Dirac terahertz plasmonics in two and three dimensions

Kelvin J.A. Ooi a,b,*, Y.S. Ang c,∗∗, Q. Zhai a,b,d, X. Sun a,b,d, P. Xing e, C.K. Ong f,g, L.K. Ang c, Dawn T.H. Tan c

a School of Energy and Chemical Engineering, Xiamen University Malaysia, Selangor Darul Ehsan 43900, Malaysia
b College of Chemistry and Chemical Engineering, Xiamen University, Xiamen 361005, China
c Science and Mathematics, Singapore University of Technology and Design, 8 Somapah Road, Singapore 487372, Singapore
d School of Electrical and Computer Engineering, Xiamen University Malaysia, Selangor Darul Ehsan 43900, Malaysia
e Photonic Systems and Devices Group, Engineering Product Development, Singapore University of Technology and Design, 8 Somapah Road, Singapore 487372, Singapore
f Department of Mathematics and Applied Mathematics, Xiamen University Malaysia, Selangor Darul Ehsan 43900, Malaysia
g Department of Physics, National University of Singapore, 2 Science Drive 3, Singapore 117551, Singapore

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A B S T R A C T

Material science at the beginning of the twenty-first century has largely been dominated by the study of graphene and other Dirac-type materials. Interesting physics manifest in the linear electronic bandstructure of Dirac-type materials, more so on the behavior of photons interacting with it. Moreover, with the recent discovery of three-dimensional Dirac semimetals (3DS), dimensionality becomes an important factor in the overall terahertz response. In this brief review, we look at some of the established physics behind the terahertz plasmonic response in Dirac materials, the contributions from their linear electronic bandstructure and dimensionality, as well as the current progress in device design and implementation. In particular, the strong terahertz response of Dirac material would have important implications in the development of terahertz-based communication systems and Internet-of-Things applications.

0. Introduction

The scientific field of electromagnetic waves has traditionally been dichotomized into two distinct domains: the low frequency spectrum belongs to the realm of radio and microwave engineers, while the high frequency spectrum is reigned by the optical and high energy physicists [1,2]. In between those two spectra lies a range of frequency which is less explored, called the terahertz band. There are many reasons that terahertz applications remain elusive till today, one of them being the dearth of terahertz sources available commercially and the absence of discovery of materials that responds well to the terahertz frequency [1–3]. For past decades till today, there has been a clarion call for the continual research efforts to bridge the terahertz gap (Fig. 1) [2–7], as the ubiquitous wireless communications technologies and the recent growth of the Internet-of-Things (IoT) had crowded out the range of usable frequency in the industrial, scientific and medical (ISM) radio bands, thereby pushing future applications into the 6G wireless technology, which would likely involve the use of the terahertz band [8].

Before the age of Dirac materials, the absence of materials with good terahertz response would only mean that these materials have to be created by the scientists and engineers. This approach has been adopted widely since the popularization of plasmonics [9], which served as the precursor to terahertz metamaterials [10]. Metamaterials are assemblies of multiple elements fashioned from composite materials such as metals and plastics, and they derive their properties from designer structures with precise shape, geometry, size, orientation and arrangement. However, since metamaterials rely on resonant patterns and arrays to create the terahertz frequency response, they suffered from narrow bandwidth, and hence most research efforts in this area were directed towards the extension of the device response bandwidth [11–16].

Everything has since changed when graphene entered the scene. In the early 1940s, graphene has been postulated to possess a unique bandstructure which is two-dimensional (2D) and linear in nature [17]. 60 years later, the first monolayer or few-layer graphene was being successfully isolated and prompted experimental investigation into its unique electronic properties [18,19]. But perhaps the most interesting development throughout graphene’s fifteen years of intense research in electronics is the companion discovery of graphene’s strong response to photons in the terahertz band [20–26]. The strong terahertz response of graphene had established a whole new area of research called graphene...
plasmonics [27], and through a confluence of factors including its linear electronic band structure, two-dimensional thin film structure, as well as environmental substrate-doping of carrier densities, places its optical response right amidst the terahertz gap.

Notwithstanding graphene’s reign as the wonder material of terahertz applications, its handling of fabrication and integration with devices and components still remained a challenge as of today. Though laboratory-scale chemical vapor deposition (CVD) graphene growth techniques has been perfected over the years [28], large-scale mass production capabilities of monolayer and defect-free films are still elusive [29–31]. Concurrent with the efforts to improve graphene’s production scalability and device integration, a separate line of thought emerged, in which the obvious solution is to search for a “three-dimensional (3D) graphene” analogue. In 2011, Burkov et al. initiated an idea of a bulk-type of topological nodal semimetals [32], where surface topologies of the semimetal would give electronic transport properties similar to graphene. In 2012, Young et al. also showed that the relativistic transport properties of graphene could theoretically be reproduced in three dimensions through ab-initio calculations [33]. Finally, recent years saw experimental discovery of these topological semimetals in cadmium arsenide [34–36], sodium bismuth [37], tantalum arsenide [38], calcium telluride [39], and platinum telluride [40]. These classes of materials are called bulk Dirac semimetals (BDS), or more familiarly referred to as three-dimensional Dirac semimetals (3DS).

In this brief review, we would look at the terahertz plasmonic response of Dirac materials in two and three dimensions. We set forth to identify and delineate the contributions from the Dirac bandstructure and material dimensionality on the strong terahertz plasmonic response, and that would serve as a guide on constructing terahertz devices based on Dirac-type materials.

1. Linear optical conductivity of Dirac materials

The description of the linear optical conductivity (the linear response to electromagnetic fields) of graphene originates from the Kubo model. Within the local response limit, it can be succinctly written as [25,26]

\[
\sigma^{(1)}(\omega) = \frac{ie^2}{\pi\hbar^2(\omega + i\nu_1)} \left[ \frac{E_F}{2k_FBE} + \ln \left( \frac{2\exp \left( -\frac{|E_F|}{k_BT} \right) + 1}{2\exp \left( -\frac{|E_F|}{k_BT} \right) - 1} \right) \right] + \frac{e^2}{4\pi} \left[ \frac{1}{2} + \frac{1}{\pi} \tan^{-1} \left( \frac{\hbar(\omega + i\nu_2) - 2|E_F|}{2k_BT} \right) \right] - \frac{e}{2\pi} \ln \left( \frac{\hbar(\omega + i\nu_2) + 2|E_F|}{\hbar(\omega + i\nu_2) - 2|E_F|} \right) \tag{1}
\]

Parameters that described the linear optical conductivity in Eq. (1) include the electronic charge, \(e\), the reduced Planck’s constant, \(\hbar\), the Boltzmann constant, \(k_B\), the environmental temperature, \(T\), the angular frequency, \(\omega\), the electronic scattering frequencies, \(\nu_1\) and \(\nu_2\), and last but not least, the Fermi level of graphene, \(E_F\).

On the other hand, recent studies on 3DS material systems have their linear optical conductivities also derived from the Kubo formula as follows [41,42]

\[
\sigma^{(1)}_{3DS}(\omega) = \frac{2}{3} \cdot \frac{ie^2(k_BT)^2}{\pi\hbar^2v_F(\omega + i\nu_1)} \left\{ 2\ln \left( \frac{2 - \exp \left( -\frac{|E_F|}{k_BT} \right)}{2 - \exp \left( -\frac{|E_F|}{k_BT} \right) + 1} \right) \right\} + \frac{e^2}{4\pi} \left[ \frac{1}{2} + \frac{1}{\pi} \tan^{-1} \left( \frac{\hbar(\omega + i\nu_2) - 2|E_F|}{k_BT} \right) \right] - \frac{e}{4\pi} \ln \left( \frac{\hbar(\omega + i\nu_2) + 2|E_F|}{\hbar(\omega + i\nu_2) - 2|E_F|} \right) \tag{2}
\]

\(\nu_1\) and \(\nu_2\) are the second-order polylogarithm. Most of the parameters that described graphene’s linear optical conductivity are applicable here. Two additional parameters that describe the linear optical conductivity of 3DS in Eq. (2) are the Fermi velocity, \(v_F\), and also the cutoff energy, \(E_C\), beyond which the Dirac spectrum is no longer linear.

Comparison of Eqs. (1) and (2) would show us that the form of the Dirac linear optical conductivities in two and three dimensions are similar. Performing dimensional analyses on both equations would yield the unit of Siemens (S) for Eq. (1) and Siemens-per-meter (S/m) for Eq. (2), which corroborate well with the physical and electronic dimensions of the respective material systems. In both equations, the
first term describes the intraband optical response of the material, which would be relevant to plasmonics, while the second term describes the interband optical response. Several interesting differences between those two equations are also noted here, for example, that graphene’s intraband conductivity is proportional to the Fermi level, while that of 3DS is proportional to the square of the Fermi level. Also, the interband optical response of 3DS changes with the angular frequency, which would cause a frequency-dependent absorption; while this parameter dependence is absent for graphene, which resulted in graphene’s iconic universal absorption coefficient of $\alpha = 2.3\%$ for all frequencies [43]. These slight differences also culminate in the different optical transition frequency points for graphene ($\hbar \omega_{F} = 1.67$) and 3DS ($\hbar \omega_{F} = 1.23$) as shown in Fig. 2 [41], at which those material systems transits from metallic to dielectric behavior. Nonetheless, such differences would not concern us within the scope of our discussion, as the plasmonic response usually occurs in the near-terahertz regime within the intraband optical response only.

2. Design freedom of plasmonic response

The plasmonic response of Dirac materials can be described by their plasmon dispersion equation. For graphene, the plasmon dispersion is two dimensional, written as [24]

$$\frac{\epsilon_1}{\sqrt{q^2 - \omega^2/c^2}} + \frac{\epsilon_2}{\sqrt{q^2 - \omega^2/c^2}} = \frac{\sigma^{(1)}(\omega)}{\epsilon_0 \omega}$$  \hspace{1cm} (3)

The parameters for the equation include the background material permittivities $\epsilon_1$ and $\epsilon_2$, permittivity of free space, $\epsilon_0$, speed of light, c, and the surface plasmon wave vector, q. The 2D plasmon dispersion has no dependence on graphene’s “thickness” with respect to its inherent nature, but one could always artificially recompose the dispersion equation into its 3D equivalent and take the limit of graphene’s thickness to its experimentally observed value (3.3 Å) [44].

Next, for 3DS the plasmonic dispersion would follow the classic equation [45]

$$\coth \left( \sqrt{q^2 - \epsilon_{3DS} d^2 c^2/\epsilon_1^2} \right) = -\frac{\epsilon_{3DS}}{\epsilon_1} \frac{\sqrt{q^2 - \epsilon_{1} \omega^2/c^2}}{\sqrt{q^2 - \epsilon_{3DS} d^2 c^2/\epsilon_1^2}}$$  \hspace{1cm} (4)

In the equation, $d$ is the thickness of the 3DS, and $\epsilon_{3DS}$ is the permittivity of the 3DS, related to the linear optical conductivity via $\epsilon_{3DS} = 1 + i\sigma^{(1)}(\omega)/\epsilon_0 \omega$. Interestingly, if we take the thickness dimension to its extremums, then for $d \rightarrow \infty$ we would obtain the interfacial surface plasmon dispersion relation, while for $d \rightarrow 0$ we can recover the dispersion relation close to that for graphene, as has been demonstrated shown in Fig. 3 [42]. We shall discuss the properties of 3DS based on the material cadmium arsenide (Cd$_3$As$_2$), which is a recently discovered 3DS that is stable and has a very high Fermi velocity of $1.5 \times 10^6$ m/s, a high electron mobility of 10,000 cm$^2$/V s at room temperature, and an electron scattering time of 0.1 ps [34,35], all phenomenological properties which closely resembled graphene.

This brings us to the discussion of the pertinent differences between graphene and 3DS: the degrees of freedom (DoF) in the design of their plasmonic response. For graphene, there are three DoFs. The first DoF is obviously the Fermi level. Electrostatic gating is a common and robust method to dynamically tune graphene’s Fermi level [19], and the relationship between the Fermi level and electric field is that of the gate-capacitor model [47]. The second DoF arises from the finite graphene widths, which produces plasmonic edge-modes as shown in Fig. 4 [46]. As long as graphene’s width is finite, the breadth of the ribbon would change the plasmonic propagation modes, with the electric fields bounded by the edge boundary conditions. The third DoF would be the finite length of graphene, when scaled to nanometer-dimensions, produces graphene quantum dots (GQD) [48–51]. The size of the GQD would affect its resonant response with the electromagnetic field frequency, and this is often employed when designing broadband graphene absorbers.

All the three DoFs for graphene plasmonics are applicable to 3DS as well. Yet, 3DS possesses a fourth DoF: the thickness of the material is also a variable in its plasmonic response. Referring back to Fig. 3, the plasmonic response of the bulk 3DS ($d \rightarrow \infty$) has a much smaller wave-vector compared to that of 3DS thin films (e.g. $d = 20$ nm). The fourth DoF would grant engineers a huge leeway to design a plethora of terahertz devices, and solve many of the terahertz free-space coupling and phase-matching issues due to the high plasmon wave-vector of graphene [52–54]. Fig. 5 shows the reconfigurable plasmon
Fig. 5. Reconfigurable plasmonic response of cadmium arsenide via (a) film thickness and (b) Fermi level. Corresponding propagation lengths in (c) and (d).

dispersion curves and propagation lengths for cadmium arsenide by both film thickness and Fermi level respectively. Confinement factors over 10 can be achieved for those film thickness and Fermi levels, while the plasmons could propagate a few micrometers to sub-millimeter distances.

3. Dirac terahertz devices

A decade of research into graphene photonics and plasmonics has yielded a sizable amount of literature, designs, and experimental prototypes of graphene-based terahertz devices. Most of the devices discussed in this section are based on the interaction between the electron and photon or plasmon, primarily through tuning the Fermi level of graphene via electrostatic-gating. These devices can be categorized into four main types: the electrical terahertz sources, which generate the plasmons or photons from electrical energy; the modulators and switches, which provide electro-optical control over the graphene plasmons; the detectors and sensors, which convert terahertz energy to electrical energy; and the resonant passive devices like filters, splitters, polarizers and couplers, which passively select the frequencies and modes of the graphene photons and plasmons via designed and patterned terahertz structures.

Electrical graphene terahertz sources can be categorized into two main types as shown in Fig. 6. The first type is plasmons generated by high energy, often relativistic swift electrons that either travel close to the surface of graphene, or bombard graphene directly [55,57–62]. The bandwidth of these plasmons are generally narrow and near the surface plasmon resonance frequency, as the swift electrons can only excite plasmons of the same or lower velocity factor [63]. The second type is plasmons generated by inelastic electron tunneling from a metallic tip to the graphene’s surface [56,64–67]. These plasmons are generally of lower intensity but have a broader bandwidth.

The design of graphene modulators and switches takes advantage of the dynamic reconfigurability of graphene’s Fermi level via electrostatic-gating, often at ultrafast speeds if the gate thickness is small enough. Modulation occurs either through the absorption and quenching of the plasmon propagation modes [69–75], or through splitting and routing the plasmons to different locations [76–78]. Interestingly, electro-optic modulation of graphene not only modulates its absorption coefficient, its phase response is significantly altered as well. Hence, combination of both effects enables design of graphene plasmonic logic gates [68,79–85]. The gate architecture making full use of both the phase and absorption modulation has been illustrated by Ooi et al. (Fig. 7) [68]. Here, the ability to dynamically tune graphene’s optical conductivity enabled a large tuning of plasmonic mode and thus simultaneously provide for cut-off and interferometric states. As a result, it is possible to construct NOR gates by utilizing the cut-off states in serial arrangement, NAND gates by utilizing cut-off states in parallel arrangement, and XNOR gates using interferometric states in Mach–Zehnder arrangement. Graphene plasmonic logic gates that rely on other forms of modulation have been studied as well, which includes magneto-optical effects [86], and all-optical modulation schemes [87–90].

On the other hand, the high unit-volume absorption coefficient of graphene made it a preferred material for photo-transduction devices like detectors and sensors [92,93]. Patterning graphene into antenna and cavity structures would give it high frequency-selective absorption of terahertz waves [94–98]. At the same time, graphene’s chemical inertness and high molecular adsorptivity, coupled with the large plasmon wave vectors, made for advantageous construction of highly sensitive biochemical plasmonic sensors (Fig. 8) [91,99]. Terahertz waves that is coupled via a Kretschmann prism excites the plasmonic modes along the gold–graphene interface. The sensitivity of the sensor is increased tremendously due to graphene’s high absorption coefficient.

Lastly, we discuss the graphene plasmonic passive elements consisting of filters [100–103], couplers [104–106], polarizers [107–109], and splitters [110]. These passive devices work the same way like their conventional metal-plasmonic or dielectric-photonic counterparts, however, due to the tight optical confinement in graphene plasmonic waveguides, they can be made very short and compact. For example, terahertz graphene plasmonic filters described in reference [100] can be made very short (400 nm) with narrow band-pass linewidths. On
the other hand, the graphene terahertz plasmonic bends and splitters described in reference [110] have very sharp 90-degree bends that exhibits unity transmittance, with overall device size in the sub-nanometer.

As for 3DS, an assortment of plasmonic devices have recently been studied and proposed. Photodetectors (Fig. 9), absorbers and sensors are the most commonly studied due to 3DS’s increased light-matter interaction compared to graphene [111–118]. In Fig. 9, Cd$_3$As$_2$ nanowires has been photo-excited with electromagnetic waves in a broad spectrum of frequencies from the terahertz to optical ranges, and the intensity of the photocurrents generated is high. Generation of electromagnetic waves is studied from the aspect of induction from electric charges [119]. Terahertz filters based on 3DS have also been proposed, capitalizing on its tunable frequency-selective plasmonic response [120,121].

4. Nonlinear optical conductivity of Dirac materials and all-optical devices

Graphene nonlinear optics has been a decade-old, established field of study. Theoretical formalism to study the nonlinear optical properties of graphene has largely been categorized into three types. The first would be the simplest semiclassical Boltzmann transport supplemented by the Dirac-equation model, which has been studied in detail by Zhang’s group [122–129]. The second type is the approach from the semiconductor Bloch equations, primarily studied by Cheng and Sipe [130–136]. While the first two types are perturbative models, which suffer from divergences when the Fermi level is low, the third type is a non-perturbative model that studies the time-dependent electron dynamics in graphene [137–140]. Nevertheless, for the simplicity of discussion, here we present a simple analytical model derived from the first type of formalism. Graphene’s intraband Kerr nonlinear optical
conductivity is written as [123]

$$\sigma^{(3)}(\omega) = \frac{3}{4} \cdot \frac{i e^2 r_F^2}{\pi \hbar^2 (\omega^2 + v^2)(2\omega + i\nu_1)} \int_0^\infty \frac{1}{\epsilon_k} \frac{\partial f_0(\epsilon_k, E_F, T)}{\partial \epsilon_k} d\epsilon_k$$  \hspace{1cm} (5)

There has also been very recent studies on the nonlinear optical properties of 3DS materials [141–144]. If we apply the same theoretical formalism as above to 3DS, the result could be written as [42]

$$\sigma^{(3)}_{3DS}(\omega) = \frac{2}{5} \cdot \frac{i e^2 e_F}{\pi^2 \hbar^2 (\omega^2 + v^2)(2\omega + i\nu_1)} \cdot \frac{1}{1 + \exp \left(-E_F/k_BT\right)}$$  \hspace{1cm} (6)

Eqs. (5) and (6) exhibits very similar forms. A plot of the nonlinear optical properties (Kerr coefficients) in Fig. 10 would reveal that their trends are similar, only differing in magnitude. Again, this shows that

the main difference between graphene and 3DS is the additional design DoF that the latter enjoy.

Graphene nonlinear optics has been applied to a multitude of all-optical terahertz device designs. Kerr nonlinear switching forms the bulk of the nonlinear graphene terahertz devices [145–153], taking advantage of its high Kerr coefficients for very low-powered switching. High harmonic generation (the three-photon process) is also an important phenomenon to generate a spectrum of terahertz waves of different frequencies [154–158]. Other nonlinear wave phenomena that are commonly seen include four-wave mixing and super-continuum generation [159–163], as well as soliton propagation [164–168]. A more detailed review on nonlinear graphene plasmonics can be found in reference [169].

On the other hand, nonlinear optics of 3DS is still a nascent field. The first devices experimentally demonstrated are the saturable absorbers [116,170]. Other devices proposed include self-phase modulation and Kerr switching [42,171,172]. Nonetheless, given the huge activity of research in 3DS nonlinear optics, the prospect for exponential growth in literature is indeed certain.

Conclusions and perspectives

We have reviewed and highlighted the interesting terahertz physics behind graphene and 3DS. Dirac-type materials, regardless of their bandstructure dimensionality, exhibit more or less the same terahertz behavior that is distinct from conventional parabolic-band dielectrics and metals. Their notable common features include the exceptionally remarkable optical response to the terahertz spectrum, and also the dynamic reconfigurability of their optical conductivities via the tuning of the Fermi-level. These two features no doubt place Dirac materials in the limelight of the 21st-century technological advancements, due to the fact it would bridge the terahertz gap to overcome present technological limitations, and also device performances based on Dirac materials are far more superior than conventional ones.

Thus the main difference between the terahertz response of graphene and 3DS lies in their physical dimensionality. 3DS materials enjoy four DoFs which consists of the three physical dimensions and a reconfigurable Fermi level. Graphene, on the other hand, is
restricted in one of the physical dimensions that limits its integrative versatility with bulk terahertz devices. This distinction manifests primarily in the highly-variable plasmonic dispersion relation of 3DS with its material thickness, which gives designers freedom to balance between optical confinement and phase-matching factors. Other benefits may include the easier handling of bulk materials in the fabrication foundries, hence, barriers-to-entry of incorporating 3DS into existing device processes would be lower.

Graphene and 3DS would inevitably take center stage in the epochal technological revolution of our times. The rivalry between these two material platforms would eventually anoint its victor, but regardless of the outcome, Dirac materials would be recorded as one of the most disruptive material technologies in history.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Kelvin J.A. Ooi: Conceptualization, Writing - original draft, Funding acquisition. Y.S. Ang: Conceptualization, Data curation, Writing - review & editing. Q. Zhai: Writing - review & editing. X. Sun: Writing - review & editing. P. Xing: Writing - review & editing. C.K. Ong: Writing - review & editing. L.K. Ang: Writing - review & editing. Dawn T.H. Tan: Writing - review & editing.

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