

Wigner Crystal State in Twisted Bilayer Graphene

An ongoing research project in solid-state physics

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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

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

One can easily find the corresponding reciprocal lattice

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

with the primitive vectors

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

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 $L \rightarrow \infty$, 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 $V(\mathbf{R}_{0,0})$; namely,

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

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

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

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

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

So,

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

The double sum inside the integrand is

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

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

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

So,

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

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

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

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

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

where dx 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 $\{\dots, 1, 1, \dots\}$ 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

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

where $d^2\mathbf{R}_{i,j}$ 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, $d^2\mathbf{R}_{i,j} = dx dy = \left(\frac{1}{2}R\right) \left(\frac{\sqrt{3}}{2}R\right) = \frac{\sqrt{3}}{4}R^2$; 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),

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

Check the unit again: both terms are of unit $\text{unit}(V) \cdot \text{unit}(k)^2 = \text{unit}(V) \cdot \text{unit}(r)^{-2}$; 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

$$V(r) = -\frac{1}{4\pi\epsilon_0} \frac{e}{r} \quad \text{with } \lim_{r \rightarrow \infty} V(r) = 0; \quad (10)$$

[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,

$$\tilde{V}(k) = -\frac{1}{4\pi\epsilon_0} \frac{2\pi e}{k} \quad \text{with} \quad \lim_{k \rightarrow \infty} \tilde{V}(k) = 0; \quad (11)$$

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 $\int_0^\infty \frac{1}{k} k dk = \int_0^\infty dk$; 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 $\mathbf{r} \rightarrow \mathbf{0}$ 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 $w \rightarrow 0$;

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

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

$$\frac{1}{(2\pi)^2} \frac{2\pi e}{4\pi\epsilon_0} \int_0^{2\pi} \int_0^\infty \frac{1}{k} e^{-\frac{1}{2}w^2k^2} k dk d\theta = \frac{1}{(2\pi)^2} \frac{2\pi e}{4\pi\epsilon_0} 2\pi \int_0^\infty e^{-\frac{1}{2}w^2k^2} dk = \sqrt{\frac{\pi}{2}} \frac{e}{4\pi\epsilon_0} \frac{1}{w}, \quad (13)$$

and the second term can be approached roughly through

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

Thus,

$$V = \quad (14)$$

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

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

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