

General Scattering Characteristics of Resonant Core-Shell Spheres

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Abstract

We present the general features and aspects regarding the electromagnetic scattering by a small core-shell sphere. First, the thickness effects on the plasmonic resonances are described in the Rayleigh limit, utilizing the MacLaurin expansion of the Mie coefficients of hollow scatterers. The results are connected with the plasmon hybridization model. A brief study regarding the core effects is given, illustrating resonant scattering peculiarities. Finally, new electrodynamic aspects of the scattering process are revealed through a the newly introduced Padé expansion of the Mie coefficients. The proposed methods can be readily applied for the studies of the dynamical dependencies for other canonical shapes, carving ways for engineering the overall properties of a single composite scatterer.

1 Introduction

One of the most studied canonical problems demonstrating a wealth of electromagnetic radiation controlling possibilities is the problem of plane wave scattering by a spherical particle; a benchmarking platform that provide insights about the nature of many single scattering phenomena [1, 2]. In this presentation we will discuss the general features regarding the electromagnetic scattering by a small core-shell sphere.

2 Results and Discussion

Thickness effects for the plasmonic resonances (collective oscillations of the free conduction charges in metals [3]) will be presented for the electrostatic limit [4], revealing a series of special characteristics, such as the resonant trends of the symmetric and antisymmetric dipole resonances [5]. For instance the Taylor expansion of the first electric Mie coefficient (a_1) for a hollow core-shell sphere gives

$$a_1^T \approx i \frac{2}{3} y^3 \frac{(\epsilon_2 - 1)(2\epsilon_2 + 1)(\eta^3 - 1)}{(\epsilon_2 + 2)(2\epsilon_2 + 1) - 2\eta^3(\epsilon_2 - 1)^2} \quad (1)$$

with pole condition described by

$$\epsilon^\pm = \frac{5 + 4\eta^3 \mp 3\sqrt{1 + 8\eta^3}}{4(\eta^3 - 1)} \quad (2)$$

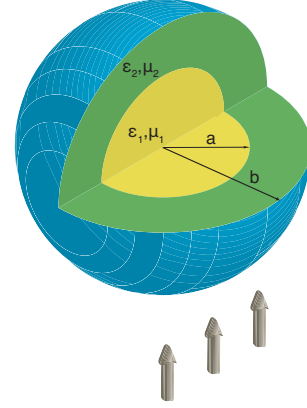


Figure 1. Problem setup: a core-shell sphere with internal radius a and external b under plane wave illumination.

where $\eta = \frac{a}{b}$ is the radius ratio. Note that we have two types of resonances, the symmetric (bonding) and antisymmetric (antibonding) [6], denoted ϵ^- and ϵ^+ , respectively.

A generalization towards higher order multipoles is given, following the same static-limit approximation the Mie coefficients for small hollow scatterers [7]. As a rule-of-thumb thick shells exhibit a volume dependence on their first dipole resonance, while thin shells have a linear dependence, even for higher multipoles.

The agreement between the static-derived results and the plasmon hybridization model will be demonstrated, illustrating the common mathematical origins of these theoretical models. For the case of a spherical hollow core-shell structure, after a significant amount of calculations, these resonant frequencies are described by the following condition [8]

$$\omega_{n\pm}^2 = \frac{\omega_B^2}{2} \left[1 \pm \frac{1}{2n+1} \sqrt{1 + 4n(n+1)\eta^{2n+1}} \right] \quad (3)$$

where ω_{1+}^2 and ω_{1-}^2 is the symmetric and antisymmetric resonances for a given background frequency (plasma frequency) ω_B and a given multipole, i.e., dipole ($n = 1$), quadrupole ($n = 2$) and so on [9].

By assuming a lossless Drude material dispersion model, i.e., $\epsilon = 1 - \frac{\omega_B^2}{\omega^2}$ and inserting it in Eq. (3) we obtain two

discrete resonances

$$\varepsilon_n^\pm = 1 - \frac{2}{1 \pm \frac{\sqrt{4n(2n+1)\eta^{2n+1}+1}}{2n+1}} \quad (4)$$

described as a function of the radius ratio η . For the dipole case the following expression is obtained

$$\varepsilon_1^\pm = 1 - \frac{2}{1 \pm \frac{1}{3}\sqrt{1+8\eta^3}} \quad (5)$$

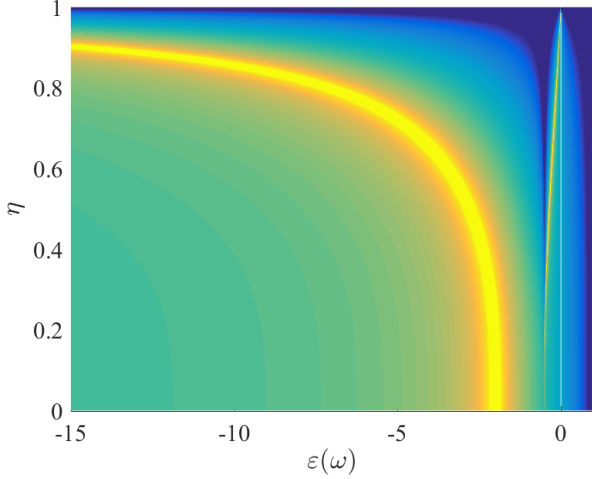


Figure 2. The extinction efficiency spectrum as a function of the core material (lossless) and the radius ratio for a small sphere ($y = 0.01$). For the plasmonic case ($b_n \approx 0$) the extinction cross section is $Q_{\text{ext}} \propto \sum_n \Re\{a_n\}$, with $n = 1, 2, \dots$. The bright lines correspond to the symmetric (left line) and antisymmetric (right line) dipole plasmonic resonances, while higher order multipoles are not visible. Note that the color scale is logarithmic and is omitted, normalized for better visualization of the resonances.

Additionally, the effects due to the (relative) permittivity of the core of the will be discussed, revealing the existence of a core-induced scattering peculiarity. Briefly, for every n -multipole there is a specific permittivity value of the core where both symmetric and antisymmetric resonances converge, for very thick shells ($\eta \rightarrow 0$), i.e.,

$$\varepsilon_{\text{branching}} = \left(\frac{n+1}{n}\right)^2 \quad (6)$$

The presentation will conclude with an analysis of some electrodynamic effects, utilizing the Padé approximants of the Mie coefficients. This expansion is able to capture the dynamic scattering mechanisms e.g., radiative damping [10, 11]. In this way both static and hybridization model are expanded, revealing promising results about the scattering process, such as the damping dependencies in both radius ratio and core material permittivities. These results can inspire further studies on the dynamical dependencies

of other canonical shapes, carving ways for engineering the radiative damping mechanism and affecting the overall properties of a single composite scatterer.

Acknowledgements

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References

- [1] X. Fan, W. Zheng, and D. J. Singh, “Light scattering and surface plasmons on small spherical particles,” *Light Sci. Appl.*, vol. 3, p. e179, Jun 2014.
- [2] B. Luk’yanchuk, N. I. Zheludev, S. A. Maier, N. J. Halas, P. Nordlander, H. Giessen, and C. T. Chong, “The Fano resonance in plasmonic nanostructures and metamaterials.,” *Nat. Mater.*, vol. 9, no. 9, pp. 707–715, 2010.
- [3] U. Kreibig and M. Vollmer, *Optical properties of metal clusters*. Springer-Verlag, 1995.
- [4] A. H. Sihvola, “Character of surface plasmons in layered spherical structures,” *Prog. Electromagn. Res.*, vol. 62, pp. 317–331, 2006.
- [5] E. Prodan, C. Radloff, N. J. Halas, and P. Nordlander, “A hybridization model for the plasmon response of complex nanostructures.,” *Science*, vol. 302, no. 5644, pp. 419–422, 2003.
- [6] E. Prodan and P. Nordlander, “Plasmon hybridization in spherical nanoparticles,” *J. Chem. Phys.*, vol. 120, no. 11, pp. 5444–5454, 2004.
- [7] A. Sihvola and I. V. Lindell, “Transmission Line Analogy for Calculating the Effective Permittivity of Mixtures with Spherical Multilayer Scatterers,” *J. Electromagn. Waves Appl.*, vol. 2, no. 8, pp. 741–756, 1988.
- [8] G. Mukhopadhyay and S. Lundqvist, “Density oscillations and density response in systems with nonuniform electron density,” *Nuovo Cim. B Ser. 11*, vol. 27, pp. 1–18, May 1975.
- [9] C. E. Román-Velázquez and C. Noguez, “Designing the plasmonic response of shell nanoparticles: Spectral representation,” *J. Chem. Phys.*, vol. 134, no. 4, 2011.
- [10] G. A. Baker and P. R. Graves-Morris, *Padé approximants*, vol. 59. Cambridge University Press, 1996.
- [11] D. C. Tzarouchis, P. Ylä-Oijala, and A. Sihvola, “Unveiling the scattering behavior of small spheres,” *Phys. Rev. B*, vol. 94, no. 14, p. 140301(R), 2016.