2024
doi:10.1017/S1431927622007851
Microsc. Microanal. 28 (Suppl 1), 2022

STEM-EELS Mapping of Eigenmodes and Coupling Effects of Photonic Silicon Nanocavities

Duncan T.L. Alexander[1*], Valentin Flauraud[2] and Frank Demming-Janssen[3]

[1.] Electron Spectrometry and Microscopy Laboratory (LSME), Institute of Physics (IPHYS), Ecole Polytechnique Fédérale de Lausanne (EPFL), Lausanne, Switzerland.
[2.] Microsystems Laboratory (LMIS1), Microengineering Institute (IMT), Ecole Polytechnique Fédérale de Lausanne, Lausanne, Switzerland.
[3.] SIMUSERV GmbH, Würzburg, Germany.
* Corresponding author: duncan.alexander@epfl.ch

During the last decade, high-permittivity dielectric nanoparticles or nanostructures that support Mie-type resonances have become of interest as an alternative to noble metal plasmonic systems for engineering the scattering of light [1]. Because their resonances are driven by displacement currents within the dielectric volume, rather than the surface charge currents of plasmonics, they offer the potential for reduced absorption losses. Moreover, by supporting a variety of both electric and magnetic resonances, they introduce novel possibilities for tuning light scattering. These dielectric nanostructures or photonic "nanocavities" therefore have a strong potential for nanophotonics applications. With electron energy-loss spectroscopy (EELS) now being a well-established tool for the near-field analysis of plasmonics, here we extend its application to the near-field probing of dielectric nanocavities. Recently, EELS was used to measure eigenmodes in crystalline Si nano-spheres [2]. By instead analysing planar amorphous Si (a-Si) nanocavities, patterned into a range of sizes and shapes according to standard photonic designs, we demonstrate the potential of EELS for a two-dimensional mapping of eigenmodes in dielectrics [3].

The samples are prepared using electron beam lithography to pattern 100 nm thick layers of a-Si deposited onto Si_3N_4 membranes. The same principle is used to pattern Ag and Au films of 30 nm thickness for certain measurements. EELS maps are recorded in scanning transmission electron microscopy (STEM) mode using a double aberration-corrected FEI Titan Themis 60-300 equipped with a Gatan GIF Quantum ERS spectrometer. A 300 keV incident electron beam is applied, monochromated to give an energy spread of 100–110 meV full width half maximum in the zero-loss peak (ZLP), and having high beam currents of ~200–260 pA for fast mapping with high spatial statistics.

In order to begin illustrating the nature of the data acquired from the dielectric resonators, in Figure 1 we contrast EELS heat intensity maps from (a) a 1000 nm Ag plasmonic wire and (b) a dielectric a-Si wire of the same length. For the Ag wire, we observe nodal patterns of increasing spatial density as energy loss increases, as expected for the standing wave Fabry–Perot type resonant modes from $m = 2$ to 5 [4]. In Figure 1(b), we present the equivalent excitations for the a-Si wire. It is seen that, while the dielectric wire gives a similar pattern of excitations, the spatial (and also spectral) signatures of its eigenmodes are significantly more extended and less well defined than for the plasmonic Ag wire. This points to fundamental differences in their respective EELS responses.

In order to explore the nature of the dielectric EELS response in more detail, we shall present the study of a-Si nanocavities that are patterned into different geometries (discs, ellipses, rectangles, square and triangle) [3]. The spatial and spectral signatures of their EELS excitations are interpreted in terms of resonant eigenmodes, as simulated using a dedicated solver in CST Studio Suite, with interpretations

supported by frequency domain dipole excitation simulations. With this approach, we decouple the signals from electric and magnetic modes across resonators of different sizes and shapes, and observe how their resonance nature translates into EELS characteristics. One example is the spectrally and spatially extended signal of the low *Q*-factor electric dipole. Another is intensity hotspots that derive from stimulating electric toroidal dipole and second-order magnetic modes within the nanocavities, with hotspot localization and size depending directly on the eigenmode's electric field nature. For certain modes, an agreement of ±0.01 eV is found between simulation and experiment. Far-field scattering simulations further connect these EELS analyses to light-optical tuning strategies for the nanocavities.

Recently, EELS has been used to study interactions in dielectric–noble metal systems [2, 5]. In Figure 1(c), we move further in this direction with the study of an Au nanorod—a-Si disc—Ag nanorod trimer. In the absence of the a-Si disc, the 110 nm gap between the 150 nm nanorods would prevent them from showing a significant modal coupling. However, the 1.32 eV EELS map suggests that the central dielectric disc enables a bonding interaction between the rods, while, at 1.51 eV, the a-Si disc and inner rod ends are instead strongly excited. Further interpretation of this data is currently in progress [6].

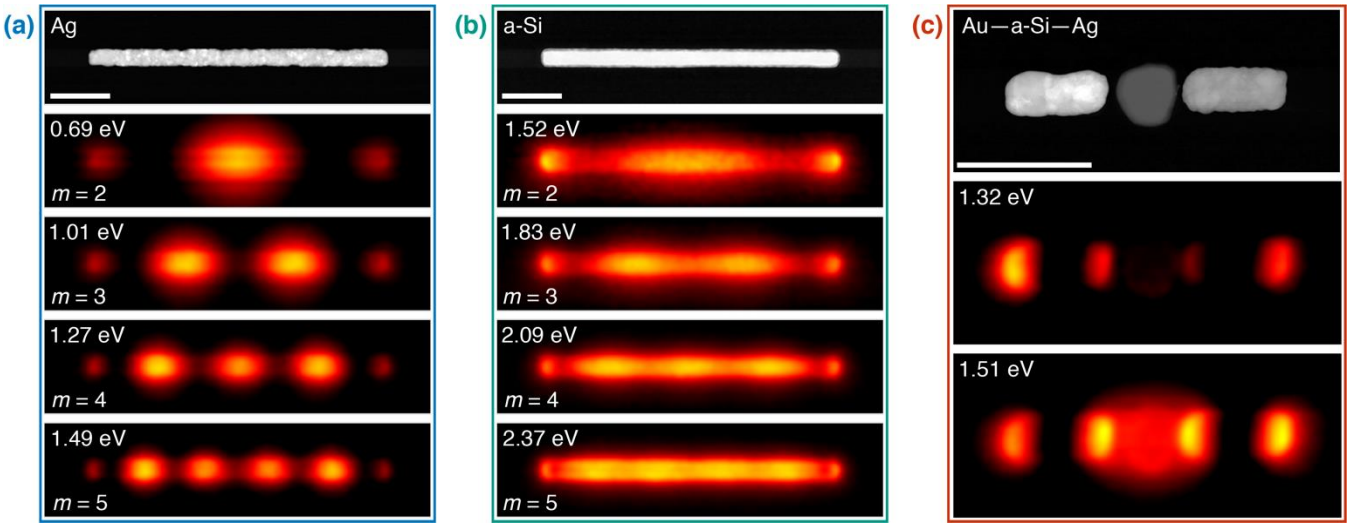

Figure 1. STEM images (top row) and EELS heat intensity maps for excitations of: (a) 1000 nm Ag wire; (b) 1000 nm a-Si wire; and (c) coupled Au nanorod—a-Si disc—Ag nanorod. The EELS maps are normalized by the ZLP intensity and integrated from 0.06 eV energy windows centered on the indicated energy-loss values. Data processing is carried out similarly to that described in [3]. Scale bars: 200 nm.

References:

[1] M Decker and I Staude, J. Opt. **18** (2016), p. 103001. doi:10.1088/2040-8978/18/10/103001
[2] A Assadillayev et al., ACS Photonics **8** (2021), p. 1582. doi:10.1021/acsphotonics.0c01554
[3] DTL Alexander, V Flauraud and F Demming-Janssen, ACS Nano **15** (2021), p. 16501. doi:10.1021/acsnano.1c06065
[4] D Rossouw et al., Nano Lett. **11** (2011), p. 1499. doi:10.1021/nl200634w
[5] J Schultz et al., Adv. Optical Mater. **9** (2021), p. 2101221. doi:10.1002/adom.202101221
[6] The CIME, CMi and J. Brugger at EPFL are thanked for support. The European Commission's 7th Framework Programme is acknowledged for early funding under grant 288263 (NANO-VISTA).