Localized surface plasmon resonance effects on the magneto-optical activity of continuous Au/Co/Au trilayers

Gaspar Armelles[1,*], Juan Bautista González-Díaz[1], Antonio García-Martín[1], José Miguel García-Martín[1], Alfonso Cebollada[1], María Ujué González[2], Srdjan Acimovic[2], Jean Cesario[2,3], Romain Quidant[2,4] and Gonçal Badenes[2]

[1]*Instituto de Microelectrónica de Madrid (CSIC), 28760-Tres Cantos, Madrid, Spain*
[2]*ICFO-Institut de Ciencies Fotoniques, Mediterranean Technology Park, 08860 Castelldefels (Barcelona), Spain*
[3]*Institut Fresnel, UMR CNRS 6133, Université Paul Cézanne Aix-Marseille III, 13397-Marseille Cedex 20, France*
[4]*ICREA-Institució Catalana de Recerca i Estudis Avançats, 08010 Barcelona, Spain*
**Corresponding author: gaspar@imm.cnm.csic.es*

Abstract: We study how the magneto-optical activity in polar configuration of continuous Au/Co/Au trilayers is affected by the excitation of localized plasmon resonances of an array of Au nanodiscs fabricated on top of them over a dielectric SiO_2 spacer. We show that the effect of the nanodiscs array is twofold. First, it optimizes the absorption of light at specific photon energies corresponding to the localized surface plasmon excitation of the array, modifying the reflectivity of the system (we define this effect as the purely optical contribution). Second, upon localized plasmon resonance excitation, the electromagnetic field in the whole system is redistributed, and an enhanced magneto-optical activity occurs at those energies where the electromagnetic field in the magnetic layer is increased (this effect is identified as the purely magneto-optical contribution of the nanodiscs array).

OCIS codes: (240.6680) Surface plasmons; (250.5403) Plasmonics; (350.4238) Nanophotonics and photonic crystals; (160.3820) Magneto-optical materials; (230.3810) Magneto-optic systems.

References and Links

1. C. F. Bohren and D. R. Huffman, *Absorption and Scattering of Light by Small Particles* (Wiley-VCH, Berlin, 1998).
2. U. Kreibig and M. Vollmer, *Optical Properties of Metal Clusters*, Vol. 25 of Springer Series in Materials Science (Springer-Verlag, Berlin, 1995).
3. H. Raether, *Surface Plasmons on Smooth and Rough Surfaces and on Gratings*, Vol. 111 of Springer Tracts in Modern Physics (Springer-Verlag, Berlin, 1988).
4. C. Genet and T. W. Ebbesen, "Light in tiny holes," Nature **445**, 39-46 (2007).
5. K. Kneipp, Y. Wang, H. Kneipp, L. T. Perelman, I. Itzkan, R. R. Dasari, and M. S. Feld, "Single Molecule Detection Using Surface-Enhanced Raman Scattering (SERS)," Phys. Rev. Lett. **78**, 1667-1670 (1997).
6. S. Nie and S. R. Emory, "Probing Single Molecules and Single Nanoparticles by Surface-Enhanced Raman Scattering," Science **275**, 1102-1106 (1997).
7. J. R. Lakowicz, "Radiative decay engineering 5: metal-enhanced fluorescence and plasmon emission," Anal. Biochem. **337**, 171-194 (2005).
8. S. Kim, J. Jin, Y.-J. Kim, I.-Y. Park, Y. Kim, and S.-W. Kim, "High-harmonic generation by resonant plasmon field enhancement," Nature **453**, 757-760 (2008).
9. M. Abe and T. Suwa, "Surface plasma resonance and magneto-optical enhancement in composites containing multicore-shell structured nanoparticles," Phys. Rev. B **70**, 235103 (2004).
10. Y. M. Strelniker and D. J. Bergman, "Optical transmission through metal films with a subwavelength hole array in the presence of a magnetic field," Phys. Rev. B **59**, R12763-R12766 (1999).
11. A. Battula, S. Chen, Y. Lu, R. J. Knize, and K. Reinhardt, "Tuning the extraordinary optical transmission through subwavelength hole array by applying a magnetic field," Opt. Lett. **32**, 2692-2694 (2007).
12. M. Diwekar, V. Kamaev, J. Shi, and Z. V. Vardeny, "Optical and magneto-optical studies of two-dimensional metallodielectric photonic crystals on cobalt films," Appl. Phys. Lett. **84**, 3112-3114 (2004).

Final pre-print — Accepted for publication in Optics Express

13. C. Clavero, G. Armelles, J. Margueritat, J. Gonzalo, M. G. del Muro, A. Labarta, and X. Batlle, "Interface effects in the magneto-optical properties of Co nanoparticles in dielectric matrix," Appl. Phys. Lett. **90**, 182506 (2007).
14. S. Melle, J. L. Menéndez, G. Armelles, D. Navas, M. Vázquez, K. Nielsch, R. B. Wehrspohn, and U. Gösele, "Magneto-optical properties of nickel nanowire arrays," Appl. Phys. Lett. **83**, 4547-4549 (2003).
15. J. B. González-Díaz, A. García-Martín, G. Armelles, D. Navas, M. Vázquez, K. Nielsch, R. Wehrspohn, and U. Gösele, "Enhanced Magneto-Optics and Size Effects in Ferromagnetic Nanowire Arrays," Adv. Mater. **19**, 2643-2647 (2007).
16. C. Hermann, V. A. Kosobukin, G. Lampel, J. Peretti, V. I. Safarov, and P. Bertrand, "Surface-enhanced magneto-optics in metallic multilayer films," Phys. Rev. B **64**, 235422 (2001).
17. J. B. González-Díaz, A. García-Martín, G. Armelles, J. M. García-Martín, C. Clavero, A. Cebollada, R. A. Lukaszew, J. R. Skuza, D. P. Kumah, and R. Clarke, "Surface-magnetoplasmon nonreciprocity effects in noble-metal/ferromagnetic heterostructures," Phys. Rev. B **76**, 153402 (2007).
18. B. Sepúlveda, A. Calle, L. M. Lechuga, and G. Armelles, "Highly sensitive detection of biomolecules with the magneto-optic surface-plasmon-resonance sensor," Opt. Lett. **31**, 1085-1087 (2006).
19. J. B. González-Díaz, A. García-Martín, J. M. García-Martín, A. Cebollada, G. Armelles, B. Sepúlveda, Y. Alaverdyan, and M. Käll, "Plasmonic Au/Co/Au Nanosandwiches with Enhanced Magneto-optical Activity," Small **4**, 202-205 (2008).
20. J. Cesario, R. Quidant, G. Badenes, and S. Enoch, "Electromagnetic coupling between a metal nanoparticle grating and a metallic surface," Opt. Lett. **30**, 3404-3406 (2005).
21. A. Christ, T. Zentgraf, S. G. Tikhodeev, N. A. Gippius, J. Kuhl, and H. Giessen, "Controlling the interaction between localized and delocalized surface plasmon modes: Experiment and numerical calculations," Phys. Rev. B **74**, 155435 (2006).
22. C. Clavero, *Size and matrix effects on the magnetic and magneto-optical properties of magnetic nanoparticles* (PhD Thesis, Universidad Autónoma de Madrid, Spain, 2007).
23. W. R. Holland and D. G. Hall, "Frequency Shifts of an Electric-Dipole Resonance near a Conducting Surface," Phys. Rev. Lett. **52**, 1041-1044 (1984).
24. G. Lévêque and O. J. F. Martin, "Optical interactions in a plasmonic particle coupled to a metallic film," Opt. Express **14**, 9971-9981 (2006).
25. A. García-Martín, G. Armelles, and S. Pereira, "Light transport in photonic crystals composed of magneto-optically active materials," Phys. Rev. B **71**, 205116 (2005).
26. C. Clavero, A. Cebollada, G. Armelles, Y. Huttel, J. Arbiol, F. Peiro, and A. Cornet, "Size effects in the magneto-optical response of Co nanoparticles," Phys. Rev. B **72**, 024441 (2005).

1. Introduction

Surface plasmons are electromagnetic modes localized at interfaces between two media with permittivity of opposite sign, such as those formed by a dielectric and a metal. For some particular geometries, photons can couple to such surface plasmon modes, modifying the optical response of the system. For example, localized surface plasmon (LSP) modes are responsible for the peaks observed in the absorption spectra of metallic nanoparticles [1,2], and propagating surface plasmon polariton (SPP) modes in a thin metallic layer on a dielectric substrate are the cause of the strong angular dependence of film reflectivity when measured in specific conditions [3]. Also, nanoperforated metallic films show an enhanced optical transmission due to the excitation of surface plasmon modes [4]. Surface plasmon excitation is associated with a strong electromagnetic field enhancement at the metal/dielectric interface [3], which is responsible of phenomena such as surface-enhanced Raman scattering [5,6], enhanced fluorescence emission [7] or high-harmonic generation [8].

Moreover, the excitation of the surface plasmon modes may also affect the magneto-optical (MO) response of metal-dielectric systems. For example, a strong enhancement of the Kerr rotation and ellipticity has been predicted in nanoparticle metallic layers due to the localized surface plasmon resonances of the nanoparticles [9]. Similarly, theoretical studies have shown that the extraordinary optical transmission of nanoperforated thin metallic films could be controlled by applying a magnetic field [10,11]. Most of these studies are based on noble metals whose intrinsic MO activity is however very small and thus require an extremely high magnetic field (tens of Tesla) to observe the predicted phenomena. The value of the needed magnetic field can be highly reduced when using instead ferromagnetic metals with large MO activity. For example a strong reduction of the magneto-optical activity has been observed in nanoperforated Co films in the spectral region corresponding to the anomalous

optical transmission [12]. On the other hand, a strong enhancement of such MO activity has been observed in Co nanoparticles and arrays of Ni nanowires, which has been attributed to the localized surface plasmon resonances of the metallic nanostructures [13-15].

Nevertheless, an important drawback for the use of plain ferromagnetic metals is their high absorption losses, since they manifest themselves as a strong damping of the surface plasmon features. A feasible way to reduce such damping without losing MO activity is to combine noble and ferromagnetic metals, forming thus a so-called magnetoplasmonic system. This way, it has been possible to obtain enhanced MO activity upon surface plasmon excitation in Au/Co/Au continuous layers when SPPs are excited under Kretschmann-Raether configuration [16-18], and also on Au/Co/Au nanodiscs [19] where localized resonances are excited at a specific wavelength. In these cases, the plasmon excitation mainly arises from the noble metal component of the system (Au), and produces an increase of the amount of electromagnetic field located in the MO active material of the system (Co) giving rise to an overall MO activity enhancement. Therefore all the components play a crucial role in the plasmonic and in the MO activity of the magnetoplasmonic material.

However, in principle it is not necessary that the plasmon excitation responsible for this enhanced MO activity occurs in intimate contact with the ferromagnetic layer. In fact, the enhancement is expected to remain as long as the evanescent field of the plasmon excitation extends long enough to reach the MO active layer. In this paper, we demonstrate this hypothesis by analyzing a set of magnetoplasmonic structures in which the MO active component and the plasmonic excitation are spatially separated. The MO region, consisting of Au/Co/Au continuous trilayers grown by magnetron sputtering, is coupled to an array of Au nanodiscs via a thin SiO_2 layer (see Fig. 1). We investigate the influence of the LSP of the Au nanodiscs on the magneto-optical response of the trilayer and we analyzed the role of the different system parameters. Varying the thickness of the dielectric spacer allows us controlling the coupling between the MO and the plasmonic components, while the density of nanodiscs allows controlling the light absorbed by the system via localized plasmon excitation.

It is important mentioning here that, in purely plasmonic media, a periodic structure placed on top of a continuous metallic layer can be used to excite the SPP of the layer at normal incidence [20,21]. In the system under consideration here this effect is very weak due to the high absorption introduced by the Co, making the interpretation more straightforward.

2. Experimental

Au/Co/Au trilayers, of thickness 16nm/10nm/6nm, were deposited by magnetron sputtering on glass substrates. A thickness of 10 nm for the Co layer guarantees the magneto-optical response of the trilayer [17]. On top of it, a SiO_2 layer of thickness d (d=20, 50 or 80 nm) has been deposited by means of electron beam evaporation. Finally, arrays of Au nanodiscs were fabricated on top of these layers by means of electron beam lithography and lift-off. The nanodiscs diameter, determined by SEM imaging, is about 110 nm, the height is 20 nm, and the array periodicity, a, has been varied between 250 and 400 nm. Figure 1 shows a sketch of the fabricated structures.

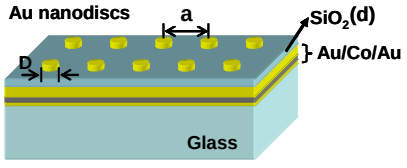

Fig. 1. Sketch of the analyzed structures, consisting of a Au/Co/Au continuous layer with magneto-optical properties separated by a SiO_2 spacer of thickness d from a layer of Au nanodiscs sustaining localized surface plasmon resonances. The Au nanodiscs, of diameter D, form a square array of period a.

The localized surface plasmon resonances of the nanodiscs have been determined by measuring the extinction spectra in a conventional optical microscope coupled to a spectrometer through an optical fiber. The magneto-optical characterization has been carried out by means of a Kerr spectrometer in the polar configuration. This system, described elsewhere [22], allows obtaining both the Kerr rotation and ellipticity spectra. The sample, placed inside an electromagnet that applies a magnetic field perpendicular to its surface high enough to saturate it, is illuminated at normal incidence by a monochromatic beam coming from a Xe lamp followed by a monochromator. By modulating the beam polarization with a photoelastic modulator, the Kerr rotation and ellipticity for each wavelength are determined.

3. Results

In a first step, we are interested in determining the LSP associated with the nanodiscs. Moreover, as it has been previously mentioned, the structures proposed in this work are similar to those used to excite SPP in a continuous underlying Au or Ag layer given that the nanodiscs array acts as a grating. To measure then the LSP and explore to what extent the SPP are excited in the Au/Co/Au trilayers, the extinction spectra of the whole system, normalized with that of a region without discs, have been measured. Figure 2 shows the corresponding extinction results for (a) two different samples with equivalent disc array periodicity (a = 300 nm) but different SiO_2 thickness (d = 20 and 50 nm), and (b) for two samples with equivalent SiO_2 separation (d = 50 nm) but two array periods (a = 250 and 350 nm). In both cases, only a single peak is observed between 1.75 and 2 eV corresponding to the Au nanodiscs LSP resonance. No peak corresponding to the SPP in the continuous trilayer is observed for any value of SiO_2 thickness or disc periodicity. This is associated with the presence of the Co, which highly damps the SPP. However, the presence of the metal trilayer below the Au nanodiscs affects the LSP position due to dipolar coupling effects which take place for very short distances between the metallic particles and the layer [23,24]. This can be seen from the dependence of the normalized absorption peak on the dielectric spacer thickness (Fig. 2(a)). First, the resonance peak shifts from 1.79 eV for 20 nm to 1.83 eV for 50 nm SiO_2 thickness, but it does not shift for further increase in the dielectric spacer (data not shown here) indicating that the interaction decreases with the distance. Second, the intensity of the absorption peak decreases when reducing the spacer thickness, indicating that the proximity of the continuous trilayer makes less effective the LSP excitation.

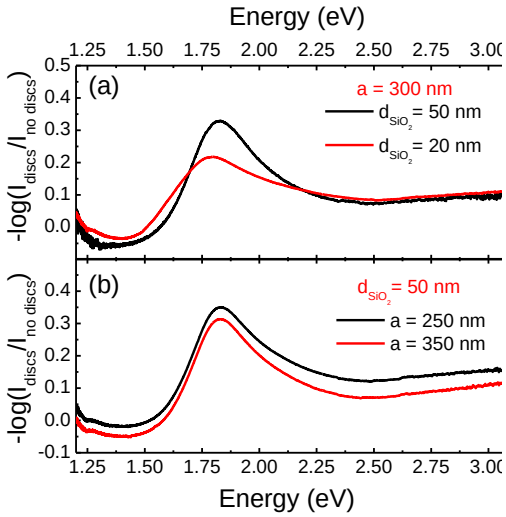

Fig. 2. Extinction spectra of the studied systems, normalized by the transmission signal through the Au/Co/Au/SiO_2 continuous layers. (a) Period of the nanodiscs array, a = 300 nm and SiO_2 layer thickness d variable. (b) SiO_2 layer thickness d = 50 nm and nanodiscs array period a variable. The nanodiscs diameter is about 110 nm in all cases.

On the other hand, Fig. 2(b) shows the effect of the disc separation for a fixed SiO_2 thickness. In this case, the interaction between each of the discs and the continuous trilayer is kept constant and the change in the array periodicity modifies the energy coupled to the SPP. However, the peak position is found to be the same in both cases (1.83 eV), corroborating the fact that no SPP excitation is observed in these systems under normal incidence. (The differences in the intensity of the absorption peaks in this case are due to the change in discs density).

Next, the MO properties of the fabricated structures were measured in polar configuration. In this configuration, normal incidence is used to measure the change in the state of polarization of the light reflected by the structure at the magnetic saturation state when the magnetic field is perpendicular to the sample surface. In Fig. 3 we present representative polar Kerr rotation and ellipticity spectra for a structure with an array period of 300 nm and a SiO_2 thickness of 50 nm. Each magnitude was measured in areas with and without discs on top of the SiO_2/trilayer structures. The strongest differences between the two zones appear in the spectral region corresponding to the LSP excitation of the Au nanodiscs (see Fig. 2).

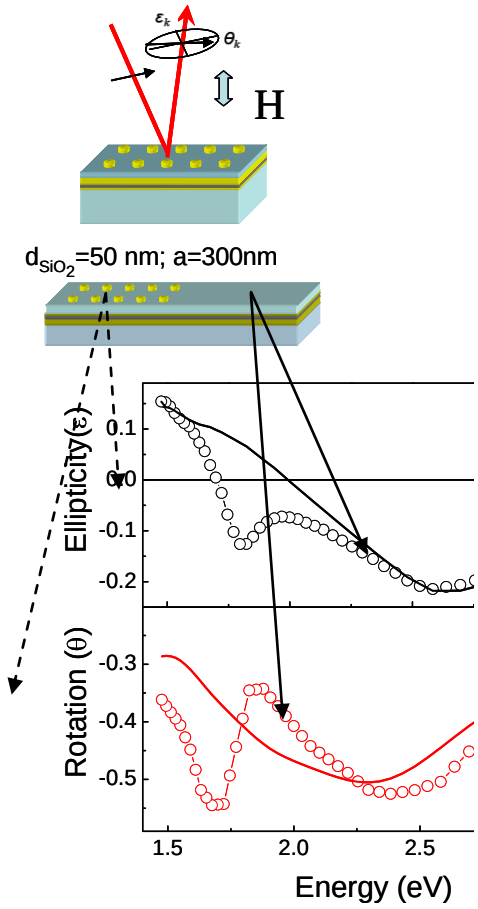

Fig. 3. Kerr rotation and ellipticity spectra, in polar configuration (see upper sketch), for a structure with a nanodiscs array period a = 300 nm and a SiO2 thickness of 50 nm. The dotted lines correspond to areas of the sample with discs on top of the SiO_2/trilayer and the continuous lines to areas without Au discs.

To interpret these findings, it is important to keep in mind that the Kerr rotation (θ) and ellipticity (φ) are related to the elements of the reflectivity matrix as follows:

$\theta + i\varphi = r_{ps}/r_{pp}$, where r_{ps} and r_{pp} are the diagonal and non-diagonal elements of the reflectivity matrix

$$\begin{pmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{pmatrix}.$$

Here, r_{ps} is the non-diagonal element of the reflectivity matrix and reflects the polarization conversion induced by the magneto-optical properties and r_{pp} is the diagonal element for p-polarized light. At normal incidence, as is the case in the measurements shown here, the elements of the reflectivity matrix satisfy $r_{pp} = r_{ss}$; $r_{sp} = -r_{ps}$. On the other hand, the modulus of the complex Kerr rotation $\phi = \sqrt{\theta^2 + \varphi^2} = |r_{ps}/r_{pp}|$ allows studying in a single magnitude all the MO activity of the system.

Considering these definitions, the observed modifications of the Kerr rotation and ellipticity can be due either to a modification of the optical part of the MO properties (r_{pp}), the magnetic part (r_{ps}), or both. To have a clearer picture on the origin of the observed effects, in Fig. 4 we present the spectral dependence of the MO activity ϕ obtained from the experimentally measured rotation and ellipticity (Fig. 4 (a)), the reflectivity r_{pp} experimentally measured without magnetic field applied (Fig. 4 (b)) and the reflectivity r_{ps} obtained as $\phi \times r_{pp}$. These magnitudes are normalized to that of the system without discs on top to isolate the contribution to the MO activity originating from the localized surface plasmons.

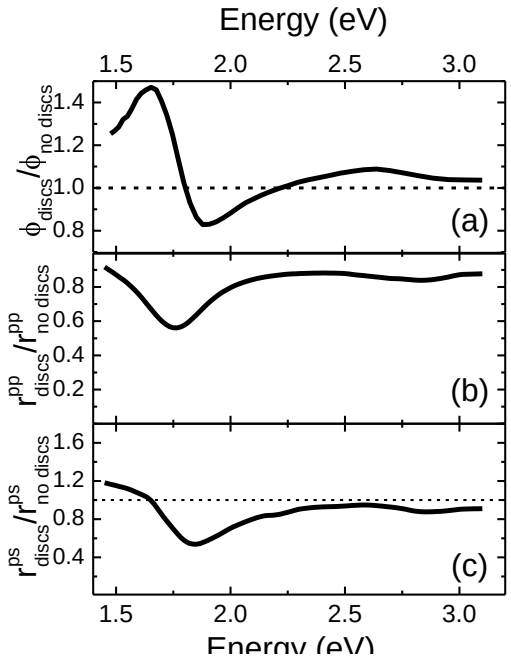

Fig. 4. (a) Complex Kerr rotation, (b) reflectivity r_{pp} and (c) non-diagonal element of the reflectivity matrix r_{ps} determined for the structure with dielectric thickness d = 50 nm and array periodicity a = 300 nm. The obtained values are normalized to those of the area of the sample without discs on top.

As it can be observed in Fig. 4(a), the MO-activity shows an S-type shape centered in the spectral region corresponding to the excitation of the nanodiscs LSP resonance (see Fig. 2(a)), with values exceeding one (MO activity larger in the region with nanodiscs than in the region without discs) in the low energy side of the absorption peak, and smaller than one (MO

activity smaller in the region with nanodiscs than in the region without discs) in the high energy side of the absorption peak. The reflectivity ratio shown in Fig. 4(b) evidences a strong decrease of reflectivity in the spectral region of the LSP peak, as expected from the excitation of the localized plasmon resonance. The purely magnetic component r_{ps}, shown in Fig. 4(c), features a strong dip located in the energy region corresponding to the excitation of the LSP resonance, indicating that, in addition to a reduction of the reflectivity (increase of the absorption) of the system, the nanodisc array also induces a noticeable modification of the "purely magnetic part" of the MO activity. The dependence of the optical absorption with spacer thickness and disc separation shown previously in Fig. 2 must have then a resemblance in that of r_{ps}. This is shown in Fig. 5 where, again, the variation of the spacer thickness (Fig. 5(a)) gives rise to an spectral shift of the dip in r_{ps} in agreement with that observed in the optical absorption, whereas the variation of array periodicity at constant spacer thickness (Fig. 5(b)) does not affect the dip position.

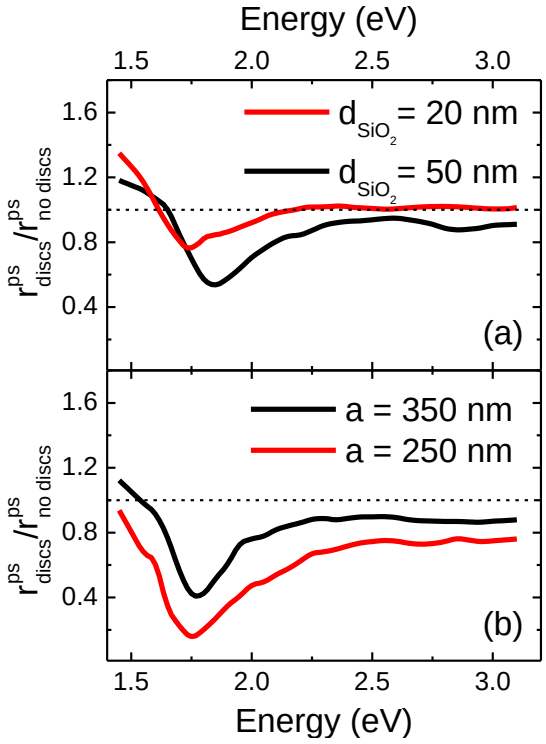

Fig. 5. Non-diagonal element of the reflectivity matrix, r_{ps}, for (a) two structures with the same array periodicity (a = 300 nm) but different dielectric spacer thickness and (b) for two structures with the same spacer thickness (d = 50 nm) but different array periodicity.

To further understand this behavior, the optical and magneto-optical properties of the studied structures have been calculated using a scattering-matrix formalism adapted to treat materials with MO activity [25]. The optical properties of Au and Co where obtained from reference [22], whereas the magneto-optical properties of Co where obtained from reference [26]. In Fig. 6 we present the results of the calculations corresponding to both absorption (Fig. 6(a)) and magneto-optical ratios (Fig. 6(b)) for a structure with a period of 300 nm and for different SiO_2 thickness. As it can be observed, the theoretical absorption spectra present a single peak, whose position redshifts and whose intensity decreases upon reduction of the spacer thickness. On the other hand, the spectra of the MO components have an S-shape structure that also redshifts as the SiO_2 thickness is reduced.

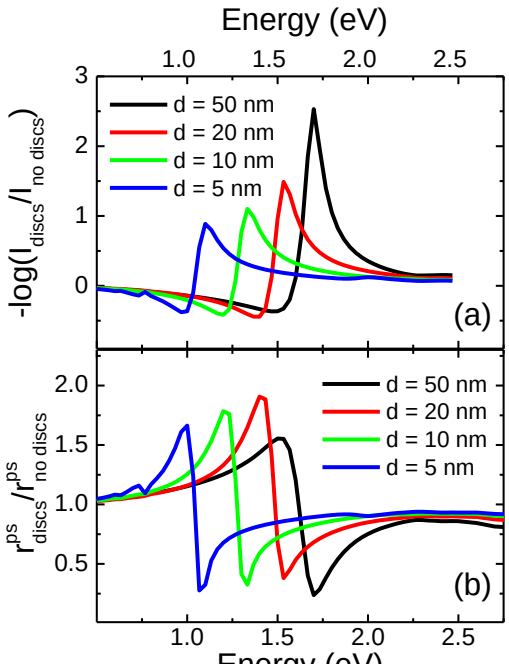

Fig. 6. (a) Calculated absorption spectra for a structure with Au nanodiscs array periodicity a = 300 nm and different spacer thickness d. (b) Calculated non-diagonal element of the reflectivity matrix for the same system as in (a).

The numerical calculations show the same trends observed in the experimental results. In the purely optical response, the dependence with the SiO_2 thickness of the spectral position of the absorption peak is particularly well reproduced, showing the previously described interaction of the localized plasmon resonance with the close continuous metallic layer [23,24]. Interestingly, the magneto-optical ratio shows a striking spectral behavior, increasing or decreasing depending on whether the energy is smaller or larger than that corresponding to the maximum of the absorption peak, respectively. This behavior can be related to the perturbation of the electromagnetic field inside the Co layer induced by the Au discs array. We could not calculate the electromagnetic field distribution inside the system for the exact structures, so to get at least a qualitative picture we have substituted the disk layer by an effective medium layer whose dielectric constant is obtained from the shape and concentration of the discs using the Maxwell–Garnett approximation. Despite the simplicity of this approach, the spectral dependence of the ratio of the electromagnetic (EM) field intensity inside the Co layer with and without discs reproduces well that of the r_{ps} ratio (see Fig. 7): For energies lower than that of the absorption peak the EM intensity inside the Co layer is enhanced compared with the same situation but without the discs layer, whereas for larger energies the EM inside the Co layer is reduced when the discs are present.

From these results we can also understand the dependence of the magnitude of the magneto-optical enhancement with the dielectric spacer thickness plotted in Fig. 5(a). Even if the nanodiscs LSP resonance intensity is decreased for d_{SiO2} = 20 nm compared to d_{SiO2} = 50 nm, the magneto-optical contribution in the lower energy area is increased due to a higher penetration of the electromagnetic field in the Co layer associated to the closer proximity between the two systems.

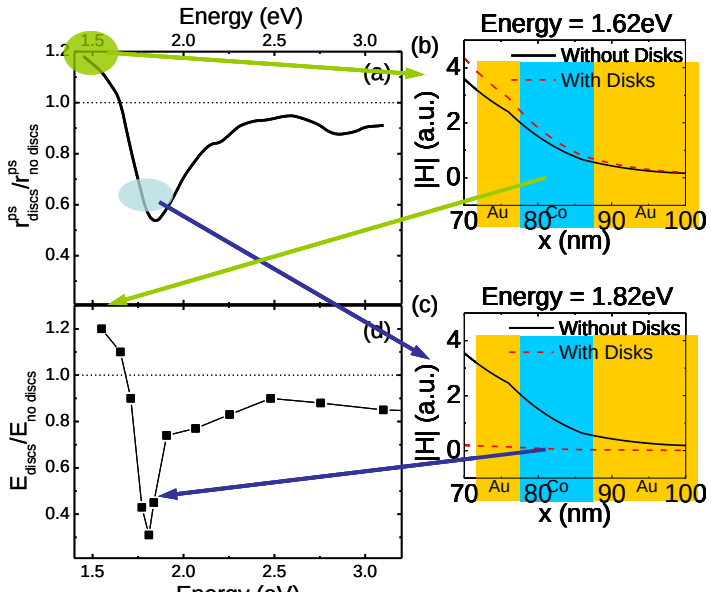

Fig. 7. (a) Non-diagonal element of the reflectivity matrix r_{ps} (magneto-optical contribution), for the structure with array periodicity a = 300 nm and dielectric thickness d_{SiO2} = 50 nm. (b) Calculated distribution of the electromagnetic field inside the Au/Co/Au continuous trilayer for the system with Au discs on top (red dashed line) and without nanodiscs (black solid line) at energy 1.62 eV (position of the maximum magneto-optical enhancement associated with the presence of the discs, r_{ps}(discs)/r_{ps}(no discs)). (c) Same as in (b) but at energy 1.82 eV (minimum magneto-optical contribution associated with the presence of the discs). (d) Spectral dependence of the ratio of the electromagnetic field inside the Co layer for the continuous SiO_2/trilayer system with and without nanodiscs on top. (Note: Calculations are done by substituting the Au nanodiscs array layer by an effective medium layer using the Maxwell-Garnett approximation.)

4. Conclusions

The results shown here validate the fact that the magneto-optical properties of a magnetic layer can be enhanced by plasmonic resonances spatially separated as far as the electromagnetic field associated with the resonances penetrates in the magnetic region. Through an appropriate design of the structure to sustain an even higher electromagnetic field in the magnetic layer at the desired energies, the magneto-optical properties of the system could be further "boosted".

Acknowledgements

This research was carried out with the financial support of the Spanish Ministry of Science and Education (NAN2004-09195-C04 and MAT2005-05524-C02-01), Comunidad de Madrid (S-0505/MAT/0194 NANOMAGNET) and the European Commission through the NoE PHOREMOST (FP6/2003/IST/2-511616). M. U. G. thanks the Spanish Ministry of Education for funding through the "Ramón y Cajal" program.