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Aharonov-Bohm-type effect in graphene tubules: A Landauer approach

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In this paper we show that (1) for graphene tubules an axial magnetic field can induce a semimetal-semiconductor transition periodically at zero bias. This is an Aharonov-Bohm-type effect and is generally true for thin cylindrical conductors; (2) the Landauer approach can be used to estimate the conductance of individual tubules measured by scanning tunneling probes as a function of applied bias, temperature, tubule geometric factors, and magnetic field. Numerical calculations are presented showing that the conductance modulation should be observable even at room temperature.

Recently graphene tubules have been produced using an arc-discharge evaporation method similar to that used for fullerene synthesis.[1-3] A monolayer graphene tubule consists of a honeycomb network of carbon atoms, and can be imagined as a cylinder rolled from a graphite sheet with a diameter of several nanometers. The synthesis of graphene tubules has stimulated many theoretical studies on their structural and electronic properties.[4-7] Tubules with different diameter and chirality are predicted to have different energy-band structures, hence different conducting properties. About one-third of the possible tubules are semimetallic with zero band gap, while the remaining two-thirds are semiconductorlike with a nonzero band gap. The first objective of this paper is to show that an axial magnetic field can induce a semimetal-semiconductor transition periodically with a period of h/eS, S being the cross section of the tubule. This is an Aharonov-Bohm-type effect arising from the dependence of the electron wave function on the vector potential,[8] and is a general phenomenon for thin cylindrical conductors.

The most direct way to check this prediction is to use a scanning tunneling probe to measure the conductance of individual tubules. It is thus important to develop a quantitative theory for the conductance of a tubule as measured between contacts. To our knowledge, all previous theoretical work has focused on calculating the band gap. This only provides qualitative information regarding the conduction properties and does not take the contacts into account. The Landauer approach[9,10] based on calculating the transmission through a finite-size sample has proved to be very useful in interpreting experiments in mesoscopic physics involving the conductance of small structures as measured between large contacts. The second objective of this paper is to show how this approach can be used to estimate the conductance of individual carbon tubules as a function of (a) applied bias, (b) temperature, (c) tubule geometric factors, and (d) magnetic field. A numerical example is presented showing that the conductance modulation in a magnetic field should be observable even at room temperature under reasonable values of bias.

Let us first explain why we expect an axial magnetic field to induce a periodic modulation in the conductance. If there is no magnetic field then the electron eigenstates can be written in the form $\sim\exp[ik_C C]\exp[ik_L L]$, where C and L represent the position along the circumference and the axis direction, respectively. The cylindrical geometry leads to quantized values of k_C given by $k_C(2\pi R)=2n\pi$, where R is the radius of the cylinder and n is an integer. A uniform magnetic field **B** applied along the axis of a cylinder leads to a vector potential **A** pointing along the circumferential direction and having a constant magnitude of $A_0=BR/2$ everywhere on the surface of the cylinder. The resulting eigenstates are obtained simply by multiplying the zero-field eigenstates by the factor $\exp[-ieA_0C/\hbar]$. This changes the quantization condition to

$$[k_C-(eA_0/\hbar)](2\pi R)=2n\pi$$
$$\Rightarrow k_C(2\pi R)=2n\pi+(eBS/\hbar)\ . \quad (1)$$

As a result the allowed values of k_C are changed by a magnetic field leading to a modulation of the density of states and hence the conductance. However, the allowed values of k_C are the same everytime the quantity $eBS/\hbar$ changes by a multiple of 2π. So we expect the conductance to change periodically with a period equal to (h/eS). This is true even if the cross section is not exactly circular.

This phenomenon should be observable in graphene tubules using a scanning tunneling probe. There are two possible configurations, but we will only consider the configuration with the axis of the tubule perpendicular to the substrate so that the current flows parallel to the axis. The other configuration (the axis of the tubule is parallel to the substrate so that the current flows along the circumference) is similar to that studied using mesoscopic rings.[11]

We choose the carbon-carbon bond direction of the honeycomb lattice as the x axis, so that the chirality angle θ is the angle between the axis of the tubule and the x axis [see Fig. 1(a)]. For simplicity, we consider only the π band, and neglect effects caused by the finite tube curvature such as the mixing of σ and π bands and the interaction between p_z orbitals. We also neglect any Peierls distortion. The dispersion relation for the tubule can be obtained from a tight-binding calculation

$$E(\mathbf{k})=\pm t_0\sqrt{1+4\cos^2(k_y b_0)+4\cos(k_y b_0)\cos(3k_x a_0)}\ , \quad (2)$$

where t_0 is the nearest-neighbor overlap integral. a_0 and b_0 are defined in Fig. 1(a).

Equation (1) defines a set of straight lines whose slope depends on the chirality angle θ and whose density depends on the radius of the tubule R. Equation (2) is used to generate a set of constant energy contours in the k_x-k_y plane. The number of intersections (within the first Brillouin zone) of the straight lines with a particular constant energy contour gives the number of allowed transverse modes $M(E)$ at that energy [see Fig. 1(b)].[12] The effect of an axial magnetic field is simply to translate the straight lines perpendicular to their length. This changes the number of modes.

Figures 2(a) and 2(b) show $M(E,B=0)$ for a tubule

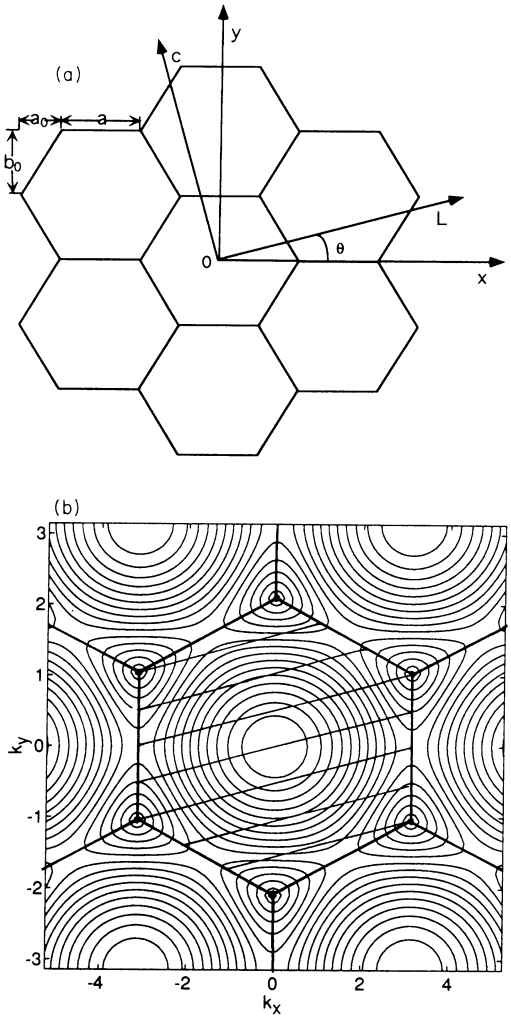

FIG. 1. (a) A two-dimensional sheet of graphite ($a=1.42$ Å). The tubule is formed by rolling the sheet into a cylinder whose axis points along $\hat{L}$. The unit vector along the circumference is denoted by $\hat{c}$. (b) Equation (1) defines a set of straight lines whose slope depends on the angle θ and whose density depends on the radius of the tubule. Equation (2) is used to generate a set of constant energy contours in the k_x-k_y plane. In the figure we show the lines for $\theta=16.1°$ and $r=2.4$ Å. The number of intersections (within the first Brillouin zone) of the straight lines with a particular constant energy contour gives the number of allowed transverse modes $M(E)$ at that energy.

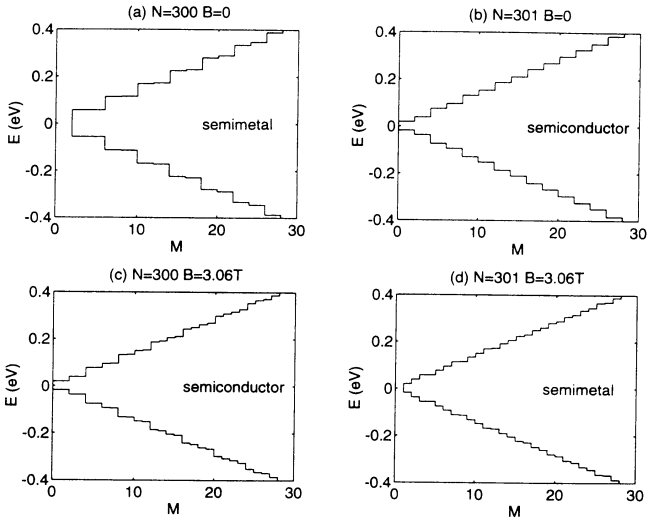

FIG. 2. The number of modes for a graphene tubule with its circumferential vector $\hat{c}$ pointing along y ($\theta=0°$) having N atoms along the circumference and a magnetic field along the axis: (a) $N=300$, $B=0$; (b) $N=301$, $B=0$; (c) $N=300$, $B=3.06$ T; (d) $N=301$, $B=3.06$ T.

having its axis along the x axis ($\theta=0$) with different numbers of atoms along the circumference ($N=300$ and 301). Whenever N is a multiple of 3, the number of modes at the Fermi energy, $M(E=0)$, is nonzero indicating a semimetal; but for other values of N, $M(E=0)$ is zero indicating a semiconductor. Tubules with other geometric factors can be analyzed in the same way. Figures 2(c) and 2(d) show the variation in $M(E=0)$ at $B=3.06$ T for $N=300$ and 301. It is clear that an axial magnetic field can turn a semimetal into a semiconductor and vice versa. The next question is whether the effect can be observed under usual experimental conditions.

The Landauer formula expresses the linear-response conductance as measured between two contacts or reservoirs. Experimental measurements using scanning tunneling probes typically involve high bias and temperature, so that a more general version of the Landauer formula is needed. This is easily done if we neglect all inelastic processes inside the tubule.[13]

We consider a single-layer graphene tubule deposited on a substrate and probed by a scanning probe [see Fig. 3(a)]. We adopt the model shown in Fig. 3(b). There is a tunneling barrier between the probe and the tubule due to the air gap. Since the nature of the contact between the tubule and the substrate is not known, we allow for a small barrier. A fraction ηeV of the applied bias is dropped across the air gap and the remainder $(1-\eta)eV$ is dropped at the tubule-substrate interface as shown in Fig. 3(c). We assume that the substrate and the probe are maintained in local equilibrium with electrochemical potentials μ_1 and μ_2, respectively. The current is given by[10,13]

$$I=\frac{2e}{h}\int dE\, M(E)t(E,V)[f(E-\mu_1)-f(E-\mu_2)]\ , \tag{3}$$

where $f(E-\mu)$ is the Fermi factor, V is the applied bias:

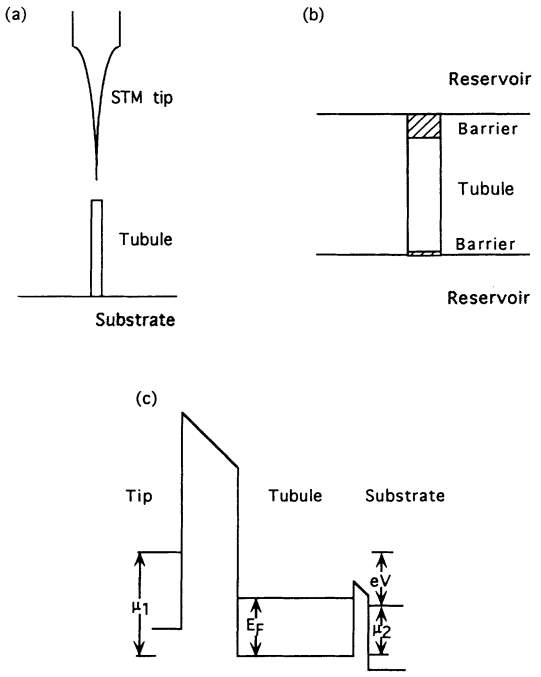

FIG. 3. (a) A graphene tubule sandwiched between the scanning tunneling microscopy tip and the substrate. (b) A model for the structure shown in Fig. 1(a). (c) Energy-band diagram where $\mu_1=E_f+eV$, $\mu_2=E_f-(1-\eta)eV$. The value of η $(0\leq\eta\leq 1)$ is determined by the ratio of the two tunneling barriers.

$\mu_1=\mu_2+eV$ and t is the average transmission probability per conducting mode.

Following Bagwell and Orlando,[13] we can express the function $[f(E-\mu_1)-f(E-\mu_2)]$ as a convolution of a bias function $[\theta(E-\mu_1)-\theta(E-\mu_2)]$ and a thermal broadening function $F_T(E)$ and rewrite Eq. (3) as

$$I(V)=\int_{\mu_2}^{\mu_1} dE\, g(E,V)\ , \tag{4}$$

where

$$g(E,V)=\frac{2e}{h}\int dE' M(E')t(E',V)F_T(E-E') \tag{5a}$$

and

$$F_T(E)=-\frac{\partial f(E)}{\partial E}=\frac{1}{4k_BT}\mathrm{sech}^2\left[\frac{E}{2k_BT}\right]\ . \tag{5b}$$

It is often more revealing to look at the conductance obtained by differentiating Eq. (4):

$$G(V)\equiv\frac{\partial I}{\partial V}=\int_{\mu_2}^{\mu_1} dE\frac{\partial g(E,V)}{\partial V}+e\eta g(\mu_1,V) +e(1-\eta)g(\mu_2,V)\ . \tag{6}$$

The first term arises from the change in the transmission $t(E,V)$ under bias. This change is primarily due to the lowering of the tunneling barrier(s) [and the consequent increase in $t(E,V)$] when an electric field is applied. Close to equilibrium this term vanishes because $\mu_1=\mu_2$. But at high bias a significant part of the conductance could be due to this term. The other two terms arise from the addition of energy channels for conduction around μ_1 and μ_2.

For quantitative calculations one could assume specific barrier heights and calculate each of the three terms. But we feel that this is not warranted since little is known experimentally about the actual barrier heights. Instead we assume that (a) the bias is low enough that the first term in Eq. (6) is negligible; (b) the voltage is dropped largely between the tip and the tubule, so that the third term is negligible. Thus Eq. (6) simplifies to

$$G(V)=\frac{2e^2}{h}\int dE\, M(E)t(E,V)F_T(E-\mu_1)\ . \tag{7}$$

At low temperatures the thermal broadening function $F_T(E)$ can be replaced by a δ function so that the conductance is given by

$$G(V)=\frac{2e^2}{h}M(\mu_1)t(\mu_1,V)\ . \tag{8}$$

Thus the conductance at low temperatures is proportional to the number of modes at an energy equal to the Fermi energy in the probe. The effect of temperature is to average the zero-temperature conductance over an energy range of several k_BT around (μ_1).

The average transmission probability $t(E,V)$ can be approximately written as the product of the probability for tunneling through the barriers and the probability for transmission through the tubule. The former could be estimated by, for example, the WKB method whereas the latter will depend on the length L of the tubule and can be written as $\lambda/(L+\lambda)$ where λ is the free path.[13] Short tubules with $L\ll\lambda$ are desirable in order to avoid conductance fluctuations associated with scatterers.

Figure 4 shows the calculated conductance of a tubule

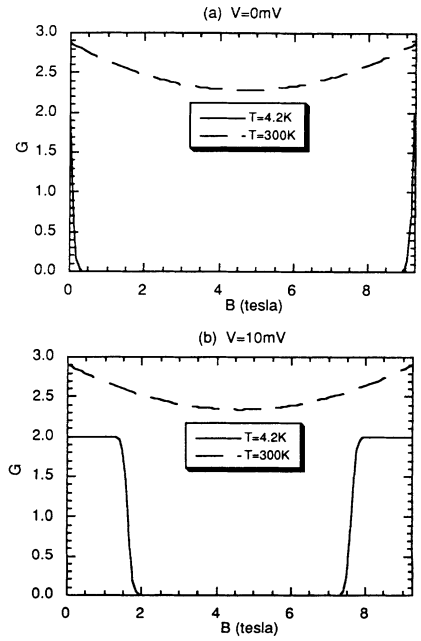

FIG. 4. The conductance of a single-layered tubule ($\theta=0°$, $N=300$, $t=$const, $\eta=1$) vs magnetic field at 4.2 and 300 K for two values of the bias voltage: (a) $V=0$ and (b) $V=10$ mV. (Conductance G is normalized to $2e^2t/h$.)

(at $V=0$ and 10 mV) with $\theta=0$ and 300 (N) atoms along the circumference. This corresponds to a tubule of radius 11.7 nm assuming a single-layered cylinder. Large tubules tend to be multilayered, but since the interlayer interaction is one-tenth of the intralyaer interaction,[14] it is reasonable to view different layers as independent conductors in parallel. Each layer will then exhibit an A-B effect with a characteristic period (and phase) determined by its diameter and chirality. This will tend to dilute the overall conductance modulation so that tubules with fewer layers are more desirable. Single-shell tubes have been synthesized but their diameter tends to be small (~ 1 nm at present stage). For such small diameters a complete cycle of A-B oscillation would take a very large magnetic field. However, for tubes with specific chirality the semimetal-semiconductor transition could occur at reasonable magnetic field. For simplicity we set the average transmission t equal to a constant and neglect any barrier on the substrate side ($\eta \sim 1$). The detailed conductance characteristics will change if these assumptions are modified, but the broad features should remain the same. The main point is that a modulation in the conductance should be observable even at room temperature under reasonable bias.

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[12]The number of allowed modes $M(E)$ plays a central role in the Landauer approach analogous to that played by the density of states $D(E)$ in the conventional approach. The latter can be obtained by weighting each mode by its corresponding one-dimensional density of states: $D(E)=\sum_m 1/(\pi|\partial E/\partial k_L|_m)$. Past work on graphene tubules has focused on the density of states $D(E)$. Since we are using the Landauer approach we will focus on the number of transverse modes $M(E)$.

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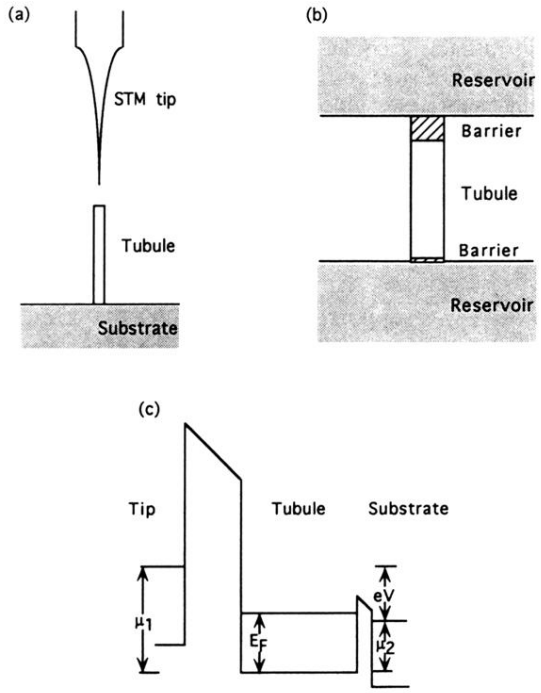

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