# Mie scattering by a charged dielectric particle: proposal for a novel plasma probe

R. L. Heinisch, F. X. Bronold, H. Fehske

*Institut für Physik, Ernst-Moritz-Arndt-Universität Greifswald, D-17489 Greifswald*

We consider how the scattering of light by a dielectric sphere (Mie scattering) is affected by a negative particle charge. Our aim is to identify the charge signatures in the Mie signal and to assess whether they could be used as a diagnostic for the particle charge. Quite generally the Mie signal contains information about surplus electrons as their electrical conductivity modifies either the boundary condition for electromagnetic fields or the polarizability of the material [1]. But to clarify to what extent and in what spectral range the particle charge influences the scattering of light requires a microscopic calculation of the electrical conductivity of the surplus electrons together with a judicious analysis of the different scattering regimes of the particle.

Where surplus electrons are trapped on the particle depends on the electron affinity $\chi$ of the dielectric [2]. For $\chi < 0$ (e.g. LiF) electrons are trapped in the image potential in front of the surface where they form a spherical two-dimensional electron gas around the grain. In this case the electrical field parallel to the surface induces a surface current $\mathbf{K} = \sigma_s \mathbf{E}_{\|}$ which is proportional to the surface conductivity $\sigma_s$. The surface current enters the boundary condition for the magnetic field. Thus, $\hat{\mathbf{e}}_r \times (\mathbf{H}_i + \mathbf{H}_r - \mathbf{H}_t) = 4\pi\mathbf{K}/c$, where $c$ is the speed of light [1].

For $\chi > 0$, (e.g. $Al_2O_3$) electrons form a space charge layer in the conduction band. Its width is typically larger than a micron. For micron sized particles we can thus assume a homogeneous distribution of the excess electrons in the bulk. The effect on light scattering is now encoded in the bulk conductivity of the excess electrons $\sigma_b$, which gives rise to a polarizability $\alpha = 4\pi i\sigma_b/\omega$, with $\omega$ the frequency, entering the refractive index $N = \sqrt{\varepsilon + \alpha}$.

The electrical conductivities $\sigma_s$ and $\sigma_b$ are limited by electron-phonon scattering. For $\chi < 0$ the coupling of the electron to the surface phonon associated with the transverse optical (TO) phonon encompasses a static part, which gives rise to the image potential, and a dynamic part, which limits $\sigma_s$. For $\chi > 0$ the $\sigma_b$ is limited by the interaction of the electron with a longitudinal optical bulk phonon. The surface and bulk conductivities are proportional to the surface or bulk electron density (see [3] for details on the calculation of $\sigma_s$ and $\sigma_b$).

The scattering behaviour is determined by the scattering coefficients. They are obtained by matching incident, reflected and transmitted partial waves at the surface of the sphere. Following

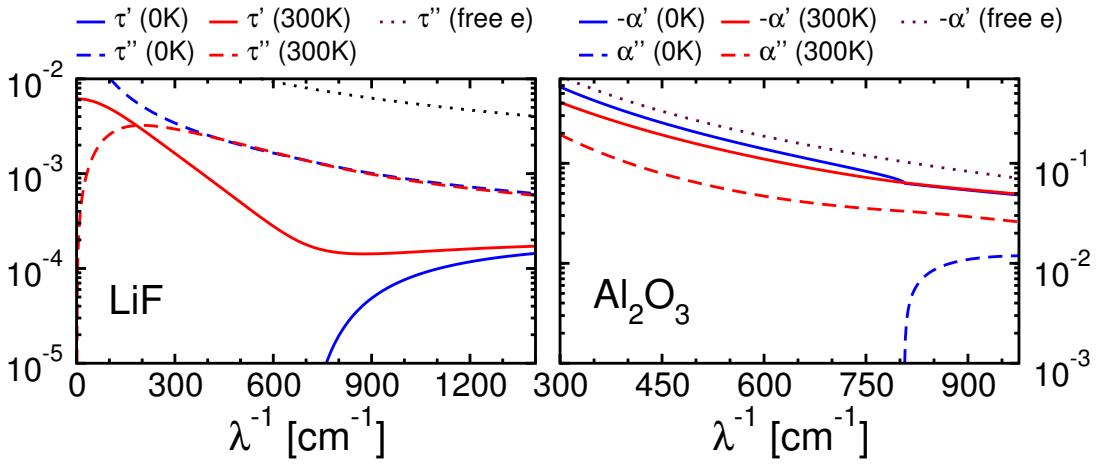


Figure 1: Dimensionless surface conductivity $\tau = \tau' + i\tau''$ for LiF for $n_s = 10^{13}$ cm$^{-2}$ (left) and polarisability of excess electrons $\alpha = \alpha' + i\alpha''$ for $Al_2O_3$ for $n_b = 3 \times 10^{17}$ cm$^{-3}$ (right) as a function of the inverse wavelength $\lambda^{-1}$.

Bohren and Hunt [1], they are given by

$$a_n^r = \frac{\psi_n(N\rho)\psi_n'(\rho) - [N\psi_n'(N\rho) - i\tau\psi_n(N\rho)]\,\psi_n(\rho)}{[N\psi_n'(N\rho) - i\tau\psi_n(N\rho)]\,\xi_n(\rho) - \psi_n(N\rho)\xi_n'(\rho)} \tag{1}$$

and

$$b_n^r = \frac{\psi_n'(N\rho)\psi_n(\rho) - [N\psi_n(N\rho) + i\tau\psi_n'(N\rho)]\,\psi_n'(\rho)}{[N\psi_n(N\rho) + i\tau\psi_n'(N\rho)]\,\xi_n'(\rho) - \psi_n'(N\rho)\xi_n(\rho)} \tag{2}$$

with $\rho = 2\pi a/\lambda$ the size parameter ($\lambda$ is the wavelength and $a$ the radius of the sphere) and $\psi_n(\rho) = \sqrt{\pi\rho/2}J_{n+1/2}(\rho)$, $\xi_n(\rho) = \sqrt{\pi\rho/2}H^{(1)}_{n+1/2}(\rho)$ with $J_n$ the Bessel and $H_n^{(1)}$ the Hankel function of the first kind. For for $\chi < 0$ ($\chi > 0$) the dimensionless surface conductivity $\tau(\omega) = 4\pi\sigma_s(\omega)/c$ ($\tau = 0$) and the refractive index $N = \sqrt{\varepsilon}$ ($N = \sqrt{\varepsilon + \alpha}$). As for uncharged particles the extinction efficiency is given by $Q_t = -(2/\rho^2)\sum_{n=1}^{\infty}(2n+1)\,\mathrm{Re}\,(a_n^r + b_n^r)$.

The charge effects in light scattering are controlled by the dimensionless surface conductivity $\tau = \tau' + i\tau''$ or the polarizability $\alpha = \alpha' + i\alpha''$. Both are shown in Fig. 1 for LiF and $Al_2O_3$ as a function of the inverse wavelength $\lambda^{-1}$. They turn out to be small even for a highly charged particle with $n_s = 10^{13}$ cm$^{-2}$ (corresponding to $n_b = 3 \times 10^{17}$ cm$^{-3}$ for $\chi > 0$ and $a = 1\mu$m). Surplus charge can thus only significantly affect light scattering where the Mie signal is particularly sensitive to a small $\tau$ or $\alpha$.

The most promising effect of the surplus electrons on the scattering of light is found at the anomalous optical resonances. They occur where the dielectric constant $\varepsilon = \varepsilon' + i\varepsilon''$ has $\varepsilon' < 0$ and $\varepsilon'' \ll 1$. For dielectric particles they lie in the infrared above the TO phonon resonance and are sensitive to small variations of $\varepsilon$ and also to $\tau$ and $\alpha$.

The extinction resonances are blue-shifted with increasing particle charge. This effect is most

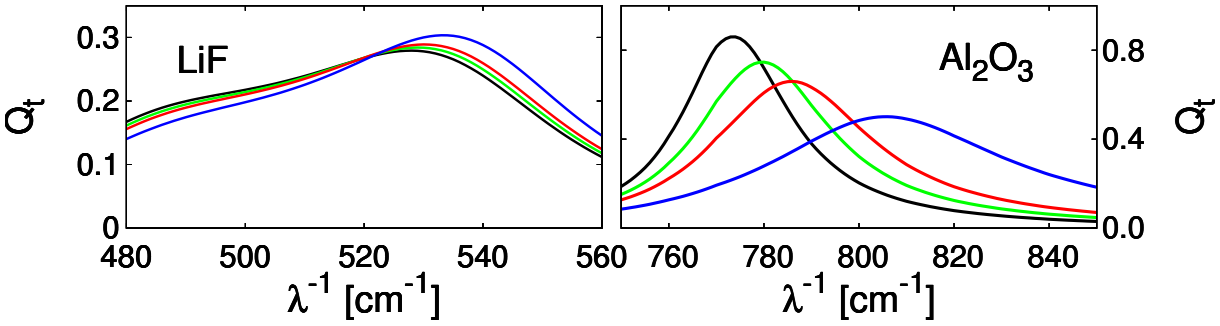


Figure 2: Extinction efficiency $Q_t$ as a function of the inverse wavelength $\lambda^{-1}$ for a LiF particle with $n_s = 0$ (black), $10^{12}$ (green), $2\times10^{12}$ (red), and $5\times10^{12}$ cm$^{-2}$ (blue) (left) and an $Al_2O_3$ particle with $n_b = 3n_s/a$ (right) for $T = 300$ K and radius $a = 0.05\mu$m.

significant for small particles. Figure 2 shows the lineshape of the extinction resonance for a LiF and an $Al_2O_3$ particle with $a = 0.05\mu$. For both materials the surplus electrons lead to a blue-shift of the resonance.

For submicron-sized particles where the resonance shift is most significant $a < \lambda$. In this small particle limit we can expand the scattering coefficients for small $\rho$. Up to $\mathcal{O}(\rho^3)$ this yields $a_1^r = a_2^r = b_2^r = 0$ and only $b_1^r \sim \mathcal{O}(\rho^3)$ contributes. Then the extinction efficiency reads

$$Q_t = \frac{12\rho\,(\varepsilon'' + \alpha'' + 2\tau'/\rho)}{(\varepsilon' + \alpha' + 2 - 2\tau''/\rho)^2 + (\varepsilon'' + \alpha'' + 2\tau'/\rho)^2}\,. \tag{3}$$

Excess charges enter either by $\tau$ ($\chi < 0$) or $\alpha$ ($\chi > 0$). For $\tau, \alpha \to 0$ this gives the limit of Rayleigh scattering. The resonance is located where

$$\varepsilon' + 2 - 2\tau''/\rho = 0 \quad \text{for} \quad \chi < 0 \quad \text{or} \quad \varepsilon' + \alpha' + 2 = 0 \quad \text{for} \quad \chi > 0, \tag{4}$$

and has a Lorentzian shape provided $\varepsilon''$ and $\tau'$ (or $\alpha''$) vary only negligibly near the resonance wavelength. Figure 2 confirms the Lorentzian lineshape for $Al_2O_3$. For LiF, however, $\varepsilon''$ has a

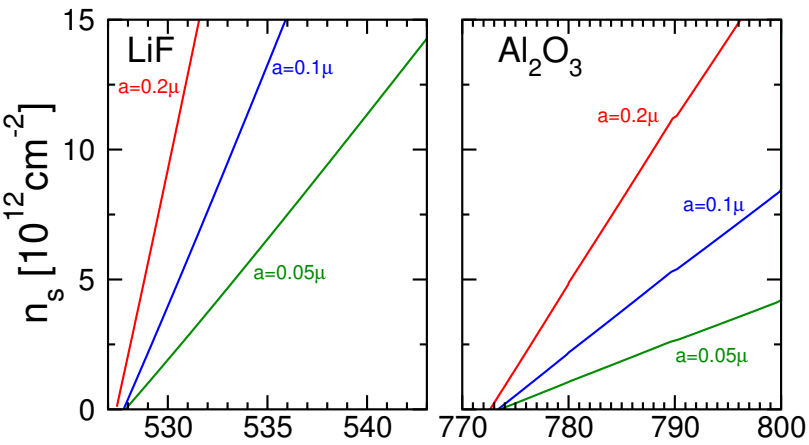


Figure 3: Position of the extinction resonance depending on the surface charge $n_s$ for LiF and $Al_2O_3$ (for equivalent bulk charge $n_b = 3n_S/a$) particles with different radii $a$.

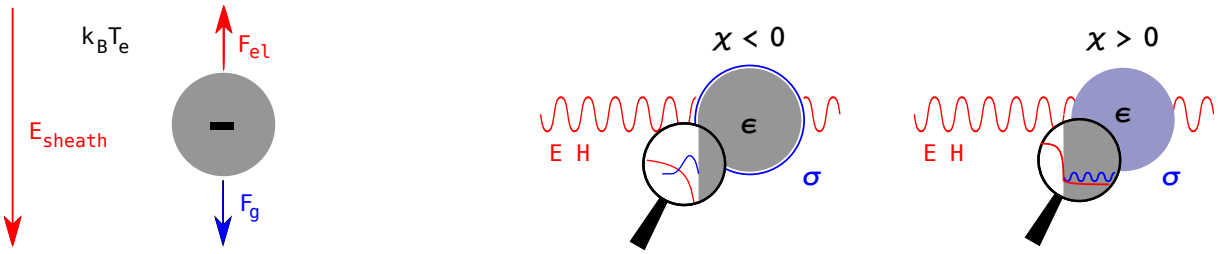


Figure 4: A conventional charge measurement (left) and the proposed optical charge measurement by Mie scattering (right). The conventional charge measurement relies on trapping the particle in the sheath and exploiting a force balance between gravity and the electric force on the particle. The knowledge of the plasma parameters is required to infer the particle charge. The optical measurement does not require the knowledge of plasma parameters.

hump close to the resonance due to a second TO phonon mode which is much weaker than the dominant TO phonon. This leads to the deviation from the Lorentzian lineshape.

The resonance shift is proportional to $n_s$ (LiF) or $n_b$ ($Al_2O_3$). This can be seen in Fig. 3 where we plot on the abscissa the shift of the extinction resonance arising from the surface electron density (or corresponding bulk electron density) given on the ordinate. Both bulk and surface electrons lead qualitatively to the same resonance shift.

We suggest to use the shift of the extinction resonance to determine the particle's charge. The most promising candidate for such an optical charge measurement is $Al_2O_3$ where the shift is strongest. Unlike for many conventional methods (see for instance [4]) this would allow the determination of the particle charge independent of the plasma parameters (see Fig. 4).

Eventually particles showing the resonance shift could be employed as minimally invasive electric probes. The particles would accumulate a charge depending on the local plasma environment. Determining their charge from Mie scattering and the forces acting on them by conventional means would then allow to infer the local electron density and temperature.

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