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Title of the Thesis

Nonlinear Electrical Transport in Dirac and Semi-Dirac Systems

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Abstract

In this thesis, we investigate some properties of nonlinear electrical transport in Dirac and Semi-Dirac systems.

In the first part, we used incident fields to derive the non-linear conductivity for a general material. More specifically, we examined the conductivity for one incident field and two incident fields with both different intensity and frequency. More recently, graphene has attracted the attention of many researchers around the world because of its superior properties. In more detail, we used a recursive method (the expansion of the Fermi-Dirac distribution up to higher orders) to find both the linear and nonlinear conductivity results for one incident field. Our result suggest that both the single frequency and frequency tripled nonlinear conductivity become different to the linear conductivity under a moderate electric field strength of $10^4$ V/cm in both their real and imaginary components. Next, we used the same method of one incident field for two incident fields in the calculation of conductivity, then we examined the transmission properties in the thin film. The results showed that the transmission of graphene is significantly altered depending on elements of electric fields and frequencies.

In the second part, we investigated the nonlinear optical response in Semi-Dirac System. The semi-Dirac material has confirmed its existence in a $TiO_2/VO_2$ multilayered structure. By using a semi-classical approach where the quantum mechanically calculated energy dispersion is used to derive the velocity in the presence of an applied field of $10^3 - 10^4 V/cm$ along the x or y direction. This unique system can find out many physical properties of bilayer graphene. The optical current is found in both Cartesian coordinates, following both the longitudinal and transverse current. It was found that the ratio of the third order non-linear response to the linear optical response is significantly dependent on both temperature and chemical potential. In more detail, in the terahertz regime, the third order response is comparable to the linear response under an applied field of $10^3 - 10^4 V/cm$. Furthermore, the non-linear response in linear direction persists at room temperature along the linear
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Chapter 1

Introduction

As far as we knew, carbon is a key factor of life and is the prime element of millions of organic chemical compounds. In a carbon atom, electrons at the outer layer can form many different types of hybridizations, so when these atoms are linked together, they also can create many forms of crystal structures, such as three-dimensional (3D), two-dimensional (2D), one-dimensional (1D) and non-dimensional (0D). This is reflected in the richness of literature about the forms of carbon materials, such as diamond, graphite, graphene, carbon nanotube and fullerenes.

The focus of material technology from 2000 to 2009 revolved around researches into two new states of carbon, as carbon nanotubes and graphene. Since it was discovered and researched in the early 1990s, scientists have been identifying that carbon nanotubes could have many practical uses. The contribution of carbon nanotubes to today's industries is plentiful, from electronics, micro-motors, memory, batteries, and in the cosmic field. But new graphene studies published in early 2009 confirmed that the new material quickly attracted the attention of scientists, being the thinnest known material at the time whilst boasting more stiffness than diamond. In addition, the electrical conductivity of graphene is ideal. One of the major causes for the different properties of graphene is the energy gap between conduction band and valence band. Dirac material is a class of condensed matter systems which can be effectively described by the Dirac equation and this equation was formulated for fermions that present within Dirac materials. Prominent examples of Dirac matter are Graphene, topological insulators, Dirac semimetals, Weyl semimetals, etc. These systems are formulated following a specific choice of the Dirac mass, the Dirac velocity, the Dirac matrices and the space-time curvature. So that these matter systems lead to a common feature with respect to the density of states, the heat capacity and impurity scattering. The optical conductivity of Dirac materials has received a huge amount of research over the years. More specifically, graphene has still been the subject of greatest interest of so many researchers and it has held an enormous
amount of scientific papers. Novoselov et al who discovered experimentally single layer graphene (2D) in 2004, unfolded a series of studies on the electric properties of graphene [1]. Fig. 1.1 is some pictures about graphene:

![Fig 1.1. (a) Graphene honeycomb lattice showing in different colors the two triangular sublattices. Also shown is the graphene Brillouin zone in momentum space. Adapted from Castro Neto et al., 2009. (b) Carbon nanotube as a rolled graphene layer. Adapted from Lee, Sharma et al., 2008. (c) Lattice structure of graphite, graphene multilayer. Adapted from Castro Neto et al., 2006. (d) Lattice structure of bilayer graphene. γ₀ and γ₁ are, respectively, the intralayer and interlayer hopping parameters t, t₁ used in the text. The interlayer hopping parameters γ₃ and γ₄ are much smaller than γ₁ = t₁ and are normally neglected. Adapted from Mucha-Kruczynski et al., 2010. Typical configuration for gated graphene [1].](image)

In recent years, two-dimensional materials such as graphene, transition-metal dichalcogenides (TMDs), and phosphorene have attracted interest for nonlinear optical applications among which graphene has shown the strongest optical nonlinearity [2-10]. More specifically, in July 2018, Tao Jiang et al had a report about an experimental study of third-harmonic generation (THG) and four-wave mixing (FWM) in graphene using gate tuning to adjust the doping level and vary the resonant condition. This contribution paves the way towards future design of graphene-based nonlinear optoelectronic devices. As we knew, Graphene is considered a record-performance nonlinear-optical material on the basis of numerous experiments [11]. Also in July 2018, Nathalie Vermeulen et al presented a
paradigm shift in the understanding of 2D-material nonlinearities and this way enables their full exploitation in next-generation nonlinear-optical devices [12]. Consequently, scientists hope that by 2020, graphene can replace silicon semiconductors. Currently, graphene has opened hope for the electronics industry. Furthermore, Terahertz (THz) technology also has many interesting applications in obtaining molecular spectral and material information [13 - 18].

1.1. The linear properties of Dirac material - graphene

Dirac materials have many linear and nonlinear properties. Graphene is known as a Dirac material in the carbon family. For many years recently, graphene has attracted many researchers by its electronic properties, such as electronic band structure, the quantum Hall effect, the minimum conductivity, cyclotron mass $m_C$ of electrons [19-22] and the strong suppression of weak localization [23-26], etc.

1.1.1. Electronic band structure of graphene

Graphene is made up of carbon atoms and arranged in a hexagonal structure on the same plane, also known as a honeycomb structure. Inside, each carbon atom linked to the three nearest carbon atoms by the linkage $\sigma$ that is formed by the overlap of the s-p hybrid lines, corresponding to the $sp^2$ hybridization state. The distance between the nearest carbon atoms is $a = 0.142$ nm. According to Pauli’s principle, the energy levels in the $\sigma$ link have been filled, so the $sp^2$ hybridization paths will characterize the degree of durability in the flat structure of the graphene membrane. The remaining p orbital of the carbon atoms is perpendicular to the planar structure of the membrane and are wrapped side by side to form the $\pi$ bond, and the energy level of the bond have not been filled yet, they are called non-localized orbitals and have an important role in formation up electrical properties of graphene.
Fig 1.2. (a) Bravais lattice of the graphene; (b) $\sigma$ and $\pi$ bonds in graphene; (c) graphene $\pi$ and $\pi^*$ band structure [27].

Following up Fig 1.2, two sublattices A and B form the unit cell. The crystal structure of graphene can be described by the lattice vectors $\vec{a}_1$ and $\vec{a}_2$ of these sublattices.

$$\vec{a}_1 = \left( \frac{3a}{2}, \frac{a\sqrt{3}}{2} \right)$$

$$\vec{a}_2 = \left( \frac{3a}{2}, \frac{a\sqrt{3}}{2} \right)$$

with $a$ is the carbon-carbon distance.

Fig 1.3. Crystal structure of single layer graphene.
At the same time, from Fig 1.3, the position between carbon atoms in lattices A and B is related to each other through vectors \( \vec{\delta}_1 \left( \frac{a}{2\sqrt{3}}, \frac{a}{2} \right), \vec{\delta}_2 \left( \frac{a}{2\sqrt{3}}, -\frac{a}{2} \right), \vec{\delta}_3 \left( -\frac{a}{\sqrt{3}}, 0 \right) \), with \( d_{c-c} = 1.42 \) Å the carbon-carbon distance in Benzene cycle, then \( a = |\vec{a}_1| = |\vec{a}_2| = d_{c-c}\sqrt{3} = 2.46 \) Å.

Next, in order to determine the energy region structure of graphene and related materials, the strong linkage approximation is often used by scientists as it is simple but a particularly effective method.

In the strong link approximation \([28]\), the energy dispersion \( E(k) \) is determined by equation

\[
\text{det} \left[ H - ES \right] = 0
\]  

(1.1)

where \( H \) is transmission matrix and \( S \) is the overlaying matrix.

The full wave function that describes the state of graphene can be written as:

\[
\psi(\vec{k}, \vec{r}) = C_A \psi_A(\vec{k}, \vec{r}) + C_B \psi_B(\vec{k}, \vec{r})
\]  

(1.2)

where the orbital wave functions are

\[
\psi_A(\vec{k}, \vec{r}) = \frac{1}{\sqrt{N}} \sum_{\delta_A} e^{i \vec{k} \cdot \vec{\delta}_A} \phi(\vec{r} - \vec{\delta}_A)
\]  

and

\[
\psi_B(\vec{k}, \vec{r}) = \frac{1}{\sqrt{N}} \sum_{\delta_B} e^{i \vec{k} \cdot \vec{\delta}_B} \phi(\vec{r} - \vec{\delta}_B)
\]  

(1.3)

Where \( N \) is total unit cells of lattice graphene, \( \vec{\delta}_\alpha \) is position vector of atoms, \( \phi(\vec{r} - \vec{\delta}_\alpha) \) is wave function that describes the state of carbon atoms in sublattices A and B (with \( \alpha = A, B \)).

The energy spectrum is determined by solving the Schrodinger equation with respect to the diagonal \( 2 \times 2 \) matrix as \( \begin{pmatrix} H_{AA} - E & H_{AB} \\ H_{BA} & H_{BB} - E \end{pmatrix} \), where \( H_{AA}, H_{BB}, H_{AB} \) are Hamiltonians of interaction between carbon atoms within the lattice A or B and between the atoms of these two lattices.

\[
H_{AA}(\vec{r}) = \frac{1}{N} \sum_{\delta, \delta'} e^{i \vec{k} \cdot (\vec{\delta} - \vec{\delta}')} \langle \varphi_A(\vec{r} - \vec{\delta}) | H | \varphi_A(\vec{r} - \vec{\delta}') \rangle
\]  

(1.4)

\[
H_{AB}(\vec{r}) = \frac{1}{N} \sum_{\delta, \delta'} e^{i \vec{k} \cdot (\vec{\delta} - \vec{\delta}')} \langle \varphi_A(\vec{r} - \vec{\delta}) | H | \varphi_B(\vec{r} - \vec{\delta}') \rangle
\]  

(1.5)
In addition, $H_{AA} = H_{BB} = E_{2p}$ with $E_{2p}$ is energy at ground state of p orbital (involved in creating $\pi$ link), and $H_{AB} = t(e^{i\vec{k} \delta_1} + e^{i\vec{k} \delta_2} + e^{i\vec{k} \delta_3}) = tf(k)$ (t is the energy required for electrons to move between adjacent atoms).

In Cartesian coordinates, we can write

$$f(k) = e^{ik_xa/\sqrt{3}} + 2e^{-ik_xa/2\sqrt{3}}\cos\left(\frac{k_ya}{2}\right)$$

(1.6)

Because $f(k)$ is complex function, $H_{AB}$ is Hermitian operator. It leads to $H_{AB} = H_{BA}^*$

$$H = \begin{pmatrix} E_{2p} & tf(k) \\ tf^*(k) & E_{2p} \end{pmatrix}$$

(1.7)

The overlaying matrices have $S_{AA} = S = 1, S_{AB} = sf(k) = S_{BA}^*$

$$S = \begin{pmatrix} 1 & sf(k) \\ sf^*(k) & 1 \end{pmatrix}$$

(1.8)

Substituting (1.7), (1.8) into the equation (1.1), we have

$$E(k) = \lambda t \sqrt{1 + 4\cos^2\left(\frac{\sqrt{3}k_xa}{2}\right) \cos\left(\frac{k_ya}{2}\right) + 4\cos^2\left(\frac{k_ya}{2}\right)}$$

(1.9)

with $\lambda = \pm 1$

As mentioned above, one of the most remarkable properties of graphene is that charges of this material do not follow the usual dispersion pattern, since energy is proportional to square of wave vector k as in conventional metals and semiconductors. Instead, at the neighbouring points K and K', the dispersion pattern is of the form:

$$E = \lambda \hbar k v_F$$

(1.10)

with $v_F = \frac{\sqrt{3}ta}{2} \approx \frac{10^6 m}{s}$ is the Fermi velocity.

The greatest dependence of energy to wave vector k can be described by the Dirac equation

$$H \nabla \psi(r) = E \psi(r)$$

(1.11)

Where $H = \hbar v_F \begin{pmatrix} 0 & k_- \\ k_+ & 0 \end{pmatrix} = \hbar v_F \sigma \vec{k}$ is Hamiltonian Dirac

$$k_\pm = k_x \pm ik_y$$
\[ \sigma = (\sigma_x, \sigma_y) \] is the Pauli matrix

Thus, since the electrons in graphene follow the Dirac equation, they can be viewed as Dirac fermions and satisfy the properties of these particle.

### 1.1.2. The minimum conductivity

Because of the special structure of the crystal lattice, graphene is a gapless semiconductor. The type of carrier in the lattice is determined by the location of the Fermi energy level and can be changed with negative or positive voltage, so there is a strong dependence between the conductivity and the supply voltage.

![Fig 1.4. The measured conductivity of graphene as a function of gate voltage \( v_g \) [29]](image)

In Fig 1.4, conductivity is minimal when \( E_F = 0 \) and lies between the conduction band and the valence band. When the voltage varies from -100 V to 100 V, the Fermi level moves from the valence band, to the centre point with the voltage equal to 0 and to the conducting region, leading to a change in the particle density and conductivity, as well as the type of electrical charges. The conductance decreases when the voltage changes from -100V to 0V, reaching the minimum and increasing again when the voltage increases from 0 to +100V. Dirac points (\( E_F = 0 \)) can be considered as having charge neutrality. Theoretically, the charge neutralization at this location results in the conductivity being suppressed (reduced to 0).
However, \( \sigma \) in fact reaches minimum with defined values \( \sigma_{\text{min-exp}} = \frac{4e^2}{h} \), in theory
\( \sigma_{\text{min-theo}} = \frac{4e^2}{\pi h} \). Currently, this problem has not been clearly explained and is being studied.

Furthermore, graphene is virtually transparent, absorbing only 2.3% of the light intensity, independent of wavelength in the optical region.

### 1.1.3. Cyclotron mass \( m_{\text{C}} \) of electrons of graphene

One of the most remarkable properties of graphene is non-observance of common dispersion formula of charge carriers in this material (the energy is proportional to the square of the wave vector \( k \) as in metal or common semiconductor). Substituting into there, at nearby location of the points \( \text{K and K}' \), the dispersion formula: \( E = \hbar k v_F \) (\( v_F \approx 10^6 \text{m/s} \) is Fermi velocity). Effective mass of electrons can be written as:

\[
m_{\text{C}} = \hbar^2 \left( \frac{\partial^2 E}{\partial k^2} \right)^{-1}
\]

At Dirac points \( E = \hbar^2 v_F \), effective mass of charge carriers in graphene is eliminated.

By the definition of effective mass (Ashcroft and Mermin cite), the formula is obtained as:

\[
m_{\text{C}} = \frac{1}{2\pi} \left[ \frac{\partial A(E)}{\partial E} \right]_{E=E_F}
\]

Where \( A \) is area of Fermi circle \( A(E) = \pi k^2 = \pi \frac{E^2}{E_F^2} \Rightarrow m_{\text{C}} = \frac{k_F}{v_F} \)

Moreover, charge density \( n \) and \( k_F \) are related by \( k_F^2 \pi = n \Rightarrow m_{\text{C}} = \frac{v_F^2}{v_F^2} \sqrt{n} \).

The electrons and holes in graphene follow the Dirac equation and have similar manifestations as Fermi particles, having an effective mass proportional to the square root of the charge density. This property has been verified by Geim's experiment cite [19].
Fig 1.5. Cyclotron mass $m_c$ of electrons and holes as a function of their concentration. Symbols are experimental data, solid curves the best fit to theory [19].

Fig 1.5 shows that between theory and experiment we have a complete fit in investigation about the relation between effective mass and charge density. This indicates that there is the existence of massless Dirac fermions within graphene.

1.1.4. The quantum Hall effect in graphene

The Quantum Hall Effect experiment in graphene was first performed in 2005 cite [19], this showed a significant difference about the quantum Hall effect between graphene and other two-dimensional materials.

The Hall conductivity can be written as:

$$\sigma_{xy} = \left(N + \frac{1}{2}\right)\frac{4e^2}{h} = \nu\frac{4e^2}{h}$$

With N is characteristic for the Landau levels.

This means that the decline of the Hall conductivity has the form of steps with equal gaps, and this change takes the semi-integer values of $\frac{4e^2}{h}$ instead of the integer values, and this effect does not change when the basis particles changes from electron to hole as Fig (1.6).
Fig 1.6. Hall conductivity $\sigma_{xy}$ and longitudinal resistivity $\rho_{xx}$ of graphene as a function of their concentration at $B = 14\, T$ and $T = 4K$, $\sigma_{xy} = (4e^2/h)v$ is calculated from the measured dependences of $\rho_{xy}(V_g)$ and $\rho_{xx}(V_g)$ as $\sigma_{xy} = \rho_{xy}/(\rho_{xx}^2 + \rho_{xy}^2)$. The behaviour of $1/\rho_{xy}$ is similar but exhibits a discontinuity at $V_g = 0$, which is avoided by plotting $\sigma_{xy}$. Inset: $\sigma_{xy}$ in ‘two-layer graphene’ where the quantization sequence is normal and occurs at integer v. The latter shows that the half-integer QHE is exclusive to ‘ideal’ graphene [19].

Expression of energy dependence $E_N$ into magnetic field B and Landau level N is shown as

$$E = v_F \sqrt{2e\hbar B \left( N + \frac{1}{2} \pm \frac{1}{2} \right)} \tag{1.15}$$

Where $v_F = 10^6m/s$ is Fermi velocity, the sign $\pm$ describes two spins.

To summarise, the results show that between the quantum Hall effect in graphene and the quantum Hall effect in other materials have the clear difference. Once again, it is shown that there is specialness in the structure of graphene energy dispersion, the appearance of the Dirac points and the charged particles are essentially identical to the Dirac fermions.
1.2. The nonlinear properties of Dirac materials

More recently, many reports both theoretically and experimentally showed that graphene is a gapless semiconductor that has the strong optical response \([30-33]\). On the 17\textsuperscript{th} of August 2009, the group of A. R. Wright et al published that within the model of massless Dirac fermions, graphene has a strong nonlinear conductivity in the terahertz regime \([32]\).

![Fig 1.7. The frequency dependent nonlinear conductivity. Both the single frequency and frequency tripling terms decrease as \(\omega^{-4}\), and with increasing temperature \([32]\).](image)

In this article, the group of authors investigated the nonlinear conductivity dependence on the frequency and the results are shown in Fig 1.7. They found out that the nonlinear effects enhanced dramatically the optical activity of mono-layer graphene in the terahertz to far infrared regime under an electric field of around \(10^3\) V/cm. Furthermore, nonlinear contribution also alters both the single frequency and frequency tripled optical response at experimentally relevant field strengths. According to the plot of the authors, we show that both nonlinear terms have a rapid decrease with frequency, and nonlinear conductivities at room temperature are lower than at zero temperature at low frequencies. In addition, when we consider them at weak fields and high frequencies, the linear current dominates and the two nonlinear terms are approximately equal in magnitude. More specifically, the frequency tripled consideration of nonlinear current opens the gateway to photonic and optoelectronic
device applications [32]. Moreover, graphene is virtually transparent, absorbing only 2.3% of the light intensity, this weak absorption can consequently be overcome by the nonlinear optical absorption.

One of simple formalism that determines the nonlinear optical properties of Dirac materials such as the intraband optical response is semiclassical electron transport theory. Here, the electron dynamics are decomposed into terms higher order in the external electric field [34, 35].

We consider a system that is described by Hamiltonian \( H(p) \).

The velocity operator is given by \( \hat{v} = \frac{\partial \hat{H}}{\partial \hat{p}} \) and the corresponding expectation value [36] can be written as \( v(p) = \frac{\partial \varepsilon(p)}{\partial p} \)

Where \( p \) is the electron momentum and the energy eigen value is given by \( \varepsilon(p) \).

In the presence of a time-dependent applied electric field in the form of

\[
E = E_0 e^{i\omega t}
\]

(1.16)

Where \( E_0 \), and \( \omega \) are the amplitude and frequency of the electric field. Ignoring the weak magnetic component, the external field is minimally coupled to the quasi-particle by performing the substitution \( p \to p - eA \), \( v(p) \to v(p - eA) \) where \( e \) is the electric charge, the vector potential \( A \) is related to the external field by \( E = -\frac{\partial A}{\partial t} \). The externally perturbed quasiparticles acquire linear and nonlinear velocities which can be separated by performing a Taylor expansion with respect to the external field:

\[
v(p, E) = \sum_{i=0} v^{(i)}(p, E)
\]

(1.17)

where \( v^{(n)}(p, E) \) represents velocity component proportional to \( E_0^n \).

The optical current response can be explicitly written as

\[
f^{(i)} = e \int v^{(i)}(p, E)f(\varepsilon)d\varepsilon
\]

(1.18)

With \( f(\varepsilon) \) is the Fermi-Dirac distribution function.

When the external electric field is strong, the equilibrium Fermi-Dirac distribution function is no longer accurate since the electron energy becomes \( \varepsilon' = \varepsilon + \Delta \varepsilon \), where \( \Delta \varepsilon \) are terms
dependent on the external electric field. In this case, we can perform a similar expansion around $\Delta \varepsilon$ as:

$$f(\varepsilon + \Delta \varepsilon) = f(\varepsilon) + \sum_{i=0}^{\infty} (\Delta \varepsilon)^i \frac{\partial f^{(i)}(\varepsilon)}{\partial \varepsilon}$$  \hspace{1cm} (1.19)

When we substitute Eq. (1.19) into Eq. (1.18), the optical current density can be found.

In addition, another formalism that also can obtain the interband optical response, is n-photon electron coupling wavefunction.

We start with solving the Schrödinger equation (1.20) to find out wavevector.

$$i\hbar \frac{\partial \psi(p)}{\partial t} = \mathcal{H} \psi(p)$$  \hspace{1cm} (1.20)

At here, we can consider a system in an external electric field that is described by Hamiltonian as:

$$\mathcal{H}(p) \rightarrow \mathcal{H}(p - eA)$$  \hspace{1cm} (1.21)

And the wavefunction can be written by

$$\Psi(p) = \sum_{n=0} \phi_n e^{i(n\omega - \varepsilon/h)t}$$  \hspace{1cm} (1.22)

where $\phi_n$ is a $\propto E^n$ (by this selecting, we can investigate the $n^{th}$ order nonlinear optical response).

In the coupling of n-photon to the electron, $\phi_n$ will show strength of that coupling and we can find it through wavevector.

Next, we can write the total optical current density as:

$$J_{\nu}^{(n)} = e \int dp \Psi^+ \vec{v} \Psi = e \int dp (\phi_0^+ + \phi_1^+ + \phi_2^+ + \cdots) \vec{v} \Psi (\phi_0 + \phi_1 + \phi_2 + \cdots)$$  \hspace{1cm} (1.23)

Where $\vec{v} = \frac{\partial \mathcal{H}}{\partial p_{\nu}}$ is the $\nu$-component velocity operator, with $\nu = (x, y)$.

In order to find the $n^{th}$ order non-linear optical current density, we can use method that select out terms $\propto E^n$. 


1.3. Introduction about the Semi-Dirac system

For a normal semiconductor, the electrons follow a parabolic dispersion due to their massive nature. Graphene is a stark contrast to this, with the electrons following a linear dispersion, indicating massless Dirac fermions [27]. This difference between a linear and a parabolic dispersion leads to drastically different properties between the two systems [1]. This difference can be seen for example when comparing the properties of single and bilayer graphene, with the latter having a parabolic dispersion [37]. Recently it was discovered that a system can exist that has an anisotropic dispersion relation, that varies smoothly between a parabolic dispersion along principal axis and linear along the other axis. This is a known as a semi-Dirac material, and has been shown to exist in a TiO$_2$/VO$_2$ multilayered structure [38, 39]. This was published by Victor Pardo et al. In this article, they considered the multilayer system TiO$_2$ and It is formed by mTiO$_2$ monolayers and nVO$_2$ monolayers grown along the rutile (001) direction. Here, group of authors used five layers of TiO$_2$ (or approximately 1.5 nm thickness) and analysed one by one different n values from 1 to 5, from insulator for small n to a metal for n of 5 or greater.

Fig 1.8. Band structure of the (TiO$_2$)$_5$/(VO$_2$)$_n$ multilayer system for n from 2 (left) to 5 (right). This shows how the transition to a metallic from an insulating state occurs through the appearance of a semi-Dirac point for n=3 and 4. The band structure is represented on the $k_x$ and $k_y$ plane, enough for the two-dimensional symmetry of the system. Different colors when two bands cross means they have different symmetry [39].
Fig 1.9. Band structure of the (TiO$_2$)$_5$/(VO$_2$)$_4$ multilayer system in a two-dimensional grid in the $k_x - k_y$ plane. The surfaces reveal a point Fermi surface, and also the semi-Dirac character of the dispersion: on the left-hand side the linear dispersion along the zone diagonal can be seen, on the right-hand side the parabolic dispersion along the perpendicular direction. The Fermi level is set at zero energy [39].

In band structure of the (TiO$_2$)$_5$/(VO$_2$)$_4$ multilayer system at Fig 1.8, we can see a representation $\varepsilon(k_x,k_y)$ of the two bands crossing the Fermi level that has the parabolic energy dispersion in the perpendicular direction and the linear dispersion along the zone diagonal as Fig 1.9 [39].

Such a dispersion can also occur when an anisotropy is introduced in the hopping parameters for a graphene monolayer. This causes pairs of Dirac cones to merge in to a single point with a semi-Dirac-like dispersion [40]. The dynamical polarisation and plasmonic properties of this system have been studied previously [41]. The semi-Dirac dispersion is also predicted theoretically to exist in an oxidized silicene layer [42]. Due to the newness of this material property, there have been only a few recent investigations into the properties of this unique system. Banerjee et. al. [43] give details on the number of physical properties such as Faraday rotation, magnetic susceptibility and heat capacity. Anderson localisation has been investigated in semi-Dirac systems [44], and the possible symmetries that allow for a semi-Dirac dispersion have also been determined [45]. Detailing as below, Banerjee et al showed that the Hamiltonian for the semi-Dirac system [46] can be written by tight-binding model as:
Where \( q = (q_x, q_y) \) is the momentum, \( m \) is the effective mass, and the Fermi velocity is \( v_F = 3t a/(2\hbar) \approx 10^6 m/s \), \( t \approx 3 eV \) is the nearest neighbour hopping bandwidth, \( a \approx 0.142 nm \) is carbon-carbon distance [47].

From experiment or from ab-initio calculations, we can find the effective mass and Fermi velocity, but they will not depend on the particular values simply appear as a scaling factor. From here, the values such as the Fermi energy, frequency, and relaxation rate also will all be given in terms of a scaling constant \( \epsilon_0 \) and it is defined as \( \epsilon_0 = 2mv_F^2 \).

By making the substitution \( \epsilon_x = \frac{p_x}{2mv_F} \) and \( \epsilon_y = \frac{p_y}{2mv_F} \), the Hamiltonian becomes:

\[
H = \epsilon_0 \begin{pmatrix}
0 & \frac{\epsilon_x^2}{\epsilon_x^2 + \epsilon_y^2} + ivF_\epsilon_y \\
\frac{\epsilon_x^2}{\epsilon_x^2 + \epsilon_y^2} - ivF_\epsilon_y & 0
\end{pmatrix}
\]

From Schrödinger equation:

\[
H \psi = \epsilon_\lambda \psi
\]

Where \( \psi = \begin{pmatrix} \alpha \\ \beta \end{pmatrix} \) is the wavevector.

We get the energy dispersion as

\[
\epsilon_\lambda = \pm \epsilon_0 \sqrt{\epsilon_x^2 + \epsilon_y^2} = 2m\lambda v_F^2 \sqrt{\frac{p_x^4}{(2mv_F)^4} + \frac{p_y^2}{(2mv_F)^2}} = \lambda v_F \sqrt{\frac{p_x^4}{(2mv_F)^4} + p_y^2}
\]

(1.27)
Fig 1.10. 3D structure of the energy dispersion in Semi-Dirac system

Fig 1.10 is 3D structure of the energy dispersion in Semi-Dirac system. It is result that we get to equation (1.27).

Some researchers previously found a similar dispersion when a pair of Dirac points in the electronic spectrum of a two-dimensional crystal may merge into a single one with the spectrum $\varepsilon = \pm \sqrt{(\Delta + \frac{q_x^2}{2m^*})^2 + c^2 q_y^2}$ (where $\Delta = 0$), following up Fig 1.11 [37].

Fig 1.11. Evolution of the spectrum when the quantity $\Delta$ is varied and changes in sign at the topological transition (arbitrary units). The low-energy spectrum stays linear in the $q_y$ direction. [37]
Where we will use the transformation \( \varepsilon_x^2 = \varepsilon \cos(\theta) \) and \( \varepsilon_y = \varepsilon \sin(\theta) \) with \( \lambda = \pm 1 \). This energy dispersion results in a symmetric upper (\( \lambda = +1 \)) and lower (\( \lambda = -1 \)) semi-Dirac cones, representing electrons and hole states respectively, and is analogy to the charge conjugation symmetry in quantum electrodynamics.

The energy dispersion will become

\[
\varepsilon_{\lambda} = \lambda \varepsilon_0 \varepsilon
\]

And the wave vector is found to be

\[
\psi = \frac{1}{\sqrt{2}} \left( \frac{1}{\varepsilon_0 (\varepsilon_x^2 - i \varepsilon_y)} \right) = \frac{1}{\sqrt{2}} \left( \frac{1}{\varepsilon_x - i \varepsilon_0 \varepsilon} \right)
\]

The velocity operator is defined as

\[
\overline{v}_\mu = \frac{1}{\rho} \frac{\partial \mathcal{H}}{\partial \varepsilon_\mu}
\]

Where \( \rho = 2 m v_F \) and \( \mu = (x, y) \)

This proves that a Semi-Dirac system can exist both a parabolic dispersion along principal axis and linear along the other axis.

1.4. Outline of this thesis

In this Thesis, we study the nonlinear electrical transport in Dirac and Semi-Dirac systems and is organised as follows:

- In the first chapter, we have shown a brief theoretical overview about the linear and nonlinear properties of Dirac system. In more detail with the linear properties of Dirac material, we present some electrical - electronic properties of graphene, such as electronic band structure, the quantum Hall effect, the minimum conductivity, Cyclotron mass \( m_C \) of electrons, etc. Next, in order to determine the nonlinear optical properties of Dirac materials such as the intraband optical response, we present some formalisms that can obtain the optical current density from semiclassical electron transport theory and an n-photon electron coupling wavefunction. Furthermore, in this chapter, we also introduce about the structure of Semi-Dirac systems.
In chapter 2, we provide some mathematical theory about linear response such as the Boltzmann transport equation (BTE), apply the Boltzmann equation for relaxation time approximation, the Kubo formula for the optical conductivity, and frequency-dependent conductivity of Graphene in clean limit.

Chapter 3 deals with the optical response of graphene in the presence of one field and two fields with different intensity and different frequency. In this chapter, we derive the general expression for the non-linear conductivity up to the 3rd order, applying the formalism specifically to graphene. Next, we find a general expression for transmission through a graphene with two fields. The result shows that the pumping field can significantly modify the conductivity and transmission of graphene.

In chapter 4, we investigate the nonlinear conductivity in an intrinsic semi-Dirac system. We will use a semi-classical approach to expand the velocity operator in a strong driving field. The third order current as function of temperature and chemical potentials will be analysed.

Finally, chapter 5 is a summary of this thesis.
Chapter 2

Linear response

2.1. The Boltzmann transport equation and relaxation time approximation

2.1.1. The Boltzmann transport equation (BTE)

In 1872, Ludwig Boltzmann devised the Boltzmann transport equation or the Boltzmann equation (BTE) [48] that describes the statistical behaviour of a thermodynamic system not in a state of equilibrium cite.

In ideal crystals, when they are not affected by the external factors, the wave function $\psi_k(\vec{r})$ will not be change over time. At the same time, the electronic distribution function $f_0(\vec{r}, \vec{K})$ over status also does not change over time and that is Fermi-Dirac distribution function

$$f_0(\vec{r}, \vec{K}) = \frac{1}{1 + \exp\left(\frac{E - EF_kB_T}{k_BT}\right)} \tag{2.1}$$

If we put in a crystal potential field $V(\vec{r})$, the status of each electronic in Brillouin zone also will change according to the following equations:

$$\frac{d\vec{p}}{dt} = -\nabla V = \vec{F}_a \tag{2.2}$$

$$\frac{d\vec{K}}{dt} = \frac{1}{\hbar} \vec{F}_a \tag{2.3}$$

Where $F_a$ is the force of the non-periodic field.

The change in momentum $\vec{p}$ after a period of time $t$ is called the momentum of force $\Delta\vec{p}$:

$$\Delta\vec{p} = \vec{p}(t) - \vec{p}_0 = \int_0^t \vec{F}_a \cdot dt = \hbar\Delta\vec{K} \tag{2.4}$$

The status of electrons at $t=0$ is determined by the distribution function $f_0(\vec{r}, \vec{K}_0) = f_0(\vec{r}, \vec{p}_0/\hbar)$, at time $t$ the electronic distribution is determined by the other distribution function as:
\[ f(\hat{r}, \vec{K}, t) = f(\hat{r}, \vec{K}_0) + \frac{1}{\hbar} \int_0^t \vec{F}_a dt \]  

(2.5)

We write the full derivative of the distribution function over time as:

\[ \frac{df(\hat{r}, \vec{K}, t)}{dt} = \frac{\partial f}{\partial t} + \frac{\partial f}{\partial \hat{r}} \frac{\partial \hat{r}}{\partial t} + \frac{\partial f}{\partial \vec{K}} \frac{\partial \vec{K}}{\partial t} = \frac{\partial f}{\partial t} + (\nabla_r f \vec{\nabla}) + \frac{1}{\hbar} \left( \nabla_k f \vec{F}_a \right) \]  

(2.6)

Due to the statistical theory of preserving the status number so it requires \( \frac{df(\hat{r}, \vec{K}, t)}{dt} = 0 \).

From (2.6):

\[ -\frac{\partial f}{\partial t} = (\nabla_r f \vec{\nabla}) + \left( \nabla_k f \frac{\vec{F}_a}{\hbar} \right) \]  

(2.7)

Equation (2.7) showed that the change of distribution function over time at each point in phase space \((\hat{r}, \vec{K})\) is due to the motion of electrons in the ordinary space \(\hat{r}\) (change of positions) and in the space of the wave vector \(\vec{K}\) (change of velocity due to force). The non-periodic field caused force \(\vec{F}_a\) (such as the electric field, the magnetic field caused the force F, and even the fields deviate from the periodic field of crystal as the missing node, the atom and ion impurities, the heat oscillations of the lattice caused the force \(F_d\)).

This means, the force \(\vec{F}_a\) can be written as:

\[ \vec{F}_a = \hat{F} + \vec{F}_d \]  

(2.8)

Equation (2.7) is then:

\[ -\frac{\partial f}{\partial t} = (\nabla_r f \vec{\nabla}) + \left( \nabla_k f \frac{\hat{F}}{\hbar} \right) + \left( \nabla_k f \frac{\vec{F}_d}{\hbar} \right) \]  

(2.9)

The change in distribution function is due to force \(F_d\), called internal force and it also is the reason that leads to many difficulties in determining the distribution functions by dynamical laws. Therefore, it can only be determined by statistical rules. That change is called scattering integral and is denoted by \(\left( \frac{\partial f}{\partial t} \right)_{scatt}\), and is given by:

\[ \left( \frac{\partial f}{\partial t} \right)_{sc} = \left( \nabla_k f \frac{\vec{F}_d}{\hbar} \right) \]  

(2.10)

We can substitute \(\frac{\hat{F}}{\hbar} = \vec{K}\) to equation (2.9):
\[
- \frac{\partial f}{\partial t} = (\nabla_r f \hat{v}) + \left( \hat{K} \nabla k f \right) + \left( \frac{\partial f}{\partial t} \right)_{\text{scatt}} 
\]  
\hspace{1cm} (2.11)

Or
\[
- \frac{\partial f}{\partial t} = v \frac{\partial f}{\partial r} + F_{\text{ext}} \frac{\partial f}{\partial p} + \left( \frac{\partial f}{\partial t} \right)_{\text{scatt}} 
\]  
\hspace{1cm} (2.12)

This is the Boltzmann transport equation in general form.

Mathematically, the stationary state is represented by the condition \( \frac{\partial f}{\partial t} = 0 \).

Therefore, the Boltzmann equation at the stationary state is shown:
\[
\nabla_r f \hat{v} + \hat{K} \nabla k f = - \left( \frac{\partial f}{\partial t} \right)_{\text{scatt}} 
\]  
\hspace{1cm} (2.13)

We need to mind some conditions that are applied for Boltzmann transport equation, such as:

Firstly, the external field must not alter the energy spectrum of the electron in the crystal, meaning that the external field must be small enough.

Secondly, the Boltzmann transport equation is a semi-classical equation, so we cannot use it to consider processes that occurs over a very short period of time and in a volume that is too small, as this will lead to uncertainty of energy and momentum.

Finally, the Boltzmann transport equation can be applied to any free-homogeneous particle system, so it is not only applicable to free-electron particle systems in metals, but also applies to electron particle system and hole particle system in semiconductors.

**2.1.2. The Boltzmann equation for relaxation time approximation**

Solving Boltzmann transport equation in general form is a very complex problem, but in cases when we give the concept about relaxation time, that problem will be solved much easier.

We consider that the electronic system is in excited status and at time \( t=0 \) we turn off excited field, the electronic system will gradually return to equilibrium. The process of returning to a status of equilibrium is called recovery or relaxation. The equation describing the relaxation process is
\[
\frac{\partial f}{\partial t} = - \left( \frac{\partial f}{\partial t} \right)_{\text{scatt}} 
\]  
\hspace{1cm} (2.14)

The simplest assumption about the occurrence of relaxation process is: the equilibrium status recovery speed is proportional to the deviation from the equilibrium position so we can write:
\[ \frac{\partial f}{\partial t} = -\left( \frac{\partial f}{\partial t} \right)_{\text{scatt}} = -\frac{1}{\tau} \left( f(\vec{r}, \vec{R}, t) - f_0(\vec{r}, \vec{R}) \right) \] (2.15)

Where \(1/\tau\) is scaling factor. Solve this equation, we have:

\[ f(\vec{r}, \vec{R}, t) = f_0(\vec{r}, \vec{R}) = \left[ f(\vec{r}, \vec{R}, 0) - f_0(\vec{r}, \vec{R}) \right] e^{-t/\tau(\vec{R})} \] (2.16)

Unit of \(\tau(\vec{K})\) is time and characteristic of relaxation process is relaxation time. Relaxation time \(\tau(\vec{K})\) allows the approximate performance of scattering integral in the Boltzmann transport equation.

At the stationary state, the Boltzmann equation in relaxation time approximation is

\[ \nabla_{\vec{r}} f \vec{\nu} + \vec{K} \nabla_k f = -\frac{f - f_0}{\tau(\vec{K})} \] (2.17)

Where \(f_0\) is distribution function in equilibrium condition, meaning is the Fermi-Dirac distribution.

Moreover, we can also express (2.17) as:

\[ f - f_0 = -\tau \left\{ \nu \frac{\partial f}{\partial r} + F_{\text{ext}} \frac{\partial f}{\partial p} \right\} \] (2.18)

If the external field is electric and magnetic field \((F_{\text{ext}} = e(E + [\nu \times H])\) Lorentz Force), the Boltzmann transport equation is now written:

\[ f - f_0 = -\tau \left\{ \nu \frac{\partial f}{\partial r} + e(E + [\nu \times H]) \frac{\partial f}{\partial p} \right\} \] (2.19)

In short, the meaning of relaxation time is that it represents the time in which the particle system returns to equilibrium.

### 2.2. The Kubo formula for the optical conductivity

The Kubo formula is an equation which expresses the linear response of an observable quantity due to a time-dependent perturbation. Among the numerous applications of linear response formula, one can mention charge and spin susceptibilities of, for instance, electron systems due to external electric or magnetic fields.

We consider a material responds to an applied electric field. The incident electric field has the form
\[ E(r, t) = E_0 e^{iqr - i\omega t} \] (2.20)

We are able to calculate linear response the conductivity by using

\[ J_\alpha(r, t) = \sigma_{\alpha\beta}(q, \omega)E_\beta(r, t) \] (2.21)

To find the current we will need to use

\[ j = -\frac{\delta H}{\delta A} \] (2.22)

Where a minimally coupled Hamiltonian can be written as

\[ H = \int d^d r \left[ \frac{1}{2m} \psi^+(x)(p - eA)^2\psi(x) - e\phi(x)\phi^+(x)\psi(x) \right] + H_{\text{int}} \] (2.23)

It is convenient when we work in the gauge \( \nabla \cdot A = 0 \) and in units \( c = 1 \)

The current is found

\[ j = -\frac{ie\hbar}{2m} (\psi^+(x)\nabla\psi(x) - \psi(x)\nabla\psi^+(x)) - \frac{e^2}{m} A\psi^+(x)\psi(x) \] (2.24)

The first term is the paramagnetic current \( j_p \) and the second term is the diamagnetic current \( j_d \).

Value of the current operator can be written as:

\[ J(r, t) = \langle j_p(r, t) \rangle - \frac{ne^2}{m} A(r, t) \] (2.25)

where \( n \) is the electron density.

\[ \langle j_{p\alpha}(r, t) \rangle = i \int_{t' < t} d^d r ' dt ' \langle [j_{p\alpha}(r, t), j_{p\beta}(r', t')] \rangle \] (2.26)

We knew that \( E = -\frac{\partial A}{\partial t} \). Therefore, formula for the conductivity is found as:

\[ \sigma_{\alpha\beta}(q, \omega) = \frac{ine^2}{m(\omega + i\delta)^2} \delta_{\alpha\beta} + \frac{1}{(\omega + i\delta)^2} \int_0^\infty dt e^{i(\omega + i\delta)t} \langle [j_\alpha(-q, t), j_\beta(q, 0)] \rangle \] (2.27)

We use terms of the retarded current-current correlation function:

\[ \Pi_{\alpha\beta}(q, \omega) = -i \int dt \theta(t - t') e^{i\omega(t - t')} \langle [j_\alpha(-q, t), j_\beta(q, t')] \rangle \] (2.28)

From (2.27):
\[
\sigma_{\alpha\beta}(q, \omega) = \frac{i}{\omega + i0^+} \left[ \Pi_{\alpha\beta}(q, \omega) + \frac{ne^2}{m} \delta_{\alpha\beta} \right] \tag{2.29}
\]

The advantage of this form is that we can use the well-established Green’s function formalism for computing the current-current correlation function. We are also only usually interested in the real part of the conductivity because it is linked to optical absorption. The imaginary part can easily be obtained from the real part using a Kramers - Kronig relation. The real part is written as

\[
\text{Re} \left[ \sigma_{\alpha\beta}(q, \omega) \right] = -\frac{1}{\omega} \text{Im} \left[ \Pi_{\alpha\beta}(q, \omega) \right] \tag{2.30}
\]

Considering at the one loop level of the current-current correlation function

\[
\Pi_{\alpha\beta}(q, \omega) = e^2 T \sum_{k,\nu_n} \left[ v_\alpha(k) G(k, \nu_n) v_\beta(k + q) G(k + q, \omega + \nu_n) \right] \tag{2.31}
\]

We use the Green’s function that is given by

\[
G(p, z) = \int \frac{d\omega}{2\pi} A(p, \omega) \frac{\delta(z - \omega)}{z - \omega} \tag{2.32}
\]

The advantage of this form is treating the problem as sustainable particles. There are many Kubo formulas in the literature that involve matrix elements of current operators divided by energy denominators. The form in terms of spectral functions for non-interacting particles is represented by

\[
A(k, \omega) = 2\pi \delta(\omega - \epsilon_k) \tag{2.33}
\]

If there are interactions described by a self-energy \( \Sigma(k, \omega) \), the form in terms of spectral functions is given by

\[
A(k, \omega) = \frac{1}{\pi} \frac{|\text{Im} \Sigma(k, \omega)|}{(\omega - \text{Re} \Sigma(k, \omega) - \epsilon_k)^2 + |\text{Im} \Sigma(k, \omega)|^2} \tag{2.34}
\]

More details on the Kubo formula are found in books by Mahan [49] or Altland and Simons [50].

### 2.3. Frequency-dependent conductivity of Graphene in clean limit

Graphene is a layer of carbon that forms a hexagonal grid has the honeycomb structure, with a carbon-carbon spacing of 0.142 nm. It is the first real two-dimensional crystalline material
and representative of a completely new family of 2D materials [51-53]. Moreover, graphene has extraordinary electron mobility that made it a promising material in the fields of nanoscale electronics and photonics. The movement of electrons in graphene is very fast, the electron does not seem to have mass and moves near the speed of light. Electrons in Graphene have speeds up to 100 times that of electrons in silicon. Electron motions do not follow the Schodinger equation but instead the Dirac equation for particles with no mass such as neutrinos. This particle is full of the properties of Dirac particles that can be called ghost particles because of its singularity. One of those properties is that Dirac particles in some cases may move backward against the direction of the electric field or the effect of force. Furthermore, the optical absorption of graphene depends on the field polarization direction. So many scientists such as Saito et al (2004) [54], Oyama et al (2006) [55] and more have chosen to study the orientation of optical absorption in graphene as the basis for some applications of this material in the electronics industry and some other industries.

Not long ago, in the article was published by Chao Zhang et al in 2008 [56], orientation response in graphene was mentioned with high frequencies.

In more detail, they studied that orientation of the optical conductivity in graphene dependents on frequency of photons that polarize along the armchair direction or along the zigzag direction.

![Polarization direction in graphene](image)

**Fig 2.1.** Polarization direction in graphene, the x axis is the armchair direction and the y axis is the zigzag direction [56].
In the honeycomb crystal structure of graphene, two sublattices A and B formed the unit cell. As shown Fig. 2.1, considering link of atoms in graphene, the x axis is along the line connecting atoms of alternating sublattice and is called the armchair direction, the y axis is along the line connecting atoms of the same sublattice and is called the zigzag direction.

From the quantum Hamiltonian of graphene \( H = \sum_{s} s \epsilon_{k,s} a_{k,s}^{\dagger} a_{k,s} + \left( \frac{1}{c} \right) A J \), where \( A = e^{-i\omega t} \), \( E = E_0 e^{-i\omega t} \), \( \epsilon_{k,s} = s \epsilon_0 \sqrt{3 + \alpha(k)} \), the components of the current density operator \((j_x, j_y)\) can be found as

\[
j_x = -2e v_F \sum_{k,s} \frac{\sin \sqrt{3} k_x \cos \frac{1}{2} k_y a}{\sqrt{3 + \alpha(k)}} a_{k,s}^{\dagger} a_{k,s} + i2e v_F \sum_{k,s} s \frac{1 + \cos \sqrt{3} k_x \cos \frac{1}{2} k_y a + \cos k_y a}{\sqrt{3 + \alpha(k)}} a_{k,s}^{\dagger} a_{k_{-s}}
\]

(2.35a)

\[
j_y = -\frac{2ev_F}{\sqrt{3}} \sum_{k,s} \frac{\cos \sqrt{3} k_x \sin \frac{1}{2} k_y a + \sin k_y a}{\sqrt{3 + \alpha(k)}} a_{k,s}^{\dagger} a_{k,s} - i \frac{2ev_F}{\sqrt{3}} \sum_{k,s} s \frac{\sin \sqrt{3} k_x \sin \frac{1}{2} k_y a}{\sqrt{3 + \alpha(k)}} a_{k,s}^{\dagger} a_{k_{-s}}
\]

(2.35b)

Here, one can easily obtain the optical conductivity from the Kubo formula

\[
\sigma_{\mu\nu}(\omega) = \frac{1}{\omega} \int_0^{\infty} e^{i\omega t} \langle j_\mu(t) j_\nu(0) \rangle dt
\]

(2.36)

Next, they substituted the components of the current density operator \( J \) that were obtained at (2.35a), (2.35b) into (2.36).

The results of the optical conductivity in the along x and y directions were found as

\[
\sigma_{xx} = \frac{e^2 v_F^2}{\pi^2 \omega} \int d \mathbf{k} \left( \frac{1 + \cos \sqrt{3} k_x \cos \frac{1}{2} k_y a + \cos k_y a}{3 + \alpha(k)} \right)^2 \left( \frac{f_{k,+} - f_{k,-}}{\hbar \omega + 2 \epsilon_0 \sqrt{3 + \alpha(k)}} - \frac{f_{k,+} - f_{k,-}}{\hbar \omega - 2 \epsilon_0 \sqrt{3 + \alpha(k)}} \right)
\]

(2.37a)

\[
\sigma_{yy} = \frac{e^2 v_F^2}{3 \pi^2 \omega} \int d \mathbf{k} \sin \frac{\sqrt{3}}{2} k_x \sin \frac{1}{2} k_y a \left( f_{k,+} - f_{k,-} \right) \left( \frac{1}{\hbar \omega + 2 \epsilon_0 \sqrt{3 + \alpha(k)}} - \frac{1}{\hbar \omega - 2 \epsilon_0 \sqrt{3 + \alpha(k)}} \right)
\]

(2.37b)

With \( \omega = \omega + i0_+ \), \( \alpha(k) = -2 + 4 \cos \frac{\sqrt{3}}{2} k_x \cos \frac{1}{2} k_y a + 4 \cos \frac{1}{2} k_y a \) and \( \epsilon_0 = |t| v_F \), \( v_F \) is the Fermi velocity.

Where \( f_{k,s} \) is Dirac-Fermi distribution function, \( \sigma_{xy} = \sigma_{yx} \) because of symmetry
In order to make a clear statement for polarization of the photons in the directions x and y, Chao Zhang et al have given some visual results as

**Fig 2.2.** (c): The real part of the optical conductivity $\sigma_{yy}$ depends on frequency at various temperatures for the Fermi energy lies on the K point. Inset: the imaginary part of the optical conductivity $\sigma_{yy}$ depends on frequency. (d): Same as (c) but the Fermi energy lies in the conduction band [56].
Following up Fig 2.2 and Fig 2.3, we can see that the real part of the optical conductivity $\sigma_{yy}$ depends on frequency and temperature with two different chemical potentials. In particularly, for a graphene with $\mu = 0$, the photon absorption starts from $\omega = 0$ at low temperature, this is consistent with the metallic absorption characteristics. But for a graphene with $\mu \neq 0$ and higher temperature, the absorption is almost negligible.

The results of article showed that the absorption of photons polarizing along the armchair direction or along the zigzag direction dependent on frequencies $\omega \sim |t|$. When the frequency is in the range of $2|t| < \omega < 6|t|$, this polarization occurs more clearly. This statement also was not violated, as recent experiments have shown that graphene is almost transparent at low frequencies under $0.5|t|$ [57, 58].

**Fig 2.3.** (b): The real part of the optical conductivity $\sigma_{yy}$ depends on chemical potential at various frequencies. (d): Same as (b) but the real part of the optical conductivity $\sigma_{yy}$ depends on temperature [56].
Chapter 3

Nonlinear conductivity in Dirac System

3.1. Introduction

There are many previous researchers, such as the group of Mikhailov et al, who used the semiclassical Boltzmann transport equation to find out the nonlinear intraband optical response of gapless graphene [30, 31]. However, those researches have a limitation that the nonlinear response usually occurs under a strong external field. But in 2014, researchers at the Lawrence Berkeley National Laboratory and the University of California at Berkeley said that they could use a pump-and-probe technique to measure the ultrafast electrons in the material respond to this light, as Fig. 3.1 [59].

Fig 3.1. Optical pump THz probe spectroscopy of Graphene with (a) the experiment on a graphene FET device, (b) transmitted THz waveforms [59].
From the Drude expression for conductivity, to the Boltzmann transport equation and Kubo formalism, most existing research is limited to linear regime under a single external field. On the other hand, commonly used technique of pump-to-probe experiments involves two fields. If the pump field is much stronger than the probing field, the response of the system to the pump field can be strongly nonlinear.

Recently, some pump-probe experiment was mentioned, typically as, in 2014, Martin Wagner et al published an article about "Ultrafast and Nanoscale Plasmonic Phenomena in Exfoliated Graphene Revealed by Infrared Pump-Probe Nanoscopy" that introduced in Fig. 3.2.

**Fig 3.2.** Pump–probe spectroscopy is central for exploring ultrafast dynamics of fundamental excitations, collective modes, and energy transfer processes. Typically carried out using conventional diffraction-limited optics, pump–probe experiments inherently average over local chemical, compositional, and electronic inhomogeneities. Here, we circumvent this deficiency and introduce pump–probe infrared spectroscopy with ∼20 nm spatial resolution, far below the diffraction limit, which is accomplished using a scattering scanning near-field optical microscope (s-SNOM). This technique allows us to investigate exfoliated graphene single-layers on SiO2 at technologically significant mid-infrared (MIR) frequencies where the local optical conductivity becomes experimentally accessible through the excitation of surface plasmons via the s-SNOM tip. Optical pumping at near-infrared (NIR) frequencies prompts distinct changes in the plasmonic behavior on 200 fs time scales. The origin of the pump-induced, enhanced plasmonic response is identified as an increase in the effective electron
temperature up to several thousand Kelvin, as deduced directly from the Drude weight associated with the plasmonic resonances [60].

Furthermore, pump–probe microscopy that is an emerging tool for functional imaging of nonfluorescent chromophores and nanomaterials. It was researched by Pu-Ting Dong and Ji-Xin Cheng in 2017. The major processes in a pump–probe experiment is shown clearly in Fig. 3.3.

**Fig 3.3.** Three major processes in a pump–probe experiment: (a) Excited state absorption, (b) stimulated emission, and (c) ground-state depletion. For ground-state depletion, the number of the molecules in the ground state is decreased upon photoexcitation, consequently increasing the transmission of the probe pulse. For stimulated emission, photons in its excited state can be stimulated down to the ground state by an incident light field, thus leading to an increase of transmitted light intensity on the detector. In the case of excited-state absorption, the probe photons are absorbed by the excited molecules, promoting them to the higher energy levels [61].

Graphene’s strong nonlinear response due to a linear energy dispersion indicates that it could be a great platform upon which to observe many theoretically predicted high energy phenomena.

Here, we use the Boltzmann transport equation and combined with the expansion of the Fermi-Dirac distribution up to higher order [62] to find a nonlinear conductivity [63] in both
cases one incident field and two incident fields of different intensity and different frequency are considered.

3.2. Conductivity of single layer Graphene in using one incident field

For the intraband conductivity we can calculate the linear result using a standard method being the Kubo formula as in previous literature [64-66] but here we use a recursive method that is the Boltzmann transport equation (BTE) to find both the linear and nonlinear conductivities. When applying this method calculates the nonlinear conductivities, we need to expand the Fermi-Dirac function to higher orders in the field.

Firstly, we begin by considering the Boltzmann transport equation in the relaxation time approximation

\[
\frac{\partial f}{\partial t} + \frac{\partial f}{\partial \mathbf{p}} \mathbf{E} e = - \frac{f-f_0}{\tau}
\]  

(3.1)

Where \( \mathbf{p} = m \mathbf{v} \), \( f_0 \) is the equilibrium Fermi-Dirac distribution, \( f = \sum_{m=0}^{n} f_m(t) \) is the non-equilibrium Fermi-Dirac distribution with \( f_m(t) \) is the component of the Fermi-Dirac distribution with total field order of \( m \), and \( \tau \) is the relaxation rate.

The electric field along the x-axis

\[
\mathbf{E} = \frac{E_0}{2} \hat{x} (e^{i\omega t} + e^{-i\omega t})
\]

From component \( \frac{\partial f}{\partial \mathbf{p}} e \mathbf{E} \) of equation (3.1), we can see that there is a relation between frequency of \( E \) and frequency of \( f \). Therefore, we can expand function \( f \) such that components of equation (3.1) that have the maximum oscillating frequency is \( n \omega \).

\[
f = \sum_{m=0}^{n} f_m(t) = \sum_{m=0}^{n} \sum_{k=-m}^{m} f_m^{k_m \omega} e^{k_i \omega t}
\]

(3.2)

With \( f_m^{(m-2l-1)} = 0 \) for \( 0 \leq l \leq \frac{m+1}{2} \)

We find that we can write the Boltzmann transport equation for the \( m^{th} \) order Fermi-Dirac distribution is:
\[
\frac{\partial f_m}{\partial t} + \frac{f_m}{\tau} = \frac{eE_0}{2} \left( e^{i\omega t} + e^{-i\omega t} \right) \frac{\partial f_{m-1}}{\partial p_x}
\]  
(3.3)

The process of recursion is completed by working up from \( m=1 \) to \( 3 \). By following this process, we find

\[
f_{1}^{\pm \omega} = \frac{eE_{0}}{(\tau^{-1} \pm i\omega)} \frac{\partial f_{0}^{0}}{\partial p_x}
\]  
(3.4a)

\[
f_{2}^{\pm 2\omega} = \frac{(eE_{0})^{2}}{(\tau^{-1} \pm i\omega)(\tau^{-1} \pm 2i\omega)} \frac{\partial^{2}f_{0}}{\partial p_x^{2}}
\]  
(3.4b)

\[
f_{2}^{0} = \frac{2(eE_{0})^{2}}{(\tau^{-2} + 2\omega^{2})} \frac{\partial^{2}f_{0}}{\partial p_x^{2}}
\]  
(3.4c)

\[
f_{3}^{\pm 3\omega} = \frac{eE_{0}}{(\tau^{-1} \pm 3i\omega)} \frac{\partial f_{2}^{\pm 2\omega}}{\partial p_x} = \frac{(eE_{0})^{3}}{(\tau^{-1} \pm 3i\omega)(\tau^{-1} \pm 2i\omega)(\tau^{-1} \pm 3i\omega)} \frac{\partial^{3}f_{0}}{\partial p_x^{3}}
\]  
(3.4d)

\[
f_{3}^{\pm \omega} = \frac{eE_{0}}{(\tau^{-1} \pm i\omega)} \left( \frac{\partial f_{2}^{\pm 2\omega}}{\partial p_x} + \frac{\partial f_{0}^{0}}{\partial p_x} \right) = \frac{(eE_{0})^{3}}{(\tau^{-1} \pm 2i\omega)(\tau^{-2} + 2\omega^{2})} \frac{\partial^{3}f_{0}}{\partial p_x^{3}}
\]  
(3.4e)

For the convenience of calculation, we make the substitutions \( \epsilon = v_{F}p \) (denote \( \epsilon_x = v_{F}p_x \)), \( \phi = \hbar v_{F} = 6.582119 \times 10^{-8} eV^2 cm/V \) and specify \( E_0 \) in units of V/cm. In addition, we will also alter \( \xi = \hbar \omega \) and \( \gamma = \hbar \tau^{-1} \) (\( \tau \) is the relaxation rate of the system). Here we will take \( \gamma = 0.0026 \), a relaxation rate used previously for conductivity in a graphene system [62].

The terms of the 1\(^{st}\) and 3\(^{th}\) order expansion for \( f \) can rewrite as

\[
f_{1}^{\pm \omega} = \phi \frac{E_{0}}{2} \frac{1}{(y \pm i\xi)} \frac{\partial f_{0}^{0}}{\partial \epsilon_x}
\]  
(3.5a)

\[
f_{3}^{\pm \omega} = \phi \left( \frac{E_{0}}{2} \right)^{3} \frac{3}{(y \pm 2i\xi)(y^{2} + \xi^{2})} \frac{\partial^{3}f_{0}}{\partial \epsilon_x^{3}}
\]  
(3.5b)

\[
f_{3}^{\pm 3\omega} = \phi \left( \frac{E_{0}}{2} \right)^{3} \frac{1}{(y \pm i\xi)(y \pm 2i\xi)(y \pm 3i\xi)} \frac{\partial^{3}f_{0}}{\partial \epsilon_x^{3}}
\]  
(3.5c)

Next, we can calculate the nonlinear conductivity by using

\[
J_{n} = \sigma_{n}E^{n} = -\frac{e v_{F}}{\pi^{2}h^{2}} \int f_{n} p d\epsilon d\cos \theta d\theta = -\frac{e}{v_{F} \pi^{2}h^{2}} \int f_{n} \epsilon d\epsilon d\cos \theta d\theta
\]  
(3.6)

We can substitute the \( n \)\(^{th}\) expansion of the Fermi-Dirac function \( f_{n} \) by component functions for the upper (+) and lower (-) bands \( f_{n}^{\pm} \) as

\[
f_{n} = f_{n}^{+} - f_{n}^{-}
\]
We can use the Newtonian formula if $E^n$ is high level

$$E^n = \left( \frac{E_0}{2} \right)^n \sum_{k=0}^{n} \frac{n!}{k!(n-k)!} e^{i(2k-n)\omega t}$$  \hspace{1cm} (3.7)

Next, we can calculate the 1\textsuperscript{st} and 3\textsuperscript{rd} order conductivities as

$$J_1 = -\frac{e}{v_F \pi^2 \hbar^2} \int f_1 e\varepsilon \cos \theta d\theta = \sigma_1 E^1$$

$$J_3 = -\frac{e}{v_F \pi^2 \hbar^2} \int f_3 e\varepsilon \cos \theta d\theta = \sigma_3 E^3$$

Furthermore, we can change the Cartesian coordinates $(x, y)$ to cylindrical coordinates $(r, \theta)$ by

$$\frac{\partial u}{\partial x} = \cos \theta, \quad \frac{\partial u}{\partial r} = \frac{1}{r} \sin \theta \frac{\partial u}{\partial \theta}$$

$$\frac{\partial f_0}{\partial \varepsilon_x} = \cos \theta \frac{\partial f_0}{\partial \varepsilon}$$

$$\frac{\partial^3 f_0}{\partial \varepsilon_x^3} = \cos^3 \theta \frac{\partial^3 f_0}{\partial \varepsilon^3} + \frac{3 \cos \theta \sin^2 \theta}{\varepsilon} \frac{\partial^2 f_0}{\partial \varepsilon^2} - \frac{3 \cos \theta \sin^2 \theta}{\varepsilon^2} \frac{\partial f_0}{\partial \varepsilon}$$

In order to simplify the expression slightly, we can introduce $\sigma_0 = \frac{e^2}{4\hbar}$.

The results are found:

$$\sigma_1(\xi) = -\frac{4\sigma_0}{\pi} \frac{1}{\gamma-i\xi} I_1$$  \hspace{1cm} (3.8a)

$$\sigma_3(\xi) = -\frac{4\sigma_0}{\pi} \phi^2 \frac{1}{(\gamma^2+\xi^2)(\gamma-2i\xi)} I_3$$  \hspace{1cm} (3.8b)

$$\sigma_3(3\xi) = -\frac{4\sigma_0}{\pi} \phi^2 \frac{1}{(\gamma-i\xi)(\gamma-2i\xi)(\gamma-3i\xi)} I_3$$  \hspace{1cm} (3.8c)

Where $I_1 = \int_0^\infty e\varepsilon \frac{\partial f_0}{\partial \varepsilon}$ and $I_3 = -\frac{3}{4} \int_0^\infty \frac{1}{\varepsilon} e\varepsilon \frac{\partial f_0}{\partial \varepsilon}$

The value at Zero temperature of $I_3$ is

$$I_1(T = 0) = -E_F$$  \hspace{1cm} (3.9a)

$$I_3(T = 0) = \frac{3}{4} \frac{1}{E_F}$$  \hspace{1cm} (3.9b)
Fig 3.4. Linear intraband conductivity depends on frequency $\omega$, with $E_F = 0.1eV$, $T=0K$.

![Graph showing linear intraband conductivity vs frequency](image)

Fig 3.5. Linear intraband conductivity depends on Fermi energy at frequency $\omega=1THz$, with $T=0K$.

![Graph showing linear intraband conductivity vs Fermi energy](image)

In Fig 3.4, we can show that at low frequency, both real and imaginary parts of the intraband conductivity have quite large differences and especially the peaks but they tend to decrease gradually with $\omega$ over 2 THz. However, when we investigate about Fermi energy dependence of the linear intraband conductivity (in Fig 3.5), both components increase gradually. Next, we can see clearly, effect of temperature to the linear conductivity. We consider ratio of linear
intraband conductivity at finite temperature to zero temperature case
\[
\frac{\sigma_1(T)}{\sigma_1(0)} = \frac{I_1(T)}{I_1(0)}
\]
that does not depend on frequency.

**Fig 3.6.** Ratio of linear intraband conductivity at finite temperature to linear conductivity depends on Fermi energy at zero temperature case with \(\omega=1\text{THz}\).

**Fig 3.7.** Dependence of ratio of linear intraband conductivity on temperature with \(\omega=1\text{THz}\), \(E_F = 0.1\text{eV}\).
From Fig 3.6, the ratio decreases as the Fermi energy increases, it approaches gradually to 1 with $E_F \gtrsim 0.05 \text{ eV}$. Furthermore, we can see clearly that, in Fig 3.7, this ratio has very weak temperature dependence. In order to make a stronger effect, we can investigate with lower values of the Fermi energy. However, with Fermi energies over 0.1 eV, the effects of temperature are not significant, so we can examine to other properties of graphene.

![Graph 3.8](image1.png)

**Fig 3.8.** *Frequency dependence of the third-order nonlinear intraband conductivity, with $E_F = 0.1\text{eV}, T=0K$.***

![Graph 3.9](image2.png)

**Fig 3.9.** *Ratio of 3rd order to 1st order conductivities depends on frequency $\omega$, with $E_F = 0.1\text{eV}, T=0K$.***
Fig 3.10. Total nonlinear intraband conductivity at zero temperature depends on frequency $\omega$, with $E_F = 0.1eV$.

From Fig 3.4, Fig 3.8, Fig 3.9 and Fig 3.10 the result shows that both the real and imaginary components of the 3rd order conductivity is negative but they are positive with the 1st order conductivity with the same frequency $\omega$. Moreover, we can see that the linear conductivity has magnitudes bigger than the nonlinear conductivity. This demonstrates that in one incident field, the linear conductivity has more advantage to the nonlinear conductivity.

Fig 3.11. Nonlinear intraband conductivity depends on frequency $3\omega$, with $E_F = 0.1eV$, $T=0K$. 
In Fig 3.11, we can see that the 3rd order conductivity with frequency $3\omega$ has a different change to both the linear and nonlinear conductivity with frequency $\omega$. In a little more detail, when we consider the 3rd order conductivity with frequency $3\omega$, both real and imaginary parts exist a frequency at which the conductivity is 0. Furthermore, the real component can be found to be positive for $\xi > \frac{\gamma}{\sqrt{11}}$, and the imaginary component can be found to be positive for $\xi > \gamma$.

Fig 3.12. *Ratio of nonlinear intraband conductivity at finite temperature to nonlinear conductivity at zero temperature depends on Fermi energy for a number of temperatures, with $w = 1\, THz$.*

Fig 3.13. *Dependence of ratio of nonlinear intraband conductivity at finite temperature to nonlinear conductivity at zero temperature on temperature, with $\omega = 1\, THz$.  


In Fig 3.12 and Fig 3.13, we investigate \( \frac{\sigma_3(T)}{\sigma_3(0)} = \frac{I_3(T)}{I_3(0)} \). The result shows that this ratio changes rapidly at low Fermi energy period. Value of Fermi energy corresponding to a peak, also will grows with temperature. To compare two ratios of the linear and nonlinear conductivity, we see in Fig 3.6, 3.7 for the linear and Figs 3.12, 3.13 for the nonlinear, they have a stronger effect with lower values of the Fermi energy and have ratio increasing sharply with high temperature at low frequency.

### 3.3. Transmission of thin films in using two incident fields

This part, we used the same method as with one incident field. This means, we extend the Fermi-Dirac distribution and apply it into the Boltzmann transport equation (BTE). As a result, we can find the current densities and transmissions in Dirac materials with the two applied fields.

We begin by considering the Boltzmann transport equation (BTE)

\[
\frac{\partial f}{\partial t} - \frac{\partial f}{\partial p} E \cdot e = - \frac{f - f_0}{\tau} \tag{3.10}
\]

Where \( f = \sum_{m=0}^{n} f_m(t) \) with \( f_m \) is the component of the Fermi-Dirac distribution with total field order of \( m \).

The total electric field is written as

\[
E = E_1(e^{i\omega_1t} + e^{-i\omega_1t}) + E_2(e^{i\omega_2t} + e^{-i\omega_2t}) \tag{3.11}
\]

Here, we considered the solution that has a rotationally symmetric system, so we can align the electric field \( E_1 \) along the x-axis as: \( E_1 = E_1 \hat{x} = E_1 \hat{r}_x \) and \( E_2 = E_2(cos \phi \hat{x} + sin \phi \hat{y}) = E_2 \hat{r}_2 \)

\[
(3.12)
\]

The electric fields are determined to be parallel to some unit vectors \( \hat{r}_j \). Therefore, we can write an expression as \( \frac{\partial}{\partial p_j} = \hat{r}_j \frac{\partial}{\partial p} \).

Next, we substitute the time-dependent electric field in expression (3.11) into the Boltzmann transport equation (BTE) (3.10) for the \( m^{th} \) order Fermi-Dirac distribution as:

\[
\frac{\partial f_m}{\partial t} + \frac{f_m}{\tau} = \frac{\partial f_{m-1}}{\partial p_1} e E_1(e^{i\omega_1t} + e^{-i\omega_1t}) + \frac{\partial f_{m-1}}{\partial p_2} e E_2(e^{i\omega_2t} + e^{-i\omega_2t}) \tag{3.13}
\]
In order to solve Eq. (3.12), we can write an expansion for the component of the Fermi-Dirac distribution with total field order of $m$, $f_m$, as

$$f_m(t) = \sum_{l,k=-m}^{m} f_{m}^{l \omega_1, k \omega_2} e^{-i(l \omega_1 + k \omega_2)t}$$

(3.14)

The process of recursion is completed by working up from $m=1$ to $3$

For $m=1$

$$f_1 = f_1^{\omega_1} e^{i \omega_1 t} + f_1^{-\omega_1} e^{-i \omega_1 t} + f_1^{\omega_2} e^{i \omega_2 t} + f_1^{-\omega_2} e^{-i \omega_2 t}$$

(3.15)

The component of the Fermi-Dirac distribution with total field order of 1st can be obtained by inserting Eq. (3.15) into (BTE).

The four solutions are:

$$f_1^{\pm \omega_1} = \frac{e E_1}{(\tau^{-1} \pm i \omega_1)} \frac{\partial f_0}{\partial p_1}$$  \hspace{1cm} (3.16a)

$$f_1^{\pm \omega_2} = \frac{e E_2}{(\tau^{-1} \pm i \omega_2)} \frac{\partial f_0}{\partial p_2}$$  \hspace{1cm} (3.16b)

For $m=2$

$$f_2 = f_2^{2 \omega_1} e^{2i \omega_1 t} + f_2^{-2 \omega_1} e^{-2i \omega_1 t} + f_2^{(\omega_1 + \omega_2)} e^{i(\omega_1 + \omega_2) t} + f_2^{-(\omega_1 + \omega_2)} e^{-i(\omega_1 + \omega_2) t} + f_2^{\omega_2} e^{i \omega_2 t} + f_2^{-\omega_2} e^{-i \omega_2 t} + f_2^{(\omega_1 - \omega_2)} e^{i(\omega_1 - \omega_2) t} + f_2^{-(\omega_1 - \omega_2)} e^{-i(\omega_1 - \omega_2) t} + 2 f_2^0$$

(3.17)

The component of the Fermi-Dirac distribution with total field order of 2nd can be obtained by inserting Eq. (3.16) into (BTE).

The 9 allowed states for $m=2$ are found to be:

$$f_2^0 = \frac{(eE_1)^2}{\tau^{-2} + \omega_1^2} \frac{\partial^2 f_0}{\partial p_1^2} + \frac{(eE_2)^2}{\tau^{-2} + \omega_2^2} \frac{\partial^2 f_0}{\partial p_2^2}$$  \hspace{1cm} (3.18a)

$$f_2^{\pm 2 \omega_1} = \frac{(eE_1)^2}{(\tau^{-1} \pm i \omega_1)(\tau^{-1} \pm 2i \omega_1)} \frac{\partial^2 f_0}{\partial p_1^2}$$  \hspace{1cm} (3.18b)

$$f_2^{\pm 2 \omega_2} = \frac{(eE_2)^2}{(\tau^{-1} \pm i \omega_2)(\tau^{-1} \pm 2i \omega_2)} \frac{\partial^2 f_0}{\partial p_2^2}$$  \hspace{1cm} (3.18c)

$$f_2^{\pm (\omega_1 + \omega_2)} = \frac{(eE_1)(eE_2)}{\tau^{-1} \pm i(\omega_1 + \omega_2)} \left( \frac{1}{(\tau^{-1} \pm i \omega_1)} + \frac{1}{(\tau^{-1} \pm i \omega_2)} \right) \frac{\partial f_0}{\partial p_1 \partial p_2}$$  \hspace{1cm} (3.18d)
\[ f_2^{\pm(\omega_1-\omega_2)} = \frac{(eE_1)(eE_2)}{\tau^{1\pm}(\omega_1-\omega_2)} \left( \frac{1}{(\tau-1\pm\omega_1)} + \frac{1}{(\tau-1\mp\omega_2)} \right) \frac{\partial}{\partial p_1} \frac{\partial f_0}{\partial p_2} \]  (3.18e)

**For \( m=3 \)**

\[ f_3 = f_3^{3\omega_1} e^{3i\omega_1 t} + f_3^{-3\omega_1} e^{-3i\omega_1 t} + f_3^{3\omega_2} e^{3i\omega_2 t} + f_3^{-3\omega_2} e^{-3i\omega_2 t} + f_3^{\omega_1} e^{i\omega_1 t} + \]
\[ + f_3^{-\omega_1} e^{-i\omega_1 t} + f_3^{\omega_2} e^{i\omega_2 t} + f_3^{-\omega_2} e^{-i\omega_2 t} + \]
\[ + f_3^{(\omega_1+2\omega_2)} e^{i(\omega_1+2\omega_2)t} + f_3^{-(\omega_1+2\omega_2)} e^{-i(\omega_1+2\omega_2)t} + f_3^{(2\omega_1+\omega_2)} e^{i(2\omega_1+\omega_2)t} + \]
\[ + f_3^{(2\omega_1+\omega_2)} e^{-i(2\omega_1+\omega_2)t} + f_3^{(\omega_1-2\omega_2)} e^{i(\omega_1-2\omega_2)t} + f_3^{-(\omega_1-2\omega_2)} e^{-i(\omega_1-2\omega_2)t} + \]
\[ + f_3^{(2\omega_1-\omega_2)} e^{i(2\omega_1-\omega_2)t} + f_3^{-(2\omega_1-\omega_2)} e^{-i(2\omega_1-\omega_2)t} \]  (3.19)

The component of the Fermi-Dirac distribution with total field order of 3rd can be obtained by inserting Eq. (3.19) into (BTE).

We have 16 terms:

\[ f_3^{\pm3\omega_1} = g_3^{\pm3\omega_1} \frac{\partial^3 f_0}{\partial p_1^3} \]  (3.20a)
\[ f_3^{\pm3\omega_2} = g_3^{\pm3\omega_2} \frac{\partial^3 f_0}{\partial p_2^3} \]  (3.20b)
\[ f_3^{\pm\omega_1} = g_3^{\pm\omega_1} \frac{\partial f_0}{\partial p_1} + g_3^{\pm\omega_1} \frac{\partial^2 f_0}{\partial p_1^2} + \frac{\partial^3 f_0}{\partial p_1^3} \]  (3.20c)
\[ f_3^{\pm\omega_2} = g_3^{\pm\omega_2} \frac{\partial^2 f_0}{\partial p_2 \partial p_1} + g_3^{\pm\omega_2} \frac{\partial^3 f_0}{\partial p_2^3} \]  (3.20d)
\[ f_3^{\pm(2\omega_1+\omega_2)} = g_3^{\pm(2\omega_1+\omega_2)} \frac{\partial^2 f_0}{\partial p_1 \partial p_1^2} \]  (3.20e)
\[ f_3^{\pm(\omega_1+2\omega_2)} = g_3^{\pm(\omega_1+2\omega_2)} \frac{\partial^2 f_0}{\partial p_1 \partial p_2^2} \]  (3.20f)
\[ f_3^{\pm(2\omega_1-\omega_2)} = g_3^{\pm(2\omega_1-\omega_2)} \frac{\partial^2 f_0}{\partial p_2 \partial p_1^2} \]  (3.20g)
\[ f_3^{\pm(\omega_1-2\omega_2)} = g_3^{\pm(\omega_1-2\omega_2)} \frac{\partial^2 f_0}{\partial p_1 \partial p_2^2} \]  (3.20h)

Where the \( g \)-terms can be determined as:

\[ g_3^{\pm3\omega_1} = \frac{(eE_1)^3}{(\tau^{-1\pm\omega_1})(\tau^{-1\pm2\omega_1})(\tau^{-1\pm3\omega_1})} \]  (3.21a)
\[
\begin{align*}
\frac{\partial^3 f_0}{\partial p_x^3} &= \left( \cos^2 \theta \frac{\partial^3 f_0}{\partial^3 p} + \frac{3}{p} \sin^2 \theta \frac{\partial^2 f_0}{\partial^2 p} - \frac{3}{p^2} \sin^2 \theta \frac{\partial f_0}{\partial p} \right) \cos \theta \\
\frac{\partial^3 f_0}{\partial p_x \partial p_y^2} &= \left[ \sin^2 \theta \frac{\partial^3 f_0}{\partial^3 p} + \frac{1}{p} (1 - 3 \sin^2 \theta) \frac{\partial^2 f_0}{\partial^2 p} - \frac{1}{p^2} (1 - 3 \sin^2 \theta) \frac{\partial f_0}{\partial p} \right] \cos \theta
\end{align*}
\]
\begin{equation}
\frac{\partial^3 f_0}{\partial p_\gamma \partial p_\delta \partial p_\varepsilon} = \frac{\partial^3 f_0}{\partial p_\gamma \partial p_\delta \partial p_x} = \left[ \cos^2 \theta \frac{\partial^3 f_0}{\partial^2 p} + \frac{1}{p} (1 - 3\cos^2 \theta) \frac{\partial^2 f_0}{\partial^2 p} - \frac{1}{p^2} (1 - 3\cos^2 \theta) \frac{\partial f_0}{\partial p} \right] \sin \theta
\end{equation}

(3.24)

\begin{equation}
\frac{\partial^3 f_0}{\partial p_\gamma^3} = \left( \sin^2 \theta \frac{\partial^3 f_0}{\partial^3 p} + \frac{3}{p} \cos^2 \theta \frac{\partial^2 f_0}{\partial^2 p} - \frac{3}{p^2} \cos^2 \theta \frac{\partial f_0}{\partial p} \right) \sin \theta
\end{equation}

(3.25)

In order to find the current, we will need to use:

\[ J_n = -\frac{e}{\pi \hbar^2} \int d^2 p f_n v \]

(3.26)

From (3.12), we can write \( \frac{\partial}{\partial p_1} = \frac{\partial}{\partial p_x}, \) and \( \frac{\partial}{\partial p_2} = \cos \varphi \frac{\partial}{\partial p_x} + \sin \varphi \frac{\partial}{\partial p_y} \) but in this solution, we consider with the perpendicular incident fields, so \( \frac{\partial}{\partial p_1} = \frac{\partial}{\partial p_x}, \) and \( \frac{\partial}{\partial p_2} = \frac{\partial}{\partial p_y} \), the velocity operator has the form \( v = v_F (\cos \vartheta, \sin \vartheta) \), and \( \epsilon = v_F p \) (denote \( \epsilon_x = v_F p_x \)).

Here, we only investigate the current density along the x-axis.

\[ J_{nx} = -\frac{e v_F}{\pi \hbar^2} \int f_n p d \epsilon \cos \vartheta \sin \vartheta \theta = -\frac{e}{v_F \pi \hbar^2} \int f_n \epsilon \epsilon \cos \vartheta \sin \vartheta \theta \]

(3.27)

To calculate the transmission, we will need the electronic field components across the boundary with frequency \( \alpha \omega_1 \), so we only investigate the terms oscillating at this frequency.

With this, we get all 1st and 3rd order currents at \( \omega_1 \) as:

\[ J_{1x}^{\pm \omega_1} = -\frac{e}{v_F \pi \hbar^2} \int f_1^{\pm \omega_1} \epsilon \epsilon \cos \vartheta \sin \vartheta \theta = -\frac{e}{v_F \pi \hbar^2} \int \epsilon \epsilon \cos \vartheta \sin \vartheta \theta \]

(3.28)

We have let \( \xi = h \omega, \gamma = h \tau^{-1}, \alpha = \pm 1 \) and \( \sigma_0 = \frac{e^2}{4h} \)

\[ J_{3x}^{\pm \omega_1} = -\frac{e}{v_F \pi \hbar^2} \int f_3^{\pm \omega_1} \epsilon \epsilon \cos \vartheta \sin \vartheta \theta = \frac{3e v_F^2}{4\pi h^2} (g_{3p_1}^{\pm \omega_1} + \frac{1}{3} g_{3p_1 p_2}^{\pm \omega_1}) l_3 \]

(3.29)

It is convenient to make the substitution \( \phi = h ev_F \cong 6.582119 \times 10^{-8} eV^2 cm/V \) and \( E_1, E_2 \) in units of V/cm
\[ J_{3x}^{\omega_1} = \frac{3\sigma_0}{\pi} \phi^2 I_3 \left\{ \frac{3}{(y^2+\xi_1^2)(y+2ai\xi_1)} E_1^3 + 2 \left[ \frac{1}{(y+ai\xi_1)(y^2+\xi_1^2)} + \frac{(y^2+ai\xi_1^2)}{(y+ai\xi_1)(y^2+\xi_1^2)} \right] + \frac{1}{(y+ai\xi_1)(y+ai\xi_1)^2+\xi_1^2} \right\} \]

(3.30)

Where \( I_3 = \int_0^\infty \frac{1}{\epsilon} \frac{\partial f_0}{\partial \epsilon} d\epsilon \)

From complete calculations of the non-linear current density, we can continue to determine the transmission properties. We begin by considering \( E_1 = E_i \) to be the incident field and \( E_2 = E_p \) to be a pumping field.

The expression relating the incident, transmitted field, and current density can be given as

\[ E_i^x = E_t^x + \frac{\pi a}{2 \sigma_0 J_{tot}} \]

(3.31)

Where \( J_{tot} = \sigma_{tot}E_t \)

\[ \vec{E}_t = (E_i^x, 0, 0) \rightarrow E_i = E_i^x, \vec{E}_t = (E_t^x, 0, 0) \rightarrow E_t = E_t^x \]

In addition, the transmission through a linear thin film is determined by the standard expression as \( T = \frac{1}{|1 + \frac{2a}{\sigma_0 \sigma_{tot}}|^2} \)

Where \( T = |t|^2 \) with \( t = \frac{E_t}{E_i} \)

Because of the boundary conditions, \( J_{tot}^x = J_{tot}^{\omega_1} \) is the total current density at frequency \( \omega_1 \).

It can be written as:

\[ J_{tot}^{\omega_1} = J_{1x}^{\omega_1} + J_{3x}^{\omega_1} \]

According to calculate before, we have results of current density as:

From (3.28):

\[ J_{1x}^{\omega_1} = -\frac{4\sigma_0}{\pi} c I_1 \]

From (3.29):

\[ J_{3x}^{\omega_1} = \frac{3\sigma_0}{\pi} \phi^2 I_3 (aE_1^3 + bE_1 E_2^2) \]

Where \( a = \frac{3}{(y^2+\xi_1^2)(y+2ai\xi_1)} \)
\[ b = 2 \left[ \frac{1}{(\gamma + a\xi_1)(\gamma^2 + \xi_1^2)} + \frac{1}{(\gamma + a\xi_1)(\gamma + \xi_1^2)(\gamma^2 + \xi_1^2)} \right] \]

\[ c = \frac{1}{\gamma + ia\xi_1} \]

At zero temperature

\[ I_1 = -E_f, I_3 = -\frac{1}{E_f} \]

After we substitute into Eq. (3.31), the result obtained as

\[ \frac{3}{2} \alpha \phi^2 a I_3 E_i^2 t^3 + \left[ \frac{3}{2} \alpha \phi^2 b l_3 E_i^2 - 2 \alpha l_4 c + 1 \right] t - 1 = 0 \]  

(3.32)

\[ A t^3 + B t - 1 = 0 \]  

(3.33)

Where \( A = \frac{3}{2} \alpha \phi^2 a I_3 E_i^2 \) and \( B = \frac{3}{2} \alpha \phi^2 b l_3 E_i^2 - 2 \alpha l_4 c + 1 \)

Eq. (3.33) is a cubic equation, the results that we would expect are 3 complex values of \( t \) that will satisfy this equation. Computing solutions to Eq. (3.33) is made difficult by the complex coefficients \( A \) and \( B \). However, this will become easier when \( E_i \rightarrow 0 \), so the cubic term and two of the roots disappear. The result that we can obtain is a usual linear root. In this case, we can understand that the roots disappeared because of merging into a single point or they can both approach \( \infty \). Now if \( E_i \) is non-zero and we attempt to solve the cubic equation we are presented with the challenge of real analysis not being applicable to the complex function in Eq. (3.33) and we expect the coefficient of \( t \) to not diverge, we should expect to convergent bistability. This however doesn’t seem to be the case. It is likely because the analysis of the real function isn’t directly applicable to the complex function. To add further difficulty, this cubic equation has coefficients \( (A, B) \) that depend on \( \omega_1 \). What this does mean however, is that if the only bistability that occurs is divergent bistability, then despite there being 3 valid roots, the only valid root should be the one that comes from the solution when \( E_i = 0 \). We can clearly see that in Fig 3.14 with the existence of multiple branches.
Fig 3.14. Dependence of transmission on $E_2$, with $\omega_1 = \omega_2 = 0.2$THz, $E_F = 0.02eV$, and $E_1 = 1kV/cm$. The principal branch here is the branch that exists when $E_i = 0$.

Fig 3.15. Transmission ratio dependence on $\omega_2$, with $\omega_1 = 1$THz, $E_F = 0.02eV$, and $E_1 = 1kV/cm$.

As the result obtains from fig 3.15, there are some cases of the transmission ratio can exceed 1. Usually, this is not valid, but in this case, we can explain them a clear way.
Specifically, we can find the transmitted field strength that is a function that depend on both the incident field strength $E_i$ and the pumping field strength $E_2$. Therefore, in this case the transmitted field strength can be larger than the incident field strength when increasing the strength of the pumping field $E_2$. Since the transmitted field obtains the extra energy from the 2nd applied field, this case doesn’t transgress any physical laws.

Fig 3.16. Transmission ratio depends on $E_2$ at various frequencies $\omega = \omega_1 = \omega_2$, $E_F = 0.02eV$, and $E_1 = 1kV/cm$.

Fig 3.17. Transmission ratio depends on $E_1$ at various $E_2$ with $\omega_1 = \omega_2 = 0.2THz$, $E_F = 0.02eV$. 
From Eq. (3.33), we can determine that the transmission ratio \( T/T_0 \) (where \( T_0 = T(E_2 = 0) \)) depends on \( E_1, E_2, \omega_1 \) and \( \omega_2 \). Therefore, its behaviour will become complicated and difficult to predict. This is shown clearly in Figs 3.15 to 3.17 and here we can investigate changes of transmission that depends on \( E_2 \).

In summary, in this chapter, we showed some results of nonlinear conductivity in Dirac system. With influencing of a single electric field, the linear conductivity is more dominant because their components have positive values, bigger magnitudes and is affected very little by temperature. On the other hand, by using two incident fields, the results that we have received were very complicated and so difficult to predict their properties exactly, but we can determine that the pumping field can significantly modifies the conductivity and transmission of graphene.
Chapter 4

Nonlinear optical response in Semi-Dirac system

4.1. Introduction

In article of Shareef et al published February 2012, they used a semi-classical approach to expand the velocity operator of the applied field and their results found out that "single layer graphene exhibits a strong nonlinear photon-mixing effect in the terahertz frequency regime. Up to room temperature, the third-order nonlinear current in graphene grows rapidly with increasing temperature. The third-order nonlinear current can be as strong as the linear current under a moderate electric field strength of $10^4 \text{ V/cm}$. Because of the unique Dirac behaviour of the graphene quasi-particles, low Fermi level and electron fillings optimizes the optical nonlinearity. Under a strong-field condition, the strong-field-induced Dirac fermion population redistribution and non-equilibrium carrier heating effects further amplify the optical nonlinearity of graphene. These results suggest that doped graphene can potentially be utilized as a strong terahertz photon mixer in the room-temperature regime.", following up Fig 4.1. [68]

![Fig 4.1. Some plots in article “Room-temperature strong terahertz photon mixing in graphene” [68].](image-url)
A finding in ultrathin (001) VO$_2$ layers embedded in TiO$_2$ was generated by Pardo et al. This new point Fermi surface system, dubbed ‘semi-Dirac,’ is a hybrid of conventional and unconventional: dispersion is linear (“massless”, Dirac-Weyl) in one of the directions of the two-dimensional (2D) layer, and is conventional quadratic (“massive” Dirac) in the perpendicular direction. At directions between the axes the dispersion is intermediate and highly direction-dependent. Interest in this unique, maximally anisotropic, dispersion arises for several reasons. They extend the semi-classical study of fermionic particle-hole symmetric semi-Dirac (more appropriately, semi- Dirac semi-Weyl) dispersion of quasiparticles, $\varepsilon_\lambda = \pm \varepsilon_0 \sqrt{\varepsilon_x^2 + \varepsilon_y^2}$ in dimensionless units, discovered computationally in oxide heterostructures. [43].

![Graphene and Semi-Dirac points](image)

**Fig 4.2. Dirac point and Semi-Dirac point.**

We can see clearly difference between Dirac and Semi-Dirac points by Fig 4.2.

In this section, we will construct the full temperature and chemical potential spectrum of the nonlinear optical response in semi-Dirac system under strong field condition. The dynamics of the quasiparticles when perturbed by a strong electric field are decomposed into linear and nonlinear components, and the optical nonlinearity of semi-Dirac system is investigated.

Here, we performed to find the expressions for the optical response in semi-Dirac system by expand the velocity operator in a strong driving field. This allows us to determine the optical response that will depend on both temperature and chemical potentials.
4.2. Optical response of Semi-Dirac System

Next, we show that the expressions for the optical response in semi-Dirac system will be first determined in terms of Cartesian coordinates, and then the general transverse and longitudinal these expressions are derived. It turns out that the general expressions for this system depends on the values of the optical response in Cartesian coordinates.

From (1.27), the energy-momentum dispersion of the Semi-Dirac system is found as:

$$\epsilon_\lambda = 2 m \lambda v_F^2 \sqrt{\frac{p_x^4}{(2m v_F)^4} + \frac{p_y^2}{(2m v_F)^2}}$$

where $\lambda = \pm 1$ is the index of valence and conduction bands, $m$ is the effective mass, $v_F = 10^6 m/s$ is the Fermi velocity, $p_x$ and $p_y$ are the respective momentums in x and y directions.

The velocity operator is given by $\vec{v} = \frac{\partial \hat{H}}{\partial \vec{p}}$ and the corresponding expectation value [17] can be written as $\overrightarrow{v_\lambda} = \frac{\partial \epsilon_\lambda}{\partial p}$

From (1.19)

$$\begin{align*}
\left\{ \begin{array}{l}
v_x = \frac{\partial \epsilon_\lambda}{\partial p_x} = \frac{\lambda p_x^3}{m F} \\
v_y = \frac{\partial \epsilon_\lambda}{\partial p_y} = \frac{2m \lambda v_F^2 p_y}{F}
\end{array} \right.
\end{align*}$$

where $\epsilon_0 = 2m v_F^2$ and $F = \sqrt{p_x^4 + 2m \epsilon_0 . p_y^2}$

We consider a time-dependent applied electric field in the form of

$$E(r, t) = \sum_\mu E_\mu \exp\left\{i(q_\mu \cdot r - \omega_\mu t)\right\}$$

(4.1)

Where $E_\mu$, $q_\mu$ and $\omega_\mu$ are the amplitude, wave vector and frequency of $\mu$th wave of the electric field. Ignoring the weak magnetic component, the external field is minimally coupled to the quasi-particle by performing the substitution $\vec{p} \rightarrow \vec{p} - e \vec{A}(r, t)$ where $\vec{u} = -e \vec{A}(r, t)$, $E(r, t) = -\partial \vec{A}(r, t)/\partial t$ and $e$ is the electric charge. The externally perturbed quasiparticle
acquires linear and nonlinear velocities which can be separated by performing a Taylor expansion with respect to the external field.

Case: \( \mathbf{u} \parallel \mathbf{e}_x \) → \( \begin{cases} p_x \to p_x + u_x \\ p_y \to p_y \end{cases} \)

A straightforward expansion of \( v_s \) up to third order of the applied field, assuming \( p(x,y) \gg u(x,y) \). The velocity becomes:

\[
\begin{align*}
\nu_x &= \frac{\lambda(p_x+u_x)^3}{m\sqrt{(p_x+u_x)^4+2\varepsilon_0p_y^2}} \\
\nu_y &= \frac{\varepsilon_0\lambda p_y}{\sqrt{(p_x+u_x)^4+2\varepsilon_0p_y^2}}
\end{align*}
\]

Up to third-order in the external field, the linear and nonlinear field–dependent velocities can be written as

\[
\begin{align*}
\left\{ \begin{array}{l}
\nu_x^{(0)} = \nu_x(0) = \frac{\lambda p_x^3}{mF} \\
\nu_y^{(0)} = \nu_y(0) = \frac{\lambda\varepsilon_0 p_y}{F}
\end{array} \right. \tag{4.2}
\end{align*}
\]

\[
\begin{align*}
\left\{ \begin{array}{l}
\nu_x^{(1)} = \frac{1}{1!} u_x \frac{\partial \nu_x}{\partial u_x}(0) = \frac{3\lambda p_x^2 u_x}{mF} - \frac{2\lambda p_x^6 u_x}{mF^3} \\
\nu_y^{(1)} = \frac{1}{1!} u_x \frac{\partial \nu_y}{\partial u_x}(0) = -\frac{2\lambda\varepsilon_0 p_y p_x^2 u_x}{F^3}
\end{array} \right. \tag{4.3a}
\end{align*}
\]

\[
\begin{align*}
\left\{ \begin{array}{l}
\nu_x^{(2)} = \frac{1}{2!} u_x \frac{\partial^2 \nu_x}{\partial u_x^2}(0) = \frac{3\lambda p_x^2 u_x^2}{mF} - \frac{9\lambda p_x^6 u_x^2}{mF^3} + \frac{6\lambda p_x^6 u_x^2}{mF^5} \\
\nu_y^{(2)} = \frac{1}{2!} u_x \frac{\partial^2 \nu_y}{\partial u_x^2}(0) = -\frac{3\lambda\varepsilon_0 p_y p_x^2 u_x^2}{F^3} + \frac{6\lambda\varepsilon_0 p_y p_x^6 u_x^2}{F^5}
\end{array} \right. \tag{4.3b}
\end{align*}
\]

\[
\begin{align*}
\left\{ \begin{array}{l}
\nu_x^{(3)} = \frac{1}{3!} u_x \frac{\partial^3 \nu_x}{\partial u_x^3}(0) = \frac{\lambda u_x^3}{mF} - \frac{17\lambda p_x^4 u_x^3}{mF^3} + \frac{36\lambda p_x^6 u_x^3}{mF^5} - \frac{20\lambda p_x^{12} u_x^3}{mF^7} \\
\nu_y^{(3)} = \frac{1}{3!} u_x \frac{\partial^3 \nu_y}{\partial u_x^3}(0) = -\frac{2\lambda\varepsilon_0 p_y p_x^2 u_x^3}{F^3} + \frac{18\lambda\varepsilon_0 p_y p_x^6 u_x^3}{F^5} - \frac{20\lambda\varepsilon_0 p_y p_x^6 u_x^3}{F^7}
\end{array} \right. \tag{4.3c}
\end{align*}
\]

Case: \( \mathbf{u} \parallel \mathbf{e}_y \) → \( \begin{cases} p_x \to p_x \\ p_y \to p_y + u_y \end{cases} \)

The velocity becomes:
\[ \begin{align*}
\begin{cases}
  v_x &= \frac{\lambda p_x^3}{m \sqrt{p_x^4 + 2m \varepsilon_0 (p_y + u)^2}} \\
  v_y &= \frac{\varepsilon_0 \lambda (p_y + u)}{\sqrt{p_x^4 + 2m \varepsilon_0 (p_y + u)^2}}
\end{cases}
\end{align*} \tag{4.4} \]

Up to third-order in the external field, the linear and nonlinear field-dependent velocities can be written as

\[ \begin{align*}
\begin{cases}
  v_x^{(0)} &= v_x(0) = \frac{\lambda p_x^3}{mf} \\
  v_y^{(0)} &= v_y(0) = \frac{\varepsilon_0 \lambda p_y}{f}
\end{cases} \tag{4.5a} \\
\begin{cases}
  v_x^{(1)} &= \frac{1}{1!} u_y \frac{\partial v_x}{\partial u_y}(0) = -\frac{2\varepsilon_0 p_y^2 u_y}{f^3} \\
  v_y^{(1)} &= \frac{1}{1!} u_y \frac{\partial v_y}{\partial u_y}(0) = \frac{\varepsilon_0 u_y}{f} - \frac{2m\varepsilon_0^2 p_y^2 u_y}{f^3}
\end{cases} \tag{4.5b} \\
\begin{cases}
  v_x^{(2)} &= \frac{1}{2!} u_y^2 \frac{\partial^2 v_x}{\partial u_y^2}(0) = -\frac{\lambda p_x^3 u_y^2}{f^3} + \frac{6m\lambda \varepsilon_0 p_x^2 p_y^2 u_y^2}{f^5} \\
  v_y^{(2)} &= \frac{1}{2!} u_y^2 \frac{\partial^2 v_y}{\partial u_y^2}(0) = -\frac{3m\varepsilon_0^2 p_y^2 u_y^2}{f^3} + \frac{6m^2\lambda \varepsilon_0^3 p_y^3 u_y^2}{f^5}
\end{cases} \tag{4.5c} \\
\begin{cases}
  v_x^{(3)} &= \frac{1}{3!} u_y^3 \frac{\partial^3 v_x}{\partial u_y^3}(0) = \frac{6m\lambda \varepsilon_0^2 p_x^2 p_y^2 u_y^2}{f^5} - \frac{20m^2\lambda \varepsilon_0^{3} p_x p_y^2 u_y^3}{f^7} \\
  v_y^{(3)} &= \frac{1}{3!} u_y^3 \frac{\partial^3 v_y}{\partial u_y^3}(0) = -\frac{m\varepsilon_0^2 u_y^3}{f^3} + \frac{12m^2\lambda \varepsilon_0^{3} p_y^2 u_y^3}{f^5} - \frac{20m^3\lambda \varepsilon_0^{4} p_y u_y^3}{f^7}
\end{cases} \tag{4.5d}
\end{align*} \]

The optical current is given as

\[ J^{(1)} = e \sum \int v^{(i)} f(\varepsilon, \mu) dp \tag{4.6} \]

Where \( f(\varepsilon, \mu) = \frac{1}{1 + \exp \left( \frac{\mu - \varepsilon}{k_B T} \right)} \) is the Fock-Dirac distribution.

From (4.6), the optical current can be written as

\[ J^{(n)}_{(x,y)} = e \int_{x_0}^{x_{max}} \int_{y_0}^{y_{max}} v^{(n)}_{(x,y)} [f(\varepsilon) - f(\varepsilon + \delta)] dp_x dp_y \tag{4.7} \]

Where \( \delta = \sum \varepsilon_{\text{photon}} \) is the total energy of the incoming photons and \( v^{(n)}_{(x,y)} \) is the n\textsuperscript{th} order velocity to the applied electric field.

From Eq. (4.7), the linear current can be explicitly written as
\[ J_{(x,y)}^{(1)} = e \int_{x_0}^{x_{\text{max}}} \int_{y_0}^{y_{\text{max}}} v_{(x,y)}^{(1)} f^{(1)}(\varepsilon) \, dp_x dp_y \]  

(4.8)

Where \( f^{(1)}(\varepsilon) = f(\varepsilon) - f(\varepsilon + \varepsilon_1) \)

And, the third order nonlinear optical current can be explicitly written as

\[ J_{(x,y)}^{(3)} = e \int_{x_0}^{x_{\text{max}}} \int_{y_0}^{y_{\text{max}}} v_{(x,y)}^{(3)} f^{(3)}(\varepsilon) \, dp_x dp_y \]  

(4.9)

Where \( f^{(3)}(\varepsilon) = f(\varepsilon) - f(\varepsilon + \varepsilon_1 + \varepsilon_2 + \varepsilon_3) \)

As far as we know, in conventional semiconductor systems with parabolic energy-momentum dispersion, the third order current density cannot compare to the linear current density. However, in strong non-linear systems, this can occur. Therefore, in this system, we can consider this relation through the ratio of the third order nonlinear optical response to the linear optical response in both x and y directions.

Firstly, we examine the ratio of the third order current density to the linear current density in case \( \vec{u} \parallel \vec{e}_x \)

\[ \frac{J_{x,y}^{(3)}}{J_{x,y}^{(1)}} = u_x \frac{ \int_{p_{x_0}}^{p_{x_{\text{max}}}} \int_{p_{y_0}}^{p_{y_{\text{max}}}} f_{(x,y)}^{(3)} G_{xx}^{(3)} \, dp_x dp_y }{ \int_{p_{x_0}}^{p_{x_{\text{max}}}} \int_{p_{y_0}}^{p_{y_{\text{max}}}} f_{(x,y)}^{(1)} G_{xx}^{(1)} \, dp_x dp_y } \]  

(4.10)

Where \( G_{xx}^{(1)} = \frac{3p_x^2}{F} - \frac{2p_x^4}{F^3} \) and \( G_{xx}^{(3)} = \frac{1}{F} - \frac{17p_x^4}{F^3} + \frac{36p_x^6}{F^5} - \frac{20p_x^{12}}{F^7} \)
Fig 4.3. Case $\vec{u} \parallel \hat{e}_x$, temperature dependent the ratio of the third order nonlinear optical response to the linear optical response at various chemical potentials. The electric field is $10^4 V/cm$, and the frequency photons are $\omega_1 = \omega_2 = 1 THz, \omega_3 = 2 THz$.

Fig 4.4. Case $\vec{u} \parallel \hat{e}_x$, the ratio of the third order nonlinear optical response to the linear optical response versus chemical potential at various temperature. The electric field is $10^4 V/cm$, and the frequency photons are $\omega_1 = \omega_2 = 1 THz, \omega_3 = 2 THz$. 
In Figs. 4.3 and 4.4, we consider the ratio of the third order non-linear optical response to the linear optical response in x direction. Here, we can see that \( \frac{J_x^{(3)}}{J_x^{(1)}} \) decreases rapidly when temperature and chemical potential increase and \( J_x^{(3)} \ll J_x^{(1)} \). This situation is expected as linear response dominates, specially at high temperatures and large chemical potentials.

Next, we investigate the ratio of the third order nonlinear optical response to the linear optical response in case \( \vec{u} \parallel \vec{e}_y \)

\[
\frac{J_y^{(3)}}{J_y^{(1)}} = 2m^2v_F^2u_y \frac{\int_{P_{x0}}^{P_{x\max}} \int_{P_{y0}}^{P_{y\max}} f^{(3)}(e)G_{yy}^{(3)} dp_x dp_y}{\int_{P_{x0}}^{P_{x\max}} \int_{P_{y0}}^{P_{y\max}} f^{(1)}(e)G_{yy}^{(1)} dp_x dp_y}
\]

Where \( G_{yy}^{(1)} = \frac{1}{f} - \frac{4m^2v_F^2p_y^2}{f^3} \) and \( G_{yy}^{(3)} = \frac{1}{f^3} + \frac{24m^2v_F^2p_y^2}{f^5} - \frac{80m^4v_F^2p_y^4}{f^7} \)

**Fig 4.5.** Case \( \vec{u} \parallel \vec{e}_y \), temperature dependent the ratio of the third order nonlinear optical response to the linear optical response at various chemical potentials. The electric field is \( 10^4 V/cm \), and the frequency photons are \( \omega_1 = \omega_2 = 1THz, \omega_3 = 2THz. \)
Fig 4.6. Case $\vec{u} \parallel \vec{e}_y$, the ratio of the third order nonlinear optical response to the linear optical response versus chemical potential at various temperature. The electric field is $10^4 V/cm$, and the frequency photons are $\omega_1 = \omega_2 = 1THz, \omega_3 = 2THz$.

From Figs. 4.5 and 4.6, we can see that the third order current density much larger than the linear current density. This shows that the third order non-linear optical response in y direction has more potential. On the other hand, there exists an optimal temperature around 120K at which the ratio of the third order nonlinear optical response to the linear optical response $J_y^{(3)} / J_y^{(1)}$ is highest and this optimal temperature increases with the chemical potential. At room temperature, the nonlinear ratio terms persist with high chemical potential and decrease insignificantly to that ratio at zero temperature.

Furthermore, these figs show that the ratio $J_y^{(3)} / J_y^{(1)}$ value in y direction (Figs.4.5 and 4.6) is much larger than that in x direction (Figs.4.3 and 4.4). This proves that the strong non-linear orientation in y direction will be predict that there are interesting applications in THz devices.
Chapter 5

Conclusions

In this thesis, we presented some results of optical conductivity in Dirac system for some incident fields (considering both one incident field and two incident fields with different intensity and frequency) and optical response in a Semi-Dirac system, an electronic system whose energy-momentum dispersion is parabolic in the x dispersion and linear in the y dispersion.

For the Dirac system, we have systematically investigated the nonlinear optical conductivity with effect of some incident fields. By using one incident field, we focused on the low frequency area of under 2 THz and the Fermi energy area of under 0.2 eV, because here both real and imaginary parts of intraband conductivity have quite large differences. Furthermore, in the case of one incident field, we can see that the linear conductivity is superior to the nonlinear conductivity because it: (1) has both the real and imaginary components that are positive at frequency \( \omega \), (2) has magnitudes much bigger than the nonlinear conductivity, and (3) is affected very little by temperature. However, when we investigate the 3\(^{rd}\) order conductivity at frequency \( 3\omega \), the real component can be found to be positive for \( \xi > \frac{\gamma}{\sqrt{11}} \).

Similarly, we examined the transmission properties of a thin film and computed the dependence on elements of electric fields and frequencies for two incident fields with different intensity and frequency. The results show that the behaviours of transmission ratio \( \frac{T}{T_0} \) (\( T_0 = T(E_2 = 0) \)) has a complicated dependence on \( E_1, E_2, \omega_1 \) and \( \omega_2 \), so we cannot determine their properties exactly. On the other hand, there is some cases where the transmission ratio can exceed 1 but this doesn’t transgress any physical laws because the transmitted field strength is a function that is depend on both the incident field strength (\( E_i \)) and the pumping field strength (\( E_p \)). Therefore, the pumping field can significantly modify the conductivity and transmission of graphene. This may contribute a technique of pump-to-probe experiments involving two fields.
For the nonlinear optical response in Semi-Dirac system, we constructed the full temperature and chemical potential spectrum of the nonlinear optical response in semi-Dirac system under the presence of an applied field of $10^3 - 10^4 \, V/cm$ along the x is parabolic dispersion and the y is linear dispersion. Our results showed that strength of the nonlinear optical response has a much larger value along y direction. The large magnitude of nonlinear optical response persists at room temperature and for large chemical potential values. Furthermore, the ratio of the third order nonlinear optical response to the linear optical response $\frac{J_y^{(3)}}{J_y^{(1)}}$ can reach the highest value at an optimal temperature around 120K and increases with the chemical potential. In conclusion, in Semi-Dirac system, the strong non-linear orientation in y direction is expected as potential applications in THz devices.
References


