The tuning of light-matter coupling and dichroism in graphene for enhanced absorption: Implications for graphene-based optical absorption devices

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Abstract
The inter-band optical absorption in graphene characterized by its fine-structure constant has a universal value of 2.3% independent of the material parameters. However, for several graphene-based photonic applications, enhanced optical absorption is highly desired. In this work, we quantify the tunability of optical absorption in graphene via the Fermi level, angle of incidence of the incident polarized light, and the dielectric constants of the surrounding dielectric media in which graphene is embedded. The influence of impurities adsorbed on the surface of graphene on the Lorentzian broadening of the spectral function of the density of states is analytically evaluated within the equilibrium Green’s function formalism. In all the cases, we find that absorption of light graphene embedded in dielectric medium is significantly higher than 2.3%. We also compute the differential absorption of right and left circularly-polarized light in graphene that is uniaxially and optically strained. The preferential absorption or circular dichroism is investigated for armchair and zigzag strain and the interplay of \( k \)-space and velocity anisotropy is examined. Finally, we relate circular dichroism to the Berry curvature of gapped graphene and explain the connection to parameters that define the underlying Hamiltonian.

Keywords: Green’s function, circular dichroism, Berry curvature, strained graphene, Dirac fermions

(Some figures may appear in colour only in the online journal)
with a description of the optical characteristics of a graphene sheet suspended in vacuum and derive the quantum of light absorption for a beam at oblique incidence (section 2.1). The calculations are repeated for a graphene sheet sandwiched between two dielectrics; the dielectric function of graphene in each case is established using a standard RPA [16] calculation. Significantly, we find that the absorption which for a sheet suspended in vacuum is a frequency-independent 2.3% shows a marked improvement as the ratio of the dielectric constants surrounding the graphene sheet is altered thus potentially offering a simple technique for enhanced light-matter interaction. Note that the standard absorption of 2.3% is not only frequency-independent, it also exhibits no functional relationship to angle of incidence. Therefore, by embedding graphene in optical media with varying dielectric constants, it is now possible to utilize the angle of incidence as a tunable parameter in studying light absorption. Further, while most calculations tacitly assume an idealized set-up in which graphene is pristine preserving its unique electronic structure, especially the linearly dispersing bands around the $K$ and $K'$ edges of the Brillouin zone, impurity atoms are usually adsorbed on the surface [17, 18] to modify the overall energy spectrum and its attendant optical response. The correction in this situation is manifest through a spectral broadening of the density of states and a changed light absorption profile. The broadening for an impurity-adsorbed graphene sheet is evaluated by employing a non-interacting Anderson-type Hamiltonian within the equilibrium Green’s function formalism (section 2.2). Here again, we observe that controlling the density of impurities adsorbed on the surface makes graphene more suited to light absorption; in particular, when the Fermi level is set close to the charge neutral point and at normal incidence, the absorption shows a sharp increase (apprx. 70%) compared to pristine graphene surrounded by dielectric constants that maintain an identical ratio in each case. We also find that an uptick in adsorbed impurity density on graphene surface leads to a degradation in absorption thus enforcing a cut-off. In the course of our study, we offer several paths for an experimentalist to modulate light absorption thus enforcing a cut-off. In the course of our study, we offer several paths for an experimentalist to modulate light absorption thus enforcing a cut-off. In the course of our study, we offer several paths for an experimentalist to modulate light absorption thus enforcing a cut-off. In the course of our study, we offer several paths for an experimentalist to modulate light absorption thus enforcing a cut-off.

Until now, we have focused exclusively on light absorption without accounting for any polarization that may exist in the incoming beam. In section 2.3 we turn our attention to explore ways to selectively absorb light of a given polarization. While detailed and accurate analyses of valley-dependent light absorption in graphene exist, particularly as presented in [19], we seek to utilize these fundamental principles relying on band topology to study experimentally feasible ways that may lead to improved polarization-dependent absorption. Since graphene is semi-metallic and symmetric about the linearly dispersing bands at the $K$ and $K'$ valley edges of the Brillouin zone, we employ optical strain (by coupling graphene to a substrate) to primarily break the inversion symmetry which in turn opens a band gap and allows for increased light absorption. A more comprehensive analysis is however carried out next by perturbing the electron–hole ($el$-$h$) symmetry of graphene through uniaxial strain. By utilizing strain-based scaling relationships that proportionately alter the hopping parameter in the tight-binding Hamiltonian for graphene, we note that the loss of $el$-$h$ symmetry tilts the Dirac cones to give anisotropic Fermi velocity components along the $x$- and $y$-axes. When uniaxial and optical strain in graphene are acting in tandem, our calculations for circular dichroism—a quantity that measures absorption preference for right- or left-circularly polarized light further show an intricate interplay between Fermi velocity and momentum-space anisotropy. Interestingly, this interplay reveals itself differently for armchair and zigzag strain. As we remarked before, circular dichroism in graphene arises because of the peculiar topology of the bands and a distinct handedness of the Bloch electrons at high-symmetry $K$ and $K'$ points. The handedness which is characterized by the Berry curvature is established in the final part of our calculations and a direct connection is drawn to circular dichroism.

We remind the reader throughout how circular dichroism is related to the finite Berry curvature of gapped graphene and is amenable to changes to the parameters that characterize the defining Hamiltonian. Numerical results are collected in section 3 and the paper concludes by briefly touching upon the implications of increased absorption for graphene-based optical devices. For ease of notation, we declare that any reference to graphene in this work refers to mono-layer graphene. We do not consider multi-layered graphene structures.

2. Optical absorption in suspended graphene

Dispersion relationships for graphene bands with linearly dispersing eigen states [20] are modeled using a two-dimensional Dirac Hamiltonian given as

$$\begin{align*}
\mathcal{H} = \hbar v_{F}(\sigma_{y}k_{x} - \sigma_{x}k_{y}) + \Delta \sigma_{z},
\end{align*}$$

where $v_{F}$ denotes the Fermi velocity of carriers, $\tilde{k}$ is the wave-vector measured relative to the Dirac points, and $\sigma_{i} (i = x, y, z)$ are the usual Pauli matrices. The corresponding wave functions, in momentum space, for the momentum around the Dirac point have the form:

$$\begin{align*}
\Psi_{\eta} = \left[ \frac{1}{\sqrt{2}} u_{\eta}(k) \exp(-i\Theta) \right],
\end{align*}$$

where $\eta = \pm$, and $u_{\eta}$ is given by

$$\begin{align*}
u_{\eta}(k) = \frac{1 \pm \frac{\Delta}{\sqrt{\Delta^2 + (\hbar v_{F}k)^2}}}{\sqrt{\Delta^2 + (\hbar v_{F}k)^2}}.
\end{align*}$$

Here $\Delta$ is the band gap induced in graphene, $\Theta = \arctan\left(\frac{k_{y}}{k_{x}}\right)$, and $\eta = \pm$ corresponds to conduction and valence bands. The band gap introduced is primarily through an interaction with the substrate on which graphene is epitaxially grown, for instance, the honeycomb lattice rigidly held on the hexagonal BN [17, 21] exhibits a band gap between 7.0 meV to 20.0 meV [22]. The band gap of pristine graphene, which hosts massless Dirac fermions, is zero.
relationship in graphene and integrating over energy space gives the absorbed flux (see appendix A) as \( \frac{\pi}{\epsilon_0} \frac{\omega^2}{k_0^2} \). The incident flux \([24]\) for an electric field is \( \frac{\pi}{4\epsilon_0} |E|^2 \). The optical interband absorption in graphene is, therefore, universally given as \( \frac{\pi}{k_0^2} = 2.3\% \), a material-independent number.

The optical absorption changes when graphene is sandwiched between two dielectric layers characterized by dielectric constants of \( \epsilon_1 \) and \( \epsilon_2 \). A transverse-magnetic [25] (TM or \( \rho \)-polarized wave) with a magnetic field along the \( y \)-axis (equation (6)) is assumed to impinge on the graphene-dielectric interface. The magnetic field corresponding to the TM polarization and propagating along the \( x \)-axis is given as

\[
H_{\perp} = (A_i e^{-ik_0z} + B_i e^{ik_0z}) e^{ik_0x},
\]

where \( k_z = k_{in} \cos \theta \) and \( k_x = k_{in} \sin \theta \). A particular physical quantity within the \( i \)th layer in the structure is identified by the subscript \( i \), \( k_{in} \) is incident wave vector, and the angle of incidence is \( \theta \). Notice that \( k_{in} \) in figure 1 is the incident wave vector \( k_{in} \). The appropriate boundary conditions at the interface \( z = 0 \) are

\[
\hat{r} \times (\vec{E}_i - \vec{E}_f) = 0,
\]
and

\[
\hat{r} \times (\vec{H}_i - \vec{H}_f) = \vec{J}_{surf},
\]

where \( \vec{J}_{surf} \) is the surface current due to the 2D charge carriers in the graphene sheet. The subscripts \( i,j \) denote a particular material layer on the left and the right side of a given interface, respectively. Note that while our analysis can be extended to multi-layered films of dielectrics stacked together, we restrict ourselves to just two layers. The subscripts \( i,j \) therefore assume values 1 and 2, respectively. The electric field can be obtained using Maxwell’s equation \( \nabla \times \vec{E} = -\frac{i}{\omega \epsilon_0} \frac{\partial \vec{B}}{\partial t} \) to yield \( \vec{E}_f = -i \frac{\omega}{\epsilon_0 c} \nabla \times \vec{H} \). For the form of the magnetic field chosen in equation (6), the corresponding electric field is given as

\[
E_{e,i} = \frac{h}{\omega \epsilon_0 \sigma \epsilon_1} (A_i e^{-ik_0z} + B_i e^{ik_0z}) e^{ik_0x},
\]
\[
E_{e,j} = \frac{h}{\omega \epsilon_0 \sigma \epsilon_2} (A_i e^{-ik_0z} - B_i e^{ik_0z}) e^{ik_0x}.
\]

Combining the above equations with the boundary condition (equation (7)) for electrical field on either side of the graphene-dielectric interface gives

\[
\frac{k_{1z}}{\epsilon_1} (A_1 - B_1) - \frac{k_{2z}}{\epsilon_2} (A_2 - B_2) = 0.
\]

The second relation using boundary conditions applied on the magnetic field (equation (7a)) is

\[
J_x = (A_1 + B_1) - (A_2 + B_2),
\]
\[
= \frac{\sigma k_{1z} (A_2 - B_2)}{\epsilon_0 \epsilon_2},
\]
Note that in the above equation, \( \sigma \) is the complex dynamical conductivity of graphene as discussed in section 2.2. All quantities on left(right) of the interface are subscripted as ‘1(2)’. In matrix notation, the amplitude of the magnetic fields on either side of the interface are related as [26]

\[
\begin{pmatrix} A_1 \\ B_1 \end{pmatrix} = M_{mm} \begin{pmatrix} A_2 \\ B_2 \end{pmatrix}
\] (10a)

The transfer matrix \( M_{mm} = M_{1,mm} + M_{2,mm} \), where

\[
M_{1,mm} = \frac{1}{2} \begin{pmatrix} 1 + \frac{\epsilon_kz_2}{\epsilon_kz_1} & 1 - \frac{\epsilon_kz_2}{\epsilon_kz_1} \\ 1 - \frac{\epsilon_kz_2}{\epsilon_kz_1} & 1 + \frac{\epsilon_kz_2}{\epsilon_kz_1} \end{pmatrix}
\] (10b)

and

\[
M_{2,mm} = \frac{1}{2} \begin{pmatrix} \sigma_kz_2 & -\sigma_kz_2 \\ \epsilon_0 \omega \epsilon_kz_2 & \epsilon_0 \omega \epsilon_kz_2 \end{pmatrix}
\] (10c)

The above results in a slightly different form were also obtained by Zhan et al in [27]. The transfer matrix \( M_{mm} \) derived in equation (10) allows us to compute reflectance and transmittance amplitudes; in particular, reflectance and transmittance amplitudes are

\[
r = \frac{M_{mm}(2,1)}{M_{mm}(1,1)},
\]

\[
t = \frac{1}{M_{mm}(1,1)}.
\]

The reflection and transmission coefficients are \( R = r^2 \) and \( T = t^2 \) which add to unity \((R + T = 1)\) for zero absorption losses.

At this point it is imperative to discuss appropriate limits that are placed on the expressions for reflectance and transmittance amplitudes derived in equations (11) and (12). The amplitudes must satisfy \( 0 \leq r \leq 1 \) and \( 0 \leq t \leq 1 \) from which follows

\[
0 \leq r = \frac{1}{M_{mm}(1,1)} \leq 1.
\] (13a)

To evaluate the constraint, we note that the matrix \( M_{mm,2} \) which describes the optical conductivity of the graphene sheet makes a small contribution to the overall matrix \( M_{mm} \) in the high-frequency limit. We are, therefore, left with the inequality

\[
0 \leq t = \frac{2 \epsilon_kz_1}{\epsilon_kz_1 + \epsilon_kz_2} \leq 1.
\] (13b)

Since all quantities in the above equation are assumed to be positive, the left half side of the above inequality is trivially true; evaluating the right hand side inequality, one arrives at the relation \( \frac{2}{\epsilon_kz_1} \leq \frac{1}{\epsilon_kz_2} \). For a case which is in violation of this condition, unphysical solutions are obtained. We also demonstrate a parallel condition by placing a similar constraint on the reflectance amplitude. Proceeding as above, we write

\[
0 \leq r = \frac{M_{mm}(2,1)}{M_{mm}(1,1)} \leq 1
\] (13c)

Ignoring the contribution of \( M_{mm,2} \) in the high-frequency limit, the right inequality (equation (13b)) is trivially satisfied while the left takes the form

\[
r = \frac{\epsilon_kz_1 - \epsilon_kz_2}{\epsilon_kz_1 + \epsilon_kz_2}.
\] (13d)

It is straightforward to see that the condition \( r \geq 0 \) leads us to \( \frac{\epsilon_kz_1}{\epsilon_kz_2} \leq \frac{z_2}{z_1} \), which means the transmittance amplitude is greater than unity. This apparent contradiction is resolved by noting that in this limiting case \( \frac{\epsilon_kz_1}{\epsilon_kz_2} \equiv \frac{\epsilon_kz_1}{\epsilon_kz_2} \), the reflected component is zero and the incident beam is fully transmitted \((t = 1)\). We have thus arrived at an analogous condition for obtaining the corresponding Brewster’ angle for a graphene sheet embedded in inhomogeneous dielectric media. Of course, the limits on the validity of the ratio \( \epsilon_kz_1/\epsilon_kz_2 \) must also account for the finite contribution from the transfer matrix \( M_{2,mm} \) in the lower frequency regime.

The transfer matrix \( M_s \), for a TE [25] wave or s—polarized wave can be similarly derived using the appropriate Maxwell’s boundary conditions. The matrix \( M_s = M_{1,se} + M_{2,se} \) is

\[
M_{1,se} = \frac{1}{2} \begin{pmatrix} 1 - \frac{\mu_kz_2}{\mu_kz_1} & 1 - \frac{\mu_kz_2}{\mu_kz_1} \\ 1 - \frac{\mu_kz_2}{\mu_kz_1} & 1 + \frac{\mu_kz_2}{\mu_kz_1} \end{pmatrix},
\]

\[
M_{2,se} = \frac{1}{2} \begin{pmatrix} \mu_0 \mu_k \omega \sigma & \mu_0 \mu_k \omega \sigma \\ \mu_0 \mu_k \omega \sigma & -\mu_0 \mu_k \omega \sigma \end{pmatrix}
\] (14b)

The magnetic permeabilities \((\mu_k, \mu_k')\) are taken to be unity for non-magnetic dielectric media.

Before closing this section, we would like to underscore the microscopic picture that is at play in modulating the change in optical absorption. In the arrangement that we investigate, light traveling through medium ‘1’ with a known dielectric constant is incident on a single-layer of graphene before exiting through medium ‘2’. The electric field distribution at the interface is at the heart of the total absorption; to explain this, consider the ensemble of electrons on graphene surface that are displaced when light shines on it, the most pronounced effect of graphene is manifest when the surface conductivity is highest, a condition that corresponds to plasmon resonance. The surface conductivity and the special case of plasmon resonance is dependent on the electric field distribution which in turn is a function of the dielectric constants of the surrounding medium/layers, expressed quantitatively through the Maxwell boundary conditions.

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We must also be cognizant of the fact that while the dielectric constants in our work have been chosen to be purely real, in principle, they have an imaginary component that represents the out-of-phase atomic oscillations and cause the electromagnetic wave to lose energy. At a microscopic level, the dielectric layers therefore set up the field distribution pattern to determine (1) the surface conductivity of graphene and (2) propagating losses via the imaginary component of the dielectric constants. As a side remark, it is pertinent to point out that mono-layer graphene magnifies plasmons and the response is highly tunable allowing resonance under various conditions and an easy transfer of energy from the incident beam to the medium with fast response times. This in a way explains the surge of interest in graphene for photonic applications.

2.2. Optical absorption in impure graphene

Electromagnetic absorption in graphene is determined by its complex dynamical conductivity. The dynamical conductivity of graphene is obtained from the dielectric function $\varepsilon(q, \omega)$ through a random phase approximation (RPA). In the long-wavelength limit ($q \rightarrow 0$), the RPA dielectric function is given as [28–30]

$$
\varepsilon(q \rightarrow 0, \omega) = 1 - \frac{2\pi e^2}{q} \frac{q^2}{2\pi \hbar \omega} \left[ \frac{2E_{F}}{\hbar \omega} + \frac{1}{2} \ln \left[ \frac{2E_{F} - \hbar \omega}{2E_{F} + \hbar \omega} \right] \right],
$$

(15)

where $E_{F}$ is the Fermi level in the graphene sheet, and $\Theta(\cdot)$ is the Heaviside function. The dynamical conductivity, $\sigma(q, \omega)$, is related to the dielectric constant as

$$
\sigma(q, \omega) = \frac{i}{2\pi q} [1 - \varepsilon(q, \omega)].
$$

(16)

Inserting equation (15) in equation (16) gives

$$
\sigma(q \rightarrow 0, \omega) = \frac{E_{F}e^2}{\pi \hbar} \frac{i}{\hbar \omega + \frac{1}{4\hbar} + e^2/4\hbar} \Theta(\hbar \omega - 2E_{F}) + \frac{i e^2}{4 \pi \hbar} \ln \left[ \frac{2E_{F} - \hbar \omega}{2E_{F} + \hbar \omega} \right].
$$

(17)

The spectral width $\Gamma$ corresponds to a Lorentzian broadening of the density of states [31, 32] and is determined by the impurities adsorbed on the graphene surface. In this work, using a retarded Green’s function approach, we evaluate the spectral width, $\Gamma$. A model non-interacting Anderson Hamiltonian [33] that captures an atom adsorbed on graphene can be written as

$$
\mathcal{H} = \sum_{k} \varepsilon_{gr} a_{g}^{\dagger} a_{g} + \sum_{s} \varepsilon_{ad} c_{s}^{\dagger} c_{s} + \sum_{k} V_{hyb} a_{g}^{\dagger} c_{s} + \sum_{k} V_{hyb} c_{s}^{\dagger} a_{g}.
$$

(18)

This Hamiltonian resembles the non-interacting Anderson impurity model for resonant impurities [34]. The energy of the adsorbed atom is given by $\varepsilon_{ad}$ while $\varepsilon_{gr}$ denotes the energy of the graphene electron states. The last two terms of the Hamiltonian describe the hybridization between the adsorbed atom and graphene. The summation over momentum vectors also denote the dual spin states. The creation (annihilation) operators for graphene and the adsorbed atom are denoted $a^{\dagger}(a)$ and $c_{s}^{\dagger}(c_{s})$, respectively.

To evaluate the spectral density function from which the overall broadening is determined, we consider the graphene-adsorbed impurity to be a non-interacting system. Using the equation of motion approach within the equilibrium Green’s function formalism, we start by writing the retarded Green’s function [35] for the adsorbed atom

$$
G^{R}(c, s, t) = -i \theta(t) \{ c_{s}(t), c_{s}^{\dagger}(0) \}.
$$

(19)

Taking the derivative of the retarded Green’s function in equation (19) and following Heisenberg’s picture ($\mathcal{H}(t) = \exp(i\mathcal{H}t) \mathcal{H} \exp(-i\mathcal{H}t)$) gives

$$
i \frac{\partial}{\partial t} G^{R}(c, s, t) = \delta(t) + \frac{i}{\hbar} \theta(t) \{ [\mathcal{H}, c_{s}(t)], c_{s}^{\dagger}(0) \}.
$$

(20)

Expanding equation (20) by evaluating the commutator (see appendix B), one obtains

$$
i \frac{\partial}{\partial t} G^{R}(c, s, t) = \delta(t) + \frac{\varepsilon_{ad}}{\hbar} G^{R}(c, s, t) + \frac{1}{\hbar} \sum_{k} V_{hyb} G^{R}(k, t),
$$

(21)

where $G^{R}(k, t)$ is the retarded Green’s function for graphene-adsorbed atom. $G^{R}(k, t)$ can be written in standard form as

$$
G^{R}(k, t) = -i \theta(t) \{ a_{g}(t), c_{s}^{\dagger}(0) \}.
$$

(22)

The corresponding equation of motion is

$$
i \frac{\partial}{\partial t} G^{R}(k, t) = \frac{\varepsilon_{gr}}{\hbar} G^{R}(k, t) + \frac{1}{\hbar} V_{hyb} G^{R}(c, s, t).
$$

(23)

The Fourier transform (equation (24)) of the two retarded Green’s functions yield a pair equations (equations (25) and (26)) which can be solved for $G^{R}(c, s, \omega)$

$$
G^{R}(c, s, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i\omega t} G^{R}(c, s, \omega),
$$

(24)

$$
\left( \omega - \varepsilon_{ad} - \frac{\varepsilon_{gr}}{\hbar} G^{R}(c, \omega) \right) = 1 + \frac{1}{\hbar} V_{hyb} G^{R}(k, \omega),
$$

(25)

and

$$
\left( \omega - \varepsilon_{gr} G^{R}(c, \omega) \right) = \frac{1}{\hbar} V_{hyb} G^{R}(c, s, \omega).
$$

(26)

Solving for $G^{R}(c, s, \omega)$ and preserving causality by making the substitution $\omega \rightarrow \omega + i0^{+}$ gives

$$
G^{R}(c, s, \omega) = \frac{\hbar}{\hbar \omega + i0^{+} - \varepsilon_{ad} - \sum_{k} \varepsilon_{gr}^{2}} + \frac{\hbar}{\varepsilon_{ad} - \sum_{k} P \varepsilon_{gr}^{2}} \delta(\hbar \omega - \varepsilon_{gr})
$$

(27)

where $P$ stands for the principal value [36] in the usual Plemelj relation: $\frac{1}{x \pm \mathbb{I}} = P \left( \frac{1}{x} \right) \pm i \pi \delta(x)$. The delta function is again the usual density of states for graphene;
putting all of them together and evaluating the spectral density
$A(c, s, ω) = -23G^R(c, s, ω)$ gives
\[ A(c, s, ω) = \frac{2\hbar}{\hbarω - \epsilon_{ad} - \sum_{k} P_{cv} |V_{\text{hyb}}|^2} + \Gamma^2, \tag{28} \]
where $d(E_\text{f}) = \frac{2|E_\text{f}|}{\pi\hbar\omega_\text{imp}}$ is the density of states for graphene at the Fermi level and the broadening parameter $\Gamma = \pi S|V_{\text{hyb}}|^2 d(E_\text{f}); S$ denotes the area of the graphene sheet under consideration and is roughly given as $S \approx 1\hbar\omega_\text{imp}$, and $n_{\text{imp}}$ is the concentration of impurity atoms adsorbed on the graphene sheet. The hybridization potential can be estimated as $V_{\text{hyb}} \approx \hbar\omega_i\left(R_0/\sqrt{\ln(R_d/R_i)}\right)$ \[37\]. Here, $R_0$ is the radius of the impurity atom, and $R_i$ is the average distance between the adsorbed impurity atoms and is approximately given as $1/\sqrt{\hbar\omega_\text{imp}}$.

Impurities therefore broaden the Dirac-delta peak, the broadening given by the hybridization potential, $V_{\text{hyb}}$. We have tacitly assumed that the system is non-interacting, and there is no Coulomb repulsion term of the form $Un_d,[n_d]_i$ that appears in a standard Hubbard Hamiltonian.

2.3. Circular dichroism in strained graphene

Circular dichroism quantifies the differential absorption of right and left circularly-polarized light. The expression for absorption coefficient due to light-induced inter-band transitions is a measure of the strength of the optical matrix element \[40\]. Inter-band absorption from a populated valence band eigen state to an empty conduction band state requires the determination of the optical inter-band transition matrix elements $|P_{cv}^x|^2$ and $|P_{cv}^y|^2$. The matrix element, for instance, $P_{cv}^x$ is nominally defined as \[40\]
\[ P_{cv}^x = \langle \Psi_x | \frac{\partial}{\partial k_x} H_{\text{opt}} | \Psi_x \rangle. \tag{29} \]
The degree of circular polarization \[41\] $\rho(k)$ is therefore
\[ \rho(k) = \frac{|P_{cv}^x|^2 - |P_{cv}^y|^2}{|P_{cv}^x|^2 + |P_{cv}^y|^2}, \tag{30} \]
where $P_{cv}^x$ and $P_{cv}^y$ for right- and left-circularly-polarized light, respectively are defined as
\[ P_{cv}^x = P_{cv}^y \pm iP_{cv}^y. \tag{31} \]

We utilize equation (29) to compute the inter-band optical matrix elements in strained graphene \[42\] and establish a relation between dichroism and the tunable Fermi level. A graphene sheet usually experiences acoustic and optical strain. Under acoustic strain, the two carbon atoms of the unit cell are displaced in phase, while for the optical case, there is a shift such that the centre-of-mass remains invariant. An out-of-plane component of optical strain renders the two sub-lattices of graphene inequivalent, a situation commonly realized when graphene is grown on a substrate, for instance, hexagonal boron nitride(h-BN). The out-of-plane optical strain component is essentially an inherent coupling of graphene to the substrate by van der Waals forces which is akin to a normal electric field that disturbs the reflection symmetry of the planar surface. The loss of reflection symmetry, in this case manifesting as breaking of the sub-lattice symmetry leads to a gap opening in the spectrum. The Hamiltonian using the method of invariants that describes this situation is written as \[43\]
\[ H_{\text{opt}} = \hbar\omega_i\{(k_x - ik_y)\sigma_x + (k_x + ik_y)\sigma_y\} + E_{\text{op}}\sigma_z, \tag{32} \]
where $\sigma_{\pm} = (\sigma_x \pm i\sigma_y)/2$ and $E_{\text{op}}$ is the optical strain. A gap, $\Delta = 2E_{\text{op}}$, in the spectrum appears now which implies the non-equivalence of the graphene sub-lattices. Interestingly, the out-of-plane optical strain component, while it breaks the reflection symmetry, does not move the Dirac points from the Brillouin zone edges $K$ and $K'$. Note that experimentally, the precise determination of the optical strain reflected by the intrinsic band gap opening has been reported through magneto-spectroscopy measurements. These measurements allow for the observation of the finite band gap by probing the Landau levels; the transition peaks between different Landau levels obtained by noting their joint density of states yield accurate data about the intrinsic band gap. We refer the reader to \[44\] for a detailed review of this method. In passing, it is worth mentioning that this technique is impervious to defects and impurities. Another useful technique that has been recently proposed involves measuring the anisotropy of photoconductivity and directly linking it to the strain induced asymmetry in graphene lattice \[45\].

The optical matrix elements and the tunable circular dichroism of an optically strained graphene sheet can be calculated by first working out the velocity components. The $x$ and $y$ components of velocity are given by
\[ \frac{\partial H_{\text{opt}}}{\partial k_x} = \begin{pmatrix} 0 & \hbar\omega_i \\ \hbar\omega_i & 0 \end{pmatrix}, \tag{33a} \]
and
\[ \frac{\partial H_{\text{opt}}}{\partial k_y} = \begin{pmatrix} 0 & -i\hbar\omega_i \\ -i\hbar\omega_i & 0 \end{pmatrix}. \tag{33b} \]

The optical matrix element corresponding to the velocity components for a finite band gap graphene can therefore be written by inserting expressions for wave functions and the velocity components from equations (2) and (33), respectively, in equation (29) for a right circularly-polarized light.
\[ P_{cv,x} = \frac{1}{2}(u_+ \exp(i\theta) u_- - u_+ \exp(-i\theta) u_-) \tag{34} \]

The matrix element corresponding to momentum operator $\hat{p}_y$ is
\[ P_{cv,y} = \frac{1}{2}(u_+ \exp(i\theta) u_- - u_+ \exp(-i\theta) u_-). \tag{35} \]
Combining both the components, the square of the right polarized optical matrix element is
\[ |P_{cv}^+|^2 = (h\nu)^2 \left[ 1 + \frac{\Delta}{(h\nu k)^2 + \Delta^2} \right]^2. \] (36)

The corresponding expression for \( |P_{cv}^-|^2 \) following an analogous procedure is
\[ |P_{cv}^-|^2 = (h\nu)^2 \left[ 1 + \frac{-\Delta}{(h\nu k)^2 + \Delta^2} \right]^2. \] (37)

The above derivation for dichroism in graphene was carried out by considering an optical strain that furnishes a finite band gap. First principle calculations [46] also show that an application of a large uniaxial strain in graphene does not destroy the semi-metallic nature of graphene but significantly impacts the Fermi velocity components. The anisotropy of Fermi velocity which manifests as tilted Dirac cones has been profitably employed to tune the optical properties of two-dimensional graphene [47]. We work out an expression for circular dichroism in graphene by considering uniaxial strain along the zigzag and armchair directions [48] in presence of a finite band gap. Writing out the Hamiltonian (equation (32)) again but with different velocity components, \( \mathbf{\hat{H}} = v_\eta p_\eta \sigma + v_\eta p_\eta \sigma \), such that \( \kappa = v_\eta / v_x \). The ratio is usually computed by resorting to a single-orbital tight-binding calculation which expresses the velocity components as [49]
\[ v_x = t_2 \delta a \sqrt{4a^2 - 1}, \]
\[ v_y = t_2 \delta a. \] (38)
Here, \( t(i = 1, 2, 3) \) denotes the kinetic energy hopping integrals between the nearest neighbours, and \( \eta = \hbar / t_2 \). The strained lattice vector is resolved in \( x \) and \( y \) components to give and \( a_x = a / 2 \) and \( a_y = \frac{\sqrt{3} a}{2} \), with \( a = \sqrt{3} \times 1.42 \) Å. Using first principles calculations [46], it has been demonstrated that a uniaxial strain along the zig-zag direction (denoted as ‘Z’ strain) in the honeycomb lattice leads to \( \eta = \hbar / t_2 \) (\( \eta < 1 \)), while for uniaxial strain in the armchair direction (denoted as ‘A’ strain), \( \eta = \hbar / t_2 \) (\( \eta > 1 \)).

The degree of velocity anisotropy, \( \kappa \), using equation (38) is given as
\[ \kappa = \frac{v_y}{v_x} = \sqrt{\frac{3}{4a^2 - 1}}. \] (39)

The degree of circular polarization (equation (30)) is modified to reflect this by an alteration to the algebraic expressions for both right and left polarized optical matrix elements. Carrying out the derivation as before gives
\[ |P_{cv}^\pm|^2 = (h\nu)^2 \left[ 1 + \frac{\pm \Delta}{\sqrt{(h\nu k_x)^2 + \Delta^2}} \right]^2, \] (40)
where \( v_x = \sqrt{a^2 + v_y^2} \). In terms of the velocity anisotropy, the above equation can be simplified as
\[ |P_{cv}^\pm|^2 = (h\nu)^2 \left[ 1 + \frac{\pm \Delta}{(h\nu k_x)^2 + \Delta^2} \right]^2, \] (41)
where \( \tan \theta_k = k_y / k_x \).

The average degree of circular polarization over a constant energy surface (free of trigonal warping effects [50, 51]) can be defined as
\[ \rho(k) = \frac{\int d^2 k \rho(k) \delta(\omega - 2\varepsilon(k))}{\int d^2 k \delta(\omega - 2\varepsilon(k))}, \] (42)
where \( \varepsilon(k) = \pm \sqrt{\hbar^2 k_x^2 + \Delta^2} \) is the energy spectrum of graphene. The frequency of the light beam must satisfy \( \omega = 2\varepsilon(k) \) which describes the energy involved in an inter-band transition from valence to conduction band.

Another possible way to look at circular dichroism is by examining its relationship to the topologically non-trivial finite Berry curvature of gapped Dirac fermions. The situation is special in case of graphene because of the presence of pseudo-spin [52] which describes the inverted sub-lattices \( A \) and \( B \) giving rise to helical bands and an intrinsic magnetic moment. When light of a certain circular polarization shines on graphene, the light wave vector \( q \) aligns parallel or anti-parallel to the pseudo-spin [53] giving rise to the markedly strong circular dichroism. For instance, a right-circularly polarized light beam would strongly couple to the pseudo-spin vector that has clockwise circulation at the \( K \) edge of the Brillouin zone giving a perfect absorption while a similar phenomenon can be observed with left-circularly polarized light at the \( K' \) valley edge. Note that \( K \) and \( K' \) are time reversed, energetically degenerate and inequivalent pairs. The Berry curvature is an analogous concept which
especially tells us about the handedness of Bloch electrons and can be seen as a measure of the geometry-dependent orbital magnetization. As light shines on graphene surface, the wave vector aligns itself parallel/anti-parallel to this orbital magnetization and as before, right-circularly polarized light, for instance, is preferentially absorbed based on the orientation of the Berry curvature. The circular dichroism is therefore directly linked to Berry curvature [54]. A very instructive reference on this subject though it mainly deals with transition metal dichalcogenides as the representative material appeared in Nature Communications in 2012, [55].

The Berry curvature for gapped Dirac fermions is given as [56]

\[ \Omega(k) = \pm \frac{\hbar^2 v_F^2 \Delta}{2 \left( \sqrt{\Delta^2 + (\hbar v_F k)^2} \right)^3}, \]  

(43)

where \( \Delta \) is the band gap. Note that the upper(lower) sign corresponds to the \( K(K') \) valley edge in the two-dimensional Brillouin zone. The opposite polarity of Berry curvature at the two valley edges is because of time reversal symmetry that connects \( K \) and \( K' \). Using equation (43), the circular dichroism is connected to Berry curvature as [57]

\[ \eta(k) = -\frac{\Omega(k) \cdot \hat{z}}{\mu_B(k)} e \frac{\Delta(k)}{2\hbar}, \]  

(44)

where \( \mu_B = e \hbar/2m^* \) is the spin Bohr magneton. We can thus easily infer from equation (44) that a change of magnitude for the Berry curvature alters the circular dichroism. The Berry curvature which is sensitive to parametric changes in momentum-space and the defining Hamiltonian therefore offers an alternative way to tune circular dichroism. For instance, the inclusion of trigonal warping effects [58, 59] in graphene would influence the overall expression for Berry curvature and circular dichroism.

3. Results

As a first check of the validity of the numerical model to compute absorption, an undoped graphene sheet with an electrically tuned Fermi level and suspended in vacuum (the dielectric constants flanking the graphene sheet are unity) is considered. The absorption coefficient, at normal angle of incidence, for a range of energies is plotted in figure 2. Two features in figure 2 stand out; when incident radiation energy is upwards of twice the Fermi level, the absorption is constant at approximately 2.3 % and the absorption coefficient has a ‘hump’ at exactly \( \hbar \omega = 2E_F \). The numerical demonstration of absorption coefficient as approximately 2.3% is significant since it is in conformity with the theoretically derived number (see appendix A) obtained directly from the wave functions of graphene’s linear Hamiltonian coupled to the incident electromagnetic field. The ‘hump’ is attributed to an onset of inter-band conductivity at \( \hbar \omega = 2E_F \), beyond which the absorption coefficient stays constant. The intra-band Drude contribution is dominant at much lower energies as can be seen by observing the optical conductivity versus energy plot of figure 3. The ‘tuning’ of absorption coefficient via the optical conductivity of graphene will concern us for rest of this section.

3.1. Absorption coefficient versus incident angle and material parameters

The amount of light absorbed by a suspended graphene sheet must be a tunable quantity for a wide range of applications such as photo-detectors and sensors. This tuning, for a given set of material constants, can be accomplished by selecting ‘control’ parameters such as frequency of the incoming light-beam, the Fermi level and the angle of incidence to alter the overall reflectance, transmission, and absorption. We show in figure 4 the relation between incident angle, \( \theta \), and the absorption coefficient for various values of \( \xi = \epsilon_1/\epsilon_2 \) at fixed values of \( E_F, \Gamma \), and \( \omega \). The two dielectric constants surrounding the graphene sheet are denoted by \( \epsilon_1 \) and \( \epsilon_2 \) as sketched in figure 1. It is easily seen that the absorption coefficient reaches up to 35.0% when the ratio of dielectric constants is 2.0 at close to normal incidence. Besides the enhanced absorption, another noteworthy feature in figure 4 is the degradation in absorption coefficient as the incident angle increases from zero to \( \pi/2 \). This is explained by examining the expressions for reflectance and transmittance given in equations (13a) and (13c); for a fixed ratio of the dielectric constants, at higher angles of incidence,
the transmitted wave vector (in medium ‘2’) $k_{2, \xi}$ changes such that a greater portion of the incident light is reflected. The transmitted wave in medium ‘2’ is simply expressed as $k_{2, \xi} = \sqrt{(\xi^2 - k_z^2)}$, where $k_z = k_0 \sin \theta$, $k_0$ and $\omega$ describe the incident wave-vector and frequency, respectively. A lesser absorption with decreasing ratio of the dielectric constants $\xi$ is also explained by utilizing equations (13a) and (13c); it is evident that a lower ratio as marked on the plot will augment the reflection coefficient.

We next turn our attention to absorption characteristics as a function of the Fermi level and the operating frequency. The functional dependence is shown in figure 5 for absorption at normal incidence, $\varepsilon_1 = 4$, $\varepsilon_2 = 2$, and the broadening parameter $\Gamma$ set to 2.6 meV. At an operating frequency of around $\omega \approx 0.01 \omega_f$, the absorption coefficient exhibits a peak for $E_t = 0.4$ eV and $E_t = 0.6$ eV as shown in figure 5. Two crucial observations can be made that will help us define an ‘optimal’ parameter space to design graphene photonic devices. We first focus on the low $\omega/\omega_f$ ratio which gives an absorption peak; at this operational frequency, the Drude inter-band conductivity dominates such that absorption increases up to approximately 70.0%, saturates, and then starts to fall. For $\omega > 0.1 \omega_f$, the Drude conductivity which is still the dominant mode until $\omega = 2 \omega_f$, rapidly drops to make a negligible contribution independent of the Fermi level (see figure 3); with all other parameters held constant, we are therefore able to explain the merging of the absorption profiles for all three Fermi levels. The inter-band scattering is zero since we increase $\omega$ until it reaches $\omega_f$.

The inset in figure 5 further shows the role of the broadening parameter in determining the absorption of light. As the broadening parameter gains strength, in regions where Drude conductivity is significant ($\omega < 0.1 \omega_f$), the conductivity is lowered for identical operational characteristics such as Fermi level, frequency etc. The lowered conductivity explains the drop in absorption coefficient. The broadening parameter in the plot is a Lorentzian and is assumed to be Fermi level independent.

We have thus identified a small window for the operating frequency for $E_t > 0.4$ eV, which enables a maximized absorption up to approximately 70.0% for the selected dielectric constants surrounding the graphene sheet for TM polarized incident light. A low value of broadening parameter is desirable to keep the absorption coefficient high. While in the above estimates for absorption, the broadening was an empirically chosen number, we present below a more accurate calculation that relates it to the impurity concentration and Fermi level of graphene.

An impure graphene sheet, which is a more realistic scenario, is considered to evaluate the spectral broadening of
states. The impurities are non-interacting as described by the Hamiltonian in equation (18). Figure 6 conveys the role of impurities in influencing the optical absorption, where the Lorentzian broadening is characterized by the hybridization potential, $V_{\text{hyb}}$, of the impurity atom with the $\pi$-bonds of graphene. The trend in figure 6 is in conformity with the behaviour shown in the inset plot of figure 5. As expected, with an increase in the impurity concentration, the broadening is enhanced which lowers the absorption. The three trend lines shown in figure 6 further demonstrate that for a low $\omega/\omega_{fy}$ ratio, the Drude conductivity is significant as indicated by the downward trend in absorption($\omega/\omega_{fy} = 0.05$). As $\omega \to \omega_{fy}$, Drude conductivity is considerably decreased in magnitude as depicted by the ‘relatively’ straight absorption curves. These absorption curves are also insensitive to the Fermi level in graphene. However, in the presence of other impurity atoms that are interacting such as charged impurities and polar phonons, absorption coefficient will indeed exhibit a dependence on the Fermi level. This aspect has not been considered. The hybridization potential used in determining the broadening is shown in the inset plot of figure 6 as a function of $n_{\text{imp}}$. For a typical impurity concentration of $n_{\text{imp}} \approx 10^{10}$ cm$^{-2}$, $V_{\text{hyb}} \approx (0.5 - 1)$ KeVÅ$^{-1}$ [60].

We have therefore identified a few key guidelines to design a graphene photonic device. Primarily, in the low-frequency regime where Drude conductivity is active, absorption saturates as a function of the ratio of dielectric constants and frequency of incident light. The corresponding Fermi level can be adjusted such that absorption is maximized. Finally, the broadening parameter must not be too high, a large spectral broadening at low frequencies degrades the overall absorption.

### 3.2. Circular dichroism

Circular dichroism or the degree of circular polarization relates to the differential absorption of right and left circularly-polarized light. In the case of graphene with its linear bands, the dichroism is analytically computed through equation (30). As is clear from equation (30), pristine graphene ($\Delta = 0$) does not exhibit circular polarization $\rho$; however, in the presence of an inversion symmetry-breaking band gap, which renders the two sub-lattices inequivalent, $\rho$ is a non-zero number. We seek to evaluate $\rho$ in the following section under a symmetry-breaking condition. The coupling of right (left) circularly polarized light (see figure 7) which is exact at the Dirac point formed at $K(K')$ edge of the Brillouin zone [19] exhibits a polarization-dependent light absorption near a particular edge, say $K$. This differential absorption quantified as the degree of circular polarization is plotted against the Fermi level. The Fermi level in this case is assumed to coincide with the energy spectrum of graphene. The $k$-vector is taken as 0.11 Å$^{-1}$. The dichroism decreases with an increasing Fermi level.

![Figure 7](image1.png) **Figure 7.** Right and left circularly polarized light couples selectively to band edges $K$ and $K'$ since they are time-reversed pairs. At other points in $k$-space, not far from the band edge, the selective coupling is lost and a varying degree of polarization-dependent absorption occurs that gives rise to the phenomenon of circular dichroism. The two curved arrows denote left and right circular polarization.

![Figure 8](image2.png) **Figure 8.** Degree of circular polarization $\rho$ in gapped graphene versus the Fermi level for two values of the band-gap: $\Delta = 5$ meV and 10 meV, $\rho$ is plotted using equation (42) over a constant energy surface defined by the respective Fermi level, which is defined by the energy spectrum of graphene. The $k$-vector is taken as 0.11 Å$^{-1}$. The dichroism decreases with an increasing Fermi level.
uniaxial strain until 24% does not contribute to an additional band gap other than that created through substrate-induced optical strain. Choi et al in [46] explain this reduction in $\kappa$ by considering the strength of the hopping integral parameters. The qualitative relationship between hopping integrals, connecting the nearest neighbors in a single-orbital tight-binding approximation under $'A'$ strain in graphene, $t_{12} > t_{13}$ is shown in the inset of figure 9. Using the velocity-anisotropy graph of figure 9, the degree of circular polarization in uniaxially-strained graphene is computed (figure 10) as a function of $'A'$ strain percentage. We immediately notice from the plot that for $\theta = \pi/4$, where $\theta = \arctan \frac{\Delta}{\Delta}$, the degree of circular polarization does not considerably vary while there is a noticeable change for $\theta = \pi/6, \pi/3$ with increasing strain. The trend for $\theta = \pi/4$ is explainable if we note that at this angle, the two components of the $k$-vectors are symmetric, the uniaxial strain does not contribute to the band gap $\Delta$ and the velocity anisotropy-factor only causes a slight change in degree of circular polarization. The slight deviation as strain increases and consequently the velocity-anisotropy factor $\kappa$ supports this reasoning. The other angles introduce additional asymmetry that augments the velocity-anisotropy to produce a larger change in the degree of circular polarization.

The same behaviour is observed again for strain of the zigzag type which is plotted in figure 11. At $\theta = \pi/4$, the degree of circular polarization is almost constant while it shows an upward trend for $\theta = \pi/6$. The important point to note is the inter-play of $k$-space and velocity anisotropy; if the two degrees of asymmetry cancel out, as it happens for $\theta = \pi/3$ in the zigzag case, there will not be a sufficient variation in circular polarisation. This assumes importance in view of our proposal to enhance light absorption of a particular polarization by selecting a suitable $k$-space asymmetry combined with velocity anisotropy we can tune the differential absorption of right or left circularly-polarized light. This idea runs parallel to the emerging field of valleytronics in transition metal dichalcogenides [61, 62].

4. Conclusions
We have demonstrated in this work the tunability of optical absorption in graphene via a combination of fabrication design techniques and external dynamic control. The insertion of dielectric films of specified permittivity that surround the active graphene layer govern the transmission and reflection coefficients at a given Fermi level. The reflection and transmission coefficients which can then be made to span the complete range, $R, T \in [0, 1]$, therefore allow, through a selection of the dielectric constants, a desired absorption window. This window can be adjusted to conform to any sought level of
absorption for a particular application. We refer to the choice of dielectric layers as a possible fabrication design technique. The effect of surface adsorbed impurity atoms is also a viable mechanism to tune the optical conductivity; by a careful choice of impurity atoms [63, 64], the spectral broadening of the density of states can be adjusted to match the proper optical absorption. Circular dichroism is considered as another possible option to selectively absorb polarization-dependent light by focusing attention to optical processes in one of the two edges \((K, K')\). Real time dynamic control, different from fabrication designs can also be exercised by a gate contact that alters the Fermi level in the graphene sheet. A changing Fermi level influences graphene’s optical conductivity and can switch the graphene-based optical device between varying degrees of absorption via modification of the reflection and transmission coefficients.

There are several parameters that can be explored to further refine the efficiency of optical absorption in graphene. We have not considered in this work the influence of strain that changes the atomic orbital overlap to give rise to an anisotropic conductivity tensor. Strain was considered in an elementary treatment to measure the degree of circular polarization assuming that the Dirac dispersion is preserved; in reality, strain alters the density of states, the Dirac Hamiltonian and underlying optical and electronic properties. A more complete investigation of optimally engineered strained graphene structures will be considered in a follow-ups work. Further, controllable doping in graphene, which provides a pathway for easy switching [65] between \(n\)-type and \(p\)-type can be considered as a fabrication design parameter to enhance optical absorption. Doping has not been explicitly considered in this work.

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Appendix A. Optical absorption in mono-layer graphene

For the sake of completeness, we derive the absorbance for a graphene sheet and show that it is independent of the incident photon frequency. The incident light is assumed to be linearly polarized along \(x\)-axis and shines normally on the surface. The electric field, using the vector potential \(A(t) = \vec{A} \exp(-i\omega t)\) is

\[
\vec{E}(t) = -\frac{1}{c} \frac{\partial \vec{A}}{\partial t}.
\]

The modified Hamiltonian after including the Peierls substitution takes the form \(\mathcal{H} = \nu \sigma \cdot \left( \vec{p} - \xi \vec{A} \right)\). The interaction part of the Hamiltonian is therefore \(\mathcal{H}_{\text{int}} = \frac{\nu \nu}{2} \vec{\sigma} \cdot \vec{A}\).

Substituting for \(\vec{A}\) from equation (A.1), \(\mathcal{H}_{\text{int}} = \frac{\nu \nu}{2} \vec{\sigma} \cdot \vec{E}\).

The factor of 0.5 comes from by retaining only the \((-i\omega t)\) term.

Using the Fermi golden rule, the transition probability for a carrier to be excited from the valence band to conduction band is \(\sum_{k,k'} \frac{2}{\hbar} \left| \langle \psi_f | \mathcal{H}_{\text{int}} | \psi_i \rangle \right|^2 \delta(E_k - E_{k'} - \hbar \omega)\) where the delta function is transformed in to the density of states.

In the above expressions, \(\psi_i\) and \(\psi_f\) represent the initial and final wave functions while \(E_k\) and \(E_{k'}\) are the energies corresponding to the bottom of the conduction band and top of the valence band, respectively. Calculating the matrix element \(M(k) = \langle \psi_f | \mathcal{H}_{\text{int}} | \psi_i \rangle\) gives

\[
M(k) = \frac{1}{2} (\exp(i\theta) - 1) \mathcal{H}_{\text{int}} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \exp(-i\theta). \quad (A.2)
\]

Inserting the expression for \(\mathcal{H}_{\text{int}}\) in equation (A.2) and noting that the \(\sigma_i\) dots with the \(x\) polarized electric field, one obtains

\[
M(k) = \frac{\nu \nu |E_k|}{4\hbar \omega} (\exp(i\theta) - 1) \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \exp(-i\theta),
\]

\[
= \frac{\nu \nu |E_k|}{2\hbar \omega} \sin \theta, \quad (A.3)
\]

where the wave functions of equation (2a) are utilized. The probability density by inserting the square of the matrix element in Fermi Golden rule can be written as

\[
P_{\text{h} \rightarrow \text{c}} = \frac{2\pi \nu \nu^2 |E_k|^2}{4\hbar^2 \omega^2} \sin^2 \theta \int d^2r. \quad (A.4)
\]

The absorbed energy flux density is therefore \(\phi = \frac{1}{A} \hbar \omega P_{\text{h} \rightarrow \text{c}}\). Summing over the density of states which is \(\frac{2\pi k_F^2}{\hbar^2}\), where energy \(\epsilon = \frac{\hbar \omega}{2}\) and using the mean value of \(\sin^2 \theta\) over a full cycle as 1/2, the final energy flux takes the following expression

\[
\phi_{\text{total}} = \frac{1}{A} \hbar \omega \frac{2\pi \nu \nu^2 |E_k|^2}{4\hbar^2} \frac{1}{2} \frac{\hbar \omega}{2\pi k_F^2} \nu^2 \frac{\hbar^2}{\hbar^2}.
\]

Note that the integral \(\int d^2r\) is equal to \(A\), the exposed graphene area in equation (A.4). The incident flux for an electric field \(|E| = \frac{\epsilon}{4\pi} |E|^2\). The absorption is therefore \(\frac{\nu \nu}{\hbar^2} = 2.3\%\). This number includes the valley degeneracy of two in graphene.

Appendix B. Evaluation of the commutator in equation (20)

The final expression for the commutator in equation (20) is derived here. The commutator is given as \([\mathcal{H}, c_s(t)]\), \(c_s(t)\). We proceed by simplifying the inner commutator \([\mathcal{H}, c_s(t)]\). The Hamiltonian \(\mathcal{H}\) is given by

\[
\mathcal{H} = \sum_k \varepsilon_k a_k^\dagger a_k + \sum_k \varepsilon_k a_k^\dagger c_k + \sum_k V_{\text{hyb}} a_k^\dagger c_k + \sum_k V_{\text{hyb}} a_k^\dagger a_k.
\]

This \(\mathcal{H}\) gives four commutators. The first commutator is
\[ \sum_s a_s^\dagger a_s c_s = \sum_s \{a_s^\dagger a_s c_s\} - \{a_s^\dagger c_s a_s\}. \]  \hspace{1cm} (B.2)

Expanding terms within the bracket, the commutator is zero. The relation \([AB, C] = A[B, C] - [A, CB]B\) is used here. In equation (B.2), the creation (annihilation) operators for graphene and the adsorbed atom are simply denoted by \(a_s^\dagger\) and \(c_s\) respectively. The subscript \(s\) describes the spin projection. More detailed subscripts, on-site energies of graphene and adsorbed impurity atom, and hybridization potentials that appear in the original Hamiltonian (equation (B.1)) are omitted for brevity.

The second commutator is
\[ \sum_s [c_s^\dagger a_s^\dagger, c_s] = \sum_s [c_s^\dagger a_s^\dagger c_s] - \{c_s^\dagger a_s^\dagger, c_s\}. \]  \hspace{1cm} (B.3)

Standard commutation relations in equation (B.4) simplify the commutator \(\sum_s [c_s^\dagger a_s^\dagger c_s]\) to \(-c_s\).
\[ \{c_s^\dagger, c_s\} = 0; \{c_s^\dagger, c_s\} = 1. \]  \hspace{1cm} (B.4)

The third commutator from the Hamiltonian is
\[ \sum_s [a_s^\dagger c_s^\dagger, c_s] = \sum_s (a_s^\dagger [c_s^\dagger c_s] - \{a_s^\dagger c_s\}). \]  \hspace{1cm} (B.5)

Evaluating each sub-anticommutator on rhs and using commutation relations in equation (B.4) yields zero.

The last commutator is
\[ \sum_s [c_s^\dagger a_s^\dagger, c_s] = \sum_s [c_s^\dagger a_s^\dagger c_s] - \{c_s^\dagger a_s^\dagger c_s\}. \]  \hspace{1cm} (B.6)

As before, evaluating the sub-anticommutators give \(\sum_s [c_s^\dagger a_s^\dagger, c_s] = -a_s\).

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