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Notes

Optical characterization of electrically charged particles using discrete dipole approximation

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ABSTRACT

The dependence of the electric potential on the absorption and scattering of light by small particles has emerged as an interesting research topic, as the unexpected amplified optical signatures of a system of electrically charged particles were satisfactory predicted recently for homogeneous, uniformly charged spheres. However, natural particles are rarely of spherical shape. A comprehensive understanding of how arbitrarily shaped, charged particles interact with electromagnetic radiation has been missing. The approach we present here attempts to fill this gap by introducing a numerical formulation of the electromagnetic scattering problem for these particles. The first results from the inter-comparison of numerical and analytical solutions for a pseudosphere show that the resonance features found are largely consistent, except for the magnitude and width of the peak amplitude, which may be due to inherent differences in the approaches used.

1. Introduction

Electromagnetic scattering is a conventional tool in rapid contactless characterization of particulate media. Optical methods for microphysical and chemical diagnoses of solid components have been developed in diverse fields of science: chemistry, biology, atmospheric and environmental sciences, astrophysics, etc. All such methods are based on the fundamental principles of electromagnetic interaction with small particles, i.e., measurement and analysis of the scattered light. Great progress in light-scattering theory discerned in the last two decades makes it possible to treat arbitrarily shaped particles with anisotropic optical properties and inclusions [3,16,20]. However, many particles may carry electric charge or become charged due to mutual interactions (collisions) or other physical or chemical processes in their natural environment.

It has been shown only recently that the optical signatures of electrically charged particles small compared to the wavelength may differ appreciably from what we can observe for equal-volume electrically neutral particles [10,17,6]. Near-field phenomena represent a specific class of optical effects that are manifested by intensity damping near a large charged particle, or intensity amplification if a charged dielectric particle has a size smaller than the illuminating wavelength [13]. The analytical solution to the Maxwell equations only exists for spherical, homogeneous, uniformly charged particles [11], while the separation of variables method (SVM) may fail if unduly applied to the electromagnetic scattering for non-uniform distributions of surface charges. For instance, Li et al. [14] have used the SVM approach to treat partially charged

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dust particles, and it is not clear if the conclusions reached using this method may be substantiated.

Many particles that occur in nature are, however, of irregular shapes. It is currently not possible to determine the optical properties of electrically charged non-spherical particles by exact analytical methods. This is why a numerical approach is needed to treat such particles. We have proceed here with the Discrete Dipole Approximation (DDA), mainly because it is well-suited to model irregular and heterogeneous material configurations by replacing a continuum target with an array of point dipoles [5]. DDA is valuable in assessing the optical properties of nanosized particles of different nature including metals [2], metal–semiconductors [19], and dielectrics [18,1], where all of the particles examined are much smaller than the illuminating wavelength and have complex morphologies. The main strength of the DDA is that it allows for modeling of non-uniform charge distributions at the particle surface through the position-depend refractive index. Such a non-uniform distribution of excess charges still remains a large source of uncertainty in the optical characterization of random particulate media.

The validation results obtained here for a pseudosphere appear promising and thus we expect this *Short Communication* can influence the research of the optics community in many aspects. An initial study on an electrically charged pseudosphere is important as it allows for a direct comparison with the analytical solution. Note that dipoles can be arranged uniformly within a discretized model of the equal-volume pseudosphere (Fig. 1). A benchmarking made in this way can uncover potential pitfalls as well as assist in remediation.

2. Theoretical considerations

It has been shown [12] that the physics incorporating net charges can be introduced through a surface-current density imposed on the boundary conditions, while resulting in new physical outcomes, for instance in unexpectedly amplified optical signatures of a system of electrically charged particles (see [12]). Surface-current conductivity is linearly proportional to a phenomenological surface conductivity σ_S and to the tangential electric field at the surface of a particle. In fact, an excess surface-charge conductivity makes the particle properties more metallic at its surface or rather in a monomolecular layer at the interface between a particle and host medium. This is why the electrical losses and impedance can be exceptionally high in the layer.

DDA is the point dipole representation of a scattering particle in 3D space, so a conductive shell can be simulated by a finite-sized layer. A smaller interdipole separation can better simulate both the surface conductive layer and the real geometry of a particle. Unfortunately, CPU and memory requirements increase as fast as the cube of the number of dipoles [4]. It is not difficult to show that the complex permittivity ϵ in the Helmholtz equation

$$0=\Delta E+i\omega\mu_0\sigma E-\omega^2\epsilon\epsilon_0\mu_0 E=\Delta E-\omega^2\mu_0\epsilon_0\left(\epsilon-i\frac{\sigma}{\omega\epsilon_0}\right)E$$

can be derived from the permittivity ϵ' of the bulk of the material:

$$\epsilon=\epsilon'-i\frac{\sigma}{\omega}, \tag{1}$$

where ω is the angular frequency of the applied radiation, E is the electric field, and μ_0 and ε_0 are the permeability and permittivity in vacuum, respectively. Volume conductivity σ of a surface layer is determined from the surface conductivity σ_S:

$$\sigma=\frac{\sigma_S}{h}, \tag{2}$$

where h is the geometrical thickness of a conductive layer, and a flow of electric charge in an otherwise infinitesimally thin conductive shell is used to model the electric current in a finite-sized surface layer of a discretized particle. It has to be emphasized that the surface conductivity depends on the frequency of the applied radiation, the plasma frequency, relaxation time of electrons, temperature, etc., so it is position dependent (see for more details [10,11]). The magnitude at which the optical effects by charged particles can deviate from those by neutral particles has relation to the parameter g that scales the coefficients in the scattered-field expansions [15] and is linearly proportional to the surface conductivity [10]:

$$g=i\omega k^{-1}\mu_0\sigma_s=\frac{x}{2}\frac{\omega_s^2}{\omega^2+\gamma_s^2}\left(-1+i\frac{\gamma_s}{\omega}\right), \tag{3}$$

where ω_s is the surface plasma frequency, $x=2\pi R/\lambda$ is the size parameter, R is the particle radius, λ is the wavelength of the incident radiation, and $k\equiv\ \omega\ \sqrt{\mu_0\varepsilon_0}$. The parameter γ_s is linearly proportional to the temperature [12], while the square of ω_s is a linear function of the surface potential [13]. Note that g is a dimensionless parameter that simply emerges from the solution of Maxwell equations taking into account the Drude model and modified boundary conditions (for more detail see Eq. 26 in [9]).

We have computed the refractive index of a conductive layer $m=\sqrt{\epsilon/\epsilon_0}$ using Eq. (1) for a model of a spherical particle of SiO_2 charged to 5 V and cooled to a temperature of 100 K (to be consistent with our previous computations, e.g. [13] or [12]). We consider the refractive index of $1.934+0.037i$ at $\lambda\approx 50$ μm [7] and γ_s is as large as $k_BT/\hbar$, where k_B is the Boltzmann constant, T is the temperature and $\hbar$ is the Planck constant. The wave phase shift $|m|kd$ over the distance d between neighboring dipoles was kept below 0.0055 to be one to two orders of magnitude below the conservative criterion suggested by Draine and Flatau [4]. However, the real and imaginary parts of the complex refractive index of a conductive shell may both exceed the value of 10 when mutual relations between size parameter, refractive index, and surface conductivity satisfy resonance conditions. Note that current implementations of the DDA do not work well when the refractive index is very large. This is why we are using a filtered coupled-dipole method (FLTRCD) near resonant frequencies [4]. Although FLTRCD is probably the best option, the accuracy of DDSCAT (i.e. the numerical implementation of DDA) is still limited and can only improve if the depth h of a conductive layer increases, thus resulting in an efficient impedance decline (see Eq. (2)). This can be reached when