Multipole Born series approach to light scattering by Mie-resonant nanoparticle structures

Nikita A Ustimenko¹,∗, Danil F Kornovan¹, Kseniia V Baryshnikova¹, Andrey B Evlyukhin¹,² and Mihail I Petrov¹

¹ School of Physics and Engineering, ITMO University, St. Petersburg 197101, Russia
² Institute of Quantum Optics, Leibniz Universitat Hannover, Hannover 30167, Germany

E-mail: nikita.ustimenko@metalab.ifmo.ru

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Abstract
Optical response of Mie-resonant nanoparticles can be modeled either by full-wave numerical simulations or by the widely used analytical coupled multipole method (CMM). However, an analytical solution in the framework of CMM can be obtained only in a limited number of cases. In this paper, a modification of the CMM in the framework of the Born series and its applicability for the simulation of light scattering by finite nanosphere structures, maintaining both dipole and quadrupole resonances, are investigated. The Born approximation simplifies an analytical analysis of various systems and helps shed light on physical processes ongoing in that systems. Using Mie theory and Green’s functions approach, we analytically formulate the rigorous coupled dipole-quadrupole equations and their solution in the different-order Born approximations. We analyze in detail the resonant scattering by dielectric nanosphere structures such as dimer and ring to obtain the convergence conditions of the Born series and investigate the influence of the physical characteristics such as absorption in particles, type of multipole resonance, and geometry of ensemble on the convergence of Born series and its accuracy.

Keywords: multiple scattering, perturbation theory, Born series, Born approximation, coupled multipoles, nanoparticles, Mie resonances

(Some figures may appear in colour only in the online journal)

1. Introduction
Rapidly developing all-dielectric nanophotonics [1–3] brings new functionalities to nanoscale devices and systems for nonlinear generation [4–8], polarization control [9, 10], sensing [11, 12], lasing [13, 14], and imaging [15]. In this way, achieving high efficiency of future devices requires well-designed nanostructures with optimized parameters, which is often connected to resource-intensive simulations, especially when it comes to irregular structures such as holographic metasurfaces [16, 17] and metalenses [18–20]. Moreover, the extensively developing machine learning algorithms for inverse design of nanophotonic structures rely upon simulations of large parameter sets for proper training of neural networks [21, 22]. Thus, fast and effective methods of the optical properties modeling are constantly required. In many cases high accuracy of numerical calculations is excessive, and approximate algorithms may be suitable [21, 23]. In this paper, we focus on the Born series approach to modeling of optical response of ensembles of resonant subwavelength scatterers.
The Born series formalism is a method for simulating wave propagation in many-body scattering problems. In nanophotonics, one can analyze the optical response of an ensemble of many nanoparticles in a perturbative manner (see figure 1) [24–30]. This method is based on constructing a convergent Born series and replacing it with a finite sum that approximating successively the interaction between particles where the accuracy depends on the number of the terms included in the sum (i.e., on the Born approximation order). Born approximations of different orders have been used to simulate tip-substrate interaction [24, 25], calculate polarizability of a non-spherical particle [26, 27], model the antireflective properties of nanoparticle coatings [29] and optimize metalens design [30]. The applicability of the Born series method, as well as its convergence, are determined by the strength of electromagnetic coupling in the system. To the best of our knowledge, there is no exhaustive physical analysis of Born series applicability to simulate Mie-resonant nanostructures [31, 32] with qualitative criteria of the series convergence. In this work, we try to provide such analysis and criteria and compare the Born series approach to the coupled multipole method (CMM) [3, 33–37].

The paper is organized as follows: in section 2, we overview a general scheme of the Born series with respect to the multipole scattering model; in section 3, we in detail investigate the Born series convergence for a nanosphere dimer (multipole Born series approach). Each such rescattering between multipoles is described by the power of Green’s function $G(r_j, r_p)$. Inset shows the scattering cross section $\sigma_{\alpha\alpha}$ of an individual nanoparticle supporting magnetic dipole (MD), electric dipole (ED), electric quadrupole (EQ), and magnetic quadrupole (MQ) resonances.

$$p^i = \alpha_p \mathbf{E}_{\text{loc}}(r_j),$$
$$m^i = \alpha_m H_{\text{loc}}(r_j),$$
$$\hat{Q}^i = \frac{\alpha_Q}{2} \left[ \nabla \mathbf{E}_{\text{loc}}(r_j) + \mathbf{E}_{\text{loc}}(r_j) \nabla \right],$$
$$\hat{M}^i = \frac{\alpha_M}{2} \left[ \nabla \mathbf{H}_{\text{loc}}(r_j) + \mathbf{H}_{\text{loc}}(r_j) \nabla \right],$$

where index $i = 1 \ldots N$, $\nabla_j$ is the nabla operator with respect to $r_j$, $\alpha_p$, $\alpha_m$, $\alpha_Q$, and $\alpha_M$ are the ED, MD, EQ, and MQ polarizabilities of a dielectric sphere, respectively (see (A.1) in appendix A). A tensor $(\nabla F + F \nabla)$ is defined as follows:

$$(\nabla F + F \nabla)_{\beta\gamma} = \frac{\partial F_{\beta}}{\partial \gamma} + \frac{\partial F_{\gamma}}{\partial \beta},$$

where $F$ is the vector of electric or magnetic field, indices $\beta = x, y, z$ and $\gamma = x, y, z$.

The local field acting on the $j$th nanoparticle is composed of the external field and fields of multipoles generated in all other nanoparticles except the multipoles $j$th nanoparticle:

$$E_{\text{loc}}(r_j) = E_{\text{loc}}(r_j) + E_{p}^j(r_j) + E_{m}^j(r_j) + E_{Q}^j(r_j) + E_{M}^j(r_j),$$

$$H_{\text{loc}}(r_j) = H_{\text{loc}}(r_j) + H_{p}^j(r_j) + H_{m}^j(r_j) + H_{Q}^j(r_j) + H_{M}^j(r_j),$$

where $E_{p}^j(r_j)$ is expressed through the all EDs generated in all nanoparticles except the $j$th one (this is highlighted by $'j$), and so on. The expressions for scattered electric $E'$ and magnetic $H'$ fields of the multipoles are provided in appendix B (see (B.1)). Thus, multipole moments of a certain nanoparticle
(1) linearly depend on the multipole moments of other nanoparticles. Hence, to calculate the multipole moments of all nanoparticles arranged in the finite nanoparticle array, a system of linear equations should be solved:

\[ \mathbf{Y} = \mathbf{Y}_0 + \mathbf{VY}. \tag{3} \]

Here \( \mathbf{Y} \) is the supervector of coupled multipole moments (1), taking into account the interaction of particles, \( \mathbf{Y}_0 \) is the supervector of multipole moments excited only by the external wave, i.e. the local fields are replaced by the incident ones in (1). The block matrix \( \mathbf{V} \) describes the interaction between multipoles. The explicit forms of vectors \( \mathbf{Y} \) and \( \mathbf{Y}_0 \), and matrix \( \mathbf{V} \) are added in appendix C.

Self-consisted CMM solution of (3) can be written as follows:

\[ \mathbf{Y} = (\mathbf{I} - \mathbf{V})^{-1}\mathbf{Y}_0. \tag{4} \]

where \( \mathbf{I} \) is the corresponding identity matrix. The Born series expansion of (4) is a matrix \( (\mathbf{I} - \mathbf{V})^{-1} \) expansion in terms of powers of the matrix \( \mathbf{V} \):

\[ \mathbf{Y} = \mathbf{IY}_0 + \mathbf{VY}_0 + \mathbf{V}^2\mathbf{Y}_0 + \mathbf{V}^3\mathbf{Y}_0 + \ldots. \tag{5} \]

Replacing series (5) by a finite sum, we can obtain the solution of (3) in the Born approximation. In the zero-order Born approximation, the interaction between multipoles is neglected:

\[ \mathbf{Y} = \mathbf{Y}_0. \tag{6} \]

The \( m \)th order Born approximation is expressed through the \((m - 1)\)th order Born approximation:

\[ \mathbf{Y}_m = \mathbf{Y}_0 + \mathbf{VY}_{m-1}. \tag{7} \]

The main criterion of applicability of the Born series is its convergence. The necessary convergence condition of series (5) is \( \det(\mathbf{I} - \mathbf{V}) \neq 0 \); otherwise, a system should be far from the condition of configurational resonance [25]. In this case, a strong electromagnetic coupling between multipoles cannot be approximated by the Born approximation of any order. The sufficient condition is \( \|\mathbf{V}\| < 1 \). Generally, series (5) converges only when all eigenvalues of matrix \( \mathbf{V} \) are inscribed in a unit circle on a complex plane. This condition is mathematically strict but cannot be applied immediately to answer the question about the Born series convergence. From a physical point of view, the Born series diverges when a strong electromagnetic interaction between the nanoparticles appears. Obviously, the electromagnetic coupling is enhanced and the question of the Born series becomes very important, at the Mie resonance inherent for each particle in the ensemble.

3. Nanosphere dimer

We start our analysis from a dimer of identical nanospheres shown in figure 2(a). On the one hand, dimer structures can be utilized for various photonic purposes such as local magnetic field enhancement [39], nonlinear generation enhancement [7, 8, 40], controlling the directionality of scattering and other purposes, and, on the other hand, offer a simple analytical solution in the framework of the CMM.

Close to a particular multipolar resonance, one can consider the dominant contribution of an isolated (specific) resonant multipole. In this case, for the ED resonance, (3) can be written as follows:

\[ p^1 = \alpha_p E_0 + \alpha_p k^2 \varepsilon^{-1} \hat{G}_{12}^{pp} p^2, \]
\[ p^2 = \alpha_p E_0 + \alpha_p k^2 \varepsilon^{-1} \hat{G}_{22}^{pp} p^1, \tag{8} \]

where \( p^1 \) and \( p^2 \) are the dipole moments of the first and second nanoparticle, respectively, \( k = 2\pi/\lambda \) is the free-space wavenumber of external plane wave, \( \hat{G}_{12}^{pp} \equiv G^{pp}(r_1, r_2) \) is the dyadic dipole Green’s function having a symmetry property \( \hat{G}_{12}^{pp} = \hat{G}_{21}^{pp} \) (see (D.1) in appendix D). The nanoparticles are separated from each other by the distance \( D \) along the y-axis as shown in figure 2(a).

Due to the structure symmetry, both dipole moments have only one non-zero component along the incident field:

\[ p_{\beta} = \frac{\alpha_p E_0}{1 - \alpha_p k^2 \varepsilon^{-1} \hat{G}_{12,\beta\beta}^{pp}}, \quad j = 1, 2, \tag{9} \]

where \( \hat{G}_{12,\beta\beta}^{pp} = G_{\beta\beta}^{pp}(r_1, r_2) \) is the \( \beta \)-element of Green’s function \( \hat{G}_{12}^{pp} \). The transverse-polarized dipole moments \( E_0 \parallel x \) and \( \beta = y \) for the longitudinal polarization \( E_0 \parallel y \).

The dipole moments calculated by (9) take into account the electromagnetic coupling between two dipoles rigorously. We can also approximate the coupling by expanding the moments...
Table 1. Conditions and parameters of Born series convergence for the dimer of ED, MD, EQ, and MQ scatterers in vicinity of isolated resonance (R) for both linear polarizations (P) of incident wave. (a) Summary of convergence criteria. (b) Solutions of inequalities in (a) for Mie resonances in non-absorptive particles ($\varepsilon'' = 0$). Here $\Delta_{\text{ED}}, a_{\text{MD}}, a_{\text{EQ}}, a_{\text{MQ}}$ are the wavelengths of ED, MD, EQ, MQ resonances, respectively; $a_0$ and $b_0$ are the Mie-coefficients; $G_{12,\text{ED}}^{pp}$ and $G_{12,\text{MQ}}^{pp}$ are the elements of dipole Green’s function $G^{pp}(r_1,r_2)$ (see (D.1) in appendix D); expressions for quantities $b_{12,\text{ED}}^{pp}$ and $b_{12,\text{MQ}}^{pp}$ are written in appendix E (see (E.1)).

(a) Convergence criteria

| R/P | $E_0 \parallel y$ | $E_0 \parallel x$ |
|-----|-----------------|-----------------|
| ED  | $6\pi|a_1||G_{12,\text{yy}}^{pp}| < 1$ | $6\pi|a_1||G_{12,\text{xx}}^{pp}| < 1$ |
| MD  | $6\pi|b_1||G_{12,\text{yy}}^{pp}| < 1$ | $6\pi|b_1||G_{12,\text{xx}}^{pp}| < 1$ |
| EQ  | $60\pi|a_2||b_{12,\text{xx}}^{pp}| < 1$ | $60\pi|a_2||b_{12,\text{xx}}^{pp}| < 1$ |
| MQ  | $60\pi|b_2||b_{12,\text{xx}}^{pp}| < 1$ | $60\pi|b_2||b_{12,\text{xx}}^{pp}| < 1$ |

(b) Critical distances ($\varepsilon'' = 0$)

| R/P | $E_0 \parallel y$ | $E_0 \parallel x$ |
|-----|-----------------|-----------------|
| ED  | $D > 0.29\lambda_{\text{ED}}$ | $D > 0.21\lambda_{\text{ED}}$ |
| MD  | $D > 0.21\lambda_{\text{MD}}$ | $D > 0.29\lambda_{\text{MD}}$ |
| EQ  | $D > 0.44\lambda_{\text{EQ}}$ | $D > 0.33\lambda_{\text{EQ}}$ |
| MQ  | $D > 0.33\lambda_{\text{MQ}}$ | $D > 0.44\lambda_{\text{MQ}}$ |

In the case of non-absorbing particles ($\varepsilon'' = 0$), the Mie-resonance condition for electric or magnetic modes is following: $a_0 = 1$ or $b_0 = 1$ correspondingly, which immediately provides the at-resonance convergence criteria of the multipole Born series. Solving inequality (12), we can find the critical distances which, for the longitudinal and transverse ED modes, are $D^{(1)} = 0.29\lambda_{\text{ED}}$ and $D^{(1)} = 0.21\lambda_{\text{ED}}$, respectively, where $\lambda_{\text{ED}}$ is the wavelength of nanoparticle ED resonance. If the distance between the centers of particles is larger than the critical one, the Born series converges, otherwise it diverges. The critical distances for other multipole resonances are specified in table 1(b). One should notice that, at the Mie resonance, the critical distance for the corresponding multipole term depends only on resonant wavelength, which can be considered as a manifestation of the scaling character of the problem as $na/\lambda$.

Once the non-zero Ohmic losses are present ($\varepsilon'' > 0$), the Mie-coefficients at the resonances become less than unity $|a_0|, |b_0| < 1$ that decreases the critical distances. Figure 2(b) shows the at-resonance absolute values of the four Mie-coefficients for varying $\varepsilon''$ of nanoparticle material. In the simulation, $\varepsilon'$ of the particle remained constant while the particle diameter $d$ was changed in order to keep the resonant wavelength constant for different $\varepsilon''$. As the convergence criteria depend on the absolute values of Mie-coefficients (see table 1(a)), with increasing of materials losses the critical distances also decrease for all multipole resonances (see figure 2(c)). This behavior has a clear physical explanation as additional losses suppress nanoparticle scattering making electromagnetic coupling between two particles weaker, therefore, improving the convergence of the Born series.

Above we considered only the isolated resonances and took into account only the contributions of corresponding resonant multipoles. In the general case, the higher order multipoles can be excited simultaneously by a linearly polarized wave and their contribution requires larger distances between particles in order to obtain convergence: $D > 0.29\lambda_0$ for the dipole resonance, and $D > 0.44\lambda_0$ for the quadrupole resonance. For absorptive particles, the critical distances can be calculated using the inequalities from table 1(a).

The critical distance values provide the applicability conditions of the Born series approximation, however, they do not provide any information about its accuracy. For this reason, we analyzed the scattering cross section and compared it with the results obtained from CMM introducing the accuracy parameter $\Delta$:

$$\Delta = \left| \frac{\sigma^{(m)} - \sigma^{(\text{CMM})}}{\sigma^{(\text{CMM})}} \right|,$$

where $\sigma^{(\text{CMM})}$ and $\sigma^{(m)}$ are the scattering cross sections of the dimer calculated using CMM multipole moments (4) and multipole moments in the $m$th Born approximation, respectively. In the dipole-quadrupole approximation, the scattering cross section of non-absorptive particle structure in a vacuum can
be calculated by the following formula [36]:

\[
\sigma = \frac{k}{\varepsilon_0 |E_0|^2} \Im \sum_{j=1}^{N} \left[ E_0^*(r_j) \cdot p + \mu_0 \frac{\nabla H_0^*(r_j)}{2} : \mathbf{M} + \mu_0 H_0^*(r_j) \cdot m + \frac{\mathbf{E}_0^*(r_j) + \mathbf{E}_0(r_j) \nabla}{12} : \mathbf{O} \right],
\]

(14)

where \(*\) denotes complex conjugation, \(\mathbf{T}\) denotes the transpose operation, and \(\cdot\) and \(:\) denote the scalar products between vectors and dyads (tensors), respectively.

We placed one particle at the origin of the coordinate system, and varied the position of the second particle, while the distance between the particles was simultaneously greater than the critical parameter (0.29\(\lambda_0\) or 0.44\(\lambda_0\)) and their diameter (200 nm). Figure 3 shows the minimal number among the Born approximation orders \(m\) one should take in order to provide the accuracy parameter error \(\Delta \leq 0.1\) for considered resonances. In figure 3, we simultaneously take into account four multipoles (ED, MD, EQ, and MQ). One can see that the convergence of the series at MD resonance is much weaker than for ED as the resonance is much more pronounced and the scattered partial fields are stronger. At the same time, the series convergence is slower along the E-field when the distance between the nanoparticles becomes small enough owing to stronger near-field interaction between particles. Very similar behavior is observed for quadrupole resonances in figures 3(c) and (d). One can also see that the diagram for the MQ resonance (figure 3(d)) has a sharp dependence on the angle showing the abrupt change of the required number of Born orders from 6 to 0 or 1 due to the strong angular dependence of MQ coupling.

4. Nanosphere ring

Another very illustrative ensemble geometry yet allowing rigorous analysis is a regular ring of \(N\) nanoparticles shown in figure 4. The ring structures often become a building block of light focusing metasurface lenses [15], thus understanding their optical response can be critical for designing metasurfaces. The interactions between the nanoparticles influence the convergence of multipole Born series close to the collective resonances. We consider system (3) in purely ED approximation solve assuming and focusing on a mode with radial polarization of dipole moments (see figure 4), which can be excited, for instance, by vectorial optical beams such as radially polarized Bessel beam [43]. In this case, (3) is:

\[
p_{j}^{p} = \alpha_p E_0(r_j) + \frac{k^2}{\varepsilon_0} \sum_{l=1,j\neq j}^{N} G_{jj}^{pp}(r_j,r_l) p_{j}^{l},
\]

(15)

where \(r_j = R \cos(\varphi_j), \sin(\varphi_j), 0\)^T is the position of the \(j\)th nanoparticle in ring of radius \(R, \varphi_j = 2\pi (j-1)/N\) is the angular coordinate of the \(j\)th nanoparticle, \(G_{jj}^{pp}\) is the component of the vacuum dipole Green’s tensor in cylindrical coordinates (see (F.2) in appendix F), and \(p_{j}^{p} = [p_{j}\mathbf{e}_1, p_{j}\mathbf{e}_2]^T\). Note that \(p_{j}\mathbf{e}_3 = 0\) for all nanoparticles in the structure.

![Figure 3. The Born approximation order as a function of the second particle position while the first particle is placed at the coordinate system origin for the (a) ED resonance (\(\lambda_{\text{ED}} = 555\) nm), (b) MD resonance (\(\lambda_{\text{MD}} = 731\) nm), (c) EQ resonance (\(\lambda_{\text{EQ}} = 407\) nm), and (d) MQ resonance (\(\lambda_{\text{MQ}} = 505\) nm). The position of the circle point indicates the second particle position, and its color corresponds to the order of Born approximation that allows calculating the extinction cross section with error (13) of less than 10%. The distance between the particles is measured in resonant wavelength \(\lambda\). The particle diameter \(d = 200\) nm, permittivity of the particle material \(\varepsilon = 12.5\). The normally incident plane wave is horizontally polarized.](image)
the ring is following:

\[ |S| = |\alpha_p k^2 \varepsilon_0^{-1} \tilde{G}_{\rho \rho}^{pp}| < 1, \]

while at the configurational resonance, when \(|S| = 1\), the Born series always diverges. Figure 4 shows the convergence parameter \(|S|\) (defined in (17)) for the radial mode of rings with different inter-particle distances and radii. It can be seen that the Born series may diverge when the distance between particles is very small or when the distance between the dipoles is equal to an integer number of resonant wavelengths. The latter case corresponds to the geometric (diffraction) resonances indicating a strong electromagnetic interaction between nanoparticles. In addition, with increasing of the number of radially oriented dipoles in the ring, the convergence parameter for the considered mode of ring approaches the dependence for a transverse mode of a linear chain (see figure 4). Indeed, in the limit of an infinite ring \(N, R \to \infty\) with fixed inter-particle distance, the dipole sum of a ring tends to the dipole sum of a chain known analytically (see appendix F).

The convergence parameter \(|S|\) of the mode can be tied with its dispersion and \(Q\)-factor. The polarizability of a single particle in the vicinity of isolated high-\(Q\) resonance \(\omega_0\) can be represented as:

\[ \alpha_p = -\alpha_0 \frac{\gamma_0/2}{\omega - \omega_0 + i\gamma_0/2}, \]

where \(\alpha_0 = 6\pi \varepsilon_0 k^{-3}\) is the resonant value of dipole polarizability, \(\gamma_0/2\) is the radiative losses rate of a single particle. Inserting (18) into (16) and using quasi-resonant approximation \(\tilde{G}_{\rho \rho}^{pp}(\omega) \approx \tilde{G}_{pp}^{pp}(\omega_0)\), we obtain the following expression for the effective polarizability:

\[ \alpha_{\text{eff}} \approx -\alpha_0 \frac{\gamma_0/2}{\omega - \omega_0 + i\gamma_0/2}, \]

where \(\Delta \omega_0\) is then detuning of resonance frequency from the resonant of an individual particle, \(\gamma/2\) is the radiative losses rate of the ring mode. At the resonance of a single particle, convergence condition (17) can be written as:

\[ |S| = \sqrt{\left( \frac{\Delta \omega_0}{\gamma_0/2} \right)^2 + \left( \frac{\gamma}{\gamma_0} - 1 \right)^2} < 1. \]

If \(|\Delta \omega_0| \ll \gamma_0/2\) (19) can be expressed through the ratio of \(Q\)-factors:

\[ |S| \approx \left| \frac{Q_0}{Q} - 1 \right| < 1, \]

where \(Q_0\) is the total \(Q\)-factor of a single-particle resonance, and \(Q\) is \(Q\)-factor of the ring mode, showing that the stronger is the collective resonance the slower does the convergence of the Born series as for \(Q > Q_0|S| \sim 1\).
5. The performance of Born series method

In the last part of our paper, we turn to analyze the performance efficiency of the Born approximation. We compare the computational time required for the rigorous solution of system (3) with the solution of (3) in the Born approximations for both dipole and dipole-quadrupole models. We consider a ring of \( N \) spherical nanoparticles (of diameter \( d = 200 \text{ nm} \)) with the inter-particle distance \( D = 0.6 \lambda_{\text{ED}} \) such that the Born series converges at the wavelength of ED resonance \( \lambda_{\text{ED}} = 615 \text{ nm} \). For the ring structure of \( N \) nanoparticles, a dimension of the matrix of system (3) is \( 6N \times 6N \) and \( 24N \times 24N \) for dipole and dipole-quadrupole models, respectively. We vary the particle number \( N \) from 20 to 120 and estimate the time required to compute the solution with help of \textit{lin.solve} function in Matlab for coupled dipoles (multipoles) and the time required for Born series summation.

The results of the comparison are presented in figure 5 in normalized units. In figure 5, \( \tau \) is the average time of \textit{lin.solve} work for the \( 1000 \times 1000 \) matrix and \( 1000 \times 1 \) right-hand side column of random complex numbers. All quantities in figure 5 were averaged after ten iterations. The computation time of CMM behaves as \( O(N^3) \) while the Born series computation time behaves as \( O(N^2) \), which provides almost one and two orders of magnitude decrease of computation time comparing to the coupled dipole and dipole-quadrupole solutions for \( N = 100 \) particles. At the same time, the numerical error \( \Delta \), provided by (13) and indicated by the circle diameter, stays well below 2\% for the third-order Born approximation.

6. Conclusion

In this paper, we analyzed the Born series approach for modeling the optical response of finite arrays of Mie-resonant nanoparticles. The Born series method allows us to approximately compute the interaction in particle systems that significantly reduce the total computation time and the usage of computational resources comparing to the rigorous coupled multipole models. We investigate the accuracy of this method and, in particular, a convergence of the Born series in the vicinity of Mie-resonances. Under the dipole and quadrupole approximation, we analytically and numerically find the convergence conditions for the nanosphere dimer and nanosphere ring. We believe that the proposed simulation method and obtained results will be useful for the optimization and modeling of nanophotonic systems such as metasurfaces, metalenses, and nanoantennas.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Acknowledgments

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Appendix A. Mie-polarizabilities of spherical particle

For Mie scattering regime, the ED \( \alpha_p \), the MD \( \alpha_m \), the EQ \( \alpha_Q \), and the MQ \( \alpha_M \) polarizabilities of spherical nanoparticles in the field of external plane wave are expressed through the analytically known Mie-coefficients \( a_1, a_2, b_1, \) and \( b_2 \) [34, 36]:

\[
\begin{align*}
\alpha_p &= \frac{6\pi\varepsilon_0\varepsilon_S}{k_S^3} a_1, \\
\alpha_m &= \frac{6\pi}{k_S^3} b_1, \\
\alpha_Q &= \frac{120\pi\varepsilon_0\varepsilon_S}{k_S^5} a_2, \\
\alpha_M &= \frac{40\pi}{k_S^5} b_2, \\
\end{align*}
\]

where \( i \) is the imaginary unit, \( \varepsilon_0 \) is the dielectric permittivity of vacuum, \( \varepsilon_S \) is the dielectric permittivity of particles surrounding medium, \( k_S = k\sqrt{\varepsilon_S} \) is the wavenumber in a medium with permittivity \( \varepsilon_S \). In the main text, \( \varepsilon_S = 1 \).

Appendix B. Multipoles fields

Electric and magnetic fields of dipoles and quadrupoles at the nanoparticle point \( r_j \) generated by nanoparticles located at the points \( r_i\) (\( i \neq j \)) [36]:

\[
\begin{align*}
E'_p (r_j) &= \frac{k^2}{\varepsilon_0} \sum_{l=1,l\neq j}^N \hat{G}'_{jl} p^l, \\
H'_p (r_j) &= \frac{ck}{\varepsilon_0} \sum_{l=1,l\neq j}^N \hat{G}'_{jl} m^l, \\
E'_q (r_j) &= \frac{ik}{\varepsilon_0} \sum_{l=1,l\neq j}^N \hat{G}'_{jl} \left( \hat{Q} n_j \right), \\
H'_q (r_j) &= \frac{ck}{\varepsilon_0} \sum_{l=1,l\neq j}^N \hat{G}'_{jl} \left( \hat{Q} n_j \right), \\
E'_M (r_j) &= 3 \frac{ik}{c\varepsilon_0} \sum_{l=1,l\neq j}^N \hat{G}'_{jl} \left( \hat{M} n_j \right), \\
\end{align*}
\]
\[ H'_M(r_j) = 3 k_\Sigma^2 \sum_{i=1, i\neq j}^{N} G_{ij}^{QO}(\hat{M} n_{ij}) , \quad \text{(B.1)} \]

where \( c \) is the light speed in vacuum, \( n_{ij} \) is the unit vector from \( j \)th nanoparticle to \( i \)th nanoparticle: \( n_{ij} = (r_j - r_i) / |r_j - r_i| \). \( G_{ij}^{pp} = \hat{G}_{pp}(r_j, r_i) \), \( \hat{G}_{ij}^{pp} = G_{ij}^{pp}(r_j, r_i) \), \( G_{ij}^{QO} = G_{ij}^{QO}(r_j, r_i) \), \( \hat{G}_{ij}^{QO} = \hat{G}_{ij}^{QO}(r_j, r_i) \). \( \hat{G}_{ij}^{QM} = \hat{G}_{ij}^{QM}(r_j, r_i) \) are the dyadic Green’s functions of the dipole and quadrupoles in the free space. The expressions for Green’s functions are provided in appendix D.

**Appendix C. System of linear equations for calculating dipole and quadrupole moments**

Introduced in (3) \( Y \) is the \( 24N \) dimensional column composed of \( N \) sub-vectors:

\[ Y = [Y^1, Y^2, \ldots, Y^N]^T , \quad \text{(C.1)} \]

where \( Y^j \) is the sub-vector of multipole moment values corresponding to the \( j \)th particle:

\[ Y^j = [p_x^j, \ldots, p_N^j, m_x^j, m_y^j, Q_x, \ldots, Q_z, M_{xx}^j, \ldots, M_{zz}^j]^T . \quad \text{(C.2)} \]

The matrix \( \hat{V} \) is composed of blocks \( V_{ij} \) describing the interaction between multipole of the \( i \)th and \( j \)th particles in the array \((i \neq j)\):

\[ \hat{V} = \begin{pmatrix} \hat{0} & \hat{V}_{12} & \hat{V}_{13} & \cdots & \hat{V}_{1N} \\ \hat{V}_{21} & \hat{0} & \hat{V}_{23} & \cdots & \hat{V}_{2N} \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ \hat{V}_{N1} & \hat{V}_{N2} & \hat{V}_{N3} & \cdots & \hat{0} \end{pmatrix} . \quad \text{(C.3)} \]

The block \( \hat{0} \) is the \( 24N \times 24N \) dimensional matrix of zeroes. The block \( \hat{V}_{ij} \) also has a dimension of \( 24N \times 24N \) and consists of several sub-blocks:

\[ \hat{V}_{ij} = \begin{pmatrix} \hat{A}_{ij}^{pp} & \hat{A}_{ij}^{pm} & \hat{A}_{ij}^{QM} & \hat{A}_{ij}^{QO} & \hat{A}_{ij}^{QM} \\ \hat{B}_{ij}^{pm} & \hat{B}_{ij}^{pp} & \hat{B}_{ij}^{QO} & \hat{B}_{ij}^{QM} \\ \hat{C}_{ij}^{pm} & \hat{C}_{ij}^{pp} & \hat{C}_{ij}^{QO} & \hat{C}_{ij}^{QM} \\ \hat{D}_{ij}^{pm} & \hat{D}_{ij}^{pp} & \hat{D}_{ij}^{QO} & \hat{D}_{ij}^{QM} \end{pmatrix} . \quad \text{(C.4)} \]

Here \( T \) denotes the transpose operation, \( \otimes \) denotes the Kronecker product explicitly defined for \( (C.5) \) as:

\[ \begin{align*}
\hat{G}_{ij}^{QO}(r_j, r_i) &= [\hat{G}_{ij}^{QO}(r_j, r_i)^x \hat{G}_{ij}^{QO}(r_j, r_i)^y \hat{G}_{ij}^{QO}(r_j, r_i)^z] , \\
\hat{G}_{ij}^{QM}(r_j, r_i) &= [\hat{G}_{ij}^{QM}(r_j, r_i)^x \hat{G}_{ij}^{QM}(r_j, r_i)^y \hat{G}_{ij}^{QM}(r_j, r_i)^z] .
\end{align*} \]

Above the components of vector \( n_{ij} \) are noted as \( n_{ij} = [\tilde{x}_{ij}, \tilde{y}_{ij}, \tilde{z}_{ij}] \), where \( \tilde{x}_{ij} = (x_j - x_i)/|r_j - r_i|, \tilde{y}_{ij} = (y_j - y_i)/|r_j - r_i|, \tilde{z}_{ij} = (z_j - z_i)/|r_j - r_i| \).

\[ \hat{A}_{ij}^{QM} = \alpha_p \frac{3ik}{c \varepsilon_0} \hat{H}_{ij}^{QM}(r_j, r_i) , \quad \text{(C.5)} \]

where

\[ \hat{H}_{ij}^{QM}(r_j, r_i) = \hat{G}_{ij}^{QM}(r_j, r_i) \otimes n_{ij}^T . \]

The matrix \( \hat{F}^{pp} \) is defined analogously.

\[ \hat{F}^{pp}(r_j, r_i) = \hat{F}^{pp}(1)(r_j, r_i) + \hat{F}^{pp}(2)(r_j, r_i) , \quad \text{(C.6)} \]
\[
\begin{align*}
\hat{F}_{\beta}^{pp(2)} &= \frac{\partial}{\partial x} (G_{jl}^{pp(2)}) \left( \frac{\partial}{\partial x} (G_{jl}^{pp(2)}) \right) \\
&= \frac{\partial}{\partial y} (G_{jl}^{pp(2)}) \left( \frac{\partial}{\partial y} (G_{jl}^{pp(2)}) \right) \\
&= \frac{\partial}{\partial z} (G_{jl}^{pp(2)}) \left( \frac{\partial}{\partial z} (G_{jl}^{pp(2)}) \right)
\end{align*}
\]

where

\[
\hat{C}_{\beta}^{pm} = \frac{\alpha_0}{2} \frac{ik}{c_0} \hat{F}_{\beta}^{pp}(r_j, r_i),
\]

and

\[
\hat{F}_{\beta}^{pp}(r_j, r_i) = \hat{F}_{\beta}^{pp(1)}(r_j, r_i) + \hat{F}_{\beta}^{pp(2)}(r_j, r_i).
\]

The tensors \(\hat{F}_{\beta}^{pp(1)}, \hat{F}_{\beta}^{pp(2)}, \hat{F}_{\beta}^{pp(3)}\) and \(\hat{F}_{\beta}^{pp(3)}\) are defined as (C.6) and (C.7) with the corresponding Green’s function, respectively.

\[
\begin{align*}
\hat{\mathcal{G}}_{\alpha\beta}^{pm}(r, r_0) &= \frac{e^{ikl}}{4\pi l} \left( 1 + \frac{i}{k_\|} - \frac{1}{k_\|^2} \right) \delta_{\alpha\beta} \\
&+ \left( -1 - \frac{3i}{k_\|=} + \frac{3}{k_\|=^2} \right) n_\alpha n_\beta,
\end{align*}
\]

Appendix D. Multipole dyadic Green’s functions, and their derivatives

The elements of dyadic dipole and quadrupole Green’s functions for the multipole sources located in the free space [36]:

\[
\begin{align*}
\hat{G}_{\alpha\beta}^{pp}(r, r_0) &= \frac{e^{ikl}}{4\pi l} \left( 1 + \frac{i}{k_\|} - \frac{1}{k_\|=^2} \right) \delta_{\alpha\beta} \\
&+ \left( -1 - \frac{3i}{k_\|=} + \frac{3}{k_\|=^2} \right) n_\alpha n_\beta,
\end{align*}
\]

\[
\begin{align*}
\hat{G}_{\alpha\beta}^{QQ}(r, r_0) &= \frac{ekl}{2\pi l} \left( -1 - \frac{3i}{k_\|=} + \frac{6}{k_\|=^2} + \frac{6i}{k_\|=^3} \right) \delta_{\alpha\beta} \\
&+ \left( 1 + \frac{6i}{k_\|=} - \frac{15}{k_\|=^2} - \frac{15i}{k_\|=^3} \right) n_\alpha n_\beta,
\end{align*}
\]

Elements of the Cartesian derivatives of dyadic Green’s functions (D.1)–(D.4):

\[
\hat{\mathcal{G}}_{\alpha\beta}^{pm}(r, r_0) = \frac{k_{\alpha\beta}}{2\pi l} \left( 1 + \frac{3i}{k_\|=} - \frac{3}{k_\|=^2} \right) n_\alpha n_\beta.
\]
The solution for isolated EQ resonance:

\[ G_{\alpha \beta}^{\text{EQ}}(r, r_0) = \frac{\alpha_e^{\text{EQ}}}{4\pi} \left\{ \left( \frac{i}{k_{sl}} - \frac{2}{k_{sl}^2} - \frac{3i}{k_{sl}^3} + \frac{3}{k_{sl}^4} \right) \delta_{\alpha \beta} r_{\gamma}, \beta \right\}, \quad (D.5) \]

The solution for isolated MQ resonance:

\[ G_{\alpha \beta}^{\text{MQ}}(r, r_0) = \frac{ik_s^{\text{MQ}}}{24\pi} \left\{ \left( -i + \frac{4}{k_{sl}} + \frac{12i}{k_{sl}^2} - \frac{24i}{k_{sl}^3} - \frac{24i}{k_{sl}^4} \right) \delta_{\alpha \beta} r_{\gamma}, \beta \right\}, \quad (D.6) \]

Appendix E. Solution of CMM equation for nanoparticle dimer at the wavelengths of isolated MD, EQ and MQ resonances

The solution of (3) in the framework of specific resonant MD response for the dimer placed in free space \((\varepsilon_5 = 1, k = k_5)\):

\[ m_j^D = \frac{\alpha_0 h_0}{1 - \alpha_0 e^{\text{MD}} G_{\alpha \beta}^{\text{MD}}}, \quad j = 1, 2, \]

where \(\beta = y\) for \(E_0\parallel x\), and \(\beta = x\) for \(E_0\parallel y\).

The solution for isolated EQ resonance:

\[ Q_{\beta}^{\text{EQ}} = \frac{\alpha_0}{1 - \alpha_0 e^{\text{EQ}} B_{12, \beta}^{\text{EQ}}} \left\{ \frac{ik_s e^{\text{EQ}}}{24\pi} \left[ -i + \frac{3i}{k_{sl}^2} - \frac{21i}{k_{sl}^3} - \frac{48i}{k_{sl}^4} - \frac{48}{k_{sl}^5} \right], \beta = y \right\}, \quad (D.1) \]

The solution for isolated MQ resonance:

\[ B_{12, \beta}^{\text{MQ}} = \frac{i k_s e^{\text{MQ}}}{12\pi} \left[ -1 + \frac{3i}{k_{sl}^2} + \frac{6i}{k_{sl}^3} + \frac{6i}{k_{sl}^4} \right], \quad \beta = x, \quad (F.1) \]

where \(\beta = y\) for \(E_0\parallel x\), and \(\beta = x\) for \(E_0\parallel y\).

Appendix F. Ring dipole sum

In order to find the fields generated at the position of the jth dipole by all other radially oriented dipole scatterers formed a ring of radius \(R\), one need to deal with the sum:

\[ G_{\beta j}^{\text{pp}} = \sum_{l=1, l \neq j}^{N} G_{\beta j}^{\text{pp}}(r_j, r_l), \quad (F.1) \]

where \(r_j = R(\cos(\varphi_j), \sin(\varphi_j), 0)^T\), and \(G_{\beta j}^{\text{pp}}\) is the component of the vacuum dipole dyadic Green’s function in cylindrical coordinates given by the following expression:

\[ G_{\beta j}^{\text{pp}}(r_j, r_l) = \frac{ke^{iD_j}}{4\pi} \left\{ \left( \frac{1}{kD_j} + \frac{i}{k^2D_j^2} - \frac{1}{kD_j^2} \right) \right\}, \quad (F.2) \]

where \(D_j = 2 \sin \left( \frac{\left| \varphi - \varphi_j \right|}{2} \right) / 2\) is the distance between the dipoles \(j\) and \(l\). Let us consider the limit of an infinitely large ring so \(N \to \infty\), but simultaneously the distance between the neighboring dipoles \(D = 2 \sin \left( \frac{\left| \varphi - \varphi_j \right|}{2} \right) / 2\) is kept fixed. First of all, notice that \(\lim_{N \to \infty} D_j = lim_{N \to \infty} 2 \sin \left( \frac{\left| \varphi - \varphi_j \right|}{2} \right) / 2\) is kept fixed. Second, in (F.2), the term \(\sin^2 \left( \frac{\varphi - \varphi_j}{2} \right) / 2\) vanishes as \(N \to \infty\), and we are left with:

\[ G_{\beta j}^{\text{pp}}(r_j, r_l) = \frac{ke^{iD_j}}{4\pi} \left\{ \left( \frac{1}{kD_j} + \frac{i}{k^2D_j^2} - \frac{1}{kD_j^2} \right) \right\}, \quad (F.3) \]

where \(D_j = 2 \sin \left( \frac{\left| \varphi - \varphi_j \right|}{2} \right) / 2\) is the distance between the dipoles \(j\) and \(l\).
which is exactly the lattice sum for an infinite periodic chain of transversally oriented dipole moments, which is known analytically [44]. Finally, this allows us to write the following:

\[
\frac{i 6 \pi}{k} \tilde{G}_p^{pp} \to 3 \left( \frac{i \text{Li}_1(e^{i k D})}{k D} - \frac{i \text{Li}_2(e^{i k D})}{(k D)^2} - \frac{i \text{Li}_3(e^{i k D})}{(k D)^3} \right),
\]  

(4.4)

where \( \text{Li}_s(e^{i k D}) \) is the polylogarithm function of order \( s = 1, 2, 3 \) and argument \( e^{i k D} \).

**ORCID iDs**

Nikita A Ustimenko  
https://orcid.org/0000-0002-5137-493X

Daniil F Kornovan  
https://orcid.org/0000-0002-4851-0697

Kseniia V Baryshnikova  
https://orcid.org/0000-0002-3753-3543

Andrey B Evlyukhin  
https://orcid.org/0000-0002-1801-6778

Mihail I Petrov  
https://orcid.org/0000-0001-8155-9778

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