Probing light emission at the nanoscale with cathodoluminescence

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Quantifying coherent and incoherent cathodoluminescence in semiconductors and metals

In this chapter we present a method to separate coherent and incoherent contributions to cathodoluminescence from bulk materials using angle-resolved cathodoluminescence spectroscopy. Using 5 and 30 keV electrons we measure the cathodoluminescence spectra for Si, GaAs, Al, Ag, Au, and Cu. We then determine the angular emission distributions for Al, GaAs, and Si. Aluminium shows a clear dipolar radiation profile due to coherent transition radiation, while GaAs shows incoherent luminescence characterized by a Lambertian angular distribution. Silicon shows both transition radiation and incoherent radiation. From the angular data we determine the ratio between the two processes and decompose their spectra. This method provides a powerful way to separate different radiative cathodoluminescence processes, which is useful for material characterization and in studies of electron-matter and light-matter interactions in metals and semiconductors.

4.1 Introduction

Cathodoluminescence (CL), the radiation excited by a ray of fast electrons, was first studied during the development of cathode tubes [80, 91]. More detailed stud-
ies proliferated after the development of scanning electron microscopes (SEMs), first with a focus on mineralogy and petrology to identify geological samples by examining mineral-specific luminescence [172, 173], later encompassing materials science in general [106, 107, 174]. One can study (band gap) luminescence and other electron transitions across a broad range of energies [108, 175, 176]. The luminescent properties can be used to examine often inaccessible details such as variations in the local composition, local dopant concentration, stress and strain, interfaces, and non-radiative recombination centers such as point or extended defects [111–114]. One can also create and excite such defect states using electron irradiation to study their nature and behavior [116, 117, 177, 178]. Cathodoluminescence studies of nanoscale structures are on the increase as well [109, 110, 179, 180].

In the last decade, CL has gained attention among the nanophotonics community, mostly centered on studies of plasmonic systems, although studies on dielectrics are proliferating. Measuring with a nanoscale excitation probe, especially when combining spectral and angular data, turns CL into a very powerful tool. Optical antennas [26, 101, 102, 130, 181–184], plasmonic nano-cavities [185, 186], waveguides [187–189], and periodic crystals [124, 135, 136] among others have been examined to study their dispersion, radiation profiles, and spatial modal distributions.

A high energy electron beam can generate radiation in a material through a multitude of processes, which can be separated into coherent and incoherent groups [92]. Coherent radiation, so-called because the emitted radiation has a fixed phase relation with the electric field of the incident electron, comprises transition radiation (TR) at the surface, generation of plasmons, and Cherenkov radiation (when applicable). These processes can be used to probe the electromagnetic behavior of nanoscale objects with great precision, but are often quite weak. In metals, TR and plasmon generation are usually the dominant processes. Incoherent radiation such as luminescence generated by electron-hole recombination in semiconductors is usually much stronger and does not interfere with coherent radiation.

CL measurements for material science have generally consisted of spectral measurements, which are very powerful in determining characteristic optical resonances and transitions for a given material. If different radiative mechanisms are at play, however, it is often not possible to separate them. Here, we present the use of angle-resolved CL spectroscopy to separate fundamental CL processes by their characteristic angular emission distributions. We investigate coherent TR and incoherent luminescence that each have very distinctive emission patterns, allowing us to discriminate between them and characterize them separately. We study Al, Ag, Au, and Cu that show strong TR as well as GaAs which shows strong incoherent luminescence. We then focus on partitioning TR and incoherent emission in Si, where we find that both mechanisms strongly contribute to the CL radiation.
4.2 Experiment

We performed measurements on polished p-type (B doping level $10^{15} - 10^{16}$ cm$^{-3}$) and n-type (P doping level $10^{15}$ cm$^{-3}$) single-crystal Si $\langle 100 \rangle$ wafers. No significant differences were found in CL measurements for these two sample types. A single-crystal wafer of Czochralski-grown Al was used to study TR and to characterize the system response of our setup. Thermal evaporation was used to grow layers of Au, Ag, and Cu on a silicon substrate. We used evaporation rates of 0.5 Å/s at a chamber pressure of $\sim 10^{-6}$ mbar. In each case the metal layers are at least 200 nm thick, such that they are optically thick. Finally, a single-crystal slab of GaAs was used as a model for a strongly incoherent emitting material. The dielectric functions of the metal films were measured using variable-angle spectroscopic ellipsometry and compared to values from Palik [190] or Johnson & Christy [191].

The experiments are all performed at room temperature in our Angle-Resolved Cathodoluminescence Imaging Spectroscopy (ARCIS) setup [128]. This consists of a FEI XL-30 SFEG scanning electron microscope (SEM) in which we place an aluminium paraboloid mirror that can be precisely positioned with a piezoelectric micromanipulation stage. We use the focused electron beam to generate radiation in our samples; the mirror collects the emission and directs it out of the microscope to an optical detection system. For spectroscopy measurements we focus the light onto a fiber connected to a spectrometer with a liquid-nitrogen-cooled Si CCD photo-detector. Alternatively, we can image the parallel beam emanating from the paraboloid mirror onto a 2D Si CCD camera, which allows us to determine the angular emission profiles of the emitted radiation [128]. In this case each emission direction from the sample will hit the mirror at a specific location and be directed onto a specific point of the CCD camera. The 2D image is then transformed into a far-field angular radiation pattern. For the angular measurements we use color filters to select certain free-space wavelength ranges (40 nm bandwidth filters, from $\lambda_0 = 400 - 900$ nm in 50 nm steps).

The spectral measurements on the single-crystal Al, evaporated Au, Ag, and Cu were performed at a beam energy of 30 keV and a current of approximately 15 nA. The integration time varied between 0.5 and 4 s. Measurements on single-crystal Si were performed at 5 and 30 keV, with the same nominal current. Data from GaAs was collected at 30 keV, but since the band gap luminescence is extremely bright, we used a much lower current of roughly 0.15 nA. CL count rates were linear with beam current in all cases. We subtract the dark spectrum measured with the electron beam blanked to correct the spectral data, which accounts for thermal and readout noise of the detector. During the measurement we scan the beam over a $200 \times 200$ nm square area, in 20 nm steps. A spectrum is measured for each pixel and the data is then averaged. We find that measurements taken on different locations on the samples are very consistent. The correction to account for the spectral sensitivity of the system is described further on. For angular measurements, the same currents and energies were used as for the spectral measurements, while the integration times were 60 s for Al and Si, and 1 s for GaAs. For the angular data...
Figure 4.1 – (a) Schematic angular emission profile for electron-beam induced radiation from a metal, in which TR is dominant. The cartoon on the right sketches this process, where the electron creates an image charge in the metal, giving rise to a vertical dipole at the surface which emits radiation with a toroidal angular shape. (b) Schematic angular emission profile for incoherent luminescence generated inside the material with a Lambertian emission profile. The cartoon on the right shows electron-hole recombination emitting light isotropically, only light emitted within the critical angle escapes from the sample. (c) Schematic emission profile for a combination of TR and luminescence, which is the case for Si. The profile is an average of those from (a) and (b).
we took 2-3 measurements for each filter wavelength in order to average them and each measurement is corrected with a dark measurement.

4.3 Results and discussion

A beam of highly energetic electrons can transfer its energy to a material or structure in different ways, leading to a variety of radiative and non-radiative processes. Figure 4.1 provides an overview of radiation processes one commonly encounters in most materials. The typical behavior of metals is shown in (a), where coherent processes such as TR and generation of surface plasmons polaritons (SPPs) are dominant [92, 99]. Due to fast non-radiative recombination of the free electrons, the beam does not tend to excite incoherent luminescence in metals. SPPs can be excited efficiently on a flat surface, but as they cannot radiate to the far field for an unstructured planar surface, the only contribution to measured radiation is from TR, which has a toroidal emission pattern similar to that of a vertical point dipole at the surface as shown in Figure 4.1(a) [92, 99, 128]. The cartoon on the right shows a simplified visualization of this process: the negatively charged electron induces a positive mirror charge in the metal that disappears when the electron transits the interface. The corresponding varying dipole moment then leads to radiation into the far field with an angular emission profile very similar to that of a radiating point dipole placed just above the metal surface. For dielectrics TR generation occurs as well, a polarization charge replacing the image charge, with the dielectric constant determining the magnitude [92].

In the case of many semiconductors and dielectrics, incoherent luminescence is the main source of radiation as it is usually orders of magnitude stronger than coherent emission such as TR. A schematic of such a luminescent material is shown in Figure 4.1(b). The energetic electron can excite a material to a range of excited states over a very broad spectral range. The impact excitation cross sections for these transitions are higher than many optical excitation cross sections [107]. Because of the high incident energy and the formation of an electron cascade, a single incident electron can lead to multiple material excitations. Creation of an electron-hole pair by an incident electron typically requires a few times the energy of the band gap [120, 121], so excitations in the visible and infrared can be generated by both the primary and secondary electrons. The low-energy secondary electrons and decelerated incident electrons have higher excitation cross sections than the primary electrons, as their localized fields can couple more strongly to such excitations than the more delocalized fields of fast electrons [92]. As this kind of CL radiation is caused by spontaneous emission, it is not coherent with the electric field of the incident electron and will not interfere with radiation that is coherent such as TR. The emission is usually due to the radiative recombination of electron-hole pairs and excitons which can recombine to the ground state or to intermediate excited defect states, which then decay to the ground state through radiative or non-radiative pathways. Incoherent emission typically occurs isotropically inside
Figure 4.2 – (a) Measured cathodoluminescence spectra from bulk samples of single crystalline Al, GaAs, and Si. Data was taken at 30 keV; for silicon also at 5 keV. The beam current for Al and Si was 15 nA, for GaAs 0.15 nA. The GaAs spectrum is divided by a factor of 20. (b) Calculated TR emission probability as a function of wavelength for Al, GaAs, and Si. (c) The spectra of Al, GaAs, and Si corrected by the system response using the TR data for Al as a reference. In this case the GaAs spectrum is divided by a factor of 3000.

a material. The resulting CL emission distribution exiting the material is Lambertian, with a cosine dependence on the zenithal angle, as shown in Figure 4.1(b). The cosine dependence occurs due to the refraction of light and follows directly
from Snell’s law [118]. The cartoon in Figure 4.1(b) illustrates these processes and also indicates the critical angle beyond which radiation is fully reflected into the substrate. Figure 4.1(c) shows a schematic of the emission pattern determined by a combination of TR and Lambertian profiles. Next, we present the experimental spectra and angular emission profiles for each of the three cases described here. We use Al as a TR emitter, GaAs as a strong incoherent emitter and Si representing both effects.

Figure 4.2(a) shows the CL spectra from bulk crystals of Al, GaAs, and Si at 30 keV. Data for Si at 5 keV is also shown. We observe that the Al and Si spectra show similar, broadband spectral shapes while the GaAs spectrum is much sharper and peaks at about $\lambda_0 = 870$ nm, corresponding to the band gap energy ($\sim$1.43 eV, or $\sim$867 nm, at 300 K).

Figure 4.2(b) shows the calculated TR spectra for the same three materials, where the TR intensity is expressed in units of photon emission probability per incoming electron per unit bandwidth. The calculation is based on the theoretical formalism described in section IV.C of Ref. [92]. In this approach Maxwell’s equations are solved for a swift electron interacting with a material, more specifically the case of an electron normally incident on a planar substrate. The moving charge induces surface charges and currents that lead to a reflected electromagnetic field at the surface that is the source of TR. The emitted TR is angle and wavelength dependent, so one can obtain angular intensity distributions and determine the total spectrum by performing the angular integral over the upper hemisphere. The variables that are of importance for the wavelength and amplitude dependence of TR are the electron energy, beam current and material permittivity. The electron energy affects the TR amplitude because the electric field of a higher energy electron extends further from its trajectory. As a result, a larger volume of material is polarized, inducing more surface currents and increasing the TR response. The TR intensity is given by an emission probability per electron, so the signal increases linearly with the number of electrons. In this way the beam current only affects the amplitude and does so in constant fashion for all wavelengths leading to a fixed factor difference in the spectrum.

As far as the wavelength dependence is concerned, TR is an interface effect based on the reflection of induced fields, so the equations contain information about light dispersion in both media, in a way similar to that of the Fresnel equations. Since in our case one medium is vacuum, the material permittivity of the sample determines the wavelength dependence of TR. Spectral features can be correlated with features in the permittivity. We use optical constants measured by ellipsometry for Al and an average of tabulated values for Si and GaAs. The inset in Figure 4.3 compares the real and imaginary part of the permittivity of Al that we measured by ellipsometry with values from Palik [190]. The trends are similar, but the absolute values of both real and imaginary parts of the permittivity differ; we attribute this to differences between the density and crystallinity of our single crystal compared to samples used by Palik. We can see that the calculated spectra for all three materials follow the same trends as their dielectric function. The TR spectra
of GaAs and Si are quite similar, in agreement with the similar permittivity. We also note that using a lower electron energy leads to a lower TR emission probability for Si.

As the CL signal from Al is purely due to TR, we now use it to calibrate our setup and determine the (relative) system response due to the spectral sensitivity of the setup. This will allow us to normalize the other experimental spectra. We obtain this system response by dividing the theoretical TR spectrum by the measured spectrum from the single crystal Al. We can then multiply the other measured spectra by this correction factor to obtain the emission probabilities for the other materials.

Figure 4.2(c) shows the corrected CL spectra for Al, GaAs, and Si. Clearly, the corrected Si spectrum at 30 keV does not correspond to the theoretical TR spectrum in Figure 4.2(b) at all, as the spectral shape is quite different and the intensities are 2 – 12 times higher than the TR spectrum. At 5 keV, the corrected spectrum for Si also exceeds the TR spectrum. It is clear that the Si spectrum cannot be explained as being only due to TR and since Si is a semiconductor, incoherent radiative processes must play a role even if non-radiative recombination is dominant. We do not expect Cherenkov radiation to play a role even though the refractive index is high enough to satisfy the emission condition, because it is emitted in the forward direction downwards into the substrate where it is fully absorbed.

In Figure 4.3, we examine the CL spectra of Au, Ag, and Cu, for which we expect the spectrum to be dominated by TR. The measured spectra are corrected using TR data from Al in the same way as above. Theoretical TR spectra of Au, Ag, and Cu are also shown as comparison. Several trends can be observed. First of all,
the experimental TR spectra for Au, Ag, and Cu have quite similar intensities, with clear kinks in the spectra for Au and Cu at $\lambda_0 = 500$ and $550$ nm respectively. The theoretical spectra show similar trends, the kinks for Au and Cu occur at the same wavelengths as for the experimental spectra. The absolute emission probabilities do not agree well between experiment and theory; they differ by up to $\sim 30\%$. We attribute this to variations between measurement sessions of the beam current, as explained in the description of Figure 4.2(b), as well as changes in the system alignment that affect the collection efficiency and thus the intensity. Repeated measurements with the same sample and measurement conditions have shown one can indeed obtain up to $\sim 30\%$ variations in intensity. Because all of the data is normalized to the intensity of Al, differences in current compared to that of the reference measurement will lead to an offset factor in the spectrum. In this case the current was higher for the measurements than for the reference, so the experimental spectra are a factor higher than the theoretical values. These results show that overall, the experimental data well represent the theoretical spectral features.

Next, we study the angular emission profiles for Al, GaAs, and Si at 30 keV. We find that the radiation profiles are azimuthally symmetric and average the data over an azimuthal range to obtain the polar profiles shown in Figure 4.4. Averaging was done over the azimuthal angle ranges $\phi = 60 - 120^\circ$ and $\phi = 240 - 300^\circ$, where $\phi = 0/360^\circ$ is the center of the mirror's open end and $\phi = 180^\circ$ corresponds to the apex at the back of the mirror. We use these ranges to avoid the open end of the mirror and the apex which contains more aberrations. To further decrease the noise for Al and Si we average the data obtained from the two angular ranges. All angular distributions are normalized to 1; no data is collected in the angular range $\theta = \pm 5^\circ$, corresponding to the hole in the parabolic mirror. The angular resolution is affected by the curvature of the mirror which modifies the solid angle of the emitted radiation compared to its distribution on the CCD camera. As described in Ref. [128] (Fig 2.c), the solid angle per pixel varies between 2–10 $\times 10^{-5}$ sr.

Figure 4.4(a) shows angular profiles for Al (at $\lambda_0 = 400$ nm) and GaAs (at $\lambda_0 = 850$ nm) together with theoretical curves for TR (Al) and a Lambertian emitter (GaAs). For Al, the measured and calculated data agree very well, with the experimental one being slightly broader, proving the emission from Al is well described by TR. The emission pattern from GaAs corresponds well to the Lambertian profile, confirming that CL from GaAs at the band gap energy is dominated by incoherent luminescence.

Figures 4.4(b–d) show the experimental angular profiles for Si at 30 keV, measured at $\lambda_0 = 400$, 550, and 900 nm, respectively. Clearly, at $\lambda_0 = 400$ nm the emission pattern is more TR-like while it becomes more Lambertian-like and thus dominated by luminescence for the longer wavelengths. The angular emission from GaAs displays a similar trend (see Figure 4.6 of the Supporting information).

For the case of incoherent luminescence it is important to keep in mind that carrier transport can play a role in determining the emission properties. Diffusion as well as photon recycling can lead to recombination well outside the area of initial generation by the electron beam. Additionally, carrier transport can be anisotropic,
Figure 4.4 – (a) Measured normalized emission patterns as a function of polar angle $\theta$ for Al and GaAs (solid lines, measured at 400 and 850 nm, respectively). The theoretical TR pattern for Al and a Lambertian pattern for GaAs are also shown (dashed lines). (b–d) Measured emission patterns of Si at 30 keV for 400, 550, and 900 nm (blue lines) shown together with fits consisting of a combination of Lambertian and TR patterns (red lines). All measurements shown here were performed with a 30 keV electron energy and the resulting data is azimuthally averaged.

further impacting the distribution of recombination and thus affecting the resulting spatial and angular CL profiles [125]. In our case there is very good agreement with the Lambertian profile, so we expect that these effects play a minor role.

To determine the relative contributions of the two processes and separate them, we model the emission pattern as a linear combination of TR and Lambertian profiles for the given wavelengths, with the relative contributions as fit parameters in a least squares fitting routine. The fitted angular profiles are shown in red in Figures 4.4(b–d) and agree well with the measured data. Next, we extend this analysis to the full 400–900 nm spectral range in steps of 50 nm, both for 30 and 5 keV electron energies. The relative contributions of TR and incoherent radiation are then determined from the fits for each wavelength; the result is shown in Figure 4.5(a). TR dominates at shorter wavelengths, while incoherent emission dominates at longer wavelengths. Similar trends are observed for 5 and 30 keV. The transition in dominance between the two radiative mechanisms is due to a combination of effects. TR has an increased intensity at shorter wavelengths as one can see from the calculation in Figure 4.2(b), while luminescence which is emitted inside the material will be absorbed much more strongly for short wavelengths than for long wavelengths, so more “red” luminescence will escape the Si. GaAs exhibits similar behavior, but with stronger relative contributions for both processes at ei-
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Figure 4.5 – (a) Relative contributions of TR and luminescence derived from fits to the Si emission patterns as in Figure 4.4, both for 5 and 30 keV electron energy (circles). The drawn lines are a guide to the eye. (b) The CL spectrum from Figure 4.2(c) (black) together with TR (blue) and incoherent luminescence (red) contributions for Si at 30 keV derived using the fractions from (a). The theoretical TR spectrum for Si at 30 keV is shown as well (blue dashed line).

Now that we have determined the relative contributions of these two radiative processes in Si, we can use this information to decompose the TR and incoherent luminescence spectra. We fit a smooth curve through the data points in Figure 4.5(a) and use this to partition the experimental spectrum for Si at 30 keV from Figure 4.2(c). The total spectrum for Si at 30 keV as well as the separated TR and incoherent contributions are shown in Figure 4.5(b). Comparing the experimentally determined TR contribution with the calculation, the overall behavior as a function of wavelength is well reproduced, while the absolute intensities differ by a factor \( \sim 1.5 \) which we attribute to a difference in beam current, as was discussed earlier.

Figure 4.5(b) shows that the incoherent Si spectrum is spectrally broad, peaks for \( \lambda_0 > 750 \) nm and extends above the TR spectrum for \( \lambda_0 > 470 \) nm. We attribute this incoherent spectrum to transitions between defect states in the direct
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band gap. Since n-type and p-type samples gave similar results, doping-related luminescence is insignificant. We note that light emission is strongly absorbed in Si, especially in the blue, so the collected spectrum does not directly reflect the emitted incoherent spectrum. Correcting for this effect the relative contribution emitted in the blue spectral range is larger than what is observed in the measured spectrum.

Our data can be compared with experiments at 200 keV performed by Yamamoto et. al. [192] at 200 keV in which the CL spectrum from Si closely follows the calculated TR spectrum, with no discernible incoherent radiation. This is due to the fact that the TR intensity is \( \sim 6 \) times stronger at 200 keV than at 30 keV. Moreover, at 200 keV the penetration depth of the electrons is much larger than at 30 keV (up to \( \sim 200 \) \( \mu \)m versus \( \sim 10 \) \( \mu \)m, as determined using Casino) [193]. Since the incoherent radiation is generated more efficiently as the electrons have decelerated deeper inside the material, it will be strongly absorbed inside the Si for higher electron energies.

4.4 Conclusions

We demonstrated a method to distinguish coherent and incoherent cathodoluminescence processes induced by a beam of fast electrons. We have shown that Al exhibits coherent transition radiation, while GaAs exhibits mainly incoherent band gap luminescence. Si cathodoluminescence is composed of both transition radiation and incoherent radiation. We distinguish the two processes by their characteristic angular profiles, namely dipolar-like lobes for transition radiation and a Lambertian angular distribution for incoherent luminescence. For silicon at 5 and 30 keV, transition radiation dominates around \( \lambda_0 = 400 \) nm, making up \( \sim 70 \% \) of the signal while incoherent luminescence becomes increasingly stronger for longer wavelengths, consisting of \( \sim 85 \% \) of the signal at \( \lambda_0 = 900 \) nm. Determining the relative strengths of these two effects allows us to decompose the experimental Si cathodoluminescence spectrum to retrieve the spectrum due to transition radiation, which agrees with calculations and the spectrum due to luminescence, which is very broadband.

Using angle-resolved cathodoluminescence to identify, separate and characterize different coherent and incoherent radiative processes is a powerful way to quantify such different forms of radiation in a multitude of materials such as metals and semiconductors. The technique is quite flexible in separating different radiative mechanisms, so long as one measures processes that do not interfere with each other (or do so in a way that can easily be deconvoluted) and have differing angular distributions. The use of antennas, (nano)structured surfaces or non-planar surfaces can all modify the coherent or incoherent distributions, but often in ways that are predictable by calculation or simulation. One can then use the modified angular patterns to separate the processes. For example, a luminescent sample with a hemispherical surface will not display a Lambertian but a hemispherical
angular distribution due to incoherent luminescence. Alternatively, one could separate the coherent emission of an antenna from the luminescence of the substrate. The presented results are relevant for material characterization and for studies of electron-matter and light-matter interaction in general.

4.5 Supporting information

Transition radiation (TR) occurs for electrons impinging on any material, so long as there is a contrast in the permittivity $\epsilon$. Examining the calculated TR emission probability in Figure 4.2(b), we note that the theoretical spectra for Si and GaAs are very similar. It is thus to be expected that GaAs exhibits TR in a similar way to Si. Angle-resolved measurements on GaAs for an electron energy of 30 keV indeed display the characteristic dipolar angular emission distribution of TR, as shown in

Figure 4.6 – Measured normalized emission patterns as a function of polar angle $\theta$ for GaAs (blue lines) and Si (red lines), for wavelengths of 400 (a), 550 (b), and 900 nm (c). Measurements were performed with a 30 keV electron energy and the data is azimuthally averaged. The black lines close to $\theta=0$ denote the angles for which we do not collect any emission with the mirror.
Figure 4.6. Here we compare the normalized emission intensity of GaAs and Si, averaging over the same azimuthal range used for Figure 4.4 ($\phi = 60 – 120^\circ$ and $\phi = 240 – 300^\circ$). The two materials follow similar emission trends, being dominated by TR at short wavelengths ($\lambda_0 = 400$ nm in Figure 4.6(a)) and by luminescence at long wavelengths ($\lambda_0 = 900$ nm in Figure 4.6(c)), with a transition in between ($\lambda_0 = 550$ nm in Figure 4.6(b)). We notice, however, that GaAs exhibits stronger extremes for both short and long wavelengths. The angular emission distribution is more dipolar in shape than for Si at both $\lambda_0 = 400$ nm and $\lambda_0 = 550$ nm, while it is more rounded and Lambertian than for Si at $\lambda_0 = 900$ nm.

This difference between the two materials can be viewed more explicitly in Figure 4.7, which shows the relative contribution of TR and luminescence to the total angular emission distribution, determined from fits as described for Figures 4.4 and 4.5. We observe that GaAs has a $\sim 15\%$ higher relative contribution from TR than Si, for wavelengths up to $\lambda_0 = 700$ nm, after which GaAs has a higher contribution from luminescence instead. These differences can explained quite intuitively. The strong dominance of the Lambertian angular profiles at these longer wavelengths is to be expected, since this matches with the spectral range of the band gap emission peak (see Figures 4.2(a,c)). In this range GaAs emits luminescence that is much more intense than TR by several orders of magnitude, whereas for Si the difference in intensity is much smaller. The luminescence of Si is quite broadband, however, contributing over the entire visible spectral range studied here. In contrast, the luminescence from GaAs is much more spectrally confined to the band gap wavelength, so at shorter wavelengths there is less competition for TR and it will play a relatively stronger role.