Scattering of light on metallic spheres

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Introduction

We introduce an alternative, but more consistent, treatment to the scattering of light at small metallic particles compared to the solution proposed by Gustav Mie in 1908.

At the beginning of the 20th century Gustav Mie introduced a new approach to scattering theory. Today, this treatment is used for applications far beyond the feasibility of that time and for systems that defy the original assumptions. Metallic nanoparticles are such a system because they are both non-dielectric and very small.

The goal of our research is to find an alternative approach to scattering theory. Using the successes of Mie-theory, while steering clear of the assumptions that are no longer appropriate, we have been able to create a consistent treatment of a Drude-metal sphere in a vacuum environment.

Mie-theory

Developed in 1908, Mie-theory has both successes and problems.

Maxwell equations → Vector spherical harmonics → Boundary: Geometry / Material: dielectric function → Solution: E & B-field → Derived quantities

Boundary: Geometry

$$\vec{D}_1 \cdot \vec{n} = \vec{D}_2 \cdot \vec{n}$$
$$\vec{B}_1 \cdot \vec{n} = \vec{B}_2 \cdot \vec{n}$$
$$\vec{E}_1 \cdot \vec{e}_t = \vec{E}_2 \cdot \vec{e}_t$$
$$\vec{H}_1 \cdot \vec{e}_t = \vec{H}_2 \cdot \vec{e}_t$$

Material: dielectric function

The dielectric function accounts for all the material properties:

$$\vec{D}_{k,\omega} = \varepsilon(k,\omega)\,\vec{E}_{k,\omega}$$
$$\vec{H}_{k,\omega} = \mu^{-1}(k,\omega)\,\vec{B}_{k,\omega}$$

SUCCESSES

- Analytical solution
- Decomposition in Vector spherical harmonics
- Easy implementation of boundary conditions
  - Dielectric function deals with charges and currents in dielectrics

PROBLEMS

- Dielectric function inappropriate for small metals
- Longitudinal waves ignored
- Treatment not physically insightful

Our treatment

Our alternative treatment solves some of the problems of Mie-theory.

Differences with Mie-theory

- We use the complete basis (both longitudinal and transversal part) of the vector spherical harmonics
- The material is directly implemented using the (Drude) equation of motion to describe the motion of electrons due to the EM-fields.
- The geometry is implemented by restricting the motion of the electron fluid in stead of the EM-fields

Maxwell equations → Vector spherical harmonics → Helmholtz decomposition → Material: Drude → Geometry: restrict movement → Solution: E & B-field → Derived quantities

Material: Drude

Solve equation of motion:

$$\mathcal{L}(\vec{\chi}(\vec{r}),\vec{v},t) = \frac{1}{2}m_e\vec{v}^2 - e\vec{A}\cdot\vec{v} + e\phi$$

$$\rightarrow \begin{cases} \rho^{(ind)}(\vec{r},\omega) = en_0\vec{\nabla}\cdot\vec{\chi}(\vec{r},\omega) \\ \vec{J}^{(ind)}(\vec{r},\omega) = i\omega en_0\vec{\chi}(\vec{r},\omega) \end{cases}$$

Geometry: restrict movement

$$\vec{\chi}(\vec{r},\omega) \rightarrow \vec{\chi}(\vec{r},\omega)\,\Theta(R-r)$$

Restrict electron movement

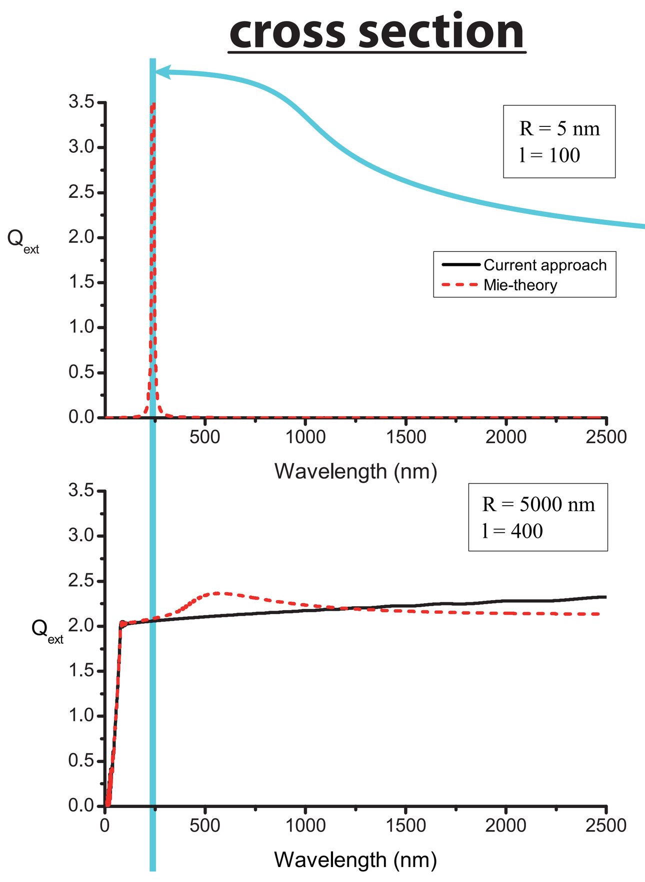
ADVANTAGES

- Consistent treatment of longitudinal and transversal waves
- Possibility to implement inhomogeneous electron densities (e.g. edge effects)
- More insightful treatment on electron level
- No need to introduce virtual D & H fields

Results

Comparing both treatments with each other shows striking differences for small nanoparticles. Larger particle sizes give better agreement, but some differences are still visible.

Relative far-field extinction cross section

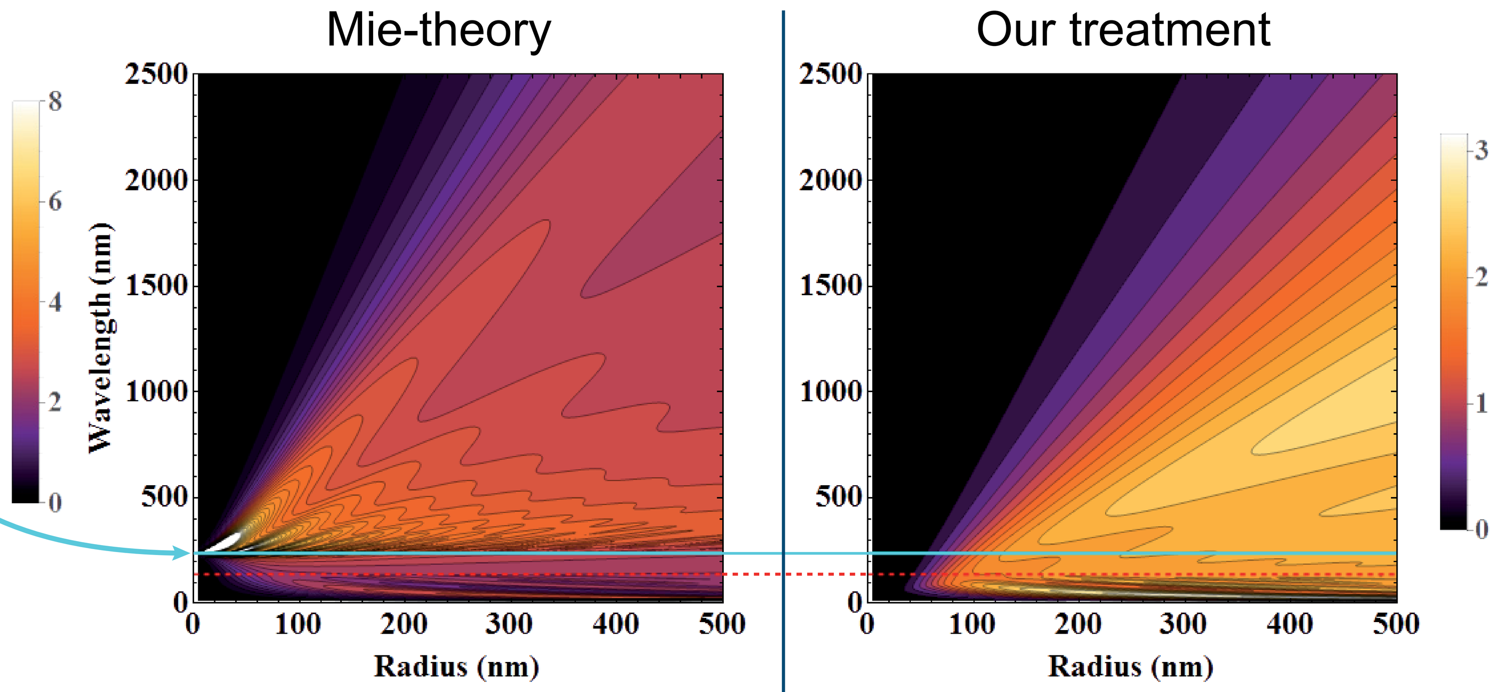


Small particles

- Mie-theory: sharp resonance at sphere plasma frequency: $\omega_{sphere} = \omega_{pl}\sqrt{\frac{l}{2l+1}}$
- Our treatment: almost no interaction -> plasma frequency only in near-field

Large particles

- Both theories become similar
- Difference: small bump around 500 nm



A single peak at the plasma frequency for a sphere is composed of different smaller peaks which fan out towards higher wavelengths for larger particles.

From low wavelengths different peaks appear and grow while fanning out towards higher wavelengths for larger particles.

Conclusion

We propose an alternative treatment of the scattering of light at metallic particles capable of circumventing some of the problems inherent to Mie-theory opening the road to a correct treatment of scattering by metallic nanoparticles.

We have demonstrated that using the successes of Mie-theory we can create an alternative way of solving the Maxwell equations circumventing some of the problems originating from Mie's assumptions. In this way we have created the basis of a theory that could surpass Mie-theory for metallic structures and small nanoparticles.

Implementing the most basic description of metals already gives some important differences with Mie-theory. The extremely high and small resonance for small particles as predicted by Mie-theory vanishes. For large particles both treatments seem to agree quite well.

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Soon to be published

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