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### Electronic edge states in graphene sheets

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#### Abstract

Electronic edge ("surface") states are investigated in semi-infinite graphene sheets and graphene ribbons (monolayers) with armchair, zig-zag or horseshoe edges within the nearestneighbor tight-binding approximation. The problem is generalized to include edge elements of the hoping (transfer) matrix distinct from the infinite-sheet ("bulk") ones. Within this model the semi-infinite graphene sheets with zig-zag or horseshow edges exhibit edge states, while the semi-infinite graphene sheet with armchair edge does not. Similarly, symmetric graphene ribbons with zig-zag or horseshoe edges have edges states, while ribbons with asymmetric edges (zig-zag and horseshoe) have not. It is also shown how to construct the "reflected" solution for the intervening equations with finite diferences both for semi-infinite sheets and ribbons, either with modified elements of the hoping matrix at the edges, or with uniform matrix elements. It is also indicated how to extend the method to rectangular, finite-size pieces of graphene sheets.

Keywords: graphene sheets, graphene ribbons, electronic edge states

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Graphene sheets (monolayers), described as early as 1947, [1]-[3] have been eventually isolated and identified in 2004-2005.[4]-[7] They are two-dimensional pieces of carbon (graphite) solids with honeycomb lattice. As it is well known, (free) two-dimensional solids cannot exist, because of atomic fluctuations. [8]-[12] In the case of graphene (as well as other two-dimensional crystals that may exist), several, more or less unknown, factors may conspire to make free-standing sheets stable, most likely their non-thermodinamic, small sizes (in this respect, independent, small-size graphene sheets can be viewed as instances of genuine quantum solids). Graphene sheets exhibit a linear spectrum of electronic excitations (Dirac massless fermions), arising from carbon  $\pi$ -electrons, which attracted much interest, especially due to their long lifetime (and mean free path). In particular, the electronic transport along well-defined sheet edges enjoys a special attention.[13]-[20] As it is well known, the identification of the edge ("surface") states requires well-defined boundary conditions. Within the tight-binding approximation, the hoping matrix elements at the edges are modified with respect to the matrix elements of the infinite sheet ('bulk").|21|-|26|In this respect, the edge states of graphene sheets have been tackled by using the tight-binding approximation within limited asymptions (see, for instance, Ref. [19]). On the other hand, using the Dirac equation for identifying edge states in graphene sheets implies the continuum limit of the solid, which is different from finite-difference equations of the tight-binding approach. [27] We give here a more advanced description of electronic edge states in graphene sheets, by using the nereast-neighbour tight-binding approximation with generalized boundary conditions. As it is

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well known, this is an old technique, employed extensively in the past in the ferromagnetism of thin films[28]-[30] and derived from previous studies of electron dynamics in crystal lattices.[31] The edge states are the counterpart in two dimensions of the Tamm or Shockley surface states in three-dimensional crystals.[21]-[23]

First we consider an infinite sheet of carbon hexagons as shown in Figs. 1-3. The position of an atom is identified by the vectors (m, n) and  $\mathbf{v}_{1,2,3}$ . For instance, the vectors  $\mathbf{v}_{1,2,3}$  in Fig. 1 are given by  $\mathbf{v}_1 = (1,0)$ ,  $\mathbf{v}_2 = (-1/2, \sqrt{3}/2)$  and  $\mathbf{v}_3 = (-1/2, -\sqrt{3}/2)$  (the hexagon side is taken equal to unity). It is easy to see that we get the hexagonal periodicity of the lattice by applying twice the vectors  $\mathbf{v}_{1,2,3}$ . The wavefunction coefficients of the tight-binding approximation (the annihilation operators of the on-site fermionic states) are denoted by  $a_{mn}$  and  $b_{mn}^{\mathbf{v}}$ , where  $\mathbf{v}$  denotes one of the vectors  $\mathbf{v}_{1,2,3}$ ; coefficients like  $a_{mn}^{\mathbf{v}_1-\mathbf{v}_2}$ , etc are coefficients of the type  $a_{m'n'}$ , due to the lattical periodicity. The equations of motion for  $a_{mn}$  and  $b_{mn}^{\mathbf{v}}$  within the nearest-neighbour tight-binding approximation are given by

$$\varepsilon a_{mn} = t(b_{mn}^{\mathbf{v}_1} + b_{mn}^{\mathbf{v}_2} + b_{mn}^{\mathbf{v}_3}) ,$$

$$\varepsilon b_{mn}^{\mathbf{v}_1} = t^* (a_{mn} + a_{mn}^{\mathbf{v}_1 - \mathbf{v}_2} + a_{mn}^{\mathbf{v}_1 - \mathbf{v}_3}) ,$$
(1)
$$\varepsilon b_{mn}^{\mathbf{v}_2} = t^* (a_{mn} + a_{mn}^{\mathbf{v}_2 - \mathbf{v}_1} + a_{mn}^{\mathbf{v}_2 - \mathbf{v}_3}) ,$$

$$\varepsilon b_{mn}^{\mathbf{v}_3} = t^* (a_{mn} + a_{mn}^{\mathbf{v}_3 - \mathbf{v}_2} + a_{mn}^{\mathbf{v}_3 - \mathbf{v}_1}) ,$$

where  $\varepsilon$  is the energy (on-site energy included) and t is the transfer (hoping) matrix element.

We get periodic solutions of this system of equations of the form  $a_{mn}^{\mathbf{v}} \sim A(\mathbf{K})e^{i\mathbf{K}[((m,n)+\mathbf{v}])}$ ,  $b_{mn}^{\mathbf{v}} \sim B(\mathbf{K})e^{i\mathbf{K}[((m,n)+\mathbf{v}])}$ , where the wavevector **K** has the components  $\mathbf{K} = (k,q)$ , for energies given by

$$|\lambda|^{2} = 3 + 2\cos \mathbf{K}(\mathbf{v}_{1} - \mathbf{v}_{2}) + 2\cos \mathbf{K}(\mathbf{v}_{1} - \mathbf{v}_{3}) + 2\cos \mathbf{K}(\mathbf{v}_{2} - \mathbf{v}_{3}) , \qquad (2)$$

where  $\lambda = \varepsilon/t$ ; we get from equation (2)

$$|\lambda|^{2} = \varepsilon^{2} / |t|^{2} = 1 + 4\cos\frac{3k}{2}\cos\frac{\sqrt{3}q}{2} + 4\cos^{2}\frac{\sqrt{3}q}{2} .$$
(3)

We have also the representation

$$\lambda A = B(e^{i\mathbf{K}\mathbf{v}_1} + e^{i\mathbf{K}\mathbf{v}_2} + e^{i\mathbf{K}\mathbf{v}_3}) , \ \lambda^* B = A(e^{-i\mathbf{K}\mathbf{v}_1} + e^{-i\mathbf{K}\mathbf{v}_2} + e^{-i\mathbf{K}\mathbf{v}_3}) ,$$
(4)

which implies  $A = B^*$ ; under these circumstances we may take t (and  $\lambda$ ) real and equations (4) become

$$\lambda A = B(e^{ik} + 2e^{-ik/2}\cos\frac{\sqrt{3}q}{2}) , \ \lambda B = A(e^{-ik} + 2e^{ik/2}\cos\frac{\sqrt{3}q}{2}) .$$
 (5)

The energy  $\varepsilon$  given by equation (3) is a well-known result; [1, 32] we can write  $\varepsilon = \pm t\sqrt{S}$ , where

$$S = 1 + 4\cos\frac{3k}{2}\cos\frac{\sqrt{3}q}{2} + 4\cos^2\frac{\sqrt{3}q}{2} ; \qquad (6)$$

the function S is positive over the Brillouin zone defined by the hexagon  $3k/2 = \pm \pi$ ,  $\sqrt{3}q/2 = \pm \pi/3$  and k = 0,  $\sqrt{3}q/2 = \pm 2\pi/3$ ; it ranges from S = 9 at the centre of the Brillouin zone to S = 0 at the hexagon corners; at these points the energy goes like  $\varepsilon = \pm (3t/2)K$ , where  $\mathbf{K} = (k,q)$  is the wavevector referred to the hexagon's corners. This is a gapless Dirac-like electronic spectrum.



Figure 1: Semi-infinite graphene sheet with "armchair" edge.



Figure 2: Semi-infinite graphene sheet with zig-zag edge.

Graphene sheets are zero-gap semiconductors (or zero-overlap semimetals). The linear electronic spectrum is similar with the electron excitations in a normal Fermi liquid (Landau quasi-particles). Of course, the degeneracy at the gapless points in the Brillouin zone may be removed by distortions of the Jahn-Teller type, if other constraints are not present.

We turn now to a semi-infinite graphene sheet with armchair edge, as shown in Fig. 1. We take the site denoted by m, 0 as a reference site for the edge sites; the elements of the transfer matrix are modified at the edge, as shown in Fig. 1; in addition, the coefficients corresponding to the sites lying on the edge line are modified into  $a_{m0}^{'v_3-v_2}$  and  $b_{m0}^{'v_1-v_2+v_3}$ . The relevant equations of motion are

$$\varepsilon b_{m0}^{\mathbf{v}_{3}} = t(a_{m0} + a_{m0}^{\mathbf{v}_{3}-\mathbf{v}_{1}}) + t'a_{m0}'^{\mathbf{v}_{3}-\mathbf{v}_{2}} ,$$

$$\varepsilon a_{m0}^{\mathbf{v}_{1}-\mathbf{v}_{2}} = t(b_{m0}^{\mathbf{v}_{1}} + b_{m0}^{2\mathbf{v}_{1}-\mathbf{v}_{2}}) + t'b_{m0}'^{\mathbf{v}_{1}-\mathbf{v}_{2}+\mathbf{v}_{3}} ,$$

$$\varepsilon a_{m0}'^{\mathbf{v}_{3}-\mathbf{v}_{2}} = t'b_{m0}^{\mathbf{v}_{3}} + t''b_{m0}'^{\mathbf{v}_{3}-\mathbf{v}_{2}+\mathbf{v}_{1}} ,$$

$$\varepsilon b_{m0}'^{\mathbf{v}_{1}-\mathbf{v}_{2}+\mathbf{v}_{3}} = t'a_{m0}^{\mathbf{v}_{1}-\mathbf{v}_{2}} + t''a_{m0}'^{\mathbf{v}_{3}-\mathbf{v}_{2}} ;$$
(7)

or, introducing the notations  $\lambda = \varepsilon/t$ ,  $t' = t(1 + \sigma)$  and  $t'' = t(1 + \rho)$ ,

$$\lambda B = A(e^{-i\mathbf{K}\mathbf{v}_3} + e^{-i\mathbf{K}\mathbf{v}_1}) + (1+\sigma)A'e^{-i\mathbf{K}\mathbf{v}_2} ,$$
  

$$\lambda A = B(e^{i\mathbf{K}\mathbf{v}_2} + e^{i\mathbf{K}\mathbf{v}_1}) + (1+\sigma)B'e^{i\mathbf{K}\mathbf{v}_3} ,$$
  

$$\lambda A' = (1+\sigma)Be^{i\mathbf{K}\mathbf{v}_2} + (1+\rho)B'e^{i\mathbf{K}\mathbf{v}_1} ,$$
  

$$\lambda B' = (1+\sigma)Ae^{-i\mathbf{K}\mathbf{v}_3} + (1+\rho)A'e^{-i\mathbf{K}\mathbf{v}_1} .$$
(8)

Making use of equations (4) we get  $A = (1 + \sigma)A'$ ,  $B = (1 + \sigma)B'$  from the first two equations (8) and

$$e^{i\mathbf{K}\mathbf{v}_3} = \sigma(2+\sigma)e^{i\mathbf{K}\mathbf{v}_2} + \rho e^{i\mathbf{K}\mathbf{v}_1} , \ e^{-i\mathbf{K}\mathbf{v}_2} = \sigma(2+\sigma)e^{-i\mathbf{K}\mathbf{v}_3} + \rho e^{-i\mathbf{K}\mathbf{v}_1}$$
(9)

from the last two equations (8). We look for damped solutions of the form  $\mathbf{K} = (k, q) \rightarrow (k, iq)$ , corresponding to edge states (q > 0); equations (9) become

$$\rho e^{i\frac{3k}{2}} + \sigma(2+\sigma)e^{-\frac{\sqrt{3}q}{2}} - e^{\frac{\sqrt{3}q}{2}} = 0 \quad , \tag{10}$$

which has no solution (except for a few isolated points in the Brillouin zone). We conclude that the semi-infinite graphene sheet with armchair edge has no edge state (within the approximation used here). Similar results have been obtained recently in Ref. [33].

We note that the ideal case of a semi-infinite sheet without edge distortion ( $\sigma = 0, \rho = 0, A' = A, B' = B$ ) has the well-known "reflected" solution

$$Ae^{i\mathbf{K}[(m,n)+\mathbf{v}]} \to A_1 e^{i\mathbf{K}[(m,n)+\mathbf{v}]} + A_2 e^{i\mathbf{K}'[(m,n)+\mathbf{v}]} , \qquad (11)$$
$$Be^{i\mathbf{K}[(m,n)+\mathbf{v}]} \to B_1 e^{i\mathbf{K}[(m,n)+\mathbf{v}]} + B_2 e^{i\mathbf{K}'[(m,n)+\mathbf{v}]} ,$$

where  $\mathbf{K} = (k, q)$  and  $\mathbf{K}' = (k, -q)$ . Indeed, the first two equations (8) are satisfied automatically (by virtue of equations (4)), while from the last two equations (8) we get

$$B_1 e^{i\mathbf{K}\mathbf{v}_3} + B_2 e^{i\mathbf{K}'\mathbf{v}_3} = 0 ,$$

$$A_1 e^{i\mathbf{K}\mathbf{v}_2} + A_2 e^{i\mathbf{K}'\mathbf{v}_2} = 0$$
(12)

which gives  $A_1, B_2 \sim e^{-i\sqrt{3}q/2}$  and  $A_2 B_1 \sim -e^{i\sqrt{3}q/2}$ . The solutions are products of plane waves along the *x*-direction (coordinate *m*) and sin-waves along the *y*-direction (coordinate *n*),

$$Ae^{ik(m+v_x)}\sin q(n-\sqrt{3}/2+v_y) , Be^{ik(m+v_x)}\sin q(n+\sqrt{3}/2+v_y) , \qquad (13)$$

exhibiting nodes on the sites along the directions perpendicular to the edge (A and B in equation (13) are undetermined constants).

We discuss now two other semi-infinite graphene sheets, one with a zig-zag edge and another with a horseshoe edge, as shown in Fig. 2 and, respectively, Fig. 3. The sheet with zig-zag edge is the sheet with armchair edge rotated by the angle  $-\pi/2$ . Equations (4) for the amplitudes are preserved, while the energy is given by

$$\lambda^2 = 1 + 4\cos\frac{3q}{2}\cos\frac{\sqrt{3}k}{2} + 4\cos^2\frac{\sqrt{3}k}{2} .$$
 (14)



Figure 3: Semi-infinite graphene sheet with horseshoe edge.

(compare with equation (3)). The equations for the relevant edge sites in this case are given by

$$\varepsilon a_{m0}^{-\mathbf{v}_{2}} = t b_{m0} + t' b_{m0}^{'-\mathbf{v}_{2}+\mathbf{v}_{1}} + t' b_{m0}^{'-\mathbf{v}_{2}+\mathbf{v}_{3}} ,$$

$$\varepsilon b_{m0}^{'-\mathbf{v}_{2}+\mathbf{v}_{1}} = t' a_{m0}^{-\mathbf{v}_{2}} + t' a_{m0}^{-\mathbf{v}_{2}+\mathbf{v}_{1}-\mathbf{v}_{3}} ,$$

$$\varepsilon b_{m0}^{'-\mathbf{v}_{2}+\mathbf{v}_{3}} = t' a_{m0}^{-\mathbf{v}_{2}} + t' a_{m0}^{-\mathbf{v}_{2}-\mathbf{v}_{1}+\mathbf{v}_{3}} ,$$
(15)

or

$$\lambda A = B e^{i\mathbf{K}\mathbf{v}_{2}} + (1+\sigma)B' e^{i\mathbf{K}\mathbf{v}_{1}} + (1+\sigma)B' e^{i\mathbf{K}\mathbf{v}_{3}} ,$$

$$\lambda B' = (1+\sigma)A(e^{-iKv_{1}} + e^{-iKv_{3}}) .$$
(16)

Making use of equations (4) we get  $B = (1 + \sigma)B'$  from the first equations (16) and

$$\sigma(2+\sigma)(e^{-i\mathbf{K}\mathbf{v}_1} + e^{-i\mathbf{K}\mathbf{v}_3}) = e^{-i\mathbf{K}\mathbf{v}_2}$$
(17)

from the second equations (16). Equation (17) can also be written as

$$2\sigma(2+\sigma)\cos\frac{\sqrt{3}k}{2} = e^{-i\frac{3q}{2}} .$$
 (18)

For  $q \rightarrow iq$  (damped solutions along the direction perpendicular to the edge) this equation becomes

$$2\sigma(2+\sigma)\cos\frac{\sqrt{3}k}{2} = e^{\frac{3q}{2}};$$
(19)

it admits solutions for  $-1 - \sqrt{2}/2 < \sigma < -1 + \sqrt{2}/2$  or  $\sigma < -1 - \sqrt{6}/2$ ,  $\sigma > -1 + \sqrt{6}/2$ . We conclude that the semi-infinite graphene sheet with zig-zag edge has electronic edge states, which are propagating, plane waves along the direction parallel with the edge (wavevector k) and damped waves along the direction perpendicular to the edge ( $\sim e^{-q(n+v_y)}$ ), for values of (k,q) given by equation (19). The energy of these edge states is given by

$$\lambda^{2} = 1 + 4\cosh\frac{3q}{2}\cos\frac{\sqrt{3}k}{2} + 4\cos^{2}\frac{\sqrt{3}k}{2} = \left[1 + \frac{1}{\sigma(2+\sigma)}\right] \left[1 + \frac{1}{\sigma(2+\sigma)}e^{3q}\right]$$
(20)

for  $0 < q < \frac{2}{3} \ln |2\sigma(2+\sigma)|$ ; for each value of q there exist two values of k in the Brillouin zone which satisfy equation (19). The energy given by equation (20) lies in the band gap. The usual case treated previously (see, for instance, Ref. [19]) corresponds to  $\sigma = -1(t' = 0$  and vanishing energy from equation (20)).

Similarly, equations (4) and (14) hold for the semi-infinite sheet with a horseshoe edge (Fig. 3) (this is a rather unrealistic situation, since the dangling bonds terminate usually with hydrogen which does not contribute to electronic states[20]). The equations for the edge sites in this case are given by

$$\varepsilon b_{m0}^{\mathbf{v}_1} = t(a_{m0} + a_{m0}^{\mathbf{v}_1 - \mathbf{v}_3}) + t' a_{m0}'^{\mathbf{v}_1 - \mathbf{v}_2} ,$$

$$\varepsilon a_{m0}'^{\mathbf{v}_1 - \mathbf{v}_2} = t' b_{m0}^{\mathbf{v}_1} .$$
(21)

Using the same technique as for the preceding case we get the equation

$$\frac{1}{2}\sigma(2+\sigma)e^{-\frac{3q}{2}} = \cos\frac{\sqrt{3}k}{2}$$
(22)

for the edge states. Equation (22) has two solutions for k for each value of q in the interval  $0 < q < \frac{2}{3} \ln \left| \frac{1}{2} \sigma(+\sigma) \right|$ , provided  $\sigma < -1 - \sqrt{3}$  or  $\sigma > -1 + \sqrt{3}$ . The energy (equation (14)) is given by

$$\lambda^{2} = 1 + 4\cosh\frac{3q}{2}\cos\frac{\sqrt{3}k}{2} + 4\cos^{2}\frac{\sqrt{3}k}{2} = (1+\sigma)^{2}\left[1+\sigma(2+\sigma)e^{-3q}\right]$$
(23)

A graphene ribbon can be treated in a similar way. Let us assume such a ribbon with *n*-rows running from n = 0 to n = N and with both zig-zag edges. We note that *a*-type unknown functions pertain to one edge, while *b*-type unknown functions pertain to the other edge. The solutions consist of "reflected" waves of the form (11), which are superpositions of direct waves of the form  $(A_1, B_1)e^{-qn}$  and reflected waves of the form  $(A_2, B_2)e^{qn}$  (*i.e.*,  $q \to iq$  in **K** and **K**'in equations (11)). The desired behaviour of the reflected solutions is ensured by the factor  $e^{-qN}$ , *i.e.* we set  $(A_2, B_2)e^{-q(N-n)}$ . The relevant equations for the N-edge are given by

$$\varepsilon b_{2mN}^{\mathbf{v}_2} = t a_{2mN} + t' a_{2mN}^{'\mathbf{v}_2 - \mathbf{v}_1} + t' a_{2mN}^{'\mathbf{v}_2 - \mathbf{v}_3} ,$$

$$\varepsilon a_{2mN}^{'\mathbf{v}_2 - \mathbf{v}_1} = t' b_{2mN}^{\mathbf{v}_2} + t' b_{2mN}^{\mathbf{v}_2 - \mathbf{v}_1 + \mathbf{v}_3} ,$$

$$\varepsilon a_{2mN}^{'\mathbf{v}_2 - \mathbf{v}_3} = t' b_{2mN}^{\mathbf{v}_2} + t' b_{2mN}^{\mathbf{v}_2 + \mathbf{v}_1 - \mathbf{v}_3}$$
(24)

(compare with equations (15)); the energy given by equation (14) remains the same, as do the equations (4) for bulk amplitudes. Equations (24) lead to the complex conjugate of equation (18), *i.e.* to the same equation (19) and the same energy for the edge states, as expected. Ribbons with armchair edges do not exhibited edge states, ribbons with horseshoe edges can be treated in likewise manner; a non-symmetric ribbon with one zig-sag edge and another horseshoe edge implies two conditions of the type of equations (19) and (22) (two distinct purely imaginary wavevectors q), which restrict appreciably the edge states.

In conclusion we may say that semi-infinite graphene sheets with zig-zag or horseshoe edges exhibit electronic edge states within the nearest-neighbour tight-binding approximation, as do the ribbons with these same edges, while the semi-infinite graphene sheet with armchair edge does not, within the same approximation. We have assumed here an infinite length along one axis (x-axis); this condition can be removed, by considering a finite length along this axis too, as for a rectangular piece of graphene sheet. The tight-binding treatment can be conducted in this case along the same lines as described above.

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