

# I Noninteracting EG-X Model and structure

## I.1 Lattice Structure of Graphene

Structure of honeycomb lattice following [yangStructureGrapheneIts2018].

Monolayer graphene forms a hexagonal lattice.

Primitive lattice vectors of the hexagonal lattice:

$$\mathbf{a}_1 = \frac{a}{2} \begin{pmatrix} 1 \\ \sqrt{3} \end{pmatrix} \quad (\text{I.1})$$

$$\mathbf{a}_2 = \frac{a}{2} \begin{pmatrix} 1 \\ -\sqrt{3} \end{pmatrix} \quad (\text{I.2})$$

with lattice constant  $a \approx 2.46 \text{ \AA}$  (distance between unit cells). Have

$$a = \sqrt{3}a_0 \quad (\text{I.3})$$

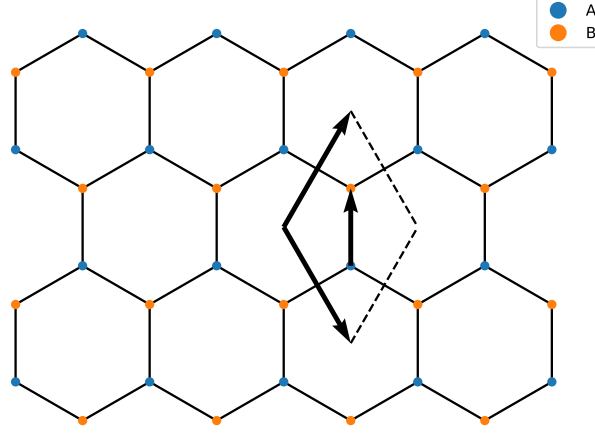
with the nearest-neighbour distance  $a_0$ .

Vectors to the nearest-neighbor  $B_i$  ( $i = 1, 2, 3$ ) atoms from atom  $A$ :

$$\boldsymbol{\delta}_{AB,1} = \begin{pmatrix} 0 \\ \frac{a}{\sqrt{3}} \end{pmatrix}, \boldsymbol{\delta}_{AB,2} = \begin{pmatrix} \frac{a}{2} \\ -\frac{a}{2\sqrt{3}} \end{pmatrix}, \boldsymbol{\delta}_{AB,3} = \begin{pmatrix} -\frac{a}{2} \\ -\frac{a}{2\sqrt{3}} \end{pmatrix} \quad (\text{I.4})$$

Vectors to the nearest-neighbor  $A_i$  ( $i = 1, 2, 3$ ) atoms from atom  $B$ :

$$\boldsymbol{\delta}_{BA,1} = \begin{pmatrix} 0 \\ -\frac{a}{\sqrt{3}} \end{pmatrix}, \boldsymbol{\delta}_{BA,2} = \begin{pmatrix} \frac{a}{2} \\ \frac{a}{2\sqrt{3}} \end{pmatrix}, \boldsymbol{\delta}_{BA,3} = \begin{pmatrix} -\frac{a}{2} \\ \frac{a}{2\sqrt{3}} \end{pmatrix} \quad (\text{I.5})$$



**Figure I.1:** Graphene lattice structure

The vectors between the Graphene A atom and the six neighbours on the same sub lattice can be found by rotating  $\mathbf{a}_1$  six times by  $1/6 * 2\pi = \pi/3$ :

$$\delta_{AA,1} = \mathbf{a}_1 = \frac{a}{2} \begin{pmatrix} 1 \\ \sqrt{3} \end{pmatrix} = a \begin{pmatrix} \frac{1}{2} \\ \frac{\sqrt{3}}{2} \end{pmatrix} = a \begin{pmatrix} \sin(\frac{\pi}{6}) \\ \cos(\frac{\pi}{6}) \end{pmatrix} \quad (\text{I.6})$$

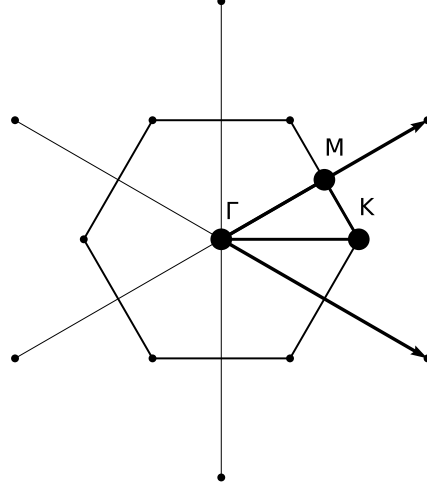
$$\delta_{AA,2} = a \begin{pmatrix} \sin(\frac{3\pi}{6}) \\ \cos(\frac{3\pi}{6}) \end{pmatrix} = a \begin{pmatrix} 1 \\ 0 \end{pmatrix} \quad (\text{I.7})$$

$$\delta_{AA,3} = a \begin{pmatrix} \sin(\frac{5\pi}{6}) \\ \cos(\frac{5\pi}{6}) \end{pmatrix} = a \begin{pmatrix} \frac{1}{2} \\ -\frac{\sqrt{3}}{2} \end{pmatrix} \quad (\text{I.8})$$

$$\delta_{AA,4} = a \begin{pmatrix} \sin(\frac{7\pi}{6}) \\ \cos(\frac{7\pi}{6}) \end{pmatrix} = a \begin{pmatrix} -\frac{1}{2} \\ -\frac{\sqrt{3}}{2} \end{pmatrix} \quad (\text{I.9})$$

$$\delta_{AA,5} = a \begin{pmatrix} \sin(\frac{9\pi}{6}) \\ \cos(\frac{9\pi}{6}) \end{pmatrix} = a \begin{pmatrix} -1 \\ 0 \end{pmatrix} \quad (\text{I.10})$$

$$\delta_{AA,6} = a \begin{pmatrix} \sin(\frac{11\pi}{6}) \\ \cos(\frac{11\pi}{6}) \end{pmatrix} = a \begin{pmatrix} -\frac{1}{2} \\ \frac{\sqrt{3}}{2} \end{pmatrix} \quad (\text{I.11})$$



**Figure I.2:** *Graphene Brillouin Zone*

The primitive reciprocal lattice vectors  $\mathbf{b}_1$ ,  $\mathbf{b}_2$  fulfill

$$\mathbf{a}_1 \cdot \mathbf{b}_1 = \mathbf{a}_2 \cdot \mathbf{b}_2 = 2\pi \quad (\text{I.12})$$

$$\mathbf{a}_1 \cdot \mathbf{b}_2 = \mathbf{a}_2 \cdot \mathbf{b}_1 = 0, \quad (\text{I.13})$$

so we have:

$$\mathbf{b}_1 = \frac{2\pi}{a} \begin{pmatrix} 1 \\ \frac{1}{\sqrt{3}} \end{pmatrix} \quad (\text{I.14})$$

$$\mathbf{b}_2 = \frac{2\pi}{a} \begin{pmatrix} 1 \\ -\frac{1}{\sqrt{3}} \end{pmatrix} \quad (\text{I.15})$$

Points of high symmetry in the Brillouin zone are:

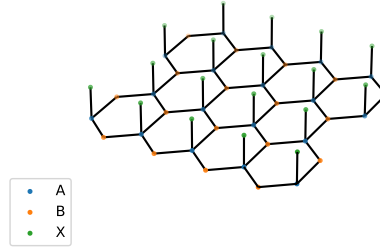
$$\Gamma = \begin{pmatrix} 0 \\ 0 \end{pmatrix} \quad (\text{I.16})$$

$$\text{M} = \frac{\pi}{a} \begin{pmatrix} 1 \\ \frac{1}{\sqrt{3}} \end{pmatrix} \quad (\text{I.17})$$

$$\text{K} = \frac{4\pi}{3a} \begin{pmatrix} 1 \\ 0 \end{pmatrix} \quad (\text{I.18})$$

## I.2 EG-X Model

Graphene lattice and a site X. Real-life motivation: layer of graphene on top



**Figure I.3:** *EG-X model*

of a substrate of another material (which provides the additional X atoms). There is no spin-orbit coupling considered in the model (but when according to Niklas: when mapping to substrates Sn or Pb, it could be necessary (but does not the qualitative result?)).

Spin-orbit coupling,  
drop second spin  
index?

Without interaction :

$$H_0 = -t_X \sum_{\langle ij \rangle, \sigma \sigma'} d_{i,\sigma}^\dagger d_{j,\sigma'} + \text{h.c.} - t_{\text{Gr}} \sum_{\langle ij \rangle, \sigma \sigma'} \left( c_{i,\sigma}^{(A),\dagger} c_{j,\sigma'}^{(B)} + c_{j,\sigma'}^{(B),\dagger} c_{i,\sigma}^{(A)} + \text{h.c.} \right) \quad (\text{I.19})$$

$$+ V \sum_{i, \sigma \sigma'} \left( d_{i,\sigma}^\dagger c_{i,\sigma'}^{(A)} + c_{i,\sigma}^{(A),\dagger} d_{i,\sigma'} \right) \quad (\text{I.20})$$

with:

- $d$  operators on the X atom
- $c^{(\epsilon)}$  operators on the graphene site ( $\epsilon = A, B$ )
- $t_X$  NN hopping for X
- $t_{\text{Gr}}$  NN hopping of Gr
- $V$  hybridization between X and Graphene B sites

We can also introduce an onsite Hubbard interaction:

$$H_{\text{int}} = U_X \sum_i d_{i,\uparrow}^\dagger d_{i,\downarrow}^\dagger d_{i,\downarrow} d_{i,\uparrow} + U_{\text{Gr}} \sum_{i, \epsilon=A,B} c_{i,\uparrow}^{(\epsilon)\dagger} c_{i,\downarrow}^{(\epsilon)\dagger} c_{i,\downarrow}^\epsilon c_{i,\uparrow}^\epsilon \quad (\text{I.21})$$

### I.2.1 Review: Hubbard model on the honeycomb lattice

### I.2.2 Band structure of the non-interacting EG-X model

To treat eq. I.20, we first write out the sums over nearest neighbours  $\langle i, j \rangle$  explicitly, writing  $\delta_X, \delta_\epsilon$  ( $\epsilon = A, B$ ) for the connections to the nearest neighbours of the X atoms and Graphene A, B sites. Doing the calculation for the example of the X atoms:

$$-t_X \sum_{\langle ij \rangle, \sigma \sigma'} (d_{i,\sigma}^\dagger d_{j,\sigma'} + d_{j,\sigma}^\dagger d_{i,\sigma'}) \quad (\text{I.22})$$

$$= -\frac{t_X}{2} \sum_{i, \sigma, \sigma'} \sum_{\delta_X} d_{i,\sigma}^\dagger d_{i+\delta_X, \sigma'} - \frac{t_X}{2} \sum_{j, \sigma, \sigma'} \sum_{\delta_X} d_{j,\sigma}^\dagger d_{j+\delta_X, \sigma'} \quad (\text{I.23})$$

$$= -t_X \sum_{i, \sigma, \sigma'} \sum_{\delta_X} d_{i,\sigma}^\dagger d_{i+\delta_X, \sigma'} \quad (\text{I.24})$$

Write review for  
Hubbard model on  
the honeycomb lat-  
tice

(The factor  $1/2$  is to account for double counting when going to the sum over all lattice sites  $i$ )

Now we can input the discrete Fourier transform (for both graphene and X operators) into eq. [I.24](#)

$$c_i = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{i\mathbf{k}\mathbf{r}_i} c_{\mathbf{k}} \quad (\text{I.25})$$

$$c_i^\dagger = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{-i\mathbf{k}\mathbf{r}_i} c_{\mathbf{k}}^\dagger \quad (\text{I.26})$$

with the completeness relation:

$$\sum_i e^{i\mathbf{k}\mathbf{r}_i} e^{-i\mathbf{k}'\mathbf{r}_i} = N\delta_{\mathbf{k},\mathbf{k}'} . \quad (\text{I.27})$$

We get:

$$-t_X \frac{1}{N} \sum_{i,\sigma,\sigma'} \sum_{\delta_X} d_{i,\sigma}^\dagger d_{i+\delta_X,\sigma'} = -t_X \frac{1}{N} \sum_{i,\sigma,\sigma'} \sum_{\delta_X} \sum_{\mathbf{k},\mathbf{k}'} e^{-i\mathbf{k}\mathbf{r}_i} d_{\mathbf{k},\sigma}^\dagger e^{i\mathbf{k}'\mathbf{r}_i} e^{i\mathbf{k}'\delta_X} d_{\mathbf{k}',\sigma'} \quad (\text{I.28})$$

$$= -t_X \frac{1}{N} \sum_{\mathbf{k},\mathbf{k}',\sigma,\sigma'} \sum_{\delta_X} d_{\mathbf{k},\sigma}^\dagger e^{i\mathbf{k}'\delta_X} d_{\mathbf{k}',\sigma'} \sum_i e^{-i\mathbf{k}\mathbf{r}_i} e^{i\mathbf{k}'\mathbf{r}_i} \quad (\text{I.29})$$

$$= -t_X \frac{1}{N} \sum_{\mathbf{k},\mathbf{k}',\sigma,\sigma'} \sum_{\delta_X} d_{\mathbf{k},\sigma}^\dagger e^{i\mathbf{k}'\delta_X} d_{\mathbf{k}',\sigma'} N\delta_{\mathbf{k},\mathbf{k}'} \quad (\text{I.30})$$

$$= -t_X \sum_{\mathbf{k},\sigma,\sigma'} d_{\mathbf{k},\sigma}^\dagger d_{\mathbf{k},\sigma'} \sum_{\delta_X} e^{i\mathbf{k}\delta_X} \quad (\text{I.31})$$

The nearest neighbours for X atoms are the vectors  $\delta_{AA,i}$  from section I.1. With that, we can calculate:

$$f_X(\mathbf{k}) = -t_X \sum_{\delta_X} e^{i\mathbf{k}\delta_X} \quad (\text{I.32})$$

$$= -t_X \left( e^{ia(\frac{k_x}{2} + \frac{\sqrt{3}k_y}{2})} + e^{iak_x} + e^{ia(\frac{k_x}{2} - \frac{\sqrt{3}k_y}{2})} \right. \quad (\text{I.33})$$

$$\left. + e^{ia(-\frac{k_x}{2} - \frac{\sqrt{3}k_y}{2})} + e^{-iak_x} + e^{ia(-\frac{k_x}{2} + \frac{\sqrt{3}k_y}{2})} \right) \quad (\text{I.34})$$

$$= -t_X \left( 2 \cos(ak_x) + 2e^{ia\frac{\sqrt{3}k_y}{2}} \cos\left(\frac{a}{2}k_x\right) + 2e^{-ia\frac{\sqrt{3}k_y}{2}} \cos\left(\frac{a}{2}k_x\right) \right) \quad (\text{I.35})$$

$$= -2t_X \left( \cos(ak_x) + 2 \cos\left(\frac{a}{2}k_x\right) \cos\left(\sqrt{3}\frac{a}{2}k_y\right) \right) \quad (\text{I.36})$$

We can do the same for the hopping between Graphene sites, for example :

$$-t_{Gr} \sum_{\langle ij \rangle, \sigma\sigma'} c_{i,\sigma}^{(A)\dagger} c_{j,\sigma'}^{(B)} = -t_{Gr} \sum_{i,\sigma\sigma'} \sum_{\delta_{AB}} c_{i,\sigma}^{(A)\dagger} c_{i+\delta_{AB},\sigma'}^{(B)} \quad (\text{I.37})$$

$$= -t_{Gr} \sum_{\mathbf{k},\sigma,\sigma'} c_{\mathbf{k},\sigma}^{(A)\dagger} c_{\mathbf{k},\sigma'}^{(B)} \sum_{\delta_{AB}} e^{i\mathbf{k}\delta_{AB}} \quad (\text{I.38})$$

We note

$$\sum_{\delta_{AB}} e^{i\mathbf{k}\delta_{AB}} = \left( \sum_{\delta_{BA}} e^{i\mathbf{k}\delta_{BA}} \right)^* = \sum_{\delta_{BA}} e^{-i\mathbf{k}\delta_{BA}} \quad (\text{I.39})$$

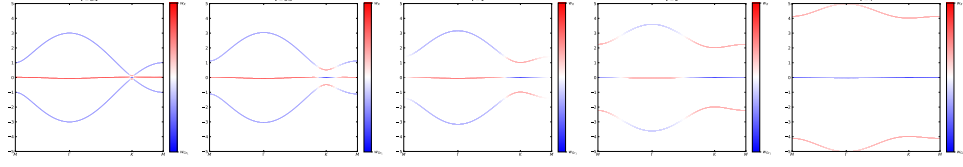
and calculate

$$f_{Gr} = -t_{Gr} \sum_{\delta_{AB}} e^{i\mathbf{k}\delta_{AB}} \quad (\text{I.40})$$

$$= -t_{Gr} \left( e^{i\frac{a}{\sqrt{3}}k_y} + e^{i\frac{a}{2\sqrt{3}}(\sqrt{3}k_x - k_y)} + e^{i\frac{a}{2\sqrt{3}}(-\sqrt{3}k_x - k_y)} \right) \quad (\text{I.41})$$

$$= -t_{Gr} \left( e^{i\frac{a}{\sqrt{3}}k_y} + e^{-i\frac{a}{2\sqrt{3}}k_y} \left( e^{i\frac{a}{2}k_x} + e^{-i\frac{a}{2}k_x} \right) \right) \quad (\text{I.42})$$

$$= -t_{Gr} \left( e^{i\frac{a}{\sqrt{3}}k_y} + 2e^{-i\frac{a}{2\sqrt{3}}k_y} \cos\left(\frac{a}{2}k_x\right) \right) \quad (\text{I.43})$$



**Figure I.4:** Bands of the non-interacting EG-X model. All the bands are spin-degenerate.

All together, we get:

$$H_0 = \sum_{\mathbf{k}, \sigma, \sigma'} \begin{pmatrix} c_{\mathbf{k}, \sigma}^{A, \dagger} & c_{\mathbf{k}, \sigma}^{B, \dagger} & d_{\mathbf{k}, \sigma}^{\dagger} \end{pmatrix} \begin{pmatrix} 0 & f_{Gr} & V \\ f_{Gr}^* & 0 & 0 \\ V & 0 & f_X \end{pmatrix} \begin{pmatrix} c_{\mathbf{k}, \sigma}^A \\ c_{\mathbf{k}, \sigma}^B \\ d_{\mathbf{k}, \sigma} \end{pmatrix} \quad (\text{I.44})$$

The band structure for the non-interacting EG-X model is easily obtained by diagonalising the matrix in eq. I.44. This was done in fig. I.4.

Values used for calculation:

- $a_0 = 1$
- $t_{Gr} = 1$
- $t_X = 0.01$

$V$  is the control parameter. (According to Niklas), a range from  $V = 0.1$  to  $V = 2$  can be mapped onto materials in experiment.