Supplementary Information for

(*n*,*m*) Assignments and Quantification for Single-Walled Carbon Nanotubes on SiO_2/Si Substrates by Resonant Raman Spectroscopy

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Supplementary Figures S1-S4

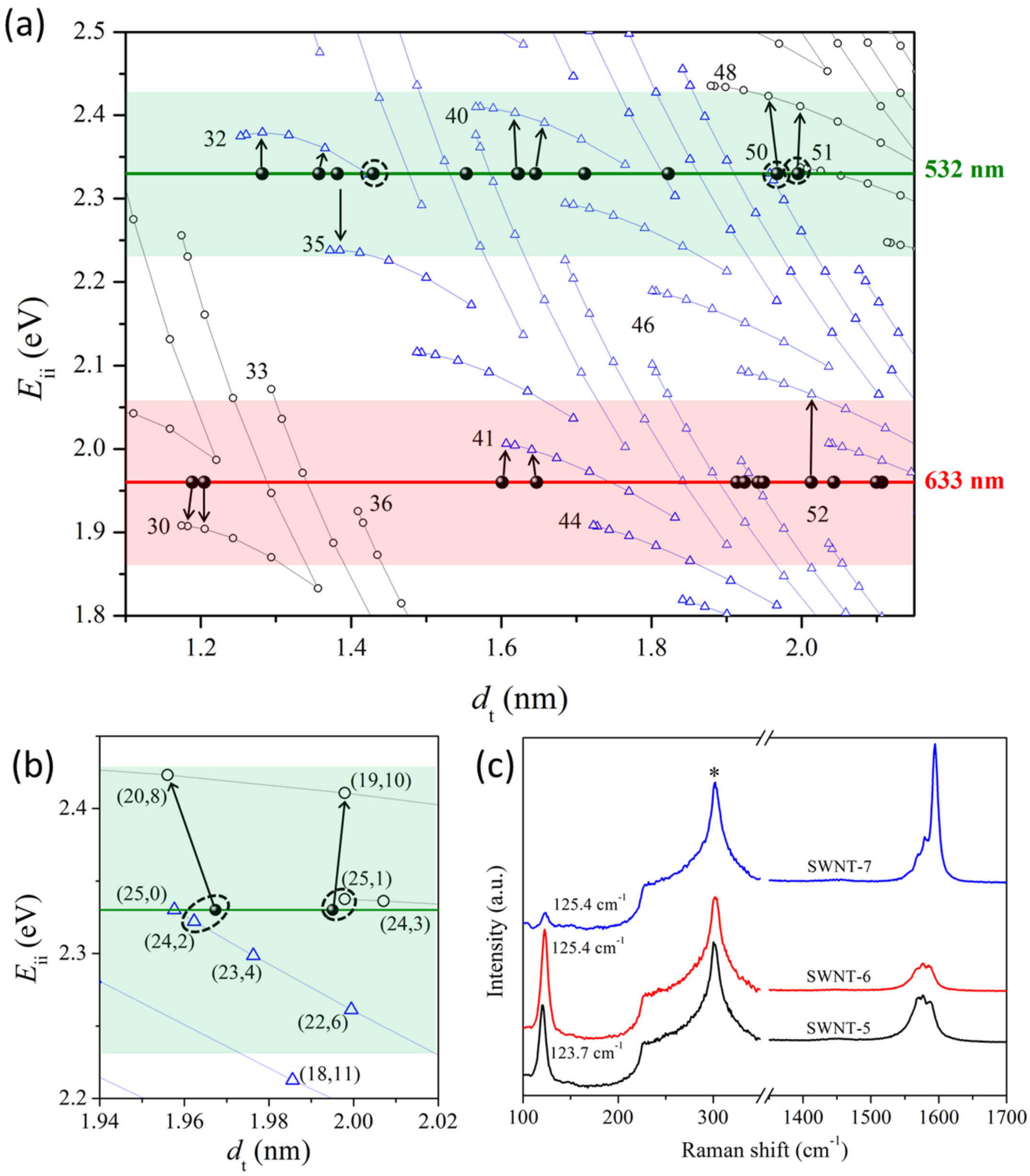

Figure S1. (a) Reference Kataura plot with data points of SWNTs on silicon substrates grown by Ni catalysts plotted according to d_t and excitation laser energy. d_t of the data points are calculated from ω_{RBM} using $d_t = 237.9 / (\omega_{\text{RBM}} - 4.1)$. The (n,m) assignments are indicated by arrows or dashed circles. (b) The enlarged Kataura plot in range of E_{ii} = 2.20-2.45 eV and d_t = 1.94-2.02 nm, showing the assignments of three SWNTs. (c) Resonant Raman spectra of the three SWNTs in (b): a metallic tube with ω_{RBM} = 123.7 cm^{-1} (SWNT-5, black), a metallic tube with ω_{RBM} = 125.4 cm^{-1} (SWNT-6, red), and a semiconducting tube with ω_{RBM} = 125.4 cm^{-1} (SWNT-7, blue). Peaks indicated by * arise from silicon substrates.

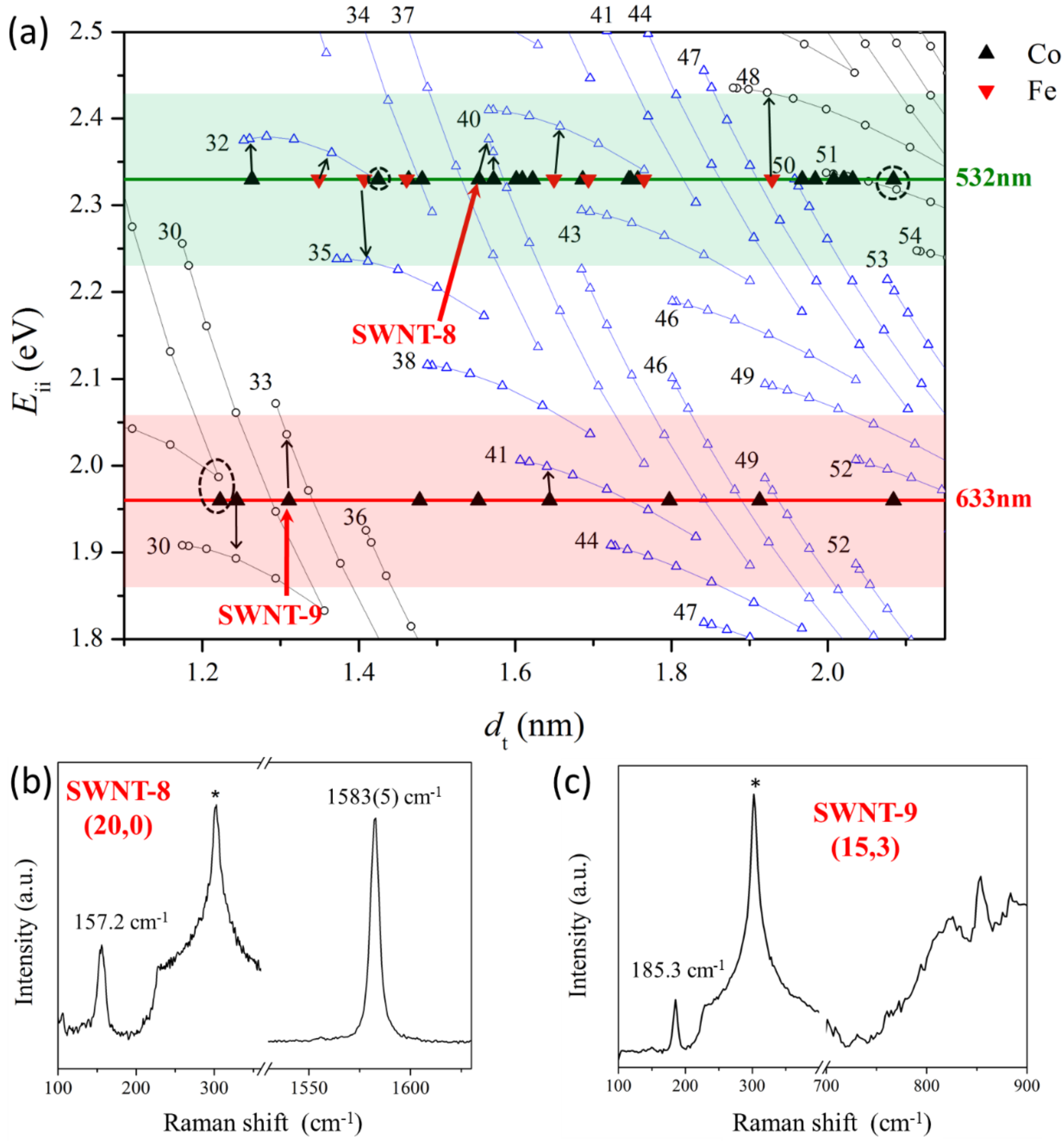

Figure S2. (a) Reference Kataura plot with data points of SWNTs on silicon substrates grown by Co and Fe catalysts plotted according to d_t and excitation laser energy. d_t of the data points are calculated from ω_{RBM} using $d_t = 237.9 / (\omega_{\mathrm{RBM}} - 4.1)$. The (n,m) assignments are indicated by arrows or dashed circles. (b) The resonant Raman spectra of SWNT-8, showing a single-component, symmetric Lorentzian G band shape and an ω_{RBM} at 157.2 cm^{-1} excited by 532 nm laser. (c) The resonant Raman spectra of SWNT-9, showing IFM features and an ω_{RBM} at 185.3 cm^{-1} excited by 633 nm laser. Peaks indicated by * arise from silicon substrates.

Previously it was found that for SWNTs in the d_t range of 0.7-1.3 nm M-SWNTs gives larger B value than S-SWNTs.[1] In addition, ω_{RBM} was also found to be θ-dependent, with near-zigzag SWNTs showing negative deviation from the linear dependence whereas near-armchair SWNTs showing positive deviation. A modified relation of $\omega_{\mathrm{RBM}} = A/d_t + B + (C + D\cos^2 3\theta) / d_t^2$ was then proposed.[1] Moreover, a nonlinear formula of $\omega_{RBM} = \frac{227}{d_t}\sqrt{1 + C_e \cdot d_t^2}$ was also reported with the constant 227 nm·cm^{-1} in agreement with the elastic property of graphite and the parameter C_e reflecting the influence of the van der Waals interaction between SWNT and its surroundings.[2]

For our case, the inset in Figure 4 plots Δd_t with respect to $\cos^2 3\theta$ for both S- (red dots) and M-SWNTs (black squares). The data points appear evenly along the $\Delta d_t = 0$ line, showing little or nearly no θ-dependence of ω_{RBM}. Moreover, we do not observe any noticeable difference in the ω_{RBM}-d_t relation between S- and M-SWNTs. Therefore, it is found that ω_{RBM} of our random SWNTs on silicon substrates depends mainly on d_t.

If the nonlinear function of $\omega_{\mathrm{RBM}} = \left(\frac{227}{d_t}\right)\sqrt{1 + C_e \cdot d_t^2}$ [2] is used instead, it is found that the calculated C_e values for **SWNT-1** to **SWNT-10** differ significantly from one another. In addition, Figure S3 plots this non-linear fitting of ω_{RBM} and d_t with a best fit C_e value of 0.065 nm^{-2} and an $R^2 = 0.988$. As can be seen, small-diameter SWNTs show position deviation and large-diameter SWNTs give negative deviation from the fitting curve, suggesting that this non-linear fitting is not suitable for our samples. If the parameter of 227 nm·cm^{-1} in the function is also allowed to vary freely, the best fit non-linear equation is $\omega_{\mathrm{RBM}} = \left(\frac{240.2}{d_t}\right)\sqrt{1 + 0.015 d_t^2}$ with an R^2=0.9991.

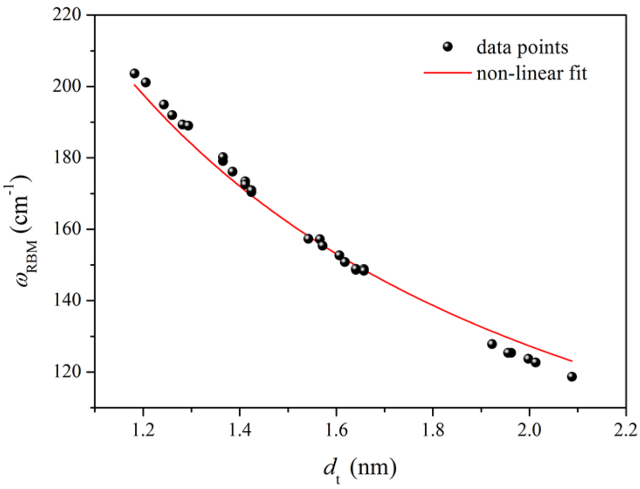

Figure S3. Non-linear fit of ω_{RBM} with respect to d_t for the same set of data points in Figure 4. The fitting curve is $\omega_{\text{RBM}} = \left(\frac{227}{d_{\text{t}}}\right)\sqrt{1 + C_e \cdot d_t^2}$ with a best fit $C_e = 0.065$ and an $R^2 = 0.988$.

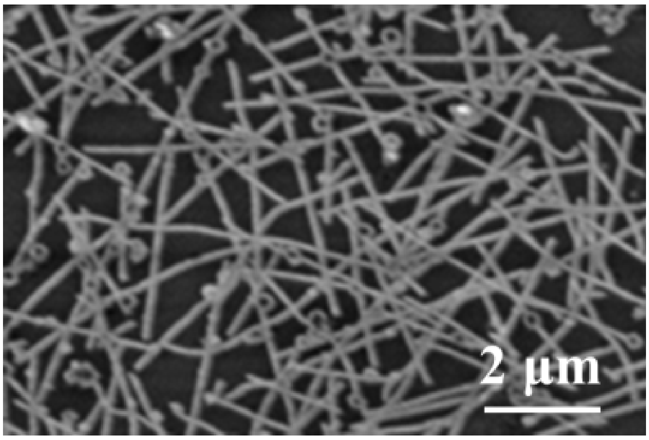

Figure S4. SEM image of a typical chirality-specified SWNT sample on silicon substrates catalyzed by W_6Co_7. The average density is about 6.7 SWNTs per spot of 1 μm diameter calculated from 7 samples.

Reference：

(1) Jorio, A.; Fantini, C.; Pimenta, M.; Capaz, R.; Samsonidze, G. G.; Dresselhaus, G.; Dresselhaus, M.; Jiang, J.; Kobayashi, N.; Grüneis, A. *Phys Rev B* **2005**, *71*, 075401.

(2) Araujo, P.; Maciel, I.; Pesce, P.; Pimenta, M.; Doorn, S.; Qian, H.; Hartschuh, A.; Steiner, M.; Grigorian, L.; Hata, K.; Jorio, A. *Phys Rev B* **2008**, *77*, 241403.