Shedding light on dark plasmons with electrons

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Introduction

- Small metallic nanoparticles can support localised surface plasmons (LSPs)
- **Bright** plasmonic modes: effective dipole moment for electromagnetic field to couple to → dipole modes, some higher order modes
- **Dark** plasmonic modes: no net dipole moment → some higher order modes, radial breathing modes (RBMs) in circularly shaped nanodisks (NDs)
  - Inaccessible with conventional optics using plane-wave excitation
  - Can be made visible via symmetry breaking or retardation effects [1]

Experimental details

- Electron-energy loss (EEL) spectroscopy (120 kV)
  - Near-field excitation and collection
  - Probes electromagnetic local density-of-states (LDOS) [2] → nanoscale study of "brightness" of plasmonic modes
- Cathodoluminescence (CL) spectroscopy (30 kV)
  - Near-field excitation but far-field collection → study of outcoupling of RBMs [1, 3]
  - Probes radiative part of LDOS

Results

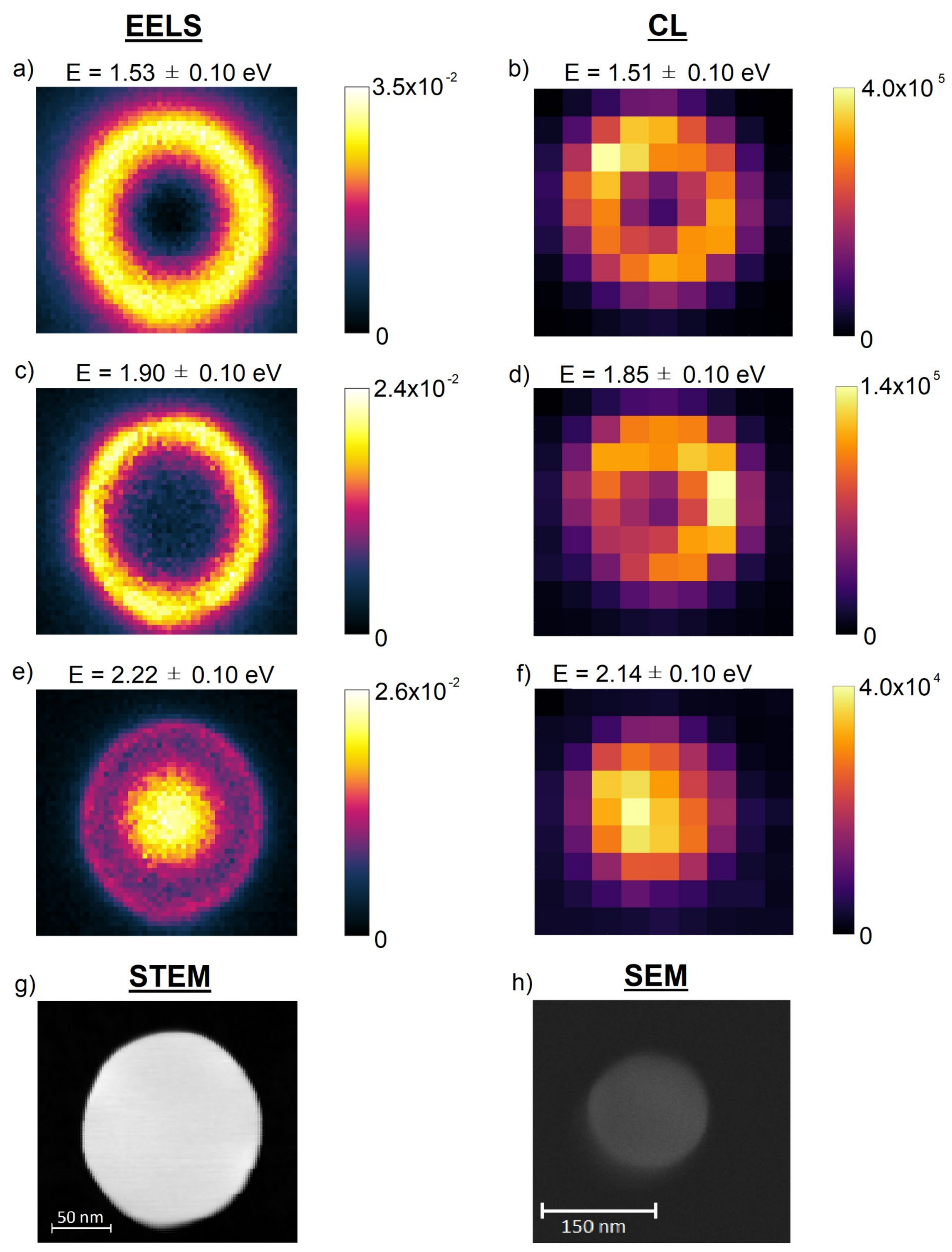

Fig. 1. EEL (left) and CL (right) spectral maps of a 20 nm thin monocrystalline Au ND with a diameter of 160 nm, integrated over (a) and (b) the dipolar plasmonic mode, (c) and (d) the quadrupolar plasmonic mode, and (e) and (f) the RBM. (g) and (h) show the corresponding STEM and SEM images, respectively.

- Typical spatial distribution for bright dipole and quadrupolar modes with greatest intensity at the rim of the ND → visible in EELS and CL (cf. fig. 1)
- Spatial distribution of RBM with maximum at centre → **dark RBM also visible in CL although only radiative part of LDOS**

Results

- Slight differences in spectral position of plasmonic modes in EELS and CL stem from non-identical NDs, e.g. imperfections in roundness
- Difference in spatial resolution in EEL and CL spectroscopy originates from significantly longer integration times needed for less efficient plasmonic radiation processes in CL → reduced pixel size though actual electron beam spot substantially smaller

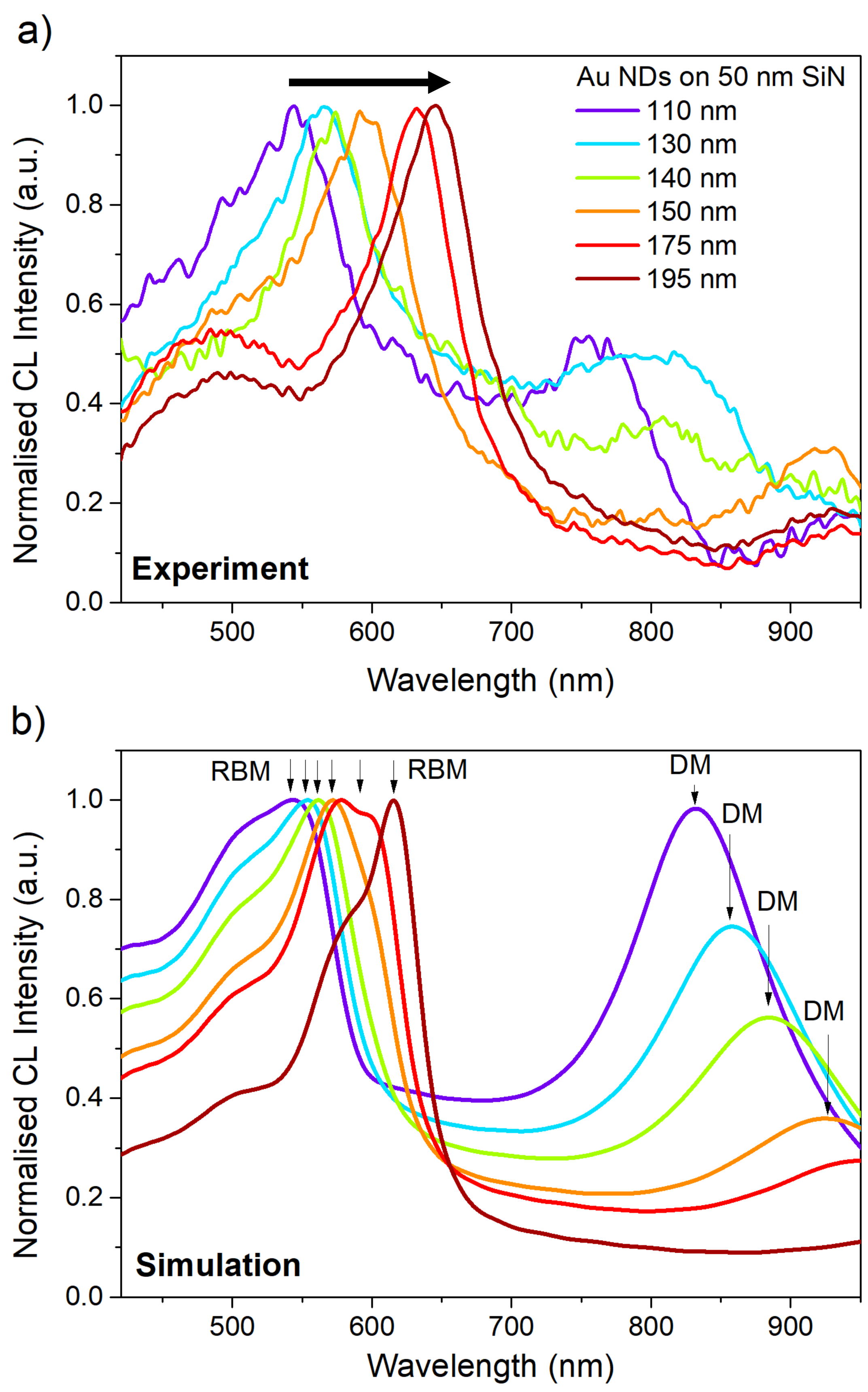

Fig. 2. (a) Experimental (normalised to RBM) and (b) simulated CL spectra of Au NDs with varying diameter, ranging from 110 nm to 195 nm, dispersed on a 50 nm thin SiN membrane. The RBMs and dipolar modes (DMs) are annotated in the numerical plots.

- RBMs visible for Au NDs with much smaller diameter than 200 nm → shown to be the required minimum size for retardation effects to be visible in CL (Ag NDs) [3]
- **Spectral position of RBMs widely tunable** (cf. fig. 2) → engineering for nanophotonic realisations
- Excellent agreement with calculated CL spectra, using the discontinuous Galerkin time-domain method [4]

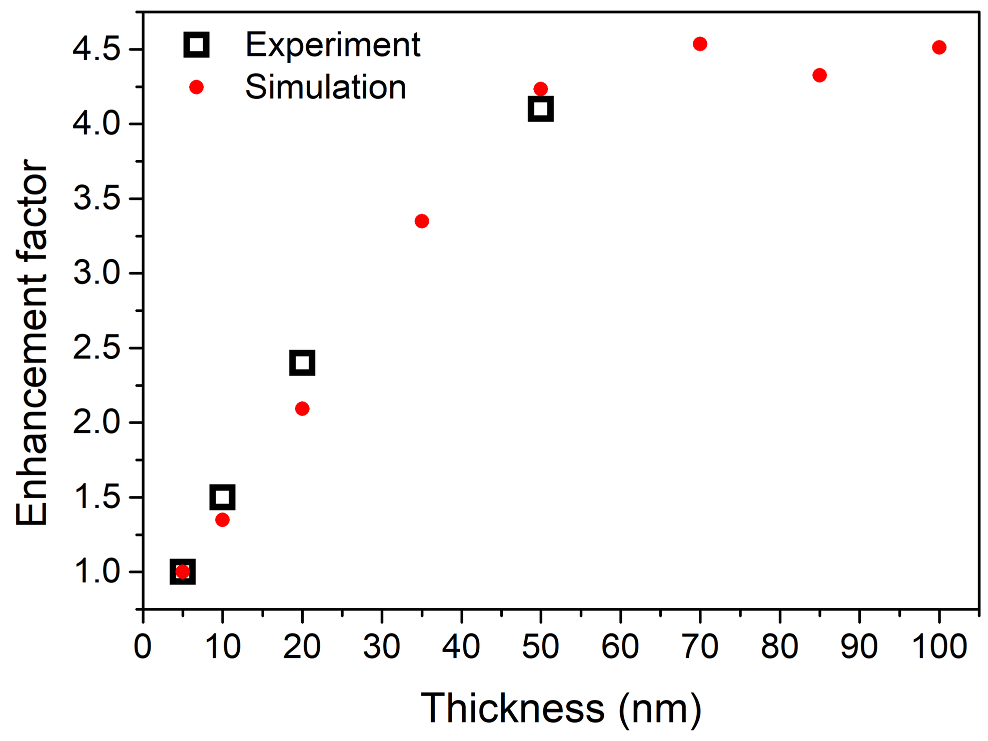

Fig. 3. Maximum CL intensity of the RBM in a 150 nm Au ND as a function of SiN thickness, normalised to 5 nm thin SiN.

Results

- Up to **4.5-fold enhanced CL** for an increased substrate thickness from 5 nm to 50 nm (experiment) and 100 nm (simulation) → **visibility influenced by substrate thickness**
- **Breaking of mirror symmetry by high-index substrate** (rotational symmetry still intact)→ effective dipole moment for electromagnetic field to couple to (cf. fig. 4)
- Simulation: CL enhancement plateaus for SiN thicknesses from 50 nm to 100 nm (cf. fig. 3) → localised electric near-field

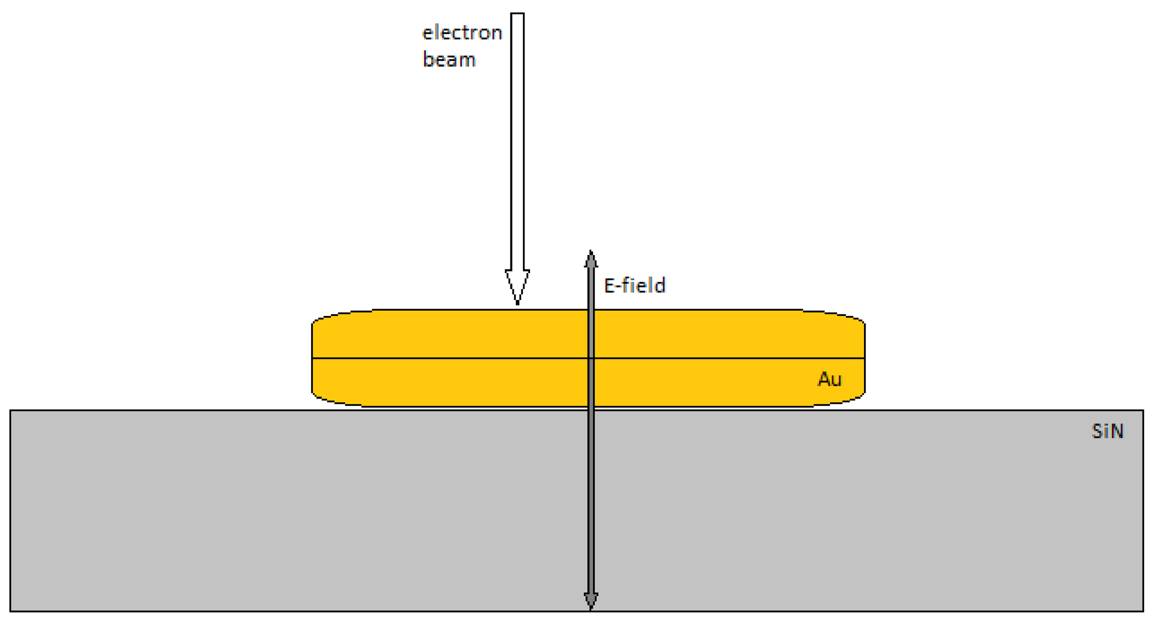

Fig. 4. Schematic of mirror symmetry breaking by high-index substrate.

Conclusion

- Dark plasmonic modes in Au NDs can be locally excited with an electron beam using EEL and CL spectroscopy, and collected in the far-field
- Up to 4.5-fold CL enhancement by increasing SiN substrate from 5 nm to 50 nm (100 nm) → **increasing substrate thickness improves visibility of dark RBMs**
- High-index substrate breaks the mirror symmetry of Au ND, creating a net dipole moment → **outcoupling into the far-field is not negligible**
- Spectral position of dark RBMs can be tuned by varying the ND's diameter → exploitation of their advantageous properties over their bright counterparts
  - longer lifetime,
  - narrower linewidth and
  - much lower radiative losses

References

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