The Aharonov-Bohm Interference and Beating in Single-Walled Carbon Nanotube Interferometers

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Relatively low magnetic fields applied parallel to the axis of a chiral single-walled carbon nanotube are found causing large modulations to the p-channel or valence band conductance of the nanotube in the Fabry-Perot interference regime. Beating in the Aharonov-Bohm type of interference between two field-induced non-degenerate sub-bands of spiraling electrons is responsible for the observed modulation with a pseudo period much smaller than that needed to reach the flux quantum $\Phi_0 = h/e$ through the nanotube cross-section. We show that single-walled nanotubes represent the smallest cylinders exhibiting the Aharonov-Bohm effect with rich interference and beating phenomena arising from well-defined molecular orbitals reflective of the nanotube chirality.

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A hallmark of the Aharonov-Bohm (AB) effect [1] is conductance oscillations of
d metallic rings or cylinders as a function of enclosed magnetic flux with a period on the
order of the flux quantum $\Phi_0 = h/e$ due to quantum interference [2,3]. Carbon nanotubes
are chemically derived cylinders with atomically well-defined structures [4,5]. Multi-
walled nanotubes (MWNT) have radius $r \approx 10$ nm and in magnetic fields parallel to the
tube axis, conductance modulations with a period of $B_0 = \Phi_0 / \pi r^2 \approx 10T$ in magnetic field
have been seen [6]. Single-walled nanotubes (SWNT) are ultra-small with $r \approx 1$ nm and
the magnetic field needed to approach $1 \Phi_0$ flux through the nanotube cross section is $B_0 \approx
1000T$, far beyond reach by experiments. We show here that in the Fabry-Perot
interference [7] regime, beating in the AB-interference between two modes of spiraling
electrons with non-degenerate wave-vectors causes conductance modulations under fields
much smaller than that needed to reach $1 \Phi_0$.

Ando and Ajiki suggested theoretically that the AB effect manifests in a SWNT
by periodically modifying its band structure with a period of $1 \Phi_0$ in magnetic flux and $\sim
1000$ T in field [8]. A periodic change in the band-gap ($\varepsilon_g$) of the nanotube with $d\varepsilon_g/dB \sim
1\text{meV/T}$ was predicted. Optical adsorption experiments were able to confirm $d\varepsilon_g/dB \sim
1\text{meV/T}$ in semiconducting SWNTs in fields up to 45 T [9]. Recently, electrical
measurements also detected similar changes in small band-gap SWNTs transport data
inside the band-gap and Coulomb blockade near the band edge [10]. Much can still be
done to probe the AB-effect in nanotubes for electronic states far away from the band-
gaps and in the ballistic quantum interference regime [7,11] with conductance near $4e^2/h$.

Here, we report the AB effect manifested in the p-channel of a chiral small-gap
SWNT in the Fabry-Perot interference regime. We clearly observe beating between two
non-degenerate modes of spiraling electrons along the nanotube ‘cylinder’ while circulating the circumference multiple-turns. Our analysis verifies the predicted band dispersion resulted from the AB effect. Also, by combining single-molecule raman spectroscopy and the AB-interference pattern from electron transport data, we can identify a (15,6) SWNT in the experimental sample. Our devices comprised of suspended SWNTs grown by chemical vapor deposition (CVD) [12] across trenches (trench height \( h \sim 300 \text{nm} \)) between pre-formed Pt contacts [13] (Fig. 1). The suspended tubes are free from substrate perturbations [13] and the high quality of the devices is responsible for the ‘clean’ experimental data in this work. Micro-raman spectroscopy [14] (Fig. 1c) performed on a \( L = 815 \text{ nm} \) long suspended tube (Fig. 1b) revealed three possible assignments to the tube \((m,n) = (14,8), (15,6) \text{ or } (11,11)\) based on resonance conditions [14,15] with the 785 nm laser used. On the other hand, electrical data suggested a small band-gap nanotube [16,17] due to a gap (with \( G \sim 0 \)) in the conductance vs. gate-voltage curve \((G-V_g, \text{ Fig. 1d})\) recorded at \( T=300 \text{ mK} \). This narrowed down the nanotube to \((14,8) \text{ or } (15,6)\) with diameter \( d \sim 1.5 \text{ nm} \).

The p-channel conductance of the SWNT is high \((\bar{G} \sim 2.3e^2/h)\) and exhibits an interference pattern in \( G \) vs. bias \((V)\) and \( V_g\), whereas the n-channel shows low conductance and Coulomb blockade (CB) (Fig. 1d). This difference is attributed to lower Schottky barrier to the p-channel of the nanotube than to the n-channel with Pt contacts [13,18]. For the p-channel, the conductance pattern is a result of Fabry-Perot like interference between two degenerate modes (sub-bands) of electrons in the nanotube ‘resonator’ confined by the two metal contacts [7].
When magnetic fields (-8T to 8T) were applied nearly parallel to the SWNT axis, we observed pronounced conductance modulations (Fig. 2) to the p-channel conductance despite the relatively low fields. The height of the conductance peaks ($\delta G$, relative to valleys) was reduced from $\sim 0.4e^2/h$ down to $\sim 0.08e^2/h$ as the field was varied from 0 to 8 T or to $-8$ T (Fig. 2b&2c). A slight shift in the positions of the conductance peaks along the $V_g$ axis was also observed as the field increased to 8 T (Fig. 2b&2c).

In zero-field, the electron wave-vector $k = (k_\perp, k_\parallel)$ is quantized along the circumference as parallel lines (Fig. 3) such that the wave-function $\Psi(r + R_{mn}) = \Psi(r)$ where $R_{mn}$ is the wrapping vector of the nanotube. For a chiral $(m,n)$ tube with $m \neq n$ and $m-n=3 \times \text{integer}$, two of the $k_\parallel$ lines cross the inequivalent yet degenerate $K_1$ and $K_2$ points at the first Brillouin zone corners (Fig. 3a) and give rise to two degenerate sub-bands with zero band-gap. Perturbations such as curvature can cause the zero-gap states deviating from $K_1$ and $K_2$ by $\pm \Delta k_\perp^0$ respectively (Fig.3a), resulting in a small band-gap for the two sub-bands (two opposite spiralling modes, Fig. 3c) at $K_1$ and $K_2$ but maintaining the degeneracy (Fig. 3b) with [8]

$$\varepsilon \left( k_\parallel \right) = \gamma \sqrt{\left( \Delta k_\perp^0 \right)^2 + k_\parallel^2},$$

where $\gamma$ is the transfer integral and $2\gamma \Delta k_\perp^0 = \epsilon_g^0$ is the band-gap.

In a magnetic field [8], the electron wave-function exhibits a phase shift by $\Psi(r + R_{mn}) = \Psi(r) \exp(i\Phi / \Phi_0)$ due to the AB effect. This causes a uniform shift in the allowed states along $k_\perp$ by $\Delta k_{AB} = \frac{2\pi \phi}{|R_{mn}| \Phi_0}$ (Fig.3d), i.e., shifting the $k_\parallel$ lines for the $K_1$ and $K_2$-related sub-bands closer to and further away from the zero-gap states (solid
circles in Fig. 3a, 3d) respectively. This leads to increased and reduced band-gaps for the two sub-bands respectively and meanwhile lifts their degeneracy (Fig. 3e),

\[
\varepsilon(k_{||}) = \sqrt{\left( \frac{\Delta k^0_\perp + 2\pi}{|R_{mn}|} \frac{\Phi}{\Phi_0} \right)^2 + k_{||}^2} \quad \text{('+' for } K_1, \text{ '-' for } K_2 \text{ sub-band)} \quad (2)
\]

The change of bandgap is \(\frac{d\varepsilon_g}{dB} \sim \pm \gamma \frac{2\pi}{|R_{mn}|} \frac{d\Phi}{dB} / \Phi_0 \sim \pm 1 \text{ meV/T.} \) Due to the lifted degeneracy between the \(K_1\) and \(K_2\) related sub-bands by the magnetic field, at a given Fermi energy \(\varepsilon\) in the p-channel, two different wave-vector amplitudes now exist (Fig. 3e&3f), i.e.,

\[
|k_{1,2}| = \sqrt{\varepsilon^2 - \left( \frac{\Delta k^0_\perp + 2\pi}{|R_{mn}|} \frac{\Phi}{\Phi_0} \right)^2} \quad \text{('+' for } K_1, \text{ '-' for } K_2 \text{ sub-band)} \quad (3)
\]

for the two modes of electrons with opposite orbiting directions around the nanotube.

We calculated \(G\) vs. \(B\) and \(V_g\) for SWNTs based on interference between non-degenerate \(\pm k_1\) and \(\pm k_2\) modes (Fig. 4) in a way similar to the Fabry-Perot interference for degenerate modes using the multi-channel Landau-Buttiker formalism and S-matrices [7,19]. Conversion of \(\varepsilon\) to \(V_g\) was based on matching experimental \(G\) vs. \(V\) and \(V_g\) under \(B=0\) (Fig. 1d lower inset) with calculated \(G\) vs \(V\) and \(\varepsilon\) (data not shown). The band-gap of the SWNT \(\varepsilon_g^0\) is calculated from \((m,n)\) indices based on the curvature induced band-gap model [17]. Numerically calculated \(G\) vs. \(B\) and \(V_g\) for the (15, 6) SWNT give excellent agreement with experimental data (Fig. 2b&2c vs. 4a) in terms of the conductance peaks height modulation vs. \(B\) and the amount of peak position shift along \(V_g\) under increased field. Calculations based on the (14,8) SWNT do not agree with
experiment with much smaller $G$ vs. $B$ modulations than experimental data (Fig. 4b vs. Fig. 2b&2c).

Up to high fields, simulations reveal that the conductance of the (15,6) SWNT is modulated by $B$ with a pseudo-period of $B_0' \sim 20-30$ T (dependent on $V_g$ or $\epsilon$) and the conductance peak-shift along $V_g$ becomes more apparent and show ‘arching’ (Fig. 4a right panel). The experimentally observed $\delta G$ vs. $B$ and $V_g$ well corresponds to such evolutions, albeit in a smaller range of $B$ field. The physics underlying the $G$ modulation with $B_0' \ll B_0$ is beating between two non-degenerate modes of spiraling electrons.

One sees that in a SWNT with greater $|\Delta k_{\perp}^0|$ or larger chiral angle (defined as $\theta=0$ for armchair and 30° for zig-zag tubes), the difference in the number of turns of circumference-orbiting between the two modes when traversing the tube length $L$ is greater than in a tube with zero or small chiral angle. For various $m$-$n=3 \times$ integer SWNTs with similar diameters, beating modulation is the most rapid in zig-zag tubes, followed by chiral tubes and is non-existent in arm-chair SWNTs (Fig. 4c), as confirmed by simulations. By setting the field-induced phase shifts between the two modes over a length of $L$ to $2\pi$, we find an approximate form of $B_0'$,

$$B_0'(\epsilon) \approx \frac{\pi r 2\epsilon}{L \epsilon_g^0} B_0 \propto \frac{\pi r 2\epsilon}{L} \frac{2\epsilon}{\sin(3\theta)} \cdot B_0$$  \hspace{1cm} (4)$$

suggesting that the beating modulation period is reduced from $B_0$ (corresponding to $1\Phi_0$) by a factor of $r/L \sim 10^{-3}$ and is highly sensitive to the tube chiral angle $\theta$ through the $1/\sin(3\theta)$ relation. Note that $\theta=14^\circ$ and $9^\circ$ for (15,6) and (14,8) SWNT respectively, and the difference in chiral angles leads to a large discernable difference in $B_0'$ according to Eq. (4) and simulations (Fig. 4a vs. 4b right panels). For an armchair nanotube, no sub-
band splitting occurs due to symmetry and thus no beating-like conductance modulation 
\((\theta=0, B_0' \sim \infty)\) by axial magnetic fields (Fig. 4c). Nevertheless, a band-gap is opened for
the two degenerate sub-bands and the band-gap change and resulting non-linearity in \(\varepsilon(k)\)
lead to shifting (or ‘arching’) (Fig. 4c right panel) of the conductance peaks along \(V_g\)
under increasing \(B\). This is the regular AB effect (in the absence of beating) with a \(1 \Phi_0\)
period in magnetic flux and is universal for nanotubes of all chirality (Fig. 4a,b and c
right panels).

The observation of quantum beats for the Aharonov-Bohm effect is to our
knowledge unprecedented in mesoscopic systems and is a result of well-defined
molecular orbitals of nanotubes in magnetic fields. Large band-gap semiconductor
SWNTs with low Schottky-barrier p-channels in the Fabry-Perot regime [18] could
exhibit much more rapid beats than the small band-gap SWNTs. Clearly, many future
opportunities exist for elucidating quantum interference and beating between well-
defined molecular orbitals.

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Figure Captions:

**Figure 1.** A suspended chiral-nanotube quantum wire. (a) Schematic device structure. Nanotubes were synthesized across Pt electrodes over trenches at 800-820°C to produce SWNTs with diameters $d < 2$ nm. (b) Electron micrographs of the device layout (left image) and actual suspended nanotube (right image, $L \sim 815$ nm) used for this work. (c) A resonance micro-raman spectrum (Renishaw, laser $\lambda=785$nm, spot size 1µm scanned over the trench) showing the radial breathing mode (RBM) of a $d \sim 1.5$ nm SWNT with possible chirality assignments of (11,11), (14,8) and (15,6) based on the RBM shift at $\omega=163 \pm 4$cm$^{-1}$. (d) $G-V_g$ characteristics recorded at $T=300$mK in a $^3$He cryostat under bias $V=1$mV. Left inset: $G$ (represented by color, dark blue: $\sim 2e^2/h$, bright white: $\sim 2.4$ $e^2/h$) vs. bias $V$ and $V_g$ for the p-channel showing Fabry-Perot interference pattern. Right inset: $G$ vs. $V$ and $V_g$ for the n-channel displaying Coulomb blockade diamonds.

**Figure 2.** Experimental data of a nanotube interferometer in magnetic fields. (a) $G-V_g$ characteristics for the suspended SWNT in magnetic fields (angle to tube axis $\sim 9^\circ$) from 0 to 8 T recorded at $T=300$ mK under $V =1$mV. The conductance peaks monotonically decrease from 0 to 8 T. (b) A zoom-in view of (a). From top to bottom curves, $G-V_g$ characteristics in field $B=0$ to 8T, in 2 T steps. Notice slight shifts in the peak positions to the left at higher fields. (c) A plot of $G$ (represented by color) vs. $V_g$ and magnetic field $B$ (-8 T to 8T) based on 160 $G-V_g$ curves from $B = -8$T to 8T in 0.1 T steps. Color scale bar unit: $e^2/h$. The slight shifts of conductance peaks positions in (b) are reflected in the slight arching of the interference stripes as highlighted by the dashed line.
Figure 3. The AB effect in a multi-mode nanotube interferometer. (a) The first Brillouin zone of a small-gap chiral SWNT (in zero magnetic field). Only two parallel lines (dashed) of the allowed states closest to the zero band-gap points (the two solid circles near the inequivalent $K_1$ and $K_2$ corner points respectively) are shown. Perturbations cause the zero band-gap states deviating from $K_1$ and $K_2$ by $\pm \Delta k_0^\perp$ due to symmetry and the opening of a small-gap along the two dashed lines ($K_1$ and $K_2$ sub-bands). (b) Dispersion $\varepsilon(k)$ relations for the two degenerate sub-bands near $K_1$ (blue curve) and $K_2$ (red curve) respectively. $k_\parallel=0$ is defined for the states where the conduction and valence band are the closest. (c) Two degenerate modes of spiraling electrons in zero-field. (d) In a magnetic field parallel to the tube axis, the electronic states (dashed lines) shift by $\Delta k_{AB}$ due to the AB effect, leading to lifting of the degeneracy between the two sub-bands. (e) Dispersion curves for the two non-degenerate sub-bands in a magnetic field of $B=8T$. (f) Two non-degenerate modes of spiraling electrons in a magnetic field.

Figure 4. Simulation of Aharonov-Bohm interference and beating versus nanotube chirality. (a), (b) and (c) are simulation results for (15,6), (14,8) and (11,11) SWNT respectively. Left panel: Calculated $G$-$V_g$ curves in fields of $B=0$ to $8T$ from top to bottom in $2T$ steps. Middle panel: Calculated 2-D plot of $G$ vs. $V_g$ and $B$ in a small field range -8T to 8T. Right panel: Calculated 2-D plot of $G$ vs. $V_g$ and $B$ over a wider field range of -30T to 30 T showing that shifting or arching of the conductance peaks is universal for the three different chirality nanotubes. Our simulations here used precise $\varepsilon(k)$ dispersions such as Eq. 2 and 3 instead of linear approximations. Note that we have
also considered and excluded the possibility of a (16,4) tube in the experimental sample. The (16,4) tube has a chiral angle of 19°, larger than the three tubes considered in this figure and exhibit more rapid beating period than the experimental data. Further, the (16,4) tube has $d \approx 1.43$ nm, outside of the range of $d=1.45$-1.53 nm determined from the Raman resonance condition$^{15}$ of $d$ (in nm)$=223.5/(\omega-12.5)$ for $\omega=163 \pm 4$ cm$^{-1}$ (Fig. 1c).
References and Notes

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Fig. 1

(a) Diagram showing the structure of the device with layers labeled Si, SiO₂, Si₃N₄, and Pt, with a local gate indicated.

(b) Images showing a scanning electron microscope (SEM) view of the device at different scales.

(c) Graph showing Raman shift intensity (A.U.) vs. Raman shift (cm⁻¹) with peaks at specific shifts.

(d) Graph showing conductance (G) vs. gate voltage (Vg) with distinct peaks and regions of interest.

(m,n) = (14,8), (15,6) or (11,11)
**Fig. 2**

(a) Graph showing the change in conductance $G$ ($e^2/h$) with respect to the inverse gate voltage $V_g^{-1}$ ($V$). The graph displays oscillatory behavior with varying magnetic field $B$.

(b) Graph showing the conductance oscillations at different magnetic fields $B$: 0 T and 8 T. The oscillations are more pronounced at $B=8$ T.

(c) Color map illustrating the variation of conductance with gate voltage $V_g$ and magnetic field $B$. The map shows a clear periodic pattern with $B=0$ T and $B=8$ T as mentioned.

*Fig. 2*
Fig. 3
Fig. 4

(a) (15,6) tube, $\theta = 14^\circ$

(b) (14,8) tube, $\theta = 9^\circ$

(c) (11,11) tube, $\theta = 0$

G ($e^2/h$) vs. $V_g (V)$ for different tubes with varying $	heta$ angles.