CONDENSED MATTER PHYSICS AND NANOTECHNOLOGY MASTER THESIS

UNIVERSIDAD AUTÓNOMA DE MADRID - UNIVERSIDAD DE OVIEDO

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# On the Universal Absorption in Two-Dimensional Systems

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# 1

# Introduction

The first isolation of graphene [**?**], a single atomic sheet of carbon atoms arranged in a honeycomb lattice made out of hexagons, has motivated a deep study of its interesting properties so far. Its particular lattice structure, with two identical atoms in the unit cell, provokes an unique energy dispersion relation near the so-called Dirac points, where the charge is neutral. In the low energies regime ($E < 1\,\mathrm{eV}$), the charge carriers in graphene behave like massless Dirac fermions which can explain many of its interesting electronic properties [**?**, **?**, **?**, **?**], and new physical phases such as the non-interger Quantum Hall Effect [**?**, **?**] or the Quantum Spin Hall Effect [**?**]. Also applications of graphene due to their optical properties [**?**] are very likely in the near future, some of which are based on the generation of long-lived plasmons [**?**, **?**].

Studies of graphene have motivated the search of other truly two-dimensional systems, either by chemical modification of graphene or exfoliation of other layered compounds, bringing up a new class of materials: heterostructures based on 2D atomic crystals [**?**]. One of these are the transition metal dichalcogenides, whose structure is characterized by the presence of three atomic planes per layer, a triangular lattice of transition metals atoms merged with two triangular lattices of oxygen family atoms (O, S, Se or Te). Among them, the molybdenum disulfide ($\mathrm{MoS_2}$) bears a special resemblance to massive graphene (graphene with an induced gap [**?**]), as its energy spectrum can be described by gapped Dirac fermions. Due to the presence of a large gap, molybdenum disulfide has attracted a lot of attention recently. Also the absence of inversion symmetry, what makes the spin orbit coupling naturally large in contrast to massive graphene. While a monolayer of $\mathrm{MoS_2}$ presents a direct band gap, the bulk is known to be an indirect semiconductor [**?**, **?**].

Dirac fermions also occur in another kind of layered materials with repeating unit cells of hexagonal structure of five layers, the 3D topological insulators. Such materials show surface states that, as in graphene, follows the massless fermions Dirac equation but with the presence of only one Dirac cone in their energy spectrum, whereas the bulk states show a large insulating gap. Breaking time-reversal invariance by, e.g., applying a magnetic field further produces a gap in the spectrum giving rise to giant magneto-optical Kerr effect and universal Faraday effect. [**?**]

Another difference between Dirac fermions in topological insulators with respect to graphene ones resides in the coupling with the momentum operator: while in the former systems it is produced by the real spin, in the latter is generated by the so-called pseudospin that appears due to the two atoms in the unit cell of the graphene lattice. This state of matter, also refered as Quantum Spin Hall State, was confirmed experimentally in 2D topological insulators by Molenkamp [?] in HgTe quantum wells grown by molecular-beam epitaxy.

In this master thesis a study of the absorbance of 2D systems is presented. Such study is motivated by the universal absorption of graphene which only depends on fundamental constants and is giving by $\pi\alpha$ with $\alpha = e^2/4\pi\epsilon_0$ the fine structure constant [?]. The proportionality to the fine-structure constant can intuitively be understood by noticing that the QED coupling of the electrons with the photonic Gauge field is determined by such a constant. The scattering rate, i.e., the absorption, should thus be proportional to $\alpha$.

Absorption measurements were recently performed on conventional 2D semiconductors structures [?]. For InAs membranes with a thickness small enough to be considered a 2D semiconductor system, they found that each set of interband transitions in the optical range was mediated by a quantum absorptance of $\pi\alpha/n_c$ whith $n_c$ an optical local field correction factor. The stairway appearance of the absorption was thus characterized by this quantum of absorption and the thickness of the sample, which is determined by the number of piled 2D monolayers.

In this master thesis, we intend to investigate the universal absorption in 2D systems in more detail. We first calculate the absorption for Dirac fermions including a mass term in the massless Dirac Hamiltonian and show that this system does not display the universal behavior for general frequencies. We further theoretically discuss the absorption of ordinary 2D semiconductors described within the effective-mass approximation. Finally we calculate the absorption of a truely 2D semiconductor crystal, $MoS_2$, experimentally measured in [?]. We close with a summary of our results and conclusions.

# 2

# Absorption in the linear regime

In a 3D material, the absorption coefficient $A(\omega)$ is characterized by the exponential attenuation of the intensity of the wave after having crossed a thickness $z$

$$I(z) = I_0 e^{-A(\omega)z} \ . \tag{2.1}$$

Since it only involves the intensity of the electromagnetic wave, direct measurements of the absorption can be easily carried out. Such measurements are of quite interest if we are studying processes near the band edge such as indirect transitions or, what interests us more, direc interband transitions. Far from the band edge, the absorption coefficients become too high for the absorption technique to be useful, so reflectivity measurements are made in such cases.

In 2D, an alternative definition of the absorption coefficient is necessary. We can define it as the power removed from the incident beam per unit volume per unit incident flux of electromagnetic energy due to an interband transition [**?**]

$$A(\omega) = \frac{\eta\hbar\omega}{\text{incident electromagnetic flux}} \ , \tag{2.2}$$

where $\eta$ = number of transitions/unit area/unit time. We will use $W_a$ to denote the numerator of equation **??**. In this work we are considering an electromagnetic wave linearly polarized with electric field $\mathbf{E}(t)$ and frequency $\omega$, propagating along the $x$ axis with normal incidence through the system of area $A$ (see figure **??**). Under this assumption, the incident electromagnetic flux ($W_i$ from now) reads

$$W_i = \frac{\varepsilon_0 c}{2}|\mathbf{E}|^2 \ . \tag{2.3}$$

On the other hand, we can directly calculate the Fresnel coefficients for the reflection ($r$) and transmission ($t$) amplitude from the scattering problem of a planar interface. The specular absorption is then defined via the continuity equation by

$$A = 1 - R - T \ , \tag{2.4}$$

with $R = |r|^2$ and $T = \sqrt{\frac{\varepsilon_2}{\varepsilon_1}}|t|^2$ where $\varepsilon_1$ and $\varepsilon_2$ the relative permittivity of the two media separated by the sample of the material. The transmission and reflection of a planar interface including graphene is then characterized by the optical conductivity $\sigma(\omega)$ and given by

$$\begin{aligned} T &= \sqrt{\frac{\varepsilon_2}{\varepsilon_1}} \frac{4(\varepsilon_1\varepsilon_0)^2}{|(\sqrt{\varepsilon_1\varepsilon_2}+\varepsilon_1)\varepsilon_0+\sqrt{\varepsilon_1}\sigma(\omega)/c|^2} \\ R &= \frac{|\sqrt{\varepsilon_1\varepsilon_2}\varepsilon_0+\sqrt{\varepsilon_1}\sigma(\omega)/c-\varepsilon_1\varepsilon_0|^2}{|\sqrt{\varepsilon_1\varepsilon_2}\varepsilon_0+\sqrt{\varepsilon_1}\sigma(\omega)/c+\varepsilon_1\varepsilon_0|^2} \end{aligned} \quad (2.5)$$

Thus, we will calculate the absorption using two different approaches: firstly we will determine the transitions rate $\eta$ using Fermi's Golden Rule and computing the absorption from equation **??**. Secondly, by means of equations **??** and **??** within the Linear Response Theory context. Both methods are detailed below.

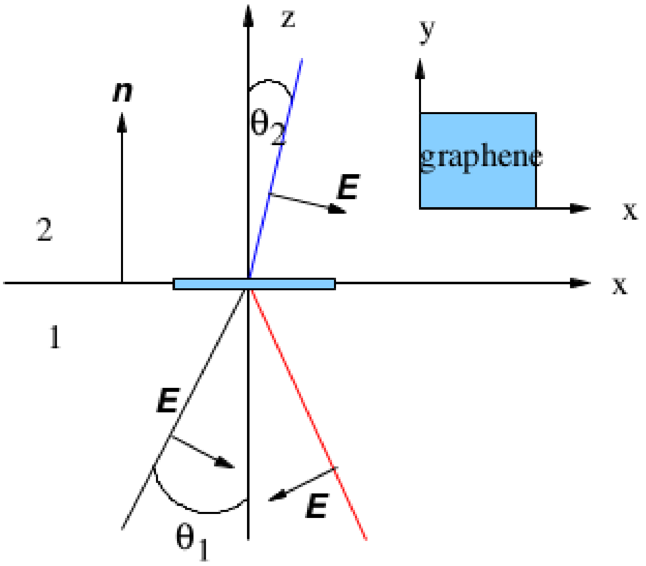

Figure 2.1: Geometry of $p$ polarized light scattering between two media with graphene separating them [**?**]

## 2.1 Fermi's Golden Rule

Time-dependent perturbation theory in leading (second) order leads to the following transition probabilities:

$$p_{m\to n}(t,t_0) = \frac{1}{\hbar^2}\left|\int_{t_0}^{t}\langle n|H_1(t')|m\rangle e^{i(\epsilon_n-\epsilon_m)t'/\hbar}dt'\right|^2 \ , \ n\neq m \quad (2.6)$$

$$p_{m\to m}(t,t_0) = 1-\sum_{m\neq n}\frac{1}{\hbar^2}\left|\int_{t_0}^{t}\langle n|H_1(t')|m\rangle e^{i(\epsilon_n-\epsilon_m)t'/\hbar}dt'\right|^2 \ , \quad (2.7)$$

where $m, \epsilon_m$ are the eigenstates and eigenenergies of the unperturbed Hamiltonian $H_0$ respectively and $H_1(t)$ a time-dependent perturbation which is switched on at $t = t_0$. These formulas are only valid if $1 - p_{m\to m} \ll 1$. In the case of a periodic perturbation

$$H_1(t) = H_1\cos(\omega t) \ , \quad (2.8)$$

the probability can be approximated to

$$p_{m\to n}(t,t_0)=(t-t_0)\frac{\pi}{2\hbar}|\langle n|H_1|m\rangle|^2\delta(\epsilon_n-\epsilon_m-\hbar\omega)\ ;\ t-t_0\to\infty\ . \tag{2.9}$$

Within a two-band model and considering momentum conservation, only transitions from the valence band $m=(\mathbf{k},-)$ to the conduction band $n=(\mathbf{k},+)$ are allowed

$$p_{\mathbf{k}}=(t-t_0)\frac{\pi}{2\hbar}|\langle\mathbf{k},+|H_1|\mathbf{k},-\rangle|^2\delta(\epsilon_{+,\mathbf{k}}-\epsilon_{-,\mathbf{k}}-\hbar\omega)\ . \tag{2.10}$$

Thus, the transition rate per unit area is given by

$$\eta_{\mathbf{k}}=\frac{\pi}{2\hbar A}|\langle\mathbf{k},+|H_1|\mathbf{k},-\rangle|^2\delta(\epsilon_{+,\mathbf{k}}-\epsilon_{-,\mathbf{k}}-\hbar\omega)\ , \tag{2.11}$$

where $A$ denote the area considered. The total transition rate is obtained by summing over all initial states

$$\eta=\frac{\pi}{2\hbar A}\sum_{\mathbf{k}}|\langle\mathbf{k},+|H_1|\mathbf{k},-\rangle|^2\delta(\epsilon_{+,\mathbf{k}}-\epsilon_{-,\mathbf{k}}-\hbar\omega)\ . \tag{2.12}$$

Because the electronic states in the Brillouin zone are quasi-continuous functions of $\mathbf{k}$ and considering a two-dimensional system, we can replace the sum by the following integral:

$$\eta=\frac{1}{8\hbar\pi}\iint d\mathbf{k}|\langle\mathbf{k},+|H_1|\mathbf{k},-\rangle|^2\delta(\epsilon_{+,\mathbf{k}}-\epsilon_{-,\mathbf{k}}-\hbar\omega)\ , \tag{2.13}$$

and by a change of variable

$$k\to\epsilon_{+,\mathbf{k}}-\epsilon_{-,\mathbf{k}}\ , \tag{2.14}$$

we can transform the integral in $k$ to an integral in the difference in energy between the two bands (eigenstates) involving the transition

$$\begin{aligned}\eta &= \frac{1}{8\hbar\pi}\iint d\varphi d(\epsilon_{+,\mathbf{k}}-\epsilon_{-,\mathbf{k}})k(\epsilon_{+,\mathbf{k}}-\epsilon_{-,\mathbf{k}})\left|\frac{\partial k}{\partial(\epsilon_{+,\mathbf{k}}-\epsilon_{-,\mathbf{k}})}\right|\times\\ &\quad |\langle\mathbf{k},+|H_1|\mathbf{k},-\rangle|^2\delta(\epsilon_{+,\mathbf{k}}-\epsilon_{-,\mathbf{k}}-\hbar\omega)\ .\end{aligned} \tag{2.15}$$

## 2.2 Linear Response

Considering an applied field $\mathbf{E}(t)=-\partial_t\mathbf{A}(t)$, with monochromatic frequency dependence $\mathbf{E}(t)=i\omega\mathbf{A}(t)$, the current operator is then given by

$$\begin{aligned}j^i_{tot} &= \frac{ine^2}{m\omega}E^i+j^i\ ,\\ j^i &= -\frac{e}{m}\sum_\alpha p^i_\alpha\end{aligned} \tag{2.16}$$

where $\mathbf{p}_\alpha$ is the momentum operator for the $\alpha$th electron. The first term of $j_{tot}$ is known as the diamagnetic contribution, related to the substitution $\mathbf{p}_\alpha\to\mathbf{p}_\alpha+e\mathbf{A}(\mathbf{r}_\alpha,t)$. The Hamiltonian of the system is

$$\begin{aligned}H &= H_0+H_1(t)\ ,\\ H_1(t) &= -\mathbf{j}\cdot\mathbf{A}(t)=(i/\omega)\mathbf{j}\cdot\mathbf{E}(t)\end{aligned} \tag{2.17}$$

where $H_0$ contains all the equilibrium properties of the system. We want to compute the current by means of the density matrix. The density matrix is perturbed away from its equilibrium value $\rho = e^{-\beta H_0/Z}$ by the field, and we want to know the correction factor $\delta\rho$ to first order in $\mathbf{E}$ (linear response). First-order time-dependent perturbation theory for $\delta\rho$ can be written in operator language using standard field-theoretic methods as

$$\delta\rho(t) = -\frac{i}{\hbar} e^{-iH_0 t/\hbar} \int_{-\infty}^{t} dt' [H_1(t'), \rho] e^{iH_0 t/\hbar} \, . \tag{2.18}$$

From this last equation we can derive Kubo formula for the conductivity in the spectral representation given a set of eigenstates $H_0|n\rangle = \epsilon_n|n\rangle$ of the system before the field is applied

$$\sigma^{ij}(\omega) = \frac{ine^2}{m\omega}\delta_{ij} + \frac{ie^2}{\omega V}\sum_{m,n} \frac{e^{-\beta\epsilon_n} - e^{-\beta\epsilon_m}}{Z} \frac{\langle n|v^i|m\rangle\langle m|v^j|n\rangle}{\hbar(\omega + i\delta) - (\epsilon_m - \epsilon_n)} \, . \tag{2.19}$$

Considering a 2D system of area $A$, the real part of the conductivity reads [**?**]

$$\mathrm{Re}\,\sigma^{ij} = \frac{4\sigma_0}{\omega A} \sum_{m\in c, n\in v} \pi\langle m|v^i|n\rangle\langle n|v^j|m\rangle\delta(\hbar\omega - (\epsilon_m - \epsilon_n)) \, , \tag{2.20}$$

where $c$,$v$ denotes the empty and occupied states respectively and $\sigma_0 = \frac{e^2}{4\hbar}$. The velocity operator is given by

$$\mathbf{v} = \frac{i}{\hbar}[H, \mathbf{r}] = \frac{\partial H}{\partial \mathbf{p}} \, . \tag{2.21}$$

If we consider that the electric field is oriented along the $x$ direction then the Kubo formula reads

$$\mathrm{Re}\,\sigma^{xx} = \frac{4\sigma_0}{\omega A} \sum_{m\in c, n\in v} \pi|\langle m|v^x|n\rangle|^2\delta(\hbar\omega - (\epsilon_m - \epsilon_n)) \, . \tag{2.22}$$

On the other hand we can transform the summatory in accesible states into a sumatory in $\mathbf{k}$ as follows

$$\begin{aligned} \mathrm{Re}\,\sigma^{xx} &= \frac{4\sigma_0\pi}{\omega A}\sum_{\mathbf{k}} |\langle \mathbf{k}, +|v^x|\mathbf{k}, -\rangle|^2\delta(\hbar\omega - (\epsilon_{+,\mathbf{k}} - \epsilon_{-,\mathbf{k}})) = \\ &= \frac{\sigma_0}{\omega\pi}\iint d\varphi d(\epsilon_{+,\mathbf{k}} - \epsilon_{-,\mathbf{k}}) k(\epsilon_{+,\mathbf{k}} - \epsilon_{-,\mathbf{k}}) \left|\frac{\partial k}{\partial(\epsilon_{+,\mathbf{k}} - \epsilon_{-,\mathbf{k}})}\right| \times \\ & \quad |\langle \mathbf{k}, +|v^x|\mathbf{k}, -\rangle|^2\delta(\hbar\omega - (\epsilon_{+,\mathbf{k}} - \epsilon_{-,\mathbf{k}})) \, . \end{aligned} \tag{2.23}$$

Thus, the Kubo Formula relates the current fluctuations of $H_0$ to the dissipative part of the conductivity $Re\sigma$. This last equation is quite similar to equation **??**. In fact, using Fermi's Golden Rule with the perturbation $H_1(t) = -\mathbf{j}\cdot\mathbf{A}(t)$ we would obtain a similar result for $\eta$.

# 3

# Massive Dirac fermions

Graphene has a honeycomb lattice structure made out of carbon atoms. The first Brillouin zone of graphene is an hexagon. Only two corners of it can not be connected by primitive vectors of the reciprobal lattice, i.e. this two points are inequivalent and are called $K$ and $K'$ points. Near these points, the electrons behaves like massless Dirac fermions $H_0 = v_F \boldsymbol{\sigma} \cdot \mathbf{p}$. This is why the $K$ and $K'$ are also called Dirac points. Including a mass term in the massless Dirac fermions Hamiltonian we have

$$H_0 = v_F \boldsymbol{\sigma} \cdot \mathbf{p} + \tau m v_F^2 \sigma^z = \begin{pmatrix} \tau m v_F^2 & \hbar v_F (k_x - i k_y) \\ \hbar v_F (k_x + i k_y) & -\tau m v_F^2 \end{pmatrix} , \tag{3.1}$$

where $\tau = \pm$ denotes the two $K$-points. Particles who follow this Hamiltonian are called *massive Dirac fermions*. For time-reversal symmetric systems, it is sufficient to only consider one valley ($\tau = +$) and include the other by the valley degeneracy factor $g_v = 2$. Since there is no coupling with the spin component, we will introduce also a degeneracy factor $g_s = 2$ in our further calculations. Such a system has the following eigenenergies

$$\epsilon_{\lambda,\mathbf{k}} = \lambda \sqrt{\hbar^2 v_F^2 (k_x^2 + k_y^2) + (m v_F^2)^2} , \tag{3.2}$$

where $\lambda$ is the band index taking on the value $\lambda = 1$ for the conduction band and $\lambda = -1$ for the valence band. The normalized wave functions read

$$\psi_{+,\mathbf{k}}(\mathbf{r}) = \frac{1}{\sqrt{A}} \begin{pmatrix} \cos(\theta/2) \\ \sin(\theta/2) e^{i\varphi} \end{pmatrix} e^{i\mathbf{k}\cdot\mathbf{r}} , \tag{3.3}$$

$$\psi_{-,\mathbf{k}}(\mathbf{r}) = \frac{1}{\sqrt{A}} \begin{pmatrix} \sin(\theta/2) \\ -\cos(\theta/2) e^{i\varphi} \end{pmatrix} e^{i\mathbf{k}\cdot\mathbf{r}} , \tag{3.4}$$

with

$$\cos(\theta) = \frac{2 m v_F^2}{\epsilon_{+,\mathbf{k}} - \epsilon_{-,\mathbf{k}}} \; ; \; e^{i\varphi} = \frac{k_x + i k_y}{k} . \tag{3.5}$$

To study the absorption of photons by electrons of massive Dirac fermions valence band we consider a light wave linearly polarized with electric field $\mathbf{E}(t)$ and frequency $\omega$, propagating along the $x$ axis with normal incidence through a graphene sheet of area $A$. Such absorption process would promote an electron from the valence band to the conduction one with a gain of energy of $\epsilon_{+,\mathbf{k}} - \epsilon_{-,\mathbf{k}} = \hbar\omega$.

Considering minimal coupling: $\mathbf{p} \to \mathbf{p} + e\mathbf{A}(t)$ with $e > 0$ gives the time-dependent perturbation $H_1(t) = ev_F \boldsymbol{\sigma} \cdot \mathbf{A}(t)$ and with $\mathbf{E}(t) = -\partial_t \mathbf{A}(t) = \mathbf{E}\cos(\omega t)$ we finally have

$$H_1(t) = \frac{ev_F}{i\omega} \boldsymbol{\sigma} \cdot \mathbf{E} \cos(\omega t) \ . \tag{3.6}$$

Using Fermi's Golden rule (**??**) we obtain

$$\begin{aligned} \eta = \frac{g_s g_v e^2 |\mathbf{E}|^2}{32\omega^2\hbar^3} \int \left( (\epsilon_{+,\mathbf{k}} - \epsilon_{-,\mathbf{k}}) + \frac{(2mv_F^2)^2}{\epsilon_{+,\mathbf{k}} - \epsilon_{-,\mathbf{k}}} \right) \delta(\epsilon_{+,\mathbf{k}} - \epsilon_{-,\mathbf{k}} - \hbar\omega) d(\epsilon_{+,\mathbf{k}} - \epsilon_{-,\mathbf{k}}) = \\ = \frac{e^2|\mathbf{E}|^2}{2\omega^2\hbar^3} \left( \frac{\hbar\omega}{4} + \frac{(mv_F^2)^2}{\hbar\omega} \right) \times \Theta(\hbar\omega - 2mv_F^2) \ , \end{aligned} \tag{3.7}$$

where we have considered the valley- and spin- degeneracy factors $g_v = g_s = 2$ .
The absorption is then given by

$$A(\omega) = \frac{W_a}{W_i} = \pi\alpha \left( 1 + \left( \frac{2mv_F^2}{\hbar\omega} \right)^2 \right) \times \Theta(\hbar\omega - 2mv_F^2) \ . \tag{3.8}$$

Let's do the same but by means of the Kubo's formula. Giving the Hamiltonian **??** the velocity is

$$v^x = \frac{\partial H}{\partial p^x} = v_F \sigma^x \ . \tag{3.9}$$

Taking this velocity, equation **??** yields

$$\begin{aligned} \mathrm{Re}\, \sigma^{xx} &= \frac{g_s g_v \sigma_0}{\omega\pi} \iint d\varphi d(\epsilon_{+,\mathbf{k}} - \epsilon_{-,\mathbf{k}}) \frac{1}{(\hbar v_F)^2} \times \\ &\quad |\langle \mathbf{k}, + | v_F \sigma^x | \mathbf{k}, - \rangle|^2 \delta(\hbar\omega - (\epsilon_{+,\mathbf{k}} - \epsilon_{-,\mathbf{k}})) \\ &= \sigma_0 \left( 1 + \left( \frac{2mv_F^2}{\hbar\omega} \right)^2 \right) \times \Theta(\hbar\omega - 2mv_F^2) \ . \end{aligned} \tag{3.10}$$

Using equations **??** and **??** and considering both media to be vacuum we obtain the absorption of massive Dirac fermions

$$A(\omega) = \left[ 1 - \frac{4\varepsilon_0^2}{\left| 2\varepsilon_0 + \frac{\sigma_0}{c} \left( 1 + \left( \frac{2mv_F^2}{\hbar\omega} \right)^2 \right) \right|^2} \right] \times \Theta(\hbar\omega - 2mv_F^2) \tag{3.11}$$

$$\simeq \pi\alpha \left( 1 + \left( \frac{2mv_F^2}{\hbar\omega} \right)^2 \right) \times \Theta(\hbar\omega - 2mv_F^2) \ . \tag{3.12}$$

Both results agree, as it must. We see that for massive Dirac fermions the absorption has a dependence on the frequency $\omega$ and on parameters of the material: $m$ and $v_F$. We have universality only for $\omega = 2mv_F^2/\hbar$, and the absorption in such a case is $A = 2\pi\alpha$. From this frequencies the absorption decays up to reaching universality again with $A = \pi\alpha$, when the mass term $(2mv_F^2/\hbar\omega)$ is negligible. Thus, we can define a crossover region where a dependence on the frequency shows up. Such behavior is expected to be seen experimentally in gapped topological insulators, which are massive Dirac fermions systems with $g_v = 1$ (since they present only one Dirac cone) and $g_s = 1$ (due to the strong spin-orbit coupling [**?**]).

# 4

# 2D Semiconductors

We will now consider a general (Galilei invariant) semiconductor with $H_0 = \frac{\mathbf{p}^2}{2m_0} + V(\mathbf{r})$ where $\mathbf{p} = -i\hbar\partial_{\mathbf{r}}$ and the periodic potential $V(\mathbf{r}) = V(\mathbf{r} + \mathbf{a}_j)$ along the lattice vector $\mathbf{a}_j$. From Bloch's theorem we can write the wave function as $\phi_{\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}}u_{n\mathbf{k}}(\mathbf{r})$ with $\mathbf{k}$ denoting the Bloch wave vector. The effective Hamiltonian for the periodic function $u_{n\mathbf{k}}(\mathbf{r}) = u_{n\mathbf{k}}(\mathbf{r} + \mathbf{a}_j)$ is thus given by

$$H_{\mathbf{k}\cdot\mathbf{p}}(\mathbf{k}) = H_0 + \frac{\hbar}{m_0}\mathbf{k}\cdot\mathbf{p} + \frac{\hbar^2k^2}{2m_0}\ . \tag{4.1}$$

This Hamiltonian shall be represented within a minimal basis set consisting of $|s\rangle$ for the conduction band and $|p_j\rangle$ for the valence band with $j = x, y, z$ which correspond to the Bloch function $u_{n\mathbf{k}}$ at $\mathbf{k} = 0$. With $\epsilon_0(k) = \frac{\hbar^2k^2}{2m_0}$ and $\langle s|\mathbf{p}|p_j\rangle \equiv i\frac{m_0}{\hbar}P$, we thus can write the Hamiltonian in matrix terms as follows

$$H_{\mathbf{k}\cdot\mathbf{p}}(\mathbf{k}) = \begin{pmatrix} \epsilon_c + \epsilon_0(k) & iPk_x & iPk_y & iPk_z \\ -iPk_x & \epsilon_v + \epsilon_0(k) & 0 & 0 \\ -iPk_y & 0 & \epsilon_v + \epsilon_0(k) & 0 \\ -iPk_z & 0 & 0 & \epsilon_v + \epsilon_0(k) \end{pmatrix}\ . \tag{4.2}$$

The valence band thus splits into a light hole with energy $\epsilon_{lh}(k) = \frac{1}{2}(\epsilon_c + \epsilon_v) + \epsilon_0(k) - \sqrt{E_g^2/4 + P^2k^2}$ and a doubly degenerated heavy hole with energy $\epsilon_{hh} = \epsilon_v + \epsilon_0(k)$ where $E_g = \epsilon_c - \epsilon_v$. The energy of the conduction band is renormalized to $\epsilon_e(k) = \frac{1}{2}(\epsilon_c + \epsilon_v) + \epsilon_0(k) + \sqrt{E_g^2/4 + P^2k^2}$.

Let us neglect the degenerate heavy hole band and approximate the other two bands for small values of $\mathbf{k}$. This yields

$$\epsilon_e(k) = \epsilon_c + \frac{\hbar^2k^2}{2m_0m_e}\ ;\ \epsilon_{lh}(k) = \epsilon_v - \frac{\hbar^2k^2}{2m_0m_{lh}}\ , \tag{4.3}$$

with the effective (dimensionless) masses $m_e^{-1} = E_P/E_g + 1$ and $m_{lh}^{-1} = E_P/E_g - 1$ where $E_P = 2m_0P^2/\hbar^2$. The reduced mass is thus given by $m_e^{-1} + m_{lh}^{-1} = 2E_P/E_g$ which is the crucial relation in order to obtain an universal optical conductivity for a 2D semiconductor.

For the optical conductivity or absorption, we need to evaluate the matrix element $\langle\mathbf{k}, c|\mathbf{e}_0\cdot\mathbf{p}|\mathbf{k}', v\rangle$ where we only consider transitions from the valence ($v$) to the conduction ($c$) band and $\mathbf{k} = k\cdot\mathbf{e}_0$. Using the above model, the full wave function is the product of the envelope function with the Bloch function at $\mathbf{k} = 0$, $\psi_{\mathbf{k}}(\mathbf{r}) \propto \chi_{\mathbf{k}}(\mathbf{r})u_{\mathbf{k}=0}(\mathbf{r})$.

For a quantum well, the envelope function can be written as $\chi_n(\mathbf{r}) = A^{-1/2}e^{i\mathbf{k}\cdot\mathbf{r}}\phi_n(z)$. For these systems the matrix element reads

$$\begin{aligned}\langle \mathbf{k}, c, n|\mathbf{e}_0\cdot\mathbf{p}|\mathbf{k}', v, m\rangle &\approx \sum_j \chi^*_{cn\mathbf{k}}(\mathbf{r}_j)\chi_{vm\mathbf{k}'}(\mathbf{r}_j)\int_{\text{unit cell}} u^*_{cn}(\mathbf{r})(\mathbf{e}_0\cdot\mathbf{p})u_{vm}(\mathbf{r})d^3r \\ &\approx \mathbf{e}_0\cdot\mathbf{p}_{cn,vm}(0)\delta_{\mathbf{k},\mathbf{k}'}\int \phi^*_{cn}(z)\phi_{vm}(z)dz \equiv p_{cn,vm}\delta_{\mathbf{k},\mathbf{k}'}\langle c, n|v, m\rangle\end{aligned} \tag{4.4}$$

To study the absorption let us consider a semiconductor with $H_0 = \frac{\mathbf{p}^2}{2m_0} + V(\mathbf{r})$. Peierls substitution $\mathbf{p}\to\mathbf{p}+e\mathbf{A}(t)$ then leads to a paramagnetic perturbation $H_{par} = \mathbf{A}\cdot\mathbf{p}/m_0$ as well as to a diamagnetic perturbation $H_{dia} = \mathbf{A}^2/2m_0$. The contribution of the diamagnetic term does not contribute at finite frequencies. Again, we parameterize the gauge potential as $\mathbf{E}(t) = -\partial_t\mathbf{A} = \mathbf{E}_0\cos(\omega t)$ and the time-dependent perturbation thus reads $V(t) = \frac{eE_0}{im_0\omega}\mathbf{p}\cdot\mathbf{e}_0\cos(\omega t)$ where we defined $\mathbf{E}_0 = \mathbf{e}_0E_0$. If we only consider transitions from the conduction band $\epsilon_c(\mathbf{k})$ to the valence band $\epsilon_v(\mathbf{k})$, the absorption obtained from Fermi's Golden rule is thus obtained as

$$\frac{W_a}{E_0^2} = \frac{\pi}{2}\frac{e^2}{m_0^2\omega}\frac{g_sg_v}{V}\sum_{\mathbf{k}}|\langle\mathbf{k}, c|\mathbf{e}_0\cdot\mathbf{p}|\mathbf{k}, v\rangle|^2\,\delta(\epsilon_c(\mathbf{k}) - \epsilon_v(\mathbf{k}) - \hbar\omega)\,, \tag{4.5}$$

where we will set $g_s = 2$ and $g_v = 1$ in the following.

Alternatively, we can consider the real part of the conductivity of a 2D semiconductor which is given by

$$\operatorname{Re}\sigma(\omega) \approx \frac{\pi e^2}{m_0^2L\omega}|p_{cn,vm}|^2|\langle c, n|v, m\rangle|^2\frac{2}{A}\sum_{\mathbf{k}}\delta(\epsilon_{c,n}(\mathbf{k}) - \epsilon_{v,m}(\mathbf{k}) - \hbar\omega) \tag{4.6}$$

where the energy bands for small $\mathbf{k}$ can be approximated by $\epsilon_{b,n}(k) = \epsilon_b \pm \epsilon_{b,n} \pm \frac{\hbar^2k^2}{2m_0m_{b,n}}$ and the upper and lower sign stands for the conduction ($b = c$) and valence ($b = v$), respectively. With the joint density-of-states

$$\frac{2}{A}\sum_{\mathbf{k}}\delta(\epsilon_{c,n}(\mathbf{k}) - \epsilon_{v,m}(\mathbf{k}) - \hbar\omega) = \frac{2m_0m_{nm}}{\pi\hbar^2}\Theta\left[\hbar\omega - (E_g + \epsilon_{c,n} + \epsilon_{v,n})\right] \tag{4.7}$$

where $m^{-1}_{nm} = m^{-1}_{c,n} + m^{-1}_{v,m}$ and $E_g = \epsilon_c - \epsilon_v$ the energy gap, the absorption shows a step-like behavior as function of the photon energy as more and more transitions from different sub-bands are involved. The height of these steps is quasi-universal if we assume $p_{cn,vm} = im_0P/\hbar$, $\langle c, n|v, m\rangle \approx 1$ and $\hbar\omega \approx E_g$:

$$\Delta\operatorname{Re}\sigma = \frac{e^2}{4\hbar L} \equiv \frac{2\sigma_0}{L} \tag{4.8}$$

Apart from the geometrical factor $L$, this is the same result as for graphene, but note that transitions at the Γ-point, i.e., one valley with $g_v = 1$, are involved. Defining the valley- and spin-dependent universal conductivity as $\sigma_0 = \frac{g_sg_v}{16}\frac{e^2}{\hbar}$, we have twice the absorption of graphene.

This universal absorption has recently been measured [**?**] for thin films of InAs. In the next chapter, we will theoretically discuss the absorption of a truely two-dimensional semiconductor crystal ($\mathrm{MoS_2}$) and analyze whether this universality still holds.

# 5

# Molybdenum disulfide

A monolayer of molybdenum disulfide [**?**, **?**, **?**], as well as graphene, has a honeycomb lattice made of molybdenum and sulfur atoms instead of carbon. Such a two-dimensional system, contrary to graphene, presents a large band gap separating valence and conduction band. A monolayer of $\mathrm{MoS_2}$ around the corners of the Brillouin zone can be described by the effective two-band model for both spin ($s \pm 1$) and valley ($\tau \pm 1$) components

$$H_0^{\tau s} = \left(\frac{\Delta}{2} + \frac{\hbar^2\beta}{4m_0}k^2\right)\sigma_z + \frac{\hbar^2\alpha}{4m_0}k^2 + \tau s\lambda\frac{1-\sigma_z}{2} + t_0a_0\mathbf{k}\cdot\boldsymbol{\sigma}_\tau \ , \tag{5.1}$$

with $\boldsymbol{\sigma}_\tau = (\tau\sigma_x, \sigma_y)$; $m_0$ as the free electron mass and the following values for the parameters

$$\Delta = 1.9\ \mathrm{eV}\ ;\ t_0 = 1.68\ \mathrm{eV}\ ;\ a_0 = 1.84\ \text{Å}\ ;\ \alpha = 0.43\ ;\ \beta = 2.21\ ;\ \lambda = 0.08\ \mathrm{eV}\ . \tag{5.2}$$

Such a system has the following eigenenergies

$$\epsilon_\pm = \frac{\hbar^2\alpha}{4m_0}k^2 + \frac{\tau s\lambda}{2} \pm \sqrt{(t_0a_0k)^2 + \left(\frac{\Delta}{2} + \frac{\hbar^2\beta k^2}{4m_0} - \frac{\tau s\lambda}{2}\right)^2}\ . \tag{5.3}$$

We will use the same eigenvectors as for massive Dirac fermions, but in this case the angle $\theta$ is defined by

$$\cos(\theta) = \frac{\Delta + \frac{\hbar^2\beta}{2m_0}k^2 - \tau s\lambda}{\epsilon_+ - \epsilon_-}\ . \tag{5.4}$$

Introducing the effect of an electromagnetic field with minimal coupling: $\mathbf{p} \to \mathbf{p} + e\mathbf{A} = \mathbf{p} + \frac{e\mathbf{E}}{i\omega}$ and with a light wave linearly polarized with electric field propagating along the $x$ axis with normal incidence we have for the time-dependent perturbation

$$H_1^\tau = \frac{\beta e|\mathbf{E}|}{4m_0\omega}\left(\frac{2\hbar}{i}k\right)\sigma_z + \frac{t_0a_0e|\mathbf{E}|\tau}{i\hbar\omega}\sigma_x + \frac{\alpha e|\mathbf{E}|}{4m_0\omega}\left(\frac{2\hbar}{i}k\right)\ , \tag{5.5}$$

where we have neglected the diamagnetic term as it is quadratic in $\mathbf{E}$.

The band structure near $k = 0$ is plotted in figure **??** where we distinguish the difference bands according to the two possible values of $\tau \cdot s = \pm 1$. It corresponds to a direct semiconductor with gap $\Delta - \lambda = 1.82$ eV. We have plotted with narrows the two possible interband transitions for a given valley. The red narrow corresponds to a transition from the valence band with $\tau \cdot s = -1$, with an energy gain of $\epsilon = 2\sqrt{(t_0 a_0 k)^2 + \left(\frac{\Delta}{2} + \frac{\hbar^2 \beta k^2}{4m_0} + \frac{\lambda}{2}\right)^2}$, and the black narrow to a transition from the $\tau \cdot s = 1$ valence band with an energy band-width of $2\sqrt{(t_0 a_0 k)^2 + \left(\frac{\Delta}{2} + \frac{\hbar^2 \beta k^2}{4m_0} - \frac{\lambda}{2}\right)^2}$.

In figure **??** we represented the total absorption of MoS$_2$ which can be written down as follows

$$
\begin{aligned}
A(\omega) &= \pi\alpha \frac{1}{\sqrt{\gamma^-}\hbar\omega} \left\{ 2a_0^2 t_0^2 \left[ \hbar\omega + \frac{16 m_0^2 \left(a_0^2 t_0^2 - \sqrt{\gamma^-}/4\right)^2}{\beta^2 \hbar^4 \hbar\omega} \right] \right. \\
&+ \left. \left(-4a_0^2 t_0^2 - \beta(\Delta - \lambda)\hbar^2/m_0 + \sqrt{\gamma^-}\right) \left[ \hbar\omega - \frac{16 m_0^2 \left(a_0^2 t_0^2 - \sqrt{\gamma^-}/4\right)^2}{\beta^2 \hbar^4 \hbar\omega} \right] \right\} \\
&\times \Theta[\hbar\omega - (\Delta - \lambda)] \\
&+ \pi\alpha \frac{1}{\sqrt{\gamma^+}\hbar\omega} \left\{ 2a_0^2 t_0^2 \left[ \hbar\omega + \frac{16 m_0^2 \left(a_0^2 t_0^2 - \sqrt{\gamma^+}/4\right)^2}{\beta^2 \hbar^4 \hbar\omega} \right] \right. \\
&+ \left. \left(-4a_0^2 t_0^2 - \beta(\Delta + \lambda)\hbar^2/m_0 + \sqrt{\gamma^+}\right) \left[ \hbar\omega - \frac{16 m_0^2 \left(a_0^2 t_0^2 - \sqrt{\gamma^+}/4\right)^2}{\beta^2 \hbar^4 \hbar\omega} \right] \right\} \\
&\times \Theta[\hbar\omega - (\Delta + \lambda)] \; ,
\end{aligned}
\tag{5.6}
$$

with

$$
\gamma^{\mp} \equiv 16 a_0^4 t_0^4 + 8 a_0^2 t_0^2 \beta \hbar^2 (\Delta \mp \lambda)/m_0 + \beta^2 \hbar^4 (\hbar\omega)^2/m_0^2 \; . \tag{5.7}
$$

We have normalized the absorption by $A_0 = \pi\alpha$, and represented for the range of energies [0-4 eV], together with the absorption of a massive Dirac fermions system with $\Delta = 2mv_F^2 = 1.9\,\text{eV}$ and degeneracy factors $g_s = g_v = 2$. We included two insets: on the top for $\hbar\omega \in [0 - 40 \text{ eV}]$, and on the bottom for the high-frequency limit. We can observe a dependence on the frequency for the absorption of both systems. We also see that the value $\pi\alpha$ and $2\pi\alpha$ is not exactly reached close to the band edge. We attribute this to renormalization effects due to the spin-orbit interaction similar to what is seen for other interactions such as the the electron-phonon interaction in graphene [**?**].

Let us discuss the high-frequency limit. Taking $\omega \to \infty$ in equation **??** we can realize that a universal absorption is recovered in massive Dirac fermions:

$$
A^{\text{MDF}}(\omega \to \infty) = \pi\alpha \; . \tag{5.8}
$$

On the other hand, doing the same for $MoS_2$,

$$
A^{\text{MoS}_2}(\omega \to \infty) = 0 \; , \tag{5.9}
$$

as we expected since for this regime the perturbation $H_1$ is dominated by the terms quadratic in $k$, i.e. the electrons behave like Schrödinger fermions, and for symmetry reasons, as we discussed in chapter **??**, the matrix element $|\langle \mathbf{k}, + | H_1 | \mathbf{k}, - \rangle$ vanishes.

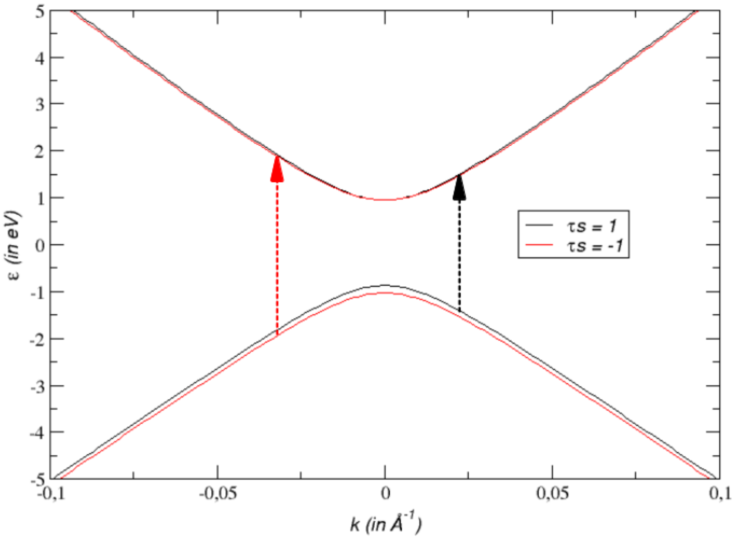

Figure 5.1: Band structure of 2D MoS$_2$ near $k = 0$ and allowed interband transitions from the valence band to the conduction band, present in the absorption process.

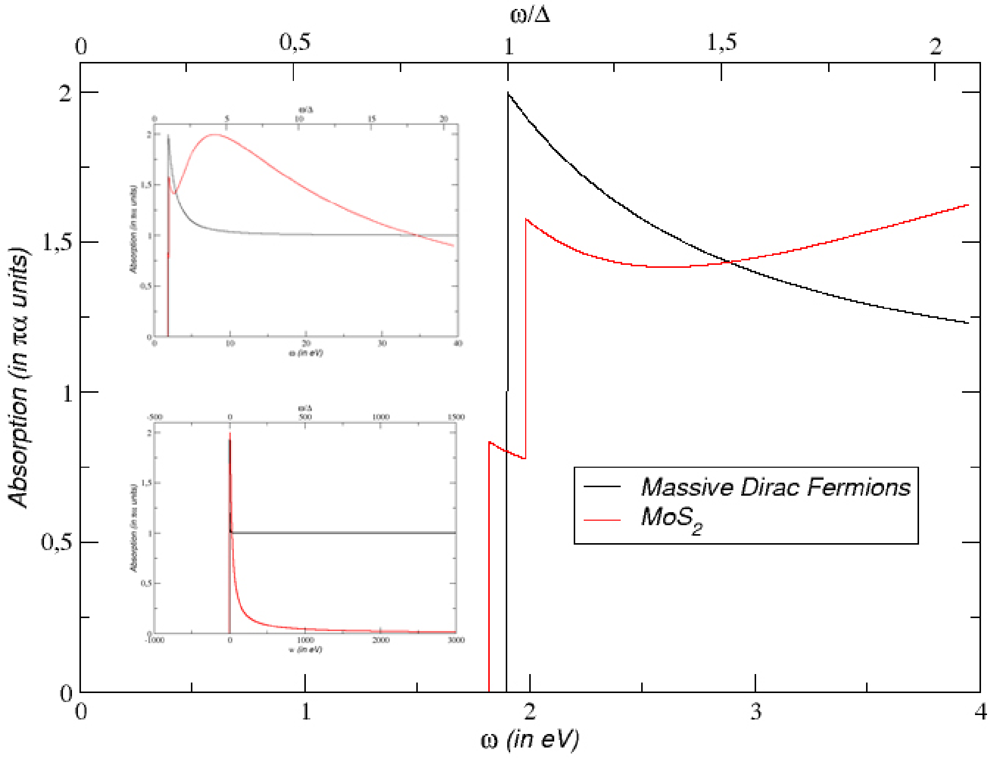

Figure 5.2: Normalized absorption as a function of the energy provided by an electromagnetic wave with normal incidence through a sheet of a massive Dirac fermions system (black) and MoS$_2$ (red). The above inset shows the absorption for $\hbar\omega \in [0 - 40]$ eV. Below is plotted for the high-frequency limit.

# 6

# Summary

In this master thesis, we have investigated the absorption coefficient of three systems: massive Dirac fermions, ordinary 2D semiconductors and a truely 2D semiconductor crystal, $MoS_2$, with two different approaches: Fermi's Golden Rule and Linear Response. The results showed an universal absorption for 2D semiconductors within the effective-mass approximation, in accord with recent experimental results [**?**], and with studies of the absorption in graphene, which remains universal even beyond the Dirac approximation [**?**].

But this universality is not obtained for massive Dirac fermions, where we got an explicit dependence on parameters of the material. In fact, the maximum in the absorption for massive graphene, a massive Dirac fermions system, was found to be twice the 2D semiconductors one. This discrepancy is due to the difference in the valley-degeneracy factor, which is 2 for massive graphene and 1 for 2D semiconductors in the $\mathbf{k} \cdot \mathbf{p}$ approximation.

Finally we study the absorption in a real 2D semiconductor, $MoS_2$, whose Hamiltonian is well-known [**?**]. We observed a similar behavior with respect to massive Dirac fermions, with a dependence of the absorption in $\omega$.

We conclude that the universality of the absorption is observed within a short range of energies close to the band edge, compatible with the limitations of the $\mathbf{k} \cdot \mathbf{p}$ perturbation theory, and also for high frequencies in the case of massive Dirac fermions, where it is reduced to a half part. A crossover region exhibits a dependence between the absorption and the frequency, as we observed in our study of massive Dirac fermions and $MoS_2$. This might be observable in 3D-topological insulators such as $Bi_2Se_3$ where a small gap is produced by a time-reversal symmetry breaking magnetic field [**?**].

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