Terahertz Response of Monolayer Graphene: Velocity Gauge Vs Length Gauge

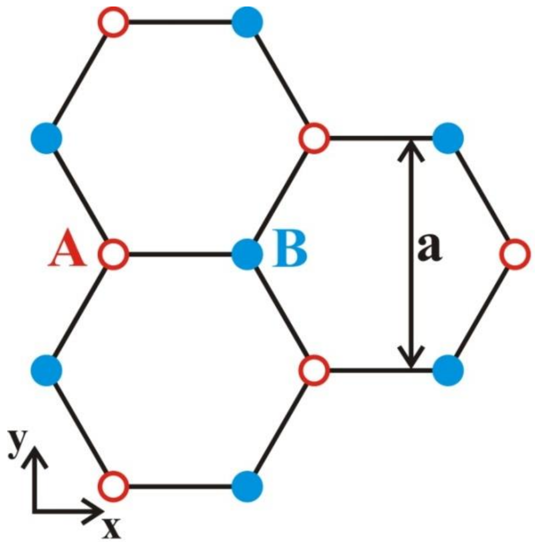
P. Navaeipour, M. M. Dignam

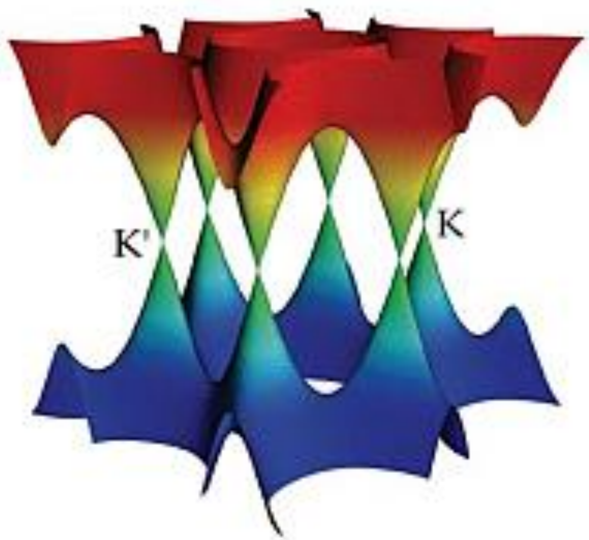
Department of Physics, Engineering Physics and Astronomy, Queen’s University, Kingston ON K7L 3N6, Canada

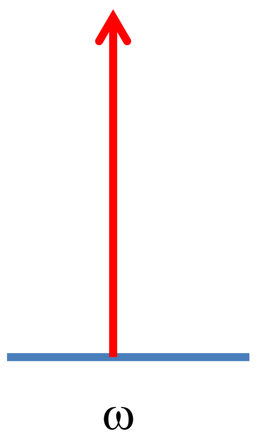
NSERC
CRSNG

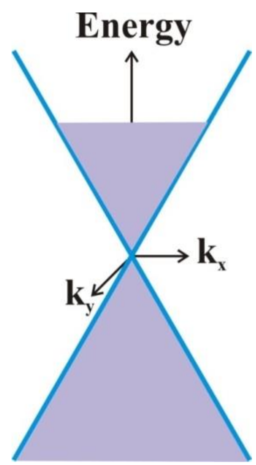
Queen’s UNIVERSITY

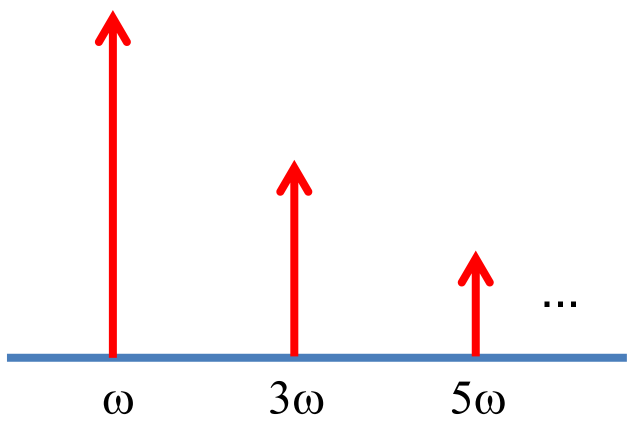
Basics of Graphene











S. A. Mikhailov, Europhysics Lett. 79, 27002 (2007)

Motivation & Outline

- Find the best method to model the nonlinear response of Graphene
- Velocity Gauge:

  F.H.M. Faisal, arXiv, 1, 0810.07881 (2008)

  A.R. Wright, Appl. Phys. 44, 083001 (2011)

- Length Gauge:

  C. Aversa and J. E. Sipe, Phys. Rev. B 52, 14636 (1995)

  K. S. Virk and J. E. Sipe, Phys. Rev. B 76, 035213 (2007)

- Response of Graphene to THz field by employing these two gauges

Velocity & Length Gauges

- Hamiltonian in the present of electric field:

$$\mathrm{H} = \mathrm{H}_0 + \mathrm{H}'$$

- Hamiltonian of the system in the absence of electric field:

$$\mathrm{H}_0 = \frac{\mathrm{p}^2}{2\mathrm{m}} + \mathbf{V}(\mathbf{r})$$

- H' is the interaction Hamiltonian.

$$\mathrm{H}' = -\frac{e}{m}\,\boldsymbol{p}.\boldsymbol{A}(t)$$ Velocity Gauge

$$\mathrm{H}' = -e\,\boldsymbol{r}.\boldsymbol{E}(\boldsymbol{r},t)$$ Length gauge

A. Chacona, J Computational Physics.1 ,1508 (2015)

Gauge Choice

Velocity vs. **Length**

- Quantum mechanics is gauge invariant and both gauges should give, in principle, identical results

- Convergence of dynamic equations using the velocity gauge requires a large number of bands

- Divergences at zero frequency arise using the velocity gauge in the nonlinear response

- The divergence can be removed by developing sum rules, which become extremely complicated at high-order in the field

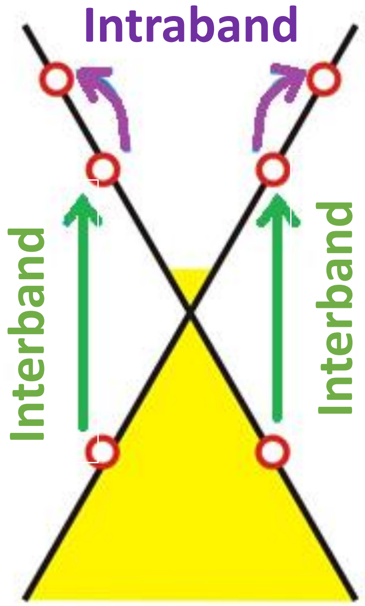
C. Aversa and J. E. Sipe, Phys. Rev. B 52, 14636 (1995)
K. S. Virk and J. E. Sipe, Phys. Rev. B 76, 035213 (2007)

Populations Dynamic Equations

Length Gauge:

$$\frac{d\rho_{nn}(\mathbf{k})}{dt} = -\frac{i\sigma_n e \cdot \mathbf{E}(t)}{\hbar}\left[\boldsymbol{\xi}_{cv}(\mathbf{k})\,\rho_{vc}(\mathbf{k}) - \boldsymbol{\xi}_{vc}(\mathbf{k})\,\rho_{cv}(\mathbf{k})\right]$$

$$-\frac{e\mathbf{E}(t)\cdot\nabla_{\mathbf{k}}\rho_{nn}(\mathbf{k})}{\hbar} - \frac{\left[\rho_{nn}(\mathbf{k}) - f_n(\mathbf{k},t)\right]}{\tau_n}$$



Velocity Gauge:

$$\frac{d\rho_{nn}(\boldsymbol{k})}{dt} = \frac{e}{\hbar}\sigma_n\,\omega_{cv}(\boldsymbol{k})(\boldsymbol{\xi}_{cv}(\boldsymbol{k})\rho_{vc}(\boldsymbol{k}) + \rho_{cv}(\boldsymbol{k})\boldsymbol{\xi}_{vc}(\boldsymbol{k})).\boldsymbol{A}(t)$$

$$-\frac{\rho_{nn}(\boldsymbol{k}) - f_n(\boldsymbol{k},t)}{\tau_n}$$

$$\boldsymbol{\xi}_{nm}(\mathbf{k}) = \left[2\delta_{n,m} - 1\right]\frac{\widehat{\boldsymbol{\theta}}}{2k}$$

$$\boldsymbol{A} = -\frac{i\boldsymbol{E}}{\omega}$$

I. Al-Naib et al., Phys. Rev. B 90, 245423, 2014

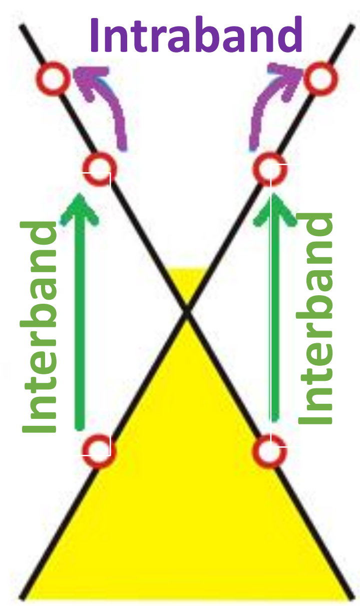
Coherences Dynamic Equations

Length Gauge:

$$\frac{d\rho_{cv}(\mathbf{k})}{dt} = \frac{ie\mathbf{E}(t)\cdot\boldsymbol{\xi}_{cv}(\mathbf{k})}{\hbar}\left[\rho_{vv}(\mathbf{k}) - \rho_{cc}(\mathbf{k})\right] - i\omega_{cv}(\mathbf{k})\,\rho_{cv}(\mathbf{k}) - \frac{e\mathbf{E}(t)\cdot\nabla_{\mathbf{k}}\rho_{cv}(\mathbf{k})}{\hbar} - \frac{\rho_{cv}(\mathbf{k})}{\tau}$$

Velocity Gauge:

$$\frac{d\rho_{cv}(\boldsymbol{k})}{dt} = -\frac{e\omega_{cv}(\boldsymbol{k})\mathbf{A}(t)\cdot\xi_{cv}(\boldsymbol{k})}{\hbar}\left(\rho_{vv}(\boldsymbol{k}) - \rho_{cv}(\boldsymbol{k})\right) - i\omega_{cv}(\boldsymbol{k})\rho_{cv}(\boldsymbol{k}) + \frac{ie\boldsymbol{A}(t)}{\hbar}\cdot\nabla_{\boldsymbol{k}}\,\omega_{cv}(\boldsymbol{k})\rho_{cv}(\boldsymbol{k}) - \frac{\rho_{cv}(\boldsymbol{k})}{\tau}$$



I. Al-Naib et al., Phys. Rev. B 90, 245423, 2014

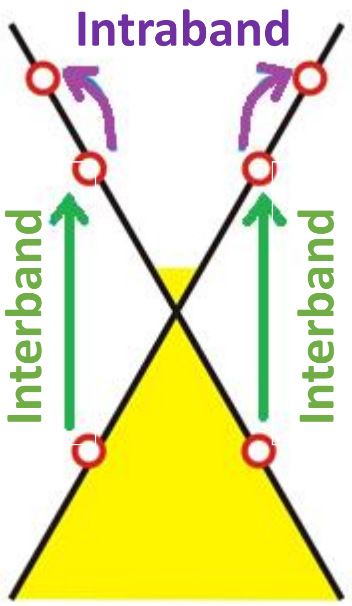
Current Density in Length Gauge

Interband Current:

$$\mathbf{J}_e(t) = \frac{8|e|}{A} Re\left\{\sum_{\mathbf{k}} \frac{\widehat{\boldsymbol{\theta}}}{2k} \frac{d\rho_{cv}(\mathbf{k},t)}{dt}\right\}$$

Intraband Current:

$$\mathbf{J}_i(t) = \frac{-4|e|v_F}{A} \sum_{\mathbf{k}} \{\rho_{cc}(\mathbf{k},t) - \rho_{vv}(\mathbf{k},t)\} \widehat{\mathbf{k}} + \frac{8|e|^2}{A\hbar} \sum_{\mathbf{k}} Re\{\rho_{cv}(\mathbf{k},t) \boldsymbol{\nabla}_{\mathbf{k}} [\mathbf{E}(t) \cdot \boldsymbol{\xi}_{vc}(\mathbf{k})]\}$$



I. Al-Naib et al., Phys. Rev. B 90, 245423, 2014

Current Density in Velocity Gauge

Interband Current:

$$\boldsymbol{J}_e(t) = \frac{4ie}{A}\sum_{\boldsymbol{k}}\left(\boldsymbol{\xi}_{cv}(\boldsymbol{k})\rho_{vc}(\boldsymbol{k}) - \boldsymbol{\xi}_{vc}(\boldsymbol{k})\rho_{cv}(\boldsymbol{k})\right)\omega_{cv}(\boldsymbol{k})$$

$$-\frac{4e^2}{\hbar}\sum_{\boldsymbol{k}}\omega_{cv}(\boldsymbol{k})\left(\xi^i_{cv}(\boldsymbol{k})\xi^j_{vc}(\boldsymbol{k}) + \xi^j_{cv}(\boldsymbol{k})\xi^i_{vc}(\boldsymbol{k})\right)\boldsymbol{A}(t)\left(\rho_{vv}(\boldsymbol{k},t) - \rho_{cc}(\boldsymbol{k},t)\right)$$

$$+\frac{4e^2}{\hbar}\sum_{\boldsymbol{k}}\left(\rho_{vc}(\boldsymbol{k},t)\xi_{cv}(\boldsymbol{k}) - \rho_{cv}(\boldsymbol{k},t)\xi_{vc}(\boldsymbol{k})\right)\nabla_k\omega_{cv}(\boldsymbol{k})\cdot\boldsymbol{A}(t)$$

Intraband Current:

$$\boldsymbol{A} = -\frac{i\boldsymbol{E}}{\omega}$$

$$\boldsymbol{J}_i(t) = -\frac{4ev_f}{A}\sum_{\boldsymbol{k}}\{\rho_{cc}(\boldsymbol{k},t) - \rho_{vv}(\boldsymbol{k},t)\}\widehat{\boldsymbol{k}}$$

$$+\frac{8e^2}{\hbar A}\sum_{\boldsymbol{k}} Im\,\{\rho_{cv}(\boldsymbol{k},t)\nabla_{\boldsymbol{k}}[\omega_{cv}(\boldsymbol{k})\boldsymbol{\xi}_{vc}(\boldsymbol{k})]\cdot\boldsymbol{A}(t)\}$$

$$-\frac{e^2}{A}\sum_{\boldsymbol{k}}\frac{1}{m}\{\rho_{vv}(\boldsymbol{k},t) + \rho_{cc}(\boldsymbol{k},t)\}\boldsymbol{A}(t)$$

Current Density

Effective Mass Sum Rule:

$$\frac{1}{m} = \frac{1}{\hbar^2}\frac{\partial^2 E_n(\boldsymbol{k})}{\partial k^x \partial k^j} - \sum_{m\neq n} \frac{\omega_{nm}^2(\boldsymbol{k})\left(\boldsymbol{\xi}_{nm}^x(\boldsymbol{k})\boldsymbol{\xi}_{mn}^j(\boldsymbol{k}) + \boldsymbol{\xi}_{nm}^j(\boldsymbol{k})\boldsymbol{\xi}_{mn}^x(\boldsymbol{k})\right)}{m^2\left(E_m(\boldsymbol{k}) - E_n(\boldsymbol{k})\right)}$$

Total Current Density:

$$\boldsymbol{J}(t) = \frac{4ie}{A}\sum_{k}\left(\boldsymbol{\xi}_{cv}(\boldsymbol{k})\rho_{vc}(\boldsymbol{k}) - \boldsymbol{\xi}_{vc}(\boldsymbol{k})\rho_{cv}(\boldsymbol{k})\right)\omega_{cv}(\boldsymbol{k})$$
$$+ \frac{4e^2}{A\hbar}\sum_{k}\left(\rho_{vc}(\boldsymbol{k},t)\xi_{cv}(\boldsymbol{k}) - \rho_{cv}(\boldsymbol{k},t)\xi_{vc}(\boldsymbol{k})\right)\nabla_k\omega_{cv}(\boldsymbol{k})\cdot\boldsymbol{A}(t)$$
$$- \frac{4ev_f}{A}\sum_{\boldsymbol{k}}\{\rho_{cc}(\boldsymbol{k},t) - \rho_{vv}(\boldsymbol{k},t)\}\hat{\boldsymbol{k}}$$
$$+ \frac{8e^2}{A\hbar}\sum_{\boldsymbol{k}} Im\,\{\rho_{cv}(\boldsymbol{k},t)\nabla_{\boldsymbol{k}}[\omega_{cv}(\boldsymbol{k})\boldsymbol{\xi}_{vc}(\boldsymbol{k})]\cdot\boldsymbol{A}(t)\}$$
$$- \frac{4e^2}{A\hbar}\sum_{\boldsymbol{k}}\left(\frac{\partial^2\omega_v(\boldsymbol{k})}{\partial\boldsymbol{k}^2}\,\rho_{vv}(\boldsymbol{k},t) + \frac{\partial^2\omega_c(\boldsymbol{k})}{\partial\boldsymbol{k}^2}\,\rho_{cc}(\boldsymbol{k},t)\right)\boldsymbol{A}(t)$$

J. Callaway, Academic Press, Second edition, (1974)

First order Intraband Conductivity

- **Length Gauge:**

$$\sigma_i^{(1)}(\omega) = \frac{2ie^2k_BT}{\pi\hbar^2(\omega + \frac{i}{\tau})}\ln(2\cosh(\frac{\beta\mu}{2}))$$

- **Velocity Gauge:**

$$\sigma_i^{(1)}(\omega) = \frac{2ie^2k_BT}{\pi\hbar^2\omega}\ln(2\cosh(\frac{\beta\mu}{2}))$$

I. Al-Naib et al., Phys. Rev. B 90, 245423, 2014

First Order Interband Conductivity

- **Length Gauge:**

$$\sigma_e^{(1)} = -\frac{ie^2}{4\pi\hbar}\int \frac{\omega}{\omega_{cv}} \frac{d\omega_{cv}}{(\omega_{cv} - \omega - \frac{i}{\tau})} \frac{\sinh\beta(\frac{\hbar\omega_{cv}}{2})}{\cosh\beta\mu + \cosh\beta(\frac{\hbar\omega_{cv}}{2})}$$

- **Velocity Gauge:**

$$\sigma_e^{(1)} = -\frac{ie^2}{4\pi\hbar}\int \frac{\omega_{cv}}{\omega} \frac{d\omega_{cv}}{(\omega_{cv} - \omega - \frac{i}{\tau})} \frac{\sinh\beta(\frac{\hbar\omega_{cv}}{2})}{\cosh\beta\mu + \cosh\beta(\frac{\hbar\omega_{cv}}{2})}$$

- $\tau \to \infty$

$$\sigma_e^{(1)} = \frac{e^2}{4\hbar}$$

Summary

- If one uses the mass sum rule and neglects scattering, two gauges yield identical linear conductivities.
- If we consider scattering term, even the linear interband conductivity is quite different in two gauges.
- One should do calculations in the interaction picture when employing velocity gauge.
- Nonlinear response can be quite different for the two approaches, due to the diverges arise at zero frequency in the velocity gauge when one uses a basis with a finite number of bands.
- One should use the length gauge for graphene when calculating the nonlinear THz response.

Thank You!

Current

- $\boldsymbol{J}(t) = e\int \frac{d\vec{k}}{(2\pi)^2} V_{nm}(k)\rho_{nm}(k,t)$
- $V_{nm}(k) = \boldsymbol{v}_{nm}(k) - e\frac{1}{m}\delta_{nm}\boldsymbol{A}(t)$
- $\langle n\mathbf{k}|\mathbf{r}\,|m\mathbf{k}'\rangle = \delta(\mathbf{k}-\mathbf{k}')\boldsymbol{\xi}_{nm}(\mathbf{k}) + i\delta_{nm}\boldsymbol{\nabla}_{\mathbf{k}}\delta(\mathbf{k}-\mathbf{k}')$

Last term in the intraband current:

$$\frac{4ie^2}{A}\sum_{\boldsymbol{k}}\frac{1}{m\omega}\{\rho_{cc}(\boldsymbol{k},t)+\rho_{vv}(\boldsymbol{k},t)\}\boldsymbol{E}(t)$$

$$\boldsymbol{E} = \boldsymbol{e}_x E_0 e^{-i\omega t} + \boldsymbol{e}_y E_0 e^{-i\omega t}$$

Mass Sum Rule

- $$\frac{1}{m} = \frac{1}{\hbar}\frac{\partial^2 E_n(k)}{\partial k^x \partial k^j} + \sum_{m\neq n} \frac{p_{nm}^x(k)p_{mn}^j(k)+p_{nm}^j(k)p_{mn}^x(k)}{m^2(E_m(k)-E_n(k))}$$

$$\langle n,k|p_e|m,k'\rangle = im\omega_{nm}(k)\langle n,k|r_e|m,k'\rangle$$
$$\langle n,k|r_e|m,k'\rangle = (1-\delta_{nm})\delta(k-k')\xi_{nm}(k,k')$$

$$\langle n\mathbf{k}|\mathbf{r}\,|m\mathbf{k}'\rangle = \delta(\mathbf{k}-\mathbf{k}')\boldsymbol{\xi}_{nm}\,(\mathbf{k}) + i\delta_{nm}\boldsymbol{\nabla}_{\mathbf{k}}\delta(\mathbf{k}-\mathbf{k}')$$

Linear Response

Velocity Gauge

$$E_c(k) = \hbar \mathrm{v_f} k \qquad \& \qquad E_v = -\hbar \mathrm{v_f} k$$

$$J_i^{(1)} = -\frac{4e^2 v_f}{\hbar A} \sum_{\boldsymbol{k}} \frac{1}{k} \left(1 - \frac{k_x^2}{k^2}\right) \left(1 - \rho_{hh}^{(0)}(\boldsymbol{k}) + \rho_{cc}^{(0)}(\boldsymbol{k})\right) A(t)$$

$$J_e^{(1)} = -\frac{8e}{A} \sum_{\boldsymbol{k}} \omega_{cv}(\boldsymbol{k}, t) Im\{\rho_{cv}^{(1)}(\boldsymbol{k}, t) \xi_{vc}(\boldsymbol{k}, t)\}$$

Length Gauge

$$J_i^{(1)} = 4ev_f \sum_{\boldsymbol{k}} \{ \rho_{cc}^{(1)}(\boldsymbol{k}) + \rho_{hh}^{(1)}(\boldsymbol{k})\} \hat{k}$$

$$J_e^{(1)} = 8e \, Re \sum_{\boldsymbol{k}} \{\boldsymbol{\xi}_{cv}(\boldsymbol{k}) \, \dot{\rho}_{cc}^{(1)}(\boldsymbol{k})\}$$

First order Intraband conductivity (ω)

- Velocity Gauge:

$$\sigma_i^{(1)}(\omega) = \frac{2ie^2k_BT}{\pi\hbar^2\omega}\ln(2\cosh(\frac{\beta\mu}{2}))$$

- Length Gauge

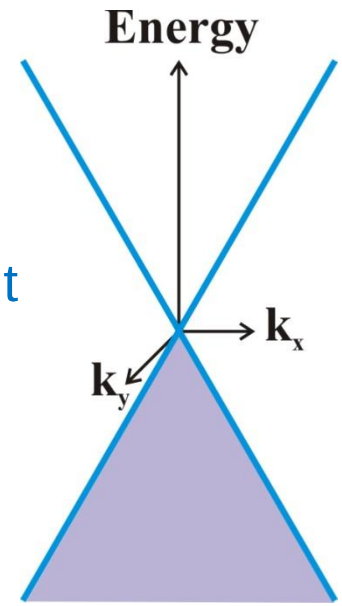
$$\sigma_i^{(1)}(\omega) = \frac{2ie^2k_BT}{\pi\hbar^2(\omega+\frac{i}{\tau})}\ln(2\cosh(\frac{\beta\mu}{2}))$$

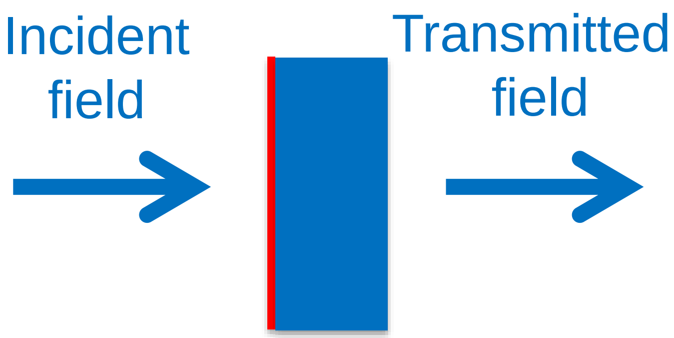
Outline

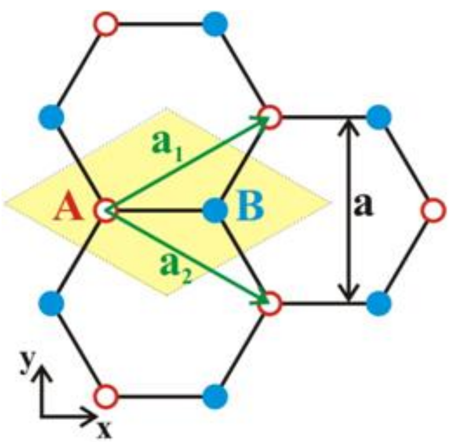
- **Motivation**
- **Velocity gauge and Length Gauge**
- **Comparing response of Graphene in Velocity gauge and Length gauge**
- **Conclusion**

System Under Study

- Graphene on a substrate
- Fermi level at the Dirac point
- Temperature of 30 K
- Scattering time of 50 fs







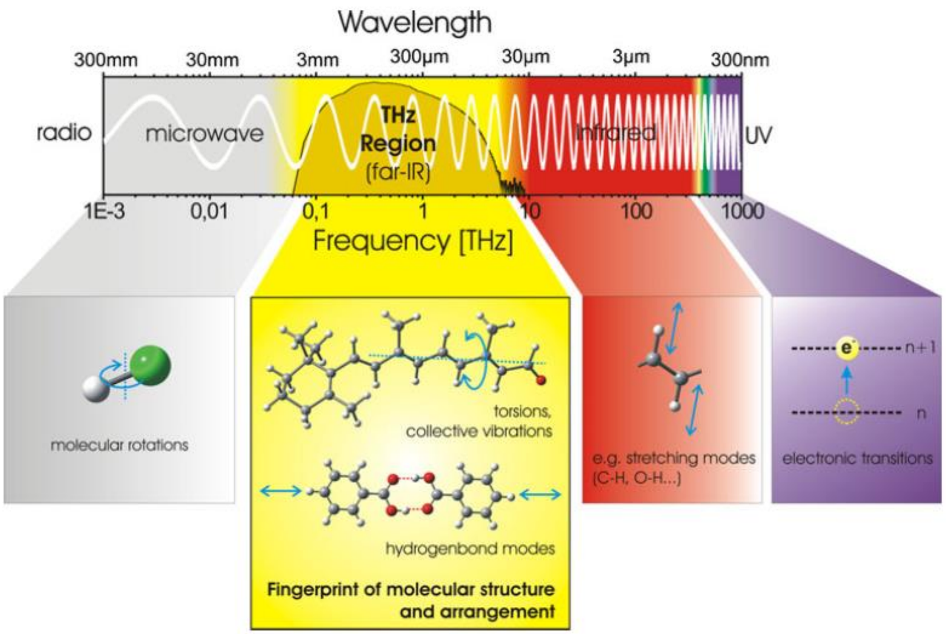
$$\phi_{n\mathbf{k}}(\mathbf{r}) = N\sum_{\mathbf{R}}[\varphi_{pz}(\delta\mathbf{r}_A) + \sigma_n e^{-i\chi(\mathbf{k})}\varphi_{pz}(\delta\mathbf{r}_B)]e^{i\mathbf{k}\cdot\mathbf{R}}$$

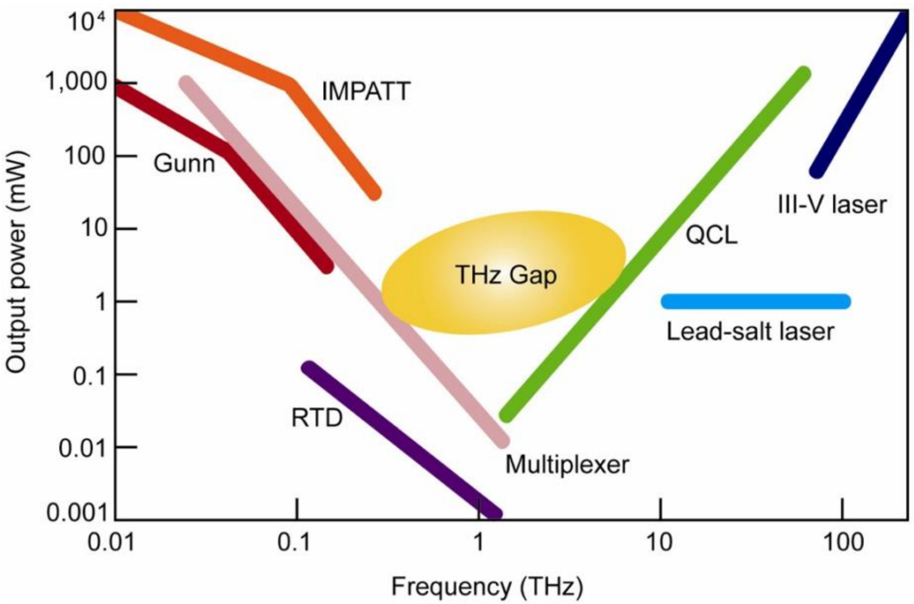
$$\sigma_v = 1 \text{ and } \sigma_c = -1$$

$$\chi(\mathbf{k}) = \arg[f(\mathbf{k})]$$

$$f(\mathbf{k}) \equiv 1 + e^{-i\mathbf{k}\cdot\mathbf{a}_1} + e^{-i\mathbf{k}\cdot\mathbf{a}_2}$$

Introduction: THz Band

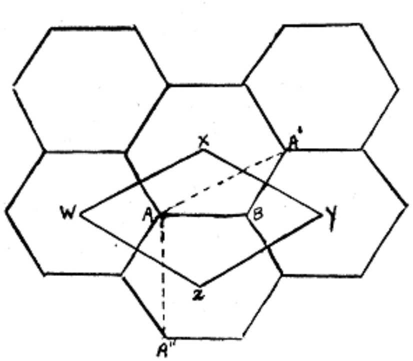




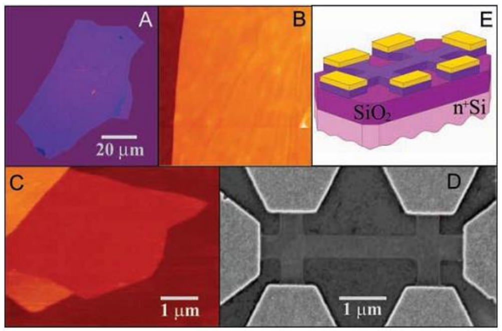
Moloney *et al.*, DOI: 10.1117/2.1201102.003523

➢ Numerous organic molecules exhibit strong absorption and dispersion due to rotational and vibrational transitions

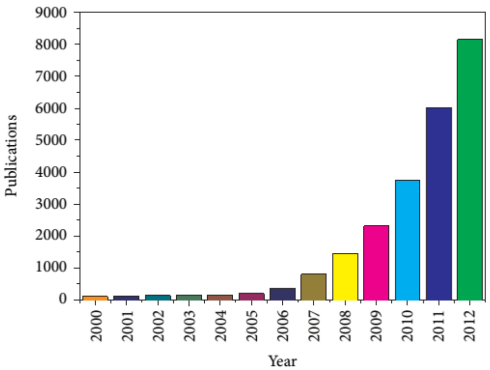
Introduction: Graphene



P. R. Wallace, Physical Review 71, 622 (1947)

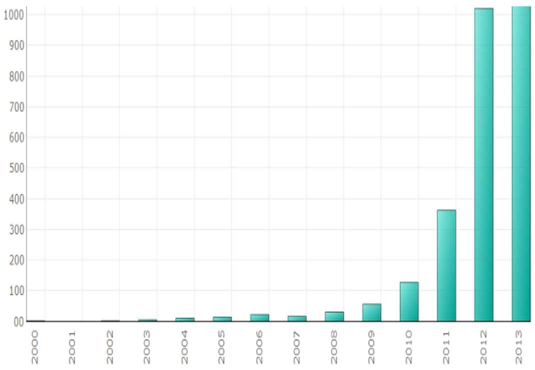


K. S. Novoselov *et al.*, Science 306, 666 (2004)



Publication trend

V. Dhand *et al.*, J. of Nanomaterials 763953 (2013)



Patents trend

Technology Insight Report: Graphene (2013)

First order Intraband Conductivity (T$\rightarrow 0$)

Velocity Gauge in the Schrodinger Picture:

$$\sigma_i^{(1)} = \frac{ie^2 E_f}{\pi\hbar^2\omega}$$

Velocity Gauge in the Interaction Picture:

$$\sigma_i^{(1)} = \frac{ie^2 E_f}{\pi\hbar^2(\omega + \frac{i}{\tau_c})}$$

Length gauge in the Schrodinger picture:

$$\sigma_i^{(1)} = \frac{ie^2 E_f}{\pi\hbar^2(\omega + \frac{i}{\tau_c})}$$

I. Al-Naib et al., Phys. Rev. B 90, 245423, 2014