Discrete Dipole Approximation for Particles near Surface: a 3D-FFT-accelerated Implementation

Maxim A. YURKIN[1-2,*] and Marcus HUNTEMANN[3]

[1] Institute of Chemical Kinetics and Combustion SB RAS, Institutskaya 3, Novosibirsk, 630090, Russia
[2] Novosibirsk State University, Pirogova 2, Novosibirsk, 630090, Russia
[3] Institute of Environmental Physics, University of Bremen, Otto-Hahn-Allee 1, Bremen, 28359, Germany

*Corresponding author: yurkin@gmail.com

Abstract

We present a modification of the discrete dipole approximation (DDA) to simulate light scattering by particles of arbitrary shape and composition located above the plane homogeneous substrate. The modification retains the 3D-FFT acceleration scheme of the free-space DDA and hence its computational complexity. The modification is implemented in the recent version of the open-source ADDA code, available for anyone to use. The test simulations, compared with the T-matrix method, confirm the correctness of the implementation.

1 Introduction

The DDA is a general method to simulate light scattering by particles of arbitrary shape and composition [1]. It is mostly applied to finite particles in a homogeneous medium (e.g. vacuum). However, there are a multitude of applications, where a particle is located near a plane surface. The extension of the DDA to such problems is possible [2,3] but has two problems. First one is the calculation of interaction of two dipoles near the substrate, related to so-called Sommerfeld integrals [4]. Efficient evaluation of such integrals is still a field of active research, but we do not discuss it here. Instead we use the reliable albeit probably not the fastest routines [4], which were also used in previous DDA implementations.

Second problem is the lack of the translational symmetry of the dipole-dipole interaction (Green's tensor), which breaks standard 3D-FFT acceleration of the matrix–vector products used in the DDA [5]. As a result, existing DDA implementation either do not use FFT at all [3] or use only 2D-FFT due to the remaining translational symmetry parallel to the surface [2]. The computational complexity of the method is then O($N_{\text{iter}}N^2$) or O($N_{\text{iter}}N^{4/3}\log N$) respectively, where N is the number of dipoles used to discretize a particle and N_{iter} is the number of iterations required for the convergence of the iterative solver (typically, $N_{\text{iter}} \ll N$). A 3D-FFT acceleration for such cases, with complexity O($N_{\text{iter}}N\log N$), was presented for a similar volume-integral equation method [6], but has never been implemented in a DDA code.

In this extended abstract we present a way to retain 3D-FFT acceleration for the DDA applied to particles above the substrate, and describe the details of its implementation in the open-source ADDA code [7]. We also performed test simulations and compare the results with that of the T-matrix method.

2 DDA formulation

In principle, generalizing the DDA to particles on surface boils down to replacing the free-space Green's tensor $\overline{\mathbf{G}}$ by $\overline{\mathbf{G}}+\overline{\mathbf{R}}$, where $\overline{\mathbf{R}}$ is the "reflected" part. The main DDA equations [1] then become:

$$\overline{\boldsymbol{\alpha}}_i^{-1}\mathbf{P}_i - \sum_{j\neq i}\left(\overline{\mathbf{G}}_{ij}+\overline{\mathbf{R}}_{ij}\right)\mathbf{P}_j = \mathbf{E}_i^{\text{inc}}, \tag{1}$$

where $\overline{\boldsymbol{\alpha}}_i$ and $\mathbf{P}_i$ is the polarizability and total polarization of the dipole i, $\mathbf{E}_i^{\text{inc}}$ is the incident field at dipole position $\mathbf{r}_i$. To determine the unknown vector $\mathbf{P}$, system of linear equations (1) is solved by an iterative solver. Then the main computational bottleneck is the evaluation of matrix–vector product, the main part of which is the sum in Eq. (1).

The first part of the sum is the same as in the free-space DDA [1] and is evaluated using the translational symmetry

$$\overline{\mathbf{G}}_{ij} = \overline{\mathbf{G}}(\mathbf{r}_i,\mathbf{r}_j) = \overline{\mathbf{G}}(\mathbf{r}_i-\mathbf{r}_j) \equiv \overline{\mathbf{G}}'_{\mathbf{i-j}}, \tag{2}$$

where we assumed that dipoles are located on a uniform cubical grid, and $\mathbf{i}$, $\mathbf{j}$ are vector indices. $\overline{\mathbf{G}}'_{\mathbf{i}} = \overline{\mathbf{G}}_{\mathbf{i0}} = \overline{\mathbf{G}}(\mathbf{i}d)$ for $|i_\mu| \leq n_\mu$ ($\overline{\mathbf{G}}'_{\mathbf{0}} \equiv \overline{\mathbf{0}}$) and is assumed periodic with period $2n_\mu$ along axis μ (d is the dipole spacing, n_μ is the size of the dipole grid). The sum is transformed into a discrete convolution

$$\sum_{j=1,j\neq i}^{N}\overline{\mathbf{G}}_{ij}\mathbf{P}_j = \sum_{\mathbf{j}=(1,1,1)}^{(n_x,n_y,n_z)}\overline{\mathbf{G}}'_{\mathbf{i-j}}\mathbf{P}_{\mathbf{j}} = \sum_{\mathbf{j}=(1,1,1)}^{(2n_x,2n_y,2n_z)}\overline{\mathbf{G}}'_{\mathbf{i-j}}\mathbf{P}'_{\mathbf{j}}, \tag{3}$$

where $\mathbf{P}'$ is the periodic (same as $\overline{\mathbf{G}}'$) zero-padded extension of $\mathbf{P}$:

$$\mathbf{P}'_{\mathbf{j}} = \begin{cases}\mathbf{P}_{\mathbf{j}},\ \forall\mu:\ 1\leq j_\mu\leq n_\mu;\\ 0,\ \text{otherwise},\end{cases} \tag{4}$$

Finally, the convolution is evaluated as:

$$[\overline{\mathbf{G}}_{ij}]\mathbf{P} = F^{-1}\left(F(\overline{\mathbf{G}}')F(\mathbf{P}')\right), \tag{5}$$

where F and F^{-1} are the direct and inverse discrete Fourier transforms applied to each component of the vector or tensor independently.

We limit ourselves to particle being fully above the semi-infinite substrate, which surface is assumed to be the xy-plane. Then the reflected part is a function of the distance between evaluation point and the image of source:

$$\overline{\mathbf{R}}(\mathbf{r},\mathbf{r}') = \overline{\mathbf{R}}(x-x', y-y', z+z') = \overline{\mathbf{R}}(\boldsymbol{\rho}, Z), \tag{6}$$

where $\boldsymbol{\rho}$ is the distance along the surface. The simplest approximation is that of a single image dipole:

$$\overline{\mathbf{R}}_{\mathrm{im}}(\boldsymbol{\rho}, Z) = \frac{1-\varepsilon_{\mathrm{s}}}{1+\varepsilon_{\mathrm{s}}} \overline{\mathbf{G}}(\boldsymbol{\rho}, Z)(\overline{\mathbf{I}} - 2\overline{\mathbf{I}}_z), \tag{7}$$

where $\overline{\mathbf{I}}_z = \hat{e}_z\hat{e}_z$ is a projector on the z-axis and ε_{s} is the complex electric permittivity of the substrate. The rigorous expression for the reflection term is

$$\overline{\mathbf{R}}(\boldsymbol{\rho}, Z) = \frac{\hat{\rho}\hat{\rho}}{\rho^2}\left(I_\rho^{\mathrm{H}} + I_\varphi^{\mathrm{H}}\right) - \left(1 - \overline{\mathbf{I}}_z\right)I_\varphi^{\mathrm{H}} + \frac{\hat{\rho}\hat{e}_z - \hat{e}_z\hat{\rho}}{\rho} I_\rho^{\mathrm{V}} \\ + \overline{\mathbf{I}}_z I_z^{\mathrm{V}} + \overline{\mathbf{R}}_{\mathrm{im}}(\boldsymbol{\rho}, Z), \tag{8}$$

where I_ρ^{H}, I_φ^{H}, I_ρ^{V}, and I_z^{V} are the Sommerfeld integrals [4] that depend on ρ, Z, and ε_{s}.

Similar to Eq. (2) we define the auxiliary vector

$$\overline{\mathbf{R}}'_{\mathbf{i}} = \overline{\mathbf{R}}_{\mathbf{i}\{0,0,0\}} = \overline{\mathbf{R}}(i_x d, i_y d, i_z d + 2h_1) \tag{9}$$

for $|i_{x,y}| \leq n_{x,y}$, $0 \leq i_z < 2n_z-1$, $\overline{\mathbf{R}}'_{\mathbf{i}} \equiv 0$ for $i_z = 2n_z-1$, and is further extended periodically (same as $\overline{\mathbf{G}}'$). h_1 is a distance from the lowest dipole layer (their centres) to the surface. The sum over z-axis is now a discrete correlation, but it can be transformed to a convolution by inverting the order of z-components of $\mathbf{P}$:

$$\sum_{j=1}^{N} \overline{\mathbf{R}}_{ij}\mathbf{P}_j = \sum_{\mathbf{j}=(1,1,1)}^{(2n_x,2n_y,2n_z)} \overline{\mathbf{R}}'_{\mathbf{i}-\mathbf{j}}\widetilde{\mathbf{P}}_{\mathbf{j}}, \tag{10}$$

where

$$\widetilde{\mathbf{P}}_{\mathbf{j}} = \begin{cases} \mathbf{P}_{\{j_x,j_y,1\}}, 1 \leq j_{x,y} \leq n_{x,y} \;\&\; j_z = 1; \\ \mathbf{P}_{\{j_x,j_y,2n+2-j_z\}}, 1 \leq j_{x,y} \leq n_{x,y} \;\&\; n_z < j_z \leq 2n_z; \\ 0, \text{otherwise}. \end{cases} \tag{11}$$

Conveniently, $\widetilde{\mathbf{P}}$ satisfies

$$F(\widetilde{\mathbf{P}}) = F_z^{-1}F_yF_x(\mathbf{P}'), \tag{12}$$

where F_μ is the 1D discrete Fourier transform along the axis μ. Combining Eqs. (5), (10), and (12) we finally obtain

$$[\overline{\mathbf{G}}_{ij} + \overline{\mathbf{R}}_{ij}]\mathbf{P} = F^{-1}\left(\left(F(\overline{\mathbf{G}}')F_z + F(\overline{\mathbf{R}}')F_z^{-1}\right)F_xF_y(\mathbf{P}')\right). \tag{13}$$

Since $F(\overline{\mathbf{G}}')$ and $F(\overline{\mathbf{R}}')$ need to be calculated only once, the computational time for matrix–vector product is only slightly (by F_z^{-1} and a few O(N) operations) larger than that for the free-space DDA. In particular, it has the same complexity order O(Nlog$N$).

Certain changes in formulae to calculate the scattered fields (based on the determined $\mathbf{P}$) are also required, but they are straightforward and were published previously [2,3,8]. Less clear is the definition of the scattering matrices, e.g. the Mueller one [9], when either incident or scattered field is in the substrate. To the best of our knowledge, this issue is not covered in the literature. We propose to generalize the standard definition of the Mueller matrix as

$$\begin{pmatrix} I_{\mathrm{sca}} \\ Q_{\mathrm{sca}} \\ U_{\mathrm{sca}} \\ V_{\mathrm{sca}} \end{pmatrix} = \frac{1}{k_{\mathrm{sca}}^2 r^2} \begin{pmatrix} S_{11} & S_{12} & S_{13} & S_{14} \\ S_{21} & S_{22} & S_{23} & S_{24} \\ S_{31} & S_{32} & S_{33} & S_{34} \\ S_{41} & S_{42} & S_{43} & S_{44} \end{pmatrix} \begin{pmatrix} I_{\mathrm{in}} \\ Q_{\mathrm{in}} \\ U_{\mathrm{in}} \\ V_{\mathrm{in}} \end{pmatrix}, \tag{14}$$

where k_{sca} is the is the wave vector for the scattering direction, and we consider scattering into substrate only for non-absorbing one, i.e. k_{sca} is always real. The definitions of the Stokes vector (both incident and scattered) through the electric fields can be found in textbooks [9,10]. Important is that this definition contains the refractive index of the medium m as factor Re(m) [10].

3 Software implementation

The formulae described in Section 2 were implemented in the open-source code ADDA. In particular, these new features are included in the recently released version 1.3b4, available online [11]. Moreover, they are fully integrated with other parts of ADDA, including employed parallelization technologies. In particular, MPI parallelization allows one to solve huge problems (up to 1 billion dipoles [7]) using a large computer cluster. OpenCL mode allows one to significantly accelerate the code for moderately-sized problems using a modern video card (GPU) [12]. And a special (non-FFT) mode is available for very porous particles [13]. Details of the implementation are described in the manual [14].

4 Test simulations

We consider a single test case – an Ag sphere (radius R = 50 nm, refractive index 0.25 + 3.14i [15]) placed on glass substrate (m_{s} = 1.5), illuminated by plane wave from below propagating at 60° relative to the surface normal (evanescent illumination). Wavelength is 488 nm, and ADDA v.1.3b4 was used with two levels of discretization (64 and 128 dipoles per sphere diameter).

Figure 1 shows perpendicular and parallel scattering intensities ($S_{11} - S_{12}$ and $S_{11} + S_{12}$ respectively) in the main scattering plane (0° correspond to upward vertical direction) in comparison with the reference T-matrix results. The latter correspond to Fig. 4.10 of [15], but were recalculated by Vladimir Schmidt using NFM-DS 1.1 [16]. Moreover, the raw results of the NFM-DS, differential scattering efficiencies for unity amplitude of the incident electric field, were multiplied by a factor $\pi(kR)^2/m_{\mathrm{s}}$ = 0.86800, where k is the free-space wave vector.

The DDA accuracy is good – relative errors of angle-resolved quantities are within a few percent. Moreover, the errors for n_x = 128 are almost exactly twice smaller than that for n_x = 64, which indicates a smooth convergence. The latter can also be used to increase the accuracy using the extrapolation technique [17]. We had to use relatively fine discretization to obtain such accuracy, but that is not surprising for metallic nanoparticles [18]. The main practical problem, however, is that it is hard to predict the accuracy of the DDA for such problems *a priori*. No detailed accuracy studies exist for particles on surface, while transferability of free-space benchmarks is at least questionable.

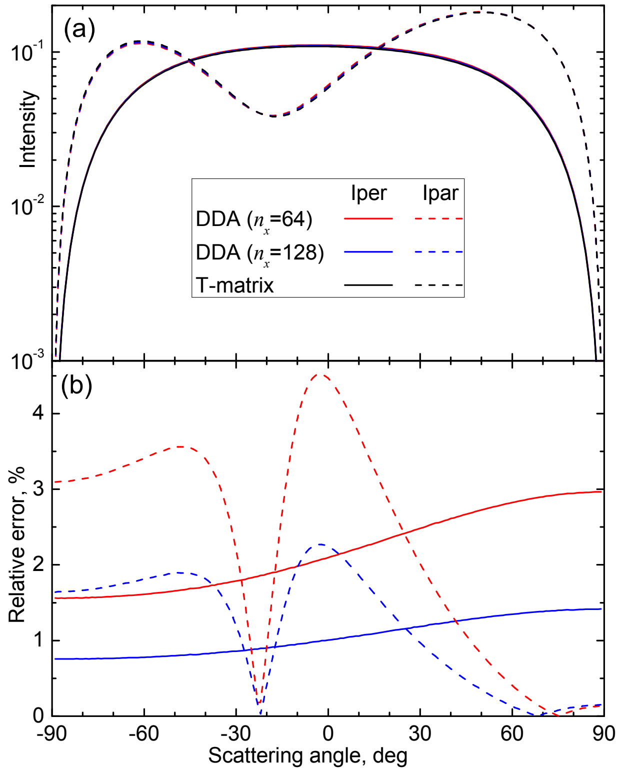

Figure 1 *(a) Light-scattering intensity for Ag sphere on a glass substrate under evanescent illumination (see text for details); (b) relative errors of DDA results in comparison with the T-matrix method.*

5 Conclusions

We presented a reliable, fast, open-source, and easy-to-use tool to simulate interaction of electromagnetic fields with particles of arbitrary shape and composition located on or near a semi-infinite plane substrate. In particular, only the particle itself needs to be discretized and the simulation time is not principally larger than that when no substrate is present. We believe this tool may find many applications in different fields, ranging, for example, from nanostructures on substrate to surface roughness on large dust particles.

6 Acknowledgement

We thank Vladimir Schmidt for providing reference T-matrix results.

7 References

[1] M.A. Yurkin and A.G. Hoekstra, "The discrete dipole approximation: an overview and recent developments," *J. Quant. Spectrosc. Radiat. Transfer* **106**, 558–589 (2007).

[2] R. Schmehl, B.M. Nebeker, and E.D. Hirleman, "Discrete-dipole approximation for scattering by features on surfaces by means of a two-dimensional fast Fourier transform technique," *J. Opt. Soc. Am. A* **14**, 3026–3036 (1997).

[3] V.L.Y. Loke, P.M. Mengüç, and T.A. Nieminen, "Discrete dipole approximation with surface interaction: Computational toolbox for MATLAB," *J. Quant. Spectrosc. Radiat. Transfer* **112**, 1711–1725 (2011).

[4] D.L. Lager and R.J. Lytle, *Fortran Subroutines for the Numerical Evaluation of Sommerfeld Integrals Unter Anderem*, Tech. Rep. UCRL-51821, Lawrence Livermore Lab., Livermore, USA (1975).

[5] J.J. Goodman, B.T. Draine, and P.J. Flatau, "Application of fast-Fourier-transform techniques to the discrete-dipole approximation," *Opt. Lett.* **16**, 1198–1200 (1991).

[6] Y.A. Eremin and V.I. Ivakhnenko, "Modeling of light scattering by non-spherical inhomogeneous particles," *J. Quant. Spectrosc. Radiat. Transfer* **60**, 475–482 (1998).

[7] M.A. Yurkin and A.G. Hoekstra, "The discrete-dipole-approximation code ADDA: capabilities and known limitations," *J. Quant. Spectrosc. Radiat. Transfer* **112**, 2234–2247 (2011).

[8] E. Bae, H. Zhang, and E.D. Hirleman, "Application of the discrete dipole approximation for dipoles embedded in film," *J. Opt. Soc. Am. A* **25**, 1728–1736 (2008).

[9] C.F. Bohren and D.R. Huffman, *Absorption and Scattering of Light by Small Particles*, Wiley, New York (1983).

[10] M.I. Mishchenko, L.D. Travis, and A.A. Lacis, *Scattering, Absorption, and Emission of Light by Small Particles*, Cambridge University Press, Cambridge (2002).

[11] ADDA – light scattering simulator using the discrete dipole approximation, http://code.google.com/p/a-dda/

[12] M. Huntemann, G. Heygster, and G. Hong, "Discrete dipole approximation simulations on GPUs using OpenCL–Application on cloud ice particles," *J. Comput. Sci.* **2**, 262–271 (2011).

[13] J. Leinonen, D. Moisseev, and T. Nousiainen, "Linking snowflake microstructure to multi-frequency radar observations," *J. Geophys. Res.: Atmos.* **118**, 3259–3270 (2013).

[14] M.A. Yurkin and A.G. Hoekstra, "User manual for the discrete dipole approximation code ADDA 1.3b4," http://a-dda.googlecode.com/svn/tags/rel_1.3b4/doc/manual.pdf (2014).

[15] A. Doicu, R. Schuh, and T. Wriedt, "Scattering by particles on or near a plane surface," in *Light Scattering Reviews 3*, A.A. Kokhanovsky, Ed., pp. 109–130, Springer, Berlin (2008).

[16] NFM-DS, http://www.scattport.org/index.php/light-scattering-software/t-matrix-codes/list/239-nfm-ds

[17] M.A. Yurkin, V.P. Maltsev, and A.G. Hoekstra, "Convergence of the discrete dipole approximation. II. An extrapolation technique to increase the accuracy," *J. Opt. Soc. Am. A* **23**, 2592–2601 (2006).

[18] M.A. Yurkin, D. de Kanter, and A.G. Hoekstra, "Accuracy of the discrete dipole approximation for simulation of optical properties of gold nanoparticles," *J. Nanophoton.* **4**, 041585 (2010).