



**UNIVERSIDAD NACIONAL AUTÓNOMA DE MÉXICO
POSGRADO EN CIENCIAS FÍSICAS**

**OPTICAL RESPONSE OF PARTIALLY EMBEDDED
NANOSPHERES**

**TESIS
QUE PARA OPTAR POR EL GRADO DE:
MAESTRO EN CIENCIAS (FÍSICA)**

PRESENTA:

JONATHAN ALEXIS URRUTIA ANGUIANO

TUTOR:

**DR. ALEJANDRO REYES CORONADO
FACULTAD DE CIENCIAS, UNAM**

MIEMBROS DEL COMITÉ TUTOR

**DRA. CITLALI SÁNCHEZ-AKÉ
INSTITUTO DE CIENCIAS APLICADAS Y TECNOLOGÍA, UNAM**

**DR. GIUSEPPE PIRRUCCIO
INSTITUTO DE FÍSICA, UNAM**

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Abstract/Resumen

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Background and Motivation

It is recommended to fill in this part of the document with the following information:

Metasurfaces are bidimensional arrays of unit cells with geometrical dimensions smaller than the wavelength of the incident light. Biosensing-aimed metasurfaces are supported onto a substrate and immersed in an aqueous medium, thus its performance is limited by its washability. One alternative to decrease a metasurface's washability is to partially embed it within the substrate while allowing the metasurface to still interact with the aqueous medium. In this work, we study the optical response of a single partially embedded metallic nanosphere and extend its behavior analytically into a disordered metasurface by employing an effective medium approach.

The incrustation of the metasurface is studied by calculating the spectral behavior of a single partially embedded nanosphere with the Finite Element Method (FEM) and proposing an effective polarizability in the elements of the metasurface.

- Your field: Context about the field your are working
Plasmonics -> Metamaterials -> Biosensing
- Motivation: Backgroung about your thesis work and why did you choose this project and why is it important.
Fabrication -> Partially embedded NPs -> No analytical (approximated) method physically introduces the incrustation degree. There are numerical solutions and Effective Medium Theories approaching the problem but the later only as a fitting method.
- Objectives: What question are you answering with your work.
Can optical non invasive tests (IR-Vis) retrieve the average incrustation degree for monolayers of small spherical particles?
- Methology: What are your secondary goals so you achieve your objective. Also, how are you answering yout question: which method or model.
Bruggeman homogenization theories on bidimensional systems?
Is the dipolar approximation is enough or do we need more multipolar terms?
Do we need the depolarization factors?
- Structure: How is this thesis divides and what is the content of each chapter.

Chapter 1

Optical Properties of Single Plasmonic Nanoparticles

The problem studied in this thesis corresponds to the theoretical analysis of the Localized Surface Plasmon Resonances (LSPR) excited on plasmonic spherical nanoparticles (NPs) when these are under realistic experimental conditions, such as those present on plasmonic biosensors, where the NPs are partially embedded into a substrate [1]. The theoretical analysis consists on the numerical calculation of the absorption, scattering and extinction cross sections of a partially embedded metal NP employing the Finite Element Method (FEM), nevertheless, to verify the validity of the obtained results, the problem of the absorption and scattering of light by an isolated particle must be addressed. In this chapter, we revisit the general solution of the light absorption and scattering by both an arbitrary particle and by a spherical particle, given by the Mie Theory [2].

1.1 The Optical Theorem: Amplitude Matrix and Cross Sections

Let $\mathbf{E}^i = \mathbf{E}_0^i \exp(i\mathbf{k}^i \cdot \mathbf{r})$ be the electric field of an incident monochromatic plane wave with constant amplitude \mathbf{E}_0^i traveling through a non-dispersive medium with refractive index n_m , denominated matrix, in the direction $\mathbf{k}^i = k\hat{\mathbf{k}}^i$, with $k = (\omega/c)n_m$ the wave number of the plane wave into the matrix, and let \mathbf{E}^{sca} be the scattered electric field due to a particle with arbitrary shape embedded into the matrix. In general, the scattered electric field propagates in all directions but for a given point $\mathbf{r} = r\hat{\mathbf{e}}_r$ the traveling direction is defined by the vector $\mathbf{k}^{sca} = k\hat{\mathbf{k}}^{sca} = k\hat{\mathbf{e}}_r$. Due to the linearity of the Maxwell's equations, the incident and scattered electric fields in the far field regime are related by the linear relation [3],

$$\mathbf{E}^{sca} = \frac{\exp(i\mathbf{k}^{sca} \cdot \mathbf{r})}{r} \mathbb{F}(\hat{\mathbf{k}}^{sca}, \hat{\mathbf{k}}^i) \mathbf{E}^i, \quad (1.1)$$

where $\mathbb{F}(\hat{\mathbf{k}}^{sca}, \hat{\mathbf{k}}^i)$ is the scattering amplitude matrix from direction $\hat{\mathbf{k}}^i$ into $\hat{\mathbf{k}}^{sca}$. Since only the far field is considered, both the incident and the scattered electric field can be decomposed into two linearly independent components perpendicular to \mathbf{k}^i and \mathbf{k}^{sca} , respectively, each forming a

right-hand orthonormal system. If the particle acting as a scatterer has a symmetric shape, it is convenient to define the orthonormal systems relative to the scattering plane, which is the plane containing \mathbf{k}^i and \mathbf{k}^{sca} , since the elements of $\mathbb{F}(\hat{\mathbf{k}}^{sca}, \hat{\mathbf{k}}^i)$ simplify when represented in these bases [3]. By defining the directions perpendicular (\perp) and parallel (\parallel) to the scattering plane, the incident and scattered electric fields can be written as

$$\mathbf{E}^i = (E_{\parallel}^i \hat{\mathbf{e}}_{\parallel}^i + E_{\perp}^i \hat{\mathbf{e}}_{\perp}^i) \exp(i\mathbf{k}^i \cdot \mathbf{r}), \quad (1.2)$$

$$\mathbf{E}^{sca} = (E_{\parallel}^{sca} \hat{\mathbf{e}}_{\parallel}^{sca} + E_{\perp}^{sca} \hat{\mathbf{e}}_{\perp}^{sca}) \frac{\exp(i\mathbf{k}^{sca} \cdot \mathbf{r})}{r}, \quad (1.3)$$

where an harmonic time dependence $\exp(-i\omega t)$ has been suppressed, and where it has been assumed that the scattered field is described by a spherical wave; the superindex ‘i’ (‘sca’) denotes the orthonormal system defined by the incident plane wave (scattered fields). Since $\{\hat{\mathbf{e}}_{\perp}^i, \hat{\mathbf{e}}_{\parallel}^i, \hat{\mathbf{k}}^i\}$ and $\{\hat{\mathbf{e}}_{\perp}^{sca}, \hat{\mathbf{e}}_{\parallel}^{sca}, \hat{\mathbf{k}}^{sca}\}$ are right-hand orthonormal systems, they are related as follows

$$\hat{\mathbf{e}}_{\perp}^i = \hat{\mathbf{e}}_{\perp}^{sca} = \hat{\mathbf{k}}^{sca} \times \hat{\mathbf{k}}^i, \quad \hat{\mathbf{e}}_{\parallel}^i = \hat{\mathbf{k}}^i \times \hat{\mathbf{e}}_{\perp}^i, \quad \text{and} \quad \hat{\mathbf{e}}_{\parallel}^{sca} = \hat{\mathbf{k}}^{sca} \times \hat{\mathbf{e}}_{\perp}^{sca}. \quad (1.4)$$

As the Eqs. (1.4) suggest, the unit vector bases of the orthonormal systems relative to the scattering plane depend on the scattering direction. For example, if the incident plane wave travels along the z axis, then $\hat{\mathbf{k}}^i = \hat{\mathbf{e}}_z$ and $\hat{\mathbf{k}}^{sca} = \hat{\mathbf{e}}_r$. Thus, according to Eqs. (1.4), the unit vector bases of the systems relative to the scattering plane are $\hat{\mathbf{e}}_{\parallel}^i = \cos \varphi \hat{\mathbf{e}}_x + \sin \varphi \hat{\mathbf{e}}_y$, $\hat{\mathbf{e}}_{\parallel}^{sca} = \hat{\mathbf{e}}_\theta$ and $\hat{\mathbf{e}}_{\perp}^i = \hat{\mathbf{e}}_{\perp}^{sca} = -\hat{\mathbf{e}}_\varphi$, with θ the polar angle and φ azimuthal angle. In Fig. 1.1 the unit vector systems (purple) based on the scattering plane (green) defined by the vectors $\hat{\mathbf{k}}^i = \hat{\mathbf{e}}_z$ and $\hat{\mathbf{k}}^{sca} = \hat{\mathbf{e}}_r$ are shown, along with the Cartesian (blue) and spherical (black) unit vector bases.

After an incident plane wave interacts with a particle with a possible complex refractive index $n_p(\omega)$, the total electric field outside the particle is given by the sum of the incident and the scattered fields. Therefore, the time averaged Poynting vector $\langle \mathbf{S} \rangle_t$, denoting the power flow per unit area, of the total field is given by

$$\langle \mathbf{S} \rangle_t = \underbrace{\frac{1}{2} \operatorname{Re} (\mathbf{E}^i \times \mathbf{H}^{i*})}_{\langle \mathbf{S}^i \rangle_t} + \underbrace{\frac{1}{2} \operatorname{Re} (\mathbf{E}^{sca} \times \mathbf{H}^{sca*})}_{\langle \mathbf{S}^{sca} \rangle_t} + \underbrace{\frac{1}{2} \operatorname{Re} (\mathbf{E}^i \times \mathbf{H}^{sca*} + \mathbf{E}^{sca} \times \mathbf{H}^{i*})}_{\langle \mathbf{S}^{ext} \rangle_t}, \quad (1.5)$$

where $(*)$ is the complex conjugate operation and where the total Poynting vector is separated into the contribution from the incident field $\langle \mathbf{S}^i \rangle_t$, from the scattered field $\langle \mathbf{S}^{sca} \rangle_t$ and from their cross product denoted by $\langle \mathbf{S}^{ext} \rangle_t$. By means of the Faraday-Lenz Law and Eq. (1.1), the contribution to the Poynting vector from the incident and the scattered fields can be rewritten as

$$\langle \mathbf{S}^i \rangle_t = \frac{\|\mathbf{E}_0^i\|^2}{2Z_m} \hat{\mathbf{k}}^i, \quad \text{and} \quad \langle \mathbf{S}^{sca} \rangle_t = \frac{\|\mathbf{E}^{sca}\|^2}{2Z_m} \hat{\mathbf{k}}^{sca} = \frac{\|\mathbb{F}(\hat{\mathbf{k}}^{sca}, \hat{\mathbf{k}}^i) \mathbf{E}^i\|^2}{2Z_m r^2} \hat{\mathbf{k}}^{sca}, \quad (1.6)$$

with $Z_m = \sqrt{\mu_m/\epsilon_m}$, the impedance of the non-dispersive matrix, while the crossed contribution

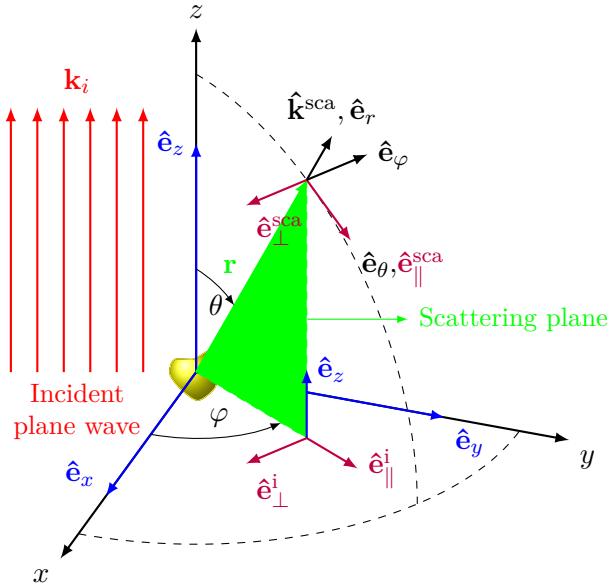


Fig. 1.1: The scattering plane (green) is defined by the vectors $\hat{\mathbf{k}}^i$, direction of the incident plane wave (red), and $\hat{\mathbf{k}}^{sca}$, direction of the scattered field in a given point \vec{r} . If the direction of the incident plane wave is chosen to be $\hat{\mathbf{e}}_z$, the parallel and perpendicular components of the incident field relative to the scattering plane are $\hat{\mathbf{e}}_||^i = \cos \varphi \hat{\mathbf{e}}_x + \sin \varphi \hat{\mathbf{e}}_y$ and $\hat{\mathbf{e}}_\perp^i = -\hat{\mathbf{e}}_\varphi$, while the components of the scattering field relative to the scattering plane are $\hat{\mathbf{e}}_\parallel^{sca} = \hat{\mathbf{e}}_\theta$, $\hat{\mathbf{e}}_\perp^{sca} = -\hat{\mathbf{e}}_\varphi$. The cartesian unit vector basis is shown in blue, the spherical unit vector basis in black, while the basis of the orthonormal systems relative to the scattering plane are shown in purple.

is given by

$$\langle \mathbf{S}^{ext} \rangle_t = \text{Re} \left\{ \frac{\exp[-i(\mathbf{k}^{sca} - \mathbf{k}^i) \cdot \mathbf{r}]}{2Z_m r^2} \left[\hat{\mathbf{k}}^{sca} (\mathbf{E}_0^i \cdot \mathbb{F}^* \mathbf{E}^{i*}) - \mathbb{F}^* \mathbf{E}^{i*} (\mathbf{E}_0^i \cdot \hat{\mathbf{k}}^{sca}) \right] + \frac{\exp[i(\mathbf{k}^{sca} - \mathbf{k}^i) \cdot \mathbf{r}]}{2Z_m r^2} \left[\hat{\mathbf{k}}^i (\mathbb{F} \mathbf{E}^i \cdot \mathbf{E}_0^{i*}) - \mathbf{E}_0^{i*} (\mathbb{F} \mathbf{E}^i \cdot \hat{\mathbf{k}}^i) \right] \right\}, \quad (1.7)$$

where the scattering amplitude matrix is evaluated as $\mathbb{F}(\hat{\mathbf{k}}^{sca}, \hat{\mathbf{k}}^i)$.

The power scattered by the particle can be calculated by integrating $\langle \mathbf{S}^{sca} \rangle_t$ in a closed surface surrounding the particle; if the scattered power is normalized by the irradiance of the incident field $\|\langle \mathbf{S}^i \rangle_t\|$, it is obtained a quantity with units of area known as the scattering cross section C_{sca} , given by

Scattering Cross Section

$$C_{sca} = \frac{2Z_m}{\|\mathbf{E}_0\|^2} \oint \langle \mathbf{S}^{sca} \rangle \cdot d\mathbf{a} = \oint \frac{\|\mathbb{F}(\hat{\mathbf{k}}^{sca}, \hat{\mathbf{k}}^i) \mathbf{E}^i\|^2}{\|\mathbf{E}_0^i\|^2} d\Omega, \quad (1.8)$$

where $d\Omega$ is the solid angle differential. In a similar manner, an absorption cross section C_{abs} can be defined as well. On the one side, the absorption cross section is given by the integral on

a closed surface of $-\langle \mathbf{S} \rangle_t$ [Eq. (1.5)] divided by the irradiance of the incident field, where the minus sign is chosen so that $C_{\text{abs}} > 0$ if the particle absorbs energy [2]. On the other side, if an Ohmic material for the particle with conductivity $\sigma(\omega) = i\omega n_p^2(\omega)$ [4] is assumed, through Joule's Heating Law [3] the absorption cross section can be computed as

Ohmic Particle - Absorption Cross Section

$$C_{\text{abs}} = \frac{1}{2} \int \frac{\text{Re}(\mathbf{j} \cdot \mathbf{E}^{\text{int}*})}{\|\mathbf{E}_0^i\|^2 / 2Z_m} dV = \int \omega Z_m \text{Im}(n_p^2) \frac{\|\mathbf{E}^{\text{int}}\|^2}{\|\mathbf{E}_0^i\|^2} dV, \quad (1.9)$$

where integration is performed inside the particle, and \mathbf{j} and \mathbf{E}^{int} , are the volumetric electric current density and the total electric field in this region, respectively. Both the scattering and the absorption cross sections are quantities related to the optical signature of a particle [5], and their relation can be made explicit by performing the surface integral representation of C_{abs} and defining C_{ext} , that is,

$$\begin{aligned} C_{\text{abs}} &= -\frac{2Z_m}{\|\mathbf{E}_0^i\|^2} \int (\langle \mathbf{S}^i \rangle_t + \langle \mathbf{S}^{\text{sca}} \rangle_t + \langle \mathbf{S}^{\text{ext}} \rangle_t) \cdot d\mathbf{a} \\ &= -C_{\text{sca}} - \frac{2Z_m}{\|\mathbf{E}_0^i\|^2} \int \langle \mathbf{S}^{\text{ext}} \rangle_t \cdot \hat{\mathbf{e}}_r d\Omega \\ &= -C_{\text{sca}} + C_{\text{ext}}, \end{aligned} \quad (1.10)$$

where the contribution of $\langle \mathbf{S}^i \rangle_t$ to the integral is zero since a non-dispersive matrix was assumed. From Eq.(1.10) it can be seen that C_{ext} takes into account both mechanisms for energy loses (scattering and absorption), thus it is called the extinction cross section. To solve the integral in Eq. (1.10) let us define θ as the angle between $\hat{\mathbf{k}}^{\text{sca}}$ and $\hat{\mathbf{k}}^i$ as the polar angle and φ as the azimuthal angle as shown in Fig 1.1. With this election of coordinates, the extinction cross section can be computed as

$$\begin{aligned} C_{\text{ext}} &= -\text{Re} \left\{ \frac{\exp(-ikr)}{\|\mathbf{E}_0^i\|^2} \oint \exp(ikr \cos \theta) (1) (\mathbf{E}^i \cdot \mathbb{F}^* \mathbf{E}^{i*}) d\Omega \right. \\ &\quad + \frac{\exp(ikr)}{\|\mathbf{E}_0^i\|^2} \oint \exp(-ikr \cos \theta) \cos \theta (\mathbf{E}^{i*} \cdot \mathbb{F} \mathbf{E}^i) d\Omega \\ &\quad \left. + \frac{\exp(ikr)}{\|\mathbf{E}_0^i\|^2} \oint \exp(-ikr \cos \theta) \sin \theta (E_{0,x}^i \cos \varphi + E_{0,y}^i \sin \varphi) (\mathbb{F} \mathbf{E}^i \cdot \mathbf{k}^i) d\Omega \right\} \end{aligned} \quad (1.11)$$

where the relations $\hat{\mathbf{k}}^{\text{sca}} \cdot \hat{\mathbf{e}}_r = 1$, $\hat{\mathbf{k}}^i \cdot \hat{\mathbf{e}}_r = \cos \theta$ and $\mathbf{E}^{\text{sca}} \cdot \hat{\mathbf{e}}_r = 0$ were employed. The integrals in Eq. (1.11) can be solved by a two-fold integration by parts on the polar angle θ and by depreciating the terms proportional to r^{-2} . This process leads to a zero contribution from the integrand proportional to $\sin \theta$ of Eq. (1.11), and after arranging the other terms in their real and imaginary parts, it follows that C_{ext} depends only in the forward direction $\hat{\mathbf{k}}^{\text{sac}} = \hat{\mathbf{k}}^i$ ($\theta = 0$). This result is known as the Optical Theorem whose mathematical expression is given by [3, 5, 6]

Optical Theorem - Extinction Cross Section

$$C_{\text{ext}} = C_{\text{abs}} + C_{\text{sca}} = \frac{4\pi}{k\|\mathbf{E}_0^i\|^2} \text{Im} \left[\mathbf{E}_0^i \cdot \mathbb{F}^*(\hat{\mathbf{k}}^i, \hat{\mathbf{k}}^i) \mathbf{E}_0^{i*} \right]. \quad (1.12)$$

From Eqs. (1.5) and (1.12) it can be seen that the extinction of light, the combined result of scattering and absorption as energy loss mechanisms, is also a manifestation of the interference between the incident and the scattered fields and that the overall effect of the light extinction can be fully understood by analyzing the amplitude of the scattering field in the forward direction. It is worth noting that Eq. (1.12) is an exact relation but its usefulness is bond to the correct evaluation of the scattering amplitude matrix \mathbb{F} [3]. Thus, in the following sections a scattering problem with spherical symmetry will be assumed, so that the exact solution to the scattering amplitude matrix can be developed; this solution is known as Mie Theory.

1.2 Mie Scattering

In the previous section, it was concluded that the extinction of light due to the interaction between a particle and a monochromatic plane wave can be determinated through the amplitude of the scattered field in the forward direction. This is stated in the Optical Theorem, which is an exact relation, but inaccuracies can arise when either the scattering amplitude matrix or extinction cross section is approximated¹. A particular case in which the scattering amplitude matrix can be exactly calculated is when the scatterer has spherical symmetry. In order to address this special case it will be introduced a vectorial basis with spherical symmetry, known as the Vector Spherical Harmonics (VSH). Once the SVH are defined, they will be used to write a monochromatic plane wave and, lastly, the scattered field by a spherical particle will be calculated by imposing the continuity of the tangential components of the electric and magnetic field.

¹See for example Section 2.4 from Ref. [3] on the Rayleigh Scattering and Section 21.7 from Ref. [7] on Thompson scattering.

The Finite Element Method

2.1 Finite Element Method and Analytical Solutions

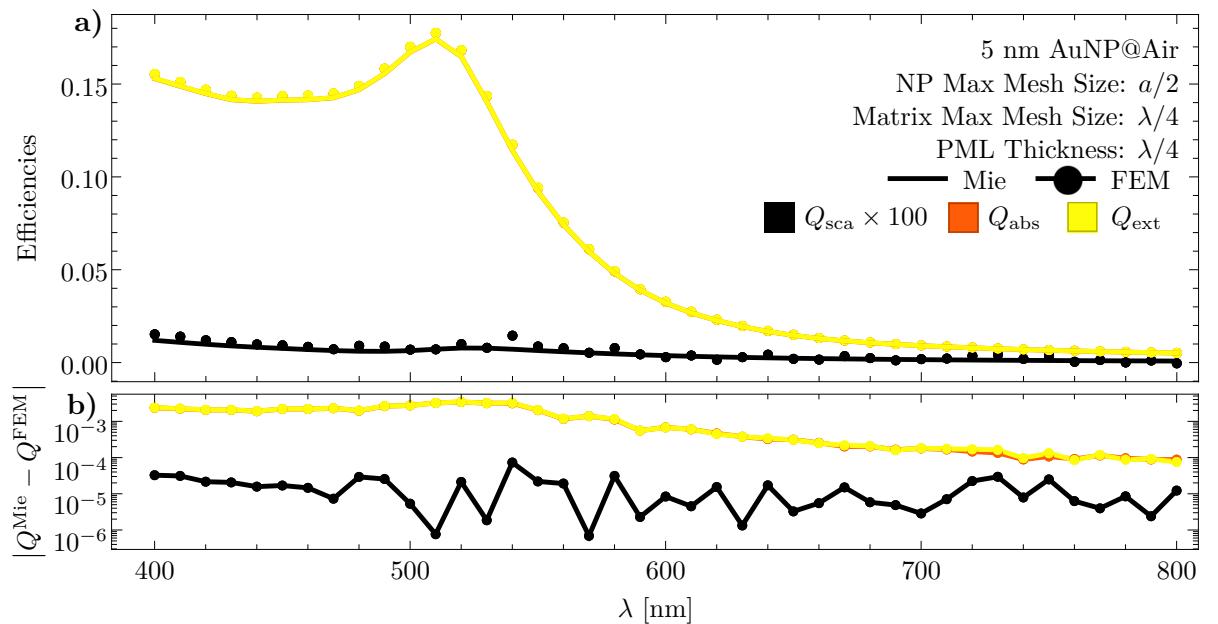
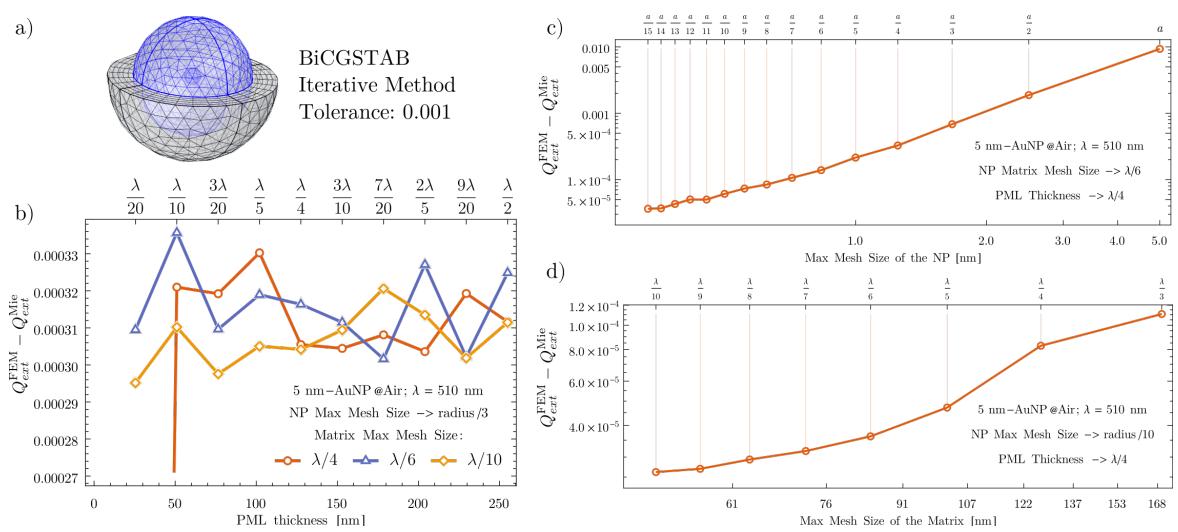
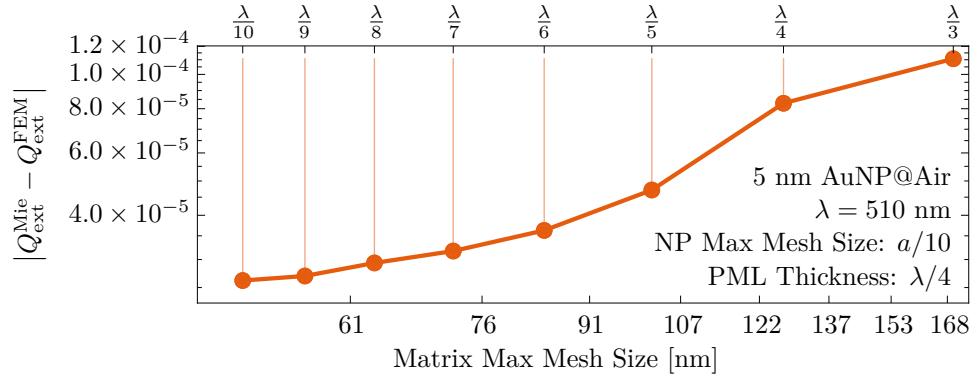
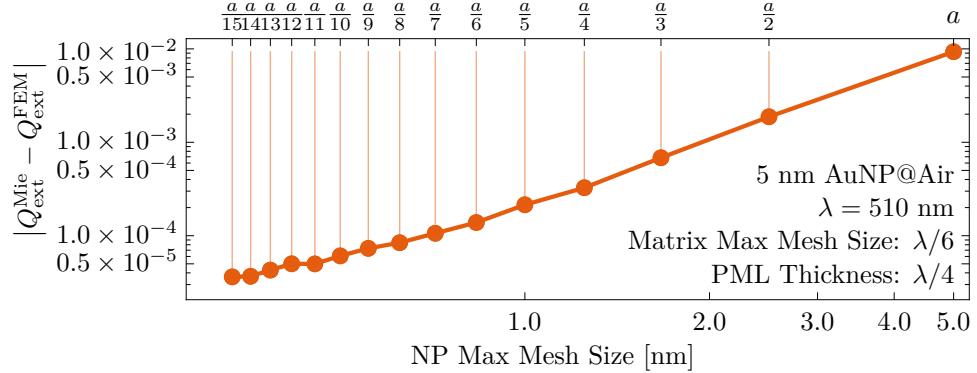


Fig. 2.1: Resonance wavelength (λ_{res}) of the scattering (orange) and extinction (black) cross sections as functions of the NPs radii when embedded 2.1a) into air and 2.1b) into water, and as function of the refractive index of the matrix for NP of radius set to ?? 12.5 nm and ?? 50 nm.

2. THE FINITE ELEMENT METHOD



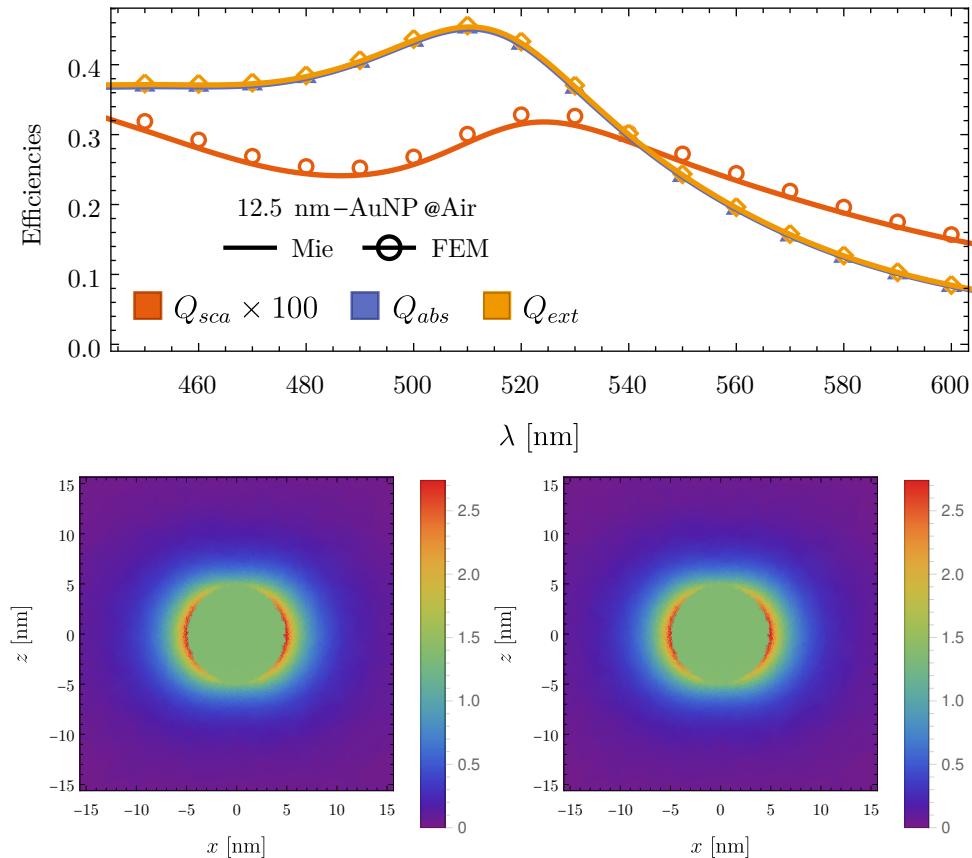


Fig. 2.5: Resonance wavelength (λ_{res}) of the scattering (orange) and extinction (black) cross sections as functions of the NPs radii when embedded 2.1a) into air and 2.1b) into water, and as function of the refractive index of the matrix for NP of radius set to ?? 12.5 nm and ?? 50 nm.

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