Anisotropic optical response in the quasi-one-dimensional topological insulators Bi_4Br_4 and Bi_4I_4

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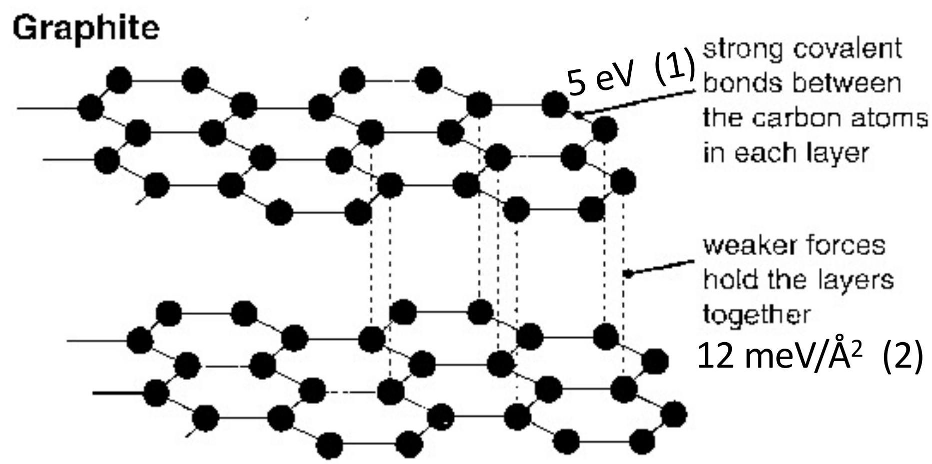
Layered van der Waals Materials

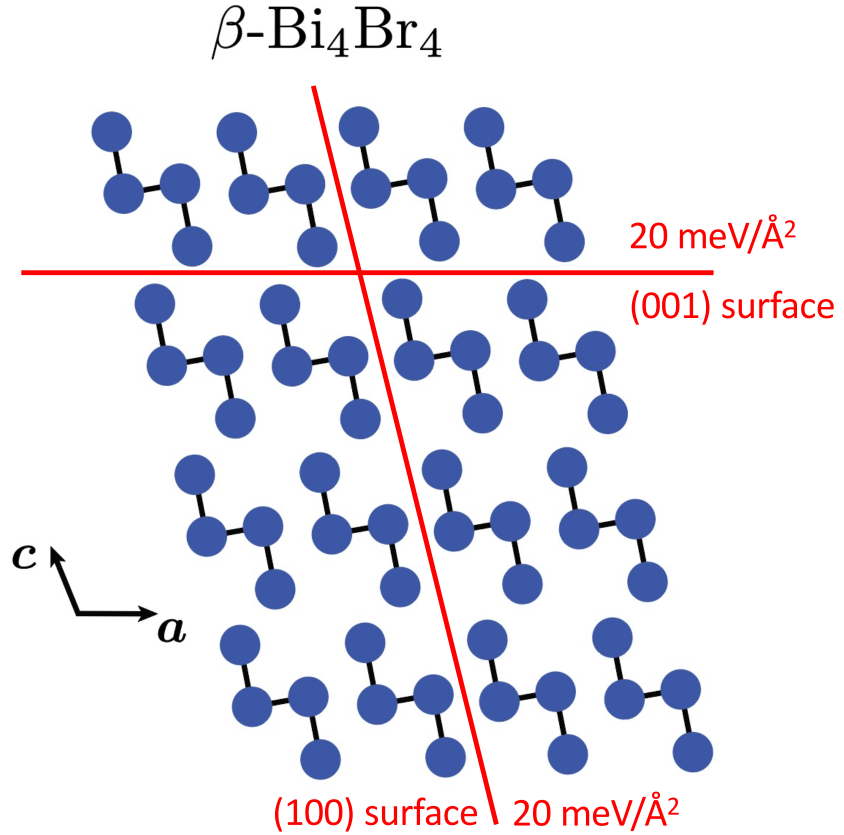
- Direction-dependent bond strength leads to natural cleavage planes
- Good cleavage planes makes for easy access to surface and edge states
- Many materials are stacked 2D layers



1. D. Brenner, et al. JPCM **14** 783 (2002)
2. Z. Liu, et al. PRB **85**, 205418 (2012)

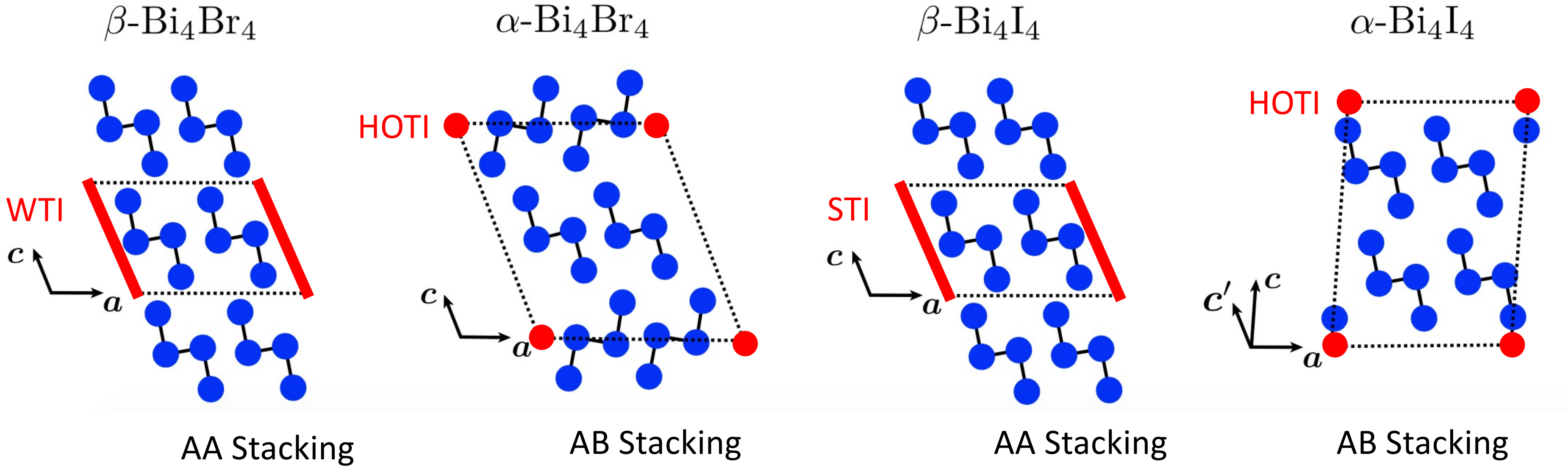
Quasi-One Dimensional Bismuth Halides

- Bi_4Br_4 and Bi_4I_4 are 1D chains that align to form 3D materials[1,2,3]
- Two natural cleavage planes
  - (001) surface: trivial insulator[1]
  - (100) surface: weak/strong TI[1]



1. C.C. Liu, et al. PRL **116**, 066801 (2016)
2. C. Yoon, et al. arXiv 2005.14710 (2020)
3. H. G. von Schnering, et al Z. Inorg. Chem. 438, **37** (1978).
   H. von Benda, et al. Z. Inorg. Chem. 438, **53** (1978).

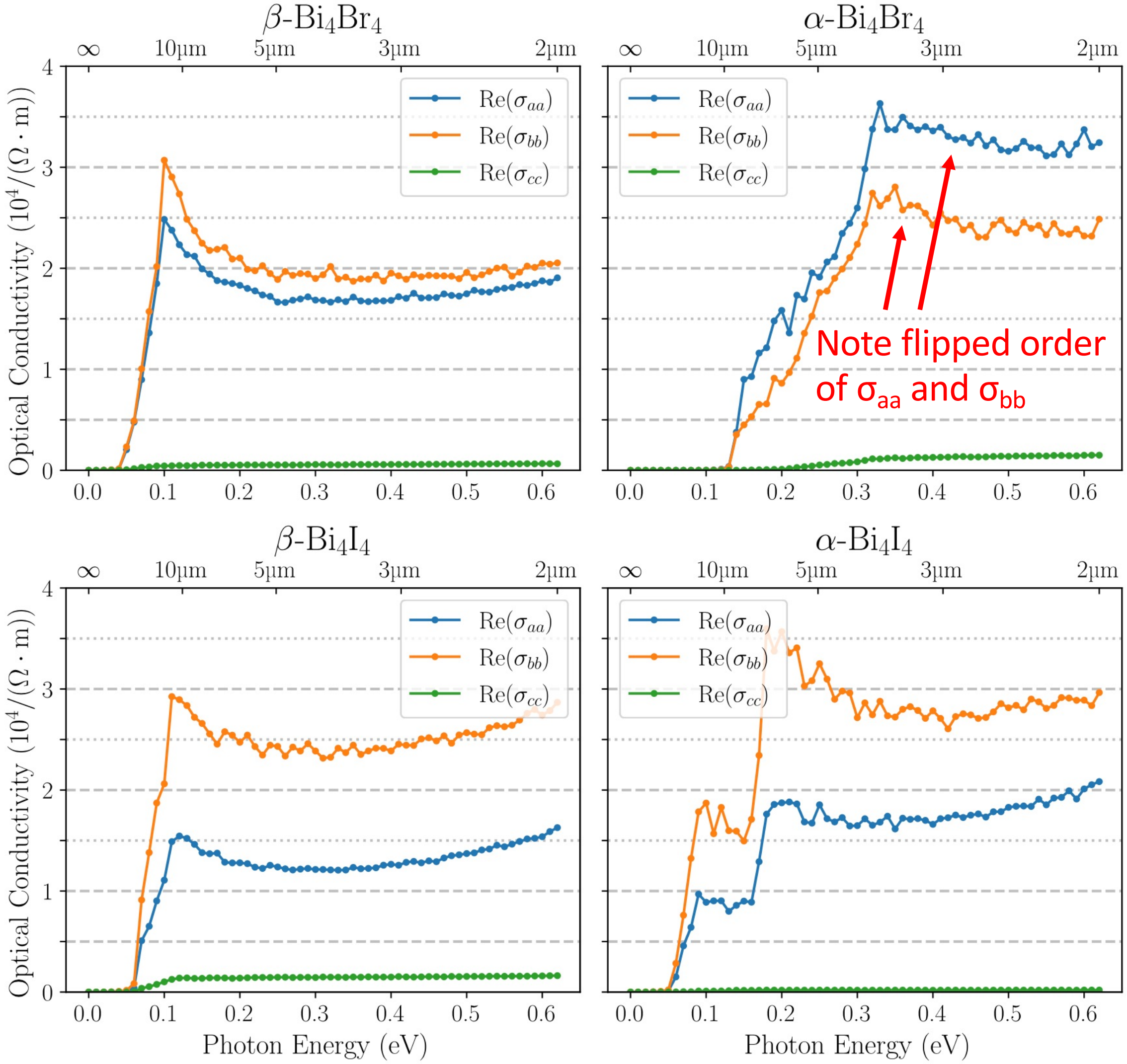
Phases of the Bismuth Halides



1. C.C. Liu, et al. PRL **116**, 066801 (2016)
2. C. Yoon, et al. arXiv 2005.14710 (2020)
3. R. Noguchi, et al. Nature **566,** 518 (2019)

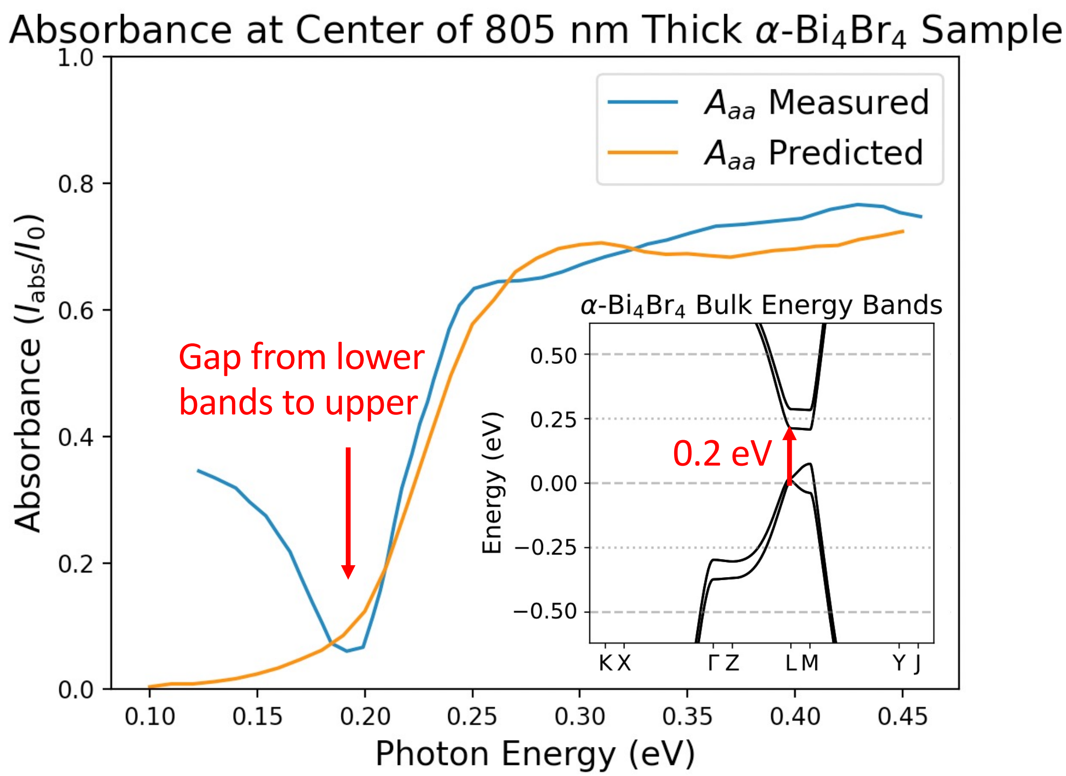
Bulk Conductivity

- Bulk optical conductivities are anisotropic
  - $\sigma_{bb} \sim \sigma_{aa} \sim$ 10-50 σ_{cc}
- Kubo formula calculation
  - Single particle
  - Low Temperature



Comparison to Experiment

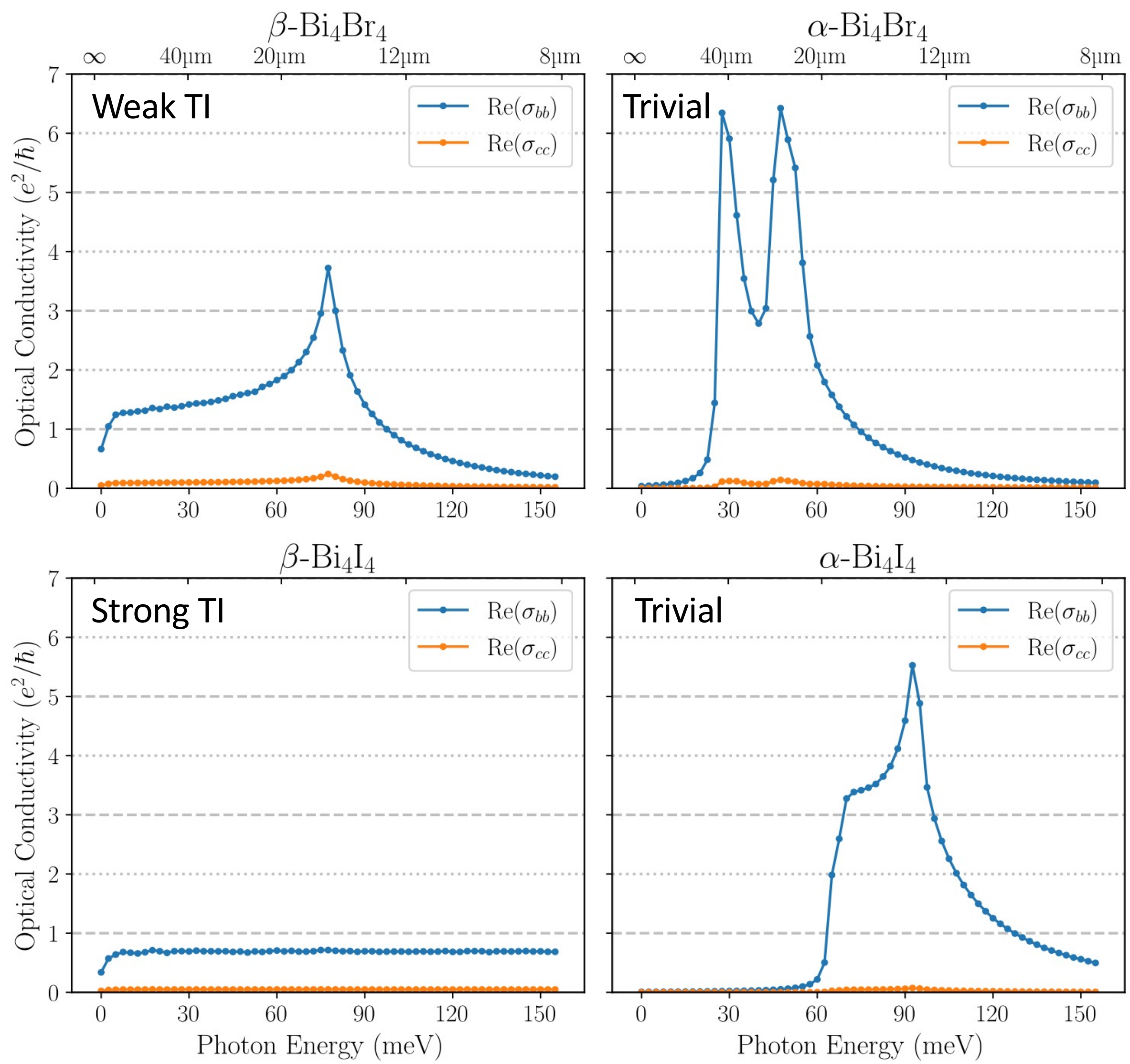
- Experiment
  - Crystals grown on CaF_2 substrate by flux method
  - Linearly polarized light shined on exposed (001) surface
  - Measured absorbance
- Model
  - Fermi energy 137 meV below center of bulk band gap
  - Beer-Lambert Law
    - $A = (1 - R)(1 - e^{-l\,\mathcal{A}})$



Experimental data from P. Mao, et al. arXiv:2007.00223 (2020).

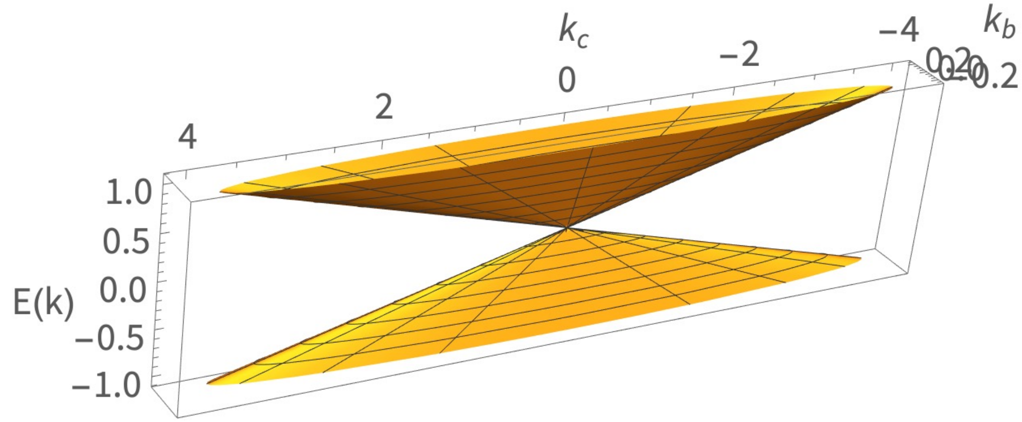
(100) Surface σ

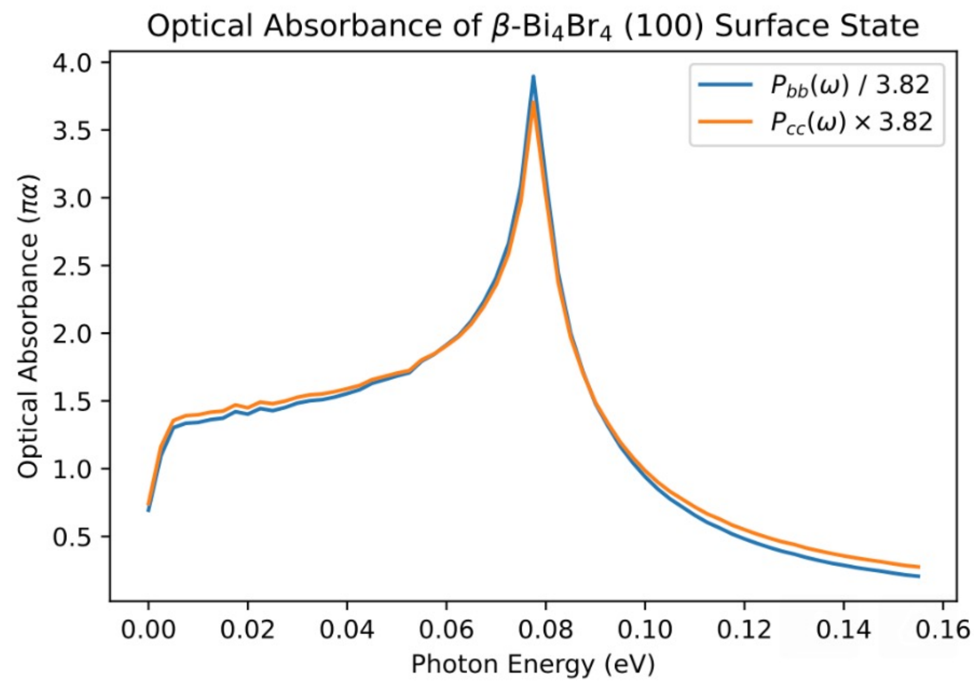
- Strand direction is much more conductive than inter-strand conduction
  - We can model as an elliptic Dirac cone
- Conductivity is large
  - Graphene in Dirac-cone limit is $e^2/4\hbar$



Elliptic Dirac Cone

- Let there be distinct Fermi velocities in x and y
- Optical absorbance
  - $A_{\text{dir}} = \pi\alpha \frac{v_{\text{dir}}^2}{v_x v_y}$
- If we multiply/divide by $v_{\text{dir}}^2/v_b v_c$, we should see the same behavior





Methods

Our group's previous work in: C. Yoon, et al. arXiv 2005.14710 (2020)

Structure

↓ First-principles DFT

MLWF tight-binding model

↓ Best-fit nearest-neighbor tight-binding model

The current work

Hamiltonians and velocity operators

↓ Integrate Kubo formula

Optical conductivity

↓ Textbook formulas

Optical response properties

Kubo Formula Integration

- Kubo formula (non-interacting, non-relativistic, low temperature):

$$\sigma_{\mu\nu} = i\frac{e^2}{\hbar}\sum_{s,s'}\int_{\mathrm{BZ}}\frac{d\boldsymbol{k}}{(2\pi)^{\mathrm{dim}}}\frac{1}{\epsilon_{s'}-\epsilon_s}\frac{\langle s,\boldsymbol{k}|\hbar\hat{v}_\mu|s',\boldsymbol{k}\rangle\langle s',\boldsymbol{k}|\hbar\hat{v}_\nu|s,\boldsymbol{k}\rangle}{\hbar\omega-(\epsilon_{s'}-\epsilon_s)+i\eta}$$

- Velocity operators (in gradient approximation):

$$\hat{v}_\mu = \frac{\partial\mathcal{H}}{\partial k_\mu}$$

- Hamiltonians were obtained in our group's previous work[1]

1. C. Yoon, et al. arXiv 2005.14710 (2020)

Conclusions

- Bi_4X_4 are quasi-1D materials with easy access to surface and edge states
- Our predictions for the bulk agree with experiment for $\hbar\omega > \text{gap}$
- We predict that the (100) side surface exhibits anisotropic optical conductivity by a factor of ~15 due to anisotropic Fermi velocities
- The peak optical conductivity for (100) surface states is predicted to exceed that of graphene in the infrared region by factors of 10-20

Acknowledgements

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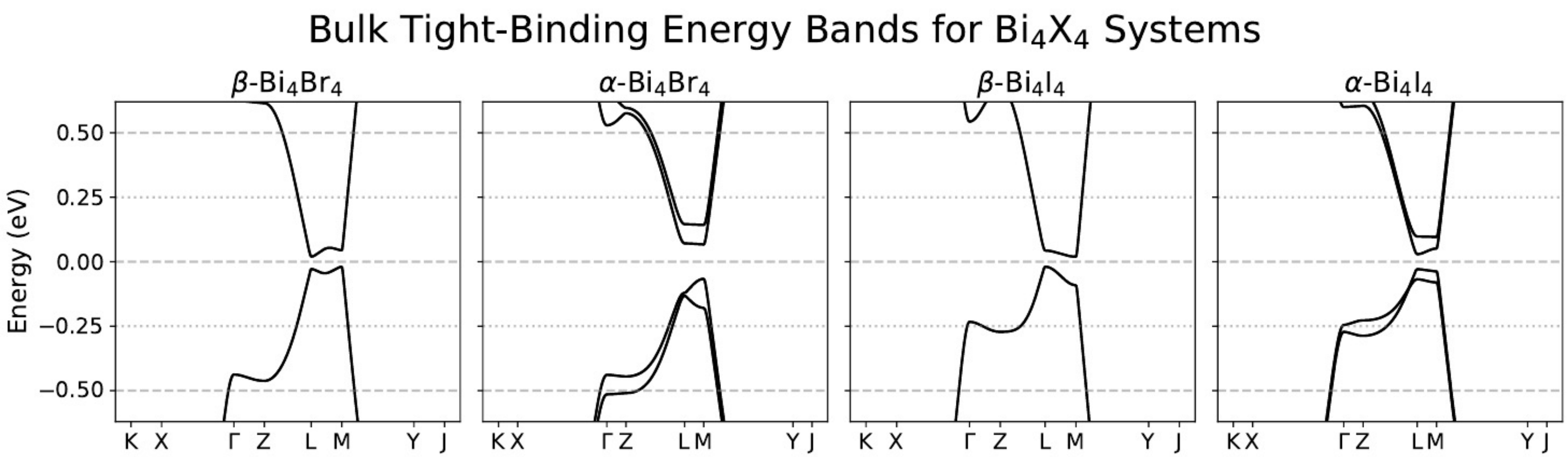
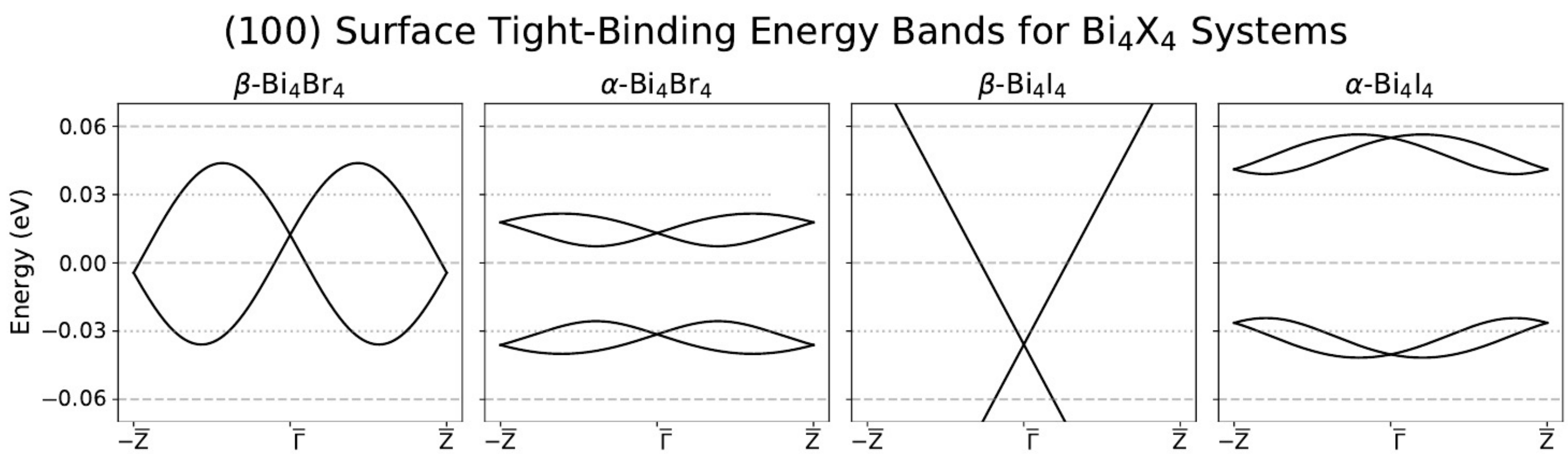
Questions?

Supplemental Slides

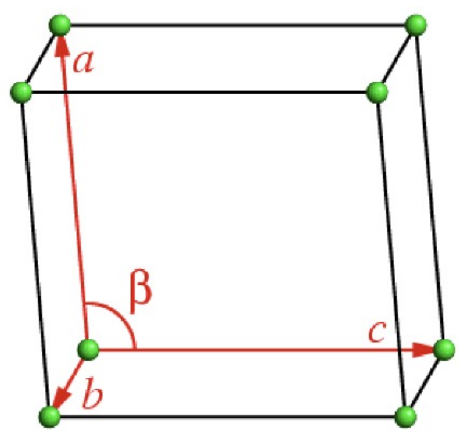
Bulk Tight-Binding Energy Bands

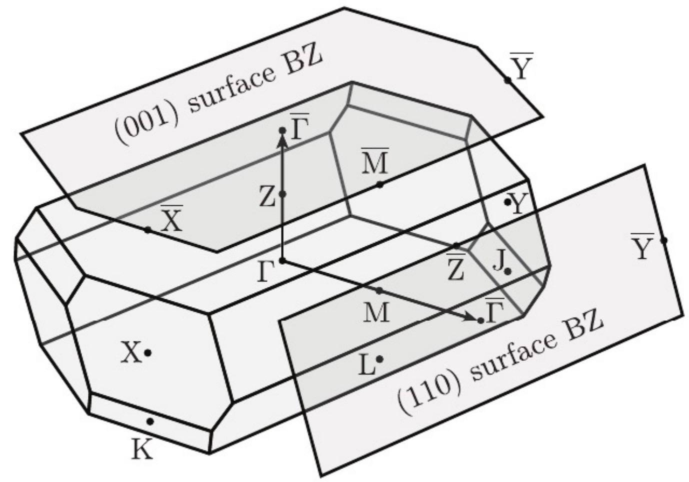


(100) Surface Tight-Binding Energy Bands

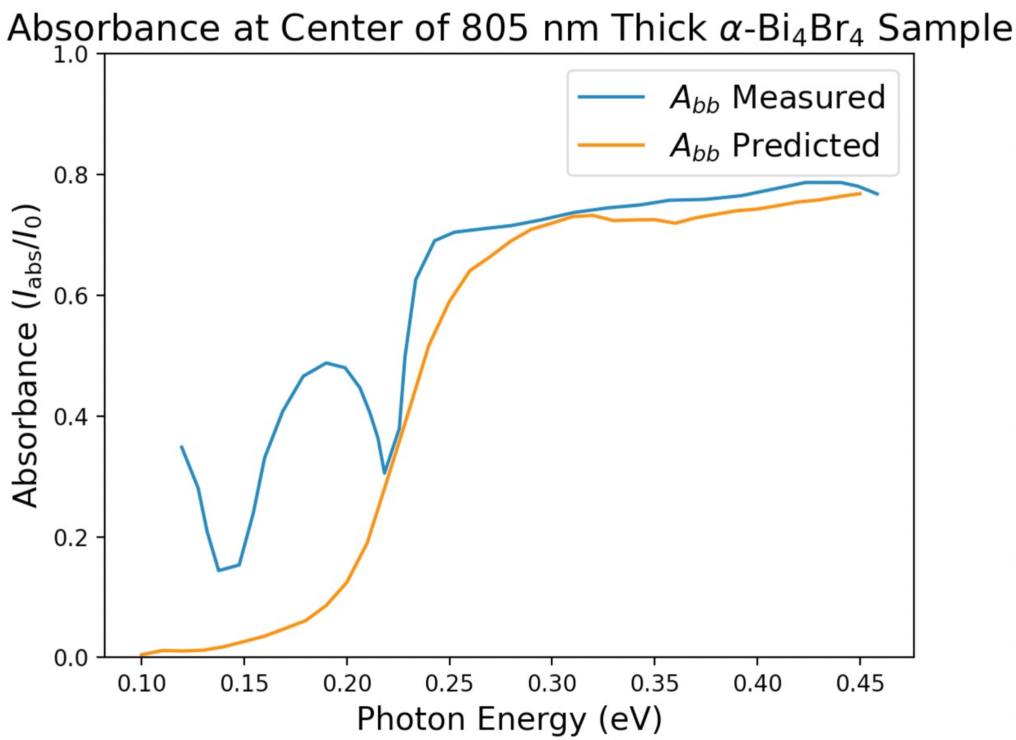


Monoclinic Lattice and Reciprocal Lattice





Bulk bb absorbance



Reprocessing Bulk σ

- Complex dielectric function

$$\epsilon = 1 + i\frac{4\pi}{\omega}\sigma$$

- Other properties follow from textbook formulas

Where the diagonal elements of the refractive index tensor are given by:

$$n_{\mu\mu}(\omega) = \sqrt{\frac{|\epsilon_{\mu\mu}(\omega)| + \mathrm{Re}[\epsilon_{\mu\mu}(\omega)]}{2}} \tag{8}$$

and the diagonal elements of the extinction coefficient tensor κ are given by:

$$\kappa_{\mu\mu}(\omega) = \sqrt{\frac{|\epsilon_{\mu\mu}(\omega)| - \mathrm{Re}[\epsilon_{\mu\mu}(\omega)]}{2}} \tag{9}$$

The absorption coefficient is:

$$A_{\mu\mu}(\omega) = \frac{2\omega}{c}\kappa_{\mu\mu} \tag{10}$$

Following the Beer-Lambert law, the absorbance per length ℓ is:

$$\mathcal{A}_{\mu\mu}(\omega) = (1 - \mathcal{R}_{\mu\mu})(1 - \exp(-\ell A_{\mu\mu})) \tag{11}$$

The reflectance is:

$$\mathcal{R}_{\mu\mu}(\omega) = \left|\frac{1 - \widetilde{n}_{\mu\mu}}{1 + \widetilde{n}_{\mu\mu}}\right|^2 \tag{12}$$

$$= \frac{(n_{\mu\mu} - 1)^2 + \kappa_{\mu\mu}^2}{(n_{\mu\mu} + 1)^2 + \kappa_{\mu\mu}^2} \tag{13}$$

Reprocessing Surface σ

$$\mathcal{A}_{\mu\mu} = 2\pi\alpha\frac{\mathrm{Re}(\sigma_{\mu\mu})}{e^2/\hbar}\left|\frac{2}{1+\widetilde{n}^{\mathrm{sub}}_{\mu\mu}+2\pi\alpha\,\sigma_{\mu\mu}/(e^2/\hbar)}\right|^2 \quad (14)$$

$$\mathcal{R}_{\mu\mu} = \left|\frac{1-\widetilde{n}^{\mathrm{sub}}_{\mu\mu}-2\pi\alpha\,\sigma_{\mu\mu}/(e^2/\hbar)}{1+\widetilde{n}^{\mathrm{sub}}_{\mu\mu}+2\pi\alpha\,\sigma_{\mu\mu}/(e^2/\hbar)}\right|^2 \quad (15)$$

$$\mathcal{T}_{\mu\mu} = \mathrm{Re}(\widetilde{n}^{\mathrm{sub}}_{\mu\mu})\left|\frac{2}{1+\widetilde{n}^{\mathrm{sub}}_{\mu\mu}+2\pi\alpha\,\sigma_{\mu\mu}/(e^2/\hbar)}\right|^2 \quad (16)$$

Bulk Operators I

For all bulk systems $v^a = v_L^a$ and $v^b = v_L^b$.

Layer

$$H_L = M\sigma_z + D + t_a\sigma_y(\sin(q_1) + \sin(q_2)) + t_b\sigma_x s_z \sin(q_2 - q_1) \tag{1}$$

$$M = m_0 + m_a(\cos(q_1) + \cos(q_2)) + m_b \cos(q_2 - q_1) \tag{2}$$

$$D = d_0 + d_a(\cos(q_1) + \cos(q_2)) + d_b \cos(q_2 - q_1) \tag{3}$$

$$v_L^a = [-(m_a\sigma_z + d_a)(\sin(q_1) + \sin(q_2)) + t_a\sigma_y(\cos(q_1) + \cos(q_2))]a/2\hbar \tag{4}$$

$$v_L^b = [-(m_b\sigma_z + d_b)\sin(q_2 - q_1) + t_b\sigma_x s_z \cos(q_2 - q_1)]b/2\hbar \tag{5}$$

$$v_L^c = 0 \tag{6}$$

Bulk Operators II

Beta

$$H_\beta = H_L + 2(d_c + m_c\sigma_z)\cos(q_3) + 2t_c\sigma_x s_y \sin(q_3) \tag{7}$$

$$v_\beta^c = [-2(d_c + m_c\sigma_z)\sin(q_3) + 2t_c\sigma_x s_y \cos(q_3)]c/\hbar \tag{8}$$

Alpha Bismuth Bromide

$$\widetilde{H}_\beta = H_L + (d_c + m_c\sigma_z)[\tau_x + (\tau_x\cos(q_3) + \tau_y\sin(q_3))] + t_c\sigma_x s_y[\tau_y - (\tau_y\cos(q_3) - \tau_x\sin(q_3))] \tag{9}$$

$$H_{\alpha\text{-Br}} = \widetilde{H}_\beta + (d_0' + m_0'\sigma_z)\tau_z + t_c'\sigma_y s_y[-\tau_x + (\tau_x\cos(q_3) + \tau_y\sin(q_3))] + (d_c' + m_c'\sigma_z)s_y[\tau_y + (\tau_y\cos(q_3) - \tau_x\sin(q_3))] \tag{10}$$

$$v_{\alpha\text{-Br}}^c = [(d_c + m_c\sigma_z + t_c'\sigma_y s_y)(-\tau_x\sin(q_3) + \tau_y\cos(q_3)) + (t_c\sigma_x - d_c' - m_c'\sigma_z)s_y(\tau_y\sin(q_3) + \tau_x\cos(q_3))]c/\hbar \tag{11}$$

Alpha Bismuth Iodide

$$H_{\alpha\text{-I}} = H_L + t\sigma_x\tau_z + t'\sigma_y s_y\tau_z + [(d_c + m_c\sigma_z)\tau_x + (d_c' + m_c'\sigma_z)(\tau_x\cos(q_3) + \tau_y\sin(q_3))] \tag{12}$$

$$+ \sigma_x s_y[t_c\tau_y - t_c'(\tau_y\cos(q_3) - \tau_x\sin(q_3))] \tag{13}$$

$$v_{\alpha\text{-I}}^c = [(d_c' + m_c'\sigma_z)(-\tau_x\sin(q_3) + \tau_y\cos(q_3)) + \sigma_x s_y t_c'(\tau_y\sin(q_3) + \tau_x\cos(q_3))]c/\hbar \tag{14}$$

(100) Surface Operators I

Beta Bismuth Bromide

$$
\begin{aligned}
h_{\beta\text{-Br}} &= (d_0 - 2d_a + d_b)1 + \eta t_b s_z q_b + 2d_c \cos(q_c) + 2\eta t_c s_y \sin(q_c) \\
v^b_{\beta\text{-Br}} &= \eta t_b s_z b \\
v^c_{\beta\text{-Br}} &= [-2d_c \sin(q_c) + 2\eta t_c s_y \cos(q_c)]c
\end{aligned}
$$

Beta Bismuth Iodide

$$
\begin{aligned}
h_{\beta\text{-I}} &= (d_0 - 2d_a + d_b + 2d_c)1 + \eta t_b s_z q_b + 2\eta t_c s_y q_c \\
v^b_{\beta\text{-I}} &= \eta t_b s_z b \\
v^c_{\beta\text{-I}} &= 2\eta t_c s_y c
\end{aligned}
$$

(100) Surface Operators I

Alpha Bismuth Bromide

$$
\begin{aligned}
h_{\alpha\text{-Br}} &= (d_0 - 2d_a + d_b)1 + \eta t_b s_z q_b + d_c(\tau_x + (\tau_x \cos(q_c) + \tau_y \sin(q_c))) \\
&\quad + \eta t_c s_y(\tau_y - (\tau_y \cos(q_c) - \tau_x \sin(q_c))) + d'_c s_y(\tau_y + (\tau_y \cos(q_c) - \tau_x \sin(q_c))) + d'_0 \tau_z \\
v^b_{\alpha\text{-Br}} &= \eta t_b s_z b \\
v^c_{\alpha\text{-Br}} &= [d_c(-\tau_x \sin(q_c) + \tau_y \cos(q_c)) + \eta t_c s_y(\tau_y \sin(q_c) + \tau_x \cos(q_c)) - d'_c s_y(\tau_y \sin(q_c) + \tau_x \cos(q_c))]c
\end{aligned}
$$

Alpha Bismuth Iodide

$$
\begin{aligned}
h_{\alpha\text{-I}} &= (d_0 - 2d_a + d_b)1 + \eta t_b s_z q_b + (d_c \tau_x + d'_c(\tau_x \cos(q_c) + \tau_y \sin(q_c))) + \eta s_y(t_c \tau_y - t'_c(\tau_y \cos(q_c) - \tau_x \sin(q_c)) + \eta t \tau_z \\
v^b_{\alpha\text{-I}} &= \eta t_b s_z b \\
v^c_{\alpha\text{-I}} &= [d'_c(-\tau_x \sin(q_c) + \tau_y \cos(q_c)) - \eta t'_c s_y(\tau_y \sin(q_c) + \tau_x \cos(q_c))]c
\end{aligned}
$$