

Wigner Crystal State in Twisted Bilayer Graphene

An ongoing research project in solid-state physics

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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, or the charge carrier 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 $\mathbf{R}_{i,j}$ be the Bravais lattice of the triangle lattice, and let $\mathbf{r}_{i,j}$ be displacement from the lattice point $\mathbf{R}_{i,j}$. Due to the electrostatic force, any small displacement feels a storing force; for the extremely small displacement, each electron can thus be considered as a harmonic oscillator. The Hamiltonian, in the real position space, of the Wigner crystal state is thus the sum of total kinetic energy, total mechanical energy, and total electrostatic energy

$$H = \sum_{i,j \in \mathbb{Z}} -\frac{\hbar^2}{2m_e} \Delta_{i,j} + \sum_{i,j \in \mathbb{Z}} \frac{1}{2} m_e \omega_{i,j}^2 r_{i,j}^2 + \frac{1}{2} \sum_{i,j \in \mathbb{Z}} (-e) V_{i,j}(\mathbf{r}_{i,j} = \mathbf{0}_{i,j}) \quad (1)$$

where $\Delta_{i,j}$ is Laplacian taken in the frame of the electron at $\mathbf{R}_{i,j}$, and $V_{i,j}$ is the electrostatic potential of the electron at $\mathbf{R}_{i,j} + \mathbf{r}_{i,j}$ where $\mathbf{r}_{i,j}$ is the displacement from the lattice point. We can see that the total potential energy at zero displacement is not zero but rather the electrostatic energy.

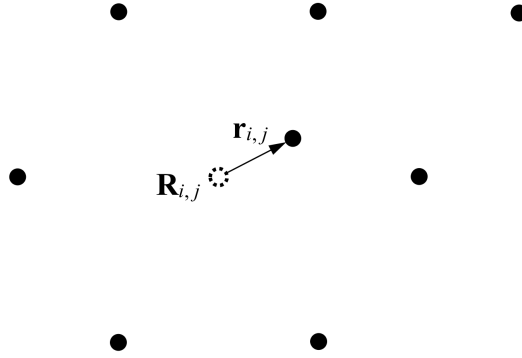


FIG. 1 The small displacement from the lattice point.

Assume the plane is infinitely large, $V_{i,j}(\mathbf{0}_{i,j}) = V$ and $\omega_{i,j} = \omega$ for all $i, j \in \mathbb{Z}$, then

$$H = \sum_{i,j \in \mathbb{Z}} \left(-\frac{\hbar^2}{2m_e} \Delta_{i,j} + \frac{1}{2} m_e \omega^2 r_{i,j}^2 \right) + \frac{1}{2} \sum_{i,j \in \mathbb{Z}} (-e) V \quad (2)$$

Since 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. The expectation of the first term per electron is just the energy of a two-dimensional harmonic oscillator, and the expectation of the second term per electron is the total electrostatic energy per electron; namely,

$$\langle H \rangle_{\text{one}} = (n+1)\hbar\omega + \frac{1}{2}(-e)V = (n+1)\frac{\hbar^2}{m_e w^2} + \frac{1}{2}(-e)V, \quad n = 0, 1, 2, \dots \quad (3)$$

where $w = \sqrt{\frac{\hbar}{m_e \omega}}$ has the unit of length. Wigner crystal needs the lowest,

$$E_{\text{one}} = \frac{\hbar^2}{m_e w^2} + \frac{1}{2}(-e)V \quad (4)$$

In other words, each electron is in the ground state of a two-dimensional harmonic oscillator with a uniform background

$$\psi_{\text{gs}}(r) = \frac{w}{\sqrt{\pi}} e^{-\frac{1}{2}w^2 r^2}, \quad \psi_{\text{gs}}(p) = \sqrt{\frac{w}{\pi \hbar}} e^{-\frac{w^2}{2\hbar^2} p^2} \quad (5)$$

Geometry of the electron lattice

Let 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

$$\mathbf{R}_{i,j} = i\mathbf{a}_1 + j\mathbf{a}_2, \quad i, j \in \mathbb{Z} \quad (6)$$

One can easily find the corresponding reciprocal lattice

$$\mathbf{K}_{l,m} = l\mathbf{b}_1 + m\mathbf{b}_2, \quad l, m \in \mathbb{Z} \quad (7)$$

with the primitive vectors

$$\mathbf{b}_1 = \frac{2\pi}{R}\left(1, -\frac{1}{\sqrt{3}}\right), \quad \mathbf{b}_2 = \frac{2\pi}{R}\left(0, \frac{2}{\sqrt{3}}\right). \quad (8)$$

of course we have

$$|\mathbf{K}_{l,m}| = \frac{4\pi}{\sqrt{3}R} \sqrt{l^2 + m^2 - lm} \quad (9)$$

Further, $d^2\mathbf{R}_{i,j}$ and $d^2\mathbf{K}_{i,j}$ stand for the area elements in Bravais lattice and reciprocal lattice space. Think about the Riemann sum and the integration in calculus

$$\int_D f(\mathbf{x}) d^2\mathbf{x} = \iint_D f(x, y) dx dy = \sum_{x^*} \sum_{y^*} f(x^*, y^*) dx dy$$

$d\mathbf{x}$ is a area element of the partition of the integrating area D and each such area element contain only one point (x^*, y^*) at which the f assumed. Similarly, $d\mathbf{R}_{i,j}$ and $d\mathbf{K}_{i,j}$ must be an area element of a partition of the lattice space and each one only contains a single lattice point. Such area element is called the **Wigner-Seitz cell**. We have

$$d^2\mathbf{R}_{i,j} = \frac{\sqrt{3}}{2}R^2, \quad d^2\mathbf{K}_{i,j} = \frac{8\pi^2}{\sqrt{3}R^2} \quad (10)$$

by elementary Euclidean geometry.

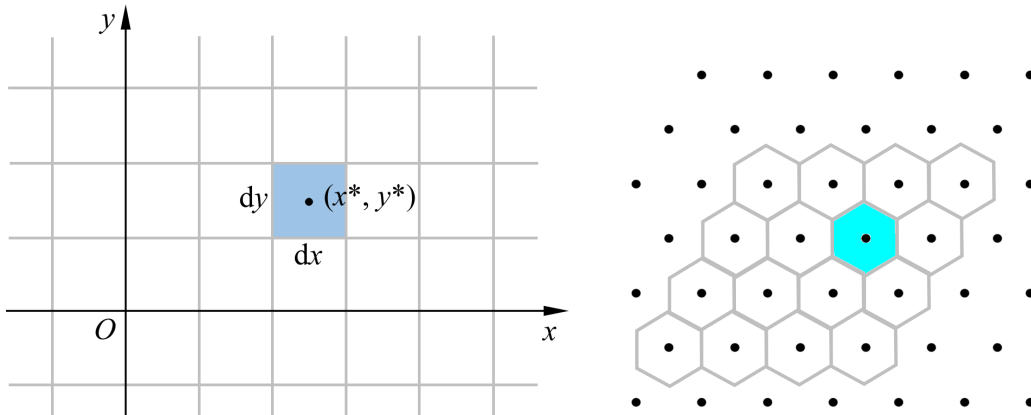


FIG. 2 (left) The partition and the unit cell of \mathbb{R}^2 of the Riemann sum; (right) the partition and the unit cell of the lattice space.

However, the following partition is not a well-defined one since each cell is not identical. The yellow one has a lattice point at the lower left vertex but the green one does not.

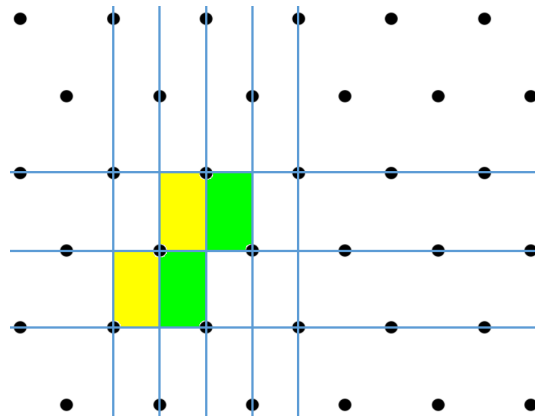


FIG. 3 An ill-defined partition and the unit cell of the lattice space.

The total potential energy per electron

In this section, we are going to find V in formula (4), the total potential energy per electron.

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,

$$V = \sum_{\substack{i,j \in \mathbb{Z} \\ (i,j) \neq (0,0)}} V(\mathbf{R}_{i,j}) \quad (11)$$

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

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

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

$$V(\mathbf{R}_{i,j}) = \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \tilde{V}(\mathbf{k}) e^{i\mathbf{R}_{i,j} \cdot \mathbf{k}} d^2\mathbf{k}. \quad (12)$$

So,

$$V = \sum_{\substack{i,j \in \mathbb{Z} \\ (i,j) \neq (0,0)}} \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \tilde{V}(\mathbf{k}) e^{i\mathbf{R}_{i,j} \cdot \mathbf{k}} d^2\mathbf{k} = \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \tilde{V}(\mathbf{k}) \sum_{\substack{i,j \in \mathbb{Z} \\ (i,j) \neq (0,0)}} e^{i\mathbf{R}_{i,j} \cdot \mathbf{k}} d^2\mathbf{k} \quad (13)$$

The double sum inside the integrand is

$$\sum_{\substack{i,j \in \mathbb{Z} \\ (i,j) \neq (0,0)}} e^{i\mathbf{R}_{i,j} \cdot \mathbf{k}} = -e^{i\mathbf{R}_{0,0} \cdot \mathbf{k}} + \sum_{i,j \in \mathbb{Z}} e^{i\mathbf{R}_{i,j} \cdot \mathbf{k}} = -1 + \frac{2(2\pi)^2}{\sqrt{3}} \frac{1}{R^2} \sum_{l,m \in \mathbb{Z}} \delta(\mathbf{k} - \mathbf{K}_{l,m}) \quad (14)$$

where we use the Fourier series of Dirac delta function δ in the last step. Then, 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

$$\sum_{\substack{i,j \in \mathbb{Z} \\ (i,j) \neq (0,0)}} e^{i\mathbf{R}_{i,j} \cdot \mathbf{k}} = -1 + (2\pi)^2 \frac{n}{-e} \sum_{l,m \in \mathbb{Z}} \delta(\mathbf{k} - \mathbf{K}_{l,m}) \quad (15)$$

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

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

To conclude,

$$\sum_{x \in \mathbb{Z}} e^{ixk} dx = 2\pi \sum_{n \in \mathbb{Z}} \delta(k - 2n\pi) \quad \text{for all } k \quad (16)$$

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

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

where $d^2\mathbf{R}_{i,j} = \frac{\sqrt{3}}{2} R^2$ is the area element in the lattice plane; plug it back into the last formula, we can then get Eq. (14).

After the calculation of the double sum, we can now plug it into (13),

$$\begin{aligned} V &= \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \tilde{V}(\mathbf{k}) \left(-1 + (2\pi)^2 \frac{n}{-e} \sum_{l,m \in \mathbb{Z}} \delta(\mathbf{k} - \mathbf{K}_{l,m}) \right) d^2\mathbf{k} \\ &= -\frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \tilde{V}(\mathbf{k}) d^2\mathbf{k} + \frac{n}{-e} \int_{-\infty}^{\infty} \tilde{V}(\mathbf{k}) \sum_{l,m \in \mathbb{Z}} \delta(\mathbf{k} - \mathbf{K}_{l,m}) d^2\mathbf{k} \\ &= -\frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \tilde{V}(\mathbf{k}) d^2\mathbf{k} + \frac{n}{-e} \sum_{l,m} \tilde{V}(\mathbf{K}_{l,m}) \end{aligned} \quad (17)$$

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

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

$$V(r) = -\frac{1}{4\pi\epsilon_0} \frac{e}{r} \quad \text{with } \lim_{r \rightarrow \infty} V(r) = 0; \quad (18)$$

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

$$\tilde{V}(k) = -\frac{1}{4\pi\epsilon_0} \frac{2\pi e}{k} \quad \text{with } \lim_{k \rightarrow \infty} \tilde{V}(k) = 0; \quad (19)$$

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

^[2] *ibid.*, p.334.

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

We see \tilde{V} is still inverse proportional to the distance in the reciprocal lattice space. This means we cannot avoid 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 (17) 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 $e^{-\frac{1}{2}w^2k^2}$, where $\text{unit}(w) = \text{unit}(k)^{-1}$, and take the limit $w \rightarrow 0$;

$$V = \lim_{w \rightarrow 0} \left(-\frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \tilde{V}(\mathbf{k}) e^{-\frac{1}{2}w^2k^2} d^2\mathbf{k} + \frac{n}{-e} \sum_{l,m} \tilde{V}(\mathbf{K}_{l,m}) e^{-\frac{1}{2}w^2K_{l,m}^2} \right) \quad (20)$$

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.

Assume the expression of V is independent of angle θ , the formula can be simplified as

$$V = \lim_{w \rightarrow 0} \left(-\frac{1}{2\pi} \int_0^\infty \tilde{V}(k) e^{-\frac{1}{2}w^2k^2} k dk + \frac{n}{-e} \sum_{l,m} \tilde{V}(K_{l,m}) e^{-\frac{1}{2}w^2K_{l,m}^2} \right) \quad (21)$$

Find (21) analytically

Plug (19) into it, the first term becomes

$$-\frac{1}{2\pi} \frac{-2\pi e}{4\pi\epsilon_0} \int_0^\infty \frac{1}{k} e^{-\frac{1}{2}w^2k^2} k dk = \sqrt{\frac{\pi}{2}} \frac{e}{4\pi\epsilon_0} \frac{1}{w}, \quad (22)$$

and the second term is

$$\begin{aligned} \frac{n}{-e} \sum_{l,m} \tilde{V}(K_{l,m}) e^{-\frac{1}{2}w^2K_{l,m}^2} &= \frac{n}{-e} \frac{-2\pi e}{4\pi\epsilon_0} \frac{\sqrt{3}R}{4\pi} \sum_{l,m} \frac{1}{\sqrt{l^2 + m^2 - lm}} e^{-\frac{1}{2}w^2 \frac{16\pi^2}{3R^2} (l^2 + m^2 - lm)} \\ &= -\frac{e}{4\pi\epsilon_0} \frac{1}{R} \sum_{l,m} \frac{1}{\sqrt{l^2 + m^2 - lm}} e^{-\frac{8\pi^2 w^2}{3R^2} (l^2 + m^2 - lm)} \\ &= -\frac{e}{4\pi\epsilon_0} \frac{1}{R} \sum_{l,m} \frac{1}{\sqrt{l^2 + m^2 - lm}} e^{-\frac{8\pi^2 \eta^2}{3} (l^2 + m^2 - lm)} \end{aligned}$$

where $\eta := \frac{w}{R}$. This infinite sum converges since it takes the form $\sum_a \frac{e^{-a}}{\sqrt{a}}$. Thus,

$$V(\eta) = \frac{e}{4\pi\epsilon_0} \frac{1}{R} \left(\sqrt{\frac{\pi}{2}} \frac{1}{\eta} - \sum_{l,m \in \mathbb{Z}} \frac{1}{\sqrt{l^2 + m^2 - lm}} e^{-\frac{8\pi^2 \eta^2}{3} (l^2 + m^2 - lm)} \right) \quad (23)$$

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

$$\frac{1}{2}(-e)V(\eta) = -\frac{e^2}{4\pi\epsilon_0} \frac{1}{2R} \left(\sqrt{\frac{\pi}{2}} \frac{1}{\eta} - \sum_{l,m \in \mathbb{Z}} \frac{1}{\sqrt{l^2 + m^2 - lm}} e^{-\frac{8\pi^2 \eta^2}{3} (l^2 + m^2 - lm)} \right) \quad (24)$$

Combine the formula (4) and (24), we have *total energy per electron*

$$E_{\text{one}} = \frac{\hbar^2}{m_e R^2 \eta^2} - \frac{e^2}{4\pi\epsilon_0} \frac{1}{2R} \left(\sqrt{\frac{\pi}{2}} \frac{1}{\eta} - \sum_{l,m \in \mathbb{Z}} \frac{1}{\sqrt{l^2 + m^2 - lm}} e^{-\frac{8\pi^2 \eta^2}{3}(l^2 + m^2 - lm)} \right) \quad (25)$$

We present the exact result here but it is hard to continue doing it analytically due to the double sum. Instead, we are going to do it numerically. ***We abandon the formula (24) and choose to code the second term in formula (21) directly. A more general computer program will compute both two terms in (21) directly; such a program will contain a function that carries the form of the potential $\tilde{V}(k)$ indicated by users.***

The total energy per electron

We are going to evaluate numerically. The first thing is the values of constants. We set

$$\hbar = m_e = e = \frac{1}{4\pi\epsilon_0} = 1 \text{ and } R = 1 \quad [\text{in computer}] \quad (26)$$

So, $\eta = w \in (0, 1)$ and $n = -\frac{2}{\sqrt{3}}$. In addition, $l = m = 0$ will typically give a divergent summand in the second term, so we have to remove it intentionally.

In our case, we need to code E_{one} as a function of η and charge density n ,

$$E_{\text{one}}(\eta, n) = T - \frac{1}{2} (V_{\text{int}} + V_{\text{sum}})$$

where

$$T = \frac{1}{\eta^2} \quad [\text{in computer}]$$

$$V_{\text{int}} = -\frac{1}{2\pi} \int_0^\infty \tilde{V}(k) e^{-\frac{1}{2}\eta^2 k^2} k dk \quad [\text{in computer}]$$

$$V_{\text{sum}} = \frac{2}{\sqrt{3}} \sum_{(l,m) \neq (0,0)} \tilde{V}(K_{l,m}) e^{-\frac{1}{2}\eta^2 K_{l,m}^2} \quad [\text{in computer}]$$

where $K_{l,m}$ is given by the formula (9). Of course, we have to define the function of potential $\tilde{V}(k)$ globally in our code.

Here is an example in Python.

```
1 def Eone(eta):
2     # T: kinetic energy
3     T = 1/(eta*eta)
4
5
6     # Vint: the integral in the expression of V
7     def integrand(k):
8         return Vtilde(k) * np.exp(-0.5*eta*eta*k*k) * k
9     I = integrate.nquad(integrand, [[0, np.inf]])
10    Vint = -1.0/(2.0*np.pi) * I[0]
11
12
13    # Vsum: the summation in the expression of V
14    def K(l,m):
15        return (4*np.pi/np.sqrt(3)) * np.sqrt(l*l + m*m - l*m)
16    S = 0
17    for l in range(-1000, 1001):
18        for m in range(-1000, 1001):
19            if l==0 and m==0:
20                S = S + 0
21            else:
22                S = S + Vtilde(K(l,m)) * np.exp(-0.5*eta*eta*K(l,m)*K(l,m))
23    Vsum = (2/np.sqrt(3)) * S
24
25
26    # returns Eone
27    return T + (-0.5)*(Vint + Vsum)
```

In vacuum,

$$\tilde{V}(k) = -\frac{2\pi}{k} \quad \text{with} \quad \lim_{k \rightarrow \infty} V(k) = 0 \quad [\text{in computer}]; \quad (27)$$

some outputs are

if we treat Eone is a function of eta and n, then

E(5,1)=-0.08533141373155004
E(1,1) = 0.3733429313326011
E(0.1,1)= 90.10665293889625
E(0.01, 0.001)=9937.281847266722
E(0.01, 0.01)=9936.809834459207
E(0.01, 0.1)=9932.08970638404
E(0.01, 1)=9884.88842563238
E(0.01, 5)=9675.104955625
E(0.01, 10)=9412.875618115773
E(0.01, 15)=9150.646280606548
E(0.01, 100)=4692.7475429497135

however, if we set R=1, then n=2/sqrt(3) and Eone is a function only of eta:

E(eta=0.01) = 9997.893564574373
E(eta=0.1) = 97.9212699456691
E(eta=1) = 0.3733429313533917
E(eta=10) = -0.05266570686577501
E(eta=100) = -0.006166570686577503

another way: only terms of R since n is related to R:

E(R=0.01) = 9792.12699456691
E(R=0.1) = 979.212699456691
E(R=1) = 97.92126994566911
E(R=10) = 9.79212699456691
E(R=100) = 0.9792126994566911

???

Quantum melting criterion

Combine