UNIVERSIDAD NACIONAL AUTÓNOMA DE MÉXICO
POSGRADO EN CIENCIAS FÍSICAS

OPTICAL RESPONSE OF PARTIALLY EMBEDDED NANOSPHERES

TESIS
QUE PARA OPTAR POR EL GRADO DE:
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Abstract/Resumen

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Contents

Introduction

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

- Your field: Context about the field your are working
  Plasmonics -> Metameterials -> 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 Amplitude Matrix and Cross Sections

Let $\mathbf{E}^{\mathrm{i}}$ be the electric field of an incident monochromatic plane wave traveling through a non-dispersive medium with refractive index n_{m}, denominated matrix, in the direction $\mathbf{k}^{\mathrm{i}} = k_{\mathrm{m}}\hat{\mathbf{k}}^{\mathrm{i}}$, with k_{m} the wave number of the plane wave into the matrix, and $\mathbf{E}^{\mathrm{sca}}$ the electric far field of the scattered 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 $\mathbf{k}^{\mathrm{sca}} = k_{\mathrm{m}}\hat{\mathbf{k}}^{\mathrm{sca}} = k_{\mathrm{m}}\hat{\mathbf{e}}_r$. Due to the linearity of the Maxwell's equations, in the far field the incident and scattered electric fields are related by a linear relation, that is,

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

where $\mathbb{F}(\hat{\mathbf{k}}^{\mathrm{sca}}, \hat{\mathbf{k}}^{\mathrm{i}})$ is the scattering amplitude matrix from direction $\hat{\mathbf{k}}^{\mathrm{i}}$ into $\hat{\mathbf{k}}^{\mathrm{sca}}$ [3]. 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 $\mathbf{k}^{\mathrm{i}}$ and $\mathbf{k}^{\mathrm{sca}}$, 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 $\mathbf{k}^{\mathrm{i}}$ and $\mathbf{k}^{\mathrm{sca}}$, since the elements of $\mathbb{F}(\hat{\mathbf{k}}^{\mathrm{sca}}, \hat{\mathbf{k}}^{\mathrm{i}})$ simplify when represented in these bases [3]. By defining the directions perpendicular ($\perp$) and parallel ($\|$) to the scattering plane, the incident and scattered electric fields can be written as

$$\mathbf{E}^{\mathrm{i}} = \left(E^{\mathrm{i}}_{\|}\hat{\mathbf{e}}^{\mathrm{i}}_{\|} + E^{\mathrm{i}}_{\perp}\hat{\mathbf{e}}^{\mathrm{i}}_{\perp}\right)\exp\left(i\mathbf{k}^{\mathrm{i}}\cdot\mathbf{r}\right), \tag{1.2}$$

$$\mathbf{E}^{\mathrm{sca}} = \left(E^{\mathrm{sca}}_{\|}\hat{\mathbf{e}}^{\mathrm{sca}}_{\|} + E^{\mathrm{sca}}_{\perp}\hat{\mathbf{e}}^{\mathrm{sca}}_{\perp}\right)\frac{\exp(i\mathbf{k}^{\mathrm{sca}}\cdot\mathbf{r})}{r}, \tag{1.3}$$

where the 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 $\{\hat{\mathbf{e}}^{\mathrm{i}}_{\perp}, \hat{\mathbf{e}}^{\mathrm{i}}_{\|}, \hat{\mathbf{k}}^{\mathrm{i}}\}$ and $\{\hat{\mathbf{e}}^{\mathrm{sca}}_{\perp}, \hat{\mathbf{e}}^{\mathrm{sca}}_{\|}, \hat{\mathbf{k}}^{\mathrm{sca}}\}$ are right-hand orthonormal systems, they are related by

$$\hat{\mathbf{e}}^{\mathrm{i}}_{\perp} = \hat{\mathbf{e}}^{\mathrm{sca}}_{\perp} = \hat{\mathbf{k}}^{\mathrm{sca}} \times \hat{\mathbf{k}}^{\mathrm{i}}, \qquad \hat{\mathbf{e}}^{\mathrm{i}}_{\|} = \hat{\mathbf{k}}^{\mathrm{i}} \times \hat{\mathbf{e}}^{\mathrm{i}}_{\perp}, \qquad \hat{\mathbf{e}}^{\mathrm{sca}}_{\|} = \hat{\mathbf{k}}^{\mathrm{sca}} \times \hat{\mathbf{e}}^{\mathrm{sca}}_{\perp}. \tag{1.4}$$

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 $\hat{\mathbf{k}}^{\mathrm{i}} = \hat{\mathbf{e}}_z$ and $\hat{\mathbf{k}}^{\mathrm{sca}} = \hat{\mathbf{e}}_r$. Thus, according to Eqs. (1.4), the unit vector bases of the systems relative to the scatterig plane are $\hat{\mathbf{e}}^{\mathrm{i}}_{\|} = \cos\varphi\hat{\mathbf{e}}_x + \sin\varphi\hat{\mathbf{e}}_y$, $\hat{\mathbf{e}}^{\mathrm{sca}}_{\|} = \hat{\mathbf{e}}_\theta$ and $\hat{\mathbf{e}}^{\mathrm{i}}_{\perp} = \hat{\mathbf{e}}^{\mathrm{sca}}_{\perp} = -\hat{\mathbf{e}}_\varphi$. In Fig. 1.1 the unit vector systems (purple) based on the scattering plane (green) defined by the vectors $\hat{\mathbf{k}}^{\mathrm{i}} = \hat{\mathbf{e}}_z$ and $\hat{\mathbf{k}}^{\mathrm{sca}} = \hat{\mathbf{e}}_r$ are shown, along with the Cartesian (blue) and spherical (black) unit vector bases.

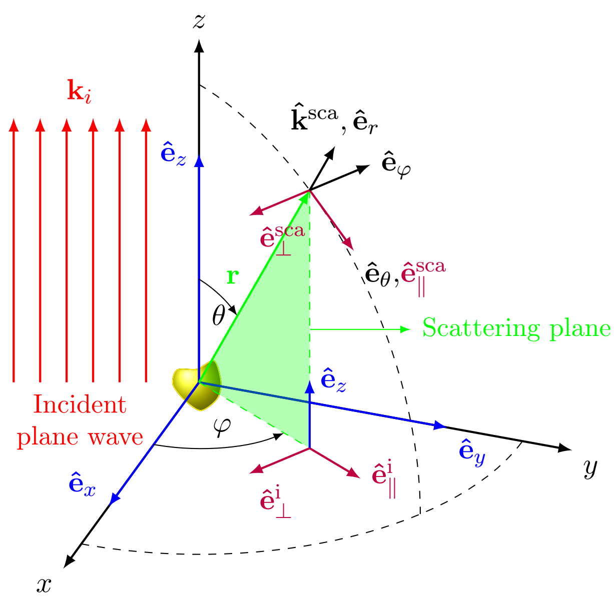


Fig. 1.1: The scattering plane (green) is defined by the vectors $\hat{\mathbf{k}}^{\mathrm{i}}$, direction of the incident plane wave (red), and $\hat{\mathbf{k}}^{\mathrm{sca}}$, direction of the scattered field in a given point $\vec{r}$. If the direction of the incident plane wave is chose to be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

After a incident plane wave interacts with a particle, the total electric field outside the particle is given by the sum of the incident and the scattered fields. Therefore the power per unit the time averaged Poynting vector, denoting the power flow per unit area, of the total field is given by

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

where it is separated into three contributions: the incident field, the scattered field and their cross product denoted by extinction field. By means of the Faraday-Lens Law and Eq. (1.1), the contribution to the Poynting vector from the incident and the scattered fields can be rewritten as

$$\langle \mathbf{S}^{\mathrm{i}} \rangle_t = \frac{\left\|\mathbf{E}^{\mathrm{i}}\right\|^2}{2Z_{\mathrm{m}}}\left\|\mathbf{E}^{\mathrm{i}}\right\|^2 \hat{\mathbf{k}}^{\mathrm{i}}, \qquad \text{and} \qquad \langle \mathbf{S}^{\mathrm{sca}} \rangle_t = \frac{\left\|\mathbf{E}^{\mathrm{sca}}\right\|^2}{2Z_{\mathrm{m}}}\hat{\mathbf{k}}^{\mathrm{sca}} = \frac{\left\|\mathbb{F}(\hat{\mathbf{k}}^{\mathrm{sca}}, \hat{\mathbf{k}}^{\mathrm{i}})\mathbf{E}^{\mathrm{i}}\right\|^2}{2Z_{\mathrm{m}} r}\hat{\mathbf{k}}^{\mathrm{sca}}, \tag{1.6}$$

with Z_{m} the impedance of the non-dispersive matrix, while the extinction field contribution is given by

$$\begin{aligned}\langle \mathbf{S}^{\mathrm{ext}} \rangle_t = \mathrm{Re}\Bigg\{ &\frac{\exp\left[-i(\mathbf{k}^{\mathrm{sca}} - \mathbf{k}^{\mathrm{i}}) \cdot \mathbf{r}\right]}{2Z_{\mathrm{m}} r^2}\left[\hat{\mathbf{k}}^{\mathrm{sca}}\left(\mathbf{E}^{\mathrm{i}} \cdot \mathbb{F}^{*}\mathbf{E}^{\mathrm{i}*}\right) - \left(\mathbb{F}\mathbf{E}^{\mathrm{i}*}\right)\left(\mathbf{E}^{\mathrm{i}} \cdot \hat{\mathbf{k}}^{\mathrm{sca}}\right)\right] \\ &+ \frac{\exp\left[i(\mathbf{k}^{\mathrm{sca}} - \mathbf{k}^{\mathrm{i}}) \cdot \mathbf{r}\right]}{2Z_{\mathrm{m}} r^2}\left[\hat{\mathbf{k}}^{\mathrm{i}}\left(\mathbf{E}^{\mathrm{i}*} \cdot \mathbb{F}\mathbf{E}^{\mathrm{i}}\right) - \left(\mathbf{E}^{\mathrm{i}*}\right)\left(\mathbb{F}\mathbf{E}^{\mathrm{i}} \cdot \hat{\mathbf{k}}^{\mathrm{i}}\right)\right]\Bigg\}, \end{aligned} \tag{1.7}$$

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

The power scattered by the particle can be calculated by integrating $\langle \mathbf{S}^{\mathrm{sca}} \rangle_t$ in a closed surface surrounding the particle and by normalizing the power scattered by $\left\|\langle \mathbf{S}^{\mathrm{i}} \rangle_t\right\|$, a quantity with units of area is obtain; this quantity is known as scattering cross section denoted by C_{sca} and given by

Scattering Cross Section

$$C_{\mathrm{sca}} = \int_{4\pi} \frac{\left\|\mathbf{E}^{\mathrm{sca}}\right\|^2}{\left\|\mathbf{E}^{\mathrm{i}}\right\|^2}\hat{\mathbf{k}}^{\mathrm{sca}} \cdot \hat{\mathbf{e}}_r r^2 \,\mathrm{d}\Omega = \int_{4\pi} \frac{\left\|\mathbb{F}(\hat{\mathbf{k}}^{\mathrm{sca}}, \hat{\mathbf{k}}^{\mathrm{i}})\mathbf{E}^{\mathrm{i}}\right\|^2}{\left\|\mathbf{E}^{\mathrm{i}}\right\|^2}\,\mathrm{d}\Omega\,. \tag{1.8}$$

In a similar manner, an absorption cross section C_{abs} can be defined. One way to define it is to consider the Joule's Heating Law [3] and assuming an Ohmic material for the particle with a conductivity $\sigma = i\omega n_p^2$ [4] and a refractive index n_p, that is,

Absorption Cross Section

$$C_{\mathrm{abs}} = \frac{1}{2}\int \frac{\mathrm{Re}(\mathbf{j} \cdot \mathbf{E}^{\mathrm{int}*})}{\left\|\mathbf{E}^{\mathrm{i}}\right\|^2/2Z_{\mathrm{m}}}\,\mathrm{d}V = \int \omega\,\mathrm{Re}(n_p)\,\mathrm{Im}(n_p)\frac{\left\|\mathbf{E}^{\mathrm{int}}\right\|^2}{\left\|\mathbf{E}^{\mathrm{i}}\right\|^2/2Z_{\mathrm{m}}}\,\mathrm{d}V\,, \tag{1.9}$$

where $\mathbf{j}$ is the electric currents inside the particle and $\mathbf{E}^{\mathrm{int}}$ the total electric field inside of the particle, where the integration is performed. Yet, another method to calculate C_{abs} is by performing the closed surface integral of Eq. (1.5) and normalizing by $\left\|\left\langle \mathbf{S}^{\mathrm{i}}\right\rangle_t\right\|$, leading to

$$\begin{aligned} C_{\mathrm{abs}} =& -\frac{2Z_{\mathrm{m}}}{\|\mathbf{E}^{\mathrm{i}}\|^2}\int \left(\left\langle \mathbf{S}^{\mathrm{i}}\right\rangle_t + \langle \mathbf{S}^{\mathrm{sca}}\rangle_t + \left\langle \mathbf{S}^{\mathrm{ext}}\right\rangle_t\right)\cdot \hat{\mathbf{e}}_r r^2\,\mathrm{d}\Omega \\ =& -C_{\mathrm{sca}} - \frac{2Z_{\mathrm{m}}}{\|\mathbf{E}^{\mathrm{i}}\|^2}\int \left\langle \mathbf{S}^{\mathrm{ext}}\right\rangle_t \cdot \hat{\mathbf{e}}_r\,\mathrm{d}\Omega \end{aligned} \tag{1.10}$$

where the contribution of $\left\langle \mathbf{S}^{\mathrm{i}}\right\rangle_t$ to the integral is zero since a non-dispersive matrix was assumed. To solve the integral in Eq. (1.10) let us define θ as the angle between $\hat{\mathbf{k}}^{\mathrm{sca}}$ and $\hat{\mathbf{k}}^{\mathrm{i}}$ as the polar angle and φ as the azimuthal angle as shown in Fig 1.1. With this election of coordinates, one can define the extinction cross section C_{ext} as

$$\begin{aligned} C_{\mathrm{ext}} = -\,\mathrm{Re}\Bigg\{ & \frac{\exp(-ik_m r)}{\|\mathbf{E}^{\mathrm{i}}\|^2}\int \exp(ik_m z\cos\theta)(1)\left(\mathbf{E}^{\mathrm{i}}\cdot \mathbb{F}^*\mathbf{E}^{\mathrm{i}*}\right)\mathrm{d}\Omega \\ & \frac{\exp(ik_m r)}{\|\mathbf{E}^{\mathrm{i}}\|^2}\int \exp(-ik_m z\cos\theta)\cos\theta\left(\mathbf{E}^{\mathrm{i}*}\cdot \mathbb{F}\mathbf{E}^{\mathrm{i}}\right)\mathrm{d}\Omega \\ & \frac{\exp(ik_m r)}{\|\mathbf{E}^{\mathrm{i}}\|^2}\int \exp(-ik_m z\cos\theta)(\sin\theta\cos\varphi + \sin\theta\sin\varphi)\left(\mathbf{E}^{\mathrm{i}*}\cdot \mathbb{F}\mathbf{E}^{\mathrm{i}}\right)\mathrm{d}\Omega\Bigg\}, \end{aligned} \tag{1.11}$$

where the relations $\hat{\mathbf{k}}^{\mathrm{sca}}\cdot\hat{\mathbf{e}}_r = 1$, $\hat{\mathbf{k}}^{\mathrm{i}}\cdot\hat{\mathbf{e}}_r = \cos\theta$, $\hat{\mathbf{E}}^{\mathrm{i}}\cdot\hat{\mathbf{e}}_r = (\sin\theta\cos\varphi + \sin\theta\sin\varphi)$ and $\mathbf{E}^{\mathrm{sca}}\cdot\hat{\mathbf{e}}_r = 0$ were employed. The integrals in Eq. (1.11) can be solved by a two fold integration by parts on the variable $\cos\theta$ and by depreciating the terms proportional to r^{-2}; this process leads to a zero contribution from the third term of Eq. (1.11). After arranging the results of the two fold integration by parts on the first two terms of Eq. (1.11), one can proof that the extinction cross section depend only in the forward direction $\theta = 0$ as follows [2]

Extinction Cross Section

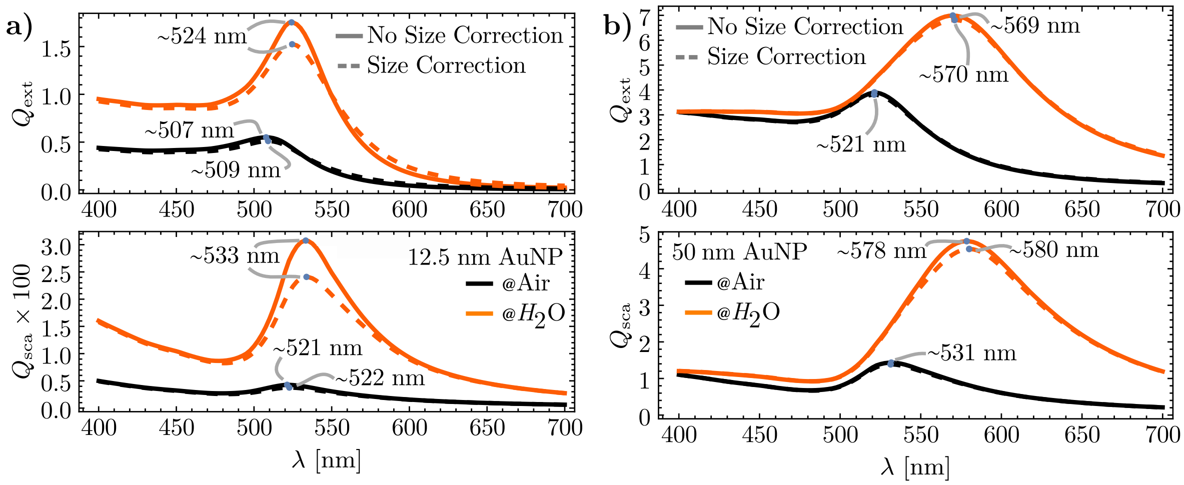
$$C_{\mathrm{ext}} = \frac{4\pi}{k_m^2\|\mathbf{E}^{\mathrm{i}}\|^2}\,\mathrm{Re}\left[\mathbf{E}^{\mathrm{i}}\cdot\mathbb{F}^*(\hat{\mathbf{k}}^{\mathrm{sca}},\hat{\mathbf{k}}^{\mathrm{i}})\mathbf{E}^{\mathrm{i}*}\right]\Bigg|_{\theta=0}. \tag{1.12}$$

Thus, the absorption cross section can be obtained from Eqs. (1.10) and (1.12), and this expression is one form of the so called optical theorem [2, 5].

Optical Theorem

$$C_{\mathrm{abs}} = C_{\mathrm{ext}} - C_{\mathrm{sca}}. \tag{1.13}$$

1.2 Mie Theory: Spherical Symmetry



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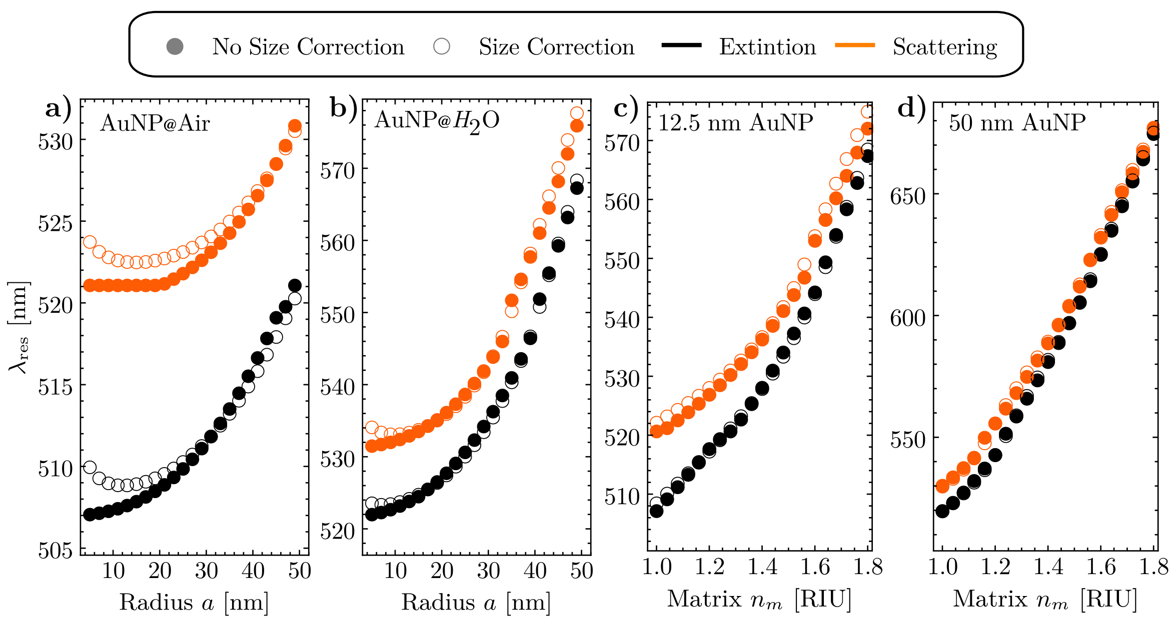

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Chapter 2

Results and discussion

2.1 Finite Element Method and Analytical Solutions

2.2 Incrustation Degree of a Spherical Particle

Chapter 3

Conclusions

3.1 Future Work: Application on Metasurfaces

Appendix A

The Finite Element Method

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List of Figures

Index