Topologically guaranteed enhancement of nonlinear optical conductivity of graphene in the presence of spin-orbit coupling

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Abstract
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Topologically guaranteed enhancement of nonlinear optical conductivity of graphene in the presence of spin-orbit coupling

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I. INTRODUCTION

Graphene is a two-dimensional honeycomb lattice of carbon atoms that has been intensely studied because of experimental progress [1–3] and many new physical features such as the half-integer quantum Hall effect [4], finite conductivity at zero charge-carrier concentration [5], perfect quantum tunneling effect [6], and ultrahigh carrier mobility [7]. The low-energy electrons in graphene offer an ideal system for studying the relativistic dynamics. A finite spin-orbit coupling (SOC) or interlayer hopping in a bilayer graphene (BLG) offers a mechanism to generate topological change of the low-energy band structure, known as the trigonal warping [8–10].

The spin-orbit coupling in graphene can be intrinsic or extrinsic. They affect the electronic properties of graphene in different ways. The intrinsic SOC [11] leads to a finite gap between the conduction and valence bands. Its modulation may turn a single layer graphene into a topological insulator [12]. The presence of extrinsic SOC results in a k-dependent energy shift and deforms the band structure [13]. The effect is manifested through the anisotropic spin splitting of the bands at the K (K′) points [9]. The extrinsic SOC can be regarded as Rashba SOC (RSOC), which is generated by the structure inversion asymmetry of the graphene sheet and has an obvious influence on the linear optical conductivity [14,15]. It can be further tuned with an external electric gate voltage, or electrostatic interaction with the substrate. The effects of intrinsic and extrinsic SOCs can be analyzed by using a tight-binding model for monolayer, bilayer, and multilayer graphene [16–18]. The role played by intrinsic and extrinsic SOI in a given physical property is quite different due to three orders of magnitude difference in their relevant energy scales. The energy of the intrinsic SOC [19] is a few μeV while that of the extrinsic SOC is in the order of 1–10 meV [20]. A Rashba splitting of 225 meV has been observed in an epitaxial graphene layer on a Ni(111) surface [13] due to the SU(2) spin symmetry breaking. Under the RSO, the low-energy bands undergo trigonal warping deformation at the K and K′ points of graphene’s Brillouin zone (BZ) in the reciprocal lattice. One Dirac point is now split into four gapless points [9,17,21]. One of them remains at the K (K′) point and the other three link up an equilateral triangle around the K (K′) point. The warping changes the topology of the low-energy bands at the K (K′) points of graphene’s BZ [22,23]. The deformation of their low-energy conical approximation is very similar to what happens in bilayer graphene [24–27]. Mathematically it can be shown that a single layer graphene with SOC and a BLG can be described by an equivalent Hamiltonian.

Graphene has superior optical properties; chief among them is the universal conductance σ0 = 4e2/πh in the low-to-visible-frequency regime [28]. Other interesting properties include the dynamic conductivity [29–32], enhanced optical conductivity of bilayer graphene nanoribbons [33] and strong terahertz conductance under a magnetic field [34]. Both SOC and the interlayer hopping change graphene’s universal conductance from σ0 to 6 σ0. This six times enhancement is independent of the strength of SOC or the interlayer hopping, confirming the topological nature of SOC and interlayer hopping [10,23,35]. Graphene is also a strong natural nonlinear material [36–39]. An important question that remains to be answered is whether a SOC changes the nonlinear response in a way that is topologically guaranteed. In this work we shall show that there exists a topologically guaranteed enhancement of the nonlinear optical conductance by SOC in graphene.

II. MODEL AND NONLINEAR CONDUCTIVITY

For the monolayer graphene with RSO the Hamiltonian is given as $H = H_0 + H_{RSO}$ [11,40], where $H_0$ is the Hamiltonian of a graphene without RSO and $H_{RSO}$ is the Hamiltonian for spin-orbit interaction. In the tight-binding approximation,

$$H_0 = \sum_{k \in BZ} \left( \hat{C}_k^+ \begin{pmatrix} h(k) & 0 \\ 0 & h^*(k) \end{pmatrix} \hat{C}_k \right),$$

(1)
\[
\mathcal{C}^+_{k_x} = \frac{1}{\sqrt{N}} \sum_{k \in BZ} e^{i \vec{k} \cdot \vec{r}} \mathcal{C}^+_{k_x, k}.
\]
\[
\mathcal{C}_x = \frac{1}{\sqrt{N}} \sum_{k \in BZ} e^{-i \vec{k} \cdot \vec{r}} \mathcal{C}_x_{k_x, k}.
\]

Here \( t_h \) is the hopping energy (\( \approx 2.7 \) eV), \( \nu = A, B, C \), \( \mathcal{C}^+_{k_x} \) and \( \mathcal{C}_x \) are creation and annihilation operators for an electron with lattice vector \( l \) in sublattice \( \nu \). \( \vec{I}_B = \vec{I}_A + \delta \), \( h(\vec{k}) = \sum_\delta e^{i \vec{k} \cdot \delta} \) and the operator \( \mathcal{C}^+_{k_x} \) is given as \( \langle \vec{r} | \mathcal{C}^+_{k_x} | 0 \rangle \equiv | \psi_{k_x}^x (\vec{r}) \rangle = \frac{1}{\sqrt{N}} \sum_{k \in BZ} e^{i \vec{k} \cdot \vec{r}} \phi_{2p}(\vec{r} - \vec{I}_A) \) with \( \phi_{2p}(\vec{r}) \) usually chosen as the \( 2p_z \) orbit wave function of the carbon atom. For a fixed Bloch wave vector \( \vec{k} \), the Hamiltonian \( \hat{H}_0 \) in the two-dimensional subspace \( \mathcal{Y}_2^{2D} \) spanned by the basis functions \( \{ | \psi_{k_x}, |, | \psi_{k_y} \rangle \} \) is
\[
\hat{H}_0^{2D}(\vec{k}) = t_h \begin{pmatrix} 0 & h(\vec{k}) \\ h^*(\vec{k}) & 0 \end{pmatrix}.
\]

When the degree of freedom of the spin is taken into account, the subspace becomes four dimensional with four base vectors,
\[
\{| \psi_{k_x}^x \rangle \otimes | \uparrow \rangle, | \psi_{k_y}^x \rangle \otimes | \uparrow \rangle, | \psi_{k_x}^y \rangle \otimes | \downarrow \rangle, | \psi_{k_y}^y \rangle \otimes | \downarrow \rangle \}
\]
\[
\equiv | \vec{k}, \mu \rangle \in \mathcal{Y}_4^{2D},
\]

\( \mu = 1, 2, 3, 4 \). The Hamiltonian \( \hat{H}_0 \) in Hilbert subspace \( \mathcal{Y}_4^{2D} \) with no spin-orbit interaction is the direct product \( \hat{H}_0^{4D} = \hat{H}_0 \otimes \hat{H}_0^{2D} \), where \( \hat{I} \) is the unit operator (matrix) in the spin space. The RSOC term can be written as \( \hat{H}_{RSO} = i \sum_{l \in T} \sum_\delta \left[ (\vec{u}_\delta \cdot \vec{\sigma}) \otimes \mathcal{C}^+_{l \delta} \mathcal{C}_{l \delta^+} - (\vec{u}_{\delta^+} \cdot \vec{\sigma}) \otimes \mathcal{C}^+_{l \delta^+} \mathcal{C}_{l \delta} \right] \).

\[
\hat{H}_{RSO} = i \vec{\sigma} \otimes \begin{pmatrix} 0 & h^*_x(\vec{k}) \\ -h_x(\vec{k}) & 0 \end{pmatrix}
\]

\[
- i \vec{\sigma} \otimes \begin{pmatrix} 0 & h^*_y(\vec{k}) \\ -h_y(\vec{k}) & 0 \end{pmatrix},
\]

where \( h_x = \frac{\lambda}{d_0} \sum_\delta e^{i \vec{k} \cdot \vec{r}} \delta_{n,0} \). The total Hamiltonian in the subspace \( \mathcal{Y}_4^{2D} \) is
\[
\hat{H}^{4D} = \hat{H}_0^{4D} + \hat{H}_{RSO}
\]

\[
= t_h \begin{pmatrix} 0 & \varphi_0 & 0 & i \varphi_+ \Delta \\ \varphi^*_0 & 0 & -i \Delta \varphi^*_+ & 0 \\ 0 & i \Delta \varphi^- & 0 & \varphi_0 \\ -i \varphi_+ \Delta & 0 & \varphi_0 & 0 \end{pmatrix}
\]

(6)

FIG. 1. (Color online) The equienergy contours at the satellite points \( (K_A, K_B, K_C) \) and original Dirac point \( K \).

\[
\varphi_+ = e^{i \frac{ak_x}{\sqrt{3}}} \left[ 1 + 2 e^{-\frac{1}{2}ak_x} \cos \left( \frac{ak_x}{2} \right) \right],
\]

\[
\varphi_- = e^{i \frac{ak_x}{\sqrt{3}}} \left[ 1 - 2 e^{-\frac{1}{2}ak_x} \cos \left( \frac{ak_x}{2} + \frac{\pi}{6} \right) \right].
\]

The eigenvalues of Eq. (6) are given by
\[
E_{\eta_1, \eta_2}(k) = \eta_1 \sqrt{W + \eta_2 \sqrt{W^2 - \left| \varphi_+^2 + \Delta^2 \varphi_- \right|^2}},
\]

(7)

where \( W = |\varphi_0|^2 + \Delta^2 (|\varphi_+|^2 + |\varphi_-|^2) \) and \( \eta_1, \eta_2 = \pm 1 \) for the different energy branches. Because the spin-orbit interaction breaks the \( SU(2) \) invariance, the twofold degenerate states for electron and hole are lifted respectively. However, for the points like \( K, K' \) possessing the high symmetry there still exists double essential degeneracy which is at the original Dirac points. Moreover, there are some double accidental degeneracy points which are dependent on the coupling parameter \( \Delta \).

In the low-energy limit a quantum system with dispersion relation geometry shaped like a Dirac cone is characterized by the effective Hamiltonian \( H = \hbar v_F \vec{\sigma} \cdot \vec{k} \), which is a massless two-dimensional (2D) Dirac equation. The Hamiltonian gives the dispersion relation \( E(k) = \hbar v_F k \). The contribution to the conductivity from a Dirac cone has been discussed in detail in Refs. [36,37]. By direct observation, one can see that the equienergy contour at \( K_A, K_B, K_C \) is a ellipselike shape as shown in Fig. 1 which at \( K_0 \) is strictly a circle. The difference between the \( K_0 \) and \( K_{A(B,C)} \) is of course more fundamental. The Chern number for each gapless point is defined as \( C_{K_i} = (1/2\pi i) \int_{K_0}^{K_i} d\phi A_\phi \), where \( A_\phi \) is the Berry
phase and \( \tan \phi = \delta k_y / \delta k_x \). For the center gapless point [15] \( A^{(K)}_\phi = -i (1/2) \), and \( C_{K_\phi} = -1/2 \). For the three satellite gapless points, \( C_{K_{\phi}} = C_{K_x} = C_{K_y} = 1/2 \). The nonzero Chern numbers of states at \( K_0, K_1, K_2, \) and \( K_3 \) manifest that the wave function can acquire a (Berry) phase \([42]\). Moreover, it is assumed that \( \zeta_x \), \( \zeta_y \), and \( \zeta_p \) are real.

The shape changing has an influence on the validity of the model Hamiltonian \( \hat{H} = \hbar v_F \vec{k} = v_F \vec{p} \cdot \vec{\sigma} \) where the dispersion relation geometry is not exactly a conic section. Considering the symmetry of the Brillouin zone we pay more attention to the band structure around the point \( K_A \) where \( (K_A) = 0 \) for simplicity. To this end, we expand the branch \( E_{k_{1,-1}}(k_x, k_y) \) to the second order at the point \( K_A = \{2 \cos^{-1}(\frac{\Delta_0}{\Delta_1 + \Delta_2}), 0 \} \) and obtain

\[
E_{k_{1,-1}}(k_x, k_y) \approx (\hbar v_F)^2 \left( \zeta_x^2 \delta k_x^2 + \zeta_y^2 \delta k_y^2 \right),
\]

where \( \delta k_x = k_x - K_{Ax}, \delta k_y = k_y - K_{Ay}, \zeta_x^2 = \Delta^2, \zeta_y^2 = 9 \Delta^2 (1 - 4 \Delta^2), \) and the relation \( \hbar v_F = \zeta_x \delta k_x a \) is used. It follows from Eq. (8) that the effective modified dispersion relation is \( E_{k_{1,-1}}(k_x, k_y) = \hbar v_F \sqrt{((\zeta_x \delta k_x)^2 + (\zeta_y \delta k_y)^2)} \) with the constant \( \Delta \neq 0 \). Therefore, the Hamiltonian of the effective quantum system characterized by the dispersion geometry in the neighborhood of the Dirac point \( K_A \) can be phenomenologically written as

\[
\hat{H}_{K_A} = v_F \vec{\sigma} \cdot \vec{p} = v_F \begin{pmatrix} p_+ & p_- \\ 0 & 0 \end{pmatrix},
\]

where \( p_+ = p'_x - i p'_y, p_+ = p'_x + i p'_y, p'_x = \zeta_x \delta k_x, \) \( p'_y = \zeta_y \delta k_y \) with defining \( p_- p'_+ = p_- p'_+ = p'_x p'_y = \zeta_x \delta k_x \). By redefining the time-harmonic external field \( \hat{A}(t) = \hat{A}_0 \exp^{-i \omega t} = \frac{\hbar \omega}{(\Delta_0)_{\text{eff}}} \) is applied on the system in terms of the principle of minimal coupling the Hamiltonian in the external field is expressed as

\[
\hat{H}_{K_{A,\text{inter}}} = v_F \begin{pmatrix} 0 & p'_+ + eA_- \\ p'_+ + eA_+ & 0 \end{pmatrix},
\]

where \( A_- = \zeta_x A_x - \zeta_y A_y, A_+ = \zeta_x A_x + \zeta_y A_y. \) For simplicity, it is assumed that \( A_x = E) = 0 \) and then \( A_- = A_+ = \zeta_x A_x = \zeta_y A_y = \delta k_{x,y} \). The current excited by the external field \( \hat{A}(t) \) is given by

\[
\vec{j} = \frac{e}{4 \pi \hbar^2} \int \langle \psi(p_x, p_y) | \hat{A}(t) | \psi(p_x, p_y) \rangle \mathrm{d}p_x \mathrm{d}p_y.
\]
take a one-sided limit to get the modified slope,

$$\lim_{k \to K^-} \frac{\partial E_{1,-1}(k, 0)}{\partial k_x} = \mp \frac{\sqrt{3}}{2} \Delta t_6 a = \mp \hbar v_F \Delta. \quad (14)$$

This slope in Eq. (14) is hinted at by the slope of the green line tangent to the cone at $K$ in the inset of Fig. 3. The effective Hamiltonian at $K$ becomes $H_K = v_F^{(0)} \begin{pmatrix} p_x & p_y \\ 0 & 0 \end{pmatrix}$ with $v_F = v_F \Delta$ and the contribution to the conductivity from the cone at $K$ reads

$$\sigma_{3,K}^M = \Delta^2 \sigma_3^D, \quad (15)$$

which is reduced into the case that $\xi_y = \xi_x = \Delta$ in Eq. (13).

For typical RSOC parameters, $\Delta < 1$ and the height of the cones are much smaller than the other two energy branches $E_{1,1,-1} \ll E_{1,1,1}$. $E_{1,1,1} - E_{1,-1,1} = \delta E$ fall into the terahertz regime. For photon energy lower than the height of the cones, the third-order nonlinear conductivity with RSOC is expressed as

$$\sigma_{3}^{\text{RSOC}} = 6 \left( \frac{3 \xi_1^2}{\xi_y} + \Delta^2 \right) \sigma_3^D. \quad (16)$$

Before we go on to analyze the feature of Eq. (16), investigating the conductivity numerically is useful to shed light on the physical mechanism of the nonlinear conductivity with RSOC. To this end we ignore the effect of the deformation for the moment and approximate the slope of the line that connects the vault and the Dirac points as the effective slope of the cones, as shown (dashed line) in the inset of Fig. 3. Denoting the angle between the dashed lines and the $k_x$ axis as $\beta_0(K)$, $\beta_1(K, \lambda)$ respectively and noting that the dimensionless slope of the standard Dirac cone ($E = v_F \hbar k$) is $\frac{\sqrt{3}}{2}$, the third-order nonlinear conductivity is numerically obtained:

$$\sigma_{3, \text{Num}}^{\text{RSOC}} = 8(\tan^2 \beta_0 + 3 \tan^2 \beta_1) \sigma_3^D. \quad (17)$$

The calculated nonlinear conductivity is shown in Fig. 3 (red solid line). It is easily found that the nonlinear conductivity with ROI is obviously enhanced for sufficiently large $\Delta$. Moreover, the enhancement is independent of frequency, which is the manifestation of topological change of the energy band from the RSOC. It is mainly due to the increase of the effective Fermi velocity (the slope) of the cones with the enhancement of coupling strength $\Delta$. It is notable that there is a restriction that the photon energy must be always lower than the cone height. The strong enhancement shown above is protected by the topology of the band structure in the presence of RSOC. In other words, it is protected by the trigonal warping at low energy. For each fixed $\Delta$ there exists a critical value of energy $E_c(\Delta)$ at which the equienergy contour has three self-intersection points shown with a red line in Fig. 4 and this energy value $E_c$ is defined as the height of the Dirac cone. When the energy of the incoming photon is larger than $E_c$ the equienergy contour contributing to the conductivity by the resonance transition becomes a simple closed curve shown in Fig. 4 (black line) which is not topologically equivalent to the one that our model holds for where the equienergy contour is composed of three discrete closed curves shown with a blue line in Fig. 4.

The comparison of nonlinear conductivity from Eq. (17) (red line) and from Eq. (16) (blue line) is shown in Fig. 5. It can be found that the results with the different methods are consistent with each other qualitatively. Moreover, for the fixed $\Delta$ the conductivity given by Eq. (16) is larger than that given by (17) where the effect of the deformation is ignored. It also indicates that the effect of the deformation of the cone is important for the enhancement of the conductivity.

To determine the value of $E_c$ we chose the derivative of the curve $\frac{a(E_{1,-1}k_{\|})}{\partial k_{\|}} = 0$ for simplicity due to the symmetry. The numerical solution to the relevant transcendental equation is shown (blue dashed line) in Fig. 3 and it follows from the curve that $E_c$ decreases monotonously with the decay of $\Delta$. When the parameter $\Delta$ changes continuously there are two classes of topologically inequivalent equienergy contours (See Fig. 4) and our model only holds for one of the two classes.
(blue contours). Therefore, the energy of the incoming photon ($h\omega_{in}$) and $\Delta$ are restricted because of the validity of our model and the constraint can be written explicitly as $h\omega_{in} < E_c(\Delta)$.

III. CONCLUSIONS

In summary we have shown that the third-order nonlinear conductivity of graphene can be distinctively enhanced in the presence of RSOC in the regime where the frequency is below the cone height defined by the spin-orbit interaction, $h\omega_{in} < E_c(\Delta)$. The enhancement originates from topologically inequivalent change of the energy bands around the original Dirac point and the shape changing of the derivative Dirac cones. The shape changing of the cones leads to a rapid increase of the density of states at low energies. While the topological change of the band structure results in a six times enhancement of the linear conductivity, it has a much stronger effect on the nonlinear conductivity. The result presented here can be readily applied to the nonlinear conductivity of a bilayer graphene.

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