Supplementary Figure S1: Schematic depicting the nanotomy process. (a) An image (taken via Nikon D60 camera) of the nanotome with mounted HOPG before commencement of the nanotomy cycle. As can be seen from the figure (and from Figure 1b), by using $\theta_v = 0$, we ensure that the whole cross section of the cutting face of HOPG is used. Bottom inset shows a schematic diagram of the diamond knife and the various knife parameters. (b) A higher magnification image of the sample holder chuck showing the HOPG rigidly attached (via ‘superglue’) to the araldite resin block. The cutting edge of the HOPG shows a metallic hue after the priming as discussed in the methods section of the manuscript. (c) Optical images immediately before and after a single cutting cycle shows production of a GNB (also see Supporting Video SV1). The cleaved GNBs float in water and are later scooped up into a glass vial. (d) Optical image of a glass vial containing scooped up GNBs for $w \sim 15$ and 25 nm. Right inset shows a cartoon depicting the structure of the GNBs, which comprise of the stacked GNRs. (e) Vials containing the stably-dispersed GNRs after the exfoliation of GNBs into GNRs using chlorosulfonic acid.
Supplementary Figure S2: Details of the nanotome (Model # PT-XL, Boeckeler Instruments Inc.; 3 mm edge width Du Pont Diamond Knife) used in the paper. A second knife of similar parameters from RMC products was also used in this report.
Supplementary Figure S3: Schematic diagram showing the atomic dimensions of a GNB block and a GNR. The C-C bond length is 0.142 nm and the interlayer spacing is 0.335 nm.
Supplementary Figure S4: Assorted FESEM and TEM images of GNRs with varying widthds demonstrating the versatility of the GNR-production process. (a, c, d) TEM images of $w \sim 15$, 30 and 40 nm GNRs immobilized on 400 mesh Lacey carbon TEM grids. Inset of (a) is an FESEM images of $w \sim 5$ nm GNRs immobilized on silicon substrates. It was interesting that the 15 nm ribbons exhibited spiral winding, which will be reported in more detail later.
Supplementary Figure S5: AFM images of w ~ 50 nm GNRs showing a typical thickness of 0.6 nm.
Supplementary Figure S6: HRTEM image of the GNRs. (a) HRTEM image of a \( w \sim 15 \) nm wide GNR showed width uniformity. A selected-area-electron-diffraction (SAED) pattern on the GNR shows a hexagonal pattern similar to that observed in graphene. (b) Aberration corrected HRTEM image of the edge of a trilayer \( w \sim 15 \) nm wide GNR showing mixed edges (superimposed cartoon showing the graphene structure is a guide for the eye). The image shows the expected hexagonal pattern attesting to the high crystallinity of the structure.
Supplementary Figure S7: High resolution XPS spectra for C-1s and S-2p levels. The scans show the sp²-carbon peak at 284.5 eV. The low levels of sulfur were attributed to the probable edge sulfonation of GNRs by chlorosulfonic acid.
Supplementary Figure S8: A typical conformal Raman mapping of a w ~ 15 nm wide GNR immobilized on a silicon dioxide surface.

Supplementary Figure S9: Inset of figure S8 shows a successful 4 peak Lorentzian fit of the 2D band of the GNR attributing bilayer nature. The I_D/G was ~ 0.4-0.5
Figure S10: Schematic diagram showing the vacuum filtration apparatus and the steps followed for fabricating GNR films. (a) A hybrid of schematic diagram and optical images showing the Büchner filtration apparatus and the polycarbonate filter on the Büchner funnel. After filtration the membrane is dried in an Argon atmosphere at room temperature for 2 days. (b) Optical image showing one such filter paper after the filtration process. Several GNR films (each similar in dimensions to the Büchner funnel pore size ~ 0.4 mm) were observed on the filter, which was then cut out for further characterization. (c) TEM image of de-laminated pristine-GNR film (by dissolution of the polycarbonate filter in 99 % chloroform) immobilized on 400 mesh Lacey carbon grids showing the random arrangement of GNRs in the film. (d) Schematic diagram of the cut out portion of a single GNR film on the polycarbonate filter substrate.
Figure S11: Typical back-gated transfer characteristics of a GNR film (the above example is for a $w \sim 15$ nm GNR film) device with $L \sim 0.2$ mm, $W \sim 0.4$ mm in air and in vacuum.
Figure S12: Typical temperature dependent transfer characteristics of the GNR-films (a shows a $w \sim 25$ nm GNR film; b shows a $w \sim 15$ nm GNR film) showing two different regimes of transport, one at high temperatures $T > T^*$ and other at low temperatures $T < T^*$. This is characteristic of VRH transport model.
Figure S13: Molecular dynamics simulations of the cutting process. a shows the simulation setup of a graphene sheet with an initial crack and a diamond rod as the knife. b and c illustrate carbon-carbon bonds in the graphitic lattice that bear maximum loads under tensile loads from a static mechanics analysis. The distribution of these bonds defines the breaking pattern as shown in Fig. 4.
Figure S14: Snapshots of the nanotomy process for cutting direction along (a) armchair, (b) zigzag and (c) chiral or mixing armchair-zigzag orientations of the graphene lattice. The color shows tensile stress $\sigma_{xx}$ in the transverse direction, which shows high stress (in red) localized in the crack front.
SUPPLEMENTARY METHODS:

**Exfoliation of GNBs:** The GNB exfoliation process was carried out via the recently reported superacid exfoliation strategy unless mentioned otherwise. The other two strategies used in this report were the Hummer’s acid exfoliation strategy and the Pyrene Carboxylic Acid (PCA) based π-intercalation exfoliation. The experimental details are as follows:

**Superacid exfoliation strategy:** The vials containing dried up GNBs were transferred into a glove box (< 1 ppm Water, < 1 ppm Oxygen) and 2 ml of Chlorosulfonic acid was pipetted into the vials. Subsequently the vials were placed on an incubator shaker at 27º C for ~ 8 hours inside the glove box for exfoliation. The liquid phase was observed to turn black, with minuscule solid residue left un-dispersed, signifying a high degree of exfoliation (Figure 1d and Supplementary Figure S1e). The GNR dispersion was quenched in DI water in a ratio of 1:200, as has been reported by Behabtu et al, for subsequent substrate immobilization or characterization.

**Hummer’s acid oxidation based exfoliation followed by hydrazine reduction:** Hummer’s acid oxidation as described in a previous report by our group (Mohanty et al, Nano Letters, 2008) was used for exfoliating the GNRs. The as-produced GONRs were reduced by the solution-phase hydrazine reduction, washed in DI water and substrate immobilized for further characterization.

**Pyrene Carboxylic Acid (PCA) based π-intercalation exfoliation:** We also used a modified version of the PCA based exfoliation strategy as reported by An et al. In brief, dehydrated GNBs were dispersed in 1 mg/ml solution of PCA in DMF, left undisturbed for ~ 8 hours and subsequently sonicated for 60 seconds resulting in the formation of light-grey colored suspension. The exfoliated GNRs were then washed several times in methanol, followed by DI water and immobilized onto substrates for further studies.

**Description of the Graphite Nanotomy Process to Produce GQDs:** An HOPG block is incorporated into wax mold to provide structural stability, and is rigidly attached to an araldite-resin-block using a cyanoacrylate adhesive, which is firmly mounted on the sample holder of an ultramicrotome (Fig 4). Using a highly-aligned, ultra-sharp diamond-
knife with ~ 1 - 2 nm edge radius\textsuperscript{26} (RMC Inc.), the HOPG-wax mold is repeatedly cut ~ 0.5 mm deep, while moving forward in small steps. The step-size can be set between 10 nm and 100 nm (Fig 3a) with 1 nm resolution. The mold is then rotated 90° and cut again in a similar manner (Fig 3b) to produce graphite nanoblocks (GNBs). This wax-mold is then evaporated by heating at 200 °C for 2 hours to obtain the pure GNBs. The GNBs are then exfoliated to obtain pristine GQD suspension (explained in section 3.5). \textbf{PARAMETERS:} The sample is precisely aligned along the edge of the diamond knife at a 5° clearance angle, and 40° rake angle (angle between the plane of the leading edge of the knife and the perpendicular to the HOPG face). The cutting speed (0.1 to 1 mm/s) and angle (2° resolution) are controllable. The ultra-small step-size of the ultramicrotome is achieved by the stepper motor mounted lead-screw mechanism.  

\textbf{PRODUCTION ANALYSIS FOR SCALE-UP:} The automated ultramicrotome produces one HOPG cut per second. The HOPG (0.4 mm X 0.4 mm X 0.5 mm) is nanotomed over an area of 0.1 X 0.1 mm\textsuperscript{2} and to a thickness of 0.5 mm. Producing 20 nm square GQDs from this HOPG will takes 2.7 hours (\textit{slowest step}) to produce 4 X 10\textsuperscript{13} GQDs from 2.5 X 10\textsuperscript{7} GNBs (each GNB has 1.6 X 10\textsuperscript{6} GQDs). Thus, 1.48 X 10\textsuperscript{13} GQDs \textit{can be produced per hour}. Commercial nanoparticle-solutions (gold, silver, polystyrene etc.) usually come in 50 ml bottle (for ~ $100) with a concentration of 10\textsuperscript{10} nanoparticles/ml.  

Description of the Process to find the efficiency of the process: The overall yield of the process was a combination of the yield of the two independent sequential processes underlying GNR production viz. (a) the cleavage of the GNBs from HOPG block (SPI Inc.) and (b) the exfoliation of the as-cleaved GNBs into constituent GNRs.  

The yield of the GNB cleaving process depends on the efficiency of cleavage and the efficiency of collection of the cleaved GNBs. After proper alignment of the nanotome, the cleavage was found to be successful in all cutting cycles (for each batch 50 cutting cycles was used). Since we collected all the GNBs from the knife boat, the yield of the GNB cleaving process was ~ 100%.  

Thus, the overall yield of the process was efficiency limited by the second step i.e. the exfoliation process.  

\textbf{Superacid exfoliation process:} In order to calculate the efficiency of this exfoliation process, the dried GNBs were weighed before being exfoliated. After, the chlorosulfonic acid
based exfoliation into constituent GNRs, the dark black colored liquid phase was quenched in DI water (1:200) and freeze dried in a Virtis Inc freeze drier (Model # 10-MR-TR). The resulting solid was again weighed for calculating the efficiency of exfoliation. The efficiency for this process was found to be ~ 80 %.

**Hummer’s acid exfoliation process:** Using a similar strategy as described above we calculate the efficiency of the hummer’s exfoliation process to be < 10 %.

**PCA based exfoliation strategy:** Using a similar strategy described above, the efficiency was found to be < 1 %.

Thus, the maximum overall efficiency of the production of GNRs was ~ 80 % (for superacid exfoliation strategy).

**Description of the XPS results:** We used XPS to study the chemical properties of the superacid exfoliated GNRs (Supplementary Figure S7). High resolution scans for the C 1s were obtained which showed the signature peak of sp² bonded Carbon at 284.5 eV. The absence of the sp³ carbon associated peaks vindicates the pristine nature of the GNRs. The XPS survey scans showed the presence of trace amounts of sulfur, which were confirmed using high resolution scans for S 2p. This is consistent with the observed up-shift of the Raman G-band peak (by ~ 2 cm⁻¹) for the w ~ 15 nm GNRs, compared to the wider GNRs: a signature of charge transfer from edge functional groups (This effect is more prominent for narrow GNRs, since they have a higher percentage of edge-associated carbon atoms than basal)⁵⁷. This presence of a negligible amount of sulfur is similar to the data reported on graphene by Behabtu et al²⁰. In view of the low penetration depth of the X-rays used in XPS (< 10 nm), we attribute the trace amounts of sulfur to the probable edge functionalization of the GNRs. Since, the edges of the GNRs, which include dangling bonds and defects, are known to be hydrogenated³⁴, ⁵⁸, and chlorosulfonic acid is known to react with phenyllic groups⁵⁹, we expect some degree of edge sulfonation⁴¹. Theoretical calculations limit the maximum incidence of sulfur at 1 atomic percentage (Based on the bottom panel of the Supplementary Figure S3, we estimated the total number of edge atoms with respect to the number of basal surface atoms for a 1 micron long, 15 nm wide GNR to be 0.0073 %. Thus, in the extreme case of full sulfonation of the edges⁴¹, the total theoretical amount of sulfur for unit mass of GNRs is expected to be < 1 %.). The atomic percentages of sulfur determined by the XPS scans were within the theoretical limits (C > 98 %, S ~ 0.92 %).
Description of the Raman Process Used: Raman spectroscopy is an excellent technique for probing the quality of the graphenic structures in terms of structural (defects), chemical (doping levels) and mechanical (strain) characteristics\(^\text{32}\). We employed Raman spectroscopy to probe the edge quality of our GNRs. We used a Renishaw inVia confocal micro-Raman spectroscope with an 100 X objective equipped with a 514.5 nm laser line to characterize our GNRs. Based on the report by Cai et al\(^\text{60}\), the diameter of the Raman laser beam is given by, \(d \approx \frac{2\lambda}{\pi \text{NA}}\) where \(\lambda\) is the wavelength of the laser and NA is the numerical aperture of the objective lens employed. The diameter of the laser beam was calculated to be \(\sim 0.4 \mu\text{m}\). Owing to the random edge crystallographies of the GNRs described in the current report (Supplementary Figure S6) and the diameter of the laser beam encompassing several individual GNRs in the GNR film, the Raman spectra (\(I_{\text{D}}/G\) ratios) is expected to probe the average roughness of the edges including the defects and dangling bonds\(^\text{34}\). Our GNRs were found to have low edge roughness as compared to most GNR fabrication strategies (Figure 3g). Further, we studied an individual \(w \sim 15\) nm GNR, first located by scanning Raman spectroscopy for the G band and then full Raman spectra being collected at the location of maximum G-band intensity (Supplementary Figure S8). The number of layer information was extracted via lorentzian fitting of the 2D peak\(^\text{32, 61}\). The \(I_{\text{D}}/G\) ratios for bilayer \(w \sim 15\) nm GNRs were found to be \(\sim 0.4 – 0.5\), comparable to that reported for similar dimension GNRs synthesized via the sonochemical unzipping strategy reported by the Dai group\(^\text{34}\). This reflects on the excellent quality of the as fabricated GNRs.

Description of the Process to study the Electrical Properties:

The sheet resistance of the thin GNR film (on polycarbonate filter substrate) was measured using the standard 4 point probe technique in which 4 tungsten probes were mechanically pressed onto the samples for contact resistance-free measurements. Sheet resistance was determined from the Van der Pauw’s formula \(R_s = \frac{\pi}{\ln 2} \frac{V}{I}\). The sheet resistance of the films was found to be \(2.2 – 5.1 \text{ M}\Omega/\square\). These results are comparable to the typical values for the reduced GO films as reported by Eda et al\(^\text{45}\) (43 k\(\Omega/\square\) - 0.06 T \(\Omega/\square\)). For surfactant stabilized graphenic films, Lotya et al\(^\text{31}\) and Hernandez et al\(^\text{24}\) have reported 0.022 M\(\Omega/\square\) and 7.1 k\(\Omega/\square\) respectively post-annealing (pre-annealing figures for Hernandez et al are 7.2 M\(\Omega/\square\); the
results reported by us are for **un-annealed samples**). In view of several randomly distributed GNR-to-GNR overlaps in the film, the contact resistance of those junctions is expected to form a major component of the sheet resistance.

Several attempts in producing large freestanding GNR films by dissolving the polycarbonate filter paper substrate in chloroform failed owing to their breakage upon detachment from the substrate. However, it might be possible to achieve this by using different filters such as alumina, which is a part of an ongoing investigation in our group. Nevertheless, delaminated GNR-films were immobilized onto a 400 mesh Lacey carbon TEM grid for TEM analysis. The TEM images showed the expected random distributions of the GNRs in the film (Supplementary Figure S10c).

In order to study the electronic properties of our GNR films, source and drain electrodes (Au:Pd = 3:2) were sputtered onto the GNR film on polycarbonate substrate (600 nm Pore size, Polycarbonate membrane, *SPI Inc.*) using a 0.2 mm wide strip of polyethylene as channel mask (Figure 4g shows an optical image of the GNR film on polycarbonate filter substrate with source and drain electrodes deposited. Figure 4e and f show FESEM and AFM images of the GNR film on the polycarbonate filter substrate.). Post-sputtering, the device is immobilized onto a gold-coated-silicon dioxide wafer via the application of pressure using a PDMS stamp. The device was annealed at 60 °C in an Argon atmosphere for 2 hours. Using the gold-coated-silicon wafer as the gate electrode and the polycarbonate filter substrate as the gate oxide, back-gated electrical studies were conducted (Figure 4d shows a schematic diagram of our back-gated FET device.). As a proof of concept, GNR films comprising of 3 different widths of GNRs were studies (*w* ~ 50 nm, 25 nm and 15 nm as determined by FESEM / TEM imaging).

Carrier mobilities were calculated using the commonly used equation,

\[
\mu = \frac{L}{W \cdot C_{OX} \cdot V_{SD}} (\frac{\Delta I_{SD}}{\Delta V_G}),
\]

where \(L\) is the channel length; \(W\) is the channel width; \(C_{OX}\) is given by \(\varepsilon_{OX} \varepsilon_0 / t_{OX}\) where \(\varepsilon_{OX}\) is the permittivity of the gate oxide, \(\varepsilon_0\) is the permittivity of free space and \(t_{OX}\) is the gate oxide thickness; \(V_{SD}\) is the source-drain voltage and \(\left(\frac{\Delta I_{SD}}{\Delta V_G}\right)\) is the transconductance or the slope of the transfer curve in the linear regime.

Transfer characteristics show clear semiconducting characteristics (Supplementary Figure S11). When the measurements were conducted in air, a strong n-type doping character was
observed for the GNR films which were attributed to the adsorbates during the fabrication process. Upon exposure to vacuum and electrical annealing as described by Moser et al\textsuperscript{51}, ambipolar character was restored resembling that of pristine graphene. This vindicates the good quality of the GNR films. Typical mobilities of the electrons and the holes were calculated to be 20 - 130 cm\textsuperscript{2}/V·S for all the devices studied in this work (8 devices were studied in all, 2 for w ~ 50 nm and 3 each for w ~ 25 and 15 nm GNRs; 1 device for w ~ 50 nm GNR shorted during gating). These values are significantly higher than that reported for the wide-functionalized-reduced-GONR films reported by Zhu et al\textsuperscript{62} (~ 0.1 – 0.5 cm\textsuperscript{2}/V·S) attributed to the pristine nature of our GNR films. However, the mobility values reported for single GNR devices are 2 orders of magnitude higher (~ 1500 cm\textsuperscript{2}/V·S in the report by Jiao et al\textsuperscript{34}). This is expected in view of the scattering at the overlap junctions of the GNRs constituting the film.

Further, the low temperature I\textsubscript{DS} versus V\textsubscript{DS} characteristics (in a Janis ST100 cryostat, 10\textsuperscript{5} Torr base pressure, 80 K) of the GNR films at the charge neutrality point (V\textsubscript{G} = 0) showed non-linearity, a characteristic of semiconductors. In order to determine the bandgap of the respective films, we analyzed their non-linear transport characteristics as described by Han et al\textsuperscript{47}. Here, the first step in the conduction process is the transition of the carriers from their non-conducting to the conducting band (at low temperature). Owing to the relatively large channel lengths in our GNR film devices, the channel length dependence of the non-linear bandgap is expected to have an insignificant effect in our devices. The edges of non linear bandgap were determined from the steep increase of the absolute value of I\textsubscript{DS} in the logarithmic scale (Figure 5a-c). A clear bandgap scaling was observed: as expected the w ~ 50 nm GNR film had a very small bandgap which could not be measured due to instrument limitations (2 devices were studied; 1 device shorted during gating), the w ~ 25 nm GNR film had a bandgap of ~ 10 meV (3 devices were studied) and the w ~ 15 nm GNR film had a bandgap of ~ 35 meV (3 devices were studied).

**Description of the Process to find the temperature dependent characteristics of the GNR-films:** In order to independently study the bandgap of the GNR films and confirm the nonlinear bandgap values determined as above, we studied the temperature dependence of the OFF current of the GNR film devices (w ~ 15 and w ~ 25 nm GNR film devices; w ~ 50 nm was not studied owing to the extremely small bandgap un-measurable by us due to instrumental limitations) on the lines of Chen et al\textsuperscript{48}. A sequential decrease in the OFF current was observed as shown in the
Figure 5h. We studied the thermally activated off-resonant conduction of the GNR films via Arrhenius plots of the $I_{\text{OFF}}$ versus temperature (Supplementary Figure S12). Two separate transport regimes were observed, one at high temperature and other at low temperatures with a crossover at a characteristic temperature $T^*$. At high temperatures ($T > T^*$), the transport is simply activated and the $I_{\text{OFF}}$ follows the simple exponential dependence on temperature given by, $I_{\text{OFF}} \propto e^{-\frac{E_g}{k_B T}}$, where $E_g$ is the bandgap, $k_B$ is the Boltzmann constant and $T$ is the temperature. Plots of $\ln(I_{\text{OFF}})$ versus $1/T$ were used to fit a straight line at high temperatures, to obtain average bandgaps of ~ 6 meV and ~ 30 meV for $w \sim 25$ nm and $w \sim 15$ nm GNR film devices respectively (2 devices were studied for each width). These compare well with the non linear bandgap. For the low temperature regime ($T < T^*$), stark deviation from simple activation behavior was observed for the $I_{\text{OFF}}$, which was now observed to decrease more slowly. This is a signature of variable range hopping (VRH) transport model. This is expected owing to the several randomly distributed GNR overlap junctions in the films which act as scattering sites. A VRH model given by, $I_{\text{OFF}} \propto e^{-\frac{T_0}{T}}$, where $T_0$ is a characteristic constant and $\gamma$ is the exponent, was fit to the data. For both $\gamma = 1/3$ and $\gamma = 1/4$, good fits were obtained with similar standard deviations. This is suggestive of transport in 2 and 3 dimensions including through multiple layers$^{49}$. More studies are currently undergoing to characterize these transport mechanisms in detail.

**Molecular dynamics (MD) simulations of the nanotomy process**

**Description of the Molecular Dynamics Simulations:** We perform MD simulations for the nanotomy process of single and multi-layered graphene sheets. The adaptive intermolecular reactive empirical bond order (AIREBO) potential is utilized to describe both intralayer bonding and interlayer van der Waals interactions$^{63}$. Although it does not give accurate prediction for graphene edge energies in comparison with from first-principles methods$^{64}$, its relatively less computational time request enables us to set up simulations with reasonable stress field during
the cutting process. A diamond nanorod with a diameter of 1 nm is set up the simulation as the knife in experiments (Supplementary Figure S13a). An initial crack with a length of 1 nm is created. A transverse force is applied to the diamond rod to cut the graphene sheet at a constant speed ($v = 0.1 \text{ m/s}$ in our simulation), starting from the crack front. Periodic boundary condition is used in perpendicular to the cutting direction. Carbon atoms in two atom chains along the cutting direction, far from the crack, are constrained by springs with a stiffness of 1602 N/m to prohibit rigid-body motion of graphene sheets, following the holding condition in the experiments. During the cutting process, the diamond knife is considered as a rigid body.

In our simulations, a time step of 0.5 fs is used. Atomic structures of are firstly relaxed using a conjugated gradient algorithm, starting from which microcanonical ensemble (NVE) MD simulations are performed. Canonical ensemble (NVT) simulations coupled with a Nosé-Hoover thermostat are also carried out from 0 K to 300 K. The results show no significant difference in the cutting processes as thermal energy $k_B T (= 0.026 \text{ eV at } T = 300 \text{ K})$ is much lower than the cohesive energy of graphene $E_c = 7.4 \text{ eV}$.

The current approach is limited in the time scale that atomistic simulations can reach, which is on the order of nanoseconds. Edge reconstruction processes occurring at larger timescale thus cannot be captured. Possible chemical reactions (hydrogenation, oxidation, etc.) are also neglected in our simulations. Moreover, although including bond orders and being benchmarked by several chemical reactions, the AIREBO potential does not give an accurate description of carbon nanostructures with dangling bonds in comparison with first-principles calculations. Although the approach here captures essential physics of the nanotomy process, new techniques, such as density functional theory (DFT) based kinetic Monte-Carlo and multi-timescale methods, should be developed to overcome aforementioned limitations.

**Simulation Results for the Nanotomy Process**

Simulations are performed for graphene sheets with lattice orientations aligning in different angles with the cutting direction. Supplementary Figure S14 shows snapshots for MD simulations where the diamond knife cuts along the armchair (Supplementary Figure S14a), zigzag (Supplementary Figure S14b) and chiral (Supplementary Figure S14c) directions. Stress component in the $x$-direction is also visualized, which suggests a localized tensile stress field.
near the crack front. Graphene edges finally cleaved for these three graphene sheets are summarized in Supplementary Figure S14d,e,f, which align globally to the cutting direction. However, the local edge is preferably along the zigzag direction in graphene sheets.

To explain these characteristics of cleaved graphene edges, MD simulations are also carried out to investigate the fracture behavior of a graphene sheet under tensile loads. A tensile strain is applied to graphene nanoribbons along both armchair, zigzag and chiral directions. In consistence with static mechanics analysis in Supplementary Figure S13b,c, simulation results show that carbon-carbon $sp^2$ bonds cleave in perpendicular to the tension load if it is applied along the armchair chains, and a zigzag crack is formed as the tensile load aligns with the zigzag chain. Additional simulations using the reactive forcefield (ReaxFF)\textsuperscript{65, 66} and density functional theory calculations show the same trend. Moreover, armchair, zigzag and chiral edges of graphene feature edge energies differing on the order of 0.1 eV\textsuperscript{64, 67}. Thus the graphene edges will relax and reconstruct during and after the cutting process, which could be even more complicate when chemical environment (hydrogenation, oxidation, etc.) are included\textsuperscript{67}. These results indicate that, unlike sublimation and chemical etching or cutting\textsuperscript{42-44, 68}, where edge-dependent reactive energy barrier defines the edge anisotropy, mechanically cutting and edge reconstruction in the nanotomy process controls the direction and shape of cleaved edges\textsuperscript{69}.

**Effects of Thermal Fluctuation and Thickness of Graphene Multilayers**

As shown in the previous paragraph, for monolayer graphene sheets, MD simulation results show remarkable rippling during the cutting, as graphene features extremely high tensile strength $\sigma_g = 120$ GPa of graphene sheet but very low bending stiffness $D = 0.91$ eV\textsuperscript{68}. It is noticeable that out-of-plane rippling is induced by thermal fluctuation, which could lead to folded edges in the final product. It modifies the cutting dynamics here but still yields straightly cleaved edges. However, the amplitude of rippling is reduced in multi-layered graphene sheet and graphite, which is verified in our simulations. Moreover, in graphene multilayers or thick HOPG blocks, because of the lattice orientation difference between graphene layers and grain boundaries within graphene sheets, nanotomy processes where graphene sheets aligns very differently with the cutting direction thus occur, yielding graphene nanoribbons of different types.
SUPPLEMENTARY REFERENCES


