Supplementary Information

Spintronic Interactions Between Topological Edge States in Chiral Carbon Nanotubes: A Natural Helical Symmetry Approach

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I. Hamiltonian Parameter Vectors of Different Edge Modes for Chiral CNTs in the 1st Nearest Neighbor Tight Binding (1NNTB) Model

I.1 Second Quantization of the 1NNTB Hamiltonian for Four Edge Modes in Graphene

Fig. 1 shows the four different basis atom selection on the graphene lattice corresponding to the four edge modes (I to IV) of chiral CNTs. The second quantized Hamiltonians for the four modes are expressed from eq(S1a) to eq(S1d). The $\hat{A}^\dagger(m_1, m_2)$ and $\hat{A}(m_1, m_2)$ represent the creation and annihilation operators of the electron at the A carbon on the lattice site $m_1 \vec{a}_1 + m_2 \vec{a}_2$. In the 1NNTB model only hopping between nearest neighbors are allowed with hopping energy $\beta_i$.

\begin{align}
\hat{H}_I &= \beta_1 \sum_{m_1, m_2} [\hat{B}^\dagger(m_1, m_2)\hat{A}(m_1, m_2) + \hat{B}^\dagger(m_1 + 1, m_2)\hat{A}(m_1, m_2) + \hat{B}^\dagger(m_1, m_2 + 1)\hat{A}(m_1, m_2)] + h.c. \\
\hat{H}_{II} &= \beta_1 \sum_{m_1, m_2} [\hat{B}^\dagger(m_1, m_2)\hat{A}(m_1, m_2) + \hat{B}^\dagger(m_1 + 1, m_2)\hat{A}(m_1, m_2) + \hat{B}^\dagger(m_1, m_2 + 1)\hat{A}(m_1, m_2) + 1)] + h.c. \\
\hat{H}_{III} &= \beta_1 \sum_{m_1, m_2} [\hat{B}^\dagger(m_1, m_2)\hat{A}(m_1, m_2) + \hat{B}^\dagger(m_1, m_2 + 1)\hat{A}(m_1, m_2) + \hat{B}^\dagger(m_1, m_2 + 1)\hat{A}(m_1, m_2)] + h.c. \\
\hat{H}_{IV} &= \beta_1 \sum_{m_1, m_2} [\hat{B}^\dagger(m_1 + 1, m_2)\hat{A}(m_1, m_2) + \hat{B}^\dagger(m_1 + 1, m_2)\hat{A}(m_1, m_2) + 1)\hat{A}(m_1, m_2) + 1)] + h.c.
\end{align}

With the Fourier transform of the creation and annihilation operators into the reciprocal space,

\begin{align}
\hat{A}^\dagger(m_1, m_2) &= \frac{1}{\sqrt{N}} \sum_{k_1, k_2} e^{-i(m_1k_1 + m_2k_2)} \hat{A}^\dagger(k_1, k_2) \\
\hat{A}(m_1, m_2) &= \frac{1}{\sqrt{N}} \sum_{k_1, k_2} e^{i(m_1k_1 + m_2k_2)} \hat{A}(k_1, k_2)
\end{align}

We derive the Hamiltonian in the reciprocal space for Mode I to Mode IV

\begin{align}
\hat{H}_I &= \beta_1 \sum_{k_1, k_2} [(1 + e^{ik_1} + e^{ik_2})\hat{B}^\dagger(k_1, k_2)\hat{A}(k_1, k_2)] + h.c. \\
\hat{H}_{II} &= \beta_1 \sum_{k_1, k_2} [(1 + e^{ik_1} + e^{i(k_2-k_1)})\hat{B}^\dagger(k_1, k_2)\hat{A}(k_1, k_2)] + h.c. \\
\hat{H}_{III} &= \beta_1 \sum_{k_1, k_2} [(1 + e^{ik_2} + e^{i(k_2-k_1)})\hat{B}^\dagger(k_1, k_2)\hat{A}(k_1, k_2)] + h.c. \\
\hat{H}_{IV} &= \beta_1 \sum_{k_1, k_2} [(e^{i(k_1+k_2)} + e^{ik_1} + e^{ik_2})\hat{B}^\dagger(k_1, k_2)\hat{A}(k_1, k_2)] + h.c.
\end{align}

And the matrix representation of the Hamiltonians in the k space for the four edge modes are expressed in eq (S4a) to (S4b)
Now we define the Hamiltonian parameter vector $H$ is the Pauli vector with the definition as shown by eq (S7)

Using the sum-difference product formula, eq (S5a) to (S5d) can be rewritten as eq (S6a) to (S6d) for more simple analysis,

$$
\hat{H}_I(k_1, k_2) = \beta_1 [(1 + \cos(k_1) + \cos(k_2))\hat{\sigma}_x + (\sin(k_1) + \sin(k_2))\hat{\sigma}_y]
$$

$$
\hat{H}_{II}(k_1, k_2) = \beta_1 [(1 + \cos(k_1) + \cos(k_2-k_1))\hat{\sigma}_x + (\sin(k_1) + \sin(k_2-k_1))\hat{\sigma}_y]
$$

$$
\hat{H}_{III}(k_1, k_2) = \beta_1 [(1 + \cos(k_2) + \cos(k_2-k_1))\hat{\sigma}_x + (\sin(k_2) + \sin(k_2-k_1))\hat{\sigma}_y]
$$

$$
\hat{H}_{IV}(k_1, k_2) = \beta_1 [(\cos(k_1 + k_2) + \cos(k_1) + \cos(k_2))\hat{\sigma}_x + (\sin(k_1 + k_2) + \sin(k_1) + \sin(k_2))\hat{\sigma}_y]
$$

Now we define the Hamiltonian parameter vector $\vec{h}$ fulfilling the condition $\hat{H} = \vec{h} \cdot \vec{\sigma}$, where $\vec{\sigma}$ is the Pauli vector with the definition as shown by eq (S7)
Then the four Hamiltonian parameter vectors for Mode I to IV are expressed as eq (S8a) to (S8d)

\[
\overrightarrow{h}(k_1, k_2) = \beta_1 \begin{pmatrix}
1 + 2 \cos(\frac{k_1 + k_2}{2}) \cos(\frac{k_1 - k_2}{2}) \\
2 \sin(\frac{k_1 + k_2}{2}) \cos(\frac{k_1 - k_2}{2}) \\
0
\end{pmatrix}
\]

(1.3 Selection of the Helical Vectors for Chiral CNTs in the NHCL scheme)

The four edge modes of graphene (Fig.1(b)) result into four nanotube terminations when rolled up into chiral CNTs in Fig.1(c). For a \((n,m)\) carbon nanotube in the natural helical crystal lattice (NHCL) scheme, the periodicity is defined by the circumferential vector \(\overrightarrow{C}_h\) and the helical vector \(\overrightarrow{T}_R\).

\[
\overrightarrow{C}_h = n \overrightarrow{a}_1 + m \overrightarrow{a}_2 \\
\overrightarrow{T}_R = t_1 \overrightarrow{a}_1 + t_2 \overrightarrow{a}_2
\]

Where the selection of \((t_1, t_2)\) in the helical vector \(\overrightarrow{T}_R\) determines the size of the unit cell of CNT in the NHCL scheme. Eq (S9a) and (S9b) can be expressed inversely as

\[
\overrightarrow{a}_1 = \frac{-m \overrightarrow{T}_R + t_2 \overrightarrow{C}_h}{N_{ucl}}
\]

\[
\overrightarrow{a}_2 = \frac{n \overrightarrow{T}_R - t_1 \overrightarrow{C}_h}{N_{ucl}}
\]

Where \(N_{ucl}\) represents the number of graphene unit cells in the unit cell of CNT in the NHCL scheme, and \(N_{ucl} = |nt_2 - mt_1|\)
Therefore the lattice vectors of the reciprocal space of graphene can be written as

\[
\begin{align*}
    k_1 &= \frac{-mk_c + t_2 k_c}{N_{ucel}} \quad (S11a) \\
    k_2 &= \frac{nk_c - t_3 k_c}{N_{ucel}} \quad (S11b)
\end{align*}
\]

Where the boundary conditions for the reciprocal lattice vectors of CNTs are \( k_c = \frac{2\pi \mu}{N_{ucel} - 1} \), and \( k_i \in [-\pi, \pi] \)

Substitute eq (S11) into eq (S8) and the Hamiltonian parameter vectors for the four edge modes can now be written in the reciprocal lattice vectors of the CNTs,

\[
\begin{align*}
    \overrightarrow{h}_I(k_i, k_c) &= \beta_1 \begin{pmatrix}
    1 + 2 \cos \left( \frac{(n-m)k_i + (t_2-t_1)k_c}{2N_{ucel}} \right) \cos \left( \frac{-(n+m)k_i + (t_1+t_2)k_c}{2N_{ucel}} \right) \\
    2 \sin \left( \frac{(n-m)k_i + (t_2-t_1)k_c}{2N_{ucel}} \right) \cos \left( \frac{-(n+m)k_i + (t_1+t_2)k_c}{2N_{ucel}} \right) \\
    0
    \end{pmatrix} \\[6pt]
    \overrightarrow{h}_{II}(k_i, k_c) &= \beta_1 \begin{pmatrix}
    1 + 2 \cos \left( \frac{-(n+m)k_i - (t_1+t_2)k_c}{2N_{ucel}} \right) \cos \left( \frac{nk_i - t_4 k_c}{2N_{ucel}} \right) \\
    2 \sin \left( \frac{-(n+m)k_i - (t_1+t_2)k_c}{2N_{ucel}} \right) \cos \left( \frac{nk_i - t_4 k_c}{2N_{ucel}} \right) \\
    0
    \end{pmatrix} \\[6pt]
    \overrightarrow{h}_{III}(k_i, k_c) &= \beta_1 \begin{pmatrix}
    1 + 4 \cos \left( \frac{(2n+m)k_i - (2t_1+t_2)k_c}{2N_{ucel}} \right) \cos \left( \frac{-mk_i + t_2 k_c}{2N_{ucel}} \right) \\
    4 \sin \left( \frac{(2n+m)k_i - (2t_1+t_2)k_c}{2N_{ucel}} \right) \cos \left( \frac{-mk_i + t_2 k_c}{2N_{ucel}} \right) \\
    0
    \end{pmatrix} \\
    \overrightarrow{h}_{IV}(k_i, k_c) &= \beta_1 \begin{pmatrix}
    -1 + 4 \cos \left( \frac{(n-m)k_i + (t_2-t_1)k_c}{2N_{ucel}} \right) \cos \left( \frac{-mk_i + t_2 k_c}{2N_{ucel}} \right) \\
    4 \sin \left( \frac{(n-m)k_i + (t_2-t_1)k_c}{2N_{ucel}} \right) \cos \left( \frac{-mk_i + t_2 k_c}{2N_{ucel}} \right) \\
    0
    \end{pmatrix}
\end{align*}
\]

(S12a) (S12b) (S12c) (S12d)

In order to simply the parameter vectors in eq (S12), we define the \( \overrightarrow{T}_R \) for the four edge modes as eq (S13a) to eq (S13d). The NHCL unit cell and the band diagrams of the chiral CNTs for the four edge modes are shown in Fig. 1(c) and Fig. S2, respectively.

\[
\begin{align*}
    \overrightarrow{T}'_R &= -\overrightarrow{a}'_1 + \overrightarrow{a}'_2 \quad (S13a) \\
    \overrightarrow{T}''_R &= -\overrightarrow{a}_2 \quad (S13b) \\
    \overrightarrow{T}'''_R &= -\overrightarrow{a}_1 \quad \text{for } m \neq 0 \quad (S13c)(i) \\
    \overrightarrow{T}''''_R &= -\overrightarrow{a}_2 \quad \text{for } m = 0 \quad (S13c)(ii) \\
    \overrightarrow{T}'_R &= -\overrightarrow{a}'_2 \quad (S13d)
\end{align*}
\]

Plug eq (S13) into corresponding Hamiltonian parameter vectors in eq(S12), and the parameter vectors are now greatly simplified as eq(S14a) to eq(S14d)
\[ \overrightarrow{h}_I(k_r, k_c) = \beta_1 \left( \begin{array}{c} 1 + 2 \cos(\frac{(n-m)k_r + 2k_c}{2(n+m)}\cos(k_f/2)) \\ 2 \sin(\frac{(n-m)k_r + 2k_c}{2(n+m)}\cos(k_f/2)) \end{array} \right) \] \tag{S14a}

\[ \overrightarrow{h}_{II}(k_r, k_c) = \beta_1 \left( \begin{array}{c} 1 + 2 \cos(\frac{(n+2m)k_r + 2k_c}{2n}\cos(k_f/2)) \\ 2 \sin(\frac{(n+2m)k_r + 2k_c}{2n}\cos(k_f/2)) \end{array} \right) \] \tag{S14b}

\[ \overrightarrow{h}_{III}(k_r, k_c) = \beta_1 \left( \begin{array}{c} 1 + 2 \cos(\frac{(n+m)k_r + 2k_c}{2m}\cos(k_f/2)) \\ 2 \sin(\frac{(n+m)k_r + 2k_c}{2m}\cos(k_f/2)) \end{array} \right) \] \tag{S14c(i)}

\[ \overrightarrow{h}_{IV}(k_r, k_c) = \beta_1 \left( \begin{array}{c} -1 + 4 \cos(\frac{(n-m)k_r - k_c}{2n}\cos(\frac{mk_r + k_c}{2n})\cos(k_f/2)) \\ 4 \sin(\frac{(n-m)k_r - k_c}{2n}\cos(\frac{mk_r + k_c}{2n})\cos(k_f/2)) \end{array} \right) \] \tag{S14d}

\vspace{1cm}

II. \quad \text{Winding Numbers of Different Edge Modes for Chiral CNTs}

II.1 Modes with Rotational Symmetry in the Parameter Vectors

As shown by Fig. S4 and Fig. S5, the plotting of the parameter vectors of different modes can be separated into two classes: those with \( C_x \) rotational symmetry where \( x \) is finite, and those without. \textbf{Mode I}, \textbf{II}, and \textbf{Mode III} when \( m \neq 0 \) exhibit \( C_x \) rotational symmetry with a rotational angle \( \Phi = \frac{2\pi}{x} \) where \( x \) equals \( n + m, n, \) and \( m, \) respectively, around the symmetry center at \( \sigma = (\beta, 0, 0), \) as shown by Fig. S4(a)–(c) and Fig. S5(a)–(c). To find the winding number, the number of the petal-shaped curves that surrounds the origin, we consider only the first petal numbered as \( \mu = 0, \) and inversely rotate the origin around \( \sigma \) for \( x \) times by the rotational angle \( \Phi = \frac{2\pi}{x} \) to get the equivalent schemes, as shown in Fig. S4(e)–(g) and Fig. S5(e)–(g). And the Hamiltonian parameter vectors in eq(S14a)–(S14c) for the \( \mu = 0 \) petal curve can be expressed in the polar coordinate as eq (S15)

\[ \overrightarrow{h}(k_r, 0) = \left( \begin{array}{c} \beta_1 + \rho(k_f)\cos(\phi(k_f)) \\ \rho(k_f)\sin(\phi(k_f)) \end{array} \right) \] \tag{S15}
where the radial function of the parameter vector fulfills \( \rho(k_i) = 2\beta_1 \cos\left(\frac{k_i}{2}\right) \). We are interested in the intersection between \( \overrightarrow{h}(k,0) \) and the circle formed by the rotation of the origin where

\[
\rho(k_i) = 2\beta_1 \cos\left(\frac{k_i}{2}\right) = \beta_1
\]

\[\text{(S16a)}\]

\[
k_i = \pm \frac{2\pi}{3}
\]

\[\text{(S16b)}\]

As shown in Fig. S4(e)–(g), the intersection occurs at \( \mu_a \) and \( \mu_b \) where \( \phi(k_i) = \phi(\pm\frac{2\pi}{3}) \). We define the “opening angle” \( \Delta \) as the angle \( \angle \mu_a \sigma \mu_b \) and \( \Delta = 2|\phi(\frac{2\pi}{3})| \) for Mode I and II. The values of these parameters corresponding to different modes are summarized in Table S1. The angle of the rotated origins are labeled as \( \mu \Phi = \frac{2\pi\mu}{x} \). The winding number is equal to the number of rotated origins within \( \Delta \). A close inspection of Fig. S4(e)–(g) and Fig. S5(e)–(g) reveals that for different modes, the opening angle \( \Delta \), marked as the red arc, may be located at the left or the right of the symmetry center \( \sigma \), which satisfies the inequality condition eq (S17) or eq (S18), respectively,

\[
\pi - \Delta < \frac{2\pi\mu}{x} < \pi + \Delta \quad \text{(S17)}
\]

\[
\frac{2\pi\mu}{x} < \Delta \quad \text{or} \quad \frac{2\pi\mu}{x} > 2\pi - \Delta \quad \text{(S18)}
\]

Now as shown by Fig. S4(g) and Fig. S5(g), for Mode III, the origin was surrounded by the \( \mu = 0 \) petal for more than one loop, i.e. \( |\phi_{III}(\frac{2\pi}{3})| = \frac{2n + m}{3m} \pi > \pi \). We define the number of loops in the \( \mu = 0 \) petal that surrounds the origin as \( \gamma = \left\lfloor \frac{2n + m}{3m} \right\rfloor \), and we define the opening angle for Mode III, \( \Delta_{III} = 2|\phi(\frac{2\pi}{3})| - 2\pi\gamma \). The condition \( \gamma \in \text{even} \) or \( \gamma \in \text{odd} \) corresponds to the red arc of \( \Delta \) located at the left (Fig.S5(g)) or right (Fig. S4(g)) of \( \sigma \), and fulfill the inequality of eq (S17) or eq (S18), respectively. All the relevant parameters for Mode I, II, and Mode III when \( m = 0 \) are summarized in Table S1.

II.2 Modes without Rotational Symmetry in the Parameter Vectors

For those modes that do not exhibit \( C_s \) rotational symmetry with finite \( x \), we have to derive different inequality conditions to calculate the winding number. First consider Mode III when \( m = 0 \), the plotting of the parameter vector appears as concentric circles centered at \( \sigma(\beta_1,0,0) \), as shown in Fig. S6(a). Therefore, the origin is surrounded by only circles with a radius greater than \( |\beta_1| \). The radii of the circles \( R(k_c) = |2\beta_1 \cos(\frac{k_c}{2n})| \) and the inequality can be written as eq(S19), as shown by Fig.S6(b)
As for **Mode IV**, as shown in Fig. S4(d), the plotting of the parameter vector also appears petal structures similar to **Mode I** but this time without rotational symmetry since the size of the petals are changing simultaneously during rotation. Therefore one also needs to derive a different inequality condition to calculate the winding number. Here \( k_i = \pm \frac{2\pi}{3} \) on each petal can be connected into two circles crossing at the origin and \( \sigma'(-\beta_1,0,0) \) and may be used in the following analysis. The origin is surrounded by the \( \mu \)-th petal curve \( h_{IV_\gamma}(k_i, k_c = 2\pi\mu) \) only when the product of \( h_{IV_\gamma}(-\frac{2\pi}{3}, k_c = 2\pi\mu) \times h_{IV_\gamma}(\frac{2\pi}{3}, k_c = 2\pi\mu) \)  is negative, as shown by eq (S20)

\[
h_{IV_\gamma}(\frac{2\pi}{3}, k_c) \times h_{IV_\gamma}(-\frac{2\pi}{3}, k_c) < 0 \tag{S20a}
\]

\[
h_{IV_\gamma}(\pm \frac{2\pi}{3}, k_c) = 2\beta_1 \sin(\frac{\pm \frac{2\pi}{3}(n-m) - k_c}{2n}) \cos(\frac{\pm \frac{2\pi}{3} m + k_c}{2n}) \tag{S20b}
\]

\[
h_{IV_\gamma}(\frac{2\pi}{3}, k_c) \times h_{IV_\gamma}(-\frac{2\pi}{3}, k_c) = 4\beta_1^2 \sin(\frac{\frac{2\pi}{3}(n-m) - k_c}{2n}) \sin(\frac{-\frac{2\pi}{3}(n-m) - k_c}{2n}) \cos(\frac{\frac{2\pi}{3} m + k_c}{2n}) \cos(\frac{-\frac{2\pi}{3} m + k_c}{2n}) \tag{S20c}
\]

Using the product-to-sum identities, eq (S20c) can be written as eq(S21)

\[
2 \sin(\frac{\frac{2\pi}{3}(n-m) - k_c}{2n}) \sin(\frac{-\frac{2\pi}{3}(n-m) - k_c}{2n}) = \cos(\frac{2\pi(n-m)}{3n}) - \cos(\frac{k_c}{n}) \tag{S21a}
\]

\[
2 \cos(\frac{\frac{2\pi}{3} m + k_c}{2n}) \cos(\frac{-\frac{2\pi}{3} m + k_c}{2n}) = \cos(\frac{2\pi m}{3n}) + \cos(\frac{k_c}{n}) \tag{S21b}
\]

Since the inequality \( \cos(\frac{2\pi m}{3n}) + \cos(\frac{k_c}{2n}) > 0 \) holds within the range \( h_{IV_\gamma}(\frac{2\pi}{3}, k_c) > 0 \), the inequality eq (S20a) can be simplified as the inequality eq (S22a)

\[
f(\mu) = \cos(\frac{2\pi(n-m)}{3n}) - \cos(\frac{2\pi\mu}{n}) < 0 \tag{S22a}
\]

\[
\mu < \frac{n-m}{3} \quad \text{or} \quad \mu > \frac{2n + m}{3} \tag{S22b}
\]

**II.3 Deriving the Winding Number from the Inequality**

So far, we have derived the inequality conditions for the number of loops in the parameter vectors that will surround the origin for different edge modes, summarized in Table S1. To derive the number of integers \( \mu, N_\mu \), that satisfy the inequality for each mode, consider the following
scenarios, for $\mu_a < \mu < \mu_b$,

\[
N_\mu = [\mu_b] - [\mu_a] \quad \mu_a \text{ and } \mu_b \in \text{fractionals} \quad (S23a)
\]

\[
N_\mu = [\mu_b] - [\mu_a] - 1 \quad \mu_a \text{ and } \mu_b \in \text{integers} \quad (S23b)
\]

\[
N_\mu = 0 \quad \mu_a = \mu_b \quad (S23c)
\]

And for $\mu < \mu_a$ or $\mu > \mu_b$,

\[
N_\mu = N_{uvel} + [\mu_a] - [\mu_b] \quad \mu_a \text{ and } \mu_b \in \text{fractionals} \quad (S24a)
\]

\[
N_\mu = N_{uvel} + [\mu_a] - [\mu_b] - 1 \quad \mu_a \text{ and } \mu_b \in \text{integers} \quad (S24b)
\]

\[
N_\mu = 0 \quad \mu_a = 0 \text{ and } \mu_b = N_{uvel} \quad (S24c)
\]

For **Mode I, II, and IV**, the winding number is equal to $N_\mu$. But only for **Mode III**, winding number equals to $N_\mu + m_\gamma$ to account for the additional $m_\gamma$ loops surrounding the origin, as shown in Fig. S4(g). For an $(n, m)$ CNT, when $n - m \neq 3p$, both the lower bound $\mu_a$ and the upper bound $\mu_b$ are fractional, and either eq (S23a) or eq (S24a) applies. Then for the cases when $n - m = 3p \neq 0$, both the lower bound $\mu_a$ and the upper bound $\mu_b$ are integers and either eq(S23b) or eq(S24b) applies. Note that for the armchair CNTs when $n = m$, the inequality condition of **Mode I, II, III, and IV** would correspond to eq (S23c), eq(S23b), eq(S23b), and eq(S24c), respectively. The inequality conditions and the winding number for the four modes are summarized in Table S1.

### II.4 Convergence of Winding Number for Large-Diameter CNTs

With the increase of the CNT diameter, when $n \to \infty$, the integer part of the Gauss function would become much larger than the fractional part and can be approximated as $\lim_{f \to \infty} \lfloor f \rfloor \approx f$. Therefore the winding number can be approximated as

\[
\lim_{n \to \infty} \nu_I(n, m) = \frac{n - m}{3} = \frac{2n + m - 3m}{6} \quad (S25a)
\]

\[
\lim_{n \to \infty} \nu_{II}(n, m) = \frac{n + 2m}{3} = \frac{2n + m + 3m}{6} \quad (S25b)
\]

\[
\lim_{n \to \infty} \nu_{III}(n, m) = \frac{2n + m}{3} \quad (S25c)
\]

\[
\lim_{n \to \infty} \nu_{IV}(n, m) = \frac{2n - 2m}{3} = \frac{2n + m - 3m}{3} \quad (S25d)
\]

The chiral angle $\theta$ is related by the chiral indices $n$ and $m$, and the tube diameter $d$, as
\[
\cos \theta = \frac{\sqrt{3}a_{cc}(2n + m)}{2\pi d_t}
\]

\[
\sin \theta = \frac{3a_{cc}m}{2\pi d_t}
\]

Therefore, at the large tube limit, the winding number would converge to eq (S27)

\[
\begin{align*}
\lim_{d_t \to \infty} \nu_I &= \frac{\pi}{3\sqrt{3}a_{cc}} \left( d_t \left( \cos \theta - \sqrt{3} \sin \theta \right) \right) = \frac{2\pi}{3\sqrt{3}a_{cc}} d_t \cos(\theta + \frac{\pi}{3}) \\
\lim_{d_t \to \infty} \nu_{II} &= \frac{\pi}{3\sqrt{3}a_{cc}} \left( d_t \left( \cos \theta + \sqrt{3} \sin \theta \right) \right) = \frac{2\pi}{3\sqrt{3}a_{cc}} d_t \cos(\theta - \frac{\pi}{3}) \\
\lim_{d_t \to \infty} \nu_{III} &= \frac{2\pi}{3\sqrt{3}a_{cc}} d_t \cos \theta \\
\lim_{d_t \to \infty} \nu_{IV} &= \frac{2\pi}{3\sqrt{3}a_{cc}} \left( d_t \left( \cos \theta - \sqrt{3} \sin \theta \right) \right) = \frac{4\pi}{3\sqrt{3}a_{cc}} d_t \cos(\theta + \frac{\pi}{3})
\end{align*}
\]

As the tube size increases, the ratio between the winding number and the diameter would gradually approach the function of \( \theta \), as shown in Fig.3.
Figure S1. (a) The energy band diagrams and (b) the Hamiltonian parameter vectors of a (5,3) CNT in **Mode I** edge termination in the TCL, MHCL and NHCL schemes. The shifting vectors $T_R$’s of the unit cell for each scheme are written explicitly. In each scheme, each pair of bands is plotted in a different color and corresponds to the colored curve segment in the Hamiltonian parameter vectors.
Figure S2. (a) the unit cell atoms and shifting vectors $T_R$’s, (b) the energy band diagrams and (c) the density of states of a (5,3) CNT for four different edge termination modes shown in red, green, blue and purple for Mode I, II, III, and IV, respectively. The shifting vectors $T_R$’s are the same for Mode II and IV.
Figure S3. The plotting of the Hamiltonian parameter vectors of (3,0)- ~ (3,3)-CNTs in four edge termination modes. Each colored closed loop corresponds to one pair of energy bands in the NHCL scheme.
Figure S4. The plotting of the Hamiltonian parameter vectors of (5,3)-CNTs in four edge termination modes calculated by (a) ~ (d) the 1NNTB model and (i) ~ (l) the 3NNTB model. (e) – (h) Simplified schemes for calculating the winding number ν in the 1NNTB model. The x and the y axes are the \( h_x \) and \( h_y \), respectively, except for (h), where the x axis represents the entries of the bands (\( \mu \)) and \( f(\mu) \) in the y axis represents the function to calculate the winding number as shown in eq.S22.
Figure S5. The plotting of the Hamiltonian parameter vectors of (53,16)-CNTs in four edge termination modes calculated by (a) ~ (d) the 1NNTB model, (i) ~ (l) the 3NNTB model, and (m)~(p) the enlarged square regions in the 1NNTB (shown as solid curves) and 3NNTB (shown as dashed curves) results for comparison. (e) – (h) Simplified schemes for calculating the winding number $\nu$ in the 1NNTB model. The x and the y axes are the $h_x$ and $h_y$, respectively, except for (h), where the x axis represents the entries of the bands ($\mu$) and $f(\mu)$ in the y axis represents the function to calculate the winding number as shown in eq.S22.
Figure S6. (a) The Hamiltonian parameter vectors for a (5,0)-CNT in Mode III containing 5 concentric circles numbered as $\mu = 0, 1, 2, 3, 4$ in which $\mu = 1$ overlaps with $\mu = 4$, and $\mu = 2$ overlaps with $\mu = 3$. (b) The radius $R$ of the 5 concentric circles. Those with $R > |\beta_1|$ encircle the origin and therefore $\nu_3 = 3$. (c) The same plot for a very large zigzag tube (101,0)-CNT in Mode III. (d) Similarly, the winding number of the (101,0)-CNT in Mode III can be calculated by those circles with $R > |\beta_1|$. In this case, $\nu_3 = 67$. 

\[ \mu_a = 1.6 \] 
\[ \mu_b = 3.3 \]

\[ R > |\beta_1| \]
\[ R < |\beta_1| \]
Figure S7. The winding numbers of an (n,m)-CNT in edge termination Mode I. The grey shaded number is the winding number corresponding to the inset Hamiltonian parameter vector of the (5,3)-CNT.
Figure S8. The winding numbers of an (n,m)-CNT in edge termination **Mode II**. The grey shaded number is the winding number corresponding to the inset Hamiltonian parameter vector of the (5,3)-CNT.
Figure S9. The winding numbers of an (n,m)-CNT in edge termination \textbf{Mode III}. The grey shaded number is the winding number corresponding to the inset Hamiltonian parameter vector of the (5,3)-CNT.
Figure S10. The winding numbers of an (n,m)-CNT in edge termination **Mode IV**. The grey shaded number is the winding number corresponding to the inset Hamiltonian parameter vector of the (5,3)-CNT.
Figure S11 (a) The tight-binding and (b) the first principles results of molecular orbital energy distribution for different lengths of a finite (5,3)-CNT in edge Mode I ($\nu_1(5,3) = 1$). The blue and the orange regions correspond to the bulk and the edge states, respectively. The molecular orbital distributions of HOMO and HOMO-1 from both methods are also shown.
Figure S12 (a) The tight-binding and (b) the first principles results of molecular orbital energy distribution for different lengths of a finite (5,3)-CNT in edge Mode II ($\nu_{II}(5,3) = 4$). The blue and the orange regions correspond to the bulk and the edge states, respectively. The molecular orbital distributions of HOMO and HOMO-4 from both methods are also shown.
Figure S13 (a) The tight-binding and (b) the first principles results of molecular orbital energy distribution for different lengths of a finite (5,3)-CNT in edge Mode III ($\nu_{III}(5,3) = 4$). The blue and the orange regions correspond to the bulk and the edge states, respectively. The molecular orbital distributions of HOMO and HOMO-4 from both methods are also shown.
Figure S14 (a) The tight-binding and (b) the first principles results of molecular orbital energy distribution for different lengths of a finite (5,3)-CNT in edge Mode IV ($\nu_{IV}(5,3) = 1$). The blue and the orange regions correspond to the bulk and the edge states, respectively. The molecular orbital distributions of HOMO and HOMO-1 from both methods are also shown.
Table S1. Summary of relevant parameters used in the derivation of winding numbers in the Hamiltonian parameter vectors for different edge termination modes of chiral CNTs.

<table>
<thead>
<tr>
<th>Mode</th>
<th>Condition</th>
<th>$\Psi$</th>
<th>$\phi(\kappa)$</th>
<th>$\delta(\kappa)$</th>
<th>Inequality condition</th>
<th>graveyard vector</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>$C_{\text{n,n}}$</td>
<td>$\pm \frac{2\pi n m}{3(\pi m)}$</td>
<td>$\pm \frac{2\pi (n-m)}{3(\pi m)}$</td>
<td>$\frac{2\pi (n+1)}{3(\pi m)}$</td>
<td>$\frac{n+1}{3} &lt; \mu &lt; \frac{2n+1}{3}$</td>
<td>$\frac{2n-1}{3}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$\frac{2n-1}{3} &lt; \mu &lt; \frac{2n+1}{3}$</td>
<td>$\frac{2n+1}{3}$</td>
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<tr>
<td>II</td>
<td>$C_{\text{n}}$</td>
<td>$\pm \frac{2\pi (2n+1)}{3\pi m}$</td>
<td>$\pm \frac{2\pi (2n+1)}{3\pi m}$</td>
<td>$\frac{2\pi (2n+1)}{3\pi m}$</td>
<td>$\frac{n+1}{3} &lt; \mu &lt; \frac{2n+1}{3}$</td>
<td>$\frac{2n+1}{3}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$\frac{2n+1}{3} &lt; \mu &lt; \frac{2n+3}{3}$</td>
<td>$\frac{2n+3}{3}$</td>
</tr>
<tr>
<td></td>
<td>$m \neq 0$</td>
<td>$y \in \text{odd}$</td>
<td>$C_{\text{n}}$</td>
<td>$\pm \frac{2\pi (2n+1)}{3\pi m}$</td>
<td>$\pm \frac{2\pi (2n+1)}{3\pi m}$</td>
<td>$\frac{2\pi (2n+1)}{3\pi m}$</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$\frac{2n+1+3y-2n}{3(n+y)} \lor \mu &gt; \frac{2n+1+3y}{3(n+y)}$</td>
<td>$m(y+1)$</td>
</tr>
<tr>
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<td></td>
<td>$\frac{2n+1-3y}{3(n+y)} \lor \mu &gt; \frac{2n+1}{3}$</td>
<td>$m(y+1)$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$\frac{2n+1}{3} \lor \mu &gt; \frac{2n+1}{3}$</td>
<td>$m(y+1)$</td>
</tr>
<tr>
<td></td>
<td>$m \neq 0$</td>
<td>$y \in \text{even}$</td>
<td>$C_{\text{n}}$</td>
<td>$\pm \frac{2\pi (2n+1)}{3\pi m}$</td>
<td>$\pm \frac{2\pi (2n+1)}{3\pi m}$</td>
<td>$\frac{2\pi (2n+1)}{3\pi m}$</td>
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<td></td>
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<td></td>
<td>$\frac{2n+1+3y-2n}{3(n+y)} \lor \mu &gt; \frac{2n+1+3y}{3(n+y)}$</td>
<td>$m(y+1)$</td>
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<td></td>
<td>$\frac{2n+1-3y}{3(n+y)} \lor \mu &gt; \frac{2n+1}{3}$</td>
<td>$m(y+1)$</td>
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<td></td>
<td></td>
<td>$\frac{2n+1}{3} \lor \mu &gt; \frac{2n+1}{3}$</td>
<td>$m(y+1)$</td>
</tr>
<tr>
<td>IV</td>
<td>$\delta(\kappa)$</td>
<td>$\pm \frac{2\pi (2n+1)}{3\pi m}$</td>
<td>$\pm \frac{2\pi (2n+1)}{3\pi m}$</td>
<td>$\frac{2\pi (2n+1)}{3\pi m}$</td>
<td>$\mu &lt; \frac{2n+1-3y}{3(n+y)} \lor \mu &gt; \frac{2n+1+3y-2n}{3(n+y)}$</td>
<td>$m(y+1)$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$\frac{2n+1+3y-2n}{3(n+y)} \lor \mu &gt; \frac{2n+1+3y}{3(n+y)}$</td>
<td>$m(y+1)$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$\frac{2n+1-3y}{3(n+y)} \lor \mu &gt; \frac{2n+1}{3}$</td>
<td>$m(y+1)$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$\frac{2n+1}{3} \lor \mu &gt; \frac{2n+1}{3}$</td>
<td>$m(y+1)$</td>
</tr>
</tbody>
</table>

Modes with $C_\downarrow$ rotational symmetry

Modes without $C_\downarrow$ rotational symmetry

$\delta(\kappa) = \left\{ \begin{array}{ll} \cos \left( \frac{2\pi (n-m)}{3n} \right) - \cos \left( \frac{2\pi m}{n} \right) < 0 & \text{if } \mu < \frac{3m-2n}{3(n+m)} \\ \mu > \frac{3m+2n}{3(n+m)} & \text{if } N/A \end{array} \right.$