Chapter

Two-Dimensional Materials for Terahertz Emission

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Abstract

The demand for ultrahigh-speed, lightweight, low-cost, and defect-tolerant electronic devices drives the industry to switch to terahertz (THz) technologies. The use of two-dimensional (2D) materials has massively increased in THz applications due to their appealing electronic and optoelectronic properties, including tunable bandgap, high carrier mobility, wideband optical absorption, and relatively short carrier lifetime. Several 2D-material-based emitters, modulators, and detectors have been fabricated and examined. In this context, considerable research has been going on for 2D-material-based THz emitting sources, including materials and device structure to understand the electronics and optoelectronics mechanisms occurring in the THz region. This chapter focuses on the 2D-material-based emitters with insights into the background, the physical principle of photoconductive THz emitters, the 2D materials' properties, and the research trends in the fabrication and characterization of the THz sources based upon 2D materials.

Keywords: 2D materials, terahertz devices, photoconductive THz emitter, 2D-material-based emitters, THz emission

1. Introduction

When looking back into the IEEE Transactions on Microwave Theory and Techniques, it is noted that "the first occurrence of the term *terahertz* (THz) in this Transactions is attributed to Fleming [1] in 1974, where the term was used to describe the spectral line frequency coverage of a Michelson interferometer" (page 1, [2]). Nowadays, THz radiation refers to those frequencies/wavelengths spanning 0.1–10 THz range (30–3000 μm) of the electromagnetic spectrum [3] (see **Figure 1**). However, due to generation and detection obstacles, the THz range remains one of the least discovered spectra [2]. For example, these broadband waves, at relatively high frequencies, cannot be emitted through conventional optical and electronic technologies due to the limitations on the wavelength band and the energy gap, respectively [3]. Nevertheless, the advancements in these technologies have been utilized toward assisting with the THz radiation by accelerating electrons and employing nonlinear mediums [4].

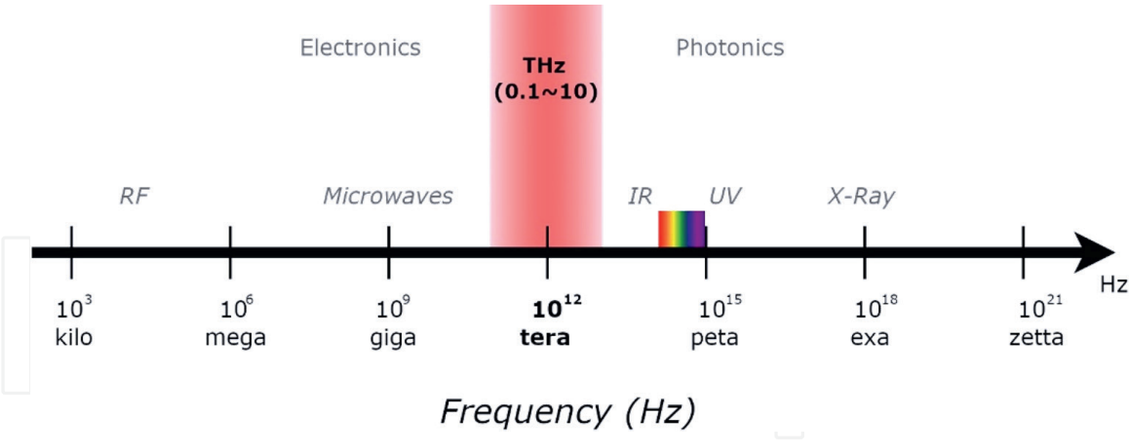

Figure 1.
Illustration of the electromagnetic spectrum in which the terahertz radiation lies in between the infrared and microwave radiations.

Terahertz technology's interest and application have grown over the past years [5]. The number of articles covering the application aspect of such a technology in different fields has exceeded that of the challenges presented in THz technology (i.e., obstacles within THz generation and detection) [6]. This technology has found itself in various scientific disciplines, from physics to biology and medical sciences [7]. The technological applications are just as vast, including outdoor/indoor communications, drug detection, security screening, semiconductor wafer inspection, semiconductor large-scale integrated (LSI) inspection, agriculture, air pollution, DNA, biometrics, medicine, and food [5]. Furthermore, this technology has continually evolved over the years as new materials and operational mechanisms are being used for the detectors and sources [8–10].

The THz sources can be classified under three categories: broadband sources (pulsed techniques), narrowband sources (continuous-wave techniques), or incoherent thermal sources [8]. Under these three categories, various sources of radiation have been investigated. Examples of such sources are mercury, silicon carbide (SiC) globar, cosmic background, quantum cascade lasers, photomixing devices (i.e., uni-traveling-carrier photodiode), free electron lasers, and backward-wave tubes or carcinotrons [5, 8, 11]. More importantly, when it comes to fundamental science/research, free electron lasers often stand out as they offer an improved signal-to-noise ratio in addition to emitting high-powered terahertz light (i.e., 10 W) for specific frequency ranges. Unfortunately, specific applications have strict limitations on the size, weight, and cost; hence such a source cannot be tolerated, and from there, researchers and scientists focused on investigating and improving other alternatives [5, 8, 11].

When looking into broadband THz emitters, it is noted that optical rectification and photoconduction device mechanisms are often employed [8]. The photoconductive emitter is an optoelectronic device that consists of photoconductive materials, photoconductive electrodes, and a lens to steer the emitted beam [3, 12]. While the THz emission by optical rectification approach can be realized by using an electro-optic (EO) medium and a nonlinear mixing, the use of ultrafast laser determines the duration of the quasi-static nonlinear response and thus the envelope of the emitted THz plus [13]. In the end, the photoconductive schemes can only be characterized by the breakdown voltage, carrier mobility, carrier lifetime, and dark resistivity [3]. Furthermore, over the years, scientists and researchers have investigated different materials for photoconductive schemes such as indium aluminum arsenide (InAlAs),

indium gallium arsenide (InGaAs), gallium arsenide (GaAs), and a combination of group III-VI materials [3]. Those examples were some of the most investigated materials for photoconductive devices. Nevertheless, progress in nanofabrication technologies has enabled the discovery of other alternative materials (i.e., two-dimensional (2D) materials) that could offer optimum photoconductive characteristics [14, 15].

Two-dimensional materials have shown massive progress over the last decade in which their compelling/appealing electronic and optoelectronic characteristics make them a potential alternative for THz applications. Such characteristics include high carrier mobility, low carrier lifetime, tunable bandgap, ultra-broadband, optical absorption, and response [14, 15]. Its electrical and optical characteristics could be altered by changing their physical and chemical properties by introducing dopants (i.e., Molybdenum disulfide), strain, or an electric field [14, 15]. Several kinds of 2D materials have been explored over the years, such as graphene, transition metal dichalcogenides (TMDs), black phosphorus, and MXenes, in addition to other prospective candidates (i.e., germanium sulphide (GeS), hexagonal boron nitride (hBN), organic-inorganic hybrid perovskites, etc.) [15]. More importantly, several kinds of 2D-material-based emitters have been fabricated and examined, as seen in the literature [16–20].

This chapter will focus on 2D-material-based THz emitters with insights into the background, the physical principle and principal components of THz emitters, and the research's trends in the fabrication and characterization of the 2D materials devices structures. In Section 2, different structure designs of photoconductive THz emitters are shown and discussed. In Section 3, the 2D material properties are reviewed. This review includes its mechanical, structural, electrical, and optical priorities. Ultimately, Section 4 presents the recent advances and development in layered 2D materials and heterostructures for THz emitters.

2. Photoconductive terahertz emitter

The THz emission by a photoconductive emitter can be performed using ultrafast laser pulses and an applied bias field, E_b, taking into consideration the bandgap and the breakdown voltage of the material. In general, The THz emission mechanism will start under the optical excitation by ultrafast (femtosecond) laser pulses when it is biased at a bias field, E_b. When the optical pulses of the laser (with a bandgap higher than the band gap of the photoconductive material) hit the photoconductive gap, the free carrier will be generated and the bias field, E_b, drives the device to emit electromagnetic radiation at the THz frequencies [3]. The emission characteristics of THz devices primarily depend on the characteristics of the photoconductive material used and the design of the device. One of the promising materials is the 2D material, which will be discussed in detail in Sections 3 and 4 of this chapter. First, the photoconductive designs are discussed in the following paragraphs. The photoconductive THz emitter design can be classified into microstructure and nano(plasmonic) structures.

In the literature concerning microstructure photoconductive emitters, several studies have presented the influence of the design on the emitter performance. Madéo *et al.* adjusted electrode spacing of the interdigitated photoconductive antennas for tunning frequency of THz. They used THz-time domain spectroscopy (TDS) to measure the pulsed electric field and emission spectra. They observed that decreasing the electrode gap from 20 to 2 μm has shifted the emitted spectra peak from 0.73 to 1.33 THz, and this demonstrated that faster space-charge screening of the bias field

occurs as the electrode spacing is reduced [21]. In addition, our previous work on the design and fabrication of photoconductive THz emitter based upon SI-GaAs and SI-InP investigated the influence of the bow-tie antenna structure on the THz emission performance and found that there is a correlation between the structure sharpness and the measured bandwidth of the photoconductive THz emitters [9].

In the nano (plasmonic) structures photoconductive emitter, Berry *et al*. presented the plasmonic photoconductive THz antenna for the first time, as shown in **Figure 2**. They related the new concept of the photoconductive antenna with the high quantum efficiency, which led to a significant increase in the optical-to-terahertz conversion efficiency [22].

Singh *et al*. fabricated an interdigitated photoconductive antenna, a THz emitter that eliminates the need for micro-lens array focusing. This avoids the saturation effect at a higher optical excitation density since photoexcitation can be done in larger areas. However, the device was fabricated as a microstructure and compared with the plasmonic structure device. The plasmonic photoconductive emitter emitted THz radiation power twice as much at 200 mW optical excitation [23].

Yardimci *et al*. reported a high THz radiation power with a notable improvement in optical-to-terahertz conversion efficiency [24]. They presented a novel design for a photoconductive THz emitter based on plasmonic (nano-structured) electrode. They enhanced optical-to-terahertz conversion efficiencies by a significant margin by incorporating plasmonic (nano-structured) electrodes within the

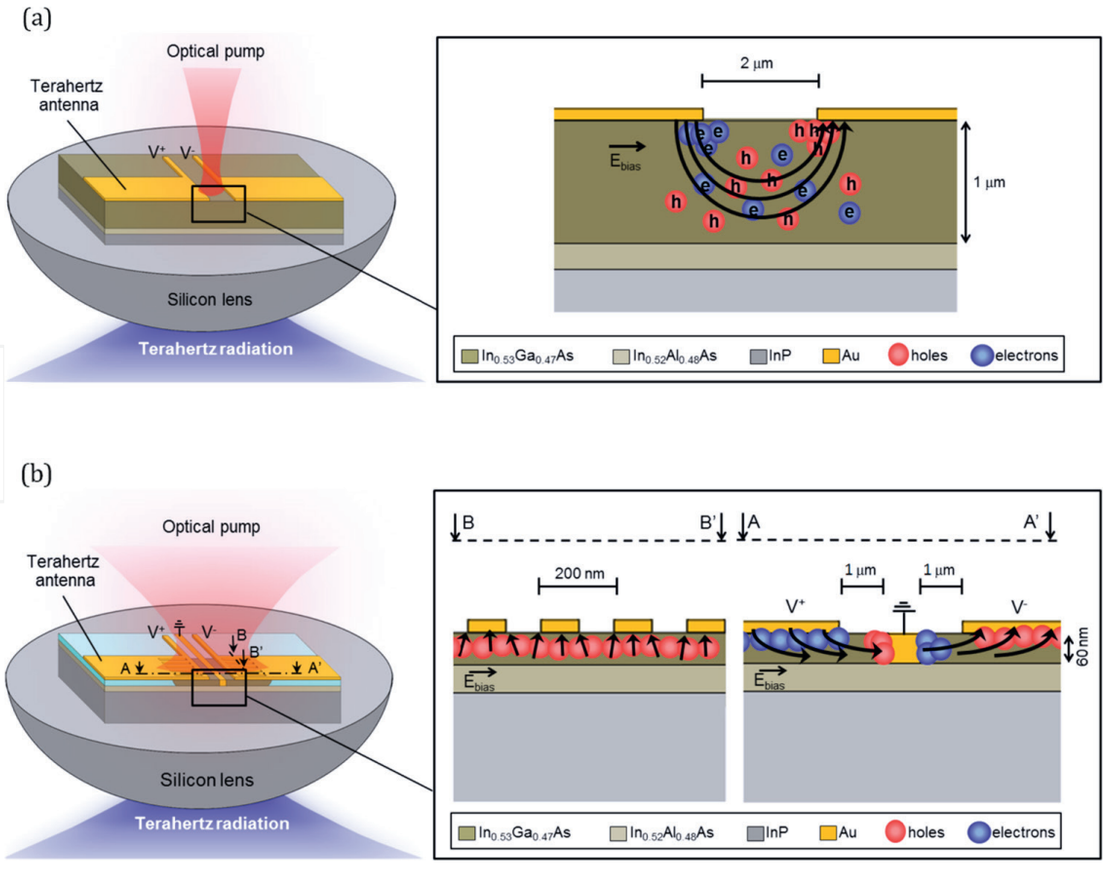

Figure 2.
(a) A microstructure photoconductive terahertz emitter with a photoconductive gap of 2 μm and (b) a nanostructure (plasmonic) photoconductive terahertz emitter based on nanoscale electrode with a spacing of 100-nm between the electrodes. © IOP publishing. Reproduced with permission. All rights reserved [22].

photoconductive gap, this increased time-varying dipole moments. As a result, about 1.6% optical-to-terahertz conversion efficiency was demonstrated, as the radiation power levels were up to 3.8 mW at an optical pump power of 240 mW over the 0.1–5-THz frequency range.

In addition, Berry et al. studied and fabricated the THz emitters within a logarithmic spiral antenna. The emitters were formed by an array of plasmonic photoconductive emitters, that were 3 by 3. Moreover, when optically pumped by 320 mW the device emitted 1.9 mW of pulsed THz radiation over the 0.1–2 THz frequency range. This design, as shown in **Figure 3**, offered much greater power THz pulses compared to other plasmonic photoconductive THz emitters [25].

The saw-toothed plasmonic contact electrode's structure has been studied by Zhang *et al*. A power enhancement is observed using a saw-toothed plasmonic logarithmic spiral photoconductive antenna (more than 10 times) [26]. The enhancement is accosted with the strong electric field obtained by the novel structure, as shown in **Figure 4**.

Chen *et al*. conducted an experiment on GeSn plasmonic THz photoconductive antenna [27]. The experiment compared a bowtie antenna with an active area of $20 \times 20\ \mu m^2$ and a gap size of 10 μm and plasmonic gratings with a spacing and pitch size of 230 nm and 480 nm, respectively. The resulting bandwidth was up to 2 THz and SNR was 50 dB. The GeSn plasmonic photoconductive antennas prototype was fabricated through a metal-oxide-semiconductor.

In addition, based on surface plasmonic resonance (SPR), Zhiguang *et al*. [28] proposed a nano cylinder array photoconductive antenna structure by analyzing the interaction between incident light and substrate. It showed an 87% optical transmission into the GaAs substrate, which was 5.8 times the optical transmission of conventional photoconductive antennas.

The field of design and fabrication of photoconductive THz emitters is massively researched using III-V semiconductors. However, using of 2D materials for these emitters is a promising field for study and research. Therefore, the upcoming section will discuss the distinct properties of the 2D materials.

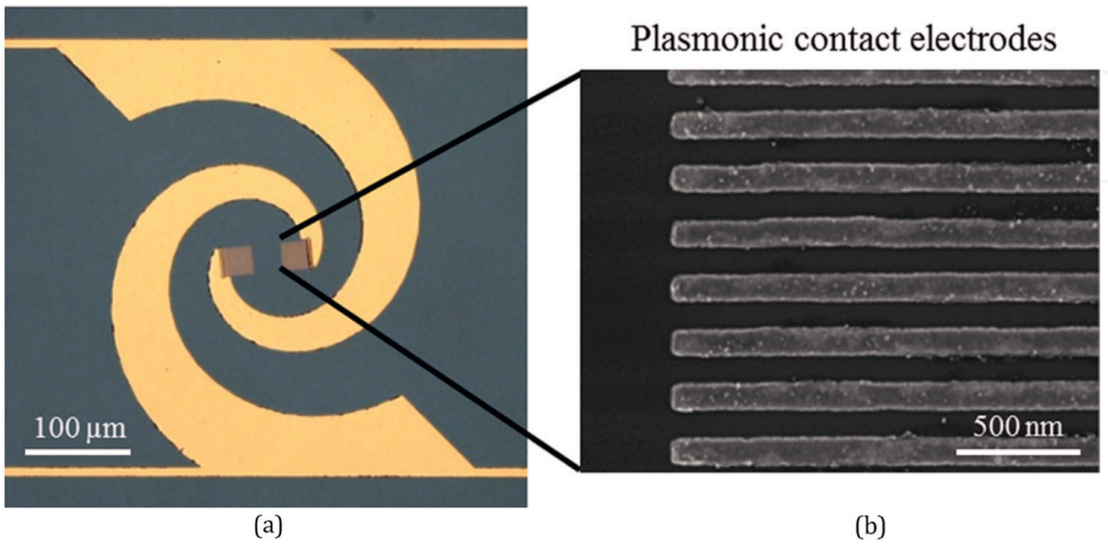

Figure 3.
The microscope (a) and SEM (b) images of the fabricated sample on an LT-GaAs substrate showing: (a) the integration/combination of the plasmonic metal electrodes within the logarithmic spiral antenna and (b) the metal electrodes. This figure is reprinted from ref. [25] with the permission of AIP publishing.

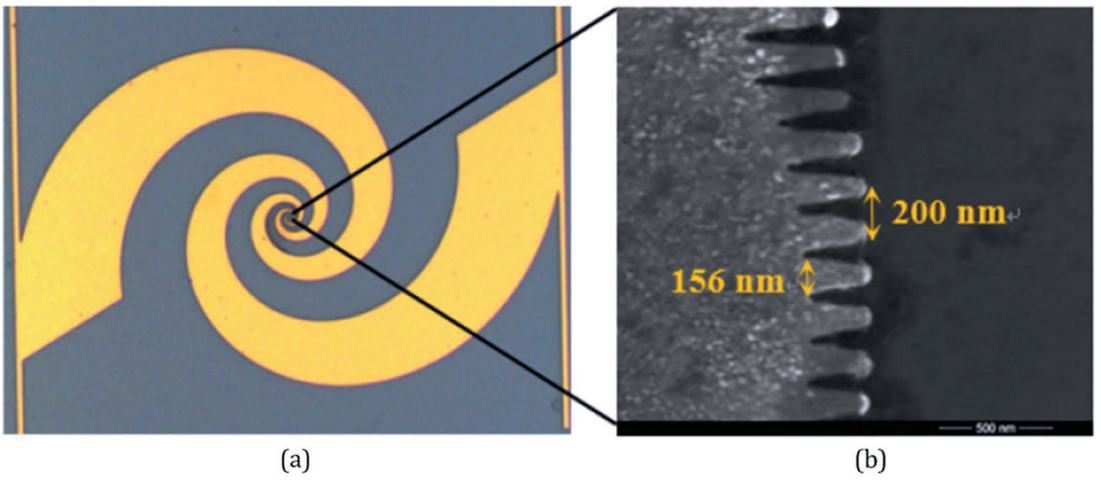

Figure 4.
The microscope (a) and SEM (b) images of the fabricated sample show: (a) the integration/combination of the saw-toothed plasmonic metal electrodes with the logarithmic spiral antenna and (b) the saw-toothed electrodes. [26].

3. Properties of 2D materials

Two-dimensional materials have recently fascinated researchers' attention due to their distinct properties. Research over the last decade has made huge progress in obtaining high-performance 2D materials. This section discusses the mechanical, structural, electrical, and optical priorities of 2D materials.

3.1 Mechanical properties of 2D materials

The mechanical properties of 2D materials are paramount to uncovering aspects of the material's behavior and how the material will act under external forces. Hence, it is vital to study the mechanical properties needed to discover the strong points in 2D materials and build novel applications and devices. The mechanical properties can be studied theoretically by the density functional theory (DFT) and experimentally. For example, graphene is a two-dimensional material. Graphene is the most robust material ever, and its bandgap energy is zero eV, so the existence of graphene in the semiconductor industry is limited. However, the TMDs, such as molybdenum disulfide (MoS_2), tungsten disulfide (WS_2), molybdenum diselenide ($MoSe_2$), and tungsten diselenide (WSe_2), are potential alternative material of graphene due to the presence of their bandgaps. As well in TMDs, bandgaps can convert from an indirect bandgap in bulk into a direct bandgap in monolayer.

The properties of 2D materials have high anisotropy between the in-plane and out-of-plane mechanical properties. The close atoms are linked together via covalent bounds in the same atomic plane leading to low defects density and strong in-plane mechanical properties. However, the out-of-plane are the interlayers that are stocked together by weak van der Waals interactions. The layers can slide easily when shear stress is employed [29]. Therefore, aside from graphene, the mechanical properties of MoS_2 also have been studied in-plane. The measured in-plane stiffness and effective Young's modulus of monolayer MoS_2 are about 182 ± 14 N.m-1 and 280 ± 21 GPa, respectively, which is less than graphene's stiffness [30]. In addition, Cooper *et al*. [31] measured the stiffness theoretically depending on DFT and found that 2D Young's modulus of monolayer MoS_2 is 130 N/m and intrinsic strength is 16.5 N/m. As a result,

it is unlocked for a broad of electronics and optoelectronics applications of monolayer TMDs such as fixable sensors and the ability to tune the bandgap by applying external strain.

3.2 Structural and electronic properties of 2D materials

Two-dimensional materials can be found in various forms, such as an element (graphene), metallic compounds (MoS_2, $MoSe_2$, WS_2, and others), nonmetallic compounds (hexagonal boron nitride (h-BN)), organics (polymer), and salt (clays) [32]. The shapes of the material are bulk, monolayer, bilayer, or multilayer. The crystal structure of TMDs is layered, and each layer consists of a hexagonal transition metal atoms sandwiched between two planes of hexagonally arranged dichalcogen atoms. The metal and chalcogens are covalently bonded within a layer stuck together by van der Waals bonds [33]. Thus, all TMDs have a hexagonal structure [34]. The atomic arrangement in the structure affects the electronic structure, vibrational modes, and optical properties. The atomic arrangement was designed with a particular phase 2H semiconductor phase, and metallic 1 T phase.

The 2D layered materials stacks are very different from the conventional bulk 3D heterostructures, as each layer acts simultaneously as the bulk material and the interface, reducing the charge displacement inside each layer. Nevertheless, the charge transfers between the 2D layers are massive. Thus, the band structure could be engineered by inducing large electric fields. The band structure can be engineered via alignment between the nearby crystals, number of layers, the type of the materials' surface recombination, and charge transfer [35].

3.3 Optical properties of 2D materials

Engineering of the 2D materials' optical properties can be achieved by changing one (or more) of the parameters, including material thickness and material type/structure, or by applying external strain [36]. The bandgap of 2D materials covers a wide range: from zero band gap (semimetal such as graphene) to the insulator (wide band gap). Thus, 2D materials are very attractive for optoelectronics applications, such as emitters, detectors, and optical modulators [37]. Additionally, our earlier work has established security applications as a promising field of optoelectronics applications of 2D materials [38]. The bandgaps of 2D materials have been subject to enormous studies experimentally (e.g., through photoluminescence) or theoretically through (DFT) modeling. Ketolainen *et al*. theoretically calculated the optical gap using the TD-HSE06 approach for a wide gap range of 1–6 eV of various 2D materials. The values agreed with the experimental optical gaps and the GW + BSE calculations. **Figure 5** shows that the theoretical absorbance depends on the TD-HSE06 method, HSE06 functional, and PBE functional for P_4, WSe_2, MoS_2, GaP, GaN, AlN, CF, and BN [39].

3.4 Synthesis and fabrication of 2D materials

Since the successful isolation of graphene from graphite in 2004, enormous efforts have been made to synthetize and fabricate 2D materials [40]. Typically, synthesis methods are classified as top-down or bottom-up. The top-down method involves isolation of a 2D layered sheet from bulk crystal using reduction methods, such as mechanical cleavage, liquid vapor exfoliation, or electrochemical reduction. In

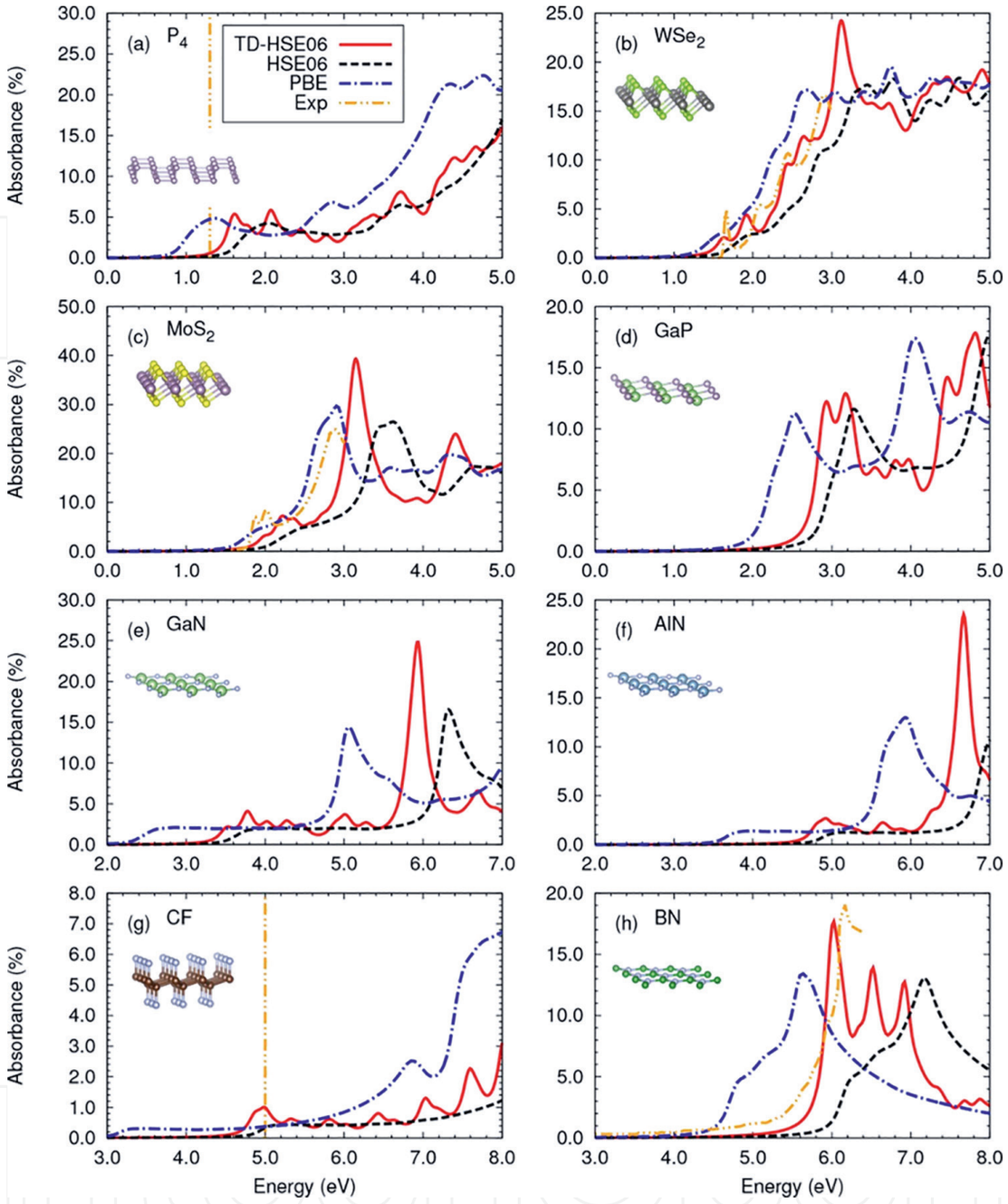

Figure 5.
This illustrates the in-plane optical absorbance using the TD-HSE06 method, HSE06 functional, and PBE functional for 2D layers: (a-h) P_4, WSe_2, MoS_2, GaP, GaN, AlN, CF, and BN, respectively. Moreover, the figure shows the experimental optical absorbance curves of WSe_2, MoS_2, and BN and the first excitation peaks in the experiments (orange curve). The atomic structures of the 2D materials are illustrated in the insets panels, "reprinted (adapted) with permission from ref. [39]. Copyright {2017} American Chemical Society."

the bottom-up method, 2D material layers are produced from atomic or molecular precursors. This method typically involves the use of advanced techniques, such as molecular epitaxial growth (MBE), chemical vapor deposition (CVD), physical vapor deposition (PVD), and pulsed laser deposition (PLD). A representative schematic of 2D materials synthesis is shown in **Figure 6** [32].

Each bulk layer of 2D material consists of dangling bond-free layers that are weakly bound to each other via van der Waals force. By using the mechanical

exfoliation method, a single layer or a few layers can be isolated from bulk materials. The applied force and adhesion to the exfoliation substrate should be controlled and overcome the van der Waals force during the exfoliation process. Typically, the mechanical exfoliation method produces the highest quality, higher crystallinity with fewer defect densities, and less impurity contamination [41]. However, the yield of this approach is very low and the method is time-consuming because of the random nature of flake thickness, size, number of flakes, and surface morphology is uncontrollable. Liquid exfoliation and chemical exfoliation techniques can be used to overcome the low yield. However, the exfoliated materials produced through these techniques are of relatively low quality and have small flake sizes. To improve the yield, we coated SiO_2 substrate with a sub-5 nm polymeric poly (vinyl alcohol) film thickness prior to exfoliation in our previous study [42]. The numbers and sizes of the produced flakes are much larger than those obtained using the conventional exfoliation method while maintaining the quality of the produced film.

As the exfoliation approach is limited in terms of large-scale production, various material growth methods have been studied to produce large-scale 2D materials. Of these, the CVD method is the most common because it allows large-scale and good-quality 2D materials to be produced. In addition, it is a low-cost method compared to MBE, PVD, and PLD. Typically, CVD is a high-temperature process used to synthesize various 2D materials on the substrate from solid, gas, or liquid precursor in a tube furnace. Typically, graphene is grown on metal substrate (e.g., Cu or Ni) using mixtures of gases. The transfer of large-area graphene films to a fabrication substrate, such as SiO_2, may lead to degradation of the electrical optical properties of the film. Alharbi et al. described high-quality, large-sized monolayer WS_2 grown on SiO_2 substrate using tungsten oxide (WO_3) and sulfur (S) solid precursors [43]. Others reported wafer-scale MoS_2 grown using metal–organic CVD [44]. Although these growth methods are scalable and produce large-size films, the quality of the films

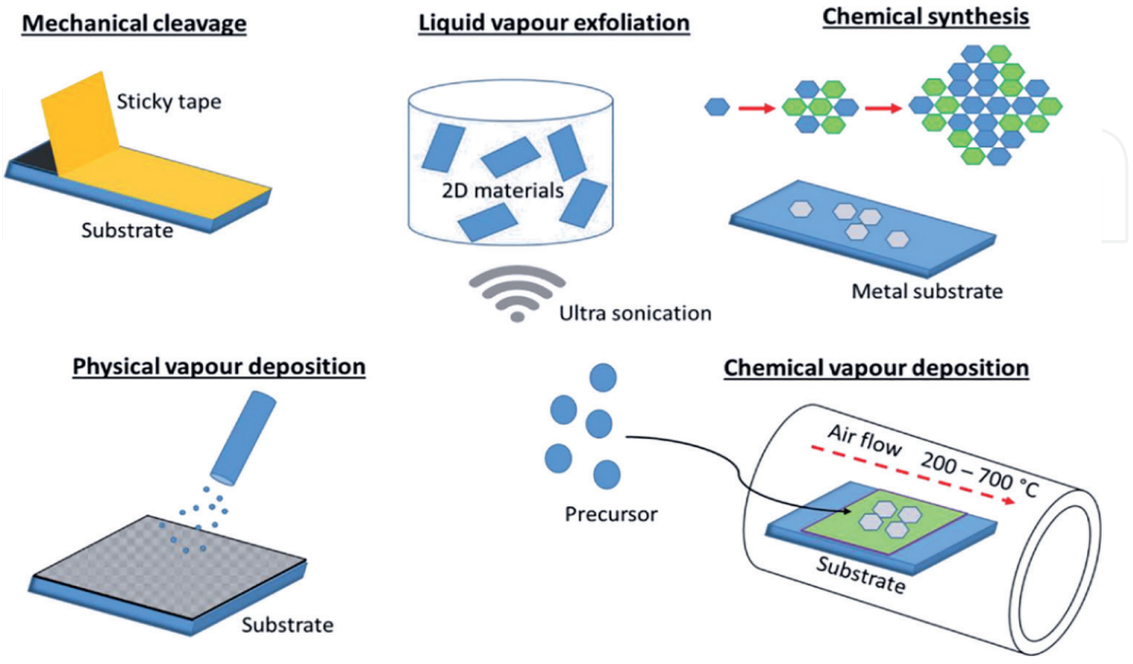

Figure 6.
A schematic of various 2D synthesis processes [32].

is lower than that of films produced using mechanical exfoliation. For example, the produced materials using CVD method are polycrystals and suffer from grain boundary and structural defects. Some of these defects may not be an issue in terms of THZ applications, the defect that suppresses the carrier lifetime.

The direct growth of heterostructure is still a major challenge. The produced heterostructures suffer from problems, such as uncontrollable growth directions, low-quality materials, and limited material combinations. Thus, most 2D material-based heterostructures are built using layer transfer methods. Suk et al. described the transfer process of CVD-grown monolayer graphene onto other substrates [45]. A deterministic dry transfer method for 2D materials that allows material positioning to be controlled has also been developed [46]. This method led to explorations of novel applications and new phenomena in 2D materials [46]. Moreover, a gold-assisted layer transfer process has been developed to overcome the adhesion of CVD-grown MoS_2 to SiO_2 and enable the controllable transfer of the film [47]. Although layer transfer methods enable many novel heterostructures to be built, the sizes of these structures remain limited.

From here, it is clear that 2D materials could become a promising alternative for THz emitters. Therefore, the upcoming section will discuss the integration and development of such materials for THz emission applications.

4. The 2D material-based THz emitters

The THz emitter is a key enabler of many THz systems. However, a wide variety of alternative techniques have been developed for THz emissions, such as nonlinear electro-optic crystal [48] and air plasma [49], to obtain THz waves of higher intensity and especially broader bandwidth. Nevertheless, these sources do not meet the current application demands. Thus, owing to their extraordinary electro-optical properties, the 2D materials-based emitter is trending in THz technologies and is expected to solve this issue. This section will focus on 2D-material-based THz emitters, including layered 2D materials and heterostructures.

4.1 Layered 2D materials

Two-dimensional materials such as graphene and MoS_2 have very high carrier mobility, surface conductivity, and a very short carrier relaxation time compared to traditional semiconductors [50]. These unique advantages can open up many new research ideas and dramatically improve device performance. Thus, 2D materials have attracted enormous research interest for THz applications. The following subsections discuss THz emitters based on layered 2D materials.

Graphene has attracted researchers' attention since its first successful separation from graphite in 2004 due to its extraordinary proprieties. Several studies have been carried out on graphene for THz applications. Graphene-based THz emitters were introduced early in 2007, where optically or electrically pumped graphene was expected [51, 52]. Then, a pumped THz laser with a Fabri-Perot resonant cavity design based on an optically pumped graphene-heterostructure was proposed in 2009 [53]. **Figure 7** illustrates the structure of the device where the optical excitation with the energy of $\hbar\Omega$ generates electrons and hole pairs in the graphene and subsequently acquires population inversion to generate THz radiation.

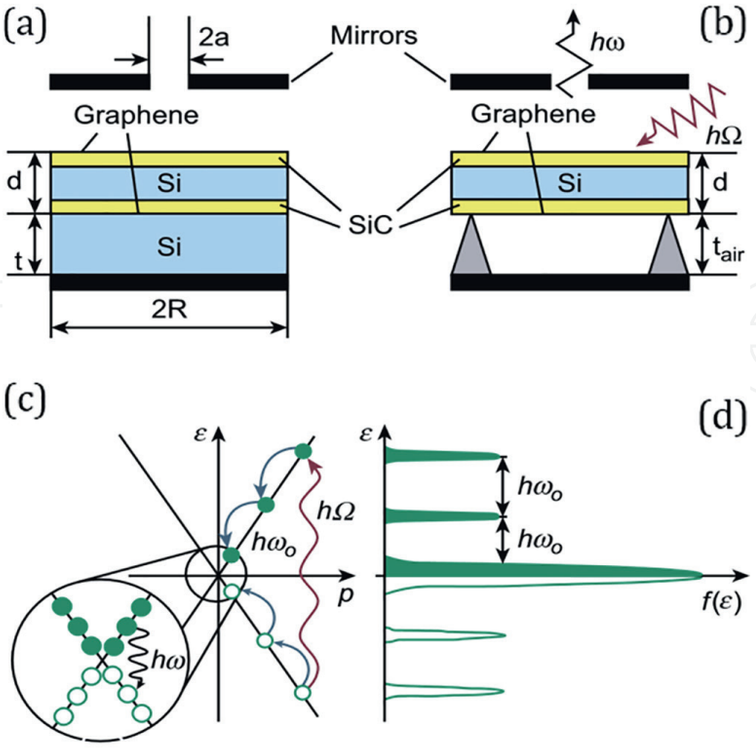

Figure 7.
Schematic view of the laser structures with (a) Si separation layer (b) air separation layer, (c) laser pumping scheme (d) electron and hole distribution functions. Figures were reprinted with permission from ref. [53].

With this in mind, a graphene-based THz laser based on optically pumped multiple-graphene-layer structures separated with a metal slot-line or a dielectric waveguide was proposed [54]. The THz lasing based on graphene P-i-n was demonstrated under certain experimental conditions [16]. Additionally, graphene-based plasmonic oscillator THz generators were proposed. Furthermore, it was found that tuning the intrinsic plasmons of graphene can generate radiation up to 1 THz [16].

Moreover, the current injection graphene-based THz emitter has been demonstrated to achieve higher power. Tong *et al*. demonstrated THz sources (1–3 THz) created with graphene field effect transistors, where the graphene has been attached to a double-patch antenna and an on-chip silicon lens [55]. The fabricated THz sources operate at room temperature and produce significantly higher power than conventional THz sources. The graphene-antenna device was fabricated on a high-resistivity silicon chip covered with a 300 nm-thick silicon oxide layer. Then, they were bonded back-to-back on an elliptical silicon lens. The designed antenna has a size of 45 × 31 μm. Graphene film was placed between the two metal patches of palladium or gold. This double patch also served as the source and drain contacts for the graphene transistor. According to their simulation results using a high-frequency structure simulator (HFSS), the impedance spectrum indicates an optimal operating frequency of 2.1THz. Therefore, the device can be used as a THz emitter or detector.

Coherent THz emission from monolayer graphene has been reported by Maysonnave *et al*. [56]. They experimentally demonstrated the process of exciting the graphene by femtosecond optical pulses to generate THz emitting in the range from 0.1 to 4 THz. The emission occurred because of a second-order nonlinear effect,

which is dynamical photon drag that is induced by the transfer of light momentum to carry through ponderomotive electric and magnetic forces [57].

Recently, Zhang *et al*. demonstrated a new hybrid photoconductive to overcome the fundamental trade-off between picosecond ultrafast pulses power output. The photoconductive switch was designed by optimizing a hot-carrier fast lane using graphene on silicon. Because of the combination of the strong absorption in bulk semiconductors with a graphene layer as a hot carrier fast lane, the proposed devices emitted a high-power THz field, with up to 80 times amplitude improvement compared to the devices without graphene [58].

4.1.1 Transitional metal dichalcogenides (TMDS)

Layered transitional metal dichalcogenides such as MoS_2 and WS_2 are a kind of 2D material that have remarkable optical properties. Unlike gapless graphene, TMDs have tunable band gaps that can be changed from indirect to direct band gaps with a number of layers.

Molybdenum disulfide (MoS_2) is the most studied TMDs material owing to its striking properties for optoelectronic applications. As the demand for the working frequency of devices increases toward the THz region for high-speed applications, the unique advantages of MoS_2 are expected to play a major role. For instance, the photoconductivity response time can reach 350 fs from CVD-grown monolayer MoS_2 and 1 Ps from trilayer MoS_2 and monolayer WSe_2 [59]. Huang *et al*. studied the THz pulse emission from layered MoS_2 film. They observed that MoS_2 can generate from 0.1 to 3.5 THz radiation time-domain surface emission spectroscopy under the excitation of linearly polarized femtosecond laser with the reflection configuration, as shown in **Figure 8**(a). The radiation amplitude has a linear dependence on the incident pump influence, which can confirm the second-order nonlinear process. **Figure 8(b)** reveals that the single-cycle THz pulses produced from the three samples (MoS_2, graphite, and InAs) have similar waveforms [60]. However, the THz radiation of MoS_2 has a longer pulse delay because of the combination of penetration depth and rugged surface. Nevertheless, the research of TMDs-based THz emitter is ongoing and most

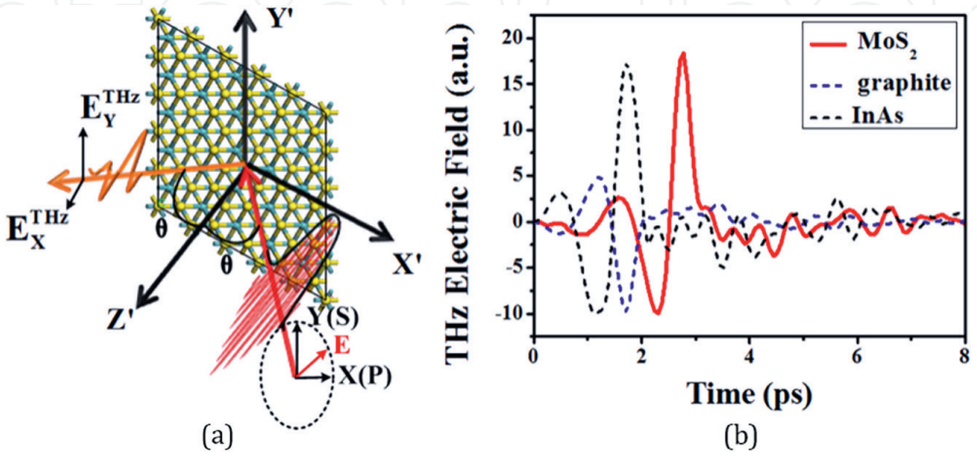

Figure 8.
(a) Schematic illustration of THz radiation in a reflection configuration. (b) THz pulses in time domain produced from MoS_2, graphite, and InAs layered crystal. "Reprinted (adapted) with permission from ref. [60]. Copyright {2017} American Chemical Society."

of the research focuses on optimizing and producing controllable various types of TMDs for THz applications.

4.1.2 Topological insulator

Topological insulators (TIs) are promising 2D materials for the next generation of optoelectronic applications. Among the TIs materials, Bi_2Se_3 has attracted tremendous research interest in the scientific community for THz emission. An efficient THz emission from the Bi_2Se_3 basal plane was observed upon femtosecond optical excitation [61]. Moreover, Giorgianni *et al*. demonstrated that Dirac surface states in Bi_2Se_3 TIs induce a strong nonlinear THz response [62]. Additionally, surface crystallography plays a major role in THz emission. For instance, Bi_2Se_3 grown on Al_2O_3 exhibits an anisotropic response with a strong modulation of the THz while in its phase, while its thin film grown on Si nanocrystals leads to THz emission in an opposite phase [63].

In another study, THz emission from Bi_2Te_3 nanofilms was studied. In their study, they used various thicknesses of Bi_2Te_3 nanofilms grown on Al_2O_3 substrates and excited the film by femtosecond laser pulses [64]. An efficient linear polarization of tunable THz radiation and high-quality chiral THz waves was demonstrated by controlling the pump laser pulse polarization, incident angle, and sample azimuthal angle.

Furthermore, 2D-TIs alloys: binary, ternary, and quaternary with a specific composition have been investigated in many studies [65, 66]. For example, Kuznetsov et al. studied the THz emission proprieties of the $Bi_{2-x}Sb_xTe_{3-y}Se_y$ (BSTS) topological insulators films grown by the MOCVD method on a sapphire substrate, with different thicknesses and chemical compositions [65]. They conclude the THz radiation intensity depends on the film thickness, and the highest THz radiation intensity observed in the island film has a total thickness of about tens nanometers with a composition close to the Ren's curve.

Overall, the TIs antenna is very proposing for THz emitters because they are low cost, extremely thin, and easy to integrate into nano-photonic devices. Also, their conversion efficiency is comparable to state-of-the-art THz emitter and could be further improved, for example, by applying plasmon gratings.

4.2 Heterostructures

Heterostructures promise to be an enabling key for novel optoelectronic devices. There are two types of heterostructures: 1) Van der Waals (vdW) heterostructures are composed of different 2D and 2) heterogeneous integration with traditional materials and devices.

Van der Waals heterostructures and devices made by stacking different 2D crystals on top of each other attract many researchers as their properties change, leading to novel hybrid properties and enhancement of electronics and optoelectronic properties [42, 67]. Many THz emitters based on van der Waals heterostructures have been proposed so far. MoS_2 monolayer sandwiched between two graphene films reveals an extraordinary enhancement in the transient THz response compared to monolayer MoS_2 and graphene [68]. The enchantment was attributed to the THz response in this heterostructure produced from the sub-bandgap below the band gap of MoS_2. Moreover, THz pulses with a bandwidth of 2.5 THz have been reported based on $MoSe_2$ and ReS_2 vdW heterostructure [69]. In this study, they used different stacking orders and measurement techniques to address the photocarrier dynamics. The emitted THz pulse is reversed by the interchange of the stack ordered, indicating that the

THz radiations originate from the plane current. The corresponding frequent spectrum of the waveforms has a peak at 1.0 THz and a bandwidth of 2.5 THz. Other vdW heterostructures such as WS_2/hBN have also been studied for THz photoconductivity [70]. Far infrared and THz emitting diode also has been proposed based on graphene/black phosphorus (BP) and graphene/MoS_2 by Ryzhii *et al*. [71].

One of the advantages of 2D materials is that they are easy to integrate with traditional optoelectronic materials, systems, and devices. These heterogeneous integrations enable many novel phenomena and applications, particularly in the THz regime. Typically, the incorporation of 2D materials into other structures is accomplished by either direct growth or thin-film transfer methods. Graphene transferred to SiO_2/Si substrate was used to study the THz emission spectroscopy on the graphene-based interface and evaluate the interface build-in potential and trapped charge dynamics [72]. In fact, various heterogeneous integrations for many THz applications including MoS_2/Si and direct growth of Bi_2Se_3 onto GaAs substrate [50, 73].

The most advanced type of heterostructure-based THz emitter is the spintronic emitter, which offers many advantages, including ultra-broad bandwidth, high efficiency, and ease of control of radiation parameters through polarization, amplitude, and phase. Typically, the THz emitter devices are made of ferromagnetic and nonmagnetic (FM/NM) thin metal film heterostructures to generate the THz radiation when the heterostructures are irradiated with optical femtosecond laser pulses [74]. One of the main advantages of 2D materials is the presence of spin-orbit coupling, which is critical for spintronics THz emitters [75, 76].

An efficient spintronic THz emitter based on ferromagnet/heavy metal/topological insulator heterostructures was demonstrated. The THz radiation amplitude of the Co/Bi/Bi_2Te_3 heterostructure increases by 198% compared to the device without an intercalated Bi film, as shown in **Figure 9** [77].

Highly efficient THz emission from the Bi_2Se_3/Co heterostructures modified with ultrafast spin injection has been reported [78]. The Bi_2Se_3/Co heterostructure reveals a much larger THz pulse amplitude compared to a single film. Moreover, efficient spin current injection from Co to monolayer MoS_2 at Co/MoS_2 structures via THz emission has been demonstrated [79]. The sample was irradiated with 50 fs linearly 800 nm laser pulses and the magnetization direction of Co was controlled

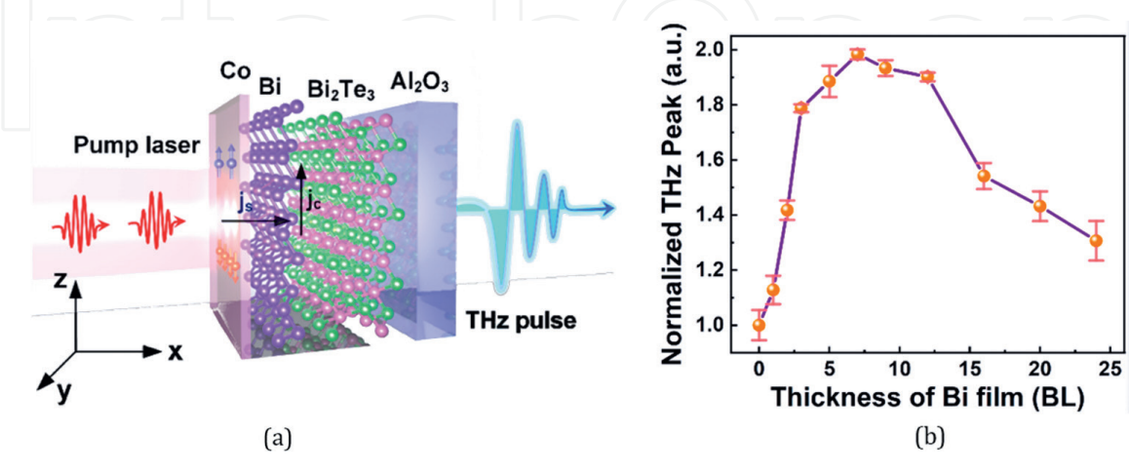

Figure 9.
(a) The schematic of the experimental measurements for THz radiation on the Co/Bi and (b) normalized THz peak amplitudes as a function of Bi film thickness. "Reprinted (adapted) with permission from ref. [77]. Copyright {2021} American Chemical Society."

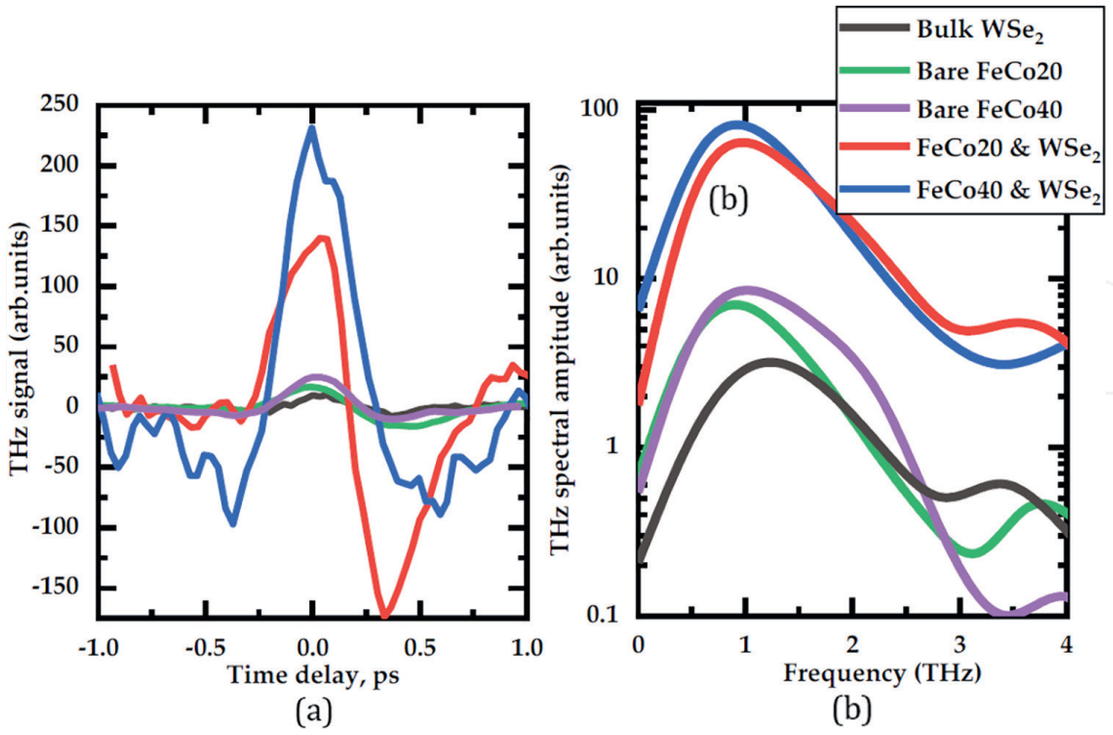

Figure 10.
(a) The THz radiation obtained by various structures in time domain and (b) the correspondent frequency spectrum [80].

by an external magnetic field. The detected THz radiation from Co/MoS_2 is strong and has an amplitude of ~2% compared to the THz emission of 1-mm thick ZnTe at the same pump fluence. Ultimately, increasing the efficiency of WSe_2/FeCo heterostructures, spintronics THz emitter has been demonstrated in reflection geometry [80]. **Figure 10** compares the THz radiation of various samples to unveil the enhancement of the THz emissions.

Overall, the 2D materials have been investigated toward high THz radiation efficiency as well as reduce the cost of the photoconductive materials. However, 2D materials are still awaiting full exploitation. Although the device fabrication is very similar to other materials-based emitters, many challenges still remain to realize efficient THz devices, such as obtaining materials with very high charge carrier mobility, low cost, and scalable. For instance, a number of research activities start on other new materials, such as gallium telluride (GaTe) [81]. Moreover, many 2D materials preparation and fabrication technologies are discussed in Section 3.4 to address many challenges related to the quality of the produced materials and scalability.

5. Conclusion

This chapter presented the 2D-material-based THz sources, particularly the photoconductive THz emitter. Several 2D materials have been employed as photoconductive materials and device structures. First, the chapter covers the basic principle of photoconductive terahertz emitter and the properties of 2D physical materials. In the end, the chapter discusses the 2D material-based THz emitters, where many forms of

2D materials and device structures are presented, including layered 2D materials and homogenous and heterogeneous heterostructures.

Conflict of interest

The authors declare no conflict of interest.

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