Introduction to the Structure and Properties of Graphene

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This manuscript is typeset by $IAT_EX 2_{\mathcal{E}}$. For better print quality, I would like to accomplish the task in English rather than Chinese. While you are reading, please note that it only gives a glimpse of recent advances of the researches on graphene. I always try to reorganize the materials so that they can be understood by an undergraduate who have already learned solid state physics but has no knowledge of Dirac's equation on relativistic electron, density functional theory, etc. If you find some errors, I will appreciate your correspondence. To get the source file, please drop me a line.

I call our world Flatland, not because we call it so, but to make its nature clearer to you, my happy readers, who are privileged to live in space.

A Square, Flatland¹.

I. BACKGROUND

In 1884, Edwin A. Abbott published an interesting novella entitled Flatland: A Romance of Many Dimensions. The narrator is a square, who leads the reader through some of the special features of two dimensions. As the geometric characters in the story come to learn, dimension matters—a lesson that has become more and more important for physicists. For many years, physicists have studied electronic properties of the twodimensional (2D) systems that occur in layered semiconductors, whose thickness typically extends from 10 to 100 atomic layers. The system can accurately be described as 2D only because of its quantum size effects that make the degrees of freedom for electron motion in the short direction irrelevant. A few years ago, a research group led by Andre K. Geim succeeded in isolating and studying the ultimate flatland—graphene, a one-atom thick sheet of carbon atoms arranged laterally in a honeycomb lattice². This flatland is not only the thinnest material in our universe, but also so charming in its properties that it had already been the object of theoretical study for more than half a century before it at last became available for experimental inspection.

According to the description in the review article³, graphene is a flat monolayer of carbon atoms tightly packed into a two-dimensional honeycomb lattice, and is a basic building block for graphitic materials of all other dimensionalities. It can be wrapped up into 0D fullerenes, rolled into 1D nanotubes or stacked into 3D graphite. Recent researches show that graphene relates to a variety of fields, including quantum electrodynamics for massless fermions^{4,5}, the anomalous quantum Hall effect⁴, Koshino-Taylor effect⁶, and even the proximityinduced superconductivity^{4,7}.

The potential applications of this fantastic material cover single molecule gas detection, graphene nanoribbons, graphene transistors, graphene biodevices, integrated circuits, transparent conducting electrodes and ultracapacitors. You can learn these applications in detail at http://en.wikipedia.org/wiki/Graphene. Here, we only mention one of the recent advances concerning its applications. In February 2010, researchers at IBM reported that they have been able to create graphene transistors with an on and off rate of 100 GHz, far exceeding the rates of previous attempts, and also the speed of silicon. The graphene transistors made at IBM were made using extant silicon-manufacturing equipment, meaning that for the first time graphene transistors are a conceivable—though still fanciful—replacement for silicon.

II. STRUCTURE

The carbon atoms in graphene are arranged in hexagonal structure (see Fig. 1). It can be seen as a triangular lattice with a basis of two atoms per unit cell. Please pay attention to the fact that atoms A, B are not equivalent. The lattice unit vectors can be written as

$$a_1 = \frac{\sqrt{3}a}{2}(\sqrt{3}\hat{x} + \hat{y}), \quad a_2 = \frac{\sqrt{3}a}{2}(\sqrt{3}\hat{x} - \hat{y}), \quad (1)$$

where $a \simeq 1.42$ Å is the carbon-carbon distance. The three nearest-neighbor vectors in real space are given by

$$\boldsymbol{\delta}_1 = a(1,0), \quad \boldsymbol{\delta}_2 = \frac{a}{2}(-1,\sqrt{3}), \quad \boldsymbol{\delta}_1 = -\frac{a}{2}(1,\sqrt{3}).$$
 (2)

Interestingly, the reciprocal space of graphene is still a hexagon, which can be obtained by rotating the real space through 30 degrees. According to the definition of reciprocal-lattice vectors $\mathbf{a}_i \cdot \mathbf{b}_j = 2\pi \delta_{ij}$, i, j = 1, 2, it is easy to get

$$\boldsymbol{b}_1 = \frac{2\pi}{3a}(\hat{x} + \sqrt{3}\hat{y}), \quad \boldsymbol{b}_2 = \frac{2\pi}{3a}(\hat{x} - \sqrt{3}\hat{y}).$$
 (3)

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FIG. 1. (Color online) Left: lattice structure of graphene, made out of two interpenetrating triangular lattices (a_1 and a_2 are the lattice unit vectors, and δ_i , i = 1, 2, 3 are the nearest-neighbor vectors). Right: the reciprocal space (blue) of graphene is still a honeycomb lattice and its first Brillouin zone (green) is also a hexagon. The Dirac cones are located at the K and K' points. These two pictures are generated by Mathematica[®].

The first three Brillouin zones are drawn in Fig. 1 (they are colored in green, yellow and magenta respectively). The first Brillouin zone is also a hexagon. Γ , K, K' and M are all high-symmetry points, among which of particular importance are K and K', i.e. Dirac points. Their positions in momentum space are given by

$$\boldsymbol{K} = \left(\frac{2\pi}{3a}, \frac{2\pi}{3\sqrt{3}a}\right), \quad \boldsymbol{K'} = \left(\frac{2\pi}{3a}, -\frac{2\pi}{3\sqrt{3}a}\right) \quad (4)$$

A similar discussion of the structure of graphene can be found in another review $\operatorname{article}^4$.

III. ELECTRONIC PROPERTIES

A. The Tight Binding Approximation

The tight binding approximation (TBA) neglects interactions between atoms separated by large distances. Suppose that, for atoms A and B, the wave functions of their localized orbitals are $\phi_A(\mathbf{r} - \mathbf{R}_A)$ and $\phi_B(\mathbf{r} - \mathbf{R}_B)$ respectively, where \mathbf{R}_A and \mathbf{R}_B are the position vectors. The the Bloch wave functions of the two kinds of distinct atoms are

$$\psi_{\alpha}(\boldsymbol{r}) = \frac{1}{\sqrt{N}} \sum_{\boldsymbol{R}_{\alpha}} e^{i\boldsymbol{k}\cdot\boldsymbol{R}_{\alpha}} \phi(\boldsymbol{r}-\boldsymbol{R}_{\alpha}), \quad \alpha = A \text{ or } B, \quad (5)$$

Therefore, the total wave function of the system can be constructed by

$$\psi(\mathbf{r}) = C_A \psi_A(\mathbf{r}) + C_B \psi_B(\mathbf{r}) \tag{6}$$

As a quantum system, $\psi(\mathbf{r})$ must satisfy the Schrödinger equation

$$H\psi(\mathbf{r}) = \varepsilon(\mathbf{k})\psi(\mathbf{r}),\tag{7}$$

where H is the Hamiltonian operator. Multiply Eq. (7) by $\psi_A^*(\mathbf{r})$ and $\psi_B^*(\mathbf{r})$ respectively and take the integration over the whole space, then we can obtain

$$H_{AA}C_A + H_{AB}C_B = \varepsilon(\mathbf{k})C_A + \varepsilon(\mathbf{k})C_BS_{AB}, \quad (8a)$$

$$H_{BA}C_A + H_{BB}C_B = \varepsilon(\mathbf{k})C_A S_{BA} + \varepsilon(\mathbf{k})C_B, \quad (8b)$$

where

$$H_{\alpha\beta} = \langle \psi_{\alpha} | H | \psi_{\beta} \rangle, \quad S_{\alpha\beta} = \langle \psi_{\alpha} | \psi_{\beta} \rangle, \tag{9}$$

and α , β can be either A or B.

Let us calculate the four elements in H:

$$H_{AA} = \frac{1}{N} \sum_{\mathbf{R}_{A}} \langle \psi(\mathbf{r} - \mathbf{R}_{A}) | H | \psi(\mathbf{r} - \mathbf{R}_{A}) \rangle$$

= $\langle \psi(\mathbf{r} - \mathbf{R}_{A}) | H | \psi(\mathbf{r} - \mathbf{R}_{A}) \rangle$
= ϵ (10)

Similarly,

$$H_{BB} = \epsilon. \tag{11}$$

These are somewhat trivial. Since $H_{AB} = H_{AB}^{\dagger}$, we only need to calculate one of them. Still, it doesn't need many efforts to arrive at the following

$$H_{AB} = \frac{1}{N} \sum_{\mathbf{R}_{A}} \sum_{\mathbf{R}_{B}} e^{i\mathbf{k}\cdot(\mathbf{R}_{B}-\mathbf{R}_{A})} \langle \psi(\mathbf{r}-\mathbf{R}_{A}) | H | \psi(\mathbf{r}-\mathbf{R}_{B}) \rangle$$
$$= t \left(e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{1}} + e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{2}+e^{i\mathbf{k}\cdot\boldsymbol{\delta}_{3}}} \right)$$
$$= t f(\mathbf{k})$$
(12)

where

$$t = \langle \psi(\boldsymbol{r} - \boldsymbol{R}_A) | H | \psi(\boldsymbol{r} - \boldsymbol{R}_A - \boldsymbol{\delta}_n) \rangle.$$
(13)

Because of the symmetry, n = 1, 2, 3 yield the same value. According to Eq. (2), we have

$$f(\mathbf{k}) = \mathrm{e}^{\mathrm{i}k_x a} + 2\mathrm{e}^{-\mathrm{i}k_x a/2} \cos\left(\frac{\sqrt{3}k_y a}{2}\right) \qquad (14)$$

Then we turn to the other part in Eqs. (8).

$$S_{AB} = \langle \psi_A | \psi_B \rangle = sf(\mathbf{k}),$$

$$s = \langle \psi(\mathbf{r} - \mathbf{R}_A) | \psi(\mathbf{r} - \mathbf{R}_A - \boldsymbol{\delta}_n) \rangle, \quad n = 1, 2, 3.$$
(15)

Now we can rewrite Eqs. (8) as

$$\begin{vmatrix} \epsilon - \varepsilon(\mathbf{k}) & tf(\mathbf{k}) - \varepsilon(\mathbf{k})sf(\mathbf{k}) \\ tf^*(\mathbf{k}) - \varepsilon(\mathbf{k})sf^*(\mathbf{k}) & \epsilon - \varepsilon(\mathbf{k}) \end{vmatrix} = 0, \quad (16)$$

$$\left[\varepsilon(\boldsymbol{k})-\epsilon\right]^2 - \left[s\,\varepsilon(\boldsymbol{k})-t\right]^2 f(\boldsymbol{k})f^*(\boldsymbol{k}) = 0$$

Referring to Eq. (2), we can easily obtain

$$\omega(\mathbf{k}) = \|f(\mathbf{k})\| = \left[3 + 2\cos\left(\sqrt{3}k_ya\right) + 4\cos\left(\frac{\sqrt{3}k_ya}{2}\right)\cos\left(\frac{3k_xa}{2}\right)\right]^{\frac{1}{2}}.$$
(17)

Finally, we come to the widely used expression in the research of graphene:

$$\varepsilon_{\pm}(\mathbf{k}) = \frac{\epsilon \pm t \,\omega(\mathbf{k})}{1 \pm s \,\omega(\mathbf{k})} \tag{18}$$

B. π and π^* Band

We will take a much more detailed discussion on the physical aspects of Eq. (18), where t is the TBA hopping parameter, s is the overlap parameter, and ϵ is the on-site energy parameter, often chosen as zero. The parameters t and ϵ are expressed in electron-volt units (eV), whereas s is dimensionless. In the expression of $\varepsilon_{\pm}(\mathbf{k})$, the plus sign corresponds to the π^* band and the minus sign corresponds to the π band. In fact, each atom in the lattice of graphene has one s and three p orbitals. The s orbital and two in-plane p orbitals are tied up in graphene's strong covalent bonding and do not contribute to its conductivity. The remaining p orbital, oriented perpendicular to the molecular plane, is odd under inversion in the plane and hybridizes to form π (valence) and π^* (conduction) bands².

To provide a visualized description, we set t = -3 eV, s = 0.13 and then plot a pseudo-3D energy dispersion and constant energy contours for the two types of bands respectively (see Fig. 2). If we only concern about the band structure around zero energy, we can assume that s = 0. Then the spectrum is symmetric and can be written as

$$\varepsilon(\mathbf{k}) = \pm t\,\omega(\mathbf{k}).\tag{19}$$



FIG. 2. (Color online) **Up**: pseudo-3D energy dispersion for the two π -bands in the first Brillouin zone of a 2D honeycomb graphene lattice. **Middle**: constant energy contours for the π -valence band. **Down**: same constant energy contours for the π^* -conduction band⁸. For figures of high quality and more interactions, we recommend you directly refer to the demonstration online.

Rewriting the momentum k as a sum of K and q, i.e.

$$\boldsymbol{k} = \boldsymbol{K} + \boldsymbol{q} = \left(\frac{2\pi}{3a} + q_x, \frac{2\pi}{3\sqrt{3}a} + q_y\right), \qquad (20)$$

we can assume that q is small and expand $\omega^2(q)$ to the leading order in this vector parameter:

$$\|f(q)\|^{2} = 3 + 2\cos\left[\sqrt{3}\left(\frac{2\pi}{3\sqrt{3}a} + q_{y}\right)a\right] + 4\cos\left[\frac{\sqrt{3}}{2}\left(\frac{2\pi}{3\sqrt{3}a} + q_{y}\right)a\right]\cos\left[\frac{3}{2}\left(\frac{2\pi}{3a} + q_{x}\right)a\right]$$

$$= 3 - \cos\left(\sqrt{3}q_{y}a\right) + \sqrt{3}\sin\left(\sqrt{3}q_{y}a\right) - 2\left[\cos\left(\frac{\sqrt{3}q_{y}a}{2}\right) + \sqrt{3}\sin\left(\frac{\sqrt{3}q_{y}a}{2}\right)\right]\cos\left(\frac{3q_{x}a}{2}\right)$$

$$\simeq 3 - \left(1 - \frac{3q_{y}^{2}a^{2}}{2}\right) + 3q_{y}a - 2\left[\left(1 - \frac{3q_{y}^{2}a^{2}}{8}\right) + \frac{3}{2}q_{y}a\right]\left(1 - \frac{9q_{x}^{2}a^{2}}{8}\right)$$

$$\simeq \frac{9}{4}\left(q_{x}^{2} + q_{y}^{2}\right)a^{2}$$

(21)

Then we arrive at the following approximation, which may be regarded as the most striking distinctiveness of graphene

$$\varepsilon(\boldsymbol{q}) \approx \pm v_F \hbar \|\boldsymbol{q}\|.$$
 (22)

For the band structure close to K', just as we expect, it will yield the same result. In Eq. (22), v_F denotes the Fermi velocity, given by $v_F = 3ta/2\hbar$. Substituting the values of the constants, we have

$$v_F = \frac{9 \times 1.602 \times 10^{-19} \times 1.42 \times 10^{-10}}{2 \times 1.055 \times 10^{-34}} = 9.7 \times 10^5 \,\mathrm{m/s}$$

For s = 0, it is possible to derive an analytical expression for the density of states per unit cell, which involves the first type complete elliptic integral⁴. Close to the Dirac points, the density of states per unit cell is given by (with a degeneracy of 4 included)

$$\rho(\varepsilon) = \frac{4A_c}{(2\pi)^2} \int \frac{\mathrm{d}l}{|\nabla_q \omega(\boldsymbol{q})|} = \frac{A_c}{\pi^2} \frac{2\pi |\boldsymbol{q}|}{v_F} = \frac{2A_c|\varepsilon|}{\pi \hbar v_F^2}, \quad (23)$$

where A_c is the unit cell area of the reciprocal space given by $A_c = 3\sqrt{3}a^2/2$, since the Bravais lattice is a diamond (see Fig. 1).

IV. 2D DIRAC EQUATION

In this section, we introduce a little knowledge about the 2D Dirac equation, mainly dealing with the intuitive deduction but not its solutions. According to the Eqs. (10, 11, 12) and the Hermitian property of H, it is easy to obtain the Dirac-like Hamiltonian

$$H = \hbar v_F \begin{pmatrix} 0 & q_x - iq_y \\ q_x + iq_y & 0 \end{pmatrix} = \hbar v_F \boldsymbol{\sigma} \cdot \boldsymbol{q}, \qquad (24)$$

where $\boldsymbol{\sigma} = (\sigma_x, \sigma_y)$ and σ_x, σ_y are the Pauli matrices. However, it should be noted that this kind of expression of Hamiltonian only holds for k around the Dirac point K. For the other point K', the corresponding expression is

$$H = \hbar v_F \boldsymbol{\sigma}^* \cdot \boldsymbol{q},\tag{25}$$

where $\boldsymbol{\sigma}^* = (\sigma_x, -\sigma_y)$. Recall the momentum operator in quantum mechnics

$$\boldsymbol{p} = -i\hbar\nabla \tag{26}$$

and the relation between the momentum p and q

$$\boldsymbol{p} = \hbar \, \boldsymbol{q},\tag{27}$$

then we can rewrite the classical Shrödinger equation $H\phi(\mathbf{r}) = \varepsilon\phi(\mathbf{r})$ as the famous 2D Dirac equation

$$-i\hbar v_F \boldsymbol{\sigma} \cdot \nabla \phi(\boldsymbol{r}) = \varepsilon \phi(\boldsymbol{r}). \tag{28}$$

Of course, the two-component wave function $\phi(\mathbf{r})$ that appears here only describes the distribution of electrons' density close to the K point.

Furthermore, let us consider the problem of a uniform magnetic field \boldsymbol{B} applied perpendicular to the graphene plane. Replacing $-i\hbar\nabla$ in Eq. (28) by $-i\hbar\nabla + e\boldsymbol{A}/c$, we obtain

$$v_{F} \big[\boldsymbol{\sigma} \cdot (-i\hbar \nabla + e\boldsymbol{A}/c) \big] \phi(\boldsymbol{r}) = \varepsilon \phi(\boldsymbol{r}).$$
(29)

Perhaps it the right time for us to stop here.

In addition, there are still two more points worthy of special attention with respect to Eq. (22). First, since most theorists would like to use units such that $\hbar = 1$, it is often convenient to regard q as the momentum, just as we have done before. Second, if you are familiar with the mass-energy relation of photon (i.e. E = pc), you may have come to the conclusion that the quasiparticles involved in the theory of graphene is massless, although it belongs to fermions. The analogy between Eq. (28) and

the classical Dirac's equation for electrons also confirms this declaration. In the following equation

$$i\hbar\frac{\partial}{\partial t}\phi(\mathbf{r}) = (-i\hbar c\mathbf{\alpha} \cdot \nabla + \beta mc^2)\phi(\mathbf{r}), \qquad (30)$$

if we set m = 0, then you can see clearly how similar these two equations are!

V. VIBRATIONAL AND THERMAL PROPERTIES

A. The Phonon Dispersion

In general, there are three methods to derive the phonon dispersion relation of graphene: the generalizedgradient approximation (GGA), the local-density approximation (LDA)⁹ and the fourth-nearest-neighbor force-constant model (4NNFC)¹⁰. The experimental results derived from the inelastic X-ray scattering can be found in Ref. 11. An introduction to such methods is far beyond our scope. However, it is still worthy of some efforts to illustrate the main results (see Fig. 3). The dispersion relation of graphene comprises three acoustic (A) and three optical (O) modes, which are either out-of-plane (Z), in-plane longitudinal (L), or transverse (T). You should pay special attention to the dispersion relations of LA, TA and ZA near Γ , for certain reasons appearing in the next subsection.



FIG. 3. *Ab initio* phonon dispersion relation of graphene. Dashed line: LDA calculation. Solid line: GGA calculation. The circles and asterisks denote the calculations by other groups. (Figure is taken from Ref. 9.)

B. The Specific Heat

At low temperature, only acoustic modes are excited. As Ref. 10 points out, in the low-temperature regime (below 20 K), the specific heat of graphene has a linear T dependence. The conclusion we can draw from this is that the low-temperature behavior of graphene cannot be merely explained by Debye's theory; otherwise, C_V is proportional to T^2 . In the following, we will provide a brief proof.

In Debye's theory, it is assumed that atoms' thermal vibration takes the form of elastic waves. For 2D system, when $\omega < \omega_D$, the following relation holds:

$$g(\omega) \,\mathrm{d}\omega = \frac{a^2}{2\pi} \left(\frac{1}{v_{\parallel}^2} + \frac{1}{v_{\perp}^2} \right) \omega \,\mathrm{d}\omega, \tag{31}$$

where $g(\omega)$ is the density of vibration modes, v_{\parallel} and v_{\perp} are the velocities of longitudinal and transverse waves respectively. Then, the specific heat can be calculated through

$$C_{V} = \frac{\hbar^{2}}{k_{B}T^{2}} \int_{0}^{\omega_{D}} \frac{\omega^{2} \exp(\hbar\omega/k_{B}T)}{\left[\exp(\hbar\omega/k_{B}T) - 1\right]^{2}} g(\omega) \,\mathrm{d}\omega$$

$$= \frac{k_{B}^{3}a^{2}}{2\pi\hbar^{2}} \left(\frac{1}{v_{\parallel}^{2}} + \frac{1}{v_{\perp}^{2}}\right) T^{2} \int_{0}^{x_{D}} \frac{x^{3}e^{x}}{(e^{x} - 1)^{2}} \,\mathrm{d}x$$
(32)

where $x = \hbar \omega / k_B T$, $x_D = \hbar \omega_D / k_B T$, k_B is the Boltzman's constant. When the temperature is very low, i.e. $\hbar \omega_D \gg k_B T$, $x_D \gg 1$, we have

$$\int_0^{x_D} \frac{x^3 e^x}{(e^x - 1)^2} \, \mathrm{d}x \approx \int_0^\infty \frac{x^3 e^x}{(e^x - 1)^2} \, \mathrm{d}x = 6\zeta(3), \quad (33)$$

where $\zeta(s) = \sum_{n=1}^{\infty} n^{-s}$ is the Riemann's zeta-function. Now, we have proven that

$$C_V = \frac{3k_B^3 a^2}{\pi \hbar^2} \left(\frac{1}{v_{\parallel}^2} + \frac{1}{v_{\perp}^2} \right) \zeta(3) T^2 \propto T^2 \qquad (34)$$

holds if the 2D system can be described by Debye's theory. Therefore, we can further conclude that it is the ZA mode that mainly contributes to graphene's thermal behavior at low temperature. Referring to Fig. 3 again, you can see that the ZA mode shows a quadratic energy dispersion near Γ while the TA and LA modes present a linear dispersion.

VI. FURTHER READING

Numerous materials on graphene can be found in the following subcategory in arXiv: cond-mat. If you want to look for a much easier and interesting introduction to graphene, you may would like to read the paper in Scientific American¹². Besides, Wikipedia also provides useful links to the information about graphene: http://en.wikipedia.org/wiki/Graphene; and the Wolfram Demonstrations Project has a dozen of demonstrations concerning the subject of graphene: http://demonstrations.wolfram.com.

¹A copy of this interesting book can be downloaded from the site: http://www.mat.ufmg.br/gaal/bibliografia/flatland.pdf.

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