OPTIMET-3D: User Manual

A software package for simulating electromagnetic wave interaction with a cluster of dielectric, metallic, and semiconducting particles embedded in a homogeneous background

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Version 2.0

5 January 2022

OPTIMET-3D user manual

OPTIMET-3D (OPTIcal METamaterials - 3Dimensional) is an *ab initio* solver for 3D electromagnetic wave scattering problem. **OPTIMET-3D** is distributed under the GPL. It is freely available on GitHub (https://GitHub.com/OPTIMET/OPTIMET) for all interested parties. It includes a description of the installation steps, as well as this usage manual.

OPTIMET-3D implements the multiple-scattering matrix (MSM) method, used to accurately describe the electromagnetic wave interaction in linear (fundamental frequency (FF)) and non-linear (second-harmonic frequency (SH)) regimes with a cluster of an arbitrary number of dielectric, metallic, or semiconducting particles (spherical or non-spherical) embedded in a homogeneous background medium. The features of **OPTIMET-3D** are instrumental for efficient modelling of light interaction with ensembles of nanoparticles. In particular, **OPTIMET-3D** can be used to investigate scientific problems arising in several different disciplines, including Engineering, Physics, Chemistry, Materials Science, and Biology.

OPTIMET-3D is written in `C++` programming language and uses the OOP framework. **OPTIMET-3D** provides the source code and an intuitive scripting system as the data input, in XML format, for running the executable. The source code and installation process are available via the GitHub public repository OPTIMET-3D. **OPTIMET-3D** employs a set of functions for efficient handling of algebraic operations and numerical computations that are related to the MSM formalism. In addition, **OPTIMET-3D** incorporates a set of routines and scientific libraries for the serial or parallel execution of the resulting system of linear equations, which can be solved directly or iteratively. For the parallel execution, it accepts several linear-systems solvers found in, for example, Scalapack for the direct solution and Belos for an iterative solution.

To use **OPTIMET-3D**, simply copy the executable `Optimet3D` to a folder of your choice, set your shell to the folder where `Optimet3D` is stored and call it using a single argument, the path to the input file. For example, `./Optimet3D input-filename.xml`.

In order to become familiar with and learn how to use **OPTIMET-3D**, refer to the detailed information provided in what follows, about the `XML` input system and simulation parameters.

1 Introduction

Computational techniques based on the multiple-scattering matrix (MSM) method have become a primary means for modelling a variety of scientific and engineering problems pertaining to electromagnetic wave interaction with systems of arbitrarily distributed particles [1-4]. In particular, computational methods based on the MSM algorithm, also called the *transfer matrix* (or *T-matrix*) method, play a central role in modelling photonic materials, metamaterials, nano-photonic devices, wireless communications devices, radar signature technology, antennae, microwave devices, bio-photonics and biomedical imaging and sensing. The versatility of the MSM method allows one to study electromagnetic wave interaction with ensembles of scatterers under a broad set of boundary conditions, material parameters and geometries.

OPTIMET-3D implements the MSM method and consequently it can be used to describe the interaction of electromagnetic waves with arbitrary distributions of dielectric, metallic and semiconducting particles.

The main part of the MSM algorithm consists in constructing and solving a system of linear equations whose unknowns are the Fourier expansion coefficients of the scattered field. The matrix defining this system is completely defined by the location, shape and material parameters of the cluster of particles. The *T*-matrix of the system has a block structure, the corresponding blocks consisting of single-particle *T*-matrices and matrices that describe bi-particle interactions (electromagnetic coupling between the two particles). As an immediate consequence of this block structure of the *T*-matrix, the MSM method can be easily parallelized on supercomputers and applied to model clusters with arbitrary number and distribution of particles.

The main advantages of **OPTIMET-3D** are:

1) The electromagnetic field is computed using a relatively small set of parameters, due to the spherical harmonic basis, which leads to a significant reduction in the computational time.
2) It inherently satisfies the far-field radiation boundary condition and therefore it is not needed to truncate the computational domain (as for example in finite element techniques).
3) It can be used to model a broad class of material properties (particles made of dielectric, metallic or semiconducting material).
4) It can be used for the computation of electromagnetic fields in linear and non-linear (second-harmonic) regimes in serial and parallel, using MPI.

2 The OPTIMET-3D XML input file manual

OPTIMET-3D uses an eXtended Markup Language (`XML`) input file to setup the numerical simulation. The `.xml` file contains the set of parameters required for a simulation and its (meta)name will be appended to the output data files, which are written in `HDF5` file format (for the case of field profile plots) or in generic data files (in the case of cross section computations). When launching a simulation, the `.xml` file name needs to be passed as an input parameter to the `Optimet3D` executable.

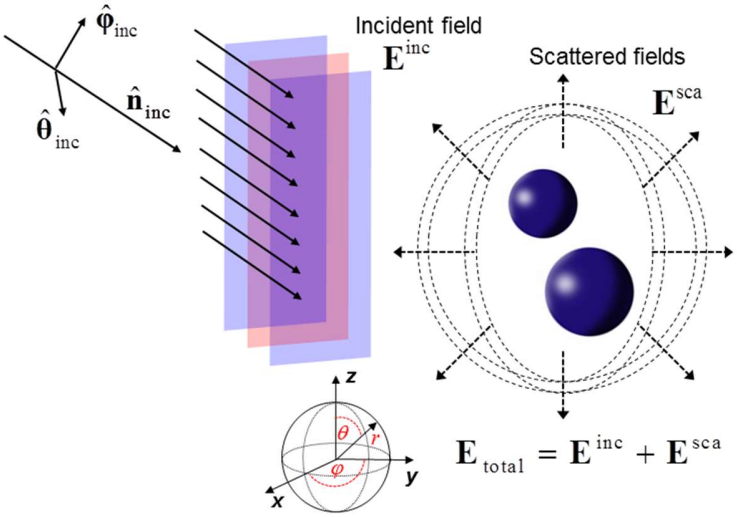

Figure 1. *Schematic representation of the incident and scattered fields at FF for a two-particle scattering system.*

The `.xml` file contains several blocks describing different aspects of the simulation problem. The order in which these blocks are defined in the `.xml` file is irrelevant. However, it is the user's responsibility to correctly define all the main blocks of a simulation.

OPTIMET-3D implements the MSM method used for the linear and second-harmonic scattering analysis, which we briefly outline in what follows. Consider a cluster of N particles being illuminated by a monochromatic plane electromagnetic wave. The origin of the global co-ordinate system of the cluster is O and to each particle (j) one associates a local co-ordinate system with the origin, O_j. The only constraint imposed on the location of the particles is that the circumscribing spheres of the particles do not touch, *i.e.* $|\boldsymbol{R}_{jl}| = |O_j - O_l| > \rho_j + \rho_l$, $j, l = 1,2,\ldots,N, j \neq l$, where ρ_j is the radius of the smallest sphere containing the jth particle.

Linear regime: The solution of the source-free Maxwell equations in 3D can be expanded in a complete basis of vector spherical wave functions (VSWFs), $\left(\boldsymbol{M}_{mn}^{(1)}, \boldsymbol{N}_{mn}^{(1)}\right)$ and $\left(\boldsymbol{M}_{mn}^{(3)}, \boldsymbol{N}_{mn}^{(3)}\right)$. In a practical implementation, the series is truncated to a certain order, n_{max}. Let $\sum_{mn} \stackrel{\text{def}}{=} \sum_{n=1}^{n_{max}} \sum_{m=-n}^{n}$; then, the incident, scattered and internal electric fields oscillating at the fundamental frequency ω, with unity amplitude can be expressed as:

incident field at FF

$$\boldsymbol{E}_0^{\omega}(\boldsymbol{R}) = \sum_{mn} \left[a_{mn}^{\omega} \boldsymbol{M}_{mn}^{(1)}(k^{\omega}\boldsymbol{R}) + b_{mn}^{\omega} \boldsymbol{N}_{mn}^{(1)}(k^{\omega}\boldsymbol{R}) \right],$$

internal field at FF

$$\boldsymbol{E}_{int}^{\omega}(\boldsymbol{R}_j) = \sum_{mn}\left[c_{mn}^{j,\omega}\mathbf{Rg}\boldsymbol{M}_{mn}^{(1)}(k_j^{\omega}\boldsymbol{R}_j) + d_{mn}^{j,\omega}\mathbf{Rg}\boldsymbol{N}_{mn}^{(1)}(k_j^{\omega}\boldsymbol{R}_j)\right],\quad R_j < \rho_j,$$

scattered field at FF

$$\boldsymbol{E}_{sca}^{\omega}(\boldsymbol{R}_j) = \sum_{mn}\left[p_{mn}^{j,\omega}\boldsymbol{M}_{mn}^{(3)}(k^{\omega}\boldsymbol{R}_j) + q_{mn}^{j,\omega}\boldsymbol{N}_{mn}^{(3)}(k^{\omega}\boldsymbol{R}_j)\right],\quad R_j > \rho_j,$$

where k^{ω} and k_j^{ω} are the wave vectors in the embedding medium and inside the *j*th particle, respectively, $\boldsymbol{R}$ and $\boldsymbol{R}_j, j = 1,2,\ldots,N$, are the position vectors of a point, *P*, in the co-ordinate systems O and O_j, respectively. It can be shown that, within the MSM method, computing the fundamental-field scattering coefficients $p_{mn}^{j,\omega}$ and $q_{mn}^{j,\omega}$, and implicitly the linear electromagnetic field and system cross-sections, amounts to solving the linear system $\boldsymbol{S}^{\omega}\boldsymbol{v}^{\omega} = \boldsymbol{u}^{\omega}$, where $\boldsymbol{v}^{\omega}$ is a vector determined by the scattering field coefficients, $\boldsymbol{u}^{\omega}$ is a known vector determined by the incoming field coefficients, and $\boldsymbol{S}^{\omega}$ denotes the scattering matrix at fundamental frequency of the system [1-4].

Second-harmonic regime: Once the linear fields are computed, **OPTIMET-3D** offers an option to extend the EM field analysis to the second-harmonic frequency Ω = 2ω. The efficient characterization of this second-order nonlinear optical effect relies on the computed fundamental electric fields internal to each particle, $\boldsymbol{E}_{int}^{\omega}(\boldsymbol{R}_j)$, from which the second-harmonic sources are constructed [5, 6]. Similarly to the linear fields computed before, the SH internal and scattered fields are decomposed into sums of VSWFs as follows:

internal field at SH

$$\boldsymbol{E}_{int}^{\Omega}(\boldsymbol{R}_j) = \sum_{mn}\left[c_{mn}^{j,\Omega}\mathbf{Rg}\boldsymbol{M}_{mn}^{(1)}(k_j^{\Omega}\boldsymbol{R}_j) + d_{mn}^{j,\Omega}\mathbf{Rg}\boldsymbol{N}_{mn}^{(1)}(k_j^{\Omega}\boldsymbol{R}_j)\right],\quad R_j < \rho_j,$$

scattered field at SH

$$\boldsymbol{E}_{sca}^{\Omega}(\boldsymbol{R}_j) = \sum_{mn}\left[p_{mn}^{j,\Omega}\boldsymbol{M}_{mn}^{(3)}(k^{\Omega}\boldsymbol{R}_j) + q_{mn}^{j,\Omega}\boldsymbol{N}_{mn}^{(3)}(k^{\Omega}\boldsymbol{R}_j)\right],\quad R_j > \rho_j,$$

and the SH excitation fields are appropriately represented through the spherical harmonic expansion of nonlinear interface conditions [7]. The linear system $\boldsymbol{S}^{\Omega}\boldsymbol{v}^{\Omega} = \boldsymbol{u}^{\Omega}$ at the SH frequency is established and solved for the unknown SH scattering field coefficients $p_{mn}^{j,\Omega}$ and $q_{mn}^{j,\Omega}$ stacked in the vector $\boldsymbol{v}^{\Omega}$ from which the field profiles and cross-sections can be calculated directly.

Constructing the system matrices $\boldsymbol{S}^{\omega}$ and $\boldsymbol{S}^{\Omega}$, and solving corresponding linear systems represent the core parts of the MSM method.

In **OPTIMET-3D**, all length values are to be given in nanometers (10^{-9} m), with the scale factor (10^{-9}) suppressed throughout. For example, the radius parameter of a sphere with radius of 500×10^{-9} m is defined as `radius="500.0"`. The angles that define the direction of an incident plane-wave are given in degrees. As a general rule, unless otherwise specified, all physical quantities are specified in SI units. In the following sub-sections, the complete procedure of defining the building blocks of an `.xml` file is described.

2.1 Simulation

2.1.1 The simulation block

The `simulation block` describes the overall properties of performing a simulation. The main property required for this block is the maximum cut-off value for field expansions, `nmax`, which must be a positive integer, `nmax` > 0, and is the same for linear and second-harmonic simulations. With the second entry in

this block we have an option to introduce the adaptive cross approximation (ACA) compression to our computations [8, 9]. This option is set to `no` by default, but we can change this by introducing `<ACA compression="yes"/>` in the `simulation block`. This directive will enable the compression algorithm in our serial and parallel computations. It is important to emphasize that the effects of the compression will start to be noticeable only for bigger particle clusters (>200 particles).

In order to accurately solve a specific scattering problem, the choice of the value of `nmax` will mostly depend on the operating wavelength, the electrical size of the particles, their electromagnetic properties and spatial distribution. Usually, electrically bigger particles require more harmonics for the fields to converge. Likewise, metallic particles need higher number of harmonics in the field expansion when compared to dielectric ones due to the higher inhomogeneity of the fields around them.

The syntax for a `simulation block` that defines a simulation with `nmax="10"` harmonics is:

```
{.xml}
<simulation>
<harmonics nmax="10"/>
<ACA compression="no"/>
</simulation>
```

2.1.2 Linear solver parameters block

Here we describe how different linear system solvers are used: `eigen_direct`, `scalapack_direct`, `Belos _GMRES and GMRES_ACA`.

Serial computations: If the code is built with the *MPI* option set to *OFF,* direct and serial linear system solution which makes use of the `eigen` solver is set by the default. If we want to introduce ACA compression to our serial calculations, then, the serial `GMRES_ACA` solver able to handle compressed matrices, specifically tuned for this purpose, is invoked.

Parallel computations: If the code is built with the *MPI* option set to *ON,* the parallel and direct solution that makes use of `scalapack` computations is enabled by the default. If ACA is invoked, parallel `GMRES_ACA` is used to solve the matrix system. The third option is to use the `GMRES` solver from Belos scientific library on an uncompressed system (ACA set to *no*). Then, in the `ParameterList block` we define the parameters of the `Belos_GMRES` solver as:

```
<ParameterList name="Belos">
     <Parameter name="Solver" type="string" value="GMRES"/>
     <Parameter name="Convergence Tolerance" type="double" value="1.0e-5"/>
     <Parameter name="Verbosity" type="int" value="8"/>
      <Parameter name="Maximum Iterations" type="int" value="500"/>
     <Parameter name="Num Blocks" type="int" value="30"/>
     <Parameter name="Block Size" type="int" value="1"/>
     <Parameter name="Maximum Restarts" type="int" value="20"/>
 </ParameterList>
```

For parallel executions, the number of processes is set in the usual `MPI` way, when calling the `Optimet3D` executable. The syntax may vary with the computing environment but will be something like `mpirun -n N /path/to/Optimet3D`, where `N` is the number of processes and `/path/to/Optimet3D/` should be the full path to the `Optimet3D` executable.

2.2 The source block

The `source block` defines the specifics of the incoming plane-wave oscillating at fundamental frequency ω that acts upon the scattering system as well as the existence of polarization second-harmonic sources.

The propagation of the pump incident plane-wave is specified in spherical coordinates $(\theta_{\text{inc}}, \varphi_{\text{inc}})$, which are defined with respect to a system with the origin located at (0, 0, 0), as shown in Fig. 1. Then, the incident electric field is defined as $\mathbf{E}^{\text{inc}} = E_{\theta}^{\text{inc}}\widehat{\boldsymbol{\theta}}_{\text{inc}} + E_{\varphi}^{\text{inc}}\widehat{\boldsymbol{\varphi}}_{\text{inc}}$, where $\widehat{\boldsymbol{\theta}}_{\text{inc}}$ and $\widehat{\boldsymbol{\varphi}}_{\text{inc}}$ are the unit vectors in the θ- and φ-directions, such that $\widehat{\mathbf{n}}_{\text{inc}} = \widehat{\boldsymbol{\theta}}_{\text{inc}} \times \widehat{\boldsymbol{\varphi}}_{\text{inc}}$ ($\widehat{\mathbf{n}}_{\text{inc}}$ is a unit vector along the direction of the wave vector of the incident wave) and E_{θ}^{inc} and E_{φ}^{inc} are the corresponding field components along $\widehat{\boldsymbol{\theta}}_{\text{inc}}$ and $\widehat{\boldsymbol{\varphi}}_{\text{inc}}$, respectively. In the definition of the `source block`, the direction of the incident wave vector is defined by the variables (`theta`, `phi`), specified in degrees, and the incident plane-wave is defined by the (complex) field projections along $\widehat{\boldsymbol{\theta}}_{\text{inc}}$ and $\widehat{\boldsymbol{\varphi}}_{\text{inc}}$, (`Etheta.real`, `Etheta.imag`) and (`Ephi.real`, `Ephi.imag`), respectively.

Furthermore, we are presented with an option to introduce the second-harmonic sources to our calculations. If we set `<SHsources condition="yes" />`, the nonlinear second-order effects will be taken into account and second-harmonic generation will be analyzed in addition to the linear scattering. Otherwise, if we set `<SHsources condition="no" />`, only linear computations at fundamental frequency will be performed.

The syntax for a `source block` that defines a linearly polarized incident plane-wave with wavelength of 1460 nm, direction of incidence (`theta`, `phi`) = (45, 90) and polarization (`Etheta.real`, `Etheta.imag`)=(1, 0) and (`Ephi.real`, `Ephi.imag`)=(0, 0), along with SH polarization sources, is:

```
{.xml}
<source type="planewave">
       <wavelength value="1460" />
       <propagation theta="45" phi="90" />
       <polarization Etheta.real="1.0" Etheta.imag="0.0"
                     Ephi.real="0.0"   Ephi.imag="0.0"/>
      <SHsources condition="yes" />
 </source>
```

We specify that in **OPTIMET-3D** the angles that define the direction of an incident wave are given in degrees. The field components `Etheta` and `Ephi` of the incoming plane-wave can have complex values. This allows one to consider incoming plane-waves that are circularly polarized.

2.3 The geometry block

The `geometry block` defines the configuration of the simulation, including electromagnetic properties of particles and the background medium at FF and SH frequencies, the type of particles (spherical or non-spherical), as well as the distribution and size of the particles. By default, the background medium is assumed to be vacuum.

The syntax used for setting the electromagnetic properties of the background to, for example, a medium with relative electric permittivity, $\varepsilon_{\text{b}} = 3$, and relative permeability, $\mu_{\text{b}} = 1$, is:

```
{.xml}
<geometry>
 <background>
       <epsilon type="relative" value.real="3.0" value.imag="0.0" />
       <mu type="relative" value.real="1.0" value.imag="0.0"/>
 </background>
</geometry>
```

To include a particle in the `geometry block`, the `object sub-block` (see ***Sec. 2.3.1** and **2.3.2***) is used to define a spherical or arbitrarily shaped, non-spherical scatterer. In these sub-blocks we can define the radius of the sphere circumscribing the particle, location and the electromagnetic properties of the target. The only constraint imposed on the location of a particle and, consequently, the distribution of the particles, is that their circumscribing spheres do not touch. For this purpose, during the simulation process and prior to solving the scattering problem, **OPTIMET-3D** first checks that this condition is satisfied. Otherwise, an error message is generated, and the simulation stops.

***Electromagnetic properties*:** in order to define the electromagnetic properties, the `geometry block` supports three classes of materials both at fundamental and second-harmonic frequencies:

a) The user defines his own frequency-independent relative permittivity and permeability values at FF and SH frequencies ($\varepsilon_r^{FF}, \mu_r^{FF}, \varepsilon_r^{SH}$), along with surface and bulk SH tensor parameters ($\chi_{\perp\perp\perp}, \chi_{\parallel\perp\parallel}, \gamma$).

b) The frequency-dependent relative permittivity for gold (Au) at FF and SH is defined by interpolating the experimental data obtained by Johnson and Christy [10] in the incident wavelengths <400 nm, 1200 nm>. The nonlinear tensor susceptibility components valid for gold are frequency dependent as well, with Rudnick-Stern parameters *a*, *b* and *d* defined by user [11].

c) We define the dispersive properties of silicon (Si) at FF and SH by interpolating the experimental data from Schinke [12] in the incident wavelength range <500 nm, 1400 nm>. The SH tensor components are frequency independent with values equal to: ($\chi_{\perp\perp\perp}$=65·10^{-19}m^{2}V^{-1}, $\chi_{\parallel\perp\parallel}$=3.5·10^{-19}m^{2}V^{-1}, γ=1.3·10^{-19}m^{2}V^{-1}).

***Location*:** in order to define the location of the particles, the `geometry block` supports three configurations:

i) `sphere object sub-block`, an object-by-object distribution of spherical particles.

ii) `arbitrary.shape object sub-block`, an object-by-object distribution of non-spherical particles.

iii) `cube or surface structure sub-block`, a predefined assembly of particles in cubic or square lattice. The particles can be spherical or arbitrarily shaped.

2.3.1 Sphere object sub-block

The `sphere object sub-block` is used to define the location of a spherical particle, as well as its radius and electromagnetic properties. The center of the sphere is defined in Cartesian coordinates. In this case, the particle *T*-matrix, both at the fundamental and SH frequency is diagonal, with entries which are computed analytically and equal to the *Mie* series coefficients.

The syntax used to define a sphere with radius of 500 nm and frequency-independent electromagnetic properties set via relative permittivities $\varepsilon_r^{FF} = 2 + 2i$ and $\varepsilon_r^{SH} = -3.2 + 5i$, relative permeability $\mu_r^{FF} = \mu_r^{SH} = 1$, and tensor parameters $\chi_{\perp\perp\perp}$(`ksippp`) = 12e-19, $\chi_{\parallel\perp\parallel}$(`ksiparppar`) = 8.5e-19 and γ(`gamma`) = 3.3e-19, whose center is located at $(100,\ 50,\ -200)$ nm, is:

```
 {.xml}
<geometry>
 <object type="sphere">
        <cartesian x="100.0" y="50.0" z="-200.0" />
        <properties radius="500.0" />
        <epsilon type="relative" value.real="2.0" value.imag="2.0" />
       <epsilon_SH type="relative" value.real="-3.2" value.imag="5.0" />
       <ksippp value.real="12e-19" value.imag="0.0" />
       <ksiparppar value.real="8.5e-19" value.imag="0.0" />
       <gamma value.real="3.3e-19" value.imag="1.0" />
```

```
        <mu type="relative" value.real="1.0" value.imag="0.0"/>
 </object>
</geometry>
```

Note that the trailing .0 in double values is optional. However, if an integer value contains a .0, **OPTIMET-3D** will trigger a warning message before rounding it to an integer.

The syntax that defines a gold (Au) sphere whose material properties are defined by the interpolation of Johnson and Christy's experimental data is:

```
 {.xml}
<geometry>
 <object type="sphere">
        <cartesian x="100.0" y="50.0" z="-200.0" />
        <properties radius="500.0" />
        <epsilon type="GoldModel">
        <parameters
         a.real="1.0" a.imag="0.0"
         b.real="-1.0" b.imag="0.0"
         d.real="1.0" d.imag="0.0" />
         </epsilon>
        <mu type="relative" value.real="1.0" value.imag="0.0"/>
 </object>
</geometry>
```

The Rudnick-Stern parameters *a*, *b* and *d* here take the value according to the hydrodynamic model, but can be set to more accurate (complex) values if needed.

The syntax that defines a silicon (Si) sphere whose material properties are determined by interpolating Schinke's experimental data is:

```
 {.xml}
<geometry>
 <object type="sphere">
        <cartesian x="100.0" y="50.0" z="-200.0" />
        <properties radius="500.0" />
        <epsilon type="SiliconModel"/>
        <mu type="relative" value.real="1.0" value.imag="0.0"/>
 </object>
</geometry>
```

Multiple `sphere object sub-blocks` can be added to the `geometry block` thus defining an array of arbitrarily distributed spheres of different sizes and material.

2.3.2 Arbitrary.shape object sub-block

The `arbitrary.shape object sub-block` is used to define the location of a non-spherical particle, the type of the particle, as well as the radius of the circumscribed sphere and its electromagnetic properties. The center of the circumscribed sphere is defined in Cartesian coordinates. **OPTIMET-3D** comes with a library of surface meshes stored in the folder ../examples/meshlib. We provide the surface meshes of different spheroids because they are especially well suited for the MSM analysis due to their axial symmetry. These meshes are used for the numerical computations of the individual scatterer *T*-matrix as well as the SH excitation vectors. In this case, the *T*-matrix of a particle is usually full.

The sub-block which defines a prolate silicon spheroid with equatorial radius $r = 50$ nm and a polar radius $R = 90$ nm centered at $(0,\ 0,\ 0)$ is:

```
{.xml}
<geometry>
 <object type="arbitrary.shape" dims="r50R90">
        <cartesian x="0.0" y="0.0" z="0.0" />
        <properties radius="90.0" />
        <epsilon type="SiliconModel"/>
        <mu type="relative" value.real="1.0" value.imag="0.0"/>
 </object>
</geometry>
```

Notice that the radius of the circumscribed sphere is 90 nm.

2.3.3 Cube/square structure sub-block

To illustrate the capabilities of **OPTIMET-3D**, we present a cubic and square (surface) particle assemblies defined by the `cube/square structure sub-block`. This structure consists of a certain number of particles arranged in a cubic or square lattice whose centers are positioned in the first octant (cubic lattice) or first quadrant (surface lattice). The user defines the specifics of a single particle (spherical or non-spherical) as well as the center-to-center distance between the neighboring particles. The distance is the same for all adjacent particles thus forming a finite periodic lattice which can be seen as a part of the metamaterial/metasurface. It is the user's responsibility to make sure that this separation distance is larger than the diameter of the circumscribed spheres, meaning that no two particles overlap.

The syntax for a `cubic structure` of spheres containing 125 gold spherical particles in the first octant (each sphere has `radius` = 100 nm) with a center-to-center separation distance of 300 nm is:

```
 {.xml}
<geometry>
 <structure type="cube">
    <object type="sphere">
    <properties radius="100.0" />
    <epsilon type="GoldModel">
    <parameters
    a.real="1.0" a.imag="0.0"
    b.real="-1.0" b.imag="0.0"
    d.real="1.0" d.imag="0.0" />
    </epsilon>
    <mu type="relative" value.real="1.0" value.imag="0.0" />
    </object>
    <properties points="5" distance="300.0" />
   </structure>
</geometry>
```

The pivotal sphere, from which the structure building begins, is located at $(0,\ 0,\ 0)$. The number of points in each direction is 5, equaling to 125 spherical scatterers. If we change the `structure type` from `"cube"` to `"surface"`, we will get a square lattice of 25 gold spheres in the first quadrant. It should also be noted that instead of the `sphere object` we can use `arbitrary.shape object` and thus form a cubic/square lattice of spheroids.

2.4 The output block

The `output block` is used to specify the way in which the output data produced by a simulation is processed and written to the output data files. **OPTIMET-3D** supports two types of `output block` components, namely `field profile output` and `spectral response output`.

2.4.1 Field profile output

The `field profile output` creates an output profile for the electric and magnetic fields for all three Cartesian components, this profile being defined on a structured 3D Cartesian grid. It provides the field cross-section plots both at FF and SH frequencies, if the SH sources are enabled in the `source block.` The grid range along each axis is specified through a minimum (`min`) and a maximum (`max`) value as well as the corresponding number of output points (`steps`) along each direction. The output data from a `field profile output` block is written in a `HDF5` file format (`.h5`), whose name is the same as that of the `.xml` file with the extension defining the FF or the SH plots (for example `TwoParticles_SH.h5`).

We give below the syntax of a `field profile output` with the following range:

On the *x*-axis, (`min`, `max`, `steps`) = (-1000.0, 1000.0, 101) nm;

On the *y*-axis, (`min`, `max`, `steps`) = (-0.1, 0.1, 2) nm;

On the *z*-axis, (`min`, `max`, `steps`) = (-1000.0, 1000.0, 101) nm.

```
{.xml}
<output type="field">
 <grid type="cartesian">
       <x min="-1000.0" max="1000.0" steps="101" />
       <y min="-0.1" max="0.1" steps="2" />
       <z min="-1000.0" max="1000.0" steps="101" />
 </grid>
</output>
```

Note that the grid of points is created in the *xz*-plane on which the FF and SH fields are computed. At least 2 output steps must be specified for any given axis.

2.4.2 Spectral response output

The `spectral response output` is used when a wavelength scan is required. An identical structure setup (particles locations, radii and electromagnetic properties) is simulated for different values of the incident (pump) wavelength (frequency sweep). The `spectral response output` is used to determine the absorption and scattering cross-sections of the system at fundamental and second-harmonic frequencies. The solution obtained will be written in two separate data (`.dat`) files for FF and SH computations, called `AbsorptionCS.dat` and `ScatteringCS.dat` for the absorption and scattering cross-sections, respectively.

The syntax used to define a `spectral response output` intended for a wavelength scan ranging from 400 nm to 1200 nm with a step size (`stepsize`) of 20 nm is:

```
{.xml}
<output type="response">
       <scan type="A+E">
              <wavelength initial="400" final="1200" stepsize="20" />
       </scan>
</output>
```

If a wavelength value is also defined in the `source block`, it will be ignored.

3 Illustrative examples

In this section, we present several examples, which illustrate how the building blocks of single-particle and multi-particle system simulations are constructed and properly assembled in the `.xml` file.

3.1 One particle scattering system

In this example, we analyse the linear and second-harmonic scattering from a single gold nanosphere with radius $r = 200$ nm and a silicon nanosphere with radius $r = 300$ nm, the calculated linear and nonlinear scattering and absorption cross sections being presented in the ***Figure 2***. In this example, spherical particles are illuminated by a plane-wave propagating along a direction characterized by angles $\theta_{inc} = {}^{\pi}/_{4}$ and $\varphi_{inc} = {}^{\pi}/_{2}$. The background medium is assumed to be vacuum. The incident electric field considered here is polarized along $\hat{\boldsymbol{\theta}}_{inc}$ direction. The number of Fourier harmonics used for solving the scattering matrix problem is set to `nmax`=13 per sphere and per frequency. The incident wavelength range is set to <400 nm, 1200 nm> for a gold sphere and <1000 nm, 1450 nm> for a silicon sphere. The syntax for `OneParticle.xml` file that describes this problem setup, is given as (the case of Si sphere):

```
<simulation>
   <harmonics nmax="13" />
<ACA compression="no" />
</simulation>

<source type="planewave">
  <wavelength value="1000" />
  <propagation theta="45" phi="90" />
  <polarization Etheta.real="1.0" Etheta.imag="0.0" Ephi.real="0.0" Ephi.imag="0.0"/>
  <SHsources condition="yes" />
</source>

<geometry>
<object type="sphere">
        <cartesian x="0.0" y="0.0" z="0.0" />
        <properties radius="300.0" />
        <epsilon type="SiliconModel"/>
        <mu type="relative" value.real="1.0" value.imag="0.0"/>
</object>
</geometry>

<output type="response">
  <scan type="A+E">
    <wavelength initial="1000" final="1450" stepsize="10" />
  </scan>
</output>
```

This value is neglected in the case of cross-section computations

Upon completion of the simulation, the cross sections are written in `OneParticle_ScatteringCS_FF.dat` and `OneParticle_AbsorptionCS_FF.dat`, for FF calculations, and `OneParticle_ScatteringCS_SH.dat` and `OneParticle_AbsorptionCS_SH.dat` for SH computations. The plots are given in the ***Figure 2***. Please note that the absorption cross-section for Si sphere at FF is not plotted since the losses are negligible in this frequency range.

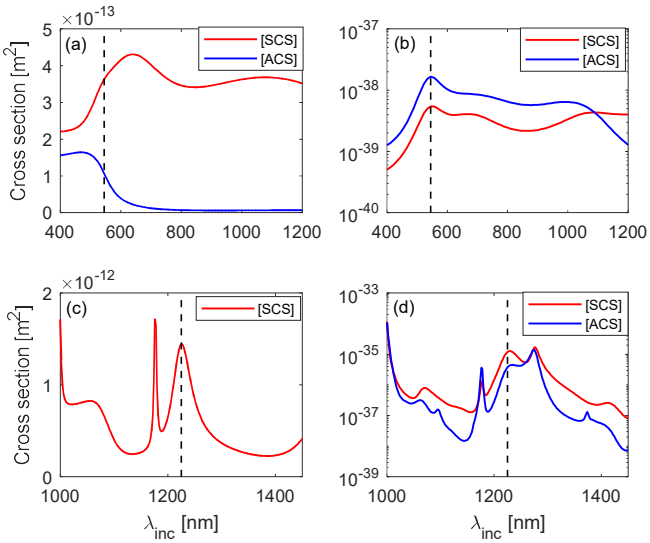

Figure 2. *Scattering and absorption cross-sections calculated at the FF (left panels) and SH (right panels). The top and bottom panels correspond to a gold nanosphere with radius r = 200 nm and a silicon nanosphere with radius r = 300 nm, respectively. The vertical lines indicate the input wavelengths at which the spatial profiles of the near-fields are calculated.*

The field profile plots at FF and SH frequencies can be obtained by running the following `OneParticle.xml` file (the case of Au sphere):

```
<simulation>
   <harmonics nmax="13" />
<ACA compression="no" />
</simulation>

<source type="planewave">
  <wavelength value="545" />
  <propagation theta="45" phi="90" />
  <polarization Etheta.real="1.0" Etheta.imag="0.0" Ephi.real="0.0" Ephi.imag="0.0"/>
  <SHsources condition="yes" />
</source>

<geometry>
<object type="sphere">
        <cartesian x="0.0" y="0.0" z="0.0" />
        <properties radius="200.0" />
        <epsilon type="GoldModel">
        <parameters
        a.real="1.0" a.imag="0.0"
        b.real="-1.0" b.imag="0.0"
        d.real="1.0" d.imag="0.0" />
        </epsilon>
        <mu type="relative" value.real="1.0" value.imag="0.0"/>
</object>
</geometry>

<output type="field">
<grid type="cartesian">
    <x min ="-400" max="400" steps="161" />
```

This is the wavelength for which the field plots are obtained

```
    <y min ="-0.1" max="0.1" steps="2" />
    <z min ="-400" max="400" steps="161" />
</grid>
</output>
```

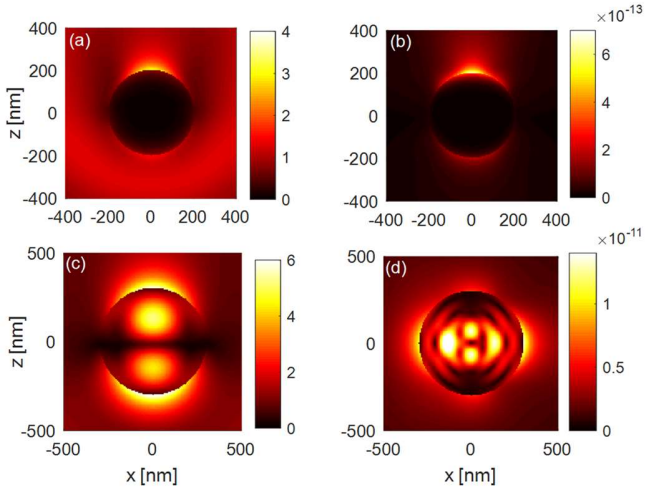

Figure 3. *Spatial distribution of the electric field amplitude at the FF (left panels) and SH (right panels), calculated for a single gold nanosphere (top panels) at the incident wavelength λ_{inc} = 545 nm and for a single silicon nanosphere (bottom panels) at the incident wavelength λ_{inc} = 1225 nm. The fields are determined in the xz-plane.*

The field profiles are stored in the corresponding `.h5` files. In order to view and analyze this data, one can use `HDFView`, a freely available software. Alternatively, once the output data from `.h5` file is displayed in a spreadsheet format and exported to, for example, `.txt` files, one can also use one's preferred visualization tool for plotting the profile of the field.

3.2 Multiple particles scattering systems

In this subsection we present several examples of multi-particle assemblies embedded in vacuum, where the full potential of the MSM method and, consequently, **OPTIMET-3D** is exploited. In all the examples, the excitation field is a $\widehat{\boldsymbol{\theta}}_{inc}$ polarized plane wave irradiating the particle systems from the direction defined by the incident angles $\theta_{\text{inc}} = {}^{\pi}/_{4}$ and $\varphi_{\text{inc}} = {}^{\pi}/_{2}$.

3.2.1 Three nanospheres

In this example, we calculate the scattering cross-sections of three nanospheres made of silicon and gold with centers located at O_1(-100, -100, 100) nm, O_2(-150, 150, 100) nm and O_3(200, 200, 100) nm, and radii equal to $r_1 = 50$ nm, $r_2 = 100$ nm and $r_3 = 150$ nm. The number of Fourier harmonics used in the simulation is set to `nmax`=8. The syntax for a `ThreeParticles.xml` file that describes this problem setup would read as (case of silicon):

```
<simulation>
 <harmonics nmax="8" />
 <ACA compression="no" />
</simulation>

<source type="planewave">
```

```
  <wavelength value="1000" />
  <propagation theta="45" phi="90" />
  <polarization Etheta.real="1.0" Etheta.imag="0.0" Ephi.real="0.0" Ephi.imag="0.0"/>
  <SHsources condition="yes" />
</source>

<geometry>
<object type="sphere">
        <cartesian x="-100.0" y="-100.0" z="100.0" />
        <properties radius="50.0" />
        <epsilon type="SiliconModel"/>
        <mu type="relative" value.real="1.0" value.imag="0.0"/>
</object>

<object type="sphere">
        <cartesian x="-150.0" y="150.0" z="100.0" />
        <properties radius="100.0" />
        <epsilon type="SiliconModel"/>
        <mu type="relative" value.real="1.0" value.imag="0.0"/>
</object>

<object type="sphere">
        <cartesian x="200.0" y="200.0" z="100.0" />
        <properties radius="150.0" />
        <epsilon type="SiliconModel"/>
        <mu type="relative" value.real="1.0" value.imag="0.0"/>
</object>
</geometry>

<output type="response">
  <scan type="A+E">
    <wavelength initial="500" final="1000" stepsize="10" />
  </scan>
</output>
```

Upon completion of the simulation defined in `ThreeParticles.xml`, the cross-section data will be written in corresponding data (`.dat`) files. The scattering cross-sections are plotted in the ***Figure 4***.

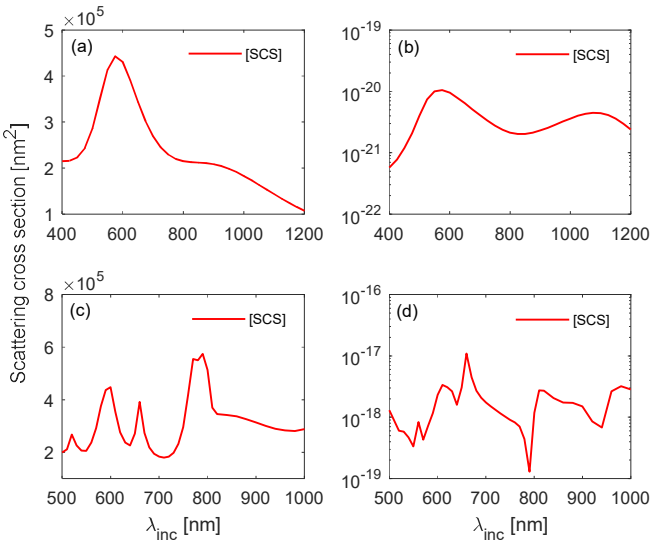

Figure 4. *Scattering cross sections at FF (left panels) and SH (right panels) for three nanospheres made of gold (upper panels) and silicon (bottom panels).*

3.2.2 Two gold spheroids

This numerical experiment presents the linear (FF) and nonlinear (SH) scattering analysis of two identical gold prolate spheroids with equatorial and polar radii equal to $r = 50$ nm and $R = 70$ nm centered at $(0,\ 0,\ 0)$ and $(0,\ 0,\ 200)$ nm. This configuration is interesting as it may find its application in the design of plasmonic nanoantennas or sensing devices since strong near-fields can be induced in the gap between the obstacles, allowing the formation of so-called *hot spots*. The syntax for a `TwoParticles.xml` file that describes the calculation of the cross-section spectra in the incident range <400 nm, 1200 nm$>$ looks as:

```
<simulation>
 <harmonics nmax="5" />
 <ACA compression="no" />
</simulation>

<source type="planewave">
  <wavelength value="1000" />
  <propagation theta="45" phi="90" />
  <polarization Etheta.real="1.0" Etheta.imag="0.0" Ephi.real="0.0" Ephi.imag="0.0"/>
  <SHsources condition="yes" />
</source>

<geometry>
<object type="arbitrary.shape" dims="r50R70">
        <cartesian x="0.0" y="0.0" z="0.0" />
        <properties radius="70.0" />
        <epsilon type="GoldModel">
        <parameters
        a.real="1.0" a.imag="0.0"
        b.real="-1.0" b.imag="0.0"
        d.real="1.0" d.imag="0.0" />
        </epsilon>
        <mu type="relative" value.real="1.0" value.imag="0.0"/>
</object>

<object type="arbitrary.shape" dims="r50R70">
        <cartesian x="0.0" y="0.0" z="200.0" />
        <properties radius="70.0" />
        <epsilon type="GoldModel">
        <parameters
        a.real="1.0" a.imag="0.0"
        b.real="-1.0" b.imag="0.0"
        d.real="1.0" d.imag="0.0" />
        </epsilon>
        <mu type="relative" value.real="1.0" value.imag="0.0"/>
</object>
</geometry>

<output type="response">
  <scan type="A+E">
    <wavelength initial="400" final="1200" stepsize="20" />
  </scan>
</output>
```

The scattering cross-sections are presented in ***Figure 5***. The dashed lines in ***Figure 5*** present the results obtained by the finite element method simulation which are used for comparison.

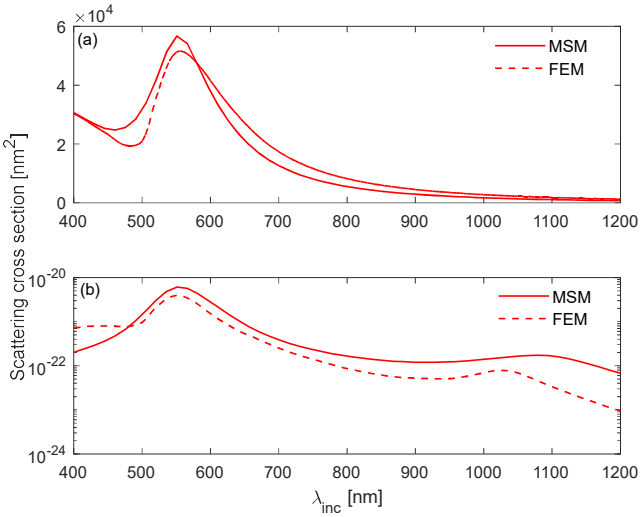

Figure 5. *Scattering cross sections at FF (upper panel) and SH (bottom panel) for two prolate spheroids made of gold.*

3.3 Cubic cluster of nanospheres

In the last example, we treat a cubic cluster of silicon nanospheres embedded in a vacuum and located in first octant of the Cartesian coordinate system, aligned with the x -, y - and z-axes. The cubic array contains $7\text{x}7\text{x}7 = 343$ spheres with same radii $r_i = 50$ nm, i $= 1, \ldots, 343$ which are equally spaced with the centre-to-centre distance $d = 190$ nm. Each sphere is anlysed with 160 harmonics in FF and SH regimes (`nmax` = 8) which tantamount to the dimensions of scattering matrices equal to $dim(\boldsymbol{S}^{\omega}) = dim(\boldsymbol{S}^{\Omega}) = 54880$. This is the computational problem of moderate size with complex system matrices. In computer memory, each matrix element occupies 16 bytes (real and imaginary parts of type double) which brings us to the total memory allocation of 48.19 GB per scattering matrix. This is a substantial memory consumption which we aim to reduce with adaptive cross approximation algorithm. The `.xml` file which describes this scattering scenario, with the ACA algorithm included in the computations, is:

```
<simulation>
 <harmonics nmax="8" />
 <ACA compression="yes" />
</simulation>

<source type="planewave">
  <wavelength value="800" />
  <propagation theta="45" phi="90" />
  <polarization Etheta.real="1.0" Etheta.imag="0.0" Ephi.real="0.0" Ephi.imag="0.0"/>
  <SHsources condition="yes" />
</source>

<geometry>
 <structure type="cube">
    <object type="sphere">
    <properties radius="50.0" />
    <epsilon type="SiliconModel"/>
    <mu type="relative" value.real="1.0" value.imag="0.0" />
    </object>
```

```
    <properties points="7" distance="190.0" />
   </structure>
</geometry>

<output type="response">
  <scan type="A+E">
    <wavelength initial="500" final="1000" stepsize="20" />
  </scan>
</output>
```

In ***Figure 6*** we plot scattering and absorption cross sections at FF (upper panels) and SH (lower panels) computed with MSM and ACA compression (dashed lines with asterisk) and compare them to the results obtained without ACA compression (full lines). Very good agreement is observed both at FF and SH frequencies. The scattering matrices compressed with ACA occupy only around 16% of the memory used by uncompressed matrices, thus making this problem amenable for ordinary PCs.

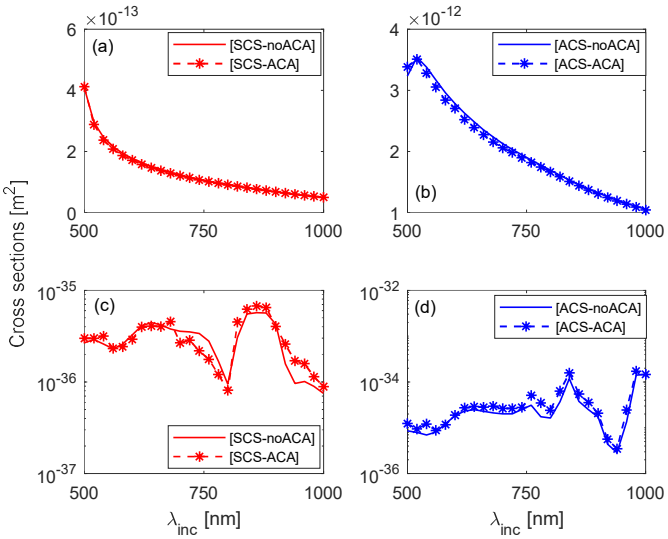

Figure 6. *Scattering and absorption cross sections at FF (upper panels) and SH (bottom panels) for a cubic lattice of nanospheres.*

In order to compute the field profiles, we replace `spectral response output` block with the `field profile output` block in our `.xml` file. In this example, the `field profile output` block is:

```
<output type="field">
<grid type="cartesian">
    <x min ="-70" max="1210" steps="171" />
    <y min ="-70" max="1210" steps="171" />
    <z min ="569.9" max="570.1" steps="2" />
</grid>
</output>
```

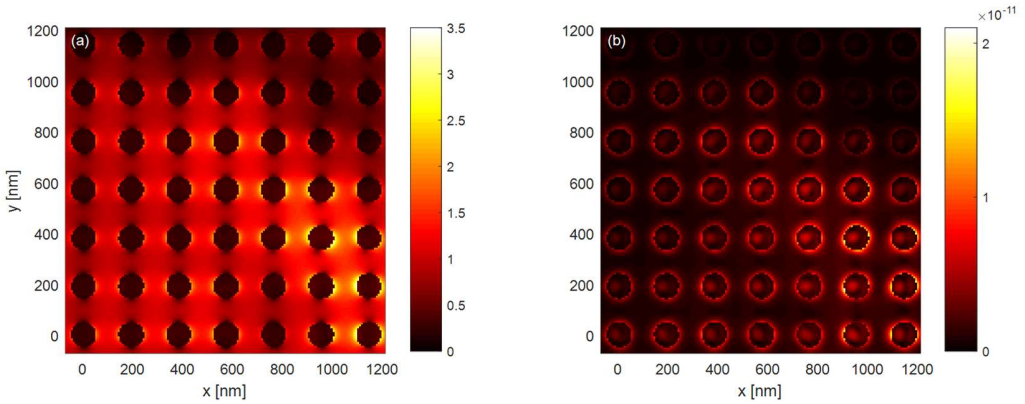

Figure 7. *Electric field amplitude plots at the FF (left panel) and SH (right panel), calculated for a cubic cluster of silicon nanospheres at the incident wavelength λ_{inc} = 800 nm. The fields are determined in the xy-plane at z=570 nm.*

4 System requirements and contact information

OPTIMET-3D is portable on most Unix 64-bit systems. **OPTIMET-3D** contains no known bugs; however, there is no guarantee that the code is bug free! If you encounter any problems while using **OPTIMET-3D**, please contact us at sekulic.ivan7@gmail.com with a brief description of the problem.

5 Installation instructions

OPTIMET-3D depends on the following external packages. In most cases, the build system will try to download and install dependencies it cannot find on the system.

- CMake: The build system. Must be installed independently.
- MPI: Required to run in parallel. Must be installed independently. Essential for non-spherical particles.
- Scalapack: (optional) Parallel linear algebra. Must be installed independently. Only useful when compiling with MPI.
- Belos: (optional) A library of iterative solvers. Must be installed independently. Only useful when compiling with MPI. Comes as a part of a Trilinos package. Version 12-10-1 of Trilinos is used (git checkout trilinos-release-12-10-1).
- Boost: (required) A set of peer-reviewed c++ libraries. At this juncture, **OPTIMET-3D** only requires the math special functions module. Automatically downloaded if unavailable.
- Eigen: (required) A c++ linear algebra library. Automatically downloaded if unavailable.
- hdf5: (required) A standard library for handling data. Automatically downloaded if unavailable. Only the C bindings are used. The c++ bindings are not necessary.
- GSL: (required) Numerical library for C and C++. Automatically downloaded if unavailable.
- F2C: (required) Fortran to C library. Required by the Amos package.

The build system is CMake (version 3.0 or higher). It can generate a build environment from a fair number of systems, from Unix makefiles to Xcode projects.

The canonical usage is as follows:

```
cd /path/to/optimet_source
mkdir build
cd build
cmake -DCMAKE_BUILD_TYPE=Release -Ddompi=ON -Ddoarshp=OFF ..
make
```

The example above will *not* compile the code responsible for the analysis of non-spherical targets. Set this option to ON if you would rather have it. The code used for the TMM analysis of non-spherical particles has to be compiled with the MPI option set to ON.
The executable `Optimet3D` should be directly in the build directory.

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OPTIMET-3D Documentation End