I Noninteracting EG-X Model and structure

I.1 Lattice Structure of Graphene

Structure of honeycomb lattice following [**Yang_Li_Lee_Ng_2018**].
Monolayer graphene forms a hexagonal lattice.
Primitive lattice vectors of the hexagonal lattice:

$$\mathbf{a}_1 = \frac{a}{2}\begin{pmatrix} 1 \\ \sqrt{3} \end{pmatrix} \tag{I.1}$$

$$\mathbf{a}_2 = \frac{a}{2}\begin{pmatrix} 1 \\ -\sqrt{3} \end{pmatrix} \tag{I.2}$$

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

$$a = \sqrt{3}a_0 \tag{I.3}$$

with the nearest-neighbour distance a_0.
Vectors to the nearest-neighbor B_i $(i = 1, 2, 3,)$ atoms from atom A:

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

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

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

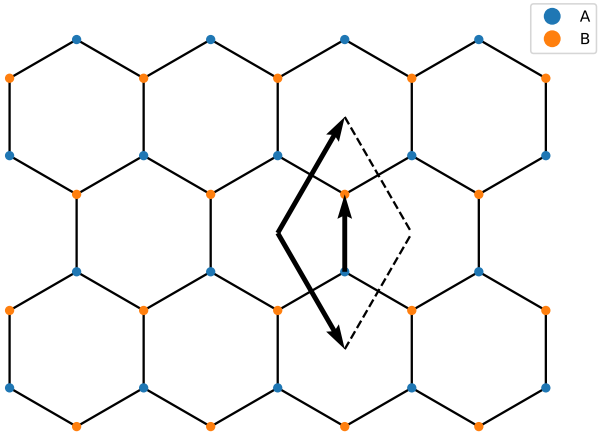

Figure I.1: *Graphene lattice structure*

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 $1/6 * 2\pi = \pi/3$:

$$\boldsymbol{\delta}_{AA,1} = \mathbf{a}_1 = \frac{a}{2}\begin{pmatrix}1\\ \sqrt{3}\end{pmatrix} = a\begin{pmatrix}\frac{1}{2}\\ \frac{\sqrt{3}}{2}\end{pmatrix} = a\begin{pmatrix}\sin\left(\frac{\pi}{6}\right)\\ \cos\left(\frac{\pi}{6}\right)\end{pmatrix} \tag{I.6}$$

$$\boldsymbol{\delta}_{AA,2} = a\begin{pmatrix}\sin\left(\frac{3\pi}{6}\right)\\ \cos\left(\frac{3\pi}{6}\right)\end{pmatrix} = a\begin{pmatrix}1\\ 0\end{pmatrix} \tag{I.7}$$

$$\boldsymbol{\delta}_{AA,3} = a\begin{pmatrix}\sin\left(\frac{5\pi}{6}\right)\\ \cos\left(\frac{5\pi}{6}\right)\end{pmatrix} = a\begin{pmatrix}\frac{1}{2}\\ -\frac{\sqrt{3}}{2}\end{pmatrix} \tag{I.8}$$

$$\boldsymbol{\delta}_{AA,4} = a\begin{pmatrix}\sin\left(\frac{7\pi}{6}\right)\\ \cos\left(\frac{7\pi}{6}\right)\end{pmatrix} = a\begin{pmatrix}-\frac{1}{2}\\ -\frac{\sqrt{3}}{2}\end{pmatrix} \tag{I.9}$$

$$\boldsymbol{\delta}_{AA,5} = a\begin{pmatrix}\sin\left(\frac{9\pi}{6}\right)\\ \cos\left(\frac{9\pi}{6}\right)\end{pmatrix} = a\begin{pmatrix}-1\\ 0\end{pmatrix} \tag{I.10}$$

$$\boldsymbol{\delta}_{AA,6} = a\begin{pmatrix}\sin\left(\frac{11\pi}{6}\right)\\ \cos\left(\frac{11\pi}{6}\right)\end{pmatrix} = a\begin{pmatrix}-\frac{1}{2}\\ \frac{\sqrt{3}}{2}\end{pmatrix} \tag{I.11}$$

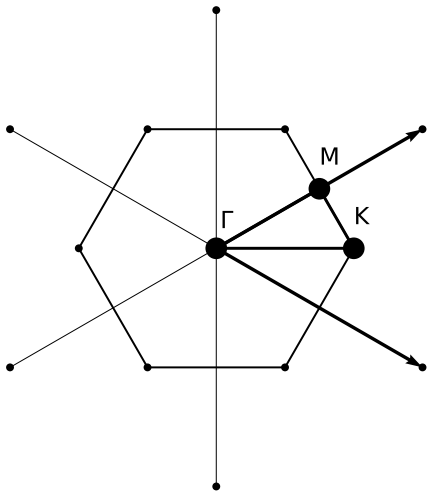

Figure I.2: *Graphene Brillouin Zone*

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

$$\mathbf{a}_1 \cdot \mathbf{b}_1 = \mathbf{a}_2 \cdot \mathbf{b}_2 = 2\pi \tag{I.12}$$

$$\mathbf{a}_1 \cdot \mathbf{b}_2 = \mathbf{a}_2 \cdot \mathbf{b}_1 = 0 \,, \tag{I.13}$$

so we have:

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

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

Points of high symmetry in the Brillouin zone are:

$$\Gamma = \begin{pmatrix} 0 \\ 0 \end{pmatrix} \tag{I.16}$$

$$\mathrm{M} = \frac{\pi}{a} \begin{pmatrix} 1 \\ \frac{1}{\sqrt{3}} \end{pmatrix} \tag{I.17}$$

$$\mathrm{K} = \frac{4\pi}{3a} \begin{pmatrix} 1 \\ 0 \end{pmatrix} \tag{I.18}$$

I.2 EG-X Model

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

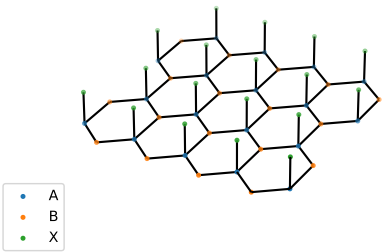

Figure I.3: *EG-X model*

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 :

$$H_0 = -t_{\mathrm{X}} \sum_{\langle ij\rangle,\sigma\sigma'} d^\dagger_{i,\sigma} d_{j,\sigma'} + \mathrm{h.c.} - t_{\mathrm{Gr}} \sum_{\langle ij\rangle,\sigma\sigma'} \left(c^{(A),\dagger}_{i,\sigma} c^{(B)}_{j,\sigma'} + c^{(B),\dagger}_{j,\sigma'} c^{(A)}_{i,\sigma} + \mathrm{h.c.} \right) \tag{I.19}$$

$$+ V \sum_{i,\sigma\sigma'} \left(d^\dagger_{i,\sigma} c^{(A)}_{i,\sigma'} + c^{(A),\dagger}_{i,\sigma} d_{i,\sigma'} \right) \tag{I.20}$$

with:

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

We can also introduce an onsite Hubbard interaction:

$$H_{\mathrm{int}} = U_{\mathrm{X}} \sum_i d^\dagger_{i,\uparrow} d^\dagger_{i,\downarrow} d_{i,\downarrow} d_{i,\uparrow} + U_{\mathrm{Gr}} \sum_{i,\epsilon=A,B} c^{(\epsilon)\dagger}_{i,\uparrow} c^{(\epsilon)\dagger}_{i,\downarrow} c^{\epsilon}_{i,\downarrow} c^{\epsilon}_{i,\uparrow} \tag{I.21}$$

I.2.1 Review: Hubbard model on the honeycomb lattice

Write review for 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 $\boldsymbol{\delta}_{\mathrm{X}}, \boldsymbol{\delta}_\epsilon$ ($\epsilon = A, B$) 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:

$$- t_{\mathrm{X}} \sum_{\langle ij\rangle,\sigma\sigma'} (d^\dagger_{i,\sigma} d_{j,\sigma'} + d^\dagger_{j,\sigma} d_{i,\sigma'}) \tag{I.22}$$

$$= -\frac{t_X}{2} \sum_{i,\sigma,\sigma'} \sum_{\delta_{\mathrm{X}}} d^\dagger_{i,\sigma} d_{i+\delta_{\mathrm{X}},\sigma'} - \frac{t_X}{2} \sum_{j,\sigma,\sigma'} \sum_{\delta_{\mathrm{X}}} d^\dagger_{j,\sigma} d_{j+\delta_{\mathrm{X}},\sigma'} \tag{I.23}$$

$$= -t_X \sum_{i,\sigma,\sigma'} \sum_{\delta_{\mathrm{X}}} d^\dagger_{i,\sigma} d_{i+\delta_{\mathrm{X}},\sigma'} \tag{I.24}$$

(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

$$c_i = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{\mathrm{i}\mathbf{k}\mathbf{r}_i} c_{\mathbf{k}} \tag{I.25}$$

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

with the completeness relation:

$$\sum_i e^{\mathrm{i}\mathbf{k}\mathbf{r}_i} e^{-\mathrm{i}\mathbf{k}'\mathbf{r}_i} = N\delta_{\mathbf{k},\mathbf{k}'} \ . \tag{I.27}$$

We get:

$$-t_X \frac{1}{N} \sum_{i,\sigma,\sigma'} \sum_{\boldsymbol{\delta}_\mathrm{X}} d_{i,\sigma}^\dagger d_{i+\boldsymbol{\delta}_\mathrm{X},\sigma'} = -t_X \frac{1}{N} \sum_{i,\sigma,\sigma'} \sum_{\boldsymbol{\delta}_\mathrm{X}} \sum_{\mathbf{k},\mathbf{k}'} e^{-\mathrm{i}\mathbf{k}\mathbf{r}_i} d_{\mathbf{k},\sigma}^\dagger e^{\mathrm{i}\mathbf{k}'\mathbf{r}_i} e^{\mathrm{i}\mathbf{k}'\boldsymbol{\delta}_\mathrm{X}} d_{\mathbf{k}',\sigma'} \tag{I.28}$$

$$= -t_X \frac{1}{N} \sum_{\mathbf{k},\mathbf{k}',\sigma,\sigma'} \sum_{\boldsymbol{\delta}_\mathrm{X}} d_{\mathbf{k},\sigma}^\dagger e^{\mathrm{i}\mathbf{k}'\boldsymbol{\delta}_\mathrm{X}} d_{\mathbf{k}',\sigma'} \sum_i e^{-\mathrm{i}\mathbf{k}\mathbf{r}_i} e^{\mathrm{i}\mathbf{k}'\mathbf{r}_i} \tag{I.29}$$

$$= -t_X \frac{1}{N} \sum_{\mathbf{k},\mathbf{k}',\sigma,\sigma'} \sum_{\boldsymbol{\delta}_\mathrm{X}} d_{\mathbf{k},\sigma}^\dagger e^{\mathrm{i}\mathbf{k}'\boldsymbol{\delta}_\mathrm{X}} d_{\mathbf{k}',\sigma'} N\delta_{\mathbf{k},\mathbf{k}'} \tag{I.30}$$

$$= -t_X \sum_{\mathbf{k},\sigma,\sigma'} d_{\mathbf{k},\sigma}^\dagger d_{\mathbf{k},\sigma'} \sum_{\boldsymbol{\delta}_\mathrm{X}} e^{\mathrm{i}\mathbf{k}\boldsymbol{\delta}_\mathrm{X}} \tag{I.31}$$

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

$$f_{\mathrm{X}}(\mathbf{k}) = -t_X \sum_{\boldsymbol{\delta}_{\mathrm{X}}} e^{\mathrm{i}\mathbf{k}\boldsymbol{\delta}_{\mathrm{X}}} \tag{I.32}$$

$$= -t_X \left(e^{\mathrm{i}a(\frac{k_x}{2}+\frac{\sqrt{3}k_y}{2})} + e^{\mathrm{i}ak_x} + e^{\mathrm{i}a(\frac{k_x}{2}-\frac{\sqrt{3}k_y}{2})} \right. \tag{I.33}$$

$$\left. + e^{\mathrm{i}a(-\frac{k_x}{2}-\frac{\sqrt{3}k_y}{2})} + e^{-\mathrm{i}ak_x} + e^{\mathrm{i}a(-\frac{k_x}{2}+\frac{\sqrt{3}k_y}{2})} \right) \tag{I.34}$$

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

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

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

$$-t_{\mathrm{Gr}} \sum_{\langle ij\rangle,\sigma\sigma'} c^{(A),\dagger}_{i,\sigma} c^{(B)}_{j,\sigma'} = -t_{\mathrm{Gr}} \sum_{i,\sigma\sigma'} \sum_{\boldsymbol{\delta}_{AB}} c^{(A),\dagger}_{i,\sigma} c^{(B)}_{i+\boldsymbol{\delta}_{AB},\sigma'} \tag{I.37}$$

$$= -t_{\mathrm{Gr}} \sum_{\mathbf{k},\sigma,\sigma'} c^{(A)\dagger}_{\mathbf{k},\sigma} c^{(B)}_{\mathbf{k},\sigma'} \sum_{\boldsymbol{\delta}_{AB}} e^{\mathrm{i}\mathbf{k}\boldsymbol{\delta}_{AB}} \tag{I.38}$$

We note

$$\sum_{\boldsymbol{\delta}_{AB}} e^{\mathrm{i}\mathbf{k}\boldsymbol{\delta}_{AB}} = \left(\sum_{\boldsymbol{\delta}_{BA}} e^{\mathrm{i}\mathbf{k}\boldsymbol{\delta}_{BA}} \right)^* = \sum_{\boldsymbol{\delta}_{BA}} e^{-\mathrm{i}\mathbf{k}\boldsymbol{\delta}_{BA}} \tag{I.39}$$

and calculate

$$f_{Gr} = -t_{Gr} \sum_{\boldsymbol{\delta}_{AB}} e^{\mathrm{i}\mathbf{k}\boldsymbol{\delta}_{AB}} \tag{I.40}$$

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

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

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

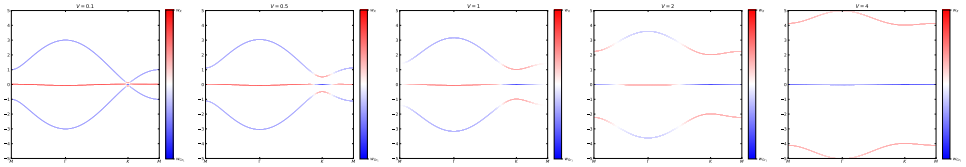

Figure I.4: *Bands of the non-interacting EG-X model. All the bands are spin-degenerate.*

All together, we get:

$$H_0 = \sum_{\mathbf{k},\sigma,\sigma'} \begin{pmatrix} c^{A,\dagger}_{k,\sigma} & c^{B,\dagger}_{k,\sigma} & d^{\dagger}_{k,\sigma} \end{pmatrix} \begin{pmatrix} 0 & f_{Gr} & V \\ f^{*}_{Gr} & 0 & 0 \\ V & 0 & f_X \end{pmatrix} \begin{pmatrix} c^{A}_{k,\sigma} \\ c^{B}_{k,\sigma} \\ d_{k,\sigma} \end{pmatrix} \tag{I.44}$$

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:

- $a_0 = 1$
- $t_{\mathrm{Gr}} = 1$
- $t_{\mathrm{X}} = 0.01$

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