Some polarimetric considerations about the interaction of metallic nanoparticles with either substrates or other particles

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Overview

- Introduction
- Theoretical background
  - Conventional Plasmon Spectroscopy
  - The linear polarization degree
- Some results
  - Particle aggregates
  - Particles on or above substrates
- Conclusions
- Future work

Introduction

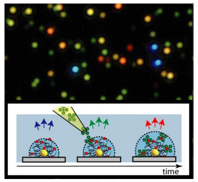
- Common experimental situations in plasmonics (nanotool+sample)
  - Volume configuration (isolated/aggregates)
  - Nanostructures on substrates
  - Nanoparticles above substrates

- Multiple scattering: influence on the plasmonic spectrum

Introduction

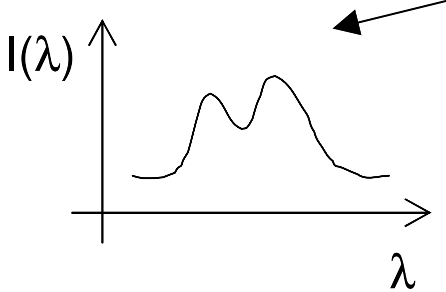
Incident light

Sample + Nanotool



Scattered light

Spectrograph



Analysis

- Peak positions
- Peak widths
- Spectral shapes

Introduction

- Spectrum: partial information (intensity)

- Mueller matrix measurement (polarimetry)

Introduction. Mueller matrices

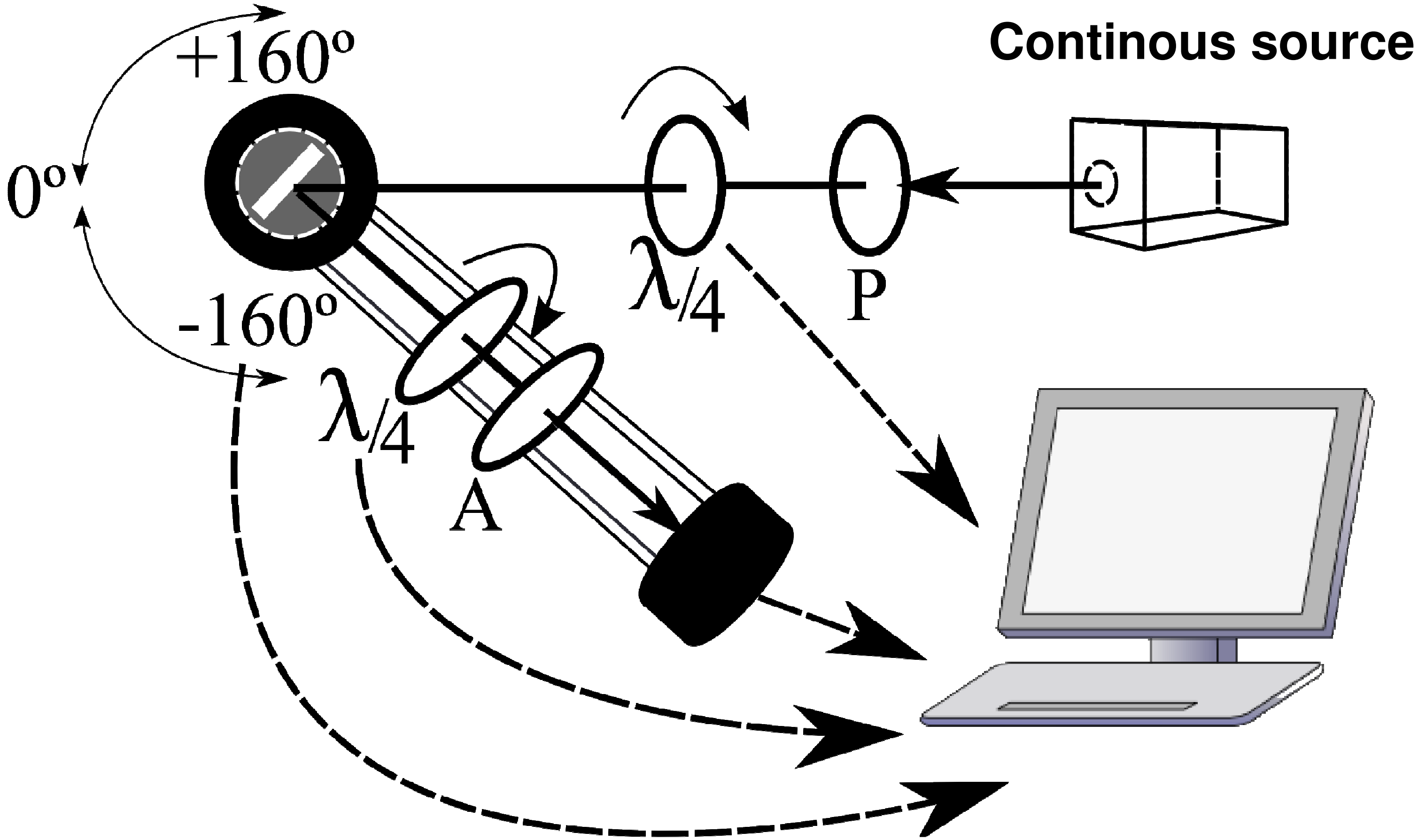
Polarimeter $\longrightarrow$ $M_{4x4} = \begin{pmatrix} m_{11} & m_{12} & m_{13} & m_{14} \\ m_{21} & m_{22} & m_{23} & m_{24} \\ m_{31} & m_{32} & m_{33} & m_{34} \\ m_{41} & m_{42} & m_{43} & m_{44} \end{pmatrix}$

$$\boxed{m_{i\,j}(\theta,\lambda)}$$

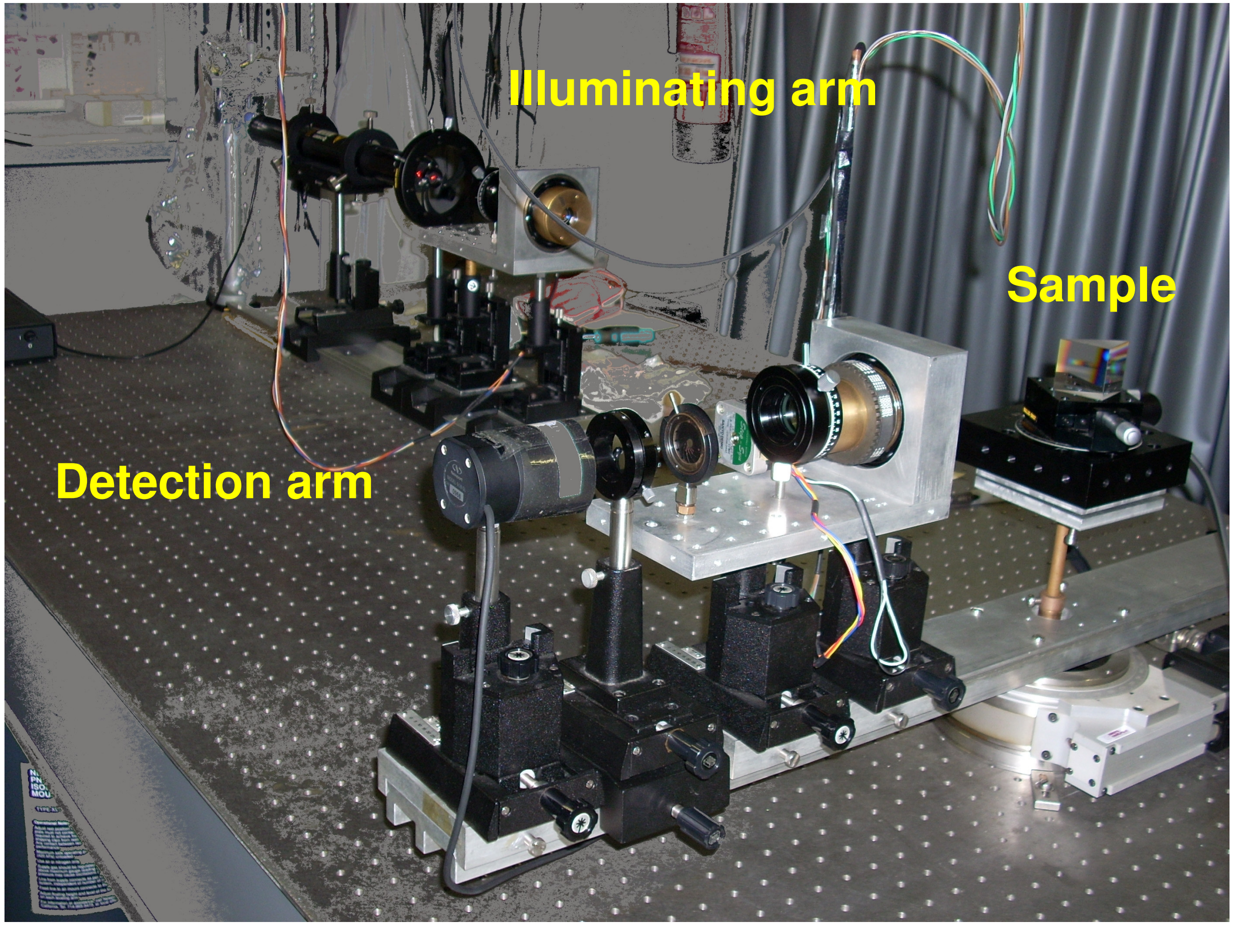
-Polar decomposition: *diatenuator, retarder and depolarizator*

$$\boldsymbol{M_{4x4} = I_{total}\, M_{\Delta}\,(d_1, d_2, d_3)\; M_R\,(\varphi, \delta, \rho)\; M_D\,(\alpha, \beta, t)}$$

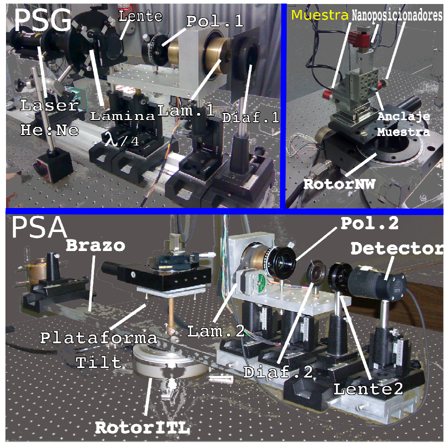
Dynamic polarimetry



Dynamic polarimetry



Some details

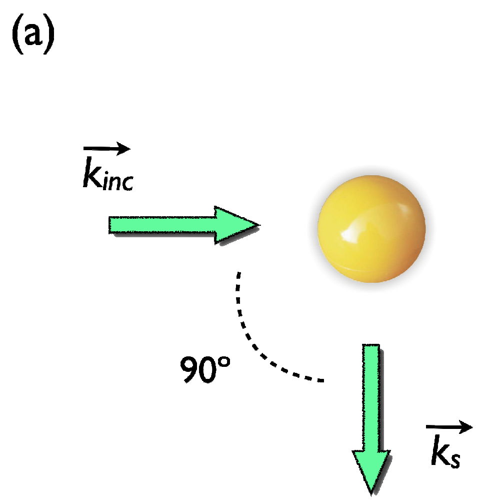


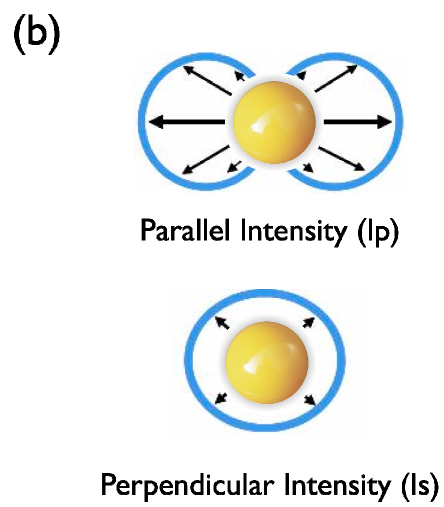
Objective

- Introduction of a complementary tool based on detecting behaviours different than the dipolar due to :
  - Size effects
  - Shape effects
  - Multiple scattering effects:
    - Particle-particle: aggregates
    - Particle-substrate
  - Heterogeneities: Nanoshells
- What's the complementary tool?

Linear polarization degree of scattered light at 90º

$$P_L = -\frac{m_{12}}{m_{11}} = \frac{I_S - I_P}{I_S + I_P}$$

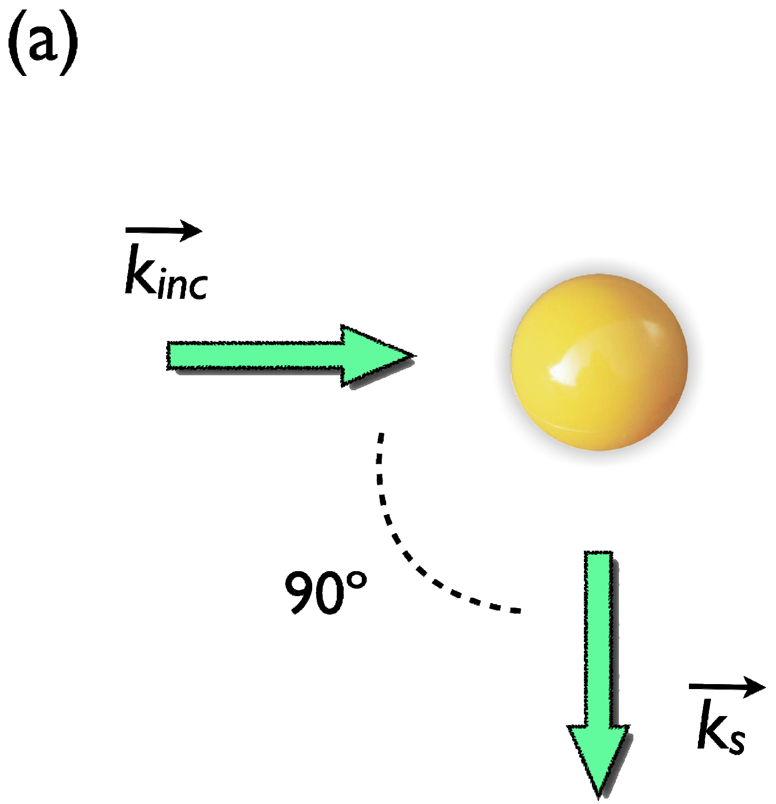


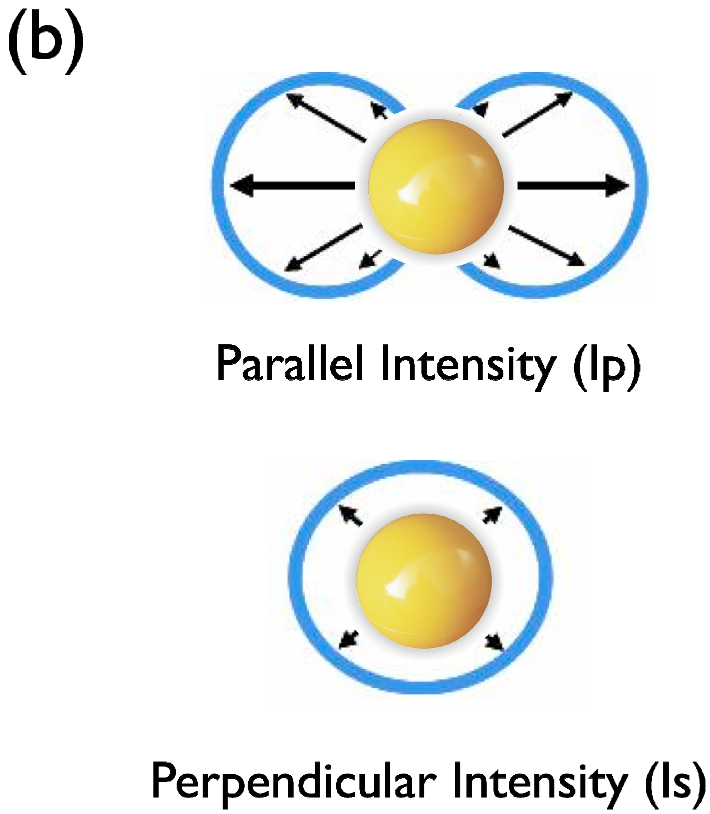


$$P_L = \begin{cases} 1 \rightarrow \text{Scattering from a dipole source} \\ < 1 \rightarrow \text{Higher order modes} \end{cases}$$

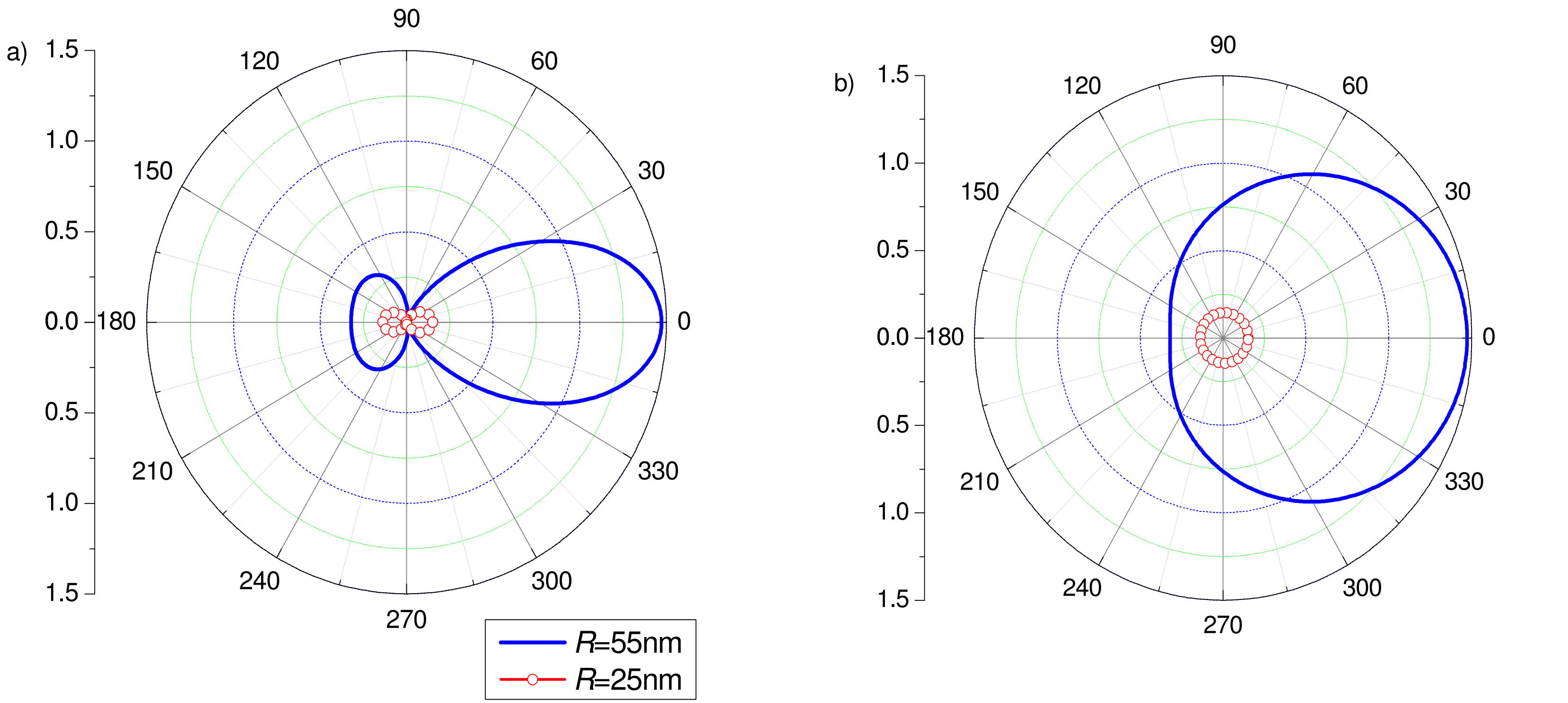
Higher order modes:
- *Particle size and shape effects*
- *Multiple scattering*
- *Anisotropy*

Linear polarization degree of scattered light

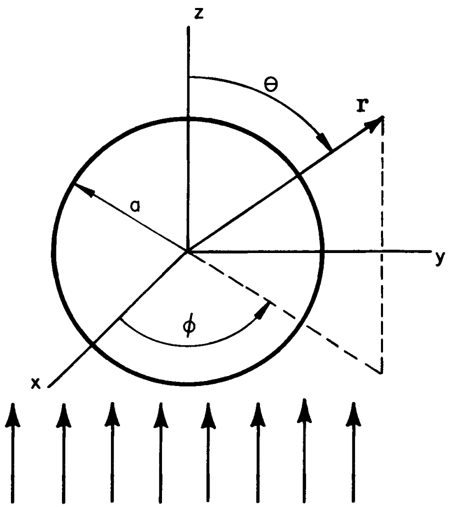




Scattering diagrams (example: Ag)



Spherical particle (Mie theory)



$$\mathbf{E}_s = \sum_{n=1}^{\infty} E_n\left(ia_n\mathbf{N}_{e1n}^{(3)} - b_n\mathbf{M}_{o1n}^{(3)}\right),$$

$$\mathbf{H}_s = \frac{\mathrm{k}}{\omega\mu}\sum_{n=1}^{\infty} E_n\left(ib_n\mathbf{N}_{o1n}^{(3)} + a_n\mathbf{M}_{e1n}^{(3)}\right),$$

$$a_n = \frac{\mu m^2 j_n(mx)[xj_n(x)]' - \mu_1 j_n(x)[mxj_n(mx)]'}{\mu m^2 j_n(mx)\left[xh_n^{(1)}(x)\right]' - \mu_1 h_n^{(1)}(x)[mxj_n(mx)]'},$$

$$b_n = \frac{\mu_1 j_n(mx)[xj_n(x)]' - \mu j_n(x)[mxj_n(mx)]'}{\mu_1 j_n(mx)\left[xh_n^{(1)}(x)\right]' - \mu h_n^{(1)}(x)[mxj_n(mx)]'},$$

Scattering cross sections

$$C_{\mathrm{sca}} = \frac{W_s}{I_i} = \frac{2\pi}{\mathrm{k}^2} \sum_{n=1}^{\infty} (2n+1)\left(|a_n|^2 + |b_n|^2\right).$$

$$C_{\mathrm{ext}} = \frac{W_{\mathrm{ext}}}{I_i} = \frac{2\pi}{\mathrm{k}^2} \sum_{n=1}^{\infty} (2n+1)\mathrm{Re}\{a_n + b_n\},$$

$$C_{abs} = C_{ext} - C_{sca}$$

<u>Resonances</u>: Zeros of denominators of a_{n} and b_n

Small particles ($\mu = 1$)

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$$a_1 = -\frac{i2x^3}{3}\frac{\varepsilon-1}{\varepsilon+2} - \frac{i2x^5}{5}\frac{(\varepsilon-2)(\varepsilon-1)}{(\varepsilon+2)^2} + O(x^7)$$

$$b_1 = -\frac{ix^5}{45}(\varepsilon-1) + O(x^7)$$

$$a_2 = -\frac{ix^5}{15}\frac{\varepsilon-1}{2\varepsilon+3} + O(x^7)$$

$$b_2 = O(x^7)$$

Dipolar Resonance ε = -2

Quad. Resonance ε = -1.5

Small particles ($\mu \neq 1$)

(metamaterials)

$$a_1 = -\frac{i2x^3}{3}\frac{\varepsilon-1}{\varepsilon+2} - \frac{i2x^5}{5}\frac{(\varepsilon-2)(\varepsilon-1)}{(\varepsilon+2)^2} + O(x^7)$$

$$b_1 = -\frac{i2x^3}{3}\frac{\mu-1}{\mu+2} - \frac{ix^5}{45}(\varepsilon-1) + O(x^7)$$

$$a_2 = -\frac{ix^5}{15}\frac{\varepsilon-1}{2\varepsilon+3} + O(x^7)$$

$$b_2 = -\frac{ix^5}{15}\frac{\mu-1}{2\mu+3} + O(x^7)$$

Dipolar Resonance ε= -2

Quad. Resonance ε= -1.5

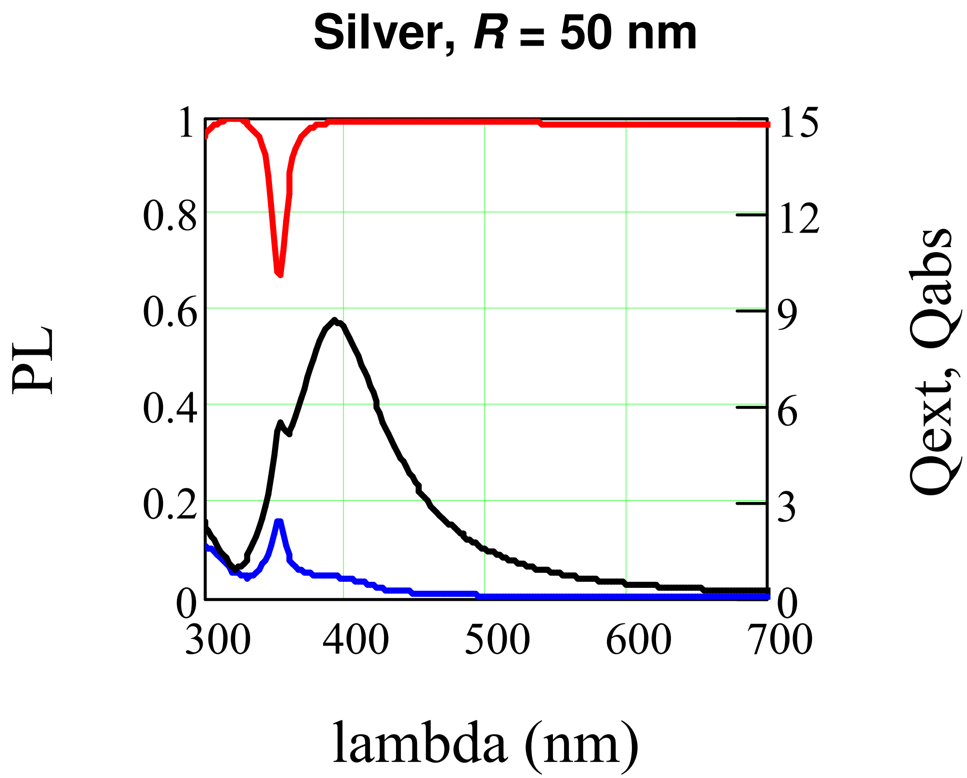
P_L for small particles

$$\begin{pmatrix} E_P^{(s)} \\ E_S^{(s)} \end{pmatrix} = \frac{e^{ik(r-z)}}{-ikr} \begin{pmatrix} S_2 & 0 \\ 0 & S_1 \end{pmatrix} \begin{pmatrix} E_P^{(i)} \\ E_S^{(i)} \end{pmatrix}$$

$$\theta = 90^{\circ}$$

$$S_1 = 1.5a_1 - 2.5b_2$$
$$S_2 = 1.5b_1 - 2.5a_2$$

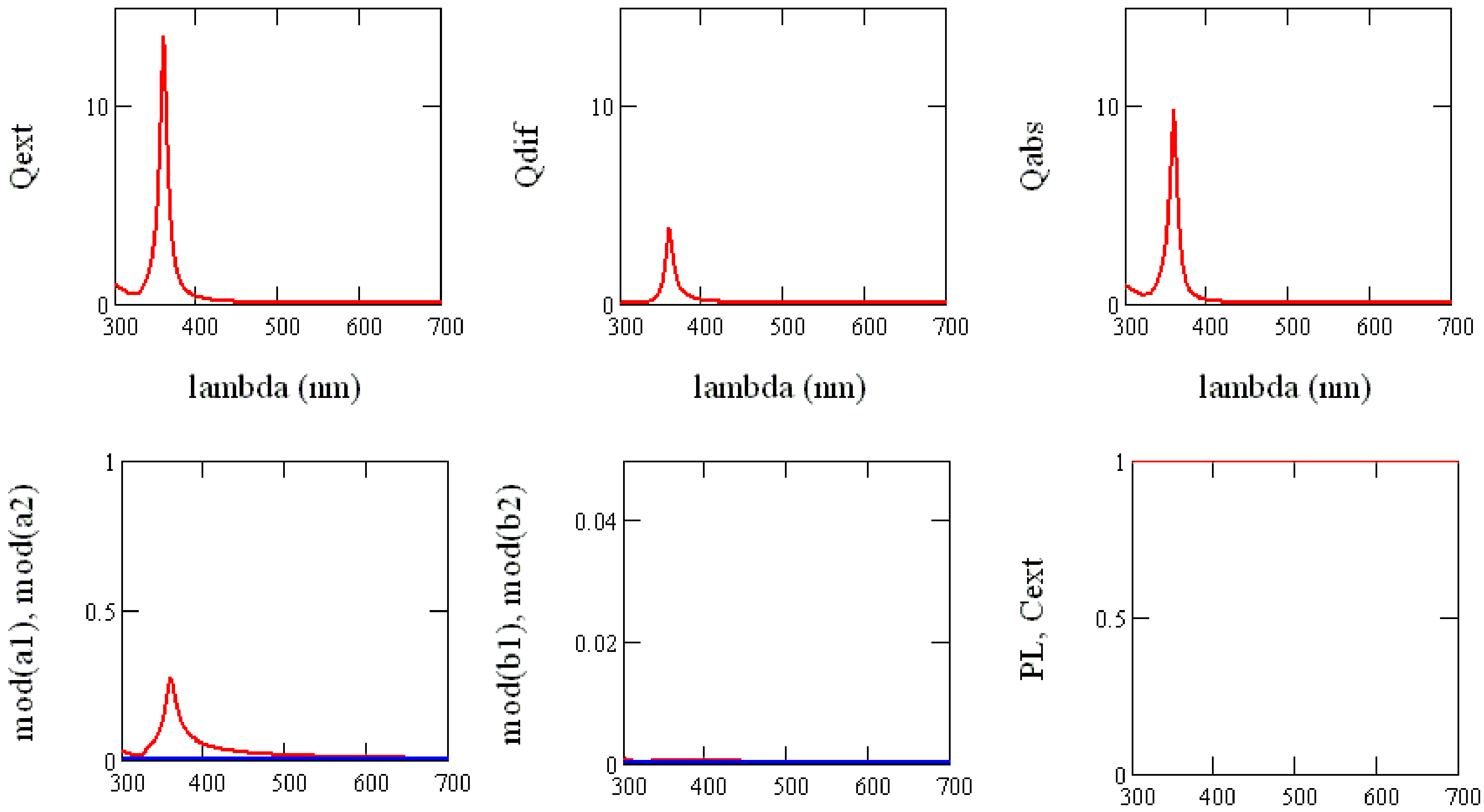
$$P_L = \frac{|S_1|^2 - |S_2|^2}{|S_1|^2 + |S_2|^2}$$



Red: P_L, Black: Q_{ext}, Blue: Q_{abs}

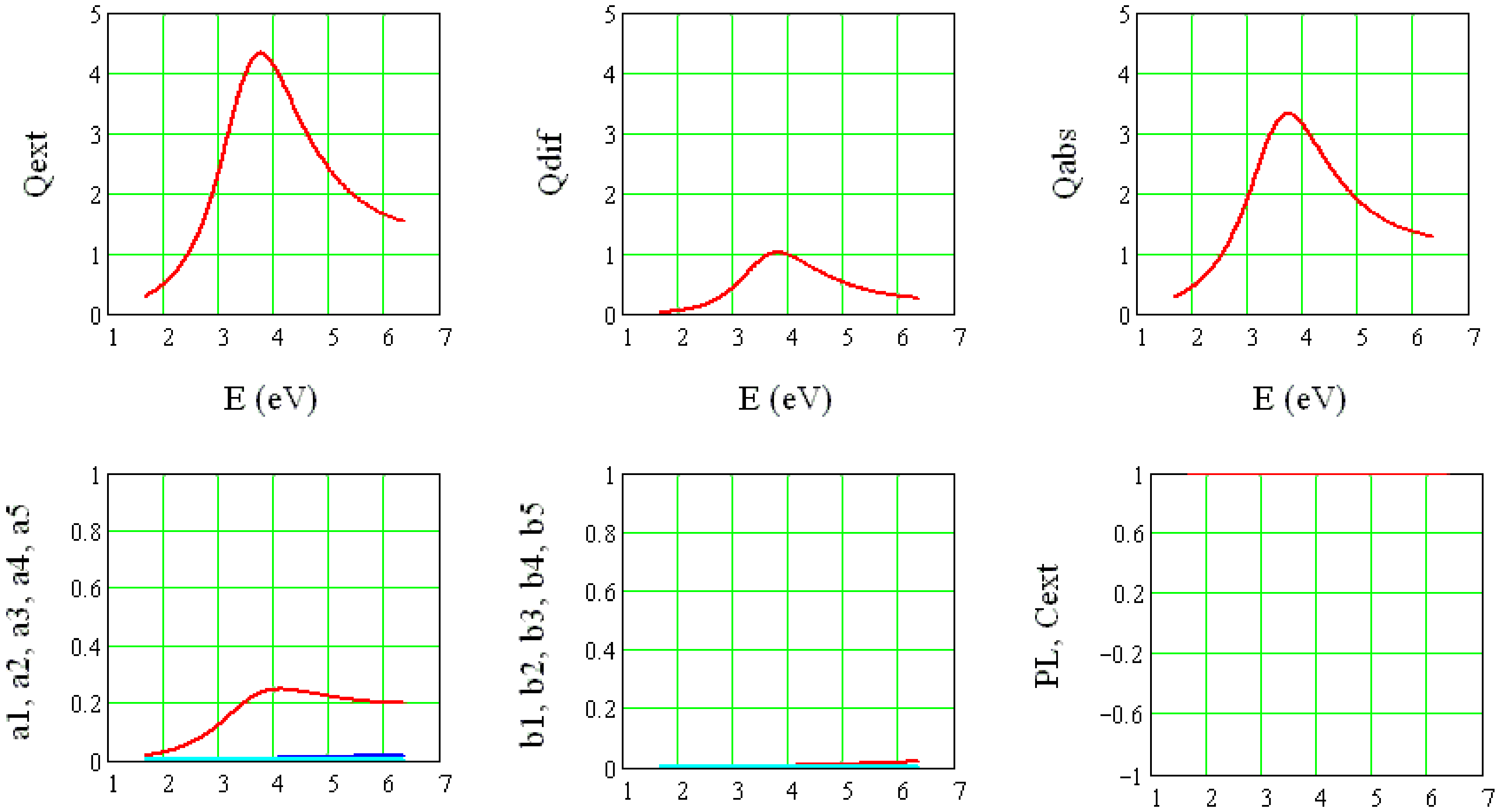
P_L for small particles (Ag)

Ag, R=20-54 nm

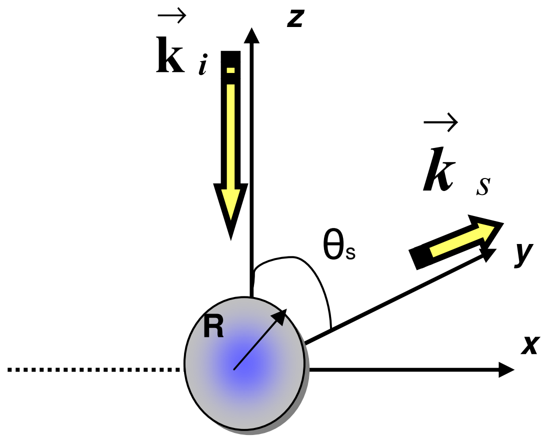


P_L for small particles (Ga)

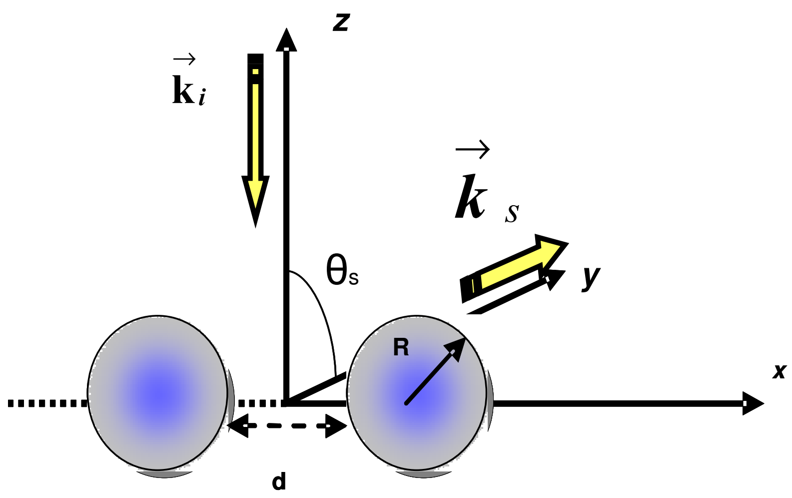
Ga, R=30 to 62 nm



Geometrical model (aggregates)

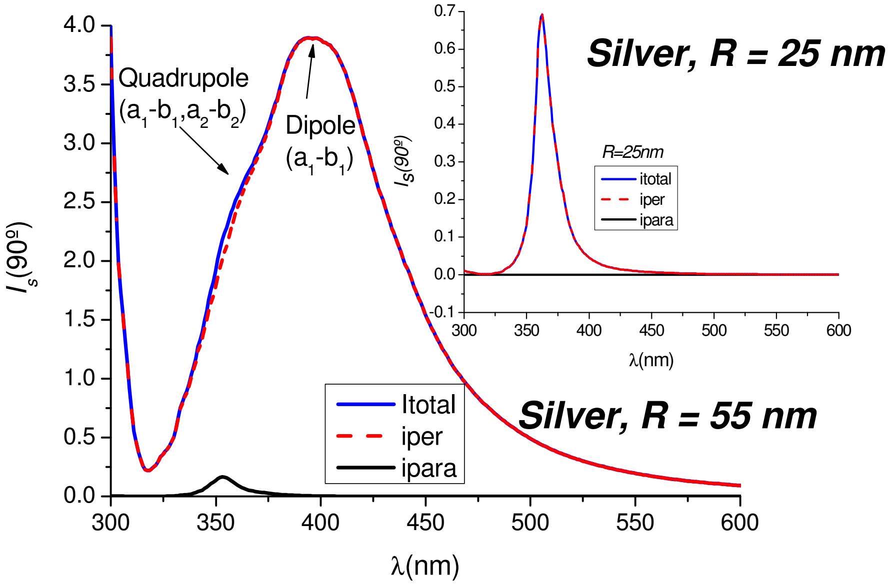


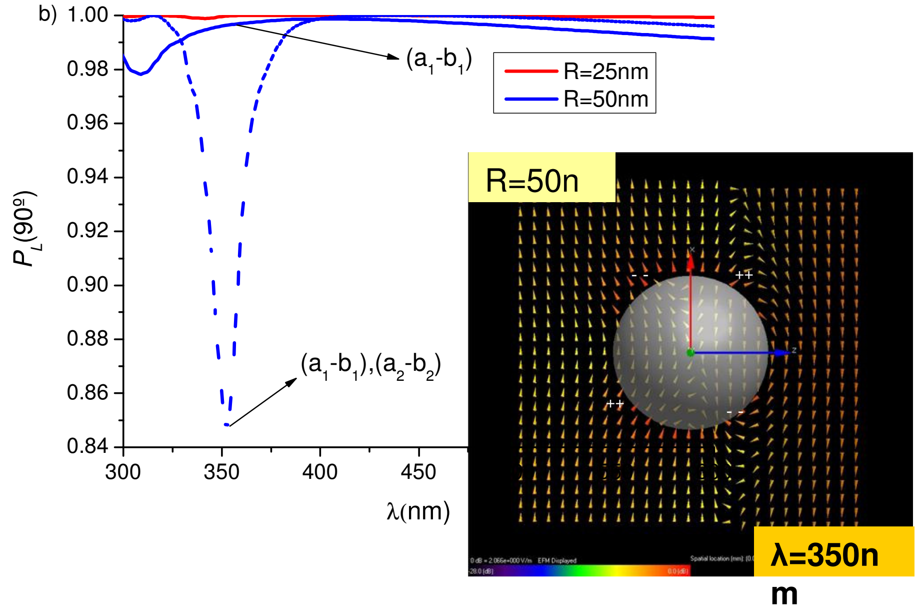
Case Isolated Particle



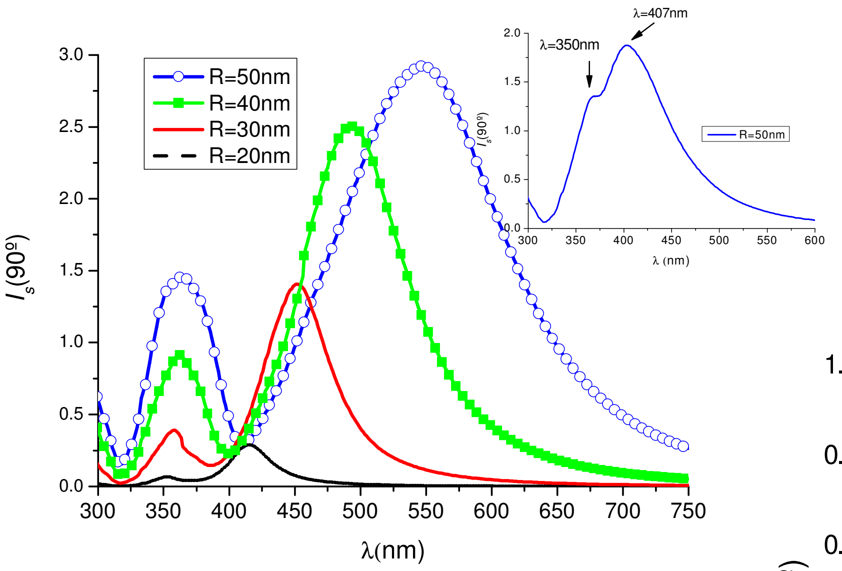
Case Dimer

P_L for isolated particles

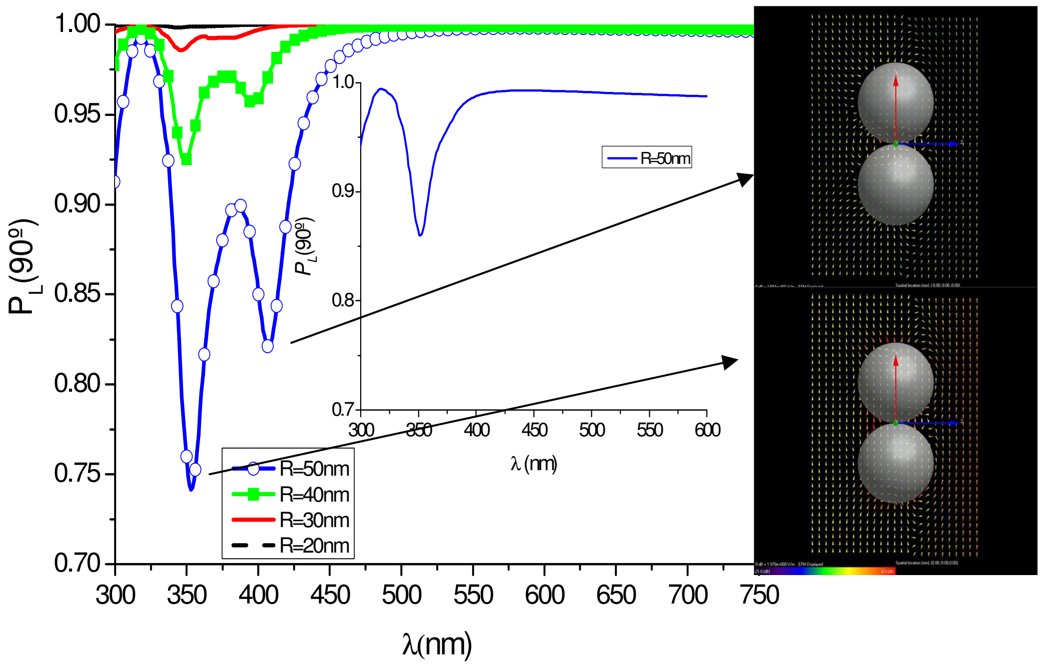




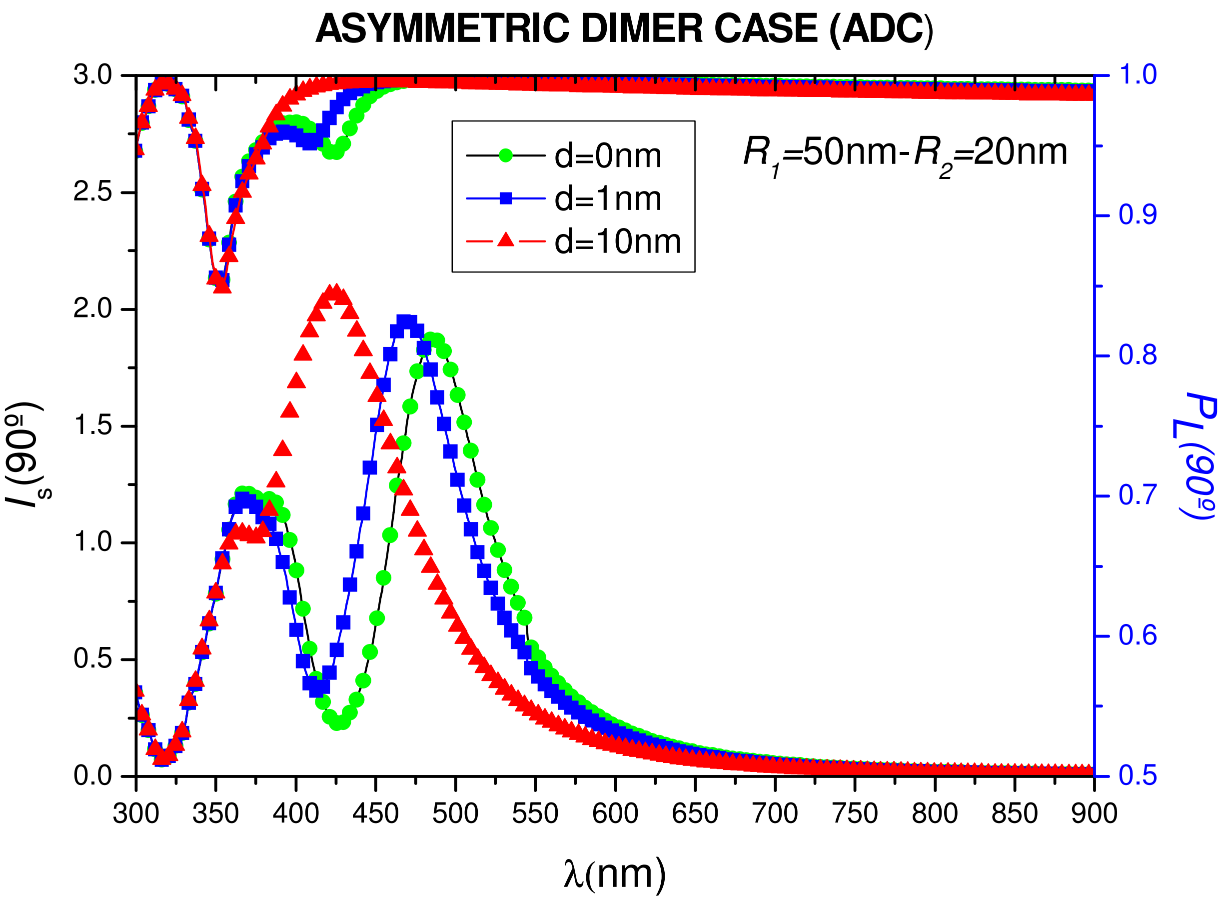
Nanoparticle dimer



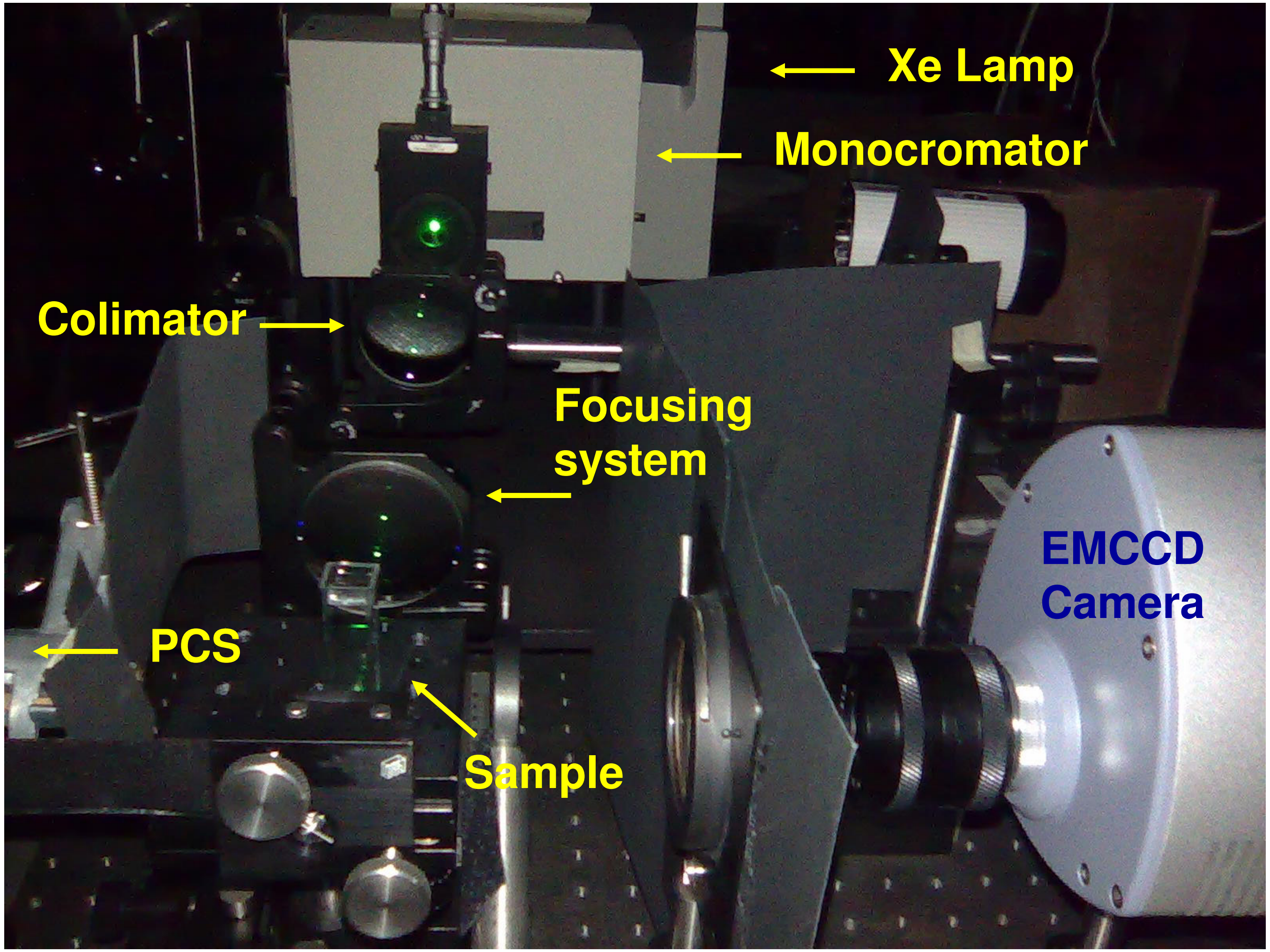
Symmetric dimer
d = 2-3 nm



Nanoparticle dimer



Experimental set-up



Nanoparticles on/above substrates

- Nanotools for spectroscopy and biosensors
- Solar cells and photodetection enhancement
- Optical communications: nanocircuits
- Surface monitoring: Plasmon optical microscopy

Conclusions

- Mueller Matrix (polarimetry) : powerful complementary method to conventional plasmon spectroscopy
- Linear polarization degree: $P_L(90^o,\lambda)$
- Information: size, shape, multiple scattering
- Implementation of the method in different applications: spectroscopy, biosensing, etc.

Future work

- Application of the determination of P_L
  - Magnetic particles
  - Nanoshells and particles with inclusions (nanoeggs)
  - Particles on magnetic substrates (MOKE)
  - Nanoparticles with unconventional optical properties
- Advances in numerical methods: DDA and DDSURF (Kirchoff integrals)
- Experimental verifications

THANKS A LOT FOR YOUR ATTENTION.

ANY QUESTION?