A Large-Scale, Omnidirectional, and Polarization-Independent Metamaterial-Based Chemical Sensor

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***Abstract*—We develop an effective method for volatile organic solutions (VOS) sensing by using metamaterial with hybrid nanopillars to form a metamaterial-based chemical sensor (MCS). It exhibits large-scale, omnidirectional, and polarization-independence characterizations, which are the most important factors for sensors applications. The experiment results indicate the sensitivity and figure-of-merit of MCS are 71 nm/RIU and 3.5, respectively. The proposed MCS can successfully recognize inorganic and organic solutions for chemical sensors in the UV-visible wavelength range.**

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

I. Introduction

Researches in electromagnetic (EM) metamaterial fields are rapidly maturing. One emerging field of them is taking advantage of the optical properties of metamaterial to characterize how these extreme EM properties achieved through the interaction of the incident wave with shape and size of designed patterns. Such inductive coupling mechanism enables the EM energy to transport in structures with transverse dimensions as well as opens an avenue to a variety of applications in practical sensors [1]. In view of metamaterial-based chemical sensor (MCS), that resonance is very sensitive on the dielectric surrounding medium [2]. The EM metamaterials are associated with an electric and magnetic fields propagating along the metal-dielectric interface. Most of energy is then confined to the metal surface which explains the remarkable sensitivity of metamaterials with respect to changes in optical parameters at the metal-dielectric boundary. Although there have already demonstrated a lot of chemical sensors in the different spectra range [1,2], detecting volatile organic solutions (VOS) with a small volume or in unclosed system are more challenging than other chemical solutions. Owing to VOS are non-ionic, non-fluorescent, easy to vapor, and do not absorb UV, the available analytical approaches for detecting VOS in microfluidic channels are quite essential. To possess potential applications of metamaterial, their angle-insensitivity and polarization-insensitivity are very important. However, most of metamaterials often exhibit angle-sensitive or polarization-sensitive behavior caused from the symmetric structure or asymmetric structure with periodic configurations, respectively [1-3]. It is a trade-off to have both optical properties. Therefore, the desire to realize high performance of angle-independence and polarization-independence has long been a research topic of interest for scientists.

In this study, we develop a novel MCS composed of hybrid nanopillars. The EM field of metamaterial is a combined effect of the incident field and radiation from each individual nanopillar, which has a significant resonance of the reflective light between these nanopillars. It exhibits large-scale, omnidirectional, and polarization-independence characterizations. Such stable optical performance permits the MCS to realize a chemical sensor and provides a greater possibility for industrial use.

II. Materials and Methods

Fig. 1(a) shows the fabrication process of MCS using direct metal deposition and post thermal annealing treatment to fabricate hybrid nanopillars. Fig. 1(b) shows the cross-sectional SEM image of MCS. It can be seen the hybrid nanopillars are distributed randomly and uniformly. The height of Au and SiO_2 are 50 nm and 200 nm, respectively. The insert top-left images are top view and cross-sectional view of simulated electric field distributions. The light field of EM metamaterial is effectively modulated between nanopillars. To verify the chemical sensing characteristic by using proposed metamaterial, the sample was encapsulated in a PDMS-based microfluidic channel, which was filled without and with various kinds of VOS to compare their refractive indexes (RIs) and then investigate the sensitivities of MCS. The schematic diagram of experimental setup is shown in Fig. 1(c).

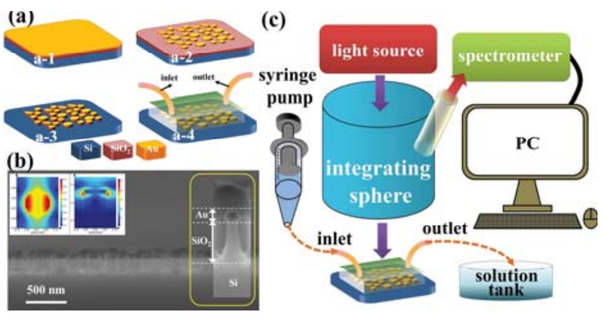

Fig. 1. (a) Fabrication process of proposed MCS. (b) Cross-sectional SEM image of metamaterial with hybrid nanopillars, while the top-left images are the top view and cross-sectional view of simulated electric field distributions. (c) Schematic drawing of experimental setup.

III. Results and Discussions

Fig. 2 shows the optical experiment results of MCS measured at TE and TM modes from normal incidence to incident angle of 80^o, respectively. The optical responses of metamaterial are

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clearly identical for both modes due to the EM field in the hybrid nanopillars are isotropic for a wide range of incident angle. The resonances and the bandwidths are very stable for all measured reflective spectra. Fig. 3 shows the reflective spectra of MCS filled with different VOS. The resonances are red-shifted 19 nm. The resonance shifts of the various VOS are plotted against their respective RI in the insert figure of Fig. 3(a). To further investigate the VOS sensing capabilities of MCS, the $C_2H_4(OH)_2$ solution (largest RI difference) with different concentration was chosen and injected into the sample as shown in Fig. 3(b). The insert figure of Fig. 3(b) plots the relationship of resonances of MCS to different concentration of $C_2H_4(OH)_2$ solution. The trend is linear for recognizing different VOS concentration. The sensitivity of MCS is 42, while figure-of-merit (FOM) is calculated as 1.4.

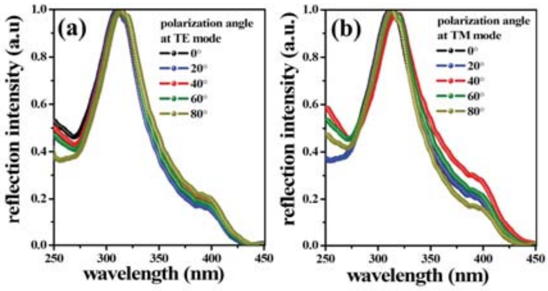

Fig. 2. The optical experiment results of MCS measured at (a) TE and (b) TM mode from normal incidence to incident angle of 80°, respectively.

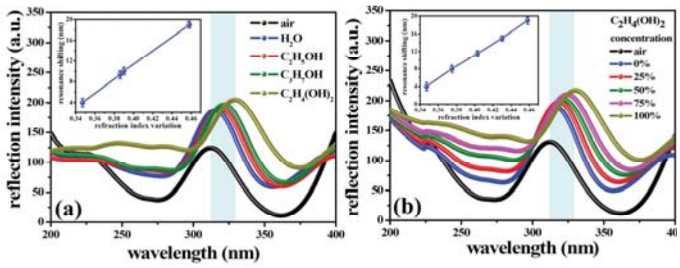

Fig. 3. (a) The resonance shifting of MCS filled without and with different VOS. (b) MCS is filled with different concentration of $C_2H_4(OH)_2$.

To further enhance the sensitivity of MCS, we designed and fabricated another MCS, which surface morphology was composed of hybrid Au/SiO_2/Si nanopillars as shown in Fig. 4(a). To distinguish two mentioned samples, we denoted the previous MCS with hybrid Au/SiO_2 nanopillars as MCS_1, while the MCS with hybrid Au/SiO_2/Si nanopillars as MCS_2. Fig. 4(b) shows the experiment results of MCS_2 under various VOS. It is a significant red-shift at the resonances as the RI of the chemical solutions increases. There are two resonances were found in MCS_2, while a single resonance was found in MCS_1, resulting from the two types of surface plasmons between two kinds of metamaterials. The first type of surface plasmon is generated by Au nanoparticles and excited by incident EM light to oscillate at the interface. In Fig. 4(b), the lower reflection intensity of resonance_1 could be explained by the deeper etching condition with around 450 nm of the total height. The interface of SiO_2/Si may give rise to the second type of surface plasmon. When MCS_2 is injected with different VOS, the resonances are red-shifted 32 nm. Fig. 4(c) shows the sensing capabilities of MCS_2 injected with different $C_2H_4(OH)_2$ concentration. The resonance shift is 32 nm compared to device without and with $C_2H_4(OH)_2$ solution. The resonance shift of the various VOS is plotted against their respective RI in Fig. 4(d). The sensitivity and highest value of FOM are 71 (nm/RIU) and 3.5, respectively. It indicates that low energy losses excite from plasmonic resonance contributed by the hybrid nanopillars. It would further prove that the hybridized resonance could enhance EM field confinement of the reflected light to arouse a significant resonance narrowing. Such a metamaterial supports the narrowband resonance as well as the strong energy confinement between hybrid nanopillars, which enables us to realize high sensitivity of chemical solution sensors.

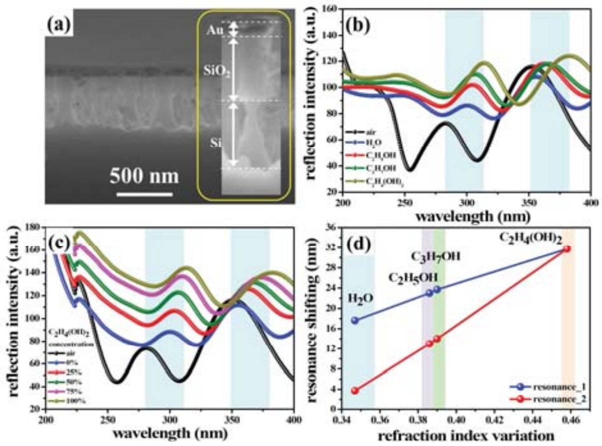

Fig. 4. (a) Cross-sectional SEM image of MCS_2. (b) The resonance shifting of MCS_2 filled without and with different chemical solutions. (c) MCS_2 is filled with different concentration of $C_2H_4(OH)_2$. (d) The relationship of RI variation and resonance shifting of MCS_2.

IV. Conclusion

A large-scale, omnidirectional, and polarization-independent MCS was demonstrated for VOS sensing. The EM resonance was supported in metamaterial constructed by randomly distributed hybrid nanopillars. The surface plasmon between nanopillars could take effect in light field modulation. It facilitates the detection of RI changes. The sensitivities of MCS_1 and MCS_2 are 42 nm/RIU and 71 nm/RIU, respectively. The proposed MCS based on the direct metal deposition and post thermal annealing treatment is simple, cost-effective, and potentially scalable for mass production. Furthermore, these state-of-the-art MCS integrated with microfluidic chips are low cost, label-free, and provide rapid detection with adequate sensitivity. Such strategy might minimize limitations of time and space in chemical sensors, biosensors, food safety and live cell monitoring, etc.

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