Probing light emission at the nanoscale with cathodoluminescence

Brenny, B.J.M.

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Introduction

1.1 Light

1.1.1 A short history of light

The science and understanding of light has gone through many stages throughout the centuries, starting out as the study of vision before the development of optical theories and then light as a separate, natural entity governed by laws of physics [1–3]. Today light still captivates both laymen and scientists alike, fueling a rapid expansion of fundamental research and applications.

We can begin the study of light with the Greek theories of vision, who regarded sight as the product of rays traveling between objects and the observer. Intramissionists such as Democritus believed that objects emanated rays while extramissionists such as the Pythagoreans and Euclid believed on the contrary that it was the eye that emitted rays that probed the surrounding world, as shown in Figure 1.1(a). Euclid established the mathematical and geometrical groundwork to study light rays, describing them as straight lines moving through space.

In the medieval period, the Arab scholar Ibn-al-Haitham (Alhacen) discarded the Greek theories and set the foundations for modern optics. He described light as a separate entity that illuminates items and then reflects away, carrying information about the object, as shown in Figure 1.1(b). Light is composed of a multitude of rectilinear rays moving through transparent materials, which allowed Ibn-al-Haitham to examine processes such as reflection, refraction and the “camera obscura”. Later, Johannes Kepler built on these principles to explain image formation, allowing him to explain the working principles of concave and convex lenses. Willebrord Snellius and René Descartes both developed the law of refraction before Francesco Grimaldi documented diffraction for the first time and defined light as a fluid substance that moves with great speed, undulating with different frequencies to produce different colors.
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After this the battle over the interpretation of light as a particle or a wave became more explicit, pitting Isaac Newton against Christiaan Huygens. Newton described light as streams of particles of different sizes moving at high speeds in straight lines. Huygens, on the other hand, interpreted light as a high-speed wave or perturbation propagating through an aether that permeates the whole universe, and described the concept of the wavefront for the first time.

Later in the 18th century, experiments on electricity and magnetism by Michael Faraday and others led to the development of new theories, which culminated with James Clerk Maxwell describing electromagnetic fields with four now-famous equations [5], establishing the direct relations between electricity and magnetism. This led to the fundamental realization that light is an electromagnetic wave, composed of rapidly oscillating electric and magnetic components. Many new theoretical and experimental concepts followed, such as polarization, which describes the direction of oscillation of the electric fields and plays an important role in many phenomena.

Experiments on the photoelectric effect in the early 20th century posed problems for Maxwell’s wave theory. Metals were shown to emit electrons when irradiated by light, but only if the frequency of the light exceeded a material-specific threshold which also determined the energy of the emitted electrons, while Maxwell’s theory predicted a dependence on radiation intensity. Albert Einstein resolved the issue by describing light as a particle with quantized energy, later named the photon [6]. The word photon is derived from the Greek word for light, φῶς (phōs). However, the wave-like properties of light remained, leading to the development of quantum mechanics, which described the particle-wave
duality and a new realm of fascinating effects such as entanglement. During the ensuing decades, research has improved the miniaturization of the objects being used to control and manipulate light, down to the scale of light itself: the nanoscale. At this small scale, light can no longer simply be described by ray optics, but one has to account for different regimes, the near and far fields. Very close to an emitter, within a (few) wavelength(s), the near field is dominant because of bound or evanescent fields, which govern the close-range interactions with other objects and emitters. At longer distances evanescent fields die out and only propagating waves survive, determining how objects interact in the far field.

1.1.2 Nanophotonics

Nanophotonics, the study of light at the nanoscale, has become an important and dynamic field of research in the last decade. At this deep-subwavelength scale, light interacts with matter in complex ways, in which not only the material properties but also their size and shape play a role in determining the interaction. At the nanoscale, one may use these interactions to mold and control different degrees of freedom of light: the temporal and spatial frequencies as well as the phase and polarization. Both fundamental new insights and a plethora of applications have followed from these efforts, a few are described below [7].

To achieve these goals, metallic and dielectric nanoscale geometries are tailored for specific applications. Photonic crystals started the rush, allowing light waves to be guided, slowed down, and even trapped [12–16]. Plasmonics, using (noble) metals to couple light to the oscillations of free electrons, permits even higher degrees of confinement and often a broadband response, albeit in exchange for greater losses [17–19]. Plasmonic structures have been used to great effect in antennas, waveguides and metamaterials [20–25]. Antennas, which connect the electromagnetic near and far fields, have a broad range of applications and can also be composed of dielectrics [26–31]. Although nanostructured materials interact most strongly with the electric field of light, precise engineering allows the enhancement of magnetic effects by using chirality and helicity of structures and/or fields [32–35]. Combining these magnetically active building blocks into 2D metasurfaces or 3D metamaterials allows for even more tailored responses, such as negative refraction [8–10, 21, 36–39]. As the optical components shrink down further, one comes into the quantum regime, opening up a host of new physics and possibilities such as quantum tunneling and single-photon processes [40–45].

All of these nanophotonic architectures and strategies have been used to develop a wide range of devices and applications. Communication/computation technology [11, 46], lasers [23, 47], solid-state lighting [48], solar energy [49–53], sensing and imaging [54–59], medical diagnostics and therapy [60–65], all of these have benefited from the developments in nanophotonics.

These initial successes underlie a vibrant and expanding universe of nano-optics, with exciting new prospects for coupling light to or tuning it with other degrees of freedom, such as mechanical vibrations, electron spins or electron motion [66–
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Figure 1.2 – Image of an electrically tunable optical metasurface, an exciting new nanophotonics application under development. Such a metasurface could be used for wavelength multiplexing, creating orbital angular momentum and computing. (Image taken from [7] and inspired by [8–11])

This can be applied to quantum processes, nonreciprocal or topologically insulated optical circuits [69–71], tunable graphene optoelectronic circuits [72, 73] or tunable optical metasurfaces [9–11]. An example of such a possible future application is shown in Figure 1.2. An electrically tunable metasurface could be modulated to achieve a response dependent on wavelength and polarization, for high speed communication and computation.

Light is an essential tool for characterizing the nanoscale world that controls it, but even by using the advantages of the nanophotonic toolbox to shrink the “size” of light, it often remains difficult to study these tiny structures in detail. Far field imaging is limited by Ernst Abbe’s optical diffraction limit, which states that light cannot be focused to a spot smaller than $\lambda_0/2$ [74], where $\lambda_0 = 400–800$ nm in the visible spectral range. While near field probes circumvent the Abbe limit, the probe itself perturbs the nanophotonic environment [75–77]. Another cornerstone of nanoscale engineering, the electron, offers an alternative.

1.2 The electron

Similarly to light, the electron has a storied history rife with the particle vs. wave duality. The word comes from the Greek word ἠλεκτρόν (elektron), meaning “amber”, since the Greeks noticed that amber attracted small objects when rubbed by cloth or fur (now known as static electricity). The centuries after the Greeks saw many experiments related to electricity, until the discovery of the electron occurred
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during research into electrical discharges in rarified gases during the 19th century. When applying a potential between a cathode and anode in a vacuum tube, a glow would appear from both the gas and the glass of the tube. Experiments by Johann Hittorf, Eugen Goldstein and Sir William Crookes showed that there appeared to be rays moving from the cathode to the anode, creating the glow [78–80]. Placing an object in the path created a shadow, suggesting the rays moved in linear fashion, similar to light, but they could be deflected by using a magnet. British and French scientists viewed the cathode rays as streams of negatively charged particles, while German physicists mostly explained them as wave-like processes in the ether. In 1897, J.J. Thomson performed several experiments that confirmed the existence of negatively charged particles that could also be deflected by electric fields and measured with high accuracy the mass to charge ratio \( m/e \) [81].

However the exact nature of the particles and their role with respect to atoms was still unknown. Numerous experiments in the ensuing two decades by Ernest Rutherford and others brought about the model of the atom as a positively charged nucleus surrounded by the negatively charged electrons. Niels Bohr posited that the electrons in this atomic model could only exist in quantized energy states, explaining atomic emission lines which tied into Einstein’s theory of the quantized photon [82]. Quantum mechanics took hold of the electron, just as it had for the photon, leading to measurements of wave-like behavior such as diffraction, the discovery of spin and processes such as superconductivity or the (quantum) spin Hall effect.

Both the photon and the electron are characterized by the particle-wave duality but, since the electron has mass, it can reach much higher momenta and correspondingly smaller wavelengths. This large energy and momentum allows for different interactions with materials and enables new light generation mechanisms. As was mentioned earlier, the relatively large wavelength of light complicates detailed studies of subwavelength nanophotonic structures. In contrast, the short wavelength of the electron wave function allows for a much smaller probe: the de Broglie wavelength is \( \sim 7\) pm for a 30 keV electron \( (\sim \lambda_{optical}/10^5) \). Electron microscopy exploits this to great effect for imaging nanoscale objects.

Electrons were first used for imaging purposes in the 1930's, by pioneers such as Ernst Ruska, Max Knoll and Manfred von Ardenne. The first electron microscopes used electrons transmitted through ultrathin samples, but the scanning electron microscope (SEM) that could image solid samples soon followed [83–87]. In an electron microscope, electrons have to be emitted, accelerated, focused and scanned over the sample of interest [88–90]. Emission sources can either use heat (thermionic emitter) or a large electrical potential gradient (field emitter) to extract electrons from a filament and use them to form a beam. The electrons are then accelerated by an anode; the acceleration voltage determines the energy and velocity of the electrons and is typically in the 1–30 kV range for a SEM. A higher electron energy corresponds to a shorter wavelength, thus affecting the resolution. Electromagnetic lenses are used to generate magnetic fields to focus the electron beam. In the final lens, coils deflect and scan the focused
electron beam over the sample surface in order to construct, point by point, an image from emitted electrons. One can detect different electron-based signals, but the most common are low-energy secondary electrons (< 50 eV) that carry information about the surface topology (forming the typical SEM image) or backscattered electrons that contain information about the atomic number and thus the composition of the sample being studied. The high-energy electrons can also generate electromagnetic radiation, which is our main point of interest: the goal of this thesis is to use electrons as a probe to generate and study the radiative properties of nanophotonic materials, structures, and devices, far below Abbe’s optical diffraction limit.

1.3 Light from electrons: cathodoluminescence

Cathodoluminescence (CL), the optical radiation emitted into the far field by a polarizable material upon excitation by a high-energy free electron, was observed long before the electron itself was known. The glow in the vacuum tubes used to study the cathode rays was the first example of CL, hence the name [91]. In essence, the electron beam is a natural source for optical excitation [92]. Just as for light, it creates a time-varying electric field that can interact with a medium, but unlike continuous wave (CW) light where the field is varying at a fixed frequency, the electron is a point charge with a field evolution that is tightly confined in space and time. Additionally, while a plane wave has $E$ and $H$ fields perpendicular to the trajectory of the light, the electron has an electric field component along the trajectory ($E_\parallel$), and radially perpendicular to it ($E_\perp$), with $H$ being azimuthally oriented. In vacuum, the electric fields of a moving electron correspond to a single oscillation, corresponding to broadband frequency components spanning the UV, visible and infrared spectral regimes. This is shown schematically in Figure 1.3. In a given medium the electron-matter interaction will become more complex due to dispersion and absorption, lengthening the time-evolution and reducing the spread in frequency. Nonetheless, CL retains a broadband character, making it a suitable candidate to study a wide selection of materials and structures.

Fast electrons carry large energies and momenta, allowing them to interact with a sample through a multitude of mechanisms and leading to several different radiation processes. We can divide these into coherent and incoherent categories [92]. Figure 1.4(a) shows a schematic of electron-sample interaction, with CL being emitted to the far field carrying information about the radiative properties. Transmitted electrons contain information about all loss mechanisms, both radiative and non-radiative, which can be determined by electron energy loss spectroscopy (EELS). In this thesis we will only discuss CL.
1.3.1 Cathodoluminescence processes

Coherent radiation

The evanescent electric fields of the moving electron can coherently couple to far field radiation when interacting with a polarizable material. The main types of coherent radiation are Cherenkov radiation (CR), diffraction radiation, transition radiation (TR), and the generation of plasmons. The emission is called coherent because it has a fixed phase relation with the fields of the impinging electron. The different coherent mechanisms can also interfere with each other [92].

Cherenkov radiation occurs when the velocity of the electrons exceeds the phase velocity of light in the medium, allowing the evanescent fields of the electron to couple to far field emission [93, 94]. CR is emitted along the forward direction, in a cone with a well defined angle determined by the electron velocity and the refractive index of the material. This process is famous for causing the blue glow in
nuclear reactors and is also used to detect subatomic particles, e.g. neutrinos.

Diffraction radiation occurs when an electron passes close to a structured surface without crossing it [95]. This is also named Smith-Purcell emission for the special case of an electron moving parallel to a grating or row of (nano)particles [96].

Transition radiation is perhaps the most general of the processes, occurring whenever an electron (or other charged particle) transits the interface between two different media [92, 97, 98]. The moving charge polarizes the media near the interface, leading to surface currents and charges that radiate into the far field. A simplified explanation in the case of metals is that the electron produces an image charge, as shown schematically in Figure 1.4(b). In all cases, the excitation corresponds very closely to a time-varying vertical electric dipole radiating into the far field, leading to a toroidal angular emission pattern (blue line in Figure 1.4(b)). Due to the highly localized excitation of the electrons and their large momenta, they can directly excite guided waves that require high in-plane momentum, such as surface plasmon polaritons (SPPs) at metal interfaces [99] (in red in Figure 1.4(b)) or waveguide modes in dielectric/semiconductor waveguides.

Finally, the evanescent electric fields of the electron can couple to localized electromagnetic modes, such as plasmon resonances [100–102] or Mie resonances [103]. Only the (induced) field parallel to the electron trajectory, at the position of the electron, can do work and produce losses such as light emission. In this way, the electron will transfer most of its energy under the influence of vertical electric fields (the electron beam is vertically oriented in the SEM). Because of this, electron beam excitations are highly sensitive to vertically polarized or oriented modes, such as SPPs, transverse magnetic (TM) waveguide modes and localized modes with a strong vertical electric field component. Field components perpendicular to the electron trajectory can also contribute however, as they can polarize the sample or structure and induce fields parallel to the trajectory that can then act back on the electron. The CL probability is, to first order, proportional to the radiative local density of optical states (LDOS) integrated along the trajectory, but it can be affected by other components and positions that act back on the electron. In certain cases, such as a high quality waveguide for example, light may be generated but not collected by the setup, leading to a dark CL signal despite the presence of a high full LDOS. While EELS is proportional to the full LDOS (both radiative and non-radiative parts), CL probes only the radiative component [104, 105].

Incoherent radiation

Fast electrons can also excite incoherent radiation in a medium, for which there is no fixed phase relation with the electric field of the incident electron. In this case, the incoming electron excites valence electrons in the material to higher energy levels through inelastic collisions [92, 106, 107]. This process can lead to x-ray emission and generates electron-hole pairs that radiatively recombine to emit luminescence in the UV/visible/infrared spectral range. A broad range of excitations, both
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Figure 1.4 – (a) Schematic showing the electron beam spectroscopies that can be used to study a sample when exciting it with a beam of fast electrons. The energy lost by transmitted electrons is measured by EELS, while the generated broadband electromagnetic radiation that is named CL can be measured by optical detection techniques, and comprises different radiation processes. (b) Schematic of coherent CL excitation mechanisms on a metallic sample. The electron can generate transition radiation (blue) and SPPs (red) at the surface. (c) Schematic of incoherent CL excitation in a semiconductor, where the electron can generate electron-hole pairs that radiatively recombine. The random emission direction inside the material combined with the narrow escape angle leads to a Lambertian angular emission pattern.

above and inside the band gap can be probed in this way: band edge recombination in semiconductors, exciton recombination, intermediate defect or donor/acceptor states and traps as well as intra-4f transitions in rare-earth ions can be probed by CL [108–111]. CL is often used to study the dependence of these radiative processes as a function of temperature, local environment, dopant or defect concentration, stress and strain, among other quantities [107, 112–117].

The electron beam is a powerful method to provide nanoscale information about the optical and electronic properties of luminescent materials. By choosing the electron beam energy one can control the lateral extent of the beam as well as the penetration depth, allowing for 3D characterization within certain limits, and determining electronic properties such as carrier diffusion or surface recombination velocities [107].

Figure 1.4(c) shows a schematic representation of electron-hole pair recombination in a semiconductor, with the radiation being emitted isotropically inside the material. A majority of the light will be internally reflected, as only the portion emitted within the critical angle can escape. The emitted intensity has a cosine dependence on the zenithal angle due to refraction at the interface, resulting in a Lambertian angular emission pattern (shown in green) [118, 119].

Due to the large momentum of the electron and its azimuthal symmetry, it can
efficiently excite transitions that are symmetry- or momentum-forbidden for plane wave excitation. In addition, the impact excitation cross sections for electrons are often higher than corresponding optical excitation cross sections [107], and a single incident electron can excite multiple electron-hole pairs, as the necessary ionization/excitation energy is typically only a few times the band gap energy [120, 121]. For most materials the band gap is on the order of a few eV, while the incident electron will usually have an energy of several keV. The large number of slower secondary electrons created by the primary electrons are also usually energetic enough to excite transitions in the optical range. All of these effects contribute to an emission probability for incoherent radiation that is usually several orders of magnitude higher than for coherent radiation.

1.3.2 Electron-matter interactions

Optical characterization of nanoscale samples using CL is advantageous for several different reasons. The technique is contactless, usually damage-free and broadly applicable to a wide range of materials. The only limitation for samples is that they must be sufficiently conductive to carry away the charge from the electron beam. Insulators can be prepared with thin conductive layers (e.g. carbon or gold) in order to circumvent this issue. Next, the large number of coherent and incoherent processes that take place over a broadband wavelength range allow for studying a wide scale of radiative mechanisms and the interaction between them. Additionally, measurements are usually performed inside an electron microscope, allowing for great control in the electron beam energy, current, spot size, positioning, and the possibility to correlate structural and optical properties. Finally, perhaps the most important advantage is the high spatial excitation resolution, which allows for deep-subwavelength characterization.

The achievable spatial resolution depends on multiple parameters. Both the electron beam itself and the interaction with the sample play an important role. For the beam, an obvious variable is the spot size of the focus at the sample surface, which depends on the beam energy and current (increasing for larger current). Typical diameters for the beam spot on the sample surface are 1–20 nm.

In addition, the evanescent electric field profile of the electron plays an important role, especially for coherent processes, as it extends away from the beam, increasingly so for larger energies. The field amplitude diverges at the electron position and falls off as a modified Bessel function of the second kind (0th-order for the component parallel to the trajectory and 1st-order for the perpendicular component) [92]. Due to the high gradient of the fields close to the electron trajectory, there is a large contrast in the interaction strength in that region. For larger distances, we can use the asymptotic behavior of the Bessel functions for large values and define a characteristic field extent: the Bohr cutoff \( \nu \gamma / \omega \) [92]. Here \( \nu \) is the electron velocity, \( \gamma \) is the Lorentz factor and \( \omega \) is the frequency. The corresponding distance for the intensity is \( \nu \gamma / 2 \omega \) [123, 124]. From this formula we can determine an “effective” diameter of the beam (twice the intensity cutoff), finding \( \sim 33 \text{ nm} \) for
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Figure 1.5 – (a) Primary electron beam trajectories in different materials calculated with the Monte Carlo simulation software Casino [122] for $10^4 30$ keV electrons impinging on Au, Si and PMMA. The superimposed partially transparent trajectories give a representation of the electron-matter interaction volume, shape and density. (b) Calculations as in (a), for Si at 5, 15 and 30 keV. (c) Histogram of the average number of electrons per $2 \times 2$ nm bin as a function of depth and radius for the 5 keV electron beam in Si. (d) Cross-cuts of (c) for different depths, showing the average number of electrons per bin as a function of radius, normalized to the same scale for comparison of the electron beam divergence (curves are multiplied by the factors shown in the figure). Both the color bar of (c) and the vertical axis of (d) correspond to the average number of electrons per bin.
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a 30 keV electron and ~118 nm for a 300 keV electron, at a wavelength $\lambda_0 = 600$ nm. Due to the divergence of the fields at the electron position, the resolution will be higher than implied by the Bohr cutoff. The field extent can be used as an advantage however, as it allows for aloof excitation of nanostructures without the electron trajectory actually passing through the structure.

Once the beam enters the material, the interaction volume of the electrons with matter will also affect the resolution, increasing the lateral and depth ranges from which radiation is being emitted. The direct interaction region due to the primary incident electrons only represents a lower bound to this volume, since both charge carriers and photons from CL emission processes can diffuse and be recycled over larger distances [107, 125]. Determining an exact spatial excitation resolution is a complex affair necessitating a convolution of all these parameters, which is strongly dependent on the beam and sample properties.

To exemplify the importance of electron-matter interactions on the resolution and extent of CL emission, we use the Monte Carlo method based software Casino [122] to calculate electron trajectories in different materials, as shown in Figure 1.5. We simulate $10^4$ electrons in each case, distributed as a 10 nm wide Gaussian incident beam, and plot all of the primary electron trajectories for different electron energies and materials. In (a) we compare Au, Si and polymethyl methacrylate (PMMA, a low density polymer used in lithography) for 30 keV electrons. We clearly see the interaction volume increasing in size by an order of magnitude and changing in shape as we go from the metal to the polymer, due to the lower material density and atomic number which both affect the electron scattering. The pear shape we see for PMMA and Si is well-known for low-atomic-number materials ($Z \leq 15$) while the hemisphere observed for Au is characteristic of large atomic numbers ($Z \geq 40$) [107]. In between these two regimes a spherical shape is expected.

In addition to the cloud formed by the primary electrons, the backscattered electrons also play a role in the resolution, as they can cover a non-negligible area when scattering out of the material. In the case of Au ~50 % of the electrons are backscattered, for Si this is ~14 % and ~4 % for PMMA. This is in agreement with the fact that materials with higher atomic number backscatter more primary electrons. The cascade of secondary electrons generated by the incident beam can also affect the interaction volume as they can diffuse through the material and excite incoherent processes, but typically they do not extend much further than the primary electron cloud due to their low energy.

Figure 1.5(b) compares 5, 15 and 30 keV electron trajectories in Si. In this case the shape of the interaction volume does not change strongly (we note a slight change due to the saturation of the trajectories) while a tenfold increase in the extent of the electron penetration is observed when comparing 5 and 30 keV.

In order to quantify the distribution of the electrons inside the material in more detail, we study the case for Si at 5 keV more closely, binning horizontal planes for each depth and then performing an azimuthal average to obtain the average number of electrons as a function of depth and radius, in $2 \times 2$ nm bins. This is
shown in Figure 1.5(c), in which we clearly see that the majority of the electrons is stopped in a small region very close to the impact position (50–100 nm depth and 25–50 nm radius), with a slower dropoff outside of this region, up to the maximum extent of the interaction volume at ∼400 nm depth and ∼300 nm radius.

Figure 1.5(d) shows cross-cuts of (c) at different depths, in order to visualize the divergence of the electron beam as it moves deeper into the sample. The curves are noisy due to the relatively small number of electrons used. While the full width at half maximum (FWHM) of the beam is ∼10 nm at 10 nm depth (unchanged from the input beam), it increases to ∼20 nm at 50 nm depth and ∼40 nm at 150 nm depth. Therefore electron-matter interactions play an important role in determining the resolution and emission characteristics of CL.

1.4 Outline of this thesis

Cathodoluminescence is a powerful, versatile and high resolution tool for the nanoscale characterization of optical and electronic properties of nanophotonic materials. In this thesis we describe different CL spectroscopy techniques, adding new functionalities to the angle-resolved CL imaging spectroscopy (ARCIS) setup that has been developed previously. This allows us to study different properties of the light emission from a variety of metallic and dielectric nanostructures, discerning the behavior of these nanophotonic structures in even greater detail.

In Chapter 2, we describe the angle-resolved cathodoluminescence imaging polarimetry and spectroscopy setup that is used throughout the thesis, giving an overview of all the different detection schemes available.

In Chapter 3, we develop a theoretical framework to describe the time-evolution of coherent CL processes that occur for an electron impinging on a single planar interface between vacuum and a given material. We show how the incident electron induces electric fields at the interface that can be emitted to the far field as transition radiation or guided along the surface as surface plasmon polaritons.

Chapter 4 shows how angle-resolved measurements can be used to recognize and separate coherent and incoherent CL emission. While metals display only dipolar TR and most luminescent materials exhibit Lambertian luminescence emission, Si shows both processes in a wavelength-dependent fashion that we can quantify and separate.

Chapter 5 introduces angle-resolved CL imaging polarimetry as a new powerful addition to the CL toolbox, allowing us to determine the full polarization state of light being emitted from CL processes. As proof of concept we study plasmonic bulls-eye and spiral antennas that exhibit strongly polarized emission as well as bulk Si and GaAs samples in which we separate polarized and unpolarized contributions.

In Chapter 6, we use CL polarimetry to study vertically grown InP nanowires of different height. We determine that the angular emission is height dependent and is dominated by azimuthally polarized rings. The emission is modeled as an incoher-
ent sum of randomly oriented dipoles radiating above a substrate, indicating that the CL luminescence emission scatters from the top of the wires. The sensitivity of measurements and calculations to the extent of the emission region enables a probe of the carrier diffusion length.

In Chapter 7, we apply CL polarimetry to horizontal GaAs nanowires of different diameter. Polarization-resolved measurements indicate that the emission behavior is dominated by the TM01 waveguide mode for both wires. The difference in diameter leads to a difference in modal dispersion, creating a subtle transition between leaky behavior and guided behavior. This is demonstrated by a change in directionality when exciting the nanowire end facets.

Chapter 8 describes CL spectroscopic tomography, a new technique that allows the 3D characterization of optical properties at the nanoscale. We study a metal-dielectric resonator, measuring 2D CL maps for different orientations and using the method of filtered back projection to reconstruct a 3D image of the CL emission. This allows us to discern material luminescence and plasmonic modes that are spatially and spectrally separated.

Finally, in Chapter 9, the CL techniques previously used in (and near) the visible spectral range are applied to the near-infrared spectral range and used to study modal fields in Si photonic crystal waveguides. We discern sharp resonances that are very strongly linearly polarized and exhibit distinct spatial distributions. The dominant emission feature corresponds to the odd (antisymmetric) TE waveguide mode of the system, demonstrating that we can couple to high-quality in-plane modes.

This thesis provides fundamental new insights into electron-induced light emission processes. It cements the status of CL spectroscopy as an essential tool in the nanophotonic and materials characterization arsenal, able to elucidate many important degrees of freedom of light at the nanoscale, for a wide range of structures and applications.