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Persistent currents in carbon nanotubes

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Abstract

Persistent currents driven by a static magnetic flux parallel to the carbon nanotube axis are investigated. Owing to the hexagonal symmetry of graphene the Fermi contour expected for a 2D lattice reduces to two points. However, the electron or hole doping shifts the Fermi energy upwards or downwards and, as a result, the shape of the Fermi surface changes. Such a hole doping leading to the Fermi level shift of (more or less) 1 eV has been recently observed experimentally. In this Letter we show that the shift of the Fermi energy changes dramatically the persistent currents and discuss the electronic structure and possible currents for zigzag as well as armchair nanotubes. © 2002 Published by Elsevier Science B.V.

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1. Introduction

There is a considerable research activity to explain physical properties of single-wall and multi-wall carbon nanotubes. Their remarkable magnetic and electrical properties stem from the unusual electronic structure of the graphene sheets—the quasi-2D material from which they are made. The influence of magnetic field on transport properties of carbon nanotubes has been explored, both in theory [1–3] and in experiment [4–6]. In our Letter we consider the influence of the magnetic field on persistent currents and structural properties of these materials induced by their topol-

ogy. The strong dependence of the band structure on the magnetic field suggests large orbital magnetic response. Indeed, large magnetic susceptibilities were calculated and experimentally observed [7,8] for magnetic fields both perpendicular and parallel to the tube axis. These results suggest the existence of persistent currents.

In a nanotube the component of the momentum along the circumference of the tube is quantized owing to the periodic boundary conditions. This raises the possibility of inducing persistent currents along the circumference of the nanotube by a static magnetic field applied parallel to the tube axis. They arise due to Aharonov–Bohm effect. Such currents, but in toroidal nanotubes, have been recently discussed theoretically [9,10].

Persistent currents in nanotubes bear some resemblance with those in metallic and semiconducting

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rings or cylinders. It is well known that persistent currents in mesoscopic cylinders depend strongly on the correlation of currents from different channels [11] and on the shape of Fermi surface [12]. The magnitude of the currents increases with increasing curvature of the Fermi surface. The current is strongest for the Fermi surface having large flat regions perpendicular to the direction in which the magnetic field changes the wave vector \mathbf{k} .

The shape of the Fermi contour expected for a 2D hexagonal lattice depends strongly on the band filling. For an undoped carbon nanotube $E_F = 0$ and the Fermi surface reduces to two isolated Fermi points instead of a whole contour. Electron or hole doping shifts the Fermi energy upwards or downwards. As a result, the shape of the Fermi surface changes and this influences the persistent currents. A substantial Fermi level shift of (more or less) 1 eV has been recently obtained [13] by the means of an electrochemical gating.

In the present Letter we study persistent currents induced by a static magnetic field parallel to the axis of a single wall carbon nanotube. We investigate the magnitude and shape of the current as well as their dependence on the hole doping. In particular, we show that for zigzag nanotubes, for certain values of the hole doping, we can obtain a large enhancement of the persistent current. Finally, we briefly discuss the possibility of creating self-sustaining currents in multiwall nanotubes.

2. The structure of a nanotube

The nanotube is a rolled up strip of a graphene sheet. The rolling vector is $\mathbf{L}_t = m_1 \mathbf{T}_1 + m_2 \mathbf{T}_2$ (the t index stands for transverse direction), where \mathbf{T}_1 and \mathbf{T}_2 form the standard basis vectors of the graphene lattice [14]. The vector of the length of the nanotube is $\mathbf{L}_l = p_1 \mathbf{T}_1 + p_2 \mathbf{T}_2$ (the l index stands for longitudinal direction). The four integers $(m_1, m_2) \times (p_1, p_2)$ therefore define the geometry of the tube. The number of lattice nodes of the nanotube is $N = m_1 p_2 - m_2 p_1$. Note that \mathbf{L}_t and \mathbf{L}_l do not have to be orthogonal (the nanotube is then twisted [14]) but for simplicity we always take in our Letter the twist angle to be 0, i.e., m_1, m_2, p_1, p_2 such that they are orthogonal.

The wave vector $\mathbf{k} = (k_x, k_y)$ of an electron on a surface of a 2D cylindrical tube obeys two boundary conditions. The first, periodic along the circumference of the nanotube, is

$$\mathbf{k} \cdot \mathbf{L}_t = k_t |\mathbf{L}_t| = 2\pi l'_t, \quad (1)$$

where, in the absence of magnetic field, l'_t is an integer. The second boundary condition, which results in vanishing of the sinusoidal wave function at the ends of the nanotube, is

$$\mathbf{k} \cdot \mathbf{L}_l = k_l |\mathbf{L}_l| = \pi l_l, \quad (2)$$

where l_l is an arbitrary integer. The first Brillouin zone of the whole graphene sheet is a hexagon whose vertices are $(\pm 2\pi/3\sqrt{3}, \pm 2\pi/3)$ and $(0, \pm 4\pi/3)$. The wave functions with l_l and $-l_l$ are linearly dependent and therefore the first Brillouin zone of the nanotube reduces to a half of the original hexagon. In our choice l_l takes only positive values.

The energy of the \mathbf{k} th state of the hexagonal lattice, in the tight binding approximation, is well known [14] to be

$$E_{\mathbf{k}} = \pm \gamma \sqrt{1 + 4 \cos^2 \frac{\sqrt{3}}{2} k_x + 4 \cos \frac{\sqrt{3}}{2} k_x \frac{3}{2} k_y}, \quad (3)$$

where γ is the resonance integral for the nearest neighbour interaction (we take $\gamma = 3.033$ eV, typical for the carbon hexagonal lattice [9]). The energy may change from -3γ to 3γ . At the half-filling, when half of the possible states are occupied, the Fermi level is at $E_F = 0$ and the Fermi surface reduces to isolated points—vertices of the original hexagon. Only two of them, e.g., $K_1 = (-2\pi/3\sqrt{3}, 2\pi/3)$ and $K_2 = (2\pi/3\sqrt{3}, 2\pi/3)$, are linearly independent and therefore define the whole Fermi surface of the nanotube at $E_F = 0$. When less than a half of the states are occupied (the nanotube is hole-doped), the Fermi level lowers and becomes a set of curves (drawn as continuous bold lines in Figs. 2–4(a) and Fig. 6(a)). At $E_F = -\gamma$ the Fermi surface is a half-hexagon whose vertices are at the centre of edges of the first Brillouin zone (cf. Fig. 3(a)). In this case the number of occupied states N_e is equal to 3/4 of the half-filling, i.e., $N_e = (3/4)N$.

After applying the magnetic field along the axis of the nanotube, all the wave functions acquire the Aharonov–Bohm phase factor $e^{i2\pi\phi/\phi_0}$, where ϕ is the magnetic flux through the nanotube and $\phi_0 = hc/e$ is

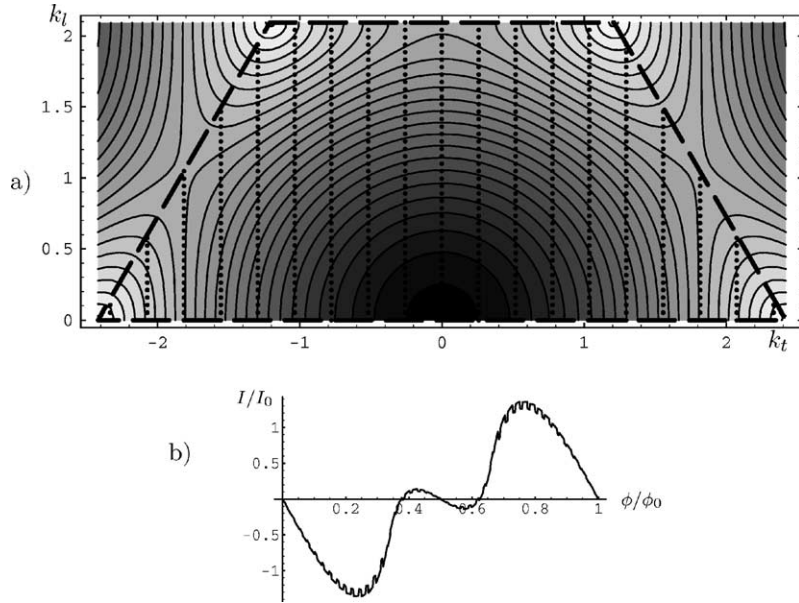


Fig. 1. (a) The Brillouin zone of a $(14, 0) \times (-100, 200)$ nanotube at $\phi = 0$ and $N_e = 2800$ (half-filling). The dotted lines represent different $k_t = \text{const}$ momentum lines. The shading in the background is the contour plot of the $E(\mathbf{k})$ relation (3). The dashed line is the edge of the Brillouin zone. (b) The persistent current $I(\phi/\phi_0)/I_0$.

the flux quantum. This phase factor is equivalent to a shift in the momentum quantum number l'_t ,

$$l'_t = l_t + \frac{\phi}{\phi_0}, \tag{4}$$

where l_t is an arbitrary integer. The magnetic field induces persistent current along the transverse direction of the nanotube

$$I_{\mathbf{k}}(\phi) = -c \frac{\partial E_{\mathbf{k}}}{\partial \phi}. \tag{5}$$

Summing over all momentum states within the first Brillouin zone and below the Fermi level, we obtain the overall current, induced by the magnetic flux:

$$I(\phi) = \sum_{\mathbf{k}_{\text{occupied}}} I_{\mathbf{k}}(\phi). \tag{6}$$

When all the shifted states remain within the Brillouin zone and below the Fermi level, the current is diamagnetic. If, due to the shift induced by ϕ , some states leave the Brillouin zone or cross the Fermi level, and others enter it or cross the Fermi level in the opposite direction, we witness a paramagnetic jump of the current [15]. This total current depends also on the temperature which suppresses its amplitude [12,15], but for simplicity we assume here that $T = 0$.

3. Persistent currents in zigzag nanotubes

We have performed numerical calculations for nanotubes of various geometries. First we discuss the case of a $(14, 0) \times (-100, 200)$ nanotube. The Brillouin zone, at zero magnetic field, for this nanotube is shown in Fig. 1(a). It is equivalent to the upper (or lower) half of the original Brillouin zone of the graphene sheet [14].

In the half-filling case ($E_F = 0$) there is one conduction electron per each lattice node, i.e., $N_e = N$ (in our case $N = 2800$). This nanotube is not metallic and there are no allowed momentum states in any of the Fermi points (the condition for metallicity of the tube is $(m_1 - m_2) \bmod 3 = 0$). The persistent current in units of $I_0 = \gamma c / \phi_0 = 1.29 \times 10^{-4}$ A, induced by the magnetic field is plotted in Fig. 1(b). The current is periodic with period ϕ_0 and diamagnetic at small ϕ .

As Ref. [13] shows, the Fermi energy can be lowered by binding some of the electrons (hole doping) and then $N_e < N$. In the following we assume that the doping lowers the number of electrons to a constant N_e , which does not depend on ϕ . Therefore, as the distribution of the momentum states in the Brillouin zone changes with ϕ , the value of E_F must be adjusted

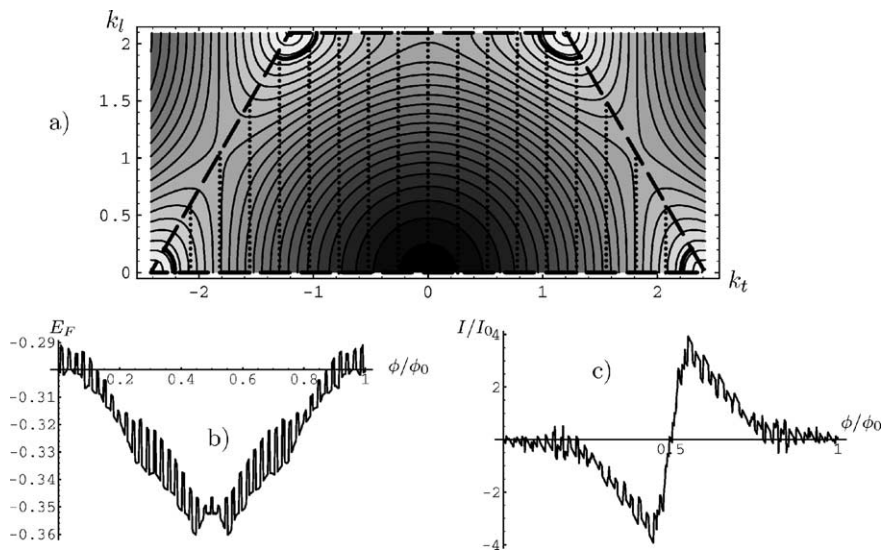


Fig. 2. (a) The Brillouin zone and Fermi surface, (b) the $E_F(\phi/\phi_0)$ plot and (c) the $I(\phi/\phi_0)$ for a $(14, 0) \times (-100, 200)$ nanotube at $N_e = 2744$, i.e., the Fermi energy lowered by about $\gamma/3$. The continuous bold contour in (a) is the Fermi surface at $\phi = 0$.

to keep N_e independent of ϕ . This is not the case for $E_F = 0$, where the number of states in the first Brillouin zone is constant ($N_e = N$) and does not depend on ϕ .

Fig. 2(a) shows the reciprocal lattice and the Fermi contour (bold line) for $N_e = 2744$ at $\phi = 0$. It corresponds to the Fermi energy shift of approximately -1 eV (or $\sim -\gamma/3$), which has been obtained experimentally [13]. The change of Fermi energy with ϕ/ϕ_0 is shown in Fig. 2(b). We see that the oscillations are around $-\gamma/3$ and have quite a substantial amplitude of approximately 0.07γ . The induced persistent current (Fig. 2(c)) has now four times higher amplitude and a different shape.

The most favourable situation for the enhancement of the current is when the Fermi level is at the value of $-\gamma$. It corresponds to the number of electrons $N_e = (3/4)N$. The Fermi surface then becomes the smaller half-hexagon in the first Brillouin zone (cf. Fig. 3(a)), with the momentum lines parallel to its vertical edges. The oscillations of the Fermi level (shown in Fig. 3(b)) are three orders of magnitude smaller compared to the previous case of $N_e = 2744$. At $\phi = 0$, a whole momentum line crosses the Fermi surface with the change of ϕ , which results in a large paramagnetic jump of the current (Fig. 3(c)).

The current's amplitude is 25–30 times higher compared with the current at the half-filling. This is the largest persistent current possible to obtain in this nanotube. By further shifting the Fermi level downwards, the Fermi surface becomes curved and again the amplitude of the current gradually decreases.

The amplitude of persistent currents, at the most favourable value of $3/4$ of half-filling, depends on the length and width of the tube. It is proportional to its length and decreases with its width. The reason for this is that the longer the tube is, the more dense are the states in the longitudinal direction, therefore, when a line of them crosses the Fermi surface, the effect is proportionally greater. On the other hand, the wider the tube is, the smaller are individual currents:

$$I_{\mathbf{k}} = -c \frac{\partial E_{\mathbf{k}}}{\partial \phi} = -c \frac{\partial E_{\mathbf{k}}}{\partial k_t} \frac{2\pi}{\phi_0 |\mathbf{L}_t|}. \quad (7)$$

Let us notice also what happens if the m_1 parameter of the nanotube (the one which determines its diameter) changes. If m_1 is even, as in the previously discussed case, there is an even number of $k_t = \text{const}$ state lines inside the Fermi contour for $N_e = (3/4)N$ (as in Fig. 3(a)) and the paramagnetic jump of the current (Fig. 3(c)) occurs at integer values of ϕ/ϕ_0 . If m_1 is odd, the number of $k_t = \text{const}$ state lines inside

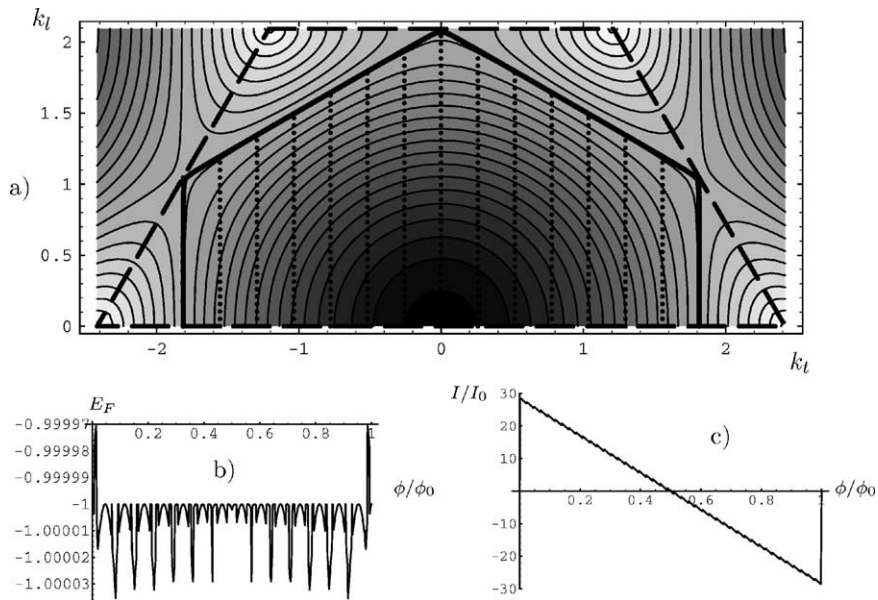


Fig. 3. (a) The plot represents the Brillouin zone and the Fermi surface (bold contour) of a $(14, 0) \times (-100, 200)$ nanotube at $N_e = 2100 = (3/4)N$; (b) is the plot of Fermi energy vs. magnetic flux; (c) is the resulting persistent current vs. magnetic flux.

the Fermi contour is odd and the paramagnetic jump of the current occurs at half-integer values of ϕ/ϕ_0 . We investigated such a situation and the result for a $(15, 0) \times (-100, 200)$ nanotube is illustrated in Fig. 4. Again, a strongly enhanced current is obtained, but this time it is diamagnetic at small ϕ . This behaviour of the current bears a strong resemblance to what happens in a one-dimensional metallic ring: the currents like the one in Fig. 3(c) (“even” nanotube) occur if the number of electrons in the ring is even and the case from Fig. 4(c) (“odd” nanotube) takes place for an odd number of electrons [15]. This analogy arises from the equivalent positions of the Fermi level with respect to the last occupied state (line of states) in these two systems.

4. Persistent currents in armchair nanotubes

At the half-filling, the principal difference between nanotubes of different chiralities is whether they are metallic or semi-conducting. We know that if the nanotube parameters obey the relation $(m_1 - m_2) \bmod 3 = 0$, the tube is metallic, otherwise it is semiconducting. This criterion follows from the specific structure

of the Brillouin zone at half-filling, and is not valid for $E_F \neq 0$. At lower, or higher, band filling all the nanotubes are more likely to be metallic because when the Fermi points become lines, the probability that some states lie on them increases.

The important feature is the shape of the tube’s momentum spectrum. The chirality of the tube varies from $(m, 0)$ for zigzag nanotubes (where the momentum lines are parallel to the sides of the smaller, $E_F = -\gamma$ hexagon) to (m, m) for armchair nanotubes (where they are parallel to one of the sides of the Brillouin zone). We now take the case of a $(9, 9) \times (-150, 150)$ nanotube which is shown in Fig. 5. Its length and width are similar to the $(14, 0) \times (-100, 200)$ tube and the number of electrons in the half-filling case is $N_e = N = 2700$. The momentum lines are parallel to the sides of the Brillouin zone and the resulting current (Fig. 5(b)) has amplitude of the order of $2I_0$.

The current in this case is much smaller compared to the similar distribution of k -states within the allowed region in Fig. 3(a). The reason for the attenuation of the current in this case is that the individual currents (7) corresponding to the states at the edge of the Brillouin zone are much smaller than the currents

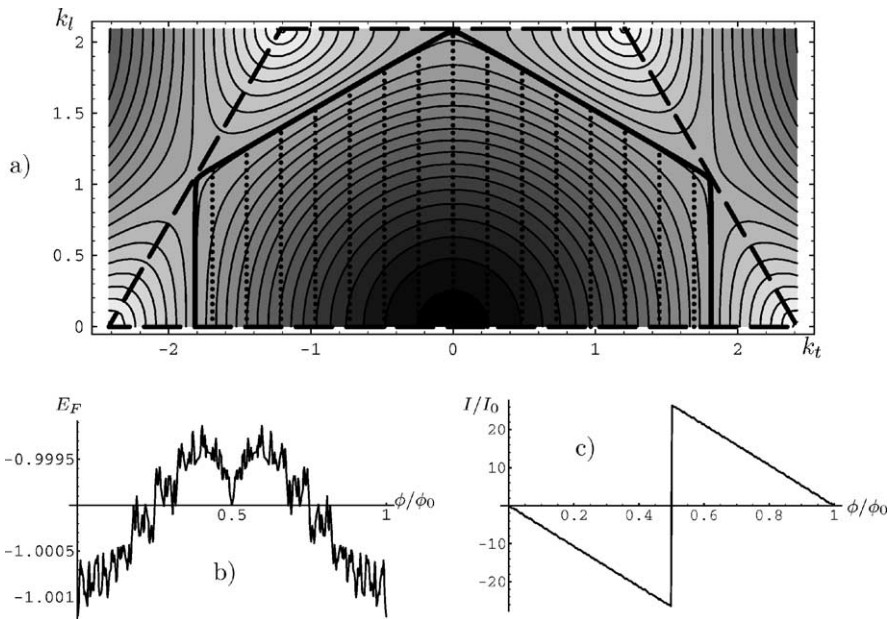


Fig. 4. The plots are all for a nanotube at $N_e = 3/4N_0$. (a) The Brillouin zone and the Fermi surface (bold line) for a $(15, 0) \times (-100, 200)$ nanotube at $\phi = 0$ —note that there is no momentum line at the Fermi level. (b) The change of Fermi energy with ϕ . (c) The resulting persistent current—note that the jump occurs at $\phi/\phi_0 = 1/2$.

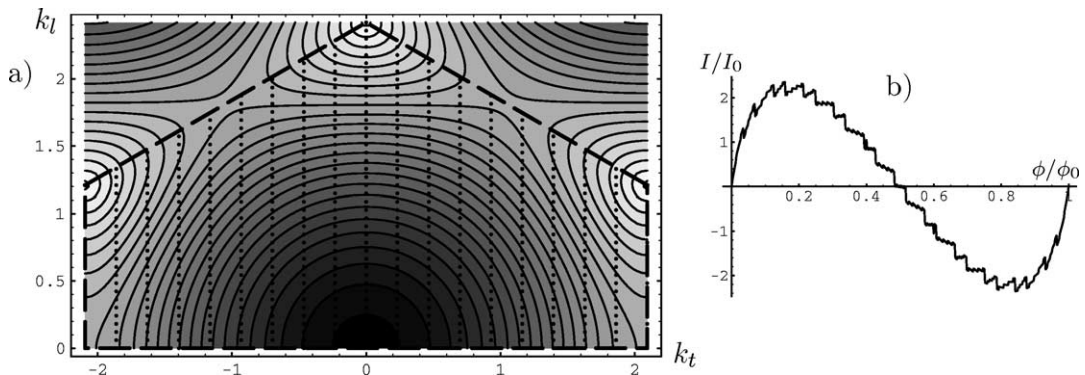


Fig. 5. (a) The Brillouin zone and momentum states for a $(9, 9) \times (-150, 150)$ nanotube at half-filling, i.e., $N_e = 2700$. (b) Persistent currents vs. magnetic flux, everything at constant number of electrons.

carried by the states at the edge of the Fermi surface. This can be seen by observing the equienergy lines at these edges (Figs. 3(a) and 5(a)).

Decreasing the number of electrons or lowering the Fermi level does not enhance the persistent currents in this case. The reason for that is that the momentum lines $k_t = \text{const}$ are not parallel to any fragment of the Fermi surface. An example of such a situation

is shown in Fig. 6, where $N_e = (3/4)N$. Both the attenuation and the oscillations of the current, as compared to the half-filling case, are caused by the lack of correlation between the states which now travel across the Fermi surface separately.

In real systems, the temperature $T > 0$ would smooth down the oscillations shown in Fig. 6(c) and the resulting current would be very small.

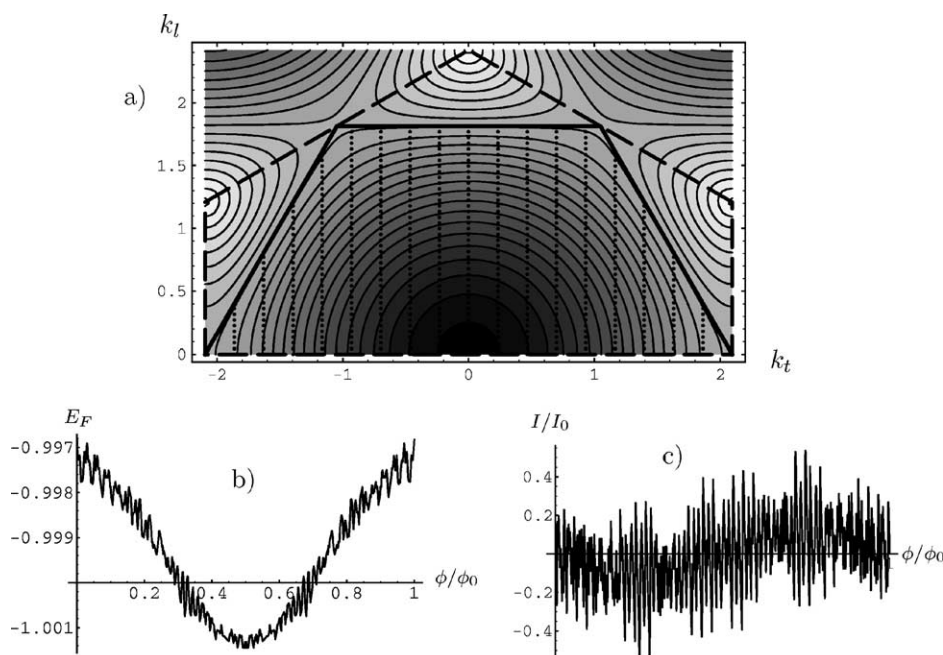


Fig. 6. (a) Brillouin zone and the Fermi surface of a $(9, 9) \times (-150, 150)$ nanotube at $N_e = (3/4)N$. (b) $E_F(\phi/\phi_0)$ plot. (c) $I(\phi/\phi_0)$.

5. Conclusions

The presence of quantized, delocalized electron states in carbon nanotubes should result in persistent currents. In carbon nanotubes, which are typically almost free of defects, these currents should be substantial and can change with doping. We have investigated the changes of the shape of the Fermi surface with the hole doping and discussed the respective changes of persistent currents in zigzag as well as in armchair nanotubes.

The currents are calculated under the assumption that the number of electrons is constant in the nanotube. They can be paramagnetic or diamagnetic depending on the position of states in the first Brillouin zone and the shape of the Fermi surface. We have shown that the largest currents can be obtained if the Fermi energy is lowered from $E_F = 0$ (half-filling case) to $E_F \cong -1\gamma$ ($N_e = (3/4)N$) for zigzag nanotubes. The Fermi surface changes then from two separate points to a flat contour and the induced persistent currents increase their amplitude 25–35 times. For chiral nanotubes such that $m_2 \ll m_1$ the enhancement of the persistent current at $E_F = -\gamma$ can also be large.

In armchair nanotubes the situation is different; namely, with hole doping the amplitude the persistent current decreases because the individual currents become less correlated.

The large amplitude of the obtained currents raises the possibility of obtaining self-sustaining currents which produce the magnetic flux capable to maintain themselves even in absence of external magnetic field [16]. However, spontaneous flux in single wall nanotubes will be very small— $\phi/\phi_0 \simeq 10^{-3}$ only. To discuss the problem of self-sustaining currents multiwall nanotubes are more suitable. This problem will be discussed in a forthcoming paper. Below we will only briefly mention various possible cases.

In the case of multi-wall nanotubes, where many tubes are arranged in a coaxial fashion, the currents produced by all tube layers should superpose. However, the electrical properties of individual tubes have been shown to vary strongly from tube to tube [17]. First, let us assume that all the tubes have the same Fermi energy $E_F = -\gamma$. The most favourable case is when all (or a majority of) the tubes are of the zigzag type discussed in Section 3.

If all tubes have the same parity (even or odd) the amplitude of the total current is greatest—it can be 10–

50 times larger than in a single-wall nanotube. This current is paramagnetic for even and diamagnetic for odd m_1 for small values of ϕ/ϕ_0 . In such situation there is a large probability of obtaining self-sustaining currents.

When tubes belonging to the multiwall nanotube have different chiralities, the total current will be smaller due to the cancellation between currents from different tubes. However, the total current will not average to zero, and the net paramagnetic current with period halving will be obtained. The least favourable case is when none of the tubes are of the zigzag type. The cancellation is then substantial and we obtain only a very small net current.

It seems that these novel and unusual properties of carbon nanotubes may have promising applications in areas such as the magnetoelectronics, e.g., in magnetic sensors.

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