Twisted bilayer NbSe$_2$

Semester 1 presentation

Conan Birkett

University of Bath department of Physics

December 13, 2021

Outline

- In 2018 Cao Y. et al realised unconventional superconductivity in 'magic-angle' twisted bilayer graphene[1], previously predicted in 2011 by R. Bistritzer and A. MacDonald[2].
- This has prompted a wave of research in the newly dubbed field of 'Twistronics', the study of the electronic properties of twisted Van der Waals heterostructures[3][4].
- Our project examines a Van der Waals heterostructure of two 2H-NbSe$_2$ layers. Our objective is to determine if a magic angle exists in this material, and if so to offer an interpretation of the resulting electronic band structure.

- For our model of NbSe$_2$ we employ a multi-orbital tight binding model from R. Habara and K. Wakabayashi [5].

- It uses the d_{z^2}, d_{xy} and $d_{x^2-y^2}$ orbitals of Nb atoms up to third nearest neighbor sites with spin up and down as a basis set to describe the electronic states of the NbSe$_2$ monolayer near the Fermi level.

- The eigenvalue equation for the TBM is

$$\hat{H}(\boldsymbol{k})\,|u_{nk}\rangle = E_{nk}\,|u_{nk}\rangle \tag{1}$$

where $k = (k_x, k_y)$ is the wave-number vector, E_{nk} is the eigenvalue and $n = 1, 2, \cdots, 6$ is the band index.

- The eigenvector is defined as

$$|u_{nk}\rangle =$$

$$\left(c_{nk,d_{z^2},\uparrow}, c_{nk,d_{xy},\uparrow}, c_{nk,d_{x^2-y^2},\uparrow}, c_{nk,d_{z^2},\downarrow}, c_{nk,d_{xy},\downarrow}, c_{nk,d_{x^2-y^2},\downarrow} \right)^T \tag{2}$$

where $(\cdots)^T$ indicates the transpose of the vector and $c_{nk\tau s}$ is the amplitude at atomic orbital τ with spin s for the nth energy band at k.

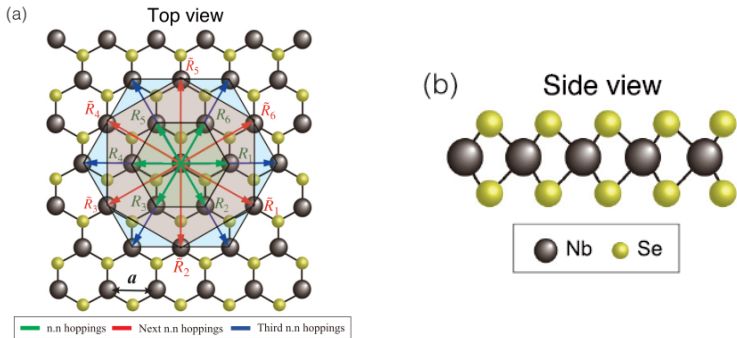# TBM Nearest Neighbors (NN)
Modelling NbSe$_2$

Figure: FIG. 1. (a) and (b) from Habara et al[5] Displaying a top-down view and side view of the structure of monolayer NbSe$_2$ consisting of Nb (black) and Se (yellow) atoms. The first, second and third nearest neighbor sites are shown in green, red and blue respectively. a is the primitive lattice constant.

TBM Hamiltonian
Modelling NbSe₂

▶ The Hamiltonian $\hat{H}(\mathbf{k})$ with spin orbit coupling can be written as

$$\hat{H}(\mathbf{k}) = \hat{\sigma}_0 \otimes \hat{H}_{\mathrm{TNN}}(\mathbf{k}) + \hat{\sigma}_z \otimes \frac{1}{2}\lambda_{\mathrm{SOC}}\hat{L}_z \tag{3}$$

▶ Which is comprised of the tight binding nearest neighbor (NN) Hamiltonian

$$\hat{H}_{\mathrm{TNN}}(\mathbf{k}) = \begin{pmatrix} V_0 & V_1 & V_2 \\ V_1^* & V_{11} & V_{12} \\ V_2^* & V_{12}^* & V_{22} \end{pmatrix} \tag{4}$$

▶ And a spin orbit coupling term

$$\hat{L}_z = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -2i \\ 0 & 2i & 0 \end{pmatrix} \tag{5}$$

▶ $\hat{\sigma}_0$ and $\hat{\sigma}_z$ are pauli matrices and $\lambda_{\mathrm{SOC}} = 0.0784$ eV is the Ising-type spin orbit coupling parameter. $V_0 \cdots V_{22}$ are functions of k [5].

1st Brillouin zone (BZ)

Modelling NbSe$_2$

- ▶ The result is a 6 by 6 block-diagonal Hamiltonian that is a function of wavevector k.
- ▶ From our eigenvalue equation (1) we can then determine the energy (eigenvalues) of the 6 electronic (eigen-)states at a given point in k-space.
- ▶ Specifically, we can take k from slices in the first brillouin zone along the standard critical points, and even a surface over k space.
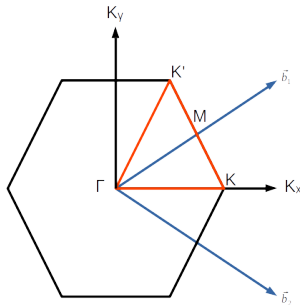

Figure: The first brillouin zone of the NbSe$_2$ lattice. Γ, K, K' and M are the standard critical points. $\vec{b_1}, \vec{b_2}$ are the reciprocal lattice vectors, shown in red is the slice $\Gamma \to K \to M \to K' \to \Gamma$.

Electronic band structure

Modelling $NbSe_2$

- ▶ As a consequence of Bloch's theorem, the eigenstates and their eigenvalues seen in the 1st brilloin zone repeat throughout the bulk lattice.
- ▶ Additionally due to reflectional symmetry the region that fully describes the electronic states can be reduced to the equilateral triangle bounded by $\Gamma \rightarrow K \rightarrow M \rightarrow K' \rightarrow \Gamma$.
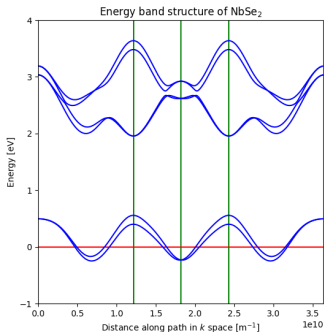

Figure: Energy band structure taken across slice $\Gamma \rightarrow K \rightarrow M \rightarrow K' \rightarrow \Gamma$. Critical points are shown in green and the Fermi level (red) is set to 0 eV.
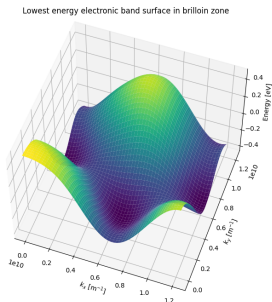

Figure: A plot of the surface of the lowest energy state in the same region. The same peaks can be seen at critical points $\Gamma, K,$ and K'.

Habara FIG 1. (d)
Modelling NbSe$_2$

- ▶ If we instead take the same slice but insead from $M \rightarrow M$ we can retrieve FIG. 1. (d) from Habara et al[5].

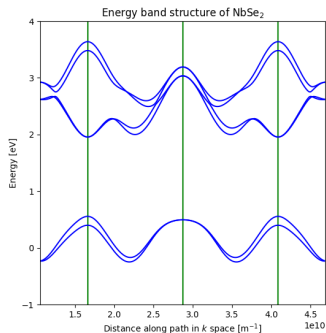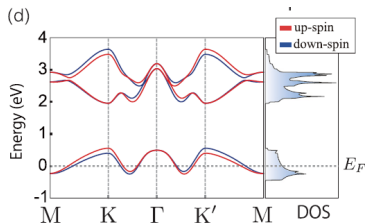- ▶ In the rest of our analysis we use $\Gamma \rightarrow \Gamma$ as Γ is the axis of rotation.

Figure: Habara et al[5] FIG. 1. (d), Energy band structure and DOS taken across slice $M \rightarrow K' \rightarrow \Gamma \rightarrow K \rightarrow M$. Note they have taken K and K' to have opposite meanings. Our recreation is shown on the right for comparison.

Rotating vectors
Adding a twist

- In order to construct a twisted bilayer, we must first figure out how to describe a twisted monolayer.

- A simple rotation in primitive lattice vectors $a_1, a_2 -> a_1', a_2'$ corresponds to the same rotation in reciprocal lattice vectors $b_1, b_2 -> b_1', b_2'$.

- We can consider this a rotation of the whole coordinate system in k space to coordinates in (k_x', k_y').

- The result of this is that in order to describe a vector $\vec{k'}$ in rotated k space (such as *Gamma*, K etc) in terms of our unrotated coordinate system (k_x, k_y) it has to be rotated *inversely* to the rotation of the layer.
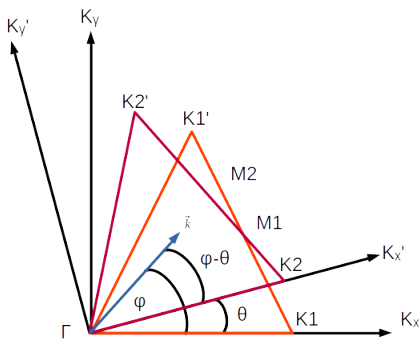
Twisted triangles
Adding a twist

Figure: The effect of rotating the layer on the path $\Gamma \rightarrow \Gamma$ and the correction that must be made to $\vec{k}$. Because of the rotation of (k_x, k_y) to (k'_x, k'_y) by θ, in order for for vector $\vec{k}$ in k' space to be represented in the unrotated k space it must be rotated clockwise by θ.
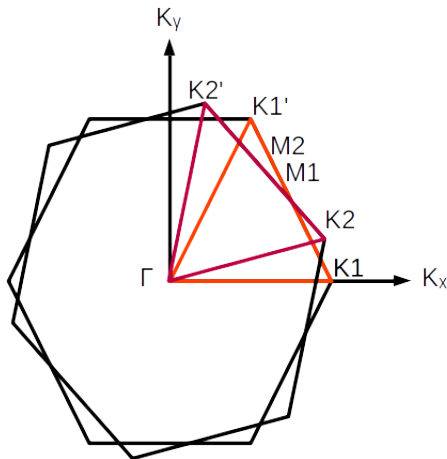
Two triangle path
Adding a twist

Figure: It is convenient to construct a path consisting of an unrotated path $\Gamma \rightarrow K_1 \rightarrow M_1 \rightarrow K_1' \rightarrow \Gamma$ followed by a rotated path (here at $15°$) $\Gamma \rightarrow K_2 \rightarrow M_2 \rightarrow K_2' \rightarrow \Gamma$. This allows us to consider the eigenvalues of a lattice from two perspectives consecutively. We will call this path $\Gamma \rightarrow \Gamma \rightarrow \Gamma$.
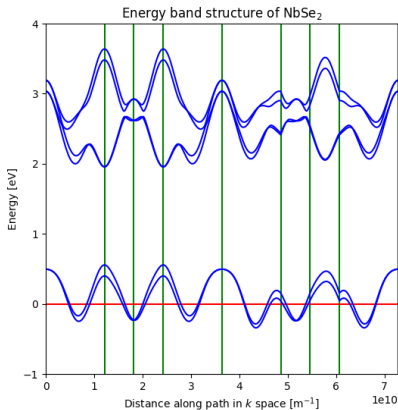
Twisted eigenvalues
Adding a twist

Figure: Our original plot of eigenvalues of an 'unrotated' layer on the new path Γ → Γ → Γ. There is clearly a substantial effect on the electronic band structure across the rotated portion of the path.

Twisted surface
Adding a twist

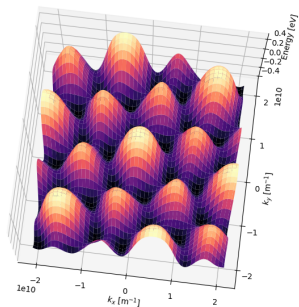Lowest energy electronic band surface in brilloin zone

Figure: A surface of the lowest energy state plotted against unrotated k_x, k_y axes. A twist of $15°$ is applied to the surface counter clockwise. The plot has been extended to an area encompassing the first brillouin zone to show this more clearly.
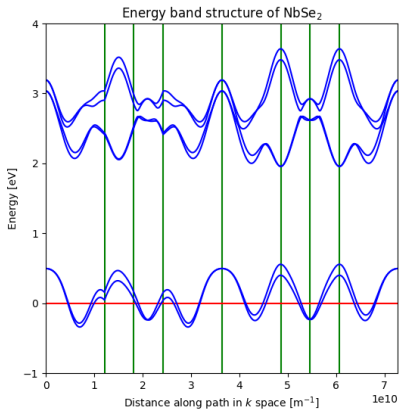
Un-twisted eigenvalues

Adding a twist

Figure: The eigenvalues of our $15°$ twisted lattice on the path $\Gamma \to \Gamma \to \Gamma$. It is flipped in the x axis when compared with the eigenvalues of the unrotated lattice.

▶ Now that we can describe a rotated layer of $NbSe_2$ we can now model two layers at once, with one rotated. A simple model assumes no interaction between states in different layers.

▶ We construct a Hamiltonian for the whole system by combining the Hamiltonians of each layer into a 12 by 12 block diagonal matrix

$$\hat{H}(\boldsymbol{k}) = \begin{pmatrix} \hat{H}_1(\boldsymbol{k}) & 0 \\ 0 & \hat{H}_2(\boldsymbol{k}') \end{pmatrix} \quad (6)$$

Where $\hat{H}_1(\boldsymbol{k})$ and $\hat{H}_2(\boldsymbol{k}')$ are the Hamiltonians of the unrotated and rotated layers respectively.

▶ Note that $\hat{H}_2(\boldsymbol{k}')$ is a function of k' which means that an implementation of this matrix of this function must rotate the k input (as described previously) before it is input into $\hat{H}(\boldsymbol{k})$.

Heterostructure eigenvalues
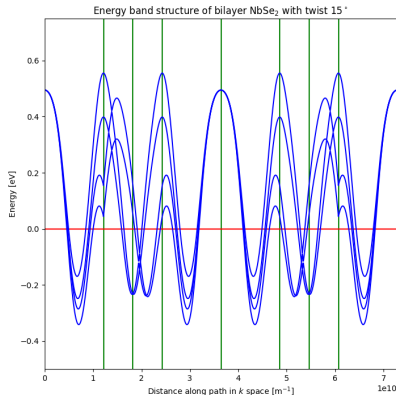
Constructing a heterostructure

Figure: The four lowest energy eigenvalues of the heterostructure with a twist of $15°$ applied to the second layer. This plot is taken in the path $\Gamma \to \Gamma \to \Gamma$. For now, we are only looking at these eigenstates because they have energy near the Fermi level, and the corresponding Fermi-surface will describe the electronic properties of the material. The other eigenvalues are still computed.

- ▶ Animation of eigenvalues at varying angles (PDF).
- ▶ Notice that the intersection of energy bands varies with the angle of rotation.
- ▶ This will become very important in our work next semester when we apply perturbation theory to a model of coupled layers.

Coupling layers - the d_{z^2} orbital
What's next

- Currently, we are assuming no interaction between the electronic states on different layers.

- Looking at the d orbitals that we're modelling with the TBM, the d_{z^2} orbital actually projects out (substantially) in the z axis. It is therefore a good candidate for describing at inter-layer coupling effects.
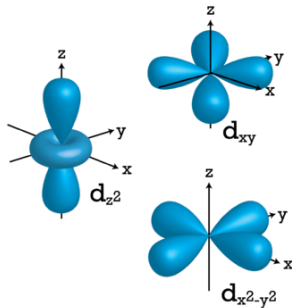

Figure: Depictions of the d orbitals from lumenlearning.com[6], note the projection of the d_{z^2} orbital into the z axis.

▶ We will make several assumptions regarding the interaction of orbitals between layers. Firstly, that the influence of the d_{xy} and $d_{x^2-y^2}$ orbitals is restricted to their corresponding xy planes i.e. their own layers.

▶ This leaves the d_{z^2} orbital. There is no quantum process that will allow for the transition into states of different spin.

▶ So we are left with four processes to consider

1. $d_{z^2,\downarrow,1} \to d_{z^2,\downarrow,2}$
2. $d_{z^2,\downarrow,2} \to d_{z^2,\downarrow,1}$
3. $d_{z^2,\uparrow,1} \to d_{z^2,\uparrow,2}$
4. $d_{z^2,\uparrow,2} \to d_{z^2,\uparrow,1}$

where $d_{z^2,s,i}$ is the d_{z^2} orbital with spin s on layer i (we take $i = 2$ as the rotated layer).

▶ All of these processes should have the same transition energy, the next step is to try and model them.

▶ Previously our 12 by 12 heterostructure Hamiltonian was block diagonal, where each 6 by 6 block corresponded to one of the layers.

$$\hat{H}(\boldsymbol{k}) = \begin{pmatrix} \hat{H}_1(\boldsymbol{k}) & 0 \\ 0 & \hat{H}_2(\boldsymbol{k}') \end{pmatrix} \tag{7}$$

▶ To account for the coupling we will add terms in the 0 blocks at positions that correspond to the four d_{z^2} orbital transitions that we are interested in.

▶ This will give us the modified Hamiltonian

$$\hat{H}(\boldsymbol{k}) = \begin{pmatrix} \hat{H}_1(\boldsymbol{k}) & T \\ T^{\dagger} & \hat{H}_2(\boldsymbol{k}') \end{pmatrix} \tag{8}$$

Where T is a 6 by 6 matrix containing our coupling term at (col, row) = (1,1) and (4,4); see the eigenvector. The nature of this coupling term $t(k, \theta)$ we are yet to determine.

▶ We construct a 'toy model' where we will set the coupling term in T to be a constant. For now we have chosen 0.3 eV, just to see what the effect could look like on our band structure.
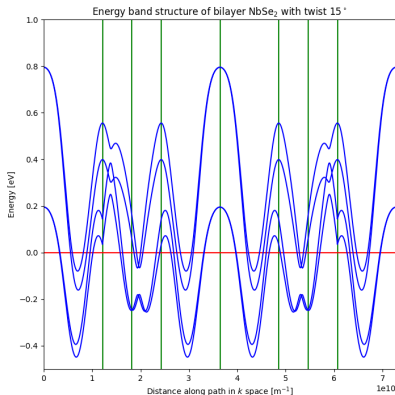

Figure: The four lowest energy eigenvalues of the heterostructure with a twist of $15°$ applied to the second layer. The 'toy model' coupling term is applied, there is a resulting effect on the electron bands that were previously unaffected by rotation.

- ▶ Our coupling terms in our more complex heterostructure Hamiltonian will naturally result in a splitting of energy levels at degenerate eigenvalues as a consequence of perturbation theory.
- ▶ The previous graph manages to naively predict the splitting of energy levels at Γ.
- ▶ A consequence of this is that electron bands become "flatter" that is, they have a lower bandwidth.
- ▶ This results in a higher density of states at certain energy levels, with lower transition energies between them.
- ▶ Things get especially interesting when this flattening happens around the Fermi level, and will be the focus of our work going into the next semester.

References

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