Simulating optical properties of extremely oblate inhomogeneous particles with the discrete dipole approximation

IOP Conf. Series: Journal of Physics: Conf. Series **1461** (2020) 012198 doi:10.1088/1742-6596/1461/1/012198

Simulating optical properties of extremely oblate inhomogeneous particles with the discrete dipole approximation

D A Smunev[1], M A Yurkin[2,3,4]

[1] Belarusian State University, Nezavisimosti av. 4, 220030 Minsk, Belarus
[2] Voevodsky Institute of Chemical Kinetics and Combustion SB RAS, Institutskaya Str. 3, 630090, Novosibirsk, Russia
[3] Novosibirsk State University, Pirogova Str. 2, 630090, Novosibirsk, Russia
[4] Corresponding author: yurkin@gmail.com

Abstract. The discrete dipole approximation (DDA) is a general volume-discretization method for simulating optical properties of arbitrary particles. We employ the formulation of the DDA with rectangular-cuboid dipoles (voxels) and analyze it in the limit of very small thicknesses of both dipole and scatterer. Taking the limit of zero thickness greatly simplifies the theoretical formulation, resulting in the Rayleigh-Debye-Gans approximation combined with the boundary conditions at the plane interface. This can also be computed in the framework of the DDA, for which we developed a specialized modification of the open-source code ADDA. We also proved that the scattering quantities computed with the standard (3D) code are independent of number of dipoles along the particle thickness, when the latter is sufficiently small. This justifies the usage of dipole monolayers for practical simulations, including those for finite-width metasurfaces.

1. Introduction

Among the wide variety of nanostructures with interesting optical properties, a separate niche is occupied by structures with significantly different dimensions along coordinate axis. In this work we consider oblate particles, although very prolate (needle-like) particles are although interesting both from application and theoretical viewpoints. A limiting case of such oblate particles is metasurfaces [1,2], when one dimension (and the scale of internal inhomogeneity) is much smaller than the wavelength, while the other two dimensions – much larger. While infinitely wide periodic metasurfaces are most easy for theoretical considerations, real experiments often deal with finite systems. For the latter the finite thickness may or may not have significant effect on the optical properties.

In principle, such systems can be simulated using general electromagnetic solvers [3,4], however, most of them perform far from optimal for very oblate shapes. Although approximate methods based on Born (or Rayleigh-Debye-Gans – RDG) approximations can be accurate for such shapes, their applicability domain (in terms of system thickness) is hard to predict a priori. This calls for specific optimization of the general numerically exact solvers for such cases.

The discrete dipole approximation (DDA) is a simple yet robust method based on discretization of volume-integral equation for the electric field inside a scattering object [5]. Typically, it uses cubic dipoles (voxels) to be compatible with FFT acceleration of the matrix-vector product. The first step towards efficient application of the DDA to oblate nanoparticles was made by introduction of

rectangular cuboid dipoles, which happened to be non-trivial due to the strong singularity of the integration kernel [6]. Moreover, the corresponding code has been recently merged into the open-source code ADDA [7,8], providing well-tested implementation for anybody to use.

The goal of this extended abstract is to push this rectangular-dipole implementation to the limit of very thin particles (and, hence, dipoles), both through the theoretical analysis and by direct numerical simulations. Moreover, we present a separate DDA code explicitly based on the theoretical formulation with infinitely thin dipoles.

2. DDA equations in the limit of flat dipoles

The DDA equations for non-magnetic particle discretized with rectangular dipoles boil down to:

$$\mathbf{E}_i = \mathbf{E}_{\mathrm{inc},i} + k^2 V_\mathrm{d} \sum_{j\neq i} \overline{\mathbf{G}}_{ij} \cdot \bar{\boldsymbol{\chi}}_j \cdot \mathbf{E}_j + (\overline{\mathbf{M}}_i - \bar{\mathbf{L}}_i) \cdot \bar{\boldsymbol{\chi}}_i \cdot \mathbf{E}_i, \tag{1}$$

which is based on [6], but using SI system of units. Here $\mathbf{E}_i \stackrel{\text{def}}{=} \mathbf{E}(\mathbf{r}_i)$ and $\mathbf{E}_{\mathrm{inc},i} \stackrel{\text{def}}{=} \mathbf{E}_\mathrm{inc}(\mathbf{r}_i)$ are the unknown electric field and given incident field at the dipole center $\mathbf{r}_i$, $\bar{\boldsymbol{\chi}}_i \stackrel{\text{def}}{=} (\bar{\boldsymbol{\varepsilon}}(\mathbf{r}_i)/\varepsilon_0) - 1$ is the electric susceptibility, $\bar{\boldsymbol{\varepsilon}}(\mathbf{r})$ is the potentially anisotropic dielectric permittivity, ε_0 – that of vacuum, and k is the wave number. The interaction term $\overline{\mathbf{G}}_{ij}$ is given by the integral over the dipole volume V_j (equal to V_d for any j):

$$\overline{\mathbf{G}}_{ij} \stackrel{\text{def}}{=} \frac{1}{V_\mathrm{d}} \int_{V_j} \mathrm{d}^3\mathbf{r}\, \overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}_i), \tag{2}$$

where

$$\overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}') = \overline{\mathbf{G}}(\mathbf{R}) = \frac{\exp(\mathrm{i}kR)}{4\pi R}\left[\left(\bar{\mathbf{I}} - \frac{\mathbf{R}\otimes\mathbf{R}}{R^2}\right) + \frac{\mathrm{i}kR - 1}{k^2R^2}\left(\bar{\mathbf{I}} - 3\frac{\mathbf{R}\otimes\mathbf{R}}{R^2}\right)\right] \tag{3}$$

is the free-space Green's dyadic, $\bar{\mathbf{I}}$ is the unity dyadic, $\mathbf{R} \stackrel{\text{def}}{=} \mathbf{r} - \mathbf{r}'$, $R \stackrel{\text{def}}{=} |\mathbf{R}|$, and $\otimes$ is a dyadic (outer) product.

Dyadics $\overline{\mathbf{M}}_i$ and $\bar{\mathbf{L}}_i$ appear from the explicit exclusion of the strong singularity of $\overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}')$ and are actually independent of i. In particular,

$$\overline{\mathbf{M}}_i \stackrel{\text{def}}{=} k^2 \lim_{V_0\to 0} \int_{V_i\setminus V_0} \mathrm{d}^3\mathbf{r}\, [\overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}_i) - \overline{\mathbf{G}}_\mathrm{st}(\mathbf{r}, \mathbf{r}_i)], \tag{4}$$

where V_0 is the infinitesimal exclusion volume and $\overline{\mathbf{G}}_\mathrm{st}$ is the static limit of the Green's dyadic

$$\overline{\mathbf{G}}_\mathrm{st}(\mathbf{R}) \stackrel{\text{def}}{=} \lim_{R\to 0} \overline{\mathbf{G}}(\mathbf{R}) = -\frac{1}{4\pi k^2 R^3}\left(\bar{\mathbf{I}} - 3\frac{\mathbf{R}\otimes\mathbf{R}}{R^2}\right). \tag{5}$$

The self-term dyadic (the source term) depends on the shape but not on the size of V_0:

$$\bar{\mathbf{L}}_i \stackrel{\text{def}}{=} \oint_{\partial V_0} \mathrm{d}^2\mathbf{r} \frac{\mathbf{n}\otimes\mathbf{R}}{4\pi R^3}. \tag{6}$$

Taking the shape of V_0 the same as that of V_i, we can replace ∂V_0 by ∂V_i in Eq. (6), while $\overline{\mathbf{M}}_i$ provides only finite-size corrections. Explicit expressions for $\bar{\mathbf{L}}_i$ and $\overline{\mathbf{M}}_i$ for dipole with size $d_1 \times d_2 \times d_3$ and diagonal $D \stackrel{\text{def}}{=} \sqrt{d_1^2 + d_2^2 + d_3^2}$ are given in [6] (different by a factor of 4π due to different system of units). Here, we only mention that

$$\bar{\mathbf{L}}_i = \mathcal{O}(1), \qquad \overline{\mathbf{M}}_i = \mathcal{O}(k^2 V_\mathrm{d}/D). \tag{7}$$

The above formulation allows straightforward limit of $(h \stackrel{\text{def}}{=} d_3) \to 0$, assuming $d_{1,2}$ and the whole dipole configuration constant, i.e. the particle thickness scales with h. Consider first a monolayer of dipoles. Then $\overline{\mathbf{G}}_{ij}$ approaches a finite limit (integral over the 2D patch), but the term V_d nullifies the sum in Eq. (1), $\overline{\mathbf{M}}_i$ also vanish, while $\bar{\mathbf{L}}_i \to \bar{\mathbf{L}}_i^{(0)} \stackrel{\text{def}}{=} \mathbf{e}_z \otimes \mathbf{e}_z$, i.e. the dyadic with only non-zero element at position 3,3 equal to 1. Thus, Eq. (1) becomes trivial:

$$\mathbf{E}_i = \overline{\mathbf{T}}_i \cdot \mathbf{E}_i^\mathrm{inc}, \ \overline{\mathbf{T}}_i \stackrel{\text{def}}{=} (\bar{\mathbf{I}} + \bar{\mathbf{L}}_i^{(0)} \cdot \bar{\boldsymbol{\chi}}_i)^{-1}, \tag{8}$$

which is similar to the RDG approximation, but with simple modification of the fields. The latter corresponds to the boundary conditions at the interface and can be obtained directly by assuming the electric field constant inside the dipole. While this assumption is generally not valid for isolated (or

boundary) dipoles in the DDA [6], it is satisfied for a flat dipole since the field variation is concentrated at corners, which are negligible with respect to the total dipole volume. Any measurable quantity can be easily computed from the obtained $\mathbf{E}_i$, but most of them scale with h. For example, absorption cross section C_{abs} is proportional to h, while scattering intensity and scattering cross section are proportional to h^2. Extinction cross section C_{ext} is proportional to h if $C_{\mathrm{abs}} \neq 0$.

The accuracy of Eq. (8) can be questioned, since only a single dipole is used per particle thickness. Typically, the DDA converges to the true solution only if many dipoles are used per any particle dimension. However, it is trivial to extend the analysis to several layers of dipoles. The only additional component is $\overline{\mathbf{G}}_{ij}$, when dipoles i and j belong to the same vertical column. Skipping the derivation,

$$k^2 V_{\mathrm{d}} \overline{\mathbf{G}}_{ij} = \overline{\mathbf{M}}_i - \left(\bar{\mathbf{L}}_i - \bar{\mathbf{L}}_i^{(0)}\right) + \mathcal{O}(h^2), \tag{9}$$

i.e. it is proportional to h and in this first order independent of the vertical separation of dipoles. For very small h, inside-column $\overline{\mathbf{G}}_{ij}$ vanish and Eq. (8) remains the same, as well as any scattering quantity. In other words, if we fix very thin particle and use different numbers of dipoles per its thickness (n_z), we get exactly the same numerical result. Thus, the result for a monolayer is exactly the same as in the limit of $n_z \to \infty$. The same equivalence holds if $\mathcal{O}(h)$ terms, including first-order interaction between the dipoles, are retained in Eq. (1) – details will be provided at the conference. Moreover, the multi-layered structures can be modelled by a monolayer of dipoles with summed polarizability.

3. A code for flat structures and test simulations

Although the simulations based on Eq. (8) are much faster than the standard DDA even for multi-layered structures (no linear system need to be solved), it can still be conveniently done within the framework of the DDA. Thus, we have implemented this capability as the development branch of ADDA: https://github.com/adda-team/adda/tree/2d_dda, as a natural extension of the existing command line options. Users should specify 0 both in dipole thickness in the corresponding relative thickness of the shape, e.g. "-shape box 1 0 -rect_dip 1 1 0". Currently only three predefined ADDA shapes are supported: box, cylinder, and prism. ADDA generates standard output files, but the computed values are scaled by the corresponding powers of h. As a simple test for the new code, we present a comparison with the standard 3D simulation with rectangular dipoles in Fig. 1. The relative differences decrease linearly with h, as expected. Other test results will be presented at the conference.

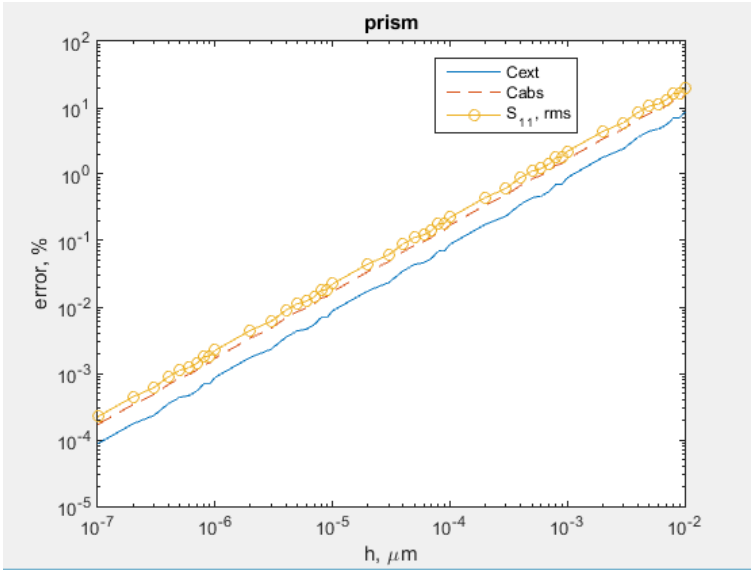

Figure 1. Relative differences of C_{abs}, C_{ext}, and first Mueller matrix element (root-mean-square over all angles) for pentagonal regular prism between standard DDA simulation and flat-limit code versus prism thickness h (in log-log scale). Both width of the prism and the wavelength are 1 μm.

4. Conclusion

The DDA with rectangular dipoles, as implemented in open-source ADDA code, is a robust and efficient method for simulating optical properties of thin inhomogeneous nanostructures, including metasurfaces of finite width. Taking the limit of infinitely thin particle and dipoles greatly simplifies the theoretical formulation, resulting in the RDG approximation combined with the boundary conditions at the plane interface. This can also be computed in the framework of the DDA using the developed specialized code. We also proved that the scattering quantities computed with the general 3D code are independent of n_z for sufficiently small particle thickness, justifying the usage of dipole monolayers for practical simulations.

Acknowledgments

This work is supported by the Russian Science Foundation (Grant No. 18-12-00052).

References

[1] Campione S, Guclu C, Ragan R and Capolino F 2014 Enhanced magnetic and electric fields via Fano resonances in metasurfaces of circular clusters of plasmonic nanoparticles *ACS Photonics* **1** 254–60

[2] Shcherbakov A A, Stebunov Y V, Baidin D F, Kämpfe T and Jourlin Y 2018 Direct S-matrix calculation for diffractive structures and metasurfaces *Phys. Rev. E* **97** 063301

[3] Correas-Serrano D, Gomez-Diaz J S, Tymchenko M and Alù A 2015 Nonlocal response of hyperbolic metasurfaces *Opt. Express* **23** 29434

[4] Wojtkiewicz S, Sawosz P, Kostecki M and Sokolowska A 2013 Optical method for characterization of nanoplates in lyosol *Microel. Engin.* **108** 121–6

[5] Yurkin M A and Hoekstra A G 2007 The discrete dipole approximation: an overview and recent developments *J. Quant. Spectrosc. Radiat. Transfer* **106** 558–89

[6] Smunev D A, Chaumet P C and Yurkin M A 2015 Rectangular dipoles in the discrete dipole approximation *J. Quant. Spectrosc. Radiat. Transfer* **156** 67–79

[7] Yurkin M A and Hoekstra A G 2011 The discrete-dipole-approximation code ADDA: capabilities and known limitations *J. Quant. Spectrosc. Radiat. Transfer* **112** 2234–47

[8] ADDA – light scattering simulator based on the discrete dipole approximation, https://github.com/adda-team/adda/