Aharonov-Bohm Oscillation and Chirality Effect in Optical Activity of Single Wall Carbon Nanotubes

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We study the Aharonov-Bohm effect in the optical phenomena of single wall carbon nanotubes (SWCN) and also their chirality dependence. Specially, we consider the natural optical activity as a proper observable and derive it’s general expression based on a comprehensive symmetry analysis, which reveals the interplay between the enclosed magnetic flux and the tubule chirality for arbitrary chiral SWCN. A quantitative result for this optical property is given by a gauge invariant tight-binding approximation calculation to stimulate experimental measurements.

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Aharonov-Bohm (AB) effect manifests the significance of the global nature of the vector potential in quantum theory. Such a geometric phase effect will result in a conductance oscillation with respect to the enclosed flux in the cylindrical conductor. This phenomenon is known as the Altshuler-Aronov-Spivak effect. Since the discovery of carbon nanotubes, it provides an ideal hollow cylindrical lattice sheet with distinguished chiral structures which stimulates extensive studies in recent years. Moreover it provides a novel system for studying the interplay between the enclosed AB magnetic flux versus the chiral symmetry of the tubule. When an applied magnetic flux is threaded through, it has been shown elegantly in the tight binding approximation (TBA) (or effective mass approximation) that the fundamental gap of a single wall carbon nanotube (SWCN) is a periodic function of magnetic flux. This result is also referred to a kind of AB effect. The corresponding spectrum feature has been further analyzed in Ref. 12, where the periodicity of Van Hove singularities with respect to the magnetic flux was also addressed. Experimental transport studies at the finite temperature, for multi-layer carbon nanotubes, also show consistently a current oscillation with a single flux quantum φ0.

Since the SWCN may behave as either an insulator or a metallic conductor depending on its chirality as well as the strength of penetrating magnetic flux, the conventional transport study might not be appropriate for exploring the generic chirality dependence of the AB effect specially for the insulating cases. Actually, as long as the AB flux appears in the hollow SWCN, all electronic canonical momenta will acquire a corresponding vector potential. As a result, the exponential phase factor associated with each C-C link induced by the threading magnetic flux together with the orientation of the chiral trident will make for an interplay between the threading flux and chirality. Therefore the AB-type effect should appear in a variety of phenomena such as the optical properties other than the transport measurements.

In this paper, we show the AB oscillation in the natural optical activity (or natural gyrotropy) of the SWCN with arbitrary chirality. We derive a generic expression for the natural gyrotropy based upon a systematic symmetry analysis and further complement a gauge invariant TBA calculation. It exhibits the interplay among the light polarization, tubule chirality and threaded magnetic flux and reveals the characteristic role of the chiral index.

FIG. 1: For given \( R_0[n_1,n_2] \), there are five chiral vectors, \( R_1[n_1+n_2,-n_1] \), \( R_2[n_2,-n_1-n_2] \), \( R_3[-n_1,-n_2] \), \( R_4[-n_1-n_2,n_1] \) and \( R_5[-n_2,n_1+n_2] \), denoting exactly the same SWCN with their chiral angle deferred \( \pi/3 \) from each other successively. The tubule described by \( R_0[n_1+n_2,-n_2] \) is the mirror image of that described by \( R_0 \). \( a_1 \) and \( a_2 \) are 2D graphite lattice basis vectors.

An ideal SWCN can be viewed as a graphite sheet rolled up into a tubule in a fixed way along the chiral vector \( R = n_1 a_1 + n_2 a_2 \) with \( a_1 \) and \( a_2 \) being the 2D graphite lattice basis vectors. Its geometric structure is hence described by a pair of integers \( [n_1,n_2] \). For given \( [n_1,n_2] \), there are six chiral vectors, i.e., \( R_0[n_1,n_2], R_1[n_1+n_2,-n_1], \ldots, R_5[-n_2,n_1+n_2] \) as shown in Fig. 1, denoting exactly the same SWCN with chiral angles (the angle between \( R \) and \( a_1 \)) as \( \theta, \theta + \pi/3, \ldots, \theta + 5\pi/3 \) respectively. If we introduce the chiral index \( \nu \equiv \text{mod}[n_1-n_2,3] \) as in Ref. 17, we find that they can be divided into two subsets, one is \( \{ R_0, R_2, R_4 \} \) with the same \( \nu \) and the other is \( \{ R_1, R_3, R_5 \} \) with an opposite \( \nu \). The essence of this obvious but nontrivial property,
the same SWCN with opposite chiral indices, lies in the
sameness of the two in-cell carbon atoms A and B. Ac-
tually this is the heritage to the carbon tubule inherited
from the 6mm symmetry of the graphite sheet which has
the 3m symmetry as its invariant subgroup with corre-
sponding quotient groups as E and C2( or σ). C2 ( or σ)
will reverse the A and B atoms and also the sign of chiral
index. As a direct result of the above analysis, the
physical quantities of SWCN can be written as a peri-
odical function of θ with period 2π/3 for ν = ±1 and
π/3 for ν = 0, which are the consequences of the 3m
symmetry and 6mm symmetry, respectively. When per-
forming Fourier expansion for these physical quantities
with respect to θ, all the expansion coefficients should be
functions of those invariant quantities such as the length
of chiral vector Λn(Λ = √n12 + n22 + n32) and the
greatest common divisor of n1 and n2 (denoted by N).

Since the rotation of the linear polarized light trav-
elling through SWCN along the tubule axis carries the
information of its chiral structure and the enclosed mag-
netic flux, we consider the optical rotation power(ORP),
i.e., the rotation angle per unit length, as an observable
to investigate the interplay between chirality and threaded
magnetic flux for the SWCN. By fixing the direction of
the incident light with frequency ω parallel to the tubule
axis (z-axis), the ORP has the expression as

$$\chi \equiv \omega^2 g_{xyz}/(2c^2)$$  \hspace{1cm} (1)

where c is the light speed and the third rank tensor g_{xyz}
is the derivative of the xy component of dielectric tensor
with respect to qz in the long wavelength limit a → 0,
i.e., $g_{xyz} = \partial e_{xy}/(i\partial q_z)_{q \rightarrow 0}$. Notice that a SWCN
and its mirror image with respect to the horizontal plane pos-
sess the same chiral index, but different chiral angle θ and
−θ, respectively (see Fig. 1). Further taking considera-
tion of the third rank tensor properties, one can see that
χ” should be an odd function of θ. Hence, it has a Fourier
series with only odd parity sine functions

$$\chi^\pm(\theta) = a_1^\pm \sin(3\theta) + a_2^\pm \sin(6\theta) + a_3^\pm \sin(9\theta) + \cdots,$$

$$\chi^0(\theta) = a_2^0 \sin(6\theta) + a_4^0 \sin(12\theta) + \cdots,$$  \hspace{1cm} (2)

where the superscripts ± or 0 denote the chiral index, and
the coefficients $a_n^\phi$ depend on the curvature, the magnetic
flux and the frequency of incident light. As a natural con-
sequence of Eq. (2), the ORP of zigzag (θ = 0) and armchair
(θ = π/6) tubules is zero. Moreover, considering the rotation of R by π/3, since $R_0$ and $R_1$ correspond
to the same SWCN with opposite ν, it is straightforward
to see $\chi^\pm(\theta + \pi/3) = \chi^\mp(\theta)$. Hence, we have

$$a_n^\phi(\omega, \Lambda; \phi) = (-1)^n a_n^\phi(\omega, \Lambda; \phi).$$  \hspace{1cm} (3)

We further adopt the scheme developed by White et al.,
which exhibits the chiral structure of SWCN ex-
plicitly with cylindrical geometry being properly built in.
In this scheme, all the lattice sites can be generated by
repeating the pure rotation $C_N$ and the screw operation
$S(α, h)$, while the latter one is shifting along the tubule
axis by h with a simultaneous rotation around the tubule
axis by α. h and α can be obtained through equation
$h = \sqrt{3N|a_1|}/2\alpha$, $\alpha = (2p_1n_1+2p_2n_2+p_3n_3+p_4n_4)/\Lambda^2$
with integers $p_1, p_2$ satisfying $p_2n_1 - p_1n_2 = N$. Hence,
the Bloch momenta $\kappa \in [-\pi, \pi]$ and $n = 0, 1, \cdot \cdot \cdot , N-1$ as
good quantum numbers can be extracted from the char-
acters of the $U(1)$-representations of $S(α, h)$ and $C_N$, re-
spectively. If the magnetic flux is threaded through the
SWCN, the space displacement groups $S(α, h)$ and $C_N$
will be replaced by the corresponding magnetic displace-
ment groups, which are again commutable to the Hamil-
tonian of the flux threaded SWCN. The Bloch momenta
then become $n + \phi/\phi_0$ and $\kappa + \alpha(\phi/\phi_0)$, respectively.
This provides the generic way of the flux dependence for
all the gauge invariant energy spectra as well as ma-
trix elements of physical quantity. As a result, the flux
dependence of the observable for intrinsic SWCNs will
exhibit an AB oscillation with a flux period $\phi_0$. This can
be easily seen from the fact that, these quantities can be
often expressed as a double summations over $\kappa$ and $n$,
the former sums over $[-\pi, \pi]$ and then smears the flux
dependence via $\kappa + \alpha(\phi/\phi_0)$ while the latter summation
will contribute the AB type flux dependence. Then we con-
clude that $a_n^\phi$ in Eq. (2) are periodic functions of $\phi$

$$a_n^\phi(\omega, \Lambda; \phi) = a_n^\phi(\omega, \Lambda; \phi + \phi_0).$$  \hspace{1cm} (4)

Equations (2), (3) and (4) complete the generic symmetry
analysis for the ORP.

Now we apply the TBA to the SWCN with arbitrary
given pair of $[n_1, n_2]$, and ignore the curvature effect for
convenience. The TBA Hamiltonian for the SWCN with
threading flux along the tubule axis can be written as a
$2 \times 2$ matrix form in the momentum space

$$\mathcal{H}(\kappa, n) = V_0 \begin{bmatrix} 0 & \gamma_n(\kappa) \\ \gamma_n(\kappa) & 0 \end{bmatrix}$$  \hspace{1cm} (5)

with V0 being the transfer integral equal to 2.6 eV. The
off-diagonal matrix element has the form $\gamma_n(\kappa) = 1 +
\cdot e^{-i\beta_3 + i\beta_2}$ with $\beta_3 = \frac{\pi}{\Lambda}(\kappa + \alpha\frac{\phi}{\phi_0}) - \frac{\pi}{\Lambda^2}(n + \frac{\phi}{\phi_0})$ and $\beta_2 =
\frac{n_2}{\Lambda}(\kappa + \frac{\phi}{\phi_0}) - \frac{2\pi n_2}{\Lambda}(n + \frac{\phi}{\phi_0})$. Then it is straightforward
to obtain the energy spectrum $E_n^{(c,v)}(\kappa) = \pm V_0|\gamma_n(\kappa)|^{1/2}$
and the one-particle wave function for the conducting or
valence band reads

$$|\kappa, n, c(v)\rangle = \frac{1}{\sqrt{2}} \left( |\kappa, n, A\rangle \pm |\gamma_n(\kappa)|^{1/2} |\kappa, n, B\rangle \right),$$  \hspace{1cm} (6)

where the plus and minus signs correspond to the con-
ducting band and the valence band, respectively. The
Bloch sum in Eq. (6) reads

$$|\kappa, n, s\rangle = \frac{1}{\sqrt{2M\Lambda}} \sum_{l=1}^{N} \sum_{m=-M}^{M} e^{i\pi n_1 m + i2\pi n_2 l/N} |m, l, s\rangle,$$  \hspace{1cm} (7)

with $s$ being the index of two in-cell atoms and $2M\Lambda$ the
tubule length. In Eq. (7) local state $|m, l, s\rangle$ of unit cell
(m, l) can be obtained from the (0, 0) unit cell [0, 0, s] by m successive screw rotations $S(\alpha, h)$ combined with l successive rotations $C_N$ as $|m, l| = T_{m, l}(0, 0)$ with $T_{m, l} \equiv S^m(\alpha, h)C_N^l$.

For purpose of ORP calculation, we start from the off-diagonal element of the dielectric tensor $\varepsilon_{xy}(q, \omega)$,

$$
\varepsilon_{xy} = \frac{8\pi^2}{m^*c^2\omega^2V} \sum_{\kappa, n} \sum_{\kappa', n'} (f(E_{\kappa'}^2(\kappa')) - f(E_{\kappa}^2(\kappa))) 
\times \frac{(\kappa n \sigma | Q_x(q) | \kappa' n' \sigma') \langle \kappa' n' \sigma' | Q_y(q) \rangle (\kappa n \sigma)}{\hbar \omega + E_{\kappa}^2(\kappa) - E_{\kappa'}^2(\kappa') + i\epsilon},
$$

(8)

with $\sigma$ the band index $c$ or $v$. Here, $m^*$ and $c$ are the electron mass and charge respectively. $V \equiv \rho \cdot \tilde{m} |\mathbf{R}|^2/(2\pi)$ is the system volume with $\rho$ as the filling factor. At zero temperature $f(E)$ is simply the step function and the vector operator $\mathbf{Q}(q)$ has the form $[\mathbf{p} + e\mathbf{A}/c, e^iqz]/2$ whose $x$ and $y$ components transform under symmetry operation $T_{m, l}$ as

$$
T_{m, l}^\dagger|Q_x(q) \pm iQ_y(q)|T_{m, l} = e^{imq\pi/2(m+1\pi/N)}|Q_x(q) \pm iQ_y(q)|.
$$

One then obtain the matrix elements of $Q_x \pm iQ_y$ between the conducting and valence bands to be

$$
\langle \kappa', n', c | Q_x(q) \pm iQ_y(q) | \kappa, n, v \rangle = \delta_{\kappa', \kappa + qh} \delta_{n', n \pm 1} 
\times \sum_{m, l} \exp \left[ -i(\kappa \pm \alpha + qh)m - i\frac{2(n \pm 1)\pi}{N} l \right] 
\times \langle m, l, c | Q_x(q) \pm iQ_y(q) | 0, 0, v \rangle,
$$

(9)

here the summation is over the nearest neighbors of the (0, 0) unit cell and it reflects the local geometric structure of SWCN. The $\delta$-function in Eq. (8) gives the selection rule for the transverse optical transition. To keep the gauge invariance, we transfer the calculation of the momentum matrix element into that of the coordinate matrix element through the commutation law $\mathbf{p} + e\mathbf{A}/c = m^* [r, H]/(\hbar^2h^2\tilde{m}^2)$ to avoid the explicit treatment of vector potential $\mathbf{A}$. The translational invariance is also rigorously kept in our calculating procedure. Based upon all the above considerations, the ORP can be calculated as

$$
\chi = \frac{\varepsilon^2\omega^2V^2}{\pi^2c^2} \sum_{n, k} \int \frac{d\kappa}{(\hbar^2 + \kappa^2)^2 - (\Delta(\kappa, n))^2} W(\kappa, n) \Delta(\kappa, n) \partial_\kappa D(\kappa, n) \partial_\kappa, D(\kappa, n) = |\gamma_{n+1}(\kappa + \alpha)|^2 - |\gamma_n(\kappa)|^2, 
$$

$$
W(\kappa, n) = 1 - \Re \left[ \frac{\gamma_n(\kappa)}{|\gamma_n(\kappa)|^2} - \frac{\gamma_{n+1}(\kappa + \alpha)}{|\gamma_{n+1}(\kappa + \alpha)|^2} e^{-i\varphi_{AB}} \right], 
$$

$$
\Delta(\kappa, n) = E_{\kappa}^+(\kappa + \alpha) - E_{\kappa}^-(\kappa),
$$

(10)

where $\varphi_{AB} = \pi(n_1 + n_2)\Delta^{-2}$ is the difference between the azimuth angles of the two atoms in one unit cell.

In the optical limit $q \rightarrow 0$ the minimum of $\Delta(\kappa, n)$ gives an indirect band gap for the transverse optical transition as $\Delta_g = \sqrt{3}\nu_0/\Delta$. This gap sets a lower bound of the incident light wavelength as $\lambda_c = 2\hbar c\Delta(\sqrt{3}\nu_0)$, below which no optical transverse absorption take place. Since this threshold is independent of chirality, it provides a means to determine the tube diameter for all kinds of SWCNs by optical measurements.

![Diagram of ORP and CHIRALITY](image)

**FIG. 2:** The dependence of the ORP on magnetic flux and chiral angle is plotted in (a) for $\nu = 1$ and in (b) for $\nu = 0$. The normalized incident light frequency is $\tilde{\omega} = 0.5$ and the tube diameter is around 37 nm. In (a) the ORP for $\nu = 1$ tubules is a periodic function of chiral angle with period $2\pi/3$ and magnetic flux with period $\phi_0$. The ORP for $\nu = -1$ tubules can be obtained through the translation of chiral angle by $\pi/3$ in (a). In (b), the ORP for the $\nu = 0$ tubules is a periodic function of chiral angle with period $\pi/3$ and magnetic flux with period $\phi_0$.

When plotting $\chi$ versus the chiral angle $\theta$ for arbitrary $[n_1, n_2]$, we found the numerical data can be interestingly regrouped into three categories in accordance with the chiral index $\nu = 0, \pm 1$. By introducing a rescaled dimensionless frequency $\omega = \hbar \omega/\Delta_g$, $\chi^c$ can be fitted perfectly by the following functions with parameter

$$
\chi^c/\hat{f} \approx a_1^\pm (\hat{\omega}, \phi_0) \sin(3\phi) + \Delta^{-1}a_2^\pm (\hat{\omega}, \phi_0) \sin(6\phi),
$$

$$
\chi^0/\hat{f} \approx \Delta^{-1}a_2^0 (\hat{\omega}, \phi_0) \sin(6\phi),
$$

(11)

where $a_1^\pm = -a_1^\mp$ and $a_2^\pm = a_2^\mp$ which, as dimensionless functions of $\hat{\omega}$ and $\phi/\phi_0$, can be determined numerically.
Eq. (11) is entirely consistent with the above symmetry analysis (Eqs. 2 and 3). In Fig. 2 we give 3D plots of $\chi^+$ and $\chi^0$ versus magnetic flux and chiral angle. Note that $\chi^-$ can be obtained through exact relation $\chi^-(\theta) = \chi^+(\theta + \pi/3)$. The results explicitly show AB oscillation in the ORP with a single flux quantum period and $\chi^\nu$ is an even function of $\phi$. For large $\Lambda$, the numerical data verifies $\chi^\nu = -\chi^\nu$ within its precision and $\chi^0$ is much smaller than $\chi^\pm$ in magnitude (see also Eq. (11)). It is interesting to note that for $\nu = \pm 1$ tubules the magnitude of $\chi^\nu$ reaches its maximum/minimum when chiral angle approach $\pi/6$ for fixed $\phi$ and $\bar{\omega}$, however for the armchair tubules ($\theta = \pi/6$), $\chi^0 = 0$. This implies that the chiral index plays a role much more sensitive than the chiral angle in the natural gyrotropy properties of SWCN.

![Graph](image)

**FIG. 3:** Coefficients $a_1^\nu$, $a_2^\nu$, and $a_0^\nu$ are plotted as functions of magnetic flux $\phi$ for different values of renormalized frequency $\bar{\omega}$ in (a), (b), and (c), respectively. The filled square, open circle and cross symbol correspond to $\omega = 0.4$, 0.5 and 0.6, respectively. Fig. (d) is the plot of $a_1^\nu$, $a_2^\nu$, and $a_0^\nu$ vs. $\omega$ denoted by triangle, circle and square, respectively, for different values of magnetic flux. The open symbols are the data for $\phi = 0$ and the filled symbols for $\phi = 0.5\phi_0$.

The fitting coefficients $a_1^\nu$, $a_2^\nu$, and $a_0^\nu$ are plotted in Figs. 3a, 3b, and 3c respectively as the functions of $\phi/\phi_0$ with $\bar{\omega} = 0.4, 0.5, 0.6$. All $a_1^\nu$ with different $\bar{\omega}$ coincide approximately at a common node $\phi/\phi_0 = 0.31$, i.e., coincide at $\pm 0.31 + \mu$ with $\mu$ being the integer, since it is a periodic even function of $\phi/\phi_0$. However, for the $\nu = 0$ tubules, the position of the zero point changes distinctively versus $\bar{\omega}$. Since $a_1^\nu$ dominates the $\chi$ function, this remarkable fact provides a way to distinguish experimentally whether the SWCN under investigation belong to $\nu = \pm 1$ or $\nu = 0$. In Fig. 3d we plotted $a_1^\nu$, $a_2^\nu$, $a_0^\nu$ as functions of $\bar{\omega}$ for fixed $\phi/\phi_0 = 0, 0.5$. It is seen that when the incident light frequency approaches the band edge the ORP for both two kinds ($\nu = 0$ and $\nu = \pm 1$) of SWCN increases rapidly. This fact may also be helpful to realize the ORP with enough intensity by tuning the proper frequency of the incident light. To have a quantitative estimation for the ORP of SWCN, we choose tubule diameter 10 Å and the incident light wavelength 1.4 $\mu$m, $\phi = 0$, and $\rho = 2$, which corresponds to $\bar{\omega} \sim 0.8$ and $\Lambda \sim 13$, then the magnitude of the rotation angle per unit length can be calculated to be 79 rad/cm $\times \nu \sin(3\theta)$ for $\nu = \pm 1$ tubules and 23 rad/cm $\times \sin(6\theta)$ for $\nu = 0$ tubules.

As the final remarks, the AB effect is conventionally investigated in connection with the transport studies for the cylindrical metallic sheet. In this paper, we show that the AB oscillation will also appear in the optical phenomena generically for arbitrary chiral SWCNs. Our arguments apply even to other kinds of chiral single wall nanotubes. Moreover, the response to the enclosed magnetic flux is dramatically affected by the chiral index which characterizes different kinds of global helicity states of SWCNs and permits distinguished chirality dependence of physical properties. The gauge invariant TBA calculation not only provides quantitative results for various quantities to stimulate the experimental measurements but also verifies all the conclusions drawn from our generic symmetry analysis.

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