Angle-Resolved Cathodoluminescence Nanoscopy
T. Coenen

The field of microscopy is an important cornerstone of physical and biological sciences. The development of high-resolutionmicroscopy/nanoscopy techniques has enabled a revolution in science and technology, greatly improving our understanding of the microscopic world around us, and forming the basis for the development of advanced nanotechnology. Similar to electrical integrated circuits there currently is a drive to miniaturize optical components with the aim to generate, detect, guide, or switch light on the nanoscale. To support this emerging field of nanophotonics, experimental techniques that can characterize optical properties on this length scale are required. In this thesis we present a novel experimental nanoscopy technique that can be used as a nanoscale characterization tool for optical properties: Angle-Resolved Cathodoluminescence (CL) Imaging Spectroscopy (ARCIS).

This technique combines electron microscopy with optical microscopy, giving it an excitation resolution greatly exceeding that of optical microscopy, while retaining optical sensitivity. Using ARCIS both the emission spectrum and the angular emission profile of optical nanostructures can be measured with great accuracy. Chapter 2 explains the operating principles of the technique. We discuss the various forms of electron-material interaction and electron-induced radiation, and give a detailed technical description of the setup. We also briefly discuss electron energy-loss spectroscopy (EELS) which is a complementary technique to ARCIS. In Chapter 3 we describe the calculation procedures for retrieving the angular emission pattern of a nanostructure using ARCIS. The technique is calibrated by studying the angle-resolved radiation pattern of a known source of radiation: transition radiation from a single-crystalline gold substrate.

In the body of the thesis we have applied the ARCIS technique to a variety of metallic and dielectric nanostructures to demonstrate its general applicability to nanophotonics. Most of these structures act as nanoantennas which are the elementary building blocks in nanophotonics. Nanoantennas form an interface between the near field and the far field and can exhibit complex angular patterns and scattering spectra which can be accurately probed by ARCIS. Throughout this thesis we have employed state-of-the-art nanofabrication techniques such as electron beam lithography and focused-ion-beam milling which allow precise engineering of optical structures on the nanoscale.

In Chapter 4 we use ARCIS to study the optical properties an array of five gold nanoparticles which acts as a subwavelength optical Yagi-Uda antenna. We demonstrate that the antenna array is strongly directional and that the directionality can be controlled by selectively driving one of the antenna elements. The angular response of the array antenna is modeled using a coupled dipole model which takes into account interparticle coupling and interaction with the reflective substrate.

In Chapter 5 we focus on the optical properties of gold plasmonic ridge antennas of different length which act as traveling wave antennas. The reflective end facets of the ridge antenna cause standing wave modes which can be visualized with ARCIS. From the data we extract a dispersion relation for the guided ridge plasmon mode. We also study the angular pattern and find that for small ridges the angular pattern can be described by a single dipole. For longer antennas we find a more complex fringe pattern, resulting from the interference of the two radiating end facets of the ridge antennas.

Chapter 6 introduces polarization-sensitive Fourier microscopy as a further extension of the ARCIS technique. We analyze the angular and polarization distributions for transition radiation from a gold surface and from a gold ridge antenna and compare these with the theoretical distributions. We find that we can accurately predict the experimentally measured polarization-filtered angular profile when the theoretical polarization and angular pattern of emission are known. Furthermore, we demonstrate that it is also possible to reconstruct the emission polarization without *a priori* knowledge of the source, enabling accurate determination of the electrical dipole moment orientation.

In Chapter 7 we use ARCIS to characterize elliptical arena cavities that are milled into a gold substrate. We image the plasmon modes inside the cavities and find that the resonance spectrum is solely determined by the major axis length and the reflection phase pickup at the cavity boundaries. We demonstrate that these elliptical cavities act as parabolic antennas and hence can show a remarkably strong directionality which can be tuned with size, ellipse eccentricity, wavelength, and the electron beam excitation position.

Chapter 8 describes how a single plasmonic nanoparticle can act as directional scatterer. Using ARCIS, we find that the angular response of the nanoparticle strongly depends on excitation position and that this can be used to beam light in a well-defined direction. The beaming is caused by interference between in-plane and out-of-plane electric and magnetic multipole components. The directionality is enhanced for larger particles, which is caused by higher-order multipole contributions to the scattering. Using a combination of full-wave simulations and analytical point-scattering theory we determine the relative contribution of different multipoles.

In Chapter 9 we study the spectral and angular properties of nanoscale holes in a gold film. We study the decay of the CL signal away from the hole to distinguish between direct near-field coupling and indirect surface plasmon polariton excitation. Furthermore, we find a striking complementarity in the directionality of the emission compared to that of the nanoparticles studied in Chapter 8. The data are compared to full-wave simulations and a simple analytical model. The data suggest that the observed directionality is caused by the Kerker effect where magnetic and electric dipole components interfere in the far-field.

In Chapter 10 we use EELS and CL measurements to gain more insight into the near-field coupling in composite plasmonic “dolmen” antennas. We demonstrate that the degree of coupling to different modes of the system can be controlled by precisely positioning the electron beam. Furthermore, we study the effect of size, particle spacing, and excitation position on the spectra. The EELS and CL measurements were performed on the same structures enabling a direct comparison between the two. We find that in some cases EELS and CL yield very similar information, while in other cases they are distinct, related to differences in the modal scattering efficiencies.

Chapters 4-10 discuss the optical properties of plasmonic metal nanostructures. In Chapters 11 and 12 we move to dielectric/semiconductor nanostructures which form an important class of structures within nanophotonics as well. In Chapter 11 we study the optical modes in 2D silicon nitride photonic crystal membranes using ARCIS. We use CL to image delocalized modes in hexagonal 2D crystals and localized modes in photonic crystal cavities. Using momentum spectroscopy we visualize the spatially-resolved dispersion in the crystal.

Finally, in Chapter 12 we study the resonant behavior of silicon nanodiscs using ARCIS. We measure resonance spectra and determine the corresponding field profiles for different disc diameters. We find that larger discs support multiple Mie resonances with complex modal profiles. Using the angular emission profiles we resolve the electric and magnetic nature of the disc resonances, and elucidate their directional properties.

Overall, this thesis introduces Angle-resolved Cathodoluminescence Imaging Spectroscopy as a novel microscopy technique to resolve optical properties at the nanoscale. By combining elements from electron and optical microscopy we are able to resolve optical phenomena that are impossible to elucidate with other techniques.