

An ultra-sensitive glucose sensor by using metamaterial-based microfluidic chip

Ji Luo, Ruijia Xu, Wenjun Chen, Jun Sha, Kanghong Yan, Dongyuan Yao, Xiaoyan Liu, Shaoquan Liao, Jitong Zhong, Shengrong Yang, Yangbin Yu, Yanlin Tong, Zefeng Xu, and Yu-Sheng Lin*

State Key Laboratory of Optoelectronic Materials and Technologies,
School of Electronics and Information Technology, Sun Yat-Sen University,
Guangzhou, China
E-mail: linyoush@mail.sysu.edu.cn

Abstract—A large-scale lithography-free metamaterial (MM) is presented, which is integrated with a microfluidic chip for effective glucose sensing. MM is composed of hybrid nanopillars fabricated with SiO₂ and Au thin-films on Si substrate. It exhibits angle-independence, and polarization-independence, which are the most important factors for color filters for industrial applications. The resonance shift is 56 nm with a stable bandwidth (~30 nm) by varying concentration of glucose solution. The sensitivity is 158 nm/RIU and the corresponding figure-of-merit is 5.3.

Keywords—metamaterials, plasmonics, lithography-free, refractive index sensors, chemical sensors

I. INTRODUCTION

Recently, the researches in metamaterial (MM) fields are rapidly maturing. One emerging field of them is taking advantage of the optical properties of nanostructural resonances for sensors applications. Owing to the method of MM-based sensors is real-time and label-free, it is considered an ideal sensing technique in live cell monitoring and biomolecules sensing [1,2]. To possess potential applications of MMs, their angle-insensitivity and polarization-insensitivity are very important. However, most of MMs often exhibit angle-sensitive or polarization-sensitive behavior caused from the symmetric structure or asymmetric structure with periodic configurations, respectively [1-4]. It is a trade-off to have both optical properties. Therefore, the desire to realize high performance of angle-independence and polarization-independence in the visible wavelength range has long been a research topic of interest for scientists. Furthermore, the requirement of easily device manufacturing is also a particular theme, which is suitable for current technology with cost-effective production.

In this study, we develop a novel MM composed of hybrid nanopillars fabricated with SiO₂ and Au thin-films on Si substrate by using natural lithography technique [5]. The proposed MM exhibits angle-independence, polarization-independence and wafer-level fabrication. Such stable optical performance permits the MM could be integrated with a microfluidic chip to realize an aqueous glucose sensor and provides a greater possibility for industrial use.

II. MATERIALS AND METHODS

Fig. 1 shows the optical simulation results of MM with varying metal and dielectric height by using software package FDTD solutions. The structural geometry of MM is shown in the

insert schematic drawing of Fig. 1(a). The distribution of nanopillars is random. The heights of metal and dielectric are denoted as H_1 and H_2 , respectively. In Fig. 1(a), the metal height (H_1) is constant as 50 nm. The reflection intensity is enhanced by increasing the dielectric height (H_2) and then saturated at $H_2 > 200$ nm. When H_2 is constant as 200 nm, the reflection intensity is enhanced and a little blue-shifted by increasing H_1 . It will be saturated at $H_1 > 50$ nm. The insert images of Fig. 1(b) show the top view and cross-sectional electric field distributions at the condition of $H_1 = 50$ nm and $H_2 = 200$ nm. The light field is effectively modulated between nanopillars.

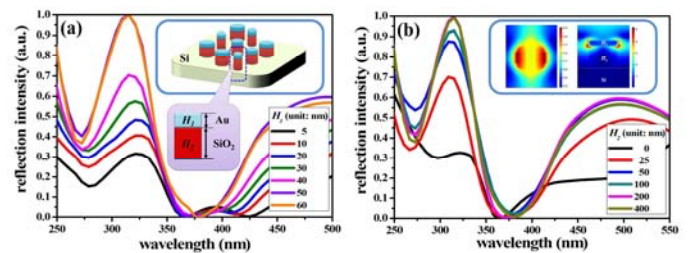


Fig. 1. The optical simulation results of MM with varying metal and dielectric height of nanopillar. (a) Reflective spectra are at the condition of constant metal height ($H_1 = 50$ nm) by varying dielectric height, H_2 . (b) Reflective spectra are at the condition of constant dielectric height ($H_2 = 200$ nm) by varying metal height, H_1 .

The fabrication process of proposed MM-based microfluidic chip is shown in Fig. 2(a). The MM was used natural lithography technique to perform the wafer-level fabrication. Finally, MM was encapsulated in a PDMS microfluidic chip and assembled with tyron tubes, syringe, and syringe pump to drive the deionized (DI) water and different concentration of glucose solution. The structural morphology of MM was characterized by using scanning electron microscopy (SEM). The optical properties of MM and MM-based microfluidic chip without and with different concentration of glucose solution were characterized by using UV-VIS spectrometer equipped on the microscopy. Fig. 2(b) and (c) are top view and cross-sectional view of SEM images of MM. It can be seen the hybrid nanopillars are distributed uniformly. Each hybrid nanopillar is composed of Au and SiO₂ with the thickness of 50 nm and 200 nm, respectively. To perform the optical responses to different concentration of glucose solution, which were diluted with deionized (DI) water to 0%, 1.25%, 2.5%, 5%, 10%, and 20%, individually, and then pipetted into a reservoir of MM-based microfluidic chip, respectively. The volume of solution is 100

μL and the reservoir area is 1 cm^2 . These glucose solutions give a good spread of refractive index values, enabling us to investigate the efficacy of glucose sensing with different concentration of glucose solution.

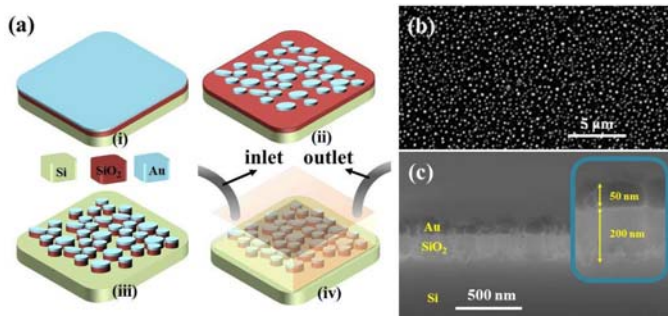


Fig. 2. (a) Fabrication process of proposed MM-based microfluidic chip. (b) and (c) are top view and cross-sectional view of SEM images of MM, respectively.

III. RESULTS AND DISCUSSIONS

Fig. 3 shows the optical experiment results of MM measured at TE and TM modes from normal incidence to different incident angle, respectively. The polarization state of incident light for both modes are also indicated in the insert schematic drawings of Fig. 3(a) and (b), respectively. In Fig. 3(a), the resonant wavelength of the sample measured at TE mode is 315 nm at normal incidence. When the polarization angle of TE mode is tilted to 80° sequentially, the resonance is identical. It is the same electromagnetic responses for the sample measured at TM mode as shown in Fig. 3(b). The optical responses of MM are clearly identical for both TE and TM modes due to the electromagnetic field in the hybrid nanopillars are isotropic for a wide range of incident angle. The resonant peaks and the bandwidths are very stable for all measured reflective spectra. According to the above mentioned, MM with the hybrid nanopillars indeed exhibits the angle-independent and polarization-independent characteristics. Such stable optical performance with wide incident angle shows the potential applications in biological and chemical sensing.

To further investigate the sensing capabilities of MM, the glucose solution with different concentration was used and injected into MM-based microfluidic chip. Fig. 4(a) shows the reflective spectra of MM-based microfluidic chip filled with different concentration of glucose solution. By plotting the resonance shift of glucose solutions with corresponding refractive indices, we obtain the sensitivity factor (S , in nm per refractive index unit (RIU)) of MM-based microfluidic chip. The refractive index of each glucose solution was measured by the refractometer (model: RA-130, KEM, Kyoto Electronics Manufacturing Co., Ltd., Tokyo, Japan). The resonances are red-shifted 56 nm from the concentration of glucose solution of 0% to 20%. The relationship of resonances to different concentration of glucose solution is plotted in Fig. 4(b). The trend is linear for recognizing different concentration of glucose solution. The sensitivity factor is 158 nm/RIU. In order to provide better quantification, figure-of-merit (FOM) is an important factor to determine the capability of MM-based chemical sensor. This FOM is calculated to evaluate the performance of device for glucose sensing. We can obtain the value of FOM as 5.3. Such FOM value is comparable to many reported on-chip bulk

plasmonic sensors [1-4]. Finally, the design of proposed MM may serve as a highly efficient glucose sensor in the visible wavelength range. This will open a route for MM applications in the nanoenvironmental conditions provided by microfluidic chips.

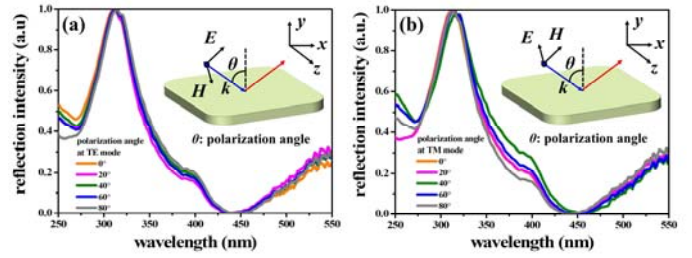


Fig. 3. The optical experiment results of MM measured at (a) TE and (b) TM mode from normal incidence to incident angle of 80° , respectively. The polarization state of incident light for TE and TM mode are indicated in the insert schematic drawings of (a) and (b), respectively.

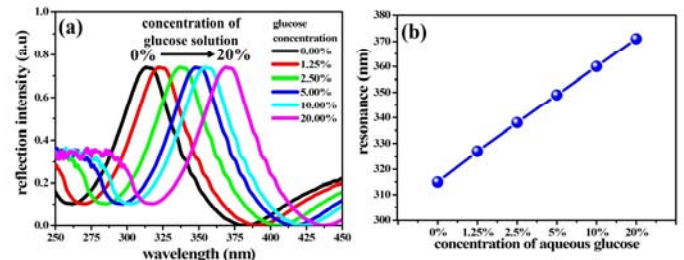


Fig. 4. The experimental results of MM-based microfluidic chip. The resonances are red-shifted when the sample immersed in DI water and different concentration of glucose solution. The insert figure shows the resonance as a function of concentration of glucose solution.

IV. CONCLUSION

In conclusion, we developed a large-scale lithography-free, wide-incident-angle, and polarization-independent MM for glucose sensing. The surface of MM is distributed hybrid nanopillars uniformly. The sensitivity and FOM of MM-based microfluidic chip are 158 nm/RIU and 5.3, respectively. It opens an avenue to a remote optical sensing mechanism by using the chemical solutions with different refractive index to adjust the resonance of MM. This sensing method with high portability, applicability, and cost-effectiveness using MM might minimize limitations of time and space in environmental monitoring, food safety and other fields.

REFERENCES

- [1] X. Li, M. Soler, C. Ozdemir, A. Belushkin, F. Yesilkoy, and H. Altug, "Plasmonic nanohole array biosensor for label-free and real-time analysis of live cell secretion," *Lab Chip*, vol.17, pp. 2208-2217, 2017.
- [2] S. Szunerits and R. Boukherroub, "Sensing using localised surface plasmon resonance sensors," *Chem. Commun.*, vol.48, pp. 8999-9010, 2012.
- [3] J. B. Lassiter, H. Sobhani, J. A. Fan, J. Kundu, F. Capasso, P. Nordlander, and N. J. Halas, "Fano Resonances in Plasmonic Nanoclusters: Geometrical and Chemical Tunability," *Nano Lett.*, vol.10, pp. 3284-3189, 2010.
- [4] J. B. Lassiter, H. Sobhani, J. A. Fan, J. Kundu, F. Capasso, P. Nordlander, and N. J. Halas, "Fano Resonances in Plasmonic Nanoclusters: Geometrical and Chemical Tunability," *Nano Lett.*, vol.10, pp. 3284-3189, 2010.
- [5] Y. S. Lin, W. C. Hsu, K. C. Huang, and J. A. Yeh, "Wafer-level fabrication and optical characterization of nanoscale patterned sapphire substrates," *Appl. Surf. Sci.*, vol.258, pp. 2-6, 2011.