Plasmon modes characterization in hybrid noble/ferromagnetic nanodisks using an effective medium approximation

C A Herreño-Fierro¹, and M Zapata-Herrera¹
¹Grupo de Instrumentación Científica y Didáctica, Universidad Distrital Francisco José de Caldas, Bogotá, Colombia
E-mail: caherrenof@udistrital.edu.co

Abstract. Magneto-plasmonic systems composed by noble/ferromagnetic metals structures can be used to control light polarization. The extraordinary optical properties arising from combining strong local enhancements of electromagnetic fields in surface plasmon excitations with the magneto-optical activity inherent to ferromagnetic materials, can be controlled by external magnetic fields that have demonstrated the possibility to control and amplify the magneto-optical properties via plasmonic excitations. In this work, the anisotropic optical and ellipsometric sensitivity to dielectric environment of multi-layered hybrid gold/cobalt magnetoplasmonic nanodisks are studied in the framework of the effective-medium approximation, where the components of the dielectric tensor of the system are modelled using Lorentz-like oscillators. Furthermore, full electromagnetic simulations were performed using the standard Finite Element Method, which allow us to characterize the modes and explore the role of the dielectric environment of the nanodisks in the optical properties of the nanostructures via the variation of a coating silicon dioxide (SiO₂) layer thickness. Our model and numerical results show a very good agreement with spectral ellipsometry measurements, where two orthogonal plasmonic modes one in-plane and one out-of-plane- are well characterized. These results lead us to conclude that the effective-medium approximation is good enough to accurately describe the plasmonic behavior of multi-layered confined nanostructures.

1. Introduction
Nanotechnology is a relatively new area of research due to the technical challenges it poses in terms of fabrication and characterization of such tiny structures. In this context, several branches of knowledge come together. Plasmonics is one of them. An electromagnetic field such as visible light impinging on a nanoscale system can excite a plasmon resonance. These resonant oscillations of the electron gas in certain metals induce a very strong electric field in the vicinity of the nanostructures through the so-called light confinement. They store the electromagnetic energy of the incoming radiation in the near field, which gives raise to, interesting interactions that modify the optical properties of the material. Plasmonics aims to study the creation and control of these resonances and systems so small that allow the excitation of plasmons in the visible spectrum of light [1,2]. Exploiting features like the high sensitivity to surrounding media of the resonances, one can think of ultrasensitive devices that look for the presence of certain molecules, for example [3]. One of these possible interactions relates plasmonics to magneto-optics. Some materials, namely ferromagnetic metals, show the ability of changing the polarization of the light that transmits through or reflects from them when a magnetic field magnetizes
the samples [4-7]. These are the magnetic Faraday and Kerr effect, respectively. It is intuitive to think that an enormous electromagnetics (EM) field concentrated in such a small space due to a plasmon resonance will modify, and hopefully enhance, these properties that can be applied to create optical devices such as rotators [8,9] in a form factor that was not possible years ago, creating the research area of flat optics for instance [10]. In this work we describe experimental and theoretically the sensitivity to the dielectric environment behavior of gold/cobalt (Au/Co) magnetoplasmonic nanodisks under the excitation of light based on ellipsometric measurements for the optical characterization and using a simple effective-media approximation model and electromagnetic simulation from the theoretical point of view. The work is arranged as follows: In the first part, we describe the system and experimental setup, then we show the plasmon modes characterization in the effective-media approximation followed by the simulation counterpart and finally we present a general discussion.

2. Experimental setup, magnetic, optical and magnetooptical characterization

The system under consideration is an array of three-layered nanodisks of gold and cobalt, randomly distributed on a glass substrate. The 10 nm thick cobalt layer is sandwiched between two gold layers of 16 nm (bottom) and 6 nm (top) thick, respectively. One 2 nm thick titanium layer is used to improve the structure addition to the glass substrate. Since the systems was growth using colloidal lithography, the actual shape of the structures is not that of cylindrical disks but truncated cones, as illustrated in Figure 1(a). Further details on the growth process and characterization of this system are given in our previous work [11]. There, a comprehensive analysis on the sensitivity to dielectric environment of the optical, magnetooptical, and ellipsometric measurements of this system was reported. The analysis was based on the evolution of such measurements against the thickness of the conformally silicon dioxide ($\text{SiO}_2$) coating deposited on the disks, from 0 nm (nude disks) to 30 nm thick. As shown in the Atomic Force Microscope image of Figure 1(b), the average bottom diameter of the nude nanodisks is 176 nm and the average distance between centers is about 300 nm, which allows us to consider the disks as isolated structures of negligible interaction between them. Their magnetic activity is demonstrated in the hysteresis loop of the magnetooptical signal obtained by means of magneto-optical Kerr effect (MOKE) experiments in the transversal geometry (see reference [11] for details) as is shown in Figure 1(c).

Figure 1. (a) Schematically draw of the system composed by magnetoplasmonic nanodisks made of Au/Co/Au layers (b) Atomic force microscopy (AFM) topography of the nude nanodisk and (c) magnetic hysteresis loop of the system obtained by transverse magnetooptical Kerr effect experiment.

In Figure 2(a) the optical transmission spectrum is shown. It reveals a deep at a wavelength ($\lambda$) close to 730 nm, related to the excitation of a dipolar surface plasmon mode. This behavior has been widely reported for this kind of nanostructures. This plasmonic effect can also be observed in the magnetooptical measurements ($\Delta R/R$) where a pick appears at the same wavelength, related to the surface enhancement of the transverse magnetooptical Kerr effect, as shown in Figure 2(b). Remarkably, the ellipsometric measurements, concerning the reflection coefficient ratio of p-polarized to s-polarized waves, confirms the plasmonic effect as can be clearly seen in Figure 2(c). While the ellipsometric amplitude (black line) exhibits a resonant like spectrum with a maximum at 730 nm, the phase difference between p-polarized to s-polarized waves (blue line) takes the value of $\pi$ (~3.14 rad) at the resonant
wavelength. This phase difference is indicative of a resonant phase shift of $+\frac{\pi}{2}$ and $-\frac{\pi}{2}$ for the p-mode to s-mode, respectively, with respect to the reference signal.

Figure 2. Transmission (a) magneto-optic (b) and ellipsometric (c) spectra for the nude nanodisks system. Evolution of the transmission (d) magneto-optic (e) and ellipsometric (f) and (g) spectra as a function of the SiO$_2$ coating thickness for 0 nm (nude disks), 10 nm and 30 nm coating.

The evolution of these four measurement spectra under thickening the SiO$_2$ coating from 0 (nude disks) to 30 nm thick is shown in Figure 2(d), Figure 2(e), Figure 2(f) and Figure 2(g), respectively. All of them exhibit the well-known redshift related to the broader coupling of the surface plasmons to the dielectric-metal structure. Nevertheless, there exists important differences in sensitivity of each measurement to changes in the dielectric environment; issue broadly discussed in [11].

3. Plasmon modes characterization in the effective-media approximation

Since the wavelength range of the incident light considered is always much larger than the average size of the disks, the incident light does not "detect" individual disks but an effective medium consisting of a mixture of multilayer nanodisks “plus” the interstitial environment that separates them from each other, as illustrated in Figure 3(a). Therefore, for the geometry illustrated in Figure 3(a), and uniquely concerning the optical response of the system, it is possible to consider an effective medium approach (EMA) [12,13] in which the system is represented as a uniform layer of thickness $d$ characterized by an effective dielectric tensor given in rectangular coordinates by Equation (1).

$$
\tilde{\varepsilon}_{\text{eff}} = \begin{pmatrix}
\varepsilon_0 & 0 & 0 \\
0 & \varepsilon_0 & 0 \\
0 & 0 & \varepsilon_0
\end{pmatrix},
$$

(1)

where, due to the cylindrical geometry of the disks, the optical response of the system depends on the polarization of the incoming light, describing uniaxial anisotropy. Superscript $||$ labels the optical response for the in-plane (x-y plane) component of electric field of light, while subscript $\perp$ is reserved for the out-of-plane component (z axis).

Using Lorentz like oscillators to model the light-matter interaction under this approach, the diagonal elements of $\tilde{\varepsilon}_{\text{eff}}$ are considered to be described as Equation (2).

$$
\varepsilon_0^\alpha = \varepsilon_0^\alpha + \frac{a_1^\alpha b_1^\alpha E_{c,1}^\alpha}{(E_{c,1}^\alpha)^2 - E^2 + ib_1^\alpha E} + \frac{a_2^\alpha b_2^\alpha E_{c,2}^\alpha}{(E_{c,2}^\alpha)^2 - E^2 + ib_2^\alpha E},
$$

(2)
with $\alpha = ||L||$, where the energy of the incident photons of wavelength $\lambda$ is given by $E = \hbar c/\lambda$, being $h$ and $c$ the Plank’s constant and the speed of light, respectively. The central energy of the $j$-th oscillator $(j = 1, 2)$ is described by $E_c^j$, while $a_j^s$ and $b_j^s$ represent, correspondingly, the amplitude and width of the resonance peak. In this model, the high frequency dielectric constant is taken as $\epsilon_\infty^a = \epsilon_\infty^b = 1$, while oscillators 1 and 2 $(j = 1, 2)$ are conceived to represent the plasmonic resonance and inter-band transitions contributions, respectively.

![Image](image_url)

**Figure 3.** (a) Schematic description of the effective medium approximation (EMA) and geometry of the system. Transmission (b), ellipsometric amplitude (c), and ellipsometric phase (d) measurements (solid line) and theoretical fit (dashed line) in the EMA.

The solid lines in Figure 3 represent the transmission (see Figure 3(b)) and ellipsometry (see Figure 3(c) and Figure 3(d)) spectra measurements for the nude (no-coated) case. With the purpose to contrast the optical response under total reflection condition, the ellipsometric measurements (Figure 3(c) and Figure 3(d)) were obtained with two different impinging light angles: 50 degrees (black lines) and 60 degrees (blue lines), which are below and above the critical angle, respectively. Dashed lines correspond to the theoretical model described by Equation (2) after fitting the parameters to the experimental curves, showing a very good agreement. The fitting process is carried out in two steps: first, we fit $\epsilon^a$ using only the transmission data for the electric field of light in-plane with respect to the nanodisks; afterwards, the fitted parameters collected in the first step are used as constant values in order to fit the out-of-the-plane component ($\epsilon^b$) to the oblique incidence data. In the latter case, the incoming electric field has both, in-plane, and out-of-the-plane components, due to the its traversal magnetic (p-wave) character. The results of the fitting process, along with the corresponding mean squared error (MSE) are presented in Table 1.

| $j$ | $a$ (eV) | $b$ (eV) | $E_c$ (eV) | MSE |
|-----|---------|---------|-----------|-----|
| $\epsilon^a$ | 1 | 1.016426 ± 0.0063041 | 0.7559 ± 0.01055 | 1.648 ± 0.002706 | 0.00313 |
| 2 | 0.168258 ± 0.0054365 | 0.9153 ± 0.11198 | 2.952 ± 0.036402 |
| $\epsilon^b$ | 1 | 0.095990 ± 0.0015050 | 0.3339 ± 0.01077 | 1.496 ± 0.002294 | 5.297 |
| 2 | 0.043011 ± 0.0007046 | 3.5916 ± 0.19142 | 3.176 ± 0.032501 |

**4. Discussion**

Our results allow us to compute the complex effective refractive index of the system defined as $N = \sqrt{\epsilon_{\text{eff}}} = n + ik$ as we shown in Figure 4. The real part, $n$, (Figure 4(a)) exhibits a strong uniaxial anisotropy. Meanwhile, the imaginary part, $k$, (Figure 4(b)) shows a resonant like peak at 730 nm for the in-plane response $k_\parallel$ (black line), associated with the excitation of a localized surface plasmon mode,
and another one more energetic and less intense peak at 450 nm, associated with inter-band transitions. These two phenomena are also identified in the out-of-plane response $k_{\parallel}$ (blue line), where the plasmonic excitation in this case is less energetic and weaker, and the inter-band transitions peak is much weaker and broader. Under SiO$_2$ coating thickening of the nanodisks, the $k_{\parallel}$ spectrum exhibits a redshift as shown in Figure 4(c), for 10 nm and 30 nm thick coating in contrast to the nude (no coating) nanodisks. As discussed before in section 1, this well-known behavior is related to the broadening of the coupling of the surface plasmons to the dielectric-metal structure, by which the resonant energy of the system is reduced [14].

![Figure 4. Real (a) and imaginary (b) part of the effective refractive index of the nude system, for the in-plane (black) and out-of-the-plane (blue) modes. (c) Evolution of the imaginary part of the effective refractive index as the thickness of the SiO$_2$ coating on the nanodisks increases from zero (black) to 30 (light blue) nm. (d) Representation of the impinging linearly polarized light at a 60° angle of incidence. (e) Optical response of the nanodisk in terms of the spectral extinction cross section for the nude (red) as well as for the coated disk varying the SiO$_2$ coating from 15 (blue) to 30 (green) nm thick. The optical response at normal incidence (magenta) is include for reference. Density charge maps of the in-plane (f) and out-of-the-plane plasmonic modes (g).](image-url)

For further analysis on the nature of the plasmonic modes and their evolution under dielectric environment changes, we have developed numerical simulations to contrast the experimental results previously described. We used the well-known commercial COMSOL Multiphysics package [15]. This kind of electromagnetic simulations allow us to characterize the excited modes with different angles of incidence of the impinging light as describe in Figure 4(d). In order to obtain this computational experiment, we used the radio frequency-electromagnetic wave module in the frequency domain.
configuration with scattering boundary conditions. The physical domain was taken as a rectangular prism surrounded of perfectly matched layers (PML’s) to prevent reflections. The model resizes according to the incoming wave parametrized by wavelength $\lambda$ to ease computation load. PML layers are swept-meshed with quadrilateral elements.

The rest of the geometry, where our system of disks is placed is discretized with free triangular elements, as well as the air and the SiO$_2$ substrate block with a $\lambda/6$ maximum size. To perform the optical description of the heterostructure in terms of the incoming light wavelength, we calculated the absorption ($\sigma_{\text{abs}}$) and scattering ($\sigma_{\text{sc}}$) cross-sections, computed by integrating, over an enclosure ellipsoid surrounding the disk, the total and the relative Poynting vector over, as is widely used in this kind of problems. Extinction cross section ($\sigma_{\text{ext}}$), collecting the information about the optical response of the system, was calculated using $\sigma_{\text{ext}} = \sigma_{\text{abs}} + \sigma_{\text{sc}}$. Optical constants were introduced through interpolation of tabulated data available in the literature [16]. Computation time varies depending on the incoming field’s wavelength, although not abruptly since the finely meshed area remains untouched, averaging 20 minutes per spectral point in our equipment. The experiment was run on a Lenovo Think Station equipped with an Intel R Xeon R CPU ES- 1650 v3 at 3.5 GHz and 128Gb of RAM memory running Windows R 7 Professional (Service Pack 1) and COMSOL Multiphysics version 5.2.

The optical response Figure 4(e) reveals the excitation of two plasmonic modes which are related to one dipolar mode parallel to the disk plane (close to 780 nm for the bare disk) that acts as a dipole and is reemitted into the far field as is expected in our experiment. A more energetic second one (close to 450 nm for the bare disk, lower wavelength) and less intense peak is weakly excited and appears because of the perpendicular contribution of the incoming light with respect to the disk’s symmetry axis. The nature of these modes is depicted in Figure 4(f) and Figure 4(g), respectively, where the density charge maps are shown. Experimental and simulation comparison allow us to elucidate a very good agreement regarding the spectral red shift for different amounts of coating layer in each case, demonstrating the extreme plasmonic resonance sensitive dependence on the dielectric constant of the surrounding medium for both, parallel and perpendicular excited modes. Further studies need to be done in order to find a mechanism which allows to geometrically manipulate these two degrees of freedom routed to design sensor devices based on magnetoplasmonic systems.

5. Conclusions

In summary, we have studied and modelled the anisotropic optical, ellipsometric, and magneto-optical surface sensitivity to the dielectric environment of a system built of multi-layered hybrid Au/Co magnetoplasmonic nanodisks, using ellipsometric measurements and theoretically analyzed by means of the effective-medium approximation and comparing with electromagnetic numerical simulations. Our results let us to identify two perpendicular plasmonic modes: one in-plane and a weaker one out-of-plane of the structure. By this means, we also obtained the diagonal elements of the effective-dielectric tensor, in which a strong uniaxial anisotropy is identify. Our study could be important for further design and manufacturing of hybrid metal-magnetic nanostructures, where the high tunability geometrical dependence on the optical response is exploited. As a novelty, the study and characterization presented in this work, allow us to show that a simple model using two Lorenz oscillators, is sufficiently enough to describe the optical response of this kind of systems. We had also found that this model provides a good description of the optical response of noble/ferromagnetic nanodisks used as a complementary technique and/or in combination with electromagnetic solvers.

Acknowledgements

This work was supported by the Research Center CIDC of “Universidad Distrital Francisco José de Caldas” and the “Departamento Administrativo de Ciencia, Tecnología e Innovación de Colombia (Colciencias)” under the “Convocatoria Programa de Estancias de Postdoctorales 784-2017” program. We expressly thank G. Armelles and A. Cebollada from “Instituto de Micro y Nanotecnología, Madrid”, Spain, for the support and orientation in performing the experiments here cited in section 2, and their contribution and enlightening discussion regarding the analysis presented in section 3.
References

[1] Maier S A 2007 Plasmonics: Fundamentals and Applications (United Kingdom: Springer Science & Business Media)

[2] Cai W and Shalaev V 2010 Negative-index metamaterials Optical Metamaterials ed Cai W and Shalaev V (New York: Springer) pp 101-122

[3] Ahmadivand A, Gerislioglu B, Manickam P, Kaushik A, Bhansali S, Nair M and Pala N 2017 Rapid detection of infectious envelope proteins by magnetoplasmonic toroidal metasensors ACS sensors 2(9) 1359

[4] Belotelov V I, Akimov I A, Pohl M, Kotov V A, Kasture S, Vengurlekar A S and Bayer M 2011 Enhanced magneto-optical effects in magnetoplasmonic crystals Nature Nanotechnology 6(6) 370

[5] Belotelov V I, Doskolovich L L and Zvezdin A K 2007 Extraordinary magneto-optical effects and transmission through metal-dielectric plasmonic systems Physical Review Letters 98(7) 077401

[6] Sepúlveda B, Calle A, Lechuga L M and Armelles G 2006 Highly sensitive detection of biomolecules with the magneto-optic surface-plasmon-resonance sensor Optics Letters 31(8) 1085

[7] Armelles G, Cebollada A, García-Martín A and González M U 2013 Magnetoplasmonics: Combining magnetic and plasmonic functionalities Advanced Optical Materials 1(1) 2

[8] Lodewijks K, Maccaferri N, Pakizeh T, Dumas R K, Zubritskaya I, Åkerman J and Dmitriev A 2014 Magnetoplasmonic design rules for active magneto-optics Nano Letters 14(12) 7207

[9] González-Díaz J B, Garcia-Martín A, Garcia-Martín J M, Cebollada A, Armelles G, Sepúlveda B and Käll M 2008 Plasmonic Au/Co/Au Nanosandwiches with enhanced magneto-optical activity Small 4(2) 202

[10] Maccaferri N, Bergamini L, Pancaldi M, Schmidt M K, Kataja M, Dijken S V and Vavassori P 2016 Anisotropic nanoantenna-based magnetoplasmonic crystals for highly enhanced and tunable magneto-optical activity Nano Letters 16(4) 2533

[11] Herreño-Fierro C A, Patiño E J, Armelles G and Cebollada A 2016 Surface sensitivity of optical and magneto-optical and ellipsometric properties in magnetoplasmonic nanodisks Applied Physics Letters 108(2) 021109

[12] Aspnes D E, Theeten J B and Hottier F 1979 Investigation of effective-medium models of microscopic surface roughness by spectroscopic ellipsometry Physical Review B 20(8) 3292

[13] Fujjwara H, Koh J, Rovira P I and Collins R W 2000 Assessment of effective-medium theories in the analysis of nucleation and microscopic surface roughness evolution for semiconductor thin films Physical Review B 61(16) 10832

[14] Homola J and Piliarik M 2006 Surface plasmon resonance (SPR) sensors Surface Plasmon Resonance Based Sensors ed Homola J (Berlin: Springer)

[15] COMSOL Group 2018 COMSOL Multiphysics® 5.2, Stockholm (United State: COMSOL Group)

[16] Johnson P B and Christy R W 1972 Optical constants of the noble metals Physical Review B 6(12) 4370