Wigner Crystal State in Twisted Bilayer Graphene

An ongoing research project under the supervision of Dr. Brian Skinner

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draft, updated randomly

General scenario

Consider a infinitely large two-dimensional graphene sheet. When we say "two-dimensional", we means the thickness is negligible and the sheet can be treated as a two-dimensional manifold; in our case, it is $\mathbb{R}^2$ or $\mathbb{C}$. The carbon atoms and the carbon-carbon bonds in this graphene form a honeycomb lattice. Next, we pour a ton of electrons onto the graphene sheet. Due the the Coulomb's force, the electrons repulse each other and form a certain arrangement. The arrangement with the lowest total energy is called the **Wigner crystal state**. It can be proved that the Wigner crystal state of electrons in two-dimensional plane is the equilateral triangle lattice. We want to find the condition under which the Wigner crystal state of electrons occurs on the two-dimensional graphene. In other words, the state we are interested in comes with two lattice on a same two-dimensional plane: the honeycomb lattice of carbon atoms and the equilateral triangle lattice of electrons. See Fig.1.

Geometry of the electron lattice

The length $R = |\mathbf{R}_{1,0}|$ be the lattice constant of the equilateral triangle lattice. The lattice can be described by the Bravais lattice with the primitive vectors

$$\begin{aligned}\mathbf{a}_1 &= \mathbf{R}_{1,0} = R\mathbf{e}_1 = R(1,0),\\ \mathbf{a}_2 &= \mathbf{R}_{0,1} = \frac{R}{2}\mathbf{e}_1 + \frac{\sqrt{3}R}{2}\mathbf{e}_2 = R\left(\frac{1}{2}, \frac{\sqrt{3}}{2}\right);\end{aligned}$$

so, the position of each electron can be labeled as

$$\mathbf{R}_{i,j} = i\,\mathbf{a}_1 + j\,\mathbf{a}_2, \quad i, j \in \mathbb{Z} \tag{1}$$

One can easily find the corresponding reciprocal lattice

$$\mathbf{K}_{i,j} = i\,\mathbf{b}_1 + j\,\mathbf{b}_2, \quad i, j \in \mathbb{Z} \tag{2}$$

with the primitive vectors

$$\mathbf{b}_1 = \frac{2\pi}{R}\left(1, -\frac{1}{\sqrt{3}}\right), \qquad \mathbf{b}_2 = \frac{2\pi}{R}\left(0, \frac{2}{\sqrt{3}}\right). \tag{3}$$

The averaged energy density of the unit cell of the electron lattice

Since we assumed the plane is an infinitely large square which means its side length $L \to \infty$, the electrostatic energy of the electron at $\mathbf{R}_{i,j}$ is approximately equal to that of the electron at $\mathbf{R}_{0,0} = \mathbf{0}$. So, our task is to first find the electrostatic potential at $\mathbf{R}_{0,0}$.

Let $V(\mathbf{R}_{i,j})$ be the electrostatic potential at $\mathbf{0}$ due to the electron at $\mathbf{R}_{i,j}$, then the total (net) electrostatic potential at $\mathbf{0}$ is just the scalar sum of all $V(\mathbf{R}_{i,j})$ except $V(\mathbf{V}_{0,0})$; namely,

$$V = \sum_{\substack{i,j\in\mathbb{Z} \\ (i,j)\neq(0,0)}} V(\mathbf{R}_{i,j}) \tag{4}$$

The direct calculation will end up with a diverge series since the plane is assumed to be infinitely large and the potential is inverse proportional to the distance.

Our approach is to utilize the Fourier transform

$$\tilde{V}(\mathbf{k}) = \int_{-\infty}^{\infty} V(\mathbf{r}) e^{-i\mathbf{k}\cdot\mathbf{r}} \mathrm{d}^2\mathbf{r}, \quad V(\mathbf{r}) = \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \tilde{V}(\mathbf{k}) e^{i\mathbf{r}\cdot\mathbf{k}} \mathrm{d}^2\mathbf{k}.$$

where $\mathbf{r}$ is a vector in $\mathbf{R}_{i,j}$ space and $\mathbf{k}$ is a vector in the corresponding reciprocal space $\mathbf{K}_{i,j}$. We are interested in the potential at $\mathbf{r} = \mathbf{R}_{i,j}$, so

$$V(\mathbf{R}_{i,j}) = \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \tilde{V}(\mathbf{k}) e^{i\mathbf{R}_{i,j}\cdot\mathbf{k}} \mathrm{d}^2\mathbf{k}. \tag{5}$$

So,

$$V = \sum_{\substack{i,j\in\mathbb{Z} \\ (i,j)\neq(0,0)}} \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \tilde{V}(\mathbf{k}) e^{i\mathbf{R}_{i,j}\cdot\mathbf{k}} \mathrm{d}^2\mathbf{k} = \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \tilde{V}(\mathbf{k}) \sum_{\substack{i,j\in\mathbb{Z} \\ (i,j)\neq(0,0)}} e^{i\mathbf{R}_{i,j}\cdot\mathbf{k}} \mathrm{d}^2\mathbf{k} \tag{6}$$

The double sum inside the integrand is

$$\begin{aligned}
\sum_{\substack{i,j\in\mathbb{Z} \\ (i,j)\neq(0,0)}} e^{i\mathbf{R}_{i,j}\cdot\mathbf{k}} &= -e^{i\mathbf{R}_{0,0}\cdot\mathbf{k}} + \sum_{i,j\in\mathbb{Z}} e^{i\mathbf{R}_{i,j}\cdot\mathbf{k}} \\
&= -1 + \sum_{i,j\in\mathbb{Z}} e^{i\mathbf{R}_{i,j}\cdot\mathbf{k}} \\
&= -1 + \sum_{i,j\in\mathbb{Z}} e^{i\mathbf{R}_{i,j}\cdot\mathbf{k}} e^{-i\mathbf{R}_{i,j}\cdot\mathbf{K}_{l,m}} \qquad \text{since } e^{i\mathbf{R}_{i,j}\cdot\mathbf{K}_{l,m}} = 1 \text{ for every } i,j,l,m \in \mathbb{Z} \\
&= -1 + \frac{1}{N} \sum_{l,m\in\mathbb{Z}} \left(\sum_{i,j\in\mathbb{Z}} e^{i\mathbf{R}_{i,j}\cdot(\mathbf{k}-\mathbf{K}_{l,m})} \right),
\end{aligned}$$

where N is the number of $\mathbf{K}_{l,m}$s multiplied. Since the plane is infinitely large, roughly every $\mathbf{K}_{l,m}$ points to an electron although they actually do not, then N can be roughly calculated from the surface charge density

$$n = \frac{N(-e)}{L^2}.$$

In addition, the Fourier series of Dirac delta function δ gives

$$\sum_{i,j\in\mathbb{Z}} e^{i\mathbf{R}_{i,j}\cdot(\mathbf{k}-\mathbf{K}_{l,m})} = \frac{4(2\pi)^2}{\sqrt{3}} \frac{1}{R^2} \delta(\mathbf{k} - \mathbf{K}_{l,m}) \quad \text{in the weak sense on a small neighborhood of } \mathbf{K}_{l,m} \tag{7}$$

So,

$$\sum_{\substack{i,j\in\mathbb{Z}\\(i,j)\neq(0,0)}} e^{i\mathbf{R}_{i,j}\cdot\mathbf{k}} = -1 + \frac{-e}{nL^2}\frac{4(2\pi)^2}{\sqrt{3}}\frac{1}{R^2}\sum_{l,m\in\mathbb{Z}}\delta(\mathbf{k}-\mathbf{K}_{l,m}) \tag{8}$$

Derivation of Eq. (7) *and the unit-consistency* First consider the following two identities

$$\int_{-\infty}^{\infty} e^{ikx}\mathrm{d}x = 2\pi\delta(k)^{[1]}$$
$$\sum_{x\in\mathbb{Z}} e^{ixk} = 2\pi\delta(k) \quad \text{in the weak sense on } k\in(-\pi,\pi)^{[2]}$$

The first one is unit-consistent since both sides has unit of x, but the second one is not since the left hand side is dimensionless yet the right hand side is of unit x. Both of them are mathematically correct, but in physics we always want the unit to be consist. So, we re-write the second one as

$$\sum_{x\in\mathbb{Z}} e^{ixk}\mathrm{d}x = 2\pi\delta(k) \quad \text{in the weak sense on } k\in(-\pi,\pi)$$

where $\mathrm{d}x$ is a unit, or the smallest allowable, step in $\mathbb{Z}$; of course, it is $+1$. One can also treat it as an approximation of the first equation or the discrete inverse Fourier transform of $\{...,1,1,...\}$ of which the result is discrete values of Dirichlet kernel and becomes the Dirac delta function when treat k as a continuous variable.

In our case, we have the unit-consistent formula

$$\sum_{i,j\in\mathbb{Z}} e^{i\mathbf{R}_{i,j}\cdot(\mathbf{k}-\mathbf{K}_{l,m})}\mathrm{d}^2\mathbf{R}_{i,j} = (2\pi)^2\delta(\mathbf{k}-\mathbf{K}_{l,m})$$

where $\mathrm{d}^2\mathbf{R}_{i,j}$ is a unit, or the smallest allowable, step in our lattice plane. Since $\sum_{i,j\in\mathbb{Z}} = \sum_{i\in\mathbb{Z}}\sum_{j\in\mathbb{Z}}$, the one unit of change has length $|\mathbf{R}_{i,j+1}-\mathbf{R}_{i,j}| = R$, the lattice constant. Thus, $\mathrm{d}^2\mathbf{R}_{i,j} = \mathrm{d}x\mathrm{d}y = \left(\frac{1}{2}R\right)\left(\frac{\sqrt{3}}{2}R\right) = \frac{\sqrt{3}}{4}R^2$; of course any allowable steps are the integer-times of it. Plug it back into the last formula, we can then get Eq. (7).

After the calculation of the double sum, we can now plug it into (6),

$$\begin{aligned} V &= \frac{1}{(2\pi)^2}\int_{-\infty}^{\infty}\tilde{V}(\mathbf{k})\left(-1+\frac{-e}{nL^2}\frac{4(2\pi)^2}{\sqrt{3}}\frac{1}{R^2}\sum_{l,m\in\mathbb{Z}}\delta(\mathbf{k}-\mathbf{K}_{l,m})\right)\mathrm{d}^2\mathbf{k} \\ &= -\frac{1}{(2\pi)^2}\int_{-\infty}^{\infty}\tilde{V}(\mathbf{k})\mathrm{d}^2\mathbf{k} + \frac{-4e}{\sqrt{3}nL^2}\frac{1}{R^2}\int_{-\infty}^{\infty}\tilde{V}(\mathbf{k})\sum_{l,m}\delta(\mathbf{k}-\mathbf{K}_{l,m})\mathrm{d}^2\mathbf{k} \\ &= -\frac{1}{(2\pi)^2}\int_{-\infty}^{\infty}\tilde{V}(\mathbf{k})\mathrm{d}^2\mathbf{k} + \frac{-4e}{\sqrt{3}nL^2}\frac{1}{R^2}\sum_{l,m}\tilde{V}(\mathbf{K}_{l,m}) \end{aligned} \tag{9}$$

Check the unit again: both terms are of unit $\mathrm{unit}(V)\cdot\mathrm{unit}(k)^2 = \mathrm{unit}(V)\cdot\mathrm{unit}(r)^{-2}$; in second term $\frac{-4e}{\sqrt{3}nL^2}$ is dimensionless.

This is the simplest form of V we can get. To keep going, we need to invoke the actual expression of the electrostatic potential V. In Bravais lattice space, let $r = |\mathbf{r}|$, then it is

$$V(r) = -\frac{1}{4\pi\varepsilon_0}\frac{e}{r} \quad \text{with } \lim_{r\to\infty} V(r) = 0; \tag{10}$$

[1] W. Strauss, *Partial Differential Equations, An Introduction*, 2nd ed (John Wiley & Sons, 2008), p.345.
[2] *ibid.*, p.334.

let $k = |\mathbf{k}|$, the Fourier transform in polar coordinate [3] then gives its form in the reciprocal lattice space,

$$\tilde{V}(k) = -\frac{1}{4\pi\varepsilon_0}\frac{2\pi e}{k} \quad \text{with } \lim_{k\to\infty}\tilde{V}(k) = 0; \tag{11}$$

We see $\tilde{V}$ is still inverse proportional to the distance in the reciprocal lattice space. This means we cannot avoid the the divergence of $\int_0^\infty \frac{1}{k}k\,\mathrm{d}k = \int_0^\infty \mathrm{d}k$; we need to find a proper way to deal with it.

The first term in (9) looks really like the inverse Fourier transform of $\tilde{V}(\mathbf{k})$, but we will not add the kernel $e^{i\mathbf{r}\cdot\mathbf{k}}$ and take the limit $\mathbf{r} \to \mathbf{0}$ to actually transform it since it will come back to where we leave. Instead, we add a Gaussian phase term centered at zero with variance $\sigma^2 = \frac{1}{w^2}$, namely $e^{-\frac{1}{2}w^2k^2}$, and take the limit $w \to 0$;

$$V = \lim_{w\to 0}\left(-\frac{1}{(2\pi)^2}\int_{-\infty}^{\infty}\tilde{V}(\mathbf{k})e^{-\frac{1}{2}w^2k^2}\mathrm{d}^2\mathbf{k} + \frac{-4e}{\sqrt{3}nL^2}\frac{1}{R^2}\sum_{l,m}\tilde{V}(\mathbf{K}_{l,m})e^{-\frac{1}{2}w^2K_{l,m}^2}\right) \tag{12}$$

where $K_{l,m} = |\mathbf{K}_{l,m}|$.

The Gaussian phase term is added to make the integral convergent; the infinite sum of discrete variables can be approximated by the corresponding integral which is also made to be convergent.

Plug (11) into it, the first term becomes

$$\frac{1}{(2\pi)^2}\frac{2\pi e}{4\pi\varepsilon_0}\int_0^{2\pi}\int_0^{\infty}\frac{1}{k}e^{-\frac{1}{2}w^2k^2}k\,\mathrm{d}k\,\mathrm{d}\theta = \frac{1}{(2\pi)^2}\frac{2\pi e}{4\pi\varepsilon_0}2\pi\int_0^{\infty}e^{-\frac{1}{2}w^2k^2}\mathrm{d}k = \sqrt{\frac{\pi}{2}}\frac{e}{4\pi\varepsilon_0}\frac{1}{w}, \tag{13}$$

and the second term can be approached roughly through

$$\begin{aligned}
\frac{-4e}{\sqrt{3}nL^2}\frac{1}{R^2}\sum_{l,m}\tilde{V}(\mathbf{K}_{l,m})e^{-\frac{1}{2}w^2K_{l,m}^2} &\approx \frac{-4e}{\sqrt{3}nL^2}\frac{1}{R^2}\frac{1}{\mathrm{d}^2\mathbf{K}_{l,m}}\int_{-\infty}^{\infty}\tilde{V}(\mathbf{K}_{l,m})e^{-\frac{1}{2}w^2K_{l,m}^2}\mathrm{d}^2\mathbf{K}_{l,m} \\
&= \frac{-4e}{\sqrt{3}nL^2}\frac{1}{R^2}\frac{1}{\mathrm{d}^2\mathbf{K}_{l,m}}\int_{-\infty}^{\infty}\tilde{V}(\mathbf{K}_{l,m})e^{-\frac{1}{2}w^2K_{l,m}^2}\mathrm{d}^2\mathbf{K}_{l,m} \\
&= \frac{-4e}{\sqrt{3}nL^2}\frac{1}{R^2}\frac{1}{\mathrm{d}^2\mathbf{K}_{l,m}}\sqrt{\frac{\pi}{2}}\frac{1}{w} \\
&=
\end{aligned}$$

Thus,

$$V = \tag{14}$$

Immediately, we can find the *average* energy of the electron at $\mathbf{0}$

$$E_{\text{avg}} = \frac{1}{2}(-e)V = \tag{15}$$

[3] N. Baddour, Adv. Imaging Electron Phys. **165**, 1 (2011).