PHOTONICS Research

All-metallic metasurfaces towards highperformance magneto-plasmonic sensing devices

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Magneto-plasmonic sensors based on surface plasmon resonance have been studied considerably in recent years, as they feature high sensitivity and ultrahigh resolution. However, the majority of such investigations focus on prism-based sandwich architectures that not only impede the miniaturization of devices but also have a weak transverse magneto-optical Kerr effect (TMOKE) in magnitude. Herein, we theoretically demonstrate a magneto-plasmonic sensor composed of Au/Co bilayer nanodisk arrays on top of optically thick metallic films, which supports a narrow surface plasmon resonance (SPR) with a bandwidth of 7 nm and allows for refractive index sensitivities as high as 717 nm/RIU. Thanks to the high-quality SPR mode, a Fano-like TMOKE spectrum with a subnanometer bandwidth can be achieved in the proposed structure, thereby giving rise to ultrahigh sensing of merit values as large as 7000 in water. Moreover, we demonstrate a large TMOKE magnitude that exceeds 0.6. The value is 1 order of magnitude larger than that of magneto-plasmonic sensors reported. We also demonstrate that the behavior of TMOKE spectra can be controlled by tuning the geometrical parameters of the device including the diameter and thickness of nanodisk arrays. This work provides a promising route for designing magneto-plasmonic sensors based on metasurfaces or metamaterials. © 2020 Chinese Laser Press

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1. INTRODUCTION

Active plasmonics is a burgeoning subfield in plasmonics, which is concerned with how to control plasmon properties by external agents such as the temperature, electric fields, or magnetic fields [1]. This branch has attracted attention because it enables novel functionalities in various devices, such as modulators, switches, sensors, and couplers [2-6]. The magnetic field is an innovative means of controlling over the plasmon properties of magnetic materials in a plasmonic structure through magneto-optical (MO) effects, which include the Faraday effect that refers to the rotation of the polarization in transmitted light and the Kerr effect that describes the changes of polarization or intensity in reflected light. In particular, the transverse magneto-optical Kerr effect (TMOKE) is to introduce the variation of intensity in reflected light when the applied magnetic field is perpendicular to the plane of incident light, which has been exploited as a new probe for creating new kinds of magneto-plasmonic sensing devices [7-9]. When surface plasmon resonance (SPR) is excited, the effect can acquire a great amplification that highly relies on the SPR excitation conditions. Therefore, the transverse MO Kerr effect can be utilized to monitor the changes of the refractive index in sensing systems.

effects varying in magnitudes, and ferromagnetic metals such as Fe, Co, and Ni show sizeable MO activity due to their large MO constants. However, the intensity of MO effects in smooth membranes of ferromagnetic metals is still not large enough for practical applications. Presently, sandwich magneto-plasmonic architectures have become widely used as a standard MO-SPR sensing configuration, but they exhibit a low MO intensity (|TMOKE| < 0.1) [10-12]. Furthermore, due to their lack of eliminating the prism-coupling mechanism, it is rather difficult to achieve miniaturization and integration of devices. With the rapid development of the nanofabrication techniques, complex nanostructures incorporated by magnetic and noble metals have been prepared experimentally [13–15]. Moreover, it has been demonstrated that nanostructures are able to enhance MO effects exploiting geometrical resonance, in which the phenomenon of SPR can be directly excited without bulky prisms [2,3]. To date, most nanostructured magneto-plasmonic systems as MO-SPR sensors adopt two-dimensional grating architectures [16,17]. However, few reports on such studies are represented to develop magneto-plasmonic sensing devices in three-dimensional

All of the materials in the natural world can exhibit MO

resonant plasmonic metasurfaces or metamaterial configurations [18].

In this paper, we report a high-performance magnetoplasmonic sensor that consists of a periodic square array of Au/Co bilayer nanodisk that is fabricated on an optically thick metallic (Au) substrate. We find that an ultranarrow SPR mode with a full width at half-maximum (FWHM) of 7 nm is formed, the high-quality resonance characteristic of which resembles that of surface lattice resonance associated with the farfield coupling between nano-objects [19-21]. However, it is worth highlighting that the physical mechanism of the ultranarrow resonance in all-metallic nanostructures is a complex combination of localized and propagating SPRs on diffractive orders in periodic arrays. Because of the high-quality SPR effect, the proposed nanosystem acquires a giant TMOKE value reaching up to 0.65 induced by the hybrid plasmon mode in a dielectric medium with refractive index of n = 1. Benefiting from the large electromagnetic field enhancement in the allmetallic nanostructure and the ultranarrow line width of the TMOKE spectrum, it is theoretically demonstrated that the novel MO-SPR sensor put forward in this work exhibits ultrahigh bulk refractive index sensitivities and incomparable sensing figure of merits (FOM) in a wide range of refractive indices.

2. THEORETICAL ANALYSIS AND SIMULATIONS

The monitor principle of the presented sensing structure is based on the transverse MO Kerr effect, the magnitude of which can be given quantitatively by the following form [5]:

TMOKE =
$$\frac{R(+H) - R(-H)}{R(+H) + R(-H)}$$
, (1)

where R(+H) and R(-H) are the reflectivity for the ppolarized light under positive and negative applied magnetic fields, respectively. The effect depicts the relative change in the intensity of the reflected light, which originates from the slight nonreciprocal variation of the SPR wavevector induced by the magnetization of ferromagnetic metals. For a smooth metallic film, the modulation of the SPR wavevector can be illustrated by using an analytical expression [11]

$$k_{\rm sp}(\pm H) = k_{\rm sp}(0) + \frac{i2d(\varepsilon_d \varepsilon_m k_0)^2}{(\varepsilon_d + \varepsilon_m)(\varepsilon_d^2 - \varepsilon_m^2)} \left(\pm \frac{\varepsilon_{yz}}{\varepsilon_{xx}}\right), \quad (2)$$

in which $k_{\rm sp}(0)$ is the SPR wavevector in a demagnetized medium; k_0 is the wavevector of the incident light in a vacuum; ε_d and ε_m are the permittivity of dielectrics and metals, respectively; d is the thickness of the metal; and ε_{yz} and ε_{xx} are the off-diagonal and diagonal components of the dielectric tensor in the magnetized metals, respectively, when the direction of applied magnetic field is along the x axis. As one can see from the formula above, the SPR wavevector relies on the nondiagonal elements of the dielectric tensor and the thickness of the metals. The generation and distribution of the nondiagonal elements highly depend on the magnitude and direction of the external magnetic field, and they can reach saturation under a certain condition. Therefore, the ultimate MO signal is dominated largely by the film thickness that affects the optical losses of the whole system, whereas it will be influenced by the overall geometrical sizes in a nanostructured system.



Fig. 1. Schematic drawing of the magneto-plasmonic sensor based on nanodisk arrays. The device's geometrical parameters include diameter *D*, Au-nanodisk thickness t_1 , Co-nanodisk thickness t_2 , and period *P*. The p-polarized illumination source propagates in the y - z plane, and the magnetization is along *x* direction. The Ausubstrate thickness is 150 nm.

Figure 1 depicts the magneto-plasmonic system based on a periodic arrangement of Au/Co nanodisk arrays standing on an Au film. Here the thickness of the underlying Au film is 150 nm. It can increase the local optical fields and reduce the optical losses of the dipolar plasmon, which is necessary to obtain narrower bandwidths. These characteristics are advantageous for obtaining high-performance biosensors. The thicknesses of the Au and Co nanodisks are t_1 and t_2 , respectively. The diameter and period of the nanodisk array are D and P, respectively. In order to produce the transverse MO Kerr effect, the p-polarized light with an incident angle of θ impinges on the metal surface, and the direction of applied magnetic field His perpendicular to the plane of incidence. Numerical simulation is performed by using a commercial finite element method (FEM) package (COMSOL Multiphysics) to calculate the optical and MO responses of the structure. The nanosheet is placed in an air medium with refractive index n = 1. The permittivity of Au arises from the experimental data by Johnson and Christy. We describe the optical properties of Co with an anisotropic permittivity tensor with $\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon_{zz} = \varepsilon$ and $\varepsilon_{yz} = -\varepsilon_{zy} = i\varepsilon Q$, where ε is taken from Ref. [22] and Q is the Voigt vector [23].

3. RESULTS AND DISCUSSION

A. Analysis for the Proposed Magneto-Plasmonic System

In Fig. 2(a), we examine the reflection spectra of the system when the alternate magnetic field is implemented, where the optimized geometrical parameters are D = 200 nm, $t_1 = 20$ nm, $t_2 = 13$ nm, and P = 400 nm, and the incident angle $\theta = 50^{\circ}$. As can be seen, the slight offset in the reflection spectrum occurs due to the nonreciprocal variation in the SPR wavevector induced by the magnetization of the Co layer. A plasmon mode arises near $\lambda = 724$ nm for these two reflection curves, the FWHM of which is only 7.748 nm. Figure 2(b) shows the dispersion of the high-quality SPR mode as a function of incident angle and wavelength, in which the white dashed lines represent (-1,0) and (1,0) diffractive orders obtained by the *k*-conservation relationship [24] $|k_{sp}|^2 = (k_x + m\mathbf{G}_x)^2 + (k_y + m\mathbf{G}_y)^2$. Here the surface plasmon



Fig. 2. Optical and MO responses of the proposed nanostructure. (a) Reflection spectra for the sample with different direction of magnetization. The FWHM of these two reflection curves is 7.748 nm. For clarity of the shift in the reflected light, the spectral range is zoomed in within a 4 nm wavelength window in the inset. (b) Corresponding TMOKE spectrum calculated by Eq. (1). The FWHM is 0.125 nm. (c) FEM simulated reflection spectra of the demagnetized sample as a function of incident angle and wavelength. The dashed lines indicate (1,0) and (-1,0) plasmon modes, which are calculated by the *k*-conservation relationship. (d) Electric field distributions in the x - y and y - z planes for four unit cells of the structure. They share the same legend.

wavevector is $k_{\rm sp} = 2\pi \sqrt{\varepsilon_m \varepsilon_d} / (\varepsilon_m + \varepsilon_d) / \lambda$, where ε_m and ε_d are the dielectric function of Au and air, respectively; $k_x = 2\pi \sin \theta \cos \phi / \lambda$ and $k_y = 2\pi \sin \theta \sin \phi / \lambda$ are the x and *y* components of the incident light wavevector, respectively, where θ is the incident angle and ϕ is the azimuth ($\phi = 0^{\circ}$ for the given configuration); (m, n) is the grating order for reciprocal lattice vectors \mathbf{G}_x and \mathbf{G}_y , where $|\mathbf{G}_x| = |\mathbf{G}_y| = 2\pi/P$ for the square lattice. The calculated result gives a good agreement with that of the simulation, which further confirms the resonance mode at $\theta = 50^{\circ}$ as the (-1, 0) SPR mode. It should be noted that the narrow SPR mode possesses complementary characteristics of localized and propagating surface plasmon polaritons together due to the presence of the Au mirror underneath. We provide the near-field distributions at the resonance minimum so as to intuitively comprehend the underlying physical mechanism of the formation of the high-quality SPR mode as shown in Fig. 2(d). The figure with green wireframe displays the profile of the electric field at 2 nm above the top surface of the structure (x - y plane), demonstrating the coupling of the surface plasmon polariton of the Au film and the dipolar mode of the nanodisks. The picture with the azure wireframe displays the electric field distribution at the y - z plane through the center of the structure. It is seen that the local electromagnetic field is largely increased in the system, which is favorable for extending deep into the medium

and increasing the interaction volume of analytes and optical fields [25]. Therefore, the proposed platform allows enhanced device performance for biosensing.

The TMOKE curve corresponding with Fig. 2(a) is plotted in Fig. 2(d), demonstrating a sharp Fano-like line shape owing to the high-quality SPR. The FWHM of the TMOKE curve with Fano-like line shape can be extracted accurately using the following analytical Fano interference model given as [26]

$$\delta(\lambda) = A + B \frac{(q\Gamma/2 + \lambda - \lambda_0)^2}{(\Gamma/2)^2 + (\lambda - \lambda_0)^2},$$
(3)

where *A* and *B* are constants representing the background and the overall peak height, respectively; *q* is the Breit–Wigner– Fano parameter determining the asymmetry of the resonance profile; Γ is the FWHM of the resonance; and λ_0 is the resonance wavelength. Through Eq. (3), the FWHM of curve in Fig. 1(d) is calculated to be 0.125 nm. It is worth mentioning that the subnanometer line width can be also achieved in the surface lattice resonance mode under appropriate geometrical parameters. However, besides such all-metallic nanostructures, the particular plasmon mode can only exist stably when nanoparticle arrays are embedded in a symmetrical dielectric environment, which is unfriendly for the large-range refractometric sensing application. Furthermore, the surface lattice resonance mode with subnanometer line width cannot be easily observed in the experiment due to some restrictions such as the numerical aperture of the focusing optics, nanofabrication techniques, and dielectric environment [27]. Fortunately, by usage of the scheme of monitoring the change of reflectivity, we effectively circumvent the limitation due to the optical losses inside metals and overcome the aforementioned barriers from which the surface lattice resonance suffers.

Because the amplitude of the TMOKE curve highly relies on the optimal SPR excitation, the spectral contrast ratio, defined as the ratio of the difference between the peak value and the dip value to the sum of these two values, namely, $CR = (R_{peak} - R_{dip})/(R_{peak} + R_{dip})$, is utilized to represent the quality of the reflection spectrum [28]. Figure 3(a) shows the reflection of the demagnetized sample (H = 0) as a function of incident angle θ , where the structural parameters are the same as those in Fig. 2. However, the CR of the reflection spectrum reaches a maximum value at $\theta = 50^\circ$, which means that the amplitude of the corresponding TMOKE spectrum has a maximum. Note that the plasmon mode cannot be excited at small-angle incidence for the proposed structure thanks to the SPR excitation going out of the wavelength range of the excitation of the dipolar mode for Au nanoparticles arranged in a periodic array with lattice constant P = 400 nm.

On the other hand, the plasmon mode in our structure is also highly sensitive to the period of the nanostructure. Figure 3(b) plots the influence of changing the lattice constant on the high-quality SPR mode at $\theta = 50^\circ$, while keeping other geometrical parameters constant. As one can see, the line width of the high-quality SPR mode is dramatically decreased as *P* increases, which is the crucial characteristic for the high-quality SPR mode in all-metallic nanostructures [27]. For an array with

a relatively small period in which particles are densely packed, the interaction of particles is mainly carried out by near-field coupling. Under this circumstance, the plasmonic modes with narrow line shapes may be formed, whereas they are usually characterized by being antisymmetric and dark, and they cannot be easily excited by incident light [29-32]. When the incident wavelength is comparable with the period of the array, under normal incidence, the high-quality SPR mode can be excited via interparticle far-field coupling also known as diffraction coupling. For our structure, the high-quality SPR mode also is excited at oblique incidence, and the bandwidth of one is increasingly larger for arrays with a small period due to the interparticle strong coupling as seen in Fig. 2(d). However, the reflection spectrum of the structure with a period P = 400 nm has a maximum value in CR, which is desirable to launch an ideal magneto-plasmonic sensor with a large TMOKE.

B. Spectral Tunability of the TMOKE

In this section, we summarize the dependence of the TMOKE response on geometrical parameters as displayed in Fig. 4. The optical and MO enhancements as well as the TMOKE spectra for devices with different geometrical parameters are discussed in detail, respectively. Figures 4(a)-4(c) represent the relationship of structural reflection and nanodisk diameter *D*, Au-disk thickness and Co-disk thickness, respectively, which correspond to the aforementioned optical enhancement. The nanostructure exhibits a distinct reflection minimum in the spectral range of interest owing to the SPR effect. When increasing the device parameters, the FWHM of the reflection spectrum is gradually increased accompanied with redshift behavior, which is due to the increased optical loss for large devices. The



Fig. 3. Influence of incident angle and period on optical responses for the magneto-plasmonic system. (a) Reflection spectra as a function of incident angle while the period P = 400 nm. (b) Reflection spectra as a function of period while the incident angle $\theta = 50^{\circ}$. Contrast ratio and FWHM of reflection spectra as a function of (c) incident angle and (d) period, respectively. The other geometrical parameters are the same as those discussed in Fig. 2.



Fig. 4. Dependence of optical and MO responses on geometrical parameters. Reflection of the hybrid nanostructure where the Co layer is demagnetized as a function of (a) nanodisk diameter D ($t_1 = 20$ nm and $t_2 = 13$ nm), (b) Au-disk thickness (D = 200 nm and $t_1 = 20$ nm), and (c) Co-disk thickness (D = 200 nm and $t_1 = 20$ nm), respectively. (d)–(f) Corresponding change of reflection with the reversal of Co magnetization. (g)–(i) Corresponding TMOKE spectra.

corresponding reflection variations of structures, R(+H)-R(-H), induced by the change of magnetization in the Co layer, are plotted in Figs. 4(d), 4(e), and 4(f). These results demonstrate that the hybrid nanosystem attains a large MO enhancement with high values of 0.01–0.015.

Figures 4(g), 4(h), and 4(i) demonstrate the evolution of TMOKE spectra for different structural parameters. The TMOKE signal representing a relative change of reflection exhibits a Fano-like line shape that becomes more extremely sharp when the reflectivity minimum tends to zero. Benefitting from the large MO enhancement, the nanostructure has a giant TMOKE amplitude of 0.6 that is much higher than that of the configurations based on a prism coupling mechanism, which is favorable to its usefulness for applications [2,5]. The TMOKE signal of sandwich configurations generally has an order of magnitude of 0.1–0.001 [8].

C. Refractive Index Sensitivity

The magneto-plasmonic platform in the ultraresolution sensing application is the main issue in this work. The refractive index sensitivity $S = \Delta \lambda / \Delta n$ is utilized to quantitatively evaluate the sensing performance of plasmon sensors, where $\Delta \lambda$ is the shift of SPR peak position that arises from the change in the environmental refractive index. Figure 5(a) shows the change of the

TMOKE spectrum for the nanosensor embedded in different media (n = 1 for air, n = 1.33 for water, n = 1.397 for butanol, and n = 1.4458 for chloroform), where the device parameters are D = 200 nm, P = 400 nm, $t_1 = 20$ nm, $t_2 = 13$ nm, and $\theta = 50^\circ$. It can be seen that the MO signal has a dramatic redshift as n increases due to the high sensitivity of the system. Furthermore, the magnitude of the MO signal is slightly different in different dielectric environments, which arises from the tiny change of amplitude in the reflection spectrum. However, the MO signal exceeding 0.4 in a wide range of refractive indices from 1 to 1.5 can sufficiently satisfy the requirement in the experiment.

The peak position for every TMOKE spectrum as a function of refractive index is plotted in Fig. 5(b), showing a linear relationship between the resonance position and the refractive index. An ultrahigh sensitivity of S = 717 nm/RIU is calculated. Such high sensitivity results from the local optical fields enhanced strongly due to the introduction of the Au mirror. Since the final accuracy of spectral position tracking depends on the magnitude and line width of the signal, the FOM, FOM = S/FWHM, becomes a most crucial parameter for the performance assessment of plasmonic sensors. Figure 5(c) shows the FOM of the magneto-plasmonic sensor in different media. Conspicuously, the nanosensor obtains a



Fig. 5. Refractive index sensitivity of Au/Co nanodisk arrays on top of Au film. (a) TMOKE spectrum for different dielectric environments as a function of wavelength. The refractive indices are air n = 1, water n = 1.33, butanol n = 1.397, and chloroform n = 1.4458. (b) Peak position of TMOKE curves as a function of refractive index. The black line indicates a linear fit to the dots, and the slope defines the sensitivity. (c) Calculated FOM for different embedding medium as a function of wavelength.

sky-high FOM close to 7000 in a water environment, which is nearly 2 orders of magnitude higher than that of Au- or Ag-based nanosensors.

The comparison of the proposed magneto-plasmonic system and the Au-based sensor is performed, where the thickness of the Au nanodisks is 33 nm for the Au-based structure, and other parameters are the same. The sensing ability of the Au-based nanosensor is examined in the same surroundings as the magneto-plasmonic system. Figure 6 compares the FOMs of these two kinds of sensors, showing an outstanding sensing performance for the proposed sensing device that benefits from the TMOKE response with subnanometer bandwidth. For the sensors based on propagating SPR, the theoretical limit of the FOM is determined by [33]

$$FOM = \frac{|\varepsilon'_m|^2}{2n\varepsilon''_m},$$
 (4)

where ε'_m and ε''_m are the real and imaginary components of the metallic permittivity, respectively, and *n* is the refractive index of the embedding medium. The black dots in Fig. 6 show the



Fig. 6. Sensing-performance comparison of MO-SPR and SPR sensors. The red and green spheres represent the FOM for the sensors based on an array of Co-Au nanodisks and based an array of Au nanodisks, respectively, in media with same gradient refractive index (n = 1, 1.33, 1.397, 1.4458). The black spheres indicate the ultimate value of FOM in propagating SPR sensors, which is calculated by Eq. (4).

theoretical limit of the FOM in SPR sensors calculated by Eq. (4) for n = 1, 1.33, 1.397, and 1.4458. The Au-based sensor's FOM reaches close to 100, which is close to the ultimate value limited solely by the material permittivity due to the narrow SPR mode. On the other hand, the magneto-plasmonic sensor based on the measurement of the MO signal effectively circumvents the limit of material losses, acquiring a tremendous improvement of sensing performance.

4. CONCLUSION

In summary, we present an MO-SPR sensor composed of a square array of Au/Co bilayer nanodisks standing on an optically thick Au mirror, which supports a narrow-line-width SPR. The special resonance mode is essentially an intricate combination of localized and delocalized SPRs on diffractive orders in periodic arrays. Assisted by the high-quality SPR mode, a giant TMOKE signal with subnanometer line width is obtained by the MO-SPR sensor, leading to the ultrahigh sensitivity and FOM. Based on the measurement of the transverse MO Kerr effect, we have an effective circumvention of limit in material properties, and we tremendously enhance the sensing performance of conventional nanostructured plasmonic sensors. With the advance of nanofabrication technologies and experimental instruments, the proposed nanosensor will exhibit its huge application potential in optical sensing with ultrahigh resolution.

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