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Surface sensitivity of optical and magneto-optical and ellipsometric properties in magnetoplasmonic nanodisks

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The optical, ellipsometric, and magneto-optical surface sensitivity to dielectric environment of magnetoplasmonic nanodisks is experimentally studied. Here, the shift of the corresponding spectral structures as a function of the thickness of a coating SiO_2 layer is characterized. Our results reveal that the so called pseudo-Brewster Angle, easily identified in the ellipsometric phase (Δ) spectrum, is up to four times more sensitive than the conventional features used in surface plasmon resonance based sensors. These results highlight the need of investigating the factual implementation of this technique to develop improved ellipsometric-phase based transducers for bio-chemical sensing purposes. © *2016 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4939772]

Bio-chemical sensing devices have been recently improved by the realization of Surface Plasmon Resonance (SPR) based sensors.[1–8] In this technique, the high sensitivity of plasmonic resonances to dielectric environment provides the sensing principle. Moreover, a significant improvement in signal-to-noise ratio, sensitivity, and limit of detection has been demonstrated in Magneto-Optical Surface Plasmon Resonance (MO-SPR) based sensors.[9–12] In this case, the magnetic actuation and the plasmonic enhancement of a Magneto-Optic (MO) signal[13] have been exploited. In these two approaches, the signal analysis involves the spectral shift of the reflection or transmission intensities alone, overlooking the phase information. On the other hand, considering the sharp phase jump undergone by any system while traversing a resonant phenomena, phase-based sensor has been proposed for propagating SPR in continuous layered structures[14,15] and localized SPR (LSPR) in nanostructured systems.[16–18] The later application not only has shown improvement of the bulk sensitivity but also sharper spectral structures compared to standard intensity based counterparts.

In the present work, we have studied the surface sensitivity to dielectric environment of optical, ellipsometric, and MO properties of a system of magnetoplasmonic nanodisks. Previously, magnetoplasmonic systems exhibiting propagating plasmons have demonstrated superior sensing capabilities with respect to standard SPR platforms.[9–11] The aim of this work is to compare this capabilities in systems supporting localized surface plasmons, which has not been explored so far.

Here, the shift of the corresponding spectral structures as a function of the dielectric thickness t is characterized. Our results reveal that the so called *pseudo*-Brewster Angle (p-BA), easily identified in the ellipsometric phase (Δ) spectrum, is up to four times more sensitive than the conventional properties. The high sensitivity of the p-BA to small changes in absorption of thin films has been used before to explore defects in materials[19,20] and for plasmonic characterization of adsorbed nanoparticles.[21] Our results open the door to the use of this technique on nanostructured transducers for bio-chemical sensing purposes. Noteworthy, this approach may collect most of the desired features for this application. Namely, high accuracy and sensitivity provided by the spectral ellipsometry technique, and tunable light-matter coupling supplied by nanofabrication tailoring techniques.

The system under consideration is a set of multilayered nanodisks with a shape of conical frustums made of Ti:2 nm/Au:16 nm/Co:10 nm/Au:6 nm giving a total height of 35 nm, bottom diameter of 180 nm, and characteristic spacing distance between them of 350 nm (Fig. 1(d)).

First, using colloidal lithography, a template of cylindrical holes is made of Poly(methyl methacrylate) (PMMA) on the glass substrate (BK7) (see Sec. 4 of Ref. 22). Afterwards, each of the layers is grown in a UHV e-gun evaporation system. The base pressure is better than 1×10^{-9} Torr, and the evaporation pressure less than 5×10^{-9} Torr.

The dielectric environment is a coating layer of SiO_2 gradually grown on top of the nanodisks. The SiO_2 deposition is done in steps of 2 and 4 nm, up to a total thickness $t = 30$ nm. The coating process is carried out by tilting the sample 45° and rotating it at angular speed of 18°/s (Fig. 1(c)). This method allows growing conformal SiO_2 layers as verified by Atomic Force Microscope (AFM) images as shown in Figs. 1(a) (stripped disks, t = 0 nm) and 1(b) (SiO_2 coated disks, t = 8 nm).

The 10 nm thick Co layer provides the in-plane magnetic character to the structure as demonstrated by Transverse MO Kerr Effect (T-MOKE) measurement in Fig. 1(e). The hysteresis loop exhibits coercive and saturation fields of 170 and 510 gauss, respectively.

The set of optical, ellipsometric, and magneto-optical, characterizations carried out on the stripped sample, i.e., without SiO_2 is illustrated in Fig. 2. After each SiO_2 layer deposition, such a set of characterizations is performed (*ex-situ*). The

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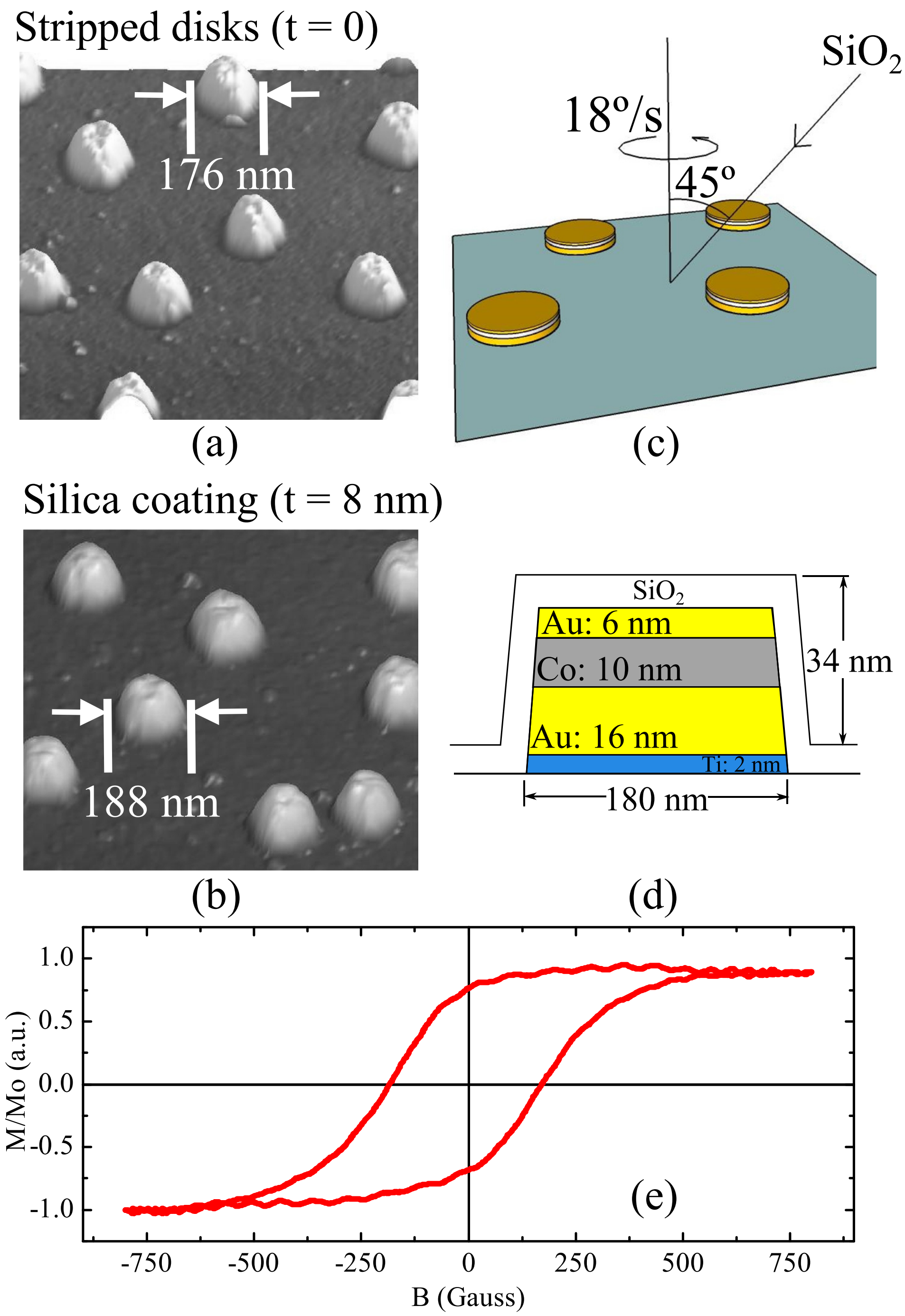

FIG. 1. The 3D AFM images of nanodisks from (a) stripped sample (t = 0) and (b) SiO_2 coated sample (t = 8 nm). (c) Orientation of SiO_2 vapor flow 45° respect to z direction grown at angular speed of 18°/s in order to obtain coating layers with conformal shape as depicted in (d). (e) Magnetic characterization obtained by T-MOKE measurement.

spectral transmission T, the ellipsometric amplitude $\tan\Psi$, and the ellipsometric phase Δ are obtained with a J.A. Woollam Co., M-2000FI™ Spectroscopic Ellipsometer. T is measured at normal incidence. Meanwhile, $\tan\Psi$ and Δ are measured in polarizer-compensator-sample-analyzer (PCSA) off-null ellipsometric configuration,[23] at incidence angles of 50° and 60°. To avoid back-side reflections, the sample is placed on a glass piece optically coupled to the substrate by using refractive index matching oil.

Angles Ψ and Δ represent the complex reflectivity ratio ρ between the two linearly polarized components of light (r_p and r_s) upon interaction with the system. Namely,

$$\rho = r_p/r_s = \tan\Psi \exp(i\Delta), \tag{1}$$

where $\Delta = \arg\{r_p/r_s\} = \Delta_p - \Delta_s$, and

$$\Delta_i = \arg\{r_i\}, \quad \text{or} \quad \tan\Delta_i = \mathrm{Im}\{r_i\}/\mathrm{Re}\{r_i\}; \quad i = p, s. \tag{2}$$

To pursuit a better signal-to-noise ratio, the spectral MOKE signal is obtained in total reflection condition (Kretschmann geometry) at 50° and 60°, as depicted in the inset of Fig. 2(d). Herein, ⊗ indicates the direction along which an oscillating magnetic field is applied to obtain the reflectivity difference ΔR between the two states of magnetic saturation ($\pm M_s$) at 390 Hz in the transverse geometry (T-MOKE).

The spectral transmission T (Fig. 2(a)) exhibits a minimum around 730 nm associated with the excitation of an in-plane LSPR mode. This is consistent with results on similar structures.[24] The ellipsometric and MO features, shown in Figs. 2(b)–2(d), indicate the excitation of the LSPR mode

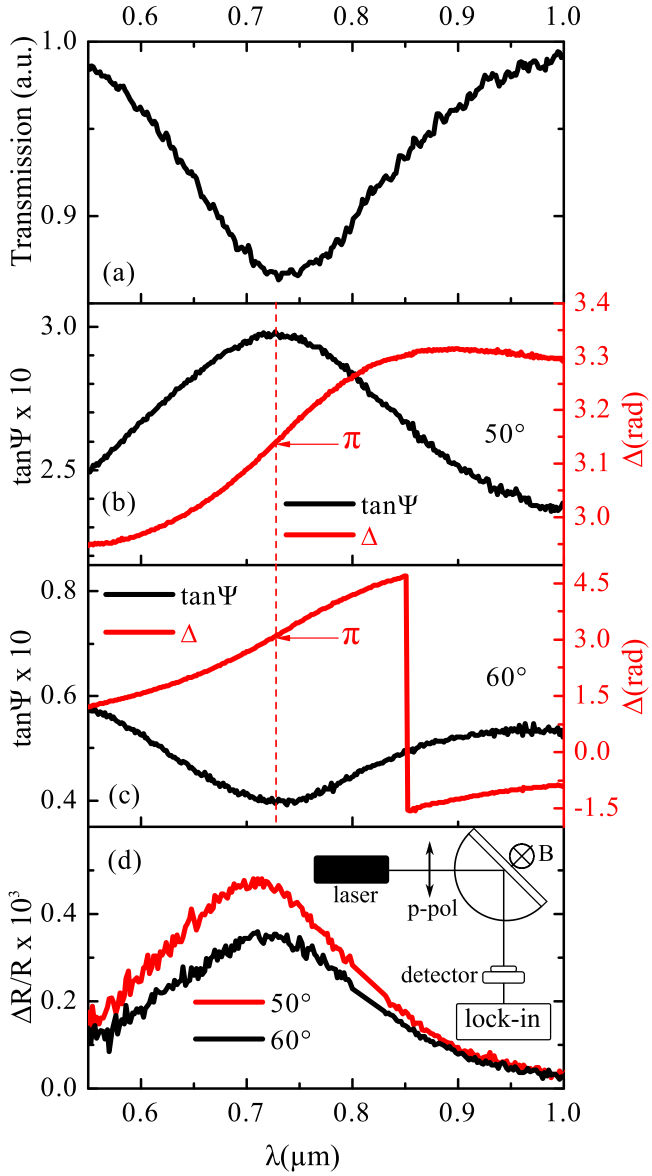

FIG. 2. Set of (a) optical, (b) and (c) ellipsometric, and (d) magneto-optical characterization of the stripped sample (t = 0). The ellipsometric measurements tan Ψ (black line, left scale) and Δ (red line, right scale) are shown at incidence angles of 50° (b) and 60° (c). The MO characterization is given in total reflection condition, i.e., Kretschmann geometry, as sketched in the inset of (d).

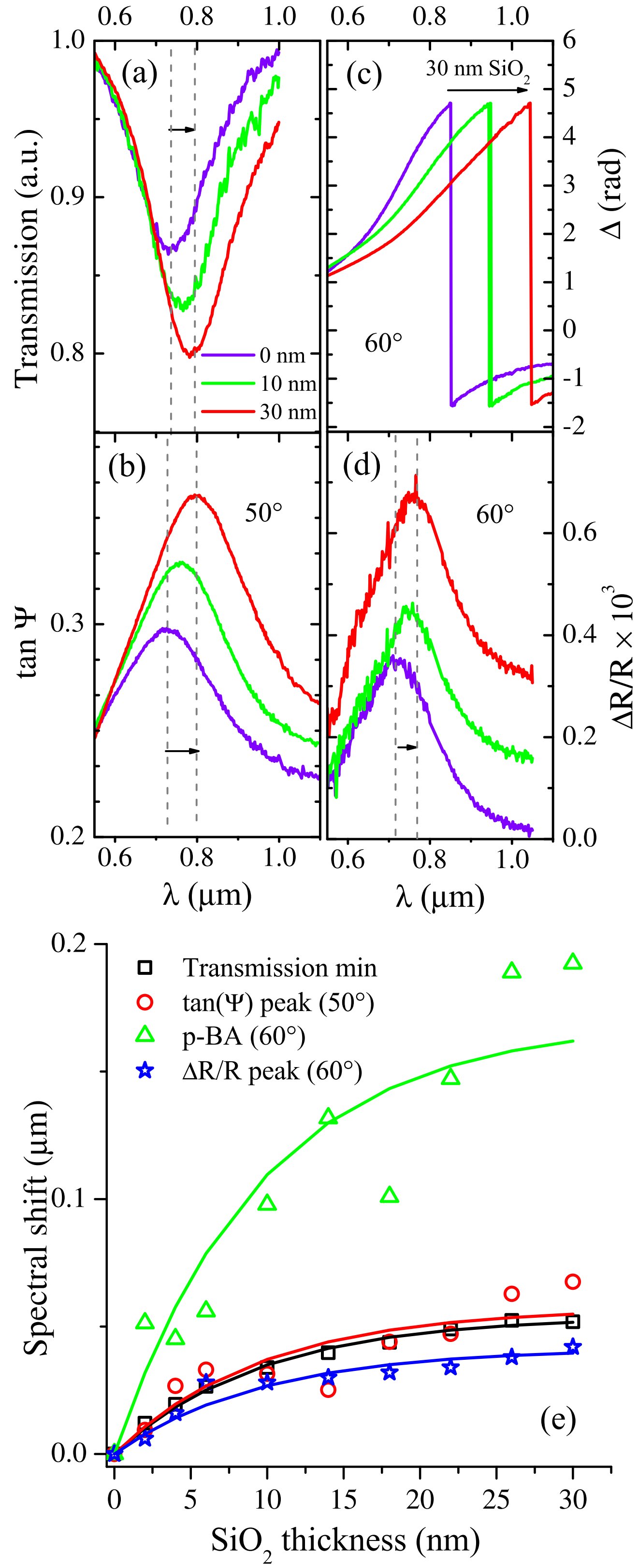

FIG. 3. (a) Optical, (b) and (c) ellipsometric, and (d) MO spectra of the nanodisks for SiO_2 thicknesses of t = 0 (violet), t = 10 nm (green), and t = 30 nm (red). (e) Spectral shift evolution of the featured structures. Solid lines correspond to the fitting as guide to the eye of the data.

responsible for the observed resonant-like spectra. The ellipsometric amplitudes tan Ψ represented in black lines (left scale) in Figs. 2(b) (at 50°) and 2(c) (at 60°) exhibit the characteristic resonant peak or dip for angles of incidence below or above the critical angle, respectively.[25,26] The ellipsometric phases Δ, represented in red lines (right scale) in Figs. 2(b) (at 50°) and 2(c) (at 60°), show a π phase difference between the *p*- and *s*-polarized waves at the plasmon resonance wavelength (∼730 nm). This fact reveals the opposite degree of coupling to the LSPR between *p*- and *s*-polarization components. This can be explained given the fact that the *s*-polarized incident field lies on the plane of the disks; meanwhile, only a projection of the incident *p*-component contributes to excite the in-plane mode of the LSPR.

At 60°, a sharp step of Δ is observed of nearly 2π at 850 nm (Fig. 2(c), red line). This is the result of the real part of the *p*-polarized component of Fresnel reflection Re$\{r_p\}$ going through zero. According to Eq. (2), zeros of Re$\{r_i\}$ give rise to 2π jumps in Δ_i. Additionally, since the sample is absorbing, r_p is complex with Im$\{r_p\} < 0$, and although

$\mathrm{Re}\{r_p\}$ nulls at certain wavelength for an incidence angle of 60°, $|r_p|$ does not vanish in such conditions. In this sense, this sharp step can be interpreted as a *pseudo*-inversion of the *p*-polarized reflected field, and therefore, we refer to this spectral structure as representative of the *p*-BA occurrence. This phenomenon was also detected in ellipsometric studies on nude gold nanodisks arrangements.[26,27] Lastly, in Fig. 2(d), we present T-MOKE spectra obtained in Kretschmann configuration. The effect of LSPR excitation is clearly seen, with an increase of the signal in the region of the resonance. This increase of the signal has also been observed for other magnetic configurations.[24,28–30]

We now proceed to describe results obtained for SiO_2 coated nanodisks. In Fig. 3, we present the optical (a), ellipsometric (b) and (c), and MO (d) spectra of the nanodisks for three selected values of *t*. Curves in (a), (b), and (d) have been shifted along the *y* axis to aid visualization. In all spectral structures, there is a red-shift as *t* increases. The detailed evolution of the red-shift of the spectral structures is presented in Fig. 3(e). Here, it is noticeable that, since *T* (black squares), $\tan\Psi$ (red circles), and $\Delta R/R$ (blue stars) are strongly dependent on the LSPR, the results of their evolutions when traversing the dielectric environment changes are commensurable.

As shown in Fig. 3(c), the shift of the spectral position of the *p*-BA is appreciably higher than that of the other three spectral structures. The fitting as guide to the eye of the spectral shift evolution (solid lines in Fig. 3(e)) reveals that the sensibility factor of *p*-BA is three times higher than that of *T* and $\tan\Psi$, and four times higher than that of $\Delta R/R$. The fact the characterization after each SiO_2 layer is made *ex-situ* (forcing us to manipulate the sample) results in an experimental error and therefore a noisy character of the spectra shift as function of thickness.

This result discloses the extraordinary sensitivity to dielectric environment of the *p*-BA for nanostructured metallic structures and suggests that ellipsometric-phase based sensor may improve the performance of bio-chemical sensing devices.

To conclude, we have studied the sensitivity to dielectric environment of optical, ellipsometric, and magneto-optical properties of magnetoplasmonic nanodisks. Our results found a sharp step of the ellipsometric phase associated to the so called *pseudo*-Brewster Angle which presents an extraordinary surface sensitivity to the dielectric environment. This sensitivity is up to four times higher than the features conventionally found in SPR based sensors. These results highlight the need of investigating the factual implementation of this technique to develop improved ellipsometric-phase based transducers.

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