Dual-Sensitivity Terahertz Metasensor Based on Lattice–Toroidal-Coupled Resonance

Jing Lou, Ruisheng Yang, Jianguang Liang,* Ying Yu, Lei Zhang, Chiben Zhang, Tangjing Li, Yuancheng Fan, Fuli Zhang, Guangming Wang, Jun Wang,* and Tong Cai*

High quality factor resonance with extremely narrow line-width offers an important platform for terahertz (THz) sensing technology as it enables strong light-matter interaction between THz waves and analyte materials. Lattice mode arising from the collective Rayleigh scattering of metamaterial periodic structures has the ability to strongly confine the electromagnetic waves on the surface that fails to radiate to the far-field. Herein, for the first time, a strategy is experimentally demonstrated to design THz metasensors, which exhibit dual-sensitivity of frequency and resonance intensity by coupling the first-order lattice mode to the toroidal resonance. The frequency sensitivity mainly results from the localized field confinement, whereas the sensitivity of resonance intensity depends on the matching degree between toroidal resonance and lattice mode. It is found that both the frequency shift and resonance intensity show exponential growth with the increase in the analyte thickness. In addition, the sensing performance between toroidal and toroidal–lattice modes is compared to verify the superiority of the dual-sensitivity property. This work would greatly improve the practical applications of THz sensing technology and open up new opportunities for the realization of slow light devices, multiband narrow filters, and nonlinear systems.

1. Introduction

Enhanced light-matter interaction has attracted tremendous attention of scientific community and has grown exponentially in the past few years. These interactions are not only significant to fundamental research such as nonlinear systems, strong coupling regimes but also display potential applications ranging from sensing, filtering, and lasing areas. Among different enhanced platforms, metamaterials, consisting of subwavelength and periodic meta-atoms, have exhibited remarkable performance due to the strong near-field coupling ability that can carry the interaction energy to the radiation field. The typical advantage of metamaterial structure is to support electromagnetic resonance at a desired frequency based on its structural parameters. Generally speaking, the field confinement ability of metamaterials is always limited by radiative and nonradiative loss mechanisms. To ensure the high performance of resonant metamaterial devices, the losses should be controlled to obtain high quality (Q) factor resonances with extremely narrow line-widths. Nonradiative losses can be improved by tailoring the conductivity of metallic resonators or selecting low-loss dielectric medium. On the other hand, radiative losses can be engineered by optimizing the subwavelength structures to excite sharp asymmetric resonant response, widely known as Fano and toroidal resonances. Fano resonances arise from the interference between bright mode and dark mode, which could concentrate the electric field in the minute Fano-gaps. Toroidal dipole viewed as the magnetic currents flowing on the surface of a torus was first considered by Zel’dovich to explain parity violation in the weak interaction. The radiative feature of toroidal geometric is quite unique, resulting from its fantastical field localization configuration in a head-to-tail manner. It was found that the toroidal metamaterials could enhance the light-matter interaction within a small volume, which subsequently produces radiation patterns that couple weakly to the free space. Optimizing the losses would certainly improve the overall performance of metamaterials and plasmonic structures. One of the direct applications demonstrated by metamaterials is about...
sensors operating at optical, infrared, and terahertz (THz) frequencies. In general, a narrow line-width metamaterial response with enough resonance intensity is essential for designing ultrasensitive metasensors. Although continued strides have been devoted to THz metasensors, most of the previous sensing performance was limited to only frequency sensitivity, which greatly restricts the freedom of THz detection. In addition, the substrates of the reported metasensors such as silicon, sapphire, and polyimide (PI), merely serve as a support of metallic structures, which also violates the philosophy for the future devices with integrated requirements. Thus, to promote their practical applications with compatible techniques, there is a strong demand on design of THz metasensors that possess flexible sensing freedom, easy fabrication technique, and reliable detection performance.

In this work, we propose and experimentally demonstrate a novel method to design THz metasensors based on the coupling effects between the toroidal resonance and the first-order lattice mode (FOLM) that is inherent in metamaterial periodic arrays. Our results indicate that the reported THz metasensors can provide two freedom degrees including frequency and resonance intensity sensitivities. It has been verified that both the frequency shift and resonance intensity increase exponentially with the increasing analyte thickness in simulations and measurements. The frequency sensitivity is mainly caused by the increase in inductance in the metamaterial structure arms together with the fringing field effect, whereas the sensitivity of resonance intensity depends on the matching degree of toroidal resonance and lattice mode. Moreover, we compare the sensing performance between toroidal and toroidal–lattice modes in simulations. Both designs display 39 GHz/RIU frequency sensitivity for 4 μm thickness analyte layers with different refractive index. The resonance intensity for toroidal–lattice mode shows an exponential increase from 0.27 to 0.88 with the increase in refractive index from 1 to 4, whereas the toroidal mode stabilizes around 0.88. Thus, coupling of the lattice mode to the toroidal resonance would open an avenue toward realization of lasing spacer, ultrasensitive sensors, low-threshold switching, and nonlinear systems. In addition, the strategy based on lattice–toroidal-coupled resonance can be extended in a wideband range from microwave to optical frequencies.

2. Results and Discussion

Due to the trapped fields, lattice mode behaves as a dark mode existing in the metamaterials and its resonance frequency can be tuned by altering the periodic of meta-atoms. The lattice modes are observed as kinks or discontinuities in the transmission or reflection spectra of the metamaterials, and also referred to as diffractive modes or Woods anomalies. The frequencies of the lattice modes for a square metamaterial structure can be evaluated as

\[ f_{LM} = \frac{c}{nP} \sqrt{p^2 + j^2} \]

where \( c \) is the speed of light in vacuum, \( n \) is the refractive index of the substrate, \( P \) is the lattice period, and \((i,j)\) are non-negative integers defining the order of the lattice mode. Equation (1) implies that the frequency of FOLM is directly associated with the refractive index of the substrate. It has been reported that both high Q factor and figure of merit (FOM) can be simultaneously obtained by coupling the FOLM to the eigenmodes of the metamaterials such as Lorentz resonances, electromagnetically induced transparency (EIT)-type resonances, and Fano resonances, which strongly enhances the resonant-field confinement and reduces the radiative loss. Inspired by these works, coupling of lattice mode to the toroidal resonance may furnish a new route to design THz metasensors.

Previous studies have indicated that the split modes excited by typical two joint metallic loops consist of lower-frequency toroidal resonance and higher-frequency Lorentz resonance (red line in Figure 1a). However, the toroidal resonance being the lower-frequency spilt mode implies that coupling to the lattice mode requires a large period to fit the FOLM frequency, which would reduce the efficiency of devices. To settle down this problem, we convert the gap-coupling (capacitive-coupling) to side-coupling (inductive-coupling) by flipping the arms of the two joint metallic loops, as shown in the inset of Figure 1a. The nature of toroidal resonance is generally identified by analyzing the opposite surface currents oscillating in the two loops. Figure 1a shows that the “flipping” adjustment realizes the transformation of toroidal resonance frequency changing from lower to higher than Lorentz frequency, which brings the toroidal resonance closer to the FOLM frequency and ensures an efficient coupling between the two modes. The simulated H-field (on XZ-plane at \( Y = 0 \)) of typical and optimized toroidal resonators at corresponding resonance frequencies can be seen in the Note 1, Supporting Information.

The proposed THz metasensor architecture is shown in Figure 1b, which describes the schematic of analyte-coated metamaterial arrays along with the unit cell dimensions shown in the inset. The electromagnetic waves ranging from 0.4 to 1.1 THz excites the sample at normal incidence, having its electric filed component parallel to the nongap arm of the meta-atoms. The samples were fabricated on silicon substrate using single-step photolithography followed by depositing 20 nm of titanium (Ti) film and 200 nm of platinum (Pt) film by magnetron sputtering. To obtain a better delay signal and enable a scan length of 40 ps in the time domain, providing an enough frequency resolution of 25 GHz, the high-resistivity (8 kΩ cm) silicon with 5 mm thickness is selected as the substrate. Each of the fabricated samples has an overall size of 0.8 cm × 0.8 cm and the microscope image of a small portion of an array is shown in Figure 1c, which meets the simulated design requirements. To systematically study the optimized structure and the coupling effects between toroidal resonance and lattice mode, we have first performed full-wave simulations using the Finite-Difference-Time-Domain (FDTD) method to explore the influence of period on the transmission spectra, as shown in Figure 1d. In our simulation, lossy silicon with dielectric constant of 11.9 was used as the substrate, and Pt metal with DC conductivity of 9.52 × 10^8 S m^-1 was adopted to design the metallic arrays. Variation in the FOLM frequency with respect to the lattice period is also plotted and shown by the green dashed–dotted lines in Figure 1d. It clearly depicts that a shift in the frequencies of the toroidal and Lorentz dip follows the frequency shift of the FOLM. As the frequency of FOLM approaches and matches the
toroidal resonance, the coupling effects narrow the resonance line-width and decrease the resonance intensity, due to the increased diffraction effects near the resonance that traps the energy in the metamaterial array and suppresses the far-field radiation energy. As observed, the critical period for toroidal resonance matching lattice mode is located at 94.8 μm. The underlying near-field effects between toroidal resonance and lattice mode are shown in the Note 2, Supporting Information.

To explicitly illustrate the influence of geometric structure on the transmission spectra, we carried out detailed simulations as well as experiments on the metamaterial arrays by varying the structure parameters. In experiments, a standard THz time-domain spectroscopy (THz-TDS) was used to measure the transmission spectra of the samples. All measurements were carried out at room temperature and relatively dry condition with a humidity of 35% to eliminate the absorption of THz waves by water vapor. The transmission amplitude was obtained by $|H(\omega)| = E_s(\omega)/E_r(\omega)$, with $E_s(\omega)$ and $E_r(\omega)$ being the electric-field amplitudes transmitted through the sample and reference substrate, respectively.

Figure 2 shows the experimental and simulated transmission spectra, which are in good agreements. Quantitative deviations in the line-widths and resonance intensity between the simulations and experiments mainly result from the lower spectral resolution of the measurement setups. The slight difference in resonance frequency is mainly due to the fabrication imperfection. The corresponding scanning electron microscope images of the fabricated metamaterials are shown in Figure S4, Supporting Information. For the sample with the structure parameters $G = 6$ μm, $P = 90$ μm (Figure 2a), a broad Lorentz-type symmetry resonance and a sharp toroidal-type asymmetry resonance center around 0.764 and 0.963 THz, respectively. As shown in Figure 2a–c, with the increase in coupling distance $G$ from 6 to 11 μm, the frequencies of the two resonances move closer to the center and the toroidal resonance intensity increases gradually. Next, when the coupling distance $G$ is fixed at 8.5 μm, resonance coupling between toroidal mode and lattice mode is observed, which significantly decreases the resonance intensity and narrows the line-width. The three-oscillator model is adopted to describe the coupling mechanism (details are provided in Note 3, Supporting Information). Further increasing the lattice period would...

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**Figure 1.** Design of THz metasensor based on the coupling effects between the optimized toroidal resonance and lattice mode. a) Simulated transmission spectra of the typical (red line) and optimized toroidal (black line) resonances. Inset figures show the simulated surface currents at respective toroidal resonance frequencies. A cyan dotted line marks the first-order lattice mode $f_{LM(0,1)}$. b) An artistic illustration of THz metasensor coated with the analyte layer on the top. Inset figure shows the unit-cell dimensions of the metamaterial structure: $K = 54$ μm, $F = 27$ μm, $W = 8$ μm. c) Microscopic image of fabricated sample with $G = 8.5$ μm, $P = 94$ μm. d) Contour plot showing the variation in the transmission amplitude of metamaterial as a function of frequency and lattice period. The green dashed–dotted curve signifies the FOLM frequency $f_{LM(0,1)}$ with varying lattice period $P$. 
continue to reduce the frequency of the lattice mode and result in the mismatch between the two modes, broadening the line-width and eliminating the typical toroidal resonance dip, as shown in Figure 2e,f. Thus, the resonance intensity demonstrates a regular and apparent trend changing with the matching degree between the two modes, which satisfies the design requirement of THz metasensors. The similar coupling features also occur in the Lorentz frequency, as shown in Figure 2g,h.

On the basis of aforementioned observations, it is clear that the toroidal–lattice mode provides another sensitivity of resonance intensity to realize sensing function apart from the fundamental frequency sensitivity when the FOLM frequency is near toroidal frequency. To present the advantages of the designed THz metasensors, we study the sensing performance on the metamaterial arrays with lattice period of 94 and 97 μm, respectively. In our experiment, a spin-coating process enables...
the uniform deposition of PI film on the metallic layer, which then solidified in a vacuum chamber at 140, 250, 350 °C for 60, 60, 120 min, respectively. PI film (n = 1.79) with different thickness can be obtained by modifying the rotation speed and repetition times during the spin-coating process. A step-ladder was used to measure the thickness of PI film by the scratch method.

**Figure 3a** shows the simulated transmission spectra for the 94 μm-period sample coated with different thickness of PI films. Without PI film, the toroidal frequency first occurred at 0.92 THz. For the PI films of 2.5, 4.8, 7.8, 10.1 μm thickness, the corresponding red-shifts of toroidal resonance are 15.5, 23.5, 30.5, 32 GHz, respectively. Importantly, it is apparent that the increase in PI thickness broadens the resonance line-width and deepens the transmission dip, which is in accordance with the aforementioned analyses. **Figure 3c** shows the measured data that agree well with the simulations. In addition, we also simulated the transmission spectra for the 97 μm-period sample, as shown in **Figure 3b**. Without PI film, the frequency of toroidal resonance is lower than the lattice mode, leading to the disappearance of toroidal dip. Once the 2.5 μm thick layer of PI film is spin-coated on the metallic structure, the frequency of toroidal resonance red-shifts and becomes lower than the lattice mode, resulting in the typical toroidal dip at 0.891 THz. With the increase in PI film thickness, the transmission spectra show the similar trend as that of the 94 μm-period samples, and the experimental results (Figure 3d) match well with the simulations (Figure 3b). The frequency sensitivity of toroidal resonance is mainly caused by the increase in inductance in the metamaterial structure arms together with the fringing field effect, whereas the sensitivity of resonance intensity is determined by the matching degree of lattice mode and toroidal resonance.

To further quantify the dual-sensitivity performance of the proposed THz metasensors, we have plotted the frequency shift and resonance intensity in respect of the analyte thickness, as shown in **Figure 4**. Both the frequency shifts of simulations (Figure 4a) and measurements (Figure 4b) increase with the exponential fit as the analyte thickness, due to per exponential decay of the localized electric field.[9,10] The exponential fit has been performed by the equation: $f_{\text{shift}} = f_{\text{sat}} + A \times \exp(-R_0 \times t)$, where $t$ is the thickness of PI film and $f_{\text{sat}}$ is the saturated frequency shift for the fitted curve. The corresponding parameters for the simulated and experimental frequency shifts are shown in Table S1, Supporting Information. It is noticed that the saturated frequency shift for the 97 μm period is slightly larger than that of the 94 μm period. This phenomenon can be attributed to that as the lattice mode is swept through the toroidal resonance (from being lower to higher than toroidal frequency), the field confinement of lattice mode with 97 μm period could be fully utilized to obtain more frequency shift. A detailed study of the resonance intensity sensitivity is also analyzed using rigorous simulations and experiments by varying the analyte thickness, as shown in **Figure 4c,d**. The resonance intensity is characterized as $(T_A - T_B)/T_A$, where $T_A$ and $T_B$ are the transmission peak and dip of toroidal resonance, respectively.

**Figure 3.** Sensing performance against lattice periods and thickness of PI films. a,b) Simulated and c,d) measured transmission spectra with different thickness of PI films coated on the top of metamaterials with lattice period of (a,c) 94 μm and (b,d) 97 μm.
Similar to the frequency sensitivity, the resonance intensity also exhibits a distinct exponential growth trend with the analyte thickness. The exponential fit has been performed by the equation: 

\[ R_{\text{Intensity}} = R_{\text{sat}} + B \times \exp(-R_1 \times t) \]

where \( t \) is the thickness of PI films and \( R_{\text{sat}} \) is the saturated resonance intensity for the fitted curve. The corresponding parameters for the simulated and experimental resonance intensity are shown in Table S2, Supporting Information. Noted that the respective saturated resonance intensity for lattice period of 94 and 97 \( \mu \)m is 0.7 and 0.56 in the simulations, whereas the corresponding experimental value is 0.3 and 0.12. The differences in resonance intensity between experiments and simulations are mainly caused by the limited resolution of instruments and the influence of testing environment, which is in accordance with the reported works \([10, 43, 48, 52]\).

Finally, we compared the sensing performance between toroidal and toroidal–lattice modes by varying the refractive index of the analyte and maintaining a constant thickness, with the results shown in Figure 5. The two metasensors have the same geometric structure \((P = 94.8 \mu\text{m})\) except flipping the two opposite resonator arms. The simulated transmission spectra in the both metasensors (Figure 5a, b) show the red-shift of resonance frequency with increase in the refractive index of 4 \( \mu\text{m} \) thickness analyte layer. The variation in frequency shifts appears to be linear in nature for the aforementioned two designs, and both the frequency sensitivity of fitted curve turns out to be 39 GHz/RIU (Figure 5c). Another comparative feature between the two designs that could be highlighted in terms of resonance intensity changing with increase in the analyte refractive index. As shown in Figure 5d, the resonance intensity of toroidal mode almost does not change with the refractive index and remains around 0.88. In contrast, with the refractive index changing from 1 to 4, the resonance intensity of the lattice–toroidal mode increases exponentially from 0.27 and eventually stabilizes at about 0.88, and the modulation depth reaches 69.3%. Therefore, toroidal–lattice mode offers a more efficient routine for interaction between matter and THz waves, which tends to be better platforms for THz sensing compared with the existing metasensors \([9, 10, 39–43]\).

3. Conclusion

In summary, based on the coupling effects between toroidal resonance and lattice mode, we experimentally demonstrate a novel THz metasensor with dual-sensitivity of frequency and resonance intensity. For practical applications, this strategy is not only superior to other methods for the preliminary detection of substances but also increases the freedom degree of terahertz sensing technology. Our results indicate that both the frequency shift and resonance intensity exhibit exponentially increased fitting with the analyte thickness. The frequency sensitivity mainly results from the localized field confinement, whereas the
resonance intensity sensitivity depends on the matching degree between toroidal and lattice modes. Furthermore, we compare the sensitivity performance of toroidal mode and toroidal–lattice mode metamaterials coated with 4 μm thickness analyte of different refractive index. The exponential fit for resonance intensity (toroidal–lattice mode) has been performed by the equation: 
\[ R_{\text{Intensity}} = R_{\text{sat}} + \frac{C}{1 + R_2 (I - 1)} \], where \( I \) is the Refractive index of analyte materials and \( R_{\text{sat}} = 0.8787 \) is the saturated resonance intensity for the fitted curve. \( C = -0.606; R_2 = 1.1 \).

Keywords
dual-sensitivities, metamaterials, mode coupling, terahertz sensing

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

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