Computer algebra solution to Mie scattering problem for the stratified sphere with nonlocal plasmonic layers

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Abstract

- Scattering by spherical particle with an arbitrary amount of concentric layers optionally featuring **spatial dispersion effects** is considered;
- Several problem statements featuring different boundary condition sets are examined with a core-shell particle as an example;
- Computer algebra code (*symMie*) based on the MATLAB/Octave symbolic engine is implemented to solve the problem for the **user-defined boundary condition set.**

Motivation

Plasmonic particles with scale less than 10 nm are known to exhibit unique scattering properties related to the quantum spatial dispersion effects [1-3]. These are not accounted for in the majority of electromagnetic solvers that are conventionally used to simulate scattering. Rigorous evaluation of such structures via i.e. Time-Dependent Density Functional Theory (TDDFT) is possible but computationally demanding for large particles ($d > 5nm$) [4]. Due to the widespread of these particles in cutting-edge applications [5], several nonlocal-corrected Maxwell-based models were recently proposed in attempt to interpret dispersive phenomena in terms of classic constitutive equations and boundary conditions (BC). Each model, i.e. Mie theory in local response approximation (**LRA**), Drude hydrodynamic theory (**HDT**), Generalized Nonlocal Optical Response (**GNOR**), or approach based on mesoscopic Feibelman formalism implies its own scattering problem statement involving possibly modified Maxwell equations and BC [1-3,6-8]. Aim of the current work is to demonstrate the advantages of a computer algebra approach capable of solving scattering by spherical layered particle for the user-defined BC set employing complete $\mathbf{M}_{\substack{e\\o}mn}, \mathbf{N}_{\substack{e\\o}mn}, \mathbf{L}_{\substack{e\\o}mn}$ functional basis.

Local response approximation (LRA)

We consider glass core (D_0) and Ag shell (D_1) sphere in free space (D_2) as an example.

$$\begin{aligned}
&\mathrm{curl}\mathbf{H}_\zeta(M) = -jk\varepsilon_\zeta \mathbf{E}_\zeta(M), && M \in D_\zeta,\\
&\mathrm{curl}\mathbf{E}_\zeta(M) = jk\mu_\zeta \mathbf{H}_\zeta(M), &&\\
&\mathrm{div}\mathbf{E}_\zeta(M) = 0, && \zeta = 0,1,2, \qquad (1)\\
&\mathrm{div}\mathbf{H}_\zeta(M) = 0, &&\\
&[\mathbf{E}_\zeta(P)\times\mathbf{n}_P] = [\mathbf{E}_{\zeta+1}(P)\times\mathbf{n}_P], && P \in \partial D_{\zeta+1},\\
&[\mathbf{H}_\zeta(P)\times\mathbf{n}_P] = [\mathbf{H}_{\zeta+1}(P)\times\mathbf{n}_P]. &&
\end{aligned}$$

Maxwell equations and boundary conditions are **the same for all layers** allowing direct analytic and numeric solutions for an arbitrary amount of layers.

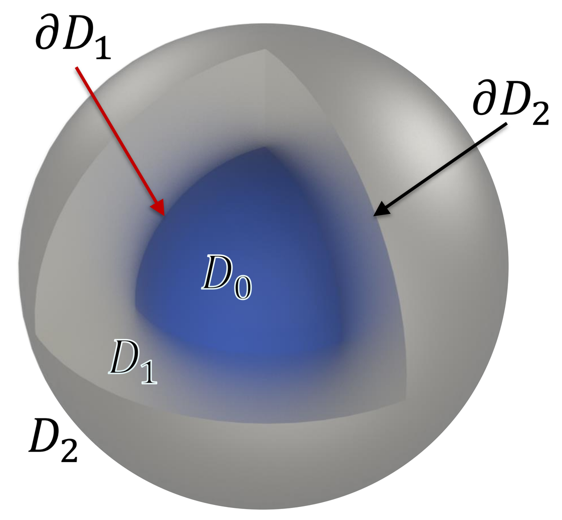

Fig. 1. Geometry of the model: plane wave scattering on the sphere with transparent core (refractive index 1.4, $r_1 = 3nm$) and silver shell ($r_2 = 6nm$) is considered.

Spatial dispersion (HDT/GNOR)

Ag shell D_2 is now nonlocal, particle is the same.

$$\mathrm{curl}\mathbf{H}_2(M) = -jk\left(\varepsilon_2 + \frac{\beta_2^2\left(\varepsilon_2' + d_2(\gamma_2 + j\omega)\right)}{\omega(\omega + j\gamma_2)}\mathrm{graddiv}\right)\mathbf{E}_2(M),$$

$$\begin{aligned}
&\mathrm{curl}\mathbf{H}_1(M) = -jk\varepsilon_1 \mathbf{E}_1(M), && M \in D_\zeta,\\
&\mathrm{curl}\mathbf{E}_\zeta(M) = jk\mu_\zeta \mathbf{H}_\zeta(M), &&\\
&\mathrm{div}\mathbf{E}_\zeta(M) = 0, && \zeta = 0,1,2,\\
&\mathrm{div}\mathbf{H}_\zeta(M) = 0, && \qquad (2)\\
&[\mathbf{E}_\zeta(P)\times\mathbf{n}_P] = [\mathbf{E}_{\zeta+1}(P)\times\mathbf{n}_P], && P \in \partial D_{\zeta+1},\\
&[\mathbf{H}_\zeta(P)\times\mathbf{n}_P] = [\mathbf{H}_{\zeta+1}(P)\times\mathbf{n}_P]. &&
\end{aligned}$$

$$\begin{aligned}
&\varepsilon_1' \mathbf{E}_1(P)\cdot\mathbf{n}_P = \varepsilon_2 \mathbf{E}_2(P)\cdot\mathbf{n}_P, && P \in \partial D_2,\\
&\varepsilon_1' \mathbf{E}_1(P)\cdot\mathbf{n}_P = \varepsilon_0 \mathbf{E}_0(P)\cdot\mathbf{n}_P, && P \in \partial D_1.
\end{aligned}$$

Maxwell equation within Ag layer **is modified** and **additional boundary conditions** are imposed on both metal-dielectric surface boundaries [1,2]. Moreover, for metal-metal surfaces another form of BC has to be used [1,7,8]. Hence **each configuration** of the stratified sphere can have a **unique problem statement** making it difficult to derive an analytic solution in general form.

Cornerstones of the HDT/GNOR

- Instead of the $\mathbf{J}(M) = \sigma\mathbf{E}(M)$ another constitutive relation is used within metal layer (D_1) [2,6,7]:
$$\left(\frac{\beta^2}{\omega(\omega+j\gamma)} + \frac{d}{j\omega}\right)\nabla[\nabla\cdot\mathbf{J}(M)] + \mathbf{J}(M) = \sigma_{\mathrm{D}}(\omega)\boldsymbol{E}(M);$$
- This leads to the presence of the **longitudinal field** $\mathrm{curl}(\mathbf{E}_\zeta^L) = 0$ in the metal layer along with the transversal field $\mathrm{div}(\mathbf{E}_\zeta^T) = 0$, $k^L/k^T \sim 10 \div 100$!
- γ is Drude damping; $\beta^2 = 3/5v_F^2$, v_F – Fermi velocity; d – electron diffusion coeff. (GNOR only);
- $\varepsilon' = \varepsilon + \omega_p^2/(\omega^2 + j\gamma\omega)$, ω_p – metal plasma freq;
- "Hard-wall" boundary condition is imposed.

Notations

- We consider plane wave $\{\mathbf{E}_i, \mathbf{H}_i\}$ scattering;
- $\{\mathbf{E}_s, \mathbf{H}_s\}$ is the scattered field in D_2;
- $\mathbf{E}_2 = \mathbf{E}_i + \mathbf{E}_s$, $\mathbf{H}_2 = \mathbf{H}_i + \mathbf{H}_s$ is the total field in D_2;
- $\{\mathbf{E}_\zeta, \mathbf{H}_\zeta\}$ are total fields in D_ζ layer;
- Within nonlocal layer $\mathbf{E}_\zeta = \mathbf{E}_\zeta^T + \mathbf{E}_\zeta^L$;
- $\mathbf{n}_P$ is a unit outward normal to the surface at point P;
- $k = 2\pi/\lambda$ is a wavenumber in vacuum, $\omega = kc$;
- ε_ζ is experimentally measured complex permittivity;
- μ_ζ is permeability; $\mathrm{Im}(\varepsilon_2) = 0$, $\mathrm{Im}(\varepsilon_\zeta) \geq 0$.
- Radiation condition at infinity (D_2) is also imposed.

Solution: Mie theory with longitudinal harmonics

$$\mathbf{M}_{\substack{e\\o}mn} = \mathrm{curl}(\mathbf{r}\psi_{\substack{e\\o}mn}),\quad \mathbf{N}_{\substack{e\\o}mn} = \mathrm{curl}(\mathbf{M}_{\substack{e\\o}mn})/k_\zeta,\quad \mathbf{L}_{\substack{e\\o}mn} = \mathrm{grad}(\mathbf{r}\psi_{\substack{e\\o}mn}),$$

Complete set of the spherical vector wave functions (SVWFs)

$$\psi_{\substack{e\\o}mn}^{(i)} = z_n^{(i)}(k_\zeta r)P_n^m(\cos\theta)\begin{cases}\cos m\varphi\\ \sin m\varphi\end{cases}$$

$$z_n^{(i)}(kr) = \left[j_n(kr), y_n(kr), h_n^{(1)}(kr), h_n^{(2)}(kr)\right]$$

Incident field: $\mathbf{E}_i = E_0\sum_{n=1}^{\infty} j^n \frac{2n+1}{n(n+1)}\left(\mathbf{M}_{o1n}^{(1)} - j\mathbf{N}_{e1n}^{(1)}\right)$, $\mathbf{H}_i = \frac{-k_2}{\omega\mu_2}E_0\sum_{n=1}^{\infty} j^n \frac{2n+1}{n(n+1)}\left(\mathbf{M}_{e1n}^{(1)} + j\mathbf{N}_{o1n}^{(1)}\right)$, (3a)

Scattered field: $\mathbf{E}_s = \sum_{n=1}^{\infty} E_n\left(ja_n\mathbf{N}_{e1n}^{(3)} - b_n\mathbf{M}_{o1n}^{(3)}\right)$, $\mathbf{H}_s = \frac{k_2}{\omega\mu_2}\sum_{n=1}^{\infty} E_n\left(jb_n\mathbf{N}_{o1n}^{(3)} + a_n\mathbf{M}_{e1n}^{(3)}\right)$, (3b)

Local core: $\mathbf{E}_0 = \mathbf{E}_0^T = \sum_{n=1}^{\infty} E_n\left(c_n\mathbf{M}_{o1n}^{(1)} - jd_n\mathbf{N}_{e1n}^{(1)}\right)$, $\mathbf{H}_0 = \frac{-k_0}{\omega\mu_0}\sum_{n=1}^{\infty} E_n\left(d_n\mathbf{M}_{e1n}^{(1)} + jc_n\mathbf{N}_{o1n}^{(1)}\right)$, (3c)

Nonlocal core: $\mathbf{E}_0 = \mathbf{E}_0^T\big|_{k_0^T} + \mathbf{E}_0^L\big|_{k_0^L}$, $\mathbf{E}_0^L = -j\sum_{n=1}^{\infty} f_n\mathbf{L}_{e1n}^{(1)}$, $\mathbf{H}_0$ does not change $\left(\mathrm{curl}\mathbf{E}_\zeta^L = 0\right)$, (3d)

Local shell: $\mathbf{E}_1 = \mathbf{E}_1^T = \sum_{n=1}^{\infty}\left(\mathrm{A}_n\mathbf{M}_{o1n}^{(1)} - j\mathrm{B}_n\mathbf{N}_{e1n}^{(1)} + \Gamma_n\mathbf{M}_{o1n}^{(2)} - j\mathrm{K}_n\mathbf{N}_{e1n}^{(2)}\right)$, $\mathbf{H}_1 = \frac{-k_1}{\omega\mu_1}\mathrm{curl}(\mathbf{E}_1)$, (3e)

Nonlocal shell: $\mathbf{E}_1 = \mathbf{E}_1^T\big|_{k_1^T} + \mathbf{E}_1^L\big|_{k_1^L}$, $\mathbf{E}_1^L = -j\sum_{n=1}^{\infty}\left(h_n\mathbf{L}_{e1n}^{(1)} + q_n\mathbf{L}_{e1n}^{(2)}\right)$, $\mathbf{H}_1$ does not change. (3f)

Computer algebra implementation & results

Within *symMie*, SVWF set is implemented into symbolic MATLAB/Octave functions, along with $\{\mathbf{E}_\zeta, \mathbf{H}_\zeta\}$ expansions (3). Expansion coefficients $a_n, b_n, \ldots, h_n, q_n$ are then evaluated symbolically using boundary conditions defined by the user. Some examples of the BC input data are provided below along with the simulation results. It is assumed that $\mathbf{E}_\zeta = \mathbf{E}_\zeta(R,\theta,\varphi), \mathbf{H}_\zeta = \mathbf{H}_\zeta(R,\theta,\varphi)$, with spherical coordinates (R,θ,φ), origin at the sphere center, $a = r_1$ and $b = r_2$.

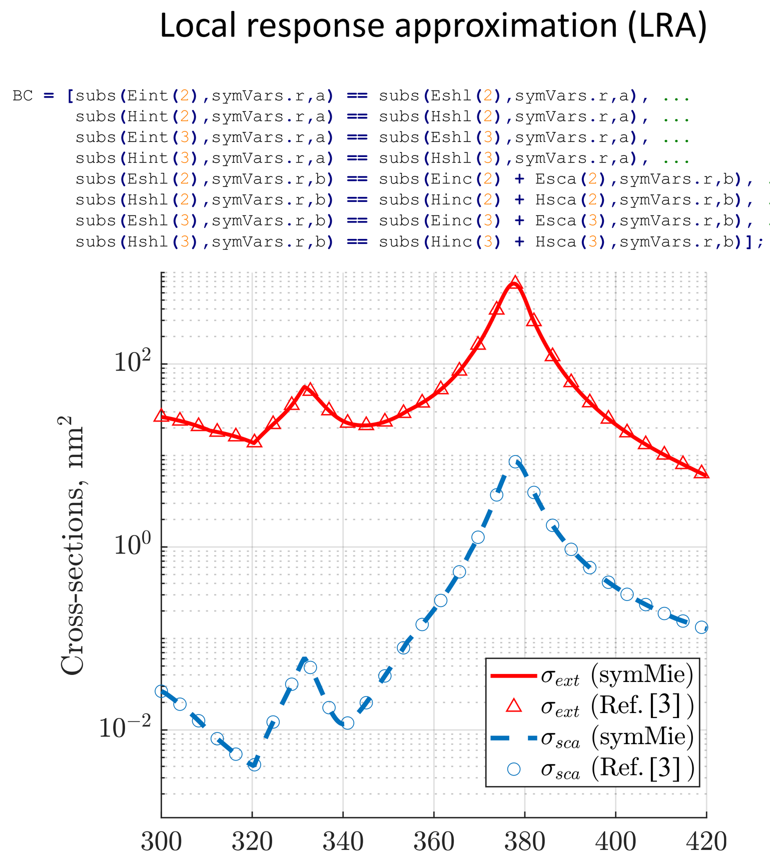

Fig. 1. Example of the boundary conditions input data for LRA problem (1) and corresponding solution ($\sigma_{ext}, \sigma_{sca}$).

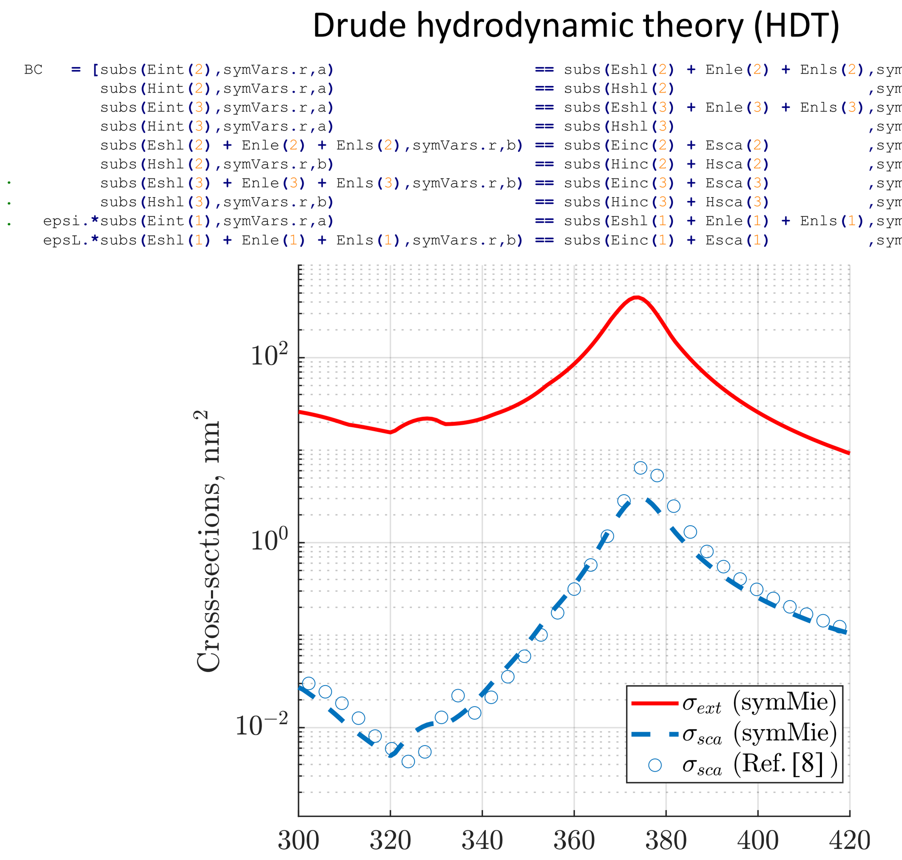

Fig. 2. Example of the boundary conditions input data for HDT problem (2) and corresponding solution ($\sigma_{ext}, \sigma_{sca}$).

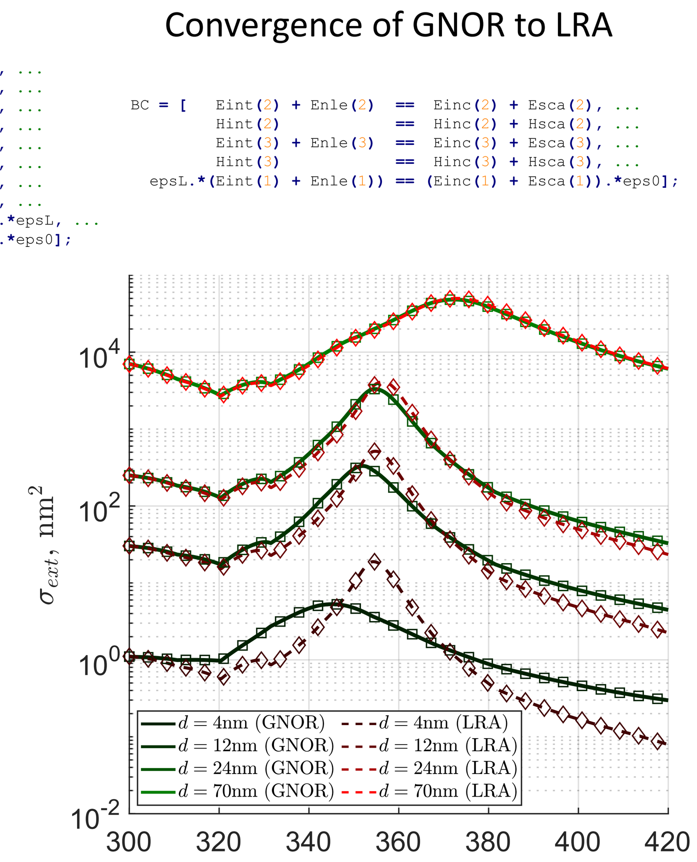

Fig. 3. Example of the boundary conditions input data for scattering by homogeneous silver sphere and demonstration of the GNOR solution convergence to the LRA solution for large particles. Symbols indicate reference data [3].

Comment: Evaluation of the expansion coefficients can be

(a) fully symbolic (result: all coefficients are expressed as functions of the input parameters, i.e. $a_n = a_n(r_1, r_2, \lambda, \varepsilon_0, \varepsilon_1, \varepsilon_1^T, \varepsilon_2, \ldots)$); **(b)** partially symbolic (some input parameters are defined numerically, i.e. $r_1 = 3nm$); **(c)** MATLAB VPA – variable precision arithmetic (both input and output data are numeric). This enables additional flexibility in i.e. rounding error analysis. This feature appears to be important due to the presence of rapidly oscillating longitudinal fields ($k^L \gg k^T$) leading to computational issues in thin plasmonic layers [3,8].

Longitudinal wavenumber: $(k^L)^2 = \frac{\omega^2 + j\gamma\omega - \omega_p^2/\varepsilon'}{\beta^2 + d(\gamma + j\omega)}$, transversal wavenumber: $(k^T)^2 = k^2\varepsilon\mu$, $\sigma_{sca} = \frac{2\pi}{k^2}\sum_{n=1}^{\infty}(2n+1)(|a_n|^2 + |b_n|^2)$, $\sigma_{ext} = \frac{2\pi}{k^2}\sum_{n-1}^{\infty}(2n+1)\mathrm{Re}(a_n + b_n)$.

Conclusion

- Implemented symbolic framework allows to accurately obtain **both analytic and numeric** solutions for the stratified sphere with nonlocal layers. Boundary conditions can be flexibly defined by user via built-in field expansions.
- **Applications** of the proposed *symMie* tool include, but are not limited to: (a) versatile comparison of the different physical approaches to the e/m scattering problem; (b) ground for verification of pure numerical high-performance techniques; (c) estimation of the solution to scattering problem without the need to implement any problem-specific code.
- This framework can potentially be expanded to the **broader scope of boundary-value problems** from any field of mathematical physics given that it allows analytic solution in terms of expansion to the complete basis function series.

References

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https://github.com/ilopushenko/symMie
(currently upon request)