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Nondissipative Orbital Currents in Finite Quantum Systems: A Comparative Study

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The idea of nondissipative, persistent currents in mesoscopic metallic or semiconducting rings and cyclinders appears counterintuitive, because it contradicts our experience with currents in macroscopic metals. On the other hand such orbital currents are well known properties of atoms. A comparative study of nondissipative ring currents in different finite quantum systems is therefore of interest. In this paper the properties of atoms, mesoscopic metallic or semiconducting rings and cylinders and elongated molecules called carbon nanotubes are dicussed and compared.

KEY WORDS: orbital current; persistent current; finite quantum systems.

1. INTRODUCTION

It is well known (Hund, 1938) that in small structures with discrete energy levels currents can flow in thermal equilibrium. These currents are flowing along closed orbits, they are frictionless, and there is no Joule heating associated with them. The magnetic moments associated with them can be paramagnetic or diamagnetic and can be quite large. Such orbital persistent currents run e.g. in atoms, molecules, mesoscopic metallic or semiconducting rings or cylinders and carbon nanotubes. A comparative discussion of these currents in different small entities is therefore of interest and will be given below.

One should stress and important difference between small, finite systems and the macroscopic ones. In a normal large conductor the initial ring currents would die down quickly due to Ohmic resistance. In small systems with discrete energy levels at temperatures T for which kT is smaller than quantum size energy gap Δ_0, the scattering to excited states is hampered and it is the reason of the persistency of the current. As Δ_0 decreases with increasing the smaple size and tends to zero in thermodynamical limit, the properties of small systems discussed below are

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absent in macroscopic samples. We shall ignore an electron spin in the following, because the incorporation of spin does not change the discussion essentially. We also assume that mesoscopic systems are made from a very clean material (ballistic regime).

2. PERSISTENT, ORBITAL CURRENTS IN ATOMS

Nondissipative orbital currents and the associated magnetic moments are well known properties of atoms with incomplete p, d, and f shells ($l = 1, 2, 3$, l is the orbital quantum number). The many-electron ground state is built by occupying single electron states with finite azimuthal quantum numbers m, $-l \leq m \leq l$.

To characterize the stationary states of an atom it is not enough to give the probability density (Ginter, 1979)

$$\varrho \equiv |\Psi(r)|^2, \tag{1}$$

one has also to consider the current density $\vec{j}(r)$.

In the presence of the magnetic field $\vec{B}(\vec{B} = rot\vec{A})$ parallel to the z axis the formula for $\vec{j}(r)$ reads

$$\vec{j}(r) = \frac{e\hbar i}{2m_e}(\Psi^* \vec{g}rad\Psi - \Psi\vec{g}rad\Psi^*) - \frac{e^2\vec{A}}{m_e}|\Psi|^2. \tag{2}$$

Let us discuss at first the current density in the absence of the magnetic field. For real Ψ, $\vec{j}(r) = 0$, however for complex Ψ it can be nonzero. If we take wave functions with finite m

$$\Psi_{nlm} = f_{nl}(r, \theta)\, e^{-im\varphi} \tag{3}$$

we obtain the nonzero current density

$$\vec{j}_{nlm}(r) = j_{nlm}(r)\vec{\hat{e}}_\varphi, \tag{4}$$

where $\vec{\hat{e}}_\varphi$ is the unit vector in the φ direction,

$$j_{nlm}(r) = \frac{e\hbar\varrho_{nlm}(r)}{m_e\sqrt{r^2 - z^2}}m, \tag{5}$$

where ϱ_{nlm} is given by (1). We see that each state with $m \neq 0$ carries the finite "spontaneous" current I_{nlm} encircling the z axis.

$$I_{nlm} = \int_S \vec{j}_{nlm}\, d\vec{S}. \tag{6}$$

By "spontaneous" we mean a current which flows without a support of any magnetic or electric field. The total current at $B = 0$ of such a many electron system follows from the appropriate superposition of occupied single electron eigenstates (e.g. according to the Hund's rule in atoms). In atoms with open p, d, or f shells such currents are often nonzero even in the ground state and their density is enormous—it is of the order of $j \sim 10^{12}$ A/cm^2. The energy difference from the ground state to the first excited state is, in atoms, of the order of 1–10 eV so these currents are persistent at ambient temperatures. In atoms with filled shells a full cancellation of "spontaneous" currents occurs.

The magnetic moment of a given state is

$$\vec{\mu} = \frac{1}{2} \int \vec{r} \times \vec{j}(\vec{r})\, d^3\vec{r}. \tag{7}$$

Assuming $A = 0$ and inserting (2) into Eq. (7) we get a nonzero orbital magnetic moments for states with nonzero "spontaneous" currents i.e. with $m \neq 0$,

$$\vec{\mu} = \mu_m \vec{\hat{z}}, \qquad \mu_m = -\mu_B m. \tag{8}$$

where $\mu_B = |e\hbar/2m_e|$. The total orbital magnetic moment (called sometimes permanent magnetic moment) equals

$$\mu = \sum_{m_{\text{occ}}} \mu_m, \tag{9}$$

where summation goes over all occupied m states for a given l. The magnetic moment of such atoms amounts to a few μ_B. Atoms with filled shells do not have permanent magnetic moments.

For $A \neq 0$ the diamagnetic current j_d is induced

$$j_d = -\frac{e^2}{m_e} A|\Psi|^2. \tag{10}$$

These diamagnetic currents are of the order of $I \sim 10^{-10}$ A for $B = 1$ T.

Thus, in the presence of the magnetic field, the magnitude of the total current and of the orbital magnetic moment changes. It is easy to check that, due to the small radius of the atom, these changes remain very small; they are of the order of 10^{-4} for the magnetic field $B = 1$ T in an atomic p state.

In atoms with "spontaneous" currents the interaction between electrons in the m-th states leads to an energy gap that stabilizes the current carrying ground state against transitions to other configurations. The interaction is via the Coulomb repulsion $e^2/4\pi\epsilon_0 r$, which acts over distances of the order of the screening length d_s, d_s is of the order of a few Å. The gain of energy with respect to the exited configurations consists in minimizing the Coulomb repulsion by occupying states with maximum m, consistent with the Pauli principle (Hund's second rule). It leads to an energy gap of the order of 1–10 eV. The corresponding transition temperature to the current carrying ground state is of the order 10^4–10^5 K, far above the melting temperature of solids. Therefore the current carrying state cannot be destroyed by simply increasing the temperature.

3. PERSISTENT, ORBITAL CURRENTS IN MESOSCOPIC RINGS AND CYLINDERS

Let us consider now the quasi 1D mesoscopic ring. The kinetic energy of an electron encircling the closed orbit of radius R in the presence of the magnetic field B perpendicular to the plane of the orbit is quantized (Cheung *et al.*, 1988)

$$E_m = \frac{\hbar^2}{2m_e R^2}\left(m - \frac{\phi}{\phi_0}\right)^2, \tag{11}$$

where $\phi = B\pi R^2$, $m = 0, \pm 1, \pm 2, \ldots,$ is the orbital quantum number for the electron going around the ring or cylinder circumference, $\phi_0 = h/e = 4.14 \times 10^{-7}$ Gscm^2, ϕ_0 is the quantum flux. For mesoscopic quasi 1D ring with N electrons $m_{\rm max} \gg 1 (m_{\rm max} = m_F \simeq N/2 \gg 1)$, $m_{\rm F}$ is the azimutal quantum number at the Fermi Surface (FS) (for atoms $m_{\rm max} = l = 1, 2, 3$).

We see that a quantum size energy gap Δ_0 exists at the FS, $\Delta_0 = \hbar^2 N/2m_e R^2$ We find $\Delta_0 \sim 270$ K for $R \sim 400$ Å and $\Delta_0 \sim 11$ K for $R \sim 1\ \mu$m.

Each electron occupying a state with $m \neq 0$ carries a finite current

$$I_m(\phi) = \frac{\partial E_m}{\partial \phi} = \frac{e\hbar}{2\pi R^2 m_e}\left(m - \frac{\phi}{\phi_0}\right). \tag{12}$$

with a current density

$$j(r) = \frac{eh\varrho}{m_e R}\left(m - \frac{\phi}{\phi_0}\right), \tag{13}$$

where ϱ is the electron density. For $\phi = 0$ we find a "spontaneous" current connected with energy levels with $m \neq 0$, $I_m = e\hbar m/2\pi m_e R^2$.

The formula for the total currents is

$$I(\phi, T) = \sum_m I_m f_{\rm FD}(T, \phi), \tag{14}$$

where $f_{\rm FD}(T, \phi)$ is the Fermi–Dirac distribution function.

For $T = 0$ only the states up to $\pm m_{\rm F}$ are occupied. The currents from occupied m levels at $\phi = 0$ have a strong tendency to cancel and the total current depends on the number of electrons (the summation goes over all occupied states).

If the number of electrons in quasi 1D ring is even, then the last level below a FS is occupied by a single electron and we get a "spontaneous" current

$$I \equiv I_0 = \frac{e\hbar m_F}{2\pi m_e R^2} = \frac{e\hbar N}{4\pi m_e R^2}. \tag{15}$$

This current has a smaller current density than in atoms. For 1D ring with $R = 400$ Å, $j \sim 10^{10}$ A/cm^2, whereas for $R = 1\ \mu$m, $j \sim 10^8$ A/cm^2.

In mesoscopic 1D rings with odd number of electrons the "spontaneous" current is zero due to the total cancelation of currents from $\pm m$ states.

In the presence of the magnetic field ($B \neq 0$) the system reacts with the diamagnetic current. For rings with an even number of particles N we get (Cheung *et al.*, 1988)

$$I = I_0 \left(sgn\phi - 2\frac{\phi}{\phi_0} \right), \qquad \phi \in (-\phi_0, \phi_0), \tag{16}$$

where $sgn\phi$ is 1 for positive ϕ and -1 for negative ϕ, whereas for rings with odd number of particles N,

$$I = -2I_0 \frac{\phi}{\phi_0}, \qquad \phi \in \left(-\frac{\phi_0}{2}, \frac{\phi_0}{2} \right). \tag{17}$$

The current amplitude decreases with increasing R. We find $I_0 \sim 10^{-6}$ A for $R = 400$ Å and $I_0 \sim 10^{-8}$ A for $R = 1\ \mu$m.

The currents in mesoscopic rings are persistent at $kT < \Delta_0$, however at finite T, the amplitude and the shape of $I(\phi, T)$ changes. In particular the sharp peak at $\phi = 0$ in Eq. (16) is smeared into the broader one and the numerical calculations (Cheung *et al.*, 1988; Stebelski *et al.*, 1998) show that we obtain for small ϕ a paramagnetic current whose amplitude decreases with increasing T (see Fig. 1). Such temperatures smearing of the "spontaneous" current is impossible in atoms because of much larger energy gaps. Persistent currents in mesoscopic rings have been detected in several experiments (Chandrasekhar *et al.*, 1991; Levy *et al.*, 1990; Mailly *et al.*, 1993).

Each current loop is equivalent to the magnetic moment. The magnetic moment of a circular current I_m is

$$\mu_m = I_m \pi R^2 = \mu_B \left(m - \frac{\phi}{\phi_0} \right). \tag{18}$$

It follows from the presented considerations that both in atoms and mesoscopic rings the currents and the associated magnetic moments change with the magnetic field. However, in atoms ϕ reaches ϕ_0 only in a field of the order of $B \sim 10^4$ T. This field is at least three orders of magnitude larger than practical laboratory fields. Therefore, we have $\phi \ll \phi_0$ for the atomic orbitals in real fields and $\phi/\phi_0 \sim 10^{-4}$ for $B \sim 1$ T. Thus in atoms the magnetic moment stays practically constant. On the other hand in mesoscopic rings with e.g. $R \sim 400$ AA, ϕ becomes of the order of ϕ_0 at $B = 2, 2$ T and for $R \sim 1\ \mu$m ϕ becomes of the order of ϕ_0 already at $B \sim 13$ Gs. It is therefore easy to shift the current from its zero field value. The current is the periodic function of ϕ/ϕ_0 at accessible fields $B < 10$ T—this phenomenon is caused by the Bohm–Aharonov effect. Thus the change of the magnetic moment μ can be easily made of the order of μ_B. Here lies an important qualitative difference between atoms and mesoscopic rings to practical magnetic fields.

As we already stated in atoms the magnetic moment amounts to a few μ_B. In mesoscopic rings with "spontaneous" current the situation is different. The total

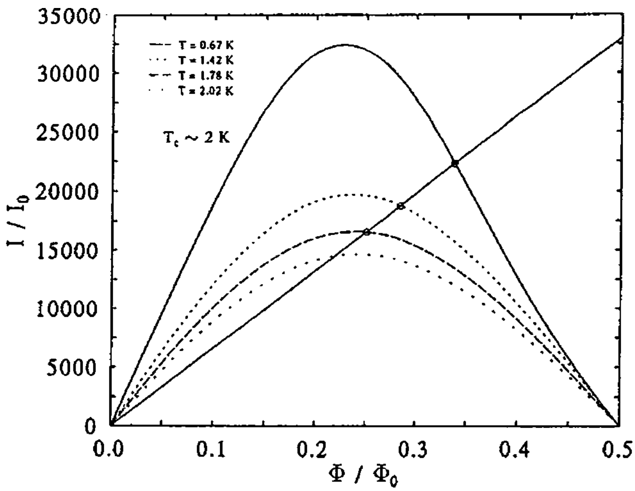

Fig. 1. The graphical solution of the self-consistent equation for the current for different temperatures. The nonzero crossings of the straight line with the current–flux characteristic corresponding to spontaneous currents are marked by open circles. The parameters of the cylinder are : $R = 5 \times 10^3$ Å, length $l_e = 10^4$ Å, and thickness $d = 300$ Å.

magnetic moment in the absence of the magnetic field is large

$$\mu = \sum_{m_{\text{occ}}} \mu_m = \frac{N}{2}\mu_B. \tag{19}$$

The 1D mesoscopic rings with last level below the FS doubly occupied are diamagnetic at $\phi < \phi_0/2$, but the induced current is periodic in ϕ/ϕ_0 and makes paramagnetic jumps at $\phi = m\phi_0/2$. We have a diamagnetic current with amplitude of the order of $I \sim 10^{-6}$ A for $R = 400$ Å and $I \sim 10^{-8}$ A for $R = 1$ μm. Thus, we see that the magnitude of diamagnetic currents is quite larger in mesoscopic rings than in atoms.

In mesoscopic metallic and semiconducting quasi 1D rings the Coulomb interaction does not influence persistent currents in the ballistic regime (Avishai and Braverman, 1995). One can study then the influence of the magnetostatic (current–current) interaction. This interaction, being very weak, is negligible in a single 1D ring. However, if we construct a system made of a set of concentric quasi 1D rings in the form of a torus or a cylinder, then the interaction of currents from different rings (channels) stabilizes the current carrying ground state (Stebelski *et al.*, 1998; Wohlleben *et al.*, 1991). The current–current interaction when taken in the self-consistent mean field approximation results (Lisowski *et al.*, 1988) in the magnetic flux $\phi_I = \alpha I$, where α is the self-inductance of the system. Thus the

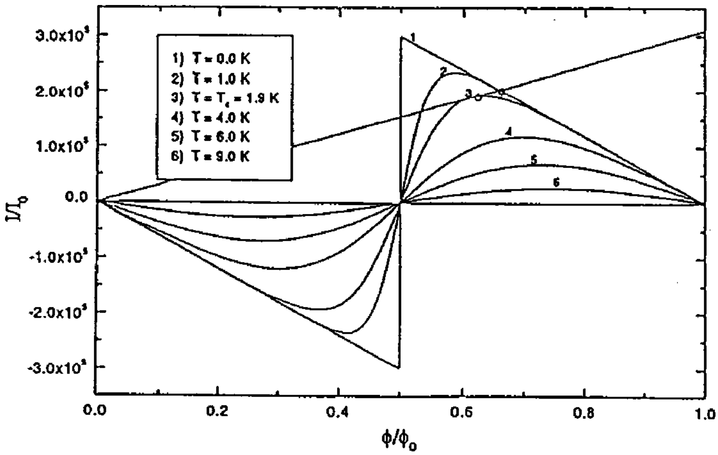

Fig. 2. The graphical solution of the self-consistent equation for the current for different temperatures. The nonzero crossings of the straight line with the current–flux characteristic corresponding to flux trapped are marked by open circles. The parameters of the cylinder are : $R = 3185$ Å, length $l_e = 4 \times 10^4$ Å, and thickness $d = 120$ Å.

total flux, which drives the current, is

$$\phi = \phi_e + \alpha I(\phi, T), \tag{20}$$

ϕ_e is the external flux, $I(\phi, T)$ is a periodic function of ϕ and its amplitude decreases with increasing T. The spontaneous current solution can be obtained by solving at $\phi_e = 0$ the self-consistent equation for the current

$$I(\phi, T) = \frac{\phi - \phi_e}{\alpha}. \tag{21}$$

We get a finite spontaneous current when the amplitude of $I(\phi, T)$ is large.

In Fig. 1 we show the spontaneous current solution for mesoscopic cylinder exhibiting a paramagnetic reaction to small ϕ.

The transition temperature to the state with spontaneous current is of the order of 2 K, it is thus possible to reach this state by reducing the temperature from above to below T_c.

In Fig. 2, we show the graphical solution of the self-consistent Eq. (21) at $\phi_e = 0$ for mesoscopic cylinder exhibiting the diamagnetic reaction to small ϕ (Lisowski, 2000; Stebelski *et al.*, 1998). The self-sustaining solutions marked by open circles are equivalent to flux trapping.

In general, the magnitude of persistent current for mesoscopic hollow cylinder depends on the shape of the FS. For spherical FS the current is weak (Cheung *et al.*, 1988; Stebelski *et al.*, 1998; Wohlleben *et al.*, 1991), whereas its amplitude increases with increasing the curvature of the FS being the strongest for the flat FS. The shape of the FS depends on the symmetry of the crystal and on the band filling. Thus, only in systems with FS having large flat parts, spontaneous currents can occur.

The presence of the magnetostatic interaction is also reflected in the energy gap $\Delta \equiv E_{m_{F+1}} - Em_F$. By making use of Eqs. (11) and (20) one finds

$$\Delta = \Delta_0 \left(1 - 2\frac{\phi_e}{\phi_0} + 2\frac{\alpha |I(\phi, T)|}{\phi_0} \right). \tag{22}$$

We see that Δ contains the term Δ_d

$$\Delta_d = \Delta_0 \frac{\alpha |I(\phi, T)|}{\phi_0}, \tag{23}$$

which is a dynamic part of the energy gap and has to be calculated in a self-consistent way.

The ground state of the system with "spontaneous" currents has a Zeeman degeneracy. Therefore, in thermal equilibrium at $\phi_e = 0$, the expectation value of this current or of the respective magnetic moment is zero if the relaxation time for transitions between the states $+\mu$ and $-\mu$ is short compared to the time scale of the measurement. This situation applies for atoms with "spontaneous" currents in a dilute gas at finite T and for 1D mesoscopic rings. However, the relaxation time increases exponentially with the number of interacting entities. Therefore, in mesoscopic rings or cylinders of finite thickness with a large number of channels the relaxation time can become much longer than the time of the experiment. The current and the magnetization are then truly spontaneous i.e. finite and in one direction in zero external field.

Mesoscopic cylinders made of a normal metal or semiconductor can be obtained e.g. by beam lithography. Such clean samples and of the required geometry are rather difficult to obtain.

4. PERSISTENT, ORBITAL CURRENTS IN CARBON NANOTUBES

Recently a new exiting material that forms small hollow cylindrical structures has been discovered (Iijima, 1991). These are carbon nanotubes (CN) that can be considered as sheets of graphite with a hexagonal lattice that have been rolled up into a tube. It turns out that 2D graphite layers are inherently unstable in the planar configurations if the number of atoms is below a certain limit; they then tend to form cage clusters such as fullerens or CN. CN are large molecules of diameter of several nanometers and length of about 1–10 μm. Their electronic properties depend sensitively on diameter and the rolling angle and slight differences in these parameters cause a shift from a metallic to semiconducting behavior (Dresselhaus *et al.*, 2001).

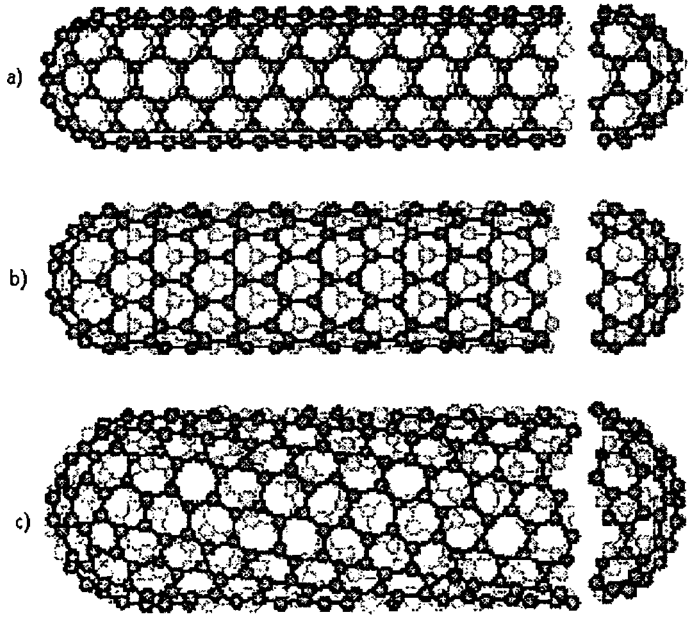

Fig. 3. Different types of carbon nanotubes: (a) armchair, (b) zigzag, and (c) chiral

It is remarkable that similarly shaped molecules consisting of only one element (carbon) may have very different electronic behavior. In general we distinguish three types of CN: armchair, zigzag, and chiral (see Fig. 3).

In a sheet of graphite, each carbon atom is very strongly bonded to three other atoms (σ bonds) and this gives CN the exeptional strengh. A fourth electron is free to move in a π band and is responsible for the conductivity.

CN can be grown in different forms:

(a) Single-wall carbon nanotubes (SWNT) which in only one atom thick with a diameter between 4 Å and 18 Å,
(b) Multiwall carbon nanotubes (MWNT) consisting of several concetric-tubes nested inside each other with outer diameter 100–1000 Å,
(c) Tori, and
(d) Springs.

In a normal metallic cylinders all electrons that contribute to conduction belong to a single parabolic band. Therefore left and right moving electrons can be easily scattered from k_F to $-k_F$ because the symmetries of the wave functions are the same.

The band structure of CN is entirely different. The two bands that cross close to the Fermi Surface are linear and have different symmetries. One band is constructed from molecular bonding states (Goldhaber-Gordon and Goldhaber-Gordon, 2001), the other from antibonding states, so the wave functions of left-and right-moving electrons are very different on an atomic scale. This has an effect on electron behavior: to switch its direction an electron must also switch from a bonding to an antibonding state (or vice versa). This restriction supresses changes in direction so an electron in a metallic CN tends to move persistently in one direction. This results in exeptional ballistic transport properties and elastic mean free paths of 10 μm or more in CN. Such long mean free paths are very difficult to obtain in rings and cylinders made of a normal metal or a semiconductor.

The magnetic field corresponding to ϕ_0 is $H = 1400$ T for nanotube of radius $R = 10$ Å (far beyond the accecible value) and $H = 8$ T for $R = 130$ Å. Thus, only in large CN with $R \geq 100$ Å, the periodicity in ϕ_0 of persistent current and of the magnetic moment (Bohm–Aharonov effect) can be observed. The Bohm–Aharonov effect being a hallmark of coherent transport has been recently observed (Bachtold *et al.*, 1999) by measuring the resistance along the multiwall nanotube in a parallel magnetic field.

The periodic boundary conditions in the circumferential direction give quantized currents in parallel magnetic field (Dresselhaus *et al.*, 2001)

$$I_m = \frac{e\hbar}{2\pi m_e R^2}\left(m - \frac{\phi}{\phi_0} - \frac{\gamma}{3}\right), \tag{24}$$

where $m = 0, \pm 1, \pm 2, \ldots, \gamma = 0$ for metallic, $\gamma = \pm 1$ for semiconducting CN. Thus persistent nondissipative currents can flow in such structures at $kT < \Delta$, Δ is the energy gap, $\Delta = \delta a/\sqrt{3}R$ where a is the lattice constant, δ is the transfer integral between the neighboring π orbitals (Ajiki and Ando, 1993). We find $\Delta \sim 5000$ K for $R \sim 10$ Å and $\Delta \sim 370$ K for $R \sim 130$ Å. Persistent currents are paramagnetic for metallic and diamagnetic for semiconducting CN at low ϕ and has been recently observed (Tsebro *et al.*, 1999). The calculations show that contrary to mesoscopic metallic cylinders the magnetic moment is independent of R for $R > 10$ Å (Ajiki and Ando, 1993).

The energy spectrum of CN looks like a double crown (Dresselhaus *et al.*, 2001) and in the ground state ($E_{\mathrm{F}} = 0$ at the half filling) only the lower one is occupied. A very interesting feature of the energy spectrum is that the FS is limited to six points at the peaks of the crown—it has important influence on the conducting properties of CN.

Pesistent current obtained in this case for the nanotube with e.g. the length $l_e = 173$ Å and the radius $R = 5.485$ Å has the amplitude $I_0 \sim 1.29 \times 10^{-4}$ A (Szopa *et al.*, 2002). By hole doping we can lower the FS and change its shape what leads to the change of persistent current amplitude. The most favorable situation is for the shift $\Delta E_{\mathrm{F}} = -3$ eV for zigzag nanotube, where the FS has a shape of a hexagon with large flat parts. The current's amplitude is then ~ 25 times higher than the current at half filling, $I_0 \sim 3 \times 10^{-3}$ Å (Szopa *et al.*, 2002). For a zigzag nanotube of the type (14, 0), the current density can reach then the value $j \sim 4 \times 10^{10}$ A/cm^2. It has also been shown that for hole doped armchair CN we do not get an enhancement of the current.

The large amplitude of persistent currents raises the possibility of getting self-sustaining currents which produce the magnetic flux capable to maintain themselves even in the absence of extrenal magnetic field. It could take place in MWNT which consists of a set of concentric tubes nested inside each other if all the tubes would be of the same kind. The currents from different walls would then superpose producing large internal flux to sustain the current. However, in MWNT produced, nowadays different tubes have in general different electric and magnetic behavior, but there is a possibility that MWNT having the desired structure will be fabricated in the future.

Finally, we can discuss the possibility of a dynamic gap in CN. The possibility of pair correlations in molecules having conducting π electrons in the presence of the σ-skeleton has been discussed in the literature (Kresin, 1967). The attraction between π electrons may be primarily due to the interaction with the vibrational degrees of freedom and it leads to the formation of the dynamic energy gap. CN, being the elongated molecules have the desired structure and such interactions may lead to superconducting correlations. First reports on superconductivity in CN have been recently published (Kociak *et al.*, 2001; Tang *et al.*, 2001; Zhao and Wang, 2001).

5. CONCLUSIONS

We have discussed some aspects of quantum coherence—nondissipative or—bital currents—in finite quantum systems. Quantum coherence is related to both small size of the sample and to electron correlations coming from Coulomb or magnetostatic interactions.

We have shown that there is no fundamental difference between persistent orbital currents in atoms, mesoscopic rings, cylinders, and carbon nanotubes, but there are some important qualitative and quantitative distinctions which have been discussed in this paper.

Mesoscopic rings and cylinders and especially carbon nanotubes are of great interest because of their possible technological applications in nanoelectronics and in quantum computers.

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