Hindawi
Advances in Condensed Matter Physics
Volume 2021, Article ID 2301222, 27 pages
https://doi.org/10.1155/2021/2301222

Review Article

Research Advance on the Sensing Characteristics of Refractive Index Sensors Based on Electromagnetic Metamaterials

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Received 10 May 2021; Revised 30 June 2021; Accepted 10 July 2021; Published 20 July 2021

Academic Editor: Shuyuan Xiao

Among different sensing platforms, metamaterials composed of subwavelength or deep subwavelength sized metal resonance elements arrays that are etched on semiconductor substrates or dielectric substrates exhibit excellent characteristics due to the strong localization and enhancement of resonance electromagnetic fields. As a new type of detection method, metamaterial sensors can break through the resolution limit of traditional sensors for a small amount of substance and have the advantages of high sensitivity, fast response, and simple measurement. Significant enhancement of the sensing characteristics of metamaterial sensors was realized by optimizing microstructures (single split-ring, double split-ring, nested split-ring, asymmetric split-ring, three-dimensional split-ring, etc.), using ultrathin substrates or low-index substrate materials, etching away local substrate, and integrating microfluidic channel, etc. This paper mainly reviews the research advance on the improvement of sensing characteristics from optimizing resonance structures and changing substrate materials and morphology. Furthermore, the sensing mechanism and main characteristic parameters of metamaterial sensors are introduced in detail, and the development trend and challenge of metamaterial sensing applications are prospected. It is believed that metamaterial sensors will have potential broader application prospects in environmental monitoring, food safety control, and biosensing in the future.

1. Introduction

Optical sensors have the advantages of high sensitivity, strong resistance to electromagnetic interference, low noise, high electrical and chemical stability, etc., so they have important applications in life science, food safety, chemical monitoring, and environment monitoring [1–5]. The generalized optical sensor can identify the measured substance or monitor the biomedical reaction process by detecting and analyzing the changes of lightwave intensity, phase, polarization, and other related information [6, 7]. The refractive index [8] is the most typical optical parameter. The real part of the refractive index affects the phase of the light wave, the imaginary part affects the intensity of the light wave, and its anisotropic distribution determines polarization and chirality. Generally, the optical refractive index sensor will introduce a resonance mechanism based on various optical effects to enhance the interaction between the light wave and the measured substance.

Metamaterials [9–12] are usually artificial electromagnetic materials composed of metal resonance elements arrays with subwavelength or deep subwavelength dimensions that are etched on semiconductor substrates or dielectric substrates according to specific rules. It has extraordinary physical properties that natural materials do not have and can control electromagnetic waves in a certain way. Metamaterials are sensitive to changes in the dielectric properties of the surrounding environment and have strong spectral characteristics for the local enhancement of the incident electromagnetic fields. When the dielectric properties (i.e., refractive index) of the surrounding environment change, the resonant characteristics (resonant amplitude, resonant frequency, phase, etc.) of the electromagnetic waves passing through the metamaterials will change accordingly. Therefore, by observing or measuring the changes, the detection of nonlinear substance around the metamaterials and the selection of a very small amount of objects to be tested can be achieved.

Metamaterial sensors [13–16] are optical sensors that can convert changes in the external refractive index into changes in optical signals. As a new type of detection method, metamaterial sensors can break through the resolution limit of traditional sensors, and have the advantages of high sensitivity, fast response, and simple measurement. In the past few decades, metamaterial sensors have made great progress in structural development and applications. Metamaterial sensors have the ability to restrict light to the nanoscale region and have high selectivity, making them have broad application prospects in the fields of environmental monitoring, drug discovery, food safety control, and temperature sensing.

Generally, the sensing characteristics of metamaterial sensors are significantly improved by optimizing microstructure and changing substrate material and morphology, which includes two main aspects, namely, improving sensitivity and improving quality factors. Improving the sensitivity of metamaterial sensors requires that the local field distribution of the resonant mode of the electromagnetic wave can overlap spatially to a greater extent with the substance being measured, that is, increasing the sensing area to enhance the interaction. Improving the quality factor of the metamaterial sensors requires lower losses in resonant mode.

The purpose of this article is to review the research advance on the sensing characteristics of metamaterial sensors. The basic principles, detecting processes, and sensing characteristics of various metamaterial sensors based on different microstructures and substrates are presented in detail, and the development trend and prospects of the metamaterials sensing technology are discussed. In addition, metamaterial sensors have potential applications in areas such as environmental sensing, homeland security, and biosensing.

2. Sensing Mechanism and Characteristic Parameters of Metamaterial Sensors

2.1. Sensing Mechanism. The operation principle of metamaterial sensors is based on the change in reflection and transmission coefficients (scattering parameters, S_{11} and S_{21}), which is induced in variation in the permittivity, permeability, or refractive index of the metamaterial resonator. The change in the dielectric constant around the sensor translates into a change in the electromagnetic signal spectrum in the form of an offset in the position of the resonance peak.

Metamaterials are artificial electromagnetic materials composed of arrays of subwavelength or deep subwavelength metal resonance elements fabricated on semiconductor substrates or dielectric substrates. The resonances are mainly divided into low-frequency resonances and high-frequency resonances. Low-frequency resonance can be understood as the coupling of capacitors and inductors, and resonant frequency can be expressed as follows [17]:

$$\omega_{LC} = (LC)^{-(1/2)} = \frac{1}{\sqrt{L}\sqrt{\varepsilon_0 \int^{\nu} \varepsilon(\nu) E(\nu) \mathrm{d}\nu}}. \quad (1)$$

As can be seen from formula (1), the resonant frequency is mainly determined by the inductance and capacitance. The inductance is mainly determined by the geometric parameters of metamaterials. If it is a nonmagnetic material, as long as the geometric parameters of the metamaterials are determined, the inductance will not change. The capacitance is related to the permittivity and electric field of the surrounding medium. As the permittivity of the surrounding environment changes, the overall capacitance will change, causing a change in the resonant frequency. For the change of electric field, high quality factor resonance and strong local field distribution can be realized by designing a special metamaterial structure, thereby improving the sensitivity of the sensor.

The high-frequency resonance of metamaterials can be understood as plasmon resonance, and its resonant frequency is expressed as follows [18]:

$$\omega_d \propto \frac{1}{\left(2d\varepsilon_{\mathrm{eff}}^{1/2}\right)}. \quad (2)$$

Here, d is mainly determined by the geometric parameters of the metamaterials, and $\varepsilon_{\mathrm{eff}}$ is the average permittivity of the environment. When the substance to be measured is connected to the metamaterial structure, it will cause the surrounding permittivity to change and then change the resonant frequency.

Since the metamaterial microstructure interacts with the substrate, and there is also capacitance between the substrate and the metamaterial microstructure, the change in the substrate also causes a shift in the resonant frequency of the metamaterials. The overall capacitance of the metamaterials can be expressed as follows [19]:

$$c = \sum_{i=1}^{4} c_i, \quad (3)$$

where c_1 is the capacitance of the substrate, c_2 is the capacitance between the substrate and the metamaterial microstructure, c_3 is the capacitance of the metamaterial microstructure itself, and c_4 is the capacitance between the metamaterial microstructure and the substance to be measured.

If the metamaterial microstructure is fabricated on a substrate with high resistance, high permittivity, and a relatively large thickness, c_1 contributes a lot to the overall capacitance, the capacitance change of the metamaterial itself is relatively small, and the sensitivity is correspondingly low. Therefore, reducing the relative contribution of the substrate, such as using a low permittivity, small absorption, and thin substrate, can also improve the sensitivity of the metamaterial sensors.

2.2. Characteristic Parameters. The sensing behaviors of metamaterial sensors are evaluated with well-known indicators, the quality factor Q, the sensitivity S, and the figure of merit FOM.

Assuming f_0 is the resonant frequency of the metamaterial sensor, it is very related to the structural parameters of the metamaterial sensor and the external environment, so the weak refractive index disturbance Δn will bring the resonant frequency shift Δf. By detecting the change of resonant frequency through the spectrum analysis system, the information of the measured substance can be obtained. However, the spectrum analysis system has its hardware limitations in terms of weak signal detection and spectral resolution, so we need to optimize the design of the metamaterial sensor structure to achieve greater changes in Δf. Generally, the refractive index frequency sensitivity of the metamaterial sensor is defined as $S(f) = \Delta f/\Delta n$, where Δf represents the change in the resonant frequency of the metamaterial sensor and Δn represents the change in the refractive index of the substance to be measured. The unit of Δn is RIU (Refractive Index Unit), which represents the unit refractive index. However, because the sensitivity $S(f)$ of the metamaterial sensor is related to the working band, the normalized sensitivity $S(f)'$ is used to exclude the influence of the working band, which is defined as follows: $S(f)' = S(f)/f_0$.

Generally, the quality factor Q reflects the resonance characteristics of the sensor. That is, the sharper the resonance peak, the greater the corresponding value, and the higher the sensitivity of the sensor. In addition, the quality factor Q also determines the resolution of the sensor. The larger the value, the higher the resolution of the sensor. The quality factor of the sensor can be defined as follows: $Q(f_0) = f_0/\text{FWHM}$ (where f_0 is the resonant frequency of the metamaterial sensor and FWHM (Full Width at Half Maximum) is the half-height width of the resonance peak).

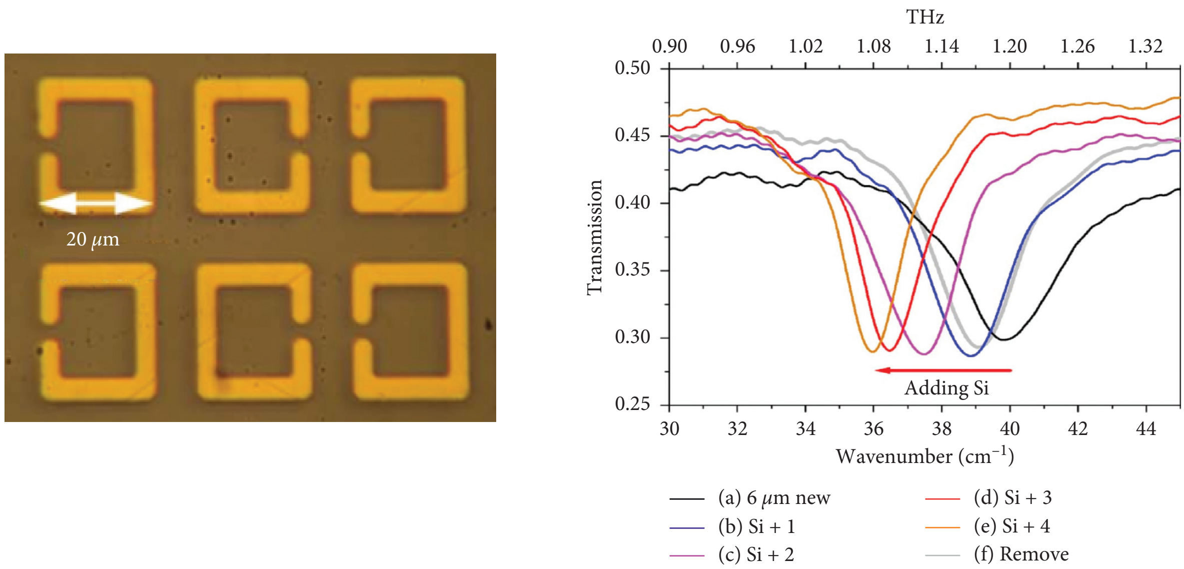
In order to make a more reasonable comparison of the characteristics of sensors working in different frequency bands, FOM (figure of merit) is usually used to describe the characteristics of the sensors. When the sensing sensitivity is the same, the larger the FOM and the better the sensing characteristics. Generally, FOM can be defined as FOM = S/FWHM, where S represents the refractive index frequency sensitivity of sensors and FWHM represents the half-height width of the resonance peak, that is, the sensing characteristics that guarantee both high sensitivity $S(f)$ and high quality factor Q is the best.

3. Research Advance in Improving the Sensing Characteristics of Metamaterial Refractive Index Sensors

Following the definition of FOM, the enhancement of the sensing capacity of metamaterial sensors can be considered from two aspects. One is to design metamaterial sensors with higher quality factor Q, while the other is to make the relevant, resonant frequency shift Δf with a larger step for the same surrounding changes. The first route is to optimize the metal resonance structures on the metamaterial surface to achieve high Q resonances, and in recent years, many novel resonance structures have been shown. Another route is to highlight the effects of changes in the surrounding dielectric by reducing substrate effects to achieve greater spectral modulation under the same surrounding dielectric changes.

3.1. Optimizing Metamaterial Structures. For metamaterials, a split-ring resonator (SRR) is the most commonly exploited topology. SRRs can be achieved by etching ring-shaped metal patterns with one or more opening gaps on the substrate surface, where the rings can be regarded as inductors, and the opening gaps can be regarded as capacitors. From the perspective of the equivalent circuit model, the SRR structure constitutes the LC oscillation circuit, and the resonant frequency is expressed as formula (1). The structural parameters of SRR determine the resonant frequency, which can be achieved at any frequency by proper selection. When the surrounding environment of the SRR changes, the equivalent capacitance and inductance corresponding to the SRR will be inevitably changed, which in turn will change the resonant frequency, so sensing can be achieved by detecting this change. In addition to SRR, metallic wires, with their simple shape, are often used as structural units in the composition of metamaterials. When the direction of the electric field of the incident electromagnetic wave is parallel to the wire, the positive and negative charges accumulate at each end of the wire, corresponding to a pair of electric dipoles. The alternating electric field causes a reciprocal movement of the charges inside the wire, so the wire is equivalent to a pair of oscillating dipoles under the electromagnetic field, and its resonant mode is dipole resonance, with the same resonant frequency as the applied electric field. Fano resonance is caused by the interference phase between a wider spectral line and a narrower discrete resonance cancellation or phase length, and Fano resonance has a higher Q-factor than dipole resonance and LC resonance.

In 2007, Driscoll et al. [18] successfully prepared tunable terahertz filters on a benzocyclobutene (BCB) film and a silicon (Si) particle film based on symmetric SRRs array metamaterial surface, and the mixed droplets of alcohol and nanosilicon spheres were coated on the surface of metamaterials. Observing the transmission spectrum of the metamaterials, it was found that its resonant frequency had changed, as shown in Figure 1(a). This is the first time that terahertz metamaterials are used in the field of sensing, which fully shows that such metamaterials can be used as sensors to detect biochemical samples with different dielectric properties. After that, metamaterials based on planar SRRs array were widely used in the field of sensing. Lee and Yook [20] applied planar SRRs metamaterials to biochemical detection for the first time in the microwave band and identified and detected avidin with a frequency shift of 40 MHz, as shown in Figure 1(b). Wu et al. [21] fabricated symmetric SRRs array on Si substrate and quartz substrate to construct terahertz metamaterial sensors. 50 μL streptavidin agarose (SA), biotin, octadecanethiol (ODT), and biotin were mixed to make a sample to be measured. The experimental detection of SA is realized based on the principle of resonant frequency movement measurement, as shown in Figure 1(c). Quartz-based metamaterials will produce two resonance peaks with a high-frequency resonant frequency of about 2 THz. After loading the sample to be measured, the resonance peak will have a frequency shift of 6.76 GHz and Q-factor of 8. As shown in Figure 1(d), in 2019, Han et al. [22] proposed a square SRRs metamaterial sensor based on



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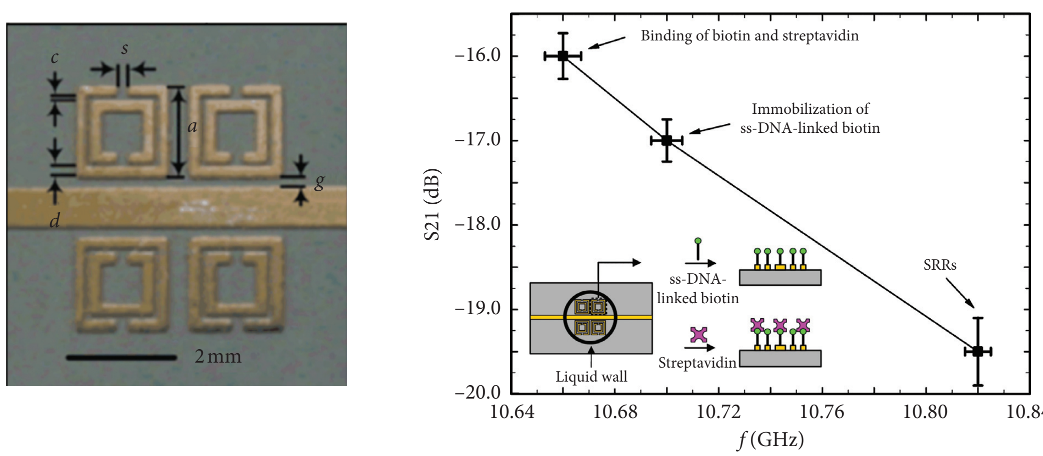

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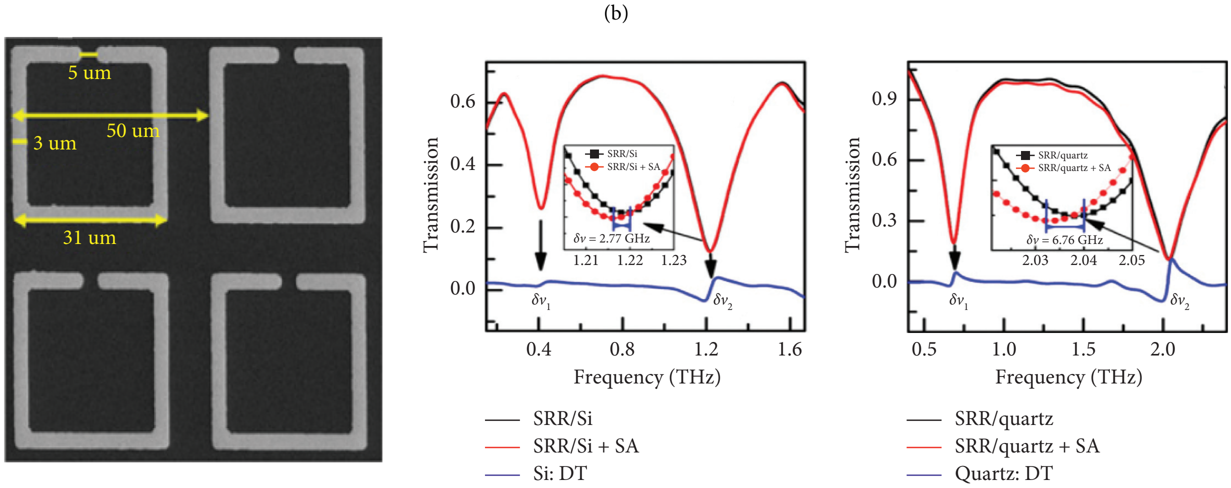

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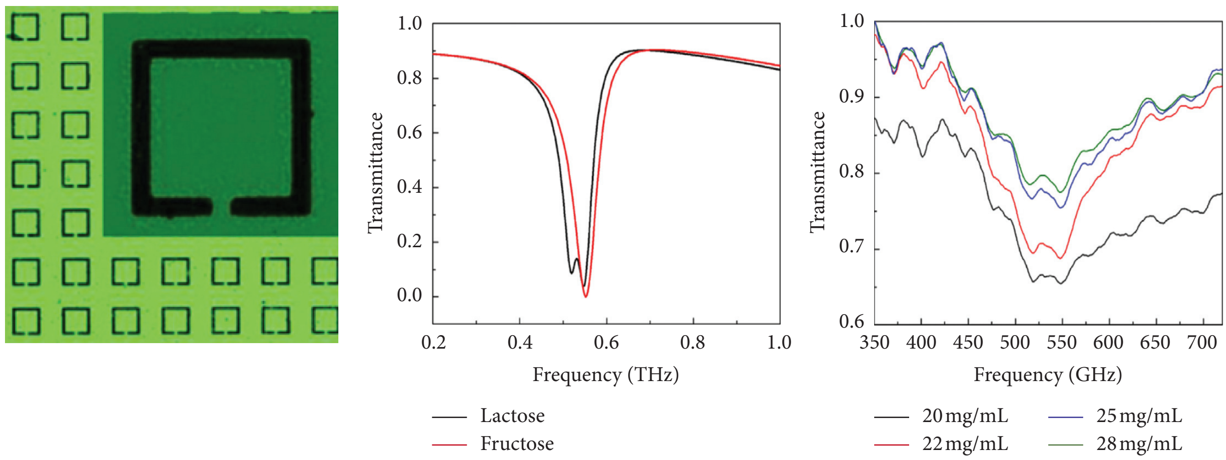


(d)

Figure 1: Schematic diagrams and transmission spectra of metamaterial sensors based on symmetric SRRs arrays. (a) Reproduced with permission from reference [18] copyright 2007, AIP publishing. (b) Reproduced with permission from reference [20] copyright 2008, AIP publishing. (c) Adapted with permission from reference [21] copyright 2015, Elsevier B.V. (d) Adapted with permission from reference [22] copyright 2018, Elsevier B.V.

resonant coupling for the fingerprint detection of lactose. The designed sensor in this work consisted of a periodic array of square-shaped copper SRRs on the top of a quartz substrate, which can achieve sensing detection of lactose concentration. It can be found from the research of these sensors that metamaterials based on SRRs structure can be used for substance detection, but the detection sensitivity is limited, and the sensing characteristics still need to be improved.

In pursuit of better sensing characteristics, such as higher quality factor Q, higher sensitivity and higher figure of merit FOM, dipole resonance, quadrupole resonance, Fano resonance, electromagnetic induced transparency (EIT)-like mode or trapped mode can be excited by metamaterials with asymmetric SRR structures. Breaking structural symmetry of metamaterial resonance structure is one of the common and efficient approaches to achieve such sharp resonances and improve sensing sensitivity.

Singh et al. [23] systematically studied the influence of the opening position of the asymmetric SRRs structure and the polarization of the incident waves. This asymmetric SRRs structure will produce three resonance mechanisms, namely LC resonance, dipole resonance, and quadrupole resonance. It is found that as the opening position is farther from the center of the SRRs, the quality factor Q resulted from quadrupole resonance is higher. In addition, Singh et al. also simulated the surface current distribution of the SRRs structure at the resonant frequency and found that the asymmetric SRRs structure has very weak electromagnetic field scattering when the quadrupole resonance occurs, which greatly reduces the energy coupled to the free space, and finally produces a very sharp resonance peak. It can be seen from Figure 2 that the quadrupole resonance has a resonance peak near 1.75 THz. When the electromagnetic wave is incident with horizontal polarization, the Q-factor can be as high as 95. Therefore, the quadrupole resonant

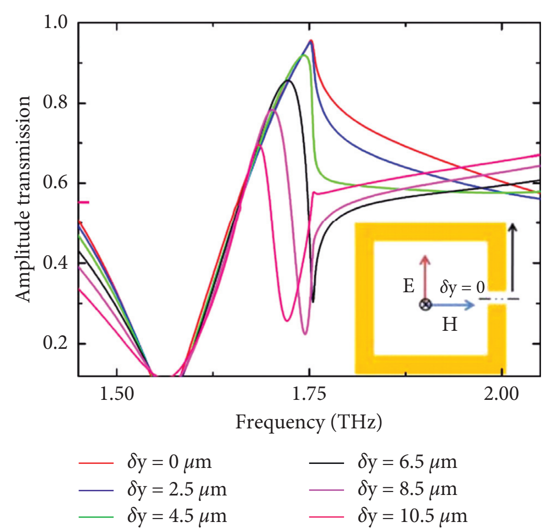


Figure 2: Transmission spectra of metamaterials based on asymmetric SRRs array; adapted with permission from reference [23] copyright 2010, Optical Society of America.

mode can achieve a good localization of the electromagnetic field and will provide new means for sensing applications.

Fano resonance has the advantages of sharp resonance profile, narrow resonance peak width, and strong localized field enhancement, so various Fano resonance metamaterial sensors based on asymmetric SRR structures have been proposed for high sensitivity refractive index sensing [24–26]. Debus and Bolivar [24] designed a double-opening asymmetric SRRs structure, as shown in Figure 3(a). For a perfect conductor material, the calculated Q-factor is as high as 40, and the electromagnetic field is localized around the double-opening SRRs.

When the substance with permittivity of 3.2 and thickness of 10 nm is deposited on the surface of the

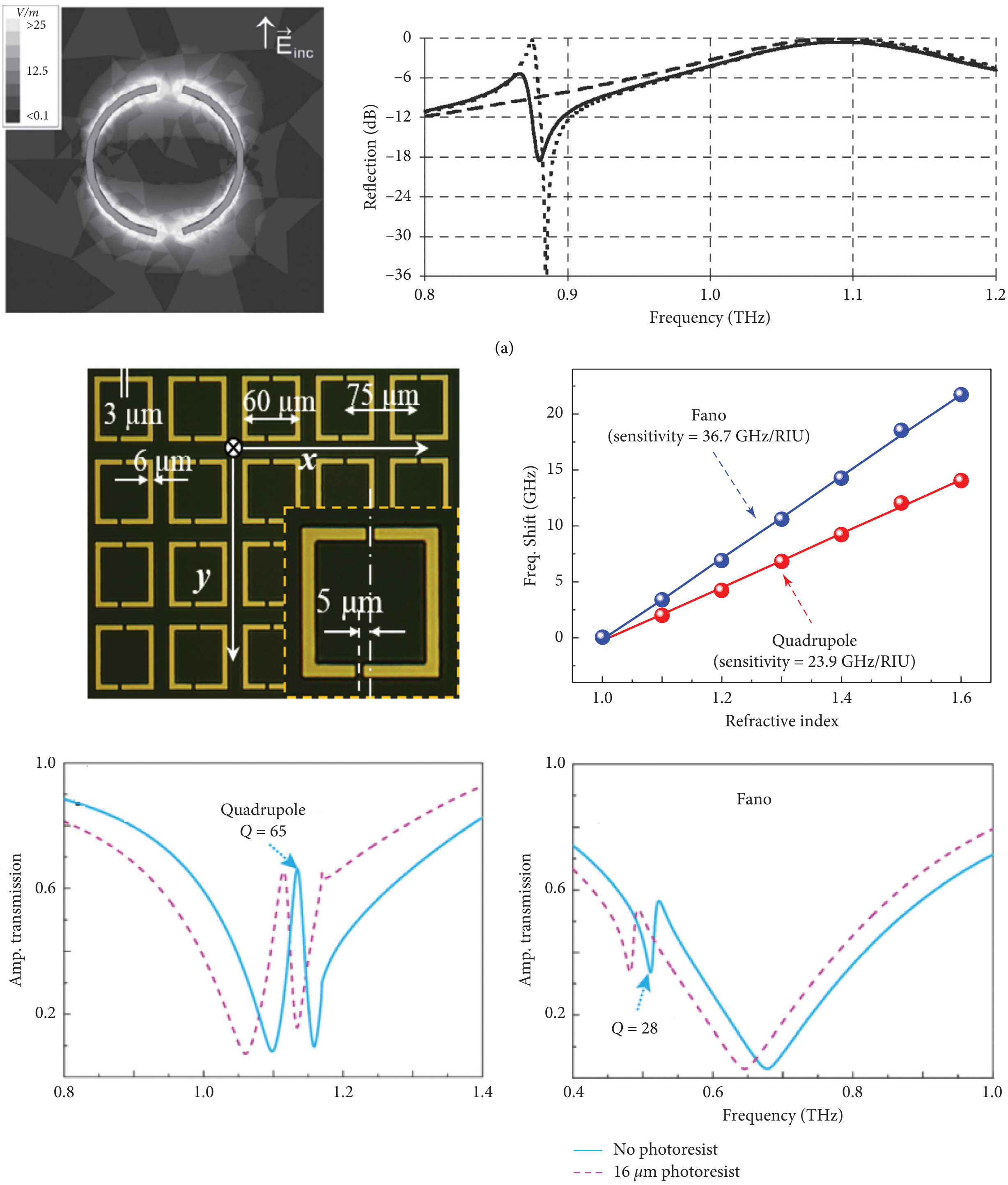


Figure 3: (a) Schematic diagrams and reflection spectra of metamaterial sensors based on double-opening asymmetrical SRRs arrays, reproduced with permission from reference [24] copyright 2007, AIP publishing. (b) Schematic diagrams and transmission spectra of metamaterial sensors based on double-opening asymmetrical SRRs arrays, reproduced with permission from reference [25] copyright 2014, AIP publishing.

metamaterial structure, it will cause the resonance peak at 867 GHz to shift by 5 GHz, which corresponds to $S = 6.3$ GHz/RIU. Singh et al. [25] analyzed the terahertz sensing characteristics of the Fano resonance and the quadrupole resonance in the double-opening asymmetric SRRs structure, as shown in Figure 3(b), the Q-factor of the quadrupole resonance is as high as 65, while the Q-factor of Fano resonance can also reach 28, which is much higher than the Q-factor of the resonance peaks produced by the previous structures. When the thickness of the substance to be measured is kept at $4\,\mu$m, the Fano resonance can achieve maximum sensitivity of 36.7 GHz/RIU, while the quadrupole resonance can achieve a sensitivity of 23.9 GHz/RIU. In 2019, Behera and Kim [26] presented FDTD simulation studies of asymmetric 2D and 3D gold resonators for different applications of high sensitivity, high figure of merit, and circular dichroism polarization and incident angle-independent refractive index sensing, respectively. It is shown that the sensitivity of the triple C-shape asymmetric Fano resonant cavity is improved to 606 nm/RIU and the FOM of the quadruple C-shape resonant is as high as 16.5 and the polarization and angle of incidence are independent.

Although highly sensitive refractive index sensing based on metamaterial sensors can rely on a single-band Fano resonant frequency shift, the effect of environmental changes (e.g., temperature and humidity) on the frequency shift of the resonance cannot be ignored. As a result, relying on a single Fano resonance alone can easily introduce errors when used for sensing applications. Therefore, the research team is now focusing on the multiband Fano resonant frequency domain as a solution to reduce the sensing errors caused by environmental variations [27, 28].

Li et al. [27] demonstrated multiple Fano resonances in an integrated single dark-mode hybrid metamaterial waveguide structure consisting of three gold tangents placed on a dielectric plate waveguide, as shown in Figure 4(a). By breaking the structural symmetry, a quadrupole mode on the middle vertical cut wire is excited and further results in multiple Fano resonances through both the diffracted waves and near-field coupling with bright dark mode. A high Q-factor refractive index sensor with FOM of 330 and 281 is realized. This study provides a method for obtaining multiple high Q-factor Fano resonances that can broaden access for the fabrication of biochemical sensor devices. As shown in Figure 4(b), Kong et al. [28] proposed a multiple Fano resonant metamaterial refractive index sensor consisting of microcavities and microstructures. By fine-tuning the microcavity thickness and microstructure configuration, multiple Fano resonances can be generated for refractive index sensing: depending on the single Fano resonance, the sensitivity can reach 831 nm/RIU and the FOM of 600; in addition, the spectral spacing of the double Fano resonance can also be used for refractive index detection with a sensitivity of 194 nm/RIU, but with significantly reduced errors caused by environmental variations. Refractive index sensors based on multiple Fano resonances have the advantages of high sensitivity, narrow Fano resonance peaks, high ease of integration, and potential to reduce environmental errors, and are expected to provide a new strategy for optical refractive index sensing.

EIT [29] is a classical quantum interference effect that occurs in coherently driven atomic systems at multiple energy levels (triple or quadruple energy levels), where this interference alters the optical response of the atomic medium, creating a clear window of transparency within a wide absorption band. In 2008, Zhang et al. [30] introduced the concept of plasma-induced transparency (PIT) in metamaterials, and a variety of metamaterial structures have since been designed to simulate the EIT effect. PIT is an EIT-like metamaterial behavior that has attracted a great deal of attention due to its wide range of practical applications. Most EIT-like metamaterials are based on two resonant modes, bright and dark or superradiative and subradiative modes. In metamaterials, EIT-like phenomena can be modelled by destructive interference between the bright and dark modes and the superradiative and subradiative modes. For EIT-like phenomena to occur in metamaterials, the two modes should have close resonant frequencies and very different Q-factor [31].

Recently, EIT-like Fano resonances have been confirmed based on cut lines [30, 32, 33] and SRRs [34–37]. Due to the strong interference between two or more resonance modes, the EIT-like-Fano resonance structure has inherent sensitive properties to changes in the local environment. Because of the asymmetric line shape, small perturbations can cause significant spectral changes, including peak-frequency shifts and linewidth changes. This sensitive nature, therefore, makes metamaterials particularly attractive as biomedical and chemical sensing platforms because of their unique advantages, including nonlabelling, nondestructive, time-saving, and low cost compared to conventional bioassays.

However, the practical application of EIT-like metamaterials for biosensing has not yet been fully promoted. On the one hand, the small theoretical sensitivity of the existing biosensors and the single criteria for evaluating their performance make it difficult to apply them in practical engineering. On the other hand, the small variations in frequency shift and miniaturised manufacturing processes of most existing metamaterial biosensors hinder the widespread use of these biosensors in reality. In practice, it is ideal that the transparent window of the EIT phenomenon can be modulated arbitrarily. The corresponding modulation methods are passive modulation and active modulation. Jin et al. [38] designed a novel terahertz metamaterial structure consisting of a pair of subwavelength inverse U-shaped SRR resonators and tangential resonators to achieve the weak coupling region EIT effect. Theoretical and simulation results show that the EIT-like phenomena can be trimmed by modulating the relative coupling distance between the broken filaments and SRRs or the mutual distance between the SRR pairs, as shown in Figure 5. This method is a passive modulation, mainly achieved by changing the geometric parameters of the resonant cavity structure, but it is determined that each modulation requires a refabrication of the structure, which limits its practical application and increases the cost of the application. In addition, the introduction of photosensitive silicon cells in the dark-mode resonant cavity unit enables active optical control of the EIT-like effect by increasing the dark-mode damping rate.

Shen et al. [29] presented a tunable electromagnetically induced reflection effect with a high Q-factor based on a complementary bulk Dirac semimetal terahertz metamaterial structure. As shown in Figure 6, the design consists of a wire-slot structure and an SRR resonator slot structure, which act as a radiating and a subradiating unit, respectively. Destructive interference between these two elements

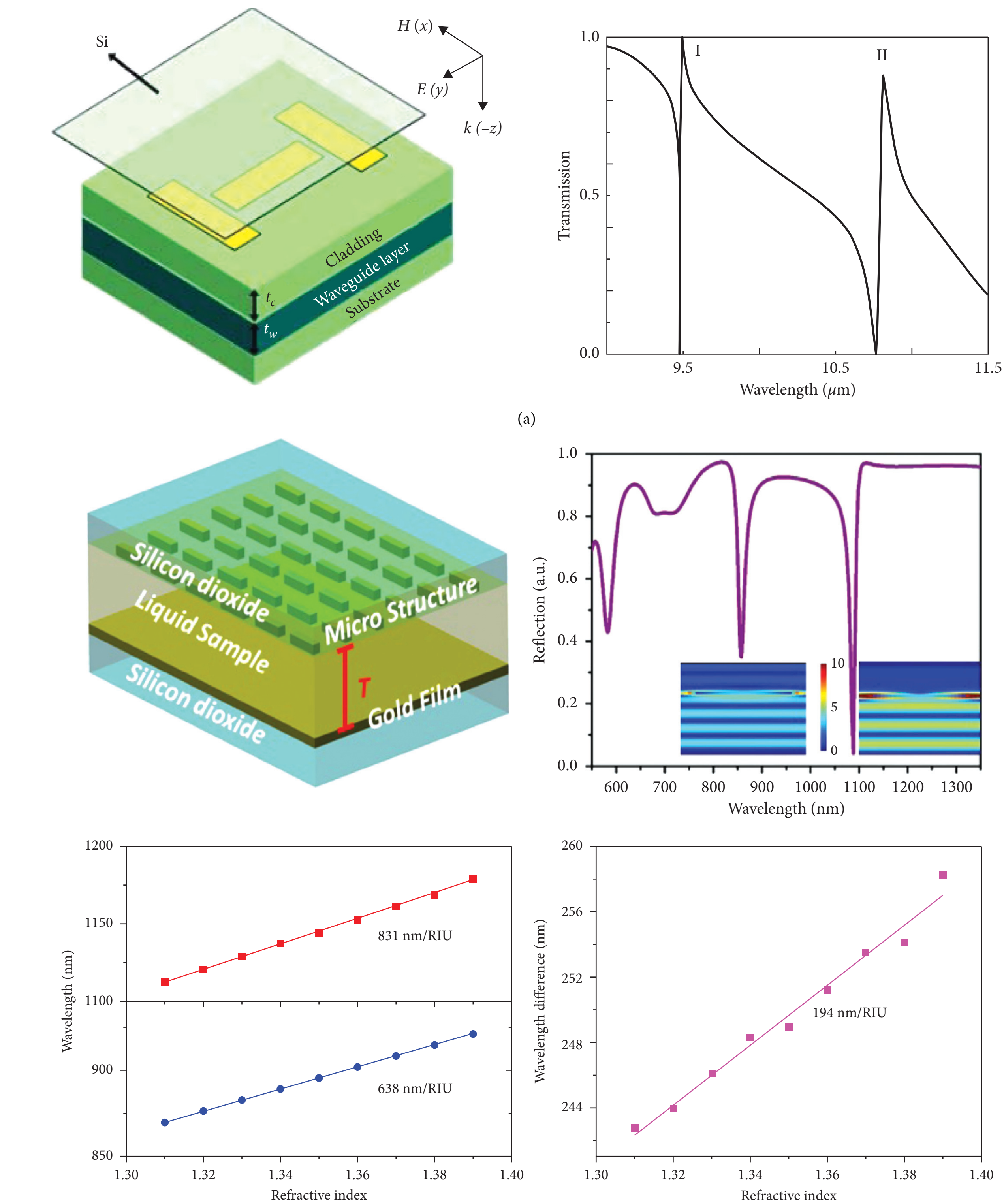


FIGURE 4: Schematic diagrams and transmission spectra of metamaterial sensors with multiple Fano resonances. (a) Reproduced with permission from reference [27] copyright 2018, IEEE. Schematic diagrams, reflection spectra, and sensitivity of metamaterial sensors with multiple Fano resonances. (b) Reproduced with permission from reference [28] copyright 2018, IEEE.

produces a reflection peak with a high Q-factor (~87.6), resulting in sensitive terahertz sensing, where the proposed bulk Dirac semimetallic sensor has a sensitivity of 302.5 GHz/RIU with a FOM of 19. This study provides an alternative approach to the design of terahertz ultrasensitive sensors, filters, and slow optical devices. The EIT-like

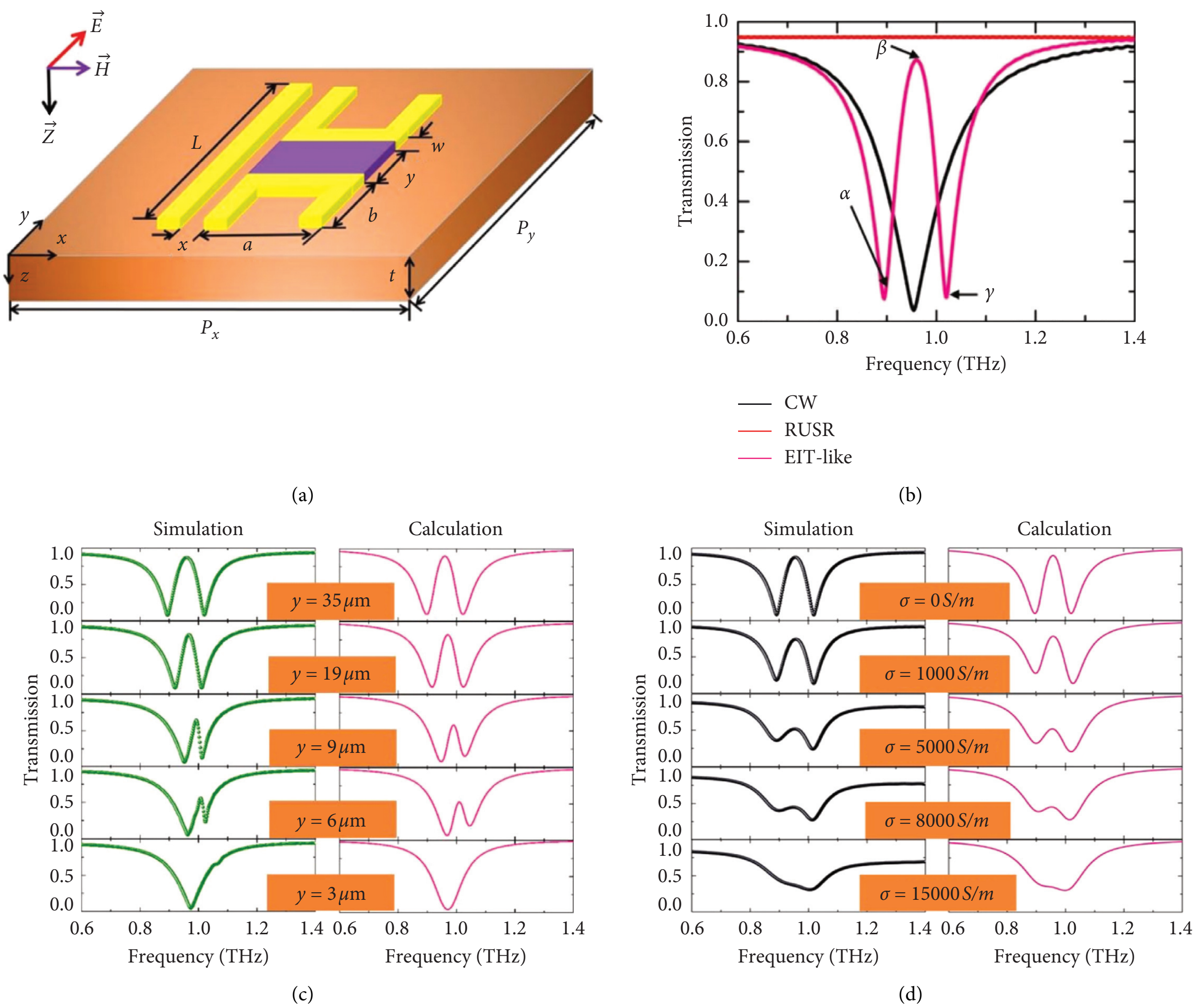
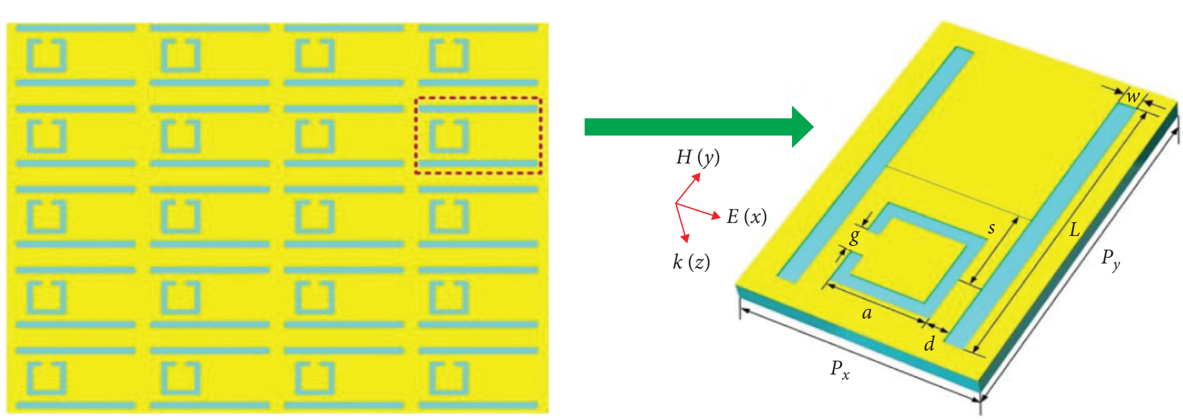


FIGURE 5: (a) Schematic diagram of EIT metamaterials. (b) Simulated transmission profile versus frequency. (c) Modulation of the transparent window when the coupling distance varies. (d) Transmission spectra at different silicon conductivities. (a–d) Reproduced with permission from reference [38] copyright 2019, Springer Nature.



(a)

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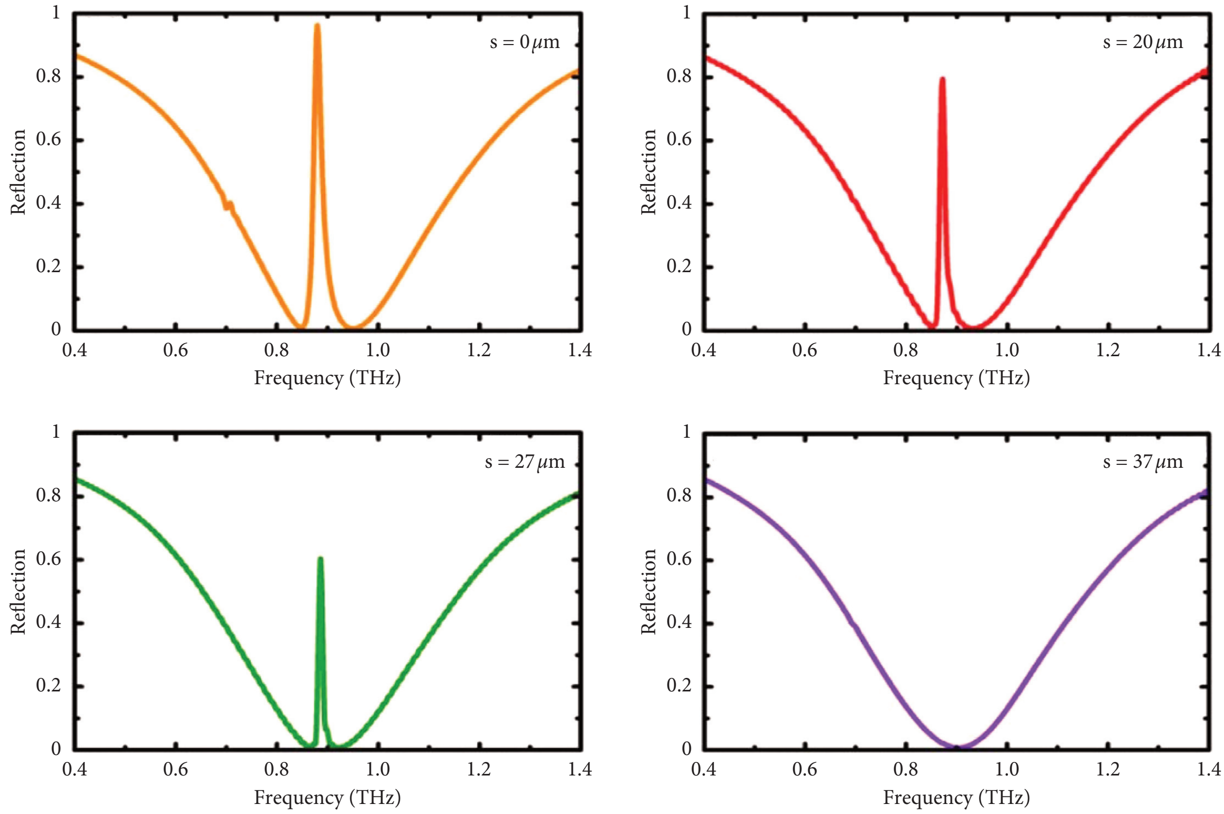

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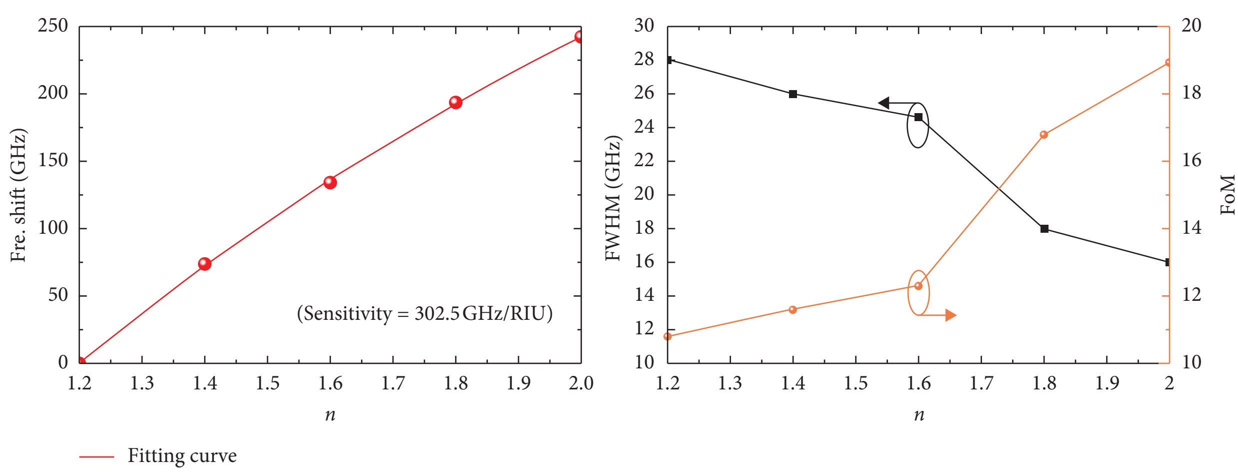

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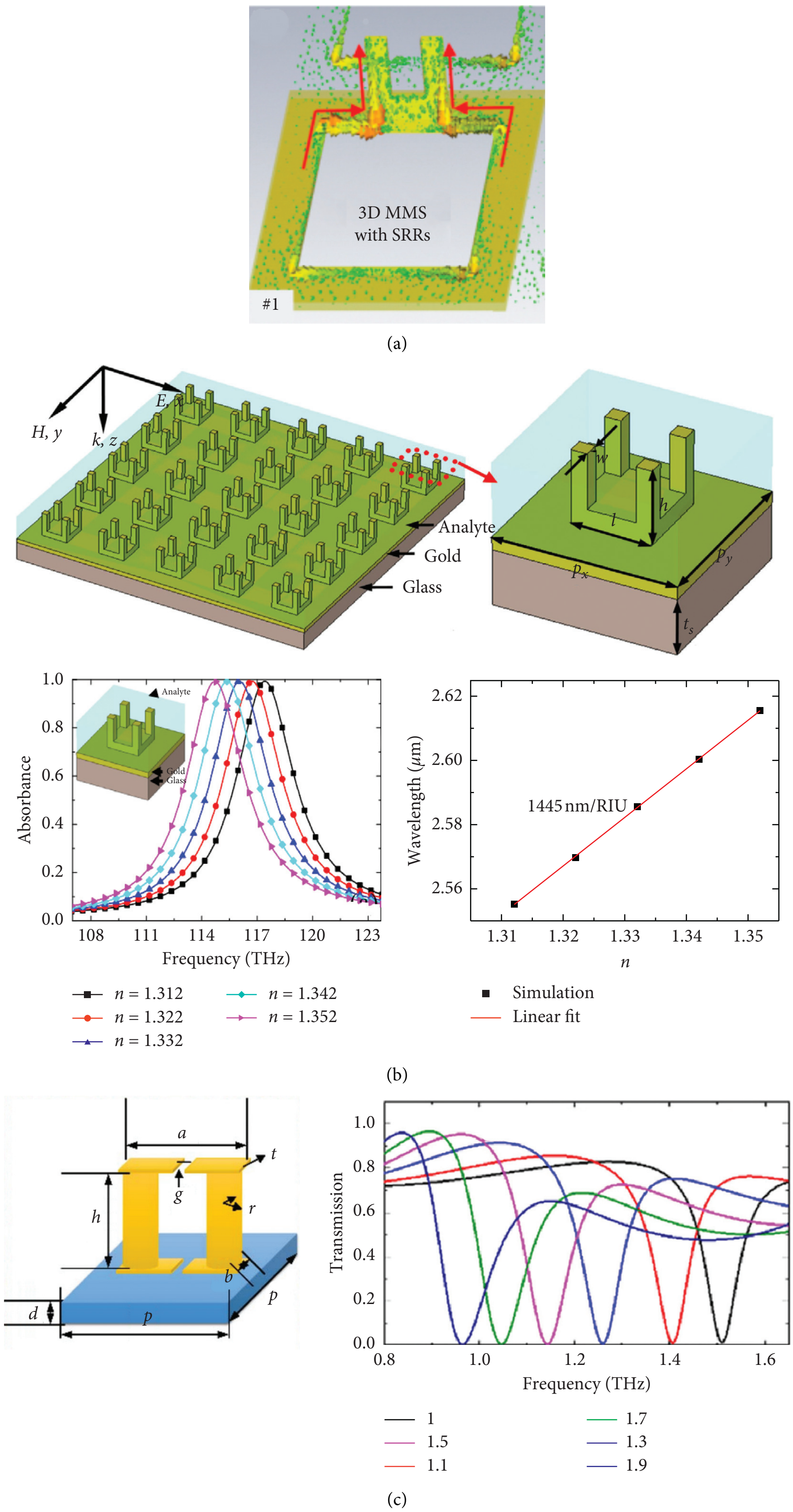
Figure 6: (a) Complementary bulk Dirac semimetal metamaterial structure. (b) Simulated reflection spectra under different lateral displacements ($E_f = 70$ meV). (c) Simulated frequency shift, FWHM, and FOM. (a, b) Reproduced with permission from reference [29] copyright 2014, IOP publishing.

transmission has promising applications in the design of slow-light devices and high sensitivity sensors.

Based on the planar SRRs structure, the resonant mode field is localized between the openings, and part of the mode field is inside the substrate, which limits the interaction between the electromagnetic field and the measured substance, so metamaterial sensors based on three-dimensional SRRs structure have been proposed. Compared with the planar SRRs structure, the electromagnetic field is extended to three-dimensional space, which can increase the mutual contact area with the substance to be measured, thereby improving the sensing sensitivity.

Bian et al. [39] proposed a metamaterial structure based on three-dimensional SRRs. The calculated surface current distribution is shown in Figure 7(a). It can be seen that the energy density is the largest at the opening, and the three-dimensional SRRs will make less energy dissipate on the substrate, which will greatly improve the interaction



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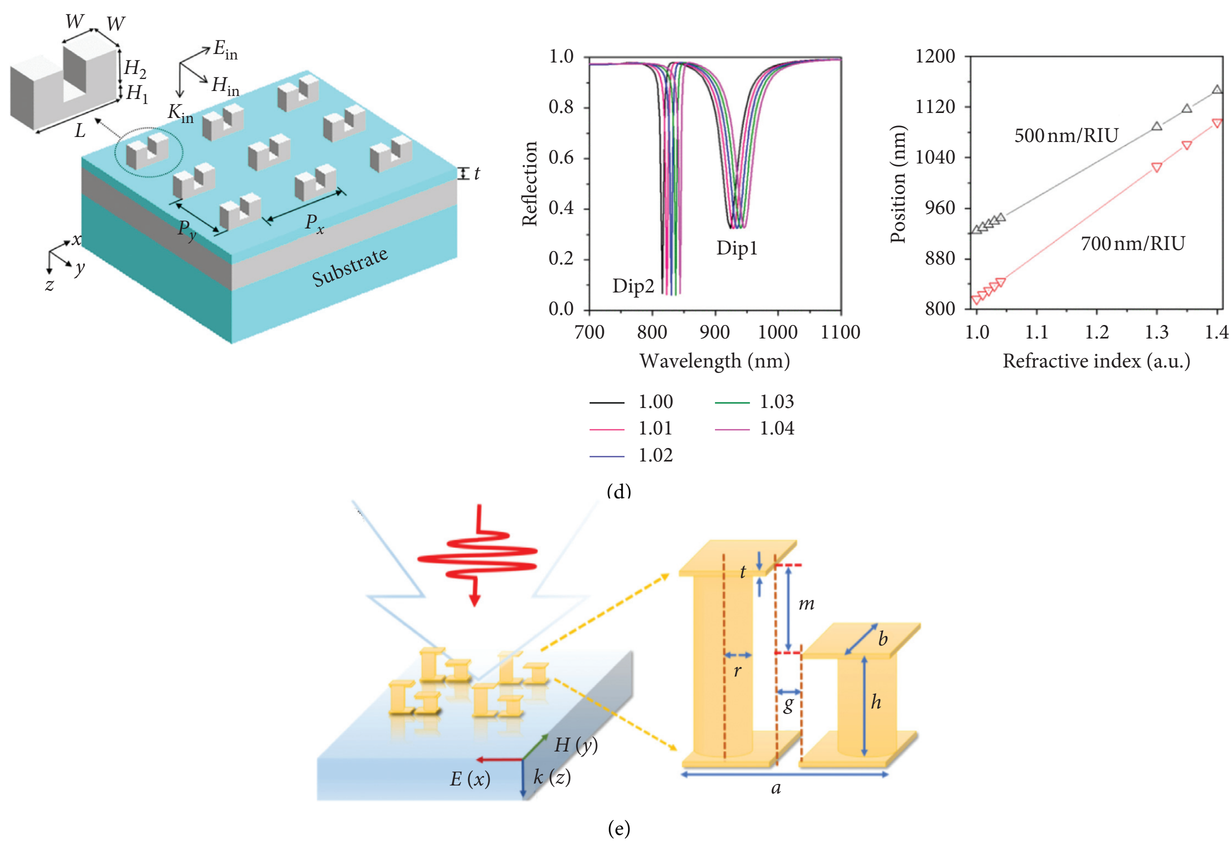


Figure 7: (a) Schematic diagrams of metamaterial sensors based on three-dimensional SRRs arrays. (b) Schematic diagrams, absorption spectra, and sensitivity of metamaterial sensors based on three-dimensional SRRs arrays, reproduced with permission from reference [40] copyright 2016, Elsevier B.V. (c) Schematic diagrams and transmission spectra of metamaterial sensors based on three-dimensional SRRs arrays, reproduced with permission from reference [41] copyright 2017, Optical Society of America. (d) Schematic diagrams, reflection, and sensitivity of metamaterial sensors based on three-dimensional SRRs arrays, reproduced with permission from reference [42] copyright 2018, IEEE. (e) Schematic diagrams of metamaterial sensors based on three-dimensional SRRs arrays, reproduced with permission from reference [43] copyright 2019, IEEE.

between the effective energy and the substance to be measured, and thus improve the sensing sensitivity. Cheng et al. [40] designed a metamaterial sensor composed of four U-shaped SRRs. Its structural schematic and transmission curve are shown in Figure 7(b). The calculated sensitivity reaches 1445 nm/RIU, the Q-factor reaches 41.2, and the FOM reaches 28.8. Wei et al. [41] designed a terahertz metamaterial sensor based on vertical split-ring with double splits. Its structural diagram and transmission curve are shown in Figure 7(c). The calculated sensitivity is 788 GHz/RIU, the Q-factor is about 20, and the FOM is about 10. In 2018, Jing et al. [42] reported an enhanced and modified 3D optical metamaterial with magnetic plasmon resonance, which consists of a periodic array of silver perpendicular SRR resonators, for an effective method of high sensitivity sensing. As shown in Figure 7(d), the sensitivity and FOM of the 3D metamaterials were as high as 700 nm/RIU and 170, respectively, indicating that the proposed 3D metamaterials have promising applications in label-free biomedical sensing. In 2019, Wang et al. [43] presented symmetry breaking introduced in vertical split-ring resonators metamaterials to excite narrow resonance, as shown in Figure 7(e). In contrast to previously reported planar asymmetric metamaterials, three-dimensional metamaterials are mainly excited by the combined magnetic and electrical components of terahertz illumination. In this case, ultranarrow linewidths (FWHM of 5.90 GHz) and sharp resonances with a high Q-factor of 327 at 1.93 THz emerge. The combination of high Q-factor resonance and high sensitivity of asymmetric vertical SRR metamaterials will lead to further inspiration for superior performance sensor design.

The metamaterial absorber is a typical structure of metamaterials. It is usually composed of a metal-dielectric-metal three-layer structure. At the resonant frequency, the upper and lower layers of metal usually form a reverse current, which in turn forms a vertical interface current loop, resulting in magnetic resonance. When the incident electromagnetic wave energy is transmitted vertically downwards and enters the dielectric layer through the top metal slit, it will form a lateral transmission. Therefore, by optimizing the structure of the metamaterial absorber and tuning the electric resonance and magnetic resonance, extremely strong electromagnetic field localization can be realized, thereby achieving zero reflection and zero transmission of the incident electromagnetic wave at the resonant frequency, that is, complete absorption.

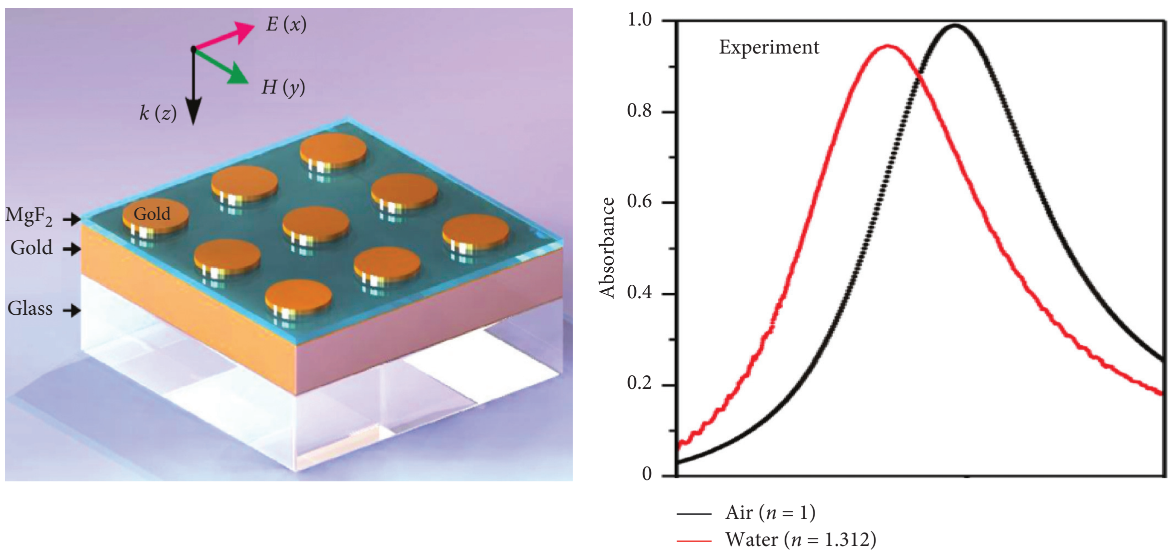
Metamaterial absorbers rely mainly on their strong resonance properties to gain significant advantages, as this type of sensor can produce a strong and measurable readout signal strong enough to accurately track the resonance absorption peak of a parameter shift in the reflection spectrum. The equivalent electromagnetic parameters of the metamaterial change with the substrate or geometry of the medium, resulting in a change in frequency. Metamaterial absorbers can be used to measure changes in the derived parameters.

Recently, there has been an increasing interest in metamaterial absorbers, providing an attractive platform for electromagnetic wave sensing applications [15, 16, 44–50]. Narrow-band or multiband metamaterial absorbers are critical in sensing and molecular detecting applications. In 2010, Na [44] first designed and implemented a metamaterial absorber working in the infrared band. The top layer of the metal structure is a periodic circular microdisk. The experiments measured the absorption spectra of air and water as the measured substance, as shown in Figure 8(a), the sensitivity reaches 400 nm/RIU and FOM reaches 87. Mirzaei et al. [48] modelled a metamaterial based label-free biosensor for DNA detection with a frequency shift of 3.6 GHz. As shown in Figure 8(b), a near-perfect metamaterial absorber for terahertz sensing applications has been proposed by Saadeldin et al. The structure has a near-perfect absorption of 99% at 2.249 THz and a narrow resonance peak with a Q-factor of 22.05. The structure has a sensitivity of 300 GHz/RIU, FOM of 2.94, and a refractive index range of 1.0 to 1.39 when applied as a sensor for sensing. In addition, the effect of the thickness of the analyte on the sensitivity was investigated over a range of 1.0 to 4.0 μm variations when the refractive index of the analyte $n = 1.35$, and the sensitivity $S = 23.7$ GHz/μm. Nanostructured plasma metamaterials are considered to be a good platform for narrow-band optical absorption and have a wide range of applications in sensing, filtering, modulation, and emission tailoring. However, achieving optical sensing and dynamic control of light in subnanometer absorption bandwidths remains a challenge. As shown in Figure 8(c), Feng et al. [49] demonstrated an asymmetric metal grating structure with perfect light absorption near 1.55 μm wavelength with a bandwidth of 0.28 nm using a propagating surface plasma mode with a low dissipation rate. By varying the structure parameters, the proposed structure provides a solution for chemical or biological sensing in the visible spectral range. The results show that the sensitivity and FOM of the sensor are 440 nm/RIU and 1333.33 RIU^{-1}, respectively.

3.2. Changing Substrate Properties. Recently, many research teams have attempted different approaches to achieve greater spectral modulation with the same medium variation, such as using low refractive index substrates, using ultrathin substrates, etching parts of high refractive index substrates, or integrating microfluidic channels or microcavities.

The dielectric layers of the metamaterial sensors mentioned previously are mostly made of GaAs and Si with high dielectric constants, which provide a large capacitance to the resonator and therefore reduce the capacitance variation caused by the target material. Recently, it has been found that the sensitivity can be significantly improved by using low refractive index materials instead of the commonly used high refractive index substrate materials and by using ultrathin substrates [51–54].

In 2010, Tao et al. [51] proposed a metamaterial biosensor based on SRRs structure and ultrathin silicon nitride (SiNx) substrate. In the experiment, 400 nm thick SiNx and 500 μm thick Si were selected as substrates, and silk fibro with different thicknesses was applied on the surface of the sensor element by spin coating. The measured transmission spectrum is shown in Figure 9(a). The test results show that when the thickness of the silk fibroin is 1500 nm, the resonance peak offset of the Si substrate sensor is 10 GHz, and the resonance peak offset of the SiNx substrate sensor is 116 GHz, indicating the sensor sensitivity of the SiNx substrate about 10 times that of Si substrate sensors. The main reason is that the permittivity of the SiNx substrate is about 7, the permittivity of the Si substrate is about 11, and the thickness of the Si substrate is much larger than the thickness of the SiNx substrate, resulting in the capacitance ratio of the Si substrate being much larger than of the SiNx substrate. Therefore, the sensitivity of the Si substrate sensor is smaller than that of the SiNx substrate sensor. Brian et al. [52] found that the refractive index sensitivity of nanopore arrays increases as the separation distance between the holes increases and redshifts the resonance. However, if a nanopore sensor is fabricated on a low refractive index substrate, additional sensitivity enhancement occurs, but the resonance is blue-shifted. They found that approximately 40% higher bulk refractive index sensitivity for a system of approximately 100 nm holes in 20 nm gold films fabricated on Teflon substrates ($n = 1.32$) compared to the case when conventional glass substrates ($n = 1.52$) were used. As shown in Figure 9(b), in order to get closer to practical applications, Hu et al. studied a paper-based terahertz metamaterial sensor [53], which can be used for quantitative analysis of different concentrations of glucose. In the experiment, 100 μL of glucose solution was deposited on the SRRs resonator, and as the glucose concentration increases, the offset of the resonant frequency increases, and the maximum offset can reach 300 GHz. For a glucose solution with a concentration of 7 mmol·L^{-1}, the sensitivity is 14.3 GHz·(mmol·L^{-1})$^{-1}$, and for a glucose solution with a concentration of 30 mmol·L^{-1}, the sensitivity is 10 GHz·(mmol·L^{-1})$^{-1}$. The flexible metamaterial based on the paper substrate has the characteristics of high sensitivity, transparency, portability, etc. and has more practical application value. In 2017, Srivastava et al. [54] experimentally demonstrated a dual-surface terahertz metamaterial sensor based on asymmetric SRRs with double splits on an ultrathin flexible polyimide substrate with a low refractive index, as shown in Figure 9(c), which can realize sensing on both sides of the structure and reveals a highly enhanced sensitivity. It has promising applications in industrial sensing systems and



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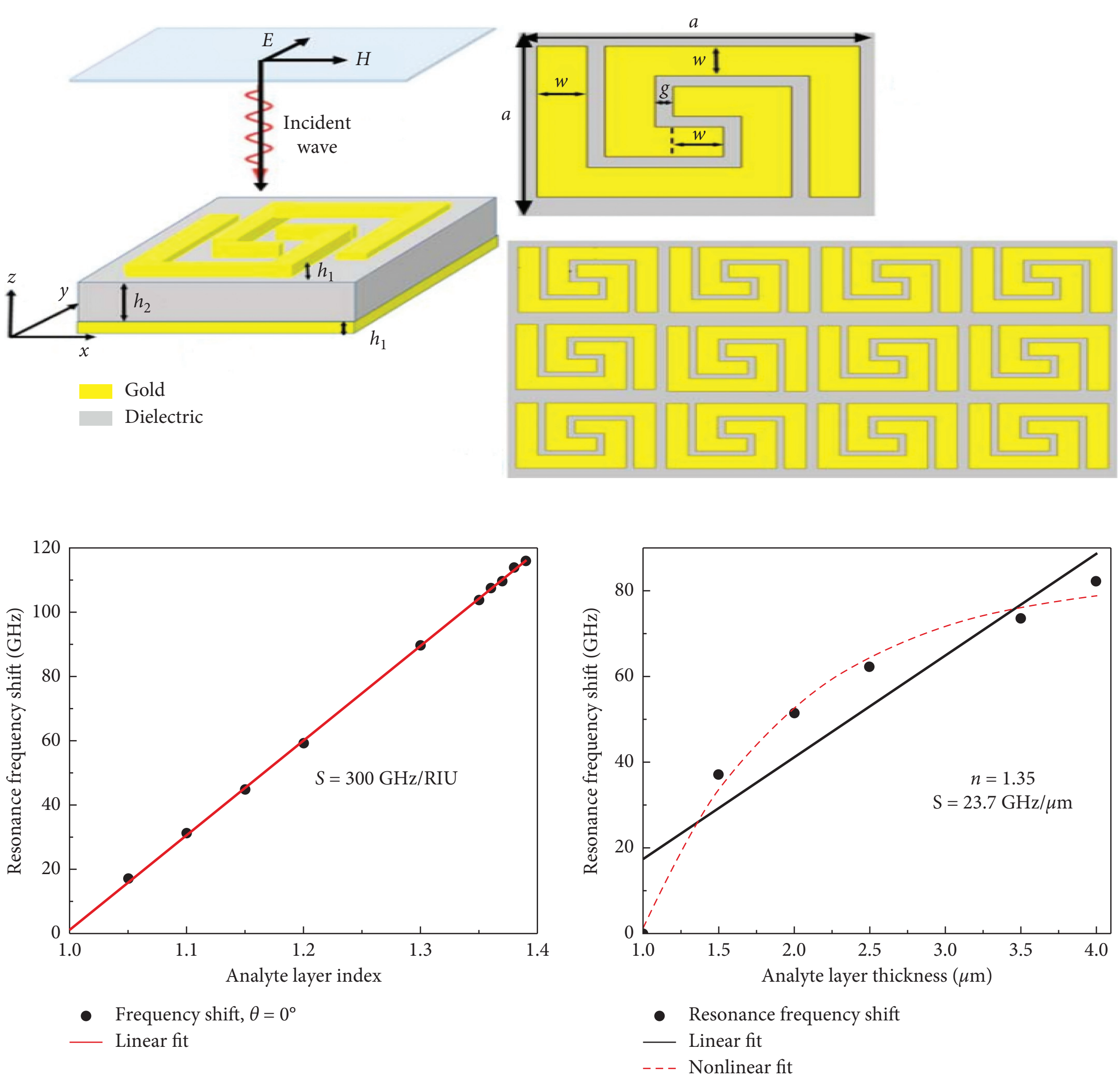

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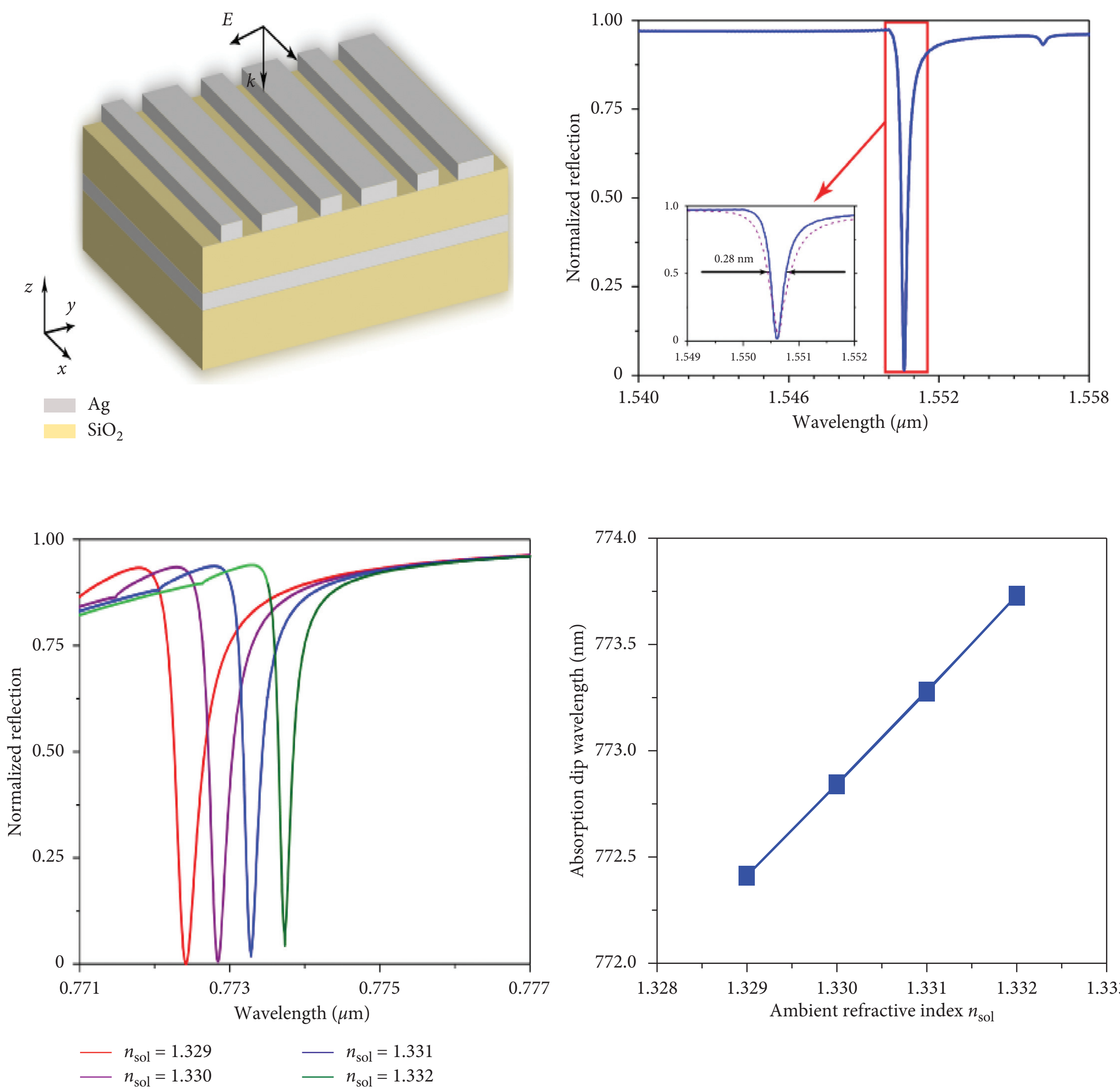


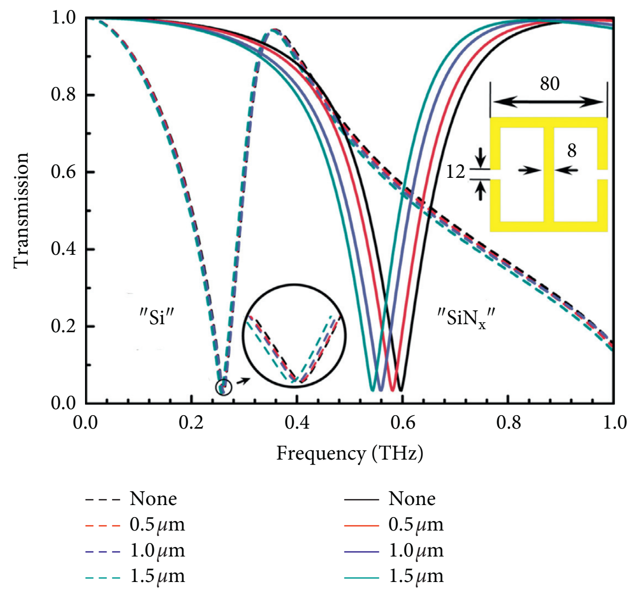
(c)

Figure 8: Schematic diagrams, reflection/absorption spectra, and sensitivity of metamaterial absorber sensors. (a) Reproduced with permission from reference [44] copyright 2010, American Chemical Society. (b) Reproduced with permission from reference [16] copyright 2019, IEEE. (c) Reproduced with permission from reference [49] copyright 2018, Optical Society of America.

improves biosensing performance at lower molecular concentrations.

However, high index semiconductor substrates are typically required to actively control the metamaterial response of the gated structure, and this is problematic with ultrathin substrates because of the integration required for the integrated fluidic channels, resulting in poor durability. In order to achieve greater spectral modulation for the same medium variation and to increase the sensing sensitivity of metamaterial sensors, the substrate effect is reduced by etching parts of the high refractive index substrate to highlight the effect of the surrounding medium variation [55–58].

In 2018, Moritake and Tanaka [57] proposed and demonstrated the use of selective and isotropic etching of substrates to eliminate the effect of substrate on plasma resonance. The concept of substrate etching is shown in Figure 10(a). After the formation of metal nanostructures on the substrate, only the substrate is selectively etched. This is shown in Figure 10(a). It is demonstrated that the use of substrate etching reduces the effective refractive index around the metal nanostructure, thereby eliminating the substrate-induced resonant red-shift and improving the refractive index sensitivity. In addition, the change in the resonance quality factor Q of the substrate etching method was investigated in detail. As shown in Figure 10(b), the



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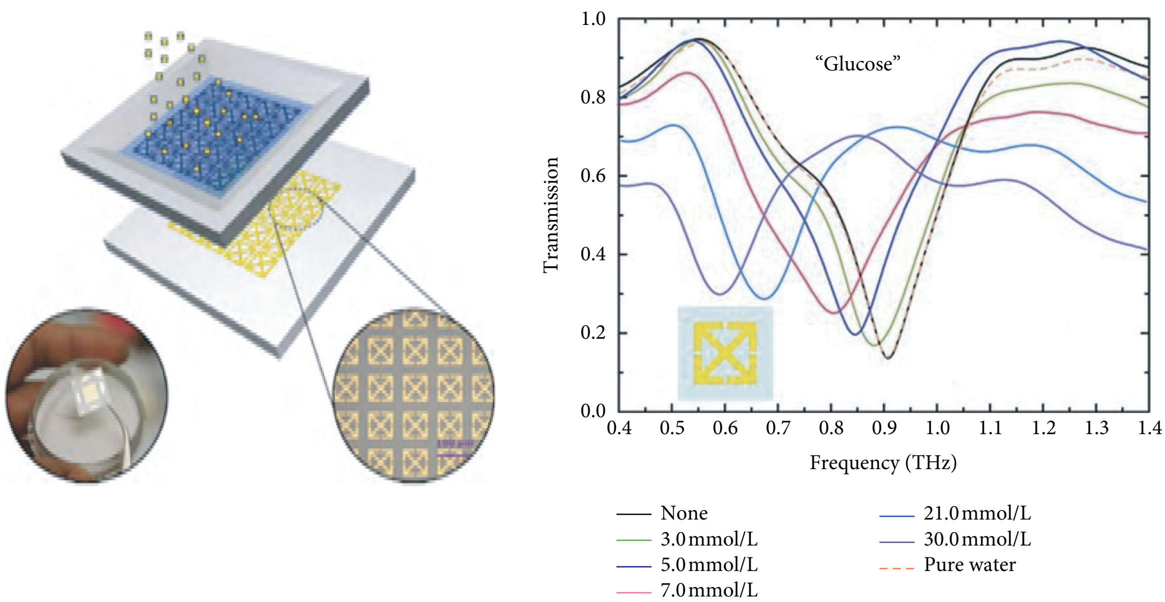

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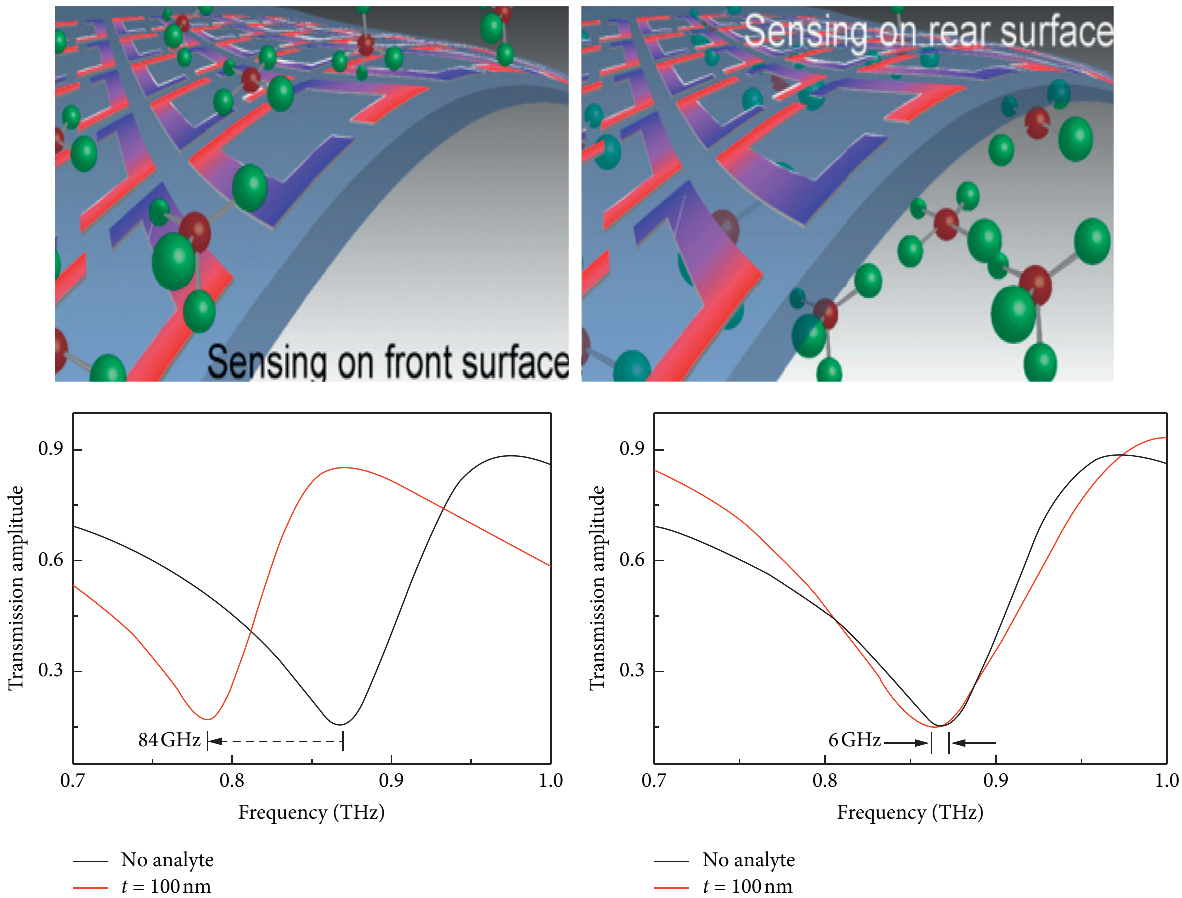
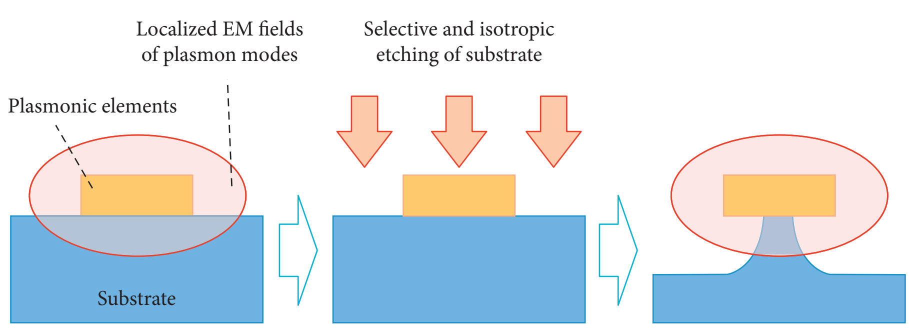


(c)

Figure 9: (a) Transmission spectra of metamaterial sensors based on ultrathin substrates, reproduced with permission from reference [51] copyright 2010, AIP publishing. (b) Schematic diagrams and transmission spectra of metamaterial sensors based on lower-index substrates, reproduced with permission from reference [53] copyright 2011, John Wiley and Sons. (c) Schematic diagrams and transmission spectra of metamaterial sensors based on an ultrathin flexible polyimide substrate, reproduced with permission from reference [54] copyright 2017, AIP publishing.



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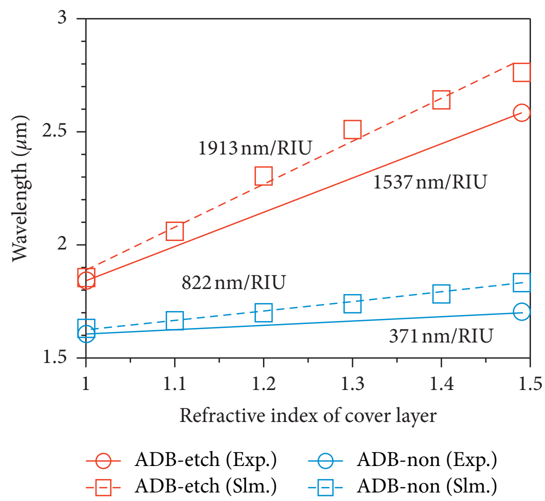


(b)

Figure 10: (a) Schematic of substrate etching for plasmonic elements formed on a substrate. (b) The wavelength shift as a function of the refractive index of the coating layer for the asymmetric double bar (ADB)-etch and ADB-non. (a, b) Reproduced with permission from reference [57] copyright 2018, Optical Society of America.

refractive index sensitivity is significantly improved to 1537 nm/RIU due to the substrate etching making the electric field accessible under the nanostructure. The refractive index sensitivity is substantially improved compared to the case without substrate etching. The method proposed in this paper is applicable to a variety of plasmonic structures to eliminate the influence of substrates on the realization of high-performance plasmonic devices.

In 2019, Meng et al. [58] proposed a metamaterial sensor with an etched trench introduced in the inductance-capacitance gap region of a split-ring resonator, and the results showed an increase in frequency shift and sensitivity when a dielectric material of up to 18 μm thickness was deposited on the sensor surface. As shown in Figure 11, Wang et al. [59] theoretically optimized the conventional fabrication process for realizing terahertz metamaterial absorbers and analyzed the feasibility of matching the existing surface micromachining process. The results showed that the sensing performance of the new metamaterial sensor was significantly enhanced due to the surface-relief design compared to the conventional metamaterial sensor. The proposed method for the design of terahertz metamaterial absorbers can be used to implement ultrasensitive metamaterial sensors.

To design a highly sensitive refractive index sensor, the degree of overlap between the electromagnetic field and the substance needs to be enhanced. Related studies have shown that when molecules are not attached to the sensor surface, the electric field strength decreases as the distance between the substance and the surface increases, which results in a weakening of sensitivity. The research team, therefore, began to investigate how to design geometrical structures to enhance the overlap between the substance and the electromagnetic field.

The emergence of microfluidics, which has received widespread attention from research teams, allows the manipulation of very small amounts of fluid (10^{-9} to 10^{-18} L) using microchannels (approximately 10–100 μm in size) [60]. It is potentially valuable in the field of biological and chemical detection because of its high sensitivity, low cost, and fast measurement. By designing different forms of microfluidic channels, the refractive index sensitivity of the sensors has been greatly enhanced [61–64].

Zhou et al. [63] demonstrated a metamaterial integrated microfluidic sensor with the capability of multiband sensing for the dielectric property of various chemicals. As shown in Figure 12(a), the sensor is composed of a symmetrical double SRRs structure, which enhances the resonant strength and sensing capability of the resonant cavity compared to a single SRRs. As shown in Figure 12(b), a dielectric model is developed by fitting a nonlinear curve to the resonance and theoretical derivation and experimentally validated to obtain a complex dielectric constant from the measured "butterfly-like" result curve, which can be used as a diagnostic indicator to identify chemicals. In addition, the sensor has been extended to integrate multiple resonators to obtain the dielectric spectrum of ethanol, enhancing its potential for chemical analysis and practical applications.

Withayachumnankul et al. [62] implemented a metamaterials microfluidic sensor based on a single SRR resonator, as shown in Figure 13. At resonance, an SRR creates a strong electric field in the deep subwavelength region. The flow of the liquid over this region can alter the local field distribution and thus affect the SRR resonance behavior. Specifically, the resonant frequency and bandwidth are influenced by the complex dielectric constant of the liquid sample. By establishing an empirical relationship between the sensor resonance and the sample dielectric constant, the complex dielectric constant of the liquid sample can be estimated. This work advances the use of SRR-based microfluidic sensors for the identification, classification and characterisation of chemical and biochemical analytes.

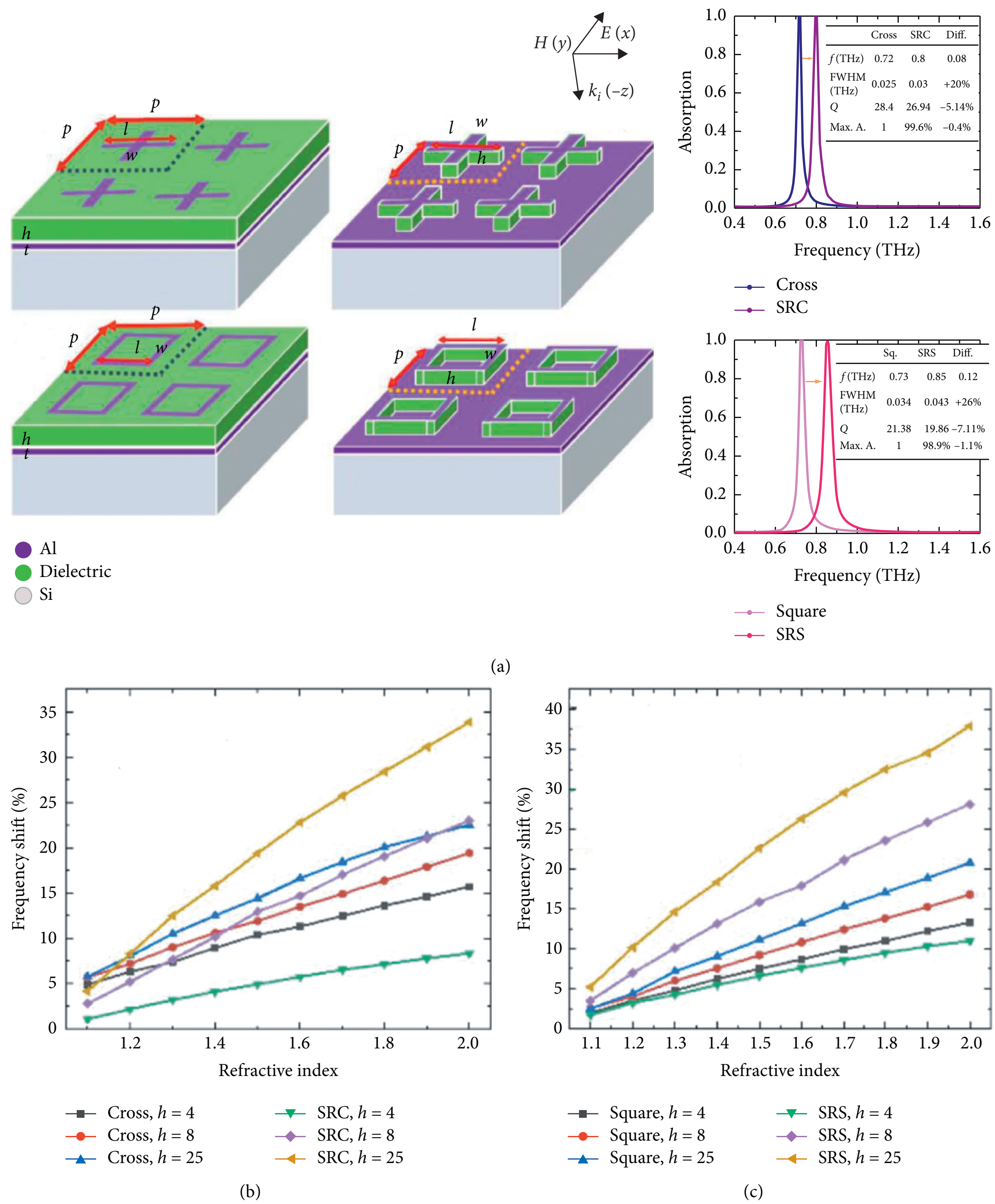


Figure 11: Comparison of the two most common terahertz metamaterial absorbers and numerical simulations in terms of absorption and sensing capabilities, reproduced with permission from reference [59] copyright 2020, Optical Society of America.

Hu et al. [64] proposed and experimentally demonstrated a novel metamaterial absorber integrated terahertz microfluidic sensor. As shown in Figure 14(a), by introducing matter into two parallel metallic structures, a transverse cavity resonance occurs inside the absorber, resulting in a significant increase in sensitivity. Furthermore, the emergence of this highly sensitive sensor provides an interesting approach to the design of subsequent highly sensitive chemical and biological sensors.

3.3. Other Ways. Furthermore, to achieve very strong magnetic resonance in metamaterials for high-performance sensing, Chen et al. [65] studied numerically photonic microcavity-enhanced magnetic plasmon resonance in metamaterials for high quality refractive index sensing, as shown in Figure 15(a). The radiation damping of the magnetic plasmon resonance is greatly reduced and the linewidth is greatly reduced due to the coupling of the magnetic plasmon resonance excited in the metallic SRRs to

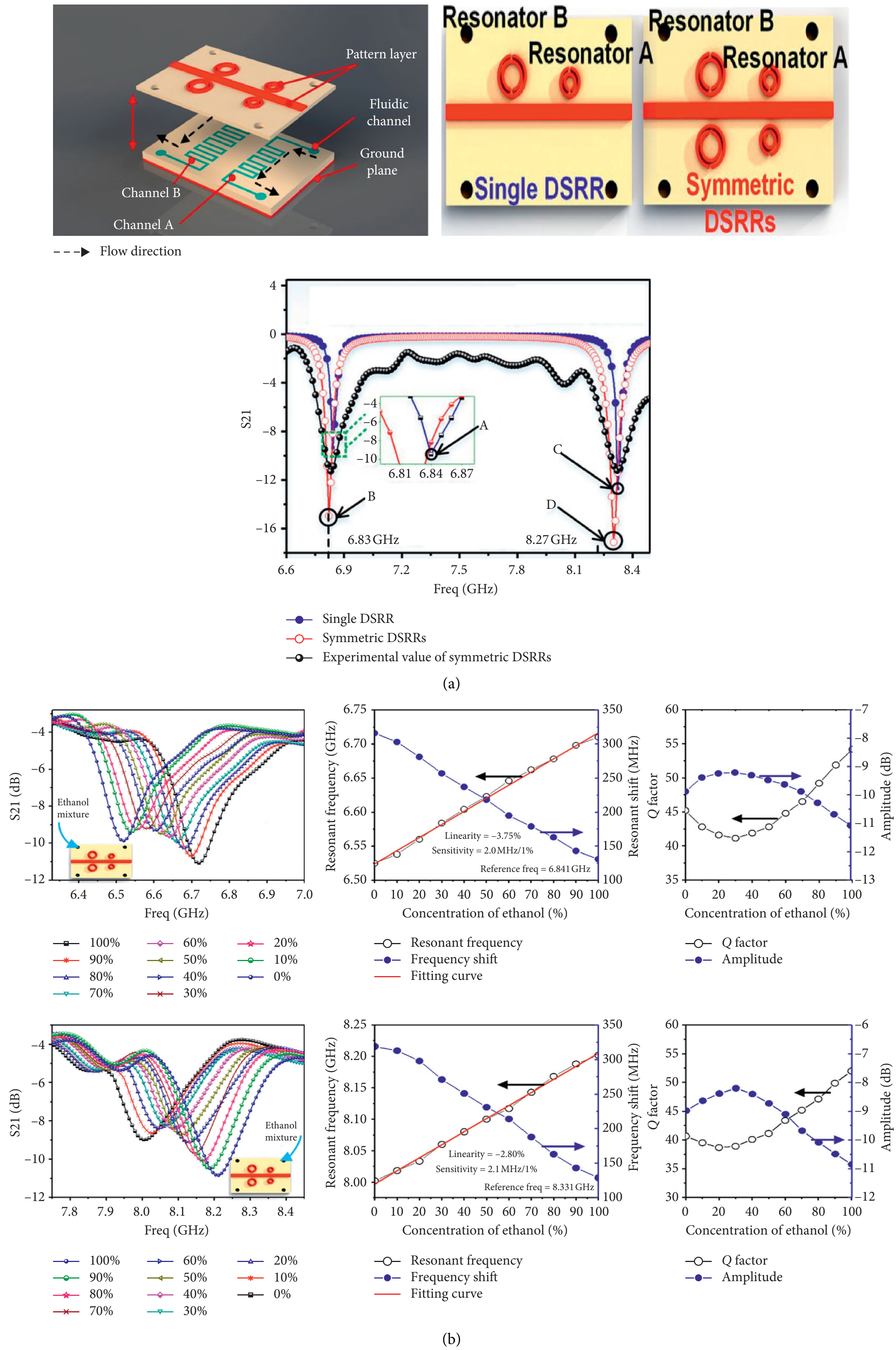


Figure 12: (a) Schematic diagrams and simulated transmission spectra of integrated microfluidic metamaterial sensors. (b) Measured results from the metamaterial sensor for ethanol with different concentrations in channels A and B. (a, b) Adapted with permission from reference [63] copyright 2018, Springer Nature.

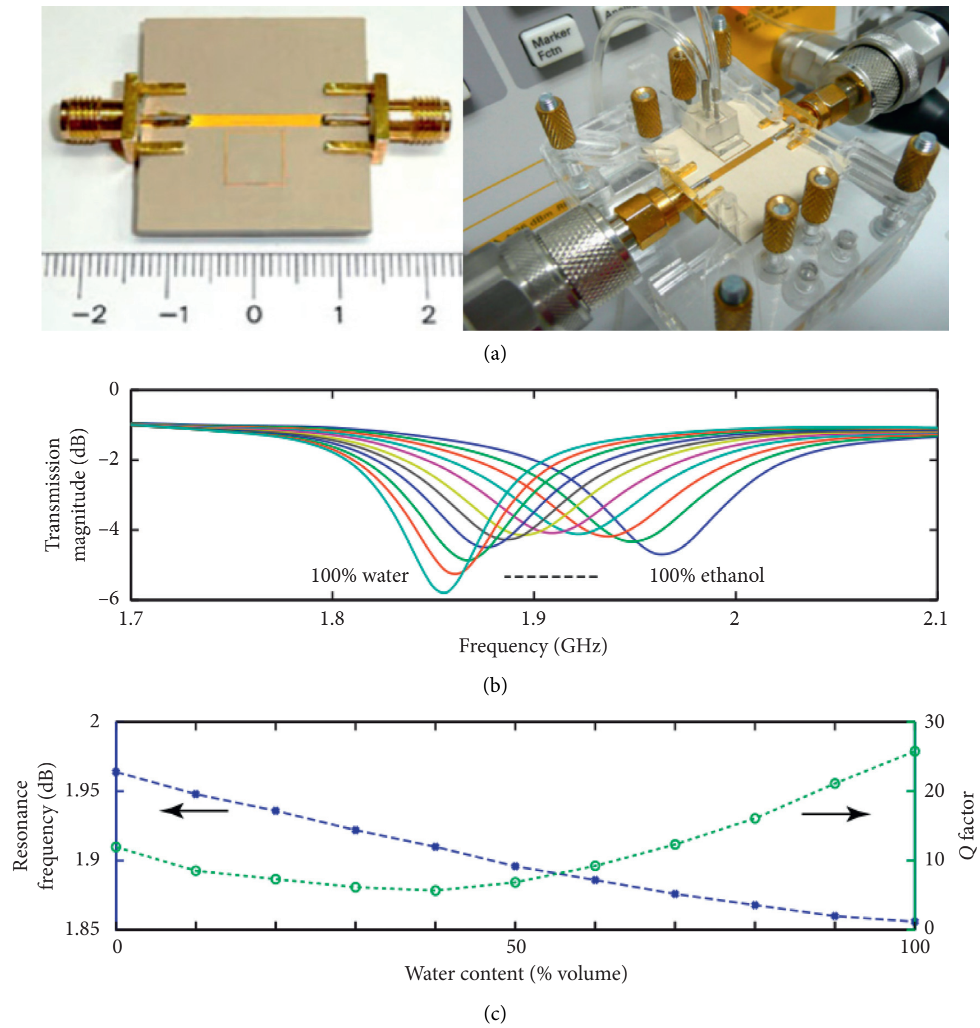
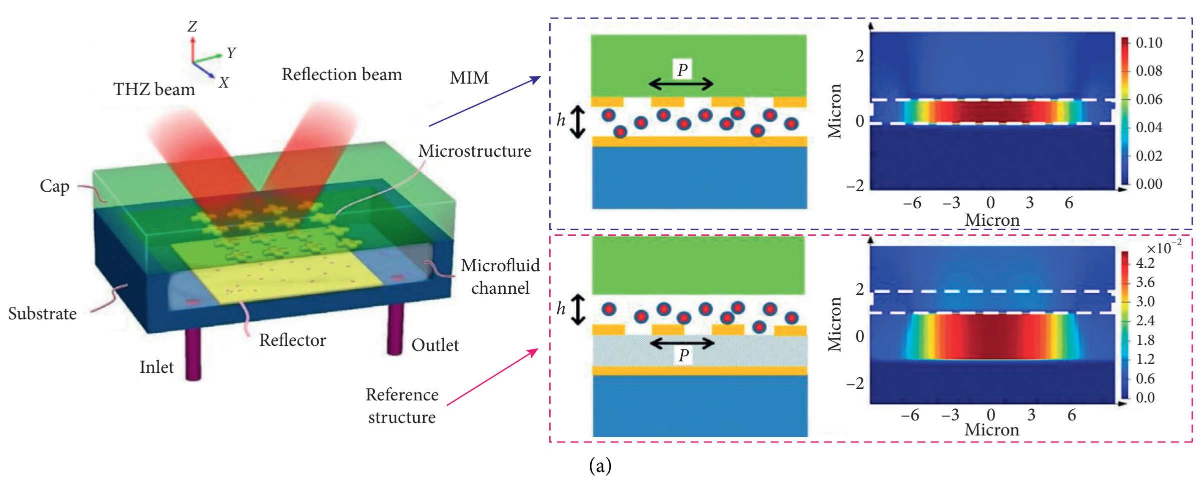


Figure 13: Assembly of microfluidic sensor modules based on microstrip-coupled SRR metamaterials and measured transmission response of water-ethanol mixtures at different volume fractions, adapted with permission from reference [62] copyright 2013, Elsevier B.V.



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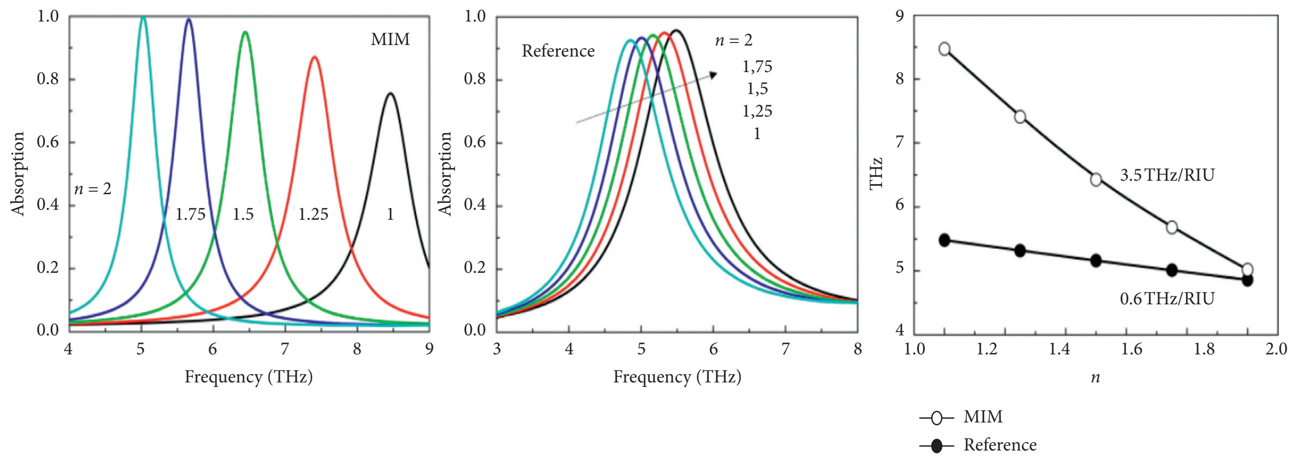
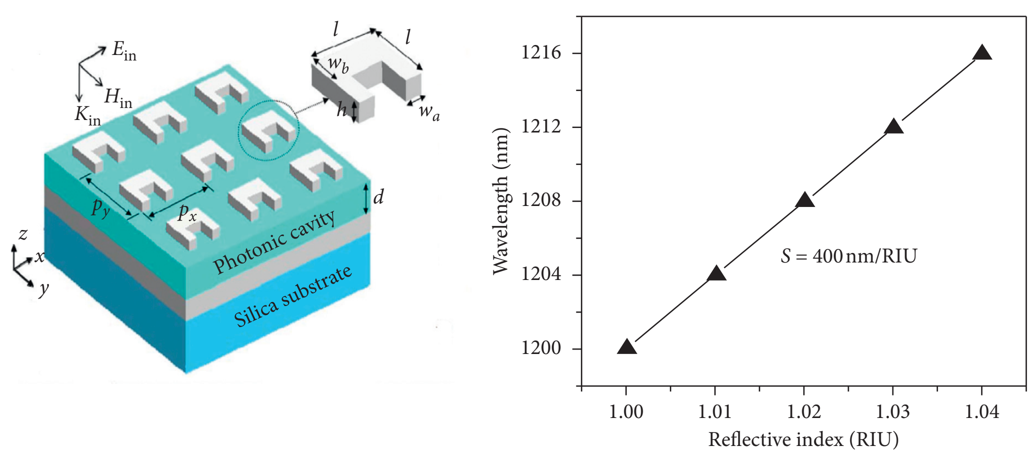


(b)

Figure 14: (a) Schematic diagram of the metamaterial absorber sensors with microfluidic channel located inside and on top of the metamaterial absorber. (b) Absorption spectra and resonance peak corresponding to different channel refractive index for MIM and reference. (a, b) Reproduced with permission from reference [64] copyright 2016, John Wiley and Sons.



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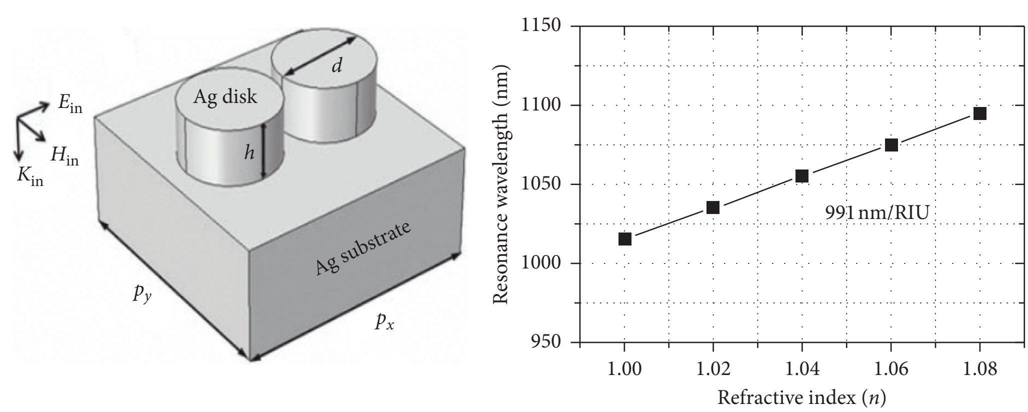


(b)

Figure 15: Schematic diagrams and refractive index wavelength sensitivity of metamaterial sensors based on strong magnetic resonance. (a) Reproduced with permission from reference [65] copyright 2019, IEEE. (b) Reproduced with permission from reference [14] copyright 2019, IOP publishing.

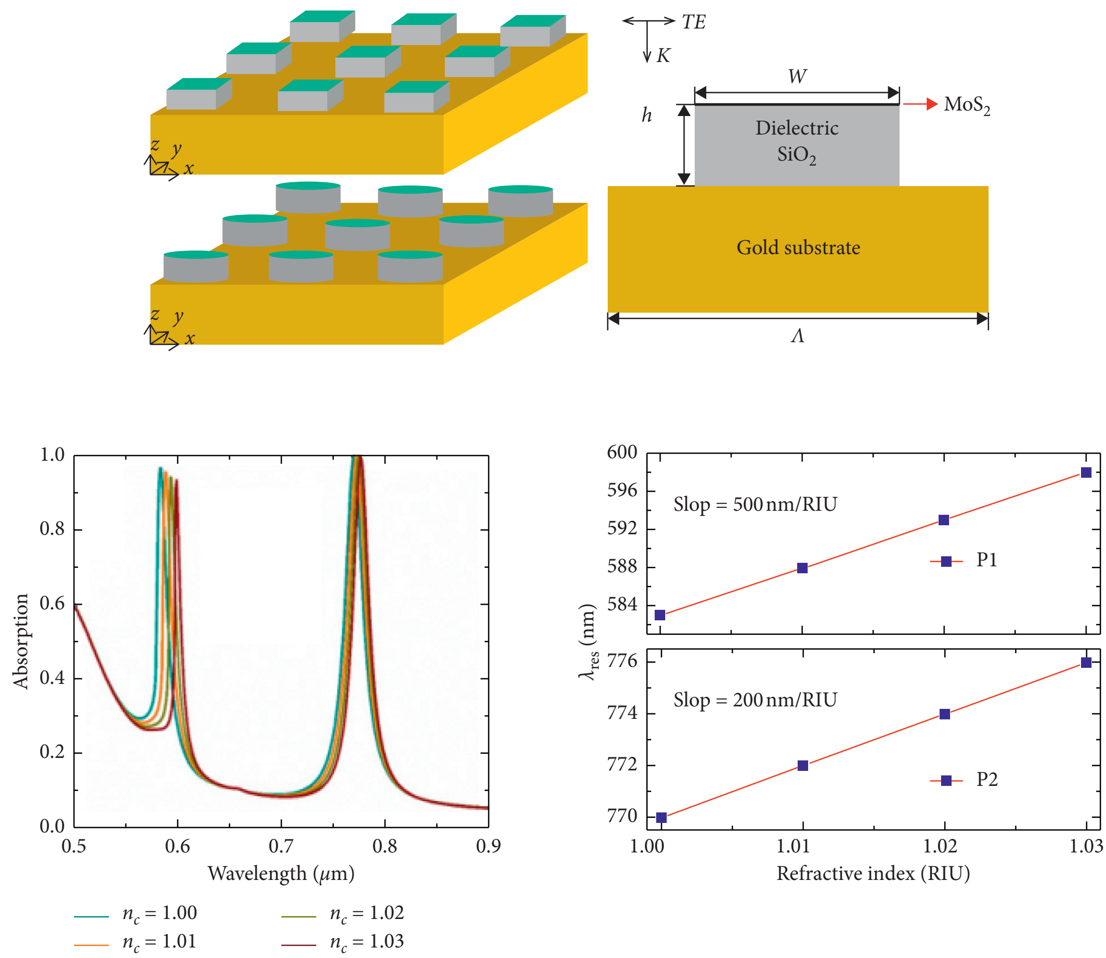


FIGURE 16: Schematic diagrams and refractive index wavelength sensitivity of metamaterial sensor based on cylinder MoS_2, reproduced with permission from reference [66] copyright 2019, Elsevier B.V.

the photonic microcavity mode supported by the photonic microcavity. The cavity-coupled metamaterial sensor has the advantages of narrow bandwidth, wide modulation range and large magnetic field enhancement at magnetic plasma resonance with high sensitivity ($S = 400$ nm/RIU and $S^* = 26$ RIU) and figure of merit (FOM = 33 and FOM* = 4215). It is shown that the proposed metamaterials have potential applications in plasma biosensors. In 2019, a metamaterial structure consisting of a pair of closely spaced metallic nanodiscs arranged on a metallic substrate was proposed by Chen et al. [14]. A very strong magnetic resonance exists in this structure, and theoretical analysis suggests that the magnetic resonance is caused by incoming plasma hybridisation within the pair of metallic nanodiscs. Under strong magnetic resonance, the electric field tends to increase, and ultranarrow bandwidths and near-perfect absorption can be achieved. It is shown that the designed sensor has a high sensitivity ($S = 991$ nm/RIU, $S^* = 47$ RIU) and FOM (FOM = 124, FOM* = 17702).

Qiu et al. [66] proposed and investigated a dual-frequency perfect metamaterial absorber consisting of a simple periodic patterned cylindrical/square MoS_2-dielectric silica arrays supported by a metal ground plane, as shown in Figure 16. The application of this metamaterial absorber in refractive index sensors is also presented in the paper. Compared to previous dual-band metamaterial absorbers, the cylinder MoS_2-based absorber is simple in shape, greatly simplifies the fabrication process and is polarization insensitive. It has great potential in the visible and near-infrared spectral range, for instance, as a plasma sensor.

As shown in Figure 17, Chen and Fan [67] demonstrated a novel terahertz metamaterial sensor with integrated microfluidic channels, where two pairs of high refractive index dielectric disks were arranged into the unit cell. The ultrahigh-strong toroidal dipole response associated with the presence of trapped modes was excited and studied by introducing a new symmetry-breaking method into the cell structure. Simulation results show that the sensor has a high Q-factor and FOM of 3189 and 515, respectively, and thus with this advantage, it will be used in a wide range of applications in liquid and gas sensing.

He et al. [68] proposed an ultrasensitive terahertz sensor based on a graphene metamaterial with a complementary structure of wire slot and split-ring resonator slot arrays, as shown in Figure 18. The sensitivity of the sensor reached 177.7 GHz/RIU with FOM of 59.3. In addition, this structure

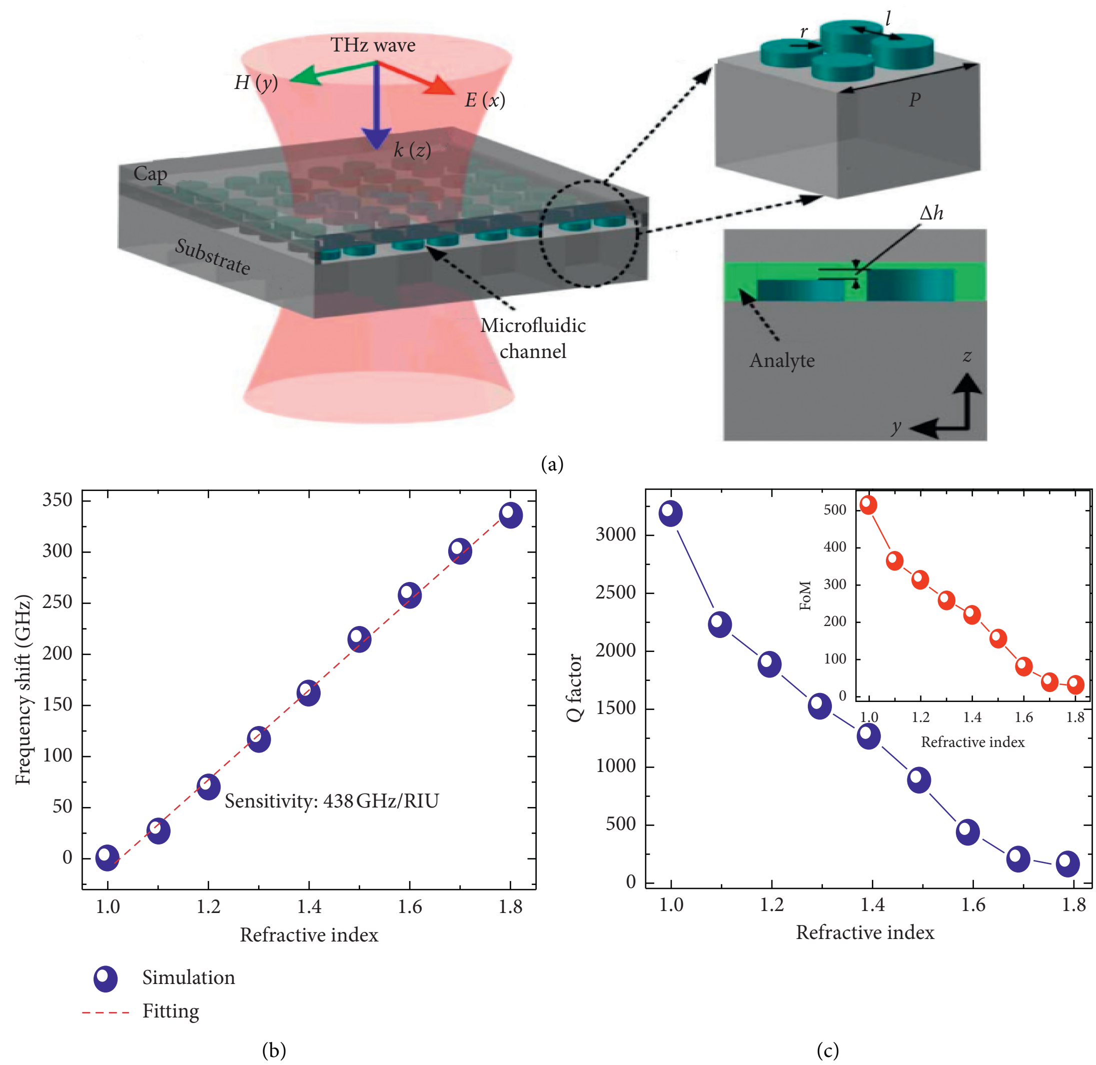
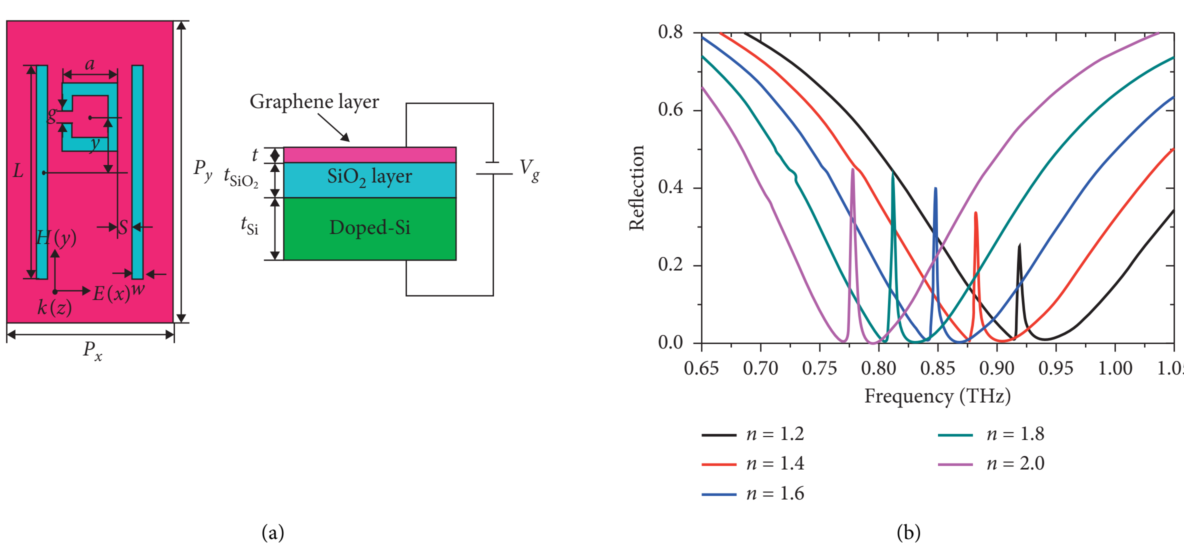


Figure 17: Schematic diagrams, refractive index frequency sensitivity, and Q-factor of all-dielectric metamaterial sensor, reproduced with permission from reference [67] copyright 2019, Optical Society of America.



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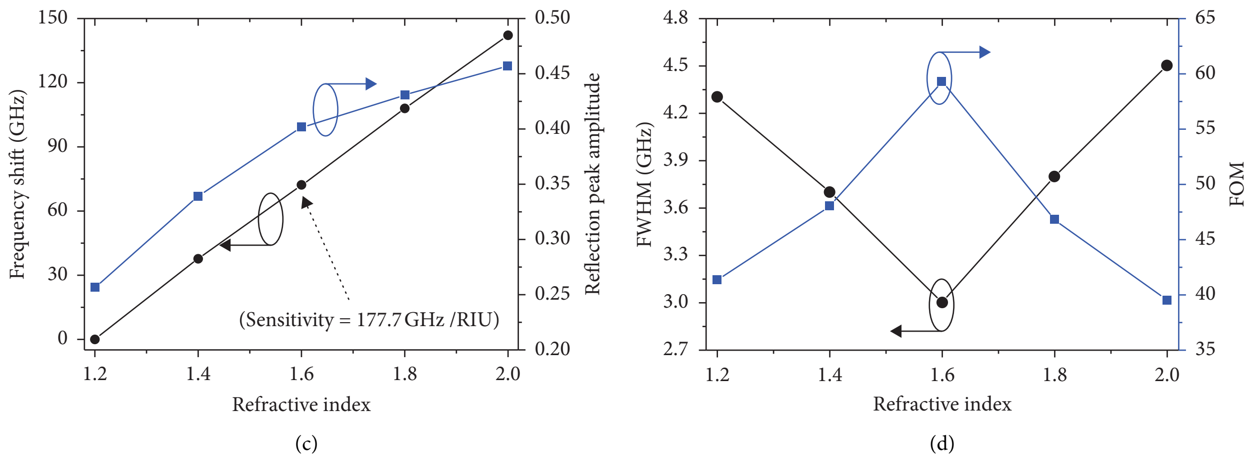

Figure 18: Schematic diagram, reflection spectra, refractive index frequency sensitivity, and FOM of sensor based on complementary graphene metamaterials, reproduced with permission from reference [68] copyright 2016, the Royal Society of Chemistry (RSC).

has the advantage of enhancing the absorption and sensing performance of biomolecules, as well as dynamically adjusting the sensing range by adjusting the Fermi energy of the graphene.

4. Challenges and Prospects

Metamaterials can manipulate electromagnetic waves in specific ways and exhibit supernormal electromagnetic properties that natural materials do not possess. In addition, metamaterials can realize strong localization and enhancement of electromagnetic field, thereby providing a novel sensing platform with the advantages of high sensitivity, high resolution, fast response, and simple measurement.

Metamaterial sensors can break through the resolution limit of traditional sensors for a small amount of substance and have higher sensing sensitivity and resolution than traditional sensors. During the past decade, researchers have been focusing on further improving the sensing characteristics of metamaterial sensors and promoting their development. Currently, metamaterial sensing technology is relatively mature. However, in order to truly move towards practical applications and explore the broad application prospects of metamaterial sensors in the fields of substance detection, environment sensing, biosensing, food safety control, and homeland security, like any emerging field, metamaterial sensing also faces many challenges:

(1) Metamaterial sensors still need improvements in sensitivity and resolution by designing special metamaterial structures or using specific dielectric materials.
(2) With the advancement of microfabrication and nanofabrication technology, the precise preparation of different metamaterial sensors is realized, creating new possibilities for its practical application.
(3) In addition to the realization of metamaterial sensors based on resonant frequency shift, the combination of the amplitude and phase changes of electromagnetic waves with the characteristics of metamaterials can expand the types of metamaterial sensors.
(4) Continue to develop the fields of metamaterial sensing, such as pressure, temperature, density, thickness, strain, and position.
(5) To address the detection limitation of only one substance at a time, multichannel metamaterial sensors should be developed.

5. Conclusion

In this review paper, we introduced metamaterial sensors from several aspects such as sensing mechanism, main characteristic parameters, and sensing characteristics improvement. It focuses on the development overview of improving the sensing sensitivity of the metamaterial sensors by optimizing the structure and changing substrate properties.

The superiority and versatility of metamaterial sensors are obvious. They usually exhibit enhanced characteristics and capabilities potential to overcome many of the limitations of conventional sensing devices. However, based on actual needs, the sensitivity, accuracy, and detection limit of metamaterial sensors need to be further improved. With the progress of microstructure processing technology and the emergence of new materials, it is believed that more and more high-performance metamaterial sensors will appear and have broader application prospects in the future.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

This work was supported and funded by the National Natural Science Foundation of China (51965047), Natural Science Foundation of Inner Mongolia (2018MS06007), and

Research Startup Fund of High-Level Talents Introduction in 2018 (21700-5185131 and 21700-5185128).

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