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1 Project objectives

In this project a tool which allows to calculate optical response from twisted bilayer graphene is presented. As this response is supposed to be linear, Kubo formula is used for it. Only inter-band transitions are included in the calculations. Gate potential and temperature are supposed to be zero: graphene is in its neutrality point and states with negative energies are occupied only.

2 Project structure

To calculate the optical response using this project one can follow this steps:

- Check Fourier amplitudes of scattering processes involving different Moire reciprocal lattice
- Calculate $\text{Re } \sigma_{xx}$ using hamiltonian with all significant processes included
- Implement Kramers-Kronig relationships for twisted bilayer graphene to calculate $\text{Im } \sigma_{xx}$
- Convert optical conductivity of thin film to its refraction and extinction coefficients

In this document main assumptions regards all this steps are covered and main procedures are documented.

3 Fourier amplitudes

3.1 Theoretical basis

As the twisted bilayer graphene structure has Moire periodicity, only scattering processes from one to another layer with Δk equal to linear combination of Moire reciprocal vectors make contribution. The proof of this can be found, for example, in [1], [2] or [3]. It starts from assumptions, typical for tight-binding models. Suppose that electronic state in this periodic structure can be represented with Bloch state function:

$$|\varphi_{\alpha k_n}^{(n)}\rangle = \frac{1}{\sqrt{N}} \sum_{R_n} \exp(ik_n(R_n + v_\alpha^{(n)})) |R_n + v_\alpha^{(n)}\rangle$$

Here N is number of electrons, summation goes over the sites of crystal structure, $v_\alpha^{(n)}$ is vector pointing on the element from the basis in sublattice α . $|R_n + v_\alpha^{(n)}\rangle$ are unperturbed atomic

p_z orbitals. "Overlapping energy" of this orbitals gives rise to the hamiltonian with matrix element:

$$\langle \varphi_{\alpha k_2}^{(2)} | \hat{H} | \varphi_{\beta k_1}^{(1)} \rangle = \frac{1}{N} \sum_{R_1 R_2} \exp(-ik_2 R_2^\alpha + ik_1 R_1^\beta) t(R_2^\alpha - R_1^\beta)$$

Here $R_2^\alpha = R_2 + v_\alpha^2$ and $R_1^\beta = R_1 + v_\beta^2$. k_1, k_2 of interest (with not too large energy) are close to Dirac point. Introduce new notations:

$$\bar{k} = \frac{k_1 + k_2}{2}, \quad \Delta k = k_2 - k_1$$

$$\langle \varphi_{\alpha k_2}^{(2)} | \hat{H} | \varphi_{\beta k_1}^{(1)} \rangle = \frac{1}{N} \sum_{R_1 R_2} \exp(-i\bar{k}(R_2^\alpha - R_1^\beta)) \exp\left(-i\Delta k \frac{R_2^\alpha + R_1^\beta}{2}\right) t(R_2^\alpha - R_1^\beta)$$

To go further we introduce clipping function which undergoes conditions

$$\begin{cases} \int_{\Omega} g(r) dr = \Omega_M, \\ g(r) \geq 0, \text{ peak at } 0, \text{ drops rapidly on scale } l_g \\ a \ll l_g \ll L_M \end{cases}$$

Here Ω_M is volume of one Moire supercell, a is length of graphene lattice vector, L_M is length of Moire supercell. This function works as delta-function if integrating with functions changing not significantly on scale of L_M . Then we put "clever unity" in matrix element:

$$\langle \varphi_{\alpha k_2}^{(2)} | \hat{H} | \varphi_{\beta k_1}^{(1)} \rangle = \frac{1}{N} \sum_{R_1 R_2} \left\{ \frac{1}{\Omega_M} \int_{\Omega} g(r - R_1^\beta) dr \right\} \exp(-i\bar{k}(R_2^\alpha - R_1^\beta)) \exp\left(-i\Delta k \frac{R_2^\alpha + R_1^\beta}{2}\right) t(R_2^\alpha - R_1^\beta)$$

Exponent with \bar{k} changes on length scale a and exponent with Δk changes on length scale L_M .

So g function acts like delta-function on second exponent.

$$\langle \varphi_{\alpha k_2}^{(2)} | \hat{H} | \varphi_{\beta k_1}^{(1)} \rangle = \frac{1}{N \Omega_M} \int_{\Omega} dr \exp(-i\Delta k r) \sum_{R_1} g(r - R_1^\beta) \sum_{R_2} \exp(-i\bar{k}(R_2^\alpha - R_1^\beta)) t(R_2^\alpha - R_1^\beta)$$

Inner sum here is a matrix element for hamiltonian with momentum fixed (no momentum transfer when scattered from one layer to another). Summing over R_1 we obtain:

$$\langle \varphi_{\alpha k_2}^{(2)} | \hat{H} | \varphi_{\beta k_1}^{(1)} \rangle = \frac{1}{\Omega_M} \int_{\Omega_M} dr \exp(-i\Delta k r) U_{\alpha\beta}(\bar{k}, \delta(r))$$

Here functional $U_{\alpha\beta}(k, \delta(r))$ is introduced:

$$\begin{aligned} U_{\alpha\beta}(k, \delta(r)) &= \frac{1}{N} \sum_{R_1 R_2} \exp(-ik(R_2^\alpha - R_1^\beta)) t(R_2^\alpha - R_1^\beta) = \\ &= \sum_{nm} \exp(-ik((na_1 + ma_2 + \delta(r))) t(na_1 + ma_2 + d_0 e_z + \delta(r)) \end{aligned}$$

From all the sum over R_2^α, R_1^β contribute only those parts where $R_2^\alpha - R_1^\beta$ is not large. So to calculate this expression one should:

- Find $\delta(r)$ - vector pointing to the closest atom from another layer
- Sum hoppings over some small n, m

As $\delta(r)$ is periodic with period L_M , Fourier integral of $U_{\alpha\beta}(k, \delta(r))$ is not zero only when condition is satisfied:

$$\Delta k = m_1 G_1^M + m_2 G_2^M$$

, where m_1, m_2 are integers, G_1^M, G_2^M - Moire reciprocal vectors. Three parts are dominating by absolute value in Fourier decomposition. To check which ones do that and to obtain their contribution to the hamiltonian one can use script `FourierComponents.py` (or `TBGFourier` in somewhat more convenient form of Jupiter notebook).

3.2 Calculation procedure

In this project we use this agreement about lattice vectors:

$$a_1 = \begin{pmatrix} \frac{\sqrt{3}}{2} \\ \frac{1}{2} \end{pmatrix} a, \quad a_2 = \begin{pmatrix} \frac{\sqrt{3}}{2} \\ -\frac{1}{2} \end{pmatrix} a$$

Graphene reciprocal lattice vectors:

$$G_1 = \begin{pmatrix} \frac{1}{\sqrt{3}} \\ 1 \end{pmatrix} \frac{2\pi}{a}, \quad G_2 = \begin{pmatrix} \frac{1}{\sqrt{3}} \\ -1 \end{pmatrix} \frac{2\pi}{a}$$

Moire reciprocal lattice vectors:

$$G_1^M = \frac{4}{\sqrt{3}} \sin \frac{\theta}{2} \begin{pmatrix} -\frac{\sqrt{3}}{2} \\ \frac{1}{2} \end{pmatrix} \frac{2\pi}{a}, \quad G_2^M = \frac{4}{\sqrt{3}} \sin \frac{\theta}{2} \begin{pmatrix} \frac{\sqrt{3}}{2} \\ \frac{1}{2} \end{pmatrix} \frac{2\pi}{a}$$

And one of the three graphene Dirac points is chosen:

$$K = \frac{2\pi}{a} \begin{pmatrix} 0 \\ -\frac{2}{3} \end{pmatrix}$$

As input parameters one needs to set the angle of twist in degrees and the number of graphene unit cells N . The algorithm goes like that:

1. For each atom in the lattice of the bottom layer the closest atom is found using `Closes-tAtomFromTop` function
2. Hoppings from bottom to top (to closest atom and its 8 closest neighbours) are summed up. Hopping function used here is based on Slater-Koster parameters taken from [1]

3. To extract Fourier amplitude obtained expression is multiplied by $\exp(-i\overline{G}\overline{r})$
4. All obtained complex numbers are summed up
5. To make program workflow faster also functions ClosestAtomFromTopAimed and Hamilt-ForKAimed are used. The principle of their work is that they use saved position of closest atom from previous step and then look for closest atom on the actual step only in small window of 4 indices

Calling FourierHeatMap function one can get heatmaps of magnitudes of Fourier amplitudes. This may take a bit of time (it was about 5 minutes per plot). The result:

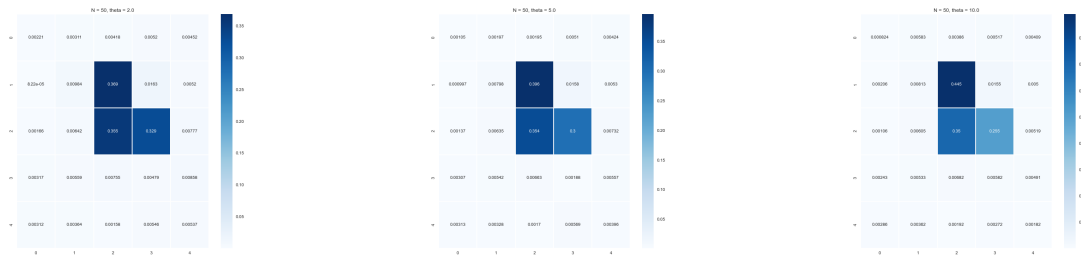


Figure 1: Heatmaps of magnitude of scattering amplitude. From these one can deduce that processes with $\Delta k = 0$, $-G_1^M$, G_2^M are dominant

Scatterings with dominant amplitudes are visible on these plots. For this scatterings one can calculate complex number representations using FourierComplexAmpl function. As this is done one can verify this form of 2×2 (here rows and columns stand for sublattice degree of freedom) matrices corresponding to each scattering processes:

$$\begin{array}{l} \Delta k = 0 \\ \Delta k = -G_1^M \\ \Delta k = G_2^M \end{array} \left| \begin{array}{c} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \\ \begin{pmatrix} 1 & e^{i\varphi} \\ e^{-i\varphi} & 1 \end{pmatrix} \\ \begin{pmatrix} 1 & e^{-i\varphi} \\ e^{i\varphi} & 1 \end{pmatrix} \end{array} \right.$$

Here $\varphi = \frac{2\pi}{3}$. Also it is possible to plot results of ClosestAtomFromTop and FourierHopping functions to visualise moire periodicity of minimal atom-to-atom distance and of hamiltonian matrix element. For visualization purposes also a tool to draw Moire structure itself is presented in MoireCrystalPlotter.

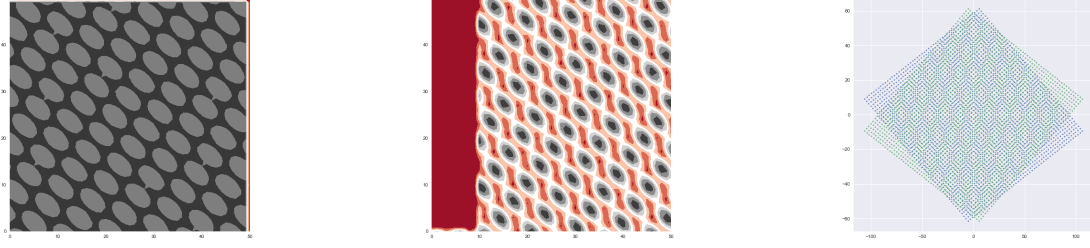


Figure 2: Moire periodicity visualisations: minimal atom-to-atom distance, amplitude of scattering, structure itself

4 Calculation of optical absorption

4.1 Hamiltonian of continuum model

As the periodicity of tBG is $\frac{L_M}{a}$ times larger than in graphene, moire brillouine zone (MBZ) is folded into a hexagon $\frac{L_M}{a}$ times less than original graphene BZ. Single-layer hamiltonian therefore should have $\frac{L_M}{a}$ more dimensions. However, for low-energy calculations only folded hexagons with not much distance from original Dirac point are important. Therefore we define tunable parameter N such that folded monolayer hamiltonian will have $2N \times 2N$ dimensions. Block matrix accounting for the second monolayer and for interlayer interaction is built in `tbg.py` in `TwistHamiltonian` function. In this program:

- All wavevector magnitudes are dimensionless as they are calculated in units of $\frac{2\pi}{a}$
- All energy quantities (and photon frequencies) are dimensionless as they are calculated in units of hopping integral t which is approximately equal to 3.1 eV

Then distance between Dirac points of two layers after rotation:

$$\Delta k = \frac{4}{3} \sin \frac{\theta}{2}$$

And primitive vector of Moire lattice is $\sqrt{3}$ times larger than Δk . In dimensionless units for monolayer its spectrum near K point is governed by absolute value of this function:

$$S(k_x, k_y) = \sqrt{3}\pi(k_y - ik_x)$$

For intralayer contributions for each monolayer k_x and k_y relative to the corresponding Dirac point is put in this formula. After zone folding electron states should be read as $|l, \alpha, n, m\rangle$, where l stands for layer 1 or layer 2, α - sublattice index, n - m - integers corresponding to translation from given point in MBZ to the point in original BZ on $nG_1^M + mG_2^M$. As the

interlayer interaction goes through three processes, in block matrix corresponding to it there will be three diagonal tapes of matrices. The resulting hamiltonian structure will be like that:

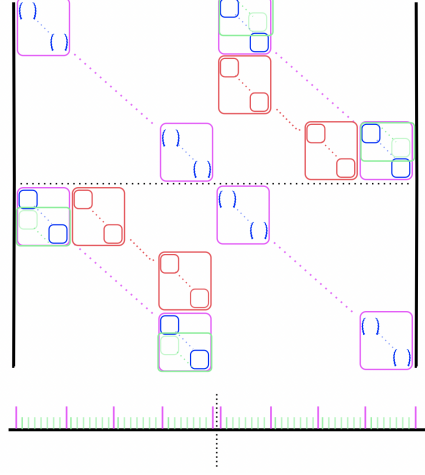


Figure 3: $4 \times (2N + 1) \times (2N + 1)$ Hamiltonian structure

Here on diagonal are monolayer 2×2 matrices. Below and above the diagonal are tapes of matrices.

$$\begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \quad \begin{pmatrix} 1 & e^{i\varphi} \\ e^{-i\varphi} & 1 \end{pmatrix} \quad \begin{pmatrix} 1 & e^{-i\varphi} \\ e^{i\varphi} & 1 \end{pmatrix}$$

First kind of matrices ($\Delta k = 0$) are painted with blue color on the picture. Second matrices ($\Delta k = -G_1^M$) stand in position of red squares. Third matrices ($\Delta k = G_2^M$) are depicted as green squares.

4.2 Kubo formula for tBG

From Kubo formula

$$\sigma_{xx}(\omega) = \frac{e^2 \hbar}{iS} \sum_{\alpha, \beta} \frac{f(\varepsilon_\alpha) - f(\varepsilon_\beta)}{\varepsilon_\alpha - \varepsilon_\beta} \frac{|\langle \alpha | v_x | \beta \rangle|^2}{\varepsilon_\alpha - \varepsilon_\beta + \hbar\omega + i\eta}$$

Here sum goes over all possible initial and final states and can therefore be splitted

$$\sum_{\alpha, \beta} = \sum_k \sum_{\text{pairs}}$$

Also it is useful to proceed firstly with the calculation of $\text{Re } \sigma_{xx}$ which is responsible for absorption of light and for high photon frequencies should give us twice of result for monolayer ([4]). Hence this expression will be useful for us:

$$\frac{1}{\varepsilon_\alpha - \varepsilon_\beta + \varepsilon + i\eta} = \text{P} \frac{1}{\varepsilon_\alpha - \varepsilon_\beta} - i\pi \delta(\varepsilon_\alpha - \varepsilon_\beta + \varepsilon)$$

Delta-function is replaced with Lorentzian for calculations:

$$\delta(\varepsilon_\alpha - \varepsilon_\beta + \varepsilon) = \frac{1}{\pi} \frac{\eta}{(\varepsilon_\alpha - \varepsilon_\beta + \varepsilon)^2 + \eta^2}$$

Here η is phenomenological parameter of line broadening. This is usually set in a range of 1 – 10 meV (i.e. 3 meV in [1]). Velocity operator $v_x = \frac{\partial H}{\partial k_x}$ is equal to diagonal matrix built from σ_x Pauli matrices extended to $4 \times (2N + 1) \times (2N + 1)$ dimensions:

$$v_x = \frac{\sqrt{3} a}{2} \hat{\sigma}_x$$

Area of a sample S is connected with the number of microscopic states involved in calculation scheme. The dimensionless distance between two closest states in k -space is:

$$k_{x \text{ step0}} = \frac{2\pi}{L_x} \bigg/ \frac{2\pi}{a} = \frac{a}{L_x}$$

Hence if k_{step0} is taken the same in all directions, then

$$\frac{1}{S} = \frac{(k_{\text{step0}})^2}{a^2}$$

Accounting for all this we can introduce the formula for dimensionless $\text{Re } \sigma_{xx}$:

$$\frac{\text{Re } \sigma_{xx}}{\sigma_{\text{mono}}} = N \sum_k \sum_{\text{pairs}} \frac{1}{\hbar\omega} \frac{|\langle \alpha | \hat{\sigma}_x | \beta \rangle|^2}{(\varepsilon_\alpha - \varepsilon_\beta - \hbar\omega)^2 + \eta^2} \eta$$

Here all energy quantities are dimensionless as they are in units of t . The numerical prefactor before this sum is defined by the grid chosen for calculation:

$$N = 12 k_{\text{step0}}^2$$

This formula is meant to be implemented on a square grid of k points close to the one of Dirac points. It was tested with monolayer hamiltonian, where this formula gave 1 as expected.

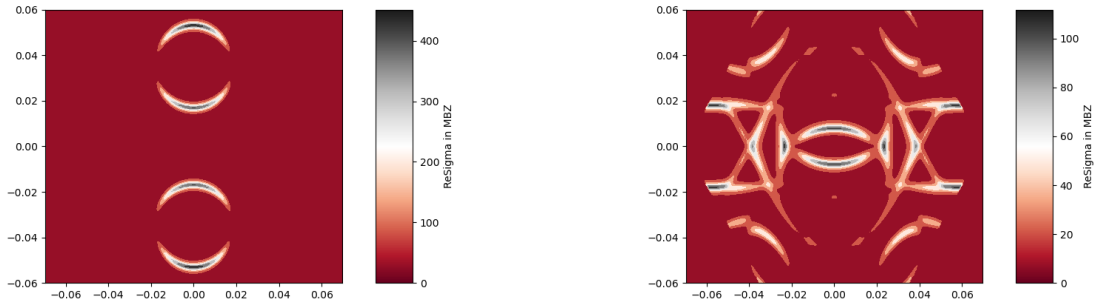


Figure 4: Example of spectral maps for two different photon energies for tBG with rotation angle of 6 degrees

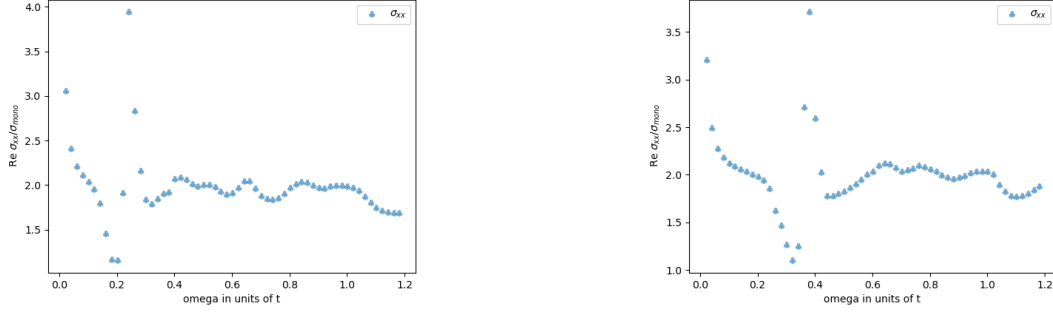


Figure 5: Two examples of $\text{Re } \sigma$ versus photon energy graphs for rotation angle of 4 and 6 degrees

4.3 Calculation procedure

The tools embedded in the project allow one to automatically (setting beforehand a range of needed angles and other parameters):

- Calculate eigensystem: eigenvalues and matrix element of velocity operator between the eigenstates. As this takes a lot of volume in the disk (more than 1 Gb per one set of calculation), eigensystem is automatically cleaned from the disk just after the calculation
- Plot density of states of twisted bilayer graphene with given configurations
- Plot spectral maps of $\text{Re } \sigma_{xx}$ in MBZ for each energy point involved in calculation of optical absorption
- Plot $\text{Re } \sigma_{xx}$ versus photon energy and save it to created folder with results. Save there also text file with calculated $\text{Re } \sigma_{xx}$

The execution time of the program is printed in terminal after each calculation. The program works much slower if increasing N (the dimensionality of hamiltonian) or decreasing $k_{\text{step}0}$ (taking more points in MBZ). For the "normal" configuration ($N = 4$, $k_{\text{step}0} \approx 0.001$) one calculation may take about an hour.

5 Optical constants of tBG

5.1 Kramers-Kronig relationships

To proceed from $\text{Re } \sigma_{xx}$ to optical constants one can use firstly Kramers-Kronig relationships for graphene. The description of those can be found in [5]. For dimensionless optical conductivities

and frequencies (in energy units of t) it can be rewritten as

$$\frac{\pi}{2} \text{Im } \sigma = \frac{1}{\omega} \int_0^{\Lambda} \frac{\nu^2}{\omega^2 - \nu^2} \text{Re } \sigma d\nu + \frac{2\Lambda}{\omega} + \log \frac{\Lambda - \omega}{\Lambda + \omega}$$

The cutoff parameter Λ here is the value where $\text{Re } \sigma$ has stabilised on level of 2. This cutoffs should be set manually in `optconsts.py` in array "cutoffs". The integral was converted to sum using trapezoidal method integration procedure. To avoid proximity of ω to Λ in the Kramers-Kronig expression an amount of calculation grid points is dropped when calculating $\text{Im } \sigma$ (set to 3 by default).

5.2 Conversion of complex σ to n, k

For a thin film a connection between complex σ and complex refractive index squared:

$$N^2 = 1 + \frac{4\pi\sigma}{\omega d}$$

d is the thickness of a thin film which can be estimated as $2 \times 3.35 \text{ nm}$ for tBG. Rewriting this formula in terms of calculated dimensionless complex σ :

$$N^2 = 1 + \alpha \pi \tilde{\sigma} \frac{c}{\omega d}$$

Here $\alpha = \frac{1}{137}$ is fine structure constant. Using also dimensionless frequencies in energy units of t and converting eV to Hz this formula can be introduced in this way:

$$N^2 = 1 + \beta \sigma, \quad \beta = \frac{\alpha \pi}{t} \frac{c [m/s]}{\tilde{\omega} \frac{1.6 \cdot 10^{-19}}{1.05 \cdot 10^{-34}} [Hz] \times d [m]}$$

Putting here all numerical values, $\beta \approx \frac{2.2}{\tilde{\omega}}$. β here depends on dimensionless frequency in units of t . A system of equations for refraction index and extinction coefficient then holds:

$$\begin{cases} n^2 - k^2 = 1 + \beta \text{Re } \sigma, \\ 2nk = \beta \text{Im } \sigma \end{cases}$$

Hence we can extract n, k using previously calculated $\text{Re } \sigma$ and $\text{Im } \sigma$. Script `optconsts.py` in the project does it and plots the dependencies of optical constants versus energy in units of t . Also boundaries of visible light photon energies are put in the graph.

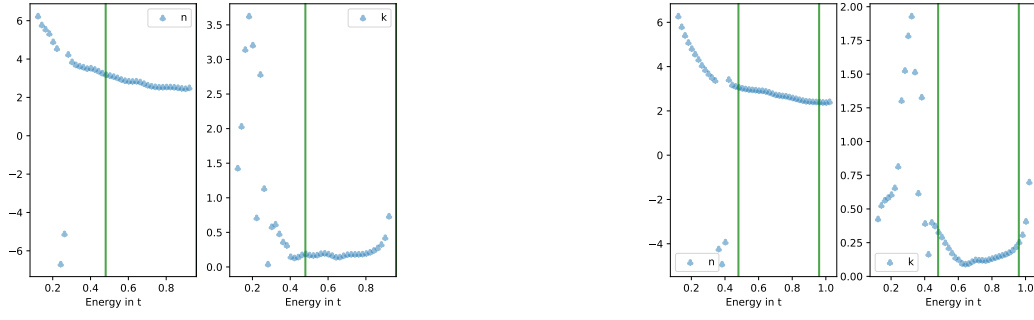


Figure 6: Two examples of optical constants versus photon energy graphs for tBG with rotation angle of 4 and 6 degrees

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