Nonlinear and anisotropic polarization rotation in two dimensional Dirac materials

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We predict nonlinear optical polarization rotation in two dimensional massless Dirac systems including graphene and 8-Pmmn borophene. When illuminated, a continuous wave optical field leads to a nonlinear steady state of photo-excited carriers in the medium. The photo-excited population inversion and the inter-band coherence gives rise to a finite transverse optical conductivity, \(\sigma_{xy}(\omega)\). This in turn leads to definitive signatures in associated Kerr and Faraday polarization rotation, which are measurable in a realistic experimental scenario.

I. INTRODUCTION

Two dimensional (2D) electronic systems have garnered incredible attention over the past decade due to their exceptional opto-electronic properties and gate tunable response.1–5 Starting with graphene6–8, several other 2D materials9, including silicene6,7, MoS\(_2\)8 and phosphorene9, have been added to the list, each with its own peculiar regime of response. A comparatively recent addition to the 2D family of graphene, is one of the many 2D polymorphs of monolayer boron, termed as 8-Pmmn borophene.10–14 Unlike graphene which has an isotropic electronic dispersion, 8-Pmmn borophene has been shown to host a tilted and anisotropic Dirac cone.10–14 Such anisotropic and tilted Dirac cone dispersion has been shown to give rise to anisotropy in plasmon dispersion and screening,15 an extra contribution to the Weiss oscillations in the longitudinal magnetoconductivity16 and anisotropic optical conductivity in a linear response regime.17 Beyond linear response, one therefore expects such materials to manifest further richness in properties and accordingly, there has been an intense search for techniques to characterize such nonlinearities and use in device applications.18–20

Here we explore nonlinear polarization rotation of a continuous wave (cw) optical field, reflected or transmitted from materials hosting quasiparticles with a tilted and anisotropic Dirac cone dispersion. Usual studies related to optical properties in such materials21–28 focus on the linear response regime, ignoring higher order field induced changes in the photo-excited carrier distribution. However, with increasing optical field strength, the photo-excited carrier distribution reaches a nonlinear steady state through competing rates of carrier excitation and various decay channels including carrier-carrier, carrier-phonon and impurity scattering.29–32 Though such nonlinearities have been studied in a transient regime33,34, a comparatively simpler steady-state study of the nonlinear transmission and reflection coefficients due to cw illumination, remains largely unexplored.

We show emergence of a finite transverse optical conductivity \(\sigma_{xy}(\omega)\), due to the nonlinear steady state population inversion, in a cw illuminated 2D massless Dirac system [see inset of Fig. 1]. We estimate the corresponding polarization rotation of reflected and transmitted cw fields and predict a measurement regime that is accessible with reasonable optical field intensities. For experimental detection, the common mode linear response contribution to the polarization rotation from other sources, can be subtracted out by using a differential measurement technique for two differing intensities [see Fig. 1]. The predicted field controlled polarization rotation can open up new possibilities in nano-structured opto-electronic devices including fast polarization switches and dynamically controlled wave plates.

This article is organized as follows: we first present the effective low energy Hamiltonian of 8-Pmmn borophene hosting a tilted and anisotropic Dirac cone and use it to calculate the corresponding optical matrix elements in Sec. II. With optical Bloch equations, next we calculate the nonlinear steady state distribution function and the corresponding density matrix in presence of a cw field. The calculation of optical conductivity, transmission and reflection coefficients along with Kerr and Faraday rotation, is presented in Sec. IV followed by a discussion of experimental implications in Sec. V. Finally we summarize our results in Sec. VI.

FIG. 1. A cartoon depicting a cw optical field undergoing polarization rotation in reflection and transmission when incident on a suspended graphene sheet. The polarization rotation occurs due to the emergence of a transverse optical conductivity (\(\sigma_{xy}\)), that bears a unique signature in the incident polarization angle (\(\theta\)) and the input field strength (\(E_1\) and \(E_2\), with \(E_2 > E_1\)) as shown in the inset.
II. HAMILTONIAN AND THE OPTICAL MATRIX ELEMENT

Hamiltonian for an electron interacting with an electric field is described in the dipole approximation as
\[ \hat{H} = \hat{H}_0 + e \mathbf{E} \cdot \hat{r}, \]
where \( \hat{H}_0 \) is the bare effective low energy Hamiltonian describing the the energy dispersion of the material, \( e \) is the magnitude of the electronic charge, and \( \mathbf{E} \) is the electric field vector. The generalized low energy effective Hamiltonian for tilted and anisotropic gapless Dirac systems in 2D, in vicinity of the Dirac point is given by,
\[ H = \sum_k H_k, \]
\[ H_k = \hbar v_F |k| \left[ \hat{v}_x \sin \phi_k + \lambda (\hat{v}_x^2 \cos^2 \phi_k + \hat{v}_y^2 \sin^2 \phi_k)^{1/2} \right], \]
with \( |k| = (k_x^2 + k_y^2)^{1/2} \), and \( \phi_k = \tan^{-1}(k_y/k_x) \), the azimuthal angle and \( \lambda \) takes values +1 and −1 for the conduction and valence band, respectively. In Eq.(3) we have defined the dimensionless velocities, \( \hat{v}_x = v_x/v_F \), \( \hat{v}_y = v_y/v_F \) and \( \hat{v}_z = v_z/v_F \).

The dipole matrix element \( r^{\lambda \lambda'} (\lambda \neq \lambda') \) in the basis formed by the eigenvectors of \( H_k \), can be written in terms of momentum matrix element (also called the optical matrix element) as\(^{39}\),
\[ r^{\lambda \lambda'} = i \mathbf{M}_k^{\lambda \lambda'} / (\omega_k), \]
where we have defined \( \hbar \omega_k = \varepsilon_k^c - \varepsilon_k^v \).

For \( H_k \) of Eq. (2), we have
\[ \mathbf{M}_k^{\lambda \lambda'} = \frac{ie \hbar v_F \hat{v}_x \hat{v}_y}{(\varepsilon_k^c \cos^2 \phi_k + \varepsilon_k^v \sin^2 \phi_k)^{1/2}} (\sin \phi_k - \cos \phi_k). \]
For the specific case of \( v_x = v_y = v_F \), as in isotropic graphene, we have \( \mathbf{M}_k^{\lambda \lambda'} = i e v_F (\sin \phi_k - \cos \phi_k) \).

III. STEADY STATE PHOTO-EXCITED CARRIERS AND COHERENCE

In this section we consider optical pumping of 2D gapless Dirac system (monolayer 8-Pmmm boronene or graphene) with a monochromatic continuous wave (CW) laser. The field at any time \( t \) is given as, \( \mathbf{E} = E_0 \cos \omega t \hat{e} \), with \( E_0 \) being the electric field amplitude, \( \omega \) the optical laser frequency and \( \hat{e} \) the polarization direction.

The dynamics of the system is best described in terms of the equation of motion for the density matrix: \( i \hbar \partial \rho(t) = [H, \rho(t)] \). We denote the diagonal elements of the density matrix as \( \rho_{11} = \rho_k^c \) and \( \rho_{22} = \rho_k^v \), where \( \rho_k^\lambda = \langle a_k^{\lambda \dagger} a_k^\lambda \rangle \) denotes the momentum resolved electron density in the \( \lambda \)-th band, and the off diagonal elements of the density matrix as \( \rho_{12} = \rho_k = \langle a_k^c a_k^v \rangle \), and \( \rho_{12} = \rho_k^c \). Here \( \rho_k \) is usually referred to as the inter-band coherence or polarization. Using these, and Eq. (1), the evolution of the density matrix is given by a set of coupled optical-Bloch equations\(^{29–32}\),
\[ \partial_t \rho_{kk} = 4 \beta_m \left[ \Omega_k^{\text{Rabi}} \rho_k^c - \gamma_1 (n_k - n_k^\text{eq}) \right], \]
\[ \partial_t \rho_k = i \omega_k \rho_k - i \Omega_k^{\text{Rabi}} n_k - \gamma_2 \rho_k. \]
In Eq. (5)-(6), \( n_k \equiv \rho_k^c - \rho_k^v \) is generally referred to as population inversion and \( h \Omega_k^{\text{Rabi}} = e \mathbf{E} \cdot \mathbf{r}^{\lambda \lambda'} \) is the interband Rabi frequency.

The last two terms in both Eqs. (5)-(6) are added phenomenologically to include the decay of the inverted population and the inter-band coherence\(^{39}\). The equilibrium population inversion (in absence of light) is expressed as \( n_k^{\text{eq}} = f_k^{(0)} - f_k^{(0)} \), where \( f_k^{(0)} \) denotes the Fermi function with band index \( \lambda \), and \( \gamma_1, \gamma_2 \) are the phenomenological relaxation rate for the momentum resolved population inversion and the inter-band coherence, respectively. Aiming for an insightful analytical solution, and for simplicity we assume \( \gamma_1 \) and \( \gamma_2 \) to be constants.

Solving Eqs. (5)-(6) in the steady state, with the assumption\(^{31}\) that, \( \rho_k = p_{1k} e^{i\omega t} + p_{2k} e^{-i\omega t} \), we have,
\[ p_{1k} = \frac{i n_k - \mathbf{E} \cdot \mathbf{M}_k^{c}}{2 \hbar \omega_k - \omega_k - i \gamma_2}, \]
\[ p_{2k} = \frac{i n_k - \mathbf{E}^* \cdot \mathbf{M}_k^{c}}{2 \hbar \omega_k + \omega_k + i \gamma_2}. \]
The parameter \( \zeta \) is the optical field strength and frequency dependent main parameter, which characterizes the ‘degree’ of the nonlinear effects and response.

It is easy to check that in the absence of incident light beam, we have \( \zeta \rightarrow 0 \), which implies \( G_k \rightarrow 1 \) and consequently \( f_{\gamma k}^{(1)} \rightarrow f_{\gamma k}^{(0)} \), with no nonlinearity in the carrier distribution function. On the other hand, for very high intensity beams we have \( \zeta \rightarrow \infty \), which leads to \( G_k \rightarrow 0 \) and this describes the optical saturation limit. Figure 2 shows the time evolution of population inversion \( n_k \) and \( \rho_k \) as a function of time (in units of \( 1/\omega \)). At \( t = 0 \) the carrier excitation probability is vanishingly small (with \( n_k = -1 \)), since the optical field is absent. With increasing time and after a few optical cycle, \( n_k \) starts increasing and finally saturates to the steady state population inversion given by \( n_k^{eq} G_k \) over timescales given by \( t \approx 1/\gamma_1 \).

In the linear response regime, the optical conductivity is calculated using the Kubo formula. In the Kubo formula, the optical field generated nonlinearity in the carrier distribution function is neglected, i.e., \( n_k \rightarrow n_k^{eq} \). Also for convenience, the calculations are usually done in the infinite coherence time limit, i.e., \( \gamma_2 \ll \omega \). To highlight the deviation in the carrier distribution function due to nonlinear optical effects in the same high frequency regime of \( \omega \gg \gamma_2 \), we expand \( G_k \) up to second order in \( \zeta \) to yield

\[
G_k^2 \approx 1 - \frac{\pi}{2} \gamma_2 \omega^2 |\mathbf{M}_{\mathbf{k}}^{ee} \cdot \hat{e}|^2 \delta(\omega_k - \omega) \tag{11}
\]

The factor \( \gamma_2 \zeta^2 \) in the above expression is actually independent of \( \gamma_2 \) as \( \zeta^2 \propto 1/\gamma_2 \). This clearly highlights the fact that in the vanishingly small optical field limit, the carrier distribution function is primarily unchanged. Deviation from the equilibrium distribution of carriers, is captured by the term proportional to \( \zeta^2 \), which in energy is centered around \( \omega_k = \omega \). The \( \zeta^2 \) term is proportional to \( |\mathbf{M}_{\mathbf{k}}^{ee} \cdot \hat{e}|^2 \), and this highlights the anisotropy in the NDF which arises from the anisotropic optical matrix element and the polarization direction.

The highly anisotropic nature of the momentum resolved population inversion, \( n_k \), for 8-\textit{Pmmn} borophene is also highlighted in Fig. 3. Similar to the case of graphene, the photo-excited carrier distribution has a maxima in a direction perpendicular to the direction of the optical field polarization. However unlike graphene, in borophene the profile of \( n_k \) is slightly elongated along the \( y \)-direction, and this can be attributed to the anisotropic band-structure of 8-\textit{Pmmn} borophene. This steady state photo-excited NDF, is what leads to nonlinear and anisotropic optical response in optical conductivity, can lead to non-equilibrium plasmons, giant and anisotropic photoconductivity, or in the polarization rotation of the reflected and transmitted beam, as discussed in the current paper.

IV. NONLINEAR OPTICAL CONDUCTIVITY AND KERR ROTATION

A. Steady state Optical conductivity

Here we focus on the steady state nonlinear optical conductivity. Following our previous work, we can express the real part of the momentum resolved current

\[
\rho_k = f_{\gamma k}^{(1)} - f_{\gamma k}^{(0)} = \begin{cases} \frac{1}{2} \left[ f_{\gamma k}^{(0)} (1 + G_k) + f_{\gamma k}^{(0)} (1 - G_k) \right], & \text{for } n_k = 0 \text{ and } n_k = -1 \text{, respectively} \end{cases}
\]
The variation of the real part of $\sigma_{xx}$ and $\sigma_{yy}$ with $\theta$ for $\zeta = 5$ is shown in (a), whereas the variation of real part of $\sigma_{xy}$ is shown in (c). The exact angular dependence more or less follows the approximate angular dependence obtained for $\text{Re}(\sigma_{xx})$, $\text{Re}(\sigma_{yy})$ and $\text{Re}(\sigma_{xy})$ in Eqs. (16), (19) and (21), respectively. The nonlinear effects has been highlighted using the exact results in panel (b) and (d), which show the $\zeta$ dependence of the longitudinal and transverse optical conductivities, respectively, for $\theta = \pi/4$. Here $\mu = 0.2 \times \hbar \omega$ and the other parameters are same as that of Fig. 2.

Thus the total current is given by,

$$J(t) = -2\text{Re}[p_k(t)M_k^{\nu}] ,$$

where $g_x$ and $g_y$ represent the spin and valley degeneracy respectively. The current in Eq. (13) is real and it has terms with time dependence of $e^{i\omega t}$ as well as $e^{-i\omega t}$. In the spirit of the linear response theory, we use the $e^{-i\omega t}$ part of the current to define the real and the imaginary components of the optical conductivity (see details of the calculation in Eq. A5 of the appendix). The nonlinear optical conductivities in 2D can be easily calculated via the relations,

$$\sigma_{ij}(\omega) = g_x g_y \frac{2}{(2\pi)^2} \int_{BZ} dk J_{ik}(\omega)/E_j ,$$

where the integral is over the Brillouin zone of the material involved.

While $\sigma_{ij}(\omega)$ is not difficult to calculate numerically, some useful insights can be obtained from the analytical expression retaining the first nonlinear term beyond the linear response regime. This is the term proportional to $\zeta^2$ or alternately to $E_0^2$, and depends on the intensity of the incident optical beam. The calculation simplifies in the $\gamma_2 \ll \omega$ limit with the lorentzian in Eq. (A14) giving way to a delta function. In this limiting case, the real part of $x$ component of the longitudinal optical conductivity is given by,

$$\frac{\text{Re}(\sigma_{xx}^\zeta)}{\sigma_0} = \Theta(h\omega - 2\mu) \int \frac{d\phi_k}{\pi} \frac{\sin^2 \phi_k v_x^2 v_y^2}{(v_x^2 \cos^2 \phi_k + v_y^2 \sin^2 \phi_k)^2} \times \left(1 - \frac{\zeta^2 v_x^2 v_y^2 \sin^2 (\phi_k - \theta)}{v_x^2 \cos^2 \phi_k + v_y^2 \sin^2 \phi_k}\right).$$

Here $\sigma_0 = e^2/4\hbar$ is the well known universal optical conductivity of graphene and $\theta$ denotes the polarization angle in an anticlockwise sense, with respect to the $\hat{x}$-axis of the crystal.

Evidently, for $h\omega < 2\mu$ there are no vertical optical transitions possible due to Pauli blocking. Consequently all optical conductivities vanish for $h\omega < 2\mu$. Thus in the rest of the manuscript, we only discuss the case of $h\omega > 2\mu$.

Performing the $\phi_k$ integration for $h\omega > 2\mu$ in Eq. (15), yields,

$$\frac{\text{Re}(\sigma_{xx}^\zeta)}{\sigma_0} = \frac{\tilde{v}_x}{8v_y} [8 - \zeta^2(3v_x^2 + v_y^2) - \zeta^2(3v_x^2 - v_y^2) \cos 2\theta].$$

As an independent check of our formalism, the longitudinal optical conductivity of graphene ($v_x = v_y = v_F$) in the linear response regime of $\zeta \to 0$ limit is given by

$$\frac{\sigma_{xx}^{\zeta \to 0}}{\sigma_0} = \Theta(h\omega - 2\mu) - \frac{i}{2\pi} \ln \left[\frac{(\omega + 2\mu/\hbar)^2}{(\omega - 2\mu/\hbar)^2}\right] ,$$

consistent with the results of Ref. [22].

Similarly the real and imaginary parts of the $y$ component of the longitudinal optical conductivity can also be calculated. They are explicitly given by

$$\frac{\text{Re}(\sigma_{yy}^\zeta)}{\sigma_0} = \frac{\tilde{v}_y}{8v_x} [8 - \zeta^2(\tilde{v}_x^2 + 3\tilde{v}_y^2) - \zeta^2(\tilde{v}_x^2 - 3\tilde{v}_y^2) \cos 2\theta],$$

and,

$$\frac{\text{Im}(\sigma_{yy}^\zeta)}{\sigma_0} = -\frac{1}{2\pi} \frac{v_y}{v_x} \ln \left[\frac{\gamma_2^2 + (\omega + 2\mu/\hbar)^2}{\gamma_2^2 + (\omega - 2\mu/\hbar)^2}\right]$$

$$-\frac{\zeta^2}{16} \frac{\gamma_2}{\omega} \frac{v_y}{v_x} \left[3v_x^2 + v_y^2 + (v_x^2 - 3v_y^2) \cos 2\theta\right].$$

Note that the anisotropy of the Dirac band-structure changes the universal isotropic optical conductivity of
graphene (within linear response) to $\Re(\sigma_{xx}) \rightarrow \sigma_0 v_x/v_y$ and $\Re(\sigma_{yy}) \rightarrow \sigma_0 v_y/v_x$.

The transverse optical conductivity $\sigma_{xy}^2$ can be obtained by replacing $\sin^2\phi_k \rightarrow -\sin\phi_k \cos\phi_k$ in the numerator of the integrand in Eq. (15), and it is given by

$$\frac{\Re(\sigma_{xy}^2)}{\sigma_0} = \frac{\Re(\sigma_{yx}^2)}{\sigma_0} = \frac{\zeta^2}{2} \tilde{v}_x \tilde{v}_y \sin \theta \cos \theta,$$

and,

$$\Im m(\sigma_{xy}^2) = \Im m(\sigma_{yx}^2) = \frac{\zeta^2}{4} \frac{\gamma_2}{\omega} \tilde{v}_x \tilde{v}_y \sin \theta \cos \theta.\tag{22}$$

We emphasize that the transverse optical conductivity $(\sigma_{xy})$ vanishes in the linear response regime, and it is finite only when the nonlinear response (proportional to $\zeta$) is significant.

Numerically calculated exact real part of longitudinal $(\sigma_{xx}, \sigma_{yy})$ and transverse interband optical conductivity, for 8-$Pmmn$ borophene, as a function of $\theta$ and $\zeta$ are shown in Fig. 4. Panels (a) and (c) of Fig. 4 have $\zeta = 5$ and finite $\mu = 10^{14}h/s = \hbar \omega/5$ and show the polarization angle dependence of the optical conductivities. Note that the exact numerical results are very close to the $\theta$ dependence predicted by approximate Eqs. (16), (19), and (21), i.e., $\Re(\sigma_{xx}) \propto \cos 2\theta$, $\Re(\sigma_{yy}) \propto \cos 2\theta$ and $\Re(\sigma_{xy}) \propto \sin 2\theta$. This is highlighted by the dashed curves in Fig. 4. In panels (b) and (d) of Fig. 4 we fix $\theta = \pi/4$ and show the $\zeta$ dependence of the optical conductivity. For vanishingly small intensity, $\zeta \rightarrow 0$, or in the linear response regime, we have $n_k \rightarrow n_k^\infty$ and therefore $\Re(\sigma_{xx}) \rightarrow \sigma_0 v_x/v_y = 1.25\sigma_0$ and $\Re(\sigma_{yy}) \rightarrow \sigma_0 v_y/v_x = 0.8\sigma_0$ and $\Re(\sigma_{xy}) \rightarrow 0$.

B. Optical transmission and reflection

Physically, optical conductivity sets the reflected, transmitted or absorbed optical spectrum along with rotation of the polarization angle in either the reflected (Kerr rotation) or in the transmitted (Faraday rotation) optical beam. Accordingly, we next derive a general expression for transmission and reflection coefficients for both $s$– and $p$– polarized incident optical beam and explore their implications for 2D massless Dirac materials, with particular emphasis on graphene and 8-$Pmmn$ borophene. For simplicity we assume the graphene or the borophene monolayer to be perfectly 2D (without ripples and defects), a reasonable assumption for an optical beam with diffraction spot size.

The transmission and reflection coefficients for $s$– (in the plane of incidence) and $p$– (perpendicular to the plane of incidence) polarized optical beam can be expressed in terms of the complex optical conductivity of the 2D monolayer\(^{38}\). For the sake of completeness, we reproduce the calculations in Appendix B. Using the fact that the optical conductivities in 2D materials are typically of the order of $\sigma_0 = e^2/(4h)$, the exact expressions for the transmission and reflection coefficients can be simplified.

Retaining only the first order terms in the small parameter, $\pi \alpha_F/2$ where $\alpha_F \sim 1/137$ is the fine structure constant, the transmission and reflection coefficients can be expressed as Eqs. (B26)-(B28). Thus the transmission coefficient is simply given by

$$|t_{ss}|^2 \approx \left( 1 - \frac{\pi \alpha_F \Re(\sigma_{xx})}{\sigma_0} \right)^2 + \frac{\pi^2 \alpha_F^2}{4} \Im m(\sigma_{xy})^2 \frac{\Im(\sigma_{xx})}{\sigma_0^2}.$$

Although both real and imaginary parts of the optical conductivity contribute to the transmittance, we can safely ignore the imaginary part for all practical purpose. In fact, keeping only first order term in $\alpha_F$ suffices and we have, $|t_{ss}|^2 \approx 1 - \pi \alpha_F \Re(\sigma_{xx})/\sigma_0$. Similarly, the diagonal component of the transmittance for the $p$– component is given as, $|t_{pp}|^2 \approx 1 - \pi \alpha_F \Re(\sigma_{yy})/\sigma_0$. Therefore, for an anisotropic system, where $\sigma_{xx} \neq \sigma_{yy}$, we have,

$$|t_{pp}|^2 - |t_{ss}|^2 \approx \frac{\pi \alpha_F}{\sigma_0} \left[ \Re(\sigma_{xx}) - \Re(\sigma_{yy}) \right]. \tag{24}$$

FIG. 5. Polarization and intensity dependence of different components of the transmission and reflection probability. (a) The variation of diagonal transmission probability for $s$– and $p$– components, $|t_{ss}|^2$ and $|t_{pp}|^2$ with $\theta$ for $\zeta = 5$. The off diagonal transmission/reflection probability is shown in (c). The $\theta$ dependence of the two diagonal components of the reflection probability are shown in (e). The approximate form of their angular variation (marked with dashed black curve) in (a), (c) and (e) are given by Eqs. (B26), (B27) and (B28). In panel (b) (d) and (e) we have shown the $\zeta$ dependence of the different components of the transmission and reflection probabilities for $\theta = \pi/4$. All parameters here are identical to that of Fig. 4.
For a 2D system like graphene which has an isotropic band-structure in vicinity of the Dirac point with $v_x = v_y = v_F$, we have $\sigma_{xx} = \sigma_{yy} = \sigma_0$ and hence, $|t_{ss}|^2_{\zeta=0} = |t_{pp}|^2_{\zeta=0} \approx 1 - \pi \alpha_F$.\textsuperscript{22,39} However for a system with anisotropic massless Dirac band-structure as in 8-\textit{P}mn\textit{m}n borophene, we have $|t_{pp}|^2_{\zeta=0} \approx 1 - \pi \alpha_F v_x/v_{x}$, while $|t_{ss}|^2_{\zeta=0} \approx 1 - \pi \alpha_F v_{x}/v_{y}$. Thus in 8-\textit{P}mn\textit{m}n borophene the transmittance for $s-$ and $p-$ polarized optical beam are not identical, even in the linear response regime. This transmission anisotropy can in fact be used to measure the ratio of the anisotropic Dirac velocities, via the relation

$$|t_{pp}|^2_{\zeta=0} - |t_{ss}|^2_{\zeta=0} \approx \pi \alpha_F \left( \frac{v_x}{v_y} - \frac{v_y}{v_x} \right).$$  

(25)

Numerically exact components of transmission and reflection probability are presented in Fig. 5. Panels (a), (c) and (e) show their dependence on the polarization angle $\theta$ for 8-\textit{P}mn\textit{m}n borophene. We have chosen $\zeta = 5$, $\theta_i = \theta_f = 0$ and $n_i = n_f = 1$. Similar to the optical conductivity, here also we see that the exact results match reasonably well with the $\theta$ dependence predicted by the approximate relations, Eqs. (B26), (B27) and (B28) as indicated by the dashed lines in panels (a), (c) and (e) of Fig. 5. The impact of varying optical field strength is shown in panels (b), (d) and (f).

C. Polarization rotation

We next use these derived reflection and transmission coefficients to calculate Kerr and Faraday polarization rotation angle. In essence, the existence of finite off-diagonal components of the coefficients, $\{r_{sp}, r_{ps}, t_{sp}, t_{ps}\}$, which in turn arise due to finite $\sigma_{xy}(\omega)$, leads polarization rotation. Such polarization rotation in the reflected beam is measurable and has been experimentally explored in graphene, in presence of a perpendicular magnetic field\textsuperscript{30,41} leading to a finite $\sigma_{xy}$. In the present context, the origin of finite $\sigma_{xy}$ is essentially the nonlinear response in the optical field strength\textsuperscript{42}. We find that while the polarization rotation is estimated to be small, it is within measurable regime of existing experimental techniques.

In general, the rotation in the polarization angle in the reflected beam can be expressed in terms of the following dimensionless complex quantities defined by

$$\chi_{Kerr}^s = -\frac{r_{sp}}{r_{ss}}, \quad \text{and} \quad \chi_{Kerr}^p = \frac{r_{sp}}{r_{pp}},$$

(26)

where the superscript $p$ and $s$ denote the $s-$ or $p-$ polarization of the incident beam. Similarly the rotation in the polarization angle in the transmitted beam can be expressed in terms of the following dimensionless complex numbers\textsuperscript{38},

$$\chi_{Faraday}^s = -\frac{t_{ps}}{t_{ss}}, \quad \text{and} \quad \chi_{Faraday}^p = \frac{t_{sp}}{t_{pp}}.$$  

(27)

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig6.png}
\caption{Kerr and Faraday rotation as a function of the polarization angle and the optical intensity of the incident beam. Panel (a) and (c) shows the exact $\Phi_{Kerr}$ and $\Phi_{Faraday}$ as a function of $\theta$ for $\zeta = 5$, respectively. The corresponding dashed black curves follow the angular dependence described by Eq. (B31) and Eq. (B32). The intensity dependence of the corresponding polarization rotation angles is shown in (b) and (d) for $\theta = \pi/4$. Note that the $s-$ and $p-$ components of Kerr and Faraday rotation angles are out-of phase with respect to each other. All parameters here are identical to that of Fig. 4.}
\end{figure}

Now the polarization angle $\Phi$, the azimuth of the major axes of the polarization ellipse of the reflected or transmitted beam, is given by\textsuperscript{38,33}

$$\tan(2\Phi_{M}^{s/p}) = \frac{2\Re[\chi_{M}^{s/p}]}{1 - |\chi_{M}^{s/p}|^2},$$  

(28)

where the subscript $M = Kerr (Faraday)$ for the reflected (transmitted) optical beam. The ellipticity $\varepsilon$, or the major - minor axis ratio of the corresponding polarization ellipse is given by\textsuperscript{38}

$$\varepsilon_{M}^{s/p} = \tan\left(\frac{1}{2}\sin^{-1}\left(\frac{2\Im[\chi_{M}^{s/p}]}{1 + |\chi_{M}^{s/p}|^2}\right)\right).$$  

(29)

We emphasize that in Eqs. (28)-(29), it is essential to include the imaginary part of the optical transmission and reflection coefficients, for an accurate evaluation of the polarization rotation angle and the ellipticity\textsuperscript{38,43-45}. Note that in the limiting case of small optical fields, such that $\zeta^2 \ll 1$, the polarization angle dependence of both $\Phi_{M}^{p/s}$ and $\varepsilon_{M}^{s/p}$, is approximately given by $\sin(2\theta)$ [see Eqs. (B19)-(B22)]. Both $\Phi_{M}^{p/s}$ and $\varepsilon_{M}^{p/s}$ can be experimentally measured by spectroscopic ellipsometry\textsuperscript{46}.

The exact dependence of the polarization rotation angle on $\theta$ and $\zeta$ is shown in Fig. 6. As expected, the angular dependence of the polarization rotation is reasonably captured by the approximate expressions in Eq. (B31) and Eq. (B32). Similarly Fig. 7, shows the $\theta$ and $\zeta$ dependence of the ellipticity. We propose that the $\theta$ dependence of the polarization rotation and the ellipticity...
An estimate of the optical field strength needed to experimentally observe the polarization rotation can be obtained from Eq. (B19). To zeroth order in \( \alpha_F \), Eq. (B19) reduces to

\[
\chi^s\text{Kerr} \approx -\zeta^2 \frac{\tilde{e}_s^2 \sin(2\theta)}{4}.
\]

Accordingly, Eq. (30) sets a practical limit \( \zeta \sin 2\theta > 4 \times 10^{-6} \) for graphene and we use this as the basis of our estimation. Furthermore, the limit translates to a requirement of \( \zeta > 2 \times 10^{-3} \), or equivalently for the optical field strength to be \( E_0 > 4 \hbar \omega \sqrt{1 - \gamma_2^2}/(\epsilon v_F) \times 10^{-6} \). Assuming \( \hbar \omega = 1.5 \) eV and \( \sqrt{1 - \gamma_2^2} \approx 10^{13} \) s\(^{-1}\), we would need the optical field to be of the order of \( E_0 \geq 3 \times 10^4 \) V/m, which is easily achievable with current technology.

In case of graphene or 8-\( Pmmn \) borophene, the measured nonlinear Kerr angle \( \theta \) will vary in a manner similar to \( \sin(2\theta) \) as a function of the polarization angle, and \( \theta \) depend on the strength of the optical field.

Experimentally, however, there are other possible effects which can also lead to polarization rotation in suspended graphene sheets. These include the presence of impurities, static folds and ripples, gate induced bending of the graphene sheet and effects of clamping at the edge of the sample. Nevertheless, it can be noted that none of these effects can lead to, 1) a \( \sin(2\theta) \) like dependence of the Kerr angle on the polarization angle and 2) the optical field strength dependence of the Kerr angle \( (E_0^2) \) for small fields. In fact, the resulting Kerr angle rotation due to all these stray effects is expected to be completely independent of the strength of optical field strength.

A simple experimental way to separate these effects from the proposed nonlinear response in the Kerr angle, is to measure the resulting Kerr rotation at a given location in the suspended graphene sheet as a function of \( \theta \) for two different values of the optical field intensity. All stray rotation signals should be common mode to both the measurements, thereby dropping off, when a difference is taken. The observation of the \( \sin(2\theta) \) like dependence in the difference would be a definitive signature of observing and estimating optical nonlinearities of 2D materials with Dirac cone like dispersion.

**FIG. 7.** The ellipticity for the reflected and transmitted light, as a function of the polarization angle and optical intensity of the incident beam. Panel (a) and (c) shows the exact \( \varepsilon_{Kerr} \) and \( \varepsilon_{Faraday} \) as a function of \( \theta \) for \( \zeta = 5 \). The corresponding dashed black curves follow the angular dependence described by Eqs. (B33)-(B34). The intensity dependence of the two are shown in panels (b) and (c) for \( \theta = \pi/4 \) respectively. All parameters here are identical to that of Fig. 4.

as shown in Figs. 6-7, and the variation of their magnitude with changing optical field strength, can be used experimentally to study nonlinear optical effects in various 2D materials.

**V. EXPERIMENTAL IMPLICATIONS**

We finally explore viability of measuring the proposed nonlinear Kerr rotation in graphene or 8-\( Pmmn \) borophene. High quality, single crystal free standing monolayer graphene samples are routinely fabricated by several groups and furthermore, magneto-optical Kerr and Faraday rotation in graphene has been measured experimentally\textsuperscript{[20,41]}. Simultaneously, with precision polarization elements (Glan-Thomson polarizers, for example) a rotation accuracy of \( 10^{-6} \) radians is routinely achieved in optics experiments, setting the limit of measurement.

An estimate of the optical field strength needed to experimentally observe the polarization rotation can be obtained from Eq. (B19). To zeroth order in \( \alpha_F \), Eq. (B19) reduces to

\[
\chi^s\text{Kerr} \approx \frac{-\zeta^2 \tilde{e}_s^2 \sin(2\theta)}{4}.
\]

VI. CONCLUSION

We have presented a study of nonlinear and anisotropic optical response of 2D massless Dirac materials like graphene and the 8-\( Pmmn \) borophene. Starting from a two band Hamiltonian and considering the carrier-field interaction in length gauge, we have obtained analytical expressions for the nonlinear steady state density matrix elements i.e., steady state population inversion and inter-band coherence, of the photo-excited carriers. The photo-excited population inversion and the interband coherence lead to a finite transverse optical conductivity \( \sigma_{xy}(\omega) \), beyond the linear response regime. This in turn modifies the transmission and reflection coefficients, giving rise to finite \{\( \tau_{sp}, \tau_{ps}, t_{sp}, t_{ps} \)\} bearing definitive signatures in Faraday and Kerr polarization rotations.

In particular, the Kerr angle has a unique \( \sin(2\theta) \) like dependence on the linear polarization angle of the incident beam along with a strong dependence on the incident field strength. We conclude that the nonlinear polarization rotation for graphene and 8-\( Pmmn \) borophene is measurable with current techniques and can lead to applications in designing polarization switches, field controlled polarization controllers and wave plates.
Appendix A: The real and imaginary part of the optical conductivity

Momentum resolved interband current for a generic two band Hamiltonian is defined as\textsuperscript{30},

\[ J_k = -2 \text{Re} \left[ p_k M_k^{vc} \right], \quad (A1) \]

where \( p_k \) is the interband coherence and \( M_k^{vc} \) is an off-diagonal component of optical transition matrix denoting a transition from valence to conduction band. Since\textsuperscript{31,32},

\[ p_k = p_{1k}e^{i\omega t} + p_{2k}e^{-i\omega t}, \quad (A2) \]

we have,

\[ J_k = -p_{1k}M_k^{cv}e^{i\omega t} - p_{2k}M_k^{cv}e^{-i\omega t} - p_{1k}^*M_k^{cv}e^{-i\omega t} - p_{2k}^*M_k^{cv}e^{i\omega t}. \quad (A3) \]

To define the real and imaginary part of the current response, it is generally expressed as \( J_k(t) = \frac{1}{2}(J_k(\omega)e^{-i\omega t} + H.c.) \). Thus we have

\[ \frac{1}{2}J_k(\omega) = -p_{2k}M_k^{cv} - p_{1k}^*M_k^{cv}, \quad (A5) \]

or more explicitly,

\[ J_k(\omega) = -\frac{in_k}{\hbar\omega_k} \left[ (E \cdot M_k^{cv})M_k^{cv} \right]_{(\omega + \omega_k + i\gamma_2)} + (E \cdot M_k^{cv})M_k^{cv} \right]_{(\omega - \omega_k + i\gamma_2)}. \quad (A6) \]

To separate the current response into its real and imaginary part, we note that in general the optical transition matrix can be a complex quantity. For 2D systems we can express it as,

\[ M_k^{vc} = (M_k^x + iM_k^y) \hat{x} + (M_k^y + iM_k^x) \hat{y}, \quad (A7) \]

and \( M_k^{vc} = (M_k^{vc})^* \). Using this we have,

\[ [(E \cdot M_k^{vc})M_k^{cv}]_x = E_x|M_x|^2 + E_y \sum_{l=r,i} M_l^x M_l^y \]

\[ + iE_y \sum_{p,q=r,i} \epsilon_{pq} M_p^x M_q^y. \quad (A8) \]

Similarly,

\[ [(E \cdot M_k^{vc})M_k^{cv}]_y = E_x \sum_{l=r,i} M_l^y M_l^y + E_y|M_y|^2 \]

\[ - iE_x \sum_{p,q=r,i} \epsilon_{pq} M_p^x M_q^y, \quad (A9) \]

where we have defined \( \epsilon_{ri} = 1 \) and \( \epsilon_{ir} = -1 \). The \( x \)- and \( y \) components of \((E \cdot M_k^{vc})M_k^{vc}\) can be found by taking complex conjugate of the above expressions. Substituting Eqs. (A8)-(A9) in Eq. (A6), the real and imaginary parts of the current can be obtained to be

\[ \text{Re}(J_x) = -\frac{n_k}{\hbar\omega_k} \left[ (E_x|M_x|^2 + E_y \sum_{l=r,i} M_l^x M_l^y) X_1 + E_y \sum_{\{p,q=r,i\}} \epsilon_{pq} M_p^x M_q^y Y_2 \right], \quad (A10) \]

\[ \text{Im}(J_x) = -\frac{n_k}{\hbar\omega_k} \left[ -E_y \sum_{\{p,q=r,i\}} \epsilon_{pq} M_p^x M_q^y X_2 + \left( E_x|M_x|^2 + E_y \sum_{l=r,i} M_l^x M_l^y \right) Y_1 \right], \quad (A11) \]

\[ \text{Re}(J_y) = -\frac{n_k}{\hbar\omega_k} \left[ \left( E_x \sum_{l=r,i} M_l^y M_l^y + E_y|M_y|^2 \right) X_1 - E_x \sum_{\{p,q=r,i\}} \epsilon_{pq} M_p^x M_q^y Y_2 \right], \quad (A12) \]

\[ \text{Im}(J_y) = -\frac{n_k}{\hbar\omega_k} \left[ E_x \sum_{\{p,q=r,i\}} \epsilon_{pq} M_p^x M_q^y X_2 + \left( E_x \sum_{l=r,i} M_l^y M_l^y + E_y|M_y|^2 \right) Y_1 \right]. \quad (A13) \]

Here we have defined the following:

\[ X_1 = \frac{\gamma_2}{(\omega - \omega_k)^2 + \gamma_2^2} + \frac{\gamma_2}{(\omega + \omega_k)^2 + \gamma_2^2}, \quad (A14) \]

\[ Y_1 = \frac{\omega - \omega_k}{(\omega - \omega_k)^2 + \gamma_2^2} + \frac{\omega + \omega_k}{(\omega + \omega_k)^2 + \gamma_2^2}. \quad (A15) \]
The corresponding band dispersion is given by

\[ X_2 = \frac{\gamma_2}{(\omega - \omega_k)^2 + \gamma_2^2} - \frac{\gamma_2}{(\omega + \omega_k)^2 + \gamma_2^2}, \]

(A16)

\[ Y_2 = \frac{\omega - \omega_k}{(\omega - \omega_k)^2 + \gamma_2^2} - \frac{\omega + \omega_k}{(\omega + \omega_k)^2 + \gamma_2^2}. \]

(A17)

Till now the formalism is very general and works for any 2D material. Below we discuss the implications for massless Dirac fermions with an anisotropic and tilted Dirac cone - as in $8\text{Pmnn}$ Borophene.

Expressing Eq. (2), in terms of the generic two band Hamiltonian of Refs. [30 and 32], \[ H = \sum_k h_k \cdot \sigma, \] we have, \[ h_{0k} = h_{v1}, \] \[ h_{1k} = h_{x}, \] \[ h_{2k} = h_{y}, \] and \[ h_{3k} = 0. \] The corresponding band dispersion is given by

\[ \epsilon_k^\lambda = h_{v1} k_y + \lambda \sqrt{h^2 v_x^2 k_x^2 + h^2 v_y^2 k_y^2}, \]

(A18)

where \( \lambda = -(+ \) for valence (conduction) band. The transition frequency is given by,

\[ \omega_k = \epsilon_k^+ - \epsilon_k^- = 2 v_F k |\sqrt{v_x^2 \cos^2 \phi_k + v_y^2 \sin^2 \phi_k}|. \]

(A19)

The corresponding 2D phase space integration factor is

\[ |k|dk = \frac{\omega_k d\omega_k d\phi_k}{4 v_F^2 \left( v_x^2 \cos^2 \phi_k + v_y^2 \sin^2 \phi_k \right)}. \]

(A20)

The corresponding optical matrix elements are given by\[30,32,]

\[ M_k^{ee} = \frac{ie}{\hbar v_F} (h_2 \nabla_k h_1 - h_1 \nabla_k h_2). \]

(A21)

For the case of Eq. (2), we have

\[ \nabla_k h_1 = \left( \frac{\partial (h_{v1} k_y)}{\partial k_x}, \frac{\partial (h_{v1} k_y)}{\partial k_y} \right) = (h_{v1}, 0), \]

(A22)

\[ \nabla_k h_2 = \left( \frac{\partial (h_{x} k_y)}{\partial k_x}, \frac{\partial (h_{x} k_y)}{\partial k_y} \right) = (0, h_{x}), \]

(A23)

finally,

\[ M_k^{ee} = i e v_F \tilde{M}_k (\sin \phi_k, -\cos \phi_k), \]

(A24)

with,

\[ \tilde{M}_k = \sqrt{v_x^2 \cos^2 \phi_k + v_y^2 \sin^2 \phi_k}. \]

(A25)

The real part of the longitudinal conductivity, \( \sigma_{xx} \) can be obtained from Eq. A10. In the regime of allowed optical transitions, i.e., \( \hbar \omega > 2 \mu \), it is given by

\[ \text{Re}(\sigma_{xx}) = \frac{\text{Re}(J_x)}{E_x} \]

(A26)

\[ = \frac{\sigma_0}{\pi^2} \int \frac{d\omega_k d\phi_k G_k M^2 \sin^2 \phi_k}{(v_x^2 \cos^2 \phi_k + v_y^2 \sin^2 \phi_k)} X_1. \]

The imaginary component can simply be obtained by replacing \( X_1 \) with \( Y_1 \) in the above expression. Note that to evaluate Eq. (A26) numerically based on a continuum model, we have to use a band cutoff. In this paper we use the energy cutoff of 8 eV for all calculations, based on half bandwidth of the tight-binding Hamiltonian of graphene. The calculation of all other conductivity components proceeds along similar lines.

**Appendix B: The reflection and transmission coefficients in terms of conductivity**

To start with we resolve the electric field vector of the incoming optical beam into \( s \) - and \( p \) - components and then we find the associated transmission and reflection coefficients. The existence of an off-diagonal (sp) component in the transmission/reflection matrix is ensured by finite non-zero value of \( \sigma_{xy} \) for an arbitrary polarization angle \( \theta \). Following Ref. [47], let \( E_i, E_p \) and \( E_t \) be the electric field vector for the incoming, reflected and the transmitted part of the light beam, respectively, and \( B_i, B_r \) and \( B_t \) be the corresponding magnetic field vectors. If the wave vectors and frequencies for the same are denoted as \( k_i, k_r \), and \( k_t \), and \( \omega_i, \omega_r \), and \( \omega_t \) respectively, then from Fig. 8, we have

\[ E_i = (E_i^s, E_i^p \cos \theta_i, E_i^p \sin \theta_i) e^{ik_i \cdot r - i \omega_i t}, \]

\[ E_r = (E_r^s, -E_r^p \cos \theta_r, E_r^p \sin \theta_r) e^{ik_r \cdot r - i \omega_r t}, \]

\[ E_t = (E_t^s, E_t^p \cos \theta_t, E_t^p \sin \theta_t) e^{ik_t \cdot r - i \omega_t t}. \]

FIG. 8. Schematic for calculation of transmission and reflection coefficients. Incident light falls on the 2D material at an angle \( \theta_i \), w.r.t. \( \hat{z} \) axis and is reflected and transmitted at \( \theta_r \) and \( \theta_t \) respectively. Both \( s \) - and \( p \) - components are shown for incident \( (E_i) \), reflected \( (E_r) \) and the transmitted \( (E_t) \) light along with their corresponding wave vectors \( k_i, k_r \), and \( k_t \). The \( p \) - polarization of the incident, reflected and transmitted beams is in the \( y - z \) plane which is also the plane of incidence. The \( s \) - polarization in all three beams is along the \( x \) axis. The dotted perpendicular line coincides with the \( \hat{z} \) axis.
Now using $\mathbf{B} = (n/c) \hat{k} \times \mathbf{E}$, with $n$ denoting the refractive index of the medium, we have

$$
\mathbf{B}_t = \frac{n_i}{c} (E_{t\parallel}^p, E_{t\parallel}^s \cos \theta_t, -E_{t\parallel}^s \sin \theta_t) e^{i(k_t \cdot r - \omega_t t)},
$$

$$
\mathbf{B}_s = \frac{n_i}{c} (E_{s\parallel}^p, E_{s\parallel}^s \cos \theta_s, -E_{s\parallel}^s \sin \theta_s) e^{i(k_s \cdot r - \omega_s t)},
$$

$$
\mathbf{B}_t = \frac{n_i}{c} (E_{t\parallel}^p, -E_{t\parallel}^s \cos \theta_t, -E_{t\parallel}^s \sin \theta_t) e^{i(k_t \cdot r - \omega_t t)}.
$$

The fields at the interface of the monolayer with air (or vacuum) satisfy the following boundary condition,

$$
\mathbf{E}_{t\parallel}^i = \mathbf{E}_{t\parallel}^s, \quad \text{and} \quad \mathbf{B}_{t\parallel}^i - \mathbf{B}_{t\parallel}^s = \mu_0 \mathbf{J} \times \hat{z}, \tag{B1}
$$

where $\mathbf{E}_i = \mathbf{E}_t + \mathbf{E}_r$, $\mathbf{E}_s = \mathbf{E}_t$, $\mathbf{B}_i = \mathbf{B}_t + \mathbf{B}_r$, $\mathbf{B}_s = \mathbf{B}_t$ and $\mathbf{J}$ is the current density produced in the monolayer due to the incident optical beam. Here we will ignore the possibility of higher harmonics generation\(^{48}\), which anyway happens for very high intensity laser beams. We also assume that the medium on either side of the monolayer is identical. The Snell’s law follows from the first boundary condition. By matching the space and time dependent exponents: $(k_t \cdot r - \omega_t t) = (k_s \cdot r - \omega_s t)$ for the monolayer located at $z = 0$ we obtain $\omega_s = \omega_t = \omega_0$, $\cos \theta_t = \cos \theta_s$, and $n_i \sin \theta_t = n_i \sin \theta_t$. Matching the $x$ and $y$ components of both the electric and magnetic fields at $z = 0$, as per Eq. (B1), we arrive at a matrix equation:

$$
\mathbf{S}_{4 \times 4} \mathbf{E}_i = \mathbf{E}_i. \tag{B2}
$$

Here $\mathbf{E}_i = [E_{t\parallel}^i, E_{s\parallel}^i, E_{t\parallel}^s, E_{s\parallel}^s]^T$ represents the $s$ and $p$ components of the outgoing fields,

$$
\mathbf{S}_{4 \times 4} = \begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & 1 & 0 & 0 \\
\mu_0 \sigma_{yz} & \mu_0 \sigma_{yx} & n_i \cos \theta_t/c + \mu_0 \sigma_{zx} & n_i \cos \theta_t/c \\
n_i \cos \theta_t/c + \mu_0 \sigma_{zx} & n_i \cos \theta_t/c & \mu_0 \sigma_{xy} \cos \theta_t & -n_i/c
\end{pmatrix}. \tag{B3}
$$

and, $\mathbf{E}_i = [E_{t\parallel}^i, E_{s\parallel}^i, E_{t\parallel}^s, E_{s\parallel}^s]^T$ comprises of the incoming fields. Solving the matrix Eq. (B2), the transmitted electric fields in the $p$- and $s$ directions can be expressed as

$$
\begin{pmatrix}
E_{t\parallel}^p \\
E_{t\parallel}^s
\end{pmatrix} = \begin{pmatrix}
\frac{t_{ps}}{E_{t\parallel}^i} & \frac{t_{ps}}{E_{t\parallel}^i} \\
\frac{t_{ps}}{E_{t\parallel}^i} & \frac{t_{ps}}{E_{t\parallel}^i}
\end{pmatrix} \begin{pmatrix}
E_{t\parallel}^p \\
E_{t\parallel}^s
\end{pmatrix}. \tag{B4}
$$

Here the diagonal part of the transmission coefficients are given by,

$$
t_{ss} = \left. \frac{E_{t\parallel}^s}{E_{t\parallel}^i} \right|_{E_{t\parallel}^s = 0} = \frac{2n_i \sigma_2}{c \mu_0 \sigma_T} \cos \theta_t, \tag{B5}
$$

and,

$$
t_{pp} = \left. \frac{E_{t\parallel}^p}{E_{t\parallel}^i} \right|_{E_{t\parallel}^p = 0} = \frac{2n_i \sigma_1}{c \mu_0 \sigma_T} \cos \theta_t. \tag{B6}
$$

Here we have defined

$$
\sigma_T = (\sigma_1 \sigma_2 - \sigma_{xy} \sigma_{yx} \cos \theta_t \cos \theta_t), \tag{B7}
$$

along with

$$
\sigma_1 = n_i \cos \theta_t/(c \mu_0) + n_t \cos \theta_t/(c \mu_0) + \sigma_{xx}, \tag{B8}
$$

$$
\sigma_2 = n_i \cos \theta_t/(c \mu_0) + n_t \cos \theta_t/(c \mu_0) + \sigma_{yx} \cos \theta_t \cos \theta_t. \tag{B9}
$$

The off diagonal elements of the transmission matrix are given by,

$$
t_{sp} = \left. \frac{E_{t\parallel}^s}{E_{t\parallel}^i} \right|_{E_{t\parallel}^s = 0} = -\frac{2n_i \sigma_{xy}}{c \mu_0 \sigma_T} \cos \theta_t \cos \theta_t, \tag{B9}
$$

1. Lowest order ($\zeta^2$) nonlinear correction in the reflection and transmission coefficients

Similar to the case of optical conductivity, while the exact numerical evaluation of the transmission and reflection coefficients is possible, it is insightful to express the different reflection and transmission coefficients analytically up to second order in the optical field strength. Using Eqs. (16)-(22) in Eqs. (B5)-(B10), we derive the expressions for the different transmission coefficients up to $\zeta^2$. The expression for $s$- and $p$- components of longitudinal transmission coefficients up to $\zeta^2$ (valid for $\zeta \ll 1$) are,
Similarly the reflection coefficients can be obtained by using above expressions in Eqs. (B11)-(B14).

Applying Eqs. (26)-(27) to the case of anisotropic 2D gapless Dirac materials, we obtain the following expressions for s- and p- components of $\chi_{\text{Kerr}}$ up to $\zeta^2$ to be,

$$\chi_{\text{Kerr}} = \zeta^2 \eta(\theta) \left[ k_3^2 - \left( 1 + \frac{2}{\pi \alpha F} \right) - 2i k_3 \left( 1 + \frac{1}{\pi \alpha F} \right) \right],$$

(B19)

and,

$$\chi_{\text{Faraday}} = -\zeta^2 \eta(\theta) \left[ k_3^2 \alpha - \left( \alpha + \frac{2}{\pi \alpha F} \right) - 2i k_3 \left( \alpha + \frac{1}{\pi \alpha F} \right) \right].$$

(B20)

Here we have defined $\eta(\theta) = v_0^2 \sin 2\theta / [2\pi \alpha F \left( k_3^2 + 1 \right)]$. Similarly the s- and p- components of $\chi_{\text{Faraday}}$ up to $\zeta^2$ are given by,

$$\chi_{\text{Faraday}}^s = -\zeta^2 \eta(\theta) \frac{1 + k_3^2}{2} \left[ \left( 1 + \frac{2}{\pi \alpha F} \right) + ik_3 \right],$$

(B21)

where $k_4 = v_x^2 + 3v_y^2$, $k_5 = v_x^2 - 3v_y^2$, and $\alpha = \tilde{v}_x / \tilde{v}_y$. Interestingly the presence of a finite $\sigma_{xy}(\omega)$, allows for the transmission of a s- (or p-) polarized beam as a p- (or s-) polarized beam. The complex off diagonal transmission coefficient is given by,

$$t^s_p = \frac{2}{\pi \alpha F} \frac{\alpha(1 + 2i k_3 \alpha + \frac{2}{\pi \alpha F})}{k_3^2 + \left( 1 + \frac{2}{\pi \alpha F} \right)^2} \left[ k_3^2 \alpha^2 + \left( \alpha + \frac{2}{\pi \alpha F} \right)^2 \right] \left[ k_3^2 + 2i \left( 1 + \frac{2}{\pi \alpha F} \alpha \right) \right].$$

(B15)

and,

$$t^p_p = \frac{2}{\pi \alpha F} \frac{\alpha(1 + 2i k_3 \alpha + \frac{2}{\pi \alpha F})}{k_3^2 + \left( 1 + \frac{2}{\pi \alpha F} \right)^2} \left[ k_3^2 \alpha^2 + \left( \alpha + \frac{2}{\pi \alpha F} \right)^2 \right] \left[ k_3^2 + 2i \left( 1 + \frac{2}{\pi \alpha F} \alpha \right) \right].$$

(B16)

Here we have defined, $k_1 = 3v_x^2 + v_y^2$, $k_2 = 3v_x^2 - v_y^2$, $k_3 = \frac{1}{2\pi} \ln \left[ (\omega + 2\mu / \hbar)^2 / (\omega - 2\mu / \hbar)^2 \right].$

Note that $t^s_p \propto \zeta^2$, and hence it is finite only due to the nonlinear optical response. Similar expressions for reflection coefficients can be obtained be using above expressions in Eqs. (B11)-(B14).

All of these $\chi^{\mu s}$, show a sin(2$\theta$) dependence on the angle of polarization of the incident beam. Thus in order to have non-vanishing polarization rotation, the polarization direction of the incoming beam should make a finite angle with the x-axis, which denotes one of the principal crystal axis of the 2D gapless Dirac material.

2. Simplified form of polarization rotation and ellipticity up to lowest order in the fine structure constant

Here we specifically discuss the case of vertical incidence, i.e., $\theta_i = \theta_r = 0$. The case of generic incidence angle can be easily obtained from Eq. (B5)-(B14). For vertical incidence we have, $r_{sp} = t_{ps}$ and $t_{pp} = 1 - t_{pp}$. Now using the fact that $\sigma_{xy} = \sigma_{yx}$, and defining the normalized conductivity $\tilde{\sigma}_{ij} = \sigma_{ij} / \sigma_0$, we have

$$t_{ss} = \frac{2}{\pi \alpha F} \frac{(2/\pi \alpha F + \tilde{\sigma}_{yy})}{(2/\pi \alpha F + \tilde{\sigma}_{xx})(2/\pi \alpha F + \tilde{\sigma}_{yy}) - \tilde{\sigma}_{xy}^2}. $$

(B23)
\[ t_{pp} = \frac{2}{\pi \alpha_F} \frac{(2/\pi \alpha_F + \tilde{\sigma}_{xx})(2/\pi \alpha_F + \tilde{\sigma}_{yy}) - \tilde{\sigma}_{xy}^2}{(2/\pi \alpha_F + \tilde{\sigma}_{xx})(2/\pi \alpha_F + \tilde{\sigma}_{yy}) - \tilde{\sigma}_{xy}^2}, \] (B24)

where \( \alpha_F = e^2/4\pi \varepsilon_0 \) \( (\ll 1) \) is the fine structure constant. Similarly we have

\[ r_{sp} = \frac{-2}{\pi \alpha_F} \frac{\tilde{\sigma}_{xy}}{(2/\pi \alpha_F + \tilde{\sigma}_{xx})(2/\pi \alpha_F + \tilde{\sigma}_{yy}) - \tilde{\sigma}_{xy}^2}. \] (B25)

Now given the fact that for 2D materials, typically we have \( \tilde{\sigma}_{ij} \) to be of the order of 1, we can do an expansion in \( \alpha_F \) to simplify. Doing this expansion of the reflection and transmission coefficient up to first order in \( \alpha_F \), we obtain the complex coefficients:

\[ t_{ss} \approx 1 - \frac{\pi \alpha_F \sigma_{xx}}{2 \sigma_0}, \] (B26)

\[ t_{pp} \approx 1 - \frac{\pi \alpha_F \sigma_{yy}}{2 \sigma_0}, \] (B27)

and,

\[ r_{sp} \approx -\frac{\pi \alpha_F \sigma_{xy}}{2 \sigma_0}. \] (B28)

Using Eqs. (B26)-(B28) in Eq. (26) and Eq. (27), we have (up to order \( \alpha_F \))

\[ \chi_{Kerr}^s \approx \frac{\sigma_{xy}}{\sigma_{xx}}, \text{ and } \chi_{Kerr}^p \approx -\frac{\sigma_{xy}}{\sigma_{yy}}, \] (B29)

and,

\[ \chi_{Faraday}^s = \frac{\pi \alpha_F}{2} \sigma_{xy}, \text{ and } \chi_{Faraday}^p = -\pi \alpha_F \sigma_{xy}. \] (B30)

Note that \( \chi_{Faraday}^s \) is in general smaller by \( \chi_{Kerr}^p \) by a factor of \( \alpha_F \approx 1/137 \). Thus the Kerr angle will always be much larger than the Faraday angle for any 2D system in general.

Using the simplified expression for \( \chi_{Kerr}^{s/p} \), the s- component of the Kerr angle can be approximated as,

\[ \tan(2\Phi_{Kerr}) \approx \frac{2}{|\sigma_{xx}|} \text{Re}[\sigma_{xy}(\sigma_{xx})^*]. \] (B31)

The corresponding Kerr rotation angle for the p- component can be obtained by replacing \( \sigma_{xx} \) with \( -\sigma_{yy} \) in the above equation. The Faraday angle for both s- and p- components are given by,

\[ \tan 2\Phi_{Faraday}^s \approx \pm \pi \alpha_F \text{ Re}(\sigma_{xy}). \] (B32)

Finally, the approximate form (lowest order in \( \alpha_F \)) of the ellipticity for s- component of the reflected beam is,

\[ \varepsilon_{Kerr}^s \approx \tan \left( \frac{1}{2} \sin^{-1} \left[ \frac{2}{|\sigma_{xx}|^2} \text{Im}[\sigma_{xy}(\sigma_{xx})^*] \right] \right). \] (B33)

From Eq. (B29), it follows that ellipticity of the p- component can be obtained by replacing \( \sigma_{xx} \) with \( -\sigma_{yy} \) in Eq. (B33), and it is given by

\[ \varepsilon_{Kerr}^p \approx \tan \left( \frac{1}{2} \sin^{-1} \left[ \frac{2}{|\sigma_{yy}|^2} \text{Im}[\sigma_{xy}(-\sigma_{yy})^*] \right] \right). \] (B34)

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1. F. Bonaccorso, Z. Sun, T. Hasan, and A. C. Ferrari, “Graphene photonics and optoelectronics,” Nature Photonics 4, 611 EP – (2010).
2. Ken Fai Mak and Jie Shan, “Photonics and optoelectronics of 2d semiconductor transition metal dichalcogenides,” Nature Photonics 10, 216 (2016).
3. Novoselov K. S., Geim A. K., Morozov S. V., Jiang D., Katsnelson M. I., Grigorieva I. V., Dubonos S. V., and Firsov A. A., “Two-dimensional gas of massless Dirac fermions in graphene,” Nature 438, 197–200 (2005).
4. Geim A. K. and Novoselov K. S., “The rise of graphene,” Nat Mater 6, 183191 (2007).
5. Shemeve Z. Butler, Shawnna M. Hollen, Linyou Cao, Yi Cui, Jay A. Gupta, Humberto R. Gutierrez, Tony F. Heinz, Seung Sae Hong, Jiaxing Huang, Ariel F. Ismach, Ezekiel Johnston-Halperin, Masaru Kuno, Vladimir V. Plashnitsa, Richard D. Robinson, Rodney S. Ruoff, Sayeef Salahuddin, Jie Shan, Li Shi, Michael G. Spencer, Mauricio Terrones, Wolfgang Windl, and Joshua E. Goldberger, “Progress, challenges, and opportunities in two-dimensional materials beyond graphene,” ACS Nano 7, 2898–2926 (2013).
6. Baojie Feng, Hui Li, Cheng-Cheng Liu, Ting-Na Shao, Peng Cheng, Yugui Yao, Sheng Meng, Lan Chen, and Ke-Hui Wu, “Observation of dirac cone warping and chirality effects in silicene,” ACS Nano 7, 9049–9054 (2013).
7. Yuehue Ruge, Yuan Yakun, Zheng Jiaxin, Wang Yangyang, Ni Zeyuan, Shi Junjie, Yu Dapeng, Yang Jinbo, and Lu Jing, “Does the Dirac Cone Exist in Silicene on Metal Substrates?” Scientific Reports 4, 5476 (2014).
8. Ken Fai Mak, Changgu Lee, James Hone, Jie Shan, and Tony F. Heinz, “Atomically thin monolayers: A new direct-gap semiconductor,” Phys. Lett. 105, 1513–1516 (2015).
9. Han Liu, Adam T. Neal, Zhen Zhu, Zhe Luo, Xianfan Xu, David Tomnek, and Peide D. Ye, “Phosphorene: An unexplored 2d semiconductor with a high hole mobility,” ACS Nano 8, 4033–4041 (2014).
10. Andrew J. Mannix, Xiang-Feng Zhou, Brian Kiraly, Joshua D. Wood, Diego Alducin, Benjamin D. Myers, Xiaojong Liu, Brandon L. Fisher, Ulises Santiago, Jeffrey R. Guest, Miguel Jose Yakaman, Arturo Ponce, Artem R. Oganov, Mark C. Hersam, and Nathan P. Guisinger, “Synthesis of borophenes: Anisotropic, two-dimensional boron polymorphs,” Science 350, 1513–1516 (2015).
Bo Peng, Hao Zhang, Hezhu Shao, Yuanfeng Xu, Rongjun Zhang, and Heyuan Zhu, “The electronic, optical, and thermodynamic properties of borophene from first-principles calculations,” J. Mater. Chem. C 4, 3592–3598 (2016).

Alejandro Lopez-Bezanilla and Peter B. Littlewood, “Electronic properties of 8−pmmn borophene,” Phys. Rev. B 93, 241405 (2016).

Baojie Feng, Osamu Sugino, Ro-Ya Liu, Jin Zhang, Ryu Yokawa, Mitsuaki Kawamura, Takashi limori, Howon Kim, Yukio Hasegawa, Hui Li, Lan Chen, Kehui Wu, Hiroshi Kumigashira, Fumio Komori, Tai-Chang Chiang, Sheng Meng, and Iwao Matsuda, “Dirac fermions in borophene,” Phys. Rev. Lett. 118, 096401 (2017).

A. D. Zabolotskiy and Amit Agarwal, “Nonlinear optical conductivity of a generic two-band system with application to doped and gapped graphene,” Phys. Rev. B 95, 155421 (2017).

A. J. Chaves, N. M. R. Peres, and Tony Low, “Pumping electrons in graphene to the m point in the brillouin zone: Emergence of anisotropic plasmons,” Phys. Rev. B 94, 195438 (2016).

Ashutosh Singh, Saikat Ghosh, and Amit Agarwal, “Nonlinear, anisotropic, and giant photoconductivity in intrinsic and doped graphene,” Phys. Rev. B 97, 045402 (2018).

Evdokia Dremetsiaka, Bruno Dublaik, Simon-Pierre Gorza, Charles Ciret, Marie-Blandine Martin, Stephan Hofmann, Pierre Seneor, Daniel Dolfi, Serge Massar, Philippe Emplit, and Pascal Kockaert, “Measuring the nonlinear refractive index of graphene using the optical kerr effect method,” Opt. Lett. 41, 3281–3284 (2016).

N. A. Savostianova and S. A. Mikhailov, “Optical Kerr Effect in Graphene: Theoretical Analysis of the Optical Heterodyne Detection Technique,” ArXiv e-prints (2018), arXiv:1801.09785 [cond-mat.mes-hall].

R.W. Boyd, Nonlinear Optics (Elsevier Science, 2003).

Claudio Aversa and J. E. Sipe, “Nonlinear optical susceptibilities of semiconductors: Results with a length-gauge analysis,” Phys. Rev. B 52, 14636–14645 (1995).

The cos(2θ) dependent terms are fitted by an expression of the form, \( y_1(\theta) = y_{1 \text{max}} \sin^2 \theta + y_{1 \text{min}} \cos^2 \theta \). On the other hand, the sin(2θ) dependent terms are fitted by a form \( y_2(\theta) = y_{2 \text{max}} - y_{2 \text{min}} \sin \theta \cos \theta \).

Toshihiko Yoshino, “Theory for oblique-incidence magneto-optical faraday and kerr effects in interfaced monolayer graphene and their characteristic features,” J. Opt. Soc. Am. B 30, 1085–1091 (2013).

R. R. Nair, P. Blake, A. N. Grigorenko, K. S. Novoselov, T. J. Booth, T. Stauber, N. M. R. Peres, and A. K. Geim, “Fine structure constant defines visual transparency of graphene,” Science 320, 1308–1308 (2008).

Shimano R., Yumoto G., Yoo J. Y., Matsunaga R., Tanabe S., Hibino H., Morimoto T., and Aoki H., “Quantum Faraday and Kerr rotations in graphene,” Nature Communications 4, 1841 (2013).

Iris Crassee, Julien Levallois, Andrew L. Walter, Markus Ostler, Aaron Bostwick, Eli Rotenberg, Thomas Seyller, Dirk van der Marel, and Alexey B. Kuzmenko, “Giant faraday rotation in single- and multilayer graphene,” Nature Physics 7, 48 (2010).

M. V. Strikha and F. T. Vasko, “Electro-optics of graphene: Field-modulated reflection and birefringence,” Phys. Rev. B 81, 115413 (2010).

Gbor Szchenyi, Mt Vigh, Andor Kormyos, and Jzsef Cserti, “Transfer matrix approach for the kerr and faraday rotation in layered nanostructures,” Journal of Physics: Condensed Matter 28, 375802 (2016).

Wang-Kong Tse and A. H. MacDonald, “Giant magneto-optical kerr effect and universal faraday effect in thin-film topological insulators,” Phys. Rev. Lett. 105, 057401 (2010).

Rahul Nandkishore and Leonid Levitov, “Polar kerr effect and time reversal symmetry breaking in bilayer graphene,” Phys. Rev. Lett. 107, 097402 (2011).

H. Fujiwara, Spectroscopic Ellipsometry: Principles and Applications (Wiley, 2007).

J. Peatross and M. Ware, Physics of Light and Optics (Brigham Young University, Department of Physics, 2011).

N. Bloembergen and P. S. Pershan, “Light waves at the
boundary of nonlinear media,” Phys. Rev. 128, 606–622 (1962).