Optical Absorbance of ß-Bi_4X_4

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Spenser Talkington (UCLA), and Fan Zhang (UTD)

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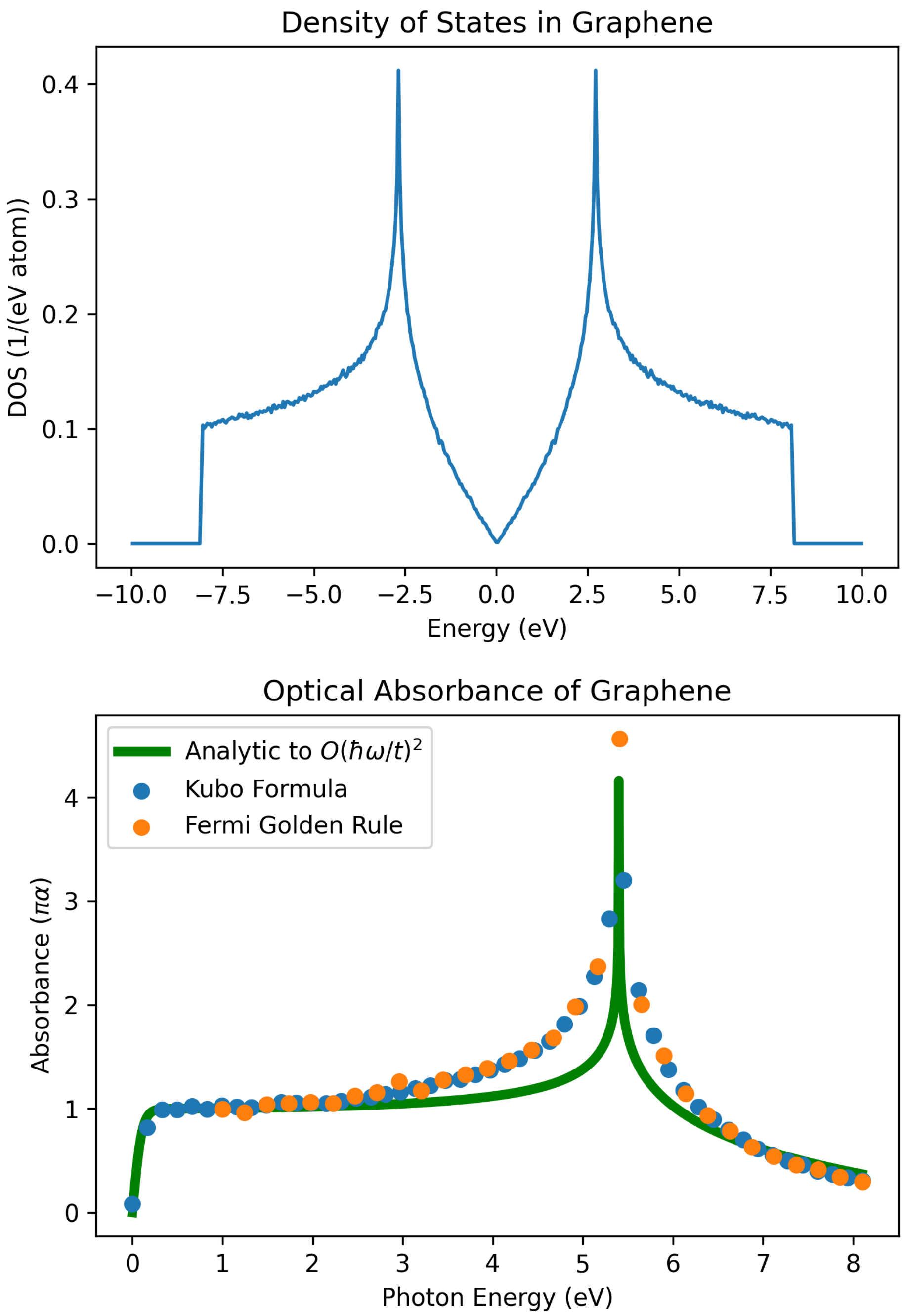
Optical Properties of Solids

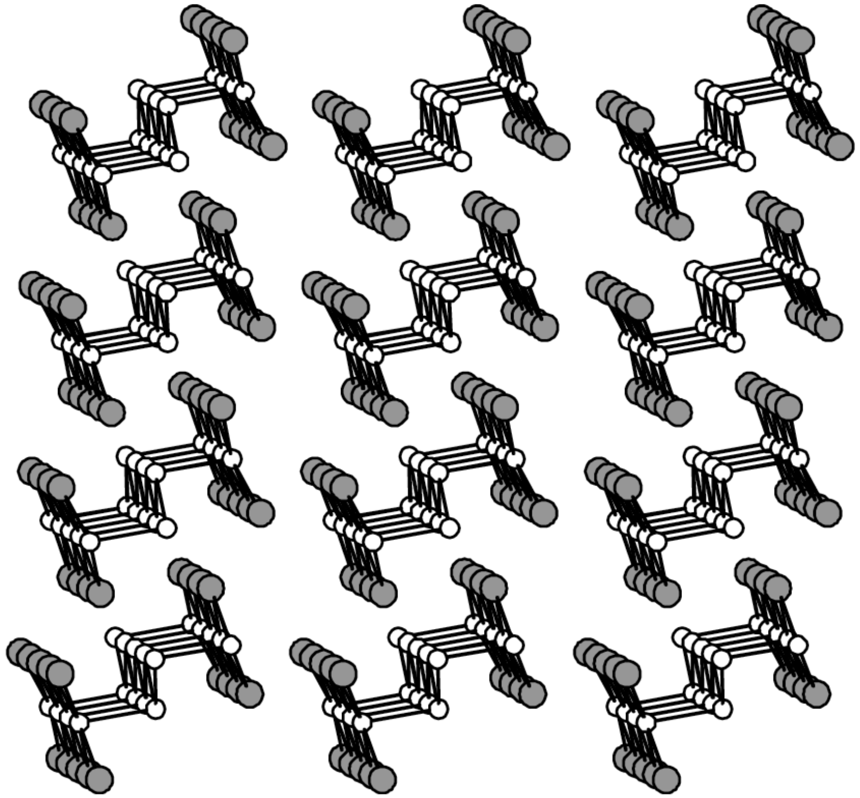
- Optical properties of solids are readily measured in experiments
- We care about optoelectronic properties because they have device applications and they help us understand materials
- Properties include absorbance, which is the amount of energy absorbed per energy incident
- Graphene's quantized absorbance[1]



[1] Nair, et al Science 320, 1308 (2008)

Bismuth Bromide and Bismuth Iodide

- Quasi-one-dimensional van der Waals materials
  - Strands that form layers that stack
  - Protected surface states in beta phase: it is a weak topological insulator[1,2]
  - Protected edge states in alpha phase: it is a higher-order topological insulator[3]
- Two forms:
  - AA stacking (beta phase)
  - AB stacking (alpha phase)



ß-Bi_4I_4 structure[4]

[1] Zhang et al PRL 116, 066801 (2016)
[2] Noguchi et al Nature 566, 518 (2019)
[3] Yoon, et al arXiv:2005.14710 (2020)
[4] Dikarev et al RCB 50(12), 2304 (2001)

The Bulk Hamiltonian (Bloch Hamiltonian)

- In lattices (discrete translational invariance), Bloch's Theorem holds, where $u_n(r)$ is a function with the periodicity of the lattice and k is a wavevector:

$$\psi_n(r) = \exp(ik \cdot r)\, u_n(r)$$

- The periodic functions $u_n(r)$ are given by the Bulk Hamiltonian, $\mathcal{H}(k)$:

$$\mathcal{H}(k)\, u_n(r) = \epsilon_n\, u_n(r)$$

- We define the valence states to be:

$$v_n = \psi_n(r) \text{ such that } \epsilon_n < 0$$

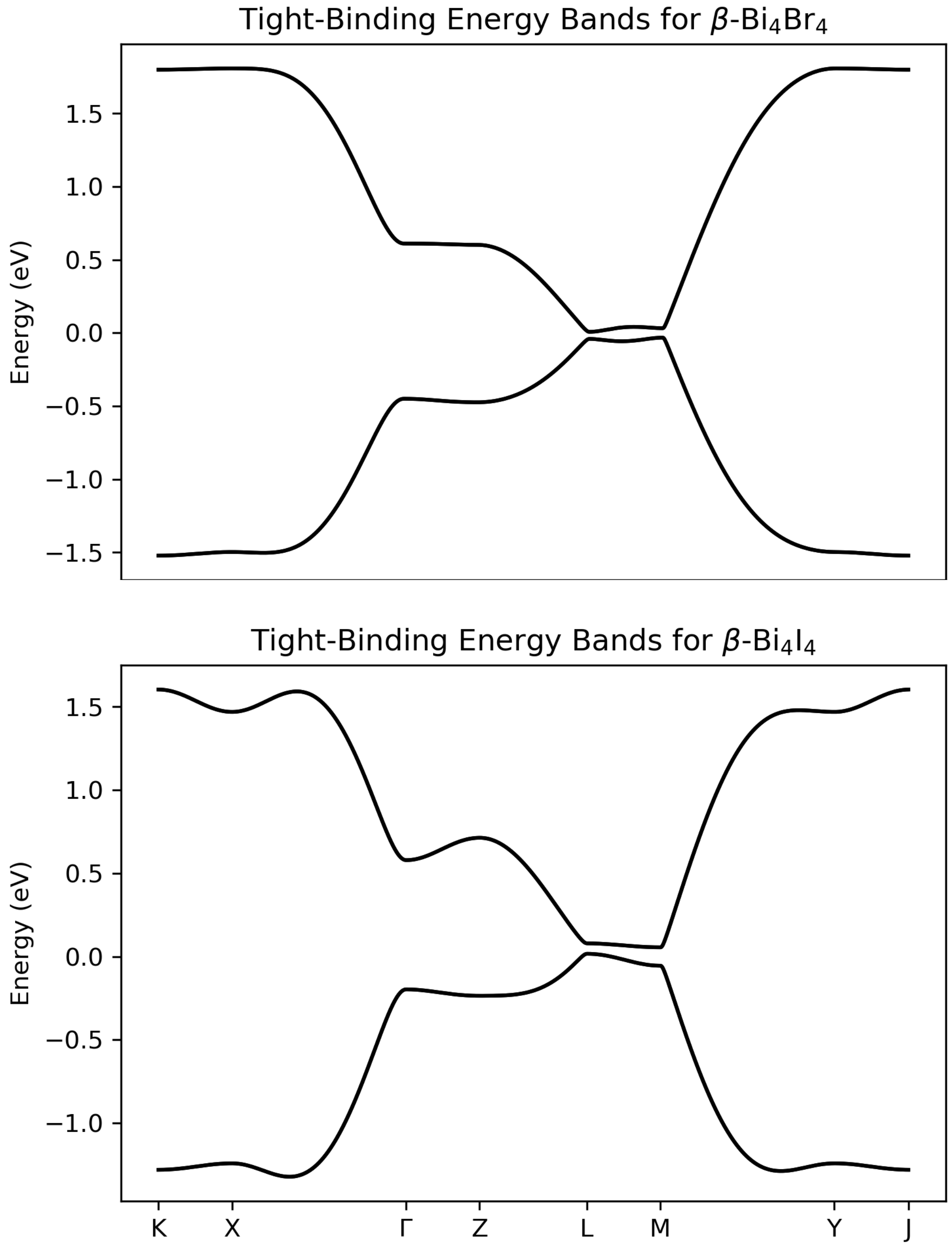
- We define the conduction states to be:

$$c_n = \psi_n(r) \text{ such that } \epsilon_n > 0$$

Energy Bands of ß-Bi_4X_4

- Energy bands: diagonalize Hamiltonian
- Tight-binding model Hamiltonian
  - Onsite energies
  - Transfer energies
  - Phase accumulation (Peierels)
- How to get a tight-binding model?
  - First principles density functional theory calculations leads to maximally localized Wannier function tight-binding models
  - Symmetry considerations and restrict to nearest neighbor transfers
  - This has been done in the literature[1]

[1] Yoon, et al arXiv:2005.14710 (2020)



Absorbance

- Absorbance is the fraction of the incident power absorbed:

$$P = \frac{W_{\text{absorbed}}}{W_{\text{incident}}}$$

- Incident light carries energy flux (Poynting Vector, in Gaussian units):

$$W_{\text{incident}} = \frac{\omega^2 |A|^2}{4\pi c}$$

- From a conductance standpoint (Ohm's Law):

$$W_{\text{absorbed}} = \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}^{\dagger} \begin{pmatrix} \sigma_{xx} & \sigma_{xy} & \sigma_{xz} \\ \sigma_{yx} & \sigma_{yy} & \sigma_{yz} \\ \sigma_{zx} & \sigma_{zy} & \sigma_{zz} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}$$

The Current Operator

- In electromagnetism, the current operator is $j = \sigma E$, or if $E = E^{\beta}\hat{\beta}$:

$$j^{\alpha} = \sigma_{\alpha\beta} E^{\beta}$$

- In the single-particle framework, the current operator is, for direction $\alpha = x, y, \mathrm{or}\ z$:

$$j^{\alpha} = ev^{\alpha}$$

- Now, we note that for the free particle Hamiltonian:

$$\frac{\partial H_0}{\partial p} = \frac{\partial p^2/2m}{\partial p} = \frac{p}{m} = v$$

- This motivates the gradient approximation:

$$v^{\alpha} = \frac{\partial \mathcal{H}(k)}{\partial p^{\alpha}}$$

- So the current operator is, with $p^{\alpha} = \hbar k^{\alpha}$:

$$j^{\alpha} = \frac{e}{\hbar} \frac{\partial \mathcal{H}(k)}{\partial k^{\alpha}}$$

Linear Response: the Kubo Formulism

- For the conductivity, integrate over the Brillouin Zone (k-space):

$$\sigma_{\alpha\beta}(\hbar\omega,\eta) = i\frac{e^2}{\hbar}\sum_{c,v}\int_{\mathrm{BZ}} \frac{\mathrm{d}^{\mathrm{dim}}k}{(2\pi)^{\mathrm{dim}}} \frac{f(\epsilon_v)-f(\epsilon_c)}{\epsilon_c-\epsilon_v} \frac{\hbar v_{vc}^{\alpha}\,\hbar v_{cv}^{\beta}}{\hbar\omega-(\epsilon_c-\epsilon_v)+i\eta}$$

- In the gradient approximation:

$$\hbar v_{vc}^{\alpha}\ \hbar v_{cv}^{\beta} = \left\langle v(k)\middle|\frac{\partial\mathcal{H}(k)}{\partial k^{\alpha}}\middle|c(k)\right\rangle\left\langle c(k)\middle|\frac{\partial\mathcal{H}(k)}{\partial k^{\beta}}\middle|v(k)\right\rangle$$

- The Fermi-Dirac distribution function is:

$$f(\epsilon) = (\exp(\epsilon/k_{\mathrm{B}}T)+1)^{-1}$$

How to Calculate Absorbance? (the kubo way)

- Choose system
- Find bulk Hamiltonian, H
- Find eigenenergies and eigenfunctions of H
- Specify polarization $\hat{E}$, frequency ω, temperature T, and broadening η
- Calculate the conductivity tensor (with Kubo)
  - At each point in the BZ, find conductivity matrix elements and evaluate; sum over these points
- Find the energy absorbed $W(\omega) = E^\dagger \sigma(\omega) E$
- Find the absorbance $P(\omega) = W_{\text{abs}}/W_{\text{inc}}$

Absorbance of ß-Bi_4X_4

sensitive figures removed

Conclusions

- Results
  - As expected from its anisotropic structure, ß-Bi_4X_4 has a correspondingly anisotropic absorbance
  - Absorbance is as would be expected from the density of states
  - The monolayer system has substantially *lower* absorbance than the full system
  - ß-Bi_4X_4 are real materials and expect our predictions to agree with experiments
- Future directions:
  - Compare and contrast alpha and beta phases
  - Address other optical properties such as the photocurrent
  - Consider the optical properties of surface/edge states

Questions?