Magnetic modulation of mid-infrared plasmons using Giant Magnetoresistance

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Abstract: We propose a novel approach to generate active mid IR plasmonic systems by incorporating giant magnetoresistance as the driving mechanism. The magnetic field induced change in resistivity in Ni81Fe19/Au multilayers directly affects the diagonal elements of the dielectric tensor of the system, mainly in the mid IR range, via the magnetorefractive effect. With magnetic fields as small as 50 Oe, we postulate the possibility to modulate the response of such continuous and patterned structures in a contactless, easy to implement fashion. The potential application impact of this modulation concept is analyzed.

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References and links
1. R. Stanley, “Plasmonics in the mid-infrared,” Nat. Photonics 6(7), 409–411 (2012).
2. S. Law, V. Podolsky, and D. Wasserman, “Towards nano-scale photonics with micro-scale photons: the opportunities and challenges of mid-infrared plasmonics,” Nanophotonics 2(2), 103–130 (2013).
3. H. H. Chen, Y. Ch. Su, W. L. Huang, Ch. Y. Kuo, W. Ch. Tian, M. J. Chen, and S. Ch. Lee, “A plasmonic infrared photodetector with narrow bandwidth absorption,” Appl. Phys. Lett. 105(2), 023109 (2014).
4. S. J. Lee, Z. Ku, A. Barve, J. Montoya, W.-Y. Jang, S. R. J. Brueck, M. Sundaram, A. Reisinger, S. Krishna, and S. K. Noh, “A monolithically integrated plasmonic infrared quantum dot camera,” Nat. Commun. 2, 286 (2011).
5. N. Yu, R. Blanchard, J. Fan, Q. J. Wang, C. Pflügl, L. Diehl, T. Edamura, S. Furuta, M. Yamanishi, H. Kan, and F. Capasso, “Plasmonics for laser beam shaping,” IEEE Trans. NanoTechnol. 9(1), 11–29 (2010).
6. H. T. Miyazaki, K. Ikeda, T. Kasaya, K. Yamamoto, Y. Inoue, K. Fujimura, T. Kanakugi, M. Okada, K. Matade, and S. Kitagawa, “Thermal emission of two-color polarized infrared waves from integrated plasmon cavities,” Appl. Phys. Lett. 92(14), 141114 (2008).
7. M. N. Abbas, Ch.-W. Cheng, Y.-Ch. Chang, M.-H. Shih, H. H. Chen, and S.-C. Lee, “Angle and polarization independent narrow-band thermal emitter made of metallic disk on SiO2,” Appl. Phys. Lett. 98(12), 121116 (2011).
8. F. Neubrech, A. Pucci, T. W. Cornelius, S. Karim, A. Garcia-Etxarri, and J. Aizpurua, “Resonant plasmonic and vibrational coupling in a tailored nanoantenna for infrared detection,” Phys. Rev. Lett. 101(15), 157403 (2008).
9. R. Adato, A. A. Yanik, J. J. Amsden, D. L. Kaplan, F. G. Omenetto, M. K. Hong, S. Erramilli, and H. Altug, “Ultra-sensitive vibrational spectroscopy of protein monolayers with plasmonic nanoantenna arrays,” Proc. Natl. Acad. Sci. U.S.A. 106(46), 19227–19232 (2009).
10. G. Armelles, A. Cebollada, A. Garcia-Martín, and M. U. González, “Magnetoplasmonics: Combining magnetic and plasmonic functionalities,” Adv. Opt. Mat. 1(1), 10–35 (2013).
11. V. V. Temnov, G. Armelles, U. Woggon, D. Guzatov, A. Cebollada, A. Garcia-Martín, J.-M. Garcia-Martín, T. Thonon, A. Leitenstorfer, and R. Bratschitsch, “Active magneto-plasmonics in hybrid metal–ferromagnet structures,” Nat. Photonics 4(2), 107–111 (2010).
12. 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(8), 1085–1087 (2006).
13. B. Sepúlveda, L. M. Lechuga, and G. Armelles, “Magnetooptic effects in surface-plasmon-polaritons slab waveguides,” J. Lightwave Technol. 24(2), 945–955 (2006).
14. B. Sepúlveda, Y. Hutel, C. Martínez-Boubeta, A. Cebollada, and G. Armelles, “Linear and quadratic magneto-optical Kerr effects in continuous and granular ultrathin monocristalline Fe films,” Phys. Rev. B 68(6), 064401 (2003).

https://doi.org/10.1364/OE.25.018784
19. Z. Jin, A. Tkach, F. Casper, V. Spetter, H. Grimm, A. Thomas, T. Kampfrath, M. Bonn, M. Kläui, and D. Pecharromán, “Field distribution near optical antennas at the subnanometer scale” in *Importance of the Close Sphere Interaction on the Surface Plasmon Resonance Absorption Peak,* Vol. 25, No. 16 | 7 Aug 2017 | OPTICS EXPRESS 18785, 477–490 (1995).

20. M. Vopsaroiu, T. Stanton, O. Thomas, M. Cain, and S. M. Thompson, “Infrared metrology for spintronic materials and devices,” Meas. Sci. Technol. 20(4), 045109 (2009).

21. R. J. Baxter, D. G. Pettifor, E. Y. Tsybukal, D. Bozec, J. A. D. Matthew, and S. M. Thompson, “Importance of the interband contribution to the magneto-refractive effect in Co/Cu multilayers,” J. Phys. Condens. Matter 15(45), L695–L702 (2003).

22. P. M. Levy, S. Zhang, and A. Fert, “Electrical conductivity of magnetic multilayered structures,” Phys. Rev. Lett. 65(13), 1643–1646 (1990).

23. S. Zhang and P. M. Levy, “Conductivity perpendicular to plane of multi-layered structures,” J. Appl. Phys. 69(8), 4786–4788 (1991).

24. G. S. Krinchik and V. A. Artem’ev, “Magneto-optical properties of Ni, Co, and Fe in the ultraviolet visible, and infrared parts of the spectrum,” Sov. Phys. JETP 26(10), 1080–1085 (1968).

25. M. Z. Alam, J. S. Aitchison, and M. A. Mojahedi, “A marriage of convenience: Hybridization of surface plasmon and dielectric waveguide modes,” Laser Photonics Rev. 8(3), 394–408 (2014).

26. T. Kaihara, T. Ando, H. Shimizu, V. Zayets, H. Saito, K. Ando, and S. Yuasa, “Enhancement of magneto-optical Kerr effect by surface plasmons in trilayer structure consisting of double-layer dielectrics and ferromagnetic metal,” Opt. Express 23(9), 11537–11555 (2015).

27. T. Kaihara, H. Shimizu, A. Cebollada, and G. Armelles, “Magnetic field control and wavelength tunability of SPP excitations using Al₂O₃/SiO₂/Fe structures,” Appl. Phys. Lett. 109(11), 111102 (2016).

28. C. Pecharromán, J. Pérez-Juste, G. Mata-Osoro, L. M. Liz-Marzán, and P. Mulvaney, “Redshift of surface plasmon modes of small gold rods due to their atomic roughness and end-cap geometry,” Phys. Rev. B 77(3), 035418 (2008).

29. C. Pecharromán, “Field distribution near optical antennas at the subnanometer scale” in *Optical Antennas,* Mario Agio ed. Pgs. 197–214, https://doi.org/10.1017/CBO9781139013475.014, (Cambridge University, 2013).

30. C. Pecharromán, “Analysis model for the dielectric response of brine-saturated rocks,” Phys. Rev. B Condens. Matter 34(8), 5145–5153 (1986).

31. D. J. Bergman, “The Dielectric Constant of a Composite Material—a Problem in Classical Physics,” Phys. Rep. 43(9), 377–407 (1978).

32. K. Ghosh and R. Fuchs, “Spectral theory for two-component porous media,” Phys. Rev. B Condens. Matter 38(8), 5222–5236 (1988).

33. R. Fuchs and F. Claro, “Spectral representation for the polarizability of a collection of dielectric spheres,” Phys. Rev. B Condens. Matter 39(6), 3875–3878 (1989).

34. T. Ogawasawa, H. Kawatsu, T. Hasama, and H. Ishikawa, “Proposal of an optical nonvolatile switch utilizing surface plasmon antenna resonance controlled by giant magnetoresistance,” Appl. Phys. Lett. 100(25), 251112 (2012).

1. Introduction

Physical phenomena as diverse as the vibrational transitions of many molecules, or the thermal emission of hot bodies, have in common the fact that their optical response take place totally or most importantly in the mid-infrared (say between 2 – 20 µm) region. These phenomena find a direct connection with our current life since they are used for the detection and quantification of chemical species like gases or liquid contaminants, organic compounds (biologic tissues, polymers, fuels, etc…), or for the remote detection and monitoring of heat sources. As a consequence, this directly translates into the interest in developing mid-IR emitters, detectors and in general mid-IR devices that can be utilized to detect or study these phenomena.

In recent years, plasmonics has proven as a powerful tool to improve the performance of around or above mid-IR devices [1,2]. In this context, noble metals have played a very important role in the development of these systems, certainly due to their ability to sustain...
(plasmon-like) optical resonances in structures such as perforated membranes, engraved metallic layers or arrays of metallic bars, resulting in plasmon enhanced light detection [3,4], plasmonic beam shaping [5], plasmonic thermal emitters [6,7], or plasmonic nanoantennas for vibrational spectroscopy [8,9]. In all these cases, the possibility of modulating the emission, propagation and/or detection of radiation in the Mid-IR and below constitutes a promising scenario to expand the limits of the currently used technologies. Now, to carry out this modulation one has to find physical mechanisms that make possible an external control of these optical resonances. Very recently, a route that allows external, fast and contactless actuation on plasmon resonances has been put forward by the inclusion of ferromagnetic components into noble metal layers and nanostructures, allowing a magnetic field control of the optical properties of the resulting magneto-plasmonic system [10]. Magneto-plasmonic structures have provided excellent results in the generation of active plasmonic platforms, yet up to now restricted to the visible and near-infrared ranges. In these structures it is possible to control the plasmon properties by applying an external magnetic field due to the magneto optical MO activity of the ferromagnet, which is directly related with the non-diagonal elements of its dielectric tensor (see Fig. 1(a), framed in red).

![Fig. 1](image_url)

**Fig. 1.** (a) Magneto Optic and Magneto Refractive effects (MOE and MRE) are related to the non-diagonal and the diagonal elements of the dielectric tensor (framed in red and blue), respectively. (b) When applied along the z axis the magnetic field modifies the wavevector of propagative plasmons via the MOE (c) In systems with GMR a magnetic field applied in the plane modifies the wavevector of propagative plasmons via the MRE.

The Magneto Optic Effect (MOE) has a linear dependence with the magnetization, and has been used in continuous film structures to modulate the wave vector of propagative surface plasmon modes (SPP) [11] due to the magnetization reversal effect on the off diagonal elements on the dielectric tensor (Fig. 1(b)). This property has already found impact in the development of sensors and MO active devices [12,13], improving the performance of currently available non-MO counterparts, mainly due to the modulation character of the detection configuration, which narrows down dramatically the spectral shape of the MO modulated features in comparison to the optical ones. However, a drawback of the MOE on plasmons is that it weakens beyond the near IR, making therefore magnetoplasmonic systems unfeasible for the implementation of active devices in the mid-IR and longer wavelengths ranges.

The former statements in addition to the the reduction in intensity of plasmon resonances at long wavelengths due to the intrinsic overdamping in standard plasmonic metals, make appealing the search for alternative mechanisms that can allow plasmon resonance modulation and as a consequence narrowing down of the characteristic spectral features in this wavelength range. To achieve this goal, one can take advantage for example of the fact that a magnetic field can act, not only on the non-diagonal elements of the dielectric tensor of a ferromagnetic system, but also on the diagonal ones (Fig. 1(a), blue framed), and as a matter of fact in more than one way. A first example of these magnetic field induced diagonal effects is the dependence of the electron dispersion curves with the orientation of the magnetization, which is quadratic with the magnetization [14]. Much more interestingly, many ferromagnetic/non-
ferromagnetic multilayered structures also exhibit strong magnetic field induced changes of the conductivity, the so called Giant Magneto Resistance (GMR), due to the different electrical conductivities characteristic of a parallel or antiparallel configuration of the individual ferromagnetic layer’s magnetizations [15,16]. This second magnetic field induced diagonal effect, known as magnetorefractive effect (MRE) [17], is also quadratic with the magnetization, and interestingly reaches sizable values in the mid-infrared and THz regimes. This phenomenon has actually been extensively used to characterize and carry out contactless tests in a wide variety of GMR systems and devices [18,19], employed for example in hard disk drives or in many other areas of our current life.

It is obvious, therefore, that the MRE can be postulated as a promising counterpart to the MOE to magnetically modulate plasmon properties in the mid IR range. As shown in Fig. 1(c), exploiting the magnetic field control on the diagonal elements of the dielectric tensor in a multilayer exhibiting GMR (distinguishing between in-plane and out of plane due to the 2D nature of the system) it should be possible to modulate for example the propagation characteristics of surface plasmons in a similar fashion than with the MOE mechanism. To proof it, one has to select a model material system, most compatible with the current mid-IR plasmonics strategies, and at the same time exhibiting sizeable values of GMR. Only if this GMR is achievable at low magnetic fields this system may become a realistic candidate for an actual application. For these reasons, and even though there exist a large number of GMR systems at hand (the classic Co/Cu multilayer system for example), for the current proof of concept we have selected the Ni$_{81}$Fe$_{19}$/Au multilayer structure, due to its well-balanced characteristics in all the factors of relevance: relative large GMR values, of up to 12%, along with saturation magnetic fields as low as 50 Oe [20]. In addition, Au is an excellent plasmonic material from the optical range up to wavelengths of around some microns.

With all this in mind, this work has been structured as follows: We first introduce the Ni$_{81}$Fe$_{19}$/Au multi-layered model system, the fabrication details and the main structural, magnetic and magnetotransport properties. The experimentally determined magnetic field dependent optical properties in the mid IR will be presented. Actually, and in spite of the large set of GMR systems whose magnetorefractive properties have been considered, no data on the IR optical and magneto-optical properties of this system have been published so far. Then, and after extracting the corresponding dielectric constants for this spectral range, a conceptual comparison of the plasmon modulation via MOE and MRE mechanisms will be developed, showing the better performance of the later for wavelengths above 4 microns. Finally, two proposals for the application of this magnetorefractive mid-IR plasmon modulation for systems exhibiting propagating and localized resonances will be foreseen.

2. Results and discussion

The layers were grown on CaF$_2$(111) single crystalline substrates. After transfer into the ultrahigh vacuum deposition system and outgas for 30 minutes at 150°C, a 2 nm Ti buffer layer was grown by electron beam evaporation. Then, 12 layers of Ni$_{81}$Fe$_{19}$ spaced by 11 layers of Au were deposited by magnetron sputtering from individual Ni$_{81}$Fe$_{19}$ and Au targets at an Ar pressure of 10$^{-3}$ mbar. Typical thickness for Ni$_{81}$Fe$_{19}$ and Au individual layers are 2.9 and 2.4 nm respectively, for which in our specific case the best GMR values were obtained. A final 5 nm thick Au layer was deposited on top of the multilayer to prevent from oxidation. The substrate temperature was kept at 150°C during the whole process.

X-ray reflectometry measurements allowed a precise determination of the individual layers thickness and interface roughness. Transverse MO Kerr hysteresis loops were recorded to confirm magnetization reversal process and magnetic fields needed to saturate the multi-layered structure. A green laser source and a magnetic field oscillating at 1 Hz with usually 50 averaged loops were used for this purpose. DC Magneto resistance measurements were performed using 4 probes in line with typically 2 mA input current and enough magnetic field to fully saturate the samples. The multilayers were then characterized in the IR using a Bruker VERTEX 70
FTIR spectrometer equipped with a MCT photovoltaic detector and magnetic coils in the sample’s enclosure, generating sufficient magnetic field to saturate the structures. In order to determine the spectral response of localized surface plasmon resonances, numerical solution of the electromagnetic problem was obtained by using the COMSOL 5.2 software package.

2.1 MRE in the Ni$_{81}$Fe$_{19}$/Au multilayers

Figure 2(a) shows the magnetic field dependent change in DC electrical resistivity ($\rho$) normalized to its resistivity at zero field for a multilayer exhibiting GMR. As it can be observed, a quadratic behaviour with the magnetization and a relative change of resistance of 4.8% for magnetic fields as low as 50 Oe are obtained. Once the presence of GMR is confirmed, we proceeded with the optical characterization in the mid-IR range. In the inset to Fig. 2(b) we present the spectral dependence of the transmission at normal incidence for such structure. As it can be seen, the transmission, of the order of $10^{-3}$, gradually decreases as the wavelength increases as expected for the considered metallic structure. As previously explained, the high resistance state in remanence characteristic for the antiparallel magnetic alignment of the individual Ni$_{81}$Fe$_{19}$ layers turns into a low resistance state upon application of a magnetic field due to the parallel magnetic alignment of the layers. This change in resistance manifests mainly in the IR in the optical properties due to the MRE, as can be seen in Fig. 2(b), where we show the relative change in the transmission ($\Delta T/T = T(M_{sat})-T(0)$) at normal incidence induced by an applied magnetic field for two different Ni$_{81}$Fe$_{19}$/Au multilayers with different GMR values (4.8% and 0.5%). The magnetic field was applied in plane and the incident light was not polarized. As it can be observed, and unlike the spectral behaviour of the transmission, the value of $\Delta T/T$ increases as we increase the wavelength and shows a maximum at around 9 µm, slightly decreasing for larger wavelengths. Very clearly, the magnitude of $\Delta T/T$ is larger for the multilayer which has a higher value of GMR, indicating the relation between both effects. This is unambiguously corroborated when comparing the magnetic field dependence of $\Delta T$ integrated over the whole spectral range, defined as:

$$\Delta T(H) = \frac{\int_{\lambda_1}^{\lambda_2} (T(\lambda,H) - T(\lambda,H_{max})) d\lambda}{\int_{\lambda_1}^{\lambda_2} (T(\lambda,0) - T(\lambda,H_{max})) d\lambda}$$

and the GMR curve. This is plotted in Fig. 2(a), showing a perfect overlap of the magnetic field dependencies of $\Delta T$ and the magnetoresistance ($\Delta \rho/\rho$).

Fig. 2. (a) Magnetic field dependent change in DC resistivity normalized to the zero field resistivity (continuous line) and the corresponding change in integrated change in IR transmission (symbols) for a Ni$_{81}$Fe$_{19}$/Au multilayer exhibiting GMR. (b) Spectral dependence of the relative change in transmission for two multilayers with 4.8% (black) and 0.5% (red) GMR respectively. The inset shows the spectral dependence of the transmission for the 4.8% GMR multilayer. (c) Spectral dependence of real and imaginary parts of the dielectric constant for the 4.8% GMR multilayer. The inset shows the magnetic field induced change in these constants for the same multilayer.
2.2 Magnetic field dependence of the diagonal elements of the dielectric tensor

With these results in hand, it is possible now to have an estimation of the diagonal elements of the dielectric tensor for the GMR multilayer as well as the changes in these diagonal elements induced by the applied magnetic field. For this, we assume that, since the thickness of the Ni$_{81}$Fe$_{19}$ and Au layers is much smaller than the wavelength of light, the optical properties of the multilayer can be described by an effective dielectric tensor, which depends on the relative orientation of the magnetization of the Ni$_{81}$Fe$_{19}$ layers:

For parallel (P):

\[
\begin{pmatrix}
\varepsilon^P_{//} & 0 & 0 \\
0 & \varepsilon^P_{//} & 0 \\
0 & 0 & \varepsilon^P_{\perp}
\end{pmatrix}
\]  

(2)

and for antiparallel (AP):

\[
\begin{pmatrix}
\varepsilon^{AP}_{//} & 0 & 0 \\
0 & \varepsilon^{AP}_{//} & 0 \\
0 & 0 & \varepsilon^{AP}_{\perp}
\end{pmatrix}
\]  

(3)

being $\varepsilon^{P,AP}_{//}, \varepsilon^{P,AP}_{\perp}$ the effective dielectric constants in the plane and perpendicular to the plane, respectively. At normal incidence only the in-plane components of the dielectric tensor, $\varepsilon^{P,AP}_{//}$, contribute to the optical properties. On the other hand, in this spectral range the main contribution to the dielectric constants comes from the conduction electrons, which, due to the dependence of the relaxation time of the electrons with the relative orientation of the spin of the electron and the magnetization of the multilayer, is different for the P, and AP states. To have a complete description, and as has also been observed in other metallic multilayer systems [21], interband transitions need also to be taken into account, whose contribution also depends on the magnetic state of the multilayer. After this consideration, and adjusting the conduction electron contribution of the AP and P magnetic states to reproduce the magnetoresistance values, we can estimate the $\varepsilon^{P,AP}_{//}$ and $\varepsilon^{P,AP}_{\perp}$ dielectric constants of the multilayer using the experimental $T$ and $\Delta T/T$ spectra. In Fig. 2(c) we present respectively the spectral dependence of the resulting real and imaginary parts of the dielectric constants (averaged for their parallel and antiparallel components) and the difference between the parallel and antiparallel components for the sample with 4.8% GMR (inset). These results show a clear metallic character and that the difference between $\varepsilon^{P}_{//}$ and $\varepsilon^{AP}_{//}$ increases as we increase the wavelength.

2.3 Mid IR plasmon modulation using the MRE

Let us now evaluate the magnitude of the magnetic field induced changes in the plasmon properties caused by the MRE. We first analyse its effect on the magnetic modulation of the wave vector of a SPP mode and compare it with that obtained using the MOE. The changes in the wavevector of a surface plasmon mode propagating at the vacuum /multilayer interface induced by the changes in the relative orientation of the magnetization of the layers can be written as:

\[
\frac{\Delta k}{k} = f_{MR//} \frac{\Delta \varepsilon_{//}}{\varepsilon_{//}} + f_{MR\perp} \frac{\Delta \varepsilon_{\perp}}{\varepsilon_{\perp}}
\]  

(4)

where $f_{\text{RM}||}$ and $f_{\text{MR}\perp}$ are parameters which depend only on the optical properties of the material:
\( f_{MR} = \frac{(\epsilon_{\perp} - 1)\epsilon_{\parallel}}{2(\epsilon_{\parallel} - 1)(\epsilon_{\perp} - 1)}; f_{MRx} = \frac{-1}{2(\epsilon_{\parallel} - 1)} \)  

\( f_{MO} = \frac{\epsilon_{MO} M_s}{\epsilon} \)  

In the far infrared range the main contribution to the optical properties comes from the conduction electrons, and the difference between \( \epsilon_\parallel \) and \( \epsilon_\perp \) is related to the difference in the scattering time of the conduction electrons. In our structures, due to the thickness of the Au and Ni81Fe19 layers and the contribution of the interfaces to the scattering, the in-plane and perpendicular scattering times should be very similar [22,23], therefore \( \epsilon_\parallel \approx \epsilon_\perp \). Moreover, in the infrared range \(|f_{MR}| \gg |f_{MRx}|\) and expression (4) can be simplified to:

\[ \frac{\Delta k}{k} = f_{MR} \frac{\Delta \epsilon}{\epsilon} \]  

where \( \Delta \epsilon = \Delta \epsilon_\parallel = (\epsilon_\parallel^p - \epsilon_\parallel^{AP}) \), \( \epsilon = \epsilon_\parallel \), and \( f_{MR} = \frac{\epsilon}{2(\epsilon^2 - 1)} \).

On the other hand [Fig. 1(b)], a magnetic field applied in the plane of the multilayer and perpendicular to the propagation direction of the SPP mode also changes its wavevector as:

\[ \frac{\Delta k}{k} = f_{MO} \frac{\epsilon_{MO}}{\epsilon} M_s \]  

here, \( \epsilon_{MO} \) is the non-diagonal element of the dielectric tensor and \( f_{MO} \) a parameter which depends only on the optical properties of the material,

\[ f_{MO} = \frac{\sqrt{\epsilon}}{(1 - \epsilon^2)} \]

In Fig. 3(a) we present the wavelength dependence of \(|\Delta k/k|\) for the two cases \((M_s = 1)\). These curves were obtained using the dielectric constants shown in Fig. 2(c). For the non-diagonal element of the effective dielectric tensor of the multilayer, \( \epsilon(Ni_{81}Fe_{19}/Au)_{MO} \), we have assumed that it is proportional to the amount of Ni81Fe19 in the multilayer. \( \epsilon(Ni_{81}Fe_{19})_{MO} \) was obtained from the far infrared values of Fe and Ni given in [24] and taking into account the relative amount of Fe and Ni in the alloy \( \epsilon(Ni_{81}Fe_{19})_{MO} = 0.81*\epsilon(Ni)_{MO} + 0.19*\epsilon(Fe)_{MO} \). As it can be observed, as we increase the wavelength, the magnetic field modulation of the surface plasmon wavevector due to the magnetorefractive effect (black curve) is larger than when it is due to the non-diagonal element of the dielectric tensor (red curve). One of the reasons of this behaviour is the difference between \( |f_{MR}| \) and \( |f_{MO}| \), which reflects the relative strength between these two effects. The wavelength dependence of the modulus of these two parameters is
presented in Fig. 3(b) and clearly show that, as far as the magnetic modulation of the wavevector in concerned, the MRE (black curve) is always more effective than the MOE (red curve). Moreover, the relative change of the optical properties, induced by the magnetorefractive effect, $|\Delta \varepsilon/\varepsilon|$, shown in Fig. 3(c) as a black curve, increases as we increase the wavelength, whereas a very small dependence with the wavelength is observed for the MOE constant $|\varepsilon_{MO}/\varepsilon|$ (red curve), resulting in the global wavelength dependence of $|\Delta k/k|$ given in Fig. 3(a).

3 Potential applications

3.1 Propagative waves

The possibility of using the magnetorefractive effect as a way to modulate the surface plasmon properties opens the door to new developments such as: magnetic modulation of the wavelength and/or of the intensity of the transmission bands in extraordinary optical transmission (EOT) metallic gratings used for plasmonic enhanced mid-IR light detectors, or magnetic modulation of the propagation properties in plasmonic waveguides and/or plasmonic lenses used in quantum cascade lasers (QCLs). As an example, we consider a system resembling the basic structure of a hybrid plasmonic waveguide: a high refractive index layer separated from a metal surface by a low refractive index spacer [25] [Fig. 4(a)] for which we present the reflectivity curves as a function of the wavelength [Fig. 4(b)].

![Fig. 4. (a) Magnetic modulation in hybrid-plasmonic waveguides can be achieved in a configuration where a GMR multilayer is separated from a high refractive index prism by a low refractive index spacer. (b) Clear magnetic field induced (red to black) shift of the reflectivity at the cut-off of the waveguide mode. The inset show the spectral dependence of the reflectivity. (c) Sensitivity of the reflectivity of this structure to a change in the refractive index of the spacer of 10$^{-5}$. The position of this minimum in $R$ can be tuned with the spacer thickness (inset). (d) Comparative sensitivity to the same change in refractive index of the magnetic field modulated reflectivity, at the adequate incidence angle, showing a sharp, derivative-like line shape.](image_url)
In our case we use a Si prism, a 15.1 microns thick $n = 1.4$ spacer layer, and a thick Ni$_{81}$Fe$_{19}$/Au multilayer with the properties shown in the previous section corresponding to a 4.8% GMR. In this structure, the minimum of the reflectivity occurs at the cut-off frequency of the waveguide mode [26,27] and, as it can be observed in the Fig., the cut-off frequency can be modulated by the magnetic field. Moreover, due to the strong localization of the light inside the spacer, the minimum of the reflectivity is very sensitive to the optical properties of the spacer and, in particular, to the optical properties near the metallic surface. Therefore, this configuration could be used for gas sensing applications, by just substituting the spacer layer by a gas cell. For example, in Fig. 4(c) we present the reflectivity curves for 19.5 microns thick spacer layer for $n = 1$ and $n = 1.00001$. As it can be observed, the minimum of the reflectivity is very sensitive to the optical properties of the spacer, with a clear redshift for an increase of $n$ as small as $10^{-5}$. On the other hand, in Fig. 4(d) we also present the magnetic field induced change of the reflectivity, $\Delta R/R$, due to the magnetorefractive effect of the Ni$_{81}$Fe$_{19}$/Au multilayer. As it is clearly seen, due to the derivative like character of this magnitude, the $\Delta R/R$ curve is much sharper than the reflectivity curve, overcoming intrinsic broadening issues for noble metals in this spectral region, and being as a consequence even more sensitive to changes in the refractive index than the reflectance. Therefore, the magnetorefractive effect could be used to increase the sensitivity of sensors in the far infrared range, in a similar way as does the MOE effect in the visible range (MOSPR sensor) [12]. Moreover, due to the dependence of the spectral position of the minimum with the thickness of the spacer (see right inset of Fig. 4(c)) we can tune the position of the minimum to match it with molecular absorption bands, which will further increase the sensitivity to this particular molecule in this proposed platform.

3.2 Nanoantennas

![Fig. 5. Resonant nanoantennas. (a) Left: Sketch of the considered system and parameters used to calculate by Finite Elements the depolarization factor $L_x$. The studied particle, which dimensions $a$ and $b$, and permittivity $\eta$ lies on a substrate of dielectric constant $\varepsilon_{\text{sub}}$. The simulation volume is much larger than the particle size ($b<<l_2$ and $a<<l_3$). Right: calculated $L_x$ as a function of axial ratio for two different substrates. (b) Calculated imaginary parts of the dielectric tensor of the rod layer for different axial ratios and two substrates.](image)

Magnetorefractive effect can also be applied on the modulation of localized resonances of optical antennas. As an example, we analyse the effect that the switch from AP to P magnetic
states in the Ni$_{81}$Fe$_{19}$/Au multilayer exhibiting 4.8% GMR has on the optical properties of rods made of this material and deposited on top of a substrate [Fig. 5(a)]. Worth to mention, internal demagnetizing fields and magnetostatic interactions may modify the effective GMR values when patterning the layer stack down to micrometer-nanometer size antennas. As these effects depend on the specific antenna dimensions and strength of the interlayer coupling responsible for the AP state, in our simulations we have opted to simply consider the optical constants for the AP and P states of our model materials system.

Oppositely to the case of a propagative plasmon, the model for a localized plasmon is properly defined when dimensions of the rods are smaller than the wavelength of the incident light, i.e. the quasistatic approximation. In this regime, effective medium models can be fully applied to determine the electromagnetic response of the system. In the particular case of high dilution, or when the rods do not interact via multipolar terms, effective models can be expanded in a Taylor series and share the same expression for $\langle \varepsilon_x \rangle$ given by

$$
\langle \varepsilon_x \rangle = \left(1 + \phi \frac{\varepsilon_x(Ni_{81}Fe_{19}/Au) - 1}{(1 - L_x) + L_x \varepsilon_x(Ni_{81}Fe_{19}/Au)}\right)
$$

Where $\varepsilon_x(Ni_{81}Fe_{19}/Au)$ is the experimentally determined in-plane effective dielectric constant of the Ni$_{81}$Fe$_{19}$/Au layers, $\phi$ the volume concentration of the rods and $L_x$ the effective depolarization factor along the rod axis, which takes into account, not only the shape of the rods, but also the effect of the substrate. For any other geometry ellipsoidal shape, $L_x$ should be obtained by means of an integral equation [28–30]. In principle, the presence of a third medium (air, metallic rod and dielectric substrate) makes the two media effective dielectric constant model (Eq. (9)) invalid. However, according to the geometry of Fig. 5(a) the particle and the substrate interact creating image charges, which can be identified as a virtual particle embedded into the substrate.

To estimate the effective depolarization factor, we have the so-called “Spectral Representation Function” procedure [31–34] (SRF or $g(\alpha)$). This method gives an exact value of the effective dielectric constant if we have accurate geometric information about the geometry of the system, given by the SRF which can be understood as a distribution function of all the depolarization factors of the particles contained in the system. Actually, Eq. (9) is the effective dielectric constant of a composite whose spectral representation function is a Dirac’s delta. One of the important properties of this function is that, if we have a method to determine $\langle \varepsilon_x \rangle$, the SRF can be analytically calculated by the Stieltjes inversion formula [30]:

$$
(\alpha) = \frac{1}{\pi V} \lim_{\delta \to 0} \text{Im} \left( \frac{\langle \varepsilon_x \rangle(\alpha, \delta, \varepsilon_m) - 1}{\varepsilon_m} \right)
$$

Where $\varepsilon_m$ is the dielectric constant of the external medium (in this case, $\varepsilon_m = 1$), $\langle \varepsilon_x \rangle(a, \delta, \varepsilon_m)$ is the effective value of the dielectric constant calculated in the region shown in Fig. 5a, $\delta$ is a small value to avoid singularities in the complex variables and $\alpha$ is a parameter given by:

$$
\frac{\varepsilon_m}{\varepsilon_m - \eta} = \alpha + i\delta \Rightarrow \eta = \varepsilon_m \left(1 - \frac{1}{\alpha + i\delta}\right)
$$

In Eq. (11), $\alpha$ is a continuous variable which runs from 0 to 1, so that the resulting magnitude $\eta$ and plays the role of a dielectric constant and varies from $-\infty$ to 0. In order to estimate the value of the effective dielectric constant $\langle \varepsilon_x \rangle$ we have employed Finite Element software (COMSOL). We have introduced the geometry as it appears in Fig. 5(a) but we have substituted the rod by a virtual material, with dielectric constant $\eta(a, \delta)$ ranging from $-\infty$ to 0, in a very similar way as the same property of a plasmonic metal. The COMSOL results were employed to determine the imaginary part of the average depolarization factor along the applied field ($E_0$), so that the effective dielectric constant is determined by:
This value is usually very low with the exception of some resonances which makes very simple to find the main depolarization factors. The value of $\alpha_{\text{max}}$ at which the largest resonance occurs (which is several orders of magnitude larger than the second) corresponds to the main depolarization factor $\alpha_{\text{max}} = L_x$. The depolarization factor obtained in such way (which depends on the particle geometry and on the dielectric constant of the substrate) can be employed to find the particle surface plasmon resonance using Eq. (9). The resulting effective depolarization factor for rods with a square section deposited on top of a CaF$_2$ and Si substrate, respectively, are presented in Fig. 5(a) as a function of the axial ratio (ratio between the long ($b$) and short axis ($a$) of the rod $b/a$). As it can be observed the effective depolarization factor decreases as we increase the axial ratio. On the other hand, the effect of the substrate is clearly seen: the depolarization factor decreases as we increase the refractive index of the substrate. In Fig. 5(b) we present the imaginary part of the effective dielectric constant along the direction of the applied field. We have assumed a 10% rod concentration. Each curve stands for rod lengths from 0.5 to 1.5 microns deposited on top of CaF$_2$ and Si substrates (the rod section is 50x50nm$^2$, and the axial ratio varies from 10 to 30). This effective dielectric constant shows a peak corresponding to the resonance of the rods. As expected, the frequency of the resonance decreases as we increase the length of the rod, and for the same rod length the resonance is located at lower energy when the rods are deposited on top of a Si substrate than when they are deposited on top of a CaF$_2$ substrate. Moreover, as the peak shifts toward lower energy it broadens and its intensity also increases due to the increase of the losses of the Ni$_{81}$Fe$_{19}$/Au layers.

With this materials system established, in Fig. 6(a) we present the transmission spectra for the same rods layers shown in Fig. 5(b) deposited on top of CaF$_2$ or Si substrates. As it can be seen, the spectra show a broad feature in the region of the rod resonances shifting towards longer wavelengths as the length of the rod increases, and being more pronounced for the Si substrate.

\[
\langle \epsilon_r \rangle = \frac{\int_V \text{Im}(D_x) \, dV}{\int_V E_0 \, dV}
\]

\( (12) \)
In addition, the corresponding magnetic field modulation effects are shown in Fig. 6(b), where we present the relative variation of transmittance under the effect of the magnetic field (named as $\Delta T/T$) due to the change from AP to P state. In contrast with the broad transmittance spectra, due to the overdamped nature of the dielectric constant of the bars at these wavelengths, the magnetic modulation of the transmittance [Fig. 6(b)] presents very well defined maxima and minima for any aspect ratio, with a zero crossing at a wavelength which approximately coincides with the resonance of the rods. All these features exhibit redshift behaviours with increasing aspect ratios similarly to the transmittance spectra. Worth to mention, the magnetic modulation of the transmittance increases notably for longer axial ratios. A similar behavior has also been proposed for arrays of nanorods made from Co/Cu GMR multilayers calculated using FDTD [35].

4. Advantages of a magnetic modulation technique

Almost all of the IR spectroscopies are nowadays done by FTIR (Fourier Transform IR) instead of dispersive gratings. While gratings allow implementing a modulation experimental set-up, in spectrophotometers based on Fourier Transformation is not straightforward. In this sense, the magnetoresistive modulation of the plasmonic material can be used to improve the traditional measurement method in comparison with standard, not modulated, FTIR platforms.

Customary, IR spectra are recorded by measuring an empty background spectrum prior to the sample data, so that it is necessary to manipulate the sample and its environment, with the consequent change in measurement conditions. Therefore, usually this process requires three measurements (we will call them $S_e$, $S_0$, and $S_B$, i.e: empty, sample without field, and sample under magnetic field respectively) and is actually one of the main sources of experimental error in IR spectroscopy, especially if the signal is small so that long times are required to improve signal to noise ratios. Instead, we propose to eliminate the $S_e$ measurement. By doing that, sample and background spectra can be taken without manipulating the experimental setup, and consequently the reproducibility of the measurement largely improves. In the presented measurement setup, the variation of the permittivity due to the magnetic modulation introduces enough contrast to obtain a signal difference large enough to be measured. Actually, the obtained signal in Fig. 6(b) is given by:

$$\frac{\Delta T}{T} = \frac{T_B - T_0}{T_0} = \frac{T_B}{T_0} - 1$$

where $T_B = S_B/S_e$ and $T_0 = S_0/S_e$.

Under the new presented experimental procedure, with only two measurements, we take the reference spectrum with the sample mounted on the sample holder but in absence of magnetic field ($S_0$). Without any manipulation of the sample and the environment, the magnetic field is applied to take the second spectrum ($S_B$). The commercial FTIR spectrophotometers customary do the quotient of both set of data to produce a transmission spectrum. In this case, this function is just:

$$\frac{S_B}{S_0} = \frac{T_B}{T_0} = 1 + \frac{\Delta T}{T}$$

This procedure has several advantages. In the first place, even when the total transmittance is very low, the magnitude $\Delta T/T$ could be measurable but more importantly, because $S_0$ and $S_B$ can be taken nearly consecutively, thermal variation of the experimental setup can be disregarded, so that, multiple $\Delta T/T$ spectra can be taken and accumulated in a nearly indefinite way, in order to increase the signal to noise ratio. Besides, the position of the rod’s resonance depends on the dielectric environment around the rod. Therefore, the modification of this
dielectric environment, because of, for example, the absorption of molecules, also induces a change in both $T$ and $\Delta T/T$; but, due to the derivative-like character of $\Delta T/T$, this magnitude is more sensitive to these absorption events than $T$, giving rise to a better sensing capability.

5. Conclusions

We have presented a novel route to achieve mid IR plasmon modulation by using the magnetorefractive effect as the driving mechanism. This is due to the change of optical constants in the mid IR range experienced by multilayers exhibiting giant magnetoresistance upon switching between the two available high and low electrical resistivity states. The plasmonic magnetorefractive transducer consists of a $\text{Ni}_{81}\text{Fe}_{19}/\text{Au}$ multilayer thin film structure which exhibits a 4.8% change in electric resistivity using magnetic fields as low as 50 Oe. With this, we envision promising applications in platforms using propagating and localized resonances, with sharp magnetic field induced resonances that make it possible, by instance, high refractive index sensitivity changes. In addition, the magnetic field external actuation and the purely optical nature of the measurement make this proposed modulation method a contactless and easy to implement alternative to modulation approaches that require patterning and electrical contacts. Finally, the implementation of magnetic modulation in a standard FTIR platform allows for an important improvement in the signal to noise ratio and reproducibility of the measurements, due to the modulation character of the measurement, and to minimize the number of successive sample manipulations for signal and background acquisitions, which can be of interest in the community using FTIR systems.

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