Magnetic modulation of surface plasmon modes in magnetoplasmonic metal-insulator-metal cavities

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Abstract: The magnetic modulation of the surface plasmon-polariton (SPP) wavevector is experimentally and theoretically studied for the plasmonic modes excited in metal-insulator-metal (MIM) magnetoplasmonic cavities. For this purpose, Ag/SiO$_2$/Ag multilayers with different SiO$_2$ layer thickness in which a thin Co layer is positioned near the top Ag/SiO$_2$ interface, near the bottom SiO$_2$/Ag one, or near both of them, are studied. The magnetoplasmonic MIM cavities present symmetric (SM) and antisymmetric (AM) plasmonic modes, of different wavevector and electromagnetic field profiles inside the MIM cavity. We show that the magnetic SPP wavevector modulation strongly depends on which mode is considered, the cavity thickness, and the number and specific location of Co layers within the structure. With only one ferromagnetic layer, a net modulation is obtained, of higher magnitude as we reduce the SiO$_2$ layer thickness. The introduction of a second Co layer in the structure reduces the modulation due to the non-reciprocal character of SPP modes under an applied magnetic field. Moreover, we demonstrate that the non-reciprocal nature of the SPP modulation can be experimentally visualized in the magnetic hysteresis loops under plasmon excitation conditions by using two Co layers with different magnetization switching fields.

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References and links

1. K. J. Vahala, “Optical microcavities,” Nature 424, 839–846 (2003).
2. A. M. Armani, R. P. Kulkarni, S. E. Fraser, R. C. Flagan, and K. J. Vahala, “Label-free, single-molecule detection with optical microcavities,” Science 317, 783–787 (2007).
3. S. A. Maier, Plasmonics: Fundamentals and Applications (Springer, 2007).
4. H. Raether, “Surface Plasmons on Smooth and Rough Surfaces and on Gratings,” in Springer Tracts in Modern Physics (Springer-Verlag, 1988), Vol. 111.
5. S. I. Bozhevolnyi, V. S. Volkov, E. Devaux, J. Y. Laluet, and T. W. Ebbesen, “Channel plasmon subwavelength waveguide components including interferometers and ring resonators,” Nature 440, 508–511 (2006).
6. B. Steinberger, A. Hohenau, H. Ditlbacher, A. L. Stepanov, A. Drezer, F. R. Aussenegg, A. Leitner, and J. R. Krenn, “Dielectric stripes on gold as surface plasmon waveguides,” Appl. Phys. Lett. 88, 094104 (2006).
7. J.-C. Weeber, Y. Lacroute, and A. Dereux, “Optical near-field distributions of surface plasmon waveguide modes,” Phys. Rev. B 68, 115401 (2003).
8. W. L. Barnes, A. Dereux, and T. W. Ebbesen, “Surface plasmon subwavelength optics,” Nature 424, 824–830 (2003).
9. S. A. Maier and H. A. Atwater, “Plasmonics: Localization and guiding of electromagnetic energy in metal/dielectric structures,” J. Appl. Phys. 98, 011101 (2005).
10. J. Homola, “Surface plasmon resonance sensors for detection of chemical and biological species,” Chem. Rev. 108, 462–493 (2008).
11. 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).
12. C. E. Talley, J. B. Jackson, C. Oubre, N. K. Grady, C. W. Hollars, S. M. Lane, T. R. Huser, P. Nordlander, and N. J. Halas, “Surface-enhanced Raman scattering from individual Au nanoparticles and nanoparticle dimer substrates,” Nano Lett. 5, 1569–1574 (2005).
13. H. Xu, E. J. Bjerneld, M. Käll, and L. Börjesson, “Spectroscopy of single hemoglobin molecules by surface enhanced Raman scattering,” Phys. Rev. Lett. 83, 4357–4360 (1999).
14. I. D. Rukhlenko, A. Paninipiyta, M. Premanatee, and G. P. Agrawal, “Exact dispersion relation for nonlinear plasmonic waveguides,” Phys. Rev. B 84, 113409 (2011).
15. E. N. Economou, “Surface plasmons in thin films,” Phys. Rev. 182, 539–554 (1969).
16. J. A. Dionne, L. A. Sweatlock, H. A. Atwater, and A. Polman, “Plasmon slot waveguides: Towards chip-scale propagation with subwavelength-scale localization,” Phys. Rev. B 73, 035407 (2006).
17. R. Zia, M. D. Selker, P. B. Catrysse, and M. L. Brongersma, “Geometries and materials for subwavelength surface plasmon modes,” J. Opt. Soc. Am. A 21, 2442–2446 (2004).
18. P. Ginzburg, D. Arbel, and M. Orenstein, “Gap plasmon polariton structure for very efficient microscale-to-nanoscale interfacing,” Opt. Lett. 31, 3288–3290 (2006).
19. J. A. Dionne, H. J. Lezec, and H. A. Atwater, “Highly confined photon transport in subwavelength metallic slot waveguides,” Nano Lett. 6, 1928–1932 (2006).
20. H. T. Miyazaki and Y. Kurokawa, “Squeezing visible light waves into a 3-nm-thick and 55-nm-long plasmon cavity,” Phys. Rev. Lett. 96, 097401 (2006).
21. S. A. Maier, “Effective mode volume of nanoscale plasmon cavities,” Opt. Quantum Electron. 38, 257–267 (2006).
22. P. Berini, “Figures of merit for surface plasmon waveguides,” Opt. Express 14, 13030–13042 (2006), http://www.opticsexpress.org/abstract.cfm?URI=oe-14-26-13030.
23. D. Pacífic, H. J. Lezec, and H. A. Atwater, “All-optical modulation by plasmonic excitation of CdSe quantum dots,” Nat. Photonics 1, 402–406 (2007).
24. T. Nikolajsen, K. Leosson, and S. I. Bozhevolnyi, “Surface plasmon polariton based modulators and switches operating at telecom wavelengths,” Appl. Phys. Lett. 85, 5833–5835 (2004).
25. M. J. Dicken, L. A. Sweatlock, D. Pacífic, H. J. Lezec, K. Bhattacharya, and H. A. Atwater, “Electrooptic modulation in thin film Barium Titanate plasmonic interferometers,” Nano Lett. 8, 4048–4052 (2008).
26. J. A. Dionne, K. Diest, L. A. Sweatlock, and H. A. Atwater, “PlasMOStor: A Metal-Oxide-Si field effect plasmonic modulator,” Nano Lett. 9, 897–902 (2009).
27. V. I. Safarov, V. A. Kosobukin, C. Hermann, G. Lampel, J. Peretti, and C. Marlière, “Magneto-optical effects enhanced by surface plasmons in metallic multilayer films,” Phys. Rev. Lett. 73, 3584–3587 (1994).
28. 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).
29. 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-magneto-plasmon nonreciprocity effects in noble-metal/ferromagnetic heterostructures,” Phys. Rev. B 76, 155402 (2007).
30. G. Armelles, J. B. González-Díaz, A. García-Martín, J. M. García-Martín, A. Cebollada, M. U. González, S. Acimovic, J. Cesario, R. Quidant, and G. Badenes, “Localized surface plasmon resonance effects on the magneto-optical activity of continuous Au/Co/Au trilayers,” Opt. Express 16, 16104–16112 (2008).
31. V. V. Temnov, G. Armelles, U. Woggon, D. Guzatov, A. Cebollada, A. García-Martín, J. M. García-Martin, T. Thomay, A. Leitenstorfer, and R. Bratschitsch, “Active magneto-plasmonics in hybrid metal- ferromagnet structures,” Nat. Photonics 4, 107–111 (2010).
32. C. Clavero, K. Yang, J. R. Skuza, and R. A. Lukaszew, “Magnetic field modulation of intense surface plasmon polaritons,” Opt. Express 18, 7743–7752 (2010), http://www.opticsexpress.org/abstract.cfm?URI=oe-18-8-7743.
33. C. Clavero, K. Yang, J. R. Skuza, and R. A. Lukaszew, “Magnetic-field modulation of surface plasmon polaritons on gratings,” Opt. Lett. 35, 1557–1559 (2010).
34. J. F. Torrado, J. B. González-Díaz, M. U. González, A. García-Martín, and G. Armelles, “Magneto-optical effects in interacting localized and propagating surface plasmon modes,” Opt. Express 18, 15635–15642 (2010), http://www.opticsexpress.org/abstract.cfm?URI=oe-18-15-15635.
35. D. Martin-Becerra, J. B. González-Díaz, V. V. Temnov, A. Cebollada, G. Armelles, T. Thomay, A. Leitenstorfer, R. Bratschitsch, A. García-Martín, and M. U. González, “Enhancement of the magnetic modulation of surface plasmon polaritons in Au/Co/Au films,” Appl. Phys. Lett. 97, 183114 (2010).
1. Introduction

Optical cavities are powerful tools to control the interaction of light with matter and have been employed in a wide range of fields from quantum optical devices [1] to ultrasensitive optical sensors [2]. These applications rely on the effective confinement of light in a small cavity mode volume. While most of cavity designs so far are based on dielectric material assemblies, the confinement of light using metallic nanostructures has recently become a topic of great interest. In contrast to dielectric cavities, metallic plasmonic ones can overcome the diffraction-limited size and in principle support a subwavelength volume mode [3]. In these structures, the optical fields are dominated by the excitation of surface plasmon polaritons (SPPs), which are electromagnetic waves coupled to oscillations of the electron plasma located on a metal–dielectric interface [4] and that are currently offering new routes to nanophotonics [5-8]. Moreover, the locally enhanced field intensities observed in plasmonic structures [9] offer promising potential for molecular biosensing [10, 11], surface enhanced Raman spectroscopy [12,13], and nonlinear optical device applications [14].

The strong confinement of light is particularly interesting for SPPs guided in a nanosized dielectric gap between two metals [15-17]. Such a metal-insulator-metal (MIM) geometry serves as a plasmonic slot waveguide, “squeezing” the SPP field into the dielectric core [18]. As a result, the wavelength along the direction of propagation is shortened significantly. MIM-SPPs can therefore be guided in waveguides with very small transverse dimensions [19] and allow the realization of nanocavities with extremely small mode volumes [20,21]. In comparison to other SPP guiding geometries, the cost of extra losses that are linked to the increase in confinement is relatively low [15,16,22].

On the other hand, a paramount issue for the future of plasmonics is the development of active elements, i.e., systems whose plasmonic properties can be modified by an external agent. For this purpose, and in addition to the use of quantum dots [23], thermo-optic [24] or electro-optic [25,26] materials, an innovative approach is the incorporation of a ferromagnetic material into the plasmonic structure, thus creating a magnetoplasmonic system. These magnetoplasmonic structures open interesting possibilities for the elaboration of active
devices in plasmonics, and in fact a great deal of intertwined plasmonic and magnetooptical (MO) phenomena have been studied in Au/Co/Au [27-36], Ag/Co/Ag [37,38] and Au/Fe/Au [39,40] heterostructures. In addition to the enhancement of the magneto-optical activity of these systems upon the excitation of plasmon resonances, a sizeable modulation of the SPP wavevector ($k_{SPP}$) has been found when an external magnetic field is applied perpendicular to the SPP propagation direction and parallel to the interface due to the MO response of the ferromagnetic layer [29,31,36]. This magnetic modulation depends on the thickness of the ferromagnetic layer and increases with the quality of the interfaces [39] or with the addition of an insulator layer [34]. Moreover, a very interesting property of the effect of the magnetic field on the SPP wavevector is the presence of non-reciprocity, i.e., the wavevector of a forward propagating SPP is different from that of a backward propagating one in the presence of a magnetic field [41]. Due to this, non reciprocity is for example of great interest for the development of integrated isolators [41-43].

In this work we present a step forward in the investigations on the effect of a magnetic field on SPPs by the incorporation of an insulator layer into the Noble Metal/ Ferromagnetic Metal/ Noble Metal heterostructure, forming magnetoplasmonic MIM (MP-MIM) systems. Thanks to the mentioned benefits of their improved confinement properties, these systems open new routes for the realization of integrated magnetic SPP modulators. In particular, here we experimentally analyze the magnetic field modulation of the wavevector for SPP MP-MIM modes of (Ag/SiO$_2$/Ag)-Co multilayered structures grown on SF10 glass substrates. The understanding and discussion of the obtained results is carried out with the help of numerical simulations based on a transfer matrix formalism with magneto-optical activity incorporated. The manuscript is organized as follows. In section 2 we will first analyze theoretically the phenomenology associated with MP-MIM structures. Section 3 describes the fabricated structures as well as their structural and magnetic characterization. Section 4 will deal with the magnetic modulation of the SPP wavevector of the different MP-MIM modes excited in the fabricated structures using Attenuated Total Reflection (ATR) configuration together with the discussion of the influence of non-reciprocity effects. Finally, section 5 summarizes the main conclusions.

2. Phenomenology of magnetoplasmonic MIM structures

Metal-Insulator-Metal (MIM) structures behave as waveguides which sustain both optical and SPP modes [16,18]. When the two metal layers are far enough each metal/dielectric interface supports a SPP mode [see the inset of Fig. 1(a)], but when both interfaces approach they become electromagnetically coupled and a more complex behavior appears. This is better seen in Fig. 1(a), where we present the evolution of the effective index of the waveguide modes for a Ag/SiO$_2$/Ag structure as a function of the SiO$_2$ layer thickness ($w_0$) for a frequency of 1.985 eV (HeNe laser). The effective index, $n_{eff}$, is defined as the ratio of a mode wavevector, $k$, to the wavevector of light in vacuum, $k_0$: $n_{eff} = k/k_0$. The modes’ wavevectors have been calculated by using a transfer matrix formalism coupled to a numerical solver for guided modes. The dielectric constant used for Ag has been taken from Ref. [38] and the SiO$_2$ layer is modeled by $n=1.464$; both values correspond to experimentally determined values of the deposited layers. As SPPs are transversal magnetic (TM) modes, only these ones are shown.

As shown in Fig. 1(a), for $w_0$ higher that 850 nm the structure supports several optical modes, that is, modes below the SiO$_2$ light line, $n=1.464$, plotted by a blue dot-dashed line, which are denoted as TM$_n$ with $n>2$. It also presents two uncoupled SPPs, one for each Ag/SiO$_2$ interface ($n_{eff} = 1.55$); these modes are above the light line because of their plasmonic nature. The number of optical modes obviously depends on the thickness of the SiO$_2$ core and decreases as $w_0$ gets smaller. More interestingly, when the core thickness decreases below 850 nm, the two surface waves start to couple through their evanescent electromagnetic fields inside the optical cavity and the degenerated SPP splits into two modes, TM$_1$ and TM$_0$. These two SPP modes are named the antisymmetric (AM) and the symmetric (SM) ones,
respectively, attending to the profiles of the \( H_y \) component of their electromagnetic fields inside the waveguide core, shown in Fig. 1(b). As the MIM core width is further reduced, the wavevector of the AM mode decreases, in such a way that this mode loses its plasmonic character (once it crosses the light line) and it is progressively less confined inside the waveguide. This mode reaches cut-off at \( w_0 = 180 \) nm. The SM mode, on the other hand, increases its in-plane wavevector as \( w_0 \) decreases, keeping always a plasmonic nature and increasing its confinement inside the waveguide [18].

Fig. 1. (a) Effective index evolution of the TM modes confined inside a Ag/SiO\(_2\)/Ag MIM structure as a function of SiO\(_2\) thickness at 1.985 eV (HeNe laser). The grey area delimits the effective index region accessible when exciting the structure by ATR configuration through a SF10 glass substrate (\( n = 1.723 \)). The patterned portion inside defines the accessible region presenting only plasmonic modes, which corresponds to the experimental area of interest for this work. Inset: Scheme of a MIM structure, with one SPP at each metal-dielectric interface.

(b) Magnetic field amplitude (\( H_y \)) distribution for both plasmonic modes, AM (red dashed line) and SM (black continuous line), inside a MIM cavity.

A thin ferromagnetic metallic layer inserted into either one or both metallic claddings of the MIM structure reduces the propagation length of the modes, but it does not modify their evolution with the dielectric core thickness, in particular the splitting of the degenerated SPP into two SPP modes with symmetric (SM) and antisymmetric (AM) character. Besides, due to the MO properties of the ferromagnetic layer, a magnetic modulation of the SPP wavevector can be obtained. This is shown in Fig. 2(a), where we present the simulated normalized magnetic modulation, \( (\Delta k/k)_{\text{SPP}} \), of the two modes induced by a magnetic field applied parallel to the interface and perpendicular to the SPP wavevector for three different MP-MIM structures: with one Co layer inserted in the upper metallic layer, labeled Co\(_{\text{top}}\) (Ag/6 nm Co/10 nm Ag/\( w_0 \) nm SiO\(_2\)/Ag); with one Co layer inserted in the bottom cladding, labeled Co\(_{\text{bottom}}\) (Ag/\( w_0 \) nm SiO\(_2\)/10 nm Ag/6 nm Co/Ag); and with two Co layers, one for each cladding, labeled 2xCo (Ag/6 nm Co/10 nm Ag/\( w_0 \) nm SiO\(_2\)/10 nm Ag/6 nm Co/Ag). \( \Delta k \) corresponds to half the difference between the SPP wavevectors when the Co layers are fully magnetized along the positive and negative direction of the applied magnetic field, respectively [31]:

\[
\frac{\Delta k}{k}_{\text{SPP}} = \frac{k_{\text{SPP}}(+H)-k_{\text{SPP}}(-H)}{2k_{\text{SPP}}(H = 0)}. \tag{1}
\]

The same formalism used for the optical analysis has been employed here, introducing the magneto-optical activity of the Co layer through its description by means of a dielectric tensor. The Co optical and magneto-optical constants were obtained from Ref. [44].

If we analyze first the behavior of the Co\(_{\text{bottom}}\) structure, we see that both the symmetric
and the antisymmetric modes present a magnetic field induced wavevector modulation of positive sign. For large $w_0$, the modulation of both modes approaches that of a single metal/SiO$_2$ interface, likewise their wavevector does. Actually, regarding $\Delta k$, the AM mode modulation moves towards that of the SiO$_2$/AgCoAg interface, with a finite value, while the SM mode $\Delta k$ approximates that of the Ag/SiO$_2$ one, of zero value as there is no ferromagnetic component. Therefore, as can be seen in Fig. 2(a), the modulation of the AM mode for large $w_0$ is slightly higher than that of the SM mode. As the SiO$_2$ layer thickness is decreased, the modulation increases for both modes, indicating the potential advantages of these MP-MIM systems from the applied point of view. This $\Delta k$ increase when $w_0$ decreases is initially smooth, but it becomes sharper when the insulator thickness reaches the values at which the modes become highly dispersive (their wavevector starts to change strongly with $w_0$). This happens near cut-off for the AM mode and for very thin cavities (below 50 nm) for the SM one.

![Graph showing wavevector modulation for AM and SM modes](image)

**Fig. 2.** (a) Simulation of the SPP wavevector modulation for 1.985 eV as a function of SiO$_2$ thickness for the symmetric (SM) and antisymmetric (AM) modes in three magnetoplasmonic MIM systems: with one Co layer inserted in the bottom metallic cladding, Co$_{\text{bottom}}$ [Ag/SiO$_2$/10 nm Ag/6 nm Co/Ag] (blue line); with one Co layer in the top cladding, Co$_{\text{top}}^+$ [Ag/6 nm Co/10 nm Ag/SiO$_2$/Ag] (grey dotted line); and with two Co layers, one in each cladding [Ag/6 nm Co/10 nm Ag/SiO$_2$/10 nm Ag/6 nm Co/Ag] (green dashed line). (b) Left: Sketch of the relations for the magnetic modulation sign in single interfaces depending on the magnetic field and wavevector directions and on the interface configuration. Right: Resulting magnetic modulation contribution to the SPP wavevector from each insulator/metal interface in a symmetric MIM structure.
Concerning the Co\textsuperscript{top} structure, we obtain exactly the same behavior and the same values of modulation as for Co\textsubscript{bottom}, but with inverted sign. This can be understood if we consider the non-reciprocal nature of the magnetic field induced modulation of SPP wavevector [41], sketched in Fig. 2(b). Here, some schematic drawings of the change of sign in the modulation for different relative orientation between the applied magnetic field, the SPP propagation direction and the metal (M) / insulator (I) interface are shown. It can be assumed that, for a M/I interface, the effect of a positive magnetic field on the wavevector of a SPP propagating in the positive direction is a reduction of wavevector, i.e. a negative modification $-|\Delta k|$. If the magnetic field orientation is reversed, so does the sign of $\Delta k$, hence it becomes positive for a negative magnetic field acting on the wavevector of a SPP propagating in the positive direction in a M/I interface. On the contrary, the system (including the applied external magnetic field) is invariant under rotation around the $x$-axis, and the wavevector modification becomes negative for a negative magnetic field on a SPP propagating in the positive direction in an I/M interface. In fact, the Co\textsuperscript{top} structure presents magnetoplasmonic activity only in the upper interface, and then it corresponds to applying a magnetic field to just the Metal/Insulator interface. The Co\textsubscript{bottom} structure, on the other hand, has magnetoplasmonic activity in the lower interface and corresponds to applying a magnetic field to the Insulator/Metal interface. Therefore, these two structures should have the same magnitude of $\Delta k$, but with opposite sign. When we couple the two M/I and I/M interfaces, both with magnetoplasmonic activity, in a MIM structure, as observed in the right sketch of Fig. 2(b), the magnetic field effects at each interface cancel each other out. Therefore, the structure with two Co layers, 2xCo, would present no magnetic field modulation of the wavevector for neither of the two modes. This is indeed the case, as can be seen in Fig. 2(a).

3. Ag/SiO\textsubscript{2}/Ag-Co magnetoplasmonic structures: morphological and magnetic characterization

In order to analyze experimentally the phenomenology introduced in the previous section, MP-MIM structures exhibiting the described plasmonic modes susceptible to be excited must be fabricated. For that, we will use a Kretschmann configuration, for which the metallic layers of the MIM structures located near the substrate should be thin enough to allow the coupling of the incident light to the SPP modes. Besides, the substrate must have a refractive index higher than that of SiO\textsubscript{2}, and for this purpose SF10 ($n = 1.723$) was selected. Under these conditions, the accessible modes of the MIM structure are those delimited by the gray area in Fig. 1(a). Moreover, since the goal of this work is the study of the magnetic field induced changes of the wavevector of the SPP modes, in order to prevent interferences with the other optical modes, the SiO\textsubscript{2} thickness will be kept below 400 nm [see Fig. 1(a)]. Consequently, the region of interest for the design of our magnetoplasmonic MIM cavities is within the 180-400 nm core width range.

Three series of Ag/SiO\textsubscript{2}/Ag-Co MP-MIM cavities with the Co layer located at different positions within the structure have been grown at RT on SF10 glass substrates. The three series, whose internal structure is schematically shown in Fig. 3, are denoted as Co\textsubscript{bottom} (Co in the lower metallic layer), Co\textsuperscript{top} (Co in the upper metallic cladding) and 2xCo (Co at both claddings). The SiO\textsubscript{2} thickness has been varied between 180 nm and 380 nm for each series. The Ag layers were deposited by magnetron sputtering whereas the Co and SiO\textsubscript{2} ones were deposited by e-beam evaporation. To prevent the chemical deterioration of Ag, the structures were capped with a 10 nm thick Au layer, deposited by magnetron sputtering.
Fig. 3. Schematic drawings of the three series of MIM-MP cavities fabricated on SF10 substrates, with different Co layer position: underneath SiO2 layer, Co\textsubscript{bottom}; on top of the SiO2 layer, Co\textsubscript{top}; and both below and above the SiO2 layer, 2xCo.

The surface morphology of all the fabricated structures was characterized by atomic force microscopy (AFM). Figure 4(a) shows a representative image for the 380 nm SiO\textsubscript{2} sample of the Co\textsubscript{bottom} series. Granular structures can be observed over the surface, presenting a clear state of coalescence between them. A comparison of representative AFM profiles for the three series of samples and a reference sample without SiO\textsubscript{2} layer is depicted in Fig. 4(b). The AFM profile for the fully metallic reference sample exhibits the flattest surface (RMS roughness value of 0.9 nm versus RMS roughness around 2 nm for the MIM samples), indicating that the surface roughness of the MP-MIM cavities is mainly due to the deposition of the insulator layer. These results suggest that the Co layer morphology could depend on its position inside the cavity.

In order to determine the magnetization reversal processes of the Co layers deposited in different places of the MIM structure, magnetic hysteresis loops at room temperature were obtained by Magneto-Optical Kerr Effect measurements in Transversal configuration (TMOKE loops). The TMOKE loops were recorded by using a linearly p-polarized 532 nm light beam (solid-state laser) at 45° incidence angle geometry with the applied magnetic field in the film plane but perpendicular to the plane of incidence, and measuring the variations in the reflected light intensity as the intensity and sign of the magnetic field are varied [see scheme in the upper part of Fig. 5].
Figure 5 shows the representative TMOKE loops of the three 380 nm SiO₂ samples. The loops are normalized to one at saturation. On the left, the TMOKE loop for the Co$_{\text{bottom}}$ sample is presented. It exhibits an almost rectangular shape and very low coercive field (around 30 Oe), that is characteristic of flat ferromagnetic films with in-plane magnetic anisotropy. On the middle graph, the TMOKE loop for the Co$_{\text{top}}$ sample can be observed. It exhibits a much higher coercive field (around 150 Oe) compared to the Co$_{\text{bottom}}$ sample loop. This difference in the magnetic behavior of the two Co position series can be ascribed to the difference of the roughness for the Co layers grown at the two different positions. The Co layer grown above the SiO₂ layer is much rougher than that grown below the SiO₂, and as a consequence it has higher density of defects. Such defects act as pinning centers preventing the movement of the magnetic domain walls, therefore higher magnetic field is required to overcome the pinning and reach the magnetization reversal. Finally on the right the TMOKE loop for the 2xCo sample is shown. The magnetization reversal process exhibits two components: The first magnetization component shows a low coercive field similar to that of Co$_{\text{bottom}}$ loops. The second magnetization component exhibits a higher coercive field similar to that of the Co$_{\text{top}}$ loop. This indicates that the Co layer underneath the SiO₂ can be reversed at low magnetic fields whereas the Co layer on top of the SiO₂ one can only be switched at higher magnetic fields.

4. Magnetic modulation of SPP modes

The SPP modes of the MP-MIM structures have been excited using a Kretschmann configuration [see Fig. 6(a)] and a He-Ne laser ($E = 1.985$ eV, $\lambda = 632$ nm). The samples were placed on the flat face of a semi-circular SF10 prism using a refractive index matching liquid. In this geometry a minimum in the angular reflectivity curve of $p$-polarized light is linked to the excitation of a SPP mode. As an example, we present in Fig. 6(b) the reflectivity of a representative sample as a function of the incidence angle. The dotted curve corresponds to the experimental data whereas the full line is a fitting from which an experimental value of the SiO₂ layer refractive index (n) of 1.464 is obtained. As it can be observed, the reflectivity curve exhibits two minima corresponding to the excitation of the AM and SM modes of the MIM cavity.
When a magnetic field is applied [see Fig. 6(a)], the wavevector of the plasmonic modes depends on the direction of the Co layers magnetization. As a result, the angle at which the reflectivity exhibits the minima ($\theta_{\text{min}}$) will also depend on the magnetization direction [see Fig. 6(c)]. Therefore the TMOKE signal, $\Delta R/R$, defined as the normalized difference between the reflectivity of the structure when the Co layers are magnetized along the positive or negative direction, $\Delta R/R = \left( R_{\text{pp}} (+H) - R_{\text{pp}} (-H) \right) / \left( R_{\text{pp}} (+H) + R_{\text{pp}} (-H) \right)$, is related to the angular derivative of the structure, $dR/d\theta$, as follows [29,38]:

$$\frac{\Delta R}{R} = \frac{dR}{d\theta} \frac{\Delta \theta}{2 R}, \tag{2}$$

where $\Delta \theta$ is connected to the magnetic modulation of the SPP wavevector, $\Delta k$, as:

$$\left( \frac{\Delta k}{k} \right)_{\text{SPP}} = \frac{\Delta \theta}{2 \cot \theta_{\text{min}}}, \tag{3}$$

being $k_{\text{SPP}}$ the SPP wavevector ($k_{\text{SPP}} = n(\lambda/2\pi)\sin \theta_{\text{min}}$), $n$ the refractive index of the SF10 substrate, and $\lambda$ the wavelength of the laser.

![Fig. 6.](image)

(a) Scheme of the TMOKE measurements under Kretschmann configuration to allow SPP excitation. (b) Experimental (blue circles) and simulated (black line) reflectivity curves for a representative MIM-MP cavity ($w_0 \text{SiO}_2 = 260 \text{ nm}$ of the Co$_{\text{bottom}}$ series) showing the antisymmetric and symmetric modes. (c) Schematic representation of the effect that the direction of the magnetization (positive-black line, negative-red line) has on the reflectivity curve for a MP-MIM structure. For the sake of clarity the effect of the magnetic field on the SPP wavevector has been exaggerated. (d) TMOKE curve showing the resonances around the excitation of the two modes and the fitting to the angular derivative of the experimental reflectivity used to obtain $\Delta \theta$.

Therefore, by comparing the TMOKE angular curves with the angular derivative of the reflectivity curves, $\Delta k$ can be obtained. Fig. 6(d) shows, as an example, the TMOKE signal
and the angular derivative of the reflectivity of the same sample as in Fig. 6(b). The only fitting parameter is $\Delta \theta$, which is different for the two SPP modes. The obtained absolute values of $\Delta k$, normalized to the SPP wavevector, are shown in the left column of Fig. 7 for both the antisymmetric and the symmetric plasmonic modes for all the fabricated samples of the three series with only one exception: the $\Delta k$ value for the AM mode of the 2xCo sample with 380 nm thick SiO$_2$ could not be obtained as the agreement between the TMOKE curve and the angular derivative of the experimental reflectivity was not good in that particular case, pointing out that other contributions to the TMOKE signal besides the wavevector modulation are sizable. For the structures with only one Co layer, i.e. the Co$_{bottom}$ [Fig. 7(a)] and Co$_{top}$ series [Fig. 7(b)], both modes show a similar modulation value for the highest $w_0$, and a strong increase in the AM mode when reducing the SiO$_2$ thickness compared to a quasi-constant value for the SM mode. These results are in good agreement with the behavior expected from the analysis presented in section 2 [Fig. 2(a)], since in the explored thickness range we are close to the cut-off thickness when reducing $w_0$ for the AM mode but far from the highly dispersive region for the SM one.

Fig. 7. Experimental (left column) and simulated (right column) absolute values of the magnetic modulation of the SPP wavevector ($|\Delta k/k|_{SPP}$) as a function of SiO$_2$ thickness for AM (red triangles and dashed lines) and SM (black symbols and solid lines) plasmonic modes in Co$_{bottom}$ (a), Co$_{top}$ (b) and 2xCo (c) MP-MIM cavities. In (c), the $|\Delta k/k|_{SPP}$ values of the 2xCo series are compared to the absolute difference of the Co$_{bottom}$ and Co$_{top}$ series modulation values for the AM (star symbols) and the SM modes (hexagonal grey symbols).
In the case of the 2xCo series [Fig. 7(c)], the magnetic modulation of the SPP wavevector is lower than that obtained for the series with only one Co layer, even though both modes yield non negligible modulations. This is in contrast to the null values expected for an ideal MIM cavity with two identical Co layers symmetrically positioned, as shown in section 2. This can be understood if we take into account that the internal structure of the fabricated MIM structures for the 2xCo series, as well as the modes excitation procedure, are asymmetric. Therefore, the distribution of the EM field intensity at each interface, which controls the magnetic modulation [31,35,36], is not exactly symmetric and the two opposing contributions to the total $\Delta k$ do not cancel out. In fact, the obtained values of $\Delta k$ for the 2xCo MP-MIM series should be almost equal to the difference between the values obtained for the $\text{Co}_{\text{bottom}}$ and $\text{Co}^{\text{top}}$ series. Such differences are also shown in Fig. 7(c), and as a matter of fact exhibit a very good agreement with the modulation of the 2xCo structures.

The values of $\Delta k$ for the fabricated structures have been also simulated with the transfer matrix formalism, using the same optical and magneto-optical constants detailed in section 2. The obtained results, shown on the right side of Fig. 7, follow the same trends as the experimental ones, including the equivalency of the modulation for the 2xCo series with the difference between those of the $\text{Co}_{\text{bottom}}$ and $\text{Co}^{\text{top}}$ ones.

It is worth noticing that non-reciprocity effects in the plasmonic modes can be further analysed by measuring the TMOKE loops under plasmon excitation (in Kretschmann configuration). Figure 8(a) shows the obtained loops for the samples with 380 nm SiO$_2$ thickness of the three series. All the loops were taken at incidence angles within the peak corresponding to the excitation of the AM mode but to the left of the absolute reflectivity minimum [see dashed line in Fig. 8(b)]. As it can be seen in the TMOKE loops, decreasing the magnetic field from magnetic saturation at positive fields leads to a sudden reduction of the reflectivity for the $\text{Co}_{\text{bottom}}$ sample when the Co magnetization switches, whereas an increase of the reflectivity takes place for the $\text{Co}^{\text{top}}$ sample. Such opposed behavior reflects the non-reciprocal nature of the magnetic modulation of the SPP wavevector, where the same direction of the magnetic field induces a $\Delta k$ of opposite sign for a M/I or an I/M interface, as shown in Fig. 2(b). This is detailed in Fig. 8(b), where we present the angular reflectivity curves for the $\text{Co}_{\text{bottom}}$ and $\text{Co}^{\text{top}}$ structures for the two magnetic orientations of the Co layer. For the $\text{Co}_{\text{bottom}}$ structure, the SPP wavevector is larger when the Co layer is magnetized along the positive direction than when it is magnetized along the negative one. Hence, the minimum of the reflectivity associated to the SPP excitation is located at higher incidence angles when the Co layer is magnetized along the positive direction. Then, for loops taken at incidence angles indicated by the vertical dashed line, the reflectivity decreases when the magnetization state of the Co layer is changed from positive to negative. The opposite behavior occurs for the $\text{Co}^{\text{top}}$ structures. Finally, the structure with two Co layers presents a more complex behavior due to the different switching fields of the bottom and top Co layers. Again decreasing the magnetic field from magnetic saturation at positive fields, we observe first a decrease of the reflectivity, which corresponds to the switching of the magnetization of the Co layer located near the substrate, resulting in an antiparallel alignment of the magnetization of the two layers (top ↑, bottom ↓); then, an increase takes place, which corresponds to the switching of the magnetization of the Co layer located near the surface, coming back to a parallel configuration (↓↑). As expected, the difference between the SPP wavevectors when the two Co layers are oriented along the same direction, ↑↑ or ↓↓ (black and red curves, respectively), is very small, as the contribution to $\Delta k$ from each metal-dielectric interface has opposite sign and the resulting wavevector modulation for the MIM plasmonic mode is very small. On the contrary, the difference in reflectivity when the two Co layers are oriented in opposite directions, ↑↓ or ↓↑ (green and blue curves, respectively), is much higher as both interfaces contribute to $\Delta k$ of the MIM mode with the same sign. The configuration ↑↓ produces a net decrease in the MIM plasmon wavevector while the reverse one, ↓↑, gives rise to an increase.
5. Conclusions

In this work MP-MIM cavities have been designed, fabricated and characterized. A systematic study of the magnetoplasmonic properties of these cavities as a function of both the SiO$_2$ thickness and the position of one or two Co layers inside the MIM structure has been performed. Experiments and simulations demonstrate the possibility of magnetic modulation of the wavevector for the AM and SM plasmonic modes. Our results show that the modulation magnitude depends on the thickness of the insulator, but also on the symmetry of the structure. In cavities with only one Co layer inserted in one metallic cladding, resulting in asymmetric interfaces, a net modulation takes place which increases as the cavity gets narrower for both modes. The increase is very strong for the highly dispersive regions of each mode, which occurs at smaller SiO$_2$ thicknesses for the SM mode than for the AM one. This behaviour has been experimentally confirmed for the AM mode, as it takes place in our accessible thickness range. The strong modulation observed for the plasmonic modes in a MIM configuration makes it an interesting alternative to be considered in plasmonic modulators or in magnetoplasmonic sensing configurations. Additionally, our results show that, in a perfectly symmetric MP-MIM cavity, as those with a Co layer at each metallic cladding, the modulation is cancelled due to non-reciprocity effects. Small asymmetries in the structure or in the modes excitation produce that the modulation is not completely cancelled, but anyway it is reduced compared to the single Co layer, as demonstrated experimentally.

Moreover, by engineering different magnetic dynamics for the different interfaces, the high modulation values for the samples with only one Co layer (Co$_{\text{bottom}}$ and Co$_{\text{top}}$ series) can
be also obtained in the 2xCo samples by simply applying a low magnetic field which allows reaching the magnetization saturation for only one of the Co layers. This offers an opportunity to magnetically switch between low and high modulation of the SPP wavevector in these MP-MIM structures with two Co layers. Finally, an experimental demonstration of non-reciprocity has been put forward in the measured hysteresis loops of the MIM structures under SPP excitation.

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