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Refractive index sensor based on magnetoplasmonic crystals

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A magneto-optical surface plasmon resonance (MOSPR) sensor based on a magnetoplasmonic crystal trilayer structure is presented. The sensitivity of the MOSPR sensor is studied as a function of ferromagnetic layer thickness and at the different modes of operation. The enhancement of the sensitivity caused by using the MOSPR sensor in magneto-optical modulation regime in comparison with reflection regime is observed.

1. Introduction

In recent decades, the miniaturization of chemical and physical sensing devices and their integration into single microchips for multichannel sensing have become a dominant goal of sensor research and development. In addition, the enhancement of the sensitivity and label-free detection has always been a challenge for sensing devices. Following the first demonstration [1], surface plasmon resonance (SPR) sensing has become a leading technique for highly sensitive and label-free detection. SPR detection devices provide real-time measurements and, coupled with micro-scale dimensions, can be applied to biochemical, physical, medical and environment analyses.

Surface plasmon polaritons (SPPs) are collective plasma oscillation phenomena that exist at the boundary of a metal (ε_1) and dielectric (ε_2) media with different signs of dielectric conductivity constants and condition of $\varepsilon_1 < 0,\ |\varepsilon_1| > \varepsilon_2$. These oscillations excite electromagnetic waves with the wave vector

$$k_{spp} = \frac{\omega}{c}\sqrt{\frac{\varepsilon_1(\omega)\varepsilon_2(\omega)}{\varepsilon_1(\omega) + \varepsilon_2(\omega)}}, \tag{1}$$

that propagate along the interface of both media; the maximum intensity is located at the metal-dielectric interface, and the waves decay exponentially into metal and dielectric. The excitation of SPPs decreases the intensity of external electromagnetic waves and can be observed as a sharp resonant dip in the reflection spectrum [2,3]. The parameters of this dip, including the position and width, strongly depend on the dielectric constants of both media and, as a consequence, on the refractive index. This dependence is commonly used for sensing purposes as a function of the refractive index of the external dielectric media.

The main performance characteristics of SPR sensors include sensitivity, accuracy, precision, repeatability and lowest detection limit. To improve these characteristics, many transducer schemes and sensing techniques have been applied. The classical SPR detector is based on the Kretschmann scheme, where the sensing layer is a thin gold film sputtered onto a glass prism. This scheme, low cost and high performance due to its simplicity, is one of the most employed. Depending on the application needs, different types of metals (Au, Ag or their combination), detection schemes (angular, spectra-angular, or phase) and modulation techniques have been used. As proven in [4], the modulation technique can enhance the performance of the sensing characteristics including sensitivity, lowest detection limit and signal-to-noise ratio (SNR); therefore, different modulation techniques have been developed (mechanical, phase, and polarization). Moreover, a new modulation technique based on magneto-optical (MO) and SPR interactions was developed very recently. This type of sensor was called a magneto-optical surface plasmon resonance (MOSPR) sensor [5,6]. It is well known that substantial enhancements of MO effects can be observed in structures that demonstrate simultaneous excitation of surface plasmon polaritons and MO effects [7–9]. Combinations of ferromagnetic materials with noble metals can be used to enhance both MO and SPP activities [8–10]. Gold and silver are the typically used metals for biosensing applications because of their satisfactory plasmonic properties. The MOSPR sensing capabilities of trilayer structures have been studied in Kretschmann scheme [5,6], and the theoretical calculations predicted an order of sensitivity magnitude enhancement.

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The objective of this work is the experimental demonstration of the possibility of using periodical magnetoplasmonic nanostructures, namely magnetoplasmonic crystals [8,11,12], for refractive index detection by using MO probes. This type of sensor represents a possible type of biosensing device. Such plasmonic crystal (PlC) biosensors based on SPR on metal grating provide good sensitivity, simple design and integration because they do not require the Kretschman scheme and can serve as a basis for the development of multichannel sensors and biochips. Another argument for PlC biosensors is that transparency of the sensing layer is not necessary to achieve stable operation. In this paper characteristics of the magnetoplasmonic crystal sensor are studied in comparison with the PlC sensor.

2. Magneto-optical surface plasmon resonance sensor

To describe the sensitivity of the MOSPR sensor on gratings, one must first consider the transversal magneto-optical Kerr effect (TMOKE) and SPR excitation conditions in periodical structures [11,13,14]. On periodical structure the phase-matching conditions of the incident electromagnetic wave vector (k_0) and the propagating SPP wave vector (k_{spp}) can be written as

$$-k_{spp} = k_0 \sin\theta + nG, \tag{2}$$

where G is the reciprocal lattice vector, n is the diffraction order, k_0 is the incident wave vector, and θ is the angle of incidence. In this letter, we consider the Voigt configuration of the MOSPR sensor when the external magnetic field is oriented along the boundary perpendicular to the SPP wave vector. Magnetization of the metallic media on air/metal boundary ($\varepsilon_2 = 1$) produces non-diagonal components in the ε_1 tensor, thereby changing Eq. (1) and the phase-matching conditions for SPP excitation [11,12]

$$k_{spp} + k_0 \sin\theta + k_0 \frac{-ig}{\sqrt{\varepsilon_1 - \varepsilon_1^2 + 1}} = -nG, \tag{3}$$

where ig is the gyration constant. Thus, the minimum in the reflection spectrum (related to SPP excitation) obtained from the magnetoplasmonic crystal shifts by the following value:

$$\Delta\lambda = \frac{-igd}{\sqrt{1 + \varepsilon_1}(1 - \varepsilon_1^2)}, \tag{4}$$

where d is the period of magnetoplasmonic crystal.

On the other hand, TMOKE in the Voigt configuration is measured by the relative change in the reflected light intensity $R(\mathbf{M})$ when the magnetization of medium $\mathbf{M}$ is switched and can be described by the following value:

$$\delta = (R(\mathbf{M}) - R(-\mathbf{M})/R(0) = \Delta R/R(0), \tag{5}$$

where $R(0)$ is the reflectance coefficient when the external magnetic field is not applied. Hence, when $\Delta\lambda/\lambda_0 \ll 1$, where λ_0 is the resonance wavelength, the value of TMOKE can be written as the derivation of the reflection spectrum with respect to wavelength:

$$\delta = \frac{\Delta R}{R(0)} = \frac{dR}{d\lambda}\frac{\Delta\lambda}{R(0)}. \tag{6}$$

Consequently, the value of TMOKE is proportional to the first derivative of the reflection spectrum with respect to wavelength. For sensing applications, we need to quantify and compare MO effects when the reflection index of the dielectric media (n_d) varies. The sensitivity of the MOSPR sensor to changes in the refractive index can be written as

$$\eta = \partial\delta/\partial n_d = \frac{\partial\delta}{\partial A}\frac{\partial A}{\partial n_d}, \tag{7}$$

where A is the wavelength (or another parameter e.g. incident angle). R could be used instead of η for the case of SPR sensor sensitivity. Substituting δ from (5) to (6) one can obtain that the sensitivity of the MOSPR sensor is proportional to the second derivative of the reflection spectrum $R(\lambda)$ with respect to wavelength:

$$\eta = \frac{\lambda}{n_d}\frac{\partial}{\partial\lambda}\left(\frac{dR}{d\lambda}\frac{\Delta\lambda}{R(0)}\right). \tag{8}$$

The sensitivity η has the dimension of inverse refractive index and is measured in RIU^1, where RIU is a refractive index unit [4,5].

3. Experiment

To evaluate the properties of the MOSPR sensor, the designed experimental setup (Fig. 1) is used. It consists of a halogen lamp that functions as a source of incident electromagnetic waves, a monochromator, a Glan Taylor prism to provide p-polarized light that is necessary for SPP excitation, a collimation system, an electromagnet and a detector.

To reduce optical noise, light reflected from the sample was collected by a fiber-optic device with a low numeric aperture. We used a photomultiplier tube operating in a spectral range of 400–860 nm to detect the collected signal, which was further registered by the lock-in amplifier at the frequency of the external magnetic field (118 Hz). For the sensing applications, a flow cell was constructed to enable control of the concentration of liquids in the mixture with high accuracy. The refractive index of the liquids for reference was also monitored by using a refractometer. As a sample for the MOSPR sensor transducer, we chose a trilayer magnetoplasmonic crystal based on a polymer substrate with a lattice period of 320 nm with Ag/Fe/SiO_2 layers sputtered onto the substrate. The other parameters of these magnetoplasmonic crystals are shown in [10]. The Ag/Fe layers were deposited to provide simultaneous SPP and MO activity. The SiO_2 coating was used to protect the sample from degradation in a liquid media and its effect on the optical properties is negligible. Three samples with different Fe layer thicknesses (5, 10, and 20 nm) were fabricated, thickness of an Ag layer was 100 nm.

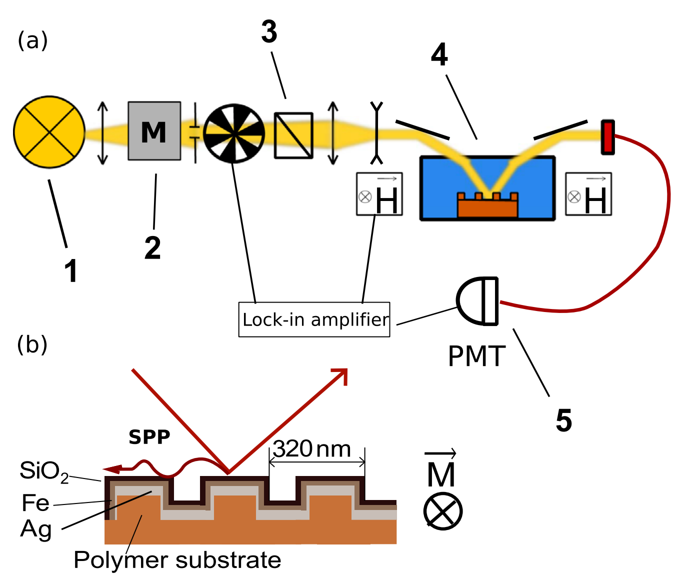

Fig. 1. (a) The scheme of the experimental setup for the magneto-optical surface plasmon resonance sensor: 1 – light source, 2 – monochromator, 3 – Glan-Taylor prism, 4 – flow cell with magnetoplasmonic crystal in an alternating magnetic field, and 5 – photomultiplier tube. (b) The scheme of the magnetoplasmonic trilayer structure and excitation of surface plasmon polaritons.

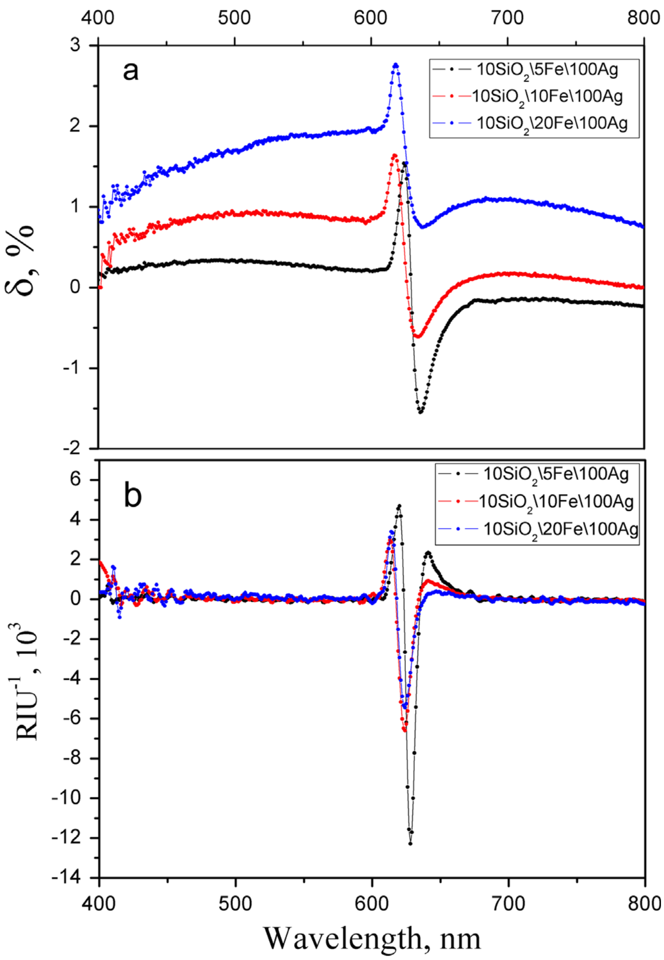

Fig. 2. (a) TMOKE spectra of magnetoplasmonic crystals with 5-, 10- and 20-nm Fe layers measured at an incident angle of 68° at the interface with air. (b) Sensitivity of MOSPR sensors calculated from the TMOKE spectra on (a) by using Eq. (7).

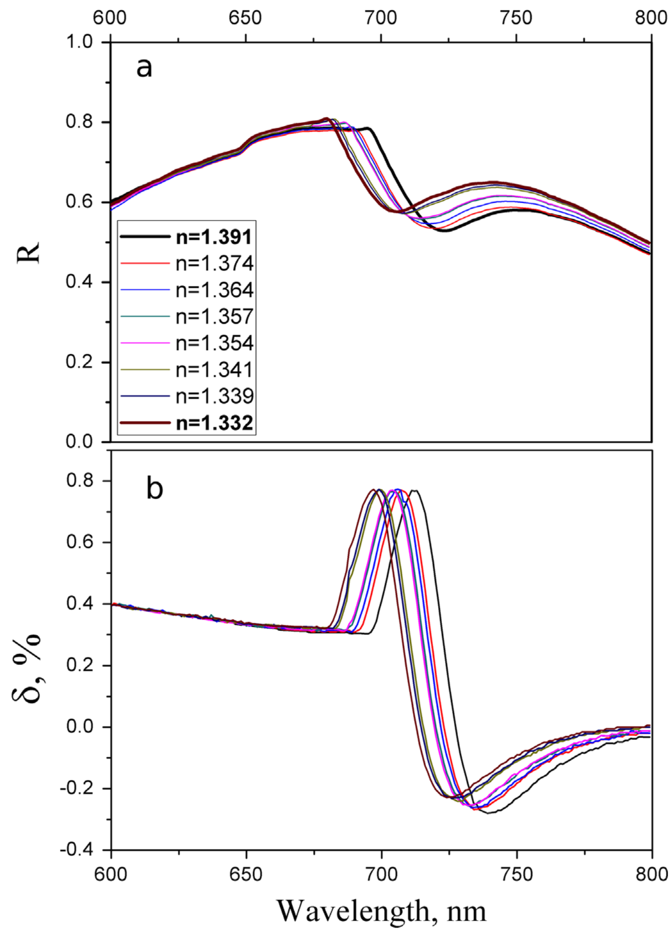

Fig. 3. The dependence of the reflectance (a) and TMOKE (b) spectra of the magnetoplasmonic crystal measured at an incident angle of 55° on the refractive index of liquid in the flow cell for the trilayer structure Ag(100 nm)/Fe(5 nm)/SiO_2(10 nm).

The value of TMOKE depends on the thickness of the ferromagnetic layer [10]. On the other hand, if we consider Eq. (6), the sensitivity of the MOSPR sensor depends on two factors: the slope of the measured curve as a function of the wavelength of the incident light and the shift of such a curve with variation of n_d. The increasing of the ferromagnetic metal thickness leads to increasing the TMOKE but reduces the quality factor of SPR. Thus, to maximize the sensitivity of MOSPR sensors, we must establish a balance between the MO and SPR activity through the selection of different layer thicknesses.

The TMOKE spectra and calculated sensitivity of the samples as a function of layer thickness are studied and compared (Fig. 2). The maximum sensitivity is obtained for the trilayer structure Ag (100 nm)/Fe(5 nm)/SiO_2(10 nm) (Fig. 2(b)) which corresponds to the sample with the smallest thickness of the ferromagnetic metal and with best quality factor of SPR. A further reduction of the ferromagnetic metal thickness was an experimental difficulty. The value of TMOKE is also measured as a function of the angle of incidence, and the maximum value for the current experimental setup is obtained at an angle of 68°. For the sensing application, due to the configuration of the flow chamber, we choose an angle of incidence of 55°.

4. Results and discussion

An MOSPR sensor based on a magnetoplasmonic crystal is studied in three different schemes of signal modulation and detection. Obtained sensitivities are compared. We perform an evaluation of MOSPR sensor in real time and by using static measurements on the same experimental setups. The MOSPR sensing performance of samples with three different Fe layer thicknesses are studied for different concentrations of glycerol–water mixture, delivered to the flow chamber. To make the static comparison of both sensing approaches, the reflectance as a function of wavelength is measured using the SPR sensing configuration, and MOKE as a function of wavelength is measured using the MOSPR scheme. Both types of spectra are measured at different concentrations of glycerol and at a fixed angle of incidence.

The increased concentrations of glycerol solute changed the optical properties of the sensing layers; therefore, this effect produced a change in the plasmon wave vector, which leads to the shift of the SPR (Fig. 3(a)) and MOSPR (Fig. 3(b)) curves. Two approaches are used for recording the signal in the MOSPR sensing configuration. The first approach is the measurement of the relative change in the reflectance when the magnetization of the medium changes in the presence of an oscillating magnetic field denoted as ΔR. The second approach is the measurement of the TMOKE value δ. The comparison of sensitivity for the MOSPR sensor for different modes of detection and choosing of maximum sensitivity points calculated by Eq. (7) are shown in Fig. 4.

We compare the SNR of MO signal and reflectance signal R for MOSPR and SPR sensors respectively for comparison of sensors sensitivities. The SNR depends on the electrical noise in the scheme as the square root of the frequency and on the optical

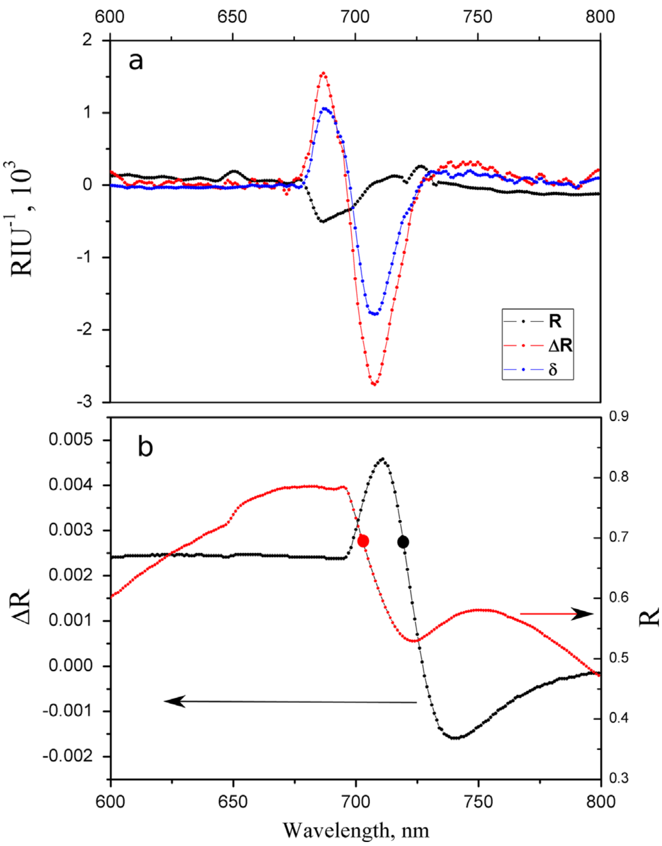

Fig. 4. (a) Spectral dependences of MOSPR sensor sensitivity calculated from experimental spectra for different modes of detection. (b) Maximum sensitivity points for different modes of operation in reflectance and ΔR spectra.

Table 1
SPR and MOSPR experimental sensitivity and the experimental limit of detection. *RIU* is a refractive index unit.

Detection mode	R	ΔR	δ
η_{exp}, RIU^{-1}	500	2750	1780
N, RIU	2×10^{-3}	3.6×10^{-4}	5.6×10^{-4}

noise. We define the minimum detectable signal (S_{min}) as five times the RMS deviation of the experimental signal recorded during 300 s [4,5]. Thereby, the experimental sensitivity can be represented as

$$\eta_{exp} = \frac{\eta}{S_{min}}, \tag{9}$$

and the experimental limit of detection can be represented as

$$N = \frac{1}{\eta_{exp}}. \tag{10}$$

The sensing performance based on the measured curves for all the approaches is listed in Table 1. The best performance is achieved in ΔR mode and exceeds R mode by 6 times in terms of both sensitivity and experimental limit of detection. The MOSPR sensor operation in real-time regime is studied in flow cell with step by step changing of refractive index of liquid. The refractive index of the liquid in flow cell is measured by a refractometer. The comparison of the SNR of the R and ΔR modes of the MOSPR sensor in the real-time regime is shown in Fig. 5(a). The operation of the MOSPR sensor in the real-time regime as a dependence of signal value on refractive index of medium is shown in Fig. 5(b).

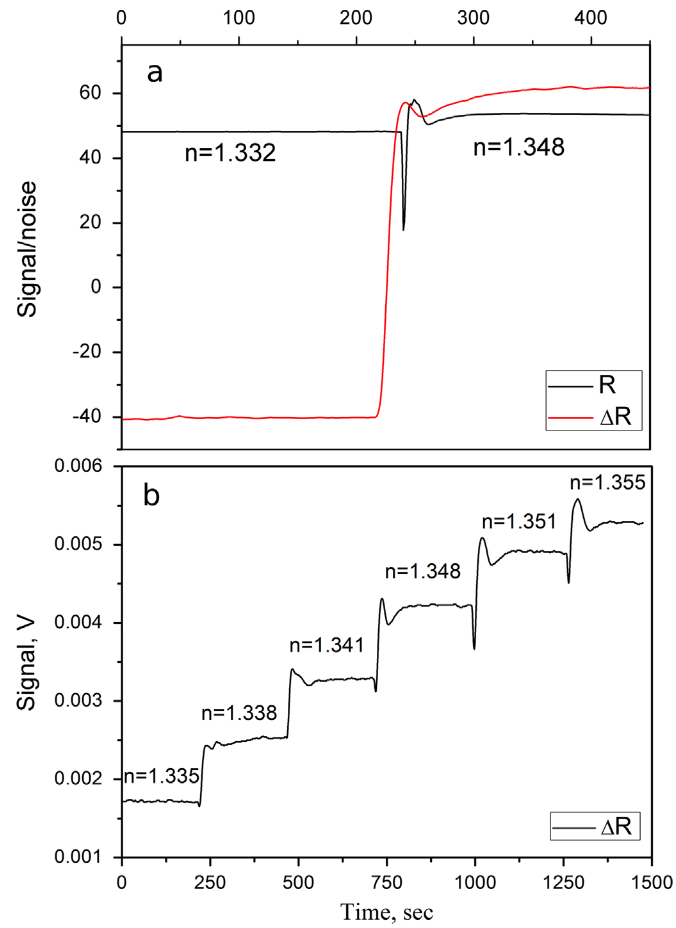

Fig. 5. (a) Signal/noise time dependence of the MOSPR sensor for the reflection and ΔR modes of detection under varying liquid concentration in the flow cell. (b) The signal of the MOSPR sensor in the ΔR-mode and real-time regime.

5. Conclusion

In conclusion, an MO sensor based on a magnetoplasmonic crystal is designed, and different modes of operation are realized. The highest sensor sensitivity is achieved for ferromagnetic layer thickness of 5 nm. A six-fold enhancement of the sensitivity and limit of refractive index detection in the MO modulation regime compared with the conventional reflection regime is observed.

Acknowledgments

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