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Mechanical reinforcement of graphene via wrinkling

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Mechanical cantilevers are central to nanotechnology, where low bending rigidity makes stability the fundamental challenge. Here, we introduce a wrinkle-induced stiffening approach that enhances the bending rigidity of monolayer graphene by several orders of magnitude, enabling the fabrication of mechanically robust graphene cantilevers. When suspended over microcavities, these wrinkled membranes exhibit significant increases in both in-plane and out-of-plane stiffness, as confirmed by nanoindentation and resonance measurements, which also reveal that enhanced bending rigidity strongly influences their vibrational response. This behavior marks a transition from tension-dominated mechanics to a regime where bending effects become prominent, even in a single atomic layer. By sculpting these structures, we realize graphene cantilevers with measured bending rigidities between 10^6 – 10^7 eV, while maintaining femtogram-scale mass. These findings establish a mechanical stabilization mechanism for realizing ultrathin cantilevers and highlight their potential for applications where high compliance is of importance.

Although graphene is widely recognized as a two-dimensional (2D) material, its lattice is never perfectly flat, practically. Instead, graphene structure exhibits a spectrum of out-of-plane deformations of distinct characteristics¹. Thermal fluctuations with subnanometric amplitudes are intrinsic deformations that spontaneously arise within the graphene lattice at finite temperatures^{2,3}. These dynamic fluctuations are fundamental to the energetic stabilization of graphene, serving as a natural mechanism that maintains its structural integrity. Moreover, they give rise to a number of exotic properties, including a negative coefficient of thermal expansion⁴ and size-dependent elastic moduli⁵.

Beyond inherent fluctuations, graphene may exhibit non-intrinsic deformations such as wrinkles, induced by external forces^{6,7}. Wrinkles are observable in a wide range of amplitudes, from a few to hundreds of nanometers. Smaller-amplitude wrinkles within this spectrum are responsive to external loading. Especially when stretched, such wrinkles would gradually be suppressed ("ironed-out"), a phenomenon that is accountable for the softening effect that is often observed in the stress-strain response of graphene membranes^{6,8–10}.

In compressive loadings, on the other hand, small-amplitude wrinkles tend to evolve to generate deformations of higher amplitudes^{11,12}. Large-amplitude wrinkles, however, may feature self-contact regions¹³ with strong van der Waals attractions between the contact surfaces. Such wrinkles are difficult to fully flatten, even after removing the external compression¹¹. Practically, these large-amplitude, stable deformations can emerge from a variety of mechanisms, including compression-induced

buckling on pre-stretched elastic substrates¹¹, thermomechanical stresses encountered during graphene growth¹⁴, or mechanical perturbations introduced during handling and transfer processes.

From the technical point of view, the inclusion of wrinkles in the graphene structure is expected to improve the mechanical stability of the material. Such irreversible, large-amplitude wrinkles can enhance the out-of-plane rigidity of planar sheets^{15,16}, enabling the formation of stable corrugated structures. In fact, free-standing monolayer graphene structures, hitherto, have been realized only in the form of the circular (perimetral clamped), rectangular (four-side clamped)^{17,18} or doubly-clamped⁴ membranes. Monolayer graphene cantilevers — strips fixed at one end and free at the other — potentially represent the ultimate limit of mechanical softness and minimal mass, making them highly attractive for nanomechanical sensing applications. However, their extreme mechanical fragility renders them inherently unstable and has thus far eluded experimental realization, except when supported in a liquid environment¹⁵.

In this work, we overcome this challenge and realize monolayer graphene cantilevers by harnessing wrinkle-induced mechanical reinforcement. We deliberately introduce a network of large-amplitude wrinkles into graphene to boost its bending rigidity and prevent collapse. Our fabrication approach involves a surfactant-mediated lateral contraction of centimeter-scale monolayer graphene at an air-liquid interface. This process generates an interconnected wrinkling pattern across the graphene sheet. The wrinkled graphene is then transferred onto a microstructured substrate containing an array of microcavities, yielding suspended graphene

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membranes that are clamped on all four sides. These four-edge-clamped membranes serve as a convenient platform to study the influence of wrinkles on mechanical properties. Using nanoindentation, we observe that the presence of wrinkles significantly enhances the membrane's 2D elastic modulus compared to flat graphene. Interestingly, we also observe an increase in the linear stiffness despite the anticipated softening from lateral contraction. This suggests that the induced out-of-plane deformations have enhanced the importance of bending stiffness compared to the membrane tension in wrinkled samples. To confirm this interpretation, we perform resonant measurements on wrinkled graphene drums. The observed vibrational response reveals a clear departure from tension-dominated dynamics, demonstrating that bending rigidity plays an important role in shaping the resonance characteristics of these membranes. Motivated by these observations, we convert the all-edge clamped membranes into one-end-anchored cantilevers while preserving the wrinkle-induced stiffening. Remarkably, despite being only one atom thick, these wrinkled graphene cantilevers exhibit effective bending rigidities on the order of 10^6 – 10^7 eV — many orders of magnitude higher than the nominal value of $\kappa \approx 1.2$ eV, known for pristine graphene⁵ — and even above the thermally renormalized bending stiffness of micron-scale graphene. Our cantilevers exhibit ultralow spring constant in the range of $\sim 10^{-2}$ N/m. At the same time, the polymer-free fabrication process allows the graphene to retain its exceptionally low mass (estimated as $\sim 10^{-13}$ – 10^{-14} g), characteristics that place them in an unprecedented regime of mechanical devices. Our findings demonstrate a viable route towards stabilizing atomically thin cantilevers by engineered corrugations. The wrinkle-assisted strategy, proposed here, yields ultra-thin, ultra-light, yet robust cantilevers, opening the door to graphene-based mechanical sensors and resonators with unprecedented sensitivity and performance.

Results

Fabrication of wrinkled graphene

Fabrication of the samples follows a protocol that we developed earlier^{19,20}. Briefly, a centimeter-scale graphene sample, chemically grown on a copper foil, was subjected to a lateral force mediated by an enclosing surfactant layer on a liquid surface, after etching away the copper foil. The stress difference at the air-liquid interface contracts the surface area of graphene to form a random network of wrinkles/folds, associated with a detectable reduction in the graphene area.

Compared to conventional wrinkling approaches based on substrate-induced strain or chemical functionalization¹, the present method relies on surfactant-mediated lateral contraction at an air-liquid interface, leading to spontaneous wrinkle formation without the need for external mechanical actuation or complex processing steps. This approach enables direct transfer of the wrinkled graphene onto arbitrary substrates, including micro-structured surfaces, which is advantageous for device integration. The typical wavelength of wrinkles in our system is in the range of a few micrometers. Our earlier experiments further suggest that the wrinkle density can be tuned by adjusting the surface pressure²⁰.

We transferred the wrinkled graphene onto a substrate with a dense array of square through-holes (side length $8\ \mu\text{m}$, corresponding to an effective circular radius $R_{\text{out}} = \frac{8}{\sqrt{\pi}}\ \mu\text{m}$) by placing the substrate in contact with the graphene. Figure 1a and b schematically illustrate the fabrication process, with the details provided in the methods and supporting information S1. The process achieves an array of freestanding membranes, which we will refer to as the “surfactant-mediated” membranes in this text. For the sake of comparison, we fabricated a separate set of membranes, referred to as “surfactant-avoided” membranes, with no surfactant involved in the fabrication process.

AFM Nanoindentation

To characterize the topography and probe the mechanical properties of graphene samples, we performed a series of scanning and nanoindentation experiments using an AFM (JPK Nanowizard 4) with an indenter radius of $R_{\text{in}} \approx 10$ nm, detailed in the methods and supporting information S2.

Figure 1c and d compare the topography of the arbitrarily selected “surfactant-mediated” and “surfactant-avoided” membranes. The presence of a network of interwoven wrinkles/folds is the salient feature of the “surfactant-mediated” membrane. The amplitude of such wrinkles can easily exceed several tens of nanometers. The “surfactant-avoided” membrane, on the other hand, still showcases a level of wrinkles, albeit very dilute and with lower amplitudes. Such wrinkles could have been generated, for example, due to the liquid sloshing during the rinsing and/or top-fishing of the graphene. Regardless of the amplitude and density of the wrinkles, the AFM mappings demonstrate the propensity of graphene for stable foldings.

Nanoindentation provides a quantitative measure of the elastic properties of graphene membranes. Briefly, the membranes are pushed in the center with an AFM tip, and the corresponding indentation depth is deduced from the AFM piezoelectric base response, albeit after post-processing and correction for the deflection of the AFM cantilever. Figure 2a illustrates the correlation between the applied force (F) and the corresponding indentation (displacement, δ) at the center of a selected membrane. We note that the work done by the applied force is stored in the bending and stretching energies of the graphene sheet and leads to the following scaling relationship^{21,22}:

$$F \cdot \delta \sim R_{\text{out}}^2 \sigma_{\text{pre}} \left(\frac{\delta}{R_{\text{out}}} \right)^2 + R_{\text{out}}^2 E \bar{h} \left(\frac{\delta}{R_{\text{out}}} \right)^4 + R_{\text{out}}^2 \kappa \left(\frac{\delta}{R_{\text{out}}} \right)^2. \quad (1)$$

where σ_{pre} , $E\bar{h}$, and κ are the pretension, 2D Young's modulus, and bending rigidity of the graphene sheet, respectively. Equation (1) involves two terms that are quadratic in the indentation depth δ : one coming from the pretension of the sheet, the other from the sheet's bending stiffness. To compare the relative importance of these two effects, we introduce a dimensionless bending stiffness, defined as $\mathcal{B} = \frac{\kappa}{\sigma_{\text{pre}} R_{\text{out}}^2}$.

Equation (1) further reveals two distinct regimes in the force-displacement response of the membrane. In the regime of small applied forces, the indentation scales linearly with force, i.e., $F \sim \delta$. In this limit, the membrane's resistance to the applied normal load is primarily governed by its in-plane pretension and/or its bending rigidity. A detailed analysis²¹ shows that which of these dominates depends on the parameter $\mathcal{B}$: when $\mathcal{B} \ll R_{\text{in}}^2/R_{\text{out}}^2$, the bending contribution is entirely negligible, and Equation (1) simplifies to a tension-dominated model. For pristine graphene membranes, with a typical bending rigidity of $\kappa \approx 1.2$ eV, this condition is generally satisfied, and the linear part of the force-displacement curve is commonly used to extract the in-plane pretension σ_{pre} ^{23–26}. However, it has also been suggested²⁷ that wrinkling or thermal crumpling of graphene may lead to an enhanced bending modulus. In this case, detailed analysis shows that the bending stiffness first plays a role when $\mathcal{B} \approx R_{\text{in}}^2/R_{\text{out}}^2$ — while the indentation stiffness is still proportional to the tension in this limit, the relevant prefactor has a logarithmic dependence on the bending stiffness parameter, $\mathcal{B}$ ²¹. When $\mathcal{B} \gg R_{\text{in}}^2/R_{\text{out}}^2$ the pretension is negligible, and it is the bending stiffness that dominates; this regime has not been reported in monolayer graphene experimentally.

To account for the combination of bending stiffness and pretension effects, we define an effective pretension parameter σ_{eff} , which captures the total linear resistance of the sheet to indentation that is measured experimentally and incorporates contributions from both the actual pretension σ_{pre} and the bending rigidity κ . In the second, so-called “cubic regime,” observed at high applied forces, the sample stiffens. Here, larger force increments are required to produce the same indentation increment, obeying: $F \sim \delta^3$. The applied force in the cubic regime stretches the chemical bonds between the carbon atoms in the basal plane; hence, the Young's modulus of the material is the dominant source of the nonlinear stiffness. These two regimes can be well-identified in Fig. 2a and b for a wrinkled membrane. In Fig. 2a, the black dashed curve corresponds to the best fit of a representative experimentally measured force-displacement curve with the model in Equation (1), incorporating dimensionless prefactors appropriately²². The values of the effective tension σ_{eff} and nonlinear

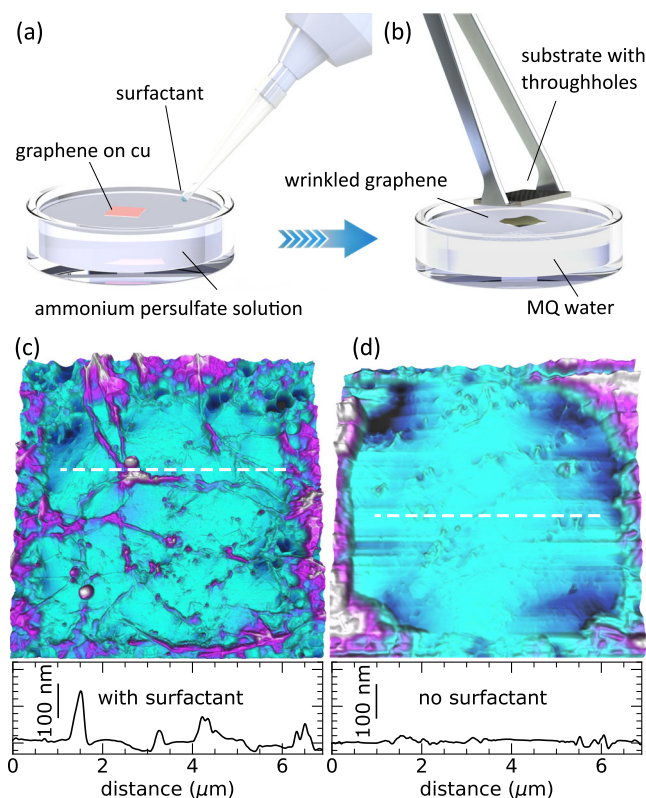


Fig. 1 | Fabrication and characterization of wrinkled graphene membranes.

a Graphene grown on copper is floated on a 0.5 M ammonium persulfate (APS) solution with a trace amount of surfactant (see Supplementary S1). **b** As the copper etches, surfactant-induced lateral forces generate wrinkles in the graphene. The APS is gradually replaced with pure water, and the wrinkled graphene is transferred onto a substrate with square through-holes and air-dried. **c** AFM image and profile of a wrinkled (“surfactant-mediated”) graphene membrane suspended over an $8 \times 8 \mu\text{m}^2$ hole. **d** Same measurement for a “surfactant-avoided” membrane prepared without surfactant. The color code in (c) and (d) indicates the local out-of-plane deformation of the membrane, with cyan corresponding to regions close to the flat reference plane and purple marking areas with the largest height deviations.

stretching stiffness, $E\bar{h}$, can be extracted by fitting the experimental nanoindentation curves. In total, 34 membranes of “surfactant-mediated” and 40 membranes of “surfactant-avoided” types have been measured.

Histograms of the estimated σ_{eff} and $E\bar{h}$ are plotted in Fig. 2c. Accordingly, we achieved an average σ_{eff} of $0.12 \pm 0.05 \text{ N/m}$ for the “surfactant-avoided” sample, which increased to $0.20 \pm 0.07 \text{ N/m}$ upon the inclusion of wrinkles: the membrane is apparently stiffened by a factor $\mathcal{S} \approx 1.7$ (or $0.8 \leq \mathcal{S} \leq 3.9$ accounting for uncertainties). The nonlinear stiffness is also prone to wrinkle-induced changes, increasing from $E\bar{h} = 140 \pm 55 \text{ N/m}$ in the “surfactant-avoided” sample to $E\bar{h} = 422 \pm 160 \text{ N/m}$ in the “surfactant-mediated” samples.

While changes in $E\bar{h}$ can be explained via nonlinear membrane stretching, a closer look at the linear regime reveals a more surprising effect: possible emergence of bending rigidity as a contributor to the nanoindentation response. More specifically, the observed increase in linear stiffness is counterintuitive: the inclusion of surfactant induces lateral contraction of the membrane²⁸, which might be expected to reduce the effective tension, and hence reduce the stiffness. Instead, we observe an enhancement, pointing to a contribution from wrinkle-induced out-of-plane reinforcement — an increase in the effective bending stiffness. A possible alternative hypothesis is that the adhesion between the folded regions leads to localized stretching of suspended regions between wrinkles, thereby enhancing the apparent stiffness. However, detailed theoretical analyses of such out-of-plane deformations show that these large-amplitude wrinkles primarily relieve in-

plane stresses, with the response governed by bending rather than by tension buildup¹³. We therefore consider it unlikely that localized stretching is a mechanism behind the observed stiffening.

To quantify the increase in indentation stiffness caused by a non-negligible bending rigidity, we first note the simpler result previously calculated for a point indenter²¹ for which the indentation stiffness is

$$\frac{k}{2\pi\sigma_{\text{pre}}} \approx \left(\gamma - \log 2 - \frac{1}{2} \log \mathcal{B} \right)^{-1} \quad (2)$$

when $\mathcal{B} \ll 1$. Here $\gamma \approx 0.577$ is the Euler-Mascheroni constant. Crucially, the indentation stiffness k depends linearly on the true pretension in the system, σ_{pre} , and is only logarithmically dependent on $\mathcal{B}$ — a dependence that is shown in Fig. 2d and that persists in more detailed calculations (see supporting information S5.1). Nevertheless, from the observed stiffening $\mathcal{S}$ of the surfactant-mediated membranes (compared to the surfactant-avoided ones) we can obtain a first estimate of the size of $\mathcal{B}$ needed to give such a large stiffening; we find that

$$\mathcal{B} \approx \frac{e^{2\gamma}}{4} \left(\frac{R_{\text{in}}}{R_{\text{out}}} \right)^{2/\mathcal{S}} \approx 5.9 \times 10^{-4}, \quad (3)$$

which represents an enhancement of the bending stiffness by a factor of around 10^4 compared to the pristine value, but with large uncertainty because of the wide range of values of $\mathcal{S}$. Note, in particular, that the exponential dependence on $\mathcal{S}$ in Eq. (3) means that even a moderate relative stiffening, $\mathcal{S}$, requires an enormous increase in the bending stiffness $\mathcal{B}$, and hence κ . Considering that the pre-tension is expected to become lower²⁸ or at least remain unchanged between the “surfactant-avoided” and “surfactant-mediated” cases, we select a representative value²⁹ of $\sigma_{\text{pre}} \sim 0.1 \text{ N/m}$ to estimate κ . Based on this, we estimate that the bending rigidity must lie in the range $8 \times 10^3 \text{ eV} \leq \kappa \leq 4 \times 10^5 \text{ eV}$ to be consistent with the observed force-displacement response. A more detailed estimate based on taking full account of the bending stiffness and finite indenter radius gives $\kappa = 2 \times 10^5 \text{ eV}$ and is provided in the Supporting Information S5.1. This value is several orders of magnitude greater than the bending rigidity of pristine graphene.

Further insight can be gained by comparing the nonlinear stiffness $E\bar{h}$ between the two cases. For the “surfactant-avoided” samples, the measured values fall below the generally accepted 2D Young’s modulus of pristine graphene ($E\bar{h} = 340 \text{ N/m}$)²³. This difference aligns with prior observations^{6,9} and is attributed to static ripples, low-amplitude out-of-plane undulations, that are flattened during indentation, costing negligible stretching energy. In contrast, the wrinkles in the “surfactant-mediated” samples are of much larger amplitude, as confirmed by AFM images in Fig. 1c and d. These large wrinkles often form self-contacted regions that are mechanically robust and energetically expensive to deform due to van der Waals adhesion¹³.

Wrinkled graphene resonator

To further investigate the presence of bending rigidity in wrinkled graphene membranes, we measured the fundamental resonance frequency of “surfactant-mediated” samples suspended over circular cavities ($3\text{--}10 \mu\text{m}$ in diameter, 285 nm deep) etched into a SiO_2 substrate. A Fabry-Pérot interferometric setup was used for measuring the motion of the suspended membranes (see ref. 30), while a modulated laser was used to thermomechanically actuate them (see Fig. 3a and Methods). A representative SEM image of a wrinkled graphene drum is further shown in Fig. 3b.

Previous studies have shown that pristine graphene drums, of diameter $d_{\text{eff}} = 2R_{\text{out}}$, behave as tension-dominated membranes, with fundamental resonance frequency $f = \frac{2.4}{\pi d_{\text{eff}}} \sqrt{\frac{\sigma_{\text{pre}}}{\rho h}}$ where ρh is the mass per unit area of the drum^{29,31}. In contrast, bending-dominated plates follow $f = \frac{20.42}{\pi d_{\text{eff}}} \sqrt{\frac{\kappa}{\rho h}}$. The different scalings of resonance frequency with diameter make resonance

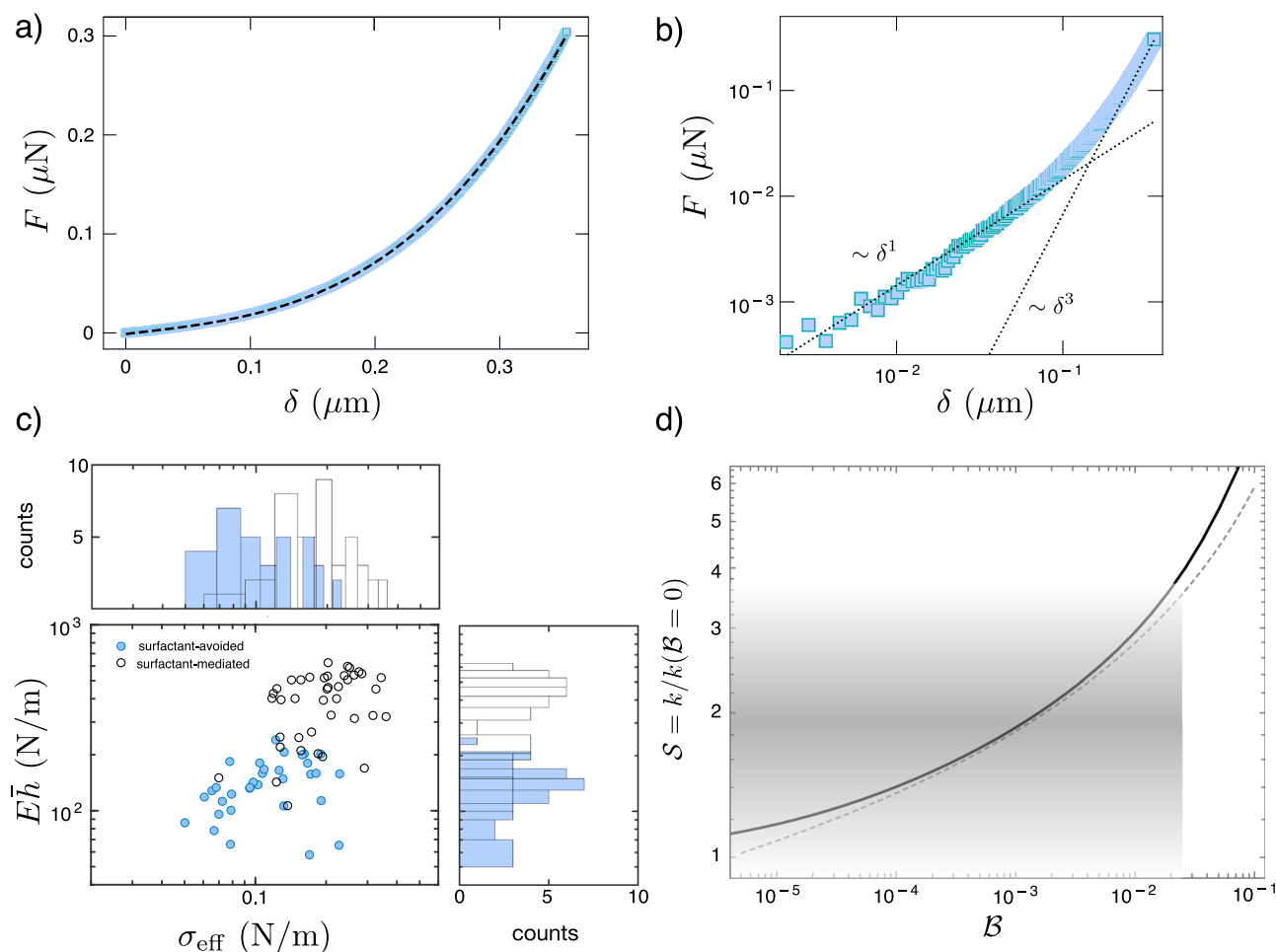


Fig. 2 | Nanoindentation of wrinkled graphene membranes. **a** Representative $F - \delta$ correlation, measured for one of the wrinkled-graphene membranes. The broken line is the best fit of the experimental data with Eq. (1). **b** The same data plotted on doubly-logarithmic scales shows the linear and cubic relationships at small and large indentations, respectively. **c** Correlation between the estimated σ_{eff} and the $E\bar{h}$ values: histograms of the pretension and the $E\bar{h}$ values (in log scale) are plotted in the side panels. **d** Enhancement of the indentation stiffness by the presence of bending

stiffness when $R_{\text{in}}/R_{\text{out}} = 2.2 \times 10^{-3}$. The analytical result for the indentation stiffness, valid for all B (see supporting information S5), is rescaled by the value when $B = 0$ to show the stiffness enhancement, S , that comes from $B > 0$ (solid curve). When $R_{\text{in}}^2/R_{\text{out}}^2 \ll 3B \ll 1$, k is given approximately by Eq. (2) (used to generate the dashed curve). The shaded region represents the range of indentation stiffening observed in our experiments, with darker regions being more likely.

measurements a sensitive probe for identifying the dominant restoring forces — especially in light of the nanoindentation results discussed earlier, which suggested the contribution of bending rigidity to the linear stiffness should not be neglected. To that end, we performed a frequency sweep across drums with a variety of diameters and fitted the resulting spectra using a harmonic oscillator model³³. As a representative example, a 7 μm -diameter drum showed a resonance frequency of 11.6 MHz with a Q-factor of 106 (see Fig. 3c). The Q-factor value is comparable to the literature reports for suspended graphene³⁴ and is indicative of moderate damping under vacuum conditions; Q-factor variation across diameters is detailed in the Supplementary Information Fig. 3. The broader dataset (Fig. 3d) shows resonance frequencies ranging from 6.7 MHz ($d_{\text{eff}} = 9 \mu\text{m}$) to 37 MHz ($d_{\text{eff}} = 3 \mu\text{m}$), revealing a scaling trend that lies between the ideal membrane and plate limits (see Fig. 3d).

To interpret this intermediate behavior, we developed a theoretical model that accounts for finite bending rigidity in membrane oscillations (see Supporting Information S5.2) and used it to fit experimental data. Assuming a pre-tension of $\sigma_{\text{pre}} = 0.1 \text{ N/m}$ —independently extracted from AFM nanoindentation for ‘surfactant-mediated’ samples and assuming that pre-tension remains unaltered when surfactant is added to the liquid–air interface, we find that the measured frequencies are best explained by bending rigidity values in

the range of 10^5 – 10^6 eV . The optimal fit corresponds to $\kappa \approx 2 \times 10^5 \text{ eV}$ for a mass density of $\rho\bar{h} = 10^{-5} \text{ kg/m}^2$, significantly exceeding the theoretical value for pristine graphene, and closely matching the value obtained from nanoindentation experiments. These results suggest that the wrinkled graphene drums operate in a hybrid mechanical regime, where both pre-tension and bending rigidity contribute to the observed dynamics.

Wrinkled graphene cantilever

Along with the increase in in-plane stiffness, the inclusion of the wrinkles is expected to make graphene stiffer in the out-of-plane direction, as shown by AFM nanoindentation and resonant measurements. However, while these methods show clearly that the effective bending stiffness of the surfactant-mediated graphene is significantly increased compared to, for example, pristine graphene, the uncertainty in the values of other parameters (notably σ_{pre} and $\rho\bar{h}$) makes determining a precise estimate of κ difficult. Within this perspective, we sought to make a direct measure of the bending rigidity; we sculpted the wrinkled membranes by removing the excess areas with a focused ion beam (see methods and Supporting Information S2 for more details) — and realized graphene cantilevers. We fabricated a total of nine cantilevers with lengths in the range of 5 to 7 μm and widths in the range of 2 to 3 μm . Figure 4a provides the scanning electron micrograph (SEM) of a

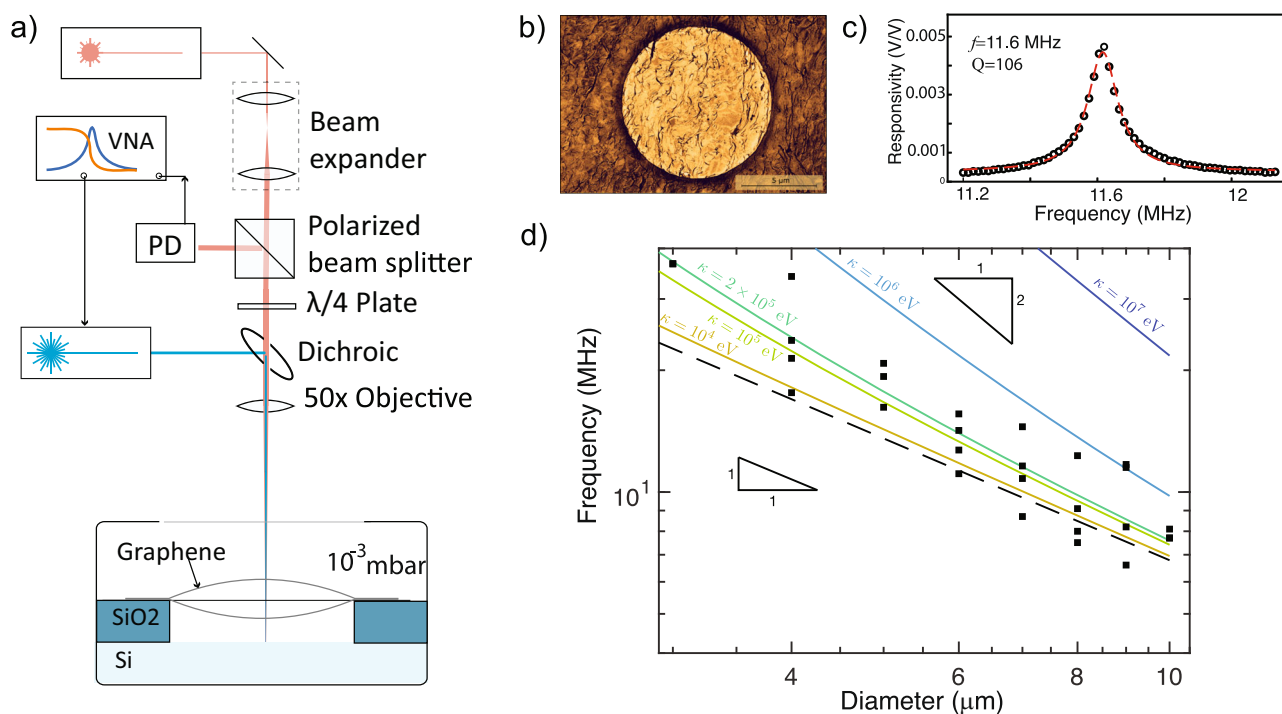


Fig. 3 | Resonance frequency measurement of wrinkled graphene drums.

a Schematic of the interferometric measurement setup used to record the vibrations. PD and VNA stand for Photodiode and Vector Network Analyzer, respectively. **b** SEM image of a wrinkled graphene drum. **c** Resonant response of a wrinkled drum with an optothermal drive of 0 dBm. The figure also includes a Lorentzian fit to the

data shown in red. **d** Diameter-dependent resonance frequencies together with fits from the theoretical model for different values of bending rigidity κ . Here, results are shown with $\bar{h} = 10^{-5}$ kg/m² and $\sigma_{\text{pre}} = 0.1$ N/m; similar plots with different values of these parameters are shown in Supplementary Fig. 5.

selected cantilever. We analyzed the bending rigidity of the graphene cantilevers further with an AFM nanoindentation in detail. Unlike fully clamped membranes, where internal tension affects the stiffness, the response of cantilevers is dominated by bending rigidity, making them a more direct probe of this property. Figure 4b illustrates a set of the force-displacement curves, measured over multiple points along the length of a graphene cantilever (note the definition of the “length” in Fig. 4a). The “approach” and “contact” regions of the curves can be respectively identified at the positive and negative sides of the “displacement” axis. Figure 4c provides a closer look at the contact region of the curves. Intuitively, it is expected that the stiffness of the graphene cantilevers decreases by increasing the length; this is translated into less steep curves in the contact region. Unlike the indentation of the fully-clamped membranes (Fig. 2a), where the force grows by higher orders of the displacement (reaches the third order at sub-micron displacements), the displacements in cantilever structures are in much smaller ranges (tens of nanometers) and preserve linear dependencies with constant, but length-dependent slopes. Importantly, the diminishing slope is evident at larger lengths along the cantilever.

To minimize the effect of local perturbations by the wrinkles in the later analysis, we mapped the graphene cantilevers in windows of width $0.5 \mu\text{m}$ along the length, using the AFM set-up. The inferred “height” and “slope” profiles are plotted in Fig. 4d. The process of sculpting induced an up-curvature in the majority of the cantilevers. Particularly for this sample, the cantilever curled up by $\sim 1.8 \mu\text{m}$ at a length of $4 \mu\text{m}$. The diminishing slope of the “contact region” — locally perturbed by the wrinkles — is evident in the bottom mapping.

Similar to the indentation of the fully clamped membranes, the spring constants of the AFM probe and graphene cantilever fall in a series configuration for the combined system (see the inset schematics in Fig. 4e). The overall spring constant reads as:

$$k_{\text{tot}}^{-1} = k_p^{-1} + k_g^{-1}, \quad (4)$$

with k_p and k_g referring to the spring constants of the probe and graphene cantilevers, respectively. We measured the constant k_p by pushing the AFM cantilever against the rigid substrate. The spring constant of the graphene cantilever, however, strongly decays with the length, as $k_g = 3\kappa w/l^3$, with κ , l , and w as the bending rigidity, the length, and the width of the wrinkled graphene cantilever. Considering the series spring model with length-dependent k_g , the measured overall spring constant of the system (k_{tot}) approaches $k_{\text{tot}} \rightarrow k_p$ and $k_{\text{tot}} \rightarrow 0$ in the extremes of $l \rightarrow 0$ and $l \gg 0$, respectively. Note that for the analyses in this section, the zero displacement ($d = 0$) is defined as the point along the length where the slope starts to deviate from the measured stiffness of the AFM cantilever (~ 0.3 N/m). Figure 4e fits the experimentally measured k_{tot} (i.e., the slope of the contact region in Fig. 4b and c) with the model of Eq. (4). The presented slope (the vertical axis) in this plot is the average over the widths ($0.5 \mu\text{m}$) of the mapped window, as in Fig. 4d. Such fittings provide estimates for the bending rigidity κ , as summarized in Fig. 4f.

Theoretically, a monolayer flat graphene sheet is postulated to show an ultimately small bending rigidity, estimated by neglecting the effect of the intrinsic thermal fluctuations of the lattice^{2,3}. At finite temperatures, anharmonic coupling between in-plane and out-of-plane thermal fluctuations renormalizes the bending rigidity as $\kappa(l) \sim \kappa l^\eta$, where l is the characteristic length scale, κ is the bending rigidity of pristine graphene, and $\eta \approx 0.8$ ³⁵. In fact, it has been experimentally shown that the thermal fluctuations in micro-scale samples (with no wrinkles involved) can stiffen the graphene by up to five orders of magnitude¹⁵ (the grey-shaded region in Fig. 4f). Our samples, with deliberately included wrinkles, on the other hand, showcase κ in the range of 10^6 to 10^7 eV, which is reasonable considering the combined hardening effects of the thermal fluctuations and large-amplitude wrinkles. These findings are consistent with our estimates of bending rigidity from the AFM nanoindentation as well as resonant measurements of all edge-clamped square membranes.

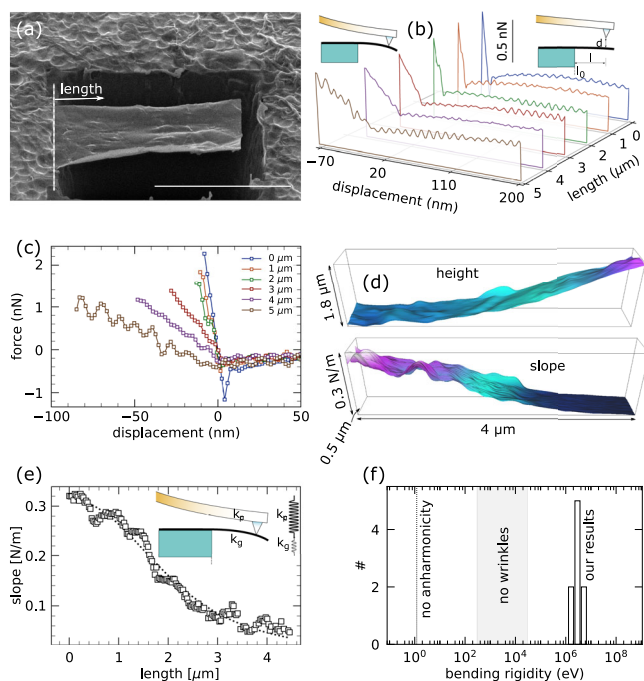


Fig. 4 | Characterization of the wrinkled graphene cantilevers. **a** Scanning electron microscopy of a graphene cantilever; The scale bar corresponds to 5 μm . **b** Force-displacement relation of the graphene cantilever in **(a)**, obtained by nanoindentation at different lengths. **c** Comparison of the slopes of the curves in the contact region in **(b)**; **d** 3D height (top) and slope (bottom) profiles measured along the same cantilever. **e** Fitting of the measured slope along the graphene cantilever with the model of Eq. (4); The data points are the average over the width of the 0.5 μm stripe in the bottom figure in **(d)**. k_p and k_g in the inset schematic refer to the bending rigidities of the probe and the graphene cantilever, respectively. **f** Bending rigidities of the wrinkled graphene cantilevers (our results), benchmarked with the published data. “No anharmonicity” corresponds to the theoretical value of the bending rigidity ($\kappa = 1.2$ eV) of graphene samples. “No wrinkles” corresponds to the microscale graphene samples experimentally measured previously¹⁵, with no extra wrinkles included.

Mechanical cantilevers offer ultrasensitive measurements across fields ranging from material science to diagnostics^{36,37}. Recent advances in miniaturization have led to devices that are only a few nanometers thick, such as Al_2O_3 cantilevers^{16,38}. Yet, their ultimate performance can only be achieved when the thickness is shrunk to the atomic limit, a regime where maintaining mechanical stability poses a fundamental challenge. The realization of monolayer graphene cantilevers, hitherto, has met the challenge of the instability of atomically thin cantilevers due to the diminishing bending rigidity associated with the extremely low thickness of the material. Our approach of increasing the bending rigidity by the inclusion of a finite level of wrinkles stands out in this respect. The graphene cantilevers reported in our work represent unprecedented examples of ultra-light, ultra-thin, and soft, yet stable, mechanical structures. Benchmarking the characteristics of the graphene cantilevers with the known system of AFM cantilevers is interesting here. The spring constant of a typical AC mode AFM cantilever (NCLR Nanoworld is arbitrarily selected here) with a 200 μm length is in the range of a few tens of N/m, while a graphene cantilever with only 5 μm length exhibits a spring constant of only 10^{-2} N/m. The bending rigidity of such an AFM cantilever, on the other hand, is on the order of 10^{13} eV; the value drops by up to seven orders of magnitude for graphene cantilevers, reaching 10^6 eV. Interestingly, the mass of the graphene cantilevers—achieved from a mono-atomic starting material—is astonishingly low. With a mass density of $\rho h \approx 10^{-5} - 10^{-6}$ kg/m²³⁴, we estimate a mass of $10^{-13} - 10^{-14}$ g for the cantilever shown in Fig. 4, which is orders of magnitude lower than that of typical AFM cantilevers ($\sim 10^{-7}$ g). Consequently, free-standing graphene cantilevers can access a previously

unexplored mechanical regime, enabling measurements at the very limits of force, mass, and displacement detection.

Discussion

In this study, we investigated the effect of wrinkling on the elastic properties of graphene and assessed its implications for nanomechanical systems. We fabricated different types of suspended structures—fully clamped drums and cantilevers—and characterized them using both nanoindentation and resonant measurements. Indentation measurements of wrinkled membranes revealed that their mechanical response is governed by a combination of tension and bending rigidity, indicating a substantial enhancement of the effective bending rigidity compared to flat graphene. Complementary resonant measurements further showed clear deviations from the conventional tension-dominated behavior, consistent with a hybrid mechanical regime in which both pre-tension and finite bending rigidity contribute to the dynamics. The consistency between static and dynamic measurements provides a coherent picture of the modified mechanical response in wrinkled graphene. Despite the self-organized and non-periodic nature of the wrinkle network, the extracted bending rigidity values remain within a relatively narrow range, indicating that the stiffening mechanism is robust and does not rely on a specific wrinkle morphology. Furthermore, the increased bending rigidity enables the realization of free-standing graphene cantilevers, overcoming the intrinsic instability of atomically thin membranes. These structures combine extremely low mass with low stiffness, placing them in a distinct mechanical regime compared to conventional doubly-clamped systems. In this context, their high compliance is particularly relevant for static nanomechanical sensing such as surface-stress based³⁹ and nanomotion sensors⁴⁰, where sensitivity is governed by displacement under applied force. Overall, this work establishes wrinkling as an effective approach for geometry-induced mechanical reinforcement in 2D materials.

Methods

Fabrication of wrinkled membranes and cantilevers

Wrinkled graphene membranes were fabricated via a surfactant-mediated process at the air-liquid interface. Graphene grown on copper foil was floated on an ammonium persulfate (APS) solution to etch away the substrate, after which a surfactant was introduced to reduce surface tension and obtain in-plane contraction of graphene. The contraction led to a network of wrinkles, with an approximate 30% reduction in membrane area. The samples were subsequently rinsed and transferred onto microcavities. To realize cantilever structures, suspended wrinkled membranes were further patterned using a stepwise focused ion beam (FIB) milling process. Initially, parallel cuts were used to obtain a doubly clamped beam geometry, and subsequent milling steps removed residual connections, releasing one end to form a cantilever. Further details about the fabrication can be found in the Supporting Information.

AFM and dynamic measurements

Atomic force microscopy (AFM) was employed for both topographical imaging and mechanical characterization using a JPK NanoWizard 4 XP system. Surface morphology and wrinkle distributions were first assessed in tapping mode to identify suspended membranes and cantilevers. Mechanical properties were then measured via nanoindentation in QI mode, where fully clamped membranes were indented at their center, and cantilevers were indented along predefined lines along their length under a controlled force. The resulting force-deflection response was recorded, and the sample deformation was obtained after correcting for the compliance of the AFM probe. Different probes were used depending on the mode (high-stiffness cantilevers for imaging and softer probes for indentation).

As per dynamic characterization, the measurements were performed in vacuum (10^{-3} mbar) at room temperature using a Fabry-Pérot interferometric setup where a power-modulated blue laser ($\lambda = 405$ nm) thermomechanically actuated the membranes, while a red laser ($\lambda = 633$ nm) detected their vibrations. The reflected intensity was then recorded with a

photodiode and analyzed via a vector network analyzer (VNA) synchronized with the actuation signal. Further details on AFM and resonance measurements are provided in the Supporting Information.

Data availability

The datasets supporting this study are not publicly available as they may be included in ongoing research projects that aim to further characterize related 2D materials, but are available from the corresponding authors upon reasonable request.

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Author contributions

H.A., G.V., D.V. and F.A. conceived the experiments and methods; H.A. and R.P. fabricated the graphene samples; R.P., H.A. and H.L. conducted the measurements, and the experimental data were analyzed by H.A., G.V., D.V. and F.A.; D.V. further built the theoretical model; F.A. supervised the project;

and the manuscript was written by H.A., D.V. and F.A. with inputs from all authors.

Competing interests

The authors declare no competing interests.

Additional information

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