ETH
Eidgenössische Technische Hochschule Zürich
Swiss Federal Institute of Technology Zurich

Department of Information Technology and Electrical Engineering

MMP Simulation of Electron Energy Loss Spectroscopy Applied to Plasmonic Particles

Ueli Koch,
Jens Niegemann, Christian Hafner

kochue@ethz.ch

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Motivation

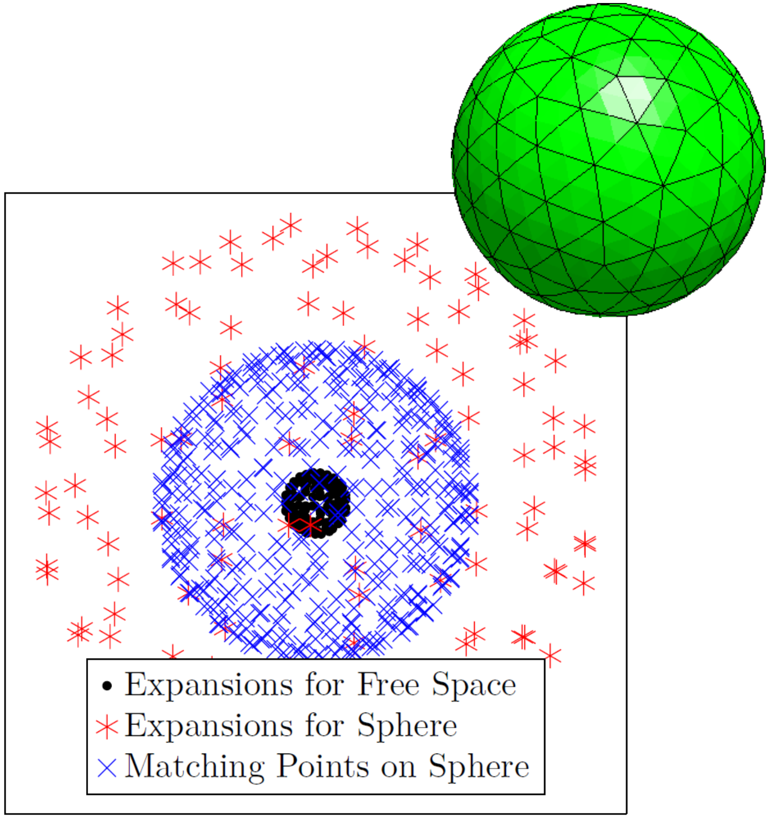
- Simulation of EELS experiments to characterize the resonances of plasmonic nanostructures
  - Spectrally (EELS spectra)
  - Spatially (EELS maps)

- Simulation with the Multiple Multipole Program (MMP)
  - Boundary discretization method to reduce memory requirements
  - Frequency domain to recycle major computations

Multiple Multipole Program (MMP)

- Electromagnetic fields are linear superpositions of multipoles (expansions)

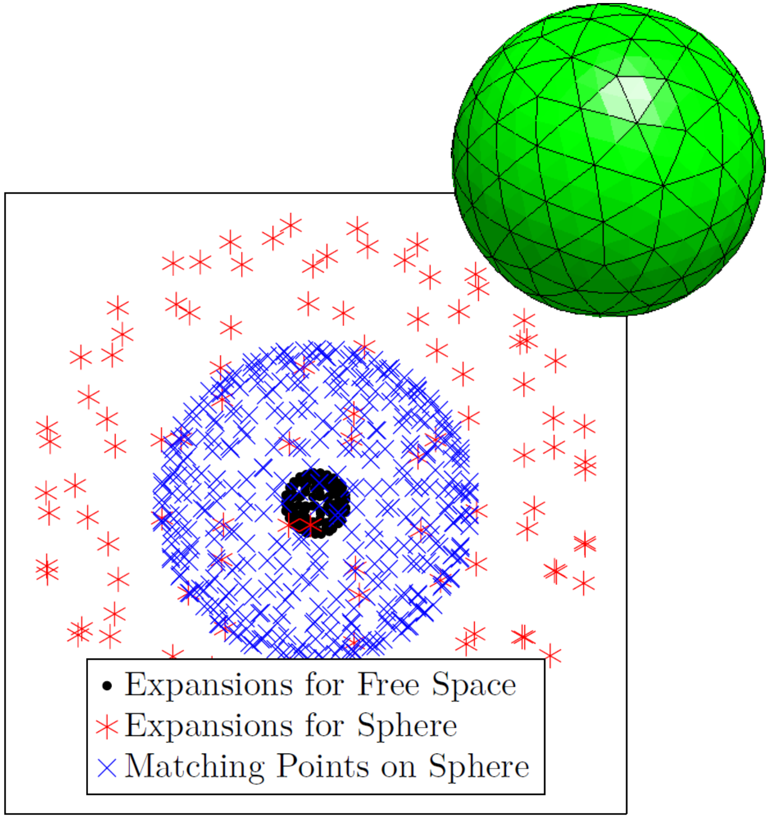
$$\vec{F}_{D_k} = \vec{F}_{ext} + \sum_{i \in \mathfrak{D}_k} \vec{F}_i$$



- Expansions
  - Fulfill Maxwell's equations
  - Fields written as linear equation
    $\vec{F} = \mathcal{A}_{e,r}\vec{x}_e$
  - Generic approach: dipoles shifted along normal direction

Multiple Multipole Program (MMP)

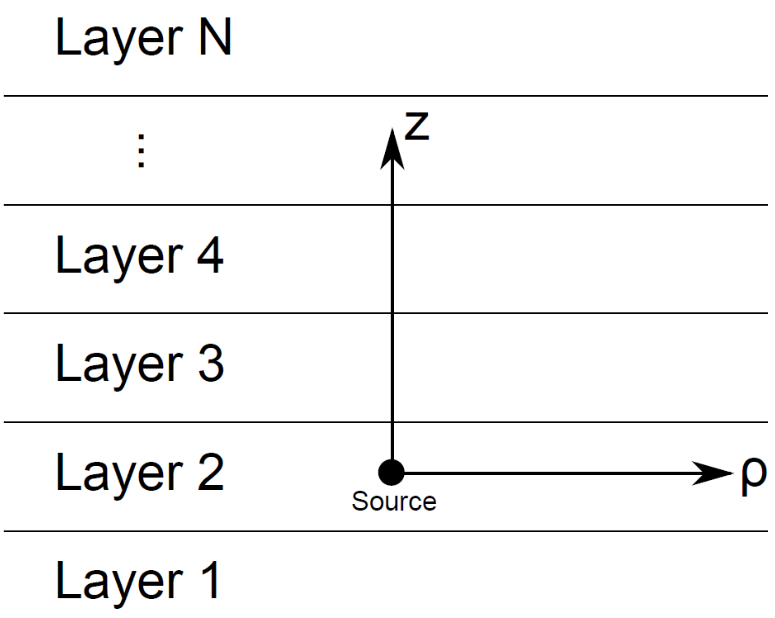
- Matching Points
  - Error minimization on particle boundaries
  - Try to fulfill boundary conditions at material interfaces
  - Generic approach: quadrature nodes on mesh elements



- Tomorrow:
  3D MMP simulations based on a FEM mesh

Layered Dipole

- Expansion which accounts for layered structure
  - Directly incorporates layers
  - Exact description of a dipole within layers
- Numerical impacts
  - No need for layer discretization
    - Reduces memory requirements
  - Requires numerical integrations
    - Increases computation time



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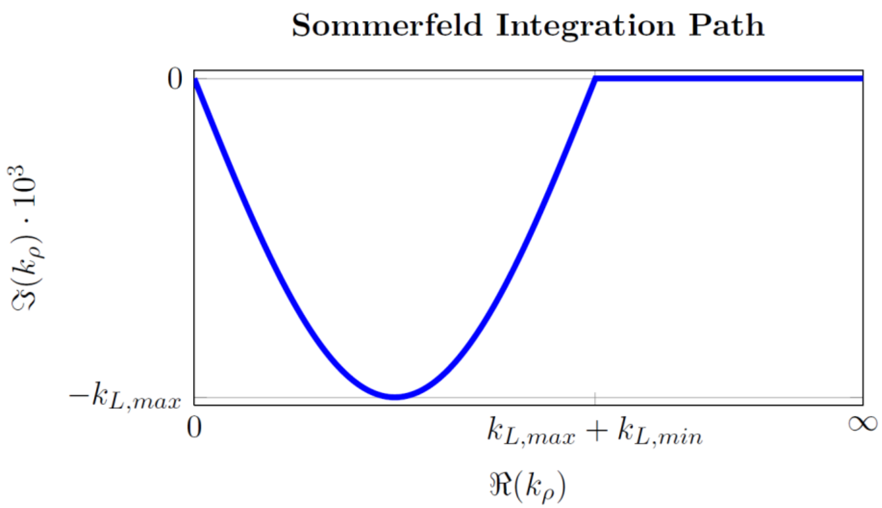
Layered Dipole

- 12 Sommerfeld integrals for each pair of expansion and observation point

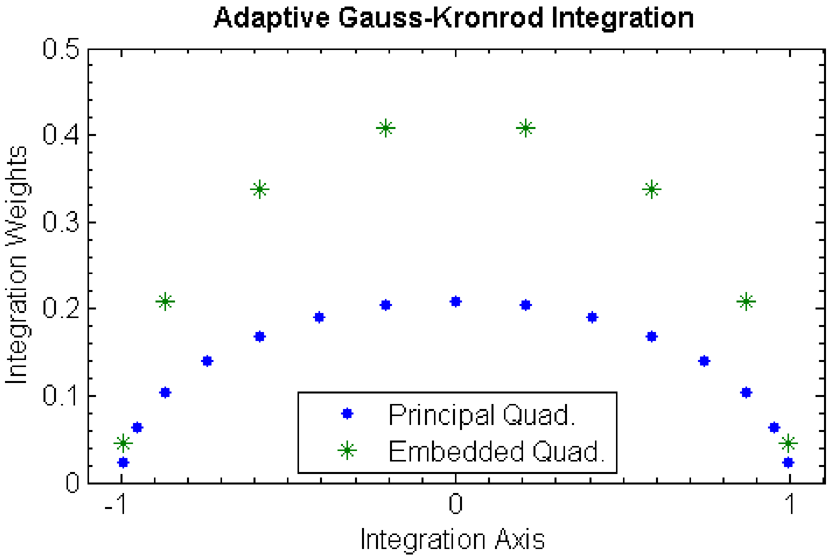
$$F_{ij} = \frac{1}{4\pi} \int_0^\infty G_{ij}^F \, J_0(k_\rho \rho) \, k_\rho \, \mathrm{d}k_\rho$$

  - Complex integration path
  - Adaptive integration / quadrature



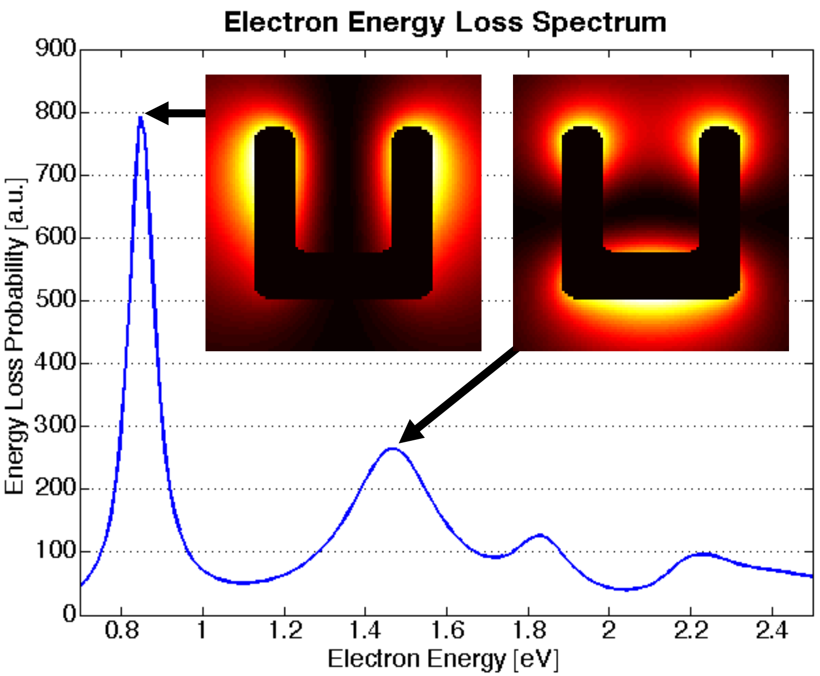
Adaptive Numerical Quadrature

- Split integration path into intervals
- Perform integration interval-wise using Gauss-Kronrod quadrature
- Estimate error within each interval
  - Low ➔ accept interval result
  - High ➔ split interval and recompute
- Repeat until convergence



Electron Energy Loss Spectroscopy (EELS)

- Excite nanoparticle with electron beam and measure electron energy loss
- Perform frequency sweeps at fixed positions to find bright and dark modes
- Generate EELS maps at fixed frequencies to find resonance patterns



Electron Energy Loss Spectroscopy

- Excite particle with electron beam

$$E_\rho = -\frac{2e\omega}{v^2\gamma\epsilon}\, e^{i\omega\frac{z}{v}}\, K_1\left(\frac{\omega\rho}{v\gamma}\right)$$

$$E_z = \frac{2e\omega}{v^2\gamma\epsilon}\, e^{i\omega\frac{z}{v}}\, K_0\left(\frac{\omega\rho}{v\gamma}\right)\frac{i}{\gamma}$$

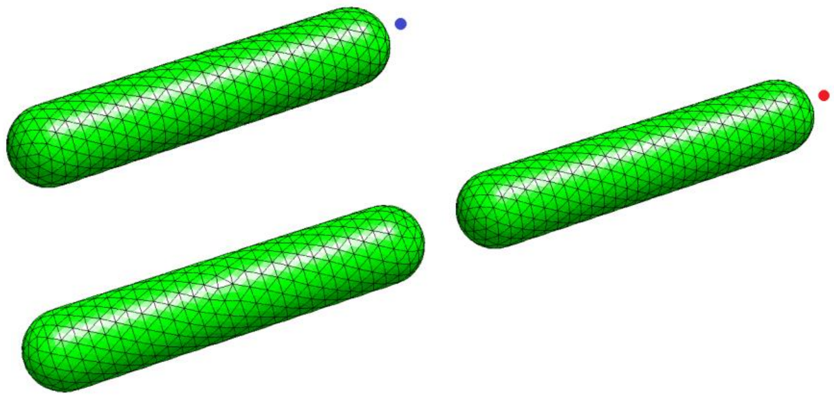
$$H_\phi = -\frac{2e\omega}{v\gamma}\, e^{i\omega\frac{z}{v}}\, K_1\left(\frac{\omega\rho}{v\gamma}\right)$$

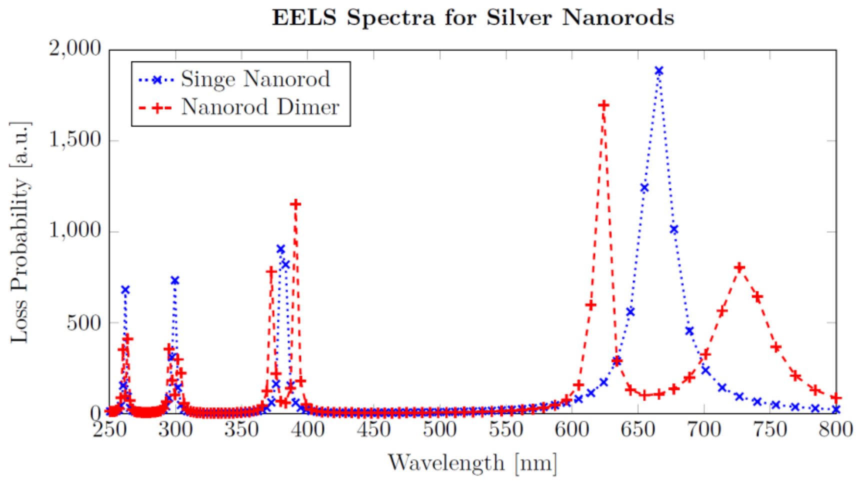
- Compute electron energy loss probability along electron path

$$P(\omega) = \frac{1}{\pi\hbar\omega}\int_{-\infty}^{\infty} \Re\left\{e^{-i\omega\frac{z}{v}} E_z(z,\omega)\right\} dz$$

Numerical Experiments: Silver Nanorod

- EELS spectra for a nanorod and a nanorod-dimer
  - Mode splitting

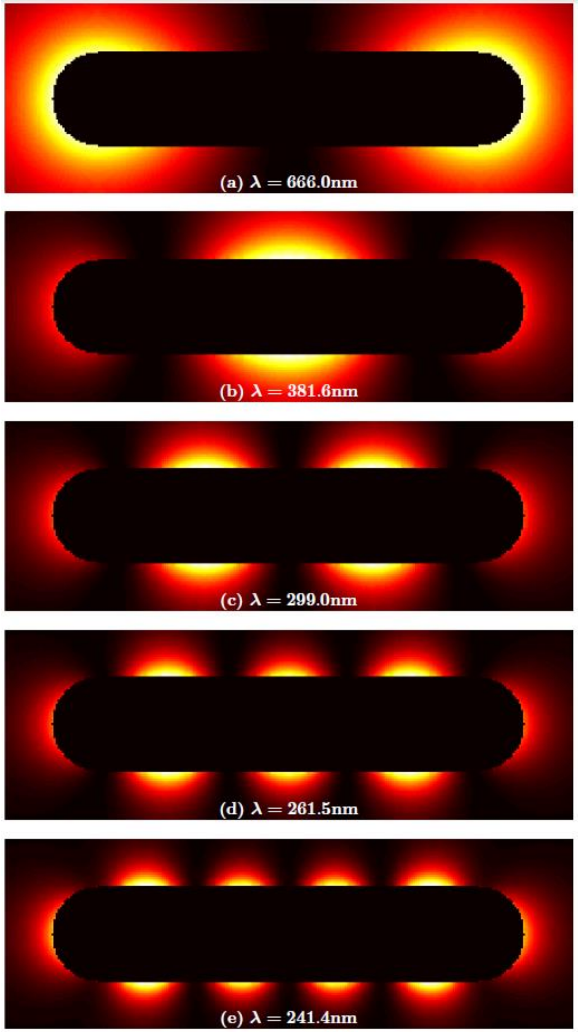




[1] Bigelow et al., ACSNANO 2012

Numerical Experiments: Silver Nanorod

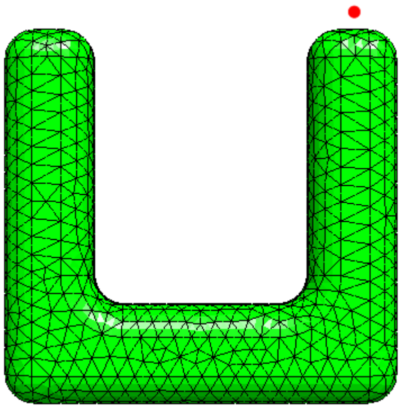
- EELS spectra for a nanorod and a nanorod-dimer
  - Mode splitting
- EELS maps show intuitive loss probability distribution for the first five modes

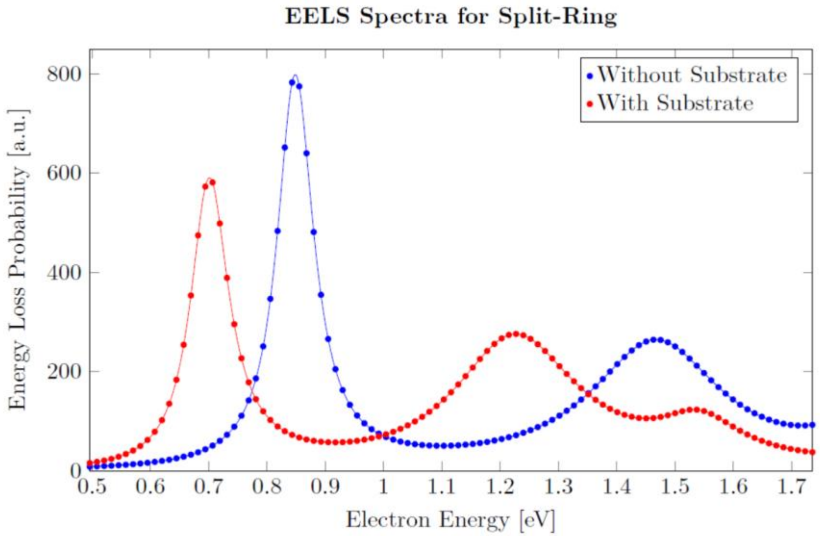


[1] Bigelow et al., ACSNANO 2012

Numerical Experiments: Golden Split-Ring

- Golden split-ring on 30nm silicon nitride substrate
- EELS spectra for electron beam excitation at position ●

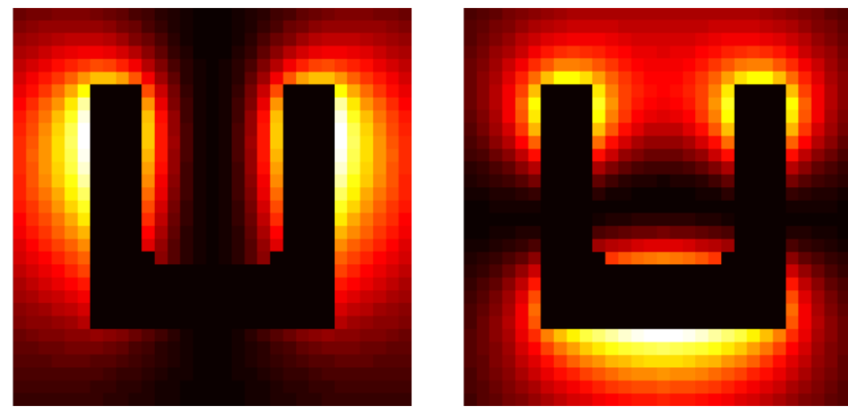


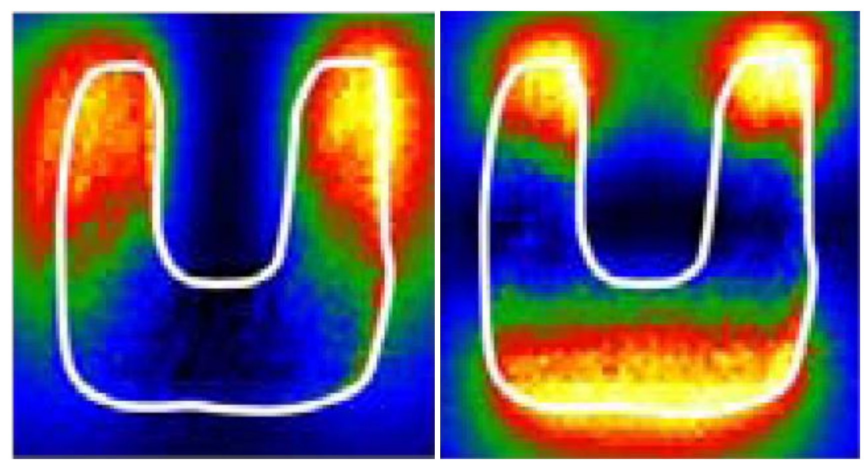


[2] von Cube et al., Opt. Mater. Expr. 1, 1009 (2011)

Numerical Experiments: Golden Split-Ring

- Golden split-ring on 30nm silicon nitride substrate
- EELS spectra for electron beam excitation at position ●
- EELS maps for first two resonance frequencies
  - In agreement with experiments





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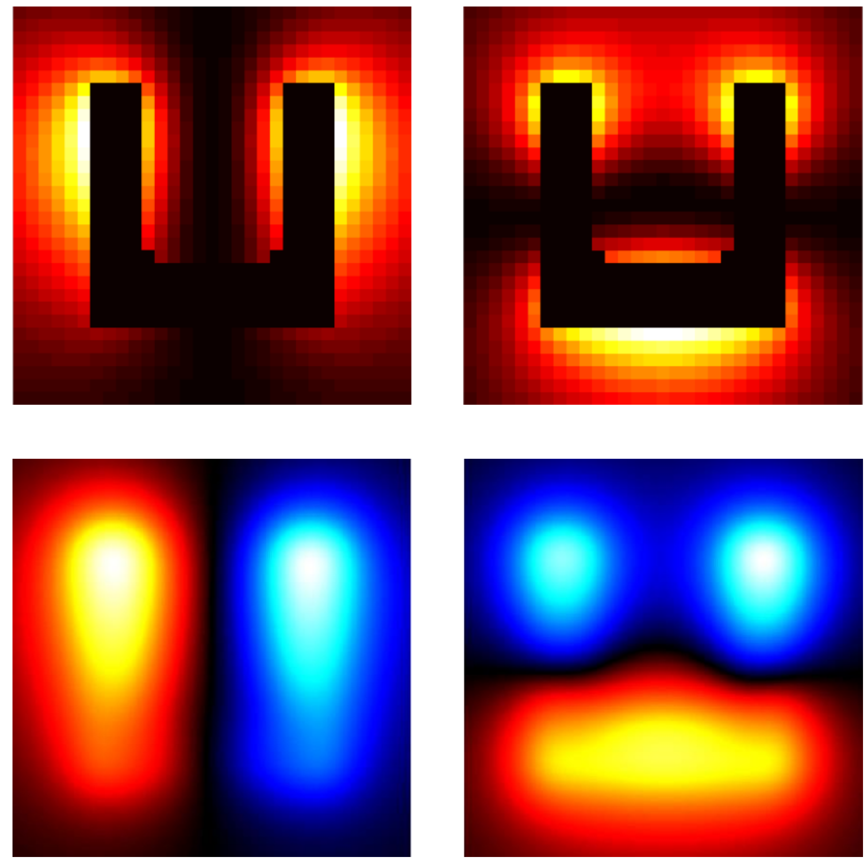
Numerical Experiments: Golden Split-Ring

- Golden split-ring on 30nm silicon nitride substrate
- EELS spectra for electron beam excitation at position ●
- EELS maps for first two resonance frequencies
  - In agreement with experiments
- Electric field in normal direction in agreement to EELS maps



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Conclusion

- MMP is a powerful simulation technique for electromagnetic problems such as EELS.
- Employing the layered dipole expansions guarantees accurate simulation of nanoparticles on layered media.
- The combination allows accurate simulation of realistic structures.
  - Simulation and experiment are in good agreement.

THANK YOU!