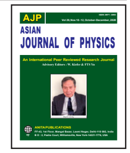
ap

Asian Journal of Physics

Vol. 29, Nos 10-12 (2020) 891-905

Available on: www.asianjournalofphysics.in



Thin film sensing with terahertz metamaterials

Dibakar Roy Chowdhury[1], Parama Pal[2], and Bishnu P Pal[1]

[1]*Department of Physics, Ecole Centrale School of Engineering - Mahindra University, Bahadurpally, Hyderabad-500 043, India*

[2]*TCS Research Labs, Bangalore-560 066, India*

This article is dedicated to Prof FTS Yu for his significant contributions to Optics and Optical information Processing

Plasmonic metamaterials-based sensing has generated a great deal of interests in recent years because of the relatively simple designs involved with metamaterials combined with strong field confinement attainable at the sub-wavelength scales. Normally high-quality factor resonance-based metamaterials are desirable to realize efficient meta sensors. In this paper, we have reviewed several metamaterials-based interesting schemes to design thin film sensors, in particular. We have also described the benefits and drawbacks of the reported sensing techniques. This review should be helpful to achieve smart designs of terahertz metamaterials-based sensors for exploitation with rich dividends.

Keywords: Metamaterials, Metasurfaces, Terahertz, Sensing, Thin films.

1 Introduction

The interaction of electromagnetic waves with structured/engineered artificial electromagnetic materials, also known as metamaterials has opened up unprecedented opportunities in photonics research. Such interactions enable manipulation of electrical permittivity and magnetic permeability, therefore making metamaterials extremely interesting for light matter interactions and photonics applications [1]. During last two decades metamaterials have become highly popular because of fascinating properties such as cloaking [2,3], negative refractive index [4], perfect focusing [5], sensing [6-22], and enabling realization of ultrafast devices [23-27], high speed communications [28], polarization manipulations [29-38] and electromagnetic wave modulations [39-51]. Typically, metamaterials operating in the terahertz regime are composed of metallic split-ring resonators (SRRs) fabricated on a dielectric substrate. Depending upon applications, numerous variations of split ring resonators are adopted by the researchers [52-62]. The biggest advantage of electromagnetic metamaterials lies in the fact that macro scale properties can be engineered through tuning at the micro level or in other words, at the basic unit cell level. This leads to unique and novel schemes in designing unit cells in order to manipulate the metamaterials properties when captured at the macro level. In other words, manipulating unit cell interactions at the near-field regime can result in effects at the far-field domain. In most of the cases, metamaterials functions most efficiently around the fundamental or higher order resonances that are excited by the probing waves. Applying intelligent design considerations at the SRR level can control the nature of the excited resonances because of several reasons. First, the major manipulations of electric and magnetic properties are mostly prominent around the fundamental resonances and second, strongest field confinements are observed at the resonances. Therefore, right from the beginning of metamaterials research, lot of efforts have been devoted to exploit these resonances for a host of applications. It involves novel designs based on single resonators as well as planar and broadside near-field coupled resonators [63-66].

Corresponding author :

e mail:bishnu.pal@mahindrauniversity.edu.in (Bishnu P Pal)

However, in this review paper, we will mainly focus on sensing based on terahertz metamaterials because of its unique ability to confine electromagnetic fields in a relatively small volume. We will discuss several sensing schemes that were demonstrated in last one and half decade involving metamaterials. Metamaterials exhibiting sharp resonances are normally considered as important candidate for sensing applications. Our discussion will not be confined within classical split ring resonators-based metamaterials. But we will extend our discussion to more complicated meta structures and will show how strong field confinement in metastructures can be leveraged to realize highly sensitive meta sensors. Metamaterials and its 2-dimensional variant metasurfaces form an excellent platform for thin film sensing mainly because of its planar nature. There are several advantages of thin-film sensing, particularly using metasurfaces operating at the terahertz regime. Thin films can also be sensed using typical terahertz spectroscopy, however, strong field confinements offered by terahertz metasurfaces, makes thin film detection much more efficient, therefore enabling sensing of minute amount of materials to be tested. This leads to practical sensing of biological materials, explosives, chemicals, hazardous materials, etc. All of these materials, if handled in large quantity, could lead to dangers for a human. However, metasurfaces can resolve this problem. Measurand materials in the form of thin films fit nicely in terms of compatibility within terahertz metasurfaces-based detection techniques.

The organization of this review article is as follows. Section 1 deals with introduction to metamaterials and thin film sensing. In section 2, we first explain basics of metasurfaces employed for thin film sensing, followed by several other meta sensing schemes. Finally, we will conclude with a discussion on the benefits and future prospects of the sensing schemes reviewed.

2 Thin film sensing using planar metamaterials

Possibility to detect dielectric materials utilizing classical metamaterials were first investigated by Drischoll *et al* in 2007 [6] (Fig 1). Strong electric field confinement inside the split gap region was explored for the first time to detect unknown materials. Split gap provides capacitance to the ring resonator. Therefore, presence of any external dielectric materials inside or in the vicinity of the split gaps can modulate the capacitive properties of the ring resonators, resulting in a shift in the resonance frequency. Such passive tuning was accomplished for the first time by Drishcholl *et al* through incorporation of dielectric material in the plane of SRRs. Typical single split gap ring resonators-based metamaterials when investigated with the probing THz radiation (Fig 1b) leads to the excitation of strong magnetic resonance in the transmitted terahertz spectrum (Fig 1c). In this pioneering work, they showed that presence of silicon nanoparticles can gradually tune metamaterials resonance when characterized in transmission mode. They attributed the observation to tuning of relative permittivity surrounding the resonators, which modifies the capacitance of the split gap. Further they demonstrated chemical sensing potential of the proposed scheme by characterizing benzocyclobutane (BCB) (Fig 1b). Several chemicals can be coated on top of metamaterials as thin films. In their study, different amount of BCB materials were coated, which resulted in shifting of the magnetic resonance frequency by variable extent. This formed the basis to determine dielectric properties through metamaterials resonance shift techniques.

In another breakthrough work, limitations in metamaterials-based thin film sensors were investigated by O'Hara *et al* [7]. In this work, authors employed double ring resonators-based metamaterials, which were characterized using Terahertz Time Domain Spectroscopy (TDS) (Fig 2a). Several resonance modes were excited due to the impinging terahertz beam when studied in the Thz-transmission mode (Fig 2a). These modes were originated from inner as well as the outer rings (Fig 2a). However, authors mainly focused on the fundamental and higher order dipole modes to study thin film detection capabilities. Fundamentally these modes were supported by the outer resonator. Photoresist of different thicknesses was coated on top of the metamaterial surface. Since photoresists can easily be removed using typical alcoholic solutions, the same meta structure was characterized in transmission mode for several coating thicknesses (Fig 2b, 2c and 2d).

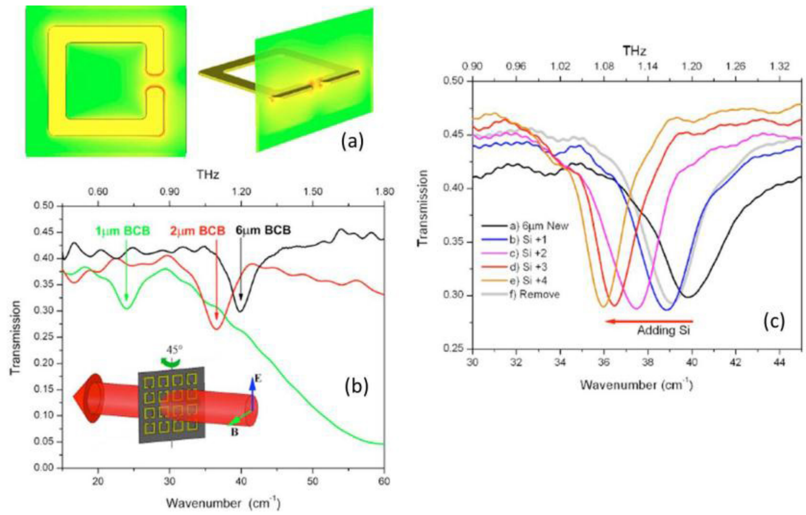


Fig 1. (a) (Top left) Variation of electric field profile in the plane of the split ring resonator and across the split gap; (b) Terahertz transmission spectra for metamaterial coated with different thicknesses of BCB (schematic shown as inset); (c) Terahertz transmission characteristics of metamaterials with different thicknesses of silicon nano particles [with permission from [6] © American Institute of Physics].

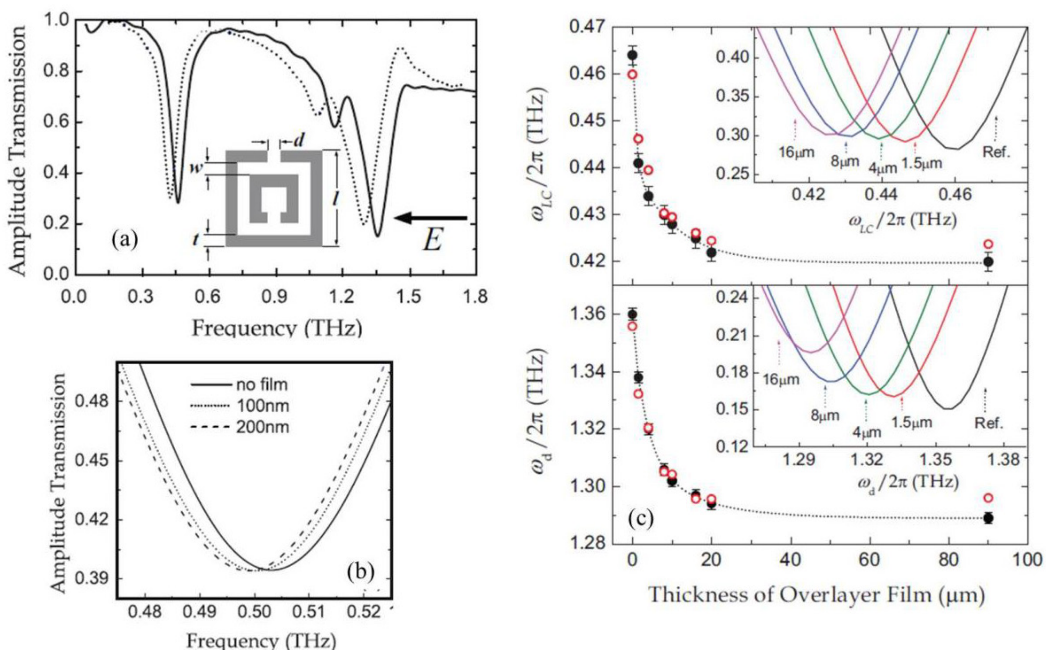


Fig 2. (a) (Top left) Transmission spectroscopy of double ring-based metamaterials, these data are generated using THz TDS, lower order (LC) resonances along with higher order modes are clearly excited in the spectrum, with coated overlayer, resonance red shifts as shown by the light color line; (b) (Bottom left) It shows the limit in detectability of fundamental resonance mode with over layer as thin as 100 nm; (c) It plots the shifts of LC and higher order modes with increasing over layer thickness, which clearly shows that beyond 20 μm thickness, resonance shift saturates, insets precisely show shifts in resonance dips with corresponding over layer thicknesses [with permission from [7] © The Optical Society].

With increasing thickness of photoresist coatings, resonances undergo red shifts because of enhanced effective dielectric constant in the vicinity of the split ring resonators. They further observed resonance shift saturates beyond a critical thickness that they attributed to dilution of fringing electric fields away from the metamaterial plane (Fig 2c & Fig 2d). This was one of the major outcomes of their study. Metamaterials were studied with electric polarization along the split gaps (Fig 2a). Materials as thin as 100 nm could be detected through resonance shift using THz TDS. On the other end, materials as thick as 16 µm demonstrated resonance shift, beyond this point resonance shift was observed to be saturated. This work formed a comprehensive basis to study the limitations of employing metamaterials as thin film sensing device while operating in the THz frequency regime.

Based on electric field polarization of probing terahertz beams, odd and even resonance orders can be excited in terahertz metasurfaces constituting single split gap-based resonators (Fig 3a). It was shown that even order resonance modes are excited for electric field parallel to side arms and odd order resonances are excited for electric field polarization parallel to the split gap [43]. In a recent work [19], sensing capabilities of these resonant modes were studied in a comprehensive manner. Loss-less dielectric materials of different thicknesses were coated over the metasurfaces and investigated in the transmission mode operating in the terahertz frequency domain. Further, refractive index of the coated films was varied to extract the sensitivity and figure of merit for different resonance modes. It was found that odd order mode resonances work better as sensors compared to the even order modes. Dimensions of the structures are derived in such a way that the lowest order resonance appears around 0.5 THz (Fig 3b). On top of the metasurface, analytes of several thicknesses are deposited. Terahertz transmission study is carried out simultaneously in order to capture the response of the samples under the influence of dielectric over layer (Fig 3a). Lowest odd order modes are excited around 0.5 THz (also called as fundamental mode or first order mode) and 1.5 THz (3rd order mode), respectively (Fig 3b). Similarly, lowest even order resonance mode is excited around 1.185 THz (Fig 3c). As dielectric over layer is applied on top of the metasurfaces, resonances undergo red shift because of the increase in effective permittivity surrounding the meta surface. In order to capture the comprehensive picture of resonance shifting, elaborate study was made for both the Electric field polarizations of the probing terahertz beam, results from which are shown in Figs 3d and 3e. It can be noted that resonance red shifts in a non-linear fashion with increase in over layer thickness. Initially resonance red shifts rapidly, however resonance shifting saturates with relatively thicker (> 12 µm) analyte layer (Figs 3d and 3e). As the analyte material fills the capacitive gaps and surrounding of the resonator array and increases in thickness above it, the net capacitance of the resonator effectively increases [34], which shifts the resonance peak progressively to lower side of the frequency spectrum. However, beyond a certain point away from the resonator surface fringing fields are almost negligible, therefore causing saturation in resonance red shift following earlier trends. In order to quantify sensing performance of a sensor, FoM (Figure of Merits) were further extracted. FoM is defined as sensitivity/full width at half maxima at resonance. Sensitivity is calculated at particular thickness by varying refractive index of the analyte from 1 to 4. This leads to a linear curve between the resonance frequency and refractive index, slope of which defines sensitivity of a meta sensor at that particular thickness. FoM calculated in such a way is plotted in Fig 3d. It can be seen that figure of merits also saturates with enhanced analyte thickness because of the same fringing field effects, as discussed earlier. An important outcome of this work is odd order resonance modes works better as a sensor in comparison to the even order modes. Precisely the lowest order mode can work as the best sensing device when compared to all the other resonance modes. Authors attribute this to stronger field confinement at the lowest order mode (also known as LC mode or magnetic mode) [6] (Fig 1). Strong electric field confinement inside the split gap region could be utilized to detect materials, which were investigated for the first time. Split gap provides capacitance to the ring resonator. Therefore, based on electric field polarization of probing terahertz beams, odd and even resonance orders can be excited in terahertz metasurfaces constituting single split gap-based resonators (Fig 3a). It was shown that even order resonance modes are excited for electric field parallel to side arms and odd order resonances are excited for electric field polarization parallel to the split gap [43]. In a recent work [19], sensing capabilities of these resonant modes were studied in a comprehensive manner. Loss-less dielectric materials of different thicknesses were coated over the metasurfaces and investigated in the transmission mode operating in the terahertz frequency domain. Further, refractive index of the coated films was varied to extract the sensitivity and figure of merit for different resonance modes. It was found that odd order mode resonances work better as sensors compared to the even order modes. Dimensions of the structures are derived in such a way that the lowest order resonance appears around 0.5 THz (Fig 3b). On top of the metasurface, analytes of several thicknesses are coated. Terahertz transmission study is carried out simultaneously in order to capture the response of the samples under the influence of dielectric over layer (Fig 3a). Lowest odd order modes are excited around 0.5 THz (also called as fundamental mode or first order mode) and 1.5 THz (3rd order mode), respectively (Fig 3b). Similarly, lowest even order resonance mode is excited around 1.185 THz (Fig 3c). As dielectric over layer is applied on top of the metasurfaces, resonances red shift because of the increase in effective permittivity surrounding the meta surface. In order to capture the comprehensive picture of resonance shifting, elaborate study was made for both the Electric field polarizations of the probing terahertz beam, results from which are shown in Figs 3d and 3e.

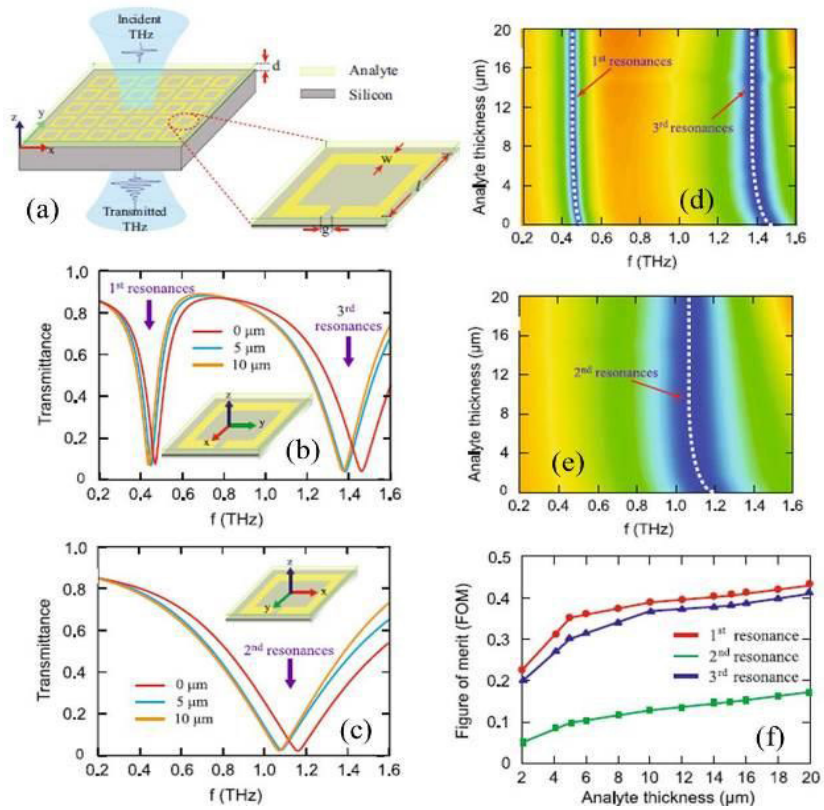


Fig 3. (a) Schematic of thin film sensing scheme with single split gap resonator-based terahertz metamaterials; (b) Demonstrates excitation of odd order modes along with resonance frequency shift with thin film over layer; (c) Excitation of even order mode along with resonance frequency shift due to over layer; (d) Comprehensive nature of resonance shift of odd order modes due to over layer film; (e) Resonance shift of even order mode due to thin film coated on meta surface; (f) Extracted Figure of Merit (FoM) for odd and even order resonance modes with increasing film thicknesses [with permission from [19] © Springer Nature].

It can be noted that resonance red shifts in a non-linear fashion with increase in over layer thickness. Initially resonance red shifts rapidly, however resonance shifting saturates with relatively thicker (> 12 µm) analyte layer (Figs 3d & 3e). As the analyte material fills the capacitive gaps and surrounding of the resonator array and increases in thickness above it, the net capacitance of the resonator effectively increases [34], which shifts the resonance peak progressively to lower side of the frequency spectrum. However, beyond a certain point away from the resonator surface fringing fields are almost negligible, therefore causing saturation in resonance red shift following earlier trends. In order to quantify sensing performance of a sensor, FoM (Figure of Merits) were further extracted. FoM is defined as sensitivity/full width at half maxima at resonance. Sensitivity is calculated at particular thickness by varying refractive index of analyte from 1 to 4. This leads to a linear curve between the resonance frequency and refractive index, slope of which defines sensitivity of a meta sensor at that particular thickness. FoM calculated in such a way is plotted in Fig 3d. It can be seen that figure of merits also saturates with enhanced analyte thickness because of the same fringing field effects, as discussed earlier. An important outcome of this work is odd order resonance modes works better as a sensor in comparison to the even order modes. Precisely the lowest order mode can work as the best sensing device when compared to all the other resonance modes. Authors attribute this to stronger field confinement at the lowest order mode (also known as LC mode or magnetic mode).

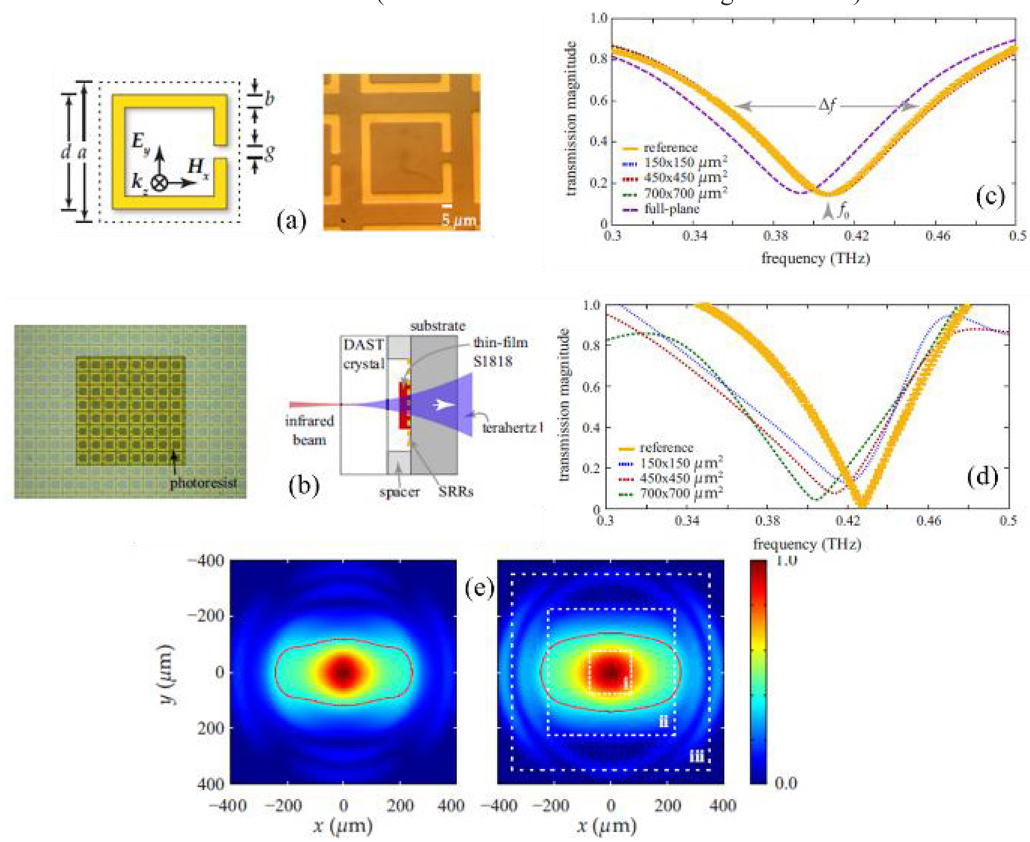


Fig 4. (a) Unit cell of single gap split ring resonators, schematic and optical photograph; (b) Near field terahertz characterization schematic and region of terahertz excitation on metasurfaces; (c) Resonance frequency shift due to coating of thin dielectric film measured in far field approximation; (d) Resonance shift measurements in near field excitation when different metasurface areas are excited with terahertz beam; (e) Numerical simulations of electric field magnitude of an impinging terahertz beam on metasurfaces have been carried out by assuming that the sample under test is placed only 50 µm away from the source [with permission from [16] © The Optical Society].

Although terahertz metasurfaces can detect thin deep sub wavelength thickness materials, however the sample area to be exposed depends on the terahertz spot size. This means with this technique, in principle, one can detect films as thin as several hundreds of nanometers. But the fact that the exposed area is of the order of several microns in diameters, it leads to an inherent limitation in thin film detection using terahertz metasurfaces. This problem was addressed by the researchers of University of Adelaide [16]. They demonstrated that using near field terahertz transmission spectroscopy, area to be sensed can dramatically reduce by more than 20 times, while still being able to sense similar material thickness. This can also be helpful in reducing volume of materials/analytes for testing significantly. Simple single gap split ring resonators array is employed to realize terahertz metamaterials for sensing purpose (Fig 4a). Instead of typical far field measurements, authors adopted near field characterization technique as shown in the schematic represented in Fig 4b. A comparison between well-established far field terahertz measurements and near field reduced active area terahertz measurements is shown in Figs (4c & 4d). Numerical simulations of excitation areas are demonstrated for near field probing in Fig (4e). It clearly shows that resonators within the reduced area metamaterials can indeed be excited effectively, which should help in thin film sensing. With this measurement technique, amount of materials sensed can be drastically reduced by an order of magnitude.

3 Thin film sensing with asymmetric metamaterials

Metamaterials consisting of double split gaps based asymmetric ring resonators turned out to be strong potential candidate for configuring thin film sensors in the terahertz regime because of its capability to excite high Q factor resonances. Therefore, during past several years trapped mode, Fano resonance, electromagnetically induced transparency effect etc. were demonstrated using asymmetric metamaterials. High Q resonance was observed for the first time in dual split gap resonators-based metamaterials [67]. Such metamaterials electromagnetically excited at a microwave frequency results in narrow, asymmetric resonance, which was termed as ‘Trapped mode’. These modes are excited through destructive interference that occurs between two split gap based asymmetric resonators. Effectively this phenomenon is similar to Fano resonance as observed in another work [68]. Trapped modes attracted lot of interests due to the strong confinement of incident electromagnetic waves and low radiation loss associated with it, which is realized through opposite magnetic dipoles excited inside the unit cell. Demonstration of such high Q factor modes motivated a great deal of interest because of potential applications in realizing ultra-sensitive sensors besides other possible applications. Because of such high potentials, several other works were carried out and reported later [68,69]. For the first time, asymmetric double split gap-based resonators were employed for thin film sensing by Ibraheem *et al* [9]. In this investigation, two different resonators were employed, both were double gap in nature. In one case they employed circular resonator with displaced gap from symmetric position (Fig 5a). In another configuration, strong field confinement was realized using tapered tip based split gaps (Fig 5b). Such tapered tips helped to confine strong electric field at the tips within a narrow region, hence establishing a way to realize highly sensitive sensors (Figs 5c & 5d). Figure 5e demonstrates that resonance frequency shifts, and consequent sensitivities are almost double for tapered tip-based rectangular resonator in comparison to a circular asymmetric resonator. Relatively stronger electric field confinement is attributable to enhanced sensitivity of the rectangular resonator, hence opens up a novel design scheme to realize improved thin film meta sensors.

Typically, Fano modes in terahertz metamaterials are attractive because of high Q factor. Another feature characterizing Fano mode is its asymmetric resonance line shapes. Enhanced Q factor results in increased photon lifetime in the meta resonator array leading to increased interaction of electromagnetic fields with the thin film placed on top of the meta surface. Therefore, series of research articles were reported to demonstrate higher Q factor metamaterials for terahertz domain [69-72]. Fano like resonances were demonstrated in rectangular asymmetric resonators-based metamaterials operating in the terahertz domains [12] (Fig 6a).

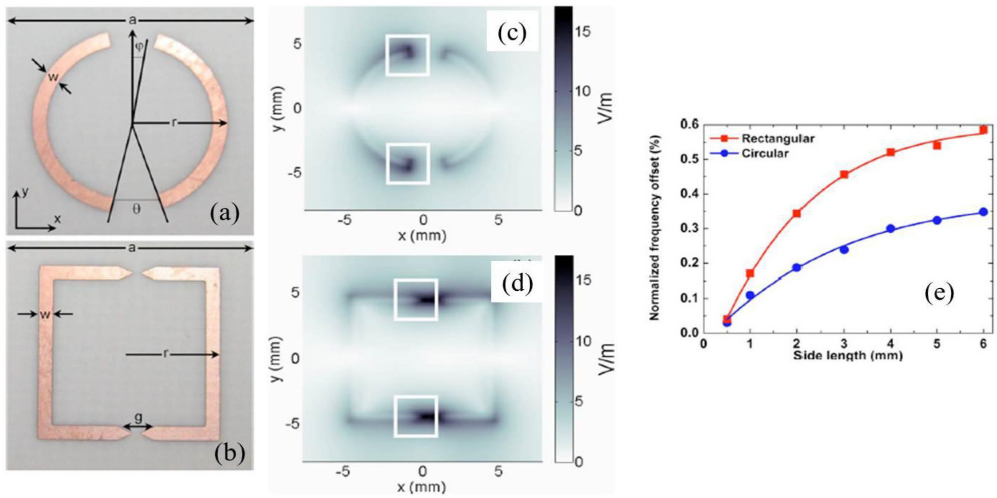


Fig 5. (a) (Top left) Optical picture of asymmetric circular double gap split ring resonator; (b) (Bottom left) Double gap sharp tip rectangular resonator; (c) (Top center) Electromagnetic simulations of electric field confinements at resonance in asymmetric gap split ring resonator; (d) (Bottom center) Simulated electric field enhancement adjacent to sharp tips in rectangular resonator; (e) (Far right) normalized resonance frequency shift for rectangular and circular resonator with materials on top [with permission [9] © American Institute of Physics].

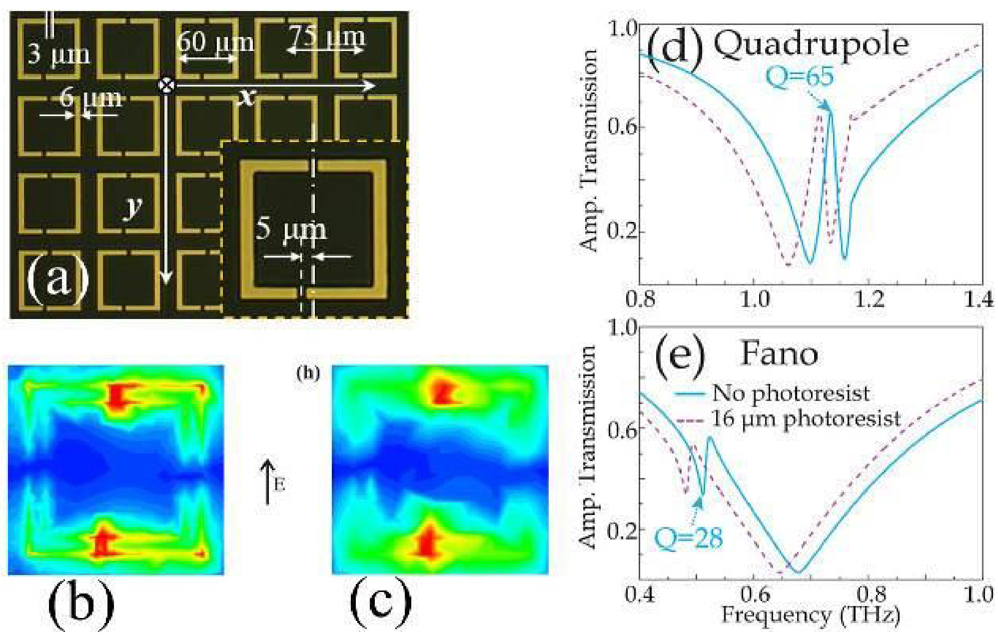


Fig 6. (a) Fabricated resonator array to realize terahertz metamaterials to demonstrate Fano resonance based thin film sensing; (b) Electric field distribution at Fano resonance without any dielectric over layer; (c) Simulated electric field distribution at Fano mode with 16 µm thick analyte on top of the meta surface; (d) Amplitude transmission with E field perpendicular to the splits gaps with and without analyte over layer; (e) Amplitude transmission with E field parallel to the split gaps with and without analyte overlayer [with permission [12] © American Institute of Physics].

Double split gap ring resonator was employed with split gaps appearing in the opposite arms of the resonator. Fano mode was excited while displacing one split gap along the arm but keeping the opposite gap intact at the middle. This leads to breaking the symmetry in the meta structure. With displacement of one gap by 5 μm from the symmetric position, strong Fano mode appeared for the E field of the THz beam parallel to the side arms. Figure 6b shows the E field confinement at the Fano mode without dielectric over layer and 6c shows the same with 16 μm overlayer. For the other polarization, quadrupole modes were excited as can be seen from Fig 6d. Figure 6e shows the transmission spectra with and without the dielectric over layers at the Fano resonance. It can be observed that a strong, high Q factor Fano resonance was excited. Employing similar technique as discussed earlier, sensitivity of different modes was found out. Fano mode demonstrated sensitivity as high as 36 GHz/RIU while quadrupole mode showed the same around 23 GHz/RIU. Therefore, Fano mode showed better potential as sensors, which is due to higher quality factor associated with it.

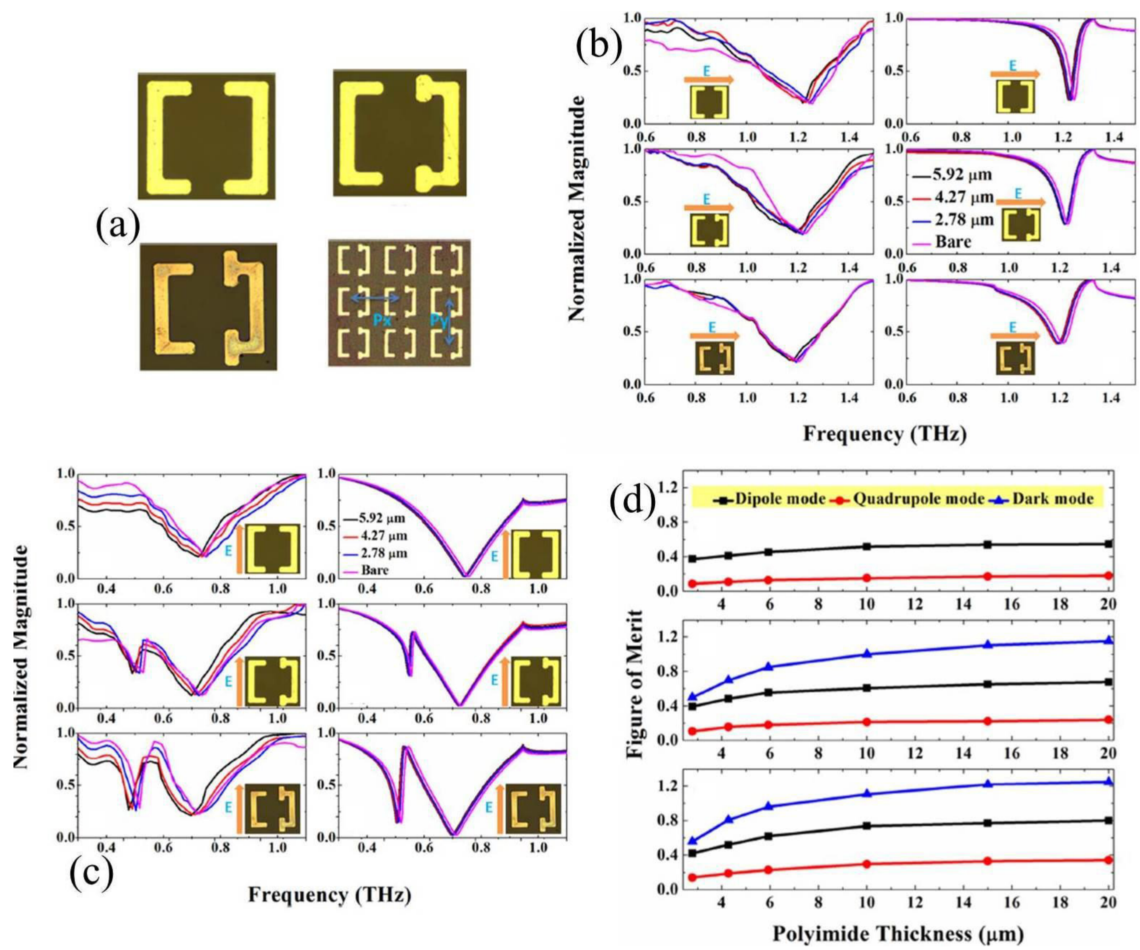


Fig 7. (a) Optical pictures of unit cell of symmetric and asymmetric metamaterials; b) experimental and numerically simulated transmission spectra of metamaterials for E field polarization parallel to split gaps, and (c) for E field polarization parallel to side arms; (d) extracted FoM values for all the three samples studied in this work, which captures the trend in FoM with increasing over layer thickness for all the three modes, dipole mode, quadrupole mode and dark mode [with permission [21] © Elsevier].

It is believed that symmetric plasmonic systems can possess dark resonance modes. These dark modes are normally originated from magnetic dipoles or higher-order electric multipoles. These modes can possess very high-quality factor resonances since they interact very weakly with the surrounding environment. In the context of terahertz metamaterials, such dark mode resonances were demonstrated through breaking the symmetry in rectangular double gap ring resonators-based unit cells. Although these modes are present in symmetric metamaterials but experimental observation is not possible unless asymmetry is introduced in such structures. Such asymmetric metamaterials can demonstrate high quality factor modes because of low radiation loses, and therefore can lead to realization of highly sensitive sensors in the terahertz domain. In this direction, thin film sensing based on dark resonant mode was reported very recently [21]. In this work, double split gap based rectangular resonators are used as metasurface unit cell (Fig 7a). Several sets of metasurfaces were fabricated and characterized where asymmetry is introduced through breaking the symmetry in the resonator structure (Fig 7a). Precisely, asymmetry is introduced via protrusion of a metal plate in one side of the both split gaps (Fig 7a). Terahertz TDS is performed through these samples in order to capture the plasmonic modes including the dark mode (Fig 7b and Fig 7c). For terahertz electric field parallel to split gaps, measured and simulated transmission characteristics are shown in figure 7b. For such excitation, quadrupole modes are excited at around 1.25 THz. Similarly, for electric field parallel to the side arms, transmission characteristics are shown in Fig 7c. Here dipole mode is excited around 0.7 THz. Importantly for the symmetric metasurfaces, only dipole mode is observed (see, Fig 7c top most panels). As an asymmetry is introduced, a sharp, asymmetric line shape mode started to appear around 0.5 THz. With increase in asymmetry, this novel mode strengthens. Q factor of this mode is relatively large (~ 12) compared to the other modes. With coating of thin dielectric films on top of the meta surfaces, modes underwent red shift. This is because of increase in effective refractive index, which was discussed earlier in details. Although all the modes red shifts but sensing performance of the modes are not same. Therefore, to elucidate this feature, FoM (Figure of Merit) is calculated for each resonant mode for all the three samples (Fig 7d). In the top most panel, FoM is shown for symmetric metasurfaces. Since dark mode cannot be excited, so FoM for dipole and quadrupole modes are only represented. Note that figure of merit is significantly low for these modes as demonstrated by the symmetric structure. In the next panel, FoM values are shown for asymmetric structures with inclusion of protrusion. FoM corresponding to dark mode are higher compared to the other modes. In the bottom most panel of Fig 7d, FoM values are shown for the metasurfaces with larger asymmetry. In this case, dark modes demonstrated higher FoM in comparison to the other modes. This study indicates that the dark resonant mode with highest Q factor demonstrates best platform as a sensing device.

4 Sensing with stacked metamaterials

Fano resonant metamaterials have garnered lot of attention recently because of its being a high-quality factor mode. Generally, Fano modes are observed in symmetry broken metamaterials. Very recently it is demonstrated that Fano resonances can also be excited in geometrically symmetric stacked terahertz metamaterials, where meta resonators are separated by an ultra-thin dielectric layer [73,74]. This allows additional degree of freedom to confine strong electromagnetic energy in deep sub-wavelength length scale [22]. This forms the basis to realize ultra-sensitive sensing through tailoring of the near field coupling inside the thin spacer region. Several sets of multi-layer metamaterial samples with variable spacer thicknesses were fabricated in order to study thin film sensing. Schematic of the proposed structures along with optical photograph of one fabricated sample is shown in Figs 8 (a & b). In Fig 8c and 8d, numerically simulated transmission spectra with the corresponding measurements are depicted. Spacer thicknesses of 2, 3, 4 and 6 μm were studied in this work. It is observed that with increasing spacer thickness both the resonances red shifts, however Fano mode shifts more rapidly compared to the dipole mode. The relatively stronger field confinement at Fano mode is responsible for the more rapid shift in resonance dip. In order to characterize the sensing potentials, sensitivities were calculated at several spacer thicknesses which were further followed by FoM calculation (Fig 8e). With increasing spacer, sensitivity saturates at around 8 μm thickness. It was further observed that around 8 μm, both the resonances (low and high frequency modes) possesses almost similar strength. Surface current analysis reveals that the higher mode is Fano but the lower one is a dipole mode. However, for the low frequency mode, bottom resonator is dominant whereas for the higher resonance mode the top resonator is dominant. This study was further extended to refractive index sensing through varying indices at wider range. As refractive index is varied from 1.4 to 2 in steps of 0.2, resonance modulation seen to be more rapid (Fig 8g). Calculation of sensitivities reveals its value as high as 1.04 THz/RIU. This is largest sensitivity reported in THz range for refractive index sensing, hence making this meta device an excellent platform for thin film sensing. Strong field confinement in between the stacked meta resonators based Fano cavity is responsible for such high sensitivities. Numerical simulations of electric fields indeed reveal high field confinement inside the spacer region.

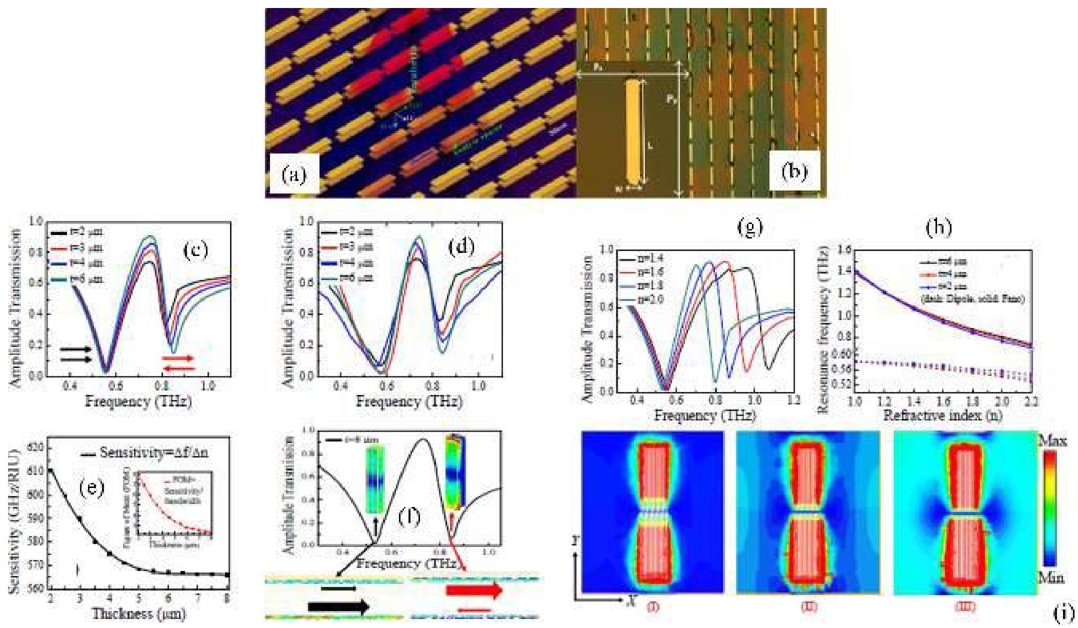


Fig 8. (a) Schematic of multi-layer dipole based terahertz Fano metamaterials; (b) Optical microscope pictures of unit cell along with array of resonators; (c) Numerical simulations of several thickness of spacers; (d) Experimental transmission spectrum of metamaterials; (e) Extracted sensitivities, inset shows the FoM values; (f) Spectrum for the 8 μm spacer thickness along with current and field distributions; (g) Scheme demonstrating refractive index sensing with transmission spectrum with changing refractive indices for various materials; (h) Extracted sensitivities for the Fano as well as dipole modes; (i) Electric field distributions inside the spacer in between the resonators [with permission Ref 73 © Wiley-VCH].

5 Conclusion

Terahertz metamaterials and metasurfaces offer a uμnique platform towards detection of materials in thin film form. Such metamaterials-based platform helps in detection of minute quantity of materials contrary to typical terahertz spectroscopy or ellipsometric techniques. Considering the unique advantages that terahertz metasurfaces provide towards thin film detection, we have focused on selective yet important works that are reported in the last 15 years. Normally, high quality factor resonances-based metamaterials are popular for thin film detection for better FoM. We have discussed several schemes to excite high Q factor resonances in terahertz metamaterials.

In this review, as a first example, we discussed thin film detection using typical single split ring resonators-based metamaterials operating in the THz domain. Magnetic resonance, also called as first order resonance mode, was investigated as a sensing platform. Presence of foreign materials in the vicinity of the split gap induce resonance shift because of modulation in effective dielectric constant. Measurement of such resonance shift can lead to sensing of the foreign materials. In the follow up work by O'Hara *et al* limitations of such frequency shift sensing technique was investigated. It is shown that resonance shift saturates beyond a certain thickness of the dielectric measurand, because of reduction in strength of fringing fields across the resonator split gap. This means beyond a certain thickness, thin film sensing is not achievable by this technique. In another work, sensing potentials of odd and even order resonance modes were investigated. It was shown that the odd order modes are always better sensor than the even order modes. So far, we discussed about single gap ring resonators-based metamaterials for sensor applications. Because of high quality factor resonances, asymmetric resonator-based terahertz metamaterials are also considered as strong candidate as sensors. Such asymmetric resonators were employed to detect thin films by several groups. In this context, we have discussed Fano resonance-based metamaterials as well as dark mode-based metamaterials for thin film sensing. These schemes of sensing were found to offer improved sensitivity as well as higher figure of merit sensors. We have also discussed tapered gap resonators for sensing. At the tip of the tapered gap ultra-strong field confinement was observed, therefore enabling sensing of very minute amount of materials. We believe that terahertz metametarials-based sensing is a matured field now and should be exploited by industries soon. Therefore, special attention should be given to commercial exploitation of thin film meta sensors in the near future.

Acknowledgement

Authors DRC and BPP would like to gratefully acknowledge the partial support from SERB, Department of Science and Technology, Government of India (CRG/2019/001656).

References

1. Pendry J B, Holden A J, Robbins D J, Stewart W J, Magnetism from conductors and enhanced nonlinear phenomena, *IEEE Trans Microw Theory Tech*, 47(1999)2075-2084.
2. Pendry J B, Schurig D, Smith D R, Controlling Electromagnetic Fields, *Science*, 312(2006)1780-1782.
3. Schurig D, Mock J J, Justice B J, Cummer S A, Pendry J B, Starr A F, Smith D R, *Science*, 314(2006)977-980.
4. Smith D R, Pendry J B, Wiltshire M C K, Metamaterials and Negative Refractive Index, *Science*, 305(2004)788-792.
5. Pendry J B, Negative Refraction Makes a Perfect Lens, *Phys Rev Lett*, 85(2000)3966; doi.org/10.1103/PhysRevLett.85.3966.
6. Driscoll T, Andreev G O, Basov D N, Palit S, Cho S Y, Jokerst N M, Smith D. R., Tuned permeability in terahertz split-ring resonators for devices and sensors, *Appl Phys Lett*, 91(2007)062511; doi.org/10.1063/1.2768300.
7. O'Hara J F, Singh R, Brener I, Smirnova E, Han J, Taylor A J, Zhang W, Thin-film sensing with planar terahertz metamaterials: sensitivity and limitations, *Opt Express*, 16(2008)1786-1795.
8. Alù A., Engheta N., Cloaking a sensor, *Phys Rev Lett*, 102(2009)233901; doi.org/10.1103/PhysRevLett.102.233901.
9. Ibraheem A Ibraheem Al-Naib, Christian Jansen, Martin Koch, *Appl Phys Lett*, 9(2008)083507; doi.org/10.1063/1.3553193.
10. Withayachumnankul W., O'Hara J. F., Cao W., Al-Naib I., and Zhang W., Limitation in thin-film sensing with transmission-modeterahertz time-domain spectroscopy, *Opt Express*, 22(2014)972-986.
11. Al-Naib I, Biomedical sensing with conductively coupled terahertz metamaterial resonators, IEEE J Selected *Topics in Quan Elec*, 23(2016)1-5.
12. Singh R, Cao W, Al-Naib I, Cong L, Withayachumnankul W, Zhang W, Ultrasensitive terahertz sensing with high-Q Fano resonances in metasurfaces, *Appl Phys Lett*, 105(2014)171101; doi.org/10.1063/1.4895595.

13. Gupta M, Srivastava Y K, Manjappa M, Singh R, Sensing with Toroidal Metamaterial, *Appl Phys Lett*, 110(2017)121108; https://doi.org/10.1063/1.4978672.
14. Withayachumnankul W, Jaruwongrungsee K, Tuantranont A, Fumeaux C, Abbott D, Metamaterial-based microfluidic sensor for dielectric characterization, *Sensors and Actuators A: Physical*, 189(2015)233-237.
15. O'Hara J F, Withayachumnankul W, Al-Naib I, A review on thin-film sensing with terahertz waves, *J Infrared Milli Terahz Waves*, 33(2012)245-291.
16. Withayachumnankul W, Lin H, Serita K, Shah C M, Sriram S, Bhaskaran M, Tonouchi M, Fumeaux C, Abbott D, Sub-diffraction thin-film sensing with planar terahertz metamaterials, *Opt Express*, 20(2012)3345-3352.
17. Li J, Shah C M, Withayachumnankul W, Ung B, Mitchell A, Sriram S, Bhaskaran M, Chang S, Abbott D, Flexible terahertz metamaterials for dual-axis strain sensing, *Opt Lett*, 38(2013)2104-2106.
18. Park S J, Hong J T, Choi S J, Kim H S, Park W K, Han S T, Park J Y, Lee S, Kim D S, Ahn Y H, Detection of microorganisms using terahertz metamaterials, *Sci Rep*, 4(2014)4988; doi.org/10.1038/srep04988.
19. Islam M, Rao S J M, Kumar G, Pal B P, Roy Chowdhury D R, Role of resonance modes on terahertz metamaterials based thin film sensors, *Sci Rep*, 7(2017)1-8; doi.org/10.1038/s41598-017-07720-9.
20. Shih K, Pitchappa P, Manjappa M, Ho C P, Singh R, Lee C, Microfluidic metamaterial sensor: Selective trapping and remote sensing of microparticles, *J Appl Phys*, 121(2017)023102; https://doi.org/10.1063/1.4973492.
21. Banerjee S, Amith C S, Kumar D, Damarla G, Chaudhary A K, Goel S, Pal B P, Chowdhury D R, Ultra-thin subwavelength film sensing through the excitation of dark modes in THz metasurfaces, *Opt Commun*, 453(2019)124366; doi.org/10.1016/j.optcom.2019.124366.
22. Karmakar S, Kumar D, Varshney R, Chowdhury D R, Strong terahertz matter interaction induced ultrasensitive sensing in fano cavity based stacked metamaterials, *J Phys D: Appl Phys*, 53(2020)415101; doi.org/10.1088/1361-6463/ab94e3.
23. Chen H-T, Padilla W J, Zide J M O, Bank S R, Gossard A C, Taylor A J, Averitt R D, Ultrafast optical switching of terahertz metamaterials fabricated on ErAs/GaAs nanoisland superlattices, *Opt Lett*, 32(2007)1620-1622.
24. Azad A K, Chen H-T, Kasarla S R, Taylor A J, Tian Z, Lu X, Zhang W, Lu H, Gossard A C, O'Hara J F, Ultrafast optical control of terahertz surface plasmons in subwavelength hole arrays at room temperature, *Appl Phys Lett*, 95(2009)011105; doi.org/10.1063/1.3168510.
25. Chowdhury D R, Singh R, Taylor A J, Chen H-T, Azad A K, Ultrafast manipulation of near field coupling between bright and dark modes in terahertz metamaterial, *Appl Phys Lett*, 102(2013)011122; doi.org/10.1063/1.4774003.
26. Lim W X, Manjappa M, Srivastava Y K, Cong L, Kumar A, MacDonald K F, Singh R, Ultrafast all-optical switching of germanium-based flexible metaphotonic devices, *Adv Mat*, 30(2018)1705331; doi.org/10.1002/adma.201705331.
27. Rao S J M, Kumar G, Azad A K, Chowdhury D R, Ultrafast relaxation of charge carriers induced switching in terahertz metamaterials, *J Infrared Millim Terahertz Waves*, 39(2018)1211-1220.
28. Shrekenhamer D, Rout S, Strikwerda A C, Bingham C, Averitt R D, Sonkusale S, Padilla W J, High speed terahertz modulation from metamaterials with embedded high electron mobility transistors, *Opt Exp*, 19(2011)9968-9975.
29. Fedotov V A, Mladyonov P L, Prosvirnin S L, Rogacheva A V, Chen L Y, Zheludev N I, Asymmetric Propagation of Electromagnetic Waves through a Planar Chiral Structure, *Phys Rev Lett*, 97(2006)167401; doi.org/10.1103/PhysRevLett.97.167401.
30. Yu N, Genevet P, Kats M A, Aieta F, Tetienne J-P, Capasso F, Gaburro Z, Light propagation with phase discontinuities: generalized laws of reflection and refraction, *Science*, 334(2011)333-337.
31. Ni X, Emani N K, Kildishev A V, Boltasseva A, Shalaev V M, Broadband Light Bending with Plasmonic Nanoantennas, *Science*, 335(2012)427-427.
32. Grady N K, Heyes J E, Chowdhury D R, Zeng Y, Reiten M T, Azad A K, Taylor A J, Dalvit D A R, Chen H-T, Terahertz Metamaterials for Linear Polarization Conversion and Anomalous Refraction, *Science*, 340(2013)1304-1307.
33. Zhou J, Chowdhury D R, Zhao R, Azad A K, Chen H-T, Soukoulis C M, Taylor A J, O'Hara J F, Terahertz chiral metamaterials with giant and dynamically tunable optical activity, *Phys Rev B*, 86(2012)035448; doi.org/10.1103/PhysRevB.86.035448.

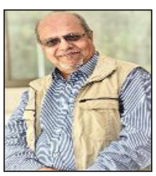
34. Wu C, Arju N, Kelp G, Fan J A, Dominguez J, Gonzales E, Tutuc E, Brener I, Shvetts G, Spectrally selective chiral silicon metasurfaces, *Nat Commu*, 5(2014)3892; doi.org/10.1038/ncomms4892.
35. Yang Y, Wang W, Moitra P, Kravchenko I I, Briggs D P, Valentin J, Dielectric meta-reflectarray for broadband linear polarization conversion and vortex generation, *Nano Lett*, 1(2014)1394-1399.
36. Wenwei L, Shuqi C, Zhancheng Li, Cheng H, Yu Ping, Li Jianxiong, Jianguo Tian, Realization of broadband cross-polarization conversion in transmission mode in the terahertz region using a single-layer metasurface, *Opt Lett*, 40(2015)3185-3188.
37. Guo T, Argyropoulos C, Broadband polarizers based on graphene metasurfaces, *Opt Lett*, 41(2016)5592-5595.
38. Rao S J M, Sarkar R, Kumar G, Chowdhury D R, Gradual cross polarization conversion of transmitted waves in near field coupled planar terahertz metamaterials, *OSA Continuum*, 2(2019)603-614.
39. Padilla W J, Taylor A J, Highstrete C, Lee M, Averitt R D, Dynamical electric and magnetic metamaterial response at terahertz frequencies, *Phys Rev Lett*, 96(2006)107401; doi.org/10.1103/PhysRevLett.96.107401.
40. Chen H T, Padilla W J, Zide J M O, Gossard A C, Taylor A J, Averitt R D, Active Terahertz Metamaterials Devices, *Nature*, 444(2006)597-600.
41. Zhang S, Genov D A, Wang Y, Liu M, Zhang X, Plasmon-induced transparency in metamaterials, *Phys Rev Lett*, 101(2008)047401; doi.org/10.1103/PhysRevLett.101.047401.
42. Reiten M T, Chowdhury D R, Zhou J, Taylor A J, O'Hara J F, Azad A K, Resonance tuning behavior in closely spaced inhomogeneous bilayer metamaterials, *Appl Phys Lett*, 98(2011)131105; doi.org/10.1063/1.3566978.
43. Chowdhury D R, Singh R, O'Hara J F, Chen H T, Taylor A J, Azad A K, Dynamically reconfigurable terahertz metamaterial through photo-doped semiconductor, *Appl Phys Lett*, 99(2011)231101; doi.org/10.1063/1.3667197.
44. Papasimakis N, Fedotov V A, Zheludev N I, Prosvirnin S L, Metamaterial analog of electromagnetically induced transparency, *Phys Rev Lett*, 101(2008)253903;
45. Chiam S Y, Singh R, Rockstuhl C, Lederer F, Zhang W, Bettiol A A, Analogue of electromagnetically induced transparency in terahertz metamaterial, *Phys Rev B*, 80(2009)153103; doi.org/10.1103/PhysRevB.80.153103.
46. Singh R, Plum E, Menzel C, Rockstuhl C, Azad A K, Cheville R A, Lederer F, Zhang W, Zheludev N I, Terahertz metamaterial with asymmetric transmission, *Phys Rev B*, 80(2009)153104; doi.org/10.1103/PhysRevB.80.153104.
47. Liu M, Hwang H Y, Tao H, Strikwerda A C, Fan K, Keiser G R, Sternbach A J, West K G, Kittiwatanakul S, Lu J, Wolf S A, Omenetto F G, Zhang X, Nelson K A, Averitt R D, Terahertz-field-induced insulator-to-metal transition in vanadium dioxide metamaterial, *Nature*, 487(2012)345-348.
48. Kuznetsov A I, Miroshnichenko A E, Fu Y H, Zhang J B, Luk'yanchuk B, Magnetic Light, *Sci Rep*, 2(2012)492; doi.org/10.1038/srep00492.
49. Chowdhury D R, Xu N, Zhang W, Singh S, Resonance tuning due to Coulomb interaction in strong near-field coupled metamaterials, *J Appl Phys*, 118(2015)023104; doi.org/10.1063/1.4926758.
50. Koshelev K, Lepeshov S, Liu M, Bogdanov A, Kivshar Y, Asymmetric metasurfaces with high-Q resonances governed by bound states in the continuum, *Phys Rev Lett*, 121(2018)193903; doi.org/10.1103/PhysRevLett.121.193903.
51. Devi K M, Islam M, Chowdhury D R, Sarma A K, Kumar G, Plasmon-induced transparency in graphene-based terahertz metamaterials, *Euro Phys Lett*, 120(2018)2700; doi.org/10.1209/0295-5075/120/27005
52. Yen T J, Padilla W J, Fang N, Vier D C, Smith D R, Pendry J B, Basov D N, Zhang X, Terahertz magnetic response from artificial materials, *Science*, 303(2004)1494-1496.
53. Shadrivov I V, Morrison S K, Kivshar Y S, Tunable split-ring resonators for nonlinear negative-index metamaterials, *Opt Exp*, 14(2006)9344-9349.
54. Azad A K, Dai J, Zhang W, Transmission properties of terahertz pulses through subwavelength double split-ring resonators, *Opt Lett*, 31(2006)634-636.
55. Azad A K, Taylor A J, Smirnova E, O'Hara J F, *App Phys Lett*, 92(2008)011119; doi.org/10.1063/1.2829791.
56. Liu N, Liu H, Zhu S, Giessen H, Stereometamaterials, *Nat Photon*, 3(2009)157-162.
57. Tao H, Strikwerda A C, Fan K, Padilla W J, Zhang X, Averitt R D, Reconfigurable terahertz metamaterials, *Phys Rev Lett*, 103(2009)147401; doi.org/10.1103/PhysRevLett.103.147401.

58. Shen X, Cui T J, Planar plasmonic metamaterial on a thin film with nearly zero thickness, *Appl Phys Lett*, 102(2013)211909; doi.org/10.1063/1.4808350.
59. Liu S, Noor A, Du L L, Zhang L, Xu Q, Luan K, Wang T Q, Tian Z, Tang W X, Han J G, Zhang W L, Zhou X Y, Cheng Q, Cui T J, Anomalous refraction and nondiffractive Bessel-beam generation of terahertz waves through transmission-type coding metasurfaces, *ACS Photonics*, 3(2016)1968-1977.
60. Karmakar S, Varshney R K, Chowdhury D R, Theoretical investigation of active modulation and enhancement of Fano resonance in THz metamaterials, OSA Continuum, 2(2019)531-539.
61. Rao S J M, Kumar D, Kumar G, Chowdhury D R, Modulating the near field coupling through resonator displacement in planar terahertz metamaterials, *J Infrared, Millimeter, and Terahertz Waves*, 38(2017)124-134.
62. Huang Y W, Chen W T, Wu P C, Fedotov V A, Zheludev N I, Tsai D P, Toroidal Lasing Spaser, *Sci Rep*, 3(2013)1237; doi.org/10.1038/srep01237.
63. Liu N, Guo H, Fu L, Kaiser S, Schweizer H, Giessen H, Three-dimensional photonic metamaterials at optical frequencies, *Nat Mater*, 7(2008)31-37.
64. Chowdhury D R, Singh R, Reiten M, Chen H-T, Taylor A J, O'Hara J F, Azad A K, A broadband planar terahertz metamaterial with nested structure, *Opt Express*, 19(2011)15817-15823.
65. Ekmekci E, Strikwerda A C, Fan K, Keiser G, Zhang X, Turhan-Sayan G, Averitt R D, Frequency tunable terahertz metamaterials using broadside coupled split-ring resonators, *Phys Rev B*, 83(2011)193103; doi.org/10.1103/PhysRevB.83.193103.
66. Chowdhury D R, Singh R, Taylor A J, Chen H-T, Zhang W, Azad A K, Coupling Schemes in terahertz planar metamaterials, *Int J Optics*, 12(2012)148985; doi.org/10.1155/2012/148985.
67. Fedotov V A, Rose M, Prosvirnin S L, Papasimakis N, Zheludev N I, Sharp Trapped-Mode Resonances in Planar Metamaterials with a Broken Structural Symmetry, *Phys Rev Lett*, 99(2007)147401; doi.org/10.1103/PhysRevLett.99.147401.
68. Singh R, Plum E, Menzel C, Rockstuhl C, Azad A K, Cheville R A, Lederer F, Zhang W, Zheludev N I, Terahertz metamaterial with asymmetric transmission, *Phys Rev B*, 80(2009)153104; doi.org/10.1103/PhysRevB.80.153104.
69. Singh R, Al-Naib I, Yang Y, Chowdhury D R, Cao W, Rockstuhl C, Ozaki T, Morandotti R, Zhang W, Observing metamaterial induced transparency in individual Fano resonators with broken symmetry, *Appl Phys Lett*, 99(2011)201107; doi.org/10.1063/1.3659494.
70. Luk'yanchuk B, N I Zheludev, Maier S A, Halas N J, Nordlander P, Giessen H, Chong C T, The Fano resonance in plasmonic nanostructures and metamaterials, *Nat Mat*, 9(2010)707-715.
71. Zheludev N I, Kivshar Y S, From metamaterials to metadevices, *Nat Mat*, 11(2012)917-924.
72. Khanikaev A B, Wu C, Shvets G, Fano-resonant metamaterials and their applications, *Nanophotonics*, 2(2013)247-264.
73. Karmakar S, Banerjee S, Kumar D, Kamble G, Varshney R K, Chowdhury D R, Deep-subwavelength coupling-induced Fano resonances in symmetric terahertz metamaterials, *Phy Stat Solidi: Rapid Research Lett*, 13(2019)1900310; doi.org/10.1002/pssr.201900310.
74. Karmakar S, Kumar D, Varshney R K, Roy Chowdhury D, Lattice-induced plasmon hybridization in metamaterials, *Opt Lett*, 45(2020)3386-3389.

[*Received*: 21.09.2020; *revised recd*: 01.10.2020; *accepted*: 14.10.2020]

Dr Dibakar Roy Chowdhury is currently Professor and Head of the Department of Physics at the Ecole Centrale School of Engineering in Mahindra University. His research interests span from electromagnetics, materials, to photonics and nanotechnology with special focus on studying light-matter interaction at the micro to nano scale levels. He has been working on design, simulations and fabrication of novel devices while simultaneously exploring their applications in information processing, sensing and energy related fields. He has extensively published his research in peer reviewed internationally reputed journals like Science, Applied Physics Letters, Scientific Reports, Optics Express, Physical Review B, Optics Letters etc. His research has been highlighted by several scientific magazines such as Worldwide Science, Phys Org, Nanotechnolgy News, Nanotechnology Now, Laser Focus World, Terahertz Technology, etc. His research contributions are also recognized by journals like Applied Physics Letters and Physica Status Solidi: Rapid Research Letters through reprinting of important results from his publications on the cover pages of certain issues of these journals. He is currently senior member of OSA and fellow of OSI and IETE.

Parama Pal is currently a Scientist with the CTO Labs, TCS. Prior to this, she has held positions in Robert Bosch Engineering and Business Solutions (RBEI) and the Robert Bosch Centre for Cyber Physical Systems at the Indian Institute of Science Bangalore where her directive was ‘Research and Translation for Impact’. She was a Research Fellow at the Wellman Center for Photomedicine, Harvard Medical School USA where her work focused on the design, development and translation of high-speed, minimally invasive, clinical imaging systems using optical techniques. She has an M.Sc. in Physics from IIT Delhi and Ph.D. in Optics from The Institute of Optics, University of Rochester, USA. Her research interests lie on femtosecond laser-based specialty fiber optic device fabrication, biophotonics, fiber optic sensors and more recently integration of AI and machine learning techniques for designing application-specific meta surfaces and meta structures.



Bishnu Pal is currently Professor of Physics and Dean of Academics at École Centrale School of Engineering in Mahindra University at Hyderabad India. He has been Former Chair of Physics Department (2007-2011) and Computer Services Centre at IIT Delhi (2003-2006) India, Member Board of Directors of OSA The Optical Society (Washington, USA, 2009-2011), Fellow of OSA the Optical Society (USA), SPIE The International Society for Optics and Photonics (USA), Distinguished Honorary Member of Royal Norwegian Academy of Sciences and Letters (DKNVS, Norway), Senior Member of IEEE (USA) and Distinguished Lecturer of IEEE Photonics Society (2005-2006), Distinguished Fellow and President of Optical Society of India (2012-March 2015), Alexander von Humboldt Fellow (1982-83, Germany) and Fulbright Scholar at NIST Boulder CO (1991, USA), PDF and latter Visiting Professor of Royal Norwegian CSIR (NTNF, Oslo, 1975-77) at Norwegian University of Science and Technology Trondheim Norway, awardee of Esther Hoffman Beller Medal of OSA The Optical Society (Centenary Year 2016, USA). His research areas span across silicon photonics, mid-IR and THz photonics, application-specific specialty optical fibers, all-fiber components and sensors.