

Enhanced dichroism in graphene nanoribbons

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Graphene Nanophotonics, Benasque 2013



NGS Graduate School for
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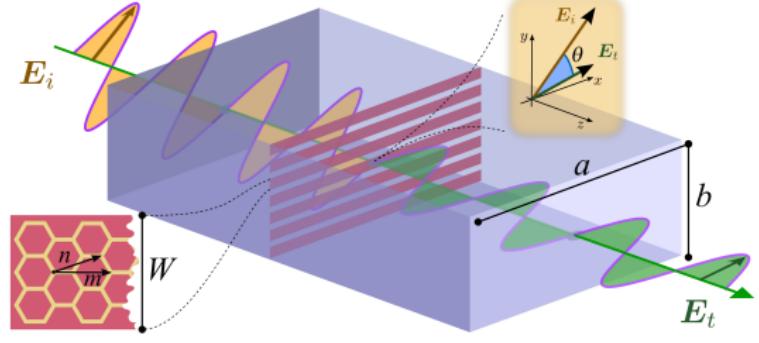
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Graphene nanoribbons

- Lateral confinement effects
- Conductivity
- Dichroic absorption

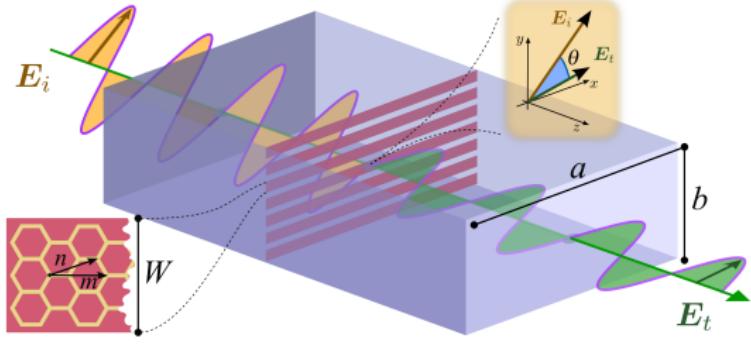


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Relevant issues

- Disorder
- Drude conductivity
- State of the art



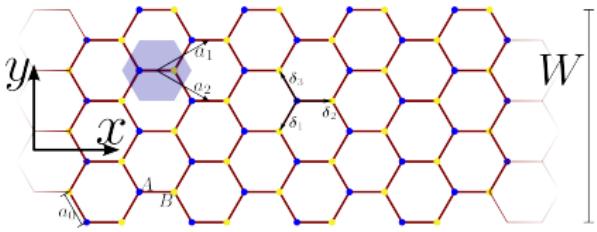
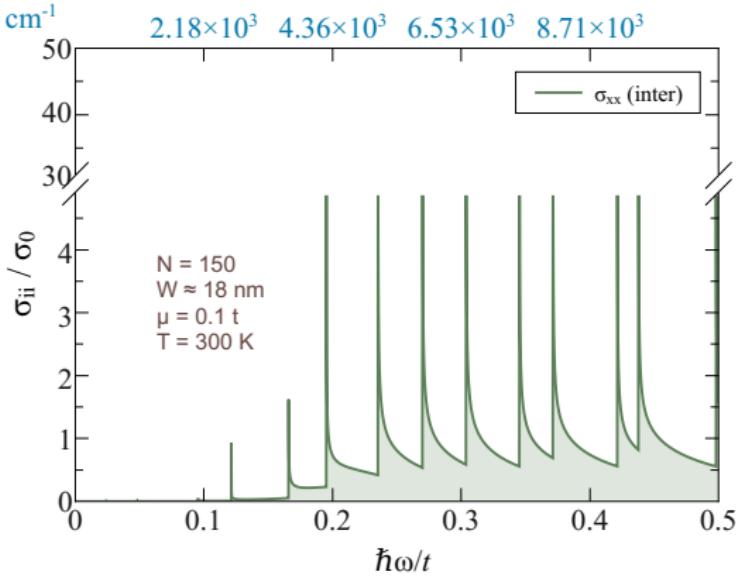
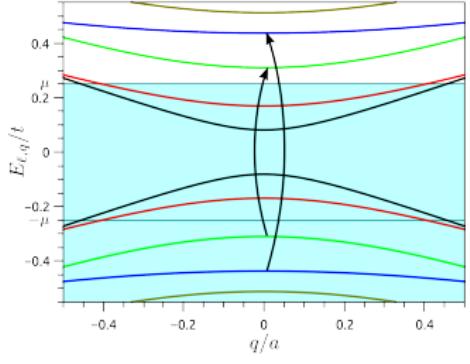
Nanoribbons

- $k_\ell = \frac{\pi\ell}{N+1}$ Saito et al. 1992; Wakabayashi et al. 2010; Ruseckas et al. 2011
- Van Hove singularities
- Kubo formula

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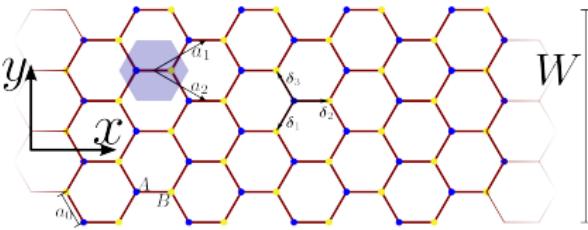
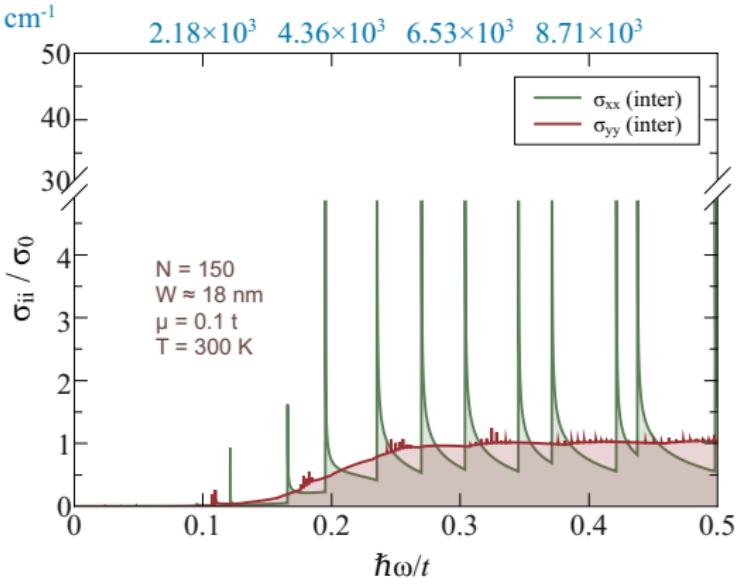
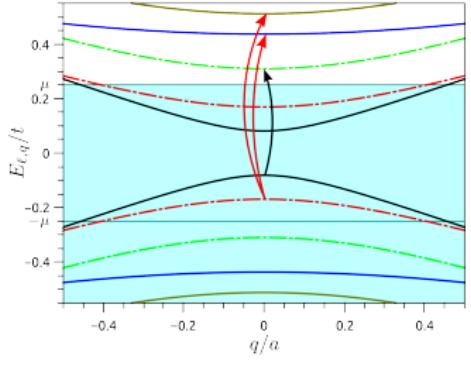
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- Selection rules for transitions



Graphene nanoribbons

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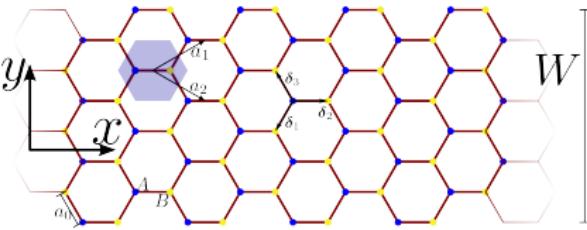
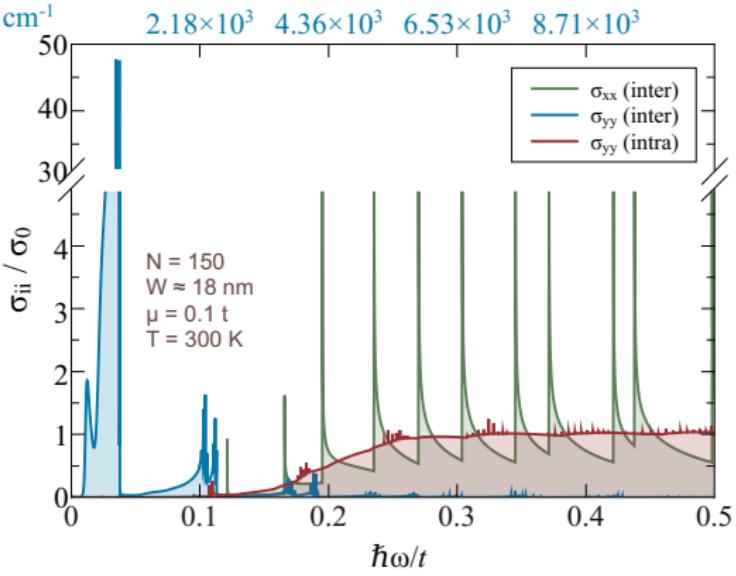
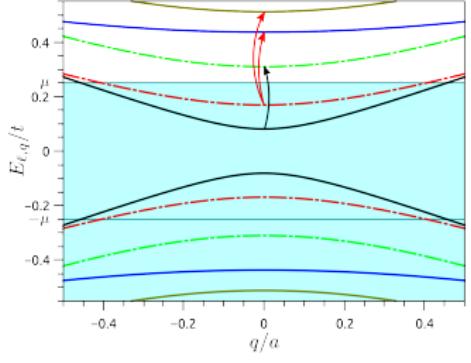
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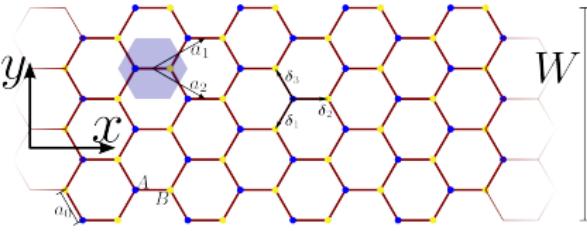
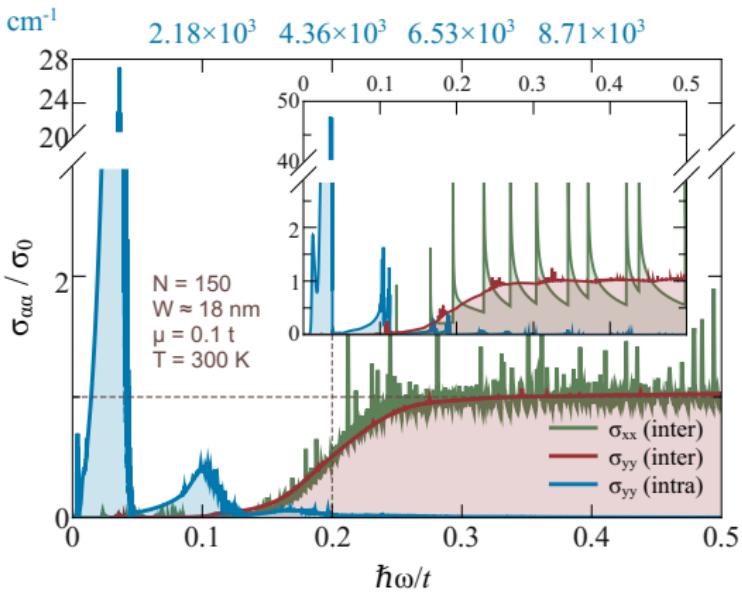
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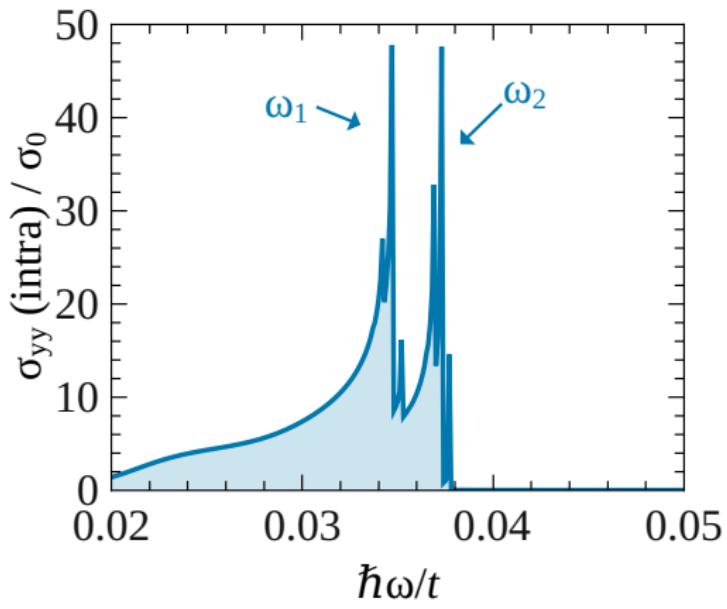
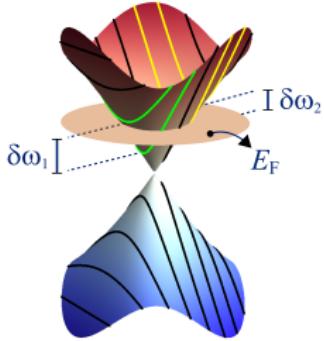
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intraband peak is resistant to level broadening



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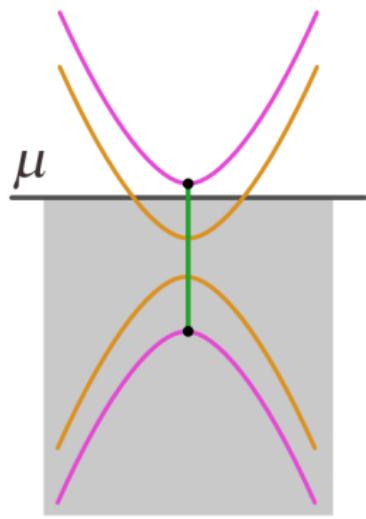
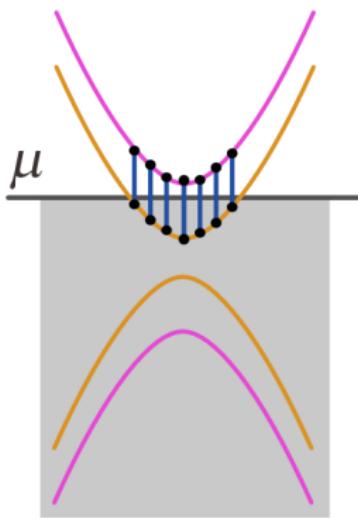
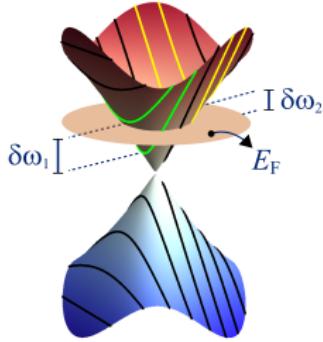


Close-up of the *intra*-band conductivity

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Independent of chirality

Absorption

- $t_\alpha(\omega) = \frac{2}{2+Z\langle\sigma_{\alpha\alpha}(\omega)\rangle}$

- $\mathcal{P}(\omega) = \frac{|t_x|^2 - |t_y|^2}{|t_x|^2 + |t_y|^2}$

Born et al. 1997; Jackson 1999

Dichroic absorption

Absorption

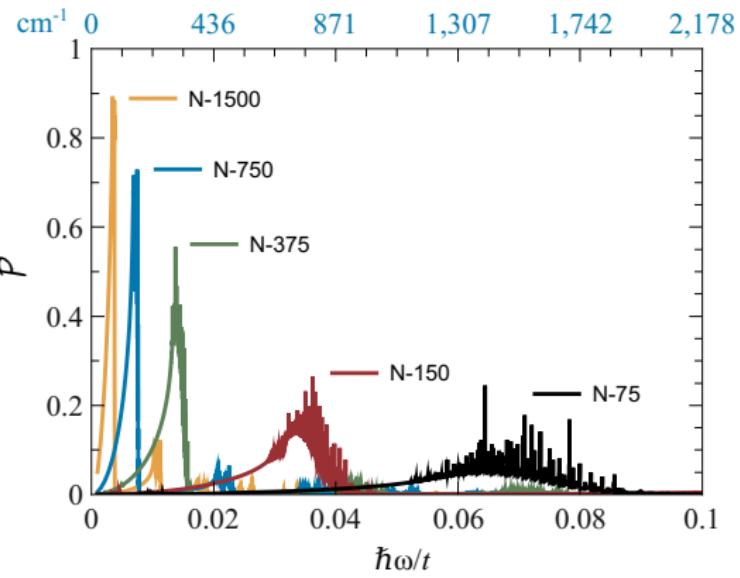
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Born et al. 1997; Jackson 1999

Dichroism

- \mathcal{P} in excess of 50% with 45 nm ribbons
- Predictability
- Narrower ribbons \rightarrow larger ω_{\max}
 \rightarrow smaller $\mathcal{P}(\omega_{\max})$



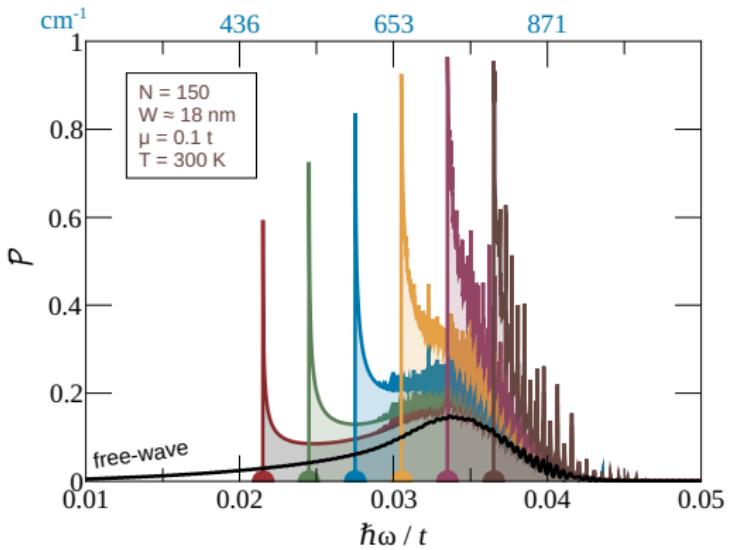
Nanoribbons

- $Z_{mm}(\omega) = Z \frac{\omega}{\sqrt{\omega^2 - \omega_{mn}^2}}$
 $\omega_{mn}^2 = 2 \frac{e^2 \pi^2}{\mu \epsilon} \left(\frac{m}{a} \right)^2$ Jackson 1999

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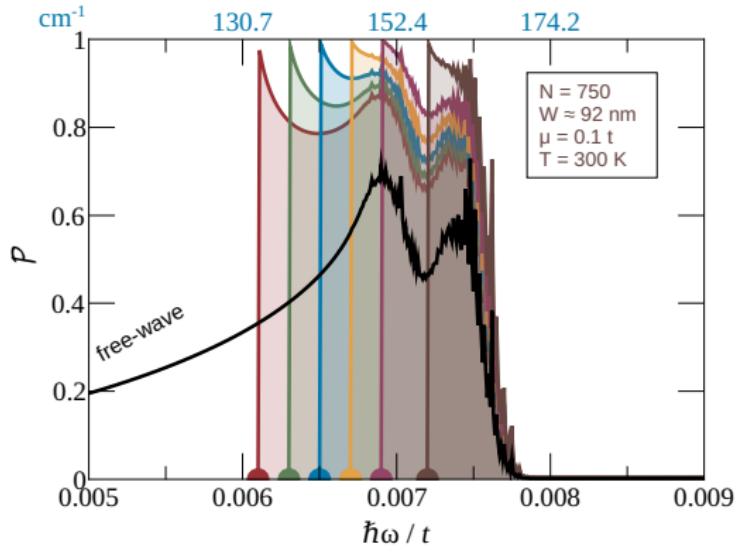
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- Large increase of DP
- Tunable threshold
- Well defined band filter



Competing effects

- Plasmon absorption resonance ($\gtrsim 1 \mu\text{m}$ range) Ju et al. 2011
- Disorder
- Drude peak at $\omega = 0$, $\gamma \sim 100 \text{ cm}^{-1}$
$$\Re \frac{\sigma_D(\omega=0)}{\sigma_0} \approx 800 |\mu|/t$$

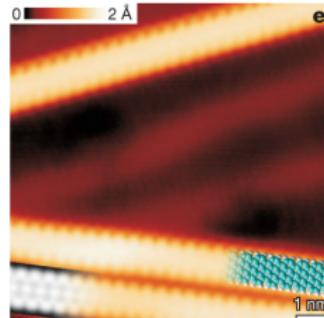
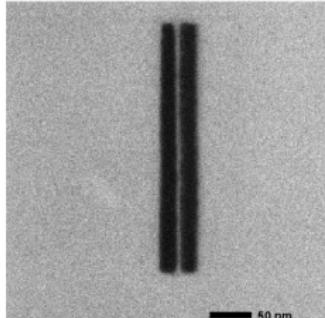
Competing factors & State of the art

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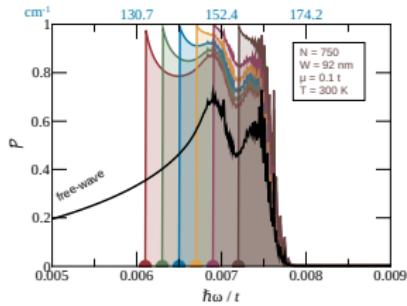
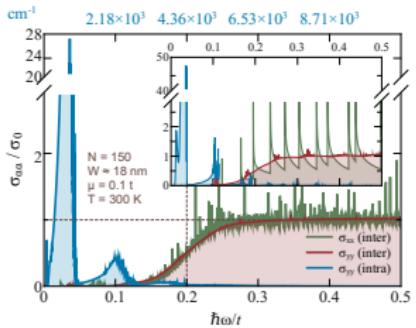
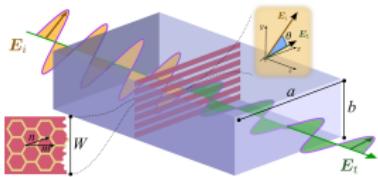
Production

- He ion lithography – features $\sim 10 \text{ nm}$ Lemme et al. 2009; Bai et al. 2009; Pickard et al. 2009
- Carbon nanotube unzipping Kosynkin et al. 2009; Jiao et al. 2010; Tao et al. 2011
- Strain engineering Pereira et al. 2009
- Self assembly – ribbons and “chevron” like ribbons $W \sim 20 \text{ nm}$ Cai et al. 2010



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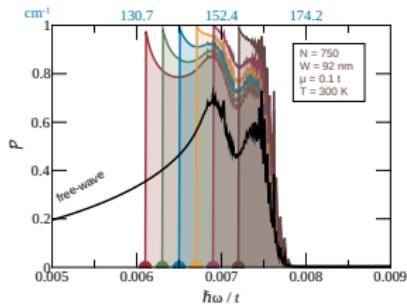
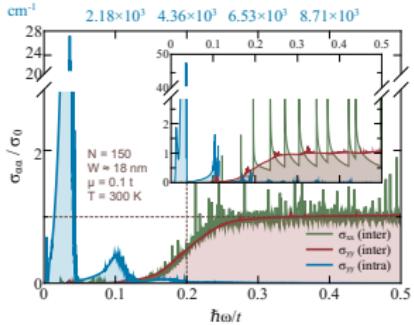
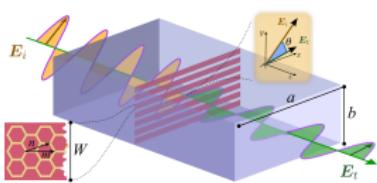
Conclusion



What have we done

- Exact optical conductivity tensor of GNRs
- Optical absorption can be made highly anisotropic
- Tunable via the ribbon width, and/or via the impedance of the medium.
- Strong anisotropy lies in a resonant feature that is simultaneously very strong and resilient to level broadening
- Very high degree of polarization $\sim 85\%$, enhanceable to near $\sim 100\%$

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Future work

- Study the influence of more specific disorder models
- Combining the intrinsic absorption response of GNRs with the geometric effects
- Interplay of the anisotropy induced here by space quantization and plasmons

References

- Bai, Jingwei et al. (2009). "Rational Fabrication of Graphene Nanoribbons Using a Nanowire Etch Mask". In: *Nano Lett.* 9, p. 2083.
- Born, M. et al. (1997). *Principles of Optics*. Cambridge University Press.
- Cai, J. et al. (2010). "Atomically precise bottom-up fabrication of graphene nanoribbons". In: *Nature* 466, pp. 470–473.
- Jackson, John David (1999). *Classical Electrodynamics*. 3rd ed. John Wiley & Sons Inc.
- Jiao, Liying et al. (2010). "Facile Synthesis of High Quality Graphene Nanoribbons". In: *Nature Nanotechnology* 5, p. 321.
- Ju, Long et al. (2011). "Graphene plasmonics for tunable terahertz metamaterials". In: *Nature Nanotechnology* 6, p. 630.
- Kosynkin, Dmitry V. et al. (2009). "Longitudinal unzipping of carbon nanotubes to form graphene nanoribbons". In: *Nature* 458, p. 872.
- Lemme, M. C. et al. (2009). "Etching of Graphene Devices with a Helium Ion Beam". In: *ACS Nano* 3, p. 2674.
- Pereira, Vitor M. et al. (2009). "All-graphene integrated circuits via strain engineering". In: *Phys. Rev. Lett.* 103, p. 046801.
- Pickard, Dan et al. (2009). "Graphene Nano-Ribbon Patterning in the ORION ® PLUS". In:
- Ruseckas, J. et al. (2011). "Spectrum of pi electrons in bilayer graphene nanoribbons and nanotubes: an analytical approach". In: *Phys. Rev. B* 83, p. 035403.
- Saito, Riiehiro et al. (1992). "Electronic structure of graphene tubules based on C_{60} ". In: *Physical Review B* 46.3, pp. 1804–1811. URL: http://prb.aps.org/abstract/PRB/v46/i3/p1804_1.
- Tao, Chenggang et al. (2011). "Spatially Resolving Edge States of Chiral Graphene Nanoribbons". In: *Nature Physics, AOP*.
- Wakabayashi, Katsunori et al. (2010). "Electronic states of graphene nanoribbons and analytical solutions". In: *Sci. Technol. Adv. Mater.* 11, p. 054504.

Appendix: Calculation of the Optical Conductivity Tensor

Wave function

$$|\Psi_{\ell,q,\lambda}\rangle = \mathcal{N} \sum_{n,m} e^{-iq(m+n/2)} \sin(k_\ell n) \times \left(|A, n, m\rangle + \lambda e^{-i\theta_{\ell,q}} |B, n, m\rangle \right)$$

Phase difference between sublattices

$$\theta_{\ell,q} = \arctan \frac{2 \cos k_\ell \sin(q/2)}{1 + 2 \cos k_\ell \cos(q/2)}$$

Conductivity tensor derived from the Kubo formula

$$\begin{aligned} \sigma_{\alpha\beta} = & \frac{2ie^2}{\omega S} \sum_{\ell_1, \ell_2, q} \sum_{\lambda_1, \lambda_2} \frac{f(E_{\ell_1, q, \lambda_1}) - f(E_{\ell_2, q, \lambda_2})}{\hbar\omega - (E_{k_2, q, \lambda_2} + E_{k_1, q, \lambda_1}) + i0^+} \\ & \times \langle \Psi_{\ell_1, q, \lambda_1} | v_\alpha | \Psi_{\ell_2, q, \lambda_2} \rangle \langle \Psi_{\ell_2, q, \lambda_2} | v_\beta | \Psi_{\ell_1, q, \lambda_1} \rangle \end{aligned}$$

Longitudinal conductivity along x -direction

$$\Re \frac{\sigma_{xx}}{\sigma_0} = \mathcal{N}_x \sum_{\ell_0} \delta f_{q_0, \ell_0} M_x^2(q_0, \ell_0)$$

$$M_x^2(q_0, \ell_0) = \frac{[\cos \theta_{\ell_0, q_0} - \cos(\theta_{\ell_0, q_0} - q_0/2) \cos k_{\ell_0}]^2}{\sin(q_0/2) \cos k_{\ell_0}}$$

$$q_0 = 2 \arccos \frac{(\Omega/2)^2 - 1 - 4 \cos^2 k_{\ell_0}}{4 \cos k_{\ell_0}}$$