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Surface plasmon excitation on magnetoactive materials

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A systematic study of the excitation of surface plasmons on ferromagnetic materials in multilayered structures composed of thin films of nickel, iron, and cobalt capped by a silver layer is presented. Electromagnetic properties of the systems are theoretically and experimentally investigated as a function of the metal layers' thickness. The critical parameters in this study of the interaction between surface plasmon waves and the magnetoactive material are discussed and an optimized structure for the investigations of spin-plasmonic effects is proposed.

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The interaction of surface plasmons (SPs)—fluctuations in the electron density at the interface between media with dielectric constants of opposite sign[1,2]—with magnetically ordered systems has attracted a lot of interest in the last ten years. The importance of this interaction has been proved, for instance, by the demonstration of a SP enhancement of magneto-optical properties such as the Faraday and Kerr effects.[3–6] Typically, in these experiments, a few nanometers of ferromagnetic material are sandwiched between two layers of silver or gold. As the result of the high optical losses of ferromagnetic metals,[7] multilayered structures need to be implemented for the excitation of SPs. The incoming electromagnetic wave, otherwise, weakly couples to the collective electron oscillations in the metal, with a consequent non-efficient excitation of surface plasmons. Reflectivity measurements thus show broad and weak absorption peaks[8–10] if compared to typical results which can be obtained in silver-based or gold-based systems, in the visible range of wavelengths.[1,11,12] Sharp features in the SP resonances are instead required, for instance, to be able to measure small angular shifts of the plasmonic resonance due to magnetically induced variations in the optical constants of the layers. The high sensitivity of surface plasmons to small variations in the dielectric constants has been widely implemented in devices, for example, plasmonic-based sensors[13] (for a review, see Ref. 14). The study of the excitation of SPs on ferromagnetic materials has recently attracted a lot of interest not only from a fundamental research point of view but also for the possibility of realizing spin-plasmonic devices, joining together the rapidly developing plasmon technology[15,16] and the field of spintronics.[17] A first proposal for a spin-plasmonic device was reported in 2004. The properties of a gigahertz magnetoplasmon optical modulator based on a bismuth-substituted yttrium iron garnet system are studied, showing the possibility of achieving a high-speed magneto-optic modulation of improved efficiency compared to bulk devices.[18] This garnet is characterized by relevant magneto-optical properties [with Faraday rotations as high as $(1°/\mu\mathrm{m})$]; however, the high absorptions of this material in the visible range of wavelengths represent one of the major limitations in its application in devices.[19] In 2007, the first experimental evidence of a spin-dependent transport in spin-plasmonic media was given, showing a magnetic manipulation of near-field-mediated light transport via the electron spin.[20] In 2008, a near-field study of vortex surface modes with a spin-dependent topological charge together with a polarization sensitive focusing in a plasmonic structure was reported.[21] This emerging field of research focused on spin effects on plasmonic structures is thus rapidly developing; however, to our knowledge, a systematic study of the excitation of SPs in ferromagnetic metal-based structures is still lacking.

Here, we present a theoretical and experimental study of the optical properties of multilayered structures composed of ferromagnetic materials (Co, Fe, and Ni) and silver and demonstrate the potentials of these systems in the investigation of plasmon-assisted magneto-optical effects. A theoretical treatment based on the solution of the electromagnetic Green's functions for layered structures[22] written in terms of 2×2 matrices[23] has been implemented to simulate the reflectivity and electromagnetic properties of the systems under study. In order to realize structures suitable to become a platform for the investigation of magnetic-induced effects in plasmonic waves, the study of the propagation depth of the electromagnetic field in the ferromagnetic layer is necessary: the overlap between the plasmonic field and the magnetoactive material needs to be maximized.

The samples composed of ferromagnetic thin films capped by a silver layer are thermally evaporated on a glass slide. The silver layer protects the magnetic material from oxidation and—more importantly—allows an efficient excitation of plasmonic waves at the silver-air interface. The optical properties of the samples are studied as a function of the layers' thickness and the surface plasmon excitation is achieved in a standard Kretschmann configuration,[1] using a fused silica prism and a He-Ne laser, emitting at 632.8 nm, as the excitation source.

Here, we present the results obtained on samples with a total metal thickness of 50 nm composed of glass/ferromagnetic layer (z)/Ag(50 nm-z)/air, where z is the thickness of the ferromagnetic layer in nanometers. The accuracy in the deposition is within 2 nm and an angular resolution of 0.18° is achieved by placing a 0.25 mm slit before the $f/2$ lens, focusing on a silicon photodiode detector. In Fig. 1, reflectivity measurements of three samples composed of a 10 nm bottom layer of either Co, Fe, and Ni capped with a 40 nm Ag layer are shown. These systems present a change in reflectivity of about 80% (measured from the baseline) of the p-polarized incident light intensity. The full width at half maximum (FWHM) measured at half the reflectivity dip of

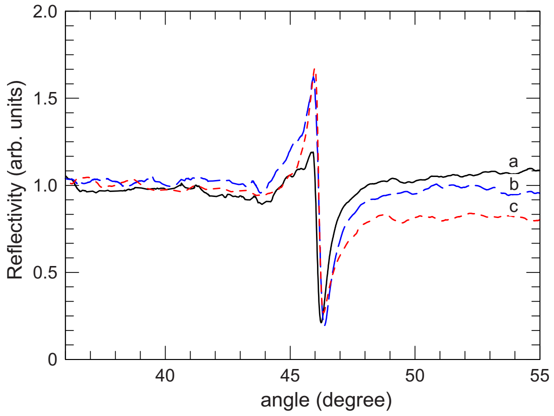

FIG. 1. (Color online) Reflectivity spectra obtained with *p*-polarized illumination and normalized to the baseline of 10 nm Ni [(a) solid line], Fe [(b) long dash line], and Co [(c) short dash line] films capped with a 40 nm Ag layer.

the absorption lines varies from 0.5° for the Ni-based sample to 1° for the Co one. Considering the optical losses for the different ferromagnetic materials at the wavelength under study—reflected by the value of the imaginary part of the dielectric constant (values taken from Refs. 7 and 24)—the best coupling between the incoming radiation and the plasmonic oscillations on the metallic surface is expected for the Ni-based sample. We thus focus our attention on this ferromagnetic material and further investigate the reflectivity of this system as a function of the ferromagnetic layer thickness z, as shown in Fig. 2(a). A good agreement between the simulated spectra [Fig. 2(b)] and the experimental results is found. The discrepancies in the absorption intensity are attributed to the nonideality of the interfaces and to the possible presence of oxidation (which would result in deviations from the pure metal optical constants used in the simulations) on the metals' surfaces. As concerns the angular position of the resonances, a difference of less than 0.8° between theory and experiment is found. This can be explained taking into account a systematic error associated with the manual calibration of the zero position of the prism (evaluated to be of the order of 0.5°) as well as considering, once again, the possible presence of residual oxidation layers on the metals.

The same measurements have been carried out for the Fe-based and Co-based samples and the results are summarized in Fig. 3, where the linewidths (FWHM) and absorption percentages are plotted as a function of the layers' thickness (where points are not presented, the broadness or low intensity of the absorption peaks does not allow an unambiguous evaluation of these quantities). The importance of the silver layer on the top surface in the excitation of the surface plasmon is evident. By reducing the silver thickness, the optical losses of the ferromagnetic layers take over and the interaction between the incident light and the electronic oscillations is dramatically reduced. Although Fe-based samples show absorptions comparable to the Co and Ni ones for a thickness of 10 nm layer, the plasmonic absorption rapidly collapses into a broad and low peak when increasing the Fe content, as a result of the high optical losses of this metal (reflected by the high imaginary part of its dielectric constant). Co-based structures, on the other side, while presenting comparable absorption percentages, present larger linewidths than Ni-based samples: this implies a wider angu-

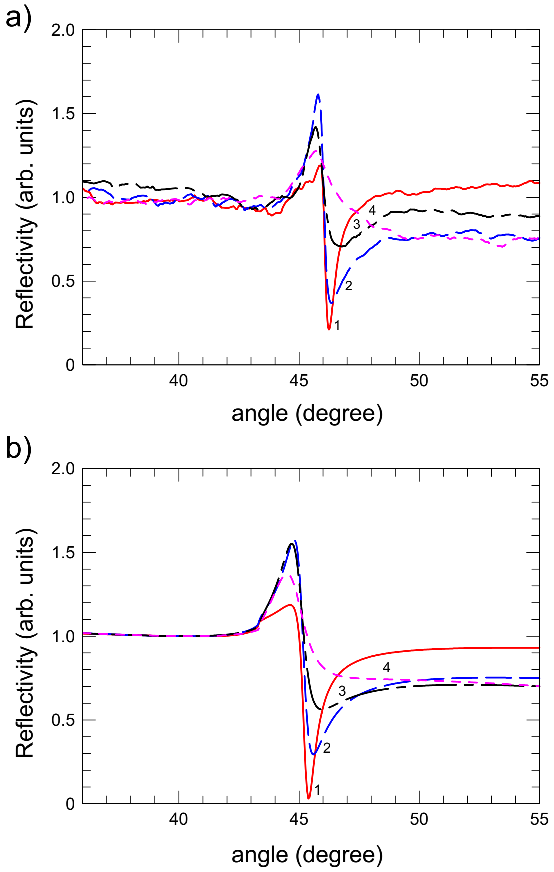

FIG. 2. (Color online) (a) Reflectivity spectra obtained with *p*-polarized illumination and normalized to the baseline of Ni(z)/Ag(50 nm-z) samples, where z is the layer thickness in nm (spectrum 1: solid line corresponds to $z=10$ nm; spectrum 2: long dash line to $z=20$ nm; spectrum 3: short dash/long dash line to $z=30$ nm; and spectrum 4: short dash line to $z=40$ nm). (b) Simulation of the reflectivity spectra presented in panel (a).

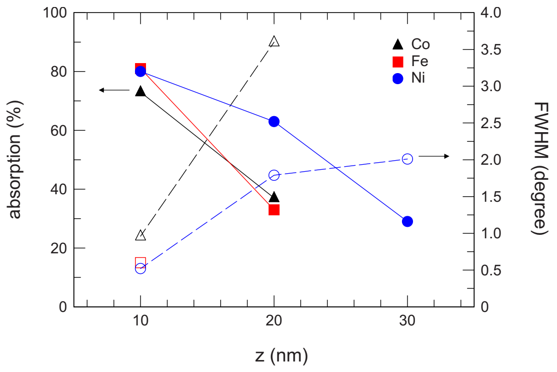

FIG. 3. (Color online) Experimental absorption percentage (full symbols) and full width at half maximum (open symbols) of the reflectivity spectra for Co, Fe, and Ni(z)/Ag(50 nm-z) samples, after normalization to the baseline.

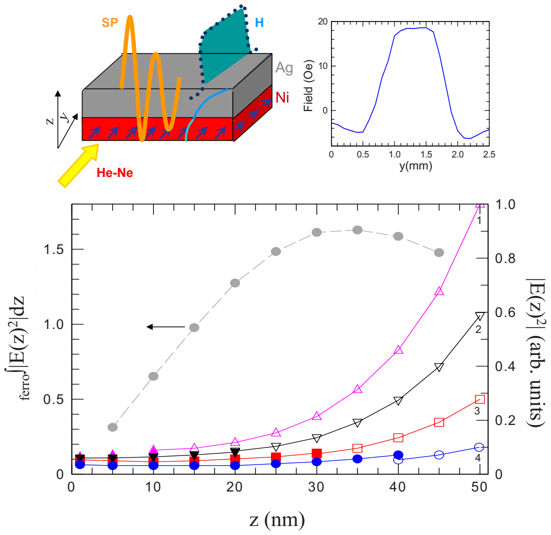

FIG. 4. (Color online) Top left: Schematic view (not to scale) of the system. Top right: Magnetic-field intensity, along the y axis, measured on the Ni(20 nm)/Ag(30 nm) sample at a distance of 1 mm from the surface (the sample lies between $y=0.5$ and 2.0 mm). Bottom: Normalized square modulus of the electric-field component along the growth axis z, for Ni(z)/Ag(50 nm-z) samples (1 corresponds to $z=10$ nm, 2 to $z=20$ nm, 3 to $z=30$ nm, and 4 to $z=40$ nm), in the ferromagnetic layer (full symbols) and in the Ag layer (open symbols). The dot-dashed line gives the value of the integral of the square modulus of the z-electric-field component over the ferromagnetic layer of thickness z.

lar spread of the excitation, which is less suitable for the investigation of small angular shifts in the resonance position.

As we have shown, nickel is the best candidate for the study of magnetoplasmonic interactions. We will thus focus our attention on structures based on this ferromagnetic material, applying the developed theoretical treatment to grant access to the electromagnetic properties of the system. In Fig. 4 (bottom panel), the intensity of the square modulus of the electric-field component along the growth axis z for Ni-based multilayered structures is presented: a typical exponential decay of the field is observed. The integral of the square modulus of the electric-field z component over the ferromagnetic layer, which gives an estimation of the degree of interaction between the plasmonic field and the ferromagnetic material, is shown too (dot-dashed line). As we can see, this integral has a maximum for a thickness of 35 nm of the Ni layer. The maximum corresponds to the best overlap of the plasmonic field with the ferromagnetic material. This overlap is not maximized by simply increasing the magnetic layer's thickness, as the damping in the ferromagnetic material strongly reduces the propagation length of the plasmonic field within the structure. Comparing these results to the reflectivity measurements of Fig. 2(a), we can see that for a Ni thickness between 20 and 30 nm, an efficient surface plasmon excitation can be obtained (absorption ranging between 30% and 60%, with linewidths between 1.8° and 2°).

The Ni(20 nm)/Ag(30 nm) structure thus represents the best compromise between efficient coupling of the external radiation to the electronic oscillations in the system and the effective overlap between the plasmonic mode and the magnetoactive layer. Additionally, the magnetizability of the systems is confirmed scanning the surface of the samples with a sensitive magnetic Hall sensor. The stray field intensity (with the in-plane premagnetized sample lying from $y=0.5$ mm to 2.0 mm) for the Ni(20 nm)/Ag(30 nm) structure is shown in Fig. 4 (top right). A detailed analysis of the magnetic properties of these multilayered systems is in progress and will be published upon completion.

The penetration depth of the SP wave along the growth axis z characterizes the interaction of the surface waves with the magnetoactive material. The structure we have proposed can thus represent a potential system for the study of spin effects in plasmonic waves. The possibility of a heat-assisted writing of magnetic bits (as small as 80 nm) on a ferromagnetic surface through a magnetic force microscopy tip, in combination with an external magnetic field has been demonstrated in Ref. 25. We foresee the application of this technique to the structures we are presenting, allowing the writing of "spin gratings" on the sample, thus opening the path to the realization of a platform for the study of spin-plasmonic devices.

In conclusion, we have presented here a systematic study of plasmonic excitation in ferromagnetic (Co, Fe, and Ni)/Ag-based structures, experimentally showing how the coupling between the incoming electromagnetic wave and the electron oscillations in the metal varies as a function of the thickness of the magnetoactive material. Furthermore, the penetration of the plasmonic field within the structures has been investigated, defining the optimal conditions for the study of the interaction between surface plasmon waves and the ferromagnetic material.

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