Highly Efficient Terahertz Generation Using 3D Dirac Semimetals

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It is shown that three-dimensional Dirac semimetals are promising candidates for highly efficient optical-to-terahertz conversion due to their extreme optical nonlinearities. In particular, it is predicted that the conversion efficiency of Cd$_3$As$_2$ exceeds typical materials like LiNbO$_3$ by >5000 times over nanoscale propagation distances. Studies show that even when no restrictions are placed on propagation distance, Cd$_3$As$_2$ still outperforms LiNbO$_3$ in efficiency by >10 times. The results indicate that by tuning the Fermi energy, Pauli blocking can be leveraged to realize a step-like efficiency increase in the optical-to-terahertz conversion process. It is found that large optical-to-terahertz conversion efficiencies persist over a wide range of input frequencies, input field strengths, Fermi energies, and temperatures. These results could pave the way to the development of ultrathin-film terahertz sources for compact terahertz technologies.

1. Introduction

High-energy, single-cycle terahertz (THz) pulses are essential for both fundamental studies and applications, such as materials analysis,[1–3] 6G communications,[4] electron acceleration,[5] and high-resolution spectroscopy.[6] A common approach to realize high-energy, few-cycle terahertz pulses from optical pulses is optical rectification, which exploits the second-order nonlinearity of materials such as LiNbO$_3$ (LN), GaAs, ZnTe, GaP, and so on.[7] In particular, LN is widely used to generate terahertz pulses in the range of 1 THz due to its relatively strong nonlinearity.[6] Alternative platforms, such as graphene and gas plasma, have been studied for few-cycle terahertz pulse generation.[8–11] In all cases, strong nonlinearity is a key requirement for generating terahertz pulses of high energy.

Three-dimensional Dirac semimetals (3D DSMs)[12]—a recently discovered class of topological materials—have been shown to exhibit extremely large optical nonlinearities that originate from their linear and gapless energy–momentum dispersion in all three dimensions. For this reason, the 3D DSM Cd$_3$As$_2$,[13] which possesses exceptionally high Fermi velocities and electron mobilities,[12,13] has been used to generate highly efficient terahertz high-order harmonics from input terahertz pulses,[14–16] and studied as a platform for nonlinear plasmonics.[19,20] Related materials like Weyl semimetals have been shown to support chiral terahertz emission and polarization manipulation.[21]

We show that the extreme optical nonlinearity of 3D DSMs can be leveraged for highly efficient optical-to-terahertz conversion over nanometer-scale propagation distances. Specifically, we predict a conversion efficiency enhancement of over 5000 times in Cd$_3$As$_2$ compared to LN, in a propagation distance of 300 nm. The state-of-the-art conversion efficiency of LN is around 1% for both single- and multi-cycle terahertz pulses with sophisticated phase-matching methods,[22,23] whereas our efficiency exceeds typical materials like LiNbO$_3$ by >5000 times over nanoscale distances. Specifically, we predict a conversion efficiency enhancement of Cd$_3$As$_2$ could deliver ~5 × 10$^{-5}$%μm$^{-1}$ under direct pump lasers incidence without any phase-matching manipulation. This is especially surprising given that we use the third-order nonlinearity in Cd$_3$As$_2$, whereas the second-order nonlinearity is used for the corresponding process in LN. Our results reveal that tuning the Fermi energy allows us to leverage Pauli blocking to achieve a step-like conversion efficiency enhancement in terahertz generation. Our findings are crucial in the development of efficient ultrathin-film terahertz sources and the development of compact terahertz driven technologies.[24,25]

2. Physics of Terahertz Generation in 3D DSMs

Terahertz generation from optical pulses in DSMs, schematically illustrated in Figure 1a (inset), occurs when two co-propagating optical pulses of the same polarization (x polarized), but with different central frequencies $\omega_1$ and $\omega_2$ (frequencies within the optical pulse centered at $\omega_0$ and $\omega_2$ are denoted by $\omega_1$ and $\omega_2$, respectively), impinge on the sample, generating an output terahertz pulse that travels in the same direction. In momentum space, the driving fields induce inter-band and intra-band carrier transitions, resulting in the absorption of two low-energy photons at $\omega_1 \approx 0.5\omega_0$ and $\omega_2$, and emitting one high-energy photon at $\omega_1$ and another low-energy terahertz photon of frequency
$\Omega = 2\omega_2 - \omega_1$. In Figure 1, we study optical-to-terahertz conversion for the specific case of the 3D DSM Cd$_3$As$_2$. We determine the linear and nonlinear material conductivities associated with the 3D Dirac cone band structure using perturbative quantum theory, and simulate the terahertz generation process by solving Maxwell’s equations using these conductivities. Our model fully considers effects including the optical Kerr effect, finite temperatures, arbitrary Fermi energies, and carrier scattering. Our model captures both inter-band and intra-band dynamics, as well as coupling between them (henceforth denoted by the term inter-intra-band). The Hamiltonian that describes the carrier dynamics within a 3D DSM is given by

$$i\hbar \partial_t \Psi(t) = \left[ H_0 + H_{\text{int}}(t) \right] \Psi(t) \tag{1}$$

where $\Psi(t)$ is the electron wave function, $H_0 = \sum_j \nu_j \sigma_j \mathbf{p}$ is the stationary Hamiltonian, $H_{\text{int}} = \mathbf{r} \cdot \mathbf{E}(t)$ is the interaction Hamiltonian in the length gauge, $\mathbf{r}$ is the position operator, $\epsilon (> 0)$ is the elementary charge, $\mathbf{E}$ is the electric field, $\hbar$ is the Planck constant, $\sigma_j$ is the Pauli matrix with $j \in x, y, z$, $v_j$ is the Fermi velocity along direction $j$ in Cartesian coordinates, and $\mathbf{p}_j$ is the momentum operator in the $j$ direction.

The length gauge is chosen for $H_{\text{int}}$ since the resulting nonlinear response is free of nonphysical zero-frequency divergences and a more transparent representation can be obtained.\cite{16,27} Due to the inversion symmetry of the Cd$_3$As$_2$, even-ordered nonlinear conductivities are zero in our configuration. We apply perturbative quantum theory to Equation (1) and obtain, for the first time, linear and nonlinear conductivities corresponding to a general 3D DSM (see Methods and Sections III and IV, Supporting Information). Our model fully considers finite temperatures, carrier scattering, and an anisotropic Dirac cone band structures with Fermi velocities corresponding to realistic 3D DSM materials.

We simulate the terahertz generation process by solving the Maxwell’s equations using a finite-difference split-step method, which captures linear and nonlinear propagation effects of paraxial pulses up to the third order in nonlinear conductivity, including the back conversion of the terahertz pulse on the optical pulses and the optical Kerr effect. The cascading effect is taken into consideration by solving the coupled equations of the terahertz and the pump pulses interactively for each numerical iteration. In particular, by defining the electric field as

$$\tilde{E}(z, t) = E(\exp(-t^2/r^2)) \exp(-i\omega_0 t) \mathbf{x} + c.c. \tag{2}$$

$$\mathcal{F}[E \exp(-t^2/r^2) \exp(-i\omega_0 t)] = E(z, \omega) \exp[ik(\omega)z] \tag{3}$$

the optical-to-terahertz conversion in DSMs is given by

$$\partial E_{\text{THz}}(z, \Omega)/\partial z = -\frac{3}{2\hbar(\mathbf{L})^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \sigma^{(3)}(\omega_2, \omega_1, -\omega_1) E_j(z, \omega_2) \times E_j(z, \omega_2 + \omega_1 - \Omega) E_j(z, \omega_1) \exp\left\{i[k(\omega_2) + k(\omega_1) - k(\Omega)z] d\omega_2 d\omega_1 \right\} \tag{4}$$

where $\mathcal{F}$ is the Fourier transform, $^*$ represents complex conjugate, $z$ is the propagation distance, $r$ is the pulse duration, $\sigma^{(3)}(\omega_2, \omega_1, -\omega_1)$ is the third-order conductivity, $\Omega$ represents the
terahertz frequency, $k(\omega_i) = n(\omega_i)\omega_i/c$ represents the angular wavenumber at frequency $\omega_i = \omega_0 + \Omega - \Omega$ (in our case both $\omega_i$ and $\omega_0$ are centered at $\omega_{0i}$). $n(\omega)$ is the refractive index, $z$ is the propagation distance, $c$ is the speed of light, and $\varepsilon_z$ is the vacuum permittivity. The terahertz pulse amplitude is given by $E_{THz}$, and the optical pulse amplitudes by $E_1$ and $E_2$.

In this work, we consider the two optical pulses of amplitudes $E_1$ and $E_2$, centered at wavelengths of $1\mu m (\lambda_{0i} = 1.24eV)$ and $2\mu m (\lambda_{0i} = 0.62eV)$, respectively, with 150 fs ($\tau = 150$ fs/$\sqrt{2\ln(2)}$) intensity full-width-half-maximum pulse duration.

3. Enhanced Optical Optical-to-Terahertz Conversion Efficiency in 3D DSMs

Figure 1a shows the optical-to-terahertz conversion efficiency as a function of propagation distance for a collinear configuration (Figure 1a inset). At a propagation distance of about 300 nm, the conversion efficiency in 3D DSM Cd$_3$As$_2$ exceeds that of LN by >5000 times. Additionally, we find that even when no restrictions are placed on propagation distance, Cd$_3$As$_2$ outperforms LN in efficiency by over ten times. Figure 1b shows the terahertz conversion efficiency with respect to optical field $E_2$, when optical field $E_1$ is fixed at $5MV/m$. Whereas LN outperforms Cd$_3$As$_2$ at low field strengths $E_2 < 5MVM/m$ (Figure 1b, inset), Cd$_3$As$_2$ rapidly surpasses LN as field strength increases. The output terahertz fields and spectra for $E_1 = 50MV/m$ (dotted circles in Figure 1b) are shown in Figures 1c and 1d, respectively. The broader terahertz spectrum generated by Cd$_3$As$_2$ in Figure 1d may be attributed to two reasons. The first reason is that convolution in the frequency domain leads to a wider final bandwidth for a third-order nonlinear process (Cd$_3$As$_2$) compared to a second-order nonlinear process (LiNbO$_3$). The second reason is that the spectra shown are captured at the peak of the conversion efficiencies, that is, the propagation distance for Cd$_3$As$_2$ and LiNbO$_3$ are approximately nanometers and micrometers, respectively. LiNbO$_3$ has larger absorption at higher frequencies (see Figure S5d, Supporting Information), which leads to a narrower spectrum since the high frequencies components are absorbed more. Since the terahertz generation process does not lead to a drastically changing spectral phase, the broader spectrum could potentially lead to a shorter pulse duration, which could enhance the peak electric field strength of the terahertz pulse.

Here, we consider the experimentally measured Fermi velocities ($v_x, v_y, v_z$) = (1.28, 1.3, 0.33) × 10^6 ms$^{-1}$. It is possible that still higher conversion efficiencies exist at larger field strengths, but that would require a non-perturbative treatment for calculating the nonlinear conductivity that falls beyond the scope of this work. In Section SVII, Supporting Information, we present an analytical estimate for the input field strengths for which our conductivity calculations remain valid.

4. Optimizing Efficiency by Tuning the Fermi Energy

Figure 2 shows that an appropriate choice of the Fermi energy $\varepsilon_f$, allows us to access a regime of enhanced terahertz generation. In Figure 2a, we see that a broad range of Fermi energies and driving field strengths exist where substantial terahertz generation efficiencies can be accessed, even within the limits of perturbation theory. The generation efficiency is defined as the ratio to the generated terahertz energy and the input pump pulse energies. $\int I(\Omega)d\Omega / \int I(0)d\omega$, where $I(\Omega, \omega) = c, n(\omega)/E(z, \Omega) \exp (iKz)E(z)E(\Omega)E(\Omega)^*/2$ is the intensity in the frequency domain as a function of terahertz frequency $\Omega$, $n(\omega)$ is the refractive index, and $\omega$ is the optical frequency. The trend in peak efficiency is partly explained through the third-order conductivity associated with terahertz generation, explaining the step-like increase of the third-order conductivity, we plot in Figure 2c,d the conductivity density corresponding to terahertz generation as a function of the energy of the electronic states, defined by $\sigma^{(3)}(\omega_1, \omega_2, -\omega_3) = \int \sigma^{(3)}(\varepsilon, \omega_1, \omega_2, -\omega_3) d\varepsilon$ at representative Fermi energy values from each regime (0.05, 0.45, 0.7). Here $\varepsilon$ denotes the eigenenergy of an electron with a given wavevector $k$.

In Figure 2c, we see that at relatively low Fermi energies, the nonlinear conductivity density corresponding to terahertz generation contains two peaks, one at $\varepsilon \approx -0.62eV$ and another at $\varepsilon \approx 0.31eV$. As the Fermi energy increases, the peaks of the conductivity density that lie within the range $\varepsilon = [-\varepsilon_f, \varepsilon_f]$ are suppressed. We infer that this is related to Pauli blocking, which occurs when an electron cannot be excited from the valence band to the conduction band due to the lack of unoccupied states in the conduction band. This can be seen in Figure 2d, where the peak at $\varepsilon \approx 0.31 eV$ disappears. Since the contribution of the peaks at $\varepsilon \approx -0.62eV$ and $\varepsilon \approx 0.31eV$ add up destructively, the disappearance of one peak has the effect of enhancing the nonlinear conductivity associated with terahertz generation, explaining the step-like increase in conductivity moving from the “No Pauli blocking” to “Enhanced terahertz” regime. At still larger Fermi energies—exemplified by the scenario in Figure 2e—both conductivity density peaks are suppressed since all transitions required for terahertz generation is forbidden, leading to a plunge in the resulting nonlinear conductivity in the “Forbidden terahertz” regime. The total nonlinear conductivity is obtained by integrating the differential conductivity over all electron energies. Since the differential conductivity consists of two peaks of opposite sign, the resulting integral is smaller in magnitude than if only one of the peaks is included in the integral. This is why the disappearance of one peak enhances the nonlinear conductivity.

In Figure 2a, we see that the optical-to-terahertz conversion efficiency generally follows the same trend as the nonlinear conductivity in Figure 2b. However, at low Fermi energies, the low electron filling in the conduction band leads to a smaller first-order conductivity at terahertz frequencies, that is, smaller terahertz absorption as shown in Equation (6). Consequently, a relatively high efficiency can be potentially attained due to lower terahertz absorption in the “No Pauli blocking” regime at very low Fermi energies.
Figure 2. Optical-to-terahertz conversion efficiency as a function of Fermi energy. a) The peak terahertz conversion efficiencies at different input field strengths $E_2$ as a function of the Fermi energy $\varepsilon_f$. The simulation results are denoted by the filled circles. The curves are visual guides. The non-perturbative regime (gray-shaded area) corresponds to $E_2 < \sqrt{|\sigma^{(1)}|/|\sigma^{(3)}|}$ (see Section SVII, Supporting Information). The pink data point marked by the dashed circle indicates the same data as in Figure 1b. b) Indicates how the third-order conductivity for terahertz generation at 1 THz ($\omega_2 = \omega_2 - \omega_1 = 1$ THz) varies as a function of $\varepsilon_f$. The "No Pauli blocking," "Enhanced terahertz," and "Forbidden terahertz" regions in (b) correspond to c) ($\varepsilon_f = 0.05$ eV), d) ($\varepsilon_f = 0.45$ eV), and e) ($\varepsilon_f = 0.7$ eV), respectively, where the conductivity density as a function of the electron energy is shown ($\sigma^{(3)}(\omega_2, \omega_2 - \omega_1) = \int \tilde{\sigma}^{(3)}(\varepsilon, \omega_2, \omega_2 - \omega_1) |E| d\varepsilon$). c) Indicates two strong conductivity density peaks, which contribute with opposite signs to the overall conductivity. Consequently, an increase in the conductivity can be observed in (b) where one of the peaks is suppressed while the other remains. d) shows that all conductivity density peaks is suppressed and thus the terahertz generation is forbidden. We consider the same simulation parameters as Figure 1. It should be noted that linear and gapless band structure of Cd$_3$As$_2$ extends up to 1 eV,[15,28] which justifies plotting up to Fermi energies of 0.8 eV in (a) and (b).

As Figure 2a shows, the high conversion efficiency of Cd$_3$As$_2$ holds over a broad range of field strengths and Fermi energies. Although we have focused on the case of $T = 77$ K here, our simulations at other temperatures (see Figure S3 in Section SV, Supporting Information) reveal stability over a broad range of temperatures ranging from 4 to 200 K. The enhanced conversion efficiency in the "Enhanced terahertz" regime, as well as the need to stay within the validity of our perturbative conductivity calculations motivated our choice of $\varepsilon_f = 0.45$ eV in Figure 1.

5. Discussion

Our model is readily extended to capture the physics of a general, anisotropic 3D Dirac cone band structure. Although Cd$_3$As$_2$ has been considered in Figures 1 and 2, our model also applies to other 3D DSMs featuring different Fermi velocities. The significance of the material's Fermi velocities can be seen from Equations (6–11) in Methods, where the linear conductivity $\sigma^{(1)}_x$ and the nonlinear conductivity $\sigma^{(3)}_{xxx}$ are directly proportional to $\nu_x/|\nu_y \nu_z|$ and $\nu_x^3/|\nu_y \nu_z|$, respectively. Figure 3 shows the peak conversion efficiency as a function of these prefactors, revealing that the combination of a small $\sigma^{(1)}_x$ and a large $\sigma^{(3)}_{xxx}$ can lead to efficient tera-
Table 1. Fermi velocities of different materials. The ZrTe$_2$ represents the averaged Fermi velocities ($v_{xx}, v_{yy}, v_{zz}$). For ZrTe$_3$, the Fermi velocity along $k$ is calculated by $v_{k} = \sqrt{\epsilon_k^2 + \frac{\pi^2 \hbar^2}{3k_B T^2}}$.

| Material       | Fermi velocity ($v_x, v_y, v_z$) [ms$^{-1}$] |
|----------------|---------------------------------------------|
| Cd$_3$As$_2$   | (1.28, 1.3, 0.33) $\times$ 10$^{11}$        |
| Na$_3$Bi      | (4.17, 3.63, 0.95) $\times$ 10$^{11}$        |
| ZrTe$_2$      | (7.46, 1.94) $\times$ 10$^{11}$             |
| ZrTe$_3$      | (4.89, 4.03, 1.94) $\times$ 10$^{12}$        |
| TlBiS$_2$     | (1.6, 1.6, 1.6) $\times$ 10$^{12}$          |

hertz generation. This can also be understood intuitively since it implies low terahertz absorption and large nonlinearity for optical-to-terahertz conversion. We show that under the given conditions, Cd$_3$As$_2$ is close to an ideal choice for optimal conversion efficiency. The large nonlinearity of Cd$_3$As$_2$ is due to its large Fermi velocities, which are obtained from the work of Liu et al. (also shown in Table 1). The values of $\sigma^{(2)}$ and $\sigma^{(3)}$ of the LN are obtained from $\chi^{(2)} = 2 \times 168$ pm/V at and $n_2 = 3 \times 10^{-12} / 4\epsilon_0 c_0 n_0$.\(^{(13)}\) where $\chi^{(3)} \approx 10^{-13}$ m$^2$/W$^{-1}$, and $n_0$ is the refractive index at the desired frequency.

Our findings suggest that even larger efficiencies can be obtained with field strengths and Fermi energies that require a non-perturbative treatment of the nonlinear conductivity. Larger conversion efficiencies for the same input fields could also potentially be obtained by considering a non-collinear interaction geometry. In particular, having obliquely incident input fields would lead to the existence of second-order nonlinearities in 3D DSMs, which could also be a promising avenue for efficient optical-to-terahertz conversion.

6. Methods

The input electric field and its positive frequency component $\exp(-i\omega_0 t)$ are shown in Equations (2) and (3), respectively. Without the loss of generality, the input fields are chosen to be linearly polarized in $\hat{x}$ direction. Note that in the following calculations, only the positive frequency part of the $\sigma(\omega)$ and $E(\omega)$ are presented. However, no approximation is made. Since with the relation $\sigma(\omega) = \sigma(-\omega)^*$ and $E(\omega) = E(-\omega)^*$, all the information is contained in the positive frequency elements.

The first-order conductivity are the followings:

$$\sigma^{(1)}_{xx}(\omega) = \frac{ie\epsilon_\nu}{6\pi^2\hbar^2 v_x (\omega + i\gamma)} \left( \epsilon^2 + \frac{\pi^2 k_B T^2}{3} \right)$$

$$\sigma^{(1)}_{xy}(\omega) = \frac{ie\epsilon_\nu}{24\epsilon_\nu \pi^2 \hbar^2} \int_{-\omega}^{\omega} \frac{n(\epsilon)}{\epsilon - (h(x + i\gamma))} d\epsilon$$

(6)

(7)

where "$i$" denotes intra-band, "$e$" denotes inter-band, $g = 4$ is the combined valley and spin degeneracy, $n(\epsilon) = f(\epsilon) - f(-\epsilon)$. $f(\epsilon) = (\exp[E - \epsilon/k_B T] + 1)^{-1}$ is the Fermi distribution function, $E$ is the energy, $k_B$ is the Boltzmann constant, $T$ is the temperature, $\gamma$ is the inter-band decay rate, and $\gamma_e$ is the coherence decay rate (see Supporting Information). In our calculations, $\gamma_e = 1/(150 \text{ fs})$. Equation (7) agrees with the work of Kotov.$^{(15)}$ (see Equation (S39), Supporting Information). Equation (6) can also be obtained from the Boltzmann transport equation (Section SVIII, Supporting Information).

Although Equation (7) may appear to be readily solved via complex analysis, one should note that $n(\epsilon)$ contains an infinite number of poles in the complex-$\epsilon$ domain. By defining $\omega_c = 2E/h$, the terms in the expression for the third-order conductivity can be written as

$$\sigma^{(3)}_{xxxx}(\omega_1, \omega_2, \omega_3) = \frac{ig e^2 \nu^3}{15 \hbar^3 \pi^2 v_x (\omega_1 + \omega_2 + i\gamma) (\omega_2 + i\gamma)}$$

$$\sigma^{(3)}_{xxxy}(\omega_1, \omega_2, \omega_3) = -\frac{ie\epsilon_\nu^3}{15 \hbar^3 \pi^2 v_x (\omega_1 + \omega_2 + i\gamma)} \int_{-\omega}^{\omega} \left[ \frac{2(\omega_2 + i\gamma) n(\epsilon)'}{\epsilon} - \frac{2(\omega_1 + \omega_2 + i\gamma) n(\epsilon)'}{\epsilon} \right] d\epsilon$$

(9)

$$\sigma^{(3)}_{xxyx}(\omega_1, \omega_2, \omega_3) = \frac{ie\epsilon_\nu^3}{30 \pi^2 v_x \pi^2 h (\omega_1 + \omega_2 + i\gamma)} \int_{-\omega}^{\omega} \frac{4n(\epsilon)'}{\epsilon} d\epsilon + \frac{M_1 (\omega, E)}{\omega_2 - \omega_1 - i\gamma_c}$$

(10)

$$\sigma^{(3)}_{xxxx}(\omega_1, \omega_2, \omega_3) = \frac{ie\epsilon_\nu^3}{30 \pi^2 v_x \pi^2 h (\omega_1 + \omega_2 + i\gamma)} \int_{-\omega}^{\omega} \frac{4n(\epsilon)'}{\epsilon} d\epsilon + \frac{M_1 (\omega, E)}{\omega_2 - \omega_1 - i\gamma_c}$$

(11)

$$M_1 (\omega, E) = \left[ \frac{\partial}{\partial \epsilon} \frac{n(\epsilon)}{\omega_2 - \omega_1 - i\gamma_c} \right] - \frac{2n(\epsilon)}{(\omega_2 - \omega_1 - i\gamma_c) E}$$

(12)

where $\sigma^{(1)}_{xxxx}$ represents the purely intra-band process, $\sigma^{(1)}_{xxxy}$ represents the inter-band process, $\sigma^{(3)}_{xxxx}$ and $\sigma^{(3)}_{xxxy}$ arise from the diagonal terms of the density matrix and the coherence terms of the density matrix, respectively, and represent the inter–intra-band process (see Supporing Information). The frequency permutation remains to be included in Equations (8–11), due to the permutation symmetry.$^{(16)}$ The final $\sigma^{(n)}$ should be an average of all $(n!)$ frequency permutations, that is, $\sigma^{(n)} = \sum P^{(n)} + \sigma^{(1)}_{xxxy} + \sigma^{(3)}_{xxxx} + \sigma^{(3)}_{xxxy}/6$, where $P$ represents the summation of 6 possible
permutations. Similar to Equation (6), Equation (8) can also be obtained by Boltzmann transport equation (Section SVIII, Supporting Information). Surprisingly, $\sigma_i^{(3)}$ is independent of $E_i^*$ and $T$. The lack of dependence on $E_i^*$ for $\sigma_i^{(3)}$ has also been discussed in the work of Cheng et al., which present linear and nonlinear conductivity expressions for isotropic 3D DSMs in the limit where $T = 0$ and carrier scattering is negligible.

Our simulation is based on a single-pulse model and thermal effects are not included. Since the thermal relaxation time and damage of the material largely depend on the pump laser fluence, wavelength, and repetition rate, the maximum input electric field strength may need to be reduced for very high repetition lasers. The relaxation lifetime of LiNbO$_3$ and Cd$_3$As$_2$ are $\tau_i \approx 0.1$ ms[42] and $\tau_i \approx 10 ps$, respectively. As a result, as long as the repetition rate of the pumping laser is smaller than $1/\tau_i$, our model should be valid.

7. Conclusion

In summary, we show that the 3D DSM Cd$_3$As$_2$ is a promising candidate for realizing highly efficient conversion of optical frequencies to terahertz frequencies over nanometer-scale propagation distances. Using theory and numerical simulations which account for finite temperatures, carrier scattering, and the anisotropic nature of realistic Dirac cones, we have shown that the conversion efficiencies achievable with Cd$_3$As$_2$ can be >5000 times larger than the conversion efficiencies of the commonly used terahertz generation platform LiNbO$_3$ over a propagation distance of 300 nm. This is surprising since the high conversion efficiencies of Cd$_3$As$_2$ are attained using its third-order nonlinearity, as opposed to the second-order nonlinearity responsible for terahertz generation in LiNbO$_3$. Furthermore, we show that the Fermi energy can be tuned to substantially enhance the nonlinearity of Cd$_3$As$_2$, thereby allowing us to increase the efficiency of the terahertz generation process. Although our exciting results are achieved within the perturbative regime, we expect similarly promising results at non-perturbative fields strengths. Our findings pave the way toward the development of highly efficient and compact terahertz light sources based on 3D DSMs.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Dirac semimetals, nonlinear optics, terahertz generation

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