doi:10.1017/S1431927615011034 Paper No. 1024 Microsc. Microanal. 21 (Suppl 3), 2015

SPARC: a Cathodoluminescence Platform for Nanoscale Plasmonics in a Scanning Electron Microscope

Toon Coenen[1,2] and Jacob Jan de Boer[1]

[1.] Delmic B. V., Delft, The Netherlands
[2.] FOM Insitute AMOLF, Center for Nanophotonics, Amsterdam, The Netherlands

In the field of plasmonics metallic nanostructures are used to confine light to deep-subwavelength volumes. Owing to the nanoscale size of such structures it is hard to resolve their optical properties with conventional optical microscopy.

Recently, electron-beam spectroscopy techniques have emerged as powerful probes in nanoscience due to their ability to generate, probe, and control light at length scales far below the diffraction limit of light. Taking advantage of the extremely high spatial resolution, novel techniques have appeared that combine electron beam excitation with optical spectroscopy. Spatially-resolved cathodoluminescence (CL) spectroscopy, in which the electron-beam-induced radiation is collected inside an electron microscope, is one of these techniques that holds great potential for nanoscience. For a long time CL spectroscopy was mainly used in geology to analyze and identify minerals, but in the past two decades its scope has expanded significantly. Recently it has been used to study fundamental optical properties of a myriad of metallic, semiconductor, and dielectric (nano)materials in the fields of materials science and nanophotonics, including plasmonics and metamaterials. We have developed a special version of CL spectroscopy in which we can both effectively measure the emitted spectrum as well as the angular emission distribution (SPARC) [1-2].

The SPARC system is integrated with a standard commercially available scanning electron microscope (SEM). SEMs are relatively easy to operate and do not require electron-transparent samples. Additionally, the vacuum chamber is more spacious providing more flexibility. As a result SEM-CL is widely applicable and easy to use. Innovations in the SPARC CL-system include an improved light collection with a piezo-controlled parabolic mirror and the development of angle-resolved CL, which have further expanded the possibilities. In Figure 1(a) we show a photograph of the SPARC CL collection system. The piezoelectric positioning of the paraboloid mirror enables efficient light collection which is critical in plasmonic studies where CL signals are typically very low (10^{-4} photons/electron). Furthermore, by measuring the CL beam profile from the paraboloid with a 2D camera we are able to measure the angular profile, as every transverse point in the beam corresponds to a unique emission angle. In Figure 1(b) an illustration is shown of how the angle-resolved measurements are performed.

The canonical structure in plasmonics is a single nanoparticle. For small nanoparticles (< 50 nm) the resonant features are relatively easy to determine as they are predominantly dipolar. However, for larger particles this can be more challenging due to retardation effects which lead to the presence of higher order multipoles (magnetic dipoles, electric quadrupoles etc.). In Figure 2(a) we show the CL spectrum of a gold nanoparticle (180 nm diameter, 80 nm high) on a silicon substrate as measured with a spectrometer. The spectrum is averaged over all scanning pixels that fell within the particle (5 nm pixel). As inset we show the spatial profile at the peak wavelength which a distinct doughnut pattern. Figure 2(b) shows the angular profiles collected at different excitation positions on the particles. Clearly the particle acts as an efficient antenna and beams light away from the excitation position in a non-dipolar pattern. The CL spectrum, the nanoscale excitation distribution as well as the angular profiles provide valuable insights into the resonant behaviour of such particles [3]. Although this is just one example we have applied the technique to many other plasmonic systems as well and shown that the SPARC is a very powerful platform for studying plasmonics at the nanoscale.

References:

[1] T. Coenen, E. J. R. Vesseur, and A. Polman, *Appl. Phys. Lett.* **99**, 143103 (2011).
[2] T. Coenen, E. J. R. Vesseur, A. Polman, and A. F. Koenderink, *Nano Lett*. **11**, 3779 (2011).
[3] T. Coenen, F. Bernal Arango, A. F. Koenderink, and A. Polman, *Nat. Commun*. **5**, 3250 (2014).

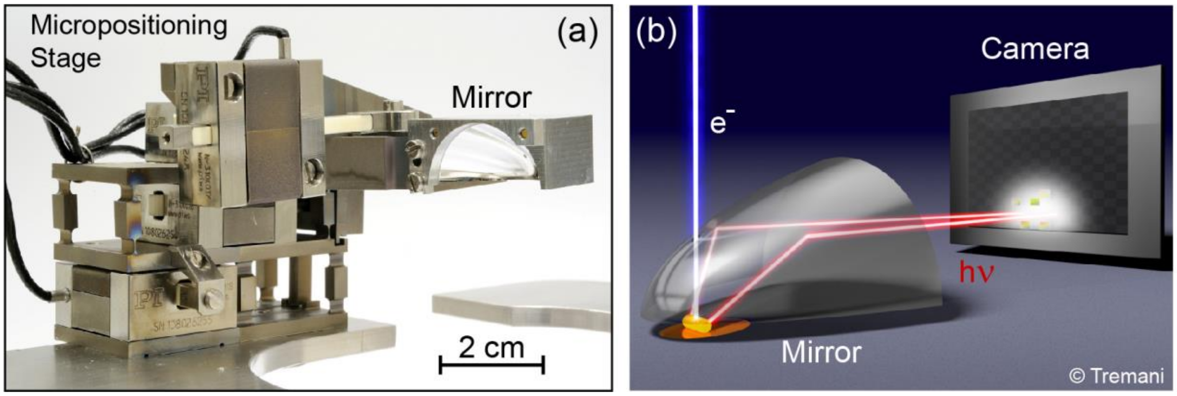

Figure 1. (a) Piezo-controlled mirror manipulator system for efficient light collection (b) Graphical representation of angle-resolved detection of CL where light coming from a parabolic mirror is projected onto a 2D silicon camera.

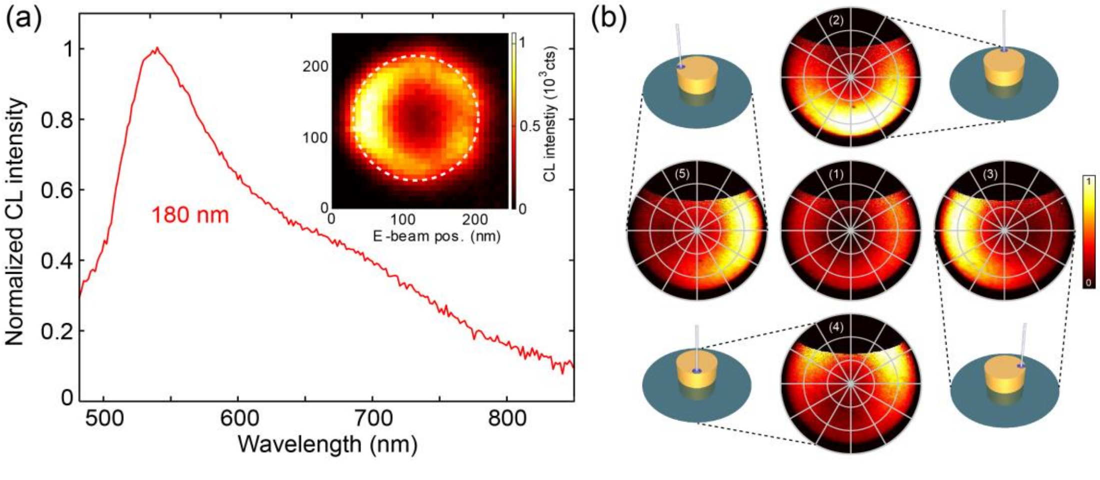

Figure 2. (a) Average CL spectrum for a 180 nm diameter gold particle on a silicon substrate. The inset shows the spatial profile at the peak wavelength (550 nm) where the white dashed line indicates the geometrical edge of the particle. (b) Angular emission profiles measured at 600 nm showing CL intensity as function of azimuthal and zenithal emission angles. for the same particle for central excitation (1) and four edge excitation positions (2-5) [3].