Polarizabilities of complex individual dielectric or plasmonic nanostructures

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When the sizes of photonic nanoparticles are much smaller than the excitation wavelength, their optical response can be efficiently described with a series of polarizability tensors. Here, we propose a universal method to extract the different components of the response tensors associated with small plasmonic or dielectric particles. We demonstrate that the optical response can be faithfully approximated, as long as the effective dipole is not induced by retardation effects, hence do not depend on the phase of the illumination. We show that the conventional approximation breaks down for a phase-driven dipolar response, such as optical magnetic resonances in dielectric nanostructures. To describe such retardation-induced dipole resonances in intermediate-size dielectric nanostructures, we introduce “pseudo-polarizabilities” including first-order phase effects, which we demonstrate at the example of magnetic dipole resonances in dielectric spheres and ellipsoids. Our method paves the way for fast simulations of large and inhomogeneous meta-surfaces.

I. INTRODUCTION

In a multitude of topical areas in contemporary physics and chemistry, the concept of the polarizability has proven to be extremely useful. In particular, in the physics of gases and surfaces, the dynamic polarizability tensor of molecules appears explicitly in the description, for example, of the Van der Waals dispersion energy, or in the description of the Raman scattering process.1–6 During the 1970s, A. D. Buckingham wrote a founding article on this subject in which an exhaustive list of linear polarizabilities is proposed.7 Although this work was restricted to atomic and molecular systems, it represents a valuable stand of the various possible contributions as well as their ranking, in terms of electric and magnetic multipolar polarizabilities.

The theoretical study of the linear optical response of small metallic or dielectric particles has also been extensively investigated in the last decades. In particular, in the context of plasmonics the concept of polarizability is often applied to the description of both far-field and near-field plasmon spectra of sub-wavelength sized noble metal particles.8–14 In many situations, single metal particles can be schematized by a sphere of radius a, in which case their optical response can be described by a scalar, frequency-dependent polarizability $\alpha(\omega_0)$. Then, the polarizability tensor is diagonal and all tensor elements are identical. In cgs units, it reads:15

$$\alpha_{ij}(\omega_0) = a^3 \left( \frac{\varepsilon_2(\omega_0) - \varepsilon_1}{2 \varepsilon_2(\omega_0) + \varepsilon_1} \right),$$

where $\varepsilon_1$ (respectively $\varepsilon_2$) is the dielectric constant of the medium (respectively the nano-sphere). From relation (1), we can extract the extinction spectrum via the imaginary part of $\alpha(\omega_0)$. Consequently, the extinction spectra of a sample containing a large number $N$ of such non-interacting nanoparticles $\alpha_i(\omega_0)$ is given by:15,16

$$I_{\text{ext}}(\lambda_0) = \frac{8 \pi^2}{\lambda_0} \sum_i ^N \text{Im} \left( \alpha_i(\omega_0) \right),$$

where $\lambda_0$ represents the incident wavelength and “Im” the imaginary part.

The sphere represents the highest symmetry, belonging to the isotropic symmetry group. As stated above, in this case, all the diagonal elements of the polarizability are identical, and the system displays a scalar response defined by $\alpha_{ij} = \alpha_{\delta_{ij}}$ (see equations (1) and (2)). When transforming the sphere into an ellipsoid of symmetry group $D_{nh}$, the polarizability must be defined with two independent components, and for even lower symmetry, all components $\alpha_{ij}$ of the polarizability tensor must be calculated. This situation corresponds to high anisotropy induced by a complex shape of particles (or nano-cavities). Note that other kinds of anisotropy can come from the intrinsic anisotropy of the dielectric constant of the particle but also from the surface of another object.17 In the latter case, the concept of effective polarizability is generally introduced, and the final symmetry of the particle is dressed by the symmetry of the surface (i.e. $D_{nh}$, for a perfectly planar surface).

As illustrated by these examples, the design of nanostructure polarizabilities starts with the conception of a reference geometry by intuitive considerations. Such an approach, however, is limited to rather simple problems. In case of complex structures or complicated phenomena, the intuitive method often fails, as unexpected effects such as polarization conversion occur in the polarizability tensors. In this work we propose a numerical method to extract the polarizability tensors for complex shaped metallic and dielectric nanostructures through a volume discretization technique, which uses the concept of a generalized propagator. Furthermore, in order to faithfully describe also magnetic optical effects in dielectric nanostructures, where the conventional dipolar polarizability approximation fails, we introduce “pseudo-polarizabilities” that include phase-induced magnetic dipole resonances, similar to some homogenization approaches for metamaterials,18,19 but at the level of a single, isolated structure. Our pseudo-polarizabilities might then be used to construct aperiodic or random meta-surface-like assemblies without periodicity.
In which the first one, $K^{EE}(r, r', \omega_0)$ that describes the electric–electric field couplings was introduced in the early beginning of near-field optics. The three other contributions, i. e. $K^{EH}(r, r', \omega_0)$, $K^{HE}(r, r', \omega_0)$ and $K^{HH}(r, r', \omega_0)$, account for coupling with the magnetic field. All these propagators are related to the corresponding mixed field–susceptibilities $S^{EE}$, $S^{EH}$, $S^{HE}$, and $S^{HH}$ associated with the source zone:

$$
\begin{align*}
K^{EE}(r, r', \omega_0) &= \delta(r-r')I + \chi_e(\omega_0) \cdot S^{EE}(r, r', \omega_0) \\
K^{EH}(r, r', \omega_0) &= \chi_m(\omega_0) \cdot S^{EH}(r, r', \omega_0) \\
K^{HE}(r, r', \omega_0) &= \chi_e(\omega_0) \cdot S^{HE}(r, r', \omega_0) \\
K^{HH}(r, r', \omega_0) &= \delta(r-r')I + \chi_m(\omega_0) \cdot S^{HH}(r, r', \omega_0)
\end{align*}
$$

As explained in references 20 and 21, these dyadic tensors can be numerically computed by performing a volume discretization of the source zone together with a Dyson sequence procedure or other numerical inversion techniques, to extract the various field-susceptibilities in the source zone.

II. A GENERALIZED ELECTROMAGNETIC PROPAGATOR FOR ARBITRARY SHAPED PARTICLES OR CAVITIES

The concept of the generalized electric field propagator previously described in reference 20 can be easily extended to the general case of meta-systems displaying both an electric and a magnetic linear response. In this case, the source zone as depicted in figure 1 is characterized by the following susceptibility tensor, where $I$ is the identity tensor:

$$
\chi(\omega_0) = \begin{pmatrix}
\chi_e(\omega_0)\ I & 0 \\
0 & \chi_m(\omega_0)\ I
\end{pmatrix};
$$

(3)

where $\chi_e(\omega_0)$ and $\chi_m(\omega_0)$ are related to the permittivity $\varepsilon_e(\omega_0)$, respectively the permeability $\mu_m(\omega_0)$ of the source zone:

$$
\chi_e(\omega_0) = \frac{\varepsilon_e(\omega_0) - \varepsilon_1(\omega_0)}{4\pi};
$$

(4)

and

$$
\chi_m(\omega_0) = \frac{\mu_m(\omega_0) - \mu_1(\omega_0)}{4\pi}.
$$

(5)

Introducing two super vectors $F_0(r, \omega_0) = (E_0(r, \omega_0), H_0(r, \omega_0))$ and $F(r, \omega_0) = (E(r, \omega_0), H(r, \omega_0))$ (where $E$ and $H$ refer to electric and magnetic fields, respectively) to describe the incident and total electromagnetic fields, we can define a unique $(6 \times 6)$ dyadic tensor $K(r, r', \omega_0)$ operating in the volume $V$ of the source zone and establishing the link between $F_0(r, \omega_0)$ and $F(r, \omega_0)$:

$$
F(r, \omega_0) = \int_V K(r, r', \omega_0) \cdot F_0(r', \omega_0) dr'.
$$

(6)

Actually, the $(6 \times 6)$ superpropagator $K(r, r', \omega_0)$ is composed of four mixed $(3 \times 3)$ dyadic tensors:

$$
K(r, r', \omega_0) = \begin{pmatrix}
K^{EE}(r, r', \omega_0) & K^{EH}(r, r', \omega_0) \\
K^{HE}(r, r', \omega_0) & K^{HH}(r, r', \omega_0)
\end{pmatrix}
$$

(7)
\[ \mathcal{M}_m(r_i, \omega_0) = \Delta v^2 \chi_m(\omega_0) \times \sum_{j=1}^{N} \left( K^{HE}(r_i, r_j, \omega_0) \cdot E_0(r_j, \omega_0) + K^{HH}(r_i, r_j, \omega_0) \cdot H_0(r_j, \omega_0) \right), \]

where the first contribution in Eq. (10), proportional to \( k_0 = \omega_0 / c \), originates from polarization vortices induced by phase changes inside the source region. These magnetic polarization effects have been extensively studied recently in the case of high index dielectric nanostructures.\(^{3,22-26}\) Note that the choice of the center of the coordinate system is important, as it has an impact on the magnetic polarization \( \mathcal{M}_m \). Usually, it is convenient to use the center of mass \( r_c \) of the nanostructure and we will adopt this choice for the following examples where we set \( r_c \) as the center of the coordinate system.

The total electric polarization \( \mathcal{P}(\omega_0) \) (respectively magnetic polarization \( \mathcal{M}(\omega_0) \)) is obtained by adding the local electric polarizations Eq. (9) (respectively the magnetic polarizations Eq. (10)) of all the elementary cells of the volume discretization. These polarizations are related to the super vector \( F_0(r_c, \omega) \) at the center of mass \( r_c \) of the nanostructure by the \((6 \times 6)\) super polarizability \( \alpha(\omega_0) \):

\[
\begin{bmatrix}
\alpha_{xx}(\omega_0) & \alpha_{xy}(\omega_0) & \alpha_{xz}(\omega_0) \\
\alpha_{yx}(\omega_0) & \alpha_{yy}(\omega_0) & \alpha_{yz}(\omega_0) \\
\alpha_{zx}(\omega_0) & \alpha_{zy}(\omega_0) & \alpha_{zz}(\omega_0)
\end{bmatrix} =
\begin{bmatrix}
\alpha_{EF}(\omega_0) & \alpha_{EH}(\omega_0) \\
\alpha_{HE}(\omega_0) & \alpha_{HH}(\omega_0)
\end{bmatrix}
\begin{bmatrix}
E_0(r_c, \omega_0) \\
H_0(r_c, \omega_0)
\end{bmatrix}
\]

where the polarizabilities \( \alpha_{EF}(\omega_0), \alpha_{EH}(\omega_0), \alpha_{HE}(\omega_0) \) and \( \alpha_{HH}(\omega_0) \) are four \((3 \times 3)\) dyadic tensors, defined by

\[
\alpha_{EF}(\omega_0) = \Delta v^2 \chi_c(\omega_0) \sum_{i,j} K^{EE}(r_i, r_j, \omega_0) e^{i k \cdot r_j}
\]

\[
\alpha_{HE}(\omega_0) = \Delta v^2 \chi_c(\omega_0) \sum_{i,j} K^{EH}(r_i, r_j, \omega_0) e^{i k \cdot r_j}
\]

\[
\alpha_{HE}(\omega_0) = \Delta v^2 \chi_c(\omega_0) \sum_{i,j} K^{HE}(r_i, r_j, \omega_0) e^{i k \cdot r_j}
\]

\[
\alpha_{HH}(\omega_0) = \Delta v^2 \chi_c(\omega_0) \sum_{i,j} K^{HH}(r_i, r_j, \omega_0) e^{i k \cdot r_j}
\]

To be more precise, these are \emph{pseudo-polarizabilities} since they depend on the direction of illumination due to the phase term \( \exp(i k \cdot r_j) \). Conventional polarizabilities depend only on the geometry and the material of the nanostructure.\(^{11,12}\)

This phase term is the direct cause of the emergence of polarization vortices, which are responsible for the existence of magnetic multipole moments in dielectric nanostructures.\(^{22,28}\) In order to be able to describe the magnetic polarization due to the mixed field susceptibility, we keep the phase term in the expression of the \emph{pseudo polarizabilities}. We note that this approximation requires that the wave vector of the incident field is known already during the calculation of \( \alpha(\omega_0) \). However, we will show later, that a further approximation can be used to generalize these pseudo-polarizabilities to any oblique illumination without prior knowledge of the angle of incidence.

For the calculation of the polarizabilities we used our own python implementation "pyGDM" of the volume discretization procedure described above.\(^{29}\)
Figure 3. Spectral variation of the real \((e)-(h)\) and imaginary part \((i)-(l)\) of the terms of the dipolar polarizability matrix for various structures represented in \((a)-(d)\). Geometries consist in \((a)\) a single isotropic pad of size \((50\text{ nm} \times 50\text{ nm} \times 25\text{ nm})\), \((b)\) an anisotropic pad of size \((50\text{ nm} \times 100\text{ nm} \times 25\text{ nm})\), \((c)\) a “F” shape structure included in the \(xOy\) plane, \((d)\) a “3D” shape structure with ramifications in the three directions of space. For the first three structures computations were performed with a discretization step \(d=2.5\text{ nm}\) while we used a step \(d=2\text{ nm}\) in the last case for a good convergence of the calculation. The color diagrams show the degenerate components of the polarizability tensor.

IV. RESULTS

A. Electric-electric polarizability for structures of arbitrary shape

In a first step, we compare the spectral variation of the imaginary part of the dipolar polarizability \(\text{Im}[\alpha^{EE}(\omega)]\) at the example of an isolated spherical gold particle (radius \(r = 5 \text{ nm}\)). Fig. 2a shows a comparison of the first diagonal term \(\alpha_{xx}\), calculated analytically (Eq. (1), blue line) or numerically (using Eq. (14a), red line). For the sphere suspended in vacuum, the diagonal terms of \(\alpha^{EE}\) are identical, and off-diagonal terms vanish. Our numerical discretization approach reproduces the well-known plasmon resonance for gold nanoparticles around \(\lambda_0 = 520 \text{ nm}\). The slight quantitative difference between the two representations is due to the inaccuracy of the analytical formula on non-atomic size scales. If we add a silica substrate in the calculation (see inset in Fig. 2b), the symmetry is reduced from spherical to a cylindrical. In consequence, the polarizability tensor is no longer diagonal and \(\alpha_{xx}^{EE} = \alpha_{yy}^{EE} \neq \alpha_{zz}^{EE}\), which is depicted in Fig. 2b.

The volume discretization allows us to treat nanostructures of arbitrary shape. Therefore, in a next step we study the evolution of the different terms of the electric-electric pseudo-polarizability tensor \(\alpha^{EE}(\omega)\), while gradually increasing the structure complexity, as illustrated in Fig. 3(a-d). Note that the polarizability tensor is symmetric (see Eqs. (14a) and (8)), so in Fig. 3(e-l) we plot only the upper triangular elements. First, we calculate the spectral variation of the polarizability matrix of a gold pad of size \((50\text{ nm} \times 50\text{ nm} \times 25\text{ nm})\), discretized with cubic cells of side length \(d = 2.5 \text{ nm}\) (cf. Fig. 3a). The real and imaginary part of each tensor component are shown in Fig. 3e, respectively 3i. Due to the symmetry of the structure the off diagonal terms of \(\alpha^{EE}\) are zero (cyan lines). Moreover, we observe that \(\alpha_{xx}^{EE} = \alpha_{yy}^{EE}\) (blue lines) which is a result of the rectangular footprint of the structure. Because the height is only half of the structure’s width, \(\alpha_{zz}^{EE}\) is significantly smaller (green line). Despite the small dimensions of the pad, localized plasmon resonances arise slightly red-shifted at around 550–600 nm. Now if we increase the size of the pad along \(\Delta x\) by a factor of two, the \(\alpha_{xx}^{EE}\) and \(\alpha_{yy}^{EE}\) terms are not equal anymore, due to the aspect ratio of the elongated pad. In this case, the resonance for excitation along the long edge is even more red-shifted to around 650 nm, which reflects the effective wavelength scaling of the localized plasmon resonance. Next, we calculate the polarizability tensor for a symmetric L-shaped gold structure (illustrated in Fig. 3c). In this structure, coupling between the horizontal and the vertical arm leads to
a non-zero off-diagonal term $\alpha^{EE}_m$, as can be seen in Fig. 3g and 3k (magenta lines). Due to this off-diagonal term, two additional resonances emerge around 690 nm and 1170 nm at which polarization conversion between the Y-arm and the X-arm of the antenna occurs. The two peaks at 690 nm and 1170 nm correspond to the anti-bonding, respectively bonding modes between the two arms. We note, that the opposite phase of the bonding and the anti-bonding mode is correctly reflected also in the spectrum of the polarizability off-diagonal element. Polarization conversion is only occurring between $X$ and $Y$, hence the other off-diagonal elements remain zero (cyan lines). Moreover, both arms are of the same length which leads to $\alpha^{EE}_x = \alpha^{EE}_y$ (blue lines). Finally, we construct a three-dimensional structure which introduces interactions between each Cartesian direction, as depicted in figure 3d. In this case, the each matrix element shows a unique spectral behavior, representing the complex interaction mechanisms between the antenna arms in different directions (Fig. 3h and 3i).

We note at this point, that the approach is also capable to deal with nano-cavities carved into a bulk medium, by using a non-unitary permittivity for the environment and $\chi_m = \chi_e = 0$ in the hollow source region.

## B. Magnetic-electric polarizability of a dielectric sphere

We now want to assess the role of the magnetic terms in the super polarizability. Since in nature no material with a significant direct magnetic optical response is known, we will assume $\chi_m = 0$, hence the magnetic field of light cannot directly interact with the nanostructure. In consequence the polarizability tensors Eqs. (14) drastically simplify. The mixed terms involving $K^{EH}$ and $K^{HE}$ all disappear, since they include the product $\chi_e \chi_m$ (see also Eq. (8)). In fact only the two terms that depend on $K^{EE}$ remain. Hence, for media with $\chi_m = 0$, the electric polarization is fully described by $\alpha^{EE}$ and the magnetic polarization is entirely governed by $\alpha^{HE}$.

As an example we show in figure 4a the extinction cross section of a dielectric nano-sphere ($n = 4$) of radius $r = 100\text{nm}$ in vacuum, calculated from the discretized electric polarization density (blue line). We show additionally the decomposition of the extinction into an effective electric and magnetic dipole moment at the sphere’s center of mass (orange, respectively green lines). The dielectric sphere has an electric dipole (ED) resonance at 600 nm and a magnetic dipole (MD) resonance at 790 nm, which are indicated by black vertical dashed lines. The real part of the electric field inside the nano-sphere at these resonances is qualitatively shown in 3D vector plots above figure 4a. In figure 4b we show the extinction cross section obtained from the effective polarizabilities $\alpha^{EE}$, respectively $\alpha^{HE}$. We compare the “static” effective polarizabilities without phase term (dashed colored lines) and the above introduced pseudo-polarizabilities including the phase term $\exp(i\mathbf{k} \cdot \mathbf{r}_j)$ (solid colored lines, see Eqs. (14)). The dotted black lines show the ED and MD response from the full internal fields. While the ED resonance in figure 4b is very well reproduced by both, the static and the phase-sensitive electric-electric pseudo-polarizability, the MD resonance cannot be reproduced if the phase term in Eq. (14c) is omitted (dashed green line in figure 4c). Only if the phase term is taken into account, the extinction calculated from the pseudo-polarizability matches the magnetic dipole resonance in the dielectric sphere (solid green line in figure 4c). This is because the magnetic dipole is induced by the vortex formed by the electric displacement current (see illustration of the MD above Fig. 4a, right), which is a direct consequence of the phase difference of the incident
field across the relatively large nano-sphere.

C. Approximation of $\alpha^{HE}$ for arbitrary angles of incidence

In contrast to “classical” static polarizabilities, the here introduced pseudo polarizabilities depend on the illumination wave-vector $k_0$ as a result of the above discussed phase term. In consequence, to solve the general problem, the pseudo-polarizability needs to be separately calculated for every incident field which limits the usefulness of the approximation. However, we can approximate arbitrary incident angles through a first order expansion of the phase term. While we keep the phase-term in the definition of the polarizabilities, we assume that the first order term of its Taylor expansion is sufficient to describe the magnetic dipolar response. Thus, while allowing retardation effects to a certain extent, we still stick with the assumption that the wavelength is large with respect to the nanostructure (i.e. $\lambda_0 \gg |r|$). Since the optical interaction is still modelled as a point-response, the wave vector of the illumination is assumed to be constant across the nanostructure. Furthermore, the approximation requires that the location of the effective dipole is independent of the wave vector. We assume here that the effective electric and magnetic dipole moments $\mathcal{P}(\omega_0)$, respectively $\mathcal{M}(\omega_0)$ lie at the particle’s center of mass $r_c$, for any angle of incidence and polarization of the illumination. Without loss of generality we now consider an incident wave vector in the $XZ$ plane, where we get:

$$\mathcal{P} \approx \left( \frac{k_x}{|k_0|} \right)^2 \alpha^{EE}_{k_x} + \left( \frac{k_z}{|k_0|} \right)^2 \alpha^{EE}_{k_z} \cdot E_0$$

(15a)

and

$$\mathcal{M} \approx \left( \frac{k_x}{|k_0|} \alpha^{HE}_{k_x} + \frac{k_z}{|k_0|} \alpha^{HE}_{k_z} \right) \cdot E_0.$$  

(15b)

For a derivation of these approximations based on a first order expansion of the phase term $\exp(ik \cdot r_c)$ in Eqs. (14), see appendices A and B. The dependence on $\omega_0$ and $r_c$ has been omitted for the sake of readability. $k_i$ is the wave vector component and $\alpha^{XX}_{k_i}$ the pseudo polarizability for the Cartesian direction $i \in \{x, z\}$. Both are evaluated at the position $r_c$ of the effective dipole (here the center of mass). Using this superposition scheme, the response of the nanostructure to any oblique plane wave illumination is described by three “pseudo super-polarizability tensors” $\alpha^{XX}_{ki}$ (one for every Cartesian coordinate axis $i$). Once the approximations Eqs. (15) for the effective dipole moments are calculated, the extinction cross sections due to the induced electric and magnetic polarizations can be calculated as:

$$I_{\text{ext.} \cdot s}(\omega_0) = \frac{8\pi^2}{\lambda_0} \text{Im} \left( E_0(r_c, \omega_0)^{\ast} \cdot \mathcal{P}(\omega_0) \right)$$

(16a)

and

$$I_{\text{ext.} \cdot p}(\omega_0) = \frac{8\pi^2}{\lambda_0} \text{Im} \left( H_0(r_c, \omega_0)^{\ast} \cdot \mathcal{M}(\omega_0) \right),$$

(16b)

where the superscript asterisk (‘∗’) indicates complex conjugation.

In figure 5 we show spectra of the extinction cross section of a dielectric spheroid (refractive index $n = 4$) in vacuum, with a diameter of $D_1 = 250 \text{nm}$ along the $OX$-oriented long axis and two identical short axes with diameters ($D_2 = 120 \text{nm}$), as illustrated at the left of figure 5. The extinction is shown for different incident angles for $s$-polarization (top row) and $p$-polarization (bottom row). Clearly, the pseudo-polarizability superposition approximation (solid lines; $\alpha^{EE}$: orange, $\alpha^{HE}$: green) yields excellent agreement with the ED and MD decomposition of the extinction from full-field simulations (dashed orange and green lines, respectively). Once again, the static polarizability approximation breaks down in case of the magnetic dipole resonance $\alpha^{HE}_{\text{static}}$ (green dotted lines). In case of the electric dipole response, the static polarizability $\alpha^{EE}_{\text{static}}$ gives a reasonable approximation. However, if the incidence direction is along the long axis of the ellipsoid,
phase effects start to play a non-negligible role, and significant deviations occur in the static polarizability approximation.

V. CONCLUSION AND PERSPECTIVES

In conclusion we introduced a mathematical scheme for a generalized description of light-matter interaction in nanostructures through both, optical electric and magnetic fields. We showed how the optical response of nanostructures can be approximated through a universal “super polarizability” tensor, which combines the optical response through electric and magnetic dipole moments. Using a volume discretization, the super polarizability can be numerically calculated for nanostructures of arbitrary shape and material. We demonstrated that our pseudo polarizability, which includes phase effects, is capable to faithfully describe also magnetic dipole resonances in dielectric nanostructures of important size, where a conventional, static point-response model is breaking down. In contrast to similar, computationally more complex multi-dipole methods, our approach of effective electric and magnetic polarizabilities is capable to capture the optical response of complex nanostructures in a single quantity, which strongly facilitates the further evaluation of the optical behavior, for instance under changing illumination conditions. We foresee that our framework can be used to calculate large assemblies of different and/or randomly positioned nanostructures. Our work paves the way to the development of powerful design methods for highly heterogeneous plasmonic and dielectric or hybrid metasurfaces.

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APPENDIX

A. Interpolation of magnetic-electric polarizabilities

For a non-magnetic nanostructure the electric-magnetic polarizability writes (see also main paper):

\[
\alpha^{HE}(\omega_0) = \sum_{i,j} A_{i,j}^{HE} e^{i k r_j} = \sum_{i,j} \alpha_{i,j}^{HE} e^{i k r_j},
\]

where we neglected the dependence on \(\omega_0\) for the sake of readability. Due to the phase term \(\exp(i k r_j)\), the polarizability \(\alpha^{HE}\) is dependent on the incident angle and writes for a wave vector \(k\) of arbitrary angle \(\theta\):

\[
\alpha^{HE}_\theta = \sum_{i,j} A_{i,j}^{HE} e^{i k r_j} = \sum_{i,j} A_{i,j}^{HE} e^{i(k_x x_j + k_y y_j + k_z z_j)}. \tag{18}
\]

Now we consider three different \(\alpha^{HE}_{\theta_i}\) corresponding to plane wave incidence along each of the three Cartesian directions:

\[
\alpha_x^{HE} = \sum_{i,j} A_{i,j}^{HE} e^{i k x_j}, \quad \alpha_y^{HE} = \sum_{i,j} A_{i,j}^{HE} e^{i k y_j}, \quad \alpha_z^{HE} = \sum_{i,j} A_{i,j}^{HE} e^{i k z_j};
\]

where \(k = \frac{2 \pi n}{\lambda_0}\) and \(n\) is the medium index.

We now develop the sum of the polarizabilities for plane wave incidence along the Cartesian coordinate axis. We define also three parameters allowing to describe an arbitrary illumination direction:

\[
\beta_x = \frac{k_x}{k}, \quad \beta_y = \frac{k_y}{k}, \quad \beta_z = \frac{k_z}{k}. \tag{20}
\]

In addition, we assumed that \(k_x^2 + k_y^2 + k_z^2 = k^2\). We can now write

\[
\beta_x \alpha_x^{HE} + \beta_y \alpha_y^{HE} + \beta_z \alpha_z^{HE} = \sum_{i,j} A_{i,j}^{HE} \left[ \beta_x e^{i k x_j} + \beta_y e^{i k y_j} + \beta_z e^{i k z_j} \right]. \tag{21}
\]

Assuming that \(\lambda_0 \gg |r|\), we can approximate the exponentials by their first order Taylor series:

\[
\beta_x \alpha_x^{HE} + \beta_y \alpha_y^{HE} + \beta_z \alpha_z^{HE} \approx \sum_{i,j} A_{i,j}^{HE} \left[ \beta_x (1 + i k x_j) + \beta_y (1 + i k y_j) + \beta_z (1 + i k z_j) \right]. \tag{22}
\]

By adding “1 – 1”, we can write

\[
\beta_x \alpha_x^{HE} + \beta_y \alpha_y^{HE} + \beta_z \alpha_z^{HE} \approx \sum_{i,j} A_{i,j}^{HE} \left[ \beta_x + \beta_y + \beta_z - 1 + i \left( k_x x_j + k_y y_j + k_z z_j \right) \right] \approx e^{i (k x_j + k y_j + k z_j)} \tag{23}
\]

The constant terms in Eq. (23) are proportional to the static magnetic-electric polarizability, which, as we have shown in
the main paper, is negligible compared to usual dipolar polarizabilities, since the vortices that generate the magnetic dipolar response in non-magnetic nanostructures cannot be described without the phase term $\exp(\text{i} k \mathbf{r})$:

$$
\sum_{i,j} \left( \text{const.} \times A_{i,j}^{HE} \right) \approx 0. \tag{24}
$$

Hence we find:

$$
\alpha_0^{HE} \approx \beta_x \alpha_x^{HE} + \beta_y \alpha_y^{HE} + \beta_z \alpha_z^{HE} \tag{25}
$$

B. Interpolation of electric-electric polarizabilities

The electric-electric polarizability writes (see also main paper):

$$
\alpha^{EE}(\omega_0) = \Delta v^2 \chi_e(\omega_0) \sum_{i,j} K_{i,j}^{EE}(\mathbf{r}_i, \mathbf{r}_j, \omega_0) e^{\text{i} k_j r_j} = \sum_{i,j} A_{i,j}^{EE} e^{\text{i} k_j r_j}, \tag{26}
$$

where we neglected the dependence on $\omega_0$ for the sake of readability.

Due to the phase term $\exp(\text{i} k \mathbf{r})$, the polarizability $\alpha^{EE}$ is dependent on the incident angle and writes for a wave vector $\mathbf{k}$ of arbitrary direction $\theta$:

$$
\alpha_0^{EE} = \sum_{i,j} A_{i,j}^{EE} e^{\text{i} k_j r_j} = \sum_{i,j} A_{i,j}^{EE} e^{\text{i}(k_j x_i + k_j y_i + k_j z_j)}. \tag{27}
$$

Now we consider three $\alpha^{EE}_{i,j}$ corresponding to plane wave incidence along each of the three Cartesian directions:

$$
\alpha_x^{EE} = \sum_{i,j} A_{i,j}^{EE} e^{\text{i} k x_j},
\alpha_y^{EE} = \sum_{i,j} A_{i,j}^{EE} e^{\text{i} k y_j},
\alpha_z^{EE} = \sum_{i,j} A_{i,j}^{EE} e^{\text{i} k z_j} \tag{28}
$$

We use the same definition of $\beta_x, \beta_y$ and $\beta_z$ as in Eq. (20). We can now write:

$$
\beta_x^2 \alpha_x^{EE} + \beta_y^2 \alpha_y^{EE} + \beta_z^2 \alpha_z^{EE} = \sum_{i,j} A_{i,j}^{EE} \left[ \beta_x^2 e^{\text{i} k x_j} + \beta_y^2 e^{\text{i} k y_j} + \beta_z^2 e^{\text{i} k z_j} \right]. \tag{29}
$$

Assuming that $\lambda_0 \gg |\mathbf{r}|$, we can approximate the exponentials by their first order Taylor series:

$$
\beta_x^2 \alpha_x^{EE} + \beta_y^2 \alpha_y^{EE} + \beta_z^2 \alpha_z^{EE} \approx \sum_{i,j} A_{i,j}^{EE} \left[ \beta_x^2 (1 + ik x_j) + \beta_y^2 (1 + ik y_j) + \beta_z^2 (1 + ik z_j) \right] = \sum_{i,j} A_{i,j}^{EE} \left[ \beta_x^2 + \beta_y^2 + \beta_z^2 + \text{const.} \right] = \sum_{i,j} A_{i,j}^{EE} \left[ 1 + i(\beta_x^2 k x_j + \beta_y^2 k y_j + \beta_z^2 k z_j) \right]. \tag{30}
$$

Now we subtract the first order Taylor expansion of equation (27)

$$
\alpha_0^{EE} \approx \sum_{i,j} A_{i,j}^{EE} \left[ 1 + i(\beta_x k x_j + \beta_y k y_j + \beta_z k z_j) \right] \tag{31}
$$

from both sides of equation (30), which yields:

$$
\beta_x^2 \alpha_x^{EE} + \beta_y^2 \alpha_y^{EE} + \beta_z^2 \alpha_z^{EE} - \alpha_0^{EE} \approx \sum_{i,j} i A_{i,j}^{EE} \left( \beta_x (\beta_x - 1) k x_j + \beta_y (\beta_y - 1) k y_j + \beta_z (\beta_z - 1) k z_j \right) = i \beta_x (\beta_x - 1) k \sum_{i,j} A_{i,j}^{EE} x_j + i \beta_y (\beta_y - 1) k \sum_{i,j} A_{i,j}^{EE} y_j + i \beta_z (\beta_z - 1) k \sum_{i,j} A_{i,j}^{EE} z_j \tag{32}
$$

To demonstrate that the expression on the right hand side in Eq. (32) is negligible, we use Eq. (24), which states that

$$
\sum_{i,j} \left( A_{i,j}^{HE} \right) = -\Delta v^2 \frac{ik_0}{2} \chi_e(\omega_0) \sum_{i,j} \{ \mathbf{r}_i \wedge \mathbf{K}_{i,j}^{EE}(\mathbf{r}_i, \mathbf{r}_j, \omega_0) \} \approx 0. \tag{33}
$$

Properly speaking, with “approximately zero” we mean that the term is negligible within the small particle approximation. Using now the symmetry of $\mathbf{K}_{i,j}^{EE}(\mathbf{r}_i, \mathbf{r}_j, \omega_0)$ and the antisymmetry of the cross product “$\mathbf{r}_i \wedge$”, we can anti-commute those two terms:

$$
\sum_{i,j} \left( A_{i,j}^{HE} \right) = \Delta v^2 \frac{ik_0}{2} \chi_e(\omega_0) \sum_{i,j} \mathbf{K}_{i,j}^{EE}(\mathbf{r}_i, \mathbf{r}_j, \omega_0) \cdot \{ \mathbf{r}_i \wedge \} = \frac{ik_0}{2} \sum_{i,j} A_{i,j}^{EE} \cdot \{ \mathbf{r}_i \wedge \} \approx 0 \tag{34}
$$

where

$$
\{ \mathbf{r}_i \wedge \} = \begin{pmatrix} 0 & -z_i & y_i \\ z_i & 0 & -x_i \\ -y_i & x_i & 0 \end{pmatrix}. \tag{35}
$$
Comparing Eq. (32) and Eq. (36) we find

\[ \frac{ik_0}{2} \sum_{i,j}^{N} A_{i,j}^x x_j \approx 0 \]

\[ \frac{ik_0}{2} \sum_{i,j}^{N} A_{i,j}^y y_j \approx 0 \]

\[ \frac{ik_0}{2} \sum_{i,j}^{N} A_{i,j}^z z_j \approx 0 \]

Comparing Eq. (32) and Eq. (36) we find

\[ (\beta_x^2 \alpha_x^{EE} + \beta_y^2 \alpha_y^{EE} + \beta_z^2 \alpha_z^{EE}) - \alpha_0^{EE} \approx 0, \]

hence

\[ \alpha_0^{EE} \approx \beta_x^2 \alpha_x^{EE} + \beta_y^2 \alpha_y^{EE} + \beta_z^2 \alpha_z^{EE}. \]
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