Theory seminar: Electronic and optical properties of graphene

Optics and Response Functions

Matthias Droth, 04.07.2013

Outline:

- Light absorption by Dirac fermions
- Intro: response functions
- The optics of Dirac fermions
- Plasmons

The fine structure constant $\alpha = \frac{e^2}{\hbar c} = \frac{1}{137}$

defines the visual transparency of graphene.

Maxwell equation: $\vec{E} = -\vec{\nabla}\phi - \partial_t \vec{A}$

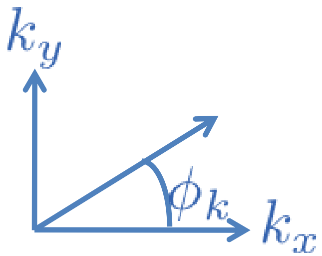
$$\phi = \text{const.}$$
$$\vec{A}(t) = \vec{A}e^{-i\omega t}$$
$$\Rightarrow$$
$$\vec{E}(t) = i\omega\vec{A}(t)$$
$$\vec{A}(t) = \frac{-i}{\omega}\vec{E}(t)$$

Hamiltonian in K valley: $H = H_0 + H_{\text{int}} = v_F \vec{\sigma}\cdot(\vec{p} - e\vec{A})$

$$H_0 = v_F \vec{\sigma}\cdot\vec{p}$$
$$H_{\text{int}} = -ev_F\vec{\sigma}\cdot\vec{A}(t) = \frac{iev_F}{\omega}\vec{\sigma}\cdot\vec{E}(t) = 2\,\frac{iev_F\vec{\sigma}\cdot\vec{E}(t)}{2\omega}$$

states in K valley:

$$\psi^{(K)}_{e(h)}(\vec{k}) = \frac{1}{\sqrt{2}}\begin{pmatrix} e^{-i\phi_k/2} \\ +(-)e^{i\phi_k/2} \end{pmatrix}$$



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Maxwell equation: $\vec{E} = -\vec{\nabla}\phi - \partial_t \vec{A}$

$\phi = \text{const.}$ $\qquad \vec{E}(t) = i\omega \vec{A}(t)$

is the Hamiltonian of the electron–photon interaction. The factor $\frac{1}{2}$ in Eq. (7.4) is necessary since the standard expression for the complex field is

$$\vec{E}(t) = \mathrm{Re}\left[\vec{E}\exp(-i\omega t)\right] = \frac{1}{2}\left[\vec{E}\exp(-i\omega t) + \vec{E}^{*}\exp(i\omega t)\right] \qquad (7.5)$$

and we take into account only the first term. This interaction induces transi-

$$H_{\text{int}} = -ev_F \vec{\sigma}\cdot\vec{A}(t) = \frac{iev_F}{\omega}\vec{\sigma}\cdot\vec{E}(t) = \cancel{2}\,\frac{iev_F \vec{\sigma}\cdot\vec{E}(t)}{2\omega}$$

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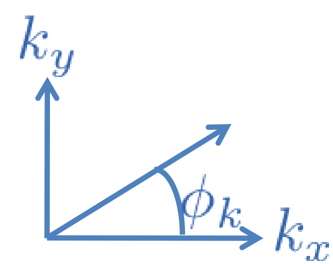


optical transition matrix element:

$$\langle\psi_h|H_{\text{int}}|\psi_e\rangle = \frac{iev_F}{2\omega}\begin{pmatrix} e^{+i\phi_k/2} \\ -e^{-i\phi_k/2}\end{pmatrix}\begin{pmatrix} 0 & \frac{E_x - iE_y}{2} \\ \frac{E_x+iE_y}{2} & 0\end{pmatrix} e^{-i\omega t}\begin{pmatrix} e^{-i\phi_k/2} \\ +e^{i\phi_k/2}\end{pmatrix} =$$

$$= \ldots = \frac{ev_F}{2\omega}\left(E_y(t)\cos\phi_k - E_x(t)\sin\phi_k\right)$$

+ for K‘

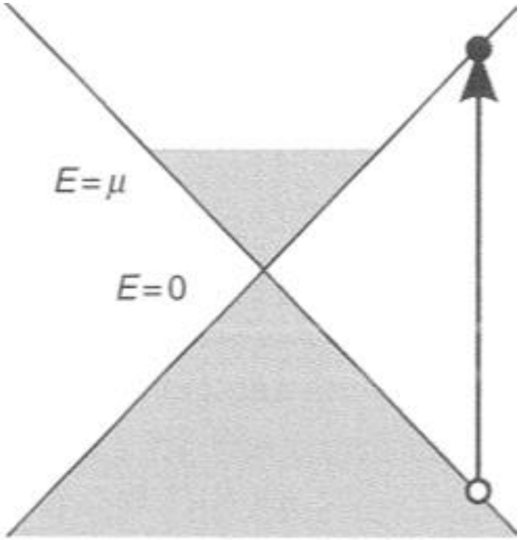
|...|^2: $$\left(\frac{ev_F}{2\omega}\right)^2\left(E_y^2\cos^2\phi_k + E_x^2\sin^2\phi_k - 2E_xE_y\cos\phi_k\sin\phi_k\right)$$

average over \phi:

$$\overline{|\langle\cdots\rangle|^2} = \frac{1}{2\pi}\int_0^{2\pi}|\langle\ldots\rangle|\mathrm{d}\phi = \left(\frac{ev_F}{2\omega}\right)^2\left(E_y^2\frac{1}{2} + 0 + E_x^2\frac{1}{2}\right) = \frac{e^2v_F^2}{8\omega^2}|\vec{E}|^2$$

assuming $E_z = 0$

i.e. photon incidence perpendicular to graphene plane



Fermi‘s Golden Rule => absorption rate:

$$\Gamma = \frac{2\pi}{\hbar}\overline{|\langle\ldots\rangle|^2}N(\epsilon = -\hbar\omega/2)$$

$$= \frac{2\pi}{\hbar}\frac{e^2 v_F^2|\vec{E}|^2}{8\omega^2}\frac{\omega}{\pi\hbar v_F^2} = \frac{e^2|\vec{E}|^2}{4\hbar^2\omega}$$

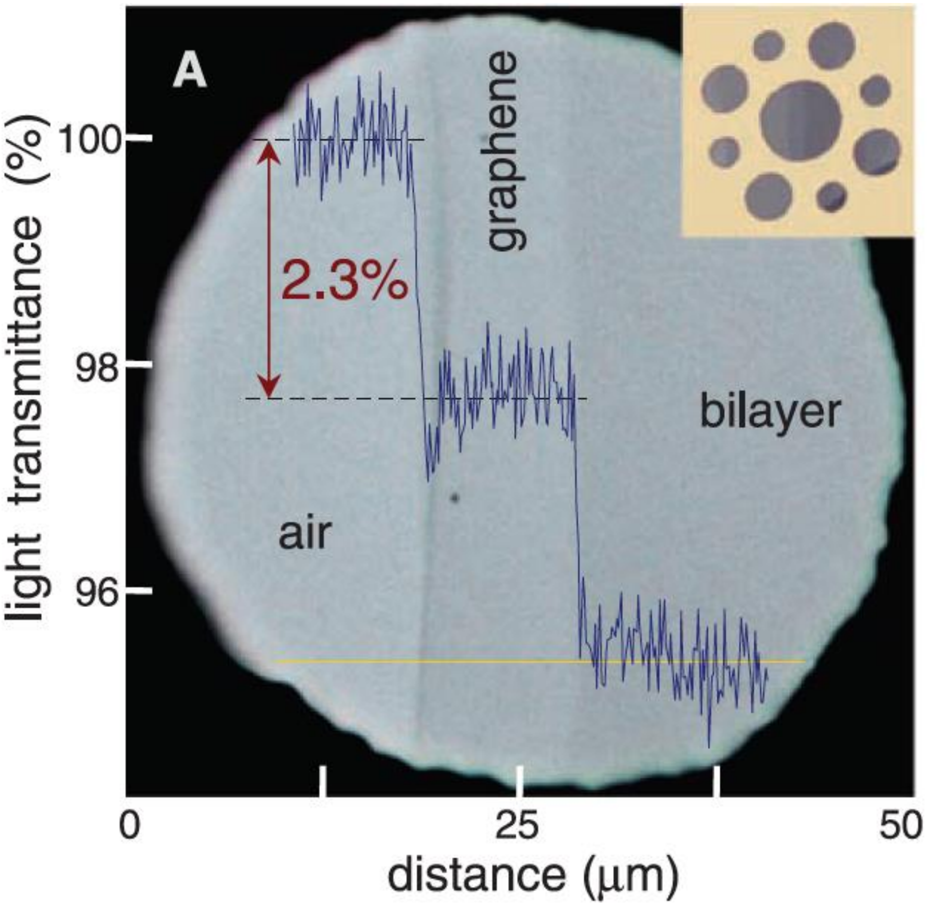
=> energy absorption rate: $W_a = \Gamma\hbar\omega = \dfrac{e^2|\vec{E}|^2}{4\hbar}$

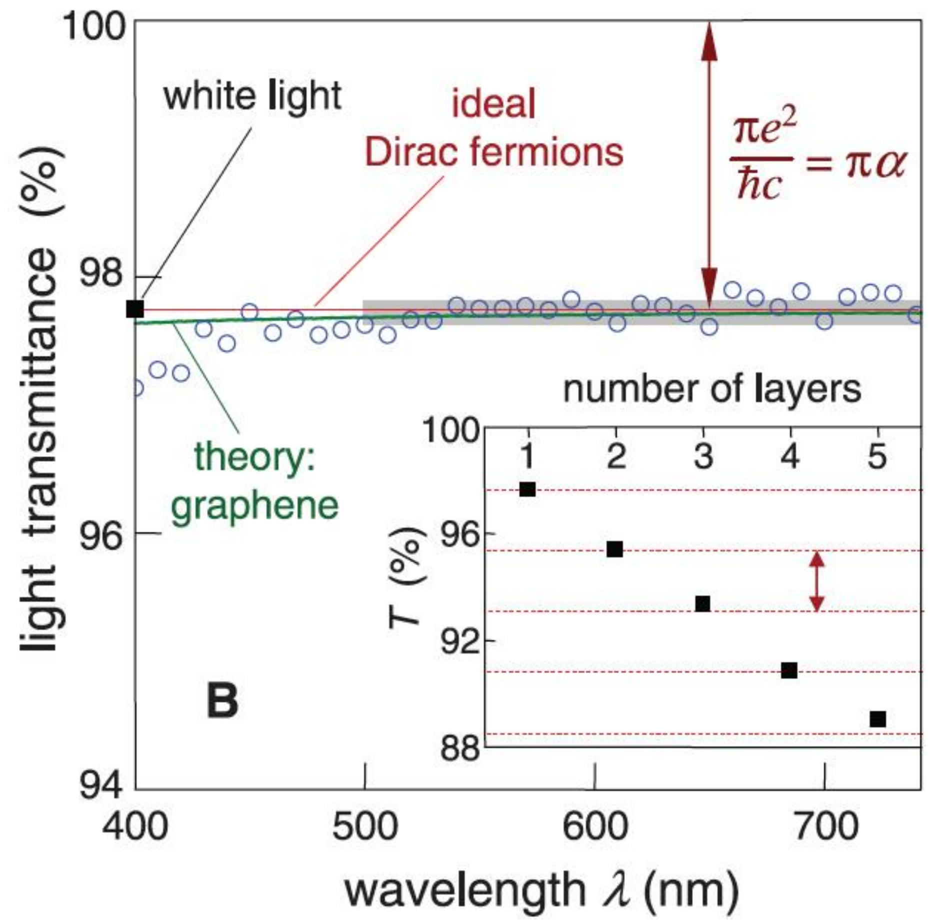
incident energy flux (Jackson, 1962): $W_i = \dfrac{c|\vec{E}|^2}{4\pi}$

=> absorption coefficient:

$$\eta = \frac{W_a}{W_i} = \frac{\pi e^2}{\hbar c} = \pi\alpha \approx \frac{3.14}{137} = 2.3\%$$

[R. Nair *et al.*, Science **320**, 1308 (2008).]





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How does a physical system react to an external perturbation?
More precise: how does the expectation value of observable $\hat{A}$ change?
According to the *response functions*!

1. conductivity
2. susceptibilty
3. heat conductivity *(retarded Green‘s functions)*

without perturbation:

$$H_0 \qquad \langle \hat{A} \rangle_0 = \mathrm{Tr}(\rho_0 \hat{A}) \qquad \rho_0 = \frac{e^{-\beta \mathcal{H}_0}}{\mathrm{Tr}(e^{-\beta \mathcal{H}_0})} \qquad \mathcal{H}_0 = H_0 - \mu \hat{N}$$

system — expectation value of observable $\hat{A}$ — density matrix — grand potential

with perturbation $V_t = F_t \hat{B}$:

$$H = H_0 + V_t \qquad \langle \hat{A} \rangle_t = \mathrm{Tr}(\rho_t \hat{A}) \qquad \rho_t = ?$$

n -> n+1
=> not solvable
=> only up to *linear* terms in V

$$\boxed{\rho_t^D(t)} = \rho_0 + \int_{-\infty}^{t} \dot{\rho}_{t'}^D(t') \mathrm{d}t' = \rho_0 - \frac{i}{\hbar} \int_{-\infty}^{t} [V_{t'}^D(t'), \boxed{\rho_{t'}^D(t')}] \mathrm{d}t'$$

Dirac picture

=> $$\rho_t^D(t) \approx \rho_0 - \frac{i}{\hbar} \int_{-\infty}^{t} [V_{t'}^D(t'), \rho_0] \mathrm{d}t'$$ linear response transform back to Schrödinger picture:

$$\rho_t \approx \rho_0 - \frac{i}{\hbar}\int_{-\infty}^{t} e^{-\frac{i}{\hbar}\mathcal{H}_0 t}[V_{t'}^D(t'),\rho_0]e^{\frac{i}{\hbar}\mathcal{H}_0 t}\mathrm{d}t'$$

=> expectation value:

$$\langle\hat{A}\rangle_t = \langle\hat{A}\rangle_0 - \frac{i}{\hbar}\int_{-\infty}^{t} F_{t'}\,\mathrm{Tr}(\rho_0[\hat{A}^D(t),\hat{B}^D(t')])\mathrm{d}t'$$

=> $\Delta A_t = \langle\hat{A}\rangle_t - \langle\hat{A}\rangle_0 = \frac{1}{\hbar}\int_{-\infty}^{\infty} F_{t'}G_{AB}^{\mathrm{ret}}(t-t')\mathrm{d}t'$

Here, $G_{AB}^{\mathrm{ret}}(t-t') = -i\theta(t-t')\langle[A(t),B(t')]\rangle_0$ is the retarded Green's function.

Fourier transform: $G_{AB}^{\mathrm{ret}}(E) = \int_{-\infty}^{\infty} G_{AB}^{\mathrm{ret}}(t-t')e^{\frac{i}{\hbar}E(t-t')}\mathrm{d}(t-t')$

transform back to Schrödinger pict

$\rho_t \approx \rho$

Usually, the Fourier transform is easier to calculate. (simple equation of motion; use spectral moments)

spectral representation: $G^{\mathrm{ret}}_{AB}(E) = \int_{-\infty}^{\infty} \frac{S_{AB}(E')}{E - E' + i0^{+}} \mathrm{d}E'$

=> further helpful properties, e.g. Kramers-Kronig relations

further reading: Nolting 7, „Viel-Teilchen-Theorie“ (chapter 3)

=> e

$\langle A$

=> $\Delta A_t = \langle$

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Fourier transform: $G^{\mathrm{ret}}_{AB}(E) = \int_{-\infty}^{\infty} G^{\mathrm{ret}}_{AB}(t-t') e^{\frac{i}{\hbar}E(t-t')} \mathrm{d}(t-t')$

the interaction of light and Dirac fermions can be described by

$$H_{\text{int}} = -e\vec{E}(t)\cdot\hat{\vec{r}} = -ie(\vec{E}(t)\cdot\vec{\nabla}_{\vec{k}})$$

=> density matrix evolves as: $i\hbar\partial_t\rho_{\vec{k}} = [H,\rho_{\vec{k}}] = \hbar v_F\vec{k}\cdot[\vec{\sigma},\rho_{\vec{k}}] - ie\vec{E}(t)\cdot[\vec{\nabla}_{\vec{k}},\rho_{\vec{k}}]$

$$= \hbar v_F\vec{k}\cdot[\vec{\sigma},\rho_{\vec{k}}] - ie(\vec{E}(t)\cdot\vec{\nabla}_{\vec{k}})$$

moreover, the density matrix can be written as $\rho_{\vec{k}} = n_{\vec{k}}1 + \vec{m}_{\vec{k}}\cdot\vec{\sigma}$

=> $n_{\vec{k}} = \mathrm{Tr}(\rho_{\vec{k}})/2$ $\qquad$ $\vec{m}_{\vec{k}} = \mathrm{Tr}(\vec{\sigma}\rho_{\vec{k}})/2$

charge density $\qquad$ pseudospin density

$$\partial_t n_{\vec{k}} = -\frac{e}{\hbar}(\vec{E}(t)\cdot\vec{\nabla}_{\vec{k}})n_{\vec{k}} \qquad \partial_t\vec{m}_{\vec{k}} = 2v_F(\vec{k}\times\vec{m}_{\vec{k}}) - \frac{e}{\hbar}(\vec{E}(t)\cdot\vec{\nabla}_{\vec{k}})\vec{m}_{\vec{k}}$$

current operator: $\hat{\vec{j}} = \sum_{\vec{k}}\hat{\vec{j}}_{\vec{k}} = \sum_{\vec{k}}\frac{e}{\hbar}\psi^{\dagger}_{\vec{k}}\left(\frac{\delta}{\delta\vec{k}}\hbar v_F\vec{\sigma}\cdot\vec{k}\right)\psi_{\vec{k}} = \sum_{\vec{k}} ev_F\psi^{\dagger}_{\vec{k}}\vec{\sigma}\psi_{\vec{k}}$

=> $\langle\hat{\vec{j}}\rangle = \mathrm{Tr}(\hat{\vec{j}}\rho) = ev_F\sum_{\vec{k}}\mathrm{Tr}(\psi^{\dagger}_{\vec{k}}\vec{\sigma}\psi_{\vec{k}}\rho) = ev_F\sum_{\vec{k}}\mathrm{Tr}(\vec{\sigma}\rho_{\vec{k}}) = 2ev_F\sum_{\vec{k}}\vec{m}_{\vec{k}}$

unitary transformation: $\psi_{\vec{k}1} = \frac{\xi_{\vec{k}1} + \xi_{\vec{k}2}}{\sqrt{2}}$ $\psi_{\vec{k}2} = e^{i\phi_{\vec{k}}} \frac{\xi_{\vec{k}1} - \xi_{\vec{k}2}}{\sqrt{2}}$

=> $H_0 = \sum_{\vec{k}} \hbar v_F \begin{pmatrix} \psi_{\vec{k}1} \\ \psi_{\vec{k}2} \end{pmatrix}^\dagger \begin{pmatrix} 0 & k_x - ik_y \\ k_x + ik_y & 0 \end{pmatrix} \begin{pmatrix} \psi_{\vec{k}1} \\ \psi_{\vec{k}2} \end{pmatrix} = \sum_{\vec{k}} \hbar v_F k (\xi^\dagger_{\vec{k}1} \xi_{\vec{k}1} - \xi^\dagger_{\vec{k}2} \xi_{\vec{k}2})$

$k e^{\mp i\phi_{\vec{k}}}$

electron hole

occupation

=> $\langle \xi^\dagger_{\vec{k}i} \xi_{\vec{k}i} \rangle = f_{\vec{k}i}$ => $\boxed{\vec{m}^{(0)}_{\vec{k}} = \frac{\mathrm{Tr}(\vec{\sigma}\rho_{\vec{k}})}{2} = \frac{\vec{k}}{2k}(f_{\vec{k}1} - f_{\vec{k}2})}$

H_{int} => pseudospin density becomes $\boxed{\vec{m}_{\vec{k}}(t) = \vec{m}^{(0)}_{\vec{k}} + \delta\vec{m}_{\vec{k}} e^{-i\omega t}}$

assume $\boxed{\vec{E}(t) = \vec{E} e^{-i\omega t} \text{ and } \delta\vec{m}_{\vec{k}} \propto \vec{E} \propto \vec{e}_x}$

[previous slide] & ... => $-i\omega\delta\vec{m}_{\vec{k}} = 2v_F(\vec{k} \times \delta\vec{m}_{\vec{k}}) - \frac{e}{\hbar}(\vec{E} \cdot \vec{\nabla}_{\vec{k}})\vec{m}^{(0)}_{\vec{k}}$

resolve $\delta m^x_{\vec{k}}$ and use $j_x = 2ev_F \sum_{\vec{k}} \delta m^x_{\vec{k}} = \sigma(\omega) E$

=> optical conductivity $\sigma(\omega)$ =...

$$\sigma(\omega) = -\frac{8ie^2v^3}{\hbar\omega}\sum_{\vec{k}} \frac{k_y}{\omega^2 - 4v^2k^2}\left(k_y \frac{\partial m_{\vec{k}}^{x(0)}}{\partial k_x} - k_x \frac{\partial m_{\vec{k}}^{y(0)}}{\partial k_x}\right). \tag{7.31}$$

On substituting Eq. (7.26) into Eq. (7.31) we find

$$\begin{aligned}\sigma(\omega) &= -\frac{4ie^2v^3}{\hbar\omega}\sum_{\vec{k}} \frac{k_y^2}{\omega^2 - 4v^2k^2}\frac{1}{k}(f_{\vec{k}1} - f_{\vec{k}2}) \\ &= -\frac{2ie^2v^3}{\hbar\omega}\sum_{\vec{k}} \frac{k(f_{\vec{k}1} - f_{\vec{k}2})}{\omega^2 - 4v^2k^2}.\end{aligned} \tag{7.32}$$

switch perturbation on adiabatically

retarded Green's function => substitute $\omega \to \omega + i0^+$

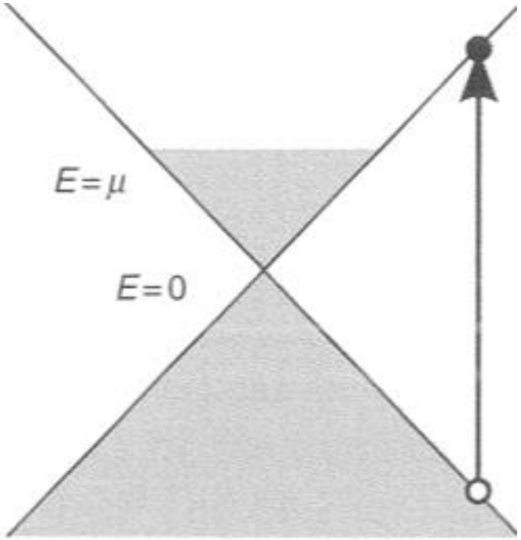
use $$\frac{1}{(\omega + i0^+)^2 - 4v_F^2k^2} = \mathcal{P}\frac{1}{\omega^2 - 4v_F^2k2} - \frac{i\pi\delta(\omega - 2v_Fk)}{4v_Fk}$$

where $\mathcal{P}\int_a^b f(x)\mathrm{d}x := \lim_{\epsilon\to 0^+}\left(\int_a^{c-\epsilon} f(x)\mathrm{d}x + \int_{c+\epsilon}^b f(x)\mathrm{d}x\right)$

defines the principal value of f(x) (Riemann-integrable in (a,c)u(c,b))

=> $$\mathrm{Re}\ \sigma(\omega) = \frac{\pi e^2 v_F^2}{2\hbar\omega}\sum_{\vec{k}}(f_{\vec{k}1} - f_{\vec{k}2})\delta(\omega - 2v_Fk) = \frac{e^2}{16\hbar}\left(f(-\hbar\omega/2) - f(\hbar\omega/2)\right)$$

include spin and valley degeneracies:



x4 => $$\mathrm{Re}\ \sigma(\omega) = \begin{cases} 0, & \omega < 2|\mu| \\ \underbrace{\frac{e^2}{4\hbar}}_{=\sigma_0 \text{ universal conductivity}}, & \omega > 2|\mu| \end{cases}$$

With the Kramers-Kronig relation

$$\mathrm{Im}\ G_{AB}^{\mathrm{ret}}(E) = \frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty} \frac{\mathrm{Re}\ G_{AB}^{\mathrm{ret}}(E')}{E - E'} \mathrm{d}E'$$

the imaginary part of the conductivity can be found:

$$\mathrm{Im}\ \sigma(\omega) = \frac{\sigma_0}{\pi}\left(\frac{4\mu}{\hbar\omega} - \ln\left|\frac{\hbar\omega + 2\mu}{\hbar\omega - 2\mu}\right|\right) \quad .$$

Note that at $\mu \to 0$, $\mathrm{Im}\ \sigma(\omega)$ vanishes for all frequencies ω (i.e. no energy loss within the graphene sheet).

The response of a material to an external charge density is described by the *dielectric function*.

$$\rho_{\mathrm{ind}}(\vec{q},E) = \left(\frac{1}{\epsilon(\vec{q},E)} - 1\right)\rho_{\mathrm{ext}}(\vec{q},E)$$

induced charge density

dielectric function

external charge density

$$\frac{1}{\epsilon(\vec{q},E)} = 1 + \frac{e^2}{V\epsilon_0 q^2} G^{\mathrm{ret}}_{\rho_{\vec{q}},\rho^\dagger_{\vec{q}}}(E)$$

In a metal, the induced charge density will typically screen the external charge density: $\epsilon(\vec{q},E) \gg 1$ => $\rho_{\mathrm{ind}}(\vec{q},E) \approx -\rho_{\mathrm{ext}}(\vec{q},E)$.

In the opposite limit, $\epsilon(\vec{q},E) \to 0$, arbitrarily small perturbations suffice to induce finite charge fluctuations within the conduction electron system.

That is, the poles of $G^{\mathrm{ret}}_{\rho_{\vec{q}},\rho^\dagger_{\vec{q}}}(E)$ correspond to resonances of that system. These collective excitations of the electron system are called *plasmons*.

In graphene, plasmons exist for $\hbar\omega \ll 2\mu$

and disperse like $\omega = \sqrt{\frac{2e^2\mu}{\hbar^2\epsilon_{\text{ext}}} q}$.

The dispersion $\omega \propto \sqrt{q}$ is a general property of plasmons in two-dimensional electron gases. However, the dependence on the electron density is special for graphene:

$\omega \propto \sqrt{\mu} \propto n^{1/4}$: graphene,
$\omega \propto n^{1/2}$: nonrelativistic electrons.

Apart from $qv_F < \omega < 2\mu$, the dielectric function has a large imaginary part such that the plasmons are suppressed due to damping.

> Each graphene layer absorbs $2.3\% = \pi\alpha$ of incoming light (independent of the photon frequency).

> Read Nolting 7, chapter 3.

> The universal conductivity of graphene is $\sigma_0 = \frac{e^2}{4\hbar}$.

> Plasmons are collective (=> quasiparticles) charge density fluctuations of the conduction electron system. Their energies are the poles of the dielectric function.

Literature:

[1] Mikhail I. Katsnelson, *Graphene*, Cambridge University Press (2012).
[2] R. Nair *et al.*, Science **320**, 1308 (2008).
[3] Wolfgang Nolting, *Grundkurs Theoretische Physik 7*, Springer (2005).
[4] J. D. Jackson, *Classical Electrodynamics*, Wiley & Sons (1962).