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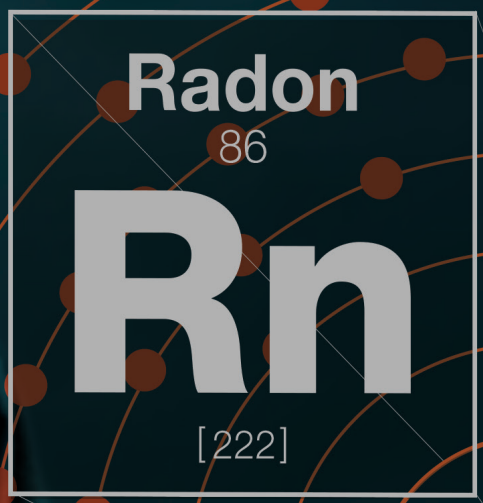

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ARTICLE

Tight-binding approximation for bulk and edge electronic states in graphene

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Abstract: The tight-binding approximation is employed here to investigate electronic bulk and edge ("surface") states in semi-infinite graphene sheets and graphene monolayer ribbons with various edge terminations (zigzag, horseshoe, and armchair edges). It is shown that edge states do not exist for a uniform hopping (transfer) matrix. The problem is generalized to include edge elements of the hopping matrix distinct from the infinite-sheet ("bulk") ones. In this case, semi-infinite graphene sheets with zigzag or horseshoe edges exhibit edge states, while semi-infinite graphene sheets with armchair edges do not. The energy of the edge states lies above the (zero) Fermi level. Similarly, symmetric graphene ribbons with zigzag or horseshoe edges exhibit edge states, while ribbons with asymmetric edges (zigzag and horseshoe) do not. It is also shown how to construct the "reflected" solutions (bulk states) for the intervening equations with finite differences both for semi-infinite sheets and ribbons.

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Résumé : Nous utilisons l'approximation des liaisons fortes pour étudier les états électroniques dans la masse et en surface (bordure) dans des feuilles semi-infinies de graphène et des rubans monocouches avec différentes terminaisons, de type zig-zag, fer à cheval ou chaise. Nous montrons que les états de surface n'existent pas pour une matrice de transfert (*hopping*) uniforme. Le problème est ici généralisé pour inclure des éléments de bordure de la matrice de transfert distincts de ceux de la feuille infinie (de masse). Dans ce cas, les feuilles semi-infinies avec bordures en zig-zag ou en fer à cheval ont des états de surface, ce que n'ont pas ceux avec bordure de type chaise. L'énergie des états de bordure–surface est au dessus du niveau (zéro) de Fermi. De même, les rubans de graphène symétriques avec bordure de type zig-zag ou fer à cheval ont des états de surface, alors que ceux avec bordures asymétriques (zig-zag ou fer à cheval) n'en ont pas. Nous montrons également comment construire les solutions réfléchies (dans la masse) pour les équations intervenantes aux différences finies, à la fois pour les feuilles semi-infinies et les rubans. [Traduit par le Rédaction]

1. Introduction

Because graphene occurs as small pieces, finite-size effects in graphene are relevant. We consider here graphene single-sheet ribbons with various edge terminations (zig-zag, horseshoe, or armchair edges). For technical rigour we consider also semi-infinite graphene sheets and, of course, the ideal case of an infinite sheet. Electronic and bonding structures of graphene have been investigated recently by various spectroscopic methods [1, 2], including a simulation of graphene edge states by photonic crystals [3].

Originally, the electronic states in (infinite) graphene sheets were studied within the tight-binding approximation [4]; massless Dirac electron states were pointed out at the corners of the Brillouin zone (zero Fermi level). It is worth emphasizing that the graphene structure consists of two identical interpenetrating triangular sublattices (honeycomb lattice) of carbon atoms.

Usually, zero-energy edge states are looked for in graphene within the tight-binding approximation [5–9], which leads to leaving aside one sublattice. Another approach is the continuum limit for the Dirac excitations with suitable boundary conditions [10–12], though the electronic elementary excitations are different from the electronic states.

The edge states in graphene are surface states [13–15] for two-dimensional solids. As is well known the tight-binding hopping (transfer) matrix elements are modified at the surface of solids. First principles calculations show that chemical modification at the edge of the graphene ribbons results in considerable changes in the bond lengths and bond angles [16, 17]. Modified edge hopping-matrix elements were considered recently in a model of graphene structure that combines the continuum limit with discrete structure [18], or in numerical studies of electronic states [9, 19, 20]. The edge modifications involve both the hopping-matrix elements and the tight-binding amplitudes. We show here that edge states in graphene may exist providing that the edge elements of the hopping matrix are different from the bulk ones. In addition, edge states exist in these circumstances only for zig-zag and horseshoe edges, with energy above the (zero) Fermi level. Making use of the well-known nearest-neighbours tight-binding technique [21–24], we derive here analytically bulk and edge states for semi-infinite graphene sheets and for (monolayer) ribbons with modified edge hopping-matrix elements.

2. Background

A typical graphene ribbon is shown in Fig. 1, exhibiting zigzag edges. The carbon atoms in the two sublattices are shown as crosses and circles in Fig. 1, the tight-binding amplitudes being denoted a_{mn} for crosses and b_{mn} for circles. The atom rows and columns are denoted m and n, respectively. We note that the units are $\sqrt{3}/2$ along the m-coordinate and 3/2 along the n-coordinate, the side of the hexagon being set equal to unity.

Leaving aside the edges, the (nearest-neighbour) tight-binding equations read

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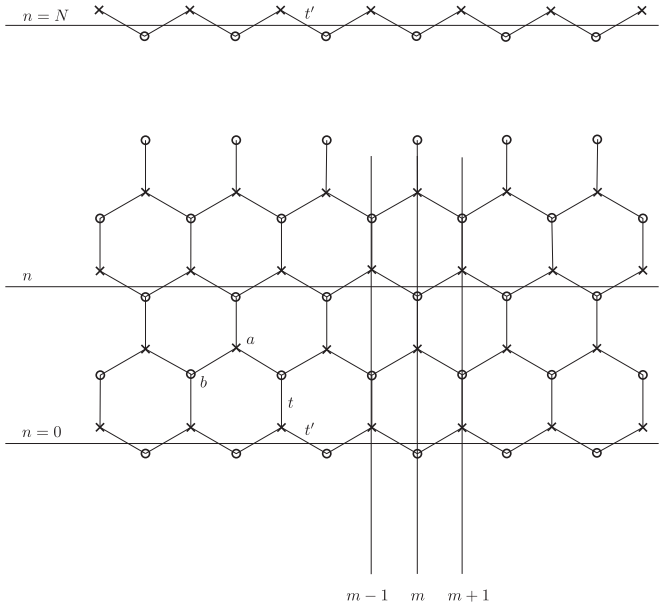
Fig. 1. Graphene ribbon with zigzag edges.



$$\varepsilon a_{mn} = t(b_{m+1n} + b_{m-1n} + b_{mn+1}) \qquad \varepsilon b_{mn} = t(a_{m+1n} + a_{m-1n} + a_{mn-1}) \tag{1}$$

where ε is the energy and t is the hopping-matrix element; we assume t to be real (phase factors can be absorbed into the amplitudes a_{mn} and b_{mn}). For travelling waves along the coordinate m we set $a_{mn} = a_n e^{ikm}$ and $b_{mn} = b_n e^{ikm}$ and (1) becomes

$$\lambda a_n = 2\cos(k) b_n + b_{n+1} \qquad \lambda b_n = 2\cos(k) a_n + a_{n-1} \tag{2}$$

where $\lambda = \varepsilon/t$. We look for solutions of the form $a_n, b_n \sim r_n$, and get

$$r = x \pm \sqrt{x^2 - 1} \qquad \lambda^2 = 4\cos^2 k + 4x\cos k + 1 \tag{3}$$

for travelling waves along the coordinate n we put $x = \cos q$ and get $r = e^{iq}$, $a_{mn} = Ae^{i(km+qn)}$, $b_{mn} = Be^{i(km+qn)}$, where

$$B = \frac{2\cos k + e^{-iq}}{\lambda} A \tag{4}$$

and $\lambda^2 = 4\cos^2 k + 4\cos k \cos q + 1$. This is a well-known result [4]. The square of the reduced energy λ^2 is positive over the whole Brillouin zone defined by the hexagon $k = \pm\pi/3$, $q = \pm\pi$ and $k = \pm 2\pi/3$, $q = 0$; it ranges from $\lambda^2 = 9$ at the centre of the Brillouin zone to $\lambda^2 = 0$ at the hexagon corners (Fermi level); nearby the corners of the Brillouin zone the energy follows $\lambda = \pm K$, where $\boldsymbol{K} = (\sqrt{3}k, q)$ is the wavevector with the origin at the hexagon's corners. This is a gapless Dirac-like electronic spectrum. Graphene sheets are zero-gap semiconductors (or zero-overlap semimetals).

Let us assume now that there exists an edge at $n = 0$, with b-atoms (the treatment is similar for a-atoms). Then, corresponding (1) reads

$$\lambda b_{m0} = a_{m+10} + a_{m-10} \tag{5}$$

which assumes the full form given in (1) providing the boundary condition $a_{mn=-1} = 0$ is satisfied. For $x = \cos q$ the solution consists of reflected waves of the form $a_n = A_1 e^{iqn} + A_2 e^{-iqn}$, $b_n = B_1 e^{iqn} + B_2 e^{-iqn}$, where

$$B_1 = \frac{2\cos k + e^{-iq}}{\lambda} A_1 \qquad B_2 = \frac{2\cos k + e^{iq}}{\lambda} A_2 \tag{6}$$

and $A_1 e^{-iq} + A_2 e^{iq} = 0$ (by the boundary condition). From (3) we may set also $x = \cosh q$, which would correspond to edge states $a_n = Ae^{-qn}$, $b_n = Be^{-qn}$ ($r = e^{-q}$, $q > 0$); unfortunately, the boundary condition $a_{n=-1} = Ae^{q} = 0$ cannot be satisfied. We conclude that edge states do not exist for a uniform hopping matrix.

There exist attempts to satisfy the boundary condition $a_{n=-1} = Ae^{q} = 0$ by setting $A = 0$ and $\lambda = 0$ (i.e., $-2\cos k = e^{q}$) such as to get a finite amplitude B from (6) [5–8]; however, the two sublattices are decoupled in this unphysical case.

We consider now a ribbon with a zigzag a-termination at $n = N$. Then, besides the previous boundary condition $a_{mn=-1} = 0$ we have also the condition $b_{mN+1} = 0$. For $x = \cos q$ we get reflected waves with amplitudes given by (6) and related, in addition, by the two boundary conditions

$$A_1 e^{-iq} + A_2 e^{iq} = 0 \qquad B_1 e^{iq(N+1)} + B_2 e^{-iq(N+1)} = 0 \tag{7}$$

Equations (6) and (7) lead to an additional constraint

$$\tan q(N+1) = -\frac{2\cos k \sin q}{2\cos k \cos q + 1} \tag{8}$$

upon the wave vector components. These equations define the bulk states in the ribbon.

The edge states in the ribbon are defined by $x = \cosh q$, $a_n = A_1 e^{-qn} + A_2 e^{qn}$, $b_n = B_1 e^{-qn} + B_2 e^{qn}$, and (6), which become

$$B_1 = \frac{2\cos k + e^{q}}{\lambda} A_1 \qquad B_2 = \frac{2\cos k + e^{-q}}{\lambda} A_2 \tag{9}$$

(where $\lambda^2 = 4\cos^2 k + 4\cosh q \cos k + 1$). The two boundary conditions give

$$A_1 e^{q} + A_2 e^{-q} = 0 \qquad B_1 e^{-q(N+1)} + B_2 e^{q(N+1)} = 0 \tag{10}$$

which, together with (9), led to $2\cos k \sinh q(N+2) + \sinh q(N+1) = 0$ ((8) for $q \to iq$). This latter equation has no solution, which means that in graphene ribbons with zigzag edges and uniform hopping-matrix elements there are no edge states. We shall see later that this is a more general conclusion.

An interesting question in this context may relate to the change brought in the density of states by a change occurring at the edges of an infinite sheet. For an infinite sheet with N_1 rows and N_2 columns, $N_{1,2} \to \infty$, we may use cyclic boundary conditions $k = (2\pi/N_1)n_1$, $q = (2\pi/N_2)n_2$, with n_1 and n_2 integers, and get the density of states $\rho = (N_1 N_2/4\pi^2)\int dS_\varepsilon 1/|\mathrm{grad}_K \varepsilon|$, where $K = (k, q)$ and the integration is performed over the curve given by equation $\varepsilon = t\lambda(k, q) = t(4\cos^2 k + 4\cos k \cos q + 1)^{1/2}$. A small change in the boundary conditions in this case amounts to a change of the form δk, $\delta q \simeq (2\pi/N_{1,2})$ in the wavevector, which leads to a relative change $\delta\rho/\rho \sim 1/N_{1,2}$ in the density of states; as is well known, this is a non-thermodynamic change, which can be neglected [25, 26]. Such a technique can be used to estimate surface quantities.

3. Ribbons and semisheets

We consider now a graphene ribbon with zigzag edges and distinct hopping-matrix elements at the edges, as shown in Fig. 1. We represent the modified hopping-matrix elements as $t' = (1 + \sigma)t$,

where σ is a parameter. The tight-binding equations for the bulk sites remain the same as (1), while the equations corresponding to the edges read now

$$\begin{aligned}\lambda a_{m0} &= (1+\sigma)\left(b'_{m+10}+b'_{m-10}\right)+b_{mn+1}\\ \lambda b'_{m0} &= (1+\sigma)(a_{m+10}+a_{m-10})\end{aligned} \tag{11}$$

and

$$\begin{aligned}\lambda a'_{mN} &= (1+\sigma)(b_{m+1N}+b_{m-1N})\\ \lambda b_{mN} &= (1+\sigma)\left(a'_{m+1N}+a'_{m-1N}\right)+a_{mN-1}\end{aligned} \tag{12}$$

we note that the edge amplitudes are also modified. We define $(1+\sigma)b'_{m0}=b_{m0}$ and $(1+\sigma)a'_{mN}=a_{mN}$, and notice that the edge equations acquire the canonical form of the bulk sites providing two boundary conditions

$$\begin{aligned}[(1+\sigma)^2-1](a_{m+10}+a_{m-10}) &= a_{mn=-1}\\ [(1+\sigma)^2-1](b_{m+1N}+b_{m-1N}) &= b_{mN+1}\end{aligned} \tag{13}$$

are fulfilled. Following the procedure described in the previous section we find solutions of the form $a_{mn}e^{ikm}a_n$, $b_{mn}=e^{ikm}b_n$, with energy given by $\lambda^2=4\cos^2k+4x\cos k+1$, where $x=\cos q$ for bulk states and $x=\cosh q$ for edge states. The bulk states are given by $a_n=A_1e^{iqn}+A_2e^{-iqn}$, $b_n=B_1e^{iqn}+B_2e^{-iqn}$, where

$$B_1=\frac{2\cos k+e^{-iq}}{\lambda}A_1 \qquad B_2=\frac{2\cos k+e^{iq}}{\lambda}A_2 \tag{14}$$

(see (6)); the two boundary conditions (13) become

$$\begin{aligned}2\sigma(2+\sigma)\cos k(A_1+A_2) &= A_1e^{-iq}+A_2e^{iq}\\ 2\sigma(2+\sigma)\cos k\left(B_1e^{iqN}+B_2e^{-iqN}\right) &= B_1e^{iq(N+1)}+B_2e^{-iq(N+1)}\end{aligned} \tag{15}$$

Equations (14) and (15) lead to the condition

$$\begin{aligned}(2\cos k+e^{iq})[2\sigma(2+\sigma)\cos k-e^{-iq}]^2e^{-iqN}\\ =(2\cos k+e^{-iq})[2\sigma(2+\sigma)\cos k-e^{iq}]^2e^{iqN}\end{aligned} \tag{16}$$

We can see that this equation reduces to (8) for $\sigma=0$; it describes bulk states. For edge states we replace q by iq in (16), which becomes

$$\begin{aligned}(2\cos k+e^{-q})[2\sigma(2+\sigma)\cos k-e^{q}]^2\\ =(2\cos k+e^{q})[2\sigma(2+\sigma)\cos k-e^{-q}]^2e^{-2qN}\end{aligned} \tag{17}$$

in contrast with the case $\sigma=0$ ($2\cos k\sinh q(N+2)+\sinh q(N+1)=0$), (17) with $\sigma\neq0$ has solutions.

For $N\gg1$ (17) reduces to

$$2\sigma(2+\sigma)\cos k-e^{q}=0 \tag{18}$$

which corresponds to a semi-infinite sheet with zigzag edge (factor $2\cos k+e^{-q}$ cannot be set equal to zero, because that would imply $\lambda=0$ and $A_1=0$ in (14)). Equation (18) admits solutions for $|2\sigma(2+\sigma)|>1$ ($-1-\sqrt{2}/2<\sigma<-1+\sqrt{2}/2$, except for $\sigma=-1$, or $\sigma<-1-\sqrt{6}/2$, $\sigma<-1+\sqrt{6}/2$). The energy of these edge states is given by

$$\begin{aligned}\lambda^2&=4\cos^2k+4\cosh q\cos k+1\\ &=\left[1+\frac{1}{\sigma(2+\sigma)}\right]\left[1+\frac{1}{\sigma(2+\sigma)}e^{q}\right]\end{aligned} \tag{19}$$

for $0<q<(2/3)\ln|2\sigma(2+\sigma)|$; for each value of q there exist two values of k in the Brillouin zone that satisfy (18). The energy given by (19) lies above the (zero) Fermi level ($\lambda^2>0$). The lowest energy values ($\lambda^2\simeq0$) correspond to the unphysical case $\sigma\simeq-1$.

The conditions given here for the occurrence of the edge states (for $N\gg1$) amount to $|t'/t|=|1+\sigma|<\sqrt{2}/2$, ($t'\neq0$) and $|t'/t|>\sqrt{6}/2$. We can see that the edge states appear for a finite change $t\to t'$, the values close to unity for the ratio t'/t not being allowed. In other words, the edge states appear for any t', except for $t'=0$ and $\sqrt{2}/2<|t'/t|<\sqrt{6}/2$.

It is worth analyzing (16) for bulk states in the presence of the modified edge hopping-matrix elements, which generate edge states. Equation (16) can be put in the form

$$\tan q(N+1)=-2\cos k\sin q\frac{M}{N+R} \tag{20}$$

where

$$\begin{aligned}M&=1+s-s^2\cos k(\cos k-\cos q)\\ N&=2\cos k\cos q+1-2s\cos k(2\cos k+\cos q)\\ R&=s^2\cos^2k(2\cos k\cos q+\cos 2q)\end{aligned} \tag{21}$$

and $s=2\sigma(2+\sigma)$. For $s=0$ this equation becomes (8), whose (approximate) solutions are of the form $q\sim1/N$ for large N; it is worth noting that they are practically independent of k (as expected in this limit). In the same limit $N\gg1$, the graphical representation of (21) shows that the solution is hardly sensitive to parameter s, so that we may take an approximate solution for non-modified edges given by (8). It follows that, practically, the density of bulk states (as given by (11)) is not modified by the edge states (and the modified edge hopping-matrix elements, which generate edge states), as expected for $N\gg1$. The density of edge states can be computed from (19); it is given by (for each value of q there exist two values of k that satisfy (18))

$$\rho_e=\frac{2N}{\pi}\varepsilon\left\{\varepsilon^2-\left[1+\frac{1}{\sigma(2+\sigma)}\right]t^2\right\}^{-1} \tag{22}$$

as expected, it is proportional to N (not N^2; and it vanishes in the thermodynamic limit).

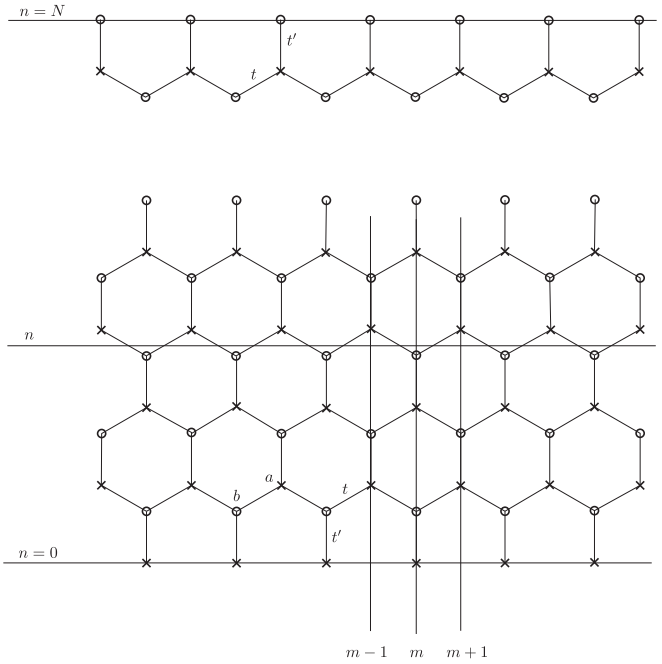
In (17), the continuum limit $q\to0$ near the (zero) Fermi level $k=\pm2\pi/3$ leads to

$$\frac{\sqrt{3}k\pm q}{\sqrt{3}k\mp q}=e^{-2qN} \tag{23}$$

which is, formally, the typical condition for edge states for massless Dirac fermions in graphene [10–12]. However, the electronic elementary excitations (envelope wavefunctions) are different from the electronic states. Elementary excitations are small variations of the already established electronic states, boundary conditions, and edge states included.

The calculations are similar for a graphene ribbon with horseshoe edges, shown in Fig. 2, though this is a rather unrealistic situation, because the dangling bonds terminate usually with hydrogen, which does not contribute to electronic states [27]. The boundary conditions in this case are

Fig. 2. Graphene ribbon with horseshoe edges.



$$a_{m+1N} + a_{m-1N} = [(1+\sigma)^2 - 1]a_{mN-1}$$
$$b_{m+10} + b_{m-10} = [(1+\sigma)^2 - 1]b_{m1} \tag{24}$$

leading to the condition

$$(2\cos k + e^{-iq})[\sigma(2+\sigma)e^{iq} - 2\cos k]^2 e^{-iqN} = (2\cos k + e^{iq})[\sigma(2+\sigma)e^{-iq} - 2\cos k]^2 e^{iqN} \tag{25}$$

For $\sigma = 0$ and $q \to iq$ we get $2\cos k \sinh q\,(N+2) + \sinh q\,(N+1) = 0$, which has no solution: edge states do not exist for horseshoe edges and a uniform hopping matrix. For $N \gg 1$ (semi-infinite sheet) the edge states are given by equation

$$\sigma(2+\sigma)e^{-q} - 2\cos k = 0 \tag{26}$$

which has two solutions for k for each value of $q > (2/3)\ln|(1/2)\sigma(+\sigma)|$, provided $\sigma < -1 - \sqrt{3}$ or $\sigma > -1 + \sqrt{3}$ and for any value of $q > 0$ otherwise ($\sigma \neq 0, -1$). The energy is given by

$$\lambda^2 = 4\cos^2 k + 4\cosh q \cos k + 1 = [1 + \sigma(2+\sigma)][1 + \sigma(2+\sigma)e^{-2q}] \tag{27}$$

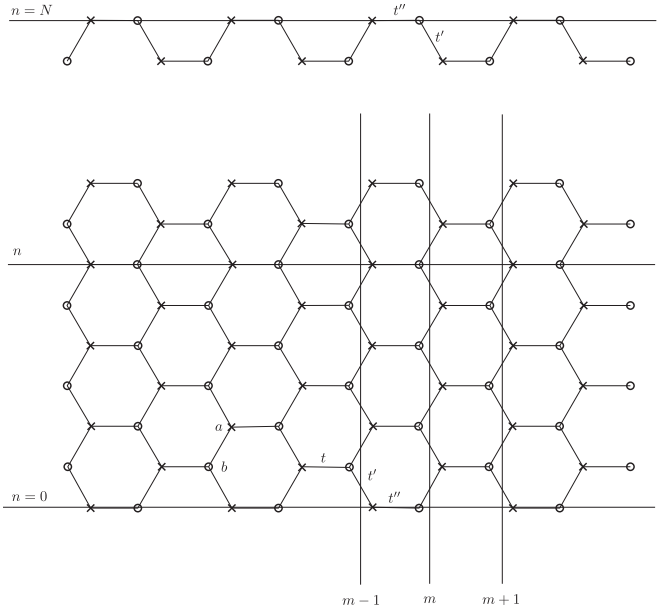
which is above the (zero) Fermi level ($\lambda^2 > 0$).

An asymmetric ribbon with one zigzag edge and the other horseshoe requires the boundary conditions

$$b_{m+10} + b_{m-10} = [(1+\sigma)^2 - 1]b_{m1}$$
$$[(1+\sigma)^2 - 1](b_{m+1N} + b_{m-1N}) = b_{mN+1} \tag{28}$$

which, together with (14), lead to two equations to be satisfied by the pair (k, q); these equations have no solutions (except, possibly, a few isolated points). We conclude that graphene sheets with zigzag or horseshoe edges exhibit edge states only for nonuniform hopping-matrix elements, while graphene ribbons with asymmetric edges (zigzag and horseshoe) do not exhibit bulk, nor edge, states.

Fig. 3. Graphene ribbon with armchair edges.



We consider a graphene ribbon with armchair edges, as shown in Fig. 3. The modified edge elements of the hopping matrix are represented by $t' = (1+\sigma)t$ and $t'' = (1+\rho)t$, where σ and ρ are parameters. The boundary conditions are given by

$$\sigma(2+\sigma)a_{m(1,N-1)} + \rho a_{m-1(0,N)} = a_{m(-1,N+1)}$$
$$\sigma(2+\sigma)b_{m(1,N-1)} + \rho b_{m+1(0,N)} = b_{m(-1,N+1)} \tag{29}$$

We seek solutions of the form $a_{mn} = e^{ikm}(A_1 e^{iqn} + A_2 e^{-iqn})$, $b_{mn} = e^{ikm}(B_1 e^{iqn} + B_2 e^{-iqn})$. The tight-binding equations for the bulk sites give

$$B_{1,2} = \frac{2\cos q + e^{-ik}}{\lambda} A_{1,2} \qquad \lambda^2 = 4\cos^2 q + 4\cos k \cos q + 1 \tag{30}$$

We can see that we have four unknowns ($A_{1,2}$, $B_{1,2}$) and six equations ((29) and (30)); in general, these equations have no solution. In the hypothetical case $\rho = 0$ ($t'' = t$), (29) are degenerate, and we have bulk reflected waves. For a semi-infinite sheet with armchair edge we get the equation

$$\sigma(2+\sigma)e^{-q} + \rho e^{ik} - e^{q} = 0 \tag{31}$$

which has no solution. We conclude that graphene sheets with armchair edges do not exhibit edge states.

4. Concluding remarks

We have shown here that a uniform hopping matrix gives no edge states for graphene monolayer ribbons or semi-infinite sheets within the nearest-neighbour tight-binding approximation. To get edge states it is necessary to introduce edge hopping-matrix elements distinct from the bulk ones. In this case, zigzag or horseshoe edges exhibit edge states, while armchair edges do not. Similarly, ribbons with asymmetric edges (one zigzag, the other horseshoe) do not have edge states. Other previous results related to edge states in graphene have also been discussed in the present paper.

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