Unveiling Single Particle Coupling of Metallic Nanoparticles and Whispering Gallery Mode Resonators

Yves Auad¹, Cyrille Hamon¹, Marcel Tencé¹, H. Lourenço-Matins^{2, 3}, Vahagn Mkhitaryan⁴, Odile Stéphan¹, F. Javier García de Abajo^{4,5}, Luiz H. G. Tizei¹, Mathieu Kociak¹

Whispering gallery mode resonators (WGMRs) confine light using total internal reflection and take advantage of its several and narrowband circulating resonances for applications in optomechanics [1], quantum electrodynamics [2] and sensing [3, 4]. The spherical symmetry and the low radiation loss of dielectric microspheres make it difficult to probe them under free-space light. However, local field enhancement from metallic nanoparticles placed at the edge of the resonators can circumvent this problem by interfacing the optical far-field with the bounded cavity modes. In this work, we study the dependency of the induced gallery-modes with the nanoparticle surface plasmon eigenmodes with spatial resolution by using electron energy loss spectroscopy (EELS) cathodoluminescence (CL) in a Scanning Transmission Electron Microscope (STEM) (Figure 1). Using both EELS and CL is mandatory to unravel the physics of these modes. Indeed, EELS offers the very wide energy range (from 1.5 eV to 9.0 eV) needed to investigate WGMs (Figure 2). However, even the very high energy resolution (~10 meV) of the used NION HERMES 200 (ChromaTEM) is insufficient to resolve modes of different polarization, only possible using CL. We also show that gallery-modes are induced along the spectral range of the dipolar mode of the nanoparticle, but absent in most of the higher order modes due to their reduced net dipolar moment. Finally, we demonstrate the dependence of the polarization of the induced gallery-mode with the induced dipole moment of the metallic nanoparticle. The present work has been recently published in ref. [5].



¹ Laboratoire de Physique des Solides, Orsay, 91405, France

² Max Planck Institute for Biophysical Chemistry, Gottingen, 37077, Germany

³ IV. Physical Institute, University of Gottingen, Gottingen 37077, Germany

⁴ ICFO-Institut de Ciencies Fotoniques, The Barcelona Institute of Science and Technology, 08860 Castelldefels (Barcelona), Spain

⁵ ICREA-Institució Catalana de Recerca i Estudis Avançats, Passeig Lluís Companys 23, 08010 Barcelona, Spain

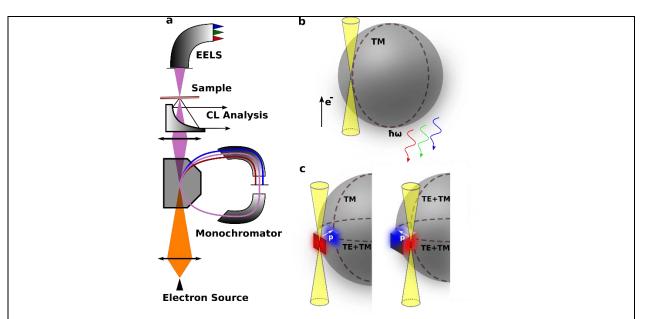


Figure 1: Scheme of the electron microscope containing the electron source, the electron monochromator, the cathodoluminescence mirror, the sample and the EELS spectrometer (a). In (b), scheme of the electron beam and the excited TM mode (dashed line) in the bare sphere. In the presence of the cube, gallery-mode polarization can be controlled by the probe position, as shown in (c).

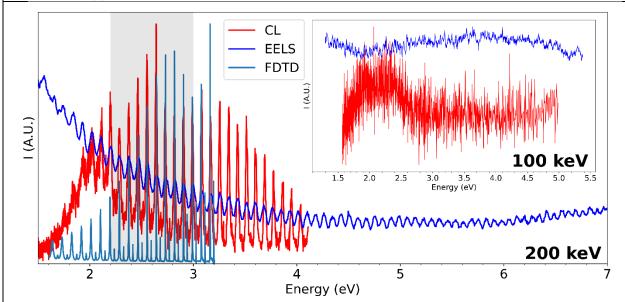


Figure 2: FDTD simulated and experimental EEL and CL spectra obtained from a bare SiO_2 sphere using 200 keV electrons. In the inset, EELS and CL under 100 keV electrons and a much weaker coupling strength.

References:

[1] Tobias Kippenberg et al. Analysis of radiation-pressure induced mechanical oscillation of an optical microcavity. Physical Review Letters, 95, 2005.

- [2] D. Vernooy et al. Cavity QED with high-Q whispering gallery modes. Physical Review A, 57, 1998.
- [3] Yu Zheng et al. Sensing and lasing applications of whispering gallery mode microresonators. Opto-Electronic advances, 1, 2018.
- [4] J. Zhu et al. Interfacing whispering-gallery microresonators and free space light with cavity enhanced Rayleigh scattering. Sciences Reports, 03, 2014.
- [5] Y. Auad et. al. Unveiling the coupling of single metallic nanoparticles to whispering-gallery microcavities. Nano Letters, 22, 2022.