Optical antennas on substrates and waveguides

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Correcting the Super Polarizability for Antennas in Scattering Environments

In this chapter we extend our method of retrieving the super polarizability of antennas to deal with scatterers placed in non trivial surroundings. We show how to implement radiative corrections to the dipole-quadrupole model, when scatterers are placed near a surface, sphere, or stratified medium, similar to the known correction of dipole polarizabilities by the local density of optical states. We demonstrate how this model allows to interpret near field excitation data of plasmon structures taken on antennas deposited on a high-index substrate that present highly directional emission.

8.1 Introduction

In nano-optics one frequently relies on first developing intuition for the functioning of antennas or Fano-resonant structures when they are in isolation, to then explore the functioning of these structures in more complex geometries. Examples are the placement of antennas in arrays, or the placement of antennas on dielectric interfaces or stratified media. Regarding the latter example, the applications based on antennas placed on substrates are abundant. For instance, plasmon antennas are used on dielectric interfaces such as glass cover slips in microscopy experiments [1–4] and sensing applications, or on high index semiconductors, for example in LEDs (III-V substrates), and solar cell applications[5, 6]. Nevertheless, it is well known that the presence of a scattering environment could drastically change the response of the antennas. Therefore, the question arises of how to take this into account for when magneto electric dipolar and quadrupolar responding antennas are placed in such an environment. We solve this
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issue by incorporating back-action effects that modify a scatterer when it is moved into a nontrivial photonic environment, such as the commonly encountered layered medium in LED and solar applications. This leads to a correction equivalent to the powerful “radiation damping corrections” for the dipole model. This radiation damping correction is fundamental to build an energy conserving multiple scattering theory [7–10]. We apply this insight to predict the response of large plasmon scatterers placed on top of a silicon substrate as encountered in recent cathodoluminescence experiments [11]. In particular, we predict strongly directive antenna action for locally excited large scatterers as consequence of the fact that the nearby interface enhances the magnetic and quadrupolar response.

8.2 Back-action correction for hybridization with environments

![Figure 8.1](image.png)

**Figure 8.1:** Sketch of the self driving on an electrically polarizable particle produced by the reflection of fields created by an interface.

Here we ask if an extracted superpolarizability for a complex antenna in isolation can be used as input for a predictive model where the antenna is placed in a complicated environment. In the dipole approximation the environment can be taken into account in the electric dipole polarizability by including induced self interactions produced by the environment, as depicted in Fig. 8.1. In this example an electric dipole

\[ \mathbf{p} = \alpha \cdot \mathbf{E}_{\text{total}}(\mathbf{r}_0) = \alpha \cdot (\mathbf{E}_{\text{in}}(\mathbf{r}_0) + \mathbf{E}_{\text{scatt}}(\mathbf{r}_0)), \]  

(8.1)

is driven in a scatterer with polarizability \( \alpha \) by an electric field \( \mathbf{E}_{\text{total}} \) at the position of the scatterer \( \mathbf{r}_0 \). The total electric field is a sum of the incident electric field \( \mathbf{E}_{\text{in}} \) plus the scattered electric field \( \mathbf{E}_{\text{scatt}} \). In the case of an environment with a substrate the scattered electric field would be the reflected field by the substrate. The scattered electric field created by the dipole, due to the substrate is given by

\[ \mathbf{E}_{\text{scatt}}(\mathbf{r}_0) = \omega^2 \mu_0 (\mathbf{G}_{\text{scatt}}(\mathbf{r}_0, r_0)) \cdot \mathbf{p} \]  

(8.2)
where $\overline{G}_{\text{scatt}}$ is the scattering part of the Green’s function for the electric field. By introducing Eq. (8.2) in Eq. (8.1) we obtain:

$$p - \alpha \omega^2 \mu \mu_0 \overline{G}_{\text{scatt}}(r_0, r_0) \cdot p = \alpha \cdot E_{\text{in}}(r_0).$$ (8.3)

Therefore we can define a corrected polarizability in the following manner:

$$\alpha_{\text{corr}} = (\mathbf{I} - \alpha \omega^2 \mu \mu_0 \overline{G}_{\text{scatt}}(r_0, r_0))^{-1} \cdot \alpha$$ (8.4)

or using the matrix identity $(A \cdot B)^{-1} = B^{-1} \cdot A^{-1}$ we can rewrite it as:

$$\alpha_{\text{corr}}^{-1} = (\alpha^{-1} - \omega^2 \mu \mu_0 (\overline{G}_{\text{scatt}}(r_0, r_0))).$$ (8.5)

The corrected polarizability is equal to the original polarizability of the particle plus a self interaction term that is essentially the scattered Green function of the embedding medium \[12–15\]. This term naturally includes radiation damping and a reactive shift in the polarizability due to the environment. This correction is of large use, as it allows us to quantitatively predict the response of arbitrary antenna geometries in complex backgrounds using as input single building blocks for which the free space polarizability is already tabulated. Here we derive a similar correction for the superpolarizability. In

$$\begin{pmatrix} E \\ H \\ \diamondsuit E \end{pmatrix} = \overline{\alpha}^S \begin{pmatrix} E \\ H \\ \diamondsuit E \end{pmatrix}$$ (8.6)

one should take into account that the driving field $(E, H, \diamondsuit E)$ should not just be the incident field, but also the field that comes back via interaction with the background to the scatterer, as quantified by the scattered part of the Green’s function of the background system. Thus the total field reads

$$\begin{pmatrix} E \\ H \\ \diamondsuit E \end{pmatrix} = \begin{pmatrix} E_0 \\ H_0 \\ \diamondsuit E_0 \end{pmatrix} + \begin{pmatrix} a_1 \overline{G}_E \cdot p + a_2 \nabla \times \overline{G}_H \cdot m + a_3 (\diamondsuit \overline{G}_E^T)^\top \cdot Q \\ \frac{a_1}{i \omega \mu \mu_0} \nabla \times \overline{G}_E \cdot p + a_4 \overline{G}_H \cdot m + \frac{a_3}{i \omega \mu \mu_0} \nabla \times (\diamondsuit \overline{G}_E^T)^\top \cdot Q \\ a_1 \diamondsuit \overline{G}_E \cdot p + a_2 \diamondsuit (\nabla \times \overline{G}_H) \cdot m + a_3 \diamondsuit (\overline{G}_E^T)^\top \cdot Q \end{pmatrix}$$ (8.7)

where in the SI unit system $a_1 = \omega^2 \mu \mu_0$, $a_2 = i \omega$, $a_3 = a_1 / 6$, $a_4 = \omega^2 \varepsilon \varepsilon_0$. This result combines the magnetoelectric Green dyadic \[16\] with the electric field radiated by a quadrupole\[17, 18\] $Q$ that is given by $E(r) = (1/6) \omega^2 \mu \mu_0 \mu (\diamondsuit \overline{G}_E^T(r, r'))^\top \cdot Q$. Equation (8.6) is of the form $\mathcal{P} = \alpha^S \varepsilon_0 + \mathcal{G} \mathcal{P}$, where $\mathcal{P}$ is the generalized induced moment, $\varepsilon_0$ is the driving field, and $\mathcal{G}$ is a generalized field propagator which includes the Green’s function, the curl of the Green’s function as well as its symmetric gradients. Hence the corrected polarizability defined through $\mathcal{P} = \overline{\alpha}_{\text{corrected}}^S \varepsilon_0$ must be of the
familiar form $\bar{\alpha}^S_{\text{corrected}}^{-1} = \alpha^S_{\text{corrected}} - \mathcal{G}$. Explicitly, we define the corrected $\bar{\alpha}^S_{\text{corrected}}$ tensor as

$$\bar{\alpha}^S_{\text{corrected}}^{-1} = \alpha^S_{\text{corrected}}^{-1} - \begin{pmatrix}
\frac{a_1}{i \omega \mu_0} \nabla \times \vec{G}_E & a_2 \nabla \times \vec{G}_H & \frac{a_3 (\hat{\nabla} \times \vec{G}_E)^T}{i \omega \mu_0} \\
\frac{a_4}{i \omega \mu_0} \nabla \times \vec{G}_E & a_2 \hat{\nabla} (\nabla \times \vec{G}_H) & \frac{a_3 (\hat{\nabla} \times \vec{G}_E)^T}{i \omega \mu_0}
\end{pmatrix}
\tag{8.8}$$

where the actual inversion again requires first casting to the 11x11 form as outlined in section 7.2, and also it requires to use the operator $\hat{\nabla}$ which is defined as $\hat{\nabla} \vec{E} = \{ (\partial_x E_y + \partial_y E_x)/2, (\partial_x E_z + \partial_z E_x)/2, (\partial_y E_z + \partial_z E_y)/2, (\partial_x E_x - \partial_z E_z), (\partial_y E_y - \partial_z E_z) \}$. In absence of quadrupolar contributions, this correction reduces to the magnetoelectric radiation damping correction derived for metamaterials by Belov [9] and Seršić [16], which in itself is a generalization of the Sipe-Kranendonk formalism [10]. Note that owing to the required matrix inverse, a nontrivial environment such as a nearby surface can induce magnetoelectric cross coupling [19], as well as mixing of dipolar and quadrupolar excitations. As a final comment, it is very important to emphasize that here we only use the scattered part of the Green’s functions, unlike Ref. [9] and [16]. The retrieved superpolarizabilities already include the self-polarization due to free space and the only additional correction needed is for the polarization created by the reflections and scattering of the new environment which are included in the scattered part of the Green’s function.

### 8.3 Single particle on a Si substrate as directional antenna

We apply the correction of the superpolarizability tensor due to a silicon substrate to a simple experiment recently performed in cathodoluminescence, on Au disks on a Si substrate [11] of height 80 nm and varying diameter up to 180 nm. We retrieve the superpolarizability for the biggest particle with height 80 nm and 180 nm diameter in a vacuum environment, and subsequently we correct $\alpha^S$ with the back-action correction (evaluated at mid-height, i.e., 40 nm above the silicon substrate).

Figure 8.2b and c summarizes as color plots the $\alpha^S$, before and after the correction, taking as wavelength 565 nm, and again scaling the superpolarizability tensor elements (Methods, section 7.2) such that equal value means equal scattered power. As for the dolmen (section 7.3), here we present a logarithmic color plot to facilitate quick identification of relevant entries; the appendix C provides numerical values. In absence of the substrate, the particle response is dominated by a strong electric polarizability along the three principal particle axes. Here $x$ and $y$ are strictly degenerate by symmetry, while the polarizability along $z$ is somewhat lower with a ratio of 0.34 owing to the smaller height. Evidently, the isolated disk has a significant magnetic dipole response and quadrupolar response owing to the big disk size. The particle symmetry,
however, implies absence of bianisotropy. Clearly once the disk is brought to the substrate, the substrate induces cross coupling between different elements, namely bianisotropy linking electric and magnetic dipoles ($p_x$ and $m_y$), and coupling of dipolar and quadrupolar responses.

To gain more insight, we highlight the polarizability tensor elements that are relevant in a typical normal incidence scattering experiment, i.e., when impinging along the ‘z’ direction with an ‘x’ polarized plane wave. The most important elements in the super polarizability tensor that play a role, i.e., $\alpha_{p_x}^{E_x}$, $\alpha_{m_y}^{E_x}$, $\alpha_{m_y}^{H_y}$, and $\alpha_{Q_{xz}}^{E_x}$, are plotted as function of wavelength in Fig. 8.2d (see methods in Sec. 7.2). For the particle in free space, the strongest contribution to the scattering is simply the purely electric dipolar
response $\alpha_{p_x}^E$, with a maximum of $1.8 \times 10^{-2} \, \mu m^3$ at a wavelength of 680 nm. The next important terms, *i.e.*, the magnetic dipole response $\alpha_{m_y}^H$ and electric quadrupole $\alpha_{Q_{xz}}^E$ are 10 to 100 times smaller, while the off-diagonal cross polarizability is another factor of 100 to 1000 smaller still.

Once the scatterer is placed on the substrate, the back action correction modifies this ordering. Fig. 8.2e reveals that the electric polarizability is enhanced and shifted in resonance due to its interaction with the silicon substrate. Remarkably, the crosspolarizability $\alpha_{m_y}^E$ and hyperpolarizability $\alpha_{Q_{xz}}^E$ are enhanced by 3 to 4 orders of magnitude, thus allowing electric fields to much more efficiently excite magnetic dipoles and quadrupoles. The physics is that if initially an electric dipole is induced in the scatterer, its image dipole has sufficient gradient for driving quadrupoles and the magnetic dipole. For the corrected case we present the values of the different elements of $\alpha^S$ in appendix C. On basis of recent experiments and theoretical proposals, it is

![Figure 8.3](image-url)

**Figure 8.3:** a) Emission patterns for a gold cylinder antenna excited with a dipolar emitter located 40 nm above the point dipole-quadrupole and 60 nm off axis. The three graphs present 3 different wavelengths 550, 600 and 700 nm. The three white circles display the angles for 30, 60 and 90 degrees. b) Cross cuts of the $|E|^2$ emission patterns of figure a). c) Polar plots of the radiated intensity by the cylindrical antennas when excited with a electrical dipolar emitter located at different positions from the center of the antenna.
expected that if one actually manages to excite magnetic dipole moments and electric quadrupoles as strongly as the fundamental electric dipole term, one can engineer complicated and directional radiation patterns [4, 20–22]. Here we predict that if the Au antenna on silicon that we analyzed is excited with a localized source, as in cathodoluminescence, strongly directional radiation patterns indeed emerge. We predict these radiation patterns simply from two ingredients: the free-space superpolarizability, and the known interface Green’s function [8], \( \text{i.e.} \), without any recourse to a full wave solution beyond extraction of the superpolarizability of the disk in free space. To obtain a prediction, we simply take as driving field the field (i.e., \( \mathbf{E}, \mathbf{H} \) and \( \mathbf{\diamond E} \)) of an electric dipole emitter as given by the known interface Green function. Next, we calculate the induced moments by multiplication of the driving field with the substrate-corrected superpolarizability tensor. Finally we find the far field radiation pattern by coherent addition of the known far fields of the induced moments, for which asymptotic expansions are likewise textbook material [8].

In Fig. 8.3a we show the total field intensity squared (\( |E|^2 \)) for the cylindrical Au antenna excited with a dipolar emitter with strength \( p_0 \), located 60 nm off axis, from the center of the cylinder, and 40 nm above the center of the multipolar scatterer antenna. Fig. 8.3a shows results for three driving wavelengths, namely 550 nm, 600 nm and 700 nm, which are the resonant wavelengths of the quadrupoles, in-plane electric dipole and out of plane electric dipole respectively. These figures together with the cross-cuts shown in Fig. 8.3b show how we can achieve a very strong directional emission with only one single particle. The strong directionality results from coherent superposition of the electric dipole, magnetic dipole, and electric quadrupole terms that are excited in such a ratio as to yield comparable far field flux. For instance, in the first panel of Fig. 8.3a the antenna acquires an electric dipole of \( |p_x| = 1.69p_0 \), \( |p_z| = 1.56p_0 \) and a magnetic dipole of \( |m_y| = 1.17p_0 \) while the dominating acquired quadrupoles have a magnitude of \( |Q_{xx}| = 3.86p_0 \), \( |Q_{yy}| = 3.61p_0 \), and \( |Q_{xz}| = 2.09p_0 \) demonstrating how a localized dipolar source can excite quadrupolar moments in single elements antennas whose radiating power exceed the radiated power from the dipolar moments.

Finally, we sweep the position of the dipole over the antenna, starting from the center moving in steps of only 5 nm. We find (Fig. 8.3c) that the angular emission changes drastically with the position of the emitter with respect to the antenna. For instance, viewing emission under a 70° angle relative to the substrate, it is possible to acquire a signal that changes up to 15% between two given points per nm of lateral position shift of the excitation source. This effect could allow using simple gold antennas on a substrate as sensitive position detectors of fluorescent molecules with resolutions better than those of STORM [23] and PALM [24] microscopy. This ultra-high sensitivity to position of a source relative to an antenna is similar in concept to a recent proposal to use a notched high index dielectric sphere with an overlapping electric and magnetic resonance [21]. However, from the practical side, fabricating an Au cylinder on a Si substrate is far simpler than preparing notched high index Mie spheres.
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Measurements

To complement the theoretical discussion we present experimental results of directionality measurements performed by Toon Coenen of cylindrical gold antennas excited with a localized source. To measure these antennas we use a tightly focused 30 keV electron beam as local excitation source to drive the cylindrical Au nanoantennas. The electromagnetic field created by electron beam provides a broadband and very localized source (~10 nm). This source allows one to do in a single experiment very fine spatially resolved measurements over a broad spectral range and to determine the scattering properties of very small nano antennas.

Figure 8.4: Experimental normalized angular CL emission patterns collected from a pillbox shaped particle with a diameter of 180 nm and a height of 80 nm. The patterns are shown at $\lambda = 600$ nm for excitations at the center of the particle, shown in fig (a), and for excitations near the edge of the particle for four angles: $0^\circ$ (b), $90^\circ$ (c), $180^\circ$ (d) and $270^\circ$ (e). The excitation positions are indicated by the cartoons next to the radiation patterns. The patterns have been normalized to the maximum intensity value for all five patterns to show the relative brightness for different excitation positions.
Cylindrical gold nanoparticles like the ones discussed in Sec. 8.3 were fabricated with diameters ranging from 50-180 nm in 5 nm steps and a height of 80 nm on a crystalline silicon substrate using electron beam lithography, thermal evaporation and a lift-off procedure. For these measurements light is collected by a half parabolic mirror which is placed between the sample and the electron microscope pole piece. This mirror directs the light either to a grating spectrometer, or to a 2D CCD camera. Therefore at each excitation position one either can collect a spectrum in the visible/nearinfrared spectral region, or an image with the information of the scattering directivity. For all the measurements a beam current of 0.8 nA was used.

To measure the far-field angular emission patterns the beam coming from the parabolic mirror was projected onto the CCD camera, to obtain the angular emission distribution. This technique has already been used and is explained in detail in Ref. [25]. Angular patterns are collected for free-space wavelengths ($\lambda$) from 400 to 750 nm using 40 nm band pass filters for all disk diameters. We will further discuss only the 180 nm disk. To optimize the signal-to-noise ratio, 120 s integration times were used for $\lambda = 400, 450, 700$ and 750 nm and 60 s for $\lambda = 500-650$ nm.

Fig. 8.4 shows the angular pattern for a $\lambda = 180$ nm particle at $\lambda = 600$ nm for excitation on four orthogonal edge positions of the nanodisk as depicted in the insets. The pattern clearly shows that the radiation patterns present strong angular asymmetries and that the particle radiates away from the excitation point when excited at the edges. If the nanodisk is excited in the center the emission intensity is lower and as expected based on symmetrical consideration the the emission is azimuthally symmetric.

We continue by studying the angular emission distribution for different wavelengths. Fig. 8.5 shows normalized angular patterns for $\lambda = 400-600$ nm for the particle disk for edge excitation on the left side. The pattern for $\lambda = 400$ nm shows a strong radiation lobe directed to the zenith. The relative contribution of this lobe strongly decreases for longer wavelength while the sideward lobe becomes dominant. At $\lambda = 600$ nm the forward scattering ratio is maximum and most of the radiation is emitted into one strong crescent-shaped lobe pointing away from the excitation position as the one shown in Fig. 8.4.

In order to unravel the multipolar composition of complex scattering phenomena several groups have opted to fit emission patterns to superpositions of multipole radiation patterns [4, 22, 26]. By using the knowledge obtained in Sec. 8.3 we can discriminate based on symmetry considerations of the excitation field and the superpolarizability tensor which are the main multipolar contributions to the scattering process. More specifically, and although the field obtained with the e-beam is not trivial to calculate, based on symmetry consideration we know that this excitation possess an electric field in the z and x directions $\mathbf{E} = \mathbf{E}_x + \mathbf{E}_z$, a magnetic field in the y direction $\mathbf{H} = \mathbf{H}_y$ and a gradient of the electric field along the ‘xz’ plane as well as one on the ‘xy’ plane which is $45^\circ$ rotated $\diamond \mathbf{E} = \diamond \mathbf{E}_{xz} + \diamond \mathbf{E}_{xx} + \diamond \mathbf{E}_{yy}$. The presence of all these fields can also be inferred by finding what are the fields created by a ‘z’ oriented electrical dipole, which share the same symmetries as the electron beam excitation. These fields when multiplied by the superpolarizability tensor indicate which are the multipolar moments involved in the scattering process i.e. $p_z$, $p_x$, $m_y$, $Q_{xz}$ and a symmetrical
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A combination of $Q_{xx} + Q_{yy}$ (which gives us a quadrupole in plane as $Q_{xy}$ but rotated 45°). We single out these components to then fit the data. The results of the fitting procedure are presented in Fig. 8.5b. It is evident that the correspondence is very good. Therefore we can be confident that the superpolarizability tensor pinpoints in an elegant and straightforward way which are the only elements needed to understand the scattering process.

We believe that the reason why these radiation patterns differ from the ones presented in Fig. 8.3 is that we do not have an accurate description of the magnitude and relative contributions of the electric and magnetic fields for the electron beam induced field, which is different from an excitation generated by an electric dipole. For completeness in Fig. 8.5c we present the radiation patterns of the multipole moments involved in the scattering process. The measurements together with the

![Figure 8.5](image_url)

**Figure 8.5:** Figure a) shows the measured experimental CL radiation patterns, while figure b) shows the numerically fitted theoretical patterns for a D=180nm Au disk excited on the ‘left’ side for $\lambda$=400-600 nm. These patterns are a result of the coherent interference of five multipole components. Figure c) shows the theoretical radiation patterns for the individual multipolar components that compose the resonances in the cylinder. These radiation patterns are calculated for $\lambda$=500nm when the multipoles are placed 40 nm above a silicon substrate. As a last panel in d) we show the corresponding charge distribution of the multipoles used. Two perspectives are used for showing the charge distributions. First, we show a top view of the disk on the left and second we show a cross section of the disk in the ‘xz’ plane, presented on the right.

theory demonstrate that the knowledge of the superpolarizability tensor for a given
structure offers a deeper insight to the complex scattering processes happening in antennas. In the case of the cylindrical antenna we show how a careful balance between the multipoles involved in scattering can give rise to an interesting “lighthouse” effect. This effect, where we can define the direction of emission by nanometric tuning of the excitation can be useful for novel types of sensors and to control directionality of LEDs and lasers.

8.4 Conclusions

To summarize, we have shown how to predict the scattering properties of a building block in a complex environment on basis of on one hand the free space building block superpolarizability and on the other hand the environment Green function. As an example we use an Au cylinder big enough to support dipolar and quadrupolar resonances placed on top of a silicon substrate. We have shown that this particle is capable of sustaining very directional scattering when driven by a localized source. This directionality is shown to be fostered by the strong interaction of the dipolar and quadrupolar resonances. The type of study presented in this chapter and performed on the Au cylinder will have a large impact on the quantitative modeling of plasmon and dielectric antennas in typical applications for solar cells, LEDs and sensors, where antennas always function in a complicated dielectric environment. Importantly, we emphasize that the complex environment can induce or enhance particular multipolar moments, yielding new methods to control directionality of scattering and emission.
References