Nonlinear plasmonics of three-dimensional Dirac semimetals

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ABSTRACT
The three-dimensional Dirac semimetal (3D DSM) is a new class of material with a slew of electronic and optical properties in common with graphene, while structurally having a bulk form like real metals. In particular, the Dirac band structure of 3D DSM conferred very high optical nonlinearities much like the case for graphene. Consequently, we found that 3D DSM has respectable nonlinear plasmonic performance in comparison with graphene, while retaining the structural benefits of bulk metals, having reduced passive plasmonic losses, and is much easier to handle in fabrication facilities. 3D DSM is expected to play a strong role in providing strong optical nonlinearities for all-optical switching and at the same time offering a superior platform for nanophotonic device integration.

INTRODUCTION
The road to all-optical computing banks on the advancement of nonlinear optical materials. The all-optical transistor demands a good medium for light-on-light control, which is availed through materials with high Kerr coefficients, low optical losses, and a small form factor. Silicon and its CMOS compatible materials are currently the mainstay for nonlinear photonics due to engineering and economic reasons.1 Recent breakthroughs in CMOS nonlinear photonics see the enhancement of Kerr coefficients to high values of 10−17 m2/W,2,3 yet their physical dimensions remain large and hence prevent any meaningful implementation of chip-scale devices. On the other hand, metal-based plasmonic devices are marked by the absence of the diffraction limit, which enabled the miniaturization of waveguides to become compatible with chip-scale integration,4 and also come with the advantage of higher Kerr coefficients.5 Unfortunately, the optical loss of plasmonic waveguides is very high, and to overcome the propagation shortfall, nonlinear plasmonics usually requires optical pumping intensities upwards of GW/cm² to achieve the required modulation depth.6–9 Recent attention is now turned towards graphene which has a unique two-dimensional Dirac band structure that confers giant optical nonlinearity upwards of 10−11 m²/W.10–17 There is growing research interest in nonlinear graphene plasmonics,18 where large propagation wave vectors and light confinement factors enhance the nonlinear performance of the graphene plasmonic waveguide,19–26 with the downside of having low light coupling from other optical components and free-space optics. To date, excitation of graphene plasmons could only be done through optical scattering off a sharp metal tip with low excitation efficiencies.27,28

Recent forays into topological properties of condensed matter systems have seen the discovery of “graphene-analogs”
in 3D bulk materials, also called three-dimensional Dirac semimetals (3D DSMs) which host gapless Dirac nodal points in the bulk (see Fig. 1). At the vicinity of the Dirac nodal point, the electronic band structure disperses linearly in all three dimensions,\textsuperscript{29,30} thus, the quasiparticles around the Dirac nodal point have exotic pseudo-relativistic characteristics. In 2014, the existence of the topological Dirac nodal point in bulk materials has been experimentally confirmed in sodium bismuth (Na3Bi) by Liu et al.\textsuperscript{31} and in cadmium arsenide (Cd2As3) by Borisenko et al.\textsuperscript{32} and Neupane et al.\textsuperscript{33} In terms of photonic applications, recent progress has seen Cd2As3 being implemented as a saturable absorber for ultrafast mid-infrared lasers, traversing the same beginnings of its predecessor graphene.\textsuperscript{34,35} As the charge carriers are described by Dirac physics in both 2D graphene and 3D DSM, it is widely expected that 3D DSM would share similar Dirac-type optical properties with graphene, such as the tunability of its optical conductivity, strong light confinement, and high nonlinear optical coefficients. Furthermore, as a bulk material, 3D DSM has an additional structural degree-of-freedom advantage over graphene in the construction of plasmonic waveguides. In this paper, we derive analytical expressions of third-order optical processes. In this paper, we derive analytical expressions of third-order optical processes.

**FIG. 1.** Schematic drawing of the Dirac energy dispersion and the intraband optical processes. The three-dimensional Dirac semimetal hosts gapless Dirac nodal points in the bulk.

**LINEAR AND NONLINEAR OPTICAL PROPERTIES OF 3D DIRAC SEMIMETALS**

To calculate the 3D DSM’s linear intraband optical conductivity, we employ the semiclassical Boltzmann transport equation under the relaxation time approximation, i.e., $\tau(\epsilon_k) = \tau$. Then, the linear intraband optical conductivity can be analytically solved as (see the supplementary material)

$$\sigma_{xx}^{(0)}(\omega) = \frac{\tau}{3\pi e^2 h} \left[ \frac{3(\epsilon_k T)^2}{\omega^2 + \frac{3}{2} \omega} \right]$$

(1)

where $\text{Li}_2(z)$ is the polylogarithm and $\sigma_0 = e^2/4\hbar$.

To calculate the third-order nonlinear conductivity, we consider an external field, $E = \frac{\epsilon_0}{2}(e^{i\omega t} + e^{-i\omega t})$, and then recursively solve the higher-order distribution functions (up to third order in $\omega$)\textsuperscript{36-38} (see the supplementary material). Finally, we obtain the third-order Kerr nonlinear optical conductivities which are given by a remarkably simple analytical expression

$$\sigma_{xx}^{(3)}(\omega) = \sigma_{yy}^{(3)}(\omega) = \sigma_{zz}^{(3)}(\omega) = \sigma_0 \frac{8\epsilon_0 e^2}{\sqrt{\epsilon_0}} \frac{\tau^3}{\omega^3} \left[ \frac{1}{1 + \exp\left(-\frac{\epsilon_k T}{k_BT}\right)} \right]$$

(2)

Note that the intraband optical conductivities derived above are valid in the optical frequency regime of $f < 2\Sigma/3$. Higher frequencies may require the interband optical conductivities to be included for an accurate description of the nonlinear optical processes.

While the intraband optical conductivities for 3D DSM and graphene are both derived from the semiclassical Boltzmann transport equation, the one important distinction between them would be the dimensionality of the conduction equation, which would strongly affect their surface plasmon dispersion relations. For graphene, the 2D optical conductivity would result in a dispersion relation for conductive surfaces, written as follows:\textsuperscript{39}

$$\frac{\epsilon_1}{\sqrt{q^2 - \frac{\epsilon_1}{c^2}}} + \frac{\epsilon_2}{\sqrt{q^2 - \frac{\epsilon_2}{c^2}}} = \frac{i\sigma_0^{(0)}(\omega)}{\epsilon_0\omega}.$$  

(3)

Notice that there is no parameter for the sheet “thickness” in the 2D dispersion relation, and consequently, there is the absence of macroscopic concepts like the refractive index and nonlinear optical Kerr coefficients. To obtain those parameters, the usual practice is to apply the effective medium theory, taking into account that graphene physically exists as a 0.3 nm carbon sheet. The effective medium theorem for graphene has been successfully applied and benchmarked with experimental results, for example, in the studies of Vakil and Engheta for linear plasmonics\textsuperscript{40} and Cheng et al. for nonlinear optics.\textsuperscript{51,52}

Whereas, for 3D DSM, the volume conductivity can be straightforwardly applied into the surface plasmon dispersion relation as\textsuperscript{8}

$$\text{coth}\left(\sqrt{q^2 - \epsilon_{DSM}\omega^2/c^2}\frac{d}{2}\right) = -\frac{\epsilon_{DSM}}{\epsilon_d} \frac{\sqrt{q^2 - \epsilon_d\omega^2/c^2}}{\sqrt{q^2 - \epsilon_{DSM}\omega^2/c^2}}.$$  

(4)
where \( d \) is the thickness of the 3D DSM waveguide.

For the simplicity of analysis, we select only the Fermi level of 0.1 eV for both graphene and 3D DSM and only the ultrafast Kerr coefficient (which corresponds to the 0.2 ps excitation pulse) for gold. For 3D DSM, our selected material of study is cadmium arsenide, which is one of the recently discovered 3D DSM which is stable and has a very high Fermi velocity of \( 1.5 \times 10^6 \) m/s, a high electron mobility of \( 10,000 \text{ cm}^2/(\text{V s}) \) at room temperature, and an electron scattering time of 0.1 ps.\(^{32,33}\) It is notable that all these figures closely resemble that of graphene. It should also be noted that cadmium arsenide possesses anisotropic conductivity in the \( z \)-direction,\(^{43}\) but for plasmonic waves, this property is not important as electromagnetic waves would not penetrate the bulk of the 3D DSM. Figure 2 shows their respective surface plasmon dispersion curves and propagation lengths. Plasmon characteristics of a waveguide is better defined as the curve moves further away from the light line, up to the limit of the surface plasmon resonance frequency, as shown in Fig. 2(a). It denotes a buildup of large surface plasmon wave-vectors and hence high plasmonic waveguide indices that leads to high optical confinement. When the waveguide’s structure is a thin film, the strongly coupled modes from the plasmonic fields on both sides of the film serve to substantially increase the plasmonic waveguide index and optical confinement. While the increase in optical confinement and waveguide index is generally welcome, the trade-off would be the associated increase in surface plasmon losses. For example, from Fig. 2(b), we observe that the surface plasmon propagation lengths of cadmium arsenide could differ by up to three orders between a bulk and a thin film form. The same is observed for bulk gold and thin gold films, while graphene, with the thickness of an atomic layer, shares the same surface plasmon propagation lengths as cadmium arsenide thin films.

The nonlinear optical properties are computed and analyzed for different classes of materials. We study cadmium arsenide for the 3D DSM material, and for graphene itself, we will study for optical nonlinearity by intraband contribution, which was primarily researched by Zhang’s group.\(^{36,44-47}\) The intraband optical nonlinearity would be a useful approximate for highly doped Dirac semimetals, while for that of low-doped or undoped Dirac semimetals, the interband nonlinear contributions will dominate. As such, here we limit our frequency range of study to only 1–5 THz for both cadmium arsenide and graphene, which is very far away from the interband transition frequency regime. Finally, for real metals, we selected gold for the analysis due to its ubiquity in the literature and the availability of data for various excitation pulse durations that was summarized by Boyd et al.\(^{5}\)

Figure 3 shows the computed nonlinear Kerr optical coefficients \( n_2 \) and \( k_2 \), respectively, for the three materials in question. \( n_2 \) is the real part of the Kerr coefficient, while \( k_2 \) is the imaginary part related to the Kerr optical losses. Figures 3(a) and 3(b) show \( n_2 \) and \( k_2 \), respectively, for cadmium arsenide, while Figs. 3(c) and 3(d) depict the same for graphene. The trends for the Kerr coefficients for these two materials are markedly similar. Both the real and imaginary coefficients are invariably negative, signifying a defocusing as well as a saturable absorption effect. Finally, for gold, we examine the case for three different excitation time scales 0.2 ps (ultrafast), 30 ps, and 710 ps, respectively. These are experimentally obtained data which are conducted only at selected frequencies; thus, we only limit our study in a narrow frequency range of around 500–560 THz. The biggest effect from the different time scales of excitation would be the presence of hot electrons, which are usually the largest contributors to the nonlinear susceptibility.\(^{38,43}\) Figures 3(e) and 3(f) depict orders of magnitude differences between the nonlinear coefficients for gold excited by short and long pulses. The ultrafast Kerr components of gold only give low
nonlinear coefficients in the range of $10^{-14}$ m$^2$/W, while the slower thermal processes can contribute up to high ranges of $10^{-10}$ m$^2$/W.

**NONLINEAR PLASMONICS OF 3D DIRAC SEMIMETALS**

The confluence of both the materials' linear and nonlinear contributions would be revealed in their nonlinear plasmonic response. The nonlinear surface plasmon indices, $n_{2\text{-plasmon}}$ and $k_{2\text{-plasmon}}$, are calculated using the standard definitions (details are available in the supplementary material) and are presented in Fig. 4. Conforming to our expectations, all the thin film DSM plasmonic waveguides, which include cadmium arsenide, graphene, and intra-graphene, show very high nonlinear surface plasmon indices. This is due to the effect of high optical confinement, and thus, the plasmons are very sensitive to the small changes in the material refractive indices. Contrariwise, if the waveguide is in bulk form, then the lower-confined plasmon would be less sensitive to the nonlinear refractive index changes, as can be observed from the four-order magnitude difference between the nonlinear plasmon indices of different cadmium arsenide waveguide structures.

However, one setback for 3D DSM is the positive-valued nonlinear surface plasmon extinction coefficient, as shown in Fig. 4(b). This means that any increase in the plasmonic waveguide index will be accompanied by a comparable amount of increase in the plasmonic losses as well. Graphene and gold in our discussion show the same positive-signed $k_{2\text{-plasmon}}$, and this would have a potential impact on limiting the nonlinear phase accumulation for the plasmonic waveguides, which will be discussed next.

Figure 5 shows the nonlinear switching performance for the respective materials and waveguide structures. For the
purpose of this analysis, the frequency is fixed at 3 THz for the Dirac semimetals and 500 THz for gold. Phase switching, i.e., the accumulation of π-phase shift in order to perform interferometric modulation, is shown in Fig. 5(a). Both cadmium arsenide and graphene would never achieve the required phase accumulation due to the in-tandem shortening of the effective propagation length. For gold, due to \( k_{2{-\text{plasmon}}} < n_{2{-\text{plasmon}}} \), π-phase accumulation is fairly attainable albeit at very high optical intensities of 10 GW/cm\(^2\).

Nevertheless, the high nonlinear extinction efficiency of 3D DSM makes it a promising extinction modulator. From Fig. 5(b), 100% of extinction modulation depth is achieved with 0.3 MW/cm\(^2\) optical intensities, and increasing to 1.0 MW/cm\(^2\) pushes this figure to almost 1000% or, in other words, over 10 times reduction of the effective propagation length. Graphene compares as favorably demonstrating high extinction modulation depths at even lower optical intensities below 0.1 MW/cm\(^2\). As for gold, the extinction modulation depth would saturate at the value of around 50%.

**CONCLUSIONS AND OUTLOOK**

There is without doubt that in terms of the optical nonlinear performance, graphene plasmonics still reigns supreme over most nonlinear materials discovered to date. However, from the standpoint of the device platform and integration, 3D DSM still has an important role to play. First, for passive optical components, graphene plasmonics is very disadvantageous for its short propagation length below 1 \( \mu \text{m} \), and this is inimitable due to its thin film structure. Whereas for 3D DSM, its bulk structure offers a great degree of flexibility to design the waveguide parameters, with possibilities of propagation lengths of over 100 \( \mu \text{m} \). Second, bulk materials are easier to handle than thin films in fabrication facilities, just like how real metals such as gold and copper have been successfully used as ingredients in the semiconductor foundries. Thus, in nanophotonic applications, we judged that 3D DSM handily outclasses its metal counterparts and will remain a strong competitor and alternative to graphene–based devices.
Finally, we note that in recent times, the study of the exotic band structure in bulk materials has been expanded to cover a whole zoo of 3D topological semimetals, for example, the Dirac nodal point, loop, net, hourglass, double-helix, knot, and many others yet to be revealed. The question of how these unusual topological electronic band structures would intertwine with their optical properties still remains to be answered. We anticipate that the unique classes of 3D topological semimetals will continue to surprise us with exotic optical phenomena that are useful for photonics applications.

SUPPLEMENTARY MATERIAL

See supplementary material for details of derivation of the linear and nonlinear conductivity of 3D and 2D Dirac semimetals, as well as formalisms for analysis of nonlinear plasmonic waveguides.

REFERENCES


