Generalized polarizabilites for an exact multipole analysis of complex nanostructures under inhomogeneous illumination

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The multipole expansion of a nano-photonic structure’s electromagnetic response is a versatile tool to interpret optical effects in nano-optics, but it requires knowledge of the internal field distribution, for which in general expensive full-field simulations are necessary. We present a generalized polarizability model based on the Green’s Dyadic Method (GDM), capable to describe resonant nanostructures, supporting both electric and magnetic dipole and quadrupole modes. Our formalism combines the recently developed exact multipole decomposition [Alaee et al., Opt. Comms. 407, 17-21 (2018)] with the concept of a generalized field propagator. It reproduces the exact multipole moments for any particle size and for arbitrary, inhomogeneous illumination fields such as focused vector beams or local light sources. After an initial computation step, our approach allows to instantaneously obtain the exact multipole decomposition for any possible illumination, and it allows to calculate spectra of the total density of multipole modes. Furthermore, the technique can be used to visualize spatial zones inside a nanostructure, where an external illumination couples strongly to a specific multipole moment. The formalism will be very useful for various applications in nano-optics like illumination-field engineering, or meta-atom design e.g. for Huygens metasurfaces. We provide a numerical open source implementation compatible with the pyGDM python package.

Keywords: polarizability, electric and magnetic resonances, dipole and quadrupole modes, Green’s Tensor, nano-optics, dielectric Huygens metasurfaces

I. INTRODUCTION

Studying the interaction of light with structures of sizes smaller or similar to the wavelength has tremendous importance for various scientific areas and related applications. Already the broad area of nano-optics covers research on a vast range of phenomena such as resonant or directional scattering,1–3 polarization conversion,4 nonlinear scattering of e.g. second or third harmonic light,5–7 optical forces,8,9 or nano-scale heat generation.10,11 These effects are used for instance to study atmospheric or astrophysical particles,12–14 and they have many practical applications, for instance in medicine for hyperthermia treatments or rapid antigen tests.15,16 Understanding and modeling of the interaction of nanostructures with light is also essential for optical metasurfaces.17

An important tool in the description and interpretation of nano-scale light-matter interaction is the modal analysis of the optical response. A powerful method is the quasinormal mode (QNM) expansion, aiming at the identification of all available resonant modes of an open system such as a photonic nanostructure.18 However, QNM expansions are often not straightforward, in particular the normalization of QNMs is a difficult task, due to the description of these modes using complex eigenfrequencies.19,20

A somewhat simpler, yet very useful modal analysis of an already excited nano-photonic system is a multipole expansion of its induced polarization density.21 The conventional expansion can be found in any electro-dynamics textbook,21 and is based on a long-wavelength approximation for the fields emitted by the multipole moments. Recently, exact expressions for the multipole expansion beyond the long-wavelength limit have been developed, that yield accurate results also in the case of larger nanostructures.22–24 While in plasmonic nanostructures usually the electric dipole dominates,25 resonant dielectric nanostructures often possess higher order modes. The latter are a result of retardation. Dielectric structures confine light less efficiently than plasmonic particles, therefore their higher order modes occur typically at sizes where the long-wavelength multipole expansion is no longer accurate.1 In such cases the exact multipole expansion is of particular relevance. From a technical point of view, the application of the multipole expansion to photonic nanostructures is straightforward, and can be done in combination with any numerical solver.26–28

The obvious drawback, compared to QNMs, is that the multipole expansion is an analysis of the internal electric polarization density, and not of the nanostructure itself. This means that an illumination needs to be chosen a priori, and a new simulation needs to be performed for every change in the illumination. Hence no general model, covering all modes of the nanoresonator is obtained. Also, because the basis of the expansion is predefined, for increasing structure size the number of multipole terms rapidly increments, hence the method is most suitable for structures of size comparable to the wavelength. In earlier work, we presented an effective polarizability model for resonant nanostructures, in which retardation has significant impact.20 However, this formalism is limited to the dipolar order and fails if the illumination is truly inhomogeneous, for instance using tightly focused vectorbeams or local sources. In the present work we there-

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fore build on the exact multipole expansion of the polarization density,\textsuperscript{22} and combine it with the concept of a generalized field propagator.\textsuperscript{30} We show that this allows to obtain a set of generalized polarizability tensors for each multipole order, establishing a direct link between an arbitrary illumination field and the induced modes in the exact multipole expansion. Once calculated, the generalized polarizabilities are a computationally very cheap approach to obtain the optical response under arbitrary illuminations and they can be stored efficiently thanks to their light memory footprint. We furthermore show that the generalized polarizabilities can be used to study local properties of light-matter interaction inside the nanostructures, and that they allow to calculate the total density of modes for the different multipoles, regardless of a specific illumination. We foresee that the technique has important applications in the analysis of single-particle light-matter interaction, as well as in the design of metasurfaces, in particular for Huygens metasurfaces.

\section{II. FORMALISM}

When studying the optical interaction of a nanostructure with an external illumination, it is usually insightful to get an approximate, but physically meaningful model for the nanostructure’s optical response. To this end, a multipole expansion of the electric polarization density inside a nanostructure can be performed.\textsuperscript{21,22,26} This gives access to the effective electric and magnetic dipole modes, quadrupole modes, etc., that are induced by the interaction of the nanostructure with an external illumination.\textsuperscript{1,25,31,32} In fact, considering just the dipolar modes of a nano-structure can already give a quite accurate picture of the physics at play, especially in the far-field to which higher order modes like quadrupoles usually couple more inefficiently. However higher order contributions readily lead to localized phenomena in the near-field and, in case of high quality factors, can still couple to the far-field in a significant manner.\textsuperscript{29,33,34}

In the case of atoms, molecules or very small nanostructures, the illumination field can be considered constant at the scale of the nanoparticle. This also allows to describe the structure with polarizability tensors, relating the illumination field $E_0(r_0, \omega)$ at the particle’s position ($r_0$) to an induced multipole moment.\textsuperscript{25,28,35} However, if a nanostructure is larger (illustrated in figure 1a), the quasistatic approximation does not hold any longer. An inhomogeneous illumination field can even lead to entirely new effects. An example are magnetic resonances in dielectric nanostructures. The latter are a result of optical vortices, that are created by a varying phase of the illumination along the nanostructure’s extension.\textsuperscript{1,29,36,37} For larger nanostructures we therefore first need to solve rigorously the light-matter interaction, before we expand the induced electric polarization density inside the particle into multipole moments.

The electric polarization density $P(r, \omega)$ inside nanoparticles of arbitrary shape can be obtained only numerically. Here we will use the Green’s Dyadic Method (GDM), a frequency domain, volume integral approach.\textsuperscript{38} In the GDM, we start with the Lippmann-Schwinger equation (cgs units)

$$E'(r', \omega) = E_0(r', \omega) + \int \frac{d\omega'}{V_{ns}} G(r', r'', \omega) \cdot \overline{\chi}(r'', \omega) \cdot E(r'', \omega)$$

that relates the induced electric field $E'(r', \omega)$ and the unperturbed illumination field $E_0(r', \omega)$ in a self-consistent way. $\overline{\chi}$ is the electric susceptibility tensor of the nanostructure, corresponding to the difference between the relative permittivities of nanostructure and environment $\overline{\chi} = (\epsilon - \epsilon_{env})/4\pi$. The integral runs over the entire volume $V_{ns}$ occupied by the nanostructure. From this it is possible to derive a so-called generalized field propagator $\overline{\chi}(r', r'', \omega)$, that directly relates the incident electric field $E_0(r'', \omega)$ to the induced local electric polarization $P(r', \omega)$.\textsuperscript{30}

$$P(r', \omega) = \overline{\chi}(r', \omega) \cdot \int \frac{d\omega'}{V_{ns}} \overline{\chi}(r', r'', \omega) \cdot E_0(r'', \omega)$$

Note that both locations $r'$ and $r''$ are inside the nanostructure and that while in our notation we use the electric polarization density, it is equivalent to using the current density, since for time-harmonic fields $P(\omega) = -j(\omega)/\omega$.\textsuperscript{39}

The generalized propagator and thus the electric polarization $P$ at any location $r'$ inside a nanostructure can now be obtained numerically by discretizing the nanostructure on a regular grid into a number of $N_c$ mesh-cells, each of volume $V_c$. Such discretization allows to numerically solve the optical Lippmann-Schwinger equation with conventional inversion techniques, giving access to the generalized propagator. For details, we refer to related literature.\textsuperscript{38,40–42} Note that the discretization also transforms the integral in equation (2) into

![Figure 1. The long wavelength approximation describes structures which are very small compared to the wavelength. If the structures become larger the phase and field distribution over its volume cannot be assumed constant any longer. Retardation effects impact the multipole expansion at two levels: (a) The true spatial variations of the illumination field and its phase across the nanostructure and (b) the spatial distribution of the field emitted by the multipoles in the expansion.](image)
a finite sum over the nanostructure mesh-cells:

\[
P(\mathbf{r}_i, \omega) = \mathbf{\overline{K}}(\mathbf{r}_i, \omega) \cdot \sum_{j=1}^{N_c} V_c \mathbf{K}(\mathbf{r}_j, \mathbf{r}_i, \omega) \cdot \mathbf{E}_0(\mathbf{r}_j, \omega) \\
= V_c \mathbf{\overline{K}}(\mathbf{r}_i) \cdot \sum_{j=1}^{N_c} \mathbf{K}_{ij} \cdot \mathbf{E}_0j
\]

(3)

In the second line of equation (3) we introduced an abbreviated notation, where indices \(i\) and \(j\) indicate evaluation at, respectively, the \(i\)th and \(j\)th mesh-cell in the discretization. For the sake of readability we also omit the dependence on the frequency \(\omega\). We will use this notation in the following for all discretized equations.

Now we have access to the distribution of the electric polarization density inside an arbitrary nanostructure, which we can subsequently expand into a series of multipole contributions. As mentioned above the conventional multipole expansion is based on a long-wavelength approximation, and valid only if the field corresponding to the multipole moments (dipole, quadrupole, etc...) can be described in this approximation over the entire nanostructure volume (see figure 1b). When the size of the nanostructure increases and begins to be comparable to the optical wavelength in its material, the conventional equations for the multipole expansion are increasingly inaccurate. For size parameters \(a/\lambda \geq 0.5\) (with \(a\) being the diameter or total length of the structure), this long-wavelength multipole expansion becomes essentially invalid.

In Refs. 22 and 23 Alae et al. have shown recently, that exact equations for the multipole moments can be derived, valid for any particle size, and not significantly more complicated than their long-wavelength counterparts. The exact electric and magnetic dipole moments \(p\) and \(m\), induced in a nanostructure are found to be:

\[
\begin{align*}
\mathbf{p}(\mathbf{r}_0, \omega) &= \mathbf{p}_0(\mathbf{r}_0, \omega) + \mathbf{p}_t(\mathbf{r}_0, \omega) \\
\mathbf{p}_0(\mathbf{r}_0, \omega) &= \int_{V_m} d\mathbf{r}^\prime \mathbf{p}^\prime(\mathbf{r}^\prime, \omega) j_0(\mathbf{k} \cdot \mathbf{r}^\prime) \\
\mathbf{p}_t(\mathbf{r}_0, \omega) &= \frac{k^2}{2} \int_{V_m} d\mathbf{r}^\prime \left( 3(\mathbf{r}^\prime \times \mathbf{p}^\prime(\mathbf{r}^\prime, \omega)) \mathbf{r}^\prime \right) \\
& \quad - \mathbf{r}^2 \cdot \mathbf{p}^\prime(\mathbf{r}^\prime, \omega) \right) \frac{j_2(\mathbf{k} \cdot \mathbf{r}^\prime)}{(\mathbf{k} \cdot \mathbf{r}^\prime)^2}
\end{align*}
\]

\[
\begin{align*}
\mathbf{m}(\mathbf{r}_0, \omega) &= -\frac{3ik}{2} \int_{V_m} d\mathbf{r}^\prime \left( \mathbf{r}^\prime \times \mathbf{p}^\prime(\mathbf{r}^\prime, \omega) \right) \frac{j_1(\mathbf{k} \cdot \mathbf{r}^\prime)}{\mathbf{k} \cdot \mathbf{r}^\prime}
\end{align*}
\]

(4)

(5)

\(k = 2\pi n_{env}/\lambda_0\) is the wavenumber in the surrounding medium (of refractive index \(n_{env}\)) and \(J_n\) is the \(n\)th order spherical Bessel function of the first kind. The electric dipole moment consists of two contributions. Its first order term \(\mathbf{p}_0\) and higher order contributions described by a second term \(\mathbf{p}_t\), often called the “toroidal” dipole moment. \(\mathbf{r}_0\) is the expansion location of the multipole series. For convenience \(\mathbf{r}_0\) is at the origin of our coordinate system. We use the nanostructure’s center of gravity as location for the series expansion.

By applying the above described volume discretization to equation (4) and combining it with equation (3), we obtain for the electric dipole moments:

\[
\begin{align*}
\mathbf{p}_0(\mathbf{r}_0, \omega) &= \sum_i V_c \mathbf{K}(\mathbf{r}_0, \mathbf{r}_i, \omega) j_0(\mathbf{k} \cdot \mathbf{r}_i) \\
\mathbf{p}_t(\mathbf{r}_0, \omega) &= \frac{k^2}{2} \sum_i \left[ \frac{3}{2} \left( \mathbf{r}_i \cdot \mathbf{K}(\mathbf{r}_0, \mathbf{r}_i, \omega) \right) j_2(\mathbf{k} \cdot \mathbf{r}_i) \right] \left( \frac{\mathbf{k} \cdot \mathbf{r}_i}{\mathbf{k} \cdot \mathbf{r}_i} \right)^2 \\
\mathbf{m}(\mathbf{r}_0, \omega) &= -\frac{3ik}{2} \sum_i \left( \mathbf{r}_i \times \mathbf{K}(\mathbf{r}_0, \mathbf{r}_i, \omega) \right) j_1(\mathbf{k} \cdot \mathbf{r}_i) / \mathbf{k} \cdot \mathbf{r}_i
\end{align*}
\]

This is the dot product between two vectors, or the scalar product between two vectors. The core idea of this work is to interchange the summation order of indices \(i\) and \(j\) and to move the illumination field at each meshcell \(\mathbf{E}_{0j}\) out of the sum over index \(i\). This will allow us to evaluate the sum over \(i\) without prior knowledge of the illumination. In Eqs. (6) this is straightforward for all terms except for the first term of the toroidal dipole \(\mathbf{p}_t\), which is a scalar product after multiplication of \(\mathbf{K}_{ij}\) with the illumination field \(\mathbf{E}_{0j}\). In this case we need to introduce a further sum over the three Cartesian vector components \((x, y, z)\) of the toroidal dipole moment, in order to perform the scalar product after the evaluation of the light matter interaction. We get for the
Figure 3. Demonstration of the accuracy of the generalized polarizabilities, compared to full field simulations. In order to obtain an inhomogeneous illumination, a chiral dipole emitter is located at a height of $h = 100\text{ nm}$ above the edge of a dielectric nano-cuboid of dimensions $240 \times 240 \times 330\text{ nm}^3$ made of a lossless material ($n = 3.5$), placed in air ($n_{\text{env}} = 1$). The top figures show the real part of the internal field distribution at selected illumination wavelengths. The bottom plots show the real and imaginary parts of the effective multipole moments, obtained from the induced internal full fields following Ref. 22 (solid lines) as well as calculated using our generalized polarizability tensors (marker symbols). We plot the first order electric dipole (top left), toroidal dipole (bottom left), magnetic dipole (top center), magnetic quadrupole (bottom center), electric quadrupole (top right) and toroidal quadrupole (bottom right) modes. Plots show arbitrary units, normalized to the illumination dipole amplitude. Besides numerical noise the values obtained from both methods match exactly. The multipole amplitudes are in arbitrary units, relative to the illumination field amplitude.

a-component of the dipole moment vectors

$$p_0^a(r_0, \omega) = V_c^2 \sum_{J}^{N_c} \left[ \sum_{l=1}^{N_c} \left( \chi_{l,j}^a K_{ij}^T \right) j_0(kr_l) \right] E_{0j}^T$$  \hspace{1cm} (7a)

$$p_t^a(r_0, \omega) = \frac{k^2 V_c^2}{2} \sum_{J}^{N_c} \sum_{l=1}^{N_c} \left[ \sum_{l=1}^{N_c} \left( 3 \left( r_l \chi_{l,j}^a K_{ij}^T \right) r_l^2 \right) - \frac{r_l^2}{3} \left( \chi_{l,j}^a K_{ij}^T \right) \right] j_2(kr_l) \frac{1}{(kr_l)^3} E_{0j}^T$$  \hspace{1cm} (7b)

Following Einstein’s convention, tensors are contracted over Greek lower case letter indices that occur twice within a product. The factor $1/3$ in the second term of Eq. (7b) comes from the conversion of the scalar product $r_j \cdot P_i$ into a sum over the three Cartesian coordinates $l$, that has been explicitly moved out of the sum over $i$ and affects now the entire term in square brackets. The terms that act on the incident electric field are thus $N_c$ rank 2 tensors for $p_0$, and $N_c$ rank 3 tensors for $p_t$:

$$\alpha_{j,k}^{p_0,a}(r_0, \omega) = V_c^2 \sum_{J}^{N_c} \left( \chi_{l,j}^a K_{ij}^T \right) j_0(kr_l)$$  \hspace{1cm} (8a)

$$\alpha_{j,k}^{p_t,a}(r_0, \omega) = \frac{k^2 V_c^2}{2} \sum_{J}^{N_c} \left( 3 \left( r_l \chi_{l,j}^a K_{ij}^T \right) r_l^2 \right) - \frac{r_l^2}{3} \left( \chi_{l,j}^a K_{ij}^T \right) \right] j_2(kr_l) \frac{1}{(kr_l)^3}$$  \hspace{1cm} (8b)

Each tensor $\overline{\alpha}_j^{p_0}$ and $\overline{\alpha}_j^{p_t}$ describes the contribution of the $j$th mesh-cell to, respectively, the electric dipole moment and the toroidal dipole moment in the multipole expansion:

$$p(r_0, \omega) = \sum_{J}^{N_c} \overline{\alpha}_j^{p_0} \cdot E_{0j} + \sum_{J}^{N_c} \overline{\alpha}_j^{p_t} \cdot E_{0j}$$  \hspace{1cm} (9)

We call these the generalized electric-electric dipole polarizability tensors. Generalized, since they allow to obtain the
effective dipole moment induced in a nanostructure by an arbitrary illumination field. In other words, \( \overline{\alpha}_i^m \) describes the strength of light-matter interaction at the location \( \mathbf{r}_j \) in the nanostructure, for inducing an effective electric dipole moment. Likewise, \( \overline{\alpha}_i^m \) describes the local coupling strength of an illumination field at \( \mathbf{r}_j \), to the toroidal dipole moment.

Proceeding in the same way with equation (5), we obtain the generalized electric-magnetic polarizabilities:

\[
m^m(\mathbf{r}_0, \omega) = -\frac{3i k V^2}{2} \sum_{j} N_i \left[ \sum_{i} \left( \epsilon_{ij}^a \epsilon_{ij}^r \epsilon_{ij}^e \epsilon_{ij}^f \right) j_i(kr_i) \right] E^E_{0j} \tag{10}
\]

\[
\alpha^{m,a}_{ij,\ell}(\mathbf{r}_0, \omega) = -\frac{3i k V^2}{2} \sum_{j} N_i \left( \epsilon_{ij}^a \epsilon_{ij}^r \epsilon_{ij}^e \epsilon_{ij}^f \right) j_i(kr_i) \tag{11}
\]

where \( \epsilon_{ij}^a \) is the Levi-Civita symbol, describing the vector product \( \mathbf{r}_i \times \mathbf{r}_j \). The tensors \( \overline{\alpha}_i^m \) link the illumination electric field distribution \( \mathbf{E}_0 \) inside the structure to the induced, total magnetic dipole moment \( \mathbf{m} \):

\[
\mathbf{m}(\mathbf{r}_0, \omega) = \sum_{j} \overline{\alpha}_i^m \cdot \mathbf{E}_0 \tag{12}
\]

This can be done in the same way also for the higher order multipoles. The expressions for the generalized polarizabilities of electric quadrupole and magnetic quadrupole are given in the Appendix VI A.

In summary, with the generalized polarizability tensors we now have a tool that allows to directly obtain the multipole expansion for arbitrary distributions of the illumination field \( \mathbf{E}_0 \) on a fixed nanostructure geometry, without requiring any further numerical simulation. For each multipole we only need to perform \( N_i \) matrix-vector multiplications of the generalized polarizability tensors with the illumination field. In case of the generalized propagator on the other hand, we require a total of \( N_i^2 \) matrix vector multiplications. In return we get the full electric polarization density inside the structure, while the generalized polarizabilities only give access to its multipole expansion. This is illustrated in figure 2.

Besides the faster evaluation, the fact that for a structure discretized in \( N_i \) mesh-cells, we require only \( N_i \) generalized polarizability tensors, instead of \( N_i^2 \) generalized field propagator tensors \( \overline{K}_{ij} \), has important implications on the memory requirements. Let’s illustrate the scaling with a structure of 10,000 mesh-cells. Storage of the electric and magnetic dipolar response with single precision floating point numbers requires only 1.72 MBytes (this is including the toroidal dipole). The electric and magnetic quadrupole moments add another 4.12 MBytes. On the other hand, 3.43 GBytes are required to store the generalized propagators for the same structure. Creating an extensive database of the generalized polarizabilities is obviously more realistic than saving the generalized field propagators for a larger set of nanostructures.

**III. BENCHMARK**

In the following we demonstrate that the generalized polarizability tensors reproduce the multipole expansion of the electric polarization density. For this test we use a large dielectric cuboid of lossless material with constant refractive index \( n = 3.5 \) and dimensions \( W \times L \times H = 240 \times 240 \times 330 \text{nm}^3 \), placed in air (\( n_{\text{env}} = 1 \)). The long wavelength approximation is not adequate for a structure of this size (c.f. supporting information (SI) figures S1 and S2). In order to create a complex illumination, we consider a right handed chiral dipolar emitter with dipole moment in the \( XY \) plane \( \mathbf{p} = p_0 (\mathbf{e}_x + i \mathbf{e}_y) \). We place this emitter at a height \( h_d = 100 \text{nm} \) above the corner of the nanostructure. The geometry is depicted in the top left of figure 3. The 3D distribution of the induced electric polarization density (its real part) is shown.
IV. DENSITY OF MULTIPLE MODES AND IMPACT OF ILLUMINATION CONDITIONS

Often, scattering spectra obtained with a fixed incident field (typically a plane wave) are used to characterize the optical properties of nanostructures. We demonstrated above that, especially in dielectric nanostructures, a large variety of multipole modes can exist. Their actual appearance, however, is strongly dependent on the illumination. Thus, in scattering spectra with fixed illumination, various multipole modes of the structure might in fact remain invisible.

The generalized polarizabilities include all multipole modes of a nanoparticle. To obtain the entirety of the theoretically available multipole moment at a given wavelength, we can sum the Frobenius tensor norms of all meshcells’ generalized polarizabilities. The obtained quantity corresponds to the sum of generalized polarizability tensor norms for the total electric dipole (blue line), the magnetic dipole (green line), the total electric quadrupole (orange line) and the magnetic quadrupole (red line). Without a detailed analysis, we can recognize a large number of resonant peaks in the different spectra. Note that we can also access the partial multipole densities, corresponding to specific components of the multipole moments (e.g. $p_\varphi$, $m_x$ or $Q_y$). For the dipole moments for instance, the partial mode density can be obtained by summation of the norms of the corresponding column vectors of the generalized polarizabilities. The distinct spectra for all isolated multipole tensor components are shown in the SI figure S3.

In figure 6 we now study the same dielectric spheroid under various illuminations. Figures 6a and 6b show scattering spectra for $p$-polarized, respectively $s$-polarized plane wave illumination. Different incident angles are indicated by different shades of the plot colors, the lightest shade corresponds to an incidence along $Z$, the darkest shade to an incidence along $X$. Figures 6c and 6d show spectra of the scattered intensity upon illumination by an electric dipole placed on top, respectively...
at the side of the spheroid. Here the different shades of the plots indicate the dipole orientation from along X (light colors) to Z (dark colors). Figure 6e finally shows the multipole expansion for the spheroid illuminated by a magnetic dipole emitter at its side, where the color shades indicate again the emitter orientation from x- (light colors) to z-direction (dark colors).

A comparison of figures 5 and 6 shows, that the fundamental field distribution plays a crucial role in the excitation of the available modes. It demonstrates that a careful choice of the incident field allows to address specific modes of a nanostructure. We see for example that the strong field gradients from a dipoic emitter placed very close to the nano-spheroid, excite more efficiently higher order multipoles, than homogeneous fields like a plane wave.

V. VISUALIZE SPATIAL SENSITIVITY TO MULTIPOLE MODE EXCITATION

The generalized polarizabilities of each mesh-cell in a discretized nanostructure correspond to the local strength with which an illumination electric field \( \mathbf{E}_0 \) induces an according multipole moment in the nanostructure. The spatial distribution of the generalized polarizability tensors can therefore be interpreted as the local coupling efficiency of an incident field to the according multipole moments. If one managed to shape an illumination field to correspond exactly to the generalized polarizability distribution, such field would ideally induce the according multipole moment in the structure. In consequence, the generalized polarizability can be used to visualize the spatial zones of strong interaction between an illumination and
Figure 7. Generalized polarizability tensors as visualization tool for the local coupling strength of the illumination to different multipole moments of a silicon disc ($R = 210$ nm, $H = 240$ nm) in air ($n_{\text{env}} = 1$). (a) sum of the tensor norms of the generalized polarizabilities of electric (blue) and magnetic (green) dipole moments. The integrated norm of the column vectors corresponding to the Cartesian components of the dipole moments are shown as thin dotted ($x$ and $y$ components) and dashed ($z$ component) lines. (b) Scattering spectra and according multipole decomposition for normal illumination with a linearly $x$-polarized plane wave. (c) same as in (b) but with oblique incidence ($45^\circ$, $s$-polarized) 
(d) Red: Optimum illumination field distributions at the wavelength $1020$ nm, with maximum possible $x$ and $y$ electric dipole moment. (e) Plane wave illumination (red) and according internal field distribution (blue). (f-g) like (d-e) but showing the optimum illumination field to induce a magnetic $m_y$ dipole moment at $\lambda_0 = 1200$ nm (f) and the internal field induced by a normal incidence plane wave (g). (h-i) Ideal field to excite a magnetic $m_z$ moment (h) compared to the internal field induced by an oblique $s$-polarized plane wave (i).

Dielectric nanostructures – Huygens sources We illustrate the possibility of such an analysis by the example of a silicon disc with radius $R = 210$ nm and height $H = 240$ nm, corresponding to the type of structure recently proposed by Decker et al. as unit cell for dielectric Huygens’ metasurfaces. A Huygens’ metasurface exploits the so-called Kerker effect resulting in forward-only scattering, to achieve unitary transmission. The permittivity of silicon is taken from literature, the disc is placed in air. 

The disc dimensions are chosen such that under normal incidence plane wave illumination, the electric and magnetic dipole resonances spectrally overlap and have similar magnitude (see figure 7b). In figure 7a we first show the mode densities for the electric and magnetic dipoles. As expected, the disc symmetry leads to degenerate $p_x$ and $p_y$ as well as $m_x$ and $m_y$ modes with resonances at $\lambda_0 = 1020$ nm, respectively $\lambda_0 = 1200$ nm. Furthermore, we see a strong magnetic dipole mode $m_z$ around $\lambda_0 = 1450$ nm. Surprisingly, compared to the mode density spectrum, under plane wave illumination the electric dipole has its maximum shifted by around 200 nm and coincides with the magnetic dipole. Furthermore, we see that the $m_z$ mode is not contributing to the scattering spectrum under normal incidence. However, the $m_z$ mode can be addressed using a plane wave at a 45° oblique incident angle and $s$-polarization (electric field parallel to the disc top surface), as shown in figure 7c.

To understand these observations we now have a look at the illumination fields that ideally induce the respective multipoles. These ideal fields are directly obtained from the generalized polarizabilities and we compare them to the internal field induced by a plane wave. In figure 7d (red quiver plot) we show the $x$-column vector of the generalized polarizabilities. This represents the illumination which maximally excites the $p_x$ dipole moment at $\lambda_0 = 1020$ nm. Figure 7e shows (blue quiver plot) that a normally incident plane wave induces an anapole, known to couple very inefficiently to far-field scattering because of destructive interference of internal field regions with opposite phase. This explains why a maximum in the mode density can occur at a minimum in the scattering spectrum (c.f. blue lines in Figs 7a-b). Going back to the optimum field for $p_x$ excitation (Fig. 7d. In contrast to a plane wave
this has a phase distribution that matches the anapole and in fact induces a strong electric dipole moment, which efficiently couples to the far-field (see also SI figure S4). 

By having a look at the column-vectors of the generalized polarizability tensors for the magnetic dipole, we find that the magnetic \( m_z \) moment at \( \lambda = 1200 \text{nm} \) can be ideally excited with an illumination field that has a vortex in the \( XZ \) plane (see bottom panel in figure 7f). An \( x \)-polarized normally incident plane wave has field components with opposite phase at top and bottom of the silicon disc (figure 7g). At the upper and lower facets of the Si disc, this is in accord with the ideal field and thus couples well to the magnetic dipole \( m_z \). Note that also a side-wards (e.g. along \( X \)) incident plane wave with polarization along \( Z \) would couple to the \( m_x \) magnetic dipole component, via the electric field components \( E_{0,z} \) of opposite phase at the left and right sides.

In figure 7h we finally show the optimum illumination field for excitation of an \( m_x \) dipole moment at the wavelength \( \lambda = 1450 \text{nm} \), and find that it corresponds to a field-vortex in the \( XY \) plane. An \( s \)-polarized oblique plane wave (polarization along \( Y \)) has an appropriate phase difference at the left and right side of the silicon disc, and thus induces the same dipole moment (see figure 7i). Note, that the vortex-like ideal field distribution is the reason why a magnetic dipole along \( z \) can be excited efficiently by an azimuthally polarized, focused vectorbeam.\(^{50,51} \) Scattering spectra of the Si disc illuminated by the optimum excitation fields shown in figures 7d, 7f and 7h are shown in the SI figure S4.

**Impact on Huygens metasurfaces** The strong dependence on the illumination of the dipole modes in dielectric nanostructures has important implications for their usage as elementary blocks in Huygens metasurfaces, as Gigli et al. have already recently discussed.\(^52 \) During the design procedure of a metasurface, a lookup table is created, for which the phase-delays of various meta-atoms are simulated. These simulations are usually done with periodic boundary conditions and using a fixed illumination angle. The phase delays of the meta-atoms are subsequently matched with the target metasurface phase map and the structures are placed accordingly. The resulting metasurface obviously does not have the periodicity, that was assumed for the simulations. In consequence the local fields are perturbed by the non-periodic structure arrangement.

The crucial point is now, that a variation of the local illumination can easily lead to the unexpected excitation of a mode that may be “invisible” for a plane wave, as for instance the \( m_y \) and \( m_x \) dipole moments in our above analysis. Furthermore, as we found in the precedent section, the broad electric dipole resonance under normal plane wave illumination (Fig. 7b) is in fact no eigenmode of the system, but rather a dressed mode, dressed by the plane wave illumination. A local source will interact very differently with the structure (c.f. Fig. 6). Also a rotation of the electric or magnetic dipole moment’s orientation can naturally occur if the effective incident angle locally deviates from the plane wave, due to scattering from surrounding structures. Also the relative magnitude between the electric and magnetic dipole moments can be significantly affected, as can be seen for instance around \( \lambda = 1200 \text{nm} \), when comparing figures 7b and 7c. In consequence Kerker’s condition will not be satisfied anymore. Reflection will occur, reducing the efficiency of the Huygens metasurface. In conclusion, a Huygens metasurface based on dielectric nanoresonators requires very delicate optimization of each single constituent, to match the local environment.

**VI. CONCLUSIONS**

In summary, by combining the exact multipole decomposition with the concept of a generalized propagator, we derived expressions for what we call generalized polarizabilities. These are defined for each meshcell of a volume discretized nanostructure and describe the contribution of the respective meshcell to the induced multipole moment. The generalized polarizability tensors allow to calculate, at basically no computational cost, the exact multipole expansion of the optical response of a nanostructure for arbitrary illuminations. We showed that the generalized polarizabilities are not only a useful quantity for fast evaluation of the multipole expansion, but they can be used to calculate spectra of the total density of multipole modes. The formalism can also be used as a tool for direct visualization of the local coupling strength of an illumination field to the different multipole moments. This is interesting for instance for beam-shaping experiments where the nature of the induced optical response in a nanostructure may be controlled through a complex illumination field. We believe that the mode-density analysis via our generalized polarizabilities formalism will be also a valuable tool to anticipate the robustness of a dielectric nanostructure as a meta-atom in a Huygens metasurface. Finally, we anticipate that the very low storage requirements will allow to use the generalized polarizabilities efficiently in lookup tables and also together with deep learning for various applications ranging from the interpretation of the optical properties of individual nanostructures to the design of complex metasurfaces.\(^{53–56} \)

**ACKNOWLEDGMENTS**

We thank Aurélien Cuche and Otto L. Muskens for fruitful discussions. A.P. acknowledges support by Airbus Defence and Space (ADS), through a Ph.D. CIFRE fellowship (No. 2008/0925). A.E.-R. thanks the Institute of Quantum Technology in Occitanie IQO and the Université Paul Sabatier Toulouse for an UPS excellence PhD grant. This work was supported by the Toulouse HPC CALMIP (grant p20010).

**SUPPORTING INFORMATIONS**

- A pdf providing a comparison of the exact multipole generalized polarizabilities and the long wavelength limit multipole expansion as well as further details on the mode-analysis of dielectric nanostructures.
- Example scripts written in python, demonstrating the
use of our method, which we implemented in the publicly available open source package pyGDM.

APPENDIX

A. Quadrupole generalized polarizabilities

The $ab$-component of the exact electric and magnetic quadrupole moments writes:\textsuperscript{22}

$$Q_{e}^{ab}(r_0, \omega) = Q_{0}^{ab}(r_0, \omega) + Q_{el}^{ab}(r_0, \omega) \quad (13a)$$

$$Q_{el}^{ab}(r_0, \omega) = 3 \int_{V_{el}} \left[ 3 \left( r^{\alpha} p_{\alpha} + r^{\beta} p_{\beta} \right) \delta^{ab} \right] \frac{f_1(kr')}{kr'} \ dV' \quad (13b)$$

$$Q_{el}^{ab}(r_0, \omega) = 6k^2V_e^2 \int_{V_{el}} \left[ 5r_{\alpha} r_{\beta} \left( \mathbf{r}' \cdot \mathbf{P}(\mathbf{r}', \omega) \right) \right]$$

$$\left[ \sum_{i} \left( r_i \mathbf{r}' \cdot \mathbf{P}(\mathbf{r}', \omega) \right) \right] \frac{j_3(kr')}{(kr')^3} \quad (13c)$$

$$Q_{m}^{ab}(r_0, \omega) = -15ik \int_{V_{el}} \left[ r^{\alpha} \left( \mathbf{r}' \times \mathbf{P}(\mathbf{r}', \omega) \right) \right]^{b}$$

$$+ r^{\alpha} \left( \mathbf{r}' \times \mathbf{P}(\mathbf{r}', \omega) \right)^{a} \frac{j_2(kr')}{(kr')^2} \quad (14)$$

$\delta^{ab}$ is the Kronecker symbol and $P^a$ is the $a$-component of the vector $\mathbf{P}(\mathbf{r}', \omega)$. $\overline{Q}_0$ and $\overline{Q}_{el}$ are, respectively, the first order term, and the toroidal quadrupole term of the total electric quadrupole moment $\overline{Q}_e$.

After discretization, substitution with Eq. (3), and reordering of the summations, we find for the electric quadrupole terms:\textsuperscript{22}

$$Q_{e0}^{ab}(r_0, \omega) = 3V_e^2 \sum_{i} \sum_{j} \left[ \sum_{l} \left( \frac{1}{3} \left( r_i \chi_{i,l,Y}^\alpha K_{j,l}^{Y} + r_i \chi_{i,l,Y}^\beta K_{j,l}^{Y} \right) \right) \right] E_{ij}^{e} \quad (15a)$$

$$Q_{el}^{ab}(r_0, \omega) = 6k^2V_e^2 \sum_{i} \sum_{j} \left[ \sum_{l} \left( 5r_{i} r_{j} \left( r_i \chi_{i,l,Y}^l K_{j,l}^{Y} \right) \right) \right]$$

$$\frac{j_3(kr_i)}{(kr_i)^3} \quad (15b)$$

$$Q_{m}^{ab}(r_0, \omega) = -15ikV_e^2 \sum_{i} \sum_{j} \left[ \sum_{l} \left( r_i \chi_{i,l,Y}^\alpha K_{j,l}^{Y} \right) \right]$$

$$\frac{j_2(kr_i)}{(kr_i)^2} \quad (15c)$$

The sum over the index $l$ is again introduced to be able to perform the scalar product $\mathbf{r}_i \cdot \mathbf{P}_l$ after summation over the index $i$. The terms in square brackets in Eqs. (15) correspond to the according $N_e$ electric quadrupolar generalized polarizabilities:

$$\alpha_{Q_{0,el}}^{ab}(r_0, \omega) = 3V_e^2 \sum_{i} \left( \frac{1}{3} \left( r_i \chi_{i,l,Y}^\alpha K_{j,l}^{Y} + r_i \chi_{i,l,Y}^\beta K_{j,l}^{Y} \right) \right)$$

$$\frac{j_3(kr_i)}{kr_i} \quad (16a)$$

$$\alpha_{Q_{el}}^{ab}(r_0, \omega) = 6k^2V_e^2 \sum_{i} \left( 5r_{i} r_{j} \left( r_i \chi_{i,l,Y}^l K_{j,l}^{Y} \right) \right)$$

$$\frac{j_3(kr_i)}{(kr_i)^3} \quad (16b)$$

which can be used to calculate the electric quadrupole for any illumination $E_0$ as

$$\overline{Q}_e(r_0, \omega) = \sum_{i=1}^{N_e} \overline{Q}_{0,i} \cdot E_{0j} + \sum_{l=1}^{N_e} \overline{Q}_{el,l} \cdot E_{0j} \quad (17)$$

Analogously, the magnetic quadrupole can be written as:

$$\overline{Q}_m(r_0, \omega) = -15ikV_e^2 \sum_{i} \left( r_i \epsilon_{\xi,K}^\alpha r_j \chi_{i,l,Y}^\xi K_{j,l,Y}^{Y} \right)$$

$$\frac{j_2(kr_i)}{(kr_i)^2} \quad (18)$$

leading to the following definition of the magnetic quadrupole generalized polarizabilities:

$$\alpha_{0,m}^{ab}(r_0, \omega) = -15ikV_e^2 \sum_{i} \left( r_i \epsilon_{\xi,K}^\alpha r_j \chi_{i,l,Y}^\xi K_{j,l,Y}^{Y} \right)$$

$$\frac{j_2(kr_i)}{(kr_i)^2} \quad (19)$$

from which we can now calculate the magnetic quadrupole moment for any illumination field:

$$\overline{Q}_m(r_0, \omega) = \sum_{j} \overline{Q}_{m,j} \cdot E_{0j} \quad (20)$$

Note that due to the scalar products occurring in Eqs. (15), we obtain $N_e$ rank 4 tensors as electric quadrupole generalized polarizabilities (additional index $l$). The magnetic quadrupole on the other hand can be expressed by $N_e$ rank 3 tensors.
