Supplementary Information for

Quantum Coupled Radial-Breathing Oscillations in Double-Walled Carbon Nanotubes

Supplementary Note S1:

Determination of m_i, m_o, k_i and k_o in double-walled carbon nanotubes (DWNTs) with known chiral indices.

Four parameters, the inner-wall unit-length mass m_i and intrinsic force constant k_i as well as the outer-wall unit-length mass m_o and force constant k_o for uncoupled constituent single-walled carbon nanotube (SWNT), can be accurately determined from the nanotube chiral indices and the isolated radial breathing mode (RBM) vibration frequency-diameter relation.

For the uncoupled two constituent SWNTs,

$$\omega_{RBM} = \frac{1}{2\pi c}\sqrt{\frac{k}{m}} = \frac{A}{D}. \quad \text{(Eq. S1)}$$

The prefactor A was found to be $228 \pm 1\ \mathrm{nm \cdot cm^{-1}}$ from both theoretical and experimental studies[21, 34]. Since m is proportional to nanotube diameter D, k will be inversely proportional to D.

$$m = c_1 D, \quad k = c_2 / D \quad \text{(Eq. S2)}$$

Here, $c_1 = 2.4\times10^{-6}$ kg•m^{-1} and $c_2 = 4.4\times10^{3}$ N•m^{-1}. In our experiment, we know the accurate chiral indices and diameter of each nanotube, thus we can directly obtain m_i, m_o, k_i and k_o for uncoupled constituent SWNTs.

Supplementary Note S2:

Quantum model of two coupled mechanical oscillators.

The Hamiltonian of the coupled oscillator model has the form:

$$H = -\frac{\hbar^2}{2m_{\mathrm{i}}}\frac{\partial^2}{\partial x_{\mathrm{i}}^2} - \frac{\hbar^2}{2m_{\mathrm{o}}}\frac{\partial^2}{\partial x_{\mathrm{o}}^2} + \frac{1}{2}k_{\mathrm{i}}x_{\mathrm{i}}^2 + \frac{1}{2}k_{\mathrm{o}}x_{\mathrm{o}}^2 + \frac{1}{2}k_{\mathrm{c}}(x_{\mathrm{i}} - x_{\mathrm{o}})^2. \qquad \text{(Eq. S3)}$$

This Hamiltonian can be diagonalized into two non-coupled harmonic oscillators describing the collective RBM oscillations in the form of

$$H = \hbar\omega_{\mathrm{L}}(q_{\mathrm{L}}^{+}q_{\mathrm{L}} + \frac{1}{2}) + \hbar\omega_{\mathrm{H}}(q_{\mathrm{H}}^{+}q_{\mathrm{H}} + \frac{1}{2}). \qquad \text{(Eq. S4)}$$

The eigenfrequencies of the two normal modes are

$$\begin{aligned} \omega_{\mathrm{L}} &= \sqrt{\frac{1}{2}\left(z_1 + z_2 - \sqrt{\Delta}\right)} \\ \omega_{\mathrm{H}} &= \sqrt{\frac{1}{2}\left(z_1 + z_2 + \sqrt{\Delta}\right)} \end{aligned}, \qquad \text{(Eq. S5)}$$

where

$$\begin{aligned} &z_1 = \frac{\omega_{\mathrm{i}}^2}{\omega_{\mathrm{c}}^2} + \sqrt{\frac{m_{\mathrm{o}}}{m_{\mathrm{i}}}}, z_2 = \frac{\omega_{\mathrm{o}}^2}{\omega_{\mathrm{c}}^2} + \sqrt{\frac{m_{\mathrm{i}}}{m_{\mathrm{o}}}}, \\ &\omega_{\mathrm{i}}^2 = \frac{k_{\mathrm{i}}}{m_{\mathrm{i}}}, \omega_{\mathrm{o}}^2 = \frac{k_{\mathrm{o}}}{m_{\mathrm{o}}}, \omega_{\mathrm{c}}^2 = \frac{k_{\mathrm{c}}}{\sqrt{m_{\mathrm{i}}m_{\mathrm{o}}}}, \Delta = (z_1 - z_2)^2 + 4 \end{aligned}. \qquad \text{(Eq. S6)}$$

The phonon creation operators of two collective RBM oscillations are superposition of those of inner- and outer-wall motions and have form of

$$
\begin{aligned}
q_{\mathrm{L}}^{+} &= a_{\mathrm{i}}^{+}\langle\omega_{\mathrm{i}}|\omega_{\mathrm{L}}\rangle + a_{\mathrm{o}}^{+}\langle\omega_{\mathrm{o}}|\omega_{\mathrm{L}}\rangle \\
q_{\mathrm{H}}^{+} &= a_{\mathrm{i}}^{+}\langle\omega_{\mathrm{i}}|\omega_{\mathrm{H}}\rangle + a_{\mathrm{o}}^{+}\langle\omega_{\mathrm{o}}|\omega_{\mathrm{H}}\rangle
\end{aligned}
. \qquad \text{(Eq. S7)}
$$

Where $a_{\mathrm{i(o)}}^{+}$ are phonon creation operators of inner(outer)-wall RBM modes; $\langle\omega_{\mathrm{i(o)}}|\omega_{\mathrm{L(H)}}\rangle$ is the inner(outer)-wall component of the coupled RBM mode $\omega_{\mathrm{L(H)}}$ with

$$
\begin{aligned}
\langle\omega_{\mathrm{i}}|\omega_{\mathrm{L}}\rangle &= \sqrt{\frac{\omega_{\mathrm{L}}}{\omega_{\mathrm{i}}}}\frac{2}{\sqrt{2(z_1-z_2)^2+8+2(z_1-z_2)\sqrt{\Delta}}} \\
\langle\omega_{\mathrm{o}}|\omega_{\mathrm{L}}\rangle &= \sqrt{\frac{\omega_{\mathrm{L}}}{\omega_{\mathrm{o}}}}\frac{z_1-z_2+\sqrt{\Delta}}{\sqrt{2(z_1-z_2)^2+8+2(z_1-z_2)\sqrt{\Delta}}} \\
\langle\omega_{\mathrm{i}}|\omega_{\mathrm{H}}\rangle &= \sqrt{\frac{\omega_{\mathrm{H}}}{\omega_{\mathrm{i}}}}\frac{2}{\sqrt{2(z_1-z_2)^2+8-2(z_1-z_2)\sqrt{\Delta}}} \\
\langle\omega_{\mathrm{o}}|\omega_{\mathrm{H}}\rangle &= \sqrt{\frac{\omega_{\mathrm{H}}}{\omega_{\mathrm{i}}}}\frac{z_1-z_2-\sqrt{\Delta}}{\sqrt{2(z_1-z_2)^2+8-2(z_1-z_2)\sqrt{\Delta}}}
\end{aligned}
, \qquad \text{(Eq. S8)}
$$

Supplementary Note S3:

Raman scattering interference between the two electronic resonance channels.

The Raman amplitudes for the low (A_{L}) and high frequency modes (A_{H}) can be respectively described as[35-37]

$$
\begin{aligned}
A_{\mathrm{L}} &= M_{\mathrm{i}}R_{\mathrm{i}}^{\mathrm{L}}\langle\omega_{\mathrm{i}}|\omega_{\mathrm{L}}\rangle + M_{\mathrm{o}}R_{\mathrm{o}}^{\mathrm{L}}\langle\omega_{\mathrm{o}}|\omega_{\mathrm{L}}\rangle \\
A_{\mathrm{H}} &= M_{\mathrm{i}}R_{\mathrm{i}}^{\mathrm{H}}\langle\omega_{\mathrm{i}}|\omega_{\mathrm{H}}\rangle + M_{\mathrm{o}}R_{\mathrm{o}}^{\mathrm{H}}\langle\omega_{\mathrm{o}}|\omega_{\mathrm{H}}\rangle
\end{aligned}
. \qquad \text{(Eq. S9)}
$$

Here $R_{\mathrm{i(o)}}$ and $M_{\mathrm{i(o)}}$ denotes, respectively, the electronic resonance factor and Raman matrix element of the inner(outer)-wall SWNT excitations.

The resonance factor $R_{\mathrm{i(o)}}$ can be obtained from Rayleigh scattering spectra of the DWNTs, which probe directly the optical resonances of both the inner- and outer-wall nanotubes. They can be described as

$$R_{\mathrm{i(o)}}^{\mathrm{L(H)}} = \frac{1}{(E_{\mathrm{ex}} - E_{\mathrm{i(o)}} + i\gamma_{\mathrm{i(o)}})(E_{\mathrm{ex}} - E_{\mathrm{i(o)}} - \omega_{\mathrm{L(H)}} + i\gamma_{\mathrm{i(o)}})}. \qquad \text{(Eq. S10)}$$

Here E_{ex} is the excitation energy; $E_{\mathrm{i(o)}}$ and $\gamma_{\mathrm{i(o)}}$ are, respectively, transition energy and energy broadening of the excited state of the inner(outer)-wall nanotubes.

Supplementary References:

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