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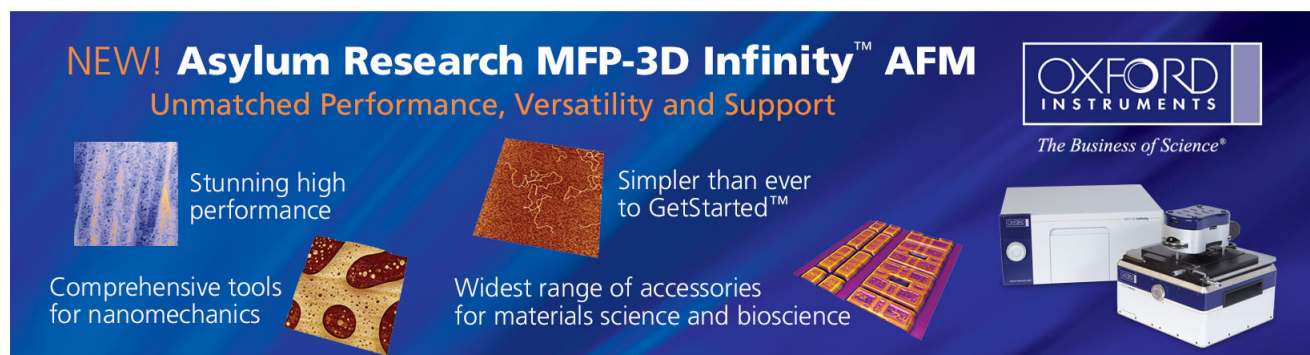
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Extremely compliant and highly stretchable patterned graphene

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Graphene is intrinsically ultra-stiff in its plane. Its huge mechanical mismatch when interfacing with ultra-compliant biological tissues and elastomers (7–9 orders of magnitude difference in stiffness) poses significant challenge in its application to functional devices such as epidermal electronics and sensing prosthesis. We offer a feasible and promising solution to this significant challenge by suitably patterning graphene into a nanomesh. Through systematic coarse-grained simulations, we show that graphene nanomesh can be made extremely compliant with nearly zero stiffness up to about 20% elongation and then remain highly compliant up to about 50% elongation. © 2014 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4874337>]

Graphene has attracted lots of attention because of its exceptional electronic, mechanical, and chemical properties.^{1–3} For example, being ultra-thin and electronically stable down to sub-nanometer, graphene is explored as a potential solution to address the physical limit of silicon-based electronics. More intriguingly, the optical transparency and mechanical durability of graphene further motivate its potential application in emerging flexible and stretchable electronics technology, such as electronic skins, smart textile, and paper-like displays.^{4–13} Existing explorations of these opportunities have mainly focused on the electronic performance of graphene devices. A crucial challenge to successful application of graphene in flexible and stretchable electronics, however, remains largely unexplored. Applications such as electronic skins and epidermal electronics require interfacing with biological tissues and elastomeric substrates, which are extremely compliant (e.g., with a stiffness ranging from ~1 KPa (tissues) to 100s KPa (elastomers)). By contrast, pristine graphene has an ultra-high stiffness (~1 TPa).³ Such a huge mechanical mismatch (7–9 orders of magnitude) between graphene and its interfacing materials poses a significant challenge in the functionality and reliability of these flexible and stretchable devices, which are often required to sustain large, repeatable deformation in operation. To address such a challenge, in this Letter, we demonstrate a feasible and promising solution by patterning pristine graphene into large area graphene nanomesh (GNM). Through a bottom-up scalable coarse-grained (CG) simulation scheme, we show that suitably patterned GNM can be made extremely compliant (with a negligible stiffness) up to elongation about 20% and then remain highly compliant (e.g., with an effective stiffness more than 20 times lower than that of pristine graphene) up to elongation about 50%. The extremely compliant and highly stretchable GNM (vs. pristine graphene with a tensile failure strain of about 20%) thus holds promise to meet the stringent operating requirements of flexible and stretchable electronics, while the associated desirable tunability of the electronic

properties of GNM could further inspire devices with multifunctionalities.

Pristine graphene has zero band-gap, an intrinsic challenge for logic applications.^{14,15} Graphene nanoribbons with a width of less than 10 nm offer a possible solution by rendering a band-gap suitable for room-temperature transistor operation.^{16–21} However, graphene nanoribbon device cannot meet the needs of practical devices and circuits that require the fabrication of dense arrays of ordered nanoribbons.^{21–24} GNM, a network of percolating graphene nanoribbons with an ordered pattern, is proposed and fabricated as an effort to overcome such difficulties of graphene nanoribbons by opening up band-gap in a large sheet of patterned graphene as a semiconducting thin film.^{25–30} It has been demonstrated that GNM can be applied as gas sensors with a high sensitivity.³¹ Enthusiasm for GNM-based devices aside, existing research on GNM mainly focuses on its electronic properties, while little study, if not none, has been reported on its mechanical properties, which are crucial for integrating GNM into flexible and stretchable electronic devices or interfacing with other materials.^{32–34} Lack of such mechanics studies can be partially attributed to the fact that the GNM fabricated and used in devices have a length scale ranging from 100s nanometer up to microns,^{27,30,31} making fully atomistic study of the mechanical behaviors of GNM computationally prohibitive.^{35,36} To overcome such a difficulty, we present a scalable bottom-up CG simulation scheme of graphene, which enables the systematic mechanics study of the GNM of practical length scale.

The uniqueness of our CG simulation scheme of graphene is twofold. First, it is a bottom-up approach, starting from the atomic structure of graphene, and scalable to any orders of CG levels. Therefore, such a CG scheme can be tailored to be suitable to simulate graphene systems of a wide range of length scales. Second, the hexagonal lattice symmetry, the origin of many exceptional properties of graphene, is maintained in all orders of CG levels. Such a feature maximizes the intrinsic similarity between the atomic structure and CG structure of graphene, the key to high precision and fidelity of CG simulations. Figures 1(a)–1(c) show the representations of the first three orders of CG levels, respectively. The building block of 1st order CG, termed as 1st order CG

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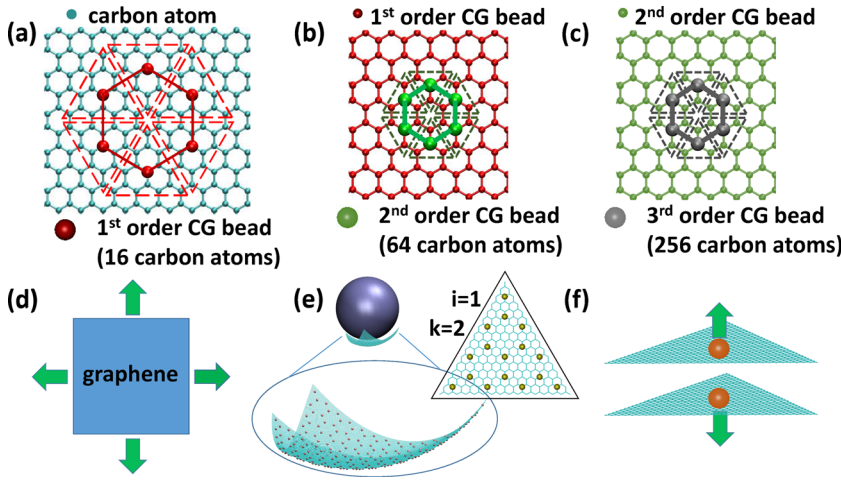


FIG. 1. A bottom-up scalable CG simulation scheme of graphene. (a)–(c) Mapping from atomic graphene (cyan) to 1st order CG beads (red), to 2nd order CG beads (green), and to 3rd order CG beads (grey). Note that the hexagonal lattice symmetry is maintained in all orders of CG. (d)–(f) Schematics of CG parameterization strategies to determine bond force constant via equi-biaxial stretching (d), bond angle constant via equi-biaxial bending (e), and van der Waals constants via out-of-plane separation (f), respectively.

bead, represents 16 carbon atoms within an equilateral triangle in the atomic graphene structure (Fig. 1(a)). As a result, the 1st order CG beads can fill up the two-dimensional space by being packed into the same hexagonal lattice as in atomic graphene, but with a larger bead-bead bond length (0.568 nm vs. 0.142 nm of carbon-carbon atomic bond length). One 2nd order CG bead represents four 1st order CG beads within an equilateral triangle (Fig. 1(b)), so that the 2nd order CG beads can fill up the two-dimensional space in a hexagonal lattice with an even larger bead-bead bond length (1.136 nm). Such a CG procedure can then be scaled up recursively (i.e., Fig. 1(c)) to any higher order, while the hexagonal lattice symmetry is maintained in the CG structure of any order. As a result, the CG scheme of N th order allows for a reduction of computation size by 4^{N+1} folds. For example, one 3rd order CG bead represents 256 carbon atoms (Fig. 1(c)), which can significantly reduce the computational expense. The mass of the CG bead and the bead-bead bond length of any CG order can be deduced recursively. The bonded energy terms of the CG scheme consist of a two-body bond energy and three-body angle energy as $U_{bonding}(r_{ij}, \theta_{ijk}) = \sum \frac{1}{2} K_b (r_{ij} - r_0)^2 + \sum \frac{1}{2} K_\theta (\cos \theta_{ijk} - \cos \theta_0)^2$, where r_{ij} is the distance between the i th and j th bonded CG beads with r_0 being its corresponding equilibrium distance; θ_{ijk} is the angle between the i - j bond and j - k bond with $\theta_0 = 120^\circ$ being its corresponding equilibrium angle; K_b and K_θ are bond force constant and angle force constant, respectively. The non-bonded term includes the van der Waals (vdW) interaction between CG beads. This bottom-up scalable CG scheme allows straightforward yet rigorous determination of the bonded energy parameters without any fitting, as to be detailed as follows. Table I lists the values of CG parameterization used in this study. All CG simulations are implemented using modeling code LAMMPS.³⁷

The bond force constant is determined recursively by equating the potential energies of a square atomic graphene and its CG counterparts of any order under equi-biaxial stretching (no change in the three-body angle energy) (Fig. 1(d)). Denote K_{bi} , ρ_i , and r_{0i} as the bond force constant, bond density (per unit area), and equilibrium bond length for the i th order CG, respectively; A_0 and A as the area of graphene before and after the equi-biaxial stretching, respectively. The strain in each bond (both atomic bond and CG bond) is therefore determined as $e = \sqrt{A/A_0} - 1$. The total

bond force energy after deformation for the i th order CG is determined as $E_{bi} = A_0 \rho_i \times \frac{1}{2} K_{bi} (e r_{0i})^2 = \frac{1}{\sqrt{3}} K_{bi} A_0 (\sqrt{\frac{A}{A_0}} - 1)^2$ (note that for a hexagonal lattice of the i th order CG, $\rho_i r_{0i}^2 = 2/\sqrt{3}$). By equating the total bond force energy for different CG orders, e.g., $E_{b(i+1)} = E_{bi}$, we have $K_{b(i+1)} = K_{bi}$. In other words, the bond force constants of CG levels of all orders are the same, another intrinsic advantage of our scalable CG scheme.

The angle force constant is determined by equating the potential energies of an atomic graphene and its CG counterpart under equi-biaxial bending (i.e., by conforming to a rigid spherical surface so that the change in the bond force energy is highly suppressed, Fig. 1(e)). Consider a unit cell of graphene in the shape of an equilateral triangle that is coarse-grained into the i th order and contains 4^k CG beads (k is independent of CG order i). Therefore, such a unit cell denotes an atomic graphene with 4^{k+i+1} carbon atoms (e.g., inset of Fig. 1(e) shows the case of $i=1$, $k=2$). Given the self-similarity in lattice structures of atomic and CG graphene, the change of the bond angle between two neighboring carbon-carbon bonds in atomic graphene is the same as that between two neighboring CG bonds in CG graphene, when conforming to the same spherical surface. Denote $K_{\theta A}$ and $K_{\theta i}$ as the angle force constant of atomic graphene and CG graphene of i th order, respectively, and the bond angle after deformation as θ . The total angle force energy in the atomic graphene is $E_{aA} = 4^{k+i+1} \times 3 \times \frac{1}{2} K_{\theta A} (\cos \theta - \cos \theta_0)^2$, while that in the CG graphene is $E_{ai} = 4^k \times 3 \times \frac{1}{2} K_{\theta i} (\cos \theta - \cos \theta_0)^2$. By $E_{aA} = E_{ai}$, we get $K_{\theta i} = 4^{i+1} K_{\theta A}$, correlating the angle force constant in CG of i th order with the atomic angle force constant of graphene.^{38–40}

TABLE I. CG parameters used in simulations.

	1st order CG	2nd order CG	3rd order CG
r_0	0.568 nm	1.136 nm	2.272 nm
K_b	47.46 eV·Å ⁻² (same for all CG orders)		
K_θ	93.23 eV	372.91 eV	1491.66 eV
σ	0.34 nm (same for all CG orders)		
ϵ	0.00666 eV	0.00483 eV	0.00337 eV

The vdW interaction between the CG beads is fitted in a Lennard-Jones pair potential as $E_{vdW} = 4\epsilon[(\frac{\sigma}{r})^{12} - (\frac{\sigma}{r})^6]$, while the CG beads for a given order are treated as triangle particles.³⁷ For all orders of CG, σ is set to be 0.34 nm. ϵ is fitted to represent the energy needed to separate two pieces of triangular atomic graphene that a given order CG bead represents (Fig. 1(f)), from equilibrium distance (0.34 nm) to infinity.

Figure 2(a) depicts the CG simulation model for GNM in experiments with hexagon-shaped holes patterned in a hexagonal lattice in a planar monolayer graphene.^{25–29,32,33} The geometry of the GNM is defined by two parameters: the spacing between two neighboring hexagonal holes (termed as GNM lattice constant and denoted by L_C) and the graphene nanoribbon width (W_R). The global size of the CG model measures $W_G \times L_G$. The choice of a proper order of CG scheme is global-size dependent in the sense that, on one hand, the CG graphene bead is small enough to capture the fine features of the deformation under loading, and on the other hand, large enough to reasonably reduce the computational cost. Hereinafter, 1st order CG is used in all simulations of GNM unless noted otherwise.

Figures 2(b)–2(g) show the deformed shape of a GNM under various applied elongation along the armchair direction of the graphene nanoribbon network (indicated by the red arrow in Fig. 2(b)). Here, $L_G = 351$ nm, $W_G = 199$ nm, $L_C = 50$ nm, and $W_R = 2$ nm, which corresponds to a patterned graphene with 353 552 carbon atoms. We implement displacement loading and the CG structure is equilibrated for every displacement increment. The CG simulation is done at 300 K by canonical ensemble and by Nose-Hoover thermostat. Figure 2(h) plots the resultant force in the GNM as a function of the applied elongation, which clearly depicts two stages of the GNM deformation. In the first stage, the GNM can readily elongate up to about 25% without appreciable resultant force. In other words, the GNM is extremely compliant with nearly zero stiffness. As shown in Figs. 2(b)–2(e), the elongation of the GNM in this stage is accommodated by deflecting and twisting the initially planar and tortuous graphene nanoribbon network out of its plane, while individual

graphene nanoribbons barely stretch. It has been shown that suitable patterning can significantly increase the stretchability of various materials under the similar deformation mechanism.^{41,42} In the second stage, the applied elongation further straightens and stretches the graphene nanoribbon network (Fig. 2(f)) until the final fracture of the GNM at about 50% elongation (Fig. 2(g)). The fracture of the GNM results from breaking of the CG bonds that are stretched beyond 22% tensile strain, which is taken as the average for the failure strains of pristine graphene in armchair (17%) and zigzag (27%) directions.^{43,44} The effective stiffness of the GNM in the second stage, defined as the average slope of the corresponding curve in Fig. 2(h) normalized by initial cross-section width (W_G), is shown to be 15.8 N/m, more than 22 times lower than that of the pristine graphene (340 N/m).³ In other words, the GNM is still highly compliant in the second deformation stage. Furthermore, the two-stage deformation behavior of GNM leads to a stretchability beyond 50%, much higher than tensile failure strain of pristine graphene.^{43,44}

The two-stage deformation behavior and the ultra-high compliance and stretchability of the GNM as revealed in Fig. 2 are shown to hold for GNMs with a wide range of pattern geometry. Figure 3 reports quantitative study on the effect of pattern geometry on GNM deformation behavior. For a given GNM lattice constant L_C , as the graphene nanoribbon width W_R decreases (Figs. 3(a)–3(c)), the stretchability of GNM increases, the range of elongation for GNM being extremely compliant increases, and the effective stiffness of GNM in the second deformation stage decreases (Fig. 3(d)), though all these changes are modest. For a given W_R , as L_C increases (Figs. 3(e)–3(g)), both the stretchability of GNM and the range of elongation for GNM being extremely compliant remain nearly unchanged, while the effective stiffness of GNM in the second deformation stage decreases modestly (Fig. 3(h)). For a given L_C/W_R ratio, as W_R decreases (Figs. 3(i)–3(k)), similar trend as shown in Figs. 3(e)–3(g) is found. These parametric studies show that a suitably patterned GNM can be made extremely compliant up to about 20% elongation and then remain highly compliant up to about

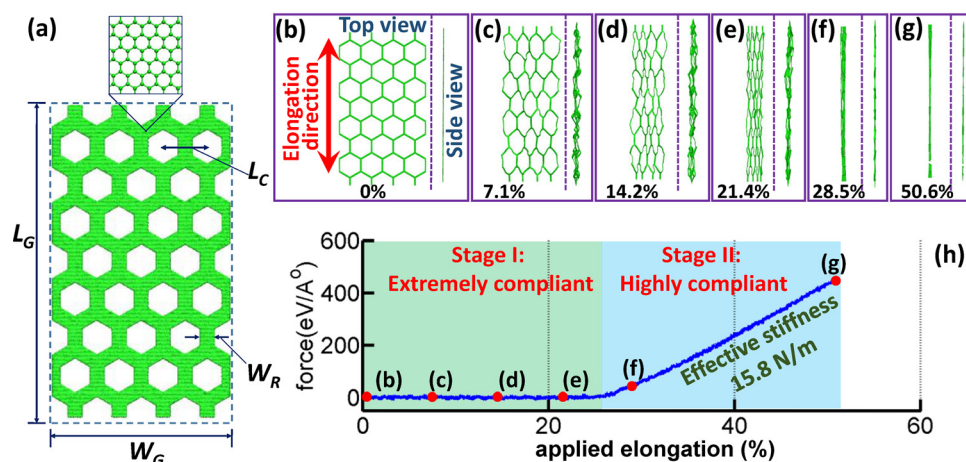


FIG. 2. (a) CG model for the patterned GNM. (b)–(g) Snapshots of the deformed GNM under various applied tensile strains. Red arrow indicates the direction of applied tensile load. (h) Resultant force in the GNM as a function of applied elongation. The light green shaded region highlights the extremely compliant region of the GNM while the light blue shaded region highlights the highly compliant region with an effective stiffness of about 15.8 N/m. Here, $L_G = 351$ nm, $W_G = 199$ nm, $L_C = 50$ nm, and $W_R = 2$ nm.

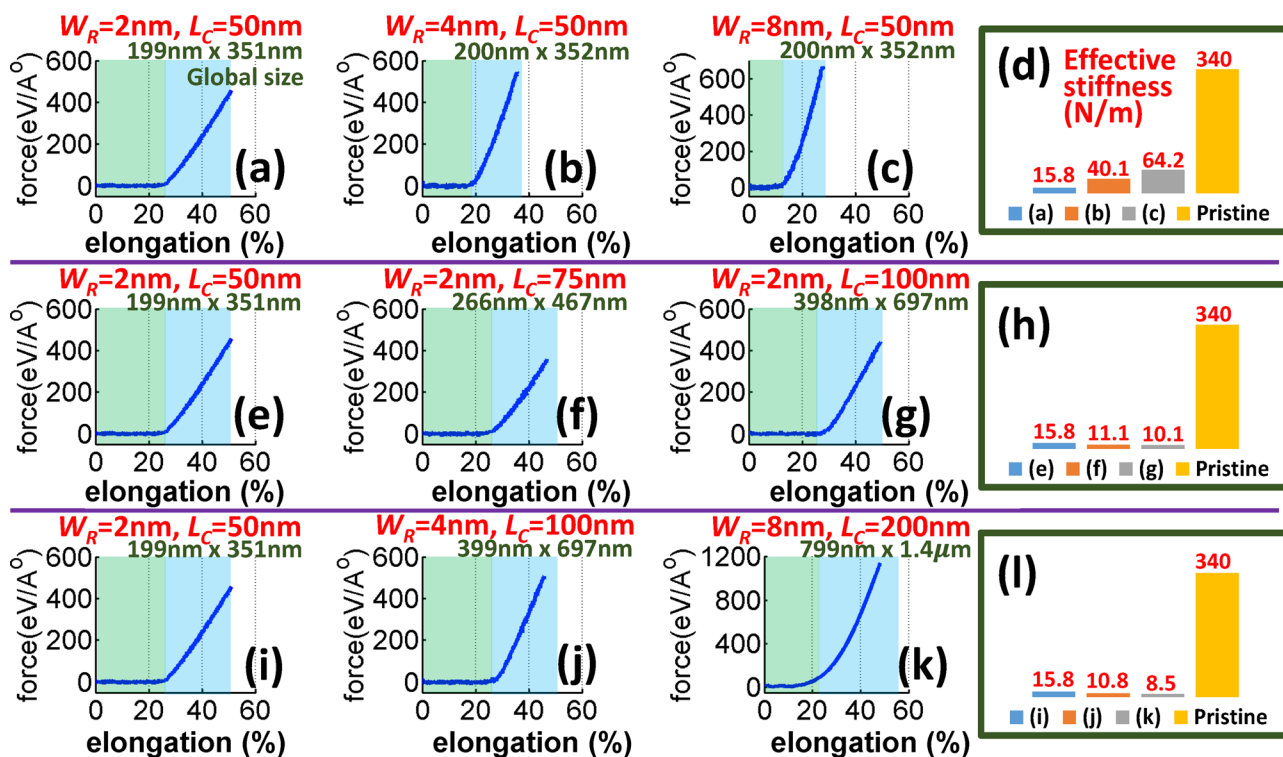


FIG. 3. Effect of pattern geometry of GNM on the deformation behavior and stretchability of GNM. (a)–(c): Resultant force in GNM as a function of applied elongation for varying W_R (2 nm, 4 nm, and 8 nm) with a fixed $L_C = 50$ nm; (e)–(g): varying L_C (50 nm, 75 nm, and 100 nm) with a fixed $W_R = 2$ nm; (i)–(k): W_R (2 nm, 4 nm, and 8 nm) with a fixed ratio $L_C/W_R = 25$. (d), (h), and (l) Compare the corresponding effective stiffness of the GNM in highly compliant stage (shaded in light blue) with that of pristine graphene. Total number of carbon atoms in the corresponding graphene network: (a) 353 552; (b) 671 536; (c) 981 040; (f) 479 408; (g) 815 408; (j) 1 041 584; (k) 4 158 272 (simulation done by CG of 2nd order given the GNM in micron size).

50% elongation (with an effective stiffness more than one order of magnitude lower than that of pristine graphene).

The significantly enhanced stretchability and ultra-high compliance of suitably patterned structures, such as GNM and other materials,^{6,41,42,45,46} find their origin in the deformation mechanism of bending and twisting, instead of pure stretching, to accommodate elongation. Such a deformation mechanism is essentially geometric, and thus independent of length scale. As suggested from the results of present study, GNM of size of microns (or even larger scale) can be made extremely compliant and highly stretchable under the same deformation mechanism. The advances in graphene patterning with nanoscale precision^{47–51} further allow applying

GNMs as structural components to enable high biaxial stretchability of the overall functional structure. For example, for biological applications,⁶ these GNMs can be attached onto the surface of biological tissues. The high stretchability and compliance of GNM can potentially accommodate the large deformation in biological tissues while keeping the electronic devices functioning. As a demonstration of the concept, Fig. 4(a) depicts a possible design with biaxial stretchability, in which biaxial tensile loads would be mainly accommodated by the elongation of GNMs along their arm-chair direction, while the elongation of the GNMs along other directions would be minimal.^{6,46} Nonetheless, to close the loop, we also study the tensile behavior of GNM under

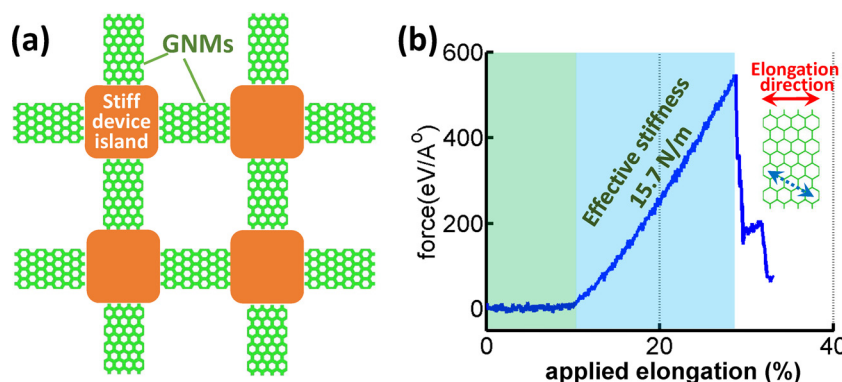


FIG. 4. (a) A possible design of a biaxially stretchable functional structure with stiff device islands (hosting device components) interconnected via GNMs. Applied tension can be largely accommodated by the extremely compliant GNMs elongating along their armchair directions, with minimal elongation along other directions. (b) Tensile behavior of the GNM in Fig. 2 under elongation along its zigzag direction, which shows the similar two deformation stages. Dashed arrow shows armchair direction, which has 30° rotation from zigzag direction.

elongation along its zigzag direction (Fig. 4(b)). It is shown that the GNM as in Fig. 2 demonstrates the same two deformation stages when elongates along zigzag direction: being extremely compliant with nearly zero stiffness up to about 10% elongation and then being highly compliant with an effective stiffness of 15.7 N/m up to $\sim 28\%$ elongation. Given the hexagonal lattice symmetry of the GNM, it is expected the tensile behaviors of the GNM along its armchair and zigzag directions (as shown in Fig. 4(b)) set the upper and lower limits of the GNM under elongation in any arbitrary in-plane direction. As the elongation of the GNM structural elements along non-armchair directions would be minimal, it is reasonable to predict that structural designs such as Fig. 4(a) can enable a percolating graphene network that remains extremely compliant and highly stretchable under large elongation.

In conclusion, we demonstrate a feasible solution that holds promise to address the challenge of interfacing intrinsically ultra-stiff graphene with highly compliant biological tissues and elastomers: by suitably patterning graphene, the resulting GNM can be made extremely compliant with nearly zero stiffness up to $\sim 20\%$ elongation and then remain highly compliant up to $\sim 50\%$ elongation (with an effective stiffness more than one order of magnitude lower than and a stretchability more than twice higher than that of pristine graphene). Such extremely compliant and highly stretchable GNMs can be used to enable biaxial stretchability of functional structures. The robustness of the deformation behaviors of GNMs against the width of the graphene nanoribbon building blocks (as shown in Fig. 3) could be further leveraged to explore other desirable electronic properties (e.g., tunable bandgap) of GNMs. We therefore call for further experimental studies to fully exploit these fertile opportunities.

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