Invited Paper

Novel materials in terahertz functional devices

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(Received February 26, 2020)

Abstract: Terahertz (THz) science and technology have been developed rapidly over the past decade due to its superiority in security, communication, imaging, and spectroscopy. In order to manipulate THz waves efficiently, many THz functional materials and devices have been proposed. Metasurfaces with subwavelength elements arranged in a periodic or quasi-periodic manner have been widely investigated. The amplitude, phase and polarization state can be controlled flexibly by designing the geometry. In this paper, several typical THz electromagnetic functional materials will be introduced, magnetic-optical semiconductors, nanoparticle liquid crystal, 3D graphene foam, carbon nanotubes, etc. These electromagnetic functional materials show unique functions for THz active modulation, polarization conversion, one-way transmission, and perfect absorption. Combined with these new materials, we designed and fabricated a series of THz metasurface device to enhance or expand the functions of these functional materials. Meanwhile, the introduction of functional materials brings THz metasurfaces into the active properties. The combination of these artificial micro-structures and electromagnetic functional materials bring new development for active or multifunctional THz devices.

Keywords: Terahertz devices, Nanomaterials, Magneto-optical materials, Liquid crystals

doi: https://doi.org/10.1051/tst/2020132041

1. Introduction

Terahertz (THz) wave has been widely used in imaging, spectroscopy, sensing, and communication with the rapid development of terahertz (THz) science and technology [1-3]. For further applications and developments of THz wave, THz functional devices such as THz modulators [4], filters [5], polarizers [6], phase shifters [7], and isolators [8] are essential. Therefore, to achieve active manipulation for THz amplitude, phase, polarization state, and wave front, it is necessary to find high-speed, low-loss THz functional materials and design novel structures, such as metasurface, photonic crystal, and plasmonics [9, 10].

In this paper, based on the research works over the last two years in our research group, we will introduce several typical THz electromagnetic functional materials, magnetic-optical semiconductors, nanoparticle liquid crystal, 3D graphene foam, carbon nanotubes, etc. These electromagnetic functional materials show unique functions for THz active modulation, polarization conversion, one-way transmission, and perfect absorption. Combined with these new materials, we have designed and fabricated a series of THz metasurface device to enhance or expand the functions of these functional materials. Meanwhile, the introduction of functional materials brings THz metasurfaces into the active properties. The combination of these artificial micro-structures and electromagnetic functional materials bring new development for active or multifunctional THz devices.

2. Semiconductor materials

The applied electric field, light field, magnetic field or thermal effect can effectively change the band structure and carrier characteristics of semiconductors, so as to change the transmission characteristics of materials to THz waves, and realize the effective manipulation and modulation of THz waves. In the THz regime, several common semiconductors, such as Si, Ge, GaAs, InSb, and HgTe, are used to implement THz sources, detectors and modulators.

2.1 Terahertz modulation enhanced by laser processed PVA film on Si substrate

An optically pumped ultrasensitive broadband terahertz (THz) wave modulator based on polyvinyl alcohol (PVA) film on Si wafer was demonstrated in this work. The THz time domain spectroscopy experiments confirm that the PVA/Si can drastically enhance the photo-induced THz wave modulation on the Si surface, especially when the PVA film is heated by a high-power laser. A modulation depth of 72% can be achieved only under 0.55W/cm^2 modulated laser power, which is superior significantly to the bare Si. The numerical simulations indicate that the laser processed PVA (LP-PVA) film increases the photo-generated carrier concentration on the Si surface in two orders of magnitude higher than that of bare Si. Moreover, the modulation mechanism and the dynamic process of laser heating on the PVA/Si have been discussed. This highly efficient THz modulation mechanism and its simple fabrication method have great application potentials in THz modulators [11].

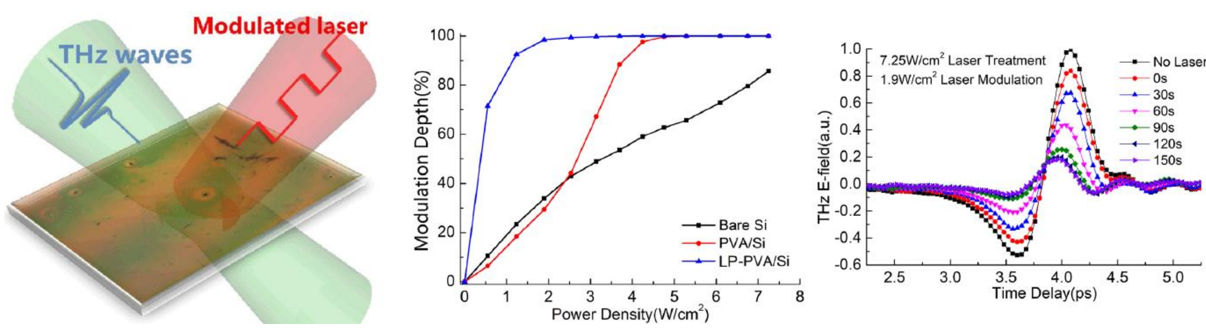

Fig. 1 laser processed PVA film on Si substrate for optically controlled THz modulation [11]

2.2 Terahertz magneto-optical polarization device based on InSb plasmonics

Researches on the magnetic properties of THz waves lag behind seriously. The unique nonreciprocal effect and magnetic tunability of magneto-optical (MO) device make it play an irreplaceable role in the high performance isolator, polarization controller, MO modulator, and magnetic field sensor, so it is necessary to fill the "terahertz gap" by not only electrical but also magnetic means. Due to the lack of high performance THz MO materials and the limitation of device fabrication, the improvement of THz MO devices is still in challenge [12].

The non-reciprocal circular dichroism and Faraday rotation effect for terahertz (THz) waves in longitudinally magnetized InSb have been investigated by the theoretical and experimental studies in the THz regime, which indicates its ability for THz circular polarized isolator, THz circular polarizer, tunable polarization converter, and polarization modulator by the manipulation of the different magnetic fields. Furthermore, we have demonstrated the InSb plasmonics based on its magneto-optical effects combined with artificial microstructure. We have found the magneto-optical enhancement mechanisms in this magneto plasmonic structure, achieving broadband perfect orthogonal linear polarization conversion modulated by the weak magnetic field in the experiment. Moreover, the magneto-optical modulation with modulation depth of 95% and one-way transmission with the isolation of 33dB can be achieved by this device under a weak magnetic field of 150mT. InSb and its magnetoplasmonic device have broadly potentials for THz isolator, magneto-optical modulator, and polarization convertor in the THz application systems [13].

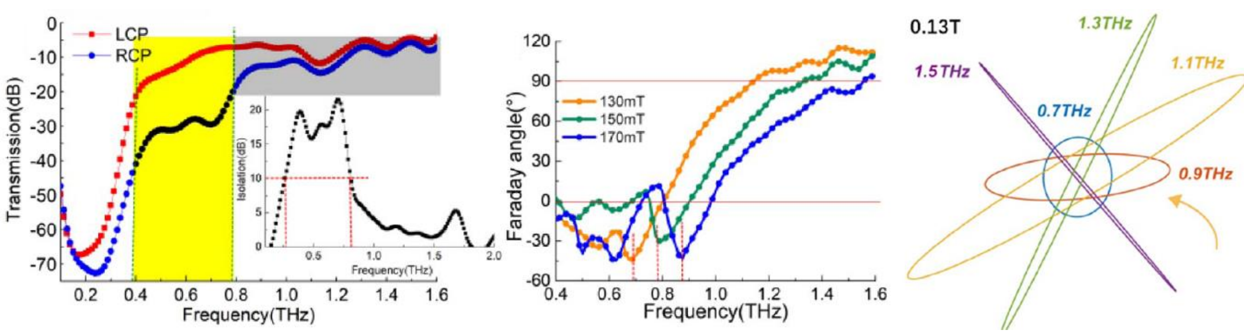

Fig. 2 THz circular dichroism and Faraday rotation effect in longitudinally magnetized InSb [13]

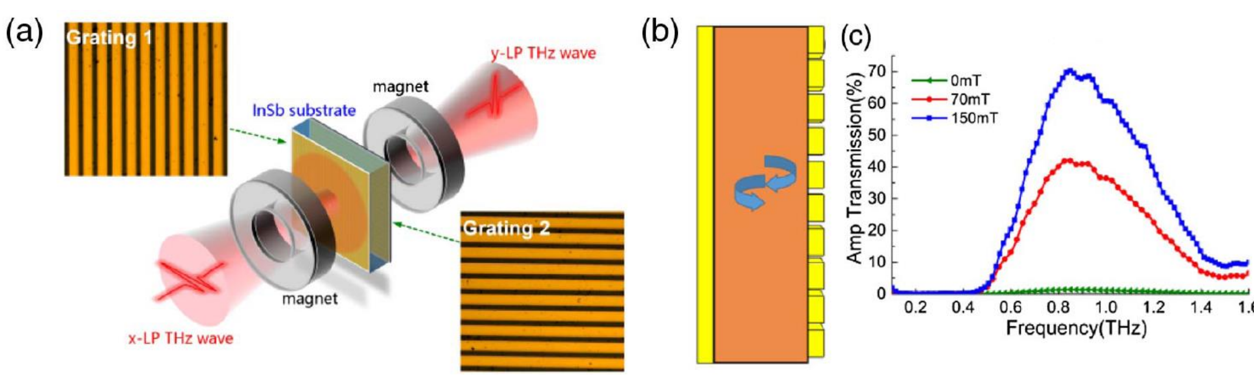

Fig. 3 Magneto-optical polarization rotation and modulation for THz waves in InSb plasmonics [13]

3. Liquid crystals

In the visible or near-infrared ranges of the electromagnetic spectrum, liquid crystals (LCs) have become one of the most important solutions for tunable phase and polarization control devices, which can be flexibly controlled by thermal, optical, electrical or magnetic field. The optical properties of LCs in the THz regime have been extensively studied in recent years, for example: Vieweg *et al.* [14] investigated the nematic mixture BL037 with high terahertz birefringence about 0.20 from 0.3 to 2.5 *THz*. Pan *et al.* [15] reported the extraordinary and ordinary indices of 5CB are n_e=1.77 and n_o=1.58, giving rise to a birefringence of 0.20±0.02 in the frequency range of 0.2-1.0 *THz*. Compared with the LC device for visible and near-infrared light, the birefringence coefficient of the currently existing LCs determines that the required thickness of the THz LC cell should be several hundred micrometers to millimeters to get enough phase shifts or modulation range (e.g., π/2 or π phase retardation), leading to several disadvantages such as poor pre-alignment, high operating voltage, and slow response. large cell gaps have been introduced, so that the LC molecules in the middle of cell are not aligned well because of the weak ability of alignment in a thick LC cell for either the rubbing-alignment or photo-alignment. These bottlenecks hinder the development of electrically controlled THz LC devices.

3.1 Terahertz liquid crystal phase shifter doped by magnetic nanoparticles

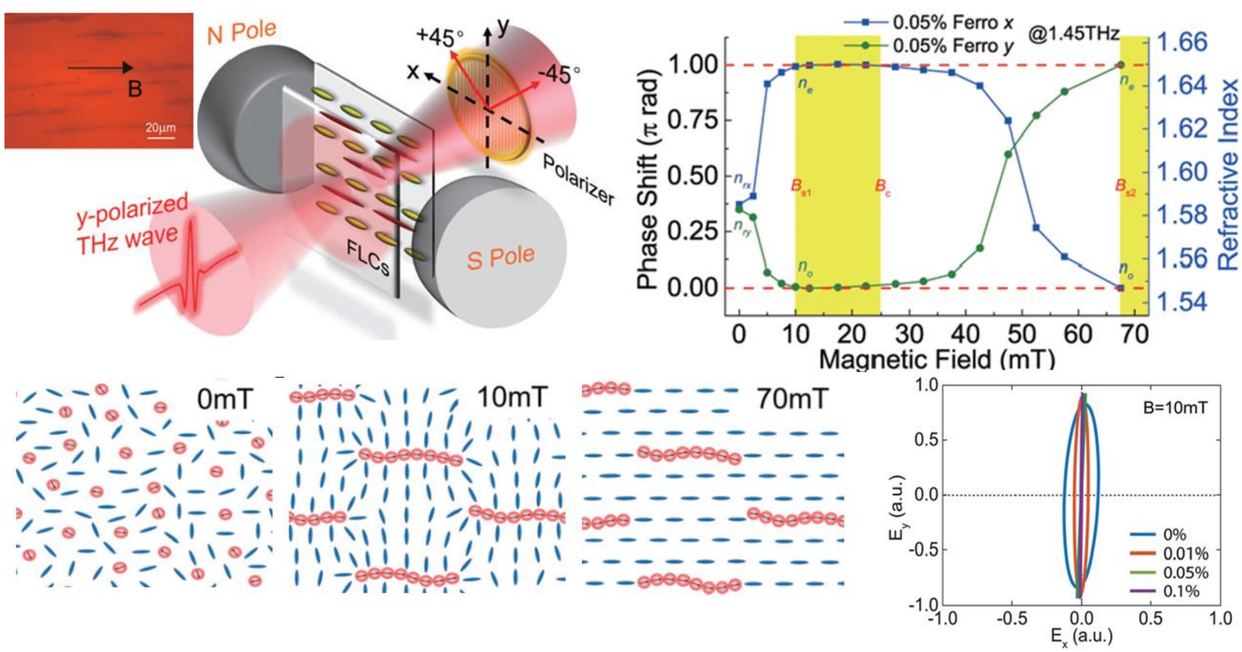

Fig.4 THz LC phase shifter doped by magnetic nanoparticles: device structure and mechanism [16]

Ferromagnetic liquid crystals (FLCs), the suspensions of ferromagnetic nanoparticle dispersed at different concentrations in liquid crystals (LCs), and its special magnetically induced birefringence characteristics has been investigated in the terahertz regime, mainly focusing on the interaction between magnetic cluster chains and LC molecules. We have experimentally demonstrated the anchoring effect of the magnetic cluster chains on LC molecules in an mm-thick LC cell under an extremely weak EMF, which leading to a uniform anchoring arrangement of the LC molecules in the entire LC cell. Unlike pure 5CB LCs, the phase shift range of the FLCs at 1.45 *THz* up to π (n_o to n_e or n_e to n_o) can achieve over the whole tunable range by simply changing the magnitude of the external magnetic field without changing its direction, and the optical axis of LC molecules can be controlled to rotate by 90°, thereby realizing a tunable THz wave plate. This work provides a new way in the development of THz magneto-optic devices and phase devices [16].

3.2 Terahertz dual-frequency liquid crystal doped by carbon nanotubes

Dual-frequency liquid crystals (DFLCs), the mixture of positive and negative liquid crystal (LC) molecules, exhibit unique alternating (AC) frequency dependent anisotropic properties. Carbon nanotube (CNT), as a novel nanomaterial with strong anisotropy, has attracted much attention in recent years. Herein, we have investigated the tunable terahertz birefringence and phase shift characteristics of the DFLC doped with CNT (CNT-LC) by using the terahertz time-domain polarization spectroscopy. The results show that the CNT-LC (1.5 wt%) can reach 0.5π at 0.793 *THz*, which can be used as a tunable THz phase shifter that is 0.12π higher than the pure DFLCs. Furthermore, through measuring the output polarization state, it is confirmed that dielectric anisotropy enhancement mechanism of CNT-LC originates from the surface interaction between CNTs and LC molecules. Therefore, a tunable quarter-wave plate with the active polarization conversion from LP to LP or LP to CP can be realized at 0.925 *THz* in the CNT-LC, while the pure DFLC cannot be achieved at the same frequency. The dielectric anisotropy enhancement of CNT-LC shows its utility in the improvement of various tunable terahertz LC phase shifter and wave plate [17].

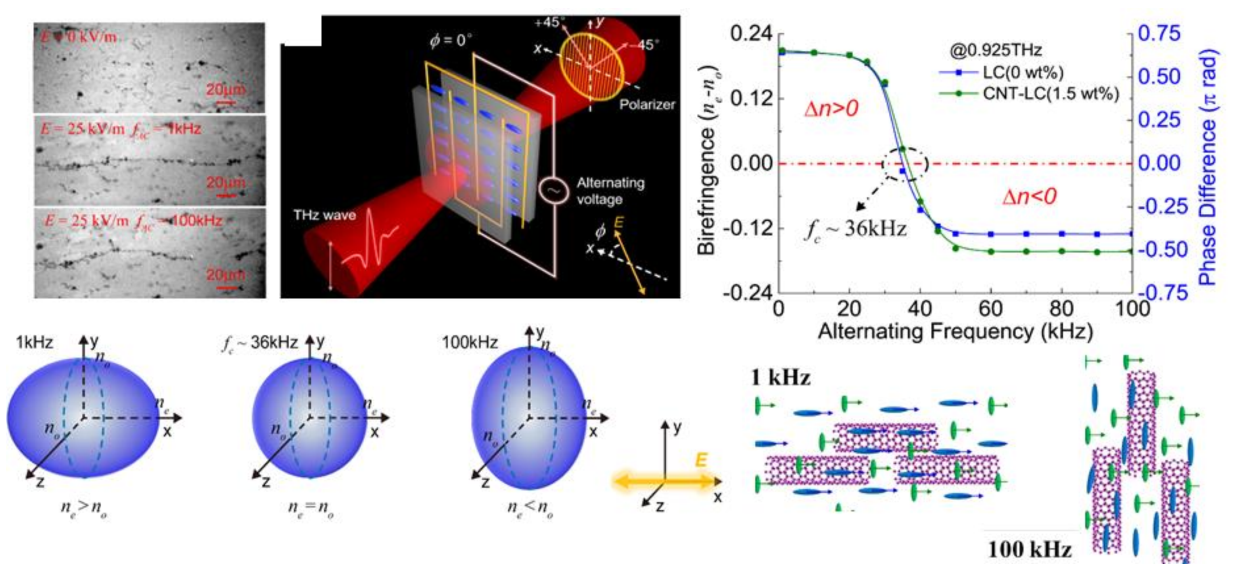

Fig. 5 DFLC doped by carbon nanotubes for THz phase shifter: device structure and mechanism [17]

3.3 Terahertz resonance switch based on liquid crystal in compound metasurface

We experimentally demonstrate an active terahertz (THz) resonance switch induced by the polarization conversion in a compound metasurface, which is a LC layer sandwiched by a metallic wire grating and resonance metamaterial (LCGM). Here, the liquid crystal (LC) plays the role of polarization conversion, which can induce the TE resonance. Moreover, there exists a localized resonance between metallic grating and metamaterial layers, and then the excited resonance will be greatly enhanced. The results show that the high extinction ratio of the resonance switch exceeds 30 *dB* at 0.82 *THz*. This work will bring new ideas for the research in developing THz phase, polarization and switch devices with LC and metasurface [18, 19].

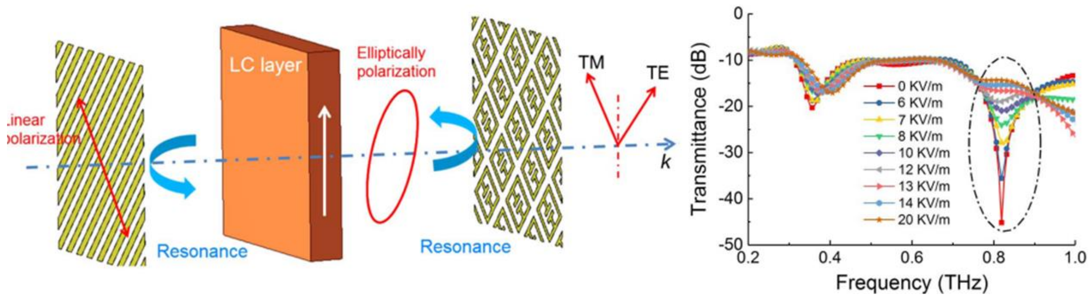

Fig.6 Mechanism and results of THz switch based on LC in compound metasurface[18]

4. 2D materials

The combination of 2D materials (*e.g.* graphene, carbon nanotubes, and MoS_2) and a metasurface holds great promise for dynamic manipulation of the electromagnetic wave from low terahertz to mid-infrared. The optical response of graphene is significantly enhanced by the highly-localized fields in the meta-atoms, and the characteristics of meta-atoms can in turn be modulated in a large dynamic range through electrical doping of 2D materials.

4.1 Mechanical Terahertz modulation based on buckled carbon nanotube sheets

Carbon nanotubes (CNT), especially super-aligned carbon nanotube sheets (SACNTS) formed by parallel arrays of CNT, have unique ultra-broadband anisotropic optical properties [27-29]. In particular, they exhibit essentially excellent polarization anisotropy in the THz regime, ideally suited for THz polarizer applications. Here, we present a mechanically tunable THz polarizer using stretchable buckled carbon nanotube sheets on natural rubber substrate (BCNTS/rubber). The transmittance and degree of polarization of THz wave can be modulated by stretching the BCNTS/rubber. The experiments showed that the degree of polarization increased from 17% to 97%, and the modulation depth reached 365% in the range of 0.2-1.2 *THz*, as the BCNTS/rubber was stretched from 0% to 150% strain. These changes can be also used for highly strain sensing up to 150% strain, with a maximum sensitivity of 2.5 *M/S*. And a spatial modulation of THz imaging was also realized by BCNTS/rubber via stretching and rotating. The theoretical analysis and numerical modeling further confirm the BCNTS/rubber changes from weak anisotropic to highly anisotropic structure, which play key roles in THz wave modulation. This approach for active THz wave manipulation can be widely used in polarization imaging, wearable material for security and highly sensitive strain sensing [20].

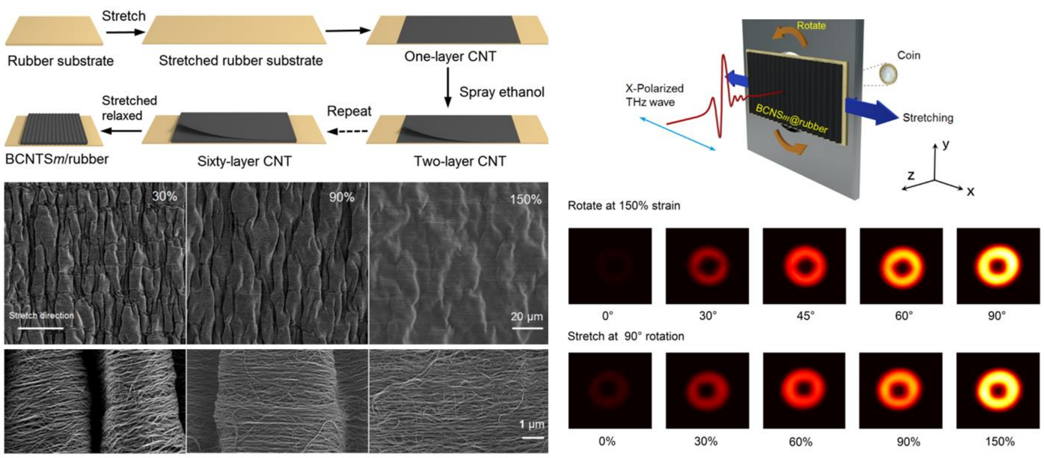

Fig. 7 Terahertz imaging modulated mechanically based on buckled carbon nanotube sheets [20]

4.2 Terahertz polarization conversion based on subwavelength grating with carbon nanotube

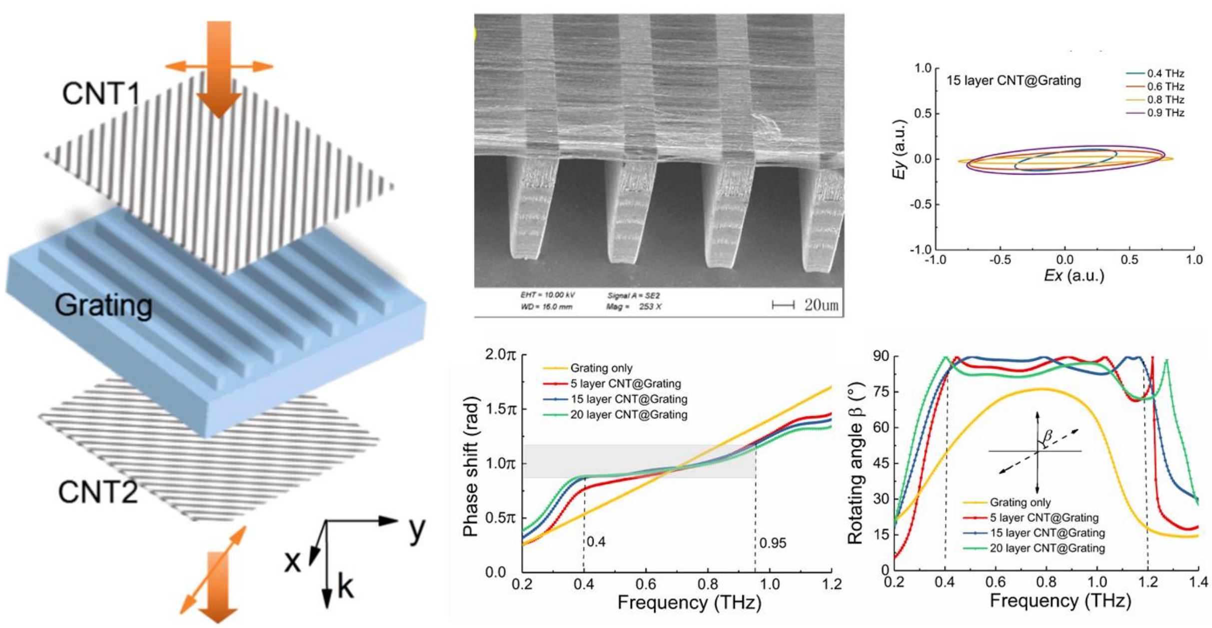

Fig. 8 THz polarization conversion based on subwavelength grating with CNTs [21]

Carbon nanotube (CNT), as a recently emerged newfangled nanomaterial, has been applied in terahertz (THz) polarizer due to its anisotropic hyper-ordered orientation. Herein, we simply adhere two orthogonal CNT sheets to the both sides of dielectric subwavelength grating to form a compound structure (i.e. CNT@Grating). Due to the subwavelength integration and cavity modes, the CNT@Grating presents local resonances between two CNT sheets, which greatly enhances the polarization rotation and expands the bandwidth. Compared with the large phase shift dispersion of single subwavelength grating, its phase shift dispersion and impedance can be manipulated by adjusting the layer number of CNT, and finally a broadband close zero dispersion from 0.4 to 0.95 *THz* has been obtained, which leads to a broadband THz polarization conversion. This work provides a new design idea towards practical applications for THz broadband polarization conversion and dispersion control [21].

4.3 Active terahertz shielding and absorption based on graphene foam

Ultralight materials for broadband terahertz (THz) shielding and absorption are promising in practical THz applications. Here, active THz shielding and absorption properties of 3D graphene foam (GF) controlled by both laser pumping and biased electric field have been investigated. The GF can be tuned from OFF-shielding state to ON-shielding state when the external field excitations are applied, and 10 *dB* shielding bandwidth expands from 0 to a broad band of 0.2-1.6 *THz*. Further researches show that the GF always keeps very low THz reflection either with or without external fields, but its absorption characteristics can be remarkably controlled from 13% to 95.4% at 0.3 *THz* by the power of the external excitations, and its specific average terahertz absorption performance increases from 3.9×10^{3} to 1.95×10^{4} $dB{\cdot}cm^{3}{\cdot}g^{-1}$. This modulation mechanism is revealed that the carrier density in GF increases one order of magnitude from 2.6×10^{14} cm^{-3} to 3.15×10^{15} cm^{-3}. Finally, the tunable THz shielding and absorption characteristics of this GF device have been demonstrated by THz transmission imaging, which shows its great potential applications in active THz imaging, radar, and electromagnetic compatibility [23].

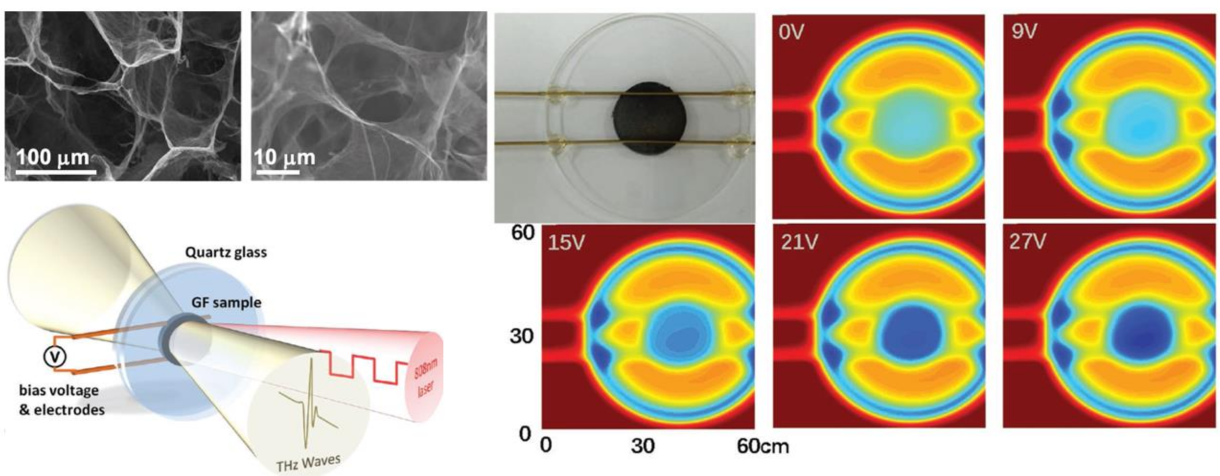

Fig. 9 THz imaging of shielding and absorption based on graphene foam controlled by *DC* electric field [22, 23]

5. Conclusion

In summary, several typical THz electromagnetic functional materials will be introduced. These electromagnetic functional materials show unique functions for THz active modulation, polarization conversion, one-way transmission, and perfect absorption. Combined with these new materials, a series of THz metasurface devices are designed and fabricated to enhance or expand the functions of these functional materials. The combination of these artificial micro-structures and electromagnetic functional materials bring new development for active or multifunctional THz devices.

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