Motivation and Introduction

In crystalline materials comprised of relatively well localized orbitals or Wannier functions, the tight binding description is appropriate. In this description, orbitals $|n\rangle$ are chosen to occupy a specific location r_n in space and electronic conduction is mediated by a hopping term $t(r_n - r_m, \sigma_n, \sigma_m)$ between these orbitals. Here σ_n describes spin and other non-location characteristics of the orbital $|n\rangle$. The tight binding model is then expressed in real space as

$$H_{\text{TB}} = - \sum_{\text{sites } n,m; \text{ unit cells } i,j} \left(t(r_{n,i} - r_{m,j}) c_{n,i}^\dagger c_{m,j} + t(r_{m,j} - r_{n,i}) c_{m,j}^\dagger c_{n,i} \right)$$

where for simplicity we suppress any dependence on σ_n since this only changes the value of t. The leading negative sign is used to indicate that this is a model of bound electrons. The i and j indices correspond to translations by lattice vectors $\alpha a_1 + \beta a_2$.

Now, this can be taken to momentum space using

$$c_{n,i}^\dagger = \frac{1}{\sqrt{\# \text{ of sites}/N}} \sum_k e^{ik \cdot r_{n,i}} c_{n,k}^\dagger$$

where N is the number of sites in the unit cell. Substituting in we find

$$H_{\text{TB}} = -\frac{1}{\# \text{ of sites}/N} \sum_{\text{sites } n,m; \text{ unit cells } i,j} \sum_{k,k'} \left(t(r_{n,i} - r_{m,j}) e^{ik \cdot r_{n,i}} e^{-ik' \cdot r_{m,j}} c_{n,k}^\dagger c_{m,k} \right.$$

$$\left. + t(r_{m,j} - r_{n,i}) e^{ik' \cdot r_{m,j}} e^{-ik \cdot r_{n,i}} c_{m,k}^\dagger c_{n,k} \right)$$

$$= - \sum_{\text{sites } n,m; \text{ unit cells } i} \sum_k \left(t(d_{nm} + d_i) e^{-ik \cdot (d_{nm} + d_i)} c_{n,k}^\dagger c_{m,k} \right.$$

$$\left. + t(d_{mn} + d_i) e^{ik \cdot (d_{mn} + d_i)} c_{m,k}^\dagger c_{n,k} \right)$$

where we used the delta function identity

$$\frac{1}{\# \text{ of sites}/N} \sum_{\text{sites; unit cells}} \sum_{k,k'} e^{i(k-k') \cdot r_\alpha} = \delta(k - k')$$

for some indices α. Here $d_{nm} = r_{n,i} - r_{m,i}$ is the vector between sites n and m in the same unit cell, and d_i is the vector from the unit cell the electrons start in to the unit cell they end up in.

Bipartite Lattice: Monolayer Graphene

Graphene's lattice is bipartite consisting of an "A" sublattice and a "B" sublattice—see Fig 1.
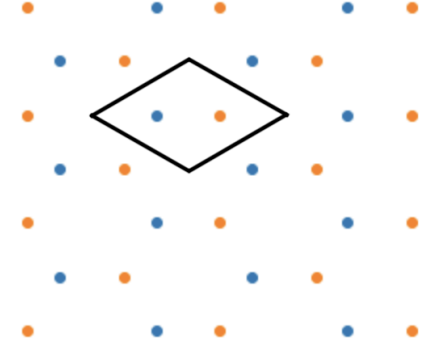

Fig 1. Structure of monolayer graphene. Unit cell is outlined in black. The distance between nearest neighbor sites of the same color is 0.246 nm, while the distance between nearest neighbor sites of different colors is 0.142 nm.

Now, the hopping term is then given by

$$t(|\vec{r}|) = \begin{cases} 2.7 \text{ eV}, & |\vec{r}| = a_0 \\ 0 \text{ eV}, & |\vec{r}| \neq a_0 \end{cases}$$

which is only non-vanishing for nearest-neighbor hopping terms. So, summing over all terms that are non-zero we find

$$H_{NN} = -\sum_k (t(d_1)e^{-ik\cdot d_1} + t(d_2)e^{-ik\cdot d_2} + t(d_3)e^{-ik\cdot d_3})a_k^\dagger b_k + (t(d_1)e^{ik\cdot d_1} + t(d_2)e^{ik\cdot d_2} + t(d_3)e^{ik\cdot d_3})b_k^\dagger a_k$$

$$= -2.7\sum_k (e^{-ik\cdot d_1} + e^{-ik\cdot d_2} + e^{-ik\cdot d_3})a_k^\dagger b_k + (e^{ik\cdot d_1} + e^{ik\cdot d_2} + e^{ik\cdot d_3})b_k^\dagger a_k$$

Or in matrix form

$$H_{NN}(k) = \begin{pmatrix} \langle a|H_{NN}(k)|a\rangle & \langle a|H_{NN}(k)|b\rangle \\ \langle b|H_{NN}(k)|a\rangle & \langle b|H_{NN}(k)|b\rangle \end{pmatrix} = -2.7 \begin{pmatrix} 0 & e^{-ik\cdot d_1} + e^{-ik\cdot d_2} + e^{-ik\cdot d_3} \\ e^{ik\cdot d_1} + e^{ik\cdot d_2} + e^{ik\cdot d_3} & 0 \end{pmatrix}$$
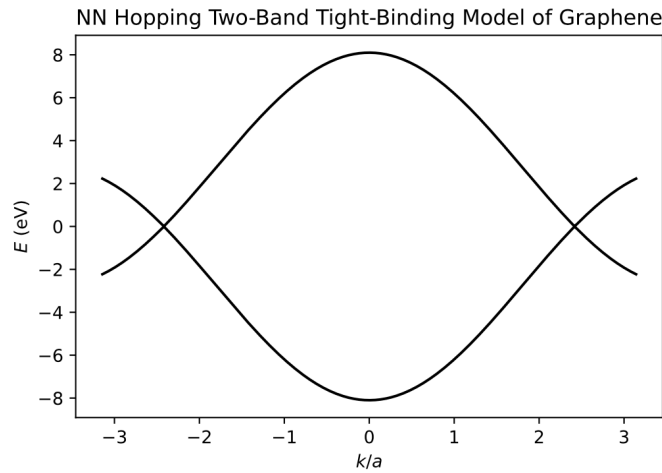
We can then plot the band structre. See Fig 2.

Fig 2. Energy bands on nearest neighbor hopping graphene model along the $\Gamma - K$ line.

Polypartite Lattice: Twisted Bilayer Graphene

As before, for N sites in the unit cell there is a corresponding N-band model that is given by a Hamiltonian that is $N \times N$ and is given by the sum over hops between sites in the unit cell

$$H = \sum_{\text{atoms in unit cell: } l,m} H_{lm}$$

The term between sites l and m is

$$H_{lm} = - \sum_{\text{unit cells: } i,j} t(r_j - r_i)(l_i^\dagger m_j + m_j^\dagger l_i)$$

$$\approx - \sum_{\text{unit cells: } i} \sum_{\alpha,\beta=-1,0,1} t((r_i + d_{lm} + \alpha a_1^M + \beta a_2^M) - r_i)(l_i^\dagger m_{r_i+d_{lm}+\alpha a_1^M+\beta a_2^M} + m_{r_i+d_{lm}+\alpha a_1^M+\beta a_2^M}^\dagger l_i)$$

where we approximate the sum as going over nearest neighboring unit cells (see Fig 3). If desired, we could sum over more unit cells. d_{lm} is the distance between sites l and m in the unit cell, and l_i is the annihilation operator of an electron on an l orbital at position r_i.
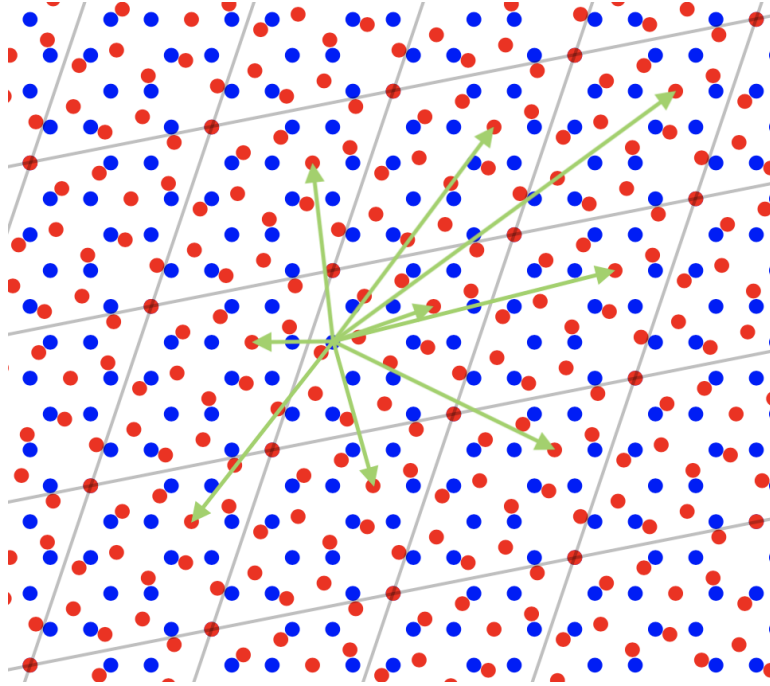

Fig 3. To sum over the relevant range it is necessary to sum over neighboring unit cells.

Substituting the momentum-space representation and evaluating the resulting delta function we obtain

$$H_{lm} \approx - \sum_{k} \sum_{\alpha,\beta=-1,0,1} t(d_{lm} + \alpha a_1^M + \beta a_2^M)e^{-ik\cdot(d_{lm}+\alpha a_1^m+\beta a_2^M)}l_k^\dagger m_k + h.c.$$

Now, if we have an expression for $t(r)$ and the atomic positions then we can calculate the band structure. Let's do this. In the Appendix, the atomic positions are listed for p_z orbitals in large-twist angle commensurate TBG, and we use the model in Nano Lett. **10**, 804 (2010):

$$t(\vec{r}) = \begin{cases} V_{pp\pi}^0 e^{-(|\vec{r}|-a_0)/\delta_0} \left(1 - \left(\frac{\vec{r}\cdot e_z}{|\vec{r}|}\right)^2\right) + V_{pp\sigma}^0 e^{-(|\vec{r}|-d_0)/\delta_0} \left(\frac{\vec{r}\cdot e_z}{|\vec{r}|}\right)^2, & |\vec{r}| \leq 4a_0 \\ 0, & |\vec{r}| > 4a_0 \end{cases}$$

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Where the parameters are given by

$$e_z = (0, 0, 1)$$
$$a_0 = a/\sqrt{3} = 0.142 \text{ nm}$$
$$d_0 = 0.335 \text{ nm}$$
$$\delta_0 = 0.184a = 0.0453 \text{ nm}$$
$$V_{pp\pi}(|\vec{r}|) = 2.7 \text{ eV}$$
$$V_{pp\sigma}(|\vec{r}|) = -0.48 \text{ eV}$$

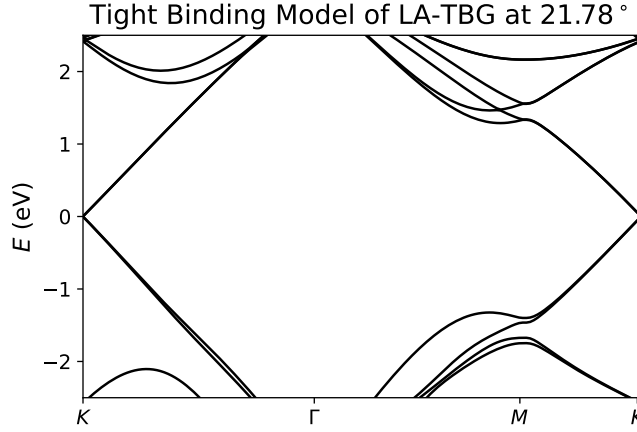We can now use this to plot the band structure of twisted bilayer graphene; see Fig 4.

FIG 4. Band structure of large twist angle commensurate TBG using 28-band tight-binding model; note that K and M are rotated by $\theta/2$ relative to monolayer graphene. Compare to Figure 9(c) of Phys. Rev. B **87**, 205404 (2013)

Appendix: Atomic Positions in $\sqrt{7} \times \sqrt{7}$ Commensurate TBG

Here are the (unrelaxed) atomic positions for a twist angle of 21.78°, corresponding to a $\sqrt{7} \times \sqrt{7}$ commensurate unit cell (distances are in nm):

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[0.        , 0.        , 0.        ],    [0.21304225, 0.123     , 0.        ],
[0.21304225, 0.369     , 0.        ],    [0.4260845 , 0.246     , 0.        ],
[0.4260845 , 0.492     , 0.        ],    [0.63912675, 0.369     , 0.        ],
[0.63912675, 0.615     , 0.        ],    [0.07101408, 0.123     , 0.        ],
[0.28405633, 0.246     , 0.        ],    [0.49709858, 0.123     , 0.        ],
[0.28405633, 0.492     , 0.        ],    [0.49709858, 0.369     , 0.        ],
[0.49709858, 0.615     , 0.        ],    [0.71014083, 0.492     , 0.        ],
[0.        , 0.        , 0.335     ],    [0.15217304, 0.19328571, 0.335     ],
[0.39564989, 0.15814286, 0.335     ],    [0.30434607, 0.38657143, 0.335     ],
[0.54782293, 0.35142857, 0.335     ],    [0.45651911, 0.57985714, 0.335     ],
[0.69999596, 0.54471429, 0.335     ],    [0.26376659, 0.10542857, 0.335     ],
[0.17246277, 0.33385714, 0.335     ],    [0.41593963, 0.29871429, 0.335     ],
[0.65941649, 0.26357143, 0.335     ],    [0.32463581, 0.52714286, 0.335     ],
[0.56811266, 0.492     , 0.335     ],    [0.7202857 , 0.68528571, 0.335     ]
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