Highly Sensitive Metamaterial Plasmonic Sensor Based on Nanoimprinting

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***Abstract*—In this paper, we propose and experimentally demonstrate a highly sensitive plasmonic nanohole metasurface sensor using nanoimprinting and electron beam evaporation. A refractive index sensitivity of 516 nm/RIU has been successfully achieved. With the help of nanoimprinting, the proposed method exhibits intrinsic advantages in a simple fabrication process, high repeatability and low cost. Furthermore, the proposed method enables large-area fabrication, making the mass-production of the metamaterial sensor highly feasible.**

***Keywords*—metasurface, nanohole, nanoimprinting, sensor**

I. Introduction

Metasurfaces have attracted significant attention due to their intriguing properties, exhibiting great potential in designing functional devices. Sensors based on plasmonic metasurfaces are very promising regarding high sensitivity and high quality factor (Q factor). Such metamaterial plasmonic sensors have the advantage of selectively absorbing light and binding the energy of electromagnetic fields to bring huge electromagnetic field enhancement. They are used in many fields, including biology, chemistry, materials, and energy [1]. However, the fabrication of metasurfaces is highly dependent on electron beam lithography or deep ultraviolet lithography. These lithography technologies are challenging to apply to large-area manufacturing and industrial production due to the expensive equipment and low fabrication efficiency. Instead, nanoimprinting offers a feasible way for large-area patterning of the metamaterial with low cost and high efficiency, enabling a more comprehensive application of the nanostructure devices [2-3]. This paper proposes and experimentally demonstrates a highly sensitive sensor based on a nanohole metasurface applying nanoimprinting and electron beam evaporation. The proposed method holds great potential for low-cost and mass-production of metamaterial sensors.

II. Design and Simulation

Fig. 1 shows the schematic diagram of the proposed metamaterial plasmonic sensor. Here, commercial software Lumerical FDTD Solutions is utilized to design the metasurface. The substrate is a curable resist patterned with nanohole array, whose diameter and depth are 190 nm and 150 nm, respectively. The substrate is thick as 5 μm, and the unit cell has a period of 500 nm.

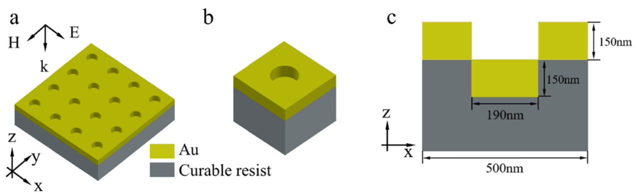


Fig.1. (a) Schematic diagram of the metamaterial plasmonic sensor; (b) Unit cell of the nanostructure; (c) Sectional view of the unit cell.

In the simulation, the optical permittivity of gold is obtained from the experimental data of Johnson and Christy [4], and the refractive index of the curable resist is set as 1.56. Periodic boundary conditions are applied to both x and y directions, while perfectly matched layers (PMLs) are assumed at the domain boundaries in the z-direction. The incident light irradiates the device along the backward z-direction, and the reflectance R of the metasurface is obtained by the frequency-domain field and power monitor.

Under the condition of wave vector matching, the electromagnetic wave irradiation on the metal surface can drive free electrons to generate collective oscillations. The oscillating electrons then excite the magnetic field coupling for electronic motion, and lead to surface plasmon resonance (SPR) due to the interaction between the electromagnetic wave and free electrons [5]. For nanohole metasurfaces, the resonant wavelength of surface plasmon polaritons (SPP) under normal incidence can be evaluated as below [6],

$$\lambda_{SPP} = \frac{P}{\sqrt{i^2 + j^2}} \sqrt{\frac{\varepsilon_d \varepsilon_m}{\varepsilon_d + \varepsilon_m}} \quad (1)$$

where P is the period of the unit cell, ε_d and ε_m are the respective dielectric constants of the environmental dielectric medium and metal, and (i, j) denote the orders of plasmonic resonance modes. It is seen from Eq. (1) that the resonant wavelength of the nanostructure highly depends on the environmental medium, revealing the physical mechanism for sensing application.

To explore the sensing performance of the metamaterial sensor, reflection spectra are recorded with four different background refractive indices (n) of 1.333, 1.361, 1.376 and 1.431, corresponding to deionized water, ethanol, isopropanol and glycol, respectively. The sensitivity (S) is then calculated by:

$$S = \frac{\Delta \lambda}{\Delta n} \tag{2}$$

where $\Delta\lambda$ denotes the wavelength shift, and Δn is the difference of refractive indices. As shown in Fig. 2(a), as the background refractive index increases, the resonant wavelength of the reflection spectra gradually redshifts. The sensitivity is assessed in Fig. 2(b) by linear fitting. It is found that the sensitivity of the metamaterial sensor reaches up to 512 nm/RIU. Fig. 2(c) and 17(d) illustrate the electromagnetic field distributions at the resonance (i.e., 737.3 nm) under the condition of deionized water (i.e., n=1.333). One can see that the electric field is intensively confined in the upper half of the hole, and the magnetic field firmly locates at the edge of the nanohole on the upper surface of the gold layer. The electromagnetic field distributions indicate intensive surface plasmon resonance, leading to a susceptible sensing property of the metamaterial nanostructure.

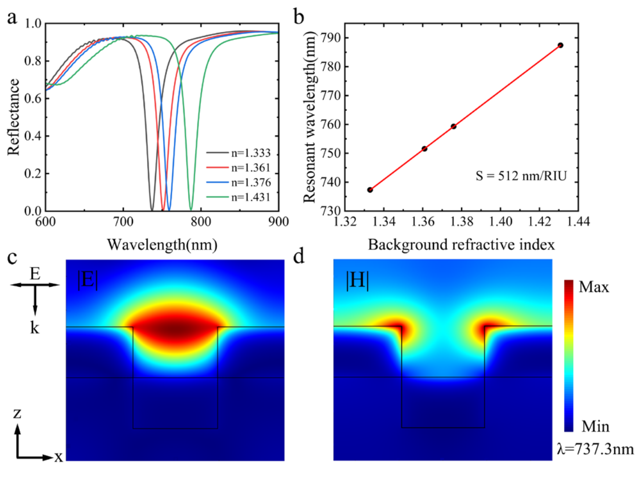


Fig. 2. (a) Simulated reflection spectra of the plasmonic nanohole sensor with different media; (b) Calculation of sensitivity; (c) Electric and (d) magnetic field distributions of the nanohole at the resonant wavelength (n=1.333).

In addition, we study the influence of the nanohole's diameter on the sensitivity of the metasurface. Fig. 3(a) shows that with the increase of the diameter in the range of 100 - 270 nm, the sensitivity correspondingly rises from 505 - 545 nm/RIU. For a wider incident wavelength range, Fig. 3(b) illustrates the reflectance as a function of both wavelength and the nanohole's diameter. Also, the wavelength difference between the two black dotted lines represents approximately the full width half maximum (FWHM) of the resonance. One can see that the larger circular diameter results in the wider resonance FWHM. At 160 - 220 nm in the diameter, the reflectance at the resonance is lower than 20%, and the FWHM remains narrower than 30 nm.

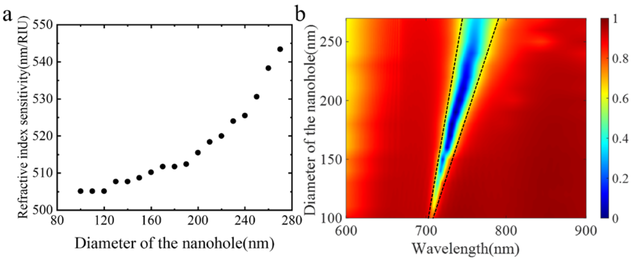


Fig. 3. (a) Refractive index sensitivity with different diameters of the nanohole; (b) Reflectance as a function of the incident wavelength and diameter of the nanohole (n=1.333).

III. Fabrication and Experiment

Fig. 4 displays the fabrication flow chart, which mainly includes nanoimprinting (steps a-c) and evaporation (step d). Specifically, the nanostructure feature is first nanoimprinted onto the transparent polymer substrate from the silicon mold (step a), in which the nanohole array has a period of 500 nm, a diameter of 190 nm, and a depth of 150 nm. After that, a UV curable resist (PAP100) is spin-coated on a SiO_2 substrate (step b). The coated substrate is then pressed by the complementarily patterned transparent polymer template and exposed via ultraviolet irradiation (step c). Ultimately, 5 nm titanium and 150 nm gold layers are deposited on the substrate using electron evaporation. In the experiment, it is worth noting that the nanoimprinted substrate is a six-inch wafer, which is tailored to 6 mm ×6 mm by laser cutting before evaporation. This in turn demonstrates the feasibility of the proposed scheme for large-scale fabrication.

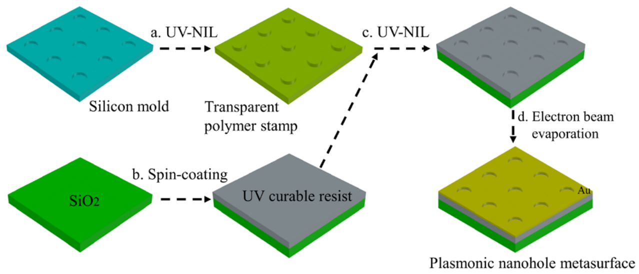


Fig. 4. Fabrication flow chart of the plasmonic nanohole metasurface sensor.

The fabricated samples are characterized by a field emission scanning electron microscope (GeminiSEM 560, Zeiss) and illustrated in Fig. 4. As shown in Fig. 5(a), the nanoholes are uniform in size after nanoimprinting. The same property is also observed for Fig. 5(b) as the electron beam evaporation was completed. The period and diameter are measured as ~ 500 nm and ~ 190 nm, respectively, showing good accordance with the design. The experimental system for reflective spectrum measurement is shown in Fig. 6. The halogen light source (HL2000, Ideaoptics) with a bandwidth of 300 - 2500 nm transmits through a 7-core Y-shaped fiber, of which six fibers in one branch work as the input channel. The light then normally incident to the sample and reflects to the rest one fiber of the 7-core Y-shaped fiber. Here, the rest one fiber performs as the receiving channel, guiding the reflective light to the UV-visible-NIR spectrometer (PG2000-pro, Ideaoptics). All reflection spectra are obtained by using a gold mirror as a reference.

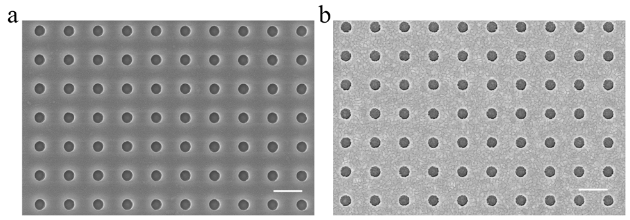


Fig. 5. SEM images of the sample after (a) nanoimprinting and (b) electron beam evaporation. The scale bar is 500 nm.

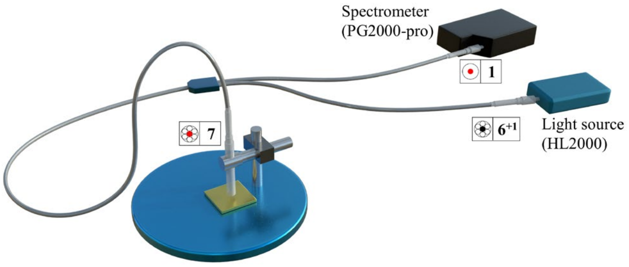
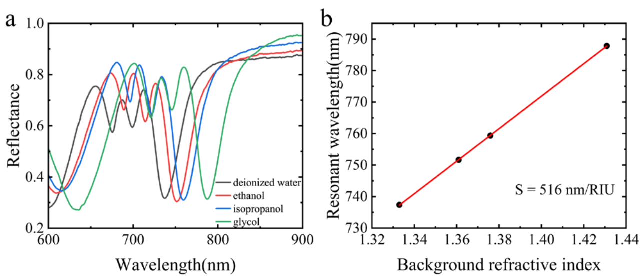


Fig. 6. Experimental system for reflective spectrum measurement.

IV. Results and Discussion

In the sensing experiment, the chip surface needs to be fully covered by the reagent. Here, we use four different kinds of reagents for sensing, including deionized water, ethanol, isopropanol, and glycol. When changing the background reagent, the sample is washed with isopropanol and deionized water and is blown dry. The measured reflection spectra under different reagents are shown in Fig. 7(a). It is seen that the resonance wavelength obviously redshifts within 730-790 nm as the refractive index of the reagent rises, matching well with the simulation results. According to the measured spectra, the evaluated refractive index sensitivity reaches to 516 nm/RIU, exhibiting better performance compared to the simulation. Figs. 7(c) and 7(d) compare the enlarged experimental and simulated reflection spectra with deionized water and ethanol, respectively. In both cases, the fabricated nanostructure is relatively more lossy than the ideal sensor, but the reflection resonances are still clear. Moreover, the wavelength difference between measurement and simulation is less than 3 nm. Considering that the large and stable wavelength shift is more important than the resonance intensity for a sensor, the proposed method can perform high-quality sensing well.



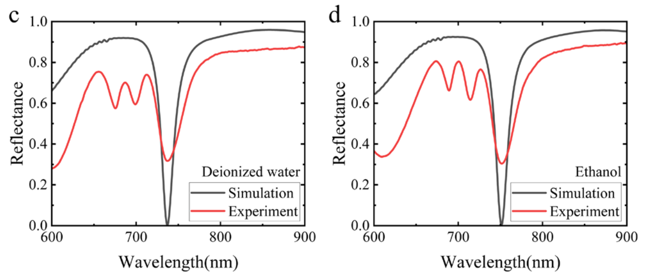


Fig. 7. (a) Experimental reflection spectra with different solutions; (b) Linear fit of the resonance wavelengths as a function of the refractive indices; Experimental and simulated reflection spectra under the background of (c) deionized water and (d) ethanol.

V. Conclusion

In summary, this work proposes and experimentally demonstrates a plasmonic metasurface sensor based on gold circular nanoholes by using nanoimprinting and electron beam evaporation. The experimental results are consistent with the simulation. Taking advantage of the intensive surface plasmon resonance, a high-quality sensitivity of 516 nm/RIU has been achieved. Additionally, the fabrication process of the proposed device is simple and cost-effective for producing large-area metamaterial plasmonic devices, which is conducive to the mass manufacturing and broad application of functional metamaterials.

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