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A New Design of a Terahertz Metamaterial Absorber for Gas Sensing Applications

Sagnik Banerjee [1], Purba Dutta [2], Snehashish Basu [2], Sunil Kumar Mishra [2], Bhargav Appasani [2,*], Sarita Nanda [2], Yadgar I. Abdulkarim [3], Fahmi F. Muhammadsharif [4], Jian Dong [5], Amitkumar V. Jha [2], Nicu Bizon [6,7,8,*] and Phatiphat Thounthong [9,10]

[1] Department of Information Engineering, Electronics and Telecommunications, Sapienza University of Rome, Via Eudossiana 18, 00184 Roma, Italy
[2] School of Electronics Engineering, Kalinga Institute of Industrial Technology, Bhubaneswar 751024, India
[3] Medical Physics Department, College of Medicals & Applied Science, Charmo University, Chamchamal 46023, Iraq
[4] Department of Physics, Faculty of Science and Health, Koya University, Koya 44023, Iraq
[5] School of Computer Science and Engineering, Central South University, Changsha 410075, China
[6] Faculty of Electronics, Communication and Computers, University of Pitesti, 110040 Pitesti, Romania
[7] ICSI Energy, National Research and Development Institute for Cryogenic and Isotopic Technologies, 240050 Ramnicu Valcea, Romania
[8] Doctoral School, Polytechnic University of Bucharest, 313 Splaiul Independentei, 060042 Bucharest, Romania
[9] Renewable Energy Research Centre (RERC), Department of Teacher Training in Electrical Engineering, Faculty of Technical Education, King Mongkut's University of Technology North Bangkok, 1518 Pracharat 1 Road, Wongsawang, Bangsue, Bangkok 10800, Thailand
[10] Group of Research in Electrical Engineering of Nancy (GREEN), University of Lorraine-GREEN, F-54000 Nancy, France
* Correspondence: bhargav.appasanifet@kiit.ac.in (B.A.); nicu.bizon@upit.ro (N.B.)

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Abstract: Metamaterial absorbers are used in the terahertz frequency regime as photo-detectors, as sensing elements, in imaging applications, etc. Narrowband absorbers, on account of their ultra-slender bandwidth within the terahertz frequency spectrum, show a significant shift in the absorption peak when an extrinsic entity relative to the absorber, like refractive index or temperature of the encircling medium, is altered. This property paves the path for the narrowband absorbers to be used as potential sensors to detect any alterations in the encircling medium. In this paper, a novel design of a terahertz metamaterial (MTM) absorber is proposed, which can sense the variations in the refractive index (RI) of the surrounding medium. The effective permeability of the structure is negative, while its permittivity is positive; thus, it is a μ-negative metamaterial. The layout involves a swastika-shaped design made of gold on top of a dielectric gallium arsenide (GaAs) substrate. The proposed absorber achieved a nearly perfect absorption of 99.65% at 2.905 terahertz (THz), resulting in a quality factor (Q-factor) of 145.25. The proposed design has a sensitivity of 2.12 THz/RIU over a range of varied refractive index from $n = 1.00$ to $n = 1.05$ with a step size of 0.005, thereby achieving a Figure of Merit (FoM) of 106. Furthermore, the sensor was found to have a polarization-insensitive characteristic. Considering its high sensitivity (S), the proposed sensor was further tested for gas sensing applications of harmful gases. As a case study, the sensor was used to detect chloroform. The proposed work can be the foundation for developing highly sensitive gas sensors.

Keywords: metamaterial absorber; sensitivity; Q-factor; figure of merit; refractive-index sensor; polarization; Refractive Index Unit (RIU); gas sensor

1. Introduction

In recent decades, periodic sub-wavelength structures have been extensively used in literature in a wide range of applications like sensing, spectroscopy, optoelectronics, antenna theory, etc. These structures, termed metamaterials (MTM) [1], have optical and mechanical properties not found in naturally occurring materials (like negative refractive index [2]) and can be tuned to attain the desired characteristics. By manipulating the optical nonlinearities, high efficiency with minimal power loss can be obtained.

The construction of a unit cell of metamaterials involves periodic arrangements of sub-wavelength units of metal resonators forming an isotropic lattice atop a dielectric medium. Low-loss photoconductive materials like polyimide (PI) and gallium arsenide (GaAs) are generally preferred to be used as the dielectric. Due to their high electron mobility and thermal stability, GaAs is widely used in high-frequency, e.g., terahertz (THz) range, applications [3,4]. In high-frequency applications like sensing and imaging, THz frequencies are preferred over microwave range due to their larger bandwidth [5,6]. In addition, it is non-ionizing, hence, does not interfere with the sensing and imaging process. Metamaterial-based THz sensing can thus guarantee an optimal performance necessary for research and development.

Label-free sensing methods are implemented for refractive index (RI) and gas sensors owing to their cost-effectiveness and high operating speed [7]. The RIU of a sensor is just a single unit of the refractive index of a given material which is very useful in quantifying the sensitivity of a surface plasmon instrument (like terahertz sensors).

Metamaterial-based THz label-free sensing was implemented by Ahn and Park [8] to detect the presence of microorganisms by observing the variation of the dielectric constant with the resonance frequency. A more significant shift can be attained by observing the variation in refractive index as the change in RI would be smaller. A metamaterial-based multi-band sensor at THz frequencies for the detection of pesticides was proposed in [9] for three different resonant frequencies. The resonant frequencies lie in the short range of 0–2 THz; for larger samples, a wider frequency range would be necessary. An RI sensor with a high sensitivity of 1317 nm/RIU was proposed in [10] by carving cross-shaped holes in a silver substrate. A one-dimensional metasurface-based RI sensor was designed in [11] to detect small amounts of samples. The sensor obtained a sensitivity of 517.9 GHz/RIU and a high quality-factor of 262. Though fabrication can be difficult, several new studies use materials like graphene and vanadium dioxide to increase performance. Coating graphene on gold nano-disks, a gas sensor was designed in [12]. The use of graphene resulted in a high sensitivity of 720 nm/RIU. However, it operates at optical frequencies.

With an accelerating increase in the number of vehicles and industries, the release of pollutants like CO_2, CO, SO_2 and chloroform tends to go unchecked on several occasions [13]. This can have adverse effects on human health and the environment. Gas sensors prove vital in curbing the ill effects of these gases by detecting their levels present in the atmosphere and enabling hazard warning systems [14]. In addition, with the onset of SARS-CoV-2, the prolonged usage of face masks has led to rebreathing of the trapped CO_2 and, consequently, induced a need to monitor the CO_2 using smart masks [15]. Aqueous chloroform ($CHCl_3$) [16] normally used in refrigerants and released from paper manufacturing industries, chlorinated water and landfills, rapidly converts into a gaseous state due to its volatile nature. It has carcinogenic properties and poses a threat to the eyes, skin, liver and lungs. At certain frequencies, the molecules of gases like $CHCl_3$ absorb radiation. The resonance frequency is measured to detect the gas. Consequently, for a high range of frequencies, a high sensitivity and, therefore greater performance can be obtained. In metamaterial absorbers (MA), when a wave is incident on the metal-dielectric interface, localized surface plasmons get excited and give rise to resonating modes. Due to this resonance, the light gets confined to the surface and converts into heat energy. The resultant attenuation enhances the electromagnetic field and increases absorption [10,17,18].

A gas sensor to detect the presence of chloroform is proposed in this paper. The design is based on a metamaterial absorber operating in the THz frequency range. The novelty of the design is exemplified by means of a comparison analysis in Table 1. In this table, quality factor (Q-factor) is the ratio of resonance frequency to the full width half maximum (FWHM) bandwidth; sensitivity is the shift in resonance frequency per unit change in the refractive index; figure of merit (FoM) is the ratio of sensitivity to the FWHM.

Table 1. Comparison of performance of sensors.

Reference No.	Q-Factor	Sensitivity (GHz/RIU)	FoM (/RIU)	Polarization Sensitive	Peak Absorption	Application in Gas Sensing
[19]	78.90	0.3537	11.0531	Yes	90%	No
[20]	13.76	851	2.927	No	99.75%	No
[21]	32.167	187	6.015	Yes	99.8%	No
[22]	44	1500	25	Yes	99.5%	No
[23]	29.5, 66.8, 59.4	540, 700, 1500	6.75, 17.5, 30	Yes	99%	No
[24]	231	186	187	Yes	97%	No
This paper	145.25	2120	106	Yes	99.65%	Yes

Metamaterials can be broadly categorized into two types vis-à-vis, double-negative (DNG) with negative values of both permittivity and permeability and single negative (SNG) for either of the negative parameters. Consequently, for negative permeability, μ, whilst $\varepsilon > 0$, the MTM structure is termed as μ-negative (MNG). This particular type can be designed by implementing artificial magnetism, thus enhancing the magnetic coupling near the resonance frequency [25]. A common example of a magnetic metamaterial is a ferromagnetic substance [26]. An approach to understanding the magnetism in nonmagnetic, conducting periodic arrays of split-ring resonators is showcased by Pendry et al. [27]. Gyrotropic and bi-gyrotropic materials have been studied for larger applicability to magnetic metamaterials [28,29].

For gas detection applications, various plasmonic sensing platforms have been used. Prism-based plasmonic sensors, which are accomplished using either Otto or Kretschmann topologies [30], and grating-based plasmonic sensors are examples of these [31,32]. From a practical point of view, prism-based configurations (Kretschmann and Otto configurations) are bulky, making them unsuitable for many real-world applications and, therefore, cannot meet the small probe size criterion for on-chip integration typically required for gas sensors [33]. Although grating-based plasmonic devices have shown more potential in on-chip implementation and tiny probe size, their sensitivity and detection limits are often insufficient to detect low gas concentrations [32]. Metamaterial absorbers are one of the most effective approaches to creating sensitive gas sensors with compact size and excellent accuracy. Recently, in [34], a gas sensor based on THz metamaterial absorber has been proposed. The design consists of concentric elliptical rings and offers a peak sensitivity of 3.59 THz/RIU. However, the design is unsymmetrical, so the absorption characteristics vary with the polarization angle. Also, the concentric rings are closely closed posing difficulty for future fabrication. This work presents a novel, highly sensitive, and innovative THz gas sensor based on Swastika shape, which is polarization angle independent. Compared to time-consuming traditional burdensome approaches such as fluorescence detection or resistive gas sensing, its comprehensive advantages open up new frontiers for label-free real-time optical gas detection.

This paper proposes a novel metamaterial absorber design that finds potential sensing applications in the terahertz regime. The proposed design is tested in the refractive index range of n = 1.00 to n = 1.05, having a step size of 0.005, and it is found to provide a sensitivity of 2.12 THz/RIU. The proposed design offers a Q-factor of 145.25 and a FoM of 106. In order to enhance the practical feasibility of the proposed sensor, we have used it to detect harmful and poisonous gases such as chloroform.

The remainder of the paper is organized as follows: the next section discusses the design of the proposed absorber, while the third section analyses the obtained results from simulations along with the description of the resonance phenomenon and parametric analysis of the design parameters. The fourth section provides an analysis of the potentiality of the proposed sensor to detect harmful gases. Eventually, the last section concludes the work.

2. Motivation and Structural Design

The idea of metamaterials first appeared in 1968, when Veselago hypothesized the possibility of elements featuring negative permittivity and permeability predicated on the famous Maxwell's equations [35]. The remarkable characteristic of metamaterials is the ability to control effective permittivity and permeability by purposefully constructing a design which is not present in naturally occurring materials. Typically, metamaterial absorbers are three-layered structures consisting of a ground plane and a dielectric spacer below the top surface with the design [36,37]. Numerous research, often including left-handed materials, photonic crystals, and super-magnetic components, have demonstrated the link between the dielectric characteristics and structural design of metamaterials. Notably, because of their excellent label-free identification and great sensitivity, metamaterials have contributed to the progress of THz sensing. Furthermore, because of the extraordinary properties of metamaterials in controlling amplitudes, phases, polarity, and impedance, terahertz metamaterial sensing delivers prospective usage in biological and other such domains [38]. The preponderance of previous research concentrated on metamaterial design modeling rather than material selection. In reality, appropriate functional choice of materials depending on the material science concepts is essential to produce an optimal "metamaterial" framework with compelling features and a variety of real-world applications.

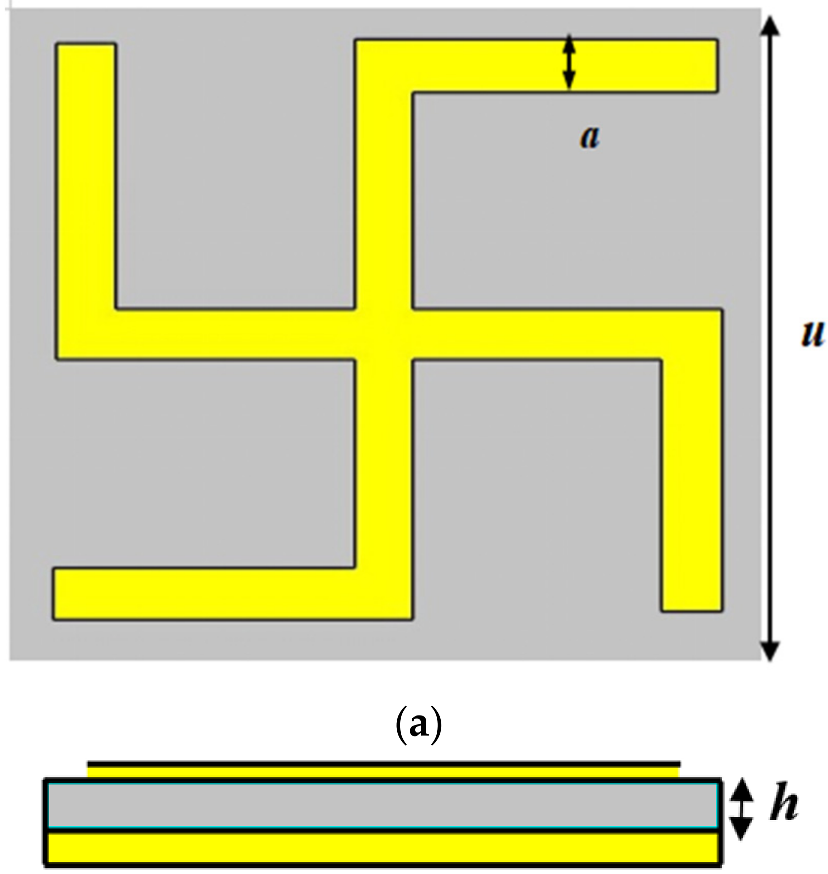
The structural design of the proposed sensor is depicted in Figure 1. The ground plane and the top surface are made of gold. Gold possesses a very high conductivity of 4.1×10^7 S/m and is less lossy at THz frequencies [34]. The sandwiched layer in between the top and ground plane acts as a dielectric spacer made of Gallium Arsenide (GaAs), which helps to provide a near-insulating region for the sensor. It is a compound semiconductor possessing a loss tangent of 0.006 and a relative permittivity of 12.94. On top of that, Gallium Arsenide is furnished with ample band gap and very high resistivity, forming a basis for its choice as a dielectric spacer. The unit cell of the proposed sensor has a height (h) of 5 µm, while the design on the top surface has a thickness (b) of 0.2 µm containing defected rectangular patches called the 'Split Ring Resonators (SRRs)', having uniform width (a) of 5 µm. As a crucial component of the sensor, the SRR helps achieve the sensor's desired magnetic susceptibility. The swastika-shaped geometry of the Split Ring Resonator generates unique electric and magnetic absorption signatures in the terahertz spectrum range. The proposed sensor has a lower substrate thickness (h) to enjoy higher mechanical pliability in the terahertz regime. The Drude's model for metals provides a method to assess the optical properties of noble metals like gold using a complex permittivity plot having real and imaginary parts [39].

$$\varepsilon_r(\omega) = \varepsilon_\infty - \frac{\omega_P^2}{\omega^2 + j\omega\gamma} \quad (1)$$

where, ω is operating frequency in rad/s, P is plasma frequency, γ is damping constant, ε_∞ is high-frequency permittivity, which is 12.94.

The unit cell of the proposed sensor has a square shape, having a side-length (u) of 100 µm. Lastly, the ground or the bottom plane has a stratum of 2 µm.

The frequency domain solver used by CST Microwave studio uses Finite Element Method to solve Maxwell's Equations. The frequency response is calculated at discrete frequency values with a step-wise separation from each other over the entire bandwidth. A frequency domain solver is used to obtain discrete frequency responses when we require the electromagnetic wave response in any material for any frequency value. It is highly useful for the calculation of electrically smaller structures. The frequency domain solver is calculated in S parameters, which stops after the calculation during simulations. Although the entire simulation takes time, the obtained accuracy is much higher using the frequency domain solver method. Periodic boundary constraints have been obtruded on either side of the unit cell, and eventually, Transverse Electromagnetic (TEM) waves are considered to be incident normally upon the top surface of the proposed sensor by propagating along the negative z-axis. Periodic boundary conditions help generate near-to-real simulations by transforming an infinite pseudo-crystal structured in a lattice. As a result, the system can interact with neighboring cells across the cell wall.



(b)

Figure 1. Proposed structure of terahertz metamaterial absorber: (**a**) top view; (**b**) side view.

3. Simulation Results and Discussions

We have used the CST Microwave Studio to carry out the design and simulation mechanisms. The absorption characteristics of the proposed sensor can be calculated using the following equation [34].

$$A = 1 - |S_{11}|^2 - |S_{12}|^2 \tag{2}$$

where, A is the absorption coefficient, S_{11} is the reflection coefficient, and S_{12} is the transmission coefficient. The transmission coefficient can be neglected as the propagation of electromagnetic waves is not allowed by the ground metal plane. THz radiation is incident on the surface of the proposed absorber, the S_{11} is measured from the ratio of reflected power to the incident power, and the absorption is measured. The proposed design achieved an absorption rate of 99.65% at the resonant frequency of 2.905 THz, as evident from Figure 2. The absorption peak maintained a Full-Width Half Maximum (FWHM) of 0.02 THz at the aforementioned resonant frequency, which eventually resulted in higher frequency selectivity due to the absorption peak having an extremely slender bandwidth. A very important index for any sensing application is the sensor's quality factor (Q-factor). The sensor exhibits better sensing performance for a higher value of the Q-factor. The Q-factor of the suggested design was found to be 145.25, making it viable in sensing applications.

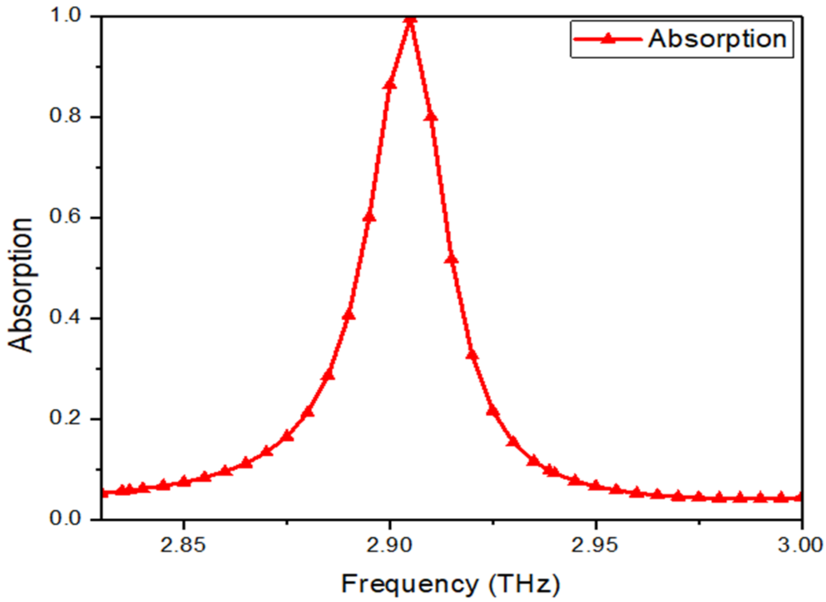


Figure 2. Absorption spectrum of the proposed sensor.

In order to have better clarity for the resonance mechanism, the surface current distribution at the resonant frequency is depicted in Figure 3. We can clearly observe that the entire SRR structure has a higher current concentration than the rest of the top plane concentration and solely contributes towards the resonance mechanism. A uniform current distribution was found all over the surface of the adjoining rectangular patches, and their cumulative effect is responsible for the appearance of the absorption bands. Thus, the concept of concatenating of the rectangular patches holds good here to bring about ultra-thin narrow bands having high quality factor within the terahertz spectrum.

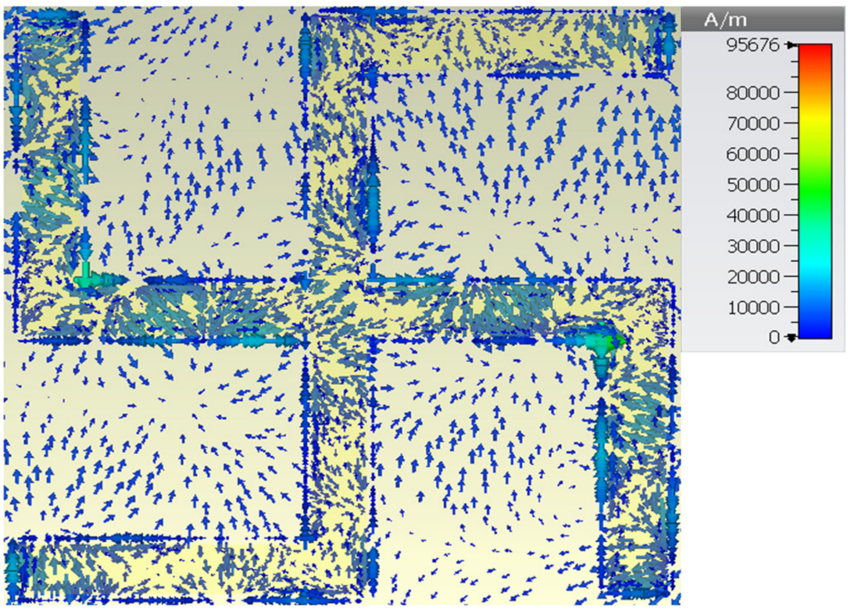


Figure 3. Surface current distribution at the resonant frequency.

The input impedance (Z_{11}) of the proposed design is 347.1–j 2.651 Ω at the resonance frequency, and the magnitude of the impedance is 347.11Ω, which is very close to the free space impedance. The impedance was calculated as a function of frequency using the following expression and is shown in Figure 4b [40].

$$Z_{11}(f) = \sqrt{\frac{(1 + S_{11}(f))^2 - S_{21}^2(f)}{(1 - S_{11}(f))^2 - S_{21}^2(f)}} \tag{3}$$

where f is the resonance frequency.

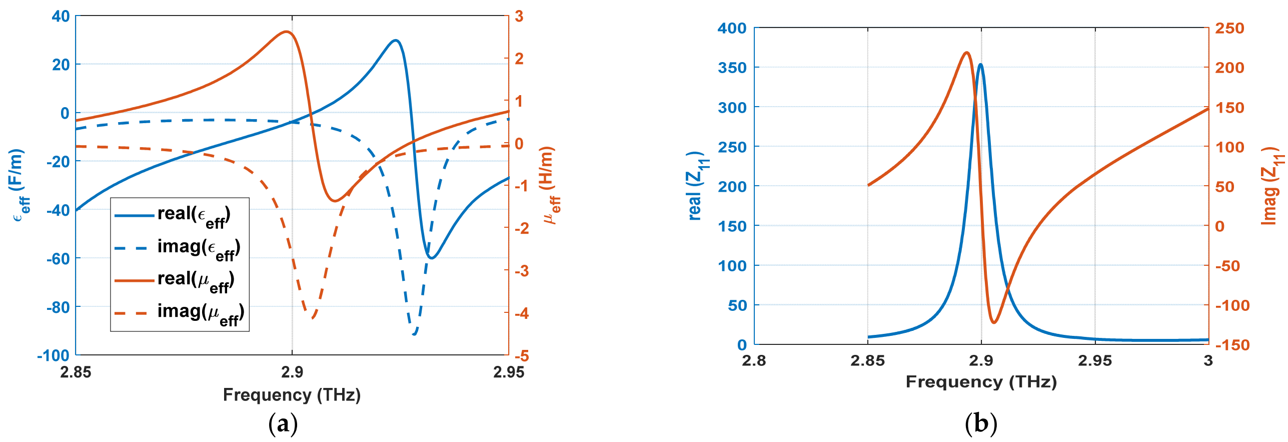


Figure 4. (**a**) Effective permittivity and permeability of the proposed design (**b**) Impedance characteristics of the design.

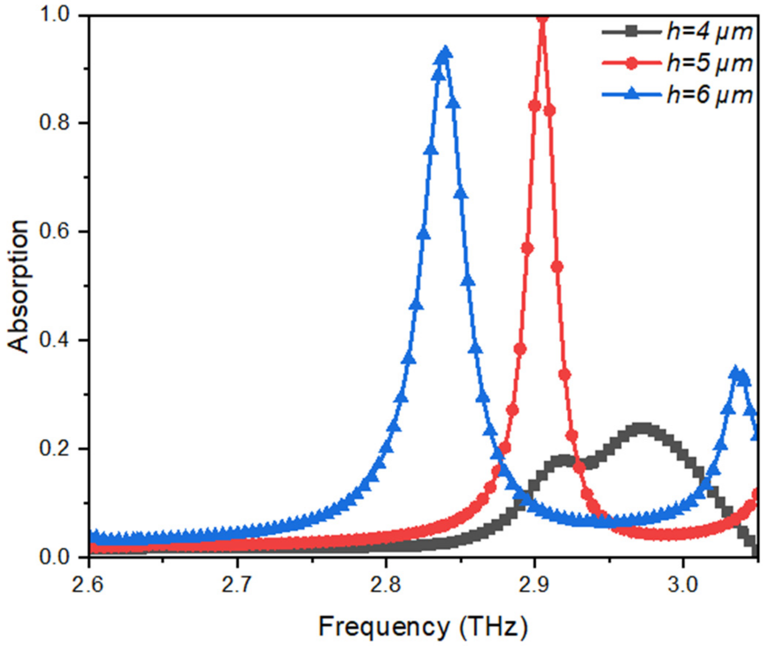
The effective permeability (ε_{eff}) and permittivity (μ_{eff}) of the proposed structure are shown in Figure 4a, to know the absorption character with enhanced clarity. These were obtained using the equations in [41]:

$$\varepsilon_{eff}(f) = \frac{c}{j\pi f d}\left(\frac{1 - S_{21} - S_{11}}{1 + S_{21} + S_{11}}\right) \tag{4}$$

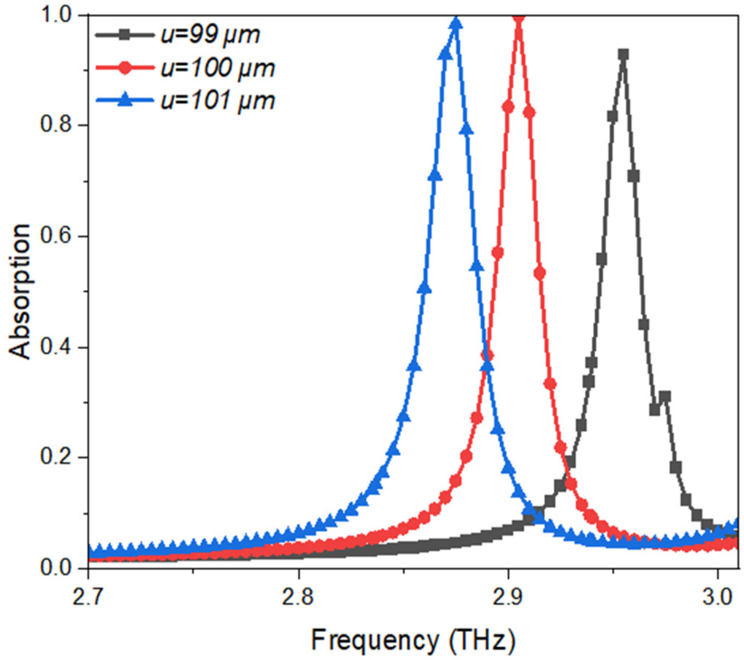
$$\mu_{eff}(f) = \frac{c}{j\pi f d}\left(\frac{1 - S_{21} + S_{11}}{1 + S_{21} - S_{11}}\right) \tag{5}$$

where c is the speed of light, and d is the thickness of the unit cell,

We find a positive magnitude for the real portion of the effective permittivity, while the real part of effective permeability has shown a negative magnitude at the resonance frequency. Hence, the proposed structure is a Single Negative Material (SNG). As we see that only the permeability is negative, so it also acts as a Mu-Negative Material (MNG). Usually, we do not find such MNG materials existing naturally; thus, it delineates the metamaterial characteristics of the proposed structure. For the sake of realizing the various parameters used in the design, we provide a parametric analysis for the height of the substrate (h), and unit cell dimensions (u). It is observed from Figure 5 that when the magnitude of the parameter increases gradually, the resonant frequency decreases and vice-versa. Hence, we obtain an inverse relationship between the parameter magnitude and the resonant frequency. Figure 5a shows that the optimal peak is obtained when the substrate thickness is h = 5 µm. A remarkable drop in the absorption rate is obtained by increasing or decreasing the substrate height further. The absorption rate in a metamaterial absorber reaches its zenith when the impedance at input becomes equal to the impedance of the free space (approximately 377 Ω) [42]. Hence, by further changing the substrate height magnitude, this impedance-matching phenomenon is disrupted, and we observe a decrement in the absorption rate. Figure 5b concludes that a unit cell dimension of u = 100 µm provides the best absorption and a slender bandwidth, resulting in a better quality factor. We observe this effect because when the unit cell dimension deviates a little from its optimum value of 100 µm, a mismatch occurs between the phase and magnitude preconditions, resulting in destructive interference due to numerous reflections degrading the absorption characteristics [23].



(**a**)



(**b**)

Figure 5. Parametric analysis for (**a**) substrate height (**b**) unit cell dimensions.

We then examine the performance of the proposed sensor for various polarization angles, as shown in Figure 6. The polarization angle was changed from 0° to 90° at 15° intervals while keeping the incidence angle perpendicular to the absorber's top plane. It is evident that with each adjustment in the polarization angle, the absorption peaks do not shift, and there is no rise or decline in the absorption rates. The suggested absorber is entirely polarization insensitive, and the design arrangement is symmetrical.

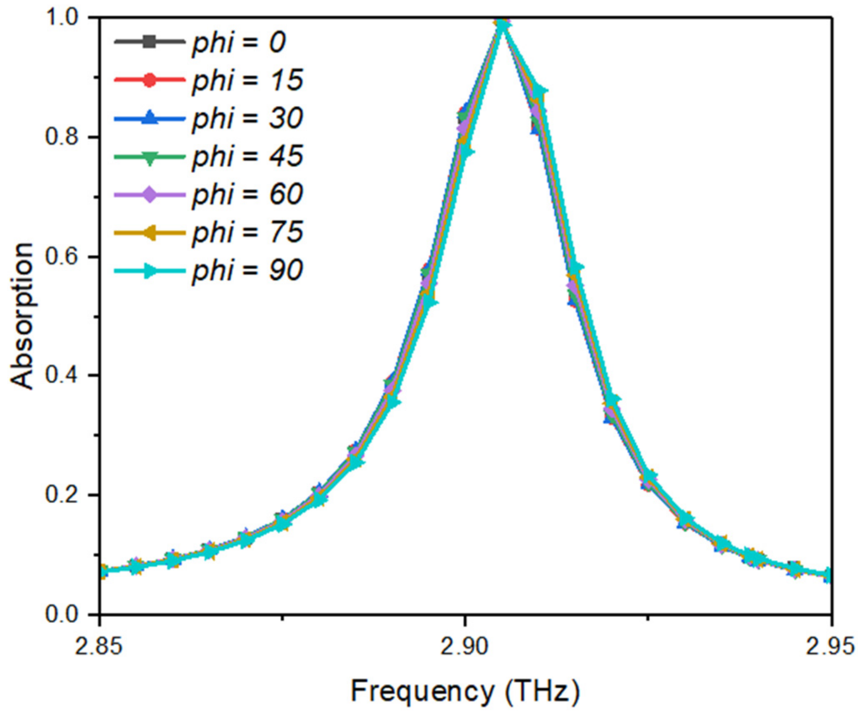


Figure 6. Absorption spectrum for different polarization angles.

Metamaterial absorber-based refractive index sensors are widely used nowadays as it remains unaffected by other external factors like temperature and pressure [2]. It should also be noted that entities like temperature and pressure are subjected to change under a wide range of conditions, but it is very difficult to alter the refractive index of a certain optical medium unless we are externally changing the color of the incident light wave from the terahertz source. The alterations in the absorption spectrum with respect to refractive index variations of the encircling media have been depicted in Figure 7, from where we can draw an easy inference that when the refractive index of the medium increases, the resonant frequency produces a leftward shift, which means it undergoes reduction and vice-versa. The variations in the refractive index have been carried out in the range of $n = 1.00$ to $n = 1.05$ by considering a very small step size or an interval of 0.005. We have used this range as it can be used for applications involving sensing harmful gases [43]. Further, it has been observed from Figure 7 that for a very minute increment in the refractive indices, the absorption peaks show a significant leftward shift which justifies that the proposed design functions as an outstanding refractive index sensor. Table 2 also provides more insights into the performance of the proposed refractive index sensor, where we do not find any significant decrement in the absorption rate when the refractive index increases.

Table 2. Variations of absorption rates with refractive indices.

Serial No.	Refractive Index (n)	Absorption (%)
1	1.00	99.65
2	1.005	97.75
3	1.01	99.50
4	1.015	99.00
5	1.02	98.75
6	1.025	98.25
7	1.03	98.25
8	1.035	97.50
9	1.04	98.00
10	1.045	98.50
11	1.05	98.50

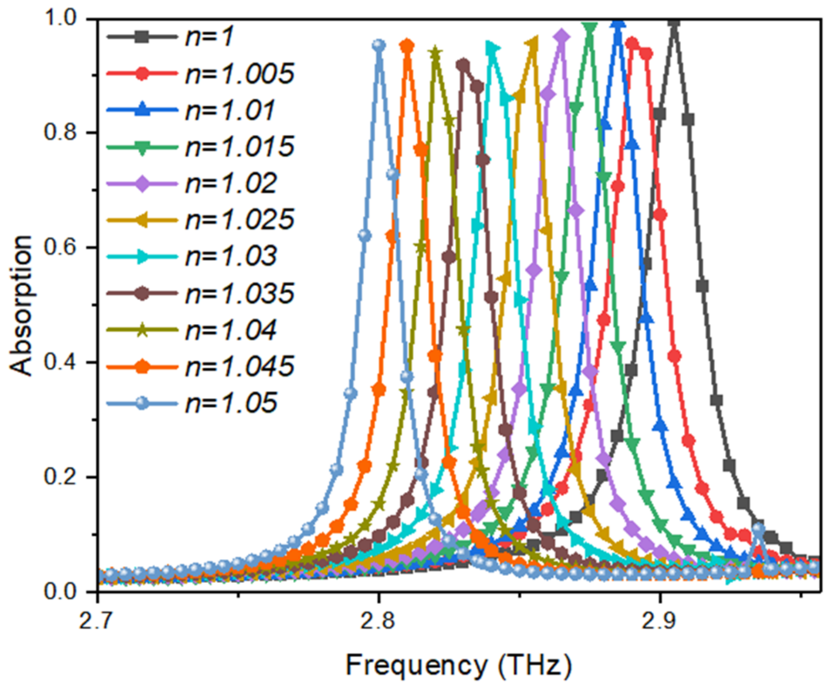
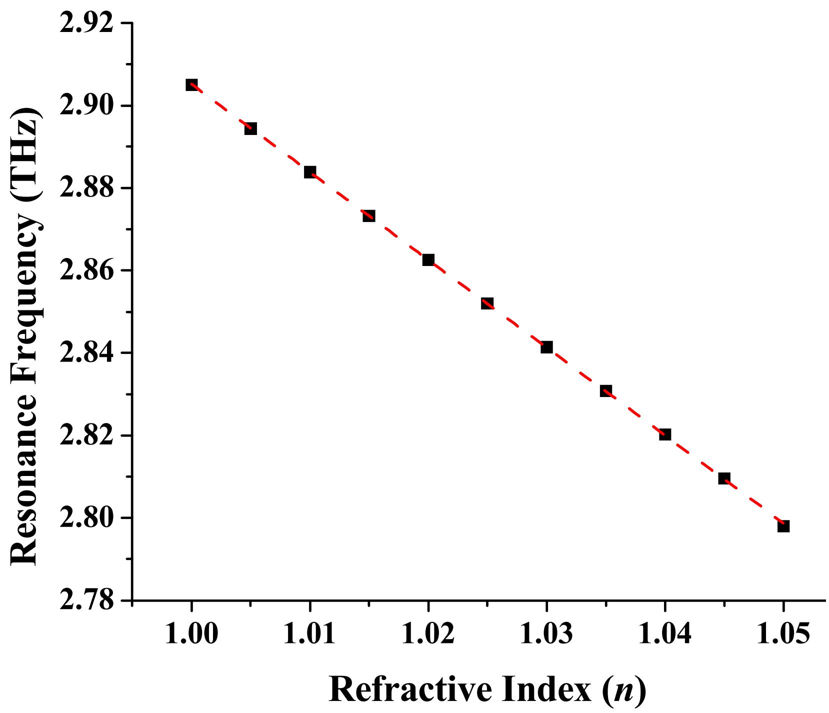


Figure 7. Absorption Spectrum for different refractive indices at the resonant frequency.

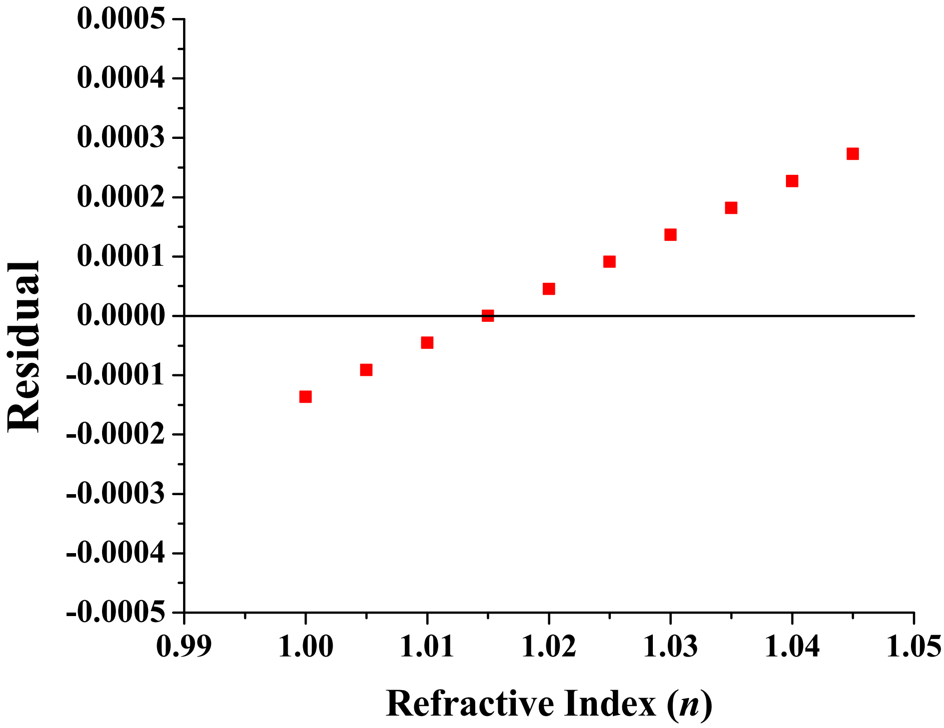
The ratio of resonance frequency alterations to the refractive index variations is called sensitivity. Figure 8 represents the sensitivity plot for the proposed sensor, and the magnitude of sensitivity comes out to be 2120 GHz/RIU, which can be determined using Equation (6), representing the equation of the sensitivity curve.

$$y = -2.120n + 5.02718 \quad (6)$$



(**a**)

Figure 8. *Cont.*



(**b**)

Figure 8. (**a**) Sensitivity plot for the proposed sensor (**b**) Residual Plot.

The proposed sensor's performance is again justified using Table 1, where we see that the sensitivity offered by our suggested model outperforms other designs.

The quantitative relationship representing the ratio between the sensitivity and the FWHM is known as the Figure of Merit (FoM) [15]. It is widely regarded as a very important metric to justify the performance of the sensors. The obtained FoM for our proposed model is 106. The higher magnitudes of Sensitivity, Q-factor, and FoM make our proposed design an outstanding refractive index sensor, which can be used for gas sensing applications.

4. Detection of Harmful Gases

Due to its impressive sensitivity, the proposed sensor can be employed to detect harmful gases in a practical scenario. An example of a very harmful gas, viz. chloroform, has been taken whose refractive index (relative) is $n = 1.0014$, taking vacuum as the reference medium ($n = 1$) at 298 K and 101.325 KPa pressure [32,44]. Chloroform ($CHCl_3$), an organic compound used as an organic solvent, poses a great danger when exposed to the human body because of its toxicity and ability to evaporate into the air quickly. Studies revealed that exposure to chloroform causes cancer [45]. Chloroform inhalation is very injurious to health and mainly affects the central nervous system of the human body, eventually leading to unconsciousness. Excess intake of chloroform can also cause hepatitis in some cases. Inhaling chloroform can induce severe acute poisoning. Inhaling intense chloroform vapor irritates mucosal membranes exposed to the air, particularly the nose and throat. Shortness of breath is also a possibility. Chloroform ingestion can result in severe acute toxicity. Gastrointestinal irritation, including abdominal pain, nausea, vomiting, and diarrhea, are some of the local consequences of chloroform intake. Hence, the detection of Chloroform gas is very important.

It is evident from Figure 9 that a frequency shifting of 0.004 THz exists between the absorption peaks as chloroform has a resonant frequency of 2.901 THz when the refractive index is increased to $n = 1.0014$, resulting in a sensitivity of 2.857 THz/RIU together with 142.50 as FoM magnitude. These lateral shifts in the absorption peaks for a minute change in the refractive index of the encircling medium (almost in the orders of 10^{-5})

make the proposed design an excellent refractive index sensor and can be used for sensing harmful gases.

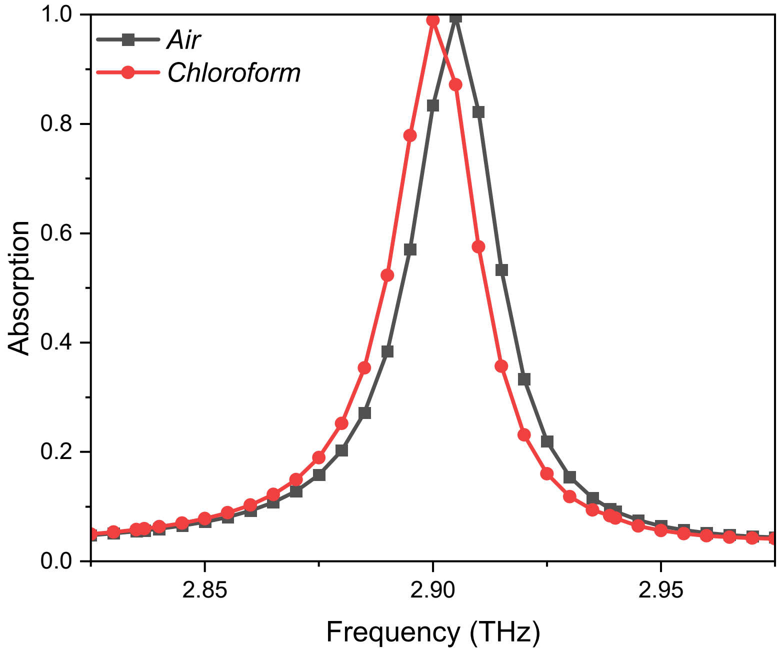


Figure 9. Absorption spectrum for refractive index of chloroform at resonant frequency.

5. Conclusions

A novel design of a terahertz metamaterial absorber has been proposed herein that uses a swastika-shaped resonator made of gold over a gallium arsenide substrate that acts as an MNG material on account of the negative-effective permeability in the resonance band. The proposed design produced an ultra-thin absorption peak with a slender bandwidth of 0.02 THz and an absorption rate of 99.65%, resulting in a high-quality factor of 145.25. It was found that the absorption peaks generate a significant lateral shift when the refractive index of the encompassing medium is altered slightly. Hence, the proposed model acts as an excellent refractive index sensor, giving rise to a sensitivity of 2120 GHz/RIU; thus, the figure of merit is 106. The suggested design is also polarization-insensitive, which adds an advantage to any design, considering the practical scenario. Due to its excellent performance, the proposed sensor has also been tested to detect poisonous gas like chloroform, producing high sensitivities of 2.857 THz/RIU for detection and effective absorption rates, quality factor, and figure of merit. Hence, our proposed model has high practical feasibility and can be used for applications involving sensing harmful gases.

The terahertz technology is still in its nascent stages, and the facilities for experimental demonstration are still expensive. So, most of the designs proposed in the literature are based on theoretical analysis done using similar software used in the study. Nevertheless, the authors will develop a working prototype in the future once the fabrication technology is economical. The authors intend to design highly sensitive topologies and incorporate machine learning for accurate specimen detection in their future work.

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