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Evaluation of amplitude difference referencing technique with terahertz metasurfaces for sub-micron analytes sensing

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Terahertz metasurfaces with subradiant modes that allow excitation of a high-quality factor are potential candidates for label-free sensing of thin-film biomolecules. Nevertheless, sensing sub-micron analytes are rather difficult due to the normalization method that has been used so far. Here, we evaluate an amplitude difference referencing technique that exploits the response of the uncoated metasurface structure as a reference. Amplitude difference percentage has been calculated by subtracting the frequency response of coated metasurface from the frequency response of the uncoated metasurface for different analyte thicknesses. To our surprise, we observe significantly large values of the amplitude difference that can be easily measured using conventional terahertz time-domain spectrometers. In future, this technique can be utilized to detect analytes with sub-micron thickness and could be exploited across the electromagnetic spectrum.

1. Introduction

The past two decades have revealed a plethora of potential terahertz (THz) applications emerging from several fields of science (Jepsen et al., 2011; Tonouchi, 2007). Applying terahertz technology in the biomedical context has invoked considerable interest (Al-Naib, 2017; Baras et al., 2003; Markelz, 2008), as rich spectroscopic features, such as collective structural vibrational modes of proteins and DNA are found to exist at these frequencies. Label-free identification of various biomedical analytes with terahertz waves has been proposed (Nagel et al., 2003, 2002; O'Hara et al., 2012). This technique would prove beneficial over present testing methods, in which the unidentified analyte is labeled with fluorescent molecules. Apart from requiring an additional preparation step (Fischer et al., 2002), which restricts the speed- and cost-efficiency, the analyte conformation might also be altered in the labeling process, lowering the yield and reliability of the method (Mickan et al., 2002). Realizing the potential of marker-free tests at terahertz frequencies is a challenging task due to the large discrepancy of the sensing wavelength and the tiny quantity of analyte, which is usually in a nanometric scale. In order to access the desired information, thin-film sensors with a very high sensitivity have to be developed. Luckily, several approaches have been introduced in recent years (Al-Naib and Withayachumnankul, 2017; Gupta et al., 2017; O'Hara et al., 2008, 2012), but the various techniques yet suffer from different limitations (Withayachumnankul et al., 2014).

In order for thin-film sensors to function efficiently, the frequency response has to feature a sharp transition to allow the recognition of small changes in the frequency response due to the modification in the dielectric environment. Conventionally, the steepness of this transition has been considered as a direct measure of the sensor sensitivity (Singh et al., 2014). Planar metamaterials or metasurfaces can be considered as filters of electromagnetic waves, consist of two-dimensional arrays of identical metallic resonators (Al-Naib et al., 2013; Fedotov et al., 2007; Singh et al., 2014). A drawback of traditional designs is the strong free-space coupling of the structural elements, which induces high radiation losses, resulting in rather low-quality (Q-) factors of the devices. Different designs have been investigated in order to suppress the radiation losses (Al-Naib et al., 2014; Singh et al., 2011b), and the most famous approach utilizes the asymmetric

Abbreviations: THz, terahertz; Q-factor, quality factor; CST, Computer Simulation Technology; RIU, refractive index unit; ADRT, amplitude difference referencing technique; ASR, asymmetric split-rectangular resonator.

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subradiant Fano resonance that can be excited by breaking the symmetry of a double-split ring resonators (Fedotov et al., 2007).

After applying the analyte onto the sensor's surface, the resonance is red-shifted due to the dielectric environment alteration. This shift of the resonance frequency is considered as a measure of the refractive index, i.e. the analyte type and its thickness. After measuring the frequency response, the results are normalized to the frequency response of a bare substrate. Measurable results were achieved when the analyte thin-film refractive index was 1.6 with a thickness of one micron or larger. For instance, a frequency shift of 10 GHz at a resonance frequency of 0.52 THz was achieved when the analyte thickness was one micron (Singh et al., 2014) and the shift increased to 29 GHz at analyte thickness of 16 μm where it is eventually saturated. In order to enable sensing of analytes with a thickness less than one micron, there were pioneering attempts to address this problem by using membranes (Chen et al., 2012; Tao et al., 2010) or very low dielectric constant substrate in reflection mode (Reinhard et al., 2012). However, these extra preparation conditions will hinder from achieving high throughput thin-film sensors. Moreover, it is highly desirable to have a fast process for the whole sensing procedure. Hence, we need to minimize the measurements scan time as much as possible. On the contrary, quite long scan times are being carried out using THz spectrometers and considered essential to achieving the required resolution in the frequency domain in order to discern close points. For instance, a quite long 200 ps scan time is required to achieve 5 GHz resolution in the frequency domain. Therefore, it will be highly desirable to utilize the standard photolithography along with a transmission mode spectroscopy and be able to discern the shift in the resonance when the analyte thickness is less than one micron using a reasonable scan time.

In this paper, we evaluate an amplitude difference referencing technique (ADRT) for thin-film sensors via considering the frequency response of an uncoated metamaterial sensor as a reference. We utilize sharp Fano resonances excited by means of metasurfaces consist of asymmetric split-rectangular resonators (ASRs) (Fedotov et al., 2007; Singh et al., 2011a). More specifically, we study the sensitivity level using the conventional method of normalization by dividing the transmission amplitude response after coating the metasurface with the analyte to its bare substrate counterpart response. More importantly, we examine the ADRT by subtracting the frequency response of the coated metasurface from the frequency response of the uncoated metasurface structure. As the refractive index of different biomolecule analytes ranges from 1.4 to 1.6 in DNA and 1.6 to 2.0 in RNA (Yahiaoui et al., 2016), an average value of the analyte refractive index of 1.6 was chosen.

2. Sensor design

The metasurface unit cell consists of double-split rectangular asymmetric metal resonators deposited on top of a dielectric substrate as shown in Fig. 1(a). The unidentified analyte is applied to the metalized side and cover the whole area. Fig. 1(b) shows the geometrical dimensions of the ASR unit cell. The relevant geometrical dimensions of the resonators are: a side length of $l = 80$ μm, a microstrip line width of $w = 10$ μm, a gap of $g = 5$ μm, an asymmetry distance from the center line of $d = 5$ μm, and a periodicity of $p = 100$ μm. The refractive index of the high resistivity silicon substrate wafer is 3.42. The thickness of the gold metalized metamaterial resonators is 200 nm on top of the silicon substrate. The simulations were conducted using the frequency domain solver of the simulation software Computer Simulation Technology (CST) Microwave Studio, which is based on the finite integration technique. Periodic boundary conditions have been utilized to mimic the actual configuration and normal incidence plane wave excitation has been applied. Since the electric field is oriented perpendicular to the two gaps, as indicated in the inset of Fig. 1, we expect an asymmetric sharp Fano resonance as well as symmetric dipole resonance to be excited.

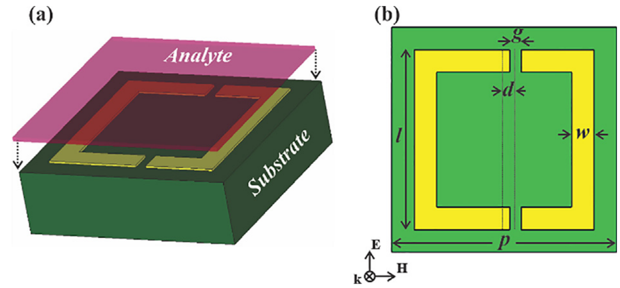

Fig. 1. Schematic of the metasurface unit cell: (a) 3D view of the asymmetric split-rectangular (ASR) metamaterial unit cell with the analyte layer applied on top of the metasurface sensor, (b) ASR unit cell with the detailed geometric dimensions. The inset shows the polarization of the electric field illumination, which is perpendicular to the gaps of the ASR.

3. Results and discussion

Conventionally, three sets of data are measured in time-domain for: (i) a bare substrate, (ii) uncoated metasurface structure, and for a (iii) coated metasurface after depositing the analyte on top of the metasurface structure. Please note that the metasurface structure in (ii) and (iii) is patterned on top of a substrate similar to the one in (i). After converting the measured data to the frequency domain, the latter two responses are normalized to the response of the bare substrate. Fig. 2(a) shows the normalized uncoated metasurface frequency response (black solid line). As expected, we observe the excitation of the Fano asymmetric resonance at 0.396 THz and the dipole resonance at 0.7 THz (not shown here). The physical mechanism of the Fano resonance line-shape is fully explained in the literature (Fedotov et al., 2007; Singh et al., 2011a). It is worth mentioning that the spectral response of the Fano resonance features a small bandwidth of 34 GHz and therefore the quality factor (defined as resonance frequency/bandwidth) of this resonance is almost 12 for the given configuration. After the analyte thickness of 0.25 μm is applied on top of the metasurface, the frequency response (red-dotted line) normalized to the bare substrate is red-shifted by 1 GHz to 0.395 THz as shown in Fig. 2(a). This red-shift in the frequency response is a consequence of the alteration in the dielectric environment of the resonators. The shift/refractive index unit is 1 GHz/(1.6–1.0) = 1.67 GHz/RIU, which represents a quite low sensitivity. It is important to note

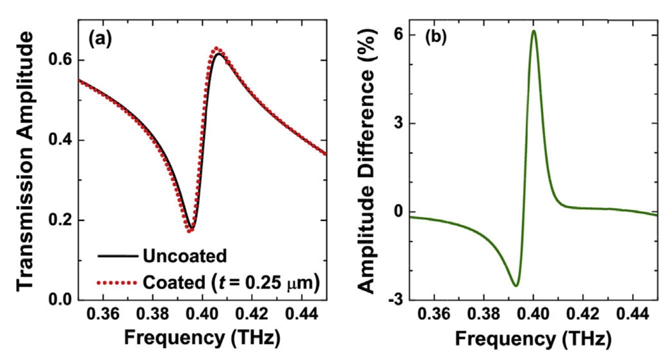

Fig. 2. (a) Transmission amplitude spectra of the uncoated and coated metasurface with 0.25 μm thick analyte using the conventional normalization method; (b) the amplitude difference percentage using the ADRT with the uncoated metasurface response as a reference.

that this value is a very small red-shift that requires a minimum of 1000 ps scan time measurements in order to discern the coated sample response from the uncoated one.

Next, we investigate the performance of the amplitude difference referencing technique in this paper. To do this, we subtract the frequency response of uncoated metasurface from the frequency response of the coated metasurface. Surprisingly, we get a very clear amplitude difference signature as shown in Fig. 2(b). The peak-to-peak difference is 8.6%, which is a significant value that can be easily measured and differentiated from the noise floor using either conventional terahertz time-domain spectrometers or even the state-of-the-art systems with a dynamic range of 90 dB (Vieweg et al., 2014). This result reflects the potential of using such a referencing technique especially in the cases of analyte thickness is in the range of sub-micron only.

In order to further evaluate the ADRT, we compare the transmission amplitude spectra for analyte thickness of 0.25, 0.5, 1, and 2 μm using the conventional normalization method and the ADRT as shown in Fig. 3. The vertical dashed line shows the location of the Fano resonance dip for the uncoated metasurface, i.e. without the analyte. Fig. 3(a)–(d) show the normalized transmission amplitude where the small red-shift increases with the increase in the analyte thickness. The corresponding red-shift in the frequency response is 1, 2, 6, and 9 GHz, respectively. For a larger analyte thickness of 5 10 μm, and 15 μm, the red-shift increased to 14.5, 18.6, and 20.1 GHz, respectively. Hence, the conventional normalization method could be useful for analyte thickness of 2 μm or more. On the contrary, the ADRT shows a large amplitude difference for the same range of the analyte thickness as shown in Fig. 3(e)–(h) ranging from 8.6% to 55.8% for analyte thickness of 0.25 μm–2 μm, respectively.

The large amplitude difference can be interpreted to the significant steepness of the flank of the amplitude response. For instance, the steepness (the frequency derivative) of the amplitude response of the uncoated sample is 6.3%/GHz, which results in an amplitude difference of 37% for an analyte thickness of 1 μm. Hence, decreasing the steepness of the amplitude response should lead to a decrease in the amplitude difference. This hypothesis is confirmed by increasing the asymmetry distance from the center line, *d*, of the ASR resonators to be 20 μm instead of 5 μm. In this case. We find that the steepness of the amplitude frequency response is decreased to 1.6%/GHz. Consequently, after analyzing the amplitude response of the coated sample with 1 μm analyte, the amplitude difference was found to be 18% only.

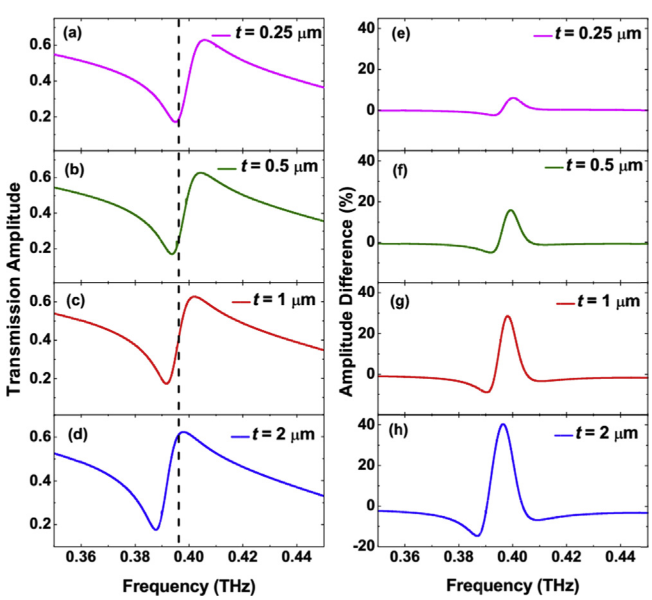

Fig. 3. Transmission amplitude spectra comparing the conventional and the ADRT for different thicknesses of the analyte deposited on the metasurface with t = 0.25 μm–2 μm: Fig. 2(a)–(d) using the conventional normalization method; Fig. 2(e)–(h) for the same thicknesses using the ADRT with the uncoated metasurface response as a reference.

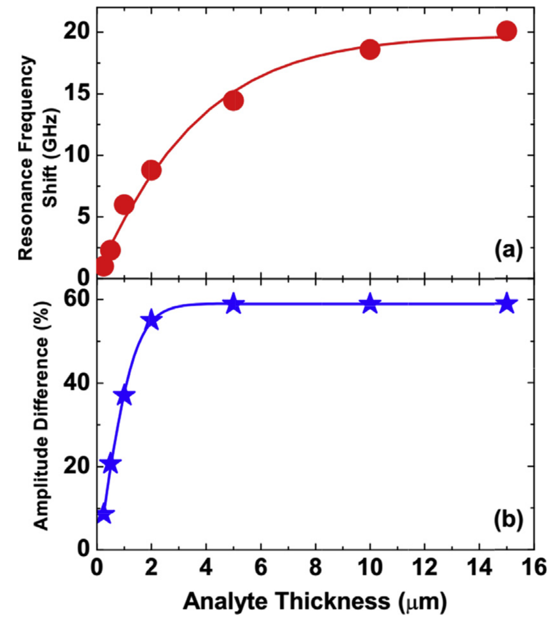

Fig. 4. (a) Fano resonance frequency shift and (b) peak-to-peak amplitude difference versus analyte thickness. The lines are meant as guides for the eye.

Finally, we compare the performance of the ADRT versus the conventional normalization method for a wide range of the analyte thickness between 0.25 μm and 15 μm. Fig. 4(a) and (b) show the resonance frequency shift and the peak-to-peak amplitude difference using the conventional normalization method and the ADRT as a function of the analyte thickness, respectively. The relation between the resonance frequency red-shift and the analyte thickness is somewhat exponential for small thicknesses less than 5 μm and gradually reaches a saturated value as revealed in Fig. 4(a).

In contrast, the saturation using the ADRT takes place for analyte thickness beyond 2 μm as shown in Fig. 4(b). It is attributed to the fact that the amplitude difference value can't be more than the difference between the peak and the dip of the amplitude response around the resonance. It is evident from these results that the ADRT can prove advantageous for sub-micron analyte thickness up to 2 μm.

4. Conclusions

In summary, the amplitude difference referencing technique has been evaluated in this paper in order to sense unknown analytes with sub-micron thickness. High *Q*-factor Fano resonance excited via asymmetric split ring metamaterial resonators has been utilized in the evaluation process of the ADRT and comparing it to the conventional normalization method. More specifically, we carefully evaluated the performance of the ADRT for a range of analyte thicknesses. The achieved amplitude difference is impressive, despite the fact that only sub-micron analyte thickness has been applied to the metamaterial sensor. In future, the ADRT can be utilized to identify analytes with sub-micron thickness and hence pave the way for a new generation of label-free biomedical sensors.

Competing interests

The authors declare no competing financial interest.

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