis

LSPRs on nanorod

- EELS measurements of 1000 x 50 nm Ag rod
- Strong EELS signal when $\vec{E}$ of probe excites LSPR with energy $\Delta E = hc/\lambda$

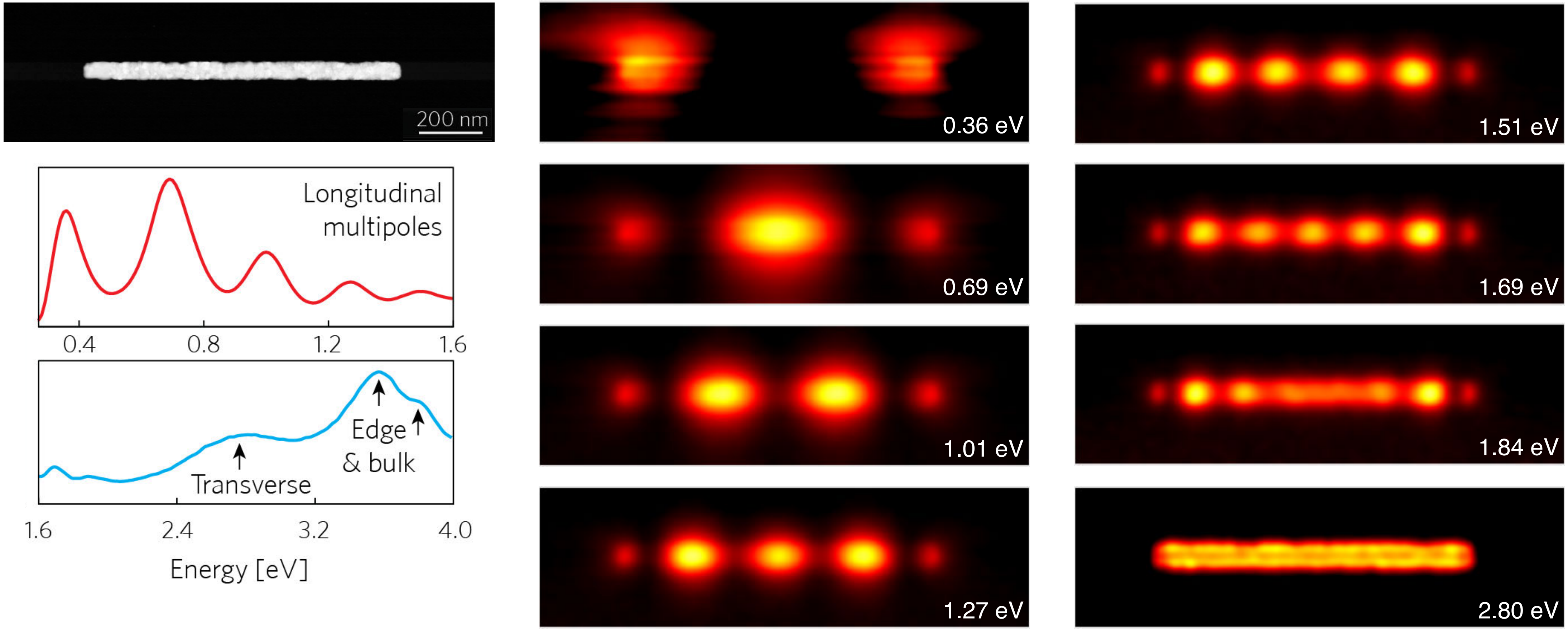


Figure by V. Flauraud and D.T.L. Alexander

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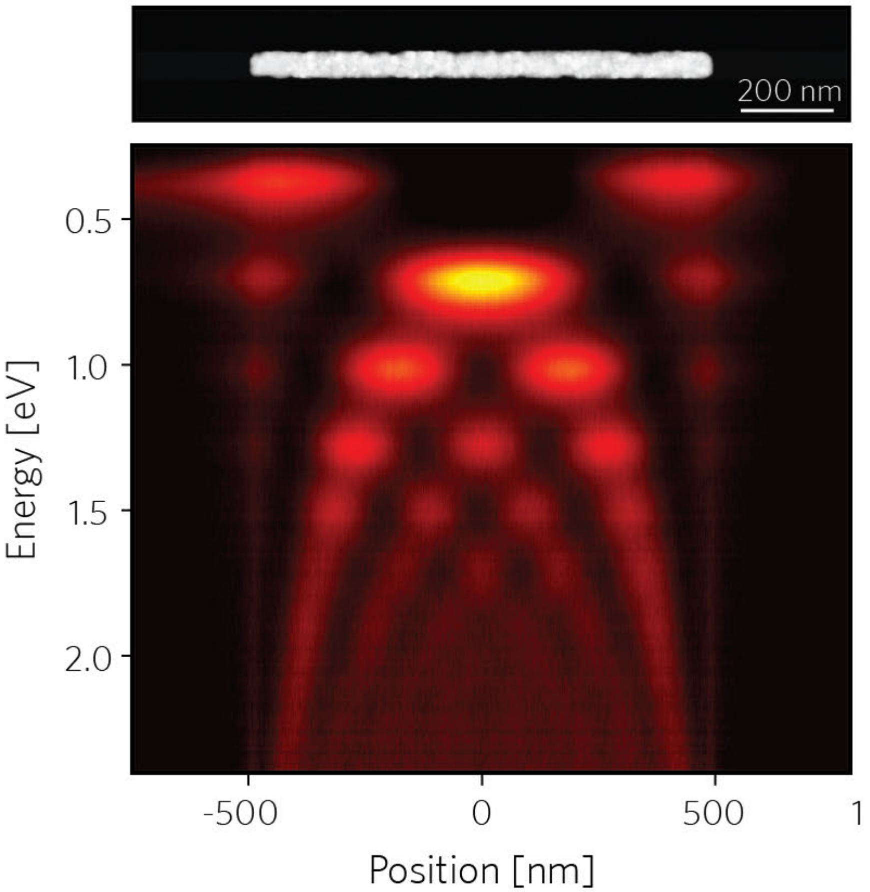
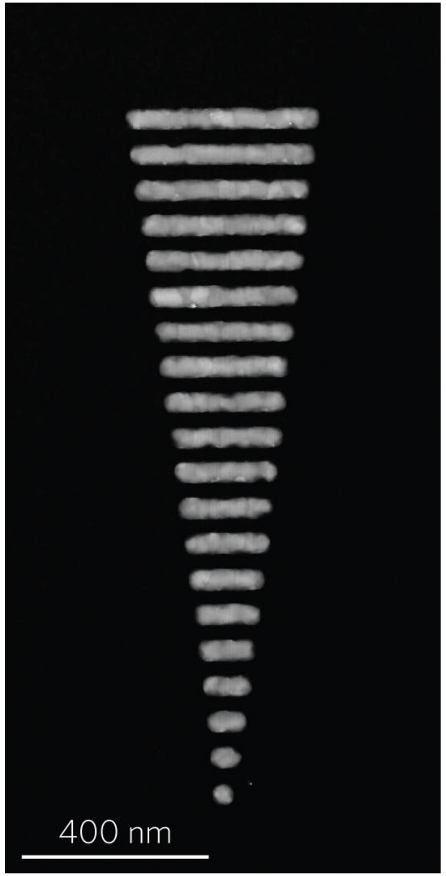
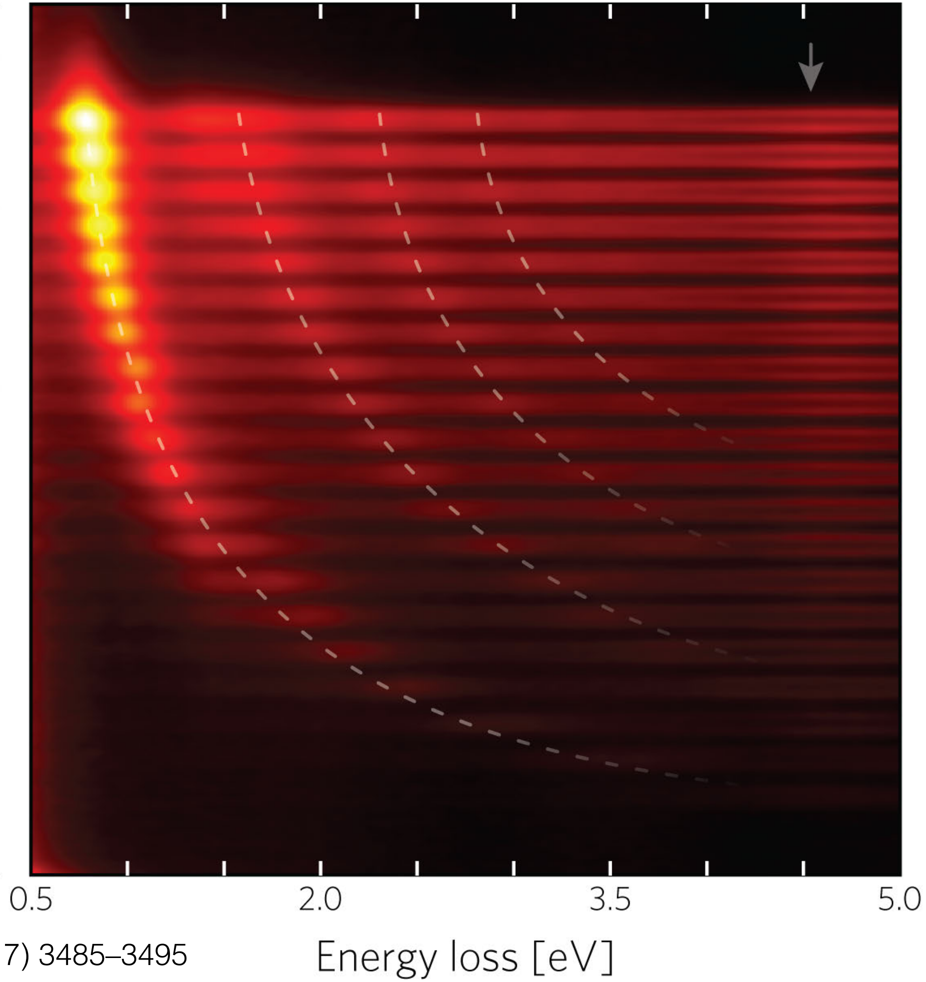


LSPRs on Al nanorods – dispersion curve

- Red-shift of resonance energies with increasing length of nanorod





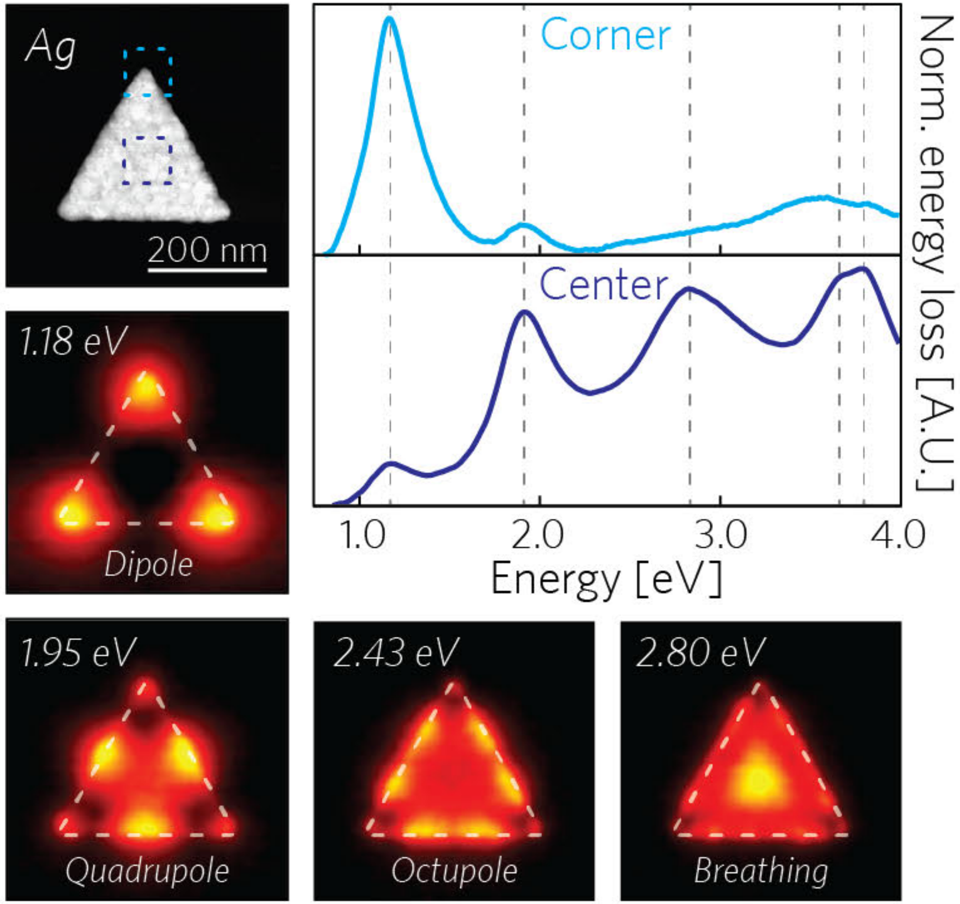
V. Flauraud et al. *ACS Nano* **11** (2017) 3485–3495

Rossow and Botton *PRL* **110** (2013) 066801

Mkhitaryan *Nano Lett.* **21** (2021) 2444–2452

LSPRs on nanotriangle

- EELS hotspots for nodes in multipolar edge modes



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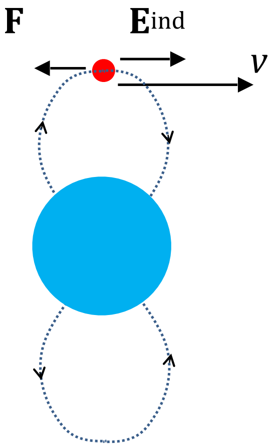
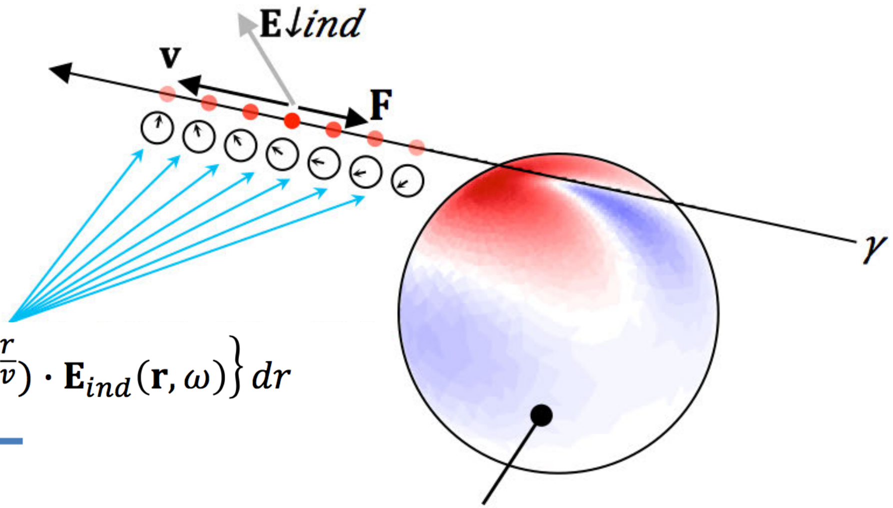
Classical: solve Maxwell's equations

- Take case of LSPR excitation on a metallic sphere
- Energy loss probability $\Gamma_{\mathrm{EELS}}(\omega)$ related to work done by Lorentz force acting back on fast transmitted e⁻ from $\vec{E}_{\mathrm{ind}}$

$$\Delta E = e \int dt\, \mathbf{v} \cdot \mathbf{E}^{\mathrm{ind}}[\mathbf{r}_e(t), t] = \int_0^\infty \hbar\omega\, d\omega\, \Gamma_{\mathrm{EELS}}(\omega)$$



$$\Gamma(\omega) = \frac{q}{\pi\hbar\omega v}\int_\gamma Re\left\{(\mathbf{v}e^{-i\omega\frac{r}{v}}) \cdot \mathbf{E}_{ind}(\mathbf{r},\omega)\right\} dr$$



Charge distribution at one frequency

Figure from: Gabriel Bernasconi, formerly NAM, EPFL

Classical approaches/numerical methods

- Analytical solutions: exist for restricted geometries (e.g. spheres).
- Discrete-dipole approximation (DDA): treat nanoparticle as discrete assembly of point dipoles which interact with each other. Time consuming.
- Finite difference time domain (FDTD) method: solve full Maxwell equations in temporal domain, throughout space (3D). Less accurate than BEM.
- Boundary element method (BEM): use boundary conditions to reformulate Maxwell equations as a boundary integral equation (BIE). Fast computation but sensitive to complex parameterisation.
- Surface integral equation (SIE) method: Green's function approach to solve the EM wave equation. Restriction to isotropic/homogeneous media but versatile surface discretisation. Developed for EELS by NAM, EPFL.
- Photonic local density of states (LDOS): context for "fundamental" interpretation

Boundary element method

- Considering LSPRs: in quasi-static limit of $kL \ll 1$ where L is nanoparticle size, retardation can be neglected and wave eq. reduces to Poisson's eq.:

$$\nabla^2 \phi(\boldsymbol{r}, \omega) = -4\pi\rho(\boldsymbol{r}, \omega)$$

- Applying boundary relations this reduces to boundary integral eq.:

$$\Lambda(\omega)(\boldsymbol{s}) = \mathcal{P} \oint_S F(\boldsymbol{s}, \boldsymbol{s}')\sigma(\boldsymbol{s}')$$

where have constant: $\Lambda(\omega) = 2\pi \frac{\epsilon_d(\omega) + \epsilon_m(\omega)}{\epsilon_d(\omega) - \epsilon_m(\omega)}$

and kernel $F(\boldsymbol{s}, \boldsymbol{s}') = \frac{-\boldsymbol{n}(\boldsymbol{s}).(\boldsymbol{s} - \boldsymbol{s}')}{|\boldsymbol{s} - \boldsymbol{s}'|^3}$

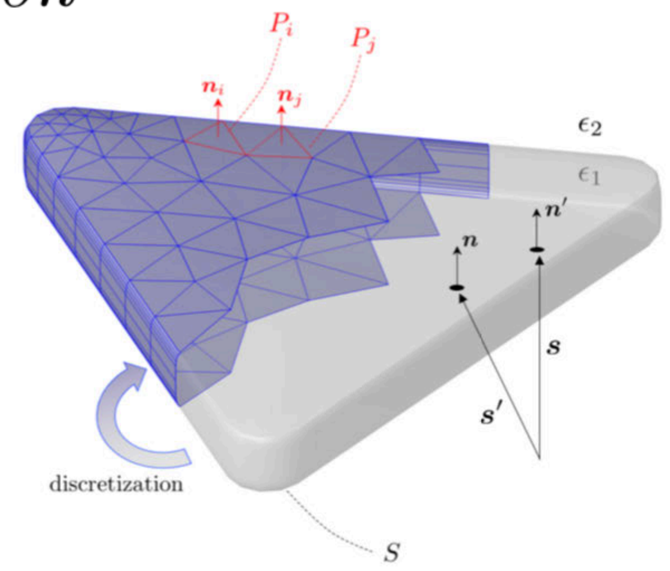
- Solutions that form infinite set $\{\lambda_n, \sigma_n\}_{n\in\mathbb{N}}$ and are solutions of eigenproblem

Boundary element method

- Boundary element method (BEM) extends this by applying EM perturbation described by scalar potential ϕ^{ext} giving:

$$\Lambda(\omega)\sigma(\boldsymbol{s},\omega) = \mathcal{P}\oint_S F(\boldsymbol{s},\boldsymbol{s}')\sigma(\boldsymbol{s}',\omega)d\boldsymbol{s}' + \frac{\partial\phi^{\text{ext}}(\boldsymbol{s}',\omega)}{\partial\boldsymbol{n}}$$

- Calculations by discretising surface and applying appropriate parameterisation

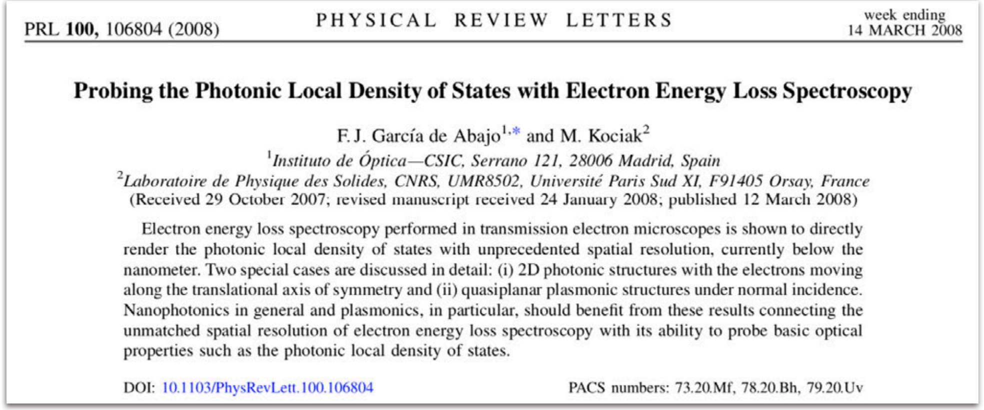


- Popular tool for applying BEM is MNPBEM toolbox by Hohenester and Trügler https://physik.uni-graz.at/de/mnpbem/

Figure from: Hugo Lourenço-Martins, PhD thesis

Photonic local density of states

- García de Abajo and Kociak showed EELS related to full photonic local density of states (LDOS) of a nanostructure, including radiative & non-radiative components



PRL **100,** 106804 (2008) PHYSICAL REVIEW LETTERS week ending 14 MARCH 2008

Probing the Photonic Local Density of States with Electron Energy Loss Spectroscopy

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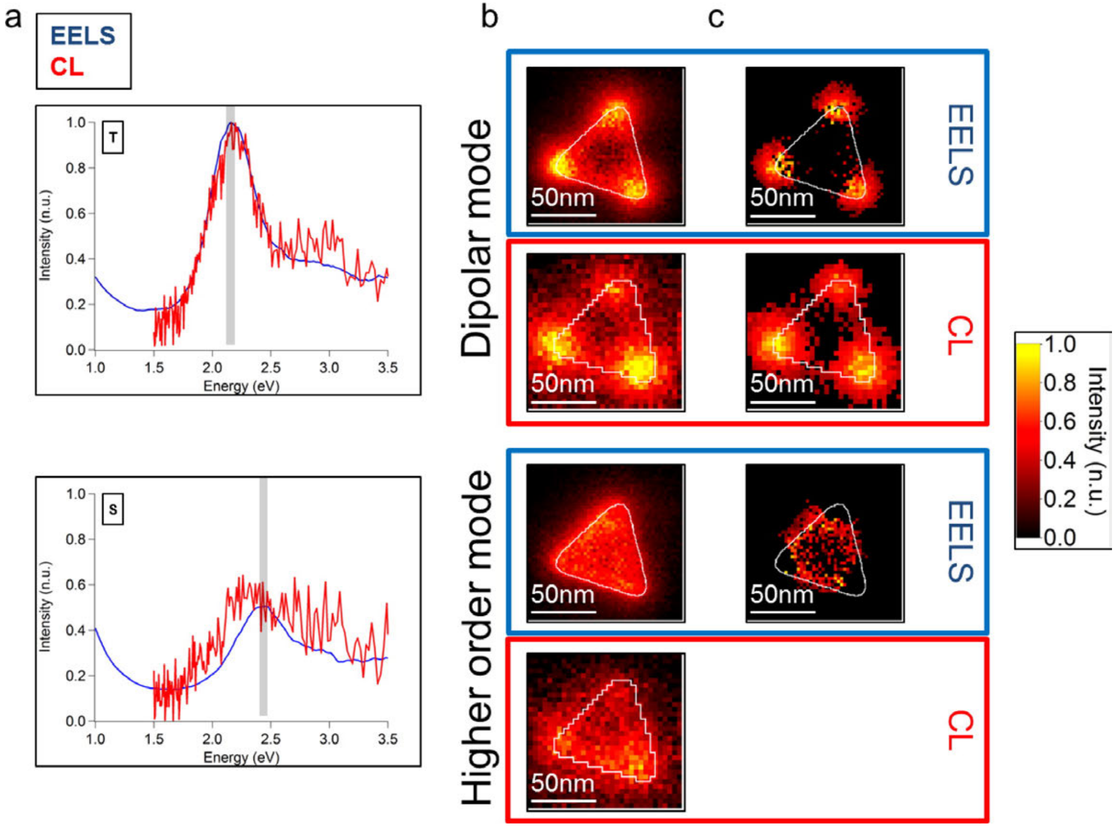
[2]*Laboratoire de Physique des Solides, CNRS, UMR8502, Université Paris Sud XI, F91405 Orsay, France*
(Received 29 October 2007; revised manuscript received 24 January 2008; published 12 March 2008)

Electron energy loss spectroscopy performed in transmission electron microscopes is shown to directly render the photonic local density of states with unprecedented spatial resolution, currently below the nanometer. Two special cases are discussed in detail: (i) 2D photonic structures with the electrons moving along the translational axis of symmetry and (ii) quasiplanar plasmonic structures under normal incidence. Nanophotonics in general and plasmonics, in particular, should benefit from these results connecting the unmatched spatial resolution of electron energy loss spectroscopy with its ability to probe basic optical properties such as the photonic local density of states.

DOI: 10.1103/PhysRevLett.100.106804 PACS numbers: 73.20.Mf, 78.20.Bh, 79.20.Uv

- Used in interpretation of correlated EELS and cathodoluminescence (CL) spectra; CL related only to radiative LDOS



STEM-CL and EELS maps of Ag nanotriangle:
A. Losquin et al. Nano Lett. **15** (2015) 1229

Photonic local density of states

- *However:* Hohenester et al. showed that, while they are intimately related, there is no direct link between EELS and LDOS maps.

PRL **103,** 106801 (2009) PHYSICAL REVIEW LETTERS week ending 4 SEPTEMBER 2009

Electron-Energy-Loss Spectra of Plasmonic Nanoparticles

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(Received 21 January 2009; revised manuscript received 21 July 2009; published 31 August 2009)

We investigate electron-energy-loss spectroscopy (EELS) on metallic nanoparticles, through simulations, and provide a comprehensive comparison between EELS and the photonic local density of states (LDOS). Most importantly, we show that there is no direct link between EELS and LDOS maps, and that EELS can even be blind to *hot spots* in the gap between coupled nanoparticles. Although intimately related, the two quantities provide complementary information. This finding is in marked contrast to recently reported results.

DOI: 10.1103/PhysRevLett.103.106801 PACS numbers: 73.20.Mf, 78.20.Bh, 79.20.Uv

- Importantly: EELS probes only component of LDOS parallel to e⁻ trajectory – scalar product of e⁻ velocity and $\vec{\boldsymbol{E}}_{\text{ind}}$
- This can make it blind to EM hotspots between coupled nanoparticles

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Coupling of plasmonic nanoparticles

- Example: coupling of Au nanorods with different separations
- Unlike illumination using planewave EM radiation:

  – observe longitudinal anti-bonding mode; a *dark* mode

  – do not observe strong excitation of longitudinal bonding mode in the gap

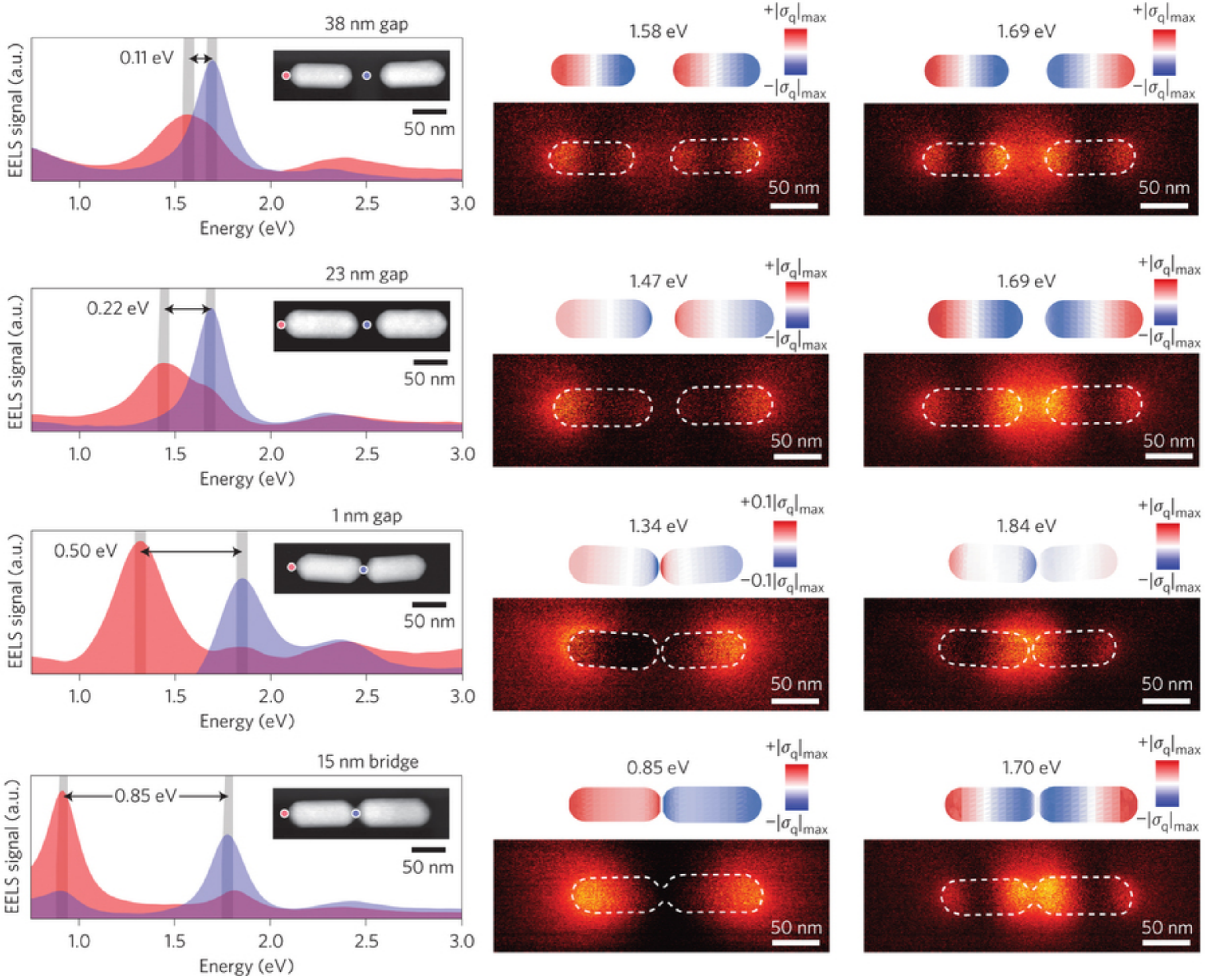
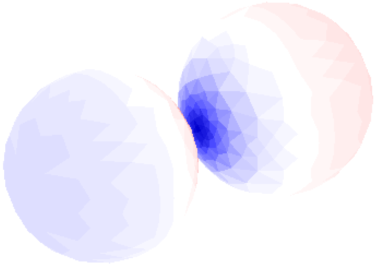


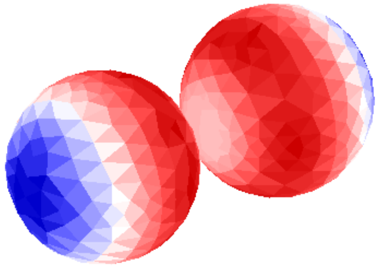
Figure from: V. Flauraud et al. Nature Nano **12** (2017) 73–80

EELS vs planewave excitation

- Planewave light: monochromatic and spatially extended
- Fast e^-: strongly localised, radially-symmetric electromagnetic field with broad spectrum $\Longrightarrow$ near-field imaging over broad frequency range
- Consider possibility to excite different resonances/charge distributions from a dimer pair of small plasmonic spheres with different geometries of incident field:



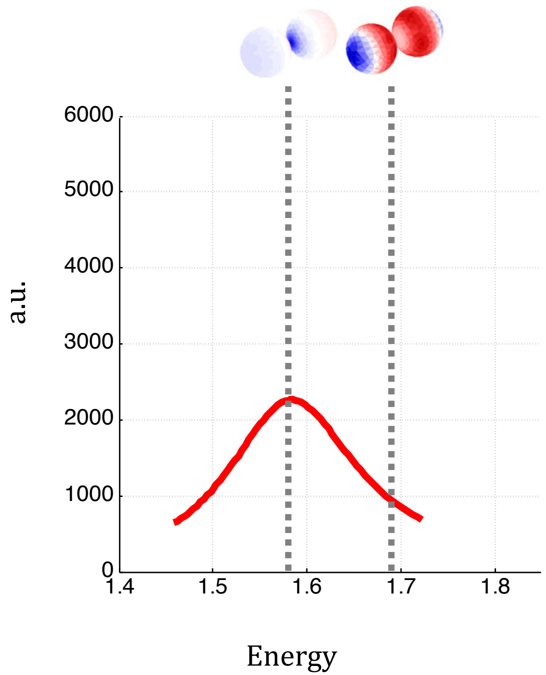
Longitudinal bonding

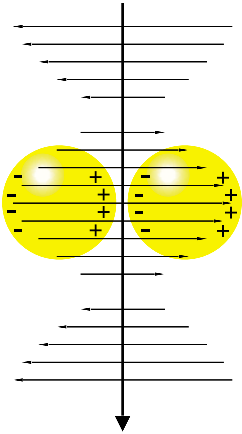


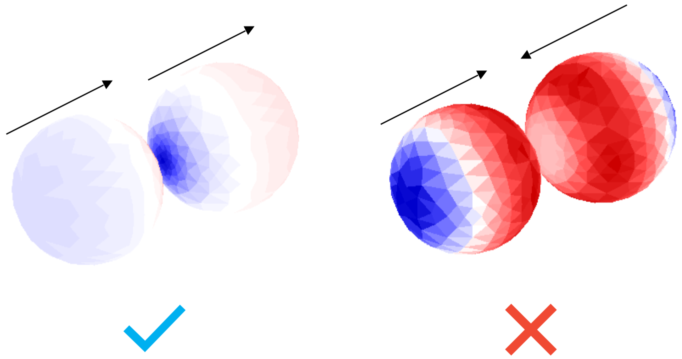
Longitudinal antibonding

Figures courtesy of Gabriel D. Bernasconi
Formerly NAM, EPFL

Planewave excitation

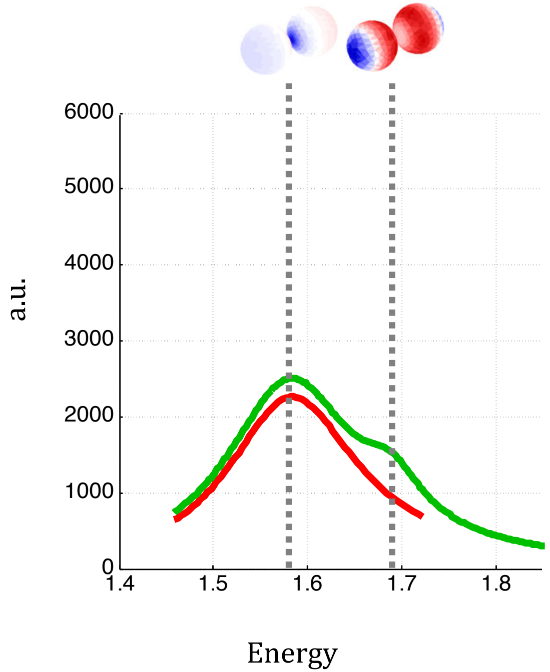


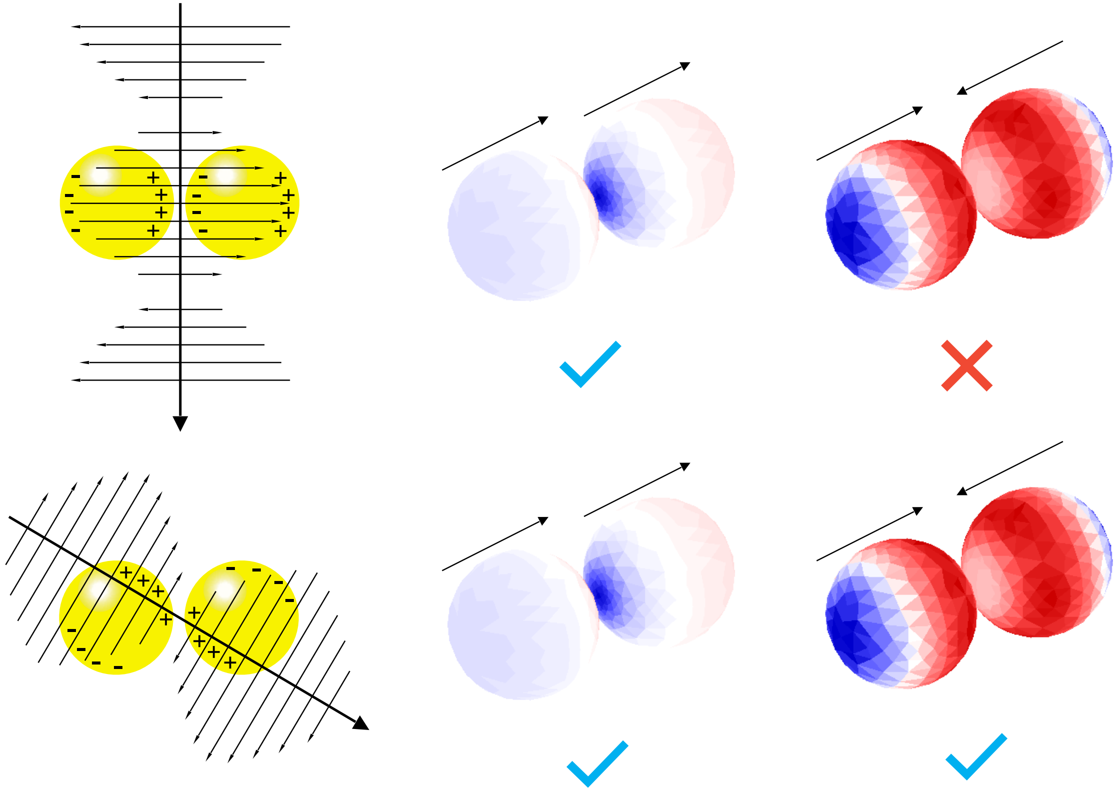




Figures courtesy of Gabriel D. Bernasconi
Formerly NAM, EPFL

Planewave excitation





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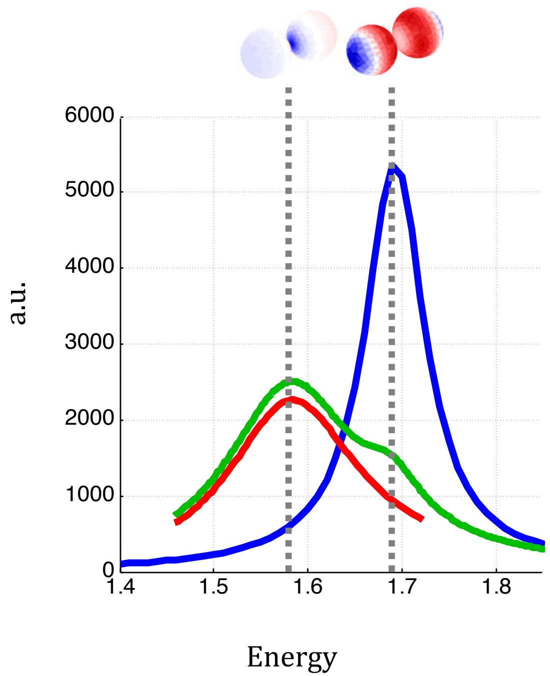
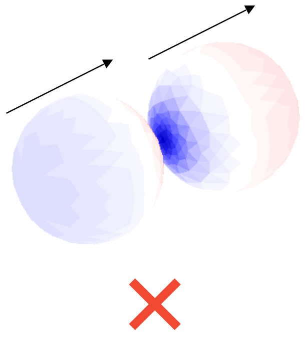
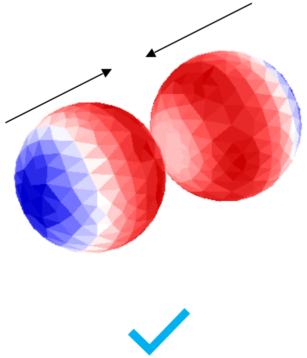
Electron excitation

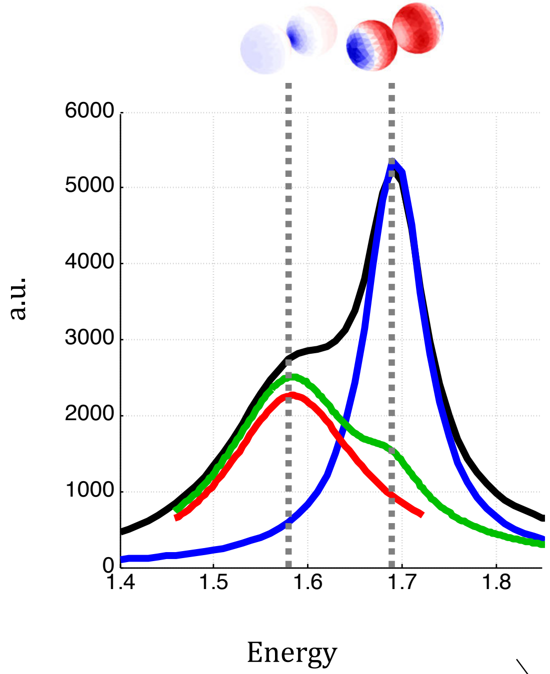


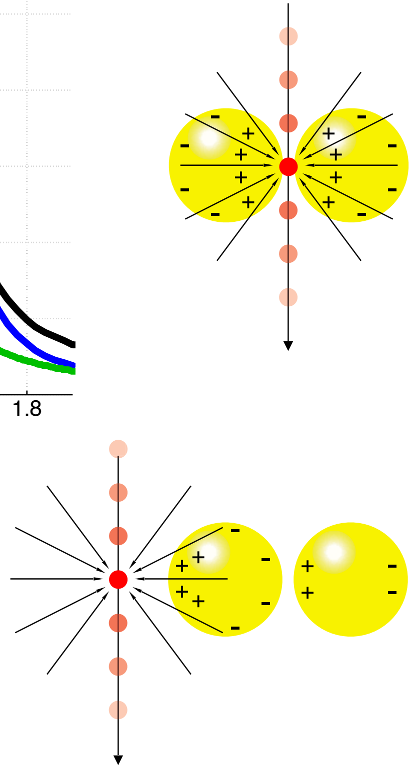


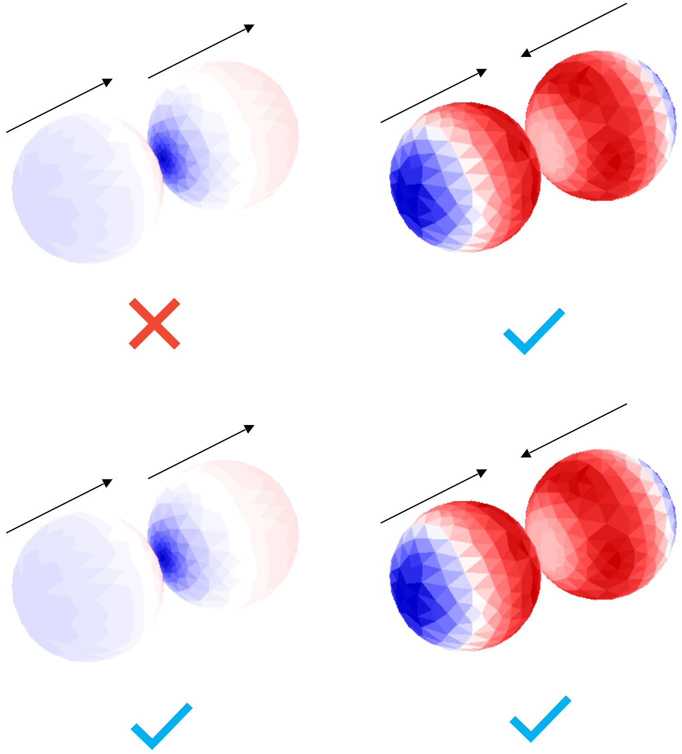


Figures courtesy of Gabriel D. Bernasconi
Formerly NAM, EPFL

Electron excitation







Figures courtesy of Gabriel D. Bernasconi
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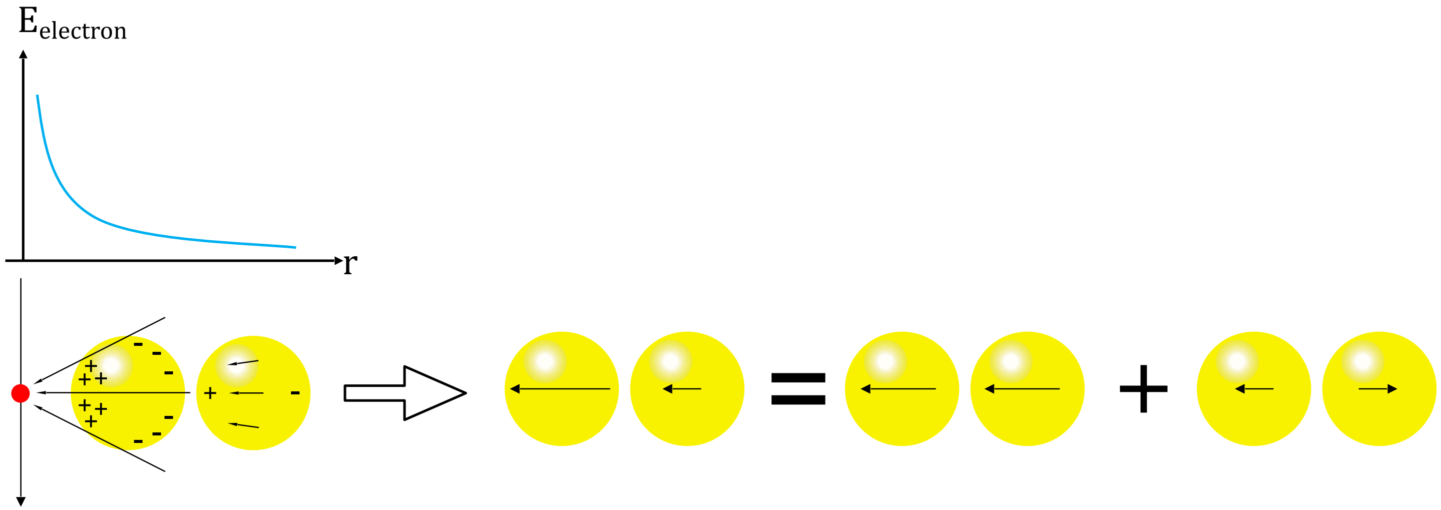
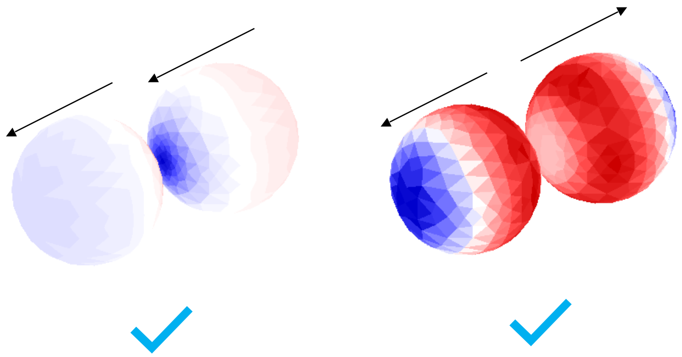
Electron excitation





Figures courtesy of Gabriel D. Bernasconi
Formerly NAM, EPFL

Electron excitation of 1 nm gap Au nano-rod dimer

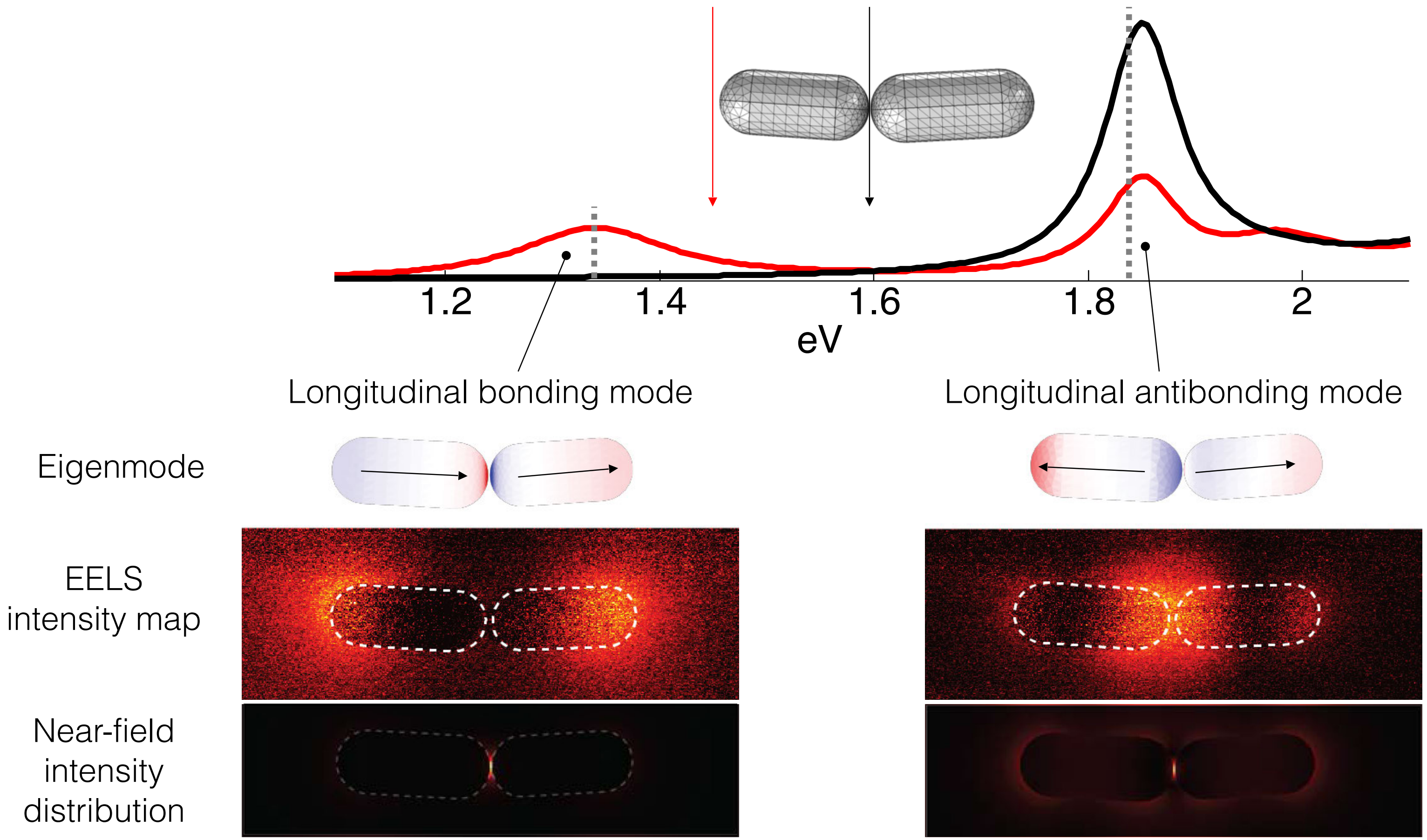


Figure by Gabriel D. Bernasconi et al. NAM, EPFL
EELS data: V. Flauraud et al. Nature Nano **12** (2017) 73–80

Electron excitation of 1 nm gap Au nano-rod dimer

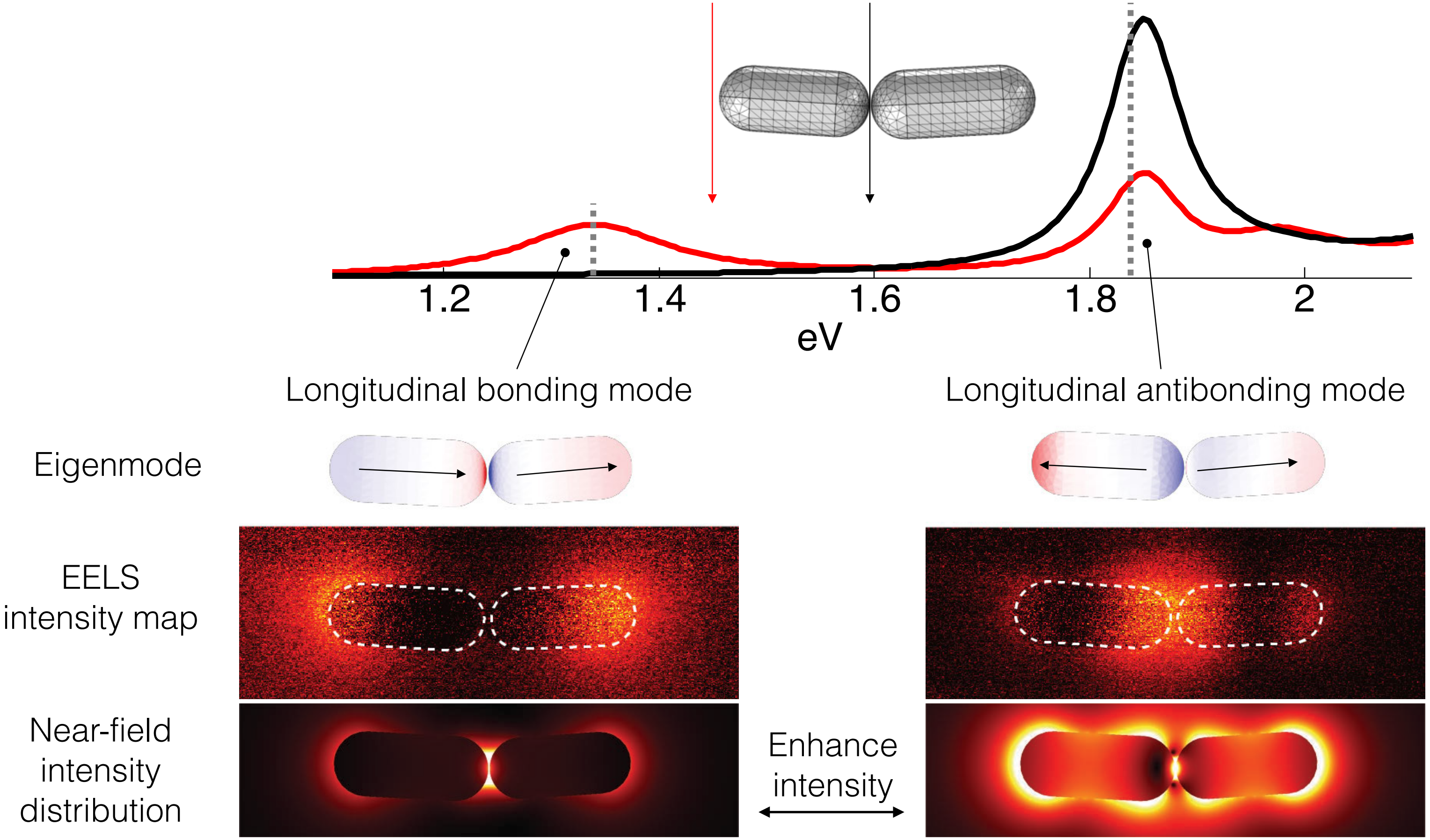


Figure by Gabriel D. Bernasconi et al. NAM, EPFL
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Electron excitation of 1 nm gap Au nano-rod dimer

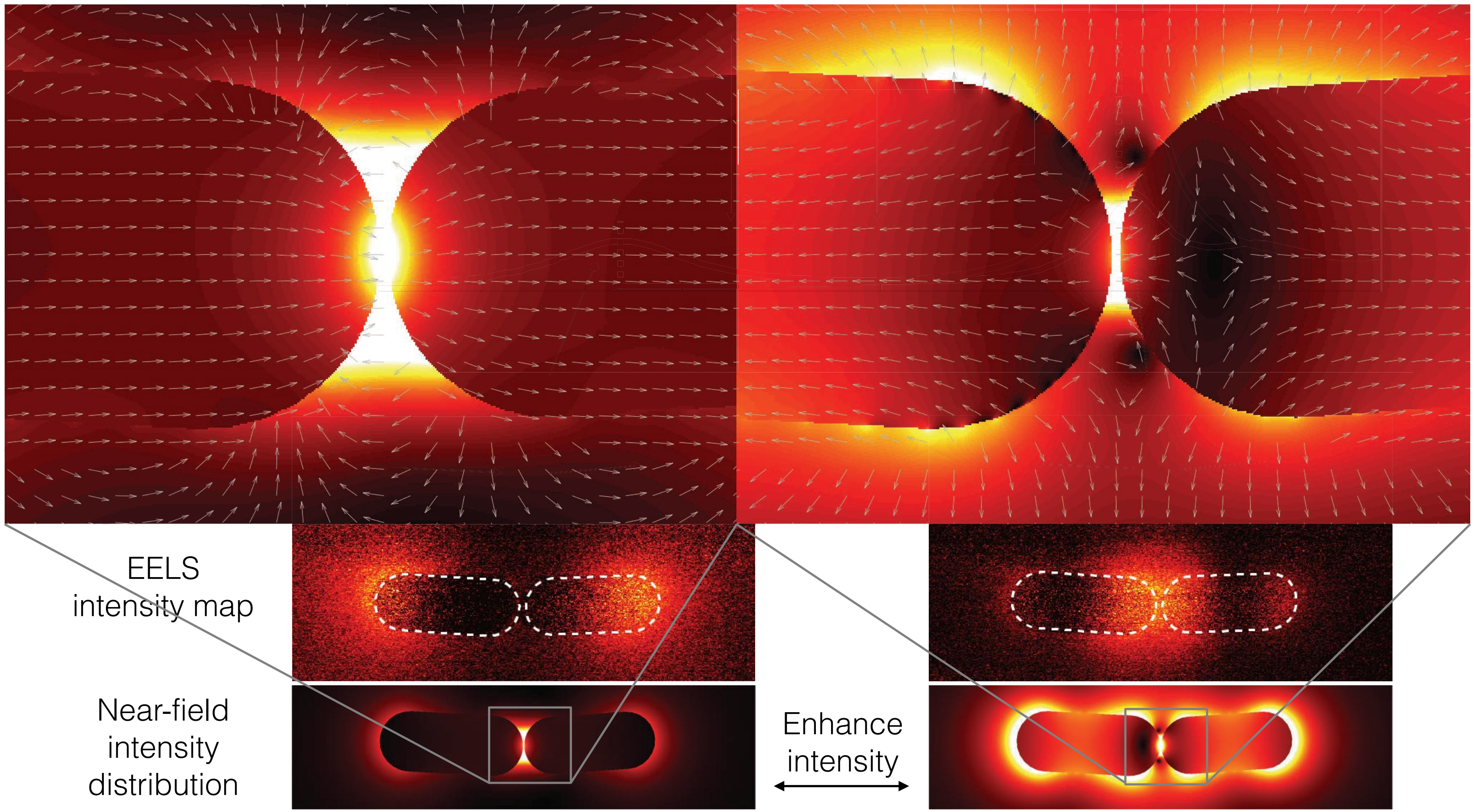
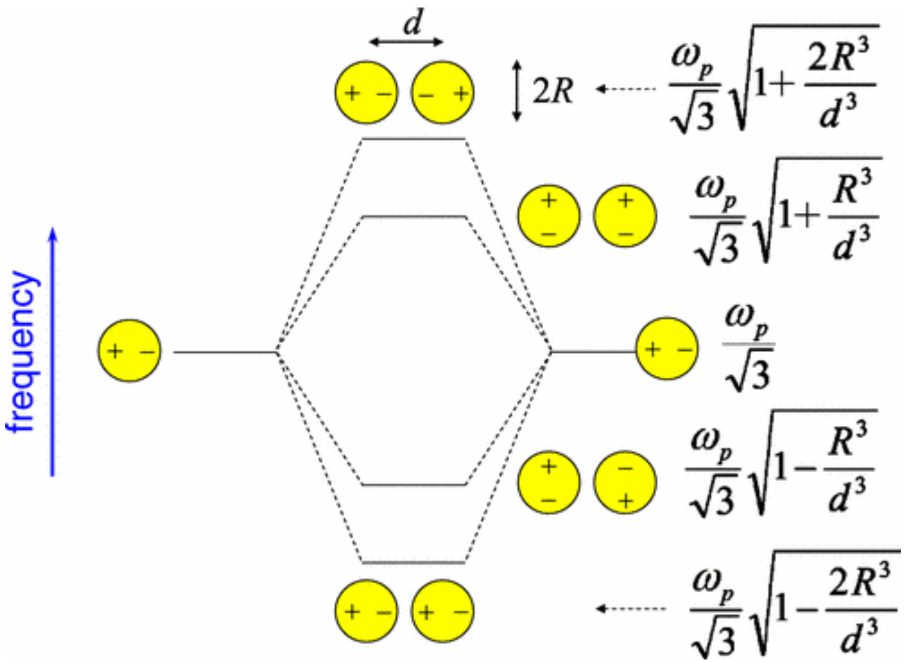


Figure by Gabriel D. Bernasconi et al. NAM, EPFL
EELS data: V. Flauraud et al. Nature Nano **12** (2017) 73–80

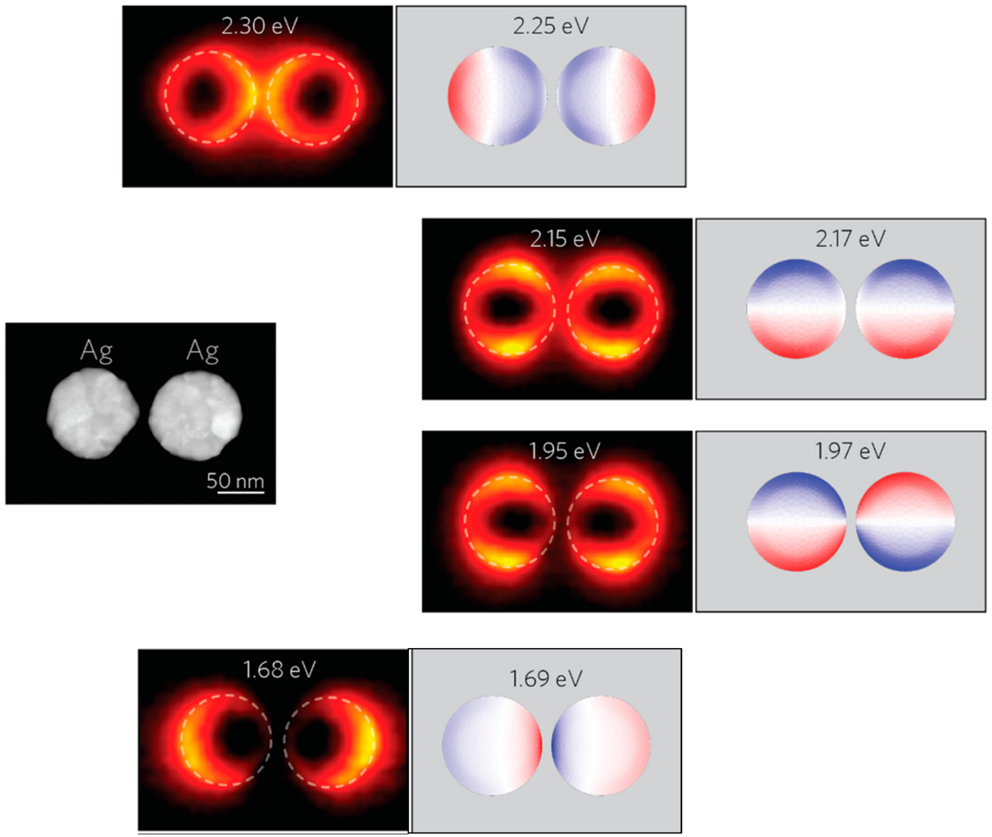
Dimer hybridisation

- From one dipole mode, dimer hybridisation of a radially symmetric nps $\Longrightarrow$ 4 modes
  EELS permits measurement of all of them

- Model (left): interacting spheres
- EELS data (right): Ag discs



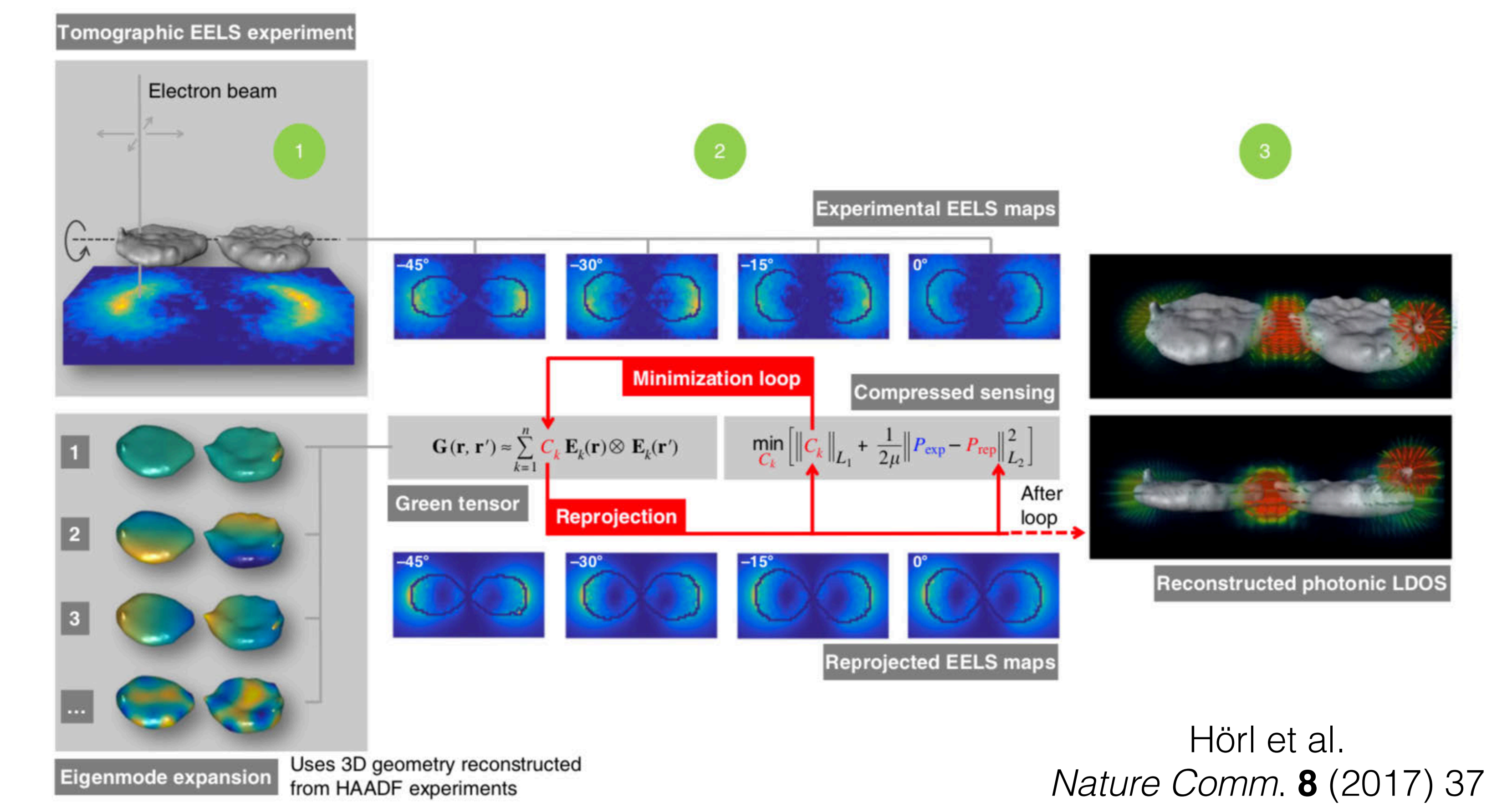
S. Riikonen et al. *PRB* **71** (2005) 235104



V. Flauraud et al. *ACS Nano* **11** (2017) 3485–3495

3D imaging of bonding mode

- Hörl et al. developed tomographic approach to imaging photonic environment of plasmonic nanoparticles through solution of an inverse problem

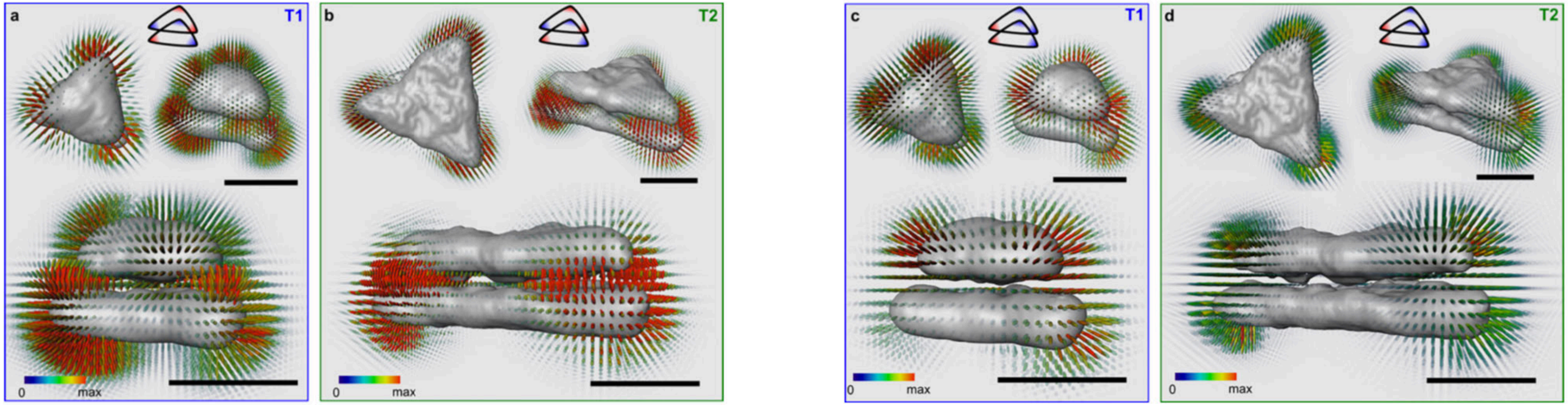


Hörl et al.
Nature Comm. **8** (2017) 37

3D imaging of bonding mode

- Applied to vertically-stacked dimer pair has allowed the gap bonding mode to be directly visualised:

Bonding modes | Anti-bonding modes



Haberfehlner et al. *Nano Lett.* **17** (2017) 6773–6777

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- Kramers-Kronig analysis and band gap measurements in visible light regime
- An evanescent probe
- Introduction to localised surface plasmon resonances (LSPRs) in EELS
- Theoretical approaches and numerical methods for EELS LSPR analysis
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- Plasmon damping; scattering and absorption
- Dielectrics and bound-states-in-the-continuum

Plasmon damping

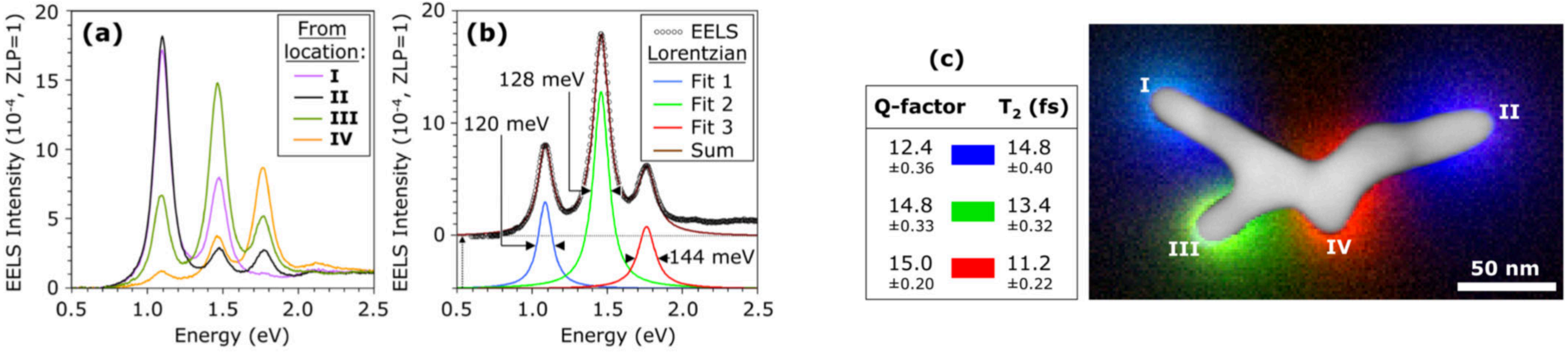
- LSPRs lose energy via:
  - scattering radiation to far-field (as could be measured by CL)
  - non-radiative channels, i.e. absorption, leading to thermal dissipation
- Decay time on the order of 10s of femtoseconds
- Can describe with damped oscillator model

Figure from: Gabriel Bernasconi, PhD thesis

Plasmon damping

- By carefully measuring width of plasmon peaks in EELS, Bosman et al. were able to measure the Q-factor and dephasing times T_2 for damping of different resonances in Au nanostructures:

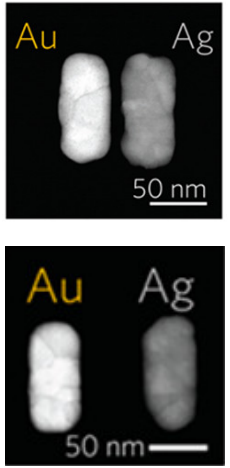
$$Q = \frac{E_p}{\Gamma}$$



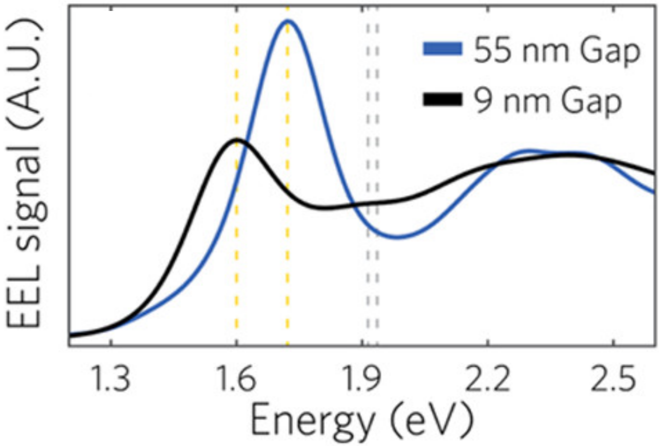
Bosman et al. *Sci. Rep.* **3** (2013) 1312

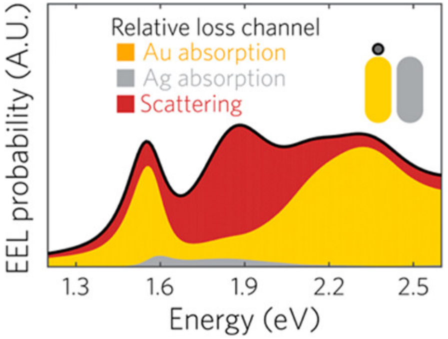
Antenna theory for scattering & absorption

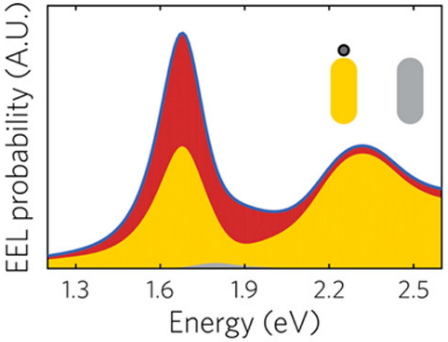
- Bernasconi et al. introduced alternative antenna-based approach to EELS
- By coupling energy conservation law with full-wave EM computations based on a SIE method, derive EEL spectra directly from dissipative processes of scattering & absorption
- Reveals interplay between absorption and scattering; i.e. where energy goes



Integrated loss over Au nanorod:







Bernasconi et al. *ACS Photonics* **4** (2017) 156–164; V. Flauraud et al. *ACS Nano* **11** (2017) 3485–3495

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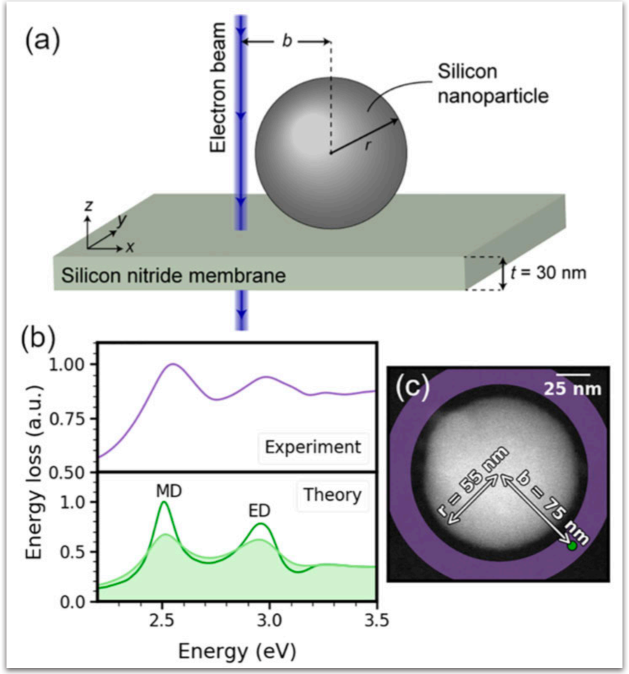
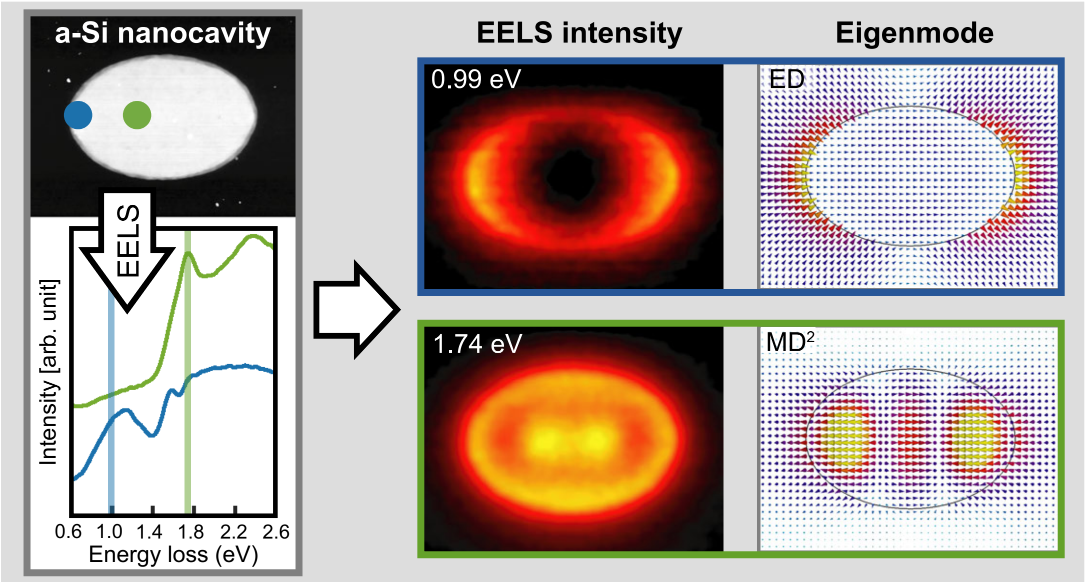
Dielectric nanostructures

- Dielectric nanostructures that support Mie-type resonances also of interest for nanophotonics applications
- Resonances correspond to charge displacement currents in particle volume
- Resonances can be excited by e⁻ probe and measured by EELS analogously to LSPRs

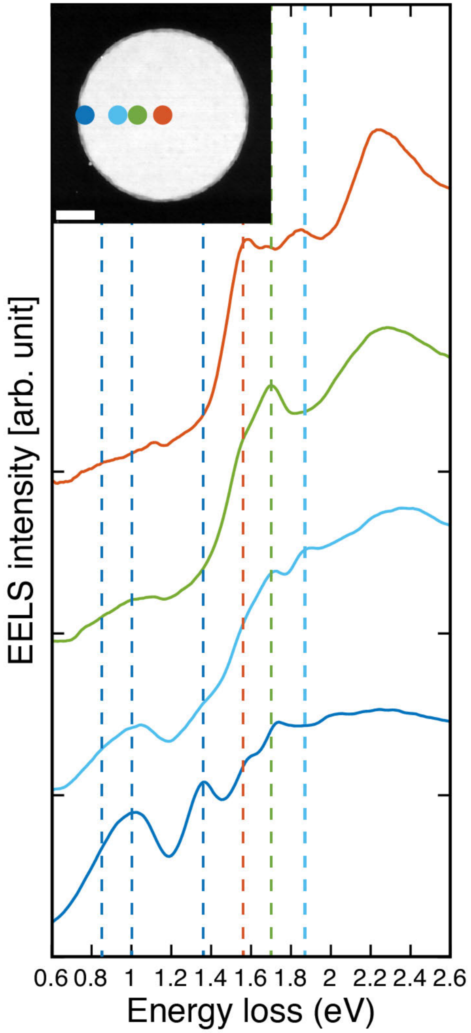


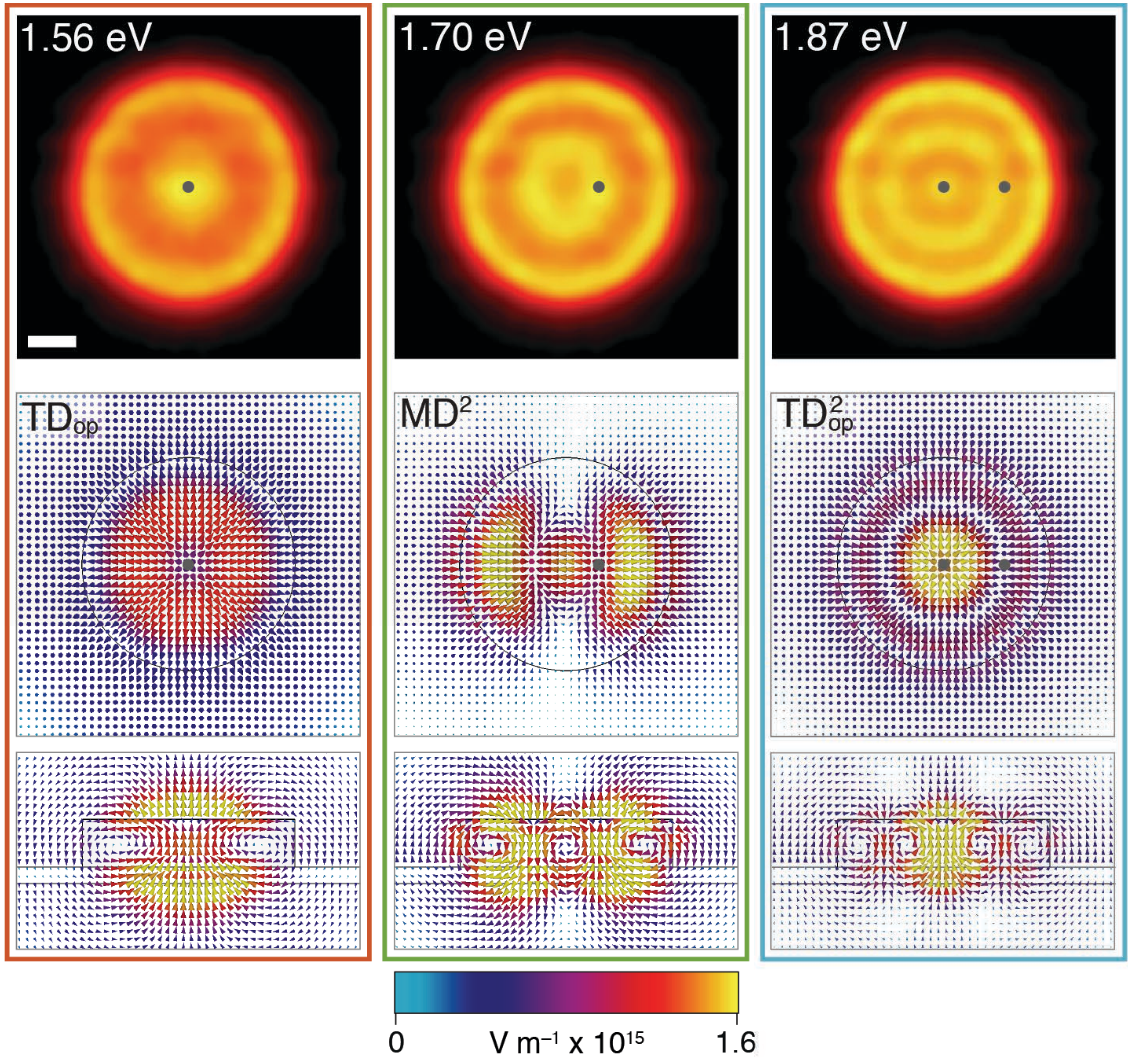
Assadillayev et al. *ACS Photonics* **8** (2021) 1582



Alexander et al. *ACS Nano* **15** (2021) 16501

Dielectric nanostructures: 450 nm a-Si disc

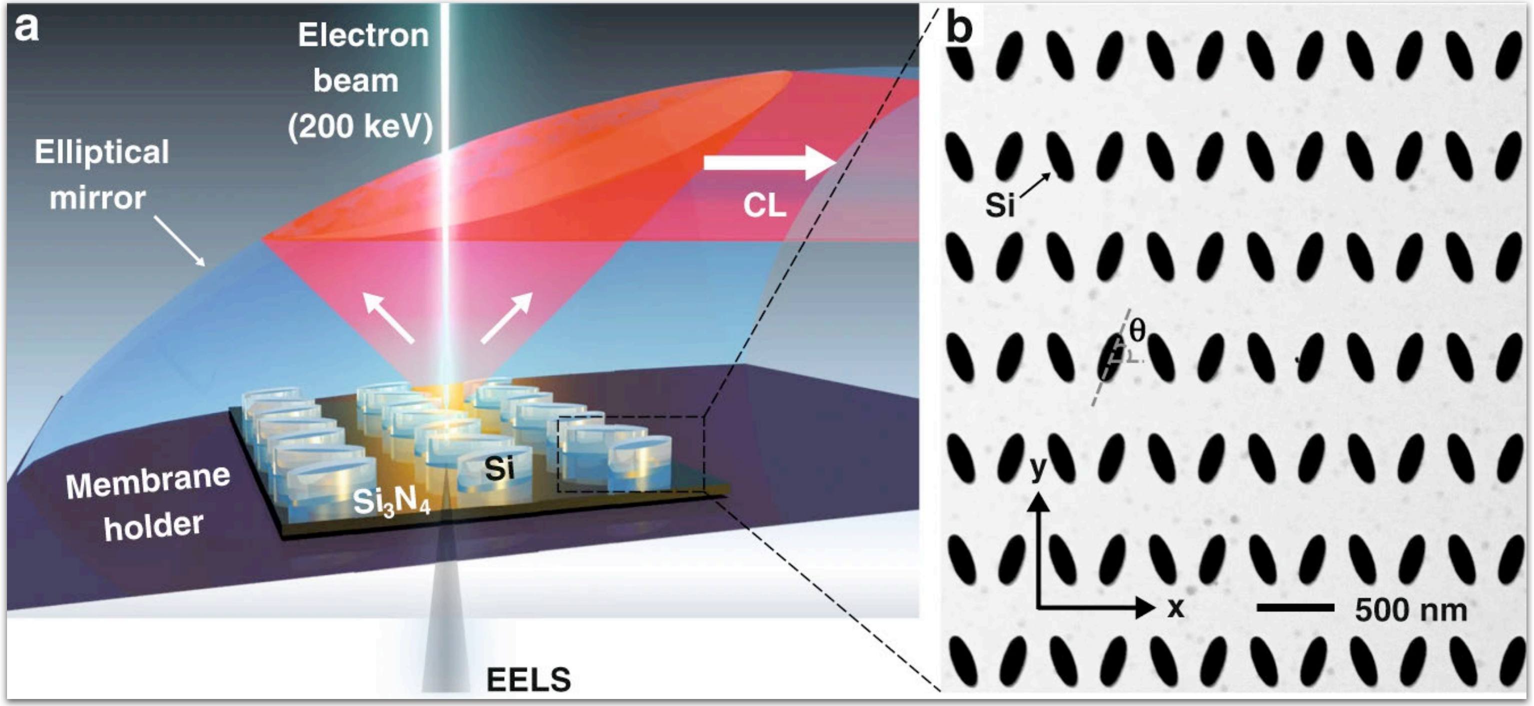




Alexander et al. *ACS Nano* **15** (2021) 16501

Bound-states-in-the-continuum (BIC)

- Optical modes with energies higher than potential wells that are still localised and bound in space ⟹ complete suppression of radiative loss in far-field
- Dong et al. studied BIC with EELS (near-field) and CL (far-field)

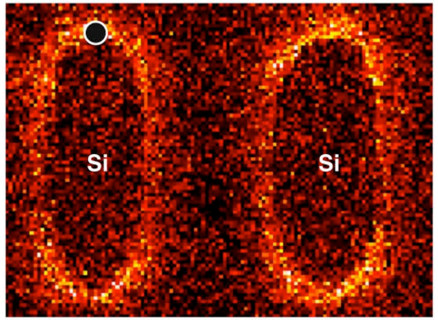


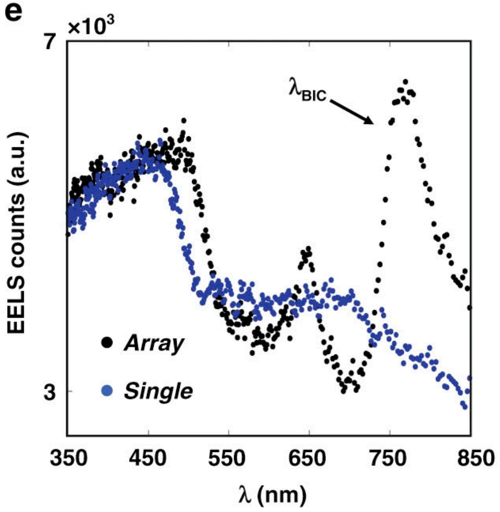
Si elliptic cylinder nanoantennas that support (quasi-) BIC modes

Dong et al. *Light: Science & Applications* **11** (2022) 20

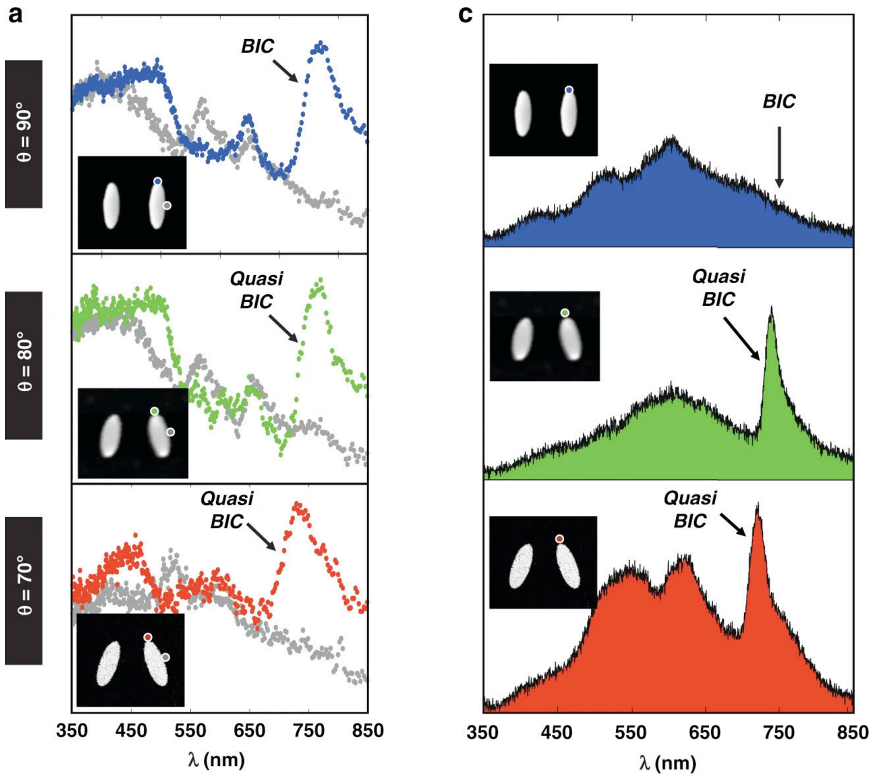
Bound-states-in-the-continuum (BIC)

- BIC excitation only seen for *array* of nanoantennas
- Tilted nanoantennas: BIC loses full confinement, some radiation escapes to far-field (Quasi BIC)





EELS



CL

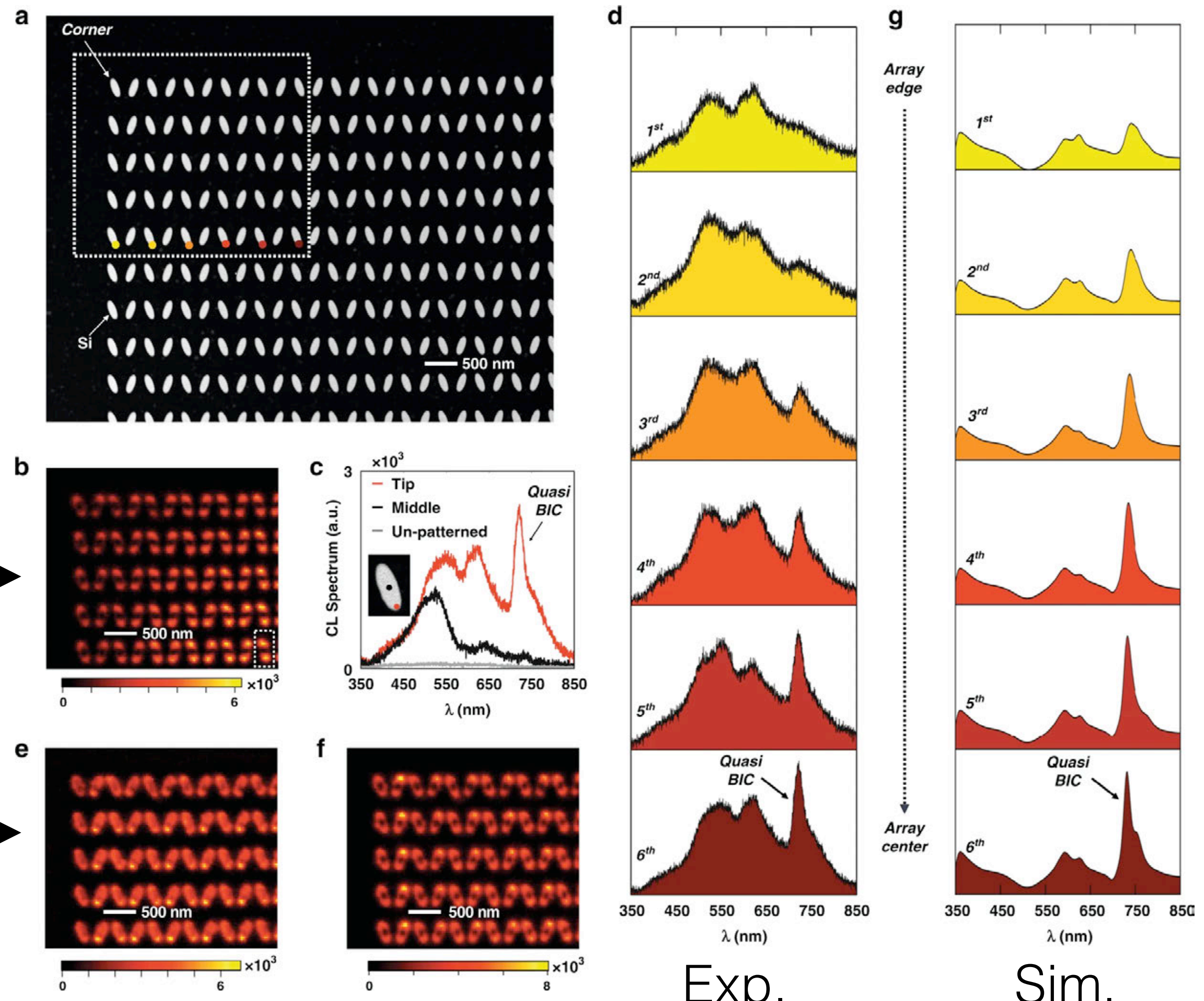
Dong et al. *Light: Science & Applications* **11** (2022) 20

Bound-states-in-the-continuum (BIC)

- Cathodoluminescence measurements across Quasi BIC array

Quasi BIC frequency →

522 nm and 614 nm Mie resonances →



Exp. Sim.

Dong et al. *Light: Science & Applications* **11** (2022) 20

Not discussed today…

- Relativistic quantum electrodynamic approach: Hugo Lourenço-Martins et al. SciPost Phys. **10** (2021) 031. E.g. for holography EELS and other phase-related experiments
- Probing LSPRs using phase-shaped electron beams. By matching wave function of e^--beam to plasmonic symmetry can selectively detect specific plasmon modes. HLM paper; Guzzinati et al. *Nat. Comm.* **8** (2017) 14999
- Study of excitations in dielectric photonic crystals. Conceptually: virtual broad band light from Cherenkov & transition radiation couples to photonic LDOS.
  García de Abajo et al. *PRL* **91** (2003) 143902, Cha et al. *PRB* **81** (2010) 113102 – TM modes measured in aloof geometry;
  Le Thomas et al. *PRB* **87** (2013) 155314 – cavity TE mode measured in bulk
- Incoherent radiation studied by EELS and STEM-CL – e.g. excitonic effects in EELS, radiation from e^--h^+ exciton recombination at quantum dots; quantum emitters; …