Wigner Crystal State in Twisted Bilayer Graphene

An ongoing research project in solid-state physics
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Last updated on 26 October 2020

draft, updated randomly

General scenario

In this project, we consider an infinitely large twisted bilayer graphene and a ton of electrons poured onto it. ***The aim of this project is to find the critical value of the electron density for which the Wigner crystal state melts on the twisted bilayer graphene.*** The first step is to do such job in vacuum. Note that the state we are interested in comes with three lattice on a same two-dimensional plane: two honeycomb lattices of carbon atoms with a twisted angle and the equilateral triangle lattice of electrons. See Fig.1.

General logic

Let $\mathrm{r}_{i,j}$ be the Bravais lattice of the triangle lattice. The Hamiltonian, in real position space, of the Wigner crystal state is

$$H = -\frac{\hbar^2}{2m_\mathrm{e}} \sum_{i,j\in\mathbb{Z}} \Delta_{i,j} + \sum_{i,j\in\mathbb{Z}} (-e) V(\mathbf{R}_{i,j}) \tag{1}$$

where $\Delta_{i,j}$ is Laplacian taken at the frame of the electron at $\mathbf{R}_{i,j}$. Since the Wigner crystal state is the state with lowest energy, we want to find the minimum of the energy per electron; apparently the variational principle gives us the upper bound

$$\frac{1}{N}E \leqslant \frac{1}{N}\langle H\rangle$$

where N is the number of electrons. The expected potential energy per electrons can be found exactly in classical way, and the expected kinetic energy per electron can be obtained by the virial theorem

$$\langle T\rangle = \frac{1}{2}\langle \mathbf{r}\cdot \mathrm{grad} U\rangle. \tag{2}$$

where $\mathbf{r}$ is a vector in $\mathbf{R}$ space. Further, if the oscillation around the lattice point of each electron is extremely small, then each electron is a two-dimensional harmonic oscillator. For the stationary state of a harmonic oscillator,

$$\langle T\rangle = \langle U\rangle = \frac{1}{2}E = \frac{1}{2}(n+1)\hbar\omega \tag{3}$$

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

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

so, the position of each electron can be labeled as

$$\mathbf{R}_{i,j} = i\,\mathbf{a}_1 + j\,\mathbf{a}_2, \quad i,j \in \mathbb{Z} \tag{4}$$

One can easily find the corresponding reciprocal lattice

$$\mathbf{K}_{i,j} = i\,\mathbf{b}_1 + j\,\mathbf{b}_2, \quad i,j \in \mathbb{Z} \tag{5}$$

with the primitive vectors

$$\mathbf{b}_1 = \frac{2\pi}{R}\left(1, -\frac{1}{\sqrt{3}}\right), \qquad \mathbf{b}_2 = \frac{2\pi}{R}\left(0, \frac{2}{\sqrt{3}}\right). \tag{6}$$

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

$$V = \sum_{\substack{i,j\in\mathbb{Z} \\ (i,j)\neq(0,0)}} V(\mathbf{R}_{i,j}) \tag{7}$$

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

$$V(\mathbf{R}_{i,j}) = \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \tilde{V}(\mathbf{k}) e^{i\mathbf{R}_{i,j}\cdot\mathbf{k}} \mathrm{d}^2\mathbf{k}. \tag{8}$$

So,

$$V = \sum_{\substack{i,j\in\mathbb{Z} \\ (i,j)\neq(0,0)}} \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \tilde{V}(\mathbf{k}) e^{i\mathbf{R}_{i,j}\cdot\mathbf{k}} \mathrm{d}^2\mathbf{k} = \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \tilde{V}(\mathbf{k}) \sum_{\substack{i,j\in\mathbb{Z} \\ (i,j)\neq(0,0)}} e^{i\mathbf{R}_{i,j}\cdot\mathbf{k}} \mathrm{d}^2\mathbf{k} \tag{9}$$

The double sum inside the integrand is

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

where we use the Fourier series of Dirac delta function δ in the last step. Now invoke the surface charge density

$$n = \frac{-e}{\text{area of one Wigner-Seitz cell}} = \frac{-e}{\frac{\sqrt{3}}{2}R^2} = \frac{2(-e)}{\sqrt{3}R^2};$$

we have

$$\sum_{\substack{i,j\in\mathbb{Z} \\ (i,j)\neq(0,0)}} e^{i\mathbf{R}_{i,j}\cdot\mathbf{k}} = -1 + (2\pi)^2 \frac{2n}{-e} \sum_{l,m\in\mathbb{Z}} \delta(\mathbf{k} - \mathbf{K}_{l,m}) \tag{10}$$

Derivation of Fourier series of Dirac delta function 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 $+1$. Further,

$$\sum_{x\in\mathbb{Z}} e^{ixk}\mathrm{d}x = 2\pi\delta(k-2\pi) \quad \text{in the weak sense on } k\in(\pi,3\pi)$$
$$\sum_{x\in\mathbb{Z}} e^{ixk}\mathrm{d}x = 2\pi\delta(k-4\pi) \quad \text{in the weak sense on } k\in(3\pi,5\pi)$$
$$\dots$$

To conclude, for all k,

$$\sum_{x\in\mathbb{Z}} e^{ixk}\mathrm{d}x = 2\pi\sum_{n\in\mathbb{Z}}\delta(k-2n\pi) \quad \text{where } n\in\mathbb{Z} \tag{11}$$

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

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

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 product of the integer-times of $\mathrm{d}x$ and of $\mathrm{d}y$. Plug it back into the last formula, we can then get Eq. (**??**).

After the calculation of the double sum, we can now plug it into (9),

$$\begin{aligned} V &= \frac{1}{(2\pi)^2}\int_{-\infty}^{\infty}\tilde{V}(\mathbf{k})\left(-1+(2\pi)^2\frac{2n}{-e}\sum_{l,m\in\mathbb{Z}}\delta(\mathbf{k}-\mathbf{K}_{l,m})\right)\mathrm{d}^2\mathbf{k} \\ &= -\frac{1}{(2\pi)^2}\int_{-\infty}^{\infty}\tilde{V}(\mathbf{k})\mathrm{d}^2\mathbf{k} + \frac{2n}{-e}\int_{-\infty}^{\infty}\tilde{V}(\mathbf{k})\sum_{l,m\in\mathbb{Z}}\delta(\mathbf{k}-\mathbf{K}_{l,m})\mathrm{d}^2\mathbf{k} \\ &= -\frac{1}{(2\pi)^2}\int_{-\infty}^{\infty}\tilde{V}(\mathbf{k})\mathrm{d}^2\mathbf{k} + \frac{2n}{-e}\sum_{l,m}\tilde{V}(\mathbf{K}_{l,m}) \end{aligned} \tag{12}$$

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 $\frac{2n}{-e}$ is of $\mathrm{unit}(r)^{-2}$.

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

$$V(r) = -\frac{1}{4\pi\varepsilon_0}\frac{e}{r} \quad \text{with } \lim_{r\to\infty} V(r) = 0; \tag{13}$$

[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,

$$\tilde{V}(k) = -\frac{1}{4\pi\varepsilon_0}\frac{2\pi e}{k} \quad \text{with} \lim_{k\to\infty}\tilde{V}(k) = 0; \tag{14}$$

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 (12) 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 $e^{-\frac{1}{2}w^2k^2}$, where $\mathrm{unit}(w) = \mathrm{unit}(k)^{-1}$, and take the limit $w\to 0$;

$$V = \lim_{w\to 0}\left(-\frac{1}{(2\pi)^2}\int_{-\infty}^{\infty}\tilde{V}(\mathbf{k})e^{-\frac{1}{2}w^2k^2}\mathrm{d}^2\mathbf{k} + \frac{2n}{-e}\sum_{l,m}\tilde{V}(\mathbf{K}_{l,m})e^{-\frac{1}{2}w^2K_{l,m}^2}\right) \tag{15}$$

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 (14) into it, the first term becomes

$$\frac{1}{(2\pi)^2}\frac{2\pi e}{4\pi\varepsilon_0}\int_0^{2\pi}\int_0^\infty \frac{1}{k}e^{-\frac{1}{2}w^2k^2}k\,\mathrm{d}k\mathrm{d}\theta = \frac{1}{(2\pi)^2}\frac{2\pi e}{4\pi\varepsilon_0}2\pi\int_0^\infty e^{-\frac{1}{2}w^2k^2}\mathrm{d}k = \sqrt{\frac{\pi}{2}}\frac{e}{4\pi\varepsilon_0}\frac{1}{w}, \tag{16}$$

and the second term can be approached roughly through

$$\begin{aligned}
\frac{2n}{-e}\sum_{l,m}\tilde{V}(\mathbf{K}_{l,m})e^{-\frac{1}{2}w^2K_{l,m}^2} &\approx \frac{2n}{-e}\frac{1}{\mathrm{d}^2\mathbf{K}_{l,m}}\int_{-\infty}^{\infty}\tilde{V}(\mathbf{K}_{l,m})e^{-\frac{1}{2}w^2K_{l,m}^2}\mathrm{d}^2\mathbf{K}_{l,m}\\
&= \frac{2n}{-e}\cdot\frac{\sqrt{3}R^2}{4\pi^2}\cdot\frac{-2\pi e}{4\pi\varepsilon_0}\int_0^{2\pi}\int_{-\infty}^{\infty}\frac{1}{k}e^{-\frac{1}{2}w^2k^2}k\,\mathrm{d}k\mathrm{d}\theta\\
&= \frac{2n}{-e}\cdot\frac{\sqrt{3}R^2}{4\pi^2}\cdot\frac{-2\pi e}{4\pi\varepsilon_0}2\pi\sqrt{\frac{\pi}{2}}\frac{1}{w}\\
&= \frac{2n\sqrt{3}R^2}{e}\sqrt{\frac{\pi}{2}}\frac{e}{4\pi\varepsilon_0}\frac{1}{w}\\
&= -4\sqrt{\frac{\pi}{2}}\frac{e}{4\pi\varepsilon_0}\frac{1}{w}
\end{aligned}$$

Again, $\mathrm{d}^2\mathbf{K}_{l,m}$ is the unit step, or the smallest allowable step, in the reciprocal lattice space. $\mathrm{d}^2\mathbf{K}_{l,m} = \mathrm{d}k_x\mathrm{d}k_y = \left(\frac{2\pi}{R}\right)\left(\frac{2\pi}{\sqrt{3}R}\right) = \frac{4\pi^2}{\sqrt{3}R^2}$, and of course any step takes the form $(a\,\mathrm{d}k_x)(b\,\mathrm{d}k_y)$ where $a, b\in\mathbb{Z}$.

Thus,

$$V = \lim_{w\to 0}\left(-3\sqrt{\frac{\pi}{2}}\frac{e}{4\pi\varepsilon_0}\frac{1}{w}\right) \tag{17}$$

Immediately, we can find the *total energy per electron*

$$\boxed{U_{\text{one}} = \frac{1}{2}(-e)V = \lim_{w\to 0}\frac{3}{2}\sqrt{\frac{\pi}{2}}\frac{1}{4\pi\varepsilon_0}\frac{e^2}{w}} \tag{18}$$

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