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Optical signatures of electrically charged particles: Fundamental problems and solutions

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ABSTRACT

The explicit solution to Maxwell's equations that satisfies the continuity equation is obtained for electrically charged spherical particles. The traditional separation-of-variables method (SVM) cannot be used to solve the vector wave equation for a non-uniformly charged spherical particle. In addition, a perturbation approach to the electromagnetic scattering problem fails if a spherical particle is occupied by electric charges that are not spatially homogeneous. By incorporating a correction to the conventional surface-current density, we have refined the conductivity model and found that the Rayleigh approximation (for mode $n=1$) is not a valid approach for modelling the optical effects by electrically charged particles much smaller than the wavelength of an incident radiation. Theoretical analyses indicate that peak enhancements of optical signatures are usually relevant in the long-wavelength limit due to the necessity to include higher-order modes of vibration ($n>1$).

1. Introduction

Scattering of electromagnetic waves by electrically neutral spheres has been well-known since the derivation of the Lorenz–Mie solution to Maxwell's equations over a century ago [1,2]. Although the optical properties of neutral particles were studied intensively in the last decades, the effect of surface charges on the electromagnetic scattering has received only minor consideration [3–5]. The fundamentals of scattering theories for charged systems date back to Bohren and Hunt [6] who introduced an excess charge surface conductivity independent of position on a spherical particle. Later Klačka and Kocifaj [7] provided exact derivations showing, e.g., that far-field scattering from neutral and electrically charged spheres differ significantly when the wavelength of the incident radiation is approximately two to three orders of magnitude larger than the diameter of the particle. Under such conditions a slightly absorbing charged spherule tends to attenuate the electromagnetic radiation more efficiently than the volume equivalent electrically neutral particle.

Incorporating the effect of charge on the light scattering is performed through the surface current density $\vec{K}=(\eta_0+\eta)\vec{u}_t$, where $\vec{u}_t$ is the tangential velocity of the surface charges, $\eta=\mathrm{Re}[\tilde{\eta}\exp(-i\omega t)]$ and η_0 is the static component of the surface charge density. The excess charge Q is confined to the surface and the excess surface

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charge density $\eta_s = (\eta_0 + \eta)$ fulfils $\int \eta_s \, dA = Q$, $\int \eta_0 \, dA = Q$, $\int \tilde{\eta} \, dA = 0$, where the integrals are taken over the surface of the sphere [6].

The surface conductivity can involve various physical processes, such as the movement of unbounded charge on metallic or semi-metallic surfaces, passing the excess electrons into the conductive band of an isolant [11] or flow of bounded electrons in the polarizable states on the particle surface.

At present, the optical effects due to excess charges on a small particle or on a collection of small particles are largely unknown; however, they are of high importance for new technologies [8–10], for correct understanding of many physical processes [11], and for the revelation of new physical phenomena [12,13]. Specifically, the interstellar extinction features that are notoriously associated with material composition also may be affected by electric charge that acts as a modulator of the optical properties of small particles. A few preliminary results have shown that very small ($\approx$nm sized) spheres exhibit significant differences between charged and uncharged states [14]. This can be understandable partly because the charge/volume is large for these tiny particles and partly because of low values of the imaginary part of the complex refractive index in some parts of the spectrum. One driving question is how the effect depends on particle microphysics and what is the role of the net charge on resonance enhancements in the far-field and near-field zones.

Optical effects related to surface charge comprise a new topic and there exist many unanswered questions. Many unknowns relate to fundamental physical considerations:

(i) The volume electric current density is proportional to the free charge density and this leads to Ohm's law in differential form. However we require a 2-dimensional reformulation of both the current density and Ohm's law. The role of η in the surface current $\vec{K} = (\eta_0 + \eta)\vec{u}_t$ also has not been thoroughly established.

(ii) Inconsistencies between the continuity equation and some of the present solutions to the electromagnetic problem for charged particles need clarification and remediation, otherwise the predicted set of optical features can be incomplete or even incorrect.

(iii) Separation of variables method (SVM) is unable to solve the electromagnetic problem for non-uniform distributions of the surface charges η_0 and may result in unphysical optical signatures if applied to partially charged spherical particles (see e.g. [15–17]). In general, the use of SVM for non-uniformly charged particles is incorrect within a linear theory and fails to satisfy the continuity equation.

Until recently, few papers have been dedicated to electromagnetic scattering by charged particles. The different approaches have resulted in some inconsistencies that need remediation. In this paper we make a thorough investigation of the fundamentals of EM scattering by charged particles, showing that the range of applicability of SVM approach is limited to uniformly charged spheres and cannot be used to treat EM scattering by, e.g., partially charged spheroids or inhomogeneously charged spheres.

The paper discusses the relations for the surface current density and the continuity equation for the surface charge densities. The present theory represents a correction to the relations published in previous papers (e.g. [6]).

2. Conventional approach to the surface current density

The conventional approach to consider the surface current density incorporates the equation of motion for a surface charge with mass m_s and electric charge q_s

$$m_s \dot{\vec{u}}_t = q_s \vec{E}_t - \beta_s \vec{u}_t, \tag{1}$$

where $\vec{u}_t$ is the velocity component tangential to the surface of the sphere of radius R, β_s is a damping coefficient, and $\vec{E}_t$ is the tangential electric field [18]. Eq. (1) is a 2-dimensional formulation of the 3-D Drude model. The oscillatory solution of Eq. (1) is

$$\vec{u}_t = \frac{q_s/m_s}{\gamma_s - i\omega} \vec{E}_t, \tag{2}$$

where $\gamma_s \equiv \beta_s/m_s$ is inversely proportional to the relaxation time τ of electrons. For highly conductive materials, such as metals, τ is long and can be determined from the formula

$$\tau^{-1} \propto \gamma_s \approx k_B T/\hbar, \tag{3}$$

where k_B is the Boltzmann constant, T is the temperature, and $\hbar$ is the reduced Planck constant. However, Eq. (3) is surprisingly valid for a wide range of materials at temperatures over tens of Kelvins [19].

The conventional surface current density $\vec{K}_c$ is given by the relations

$$\vec{K}_c = (\eta_0 + \eta)\vec{u}_t, \tag{4}$$

and

$$\vec{K}_c = \sigma_s \vec{E}_t, \tag{5}$$

see e.g. [20]. Eqs. (4) and (5) correspond to the 2-dimensional differential form of Ohm's law.

Eqs. (1)–(2) and (4)–(5) have been presented by Bohren and Hunt [6], while Eq. (3) has been presented by Klačka and Kocifaj [7].

3. Generalized model

The explicit solution to Maxwell's equations for an uncharged, homogeneous spherical particle is known as Lorenz–Mie theory that reduces the complexity of the vector wave equation by introducing SVM. This concept makes a transition to the scalar wave equation through three equations, one for each coordinate [21], which makes satisfying the boundary conditions straightforward. The boundary conditions for the case $\eta_0 = 0$ are given by the following relations for electric and magnetic intensities, the surface current and charge densities:

$$\left(\varepsilon_0 \vec{E}_2 - \varepsilon_1 \vec{E}_1\right) \cdot \vec{n} = \eta,$$

$$\left(\mu_0 \vec{H}_2 - \mu_1 \vec{H}_1\right) \cdot \vec{n} = 0,$$