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Recent Progress of Two-Dimensional Materials for Terahertz Protection

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Abstract

With the wide applications of Terahertz (THz) devices in future communication technology, the THz protection materials are essential to overcome potential threats. Recently, THz metamaterials (MMs) based on two-dimensional (2D) materials (e.g., graphene, MXenes) have been extensively investigated due to their unique THz response properties. In this Review, the THz protection theories are briefly presented firstly, including reflection loss and shielding mechanisms. Then, the research progress of graphene and other 2D materials based THz MMs and intrinsic materials are reviewed. The MMs absorbers in the forms of single layer, multiple layers, hybrid and tunable metasurfaces show excellent THz absorbing performance. These studies provide a sufficient theoretical and practical basis for THz protection, and the superior properties promised the wide application prospects of 2D MMs. The three-dimensional intrinsic THz absorbing materials based on porous and ordered 2D materials also show exceptional THz protection performance, which effectively integrate the advantages of intrinsic properties and structural characteristics of 2D materials. These special structures could optimize the surface impedance matching, and form THz multiple scatterings and electric transmission loss, which could realize high-efficiency absorption loss and active controllable protection performance in ultra-wide THz waveband. Finally, the advantages and existing problems of current THz protection materials are summarized, and the possible future development and applications are prospected.

Keywords: 2D materials; Terahertz; THz shielding; THz absorption
1. Introduction

As the most possible waveband for next generation communication (6G) applications, the THz waves have attracted extensive attention [1-4]. However, for the potential threats of this unknown waveband to human health, electromagnetic interference, information security and military stealth, the essential THz protection materials should be fully studied before the large-scale application. To achieve effective reflection loss or transmission shielding of the surplus THz energy, the THz protection materials are mainly THz absorbing and shielding materials. From the view of the interaction between electromagnetic (EM) waves and materials, the response mechanism of natural materials is not continuous across the whole spectrum. Within hundreds of gigahertz waveband, the electrons dominate the response process to the EM wave. And in the infrared to ultraviolet waveband, the EM wave mainly interacts with photons of the materials. However, between the two response areas, the 0.1 - 10 THz band is a “terahertz gap” which lacks material response, so it is an important topic to develop effective THz protection materials [5, 6].

Two-dimensional (2D) materials such as graphene and MXene, due to their high conductivity, ultra-thin, large specific surface area, ultra-lightweight, high strength and adjustable electromagnetic response, have shown excellent microwave attenuation performance [7-9], which encourage researchers to realize their THz protection abilities [10-13]. However, single atomic layered 2D materials could not absorb the incident THz energy effectively, and simply stacked materials would lead to impedance mismatching [14, 15]. Therefore, in order to make full use of the advantages of 2D materials, it is very important to reasonably design the form of 2D materials in THz protective materials, such as being patterned as the resonance unit of MMs absorbers and forming nanostructured materials [13, 16-18]. Since MMs have been proved to be able to respond to THz radiation [5] and fully absorb microwave energy [19, 20], the THz MMs are promised to be perfect THz absorbers. MMs are artificial EM dielectric materials, and usually composed of a MMs layer (periodically repeated patterned high conductivity materials), a dielectric spacer and a metal ground layer. Electrons resonated in these subwavelength scale unit structures can form THz absorption [21], so the THz response of the MMs can be controlled by designing the unit structure and component materials. Similar to traditional metal resonant MMs layers [22-27], coherent and patterned graphene 2D materials
applied as the MMs layers have been widely studied, because of their high conductivities and unique EM responses (such as Surface Plasmon Polaritons (SPPs)) \cite{17, 18, 28-30}. Otherwise, for the single atomic layer thickness and tunable chemical potential, 2D materials are easy to composite with other materials and their EM properties can be controlled by external stimuli. Recently, by optimizing the composite structures and active control approaches of the MMs, many THz protection MMs with a wide response band and adjustable absorption performance have been designed \cite{20, 31, 32}.

Besides 2D materials based THz protection MMs, the nanostructured 2D materials are another effective THz protection materials in the form of intrinsic loss \cite{10, 16, 33}. The enhanced absorption of the nanostructured materials is mainly contributed by the porous structure and long distance conductive networks. On the one hand, the porous structure ensures proper effective permittivity, which give a good impedance match to reduce the surface reflection. On the other hand, the cross-linked 2D materials can generate large surface induced current, so that the THz radiation rapidly decay and turn into heat in the resistance network. Furthermore, by compositing the nanostructured 2D materials with other THz loss materials, the THz response properties would be further regulated to achieve a wider effective waveband and stronger attenuation capability\cite{16}.

In this Review, we focus on recent progresses of THz protection materials based on 2D materials, including THz protection MMs and intrinsic loss materials, as shown in Fig. 1. The 2D materials discussed here are mainly graphene and MXenes, along with phosphorene and other 2D materials. Firstly, the THz protection theories are briefly presented, including reflection loss and shielding mechanisms. Secondly, the research progresses of graphene and other 2D materials based THz protection MMs and intrinsic materials are reviewed. Finally, the advantages and existing problems of current THz protection materials are summarized, and the future development and applications are prospected.
**Figure 1.** Schematic illustration of THz protection materials based on 2D materials.

### 2. THz Protection Theory

For different applications, THz protection materials should show different properties. In many cases, the THz protection materials need to efficiently absorb THz radiation, so as to reduce the THz echo as much as possible and form reflection loss. In some other cases, the THz transmittance must be strictly low, which could shield the incident THz radiation. The two THz protection theories were briefly presented as follows.

#### 2.1 THz Reflection Loss

According to the electromagnetic transmission theory, the simplified transmission model of THz reflection loss could be established, as shown in **Fig. 2a** [34, 35]. The incident THz radiation $E_0$ transmitted into the absorbing material from the air. The absorbing material was an infinite flat plate with a thickness of $d$, which was covered with a total reflection metal plate on the back. The $E_{r1}$, $E_{r2}$ to $E_{rn}$ represented the THz radiation intensity of the first reflection, the second reflection till the $n$-th reflection respectively, which were formed after multiple reflections of incident THz through the metal back plate and absorbing material surface. And the accumulation of multiple reflection energy formed the total reflection.
Figure 2. Schematic models of (a) THz reflection loss and (b) THz shielding theories.

When the THz radiation was normally incident to medium 2 (absorbing material) from medium 1 (air), according to the Fresnel formula, the reflectivity $R$ and transmittance $T$ at the interface could be expressed as follows [35-37]:

$$R_{12} = \frac{n_2 - \tilde{n}_1}{n_1 + \tilde{n}_2}$$

(1)

$$T_{12} = \frac{2\tilde{n}_1}{n_1 + \tilde{n}_2}$$

(2)

$$R_{21} = \frac{n_1 - \tilde{n}_2}{n_1 + \tilde{n}_2}$$

(3)

$$T_{21} = \frac{2n_2}{n_1 + n_2}$$

(4)

where $\tilde{n}_1$ and $\tilde{n}_2$ are the complex refractive index of medium 1 and medium 2, respectively. For lossy medium, $\tilde{n} = n - \kappa i$, where $n$ and $\kappa$ represented the real and imaginary parts of the refractive index, respectively.

The THz wave would be attenuated with propagating a certain distance $L$ in the lossy medium, so the intensity of THz radiation $E_L(\omega)$ could be expressed as follow [35]:

$$E_L(\omega) = E_0(\omega) \cdot e^{-\tilde{n}(\omega) L/c}$$

(5)

where $c$ is the speed of light ($3 \times 10^8$ m·s$^{-1}$), $\omega$ is the angular frequency (rad·s$^{-1}$), $E_0(\omega)$ is the initial THz signal intensity. And the propagation loss $P_n(\omega L)$, which was defined as the power ratio of THz signal after a propagation length to the initial THz signal, could be expressed as follow [35]:

$$P_n(\omega L) = e^{-\tilde{n}(\omega) L/c}$$

(6)
In such case, the total THz energy reflected by the absorbing material could be expressed as follow [34, 35]:

\[
E_T^R(\omega) = E_{T1}^R(\omega) + E_{T2}^R(\omega) + E_{T3}^R(\omega) + \cdots + E_{Tn}^R(\omega)
\]

\[
= E_0^R(\omega) \cdot R_{T1}^2 + E_0^R(\omega) \cdot T_{T1}^2 \cdot T_{T2}^2 \cdot P_n^0(\omega,L) + E_0^R(\omega) \cdot T_{T1}^2 \cdot T_{T2}^2 \cdot R_{T1}^2 \cdot P_n^0(\omega,L) + \cdots + E_0^R(\omega)
\]

\[
\cdot T_{T1}^2 \cdot T_{T2}^2 \cdot R_{T2}^n \cdot R_{T1}^n - 4 \cdot P_n^{4n-4}(\omega,L)
\]

(7)

The whole reflectivity (R), which represented the ratio of the total reflected power (P_R) to incident power (P_0), could be expressed as follow [35-37]:

\[
R = \frac{P_R}{P_0} = \lim_{n \to \infty} \frac{E_T^R(\omega)}{E_0^R(\omega)} = \left[ \frac{(n(\omega) - 1)^2 + \kappa^2(\omega))^2}{(n(\omega) + 1)^2 + \kappa^2(\omega))^2} \right]^{\frac{16}{n^2(\omega) + \kappa^2(\omega) - 1}} \cdot \exp\left( -\frac{4\omega \cdot k(\omega) \cdot d}{c} \right)
\]

(8)

Therefore, the reflection loss (R_L) could be written in decibels (dB) as follow [16, 34, 35]:

\[
R_L = -10 \cdot \log \left[ \lim_{n \to \infty} \frac{E_T^R(\omega)}{E_0^R(\omega)} \right]
\]

(9)

2.2 THz Shielding

Similar to the THz reflection loss, the THz shielding model was built with a slight difference, as shown in Fig. 2b [34]. The THz shielding material was also an infinite flat plate, but without a metal layer on the back. Therefore, in addition to the multiple reflections, there were also THz transmission wave correspondently, labeled as E_{i1}, E_{i2} to E_{in}.

Accordingly, the total THz transmission of the shielding material could be expressed as the sum of all multiple transmission energy [34, 36]:

\[
E_T^T(\omega) = E_{T1}^T(\omega) + E_{T2}^T(\omega) + E_{T3}^T(\omega) + \cdots + E_{Tn}^T(\omega) = E_0^T(\omega) \cdot T_{T1}^2 \cdot T_{T2}^2 \cdot P_n^0(\omega,L) + E_0^T(\omega) \cdot T_{T1}^2 \cdot T_{T2}^2 \cdot R_{T1}^2 \cdot P_n^0(\omega,L) + \cdots + E_0^T(\omega) \cdot T_{T1}^2 \cdot T_{T2}^2 \cdot R_{T2}^n \cdot R_{T1}^n - 4 \cdot P_n^{4n-2}(\omega,L)
\]

(10)

Thus, the whole THz transmittance (T) could be written by the total transmitted power (P_T) and the incident power (P_0) [34, 36]:

\[
T = \frac{P_T}{P_0} = \lim_{n \to \infty} \frac{E_T^T(\omega)}{E_0^T(\omega)} = \frac{16 \cdot \left[ \frac{(n(\omega) - 1)^2 + \kappa^2(\omega))^2}{(n(\omega) + 1)^2 + \kappa^2(\omega))^2} \right]^{\frac{16}{n^2(\omega) + \kappa^2(\omega) - 1}} \cdot \exp\left( -\frac{4\omega \cdot k(\omega) \cdot d}{c} \right)}
\]

(11)

Therefore, the electromagnetic shielding effectiveness (EMI SE) could be written in dB as follow [16, 34, 35]:
7

3. Graphene based THz Protection Materials

3.1 Graphene based THz Protection MMs

3.1.1 Single-layer Graphene MMs

Because of the obvious SPPs in the THz band [30], graphene had been widely studied for THz protection [38-54]. However, the atomic monolayer graphene is almost transparent to THz wave, so the overall intrinsic absorption of monolayer graphene is highly limited [14, 15]. Therefore, originated from the SPPs effect, the well-designed continuous or patterned graphene in the MMs layer could generate the collective oscillation of charge density and light on the interface between graphene and its surroundings, which were beneficial to effective THz absorption and protection [39]. The THz response of continuous films was extensively studied, which promising a further application in THz protection[17, 43, 55, 56]. By using polymethyl methacrylate (PMMA), Min et al. [56] transferred the continuous 2D graphene sheet grown on nickel to a polymer substrate with a gold bottom film. The whole structure built up a Salisbury screen MMs as shown in Fig. 3a, in which the graphene sheet worked as the resistance layer. The conductivity of the graphene sheet could be regulated by chemical doping to match the free space impedance, so that the incident THz attenuated directly in the MMs by the way of destructive interference. And the measured results showed that, when the square resistance of graphene sheet was $689 \Omega \cdot \square^{-1}$, the maximum absorption of 0.95 and 0.97 could be achieved at 0.97 and 1.5 THz (Fig. 3b). In addition, some patterned graphene, such as square [38, 57], cross [58-61], disk [60, 62], ribbon [63, 64], ring [39, 65, 66] and pixel [67] shapes, had also been designed and simulated for THz absorption. Xiao et al. [59] designed rectangular and cross-shaped graphene metasurfaces for THz absorption (Figs. 3c, e). The full-wave simulation showed that both the two MMs indicated effective absorbing ability, and the effective absorption (absorption above 90 %) bandwidth of the cross-shaped MMs could reach up to 1.13 THz (Figs. 3d, f). Furthermore, generated from higher real part conductivity of graphene surface and larger propagation loss of SPPs, the absorbing range in the low-frequency was broadened. As shown in Fig. 3g, the electric field distribution at the absorption peak revealed that the localization fields on the edges of the patterned graphene were obviously stronger,
because of the charges accumulation excited by the electric dipole resonances.

**Figure 3.** (a) Diagram of graphene based Salisbury screen MMs. (b) THz absorption of the MMs illustrated in (a) with different square resistances (sample A: $1295 \ \Omega \cdot \square^{-1}$ (●), sample B: $817 \ \Omega \cdot \square^{-1}$ (■), and sample C: $689 \ \Omega \cdot \square^{-1}$ (▲)). (Reproduced with permission from Ref. [56]. Copyright 2014 AIP Publishing.) (c) Unit cell of MMs with the rectangular array. (d) THz absorption of the MMs illustrated in (c) for various values of length $L$. (e) Unit cell of MMs with the cross-shaped array. (f) THz absorption of the MMs illustrated in (e) for various values of length $L$. (g) Electric field distributions of the MMs illustrated in (e) at the absorption peak for different $L$. (Reproduced with permission from Ref. [59]. Copyright 2017 The Optical Society.)

Meanwhile, some complex or composite graphene patterns were proposed to enhance THz absorption[39, 60, 66, 68, 69], so that the absorbing bandwidth could be broadened by the multiple resonant peaks generated from the multi-structures. Mou et al. [66] designed a patterned graphene concentric double rings (GCDR) metasurface to enlarge the absorbing bandwidth by forming plasmonic hybridization between two graphene rings (**Fig. 4a**). The calculated absorption results shown in **Fig. 4b** revealed that the MMs with GCDR can effectively absorb THz energy above 90% ranging from 1.18 to 1.64 THz with two isolated...
absorption peaks located at 1.26 and 1.54 THz. While the MMs with only inner or outer graphene single ring exhibited single absorption peak and narrow bandwidth, corresponding to the high-frequency and low-frequency peak of the GCDR structures. The coupling effect between the two graphene rings obviously enhanced the THz absorption with the stronger dissipating ability and wider bandwidth. Moreover, to further broaden the absorption bandwidth, Wu et al. [69] proposed the broadband MMs based on the self-affine fractal Hilbert curves, in which the self-affine multi-scale structures could induce different resonant frequencies, and the first five (I - V) iterative levels of the Hilbert curves were shown in Fig. 4c. As shown in Fig. 4d, with the increment of the Hilbert curve level, the responsive absorption bandwidth was clearly broadened for the multiple resonant behaviors, which were originated from the self-affine structures.

Figure 4. (a) Schematic of the THz absorber consisting of the GCDR. (b) Simulated absorption of the proposed GCDR absorber with different graphene patterns [66]. (c) Iterative construction of the Hilbert curve for the first five (I - V) levels. (d) Simulated absorption for graphene Hilbert structures of levels I, III, and V [69].
3.1.2 Multi-layer Graphene MMs

Multi-layer graphene MMs were also commonly introduced to achieve broadband THz protection. The resonant frequencies and coupling effects could be manipulated by designing the graphene patterns of each metasurface, so that the multi-layer graphene MMs could respond to more frequencies and wider THz bandwidth[15, 28, 70-78]. Rahmanzadeh et al. [15] designed the three-layer MMs composed of different graphene metasurfaces with square, cross and circular shaped patterns, respectively. The multi-layer structures and graphene patterns of each layer were shown in Figs. 5a-d. According to the equivalent circuit model, the patterned graphene layers could be simulated as the dispersion complex impedances (Fig. 5e), in which the structural parameters could be theoretically designed for the optimal absorption performance. The THz absorption results of the full-wave simulation shown in Fig. 5f revealed that the multi-layer graphene MMs achieved ultra-wide effective bandwidth, ranging from 0.55 to 3.12 THz. By analyzing the induced electric field, surface current and power loss density of each patterned graphene metasurface, it was clear that the SPPs played a dominant role in the THz dissipation process, and formed the physical absorption mechanism of the multi-layer graphene MMs. Meanwhile, the simulation results also verified that the designed structure was omnidirectional and polarization insensitive, so that the MMs could effectively protect the device under multi-polarizations THz incidence from wide incident angles.
3.1.3 Hybrid Graphene MMs

In order to further expand the THz response bandwidth and strengthen the THz absorption capability of the THz protection materials, it was also an effective method to combine graphene with other THz responsive materials, such as metals [18, 79-88], Si [89-93], water [94] and other materials [95, 96]. By reasonably designing the meta-structures of each component, the comprehensive advantages of each material could possibly be utilized. Peng et al. [85] designed the hybrid MMs based on four graphene fishing net structures and double metal rings (Figs. 6a-e). As the simulated results shown in Fig. 6d, the effective absorption bandwidth of the graphene-metal hybrid MMs could reach as high as 6.46 THz (1.24 - 7.70 THz). Meanwhile, the absorber was insensitive to different polarizations and incident angles of the THz incidence. The ultra-wide effective THz absorption bandwidth of the graphene-metal hybrid MMs was formed by the following three factors: (i) The SPPs of the graphene metasurface were enhanced by the metal metasurface, leading to the enhancement of the confined field. (ii) The resonance coupling of the metal and graphene metasurface could trap the incident THz wave between the...
two metasurfaces, which could form energy exchange and dissipation. (iii) The multiple THz reflections and their superposition between the metasurface and gold substrate further enhanced the THz absorption. In addition, the absorption bandwidth could also be broadened by composing graphene MMs with water, by taking advantage of the intrinsic dispersive permittivity of water. Zhang et al. [94] designed a graphene-water hybrid MMs THz absorber with ultra-wide effective waveband, in which the water was encapsulated in the polytetrafluoroethylene (PTFE) dielectric material with the patterned graphene metasurface on the top (Figs. 6e,f). The simulated results shown in Fig. 6g presented a broad THz effective bandwidth covering 4.52 - 9.02 THz. It also could be seen that the broadband absorption was superimposed by three absorption peaks, in which two low-frequency absorption peaks were excited by graphene metasurface, and the high-frequency peak was induced by the water. Moreover, by introducing the filling water, the absorption bandwidth of the hybrid absorber was obviously increased by 57.34 % compared to the absorber without water, while the absorption peak of the graphene metasurface was not shifted. Otherwise, the graphene-water MMs absorber was confirmed insensitive to the incident angles and polarizations.

![Graphene-water hybrid MMs THz absorber schematic](image)

**Figure 6.** (a-c) Schematic view of graphene-metal MMs THz absorber. (d) Simulated THz
absorption and reflection of graphene-metal MMs [85]. (e, f) Schematic view of graphene-water MMs THz absorber. (g) simulated THz absorption of graphene-water MMs (A_{12}: graphene-water hybrid MMs, A_1: A_{12} MMs without water, A_2: A_{12} MMs without patterned graphene metasurface) [94].

3.1.4 Tunable Graphene MMs

As a typical 2D semiconductor, the electron mobility and Fermi level of the graphene could be controlled by the external stimulations, such as electrostatic field [59, 90, 97-110], magnetic field [111, 112], optical pump [113] and temperature [83, 114]. So that the resonance frequencies of the designed graphene structures could be regulated by external stimulations, which could form effective and broadband THz absorption. According to Kubo formula, the surface conductivity of graphene could be expressed as follow [103]:

\[
\sigma_g = i \frac{e^2 K_B T}{\pi h^2 (\omega + i\tau)} \left[ \frac{E_F}{K_B T} + 2 \ln \left( \exp \left( - \frac{E_F}{K_B T} \right) + 1 \right) + i \frac{e^2}{4\pi h} \ln \left[ \frac{2|E_F| - h(\omega + i\tau)}{2|E_F| + h(\omega + i\tau)} \right] \right]
\]

where \( K_B, e, h, \tau, T \) and \( E_F \) are the Boltzmann constant, electron charge, reduced Planck’s constant, relaxation time, temperature and Fermi level, respectively. From the formula, it is obvious that the surface conductivity of graphene can be actively controlled by the Fermi level and temperature. Sensale-Rodriguez et al. designed an extraordinary controllable THz modulator by transferring a single-layer graphene onto a SiO_2/p-Si substrate (Fig. 7a) [110]. The carrier concentration and Fermi level in graphene could be tuned, when the voltage employed between the graphene and back metal was changed. While the Fermi level was modulated to the Dirac point (\( V = V_{\text{CNP}} \)), the THz absorption was at its minimum. And the THz absorption increased as the Fermi level shifted into valence or conduction band, for the intraband transition of the available density of states. The calculated power reflectance in Fig. 1b showed that the field intensity in graphene is maximum when the optical thickness of the substrate was an odd-multiple of the THz wavelength, in which obvious absorption swings occurred when the conductivity in graphene was extraordinary controlled. However, the THz absorption was hardly absorbed and did not change, when the substrate thickness was an even-multiple of the THz wavelength. Furthermore, by integrating the metal grating into the chemical vapor deposition (CVD) grown graphene sheet Salisbury screen MMs, Chen et al. [103]
realized an electrically tunable hybrid THz MMs absorber based on Ion Gel/graphene/Al/Polyimide(PI)/Al thin film structure (Figs. 7c, d). The Fermi level of the graphene layer could be changed by the external bias voltage, which could lead to controllable THz absorbing ability. The experimental results showed that it is feasible to achieve tunable THz absorption based on graphene grown by CVD with low carrier mobility, and the modulation depth of the THz absorption could reach 25% by applying relatively small bias voltage (-2 - 2 V), as shown in Fig. 7e. Then the numerical simulations based on transmission line theory and full-wave simulation software also verified the measured results. Besides graphene nanosheets, some patterned graphene structures were also designed and prepared for tunable THz absorption. Jin et al. [104] designed a coherent perfect absorber based on the square patterned graphene structure, which was composed of two alternately arranged graphene MMs layers (Figs. 7f-h). The Fermi level of each staggered metastructure could be independently controlled by the applied bias voltage distributed on the side. The numerical simulation results showed that the absorbing frequency peaks can be effectively controlled by changing the applied voltage values, and the absorber presented two independent tunable absorption peaks when the applied voltages of each staggered metastructure were different (Fig. 7i). Moreover, when the absorber was stacked by the metastructures in layer, multiple THz absorption peaks would be formed to achieve an ultra-wide THz absorbing waveband. Although the preparation of the tunable patterned graphene MMs was difficult now, it also could be achieved by template selective etching and some other new methods [101].
Figure 7. (a) Schematic structure, operating principle and (b) calculated power reflectance of the graphene THz absorption modulator [110]. (c, d) Schematic view and (e) simulated THz reflection of the tunable MMs THz absorber based on graphene film [103]. (f-h) Schematic view and (i) simulated THz absorption of the tunable MMs THz absorber based on patterned graphene [104].

3.2 Graphene Intrinsic THz Protection Materials

3.2.1 Graphene Foams

As a advanced carbon material with high thermal conductivity, high strength, large specific surface area, optical transparency and adjustable electromagnetic response, graphene had been widely used in radar wave attenuation and shown excellent loss performance [7], so its THz protection applications attracted a lot of attention [10-12, 35]. Different from the traditional
metal THz shielding materials for the reflection mechanism, Zdrojek et al. [11] prepared flexible THz shielding materials by dispersing graphene nanosheets in a polydimethylsiloxane (PDMS) matrix, whose shielding effect was mainly contributed by absorption. The shielding material showed excellent unit shielding efficiency exceeding $30 \text{ dB} \cdot \text{cm}^{-3} \cdot \text{g}^{-1}$. However, because of the inhomogeneous dispersion and interface mismatch, the THz shielding materials of this type would produce large surface reflection, which made the maximum absorption hardly to reach 80%. Therefore, adjusting the THz characteristics and improving THz attenuation were still challenging [10]. In recent years, with the fabrication development of 3D porous materials, such as graphene foams, the THz absorbers with conductive networks and large surface area could be established, which could provide excellent impedance matching and THz energy attenuation [7, 10, 35, 115]. Huang et al. [10] prepared a 3D porous graphene foam with a density of only 0.8 g/cm$^3$ by a solvothermal method (Fig. 8a). The optimal reflection loss of the THz absorber could reach 19 dB at 0.88 THz, and the effective absorbing bandwidth (reflection loss above 10 dB) could cover 95% of the test band (0.1 - 1.2 THz) under normal conditions, as shown in Fig. 8b. Moreover, when the incident angle was 45°, the effective bandwidth covered the whole test bandwidth while the optimal reflection loss was 28.6 dB at 0.64 THz (Fig. 8c). It was found that the outstanding THz absorbing ability of 3D graphene foam is mainly originated from the porous structure and long distance conductive network. On the one hand, the effective dielectric constant and impedance matching degree could be optimized by designing the porous structures to reduce the surface reflection. On the other hand, the 3D graphene sheets could induce surface currents under the THz radiation, so that the incident THz energy would be rapidly decayed in the resistance network by being converted into heat.
Figure 8. (a) Cross-section SEM image of the 3D graphene foams. (b) Reflection loss curves of some graphene foams samples. (c) Reflection loss curves of the T1000 samples with 4mm thickness at different incident angles [10]. (d) SEM image, structural diagram, (e) EDS maps and (f) reflection loss curve of the Graphene/Fe$_3$O$_4$ hybrid foams [35].

3.2.2 Graphene Hybrid Foams

To further enhance the THz protection properties, 3D graphene foams composited with carbon nanotubes [116], Fe$_3$O$_4$ [35] and other dielectric/magnetic particles were proposed to perform multiple attenuation mechanisms. Chen et al. [35] prepared the high-performance graphene/Fe$_3$O$_4$ 3D porous electromagnetic protection foams with ultra-lightweight and ultra-wide effective waveband (Figs. 8d, e). The measured results showed that the 3D foams could achieve an effective absorption waveband covering from 3.4 GHz to 2.5 THz, while the average reflection loss reached -38 dB (Fig. 8f). In addition, the 3D composite foams also exhibited excellent absorption properties under oblique incidence and different compression strains, while the absorber still maintained a stable absorption ability after 200 repeated compression/release cycles. The addition of magnetic particles further regulated the electromagnetic characteristics of the 3D graphene porous cross-linked structures, which greatly broadened the response waveband, and provided a strong technological foundation for
the active protection materials according to the increasingly complex electromagnetic environment in the future.

3.2.3 Tunable Graphene THz Protection Materials

In addition, for the Fermi level and carrier concentration of graphene could be conveniently regulated by some external stimuli, such as electric field and optical radiation, the THz absorbing modulation of the graphene foams was also available. Xu et al. [36] studied the active modulation behaviors of 800 nm laser and bias electric field on graphene foams, and realized the flexible regulation of THz shielding and absorbing performances. The schematic diagram of external stimulus control and the microstructure of graphene foams were shown in Figs. 9a, b. The absorptivity curves shown in Fig. 9c revealed that the THz absorption was strongly related to the frequency when there was no external field, with an absorptivity change from 0.04 at 0.2 THz to 0.8 at 1.6 THz. Furthermore, the absorption raised sharply with the increment of excitation intensity. The absorptivity changed rapidly from 0.13 to 0.954 at the low frequency of 0.3 THz, while the absorptivity at the high frequency of 1 THz varied from 0.62 to 0.995. The excellent THz absorbing modulation properties could be explained as follows: (i) Without external stimulation, the cross-linked graphene networks could be regarded as numerous coupling circuits of resistance, inductance and capacitance, so the time-varying THz field could excite induced currents on the cell wall of graphene. These long-range induced currents attenuated rapidly in the high resistance networks, resulting in sharp decay of the incident THz wave (Fig. 9d). (ii) When the graphene foams were stimulated by the laser or electric field, the nonequilibrium carriers could be generated to raise the Fermi level to a higher conduction band and enhance the THz absorption (Figs. 9e, f). (iii) Fundamentally, because the laser radiation could not penetrate the interior of the graphene foams, the photo-generated carriers could only be generated in the shallow layer of the foams. However, the electric field could pass through the whole graphene foams, resulting in more nonequilibrium carriers on the graphene sheets, which caused a superior regulating effect than the laser radiation.
Figure 9. (a) Schematic diagram of electrically and optically controlled graphene foams for THz modulation and absorption. (b) SEM image of tunable THz absorbing graphene foams. (c) The absorptivity curves of tunable graphene foams stimulated by different electrical voltages and laser excitations. Schematic diagram of THz waves propagation and interaction with free electrons in the graphene foams (d) without external field excitations, (e) excited by the laser light and (f) excited by the electric field [36].

4. Other 2D THz Protection Materials

4.1 MXene based Intrinsic THz Protection Materials

Besides graphene, MXenes were also commonly used 2D THz protection materials in recent years [13, 16, 37, 117]. MXenes are 2D transition-metal carbides and nitrides materials with the general formula $M_{n+1}X_nT_x$. MXenes had attracted extensive attention for their unique mechanical, structural, physical and chemical properties, among which the ultra-high conductivity (1500 S·cm$^{-1}$) and hydrophilic surface endowed them with excellent electromagnetic response characteristics and controlable structures [9, 118, 119]. 2D MXene nanosheets could induce the incident electromagnetic waves to conduct between their nanosheets, due to their good internal conductivity and certain layer spacing, which had been
proved to enhance the electromagnetic absorption effectively \[9, 120\]. Furthermore, Jhon et al. \[121\] theoretically confirmed the intrinsic THz response characteristics of MXene through first principles. Thus, Ma et al. \[37\] prepared ultra-light and compressible THz absorbing 3D porous MXene/Graphene oxide (GO) foams by solvothermal method (Figs. 10a-c). The absorber combined the advantages of MXene and GO, forming the tunable and ultra-wideband THz absorption properties. MXene/GO foams showed high THz loss efficiency during the whole test range (0.2 - 2.0 THz) with the maximum reflection loss of 37 dB at 2 THz. And when the mass ratio of MXene and GO was 1:5, the absorber presented the maximum average absorption loss of 30.6 dB. Moreover, after 200 times repeated compression, the THz absorption performance of composite foams unchanged (Fig. 10d). Similar to graphene foam structures, 3D porous MXene/GO foams could produce better impedance match and larger induced current loss, and the introduction of MXenes with high THz response could further improve the THz absorption performance. By utilizing the cross-linking reinforcement of the multivalent metal ions to MXene and GO nanosheets, Lin et al. \[16\] prepared the free-standing, lightweight, foldable and high stable MXene/GO/Zn\(^{2+}\) THz shielding foams by ion-diffusion-induced gelation method (Fig. 10e). The measured morphology results showed that a small amount of Zn components were evenly distributed in the homogenous foams, and the nanosheets were tightly packed in the cell wall of the foams (Figs. 10f-h). The unique cross-linked porous structures and moderate conductivity endowed the foams with higher THz shielding ability and lower surface reflection (Fig. 10i). The THz protection test results showed that when the thickness of the 3D foams was only 85 \(\mu\)m, the electromagnetic shielding effectiveness could reach 51 dB, while the reflection loss bellow -10 dB could cover 0.86 - 2.0 THz (Figs. 10j, k).
Figure 10. (a) Digital images, (b) structural diagram and (c) SEM image of MXene/GO 3D foams. (d) Reflection loss curves of MXene/GO 3D foams with different thicknesses after 0, 100 and 200 times compression [37]. (e) Structural diagram, (f) sectional SEM image, (g) EDS maps, (h) HAADF-STEM image, (i) THz shielding schematic diagram, (j) THz shielding efficiency curves and (k) reflection loss curves of MXene/GO/Zn$^{2+}$ composite 3D foams [16].

4.2 Tunable MXene based THz Protection Materials

For the electric conductivity of the MXenes could be inhibited by the external optical stimulation, it is possible to modulate their THz shielding properties. Therefore, Li et al. [117] studied the THz shielding regulation of the MXenes film stimulated by the ultrafast optical pulse (Fig. 11a). Due to the synergistic effects of THz absorption and reflection, the THz shielding effectiveness per unit thickness of the highly conductive Ti$_3$C$_2$T$_x$ thin film with a thickness of only 25 nm (Fig. 11b) could reach $4 \times 10^5$ dB·cm$^{-2}$·g$^{-1}$, which meant that the THz shielding effectiveness of a micron thickness film could come up to tens of decibels. After the
stimuli of 400 and 800 nm optical pulses, the transient THz transmittance of the MXene film could be enhanced within 2 ps. The THz shielding was suppressed by the photoinduced transient decrease of the real conductivity components which were proportional to the THz absorption, leading to more THz transmission. And with the increase of irradiation time, the influence on THz transmittance and shielding gradually weakened (Figs. 11c, d). Moreover, the measured results revealed that the THz shielding effectiveness could be reduced within nanoseconds through ultrafast optical pulses ranging from 95 K to room temperature (Figs. 11e-g). These unique properties made MXenes promising for the dynamic control of THz shielding and detection devices.

Figure 11. (a) THz shielding modulation schematic diagram of the MXene film stimulated by optical radiation. (b) AFM micrograph of a 25 nm thick Ti$_3$C$_2$T$_y$ film. (c) THz transmittance and (d) shielding effectiveness changes under different optical radiation time. (e) THz shielding effectiveness of the MXene film at 290 and 95 K without external stimuli. (f) THz transmittance change after excitation with 800 nm, 950 μJ·cm$^{-2}$ laser pulse at 290 and 95 K. (g) THz shielding effectiveness change 2 ps after photoexcitation at 290 and 95 K with 800 nm, 950 μJ·cm$^{-2}$ pulse [117].
4.3 Other 2D THz Protection Materials

Because of the resemble properties observed in 2D materials, other 2D materials such as phosphorene (2D black phosphorus) [122-130] and molybdenum disulfide (MoS$_2$) [131] were also studied for THz protection in recent research works. Based on the SPPs and localized plasmons resonance in phosphorene, it was convenient to produce THz absorbing MMs comprised of period structural phosphorene arrays. Wang et al. [127] designed the ultra-wideband MMs by assembling dozens of phosphorene/dielectric pairs with different widths, in which the meta-structures were divided into 4 groups and distributed in 4 layers (Figs. 12a-c). The widths of the phosphorene stripes were identical in each group, and different in distinct groups. Because of the asymmetrical structure and strong anisotropy, the simulated THz absorption results of the phosphorene MMs absorber showed that the absorption above 90 % could cover 3.4 - 9.6 THz for normal incidence, while the absorber could remain ultra-wideband and high absorption within the incident angular around 40° (Fig. 12d). In addition, as a typical 2D semiconductor with unique electrical, optical and magnetic properties, transition-metal dichalcogenides MoS$_2$ was also studied to design THz absorbing MMs. Wang et al. [131] proposed the angle insensitive THz absorbing MMs composed of a monolayer MoS$_2$ concentric double ring (MCDR) metastructure and a metal film separated by a dielectric layer (Fig. 12c). For the plasma hybridization caused by the lateral coupling between the two separate MoS$_2$ rings, the THz absorption bandwidth and effectiveness could be both improved. Moreover, the special concentric structures made the absorber insensitive to the incident angles and polarizations of the THz sources. Comparing to the MoS$_2$ single ring (MSR) MMs with only separate outer or inter ring, the calculated results confirmed that the MCDRs absorber performs obviously superior THz absorption in wider THz waveband (Fig. 12f).
Figure 12. (a) Unit cell, (b) structural details in each group, (c) top view across one phosphorene-section and (d) THz absorption curves of the phosphorene MMs [127]. (e) Schematic structure of the proposed THz absorbing MMs comprising MCDRs and metal gold separated by a thin layer of silicon dioxide. (f) THz absorption curves of the designed MCDR absorber and MSR absorbers [131].

5. Summary and Perspectives
With unique THz response characteristics, the MMs absorbers based on 2D materials in the forms of single layer, multiple layers, hybrid and tunable metasurfaces showed excellent THz absorbing performance. Recent advances in this field provided a sufficient theoretical and practical basis for THz protection, and the superior properties promised the 2D materials MMs wide application prospects. However, due to the preparation limitations of the patterned 2D materials metasurfaces, current researches were mostly focused on theoretical designs and simulations, which slowed down the practical applications of the 2D material MMs. In addition, the 3D intrinsic THz absorbing materials based on the porous or ordered 2D materials also showed exceptional THz protection performance, which effectively integrated the advantages of intrinsic properties and structural characteristics of 2D materials. These special structures
could optimize the surface impedance matching, and formed THz multiple scattering and electric transmission loss, which could realize high-efficiency absorption loss and active controllable protection performance in wide THz waveband. Without considering the material thickness, the 3D porous THz protection materials were more suitable to be practically applied in high-performance THz protection devices. Some of typical THz protection materials and their performances are summarized and listed in Table 1.

Table 1. Typical THz protection materials.

| Sample | Absorption | Effective bandwidth | Shielding effectiveness | Active control excitations | Ref. |
|--------|-------------|---------------------|-------------------------|---------------------------|------|
| Graphene/SiO$_2$/p-Si/Au MMs | $\sim$0.8 @ 620 GHz | - | - | Electric field | [110] |
| Graphene/polymer/Au MMs | 0.95 @ 0.5 THz | - | - | - | [56] |
| Porous graphene/cross-shape Au/liquid crystal/Au MMs | 0.96 @ 1 THz (TM) | 0.97 @ 0.87 THz (TE) | - | Electric field | [95] |
| MXene/Au nano-slot MMs | - | - | 20 dB @ 1.0 THz | - | [13] |
| Patterned graphene/PI/Au MMs | - | 1.54 – 2.23 THz | - | | [108] |
| Ion gel/graphene/stripe Al/PI/Al MMs | ~20 dB @ 0.43 & 0.75 THz | - | - | Electric field | [103] |
| rGO paper | 17.6 dB @0.7 THz | - | 72.1 dB @ 0.6 THz | - | [132] |
| 3D rGO foam | 28 dB @ 1.6 THz | 0.2 – 1.6 THz | 28 dB @ 1.6 THz | Electric and optical field | [10, 36] |
| WCNT/rGO foam | 30 dB | 1.5 THz (0.1 – 1.6 THz) | 61 dB | | [33] |
| Fe$_3$O$_4$/rGO foam | 38 dB (Average) | 3.4 GHz – 2.5 THz | - | | [35] |
| MXene/rGO foam | 30.6 dB (Average) | 0.2 – 2.0 THz | | | [37] |
| MXene/GO/Zn$^{2+}$ foam | 0.86 -2.0 THz | 51 dB | | | [16] |

The THz protection materials in future 6G applications should meet the application requirements, such as reduced dimensions, light-weight, wide effective absorption bandwidth
and high absorbing ability. Meanwhile, the THz protection materials should also realize tunable THz protection performance. For THz protection MMs, the advances in nanomaterials science and nano-manufacturing technology should be adopted to realize enhanced THz protection and tunable characteristics of 2D materials, and the device integration fabrication ability should be fully considered to improve their performance in practical applications. For THz intrinsic protection materials, the influence of intrinsic properties (scale, defects, functional groups and flake sizes, etc.), inner nanostructures (hole structure or layer spacing, etc.) and doping behaviors with other loss materials (dielectric or magnetic particles, etc.) on the THz responses should be systematically studied.

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