Spontaneously ordered hierarchical two-dimensional wrinkle patterns in two-dimensional materials

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ABSTRACT

To achieve two-dimensionally (2D) ordered surface wrinkle patterns is still challenging not only for the atomic-thick 2D materials, but also general for all the soft surfaces. Normally disordered 2D wrinkle patterns on isotropic surfaces can be rendered via biaxial straining. Here, we report the 1D and 2D ordered wrinkle patterns in 2D materials can be produced by sequential wrinkling controlled by thermal straining and vertical spatial confinement. The various hierarchical patterns in 2D materials generated by our method are highly periodic, and the hexagonal crystal symmetry is obeyed. More interestingly, these patterns can be maintained in suspended monolayers after delamination from the underlying surfaces which shows the great application potentials. Our new approach can simplify the patterning processes on 2D layered materials and reduce the risk of damage compared to conventional lithography methods, and numerous engineering applications that require nanoscale ordered surface texturing could be empowered.

MAIN TEXT

The surface instability induced spontaneous wrinkling prevails in lots of soft material surfaces.[1-8] Surface wrinkles can be developed by different strain conditions on surfaces with respect to the interior bulk parts, under direct straining,[9-19] thermal treatments,[1, 20, 21] chemical or environment changes.[22] Particularly, 1D compressive surface strain is known to generate instability and well assembled 1D periodic wrinkle patterns,[1, 9, 14, 16, 19, 23] however, it is more difficult to generate 2D ordered wrinkle patterns.[7, 8] The intrinsic disorder in 2D wrinkle patterns mainly comes from the mixed modes of 2D instability, as well as the interactions between

wrinkles in different orientations, especially the complex strain fields and ultra-sensitive morphologies at the crossings of wrinkles.[3, 4, 11, 12, 23]

Wrinkles are also frequently observed in the atomic-thick 2D materials,[24] such as graphene[13, 21, 25-27] and 2D transition metal dichalcogenides.[2, 28] The intentional 1D wrinkling in 2D materials controlled by external compressive loading (*e.g., via* flexible substrates) was the most accessible method in previous literatures.[4, 12, 13, 15, 26] In specific, the periodic 1D wrinkle wavelengths in 2D materials have been exploited to understand the flexural properties, *e.g.*, the bending rigidities.[18, 19, 26] Recently, we have identified and measured the critical length for transition between stable ('clipped') and unstable ('unclipped') 1D wrinkles of 2D materials.[24] The wrinkle stabilization relied on that two side walls of wrinkle were 'clipped' by van der Waals interaction.[24] Although the 1D wrinkles in 2D materials have been extensively studied, the 2D wrinkle patterns in 2D materials remain largely unexplored. Some polymer films like ethylene glycol diacrylate (EGDA) and 2-hydroxye-thyl methacrylate (HEMA) can generate 2D periodic wrinkles on biaxial pre-stretched substrate.[18] However, biaxial compressive stress on monolayer (1L) graphene was demonstrated to yield the irregular wrinkle patterns,[11-13] or totally disordered crumpled surface morphologies. [11, 12] Additional (lithography) masks on graphene could enable the ordered patterns, however, the wrinkle themselves were still disordered or just following the underlying disordered surface undulations of polymer substrates.[13]

Albeit in the continuum context graphene (or other isotropic 2D materials) exhibits isotropic in-plane mechanical responses, in the atomic scale these wrinkles indeed have the favored crystal orientations.[15, 22] Controlling ordered 2D wrinkles on graphene demands understanding effects of

crystallographic orientation on wrinkle formation.[7, 8] For instance, wrinkles in 1L-graphene along zigzag or armchair directions (in honeycomb lattice) have lower formation energies than the other directions, particularly the zigzag direction.[26] Therefore, spontaneous wrinkles under equal-biaxial stress in graphene can potentially follow preferential directions which might be able to develop into regular patterns, predicted by several simulation works.[6, 12, 17] However, the ordered 2D wrinkles in graphene or any other 2D materials have not been achieved yet, primarily owning to the randomly merging of wrinkles under the biaxial compressions.[7, 8] Here we introduce a novel strategy that can spontaneously generate 2D ordered wrinkle patterns in graphene and other 2D materials, on the basis of sequential wrinkling control in different directions and spatial confinement of wrinkles in the vertical direction. Various pattern morphologies can be produced and maintained even in suspended graphene layers.

The graphene and 2D rhenium disulfide (ReS_2) used in the experiments were grown by chemical vapor deposition (CVD) (see Methods).[24] The growth condition for graphene was optimized so that 1L continuous full film graphene could be obtained on copper foil. Basic characterizations on the quality of 1L-graphene have been shown in Figure S1. The 1L-graphene was then transferred onto a uniaxially pre-stretched polydimethylsiloxane (PDMS) substrate using the polymethylmethacrylate (PMMA) method (see Methods).[2] The wrinkle patterns in 1L graphene was triggered by a simple thermal treatment (annealing) when graphene was encapsulated between PDMS and PMMA (see schematic Figure 1a). After treatment, the PMMA on the top of graphene was removed by acetone (see Methods). The monoaxial pre-strain in PDMS was either maintained or released followed by further characterizations on the post-treatment graphene samples.

The typical graphene surface morphologies (images taken by scanning electron microscopy (SEM), see Methods) after thermal annealing on the pre-strained PDMS (Figure S2) have been presented in Figure 1b-e. The wrinkle patterns were apparently generated in the 1L-gaphene. Besides, the patterns are two dimensionally ordered. The angles between the three principle directions in the wrinkle patterns were distinctly 60 °, in agreement with the favored zigzag directions in graphene. Upon closer inspection, the wrinkles were all "clipped wrinkles" that are mechanically stable (Figure 1e, g and Figure S3),[24] with a nearly constant period (wavelength). The gaps between wrinkles hence remained flat (Figure 1e). Particularly, the wrinkling directions (*viz.* up or down with respect to the basal plane) varied with wrinkle (line) directions (Figure 1f, h). Flat zones at different heights can also be observed in some wrinkle patterns (Figure 1d). These well ordered 2D wrinkle features suggested a different wrinkle formation mechanism from the previous 2D disordered wrinkle patterns in graphene.[11, 12, 25]

In our samples, the encapsulated 1L-graphene layer by PDMS and PMMA experienced a biaxial stress field upon thermal annealing. Due to the uniaxial pre-stretching (30 %) loaded on PDMS, the initial tensile (longitudinal, stretching direction) and contractive (lateral) strain are in presence (see Figure S4, S5). After the graphene with PMMA layer was transferred onto the PDMS surface, the double van der Waals (vdW) interfaces (PMMA/graphene/PDMS) were formed (see Figure 1a and Figure 2a). Further thermal annealing (60 ~ 100 ^{o}C) could partially relax the pre-strain in PDMS along both directions (Figure 2a). During relaxation, contraction in the longitudinal direction as well as expansion in lateral direction were additionally imposed on PDMS. Moreover, considering the relatively low vdW interactions at the interfaces

(graphene/PMMA and graphene/PDMS),[5,6,20] layer sliding between graphene/PDMS or graphene/PMMA was easily triggered during strain relaxation upon thermal annealing.

The initialization of wrinkles here are in analogous to the 1D instability on surfaces,[5] however, owning to the spatial confinement in vertical direction between PDMS and PMMA, the 1D wrinkles would not develop into the common sinusoidal shapes. Rather, the curvatures would be immediately focused at the nodes (clipped wrinkles) separated by the periods of initial surface instabilities. By using the vdW forces (static frictional force) between PDMS and graphene (which is the maximum compressive stress could be applied in graphene) and the basal plane rigidity of graphene, the instability wavelengths (*viz.* the period in wrinkle patterns) can be estimated as *ca.* 240 nm, by the rule of Euler instability.[24] This result is very close to our experimental observations above (Figure 2b). Accordingly, the period (wavelength) of such graphene wrinkles can be modulated by the selection of flexible substrates and cover layers (hence different max. interfacial vdW interactions), as well as the selection of 2D materials. We have carried out similar wrinkling experiments on 2D ReS_2 flakes and observed the similar phenomena (see supplementary Figure S6). Figure 2c compared the wrinkle wavelengths of graphene with different preparation methods (CVD growth or mechanical exfoliation, see supplementary Figure S7-S9) and some other 2D transition metal dichalcogenides (TMD) like 2D ReS_2 by our approach.

More importantly, as the longitudinal and lateral thermal stress were not equal, the instability induced wrinkles along different directions in 2D were actually sequentially generated. The wrinkles in the second direction were initialized and developed in the flat areas between the

parallel wrinkles along the first direction, so did the third directions. Eventually, all the three equal zigzag directions in graphene could be occupied by the three set of parallel periodic wrinkles (see Figure 1c-d, and inset of Figure 2b), assembled into the hierarchical periodic 2D patterns as shown above. Distinctly, in 2D ReS_2, due to the crystal anisotropy (hence different elasticity along each direction), the wrinkle wavelengths in the three directions are non-equal (Figure S6 and S10).

The "up" and "down" wrinkles (Figure 1e,g) were respectively associated with the relative contraction/expansion between PDMS and graphene, along different directions. If the 2D stresses were equally applied, *e.g.*, a 2D symmetrical biaxial pre-strain was applied on PDMS (Figure 3a) while the other conditions kept the same as above, then the generated wrinkle patterns would become random and disordered (Figure 3a and Figure S11), which should be attributed to the mode mixing on the surface instability and wrinkle interactions in 2D, similar to the previous reports. On the other hand, the wrinkles in three principle orders have similar height without effect of pre-strain relaxation (Figure S12, S13), due to its formation merely favored to crystal isotropy of graphene (Figure S14).

In addition, if the pre-strain condition in PDMS was set close to the pure uniaxial condition (Figure 3b, c), the 1D or close-to-1D wrinkle patterns with hierarchical structures (including first and second generation of instability) can be generated (Figure 3b-d). For 1D case, the first generation and second-generation wrinkles were in parallel and alternatively assembled (second generation wrinkles located at the center between two first generation wrinkles), and wrinkle heights (intensity) could be clearly observed to split into two levels, corresponding to the two

generations of wrinkles, respectively (Figure 3e). Therefore, our experiments also suggested the shape of 2D patterns could be tunable by controlling the symmetry of the pre-strain field in these flexible polymer substrates.

In all the wrinkle patterns in graphene, we have observed many different hierarchy levels in final products (Figure 4a). They were averaged along the single directions and over large areas (100 × 100 μm). The dependent hierarchy levels should be attributed to the magnitude of the pre-straining in PDMS. Besides that, the tensile stress in this range (10 – 50 %) was linear dependence on pre-strain magnitude regardless of thickness of PDMS.[29] For a higher pre-straining, the thermal relaxation induced interfacial stress thereafter could generate more times of Euler instabilities.

According to our previous understandings in the mechanical stability of wrinkles in 2D materials, the clipped wrinkles here should be stable against thermal or mechanical perturbations.[24] It was indeed discovered the 2D wrinkle patterns formed here were sufficient robust and stable, during the multiple post-processes including mechanical unloading, surface Au coating (for SEM imaging) and annealing, etc. In some extreme cases, as exhibited in Figure 4b, the 2D wrinkle patterns in graphene were even intact by the delamination from the straining substrates (1L graphene membrane bending and suspension globally can be clearly observed with the 2D wrinkle patterns). The global bending and suspended graphene layer were created by further straining control in PDMS substrate, *e.g.*, release of the pre-straining mechanically (see Methods for details). Due to the nm-thick surface Au coating to reduce charging effect (non-conductive substrates were used here) prior to SEM observations, the curvatures of graphene

wrinkles have been blurred in the SEM pictures. Nevertheless, the survival of these wrinkle patterns after suspension have been confirmed. In this instance, these 2D ordered wrinkle patterns made of 2D materials are appealing, because the patterns can be generated on certain flexible substrates and then suspended or transferred to other targeted locations.

In conclusion, we have developed an efficient approach for spontaneously generating the ordered 2D wrinkle patterns in 2D materials, including monolayer graphene and transition metal dichalcogenides. The innovative sandwich structure, by encapsulating the soft polymer surfaces on both sides of graphene, can provide the unique vertical confinement, and enable the ordered 2D hierarchical wrinkle patterns. The regular wrinkle structures are basically developed sequentially along different directions without interactions which may lead to disordering. A variety of application potentials for such spontaneously 2D-ordered wrinkle patterns of graphene and other 2D materials, such as the photonic and meta-material surfaces,[9] hydrophobic surfaces[3, 12] as well as the nano-fluidics using 2D materials,[4, 25] could be highly attractive to the relevant fields.

METHODS

Sample Preparation

The graphene was grown on 75 μm-thick copper foil (99.95 %, GoodFellow, England) using CVD system at atmospheric pressure. The foil was inserted to center of the tube furnace with flow of Ar/H_2 (ratio of 20:1) and 10 sccm CH_4 (1 % of volume) for 45 minutes at 1060 ºC. 2D ReS_2 was grown on a c-face sapphire substrate by the atmospheric CVD system. Ammonium

perrhenate (NH_4ReO_4) (Aldrich, 99.999 %) and Sulfur powder (Aldrich, 99.998 %) were used as precursors with weight ratio 1:50. A two-zone splitting tube furnace was used to control accurately Sulfur and substrate zone temperature, respectively. During the deposition process, argon gas (80 sccm) was used as the carrier gas to transport sulfur vapor to substrate zone. The sulfur zone was ramped to 200 °C, at the same time, the substrate zone reaching 850 °C in 30 minutes and then held for 10 minutes. The as-grown graphene was coated by a thin layer of PMMA (A4, Chem) using spin-coater in speed of 3000 rpm for 1 minute. Afterwards, underlying copper foil was etched away copper etchant (Aldrich, 667528). The remained PMMA/graphene was rinsed by transferring to DI water (×3 times) before transferred onto the stretching PMDS substrate. The stretching stage consist two separate monoaxial moving bars. 500 µm-thick PDMS film was fixed onto these bars, the un-strained suspended PDMS between two bars was around 1 × 1 cm. The uniaxial strain (10 – 50 %) was applied on PDMS by moving two bars apart. The PMMA/graphene after transferred onto stretching PDMS was dried in oven at 60 – 100 ºC for 30 minutes. After drying, sample was cooled down to room temperature then PMMA was remove by acetone.

Scanning Electron Microscope (SEM). The wrinkle topography was observed using Environment controlled SEM (Quattro ESEM, Thermo Scientific™) under various accelerated voltages. Prior to measurement, a few-nm-thick Au film was sputtered on sample to reduce charging effect.

Raman Spectroscopy. The quality of graphene transferred on PDMS was characterized using Renishaw Raman Spectroscopy 2000 under laser 514 nm excitation with approximately 500 nm spot size.

Atomic Force Microscope (AFM). The topographic images were carried out using using AFM5300E system (Hitachi, Japan). In order to reduce surface deformation, tapping mode was applied for observation of the topography, using a Si-DF3P2 cantilever (Hitachi, Japan) with approximately 10 nm tip curvature.

X-ray Diffraction (XRD). The XRD characterization of graphene on PDMS samples were measured using Bruker D2 Phaser with LYNXEYE XE-T detector, which scanned over the sample in the 2-theta range of 5 – 80 °, with the resolution of 0.02 °.

FIGURES AND FIGURE LEGENDS

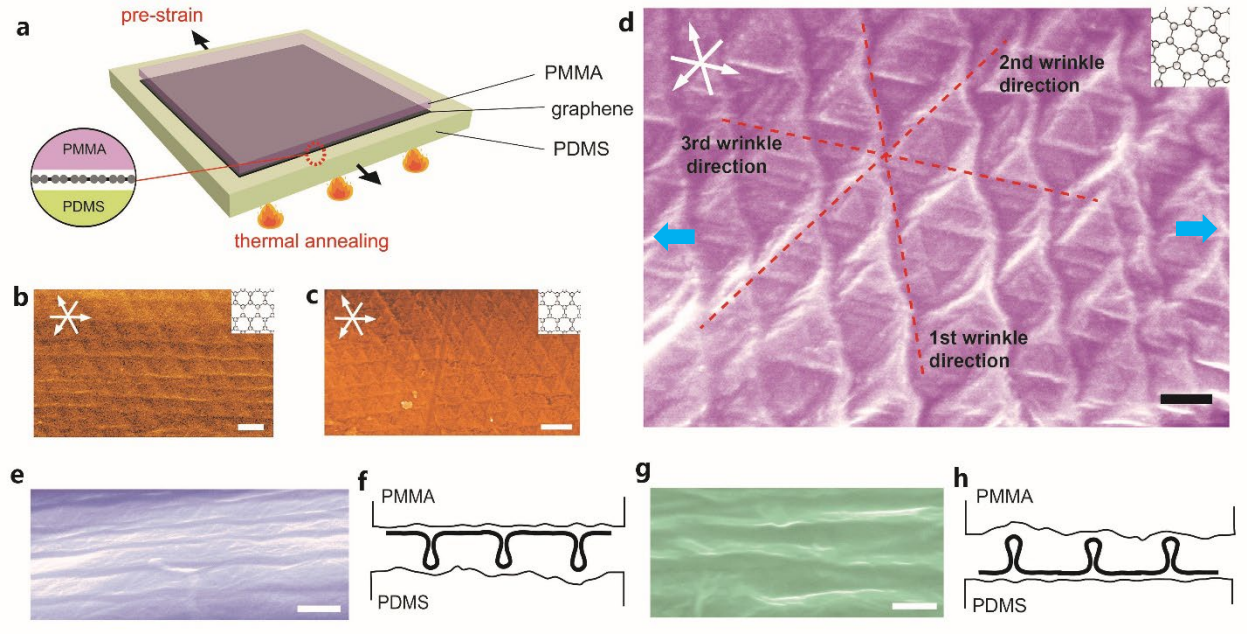

Figure 1. 2D ordered wrinkles in graphene. (a) Schematic illustrates graphene sample sandwiched between PMMA and pre-strained PDMS. **(b-d)** SEM images of spontaneously ordered hierarchical two-dimensional wrinkle patterns following the crystal orientation in graphene. The white arrows (upper-left) and crystal structures (upper-right) highlight the favored wrinkle directions. The blue arrows show pre-strained direction. **(e-h)** Two modes (**e,f** for down and **g, h** for up) of periodic "clipped wrinkles" formation in 1L graphene. Scalebars in (**b,c**) = 1 μm. Scalebars in (**d,e,g**) = 300 nm.

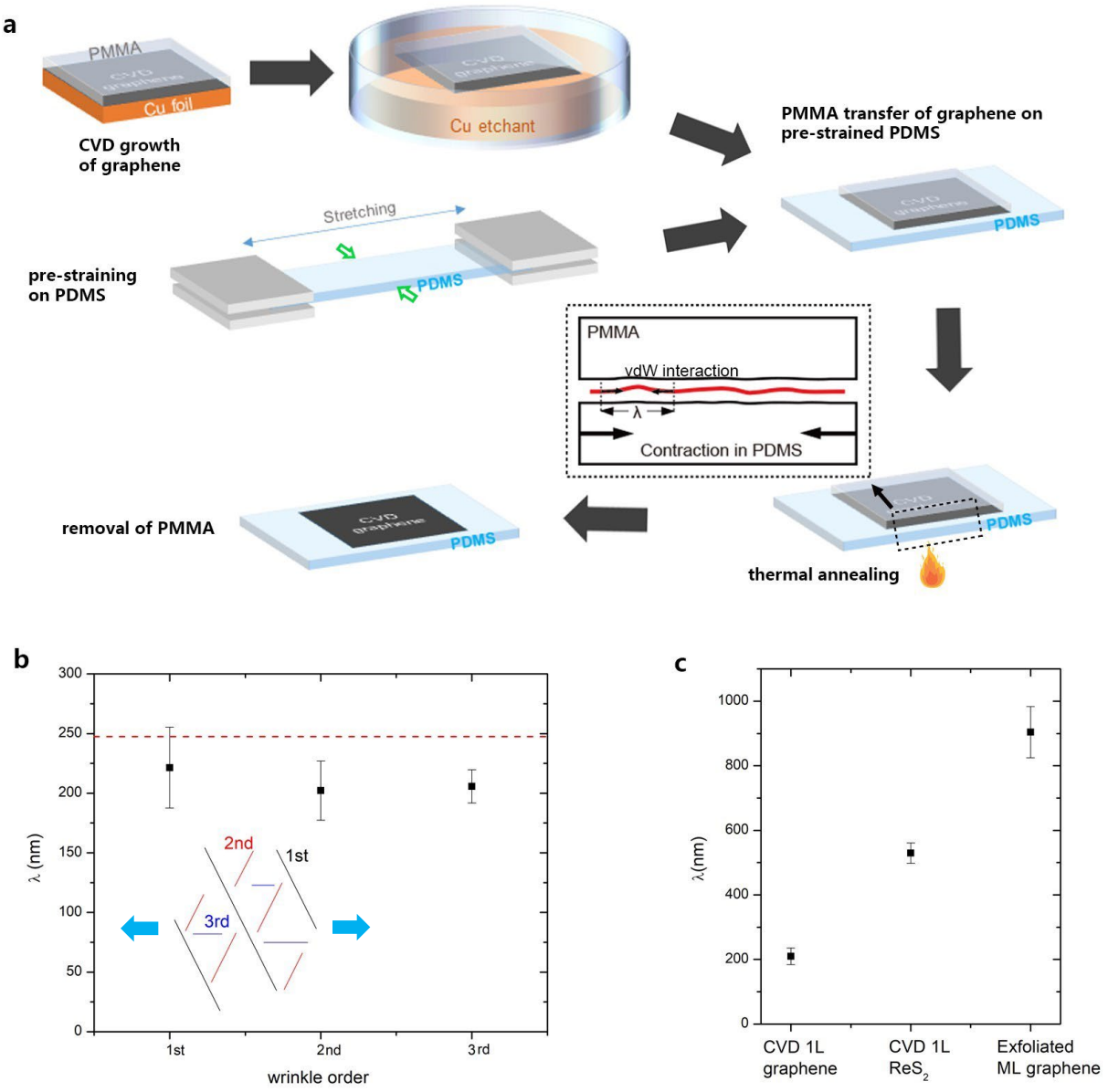

Figure 2. Formation of spontaneously ordered 2D winkles in graphene. (a) Schematic illustrates the preparation process to generate the hierarchical 2D wrinkles on graphene. **(b)** The wavelength of individual wrinkle order in graphene, compared with the theorical instability wavelength of graphene on PDMS (red dash line) calculated according to Euler instability. The blue arrows show pre-strained direction upon the wrinkle formation. **(c)** The wavelength of wrinkle formation on CVD monolayer (1L) graphene, 1L ReS_2 and exfoliated multilayer (ML) graphene on PDMS.

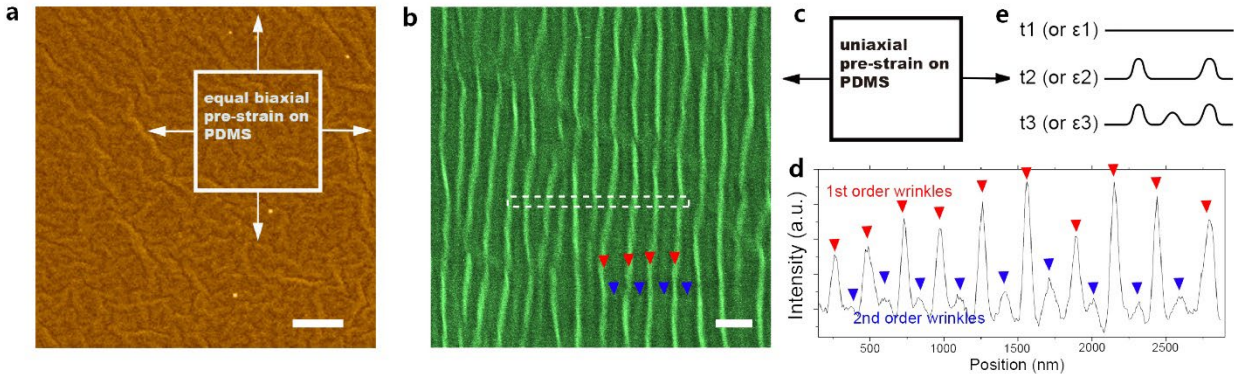

Figure 3. Hierarchical wrinkle formation on PDMS dependent on pre-strain field. (a) SEM images of 1L-graphene equally biaxial pre-strain and **(b)** uniaxial pre-strain PDMS, scale bar = 0.5 µm. **(c)** Direction of uniaxial pre-strain in (b). **(d)** Line profile shows 1st order (red arrows) and 2nd order (blue arrows) wrinkles generated by the uniaxial pre-strained graphene pattern in (b). **(e)** Formation mechanism of the spontaneously ordered wrinkles with different generations (hierarchy no.).

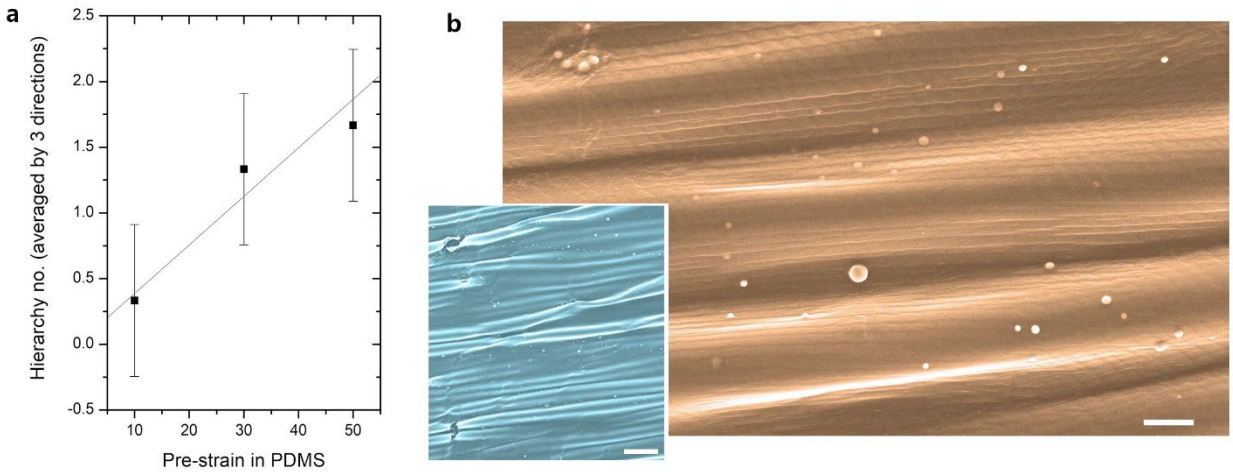

Figure 4. Dependence on the pre-strain amplitude and suspended 2D wrinkle patterns. (a) Hierarchical number (no.) under the effect of pre-strain amplitude in PDMS. **(b)** SEM images of graphene experiencing global curvature (pre-strain completely unloaded in PDMS), showing that the initial hierarchical 2D wrinkle patterns (the lattice features with periods of around 200 nm) in 1L-graphene survived after delamination, wrinkling and suspension. Scale bar = 2 μm. (Lower-left) The low-magnification image for (b), scale bar = 6 μm.

ASSOCIATED CONTENT

Supporting Information.

Supporting information includes Figure S1 to S14. This material is available free of charge via the internet at http://pubs.acs.org.

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TOC Figure