

VibeTube: a code for accurate extraction of CNTs' chirality from TEM images

Daiming Tang, Gaurav Raj, Emmanuel Picheau, Shaaista Hasan, Peng-Xiang Hou & Chang Liu

To cite this article: Daiming Tang, Gaurav Raj, Emmanuel Picheau, Shaaista Hasan, Peng-Xiang Hou & Chang Liu (20 Jul 2026): VibeTube: a code for accurate extraction of CNTs' chirality from TEM images, Science and Technology of Advanced Materials, DOI: [10.1080/14686996.2026.2705816](https://doi.org/10.1080/14686996.2026.2705816)

To link to this article: <https://doi.org/10.1080/14686996.2026.2705816>



© 2026 The Author(s). Published by National Institute for Materials Science in partnership with Taylor & Francis Group.



Accepted author version posted online: 20 Jul 2026.



Submit your article to this journal [↗](#)



View related articles [↗](#)



View Crossmark data [↗](#)

Publisher: Taylor & Francis & The Author(s). Published by National Institute for Materials Science in partnership with Taylor & Francis Group.

Journal: *Science and Technology of Advanced Materials*

DOI: 10.1080/14686996.2026.2705816

VibeTube: a code for accurate extraction of CNTs' chirality from TEM images

Daiming Tang^{a,b,#*}, Gaurav Raj^{a,c,#}, Emmanuel Picheau^a, Shaaista Hasan^{a,b}, Peng-Xiang Hou^d, Chang Liu^{d,e*}

a, Research Center for Materials Nanoarchitectonics (MANA), National Institute for Materials Science (NIMS), Tsukuba, 305-0044, Japan

b, Institute of Pure and Applied Sciences, University of Tsukuba, Tsukuba, 305-8571 Japan

c, Department of Metallurgical and Materials Engineering, Indian Institute of Technology Roorkee, Roorkee, 247667, Uttarakhand, India

d, Shenyang National Laboratory for Materials Science, Institute of Metal Research (IMR), Chinese Academy of Sciences, 72 Wenhua Road, Shenyang, 110016, PR China

e, School of Materials Science and Engineering, University of Science and Technology of China, Hefei 230026, PR China

Contributed equally.

* Corresponding authors: Tang.Daiming@nims.go.jp, cliu@imr.ac.cn

Abstract

Accurate determination of the chiral indices of carbon nanotubes (CNTs) is crucial for their controllable synthesis and practical applications. Transmission electron microscopy (TEM) combined with electron diffraction has been one of the most reliable methods for characterizing the chiral indices of CNTs. However, it is still a challenge to analyze TEM images to extract chirality with high efficiency and accuracy, especially for nanotubes with a diameter larger than 2 nanometers. In this work, a Python code is developed with assistance from an artificial intelligence model (Claude Code) for the auto-extraction of CNTs' chirality from TEM images. Precise and reliable measurement of the diameter was realized by using a new method to determine the average distance between the bright and dark fringes. Symmetry and geometry-based rules enabled measurements of layer lines in the Fourier transformation of TEM images with a sub-pixel resolution. As a result, an accuracy higher than 90% for CNTs with diameters ranging from 0.5 nm to 3.0 nm was achieved. High robustness has been validated by experimentally analyzing the chiralities of CNTs grown by a floating catalyst chemical vapor deposition method. The open-source codes and tools will be valuable for the research community to quantitatively identify the chirality distribution of CNTs with high efficiency.

Keywords

carbon nanotube; chirality; diameter; transmission electron microscope; fast Fourier transform; AI for Science; python code

Impact statement

An open-source Python code for automated, accurate, robust and fast extraction of carbon nanotubes chiralities from TEM images is developed and shared.

Introduction

Electronic properties of carbon nanotubes (CNTs) are determined by the so-called chirality, that is the chiral vector associated with quantization along the circumference. Depending on the chiral indices (n, m) , CNTs can be metallic or semiconducting (1). Therefore, it is critical to accurately determine the chirality for understanding the precise chirality-related fundamental properties and their practical applications in electronics and optics (2-4).

Spectroscopic methods have been developed for characterization of CNTs' chiralities, including resonant Raman scattering (5, 6), photoluminescence (PL) (7, 8), Rayleigh scattering (9, 10). Optical spectroscopic techniques have advantages of being non-destructive, high throughput, high sensitivity to individual nanotubes, and low cost. Microscopic techniques were applied to determine the atomic structure of CNTs. By using scanning tunneling microscopy (STM) combined with scanning tunneling spectroscopy (STS), not only atomic configurations but also electronic structures of individual nanotubes including nanotube junctions were obtained (11). Transmission electron microscope (TEM) has been one of the most powerful instruments to characterize the atomic structure and properties of CNTs. In 1991, Iijima discovered these small graphitic tubes and identified their helical structures by electron diffraction (ED) (12). In recent years, TEM resolution has been greatly enhanced to sub-angstrom level by aberration correctors (13). Since then, TEM has provided detailed information about CNT atomic structures, such as atomic defect (14, 15), chiral angle (16), atomic strain (17), interface structure (18) and handedness (19). Complementary to the local atomic structures in real space, ED provides structural information in reciprocal space and has been used for analyzing the chirality of individual CNTs (20-25). By analyzing layer line patterns, detailed helical structures could be understood. For example, chiral angle could be calculated from the ratio of distances of principal layer lines (D1, D2, D3) to the equator line. And by analyzing intensity oscillations in the equator line, CNT diameter could be calculated. Up to now, such microscopic analysis has been done manually, which is time-consuming and prone to human error.

In 2020, Förster et al. developed a deep learning approach for determining the chiral indices of CNTs from high-resolution transmission electron microscopy (HRTEM) images (26). Automated assignment of the chiralities from the deep learning model was found to be in good agreement

(71%) with manual analysis results. It was also noticed that the accuracy was still not sufficient, especially for nanotubes with relatively large diameters of ~ 2 nm.

Analytical approach, with better interpretability, has been tried to analyze the diameters from TEM images and chiral angles from Fast Fourier Transform (FFTs), however, with less success. It is in principle a simple process to analyze TEM images for CNTs chiralities, essentially by matching the diameter and chiral angle with a table. However, in practice, it is very challenging to accurately assign the chiral indices, mainly for two reasons. Firstly, it is still difficult to measure the diameter of CNTs from TEM images with high precision, due to the interference nature of the fringes (27, 28). Secondly, there is a compromise between resolution in real and reciprocal space. Due to the small field of view (FOV) of HRTEM images, the FFTs suffer from a noisy background.

In this work, a Python code “VibeTube” is developed by using the analytical approach to extract chirality of CNTs from TEM images. Diameters are measured precisely by considering the influence of defocus and by implementing symmetry-based algorithms. Precise measurements in the reciprocal space are realized by a combination of masking, background subtraction, and iterative multiple peak fitting process. As a result, a high accuracy of 84.5% is achieved, tested against a simulated dataset, including 6930 TEM images from 330 chiralities ranging from (6, 0) to (25, 25), diameters from 0.5 nm to 3.0 nm, defocus from -10 nm to +10 nm. In addition, by limiting the defocus value to be larger than 1 nm, to avoid the lack of contrast at near focus, the accuracy is further enhanced to 92.3%. Robustness of the code was tested against an experimental dataset with defocus ranging from -22.8 nm to +22.8 nm. With defocus limited to a reasonable range from -11.4 nm to 7.6 nm, 13 out of 14 outputs were consistent, proving the high robustness. The code has been applied to analyze the chirality distribution of CNTs grown by using a floating catalyst chemical vapor deposition (FCCVD) method with diameters ranging from ~ 1.4 nm to ~ 2.8 nm.

Results & discussions

1. “VibeTube” code architecture and pipelines

The purpose of “VibeTube” codes is to provide the research community with an open-source tool for accurate, robust, fast, and automated determination of carbon nanotube (CNT) chirality

indices (n, m) from TEM images. It is designed to be easy to use, with the main architecture that includes interactive Jupyter notebooks and helper modules (Figure S1). In the notebooks, the user can set parameters, call helper functions, and display results. All physics and computation logics live in helper modules. Parameters are included in a metadata file, which is shared between pipeline stages, for example, between TEM simulation and chirality analysis.

The development of “VibeTube” Python codes was carried out on a Windows 11 Pro system for Workstations. The hardware includes an Intel(R) Xeon(R) W-2195 CPU (2.30 GHz), a RAM of 256 GB, and a graphics processing unit (GPU) of NVIDIA Quadro GV100 with a GPU memory of 32 GB. The codes were developed on Visual Studio (VS) Code with the assistance of artificial intelligence (AI) models, mainly Claude Code.

The codes have been designed for high accuracy, high robustness and high efficiency. By analyzing the physics and contrast transfer functions (CTF) of electron microscopy, a new method is developed to measure the diameter precisely and to be robust against the change of imaging conditions, such as the defocus. Symmetry-based criterion helped to determine the fringes corresponding to the CNTs walls. Quantitative measurement and fitting enable reliable diameter offset for calculating the diameter precisely. Insight into the TEM simulation process resulted in enhanced efficiency, where computation-expensive multi-slice simulation was conducted once and combined with a CTF to calculate TEM images of defocus series.

For AI-assisted coding, or “vibe coding”, it is important to define the architecture and conventions clearly. There are two particularly important conventions in this work: coordinates and defocus. During TEM simulations in abTEM, the coordinate mapping was as follows: nanotube axis was along x and perpendicular to y, and beam propagation was along direction z (Figure S2). In this work, defocus values were referenced to the nanotube center plane, not the cell bottom which is the native reference in abTEM. The conversion is: $df_{abTEM} = df_{user} + cell_width / 2$, where $cell_width$ is the supercell side length. In abTEM, positive defocus = underfocus, while in Kirkland convention: $C10 = -defocus$. This convention was verified by the contrast reversal (d_{max}/d_{min} crossover) occurring at $df \approx -0.5$ nm for all tested nanotubes, confirming that $df = 0$ corresponds to Gaussian focus at the tube center.

The basic pipeline for extracting chirality from TEM images follows (Figure 1): TEM image $\rightarrow$ diameter + FFT D-values $\rightarrow$ chirality lookup (n, m). Simulated TEM images and experimental TEM images are preprocessed into a common normalized representation, then passed through a shared analysis pipeline that extracts chirality from diameter and FFT layer-line measurements. Pipeline for simulated images includes (1) multislice simulations to generate TEM images via abTEM, (2) chirality analysis to extract chirality from simulated TEM images. Pipeline for experimental TEM images includes: (1) preprocessing TEM images, (2) chirality analysis to extract chirality from experimental TEM images. Both pipelines share the same chirality analysis algorithms.

2. Diameter measurements

To accurately assign the chiral indices of CNTs, the diameter must be precisely measured. Under weak-phase object approximation (WPOA), the TEM image of a carbon nanotube is a 2D projection of the 3D cylinder potential. At the optimum imaging condition, the focus is slightly negative to compensate for the positive spherical aberration. The focus condition is the so-called Scherzer focus. Under this condition, atoms show dark contrast in TEM images, and carbon nanotube walls are featured by two parallel dark fringes, along with bright fringes outside. Therefore, measurements of CNTs' diameters have been done commonly by measuring the distance between the dark fringes, which is defined as $d_{\min}$. However, $d_{\min}$ has been reported to be not precisely equal to the geometric diameter of CNTs, which is defined as d_{cal} in this work (26). Förster et al. applied a diameter offset (0.06 nm) to $d_{\min}$ to measure the diameter of CNTs (26). It was reported that the distance between the so-called inflection points (d_{inf}) between the dark and bright fringes are closer to the actual diameter, however, such inflection points are not straightforward to measure and not easily implemented experimentally.

Position of $d_{\min}$ is closely dependent on imaging conditions, especially the defocus value. More importantly, when the imaging condition is changed from under focus to over focus conditions, there is a contrast reversal. Instead of dark fringes, CNT walls will show dominant bright fringes with less dominant dark fringes outside, which is the opposite of the image taken at underfocus condition. The reason for the contrast reversal is due to the CTF, which describes the phase shift due to the objective lens: $\chi(k) = -\pi \lambda \Delta f k^2 + (\pi/2) Cs \lambda^3 k^4$, where λ is the electron wavelength, Δf the defocus, and Cs the spherical aberration coefficient. For an aberration

corrected TEM, Δf dominates the CTF. The image intensity of a weak phase object is modulated by $\sin(\chi)$, which changes sign when Δf passes through near zero (Gaussian focus), leading to the contrast reversal.

In this work, the measurement of CNTs diameters is improved by measuring d_{ave} , which is the average distance between the dark and bright fringes along CNT walls (d_{min} and d_{max}). While d_{max} and d_{min} swap roles at contrast reversal, $d_{ave} = (d_{max} + d_{min})/2$ is largely immune to the swap. It was noticed that the fringes of CNT walls show symmetric patterns, that is “max-min-min-max” for underfocus condition, and “min-max-max-min” for overfocus condition. In addition, after subtraction of the background the variance of the nanotube area is much higher than that of background. Therefore, an algorithm based on local variance and symmetry was designed to identify the wall fringes reliably. Then d_{ave} was calculated by averaging d_{min} and d_{max} , and d_{inf} was obtained by calculating the second derivative between each max–min pair.

Diameter measurement results of the (20, 10) nanotube are presented in Figure 2 and Figure S3, for diameter modes of d_{ave} and d_{min} , respectively. Simulated TEM images at under and over focus conditions are shown in Figure 2a-b, and Figure S3a-b. At a defocus of 4 nm, the nanotube shows a dark fringe with a bright fringe outside. Such contrast is reversed for the image simulated at a defocus of -4 nm. Such contrast features could be clearly seen in the projected intensity profiles (Figure 2c-d and Figure S3c-d). Symmetric but opposite patterns could be detected by images at under and over focus conditions. For conventional d_{min} method, the diameter was measured to be 2.023 nm and 2.468 nm. While the measured value is close to the ground truth of 2.072 nm at under focus condition, there was a big error (19.1%) for the d_{min} value at over focus condition. On the other hand, the d_{ave} values were calculated to be 2.085 nm and 2.059 nm, with a small error of 0.6% and -0.6%, for under and over focus conditions.

Diameter modes and diameter offsets from the geometric diameters of CNTs are tested by using a simulated dataset, including 330 CNT species ranging from (6, 0) to (25, 25), and defocus values $df = -10$ to $+10$ nm, therefore 6,930 simulated TEM images. Diameter offsets, defined as the differences between measured and calculated diameters were evaluated for both under and over focus conditions to determine the most stable, reliable and implementable diameter mode.

Optimum diameter offsets for each diameter mode have been determined from the average offsets.

Figure 3 presents the diameters determined by different modes against the geometric diameters of the simulated dataset at under (a) and over (b) focus conditions, for a defocus values of 2 nm and -2 nm, respectively. Except for a few points, most of the points showed linear relations with the actual nanotube diameters, for both under and over focus conditions. It is interesting to note that for $d_{\min}$ (blue) and $d_{\max}$ (orange), there was a swap between them for the two focus conditions. Such swap can be understood by contrast reversal due to the change in sign of phase shift in the CTF. On the other hand, d_{ave} and d_{inf} are almost overlapped, and the offsets of these two modes did not change the sign along with the change of focus. Therefore, for the conventional $d_{\min}$ mode, it is only reliable for the underfocus condition, with an offset of 0.10 nm. In this work, we use d_{ave} to measure CNT diameters, with an offset of -0.12 nm. This optimum diameter offset was found largely independent of defocus condition for a [-10, +10] nm defocus range.

3. FFT measurements

In addition to diameter measurements in real space (TEM images), the chiral angles are calculated based on layer line measurements in reciprocal space, which are the Fourier transform of TEM images. Because of the small area of a TEM image, the FFT is typically noisy, making it difficult to precisely measure the featured lines corresponding to CNT diffractions. In conventional FFTs, periodic boundary conditions are assumed for the systems, which is not the case for the TEM image. As a result, artifacts due to the edges obscure the weak layer-line from CNTs. We have used Moisan's periodic + smooth decomposition method to eliminate the edge artifacts (29). A 2D polynomial background is fitted to the masked square region ($\pm 6.5 \text{ nm}^{-1}$) and subtracted to flatten the residual gradient. Clear layer lines could be seen from processed FFTs. As expected, FFTs are not sensitive to the focus conditions (Figures S4-5).

The distances of layer lines (D_1 , D_2 , D_3) to the equator were measured by fitting the projected intensity profile with multi-peak pseudo-Voigt model plus a linear baseline. D_1/D_2 and D_3 regions were separately fitted in an iterative algorithm up to 6 iterations. One of the difficulties is the small separation between D_1 and D_2 for near armchair type region. Peaks with fitted width

larger than 3 px were split into two narrower peaks, to resolve close peaks near armchair chiralities.

D1, D2 layer lines were tentatively assigned to two strong peaks within D1/D2 range [2.20, 4.84] nm⁻¹. Then the assignment was validated by calculating the chiral angle $\alpha = \arctan ((2D2 - D1) / (\sqrt{3} D1))$ to check if α lies in [0°, 30°]. Another cross-check was to compare $D3 = D1 - D2$ with measured D3 peak and flag the consistency between the two. Finally, special case of armchair ($D1 \approx D2$) was considered if there is no D3 peak detected corresponding to the D1-D2, and if there is only a dominant peak in D1/D2 range. Chiral angles of the CNTs are calculated by using the layer line method (24).

Example of the layer line measurement are presented in Figure 4 and Figure S6 for the simulated TEM image of (20,10) nanotube. In addition to the equator line, 4 peaks were detected on either side of the projected profile of the processed FFT (a), as marked by the dotted lines. The profile was further fitted in D1/D2 and D3 regions, where it is expected to find two peaks in D1/D2 region for calculating the chiral angle and to find a peak in D3 region to validate the two peaks in D1/D2 region. The FFT in the central area along with detected layer line positions, corresponding to the peaks in Figure 4(a) is plotted in Figure 4(d). Among the four detected peaks, three of them were identified as D1, D2 and D3 to satisfy the $D1-D2=D3$ requirement.

4. Chirality analysis of simulated TEM set

Based on precise measurement of diameter, layer lines and subsequently chiral angles, chiral indices of the CNTs were determined. Experimental circumference was calculated by $Ch_exp = diameter_nm \times \pi$, and experimental chiral angle using $\alpha_exp = \arctan ((2D2 - D1) / (\sqrt{3} D1))$. Then the experimental circumference and chiral angle was compared with pre-built lookup table, to find the best matched indices that satisfied with the following conditions: $|Ch_calc - Ch_exp| / Ch_exp < 7\%$ AND $|\alpha_calc - \alpha_exp| < 2.0^\circ$. The tolerance of acceptable circumference and angles can be adjusted in the analysis Jupyter notebooks, depending on specific calibration of the electron microscopes.

An example of chirality analysis of a simulated TEM image from CNT (20, 10) model is presented in table 1. Analysis results from conventional d_min and the measurement mode used in this work (d_ave) are compared for under and over focus conditions. As the defocus value was

varied from -4 nm to 4 nm, the measured chiral angle was very robust, showing a small deviation of 0.02 degrees, and very close to the actual angle of 19.1 degrees. On the other hand, the measured diameters were dependent on measurement modes and defocus values. For conventional d_min mode, while the measured diameter was close to the ground truth at underfocus condition, it showed a large error for the overfocus condition. On the other hand, d_ave values were robust against the change in defocus. As a result, the chiral indices were correctly assigned for d_ave mode at both focus conditions, while the result for d_min mode was only correct at underfocus condition.

Accuracy, robustness and efficiency of the “VibeTube” code have been tested using the simulated dataset, including 330 chiralities, ranging from (6, 0) to (25, 25), and 6930 simulated images in total. Statistical analysis results are presented in Figure 5, and Figures S7-S9, for different diameter modes and defocus ranges. For d_ave diameter mode, the overall accuracy was 84.5% (Figures S7), which was significantly higher than the accuracy of 44.9 % by using the conventional d_min mode (Figure S8). It is interesting to notice that the accuracy of d_min mode was very high (>90 %) in underfocus conditions, however the error rate was nearly 100 % for overfocus condition, due to the error in diameter measurement.

For d_ave mode used in the current work, the error rate was also very high when the defocus is near zero. It is understandable that when the focus is close to the Gaussian point, the contrast is minimized, causing confusion in diameter measurement. Therefore, it is recommended to avoid the near focus condition to take TEM images of CNTs. Within the range $2 \text{ nm} < |df| < 10 \text{ nm}$, the accuracy was as high as 92.3% (Figure 5). The accuracy was rather robust against the change in defocus (Figure 5a), in contrast to the behavior of d_min mode (Figure S9). As the diameter increases, it becomes more difficult to differentiate neighboring indices, and the error rate increases for nanotubes near 3 nm in diameter (Figure 5b). Nanotubes near zigzag and armchair regions are more challenging than chiral nanotubes (Figure 5c), mainly because of the special diffraction patterns with layer lines that are difficult to separate. Such chiral angle-dependent error rate was also clear from the chiral map (Figure 5d).

Within the recommended window ($2 < |df| < 10 \text{ nm}$, accuracy 92.3 %), the residual error is structural and concentrates on near-armchair and near-zigzag tubes (Fig. 5c,d). The chiral angle is obtained from the positions of the D1, D2 and D3 layer lines ($D3 = D1 - D2$). And these lines

coalesce at the two limits of the chiral-angle range — D1 and D2 merge toward armchair ($\alpha \rightarrow 30^\circ$, $D3 \rightarrow 0$), and D2 and D3 merge toward zigzag ($\alpha \rightarrow 0^\circ$, $D1 \rightarrow 2 \cdot D2$). When the layer lines get too close, they cannot be resolved reliably. The diameter, by contrast, is measured within tolerance. So, it is the chiral angle measurement, not the diameter, that limits accuracy in this regime. The pipeline handles these special cases with symmetry constraints, a $D1 - D2 = D3$ consistency check, and a dedicated armchair branch, and flags ambiguous cases for user inspection; a fully automatic treatment of the armchair/zigzag limit is left to future work.

5. Chirality analysis of experimental TEM set

Accuracy and robustness of the “VibeTube” code was further tested on an experimental dataset of HRTEM images of CNTs synthesized by a FCCVD method. Figure 6 shows the step-by-step treatment and analysis of one TEM image example after preprocessing. The background corrected image in Figure 6a shows extremely clear CNT features on a very homogeneous background. Normalized image intensity is then projected along the tube axis and used for the walls detection as shown in Figure 6b. Several oscillations corresponding to the two wall regions are clearly visible above a flat background. Those oscillations are used to determine $d_{\max}$, $d_{\min}$, $d_{\inf}$. While in this example the detected peaks correspond to the global maxima, the peaks are not simply assigned by using the maximal oscillation variation on each side. Here the selected fringes happen to be the global maxima, but they are not chosen simply as the largest oscillation on each side. As described in Section 2, all fringe peaks above a prominence threshold are considered, and the pair that best matches the expected wall symmetry (max–min–min–max at underfocus, reversed at overfocus) is selected. Following their detection, the average between $d_{\max}$ and $d_{\min}$ was calculated for d_{ave} . Following the CNT diameter extraction, boundary corrected and center-masked FFT of the preprocessed image was generated (Figure 6f) and its intensity projected along the equatorial line (Figure 6c). The signal is then smoothed and fitted to detect peaks in three distinct ranges of the projection. The first two ranges correspond to the two pairs of so-called D1 and D2 lines, on both sides of the equatorial line. Figure 6d shows an enlarged view of one range and the fits corresponding to D1 and D2. The third range is centered on the equator and includes the D3 lines pair, as enlarged on Figure 6e. Symmetry conditions on the pair peaks are applied here as well to account for the physics of electron diffraction of tubes. As visible on Figures 6c and f, the lines reported on the FFT and its projection, allowing users visual validation, are properly assigned. The chiral angle α is then calculated from the D1 and D2

line positions. Finally, the as mentioned determined diameter and angle couple is compared to the theoretical couple list for CNTs, and the chirality extracted from the best matching couple. In this example, a d_{ave} of 1.678 nm has been detected, D1 and D2 positions have been determined at 4.468 nm^{-1} and 3.834 nm^{-1} giving $\alpha = 22.5^\circ$, best matching a (15, 9) chirality.

5.1 Robustness: defocus

Robustness against defocus of the “VibeTube” code was tested with 31 images of one CNT, recorded at 14 different focus conditions, with defocus value ranging from -22.8 nm to +22.8 nm. The measured diameters are about ~ 2.2 nm for the defocus in the range from -11.4 nm to 7.6 nm. When the defocus was out of the range, the measured diameters showed a jump to ~ 2.6 nm (Figure 7a). It is understandable that as the focus is increased either in the under or over focus directions, more fringes will appear, making the identification of the correct wall positions difficult. Chiral angles are distributed within a narrow range of $25.1 \pm 0.4^\circ$, showing a higher robustness against the focus conditions (Figure 7b). Extracted (n, m) indices are presented in Figure 7c. Since diameter measurements are correct only within the defocus ranging from -11.4 nm to 7.6 nm, it is not surprising that the chiral indices also showed discontinuity at these defocus values. Within the defocus range [-11.4, 7.6] nm, 13 out of 15 images were assigned to (19, 14) and 2 were assigned to (18, 14). Since one of the (18, 14) results from the image with defocus at 0 nm, at which point the contrast is very low, the accuracy of the code in the proper defocus range is estimated to be $13/14 = 92.9\%$, which is consistent with the estimated accuracy from simulated dataset.

5.2 Statistical analysis and chirality map

The “VibeTube” code was used to analyze the chirality distribution of CNTs grown from FCCVD method, with a dataset of 67 TEM images from 50 unique CNTs. Images have been selected based on their quality, so that clear FFT patterns can be seen. As can be seen in Figure 8, the code outputs a distribution of 42 chiralities, with diameters ranging from ~ 1.4 nm to ~ 2.8 nm and chiral angles spread from $\sim 1.6^\circ$ to 30° . For 17 of the CNTs, two different images of the same tube have been included to test the consistency. In all the 17 cases, extracted chiralities are consistent for the two input images, showing the high reliability for practical applications.

6. Extension to double-walled CNTs and bundles

The pipeline of chirality extraction from TEM images is designed and validated for isolated single-walled carbon nanotubes. It detects the two wall fringes that define one diameter and the three layer lines on each side of the FFT equator that define one chiral angle. In practice, there are double-walled CNTs, and SWCNTs typically exist in the form of bundles. Accurate chirality assignment of more complex CNT materials is the basis to investigate the rich physics of the interactions, and to realize their chirality-dependent applications. Previously, electron diffraction has been applied to extract the chirality of each shell in multi-walled CNTs (22, 30, 31). Here, we have explored an extension of the method from isolated single-wall CNTs to extract the chirality of double-walled CNTs and bundles from simulated TEM images.

Although the analytical pipeline developed based on isolated SWCNTs does not apply directly to cases with more than one nanotube shell such as DWCNTs and bundles, the underlying measurements can be extended naturally. In the TEM image of a multi-wall CNT or multi-nanotubes, the number of wall fringes and layer lines in FFTs is proportional to the number of nanotube shells. One of the difficulties is detecting and pairing the wall fringes in TEM images with the layer lines in FFTs. It is noticed that the larger-diameter shell contains more atoms per projected length and therefore produces both the deeper wall fringes in TEM images and the higher-intensity layer lines in FFTs, so a larger diameter can be paired with the stronger layer-line set to determine the chirality of each shell. The above process was implemented as an additive module, leaving the validated single-wall pipeline unchanged. The analysis was tested on frozen-phonon multislice TEM simulations of a (10,5)@(20,3) DWCNT and a (10,5)+(20,3) bundle under different overlapping conditions.

Chirality indices of the nanotube shells in the DWCNT and bundle were extracted (Figures S10 and S11). For the DWCNT, the four dark wall fringes were detected in the projected wall profile and grouped by contrast depth. At underfocus condition, the diameters were measured by using the dark-fringe $d_{\min}$ mode, which avoids the ambiguous bright/dark pairing when several walls overlap. This gives two correct diameters (1.73 nm and 1.07 nm), without prior knowledge of how the walls are arranged in projection. The FFT shows six layer-lines (two in each of the D1, D2 and D3 sets). Taking the two strongest peaks per set and pairing them by the same $D3 = D1 - D2$ consistency yields two (D1, D2) triplets and hence two chiral angles (6.9° and 19.3°). The

larger measured diameter was paired with the stronger layer-line triplet. The result was (20,3) for the outer shell and (10,5) for the inner shell, matching the model structure exactly.

For the bundle, the chirality was extracted through a similar procedure. The chiral angles were robust against the overlapping conditions, because the FFTs are essentially identical irrespective of projected overlap, with the layer lines of both tubes superimposed. The diameters were measured correctly for the full-overlap and no-overlap cases, giving (20,3) and (10,5) for the two nanotubes in the bundle. Partial overlap was the most difficult case: the wall fringes interfere in the overlapped region, so the measured diameters were slightly off (1.74 nm and 1.08 nm). The larger tube was then assigned (21,3) rather than (20,3), one index off, while the smaller tube's chirality remained correct. These results show that, in principle, the pipeline designed for isolated SWCNTs can be extended to more complex CNT materials such as double-walled tubes and bundles. However, further work, using both simulations and experiments, is needed to address the wider range of conditions encountered in practice.

7. Practical usage guideline

For practical applications of the VibeTube codes to analyze TEM images to extract CNTs' chiralities, it is important to understand how the imaging parameters affect the data quality and accuracy. Practical usage guidelines have been made based on the investigations of the influence of three parameters, electron dose, pixel size and field of view (Figure S12), in addition to defocus influence which has been discussed in section 4.

The robustness test was based on simulation and analysis of a dataset with nanotube indices ranging from (20, 1) to (20, 19), spanning from near-zigzag to near-armchair ranges. The electron dose was applied as Poisson shot noise on the intensity. The pixel size was coarsened by area-averaged down-sampling. The field of view was reduced by re-simulating the tube at each length. The full procedure and code are provided with the released analysis notebook.

Electron dose (Fig. S12a) sets the overall signal-to-noise: the assignment is fully correct for doses $\geq 1000 \text{ e}/\text{\AA}^2$ — the range of standard HRTEM imaging (500–2000 $\text{e}/\text{\AA}^2$, shaded) — and degrades below a few hundred $\text{e}/\text{\AA}^2$, where the weak layer lines are the first features to disappear under the shot noise background. Pixel size (Fig. S12b) limits the diameter measuring precision: as the sampling coarsens the wall fringes are blurred, and the diameter is overestimated. The

accuracy stays $\sim 95\%$ up to 0.05 nm/px and collapses beyond 0.06 nm/px . Tube length (Fig. S12c) limits the layer-line measuring precision: a shorter tube has fewer lattice periods, so the layer lines broaden. The accuracy is 100% for lengths $\geq 15\text{ nm}$ but falls sharply for shorter tubes. The two-parameter maps (Fig. S12d,e) show that a high accuracy is associated with a high dose $\geq \sim 1000\text{ e/\AA}^2$, a small pixel size $\leq 0.05\text{ nm/px}$, and a long nanotube $\geq 15\text{ nm}$. Together with defocus $2 < |df| < 10\text{ nm}$, these parameters are the recommended conditions for accurate analysis of CNTs' chirality from TEM images.

Conclusions

In conclusion, an open-source Python pipeline is developed for automated chirality extraction of carbon nanotubes from TEM images with an accuracy exceeding 90% across diameters from 0.5 to 3.0 nm against a simulated dataset. The method combines symmetry-based diameter measurement with sub-pixel layer line analysis in Fourier space, enabling reliable extraction of chiral indices (n, m) . Validation with CNTs synthesized by floating catalyst CVD confirms the practical applicability of the codes to real experimental datasets. By releasing the “VibeTube” code as an open-source tool, we provide the CNT research community with an accurate, robust, efficient method for chirality characterization that can accelerate chirality-controlled growth and structure–property relationships.

Data availability

The data that support the findings of this study are openly available in NIMS-operated Materials Data Repository (MDR).

Code deposition

The code will be open source, and shared through GitHub, after publication of the paper.

Acknowledgements

Ms. Anping Wu is acknowledged for preparing carbon nanotube samples.

Funding details

This work was supported by the JSPS Kakenhi; under Grant [JP25820336], [JP20K05281], [JP23H01796], [JP26K17753]; JST-FOREST Program under Grant [JPMJFR223T]; WPI-MANA under [Challenging Research Program (CRP)]; NIMS under [Support system for curiosity-driven research program] and the “Advanced Research Infrastructure for Materials and Nanotechnology in Japan (ARIM)” program of the Ministry of Education, Culture, Sports, Science and Technology (MEXT) under Grant [JPMXP1225NM5167].

The authors report that there are no competing interests to declare.

References

1. R. Saito, M. Fujita, G. Dresselhaus, M. S. Dresselhaus, Electronic structure of chiral graphene tubules. *Appl Phys Lett* **60**, 2204-2206 (1992).
2. M. F. L. De Volder, S. H. Tawfick, R. H. Baughman, A. J. Hart, Carbon Nanotubes: Present and Future Commercial Applications. *Science* **339**, 535-539 (2013).
3. D.-M. Tang *et al.*, Semiconductor nanochannels in metallic carbon nanotubes by thermomechanical chirality alteration. *Science* **374**, 1616-1620 (2021).
4. D.-M. Tang *et al.*, Chirality engineering for carbon nanotube electronics. *Nature Reviews Electrical Engineering* **1**, 149-162 (2024).
5. A. Jorio *et al.*, Structural (n,m) determination of isolated single-wall carbon nanotubes by resonant Raman scattering. *Phys Rev Lett* **86**, 1118-1121 (2001).
6. A. Jorio *et al.*, Resonance Raman spectroscopy (n,m)-dependent effects in small-diameter single-wall carbon nanotubes. *Phys Rev B* **71**, 075401 (2005).
7. S. M. Bachilo *et al.*, Structure-assigned optical spectra of single-walled carbon nanotubes. *Science* **298**, 2361-2366 (2002).
8. D. I. Levshov *et al.*, Photoluminescence from an individual double-walled carbon nanotube. *Phys Rev B* **96**, 195410 (2017).
9. M. Y. Sfeir *et al.*, Probing Electronic Transitions in Individual Carbon Nanotubes by Rayleigh Scattering. *Science* **306**, 1540-1543 (2004).
10. K. Liu *et al.*, An atlas of carbon nanotube optical transitions. *Nat Nanotech* **7**, 325-329 (2012).
11. J. W. G. Wilder, L. C. Venema, A. G. Rinzler, R. E. Smalley, C. Dekker, Electronic structure of atomically resolved carbon nanotubes. *Nature* **391**, 59-62 (1998).
12. S. Iijima, Helical microtubules of graphitic carbon. *Nature* **354**, 56-58 (1991).
13. M. Haider *et al.*, Electron microscopy image enhanced. *Nature* **392**, 768-769 (1998).
14. A. Hashimoto, K. Suenaga, A. Gloter, K. Urita, S. Iijima, Direct evidence for atomic defects in graphene layers. *Nature* **430**, 870 (2004).
15. K. Suenaga *et al.*, Imaging active topological defects in carbon nanotubes. *Nat Nano* **2**, 358-360 (2007).

16. Y. Sato *et al.*, Chiral-Angle Distribution for Separated Single-Walled Carbon Nanotubes. *Nano Lett* **8**, 3151-3154 (2008).
17. J. H. Warner, N. P. Young, A. I. Kirkland, G. A. D. Briggs, Resolving strain in carbon nanotubes at the atomic level. *Nat Mater* **10**, 958 (2011).
18. K. Mustonen *et al.*, Atomic-Scale Deformations at the Interface of a Mixed-Dimensional van der Waals Heterostructure. *ACS Nano* **12**, 8512-8519 (2018).
19. Y. Yu *et al.*, Determine the Complete Configuration of Single-Walled Carbon Nanotubes by One Photograph of Transmission Electron Microscopy. *Advanced Science* **10**, 2206403 (2023).
20. P. Lambin, A. A. Lucas, Quantitative theory of diffraction by carbon nanotubes. *Phys Rev B* **56**, 3571-3574 (1997).
21. L. C. Qin, T. Ichihashi, S. Iijima, On the measurement of helicity of carbon nanotubes. *Ultramicroscopy* **67**, 181-189 (1997).
22. M. Kociak, K. Hirahara, K. Suenaga, S. Iijima, How accurate can the determination of chiral indices of carbon nanotubes be? *European Physical Journal B: Condensed Matter Physics* **32**, 457-469 (2003).
23. Z. Liu, L.-C. Qin, Symmetry of electron diffraction from single-walled carbon nanotubes. *Chem Phys Lett* **400**, 430-435 (2004).
24. L.-C. Qin, Electron diffraction from carbon nanotubes. *Rep Prog Phys* **69**, 2761 (2006).
25. H. Jiang, A. G. Nasibulin, D. P. Brown, E. I. Kauppinen, Unambiguous atomic structural determination of single-walled carbon nanotubes by electron diffraction. *Carbon* **45**, 662-667 (2007).
26. G. D. Förster *et al.*, A deep learning approach for determining the chiral indices of carbon nanotubes from high-resolution transmission electron microscopy images. *Carbon* **169**, 465-474 (2020).
27. C. Qin, L. M. Peng, Measurement accuracy of the diameter of a carbon nanotube from TEM images. *Phys Rev B* **65**, 155431 (2002).
28. R. Fleurier, J.-S. Lauret, U. Lopez, A. Loiseau, Transmission Electron Microscopy and UV-vis-IR Spectroscopy Analysis of the Diameter Sorting of Carbon Nanotubes by Gradient Density Ultracentrifugation. *Adv Funct Mater* **19**, 2219-2223 (2009).
29. L. Moisan, Periodic Plus Smooth Image Decomposition. *J Math Imaging Vis* **39**, 161-179 (2011).
30. Z. Liu, Q. Zhang, L.-C. Qin, Accurate determination of atomic structure of multiwalled carbon nanotubes by nondestructive nanobeam electron diffraction. *Appl Phys Lett* **86**, 191903 (2005).
31. H. Deniz, A. Derbakova, L.-C. Qin, A systematic procedure for determining the chiral indices of multi-walled carbon nanotubes using electron diffraction—each and every shell. *Ultramicroscopy* **111**, 66-72 (2010).
32. S. Jiang *et al.*, Ultrahigh-performance transparent conductive films of carbon-welded isolated single-wall carbon nanotubes. *Sci Adv* **4**, (2018).
33. J. Madsen, T. Susi, abTEM: A Fast and Flexible Python-based Multislice Simulation Package for Transmission Electron Microscopy. *Microsc Microanal* **29**, 680-680 (2023).

Figures and table

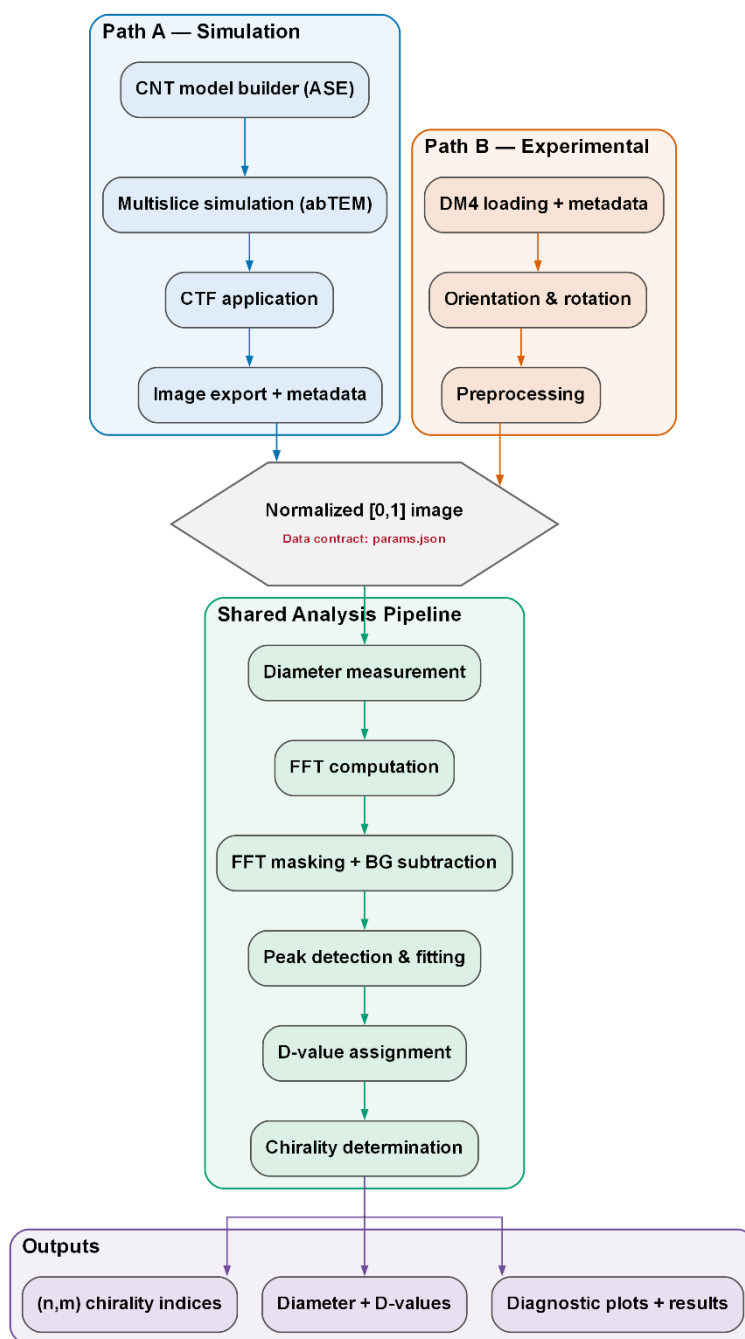


Figure 1. Pipelines of “VibeTube” codes for extracting chirality from CNTs’ TEM images.

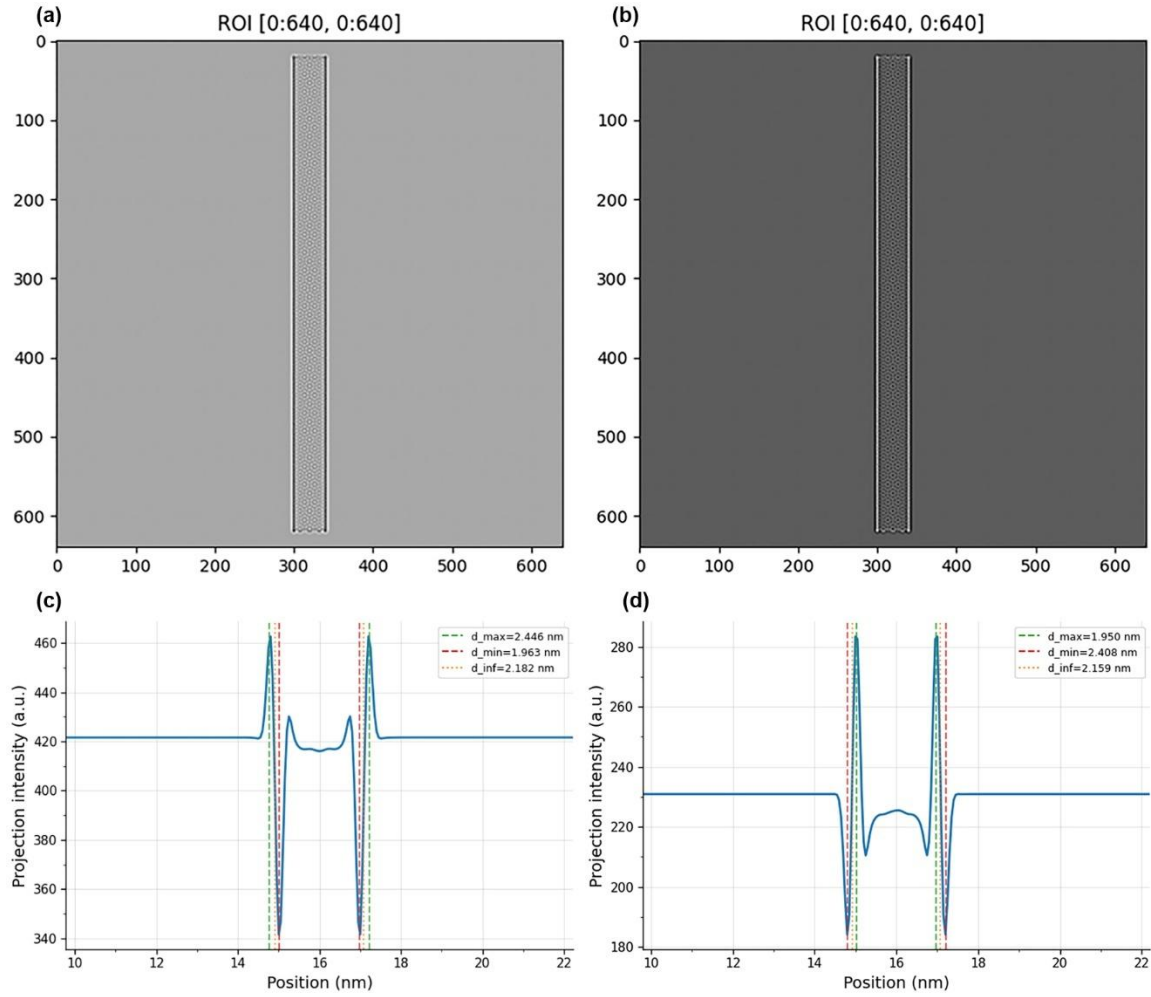


Figure 2. Diameter measurement by using d_{ave} mode. (a-b) Simulated TEM images of (20, 10) CNT at under ($df = 4$ nm) and over ($df = -4$ nm) focus conditions. (c-d) Projected intensity profiles and detected max, min and inflection points.

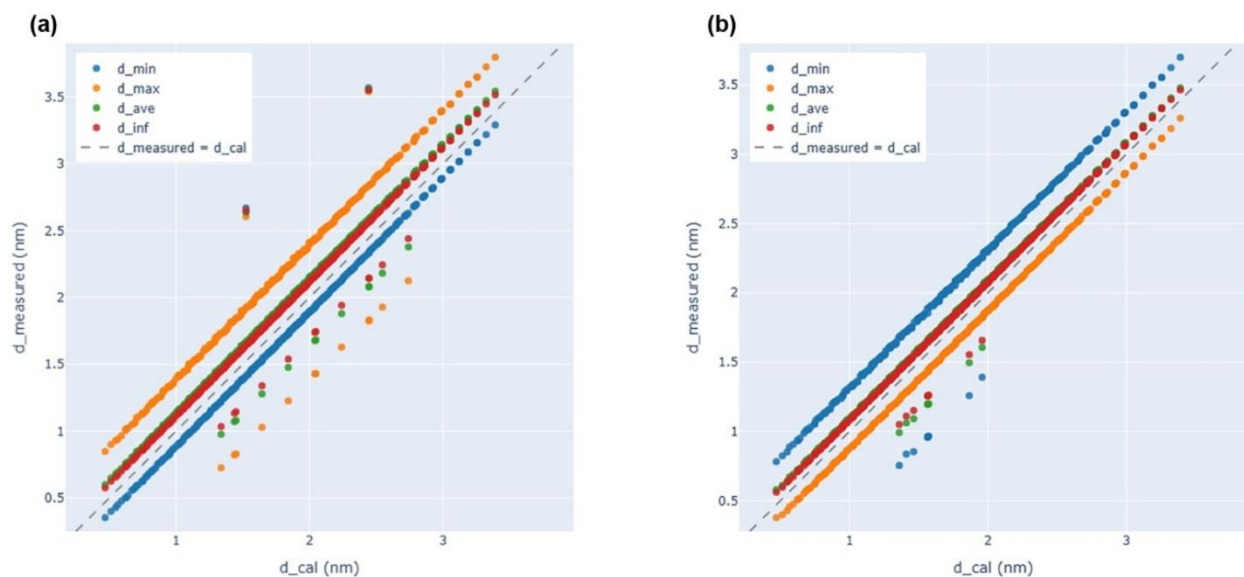


Figure 3. Diameter-offsets for different measurement methods. (a) Measured d_{min} , d_{max} , d_{ave} and d_{inf} as functions of d_{cal} with defocus of 2 nm (underfocus). (b) Measured d_{min} , d_{max} , d_{ave} and d_{inf} as functions of d_{cal} with defocus of -2 nm (overfocus).

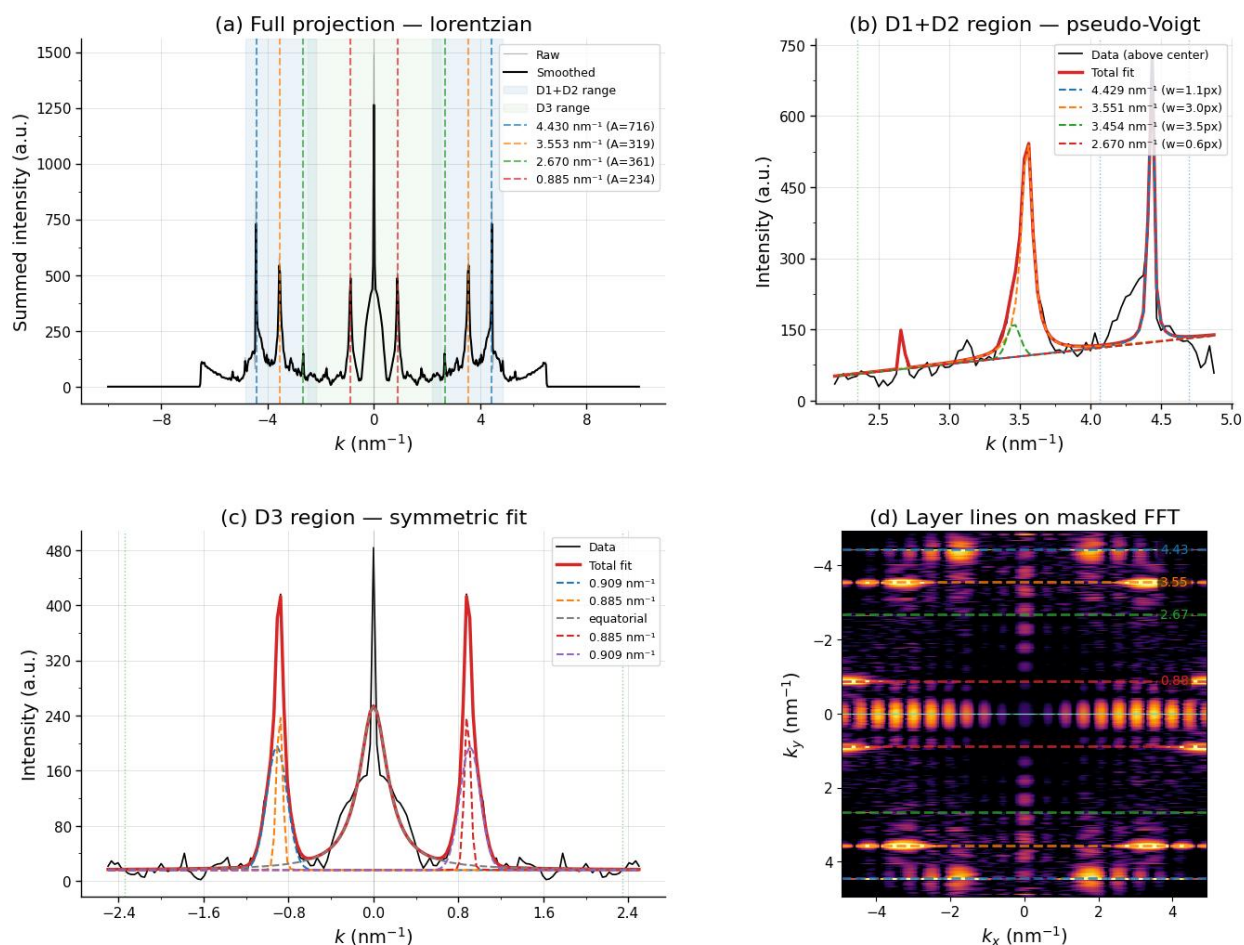


Figure 4. Layer line detection and measurement of the FFT from (20, 10) CNT at an underfocus condition. (a) Projection profile of the processed FFT with multiple peaks detected. (b) Fitting and peak detection in D1/D2 region by using a pseudo-Voigt method. (c) Fitting in D3 region by using the symmetric algorithm. (d) FFT in the central area with detected peak positions marked, corresponding to the peaks in (a).

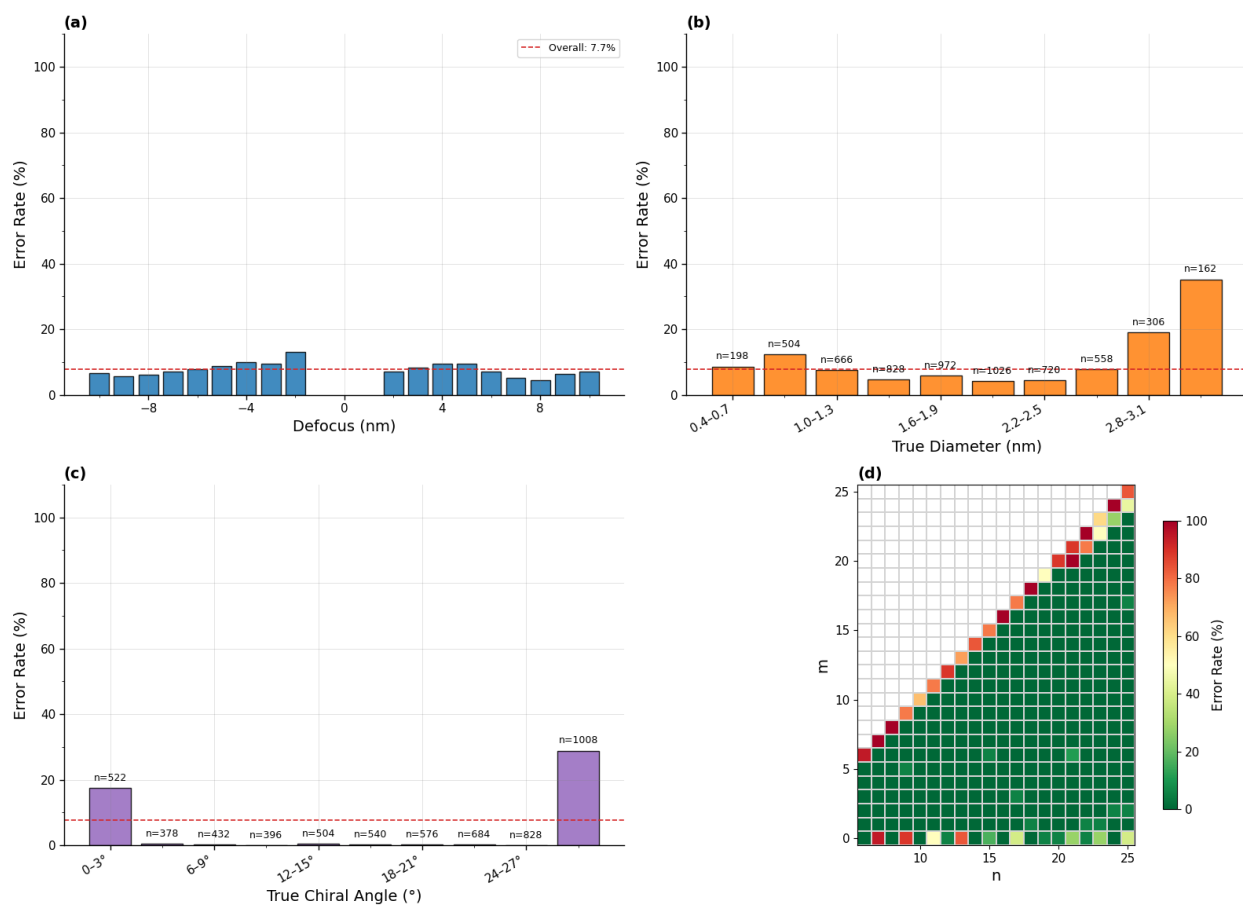


Figure 5. Error analysis for simulated dataset with diameter measured by using d_{ave} mode and excluding $|df| < 2$ nm. (a) Error rate as a function of defocus. (b) Error rate as a function of tube diameter. (c) Error rate as a function of chiral angle. (d) Error rate as a function of chiral indices (n, m).

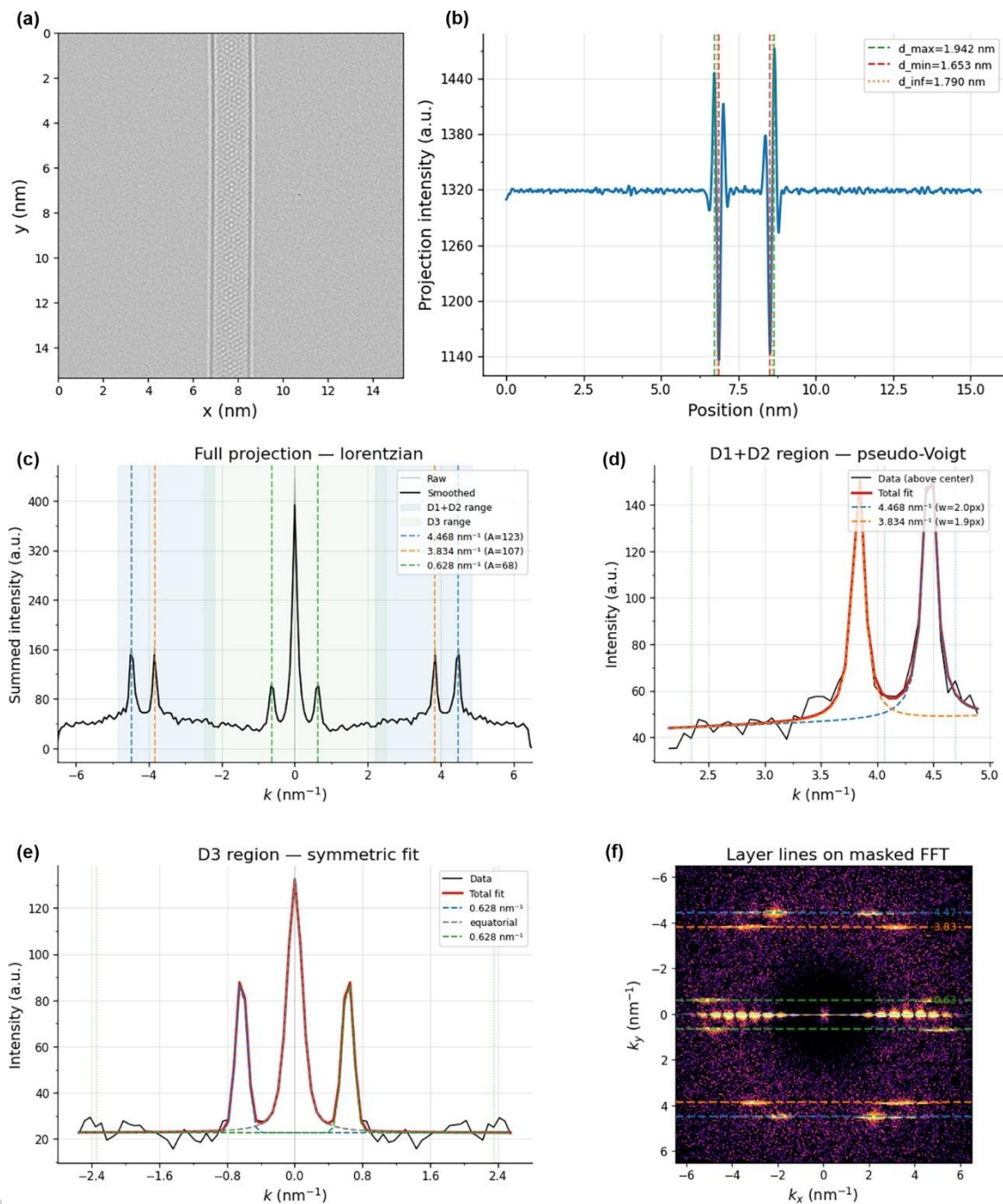


Figure 6. Chirality analysis of an experimental TEM image of CNT. (a) Preprocessed TEM image. (b) Projected profile and detected $d_{\min}$, $d_{\max}$, and $d_{\inf}$ for evaluating the diameter. (c) Projected profile of FFT layer lines and detected peaks. (d-e) Peak fitting and detection of D1, D2 and D3 layer lines. (f) Processed FFT with detected D1, D2 and D3 layer lines labelled.

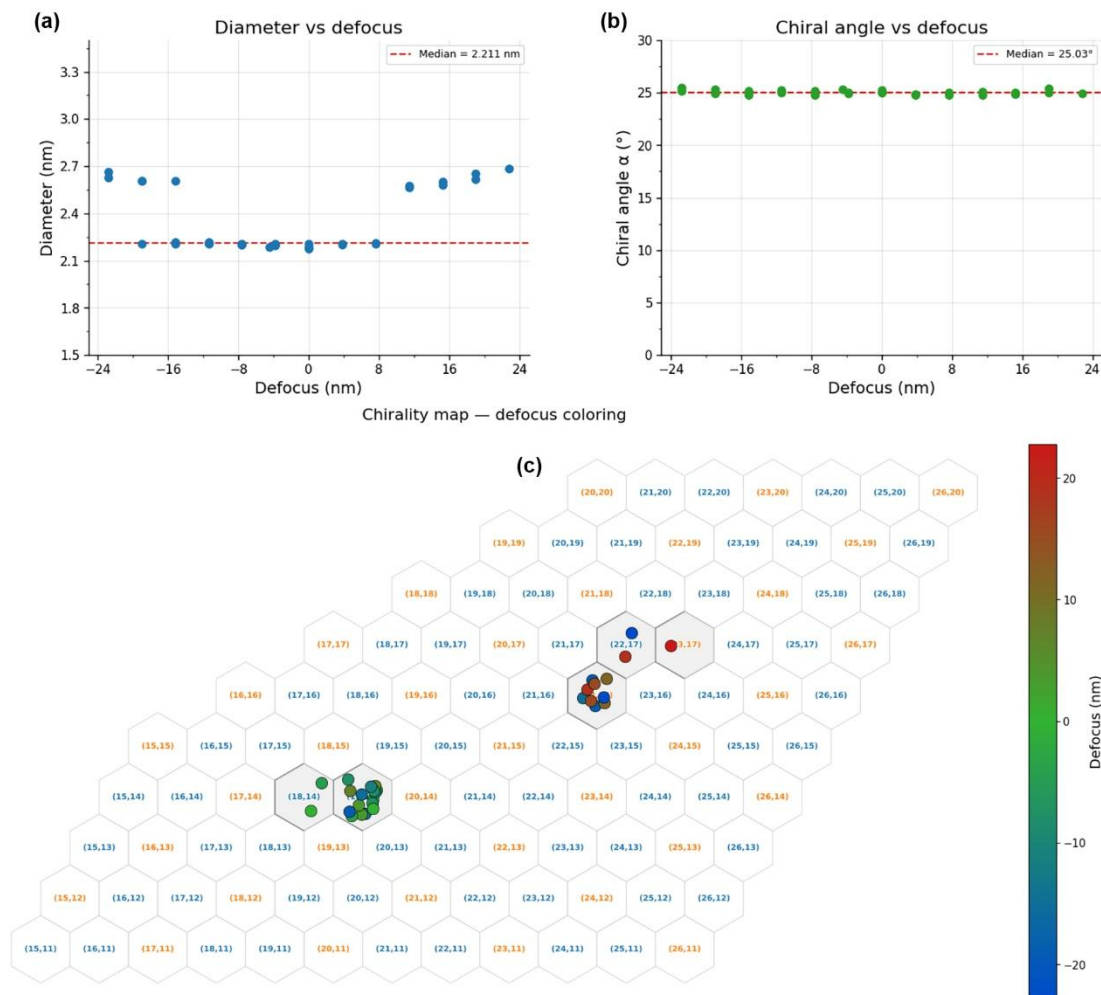


Figure 7. Robustness test of “VibeTube” codes on a CNT (experimental image) with defocus series. (a) Measured diameter as a function of defocus. (b) Measured angle as a function of defocus. (c) Calculated (n, m) as a function of the defocus, where the defocus value is indicated by a color bar.

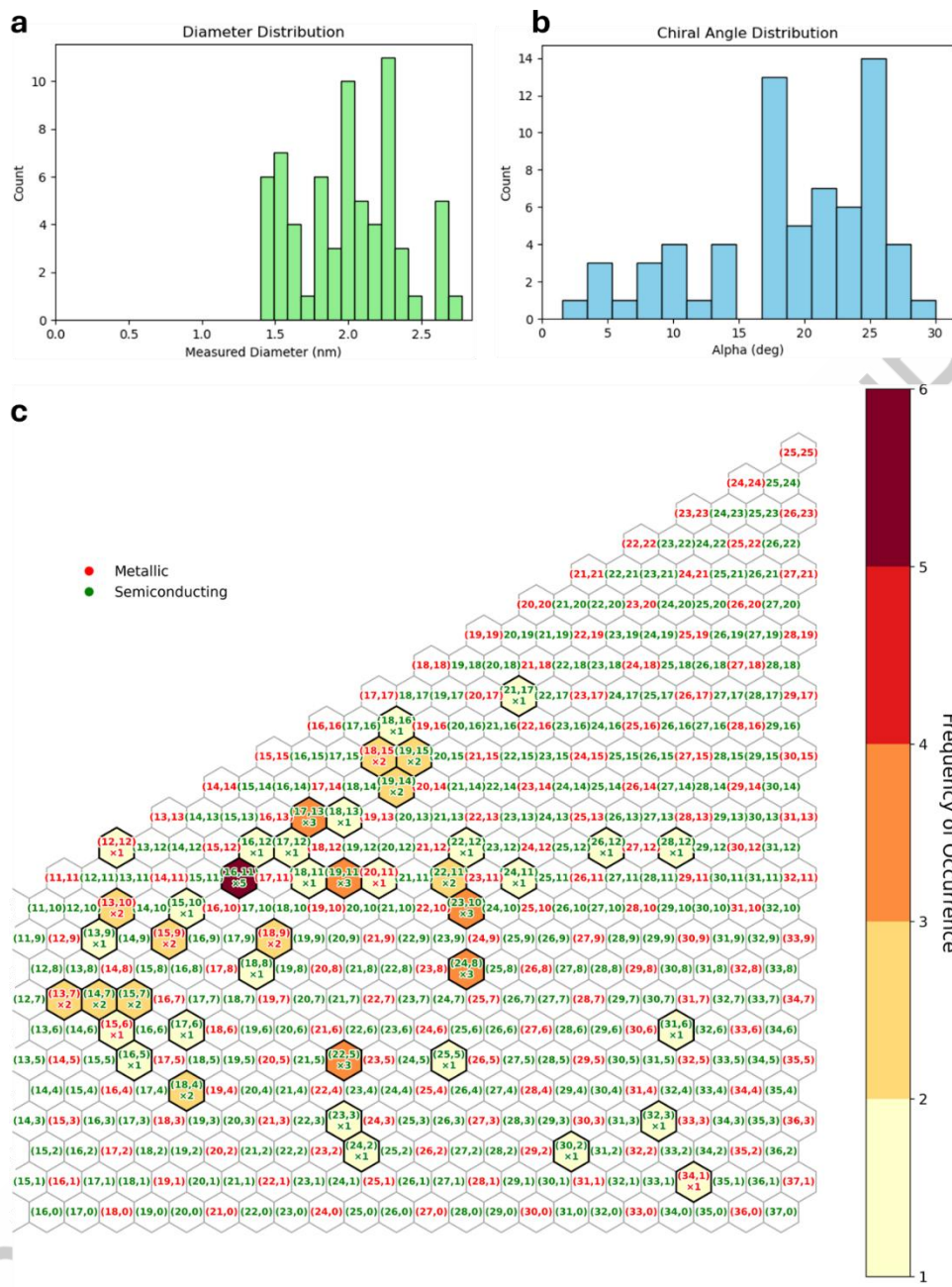


Figure 8. Statistical analysis of chirality distribution of CNTs grown by FCCVD. (a-b) Distribution of measured diameters and chiral angles. (c) Chirality distribution with the frequency of occurrence indicated by the color.

Table 1. Chirality analysis of a simulated TEM from CNT (20, 10).

CNT model	Defocus (nm)	Diameter mode	Diameter (nm)	Alpha (deg)	n	m	Match
(20, 10)	4	d_ave	2.08	19.22	20	10	1
(20, 10)	-4	d_ave	2.06	19.24	20	10	1
(20, 10)	4	d_min	2.06	19.22	20	10	1
(20, 10)	-4	d_min	2.51	19.24	24	12	0

Supporting information

Methods

1. SWCNTs growth and collection

SWCNT networks were prepared by an injection floating catalyst chemical vapor deposition (FCCVD) method and transferred onto TEM grids directly. The detailed synthesis process can be found in previous report (32). Briefly, CVD growth was conducted inside a horizontal tubular furnace. C_2H_4 and H_2 were used as the carbon source and carrier gas, respectively. Floating catalyst precursor (ferrocene) was injected into the reactor by syringe pump. Temperatures in the furnace and at the position of the injection needle were 1100 °C and 83 °C, respectively. Collection time was about 20 seconds to obtain SWCNT networks containing high-purity, individual nanotubes.

2. Preprocessing TEM images

The TEM used in this work was a JEOL JEM-ARM300F operated at 80 kV, with aberration correctors for both TEM and STEM modes. TEM images were taken using a Gatan OneView camera, with 4k x 4k resolution. To take lattice-resolved TEM images of carbon nanotubes, the magnifications were set to 1 million to 2 million times. To obtain TEM image of defocus series, the focus was changed by objective lens focus.

Experimental TEM images of CNTs were preprocessed as a separate step, to produce the same normalized format as simulated images, and to prepare an experimental dataset that can be reused for various analysis. Through preprocessing experimental and simulated TEM images can use identical analysis pipeline to extract CNT chirality.

The pipeline for preprocessing mainly includes (1) tube orientation in coarse and fine modes, (2) rotation to vertical orientation, (3) Region of Interest (ROI) selection, (4) Background subtraction and normalization. A two-stage orientation detection was used to precisely rotate the TEM image so that the nanotube axis is along the vertical direction. In stage 1 (coarse): the user clicks two points along the tube axis in a matplotlib GUI window. The orientation angle is computed as $\text{atan2}(dy, dx)$ from the clicked points. In stage 2 (fine sweep): an area defined by the user is rotated within 5° around the coarse angle in 0.1° steps. At each step the projection standard deviation was computed. The orientation angle was determined at the angle with

maximum standard deviation in the projection profile. As a result, the orientation was accurate to $\sim 0.1^\circ$. Two measures have been taken to improve robustness: (1) a margin of $\max(H,W) \times \sin(\theta)/2$ pixels was cropped to eliminate edge artifacts; (2) Gaussian background subtraction ($\sigma=10$) is applied to remove gradients in background that would otherwise dominate the signal.

3. Simulation of TEM images

High-resolution transmission electron microscopy (HRTEM) images of single-walled carbon nanotubes (SWCNTs) were simulated using the multislice method. The simulated images were used as a dataset for testing diameter measurement methods and for calibrating the diameter offset during chirality analysis. The dataset was used for testing the accuracy, speed and robustness of chirality analysis codes.

Nanotube models were constructed by using Atomic Simulation Environment (ASE), from chirality indices (n, m) with C–C bond length 1.42 Å. The original ASE's `ase.build.nanotube()` function has a hard-coded limit $nk = 6000$ that prevents construction of large-index tubes. A runtime patch was made in this project to avoid the nk limit, so that all (n, m) nanotube models could be constructed. The size of the cube-shaped supercell was 32 nm. The real space sample was 0.5 Å/px. And the lengths of nanotubes were 30 nm for all chiral indices.

The chiral indices ranged from $(6, 0)$ to $(25, 25)$, with a diameter ranging from 0.47 nm to 3.39 nm, corresponding to 330 unique chiralities. For each nanotube, the defocus varied from -10 to +10 nm. Therefore, 21 TEM images were simulated for each nanotube, and the dataset included 6,930 simulated TEM. An incremental save mechanism was used to write the `summary.csv` every 50 tubes to avoid data loss in case of crash.

The simulation uses the frozen phonon, multislice algorithm implemented in abTEM (33). Multiple atomic configurations with random thermal displacements (RMS $\sigma = 0.1$ Å) were generated to simulate thermal diffuse scattering. A plane electron wave at the specified energy (60 kV) is propagated slice-by-slice, alternating transmission (phase grating) and free-space propagation (Fresnel propagator via FFT) to obtain a complex wave function at the exit surface. TEM images were generated by converting the exit wave through the contrast transfer function (CTF): $I(r) = |\text{FT}^{-1}[\text{FT}[\psi_{\text{exit}}] \times \text{CTF}(k)]|^2$, where mainly the influences of defocus (Δf) and spherical aberration (C_s , 1 μm) were included.

It is noticed that the exit wave does not depend on the CTF. For a defocus series of N values on the same (n, m) tube, the exit wave was simulated once and then applied N times with the CTF including a defocus value. The defocus convention in this project is based on the reference of the nanotube center, instead of the bottom surface of the supercell, which is the convention of abTEM software. Therefore, corresponding to the defocus ranging from -10 nm to 10 nm, the CTF defocus at cell bottom was from +6.0 nm to +26.0 nm.

Since there are multiple Fourier transformations, GPU has advantage over CPU for TEM image simulations. On the specific hardware platform, the GPU was tested to be 14.5 times faster than the CPU with 1 core, and about 4.5 times faster than the CPU with 16 cores. For the simulations carried out by CPU, the performance plateaued at 16 workers due to saturated memory bandwidth. The time to simulate the 21 TEM images for one nanotube was about 7.8 seconds. And the total time to simulate the 6930 TEM images for the 330 nanotubes was about 43 minutes.

4. Measurement of CNT diameter

Precise measurement of CNT diameter is basic but practically complex and challenging. In this work, different approaches have been tested to get accurate and robust diameter measurements. The four diameter modes are: (1) d_{max} : distance between two intensity maxima, corresponding to the bright-fringe spacing, (2) d_{min} : distance between two intensity minima, corresponding to the dark-wall spacing, (3) d_{inf} : distance between inflection points, corresponding to the steepest-gradient positions, and (4) d_{ave} : $(d_{\text{max}} + d_{\text{min}}) / 2$, the average value of d_{max} and d_{min} .

The most challenging task for diameter measurement is the detection of the correct fringes that are associated with CNT walls. With the focus condition to be under focus ($df > 0$) it is expected that the CNT wall is dominated with a dark fringe along with a weaker bright fringe outside. The contrast is reversed when the focus is changed to be over focus condition. What makes it more complex is that between CNT walls, there are also interference fringes due to the lattices of upper and lower walls. Such fringes may show strong contrast, and sometimes even stronger than the wall fringes. Therefore, simple algorithms based on global detection of intensity maxima do not work.

In this work, a new algorithm was designed to detect the correct wall fringes by using symmetric patterns and local variance. The pipeline for diameter measurement mainly includes: (1) Smooth projection with Gaussian filter ($\sigma = 2$ px), (2) Estimate wall regions from local variance, (3) Dual search for wall fringes: Path A anchors on wall minima and finds symmetric maxima; Path B anchors on wall maxima and finds symmetric minima — the path with the higher symmetry score wins; (4) Sub-pixel refinement by a parabola fitting; (5) $d_{\min}$, $d_{\max}$ and d_{ave} calculation with detected maxima fringes; (6) d_{inf} was obtained by finding the zero-crossing of second derivative between max–min pair.

5. Detection of FFT layer-lines and chirality assignment

The chiral angle is calculated in reciprocal space from the layer lines of the fast Fourier transform (FFT) of the TEM image. The pipeline for FFT layer-line detection mainly includes: (1) a boundary-corrected FFT computed with Moisan's periodic-plus-smooth decomposition to remove the edge-artifact cross; (2) refinement of the equator to the brightest row within ± 2 px of the geometric center; (3) masking to a square of half-width 6.5 nm^{-1} about the center, followed by a two-dimensional polynomial background fit and subtraction to flatten the residual gradient; (4) projection of the processed FFT onto the layer-line axis (perpendicular to the equator) to give a one-dimensional profile; (5) fitting of the profile with a sum of pseudo-Voigt peaks plus a linear baseline, with the D1/D2 and D3 regions fitted separately and iteratively (up to 6 iterations, initial peak width 2 px). Detected peaks are kept only within $[0.5, 4.85] \text{ nm}^{-1}$, with width 0.3–5.0 px and amplitude above 5 % of the profile maximum; any peak wider than 3 px is split into two narrower peaks so that close near-armchair D1/D2 lines can be resolved.

The layer lines are then assigned as follows: (1) peaks within the combined D1/D2 range $[2.20, 4.84] \text{ nm}^{-1}$ are taken as layer-line candidates; (2) the two strongest by intensity are selected, and the higher-frequency one is assigned D1, the lower D2 ($D1 \geq D2$ by definition); (3) the assignment is validated by requiring $\alpha = \arctan[(2D2 - D1)/(\sqrt{3} D1)]$ to lie in $[0^\circ, 30^\circ]$; (4) D3 is cross-checked in the range up to 2.35 nm^{-1} : a measured D3 peak that matches $D1 - D2$ within $2 \Delta k$ confirms the (D1, D2, D3) consistency; (5) special cases near armchair region are handled explicitly — if $D1 \approx D2$, or if a single dominant peak is found in the D1/D2 range with no real peak in the D3 region, the tube is flagged as armchair type ($D2 = D1$, $D3 = 0$); (6) the chiral

angle is computed from the validated D1 and D2, and the (n, m) indices are assigned from the diameter and chiral angle.

6. Simulation and analysis of DWCNT and CNT bundle

Multi-tube models were built from the same fixed-geometry single-wall builder (ASE, with C–C bond length 1.42 Å). The DWCNT was constructed as two concentric shells, inner (10,5) and outer (20,3). The bundle was constructed from tubes (10,5) and (20,3) placed parallel and kept in contact at a fixed centre-to-centre distance of 1.706 nm. The projected (in-image) separation Δy was set to 0, 0.85 and 1.60 nm to give full, partial and no projected overlap.

TEM images were simulated with the frozen-phonon multislice method with 8 configurations, RMS displacement $\sigma = 0.1$ Å, plane-wave illumination, exit wave computed then combined with the contrast transfer function. The accelerating voltage was 60 kV, and the pixel size was 0.0127 nm/px, fine enough to resolve the ~ 0.33 nm inter-wall spacing. Images were simulated at +4 nm defocus (underfocus).

The projected wall profile was obtained. A DWCNT or two-tube bundle presents four dark wall fringes rather than two. These were detected and grouped by contrast depth, the two deepest fringes belonging to the larger-diameter shell and the two shallowest to the smaller. Each shell diameter was taken as the separation of its two dark fringes (d_min mode) plus a fixed 0.10 nm offset, which is accurate at underfocus condition.

The FFT was computed and cleaned as for a single tube following periodic-plus-smooth decomposition, central-area masking, and polynomial background subtraction. A two-shell nanotube (either DWCNT or bundle) produces six layer-lines, two in each of the D1, D2 and D3 sets. In each set the two strongest peaks were retained, and the six peaks were paired into two (D1, D2, D3) triplets using the same $D3 = D1 - D2$ consistency as the single-wall method. Each triplet gives one chiral angle through the standard relation $\alpha = \arctan[(2D2 - D1)/(\sqrt{3} D1)]$.

The two diameters and two chiral angles were paired by a physical rule: the larger-diameter shell contains more atoms per projected length and therefore produces deeper wall fringes and higher-intensity layer lines, so the larger measured diameter was paired with the stronger layer-line triplet. Each (diameter, angle) pair was then assigned (n, m) with the same geometry lookup used for SWCNTs.

ACCEPTED MANUSCRIPT

7. Robustness analysis by simulation

In addition to the experimental repeatability test, robustness was assessed by simulation to consider the influence from electron dose (shot noise), camera pixel size, and field of view (tube length). Dose and pixel size are detector effects, so they were applied as CPU post-processing to one clean simulation per tube. Tube length changes the specimen model (atom count, ends, number of lattice periods) and so requires a fresh multislice per length on the GPU. The test set was the single family (20,1)–(20,19), which sweeps the chiral angle from 2.42° to 29.15° and the diameter from 1.61 to 2.65 nm. Every image was analyzed by the existing pipeline without modification — automatic region-of-interest selection, wall-projection diameter (d_ave mode), FFT with center finding, central-area masking and background subtraction, Lorentzian layer-line fitting, D-value assignment, and geometry lookup.

One clean image per tube was produced by frozen-phonon multislice (8 configurations, RMS displacement $\sigma = 0.1$ Å, plane-wave illumination, exit wave then combined with the CTF) at 60 kV, $C_s = 1$ μm, $C_c = 1$ mm, energy spread 0.35 eV and objective semi-angle cutoff 45 mrad, for a 30 nm tube in a 32 nm supercell at 0.0127 nm/px (2520 × 2520 px). Images were formed at +4 nm defocus (underfocus) referenced to the tube-center plane.

To simulate the influence from electron dose, Poisson shot noise was applied to the clean image. For a dose D ($e/\text{Å}^2$) and pixel size p (nm) the counts per pixel are $N = D \cdot (10p)^2$, drawn from a Poisson distribution, so the signal-to-noise ratio scales as $\sqrt{N}$. Doses from 10 to 10 000 $e/\text{Å}^2$ were tested at 0.05 nm/px; for reference, standard HRTEM imaging uses of order 500–2000 $e/\text{Å}^2$, low-dose imaging tens to a hundred, and ultra-low-dose work about 8. To simulate the influence of camera pixel size, a coarser camera was modelled by area-weighted, anti-aliased down-sampling of the fine clean image to the target pixel size. Ten pixel-sizes from 0.0127 to 0.10 nm were tested. The influence of field of view was simulated with nanotubes of different lengths. Each length was re-simulated, with all parameters unchanged, except for the length. Five lengths from 5 to 30 nm were tested at 0.05 nm/px.

Supporting figures

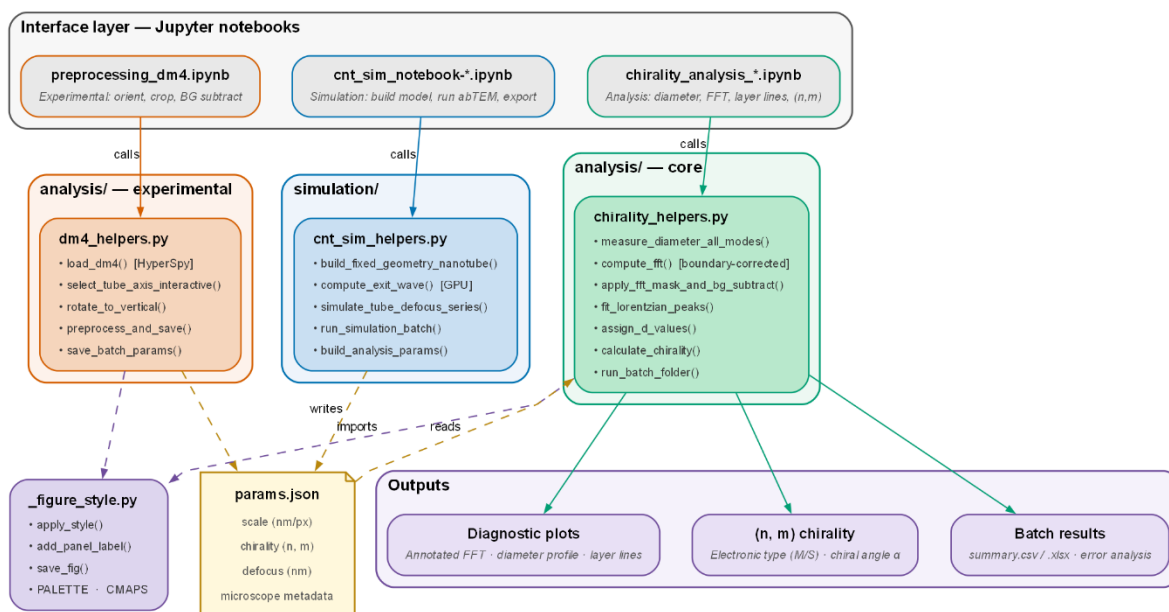


Figure S1. Architecture of the “VibeTube” code for extracting CNTs’ chirality from TEM images.

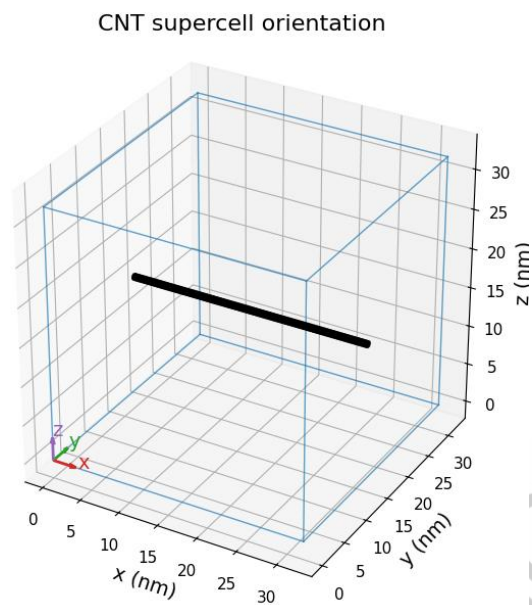


Figure S2. CNT supercell for TEM image simulation. The supercell is a cube with a size of 32 nm, with the nanotube at the middle, which defines the reference level of defocus. The lengths of nanotubes are fixed at 30 nm for all tubes, regardless of the size of the unit cell, which depends closely on the chiral indices (n , m). Nanotube axis is along x direction and perpendicular to y direction. The electron beam propagates along z direction.

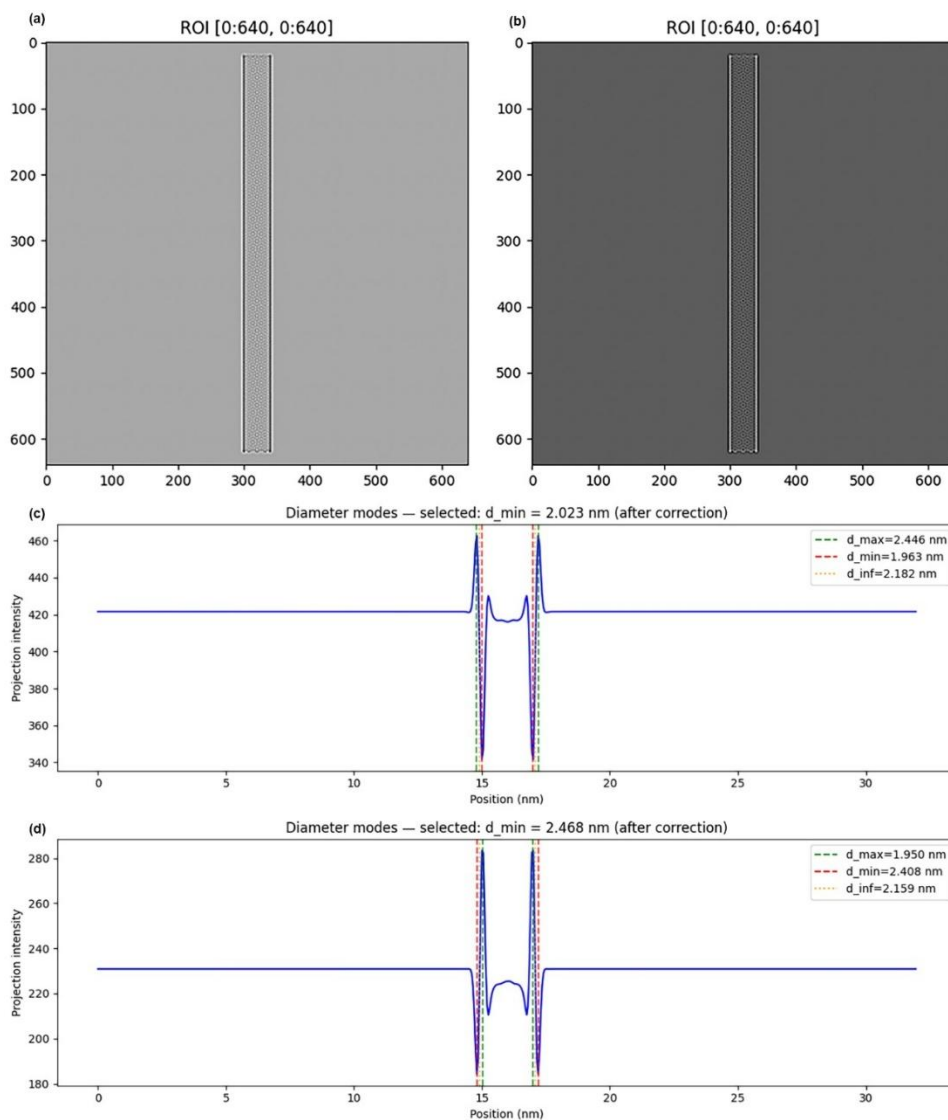


Figure S3. Diameter measurement by using $d_{\min}$ at under and over focus conditions. (a-b) simulated TEM images of (20, 10) CNT at under and over focus conditions, with df at 4 nm and -4 nm, respectively. (c-d) Projected intensity profile, detected max and min positions corresponding to the CNT wall fringes, and diameters measured by using $d_{\min}$ method.

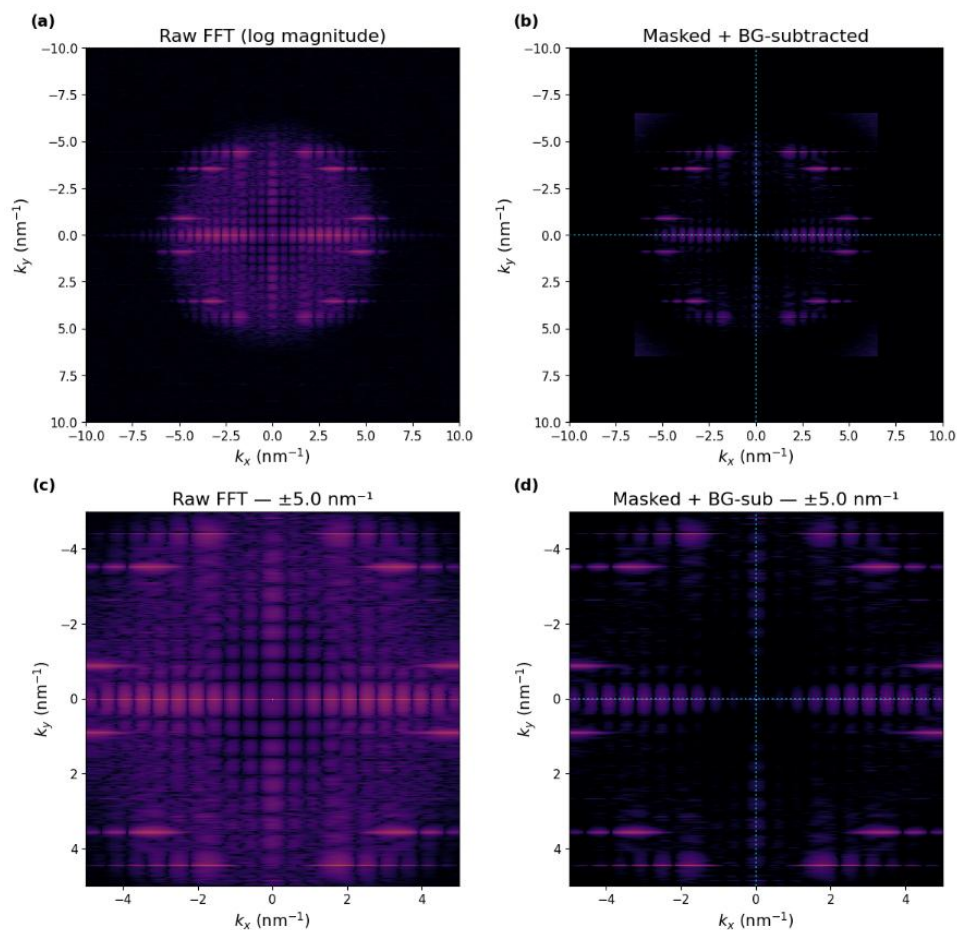


Figure S4. Processing FFT for simulated (20, 10) CNT with defocus at +4 nm. (a-b) and (c-d) show the FFT in full size and in the central area, respectively. The raw FFT is plotted in logarithmic scale (a, c). After masking the central area with a square of 6.5 nm^{-1} , and subtracting the background gradients by Gaussian method, the layer lines appear clearly in the central area (b, d).

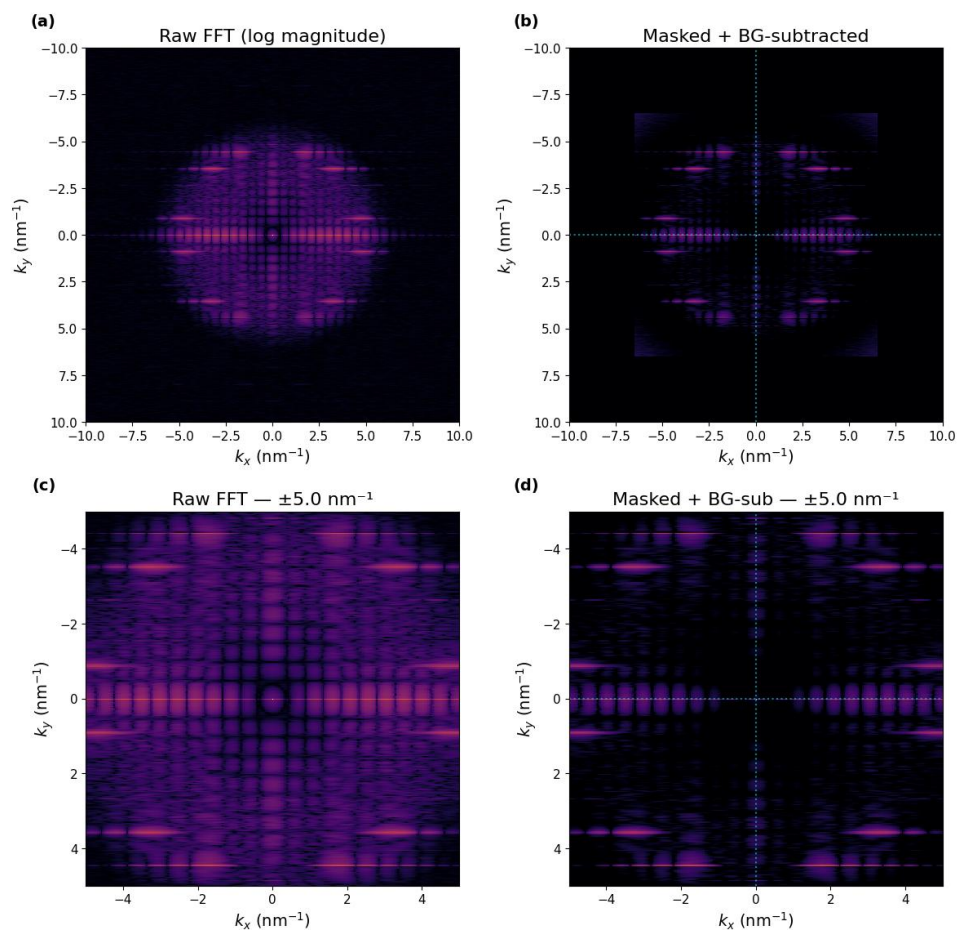


Figure S5. Processing FFT for simulated (20, 10) CNT with defocus at -4 nm. (a-b) and (c-d) show the FFT in full size and in the central area, respectively. The raw FFTs are plotted in logarithmic scale (a, c). After masking the central area with a square of 6.5 nm^{-1} , and subtracting the background gradients by Gaussian method, the layer lines appear clearly in the central area (b, d).

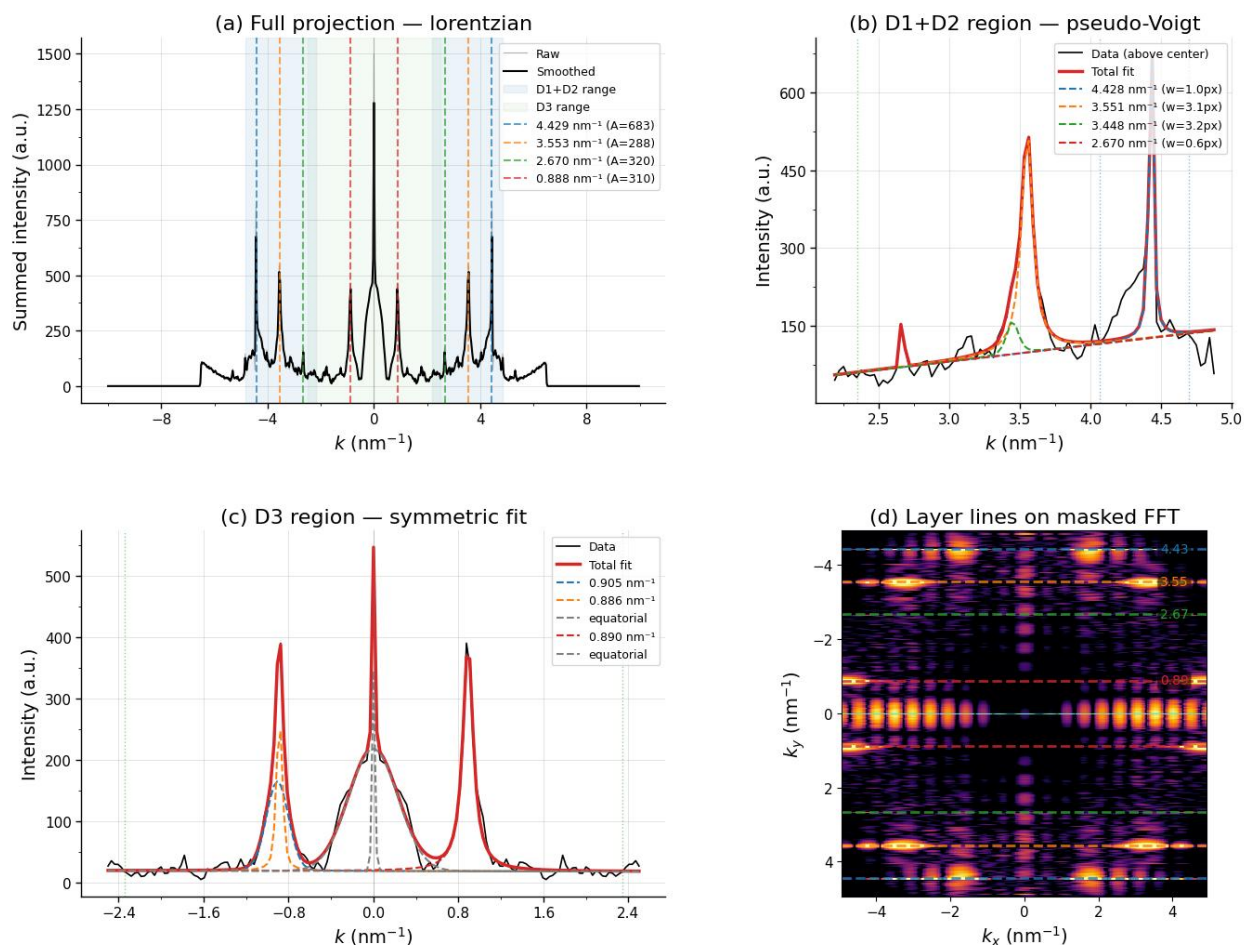


Figure S6. Layer line detection and measurement of the FFT from (20, 10) CNT at an overfocus condition. (a) Projection profile of the processed FFT with multiple peaks detected. (b) Fitting and peak detection in D1-D2 region by using a pseudo-Voigt method. (c) Fitting in D3 region by using the symmetric algorithm. (d) FFT in the central area with detected peak positions marked, corresponding to the peaks in (a).

Chirality Analysis — 5853/6930 correct (84.5%)

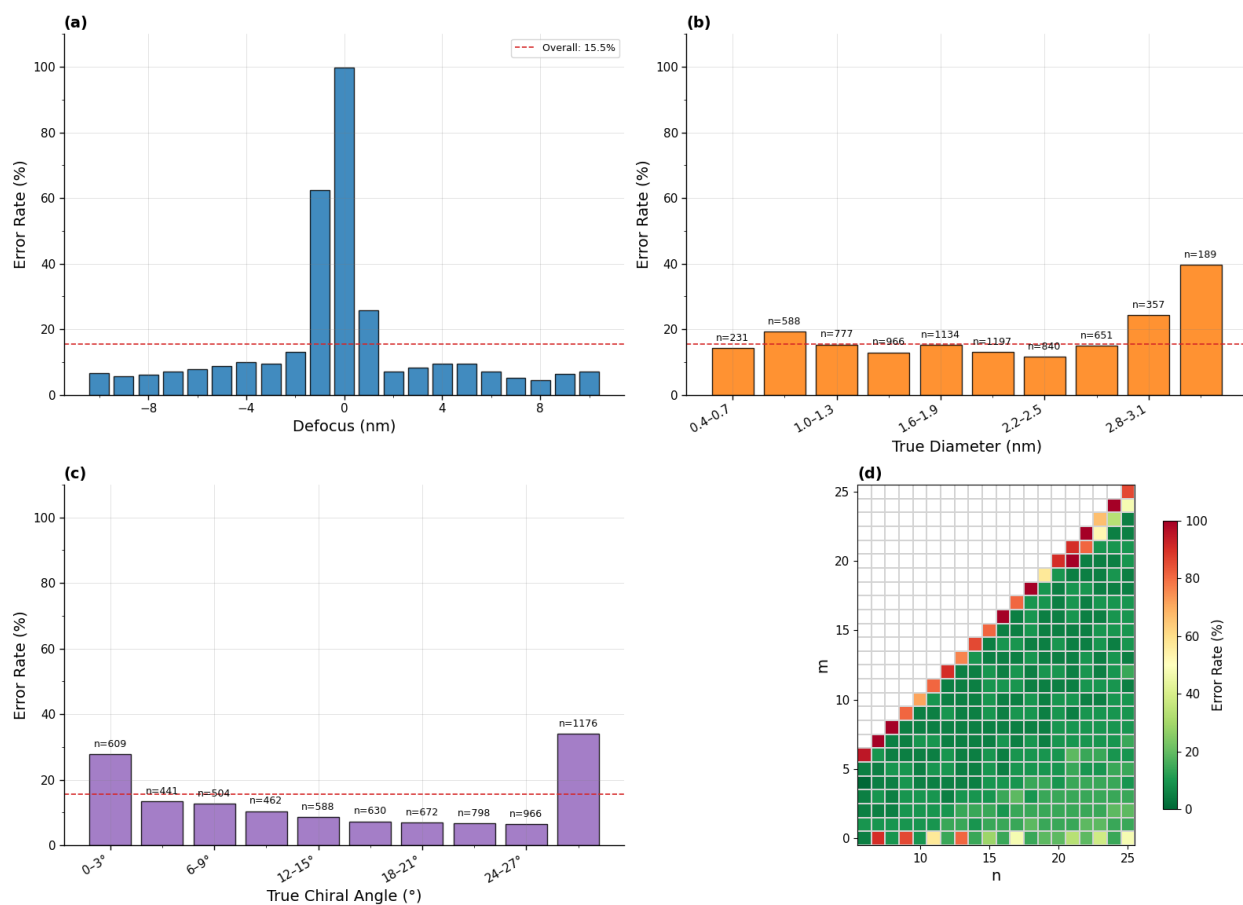


Figure S7. Error analysis for chirality extraction from simulated dataset. (a) Error rate as a function of defocus. (b) Error rate as a function of tube diameter. (c) Error rate as a function of chiral angle. (d) Error rate as a function of chiral indices (n, m).

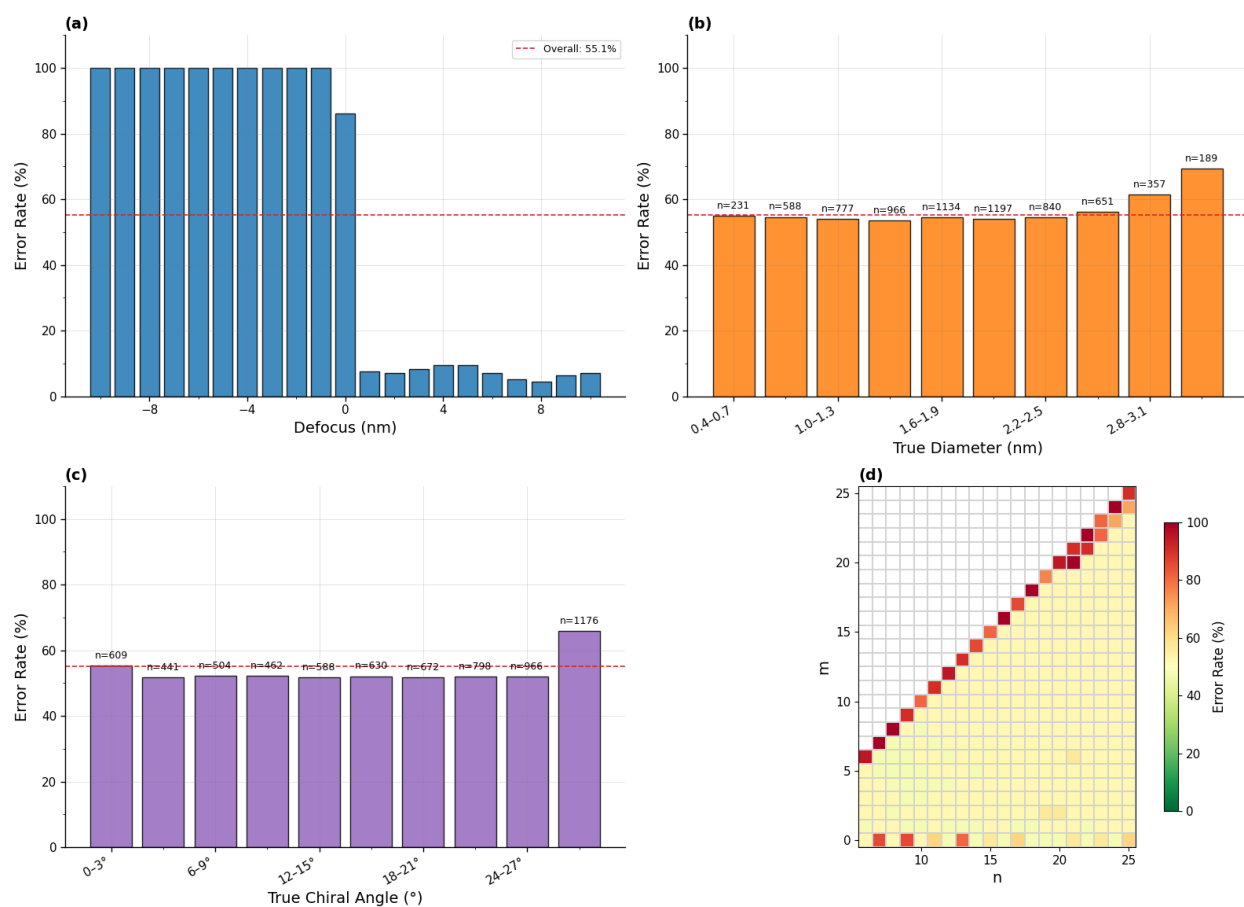


Figure S8. Error analysis for chirality extraction from simulated dataset with diameter measured by using d_{min} mode. (a) Error rate as a function of defocus. (b) Error rate as a function of tube diameter. (c) Error rate as a function of chiral angle. (d) Error rate as a function of chiral indices (n, m).

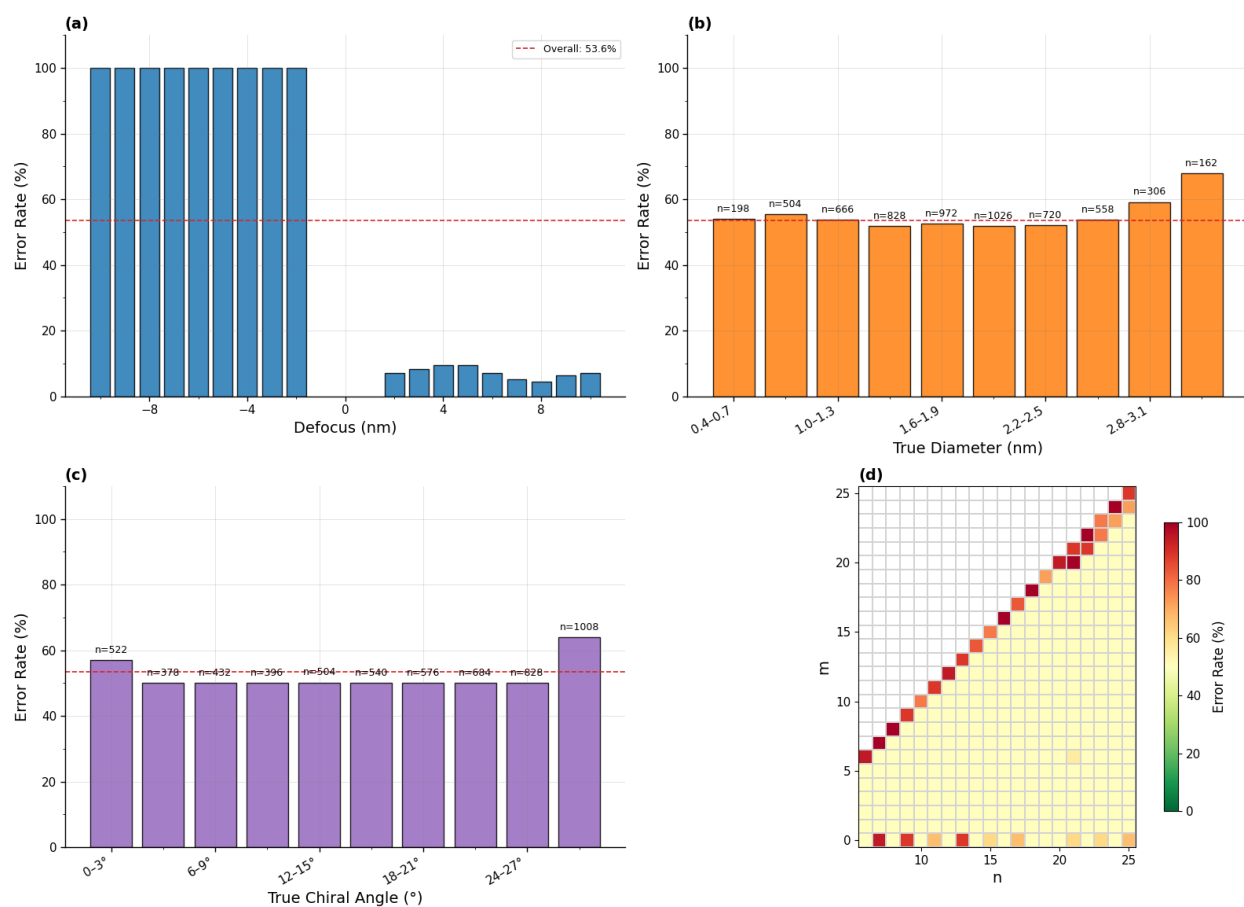


Figure S9. Error analysis for chirality extraction from simulated dataset with diameter measured by using d_{min} mode and excluding $|df| < 2$ nm. (a) Error rate as a function of defocus. (b) Error rate as a function of tube diameter. (c) Error rate as a function of chiral angle. (d) Error rate as a function of chiral indices (n, m).

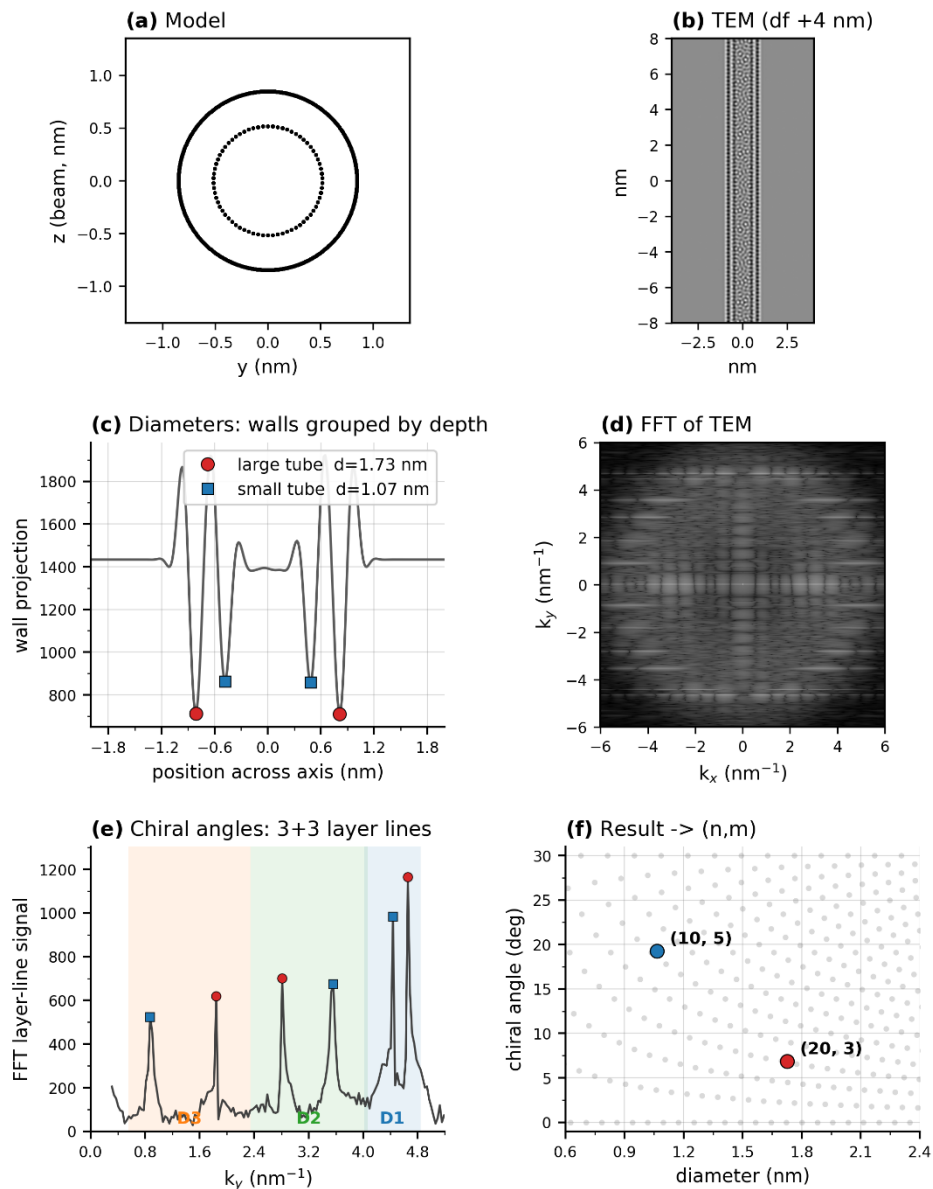


Figure S10. Chirality analysis of a double-walled CNT (10,5)@(20,3) from a simulated TEM image (60 kV, 0.0127 nm/px). (a) Atomic-model cross-section (beam along z). (b) Simulated TEM image (df = +4 nm). (c) Four dark-wall minima grouped by depth: the two deepest (red) span the larger shell (1.73 nm), the two shallowest (blue) the smaller shell (1.07 nm). (d) FFT of the image. (e) Six FFT layer lines (two per D-set); the two triplets, paired by $D3 = D1 - D2$, give chiral angles of 6.9° and 19.3°. (f) Pairing the larger diameter with the stronger layer-line triplet assigns (20,3) and (10,5), matching the isolated single-wall references.

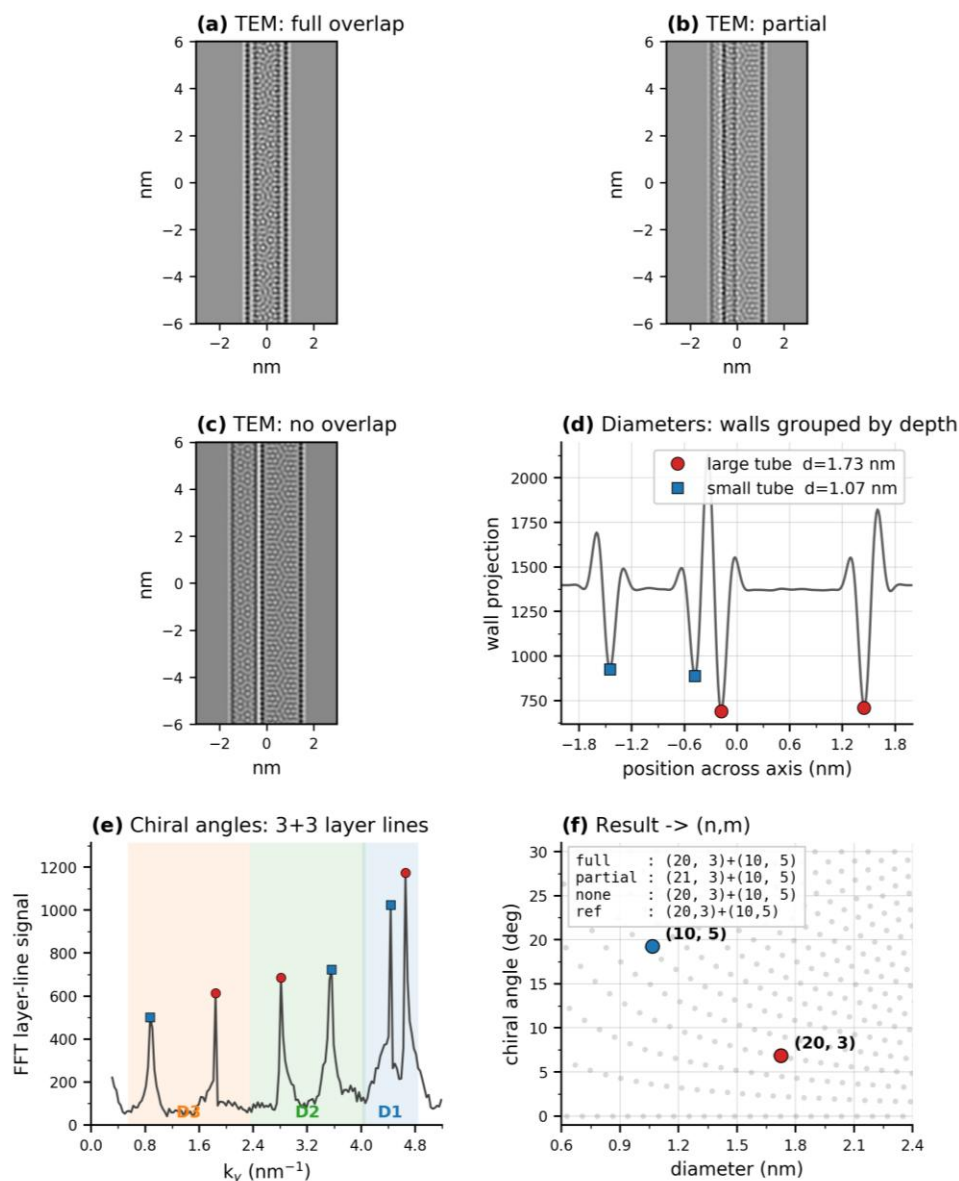


Figure S11. Chirality analysis of a (10,5)+(20,3) bundle across three projected-overlap regimes. (a–c) Simulated TEM images for full overlap ($\Delta y = 0$), partial overlap ($\Delta y = 0.85$ nm) and no overlap ($\Delta y = 1.6$ nm). (d) Depth-grouped wall profile (no-overlap case) giving the two diameters. (e) FFT layer-line projection giving the two chiral angles. (f) (n,m) assignment per regime: full and no-overlap recover (20,3)+(10,5) exactly; partial overlap gives (21,3)+(10,5), the outer index shifted by one because the walls interfere in the overlap region. The chiral angles are correct in all regimes.

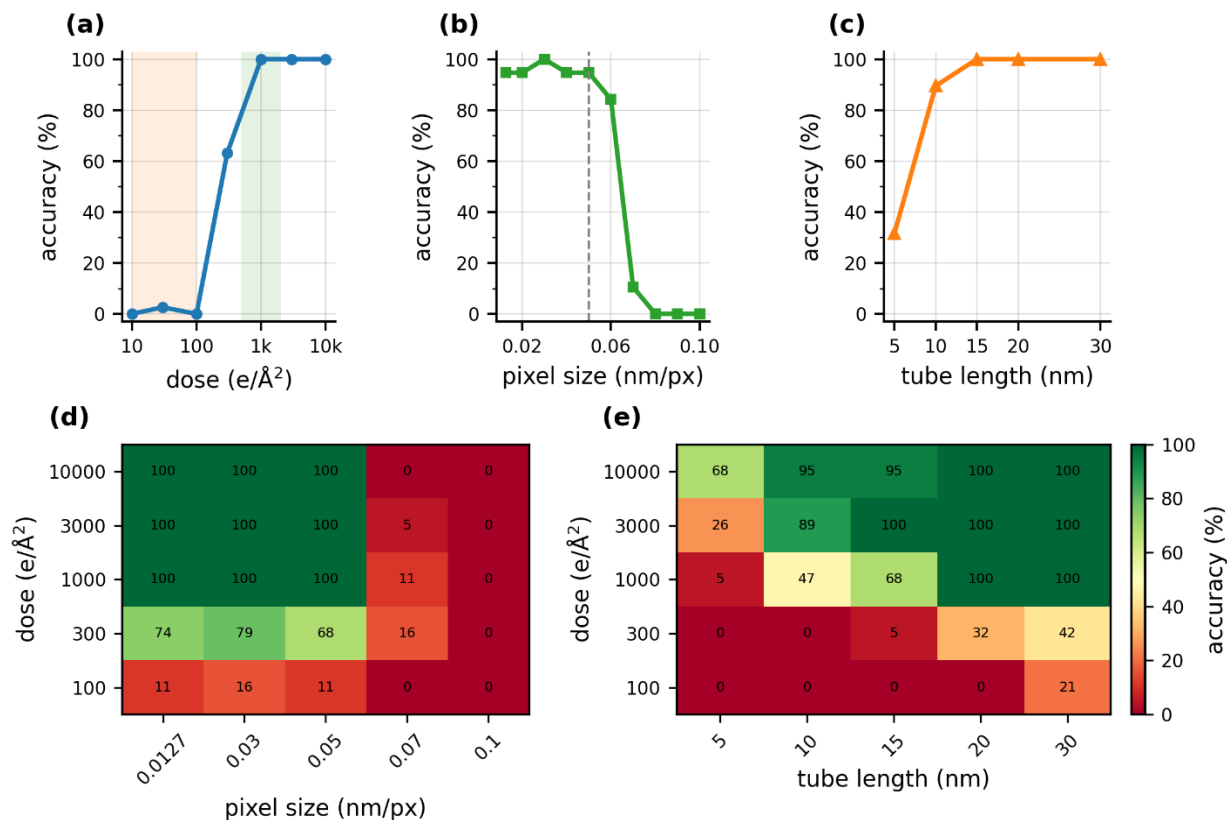
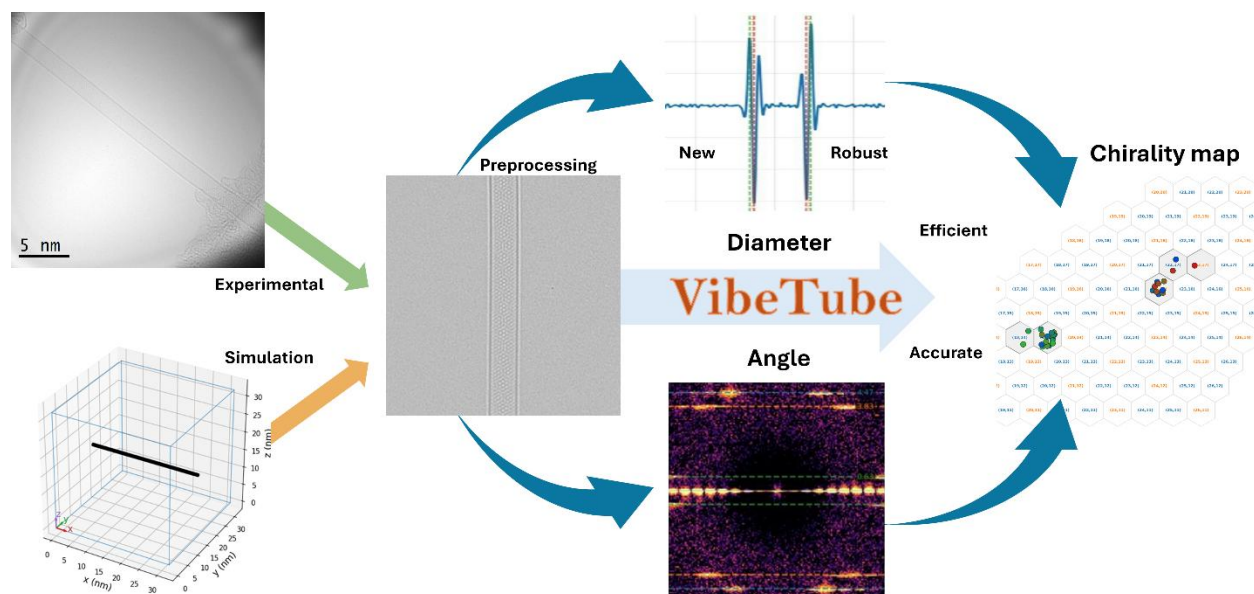


Figure S12. Robustness of the automated (n, m) assignment to acquisition conditions, for the (20,1)–(20,19) family at $df = +4$ nm, 60 kV, $C_s = 1$ μ m. (a) Accuracy vs electron dose at 0.05 nm/px. Shaded bands mark standard-HRTEM (500–2000 $e/\text{\AA}^2$) and low-dose (10–100 $e/\text{\AA}^2$) regimes. (b) Accuracy vs pixel size. The diameter is over-estimated as the walls are under-sampled, resulting error in chirality identification. (c) Accuracy vs simulated tube length at 0.05 nm/px. (d, e) Accuracy maps for dose $\times$ pixel size and dose $\times$ tube length. Pixel size limits the diameter measurement precision, tube length limits the chiral angle precision, and electron dose affects both.



Graphical Abstract

Developed with an AI co-scientist (Claude Code), "VibeTube" is an open-source, interpretable and physics-based Python code for automated, accurate, and robust extraction of carbon nanotube chiralities from TEM images.