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Nanostructures in suspended mono- and bilayer epitaxial graphene

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A B S T R A C T

Suspended graphene membrane presents a particular structure with fundamental interests and applications in nanomechanics, thermal transport and optoelectronics. Till now, the commonly used geometries are still quite simple and limited to the microscale. We propose here to overcome this problem by making nanostructures in suspended epitaxial bilayer graphene on a large scale and with a large variety of geometries. We also demonstrate a new hybrid thin film of SiC-graphene with an impressive robustness. Since the mechanics and thermal dissipation of a suspended graphene membrane are strongly related to its own geometry, we have in addition focused on thermal transport and strain engineering experiments. Micro-Raman spectroscopy mapping was successfully performed for various geometries with intrinsic properties measurements at the nanoscale. Our engineering of graphene geometry has permitted to reduce the thermal transport, release and modulate the strain in our structures.

1. Introduction

The first realization of suspended graphene was done in 2007 [1,2], just two years after the first graphene Hall bar samples. It was in fact an incredible feat, and is still today representative of the quality that can be achieved: small monolayer membranes of few microns length which are suspended and resonate with good nanomechanical properties. Until now, the proposed geometries remain quite simple without specific structure in most of the cases. However, nanostructuring of suspended atomically thin material has emerged recently and found various applications: real-time DNA detection, proton, molecular or water filtering, nanoscale kirigami using graphene to obtain stretchable transistors with up to 240% of elongation, a matter-wave beam splitter for molecules trough atomically thin material [3–6]. This can be achieved by lithography and graphene etching, by placing the 2D material on prepatterned micropillar arrays [7,8], or by mechanical buckling of the membrane [9]. There are important motivations to develop nanofabrication techniques of 2D materials. For example, nanostructured graphene with high porosity will permit to combine the thermal transport engineering and the highest thermal conductive material [10]. Nanostructuring can be a straightforward path to control strain gradient at the nanoscale. It is well known to induce strong band structure variation in 2D materials with possibilities to engineer novel systems in electronics and optics [11].

Currently, there is a bottleneck in the design complexity due to low sample numbers, statistical problems, and/or small graphene monodomains. Here we propose an approach allowing fast and simple design of suspended graphene including 2D nanostructuring patterns based on e-beam lithography and epitaxial graphene. Epitaxial graphene offers large scale monodomains graphene and high yield. We have explored the limits of nanostructuring: a quasi-freestanding graphene maintained only by a thin graphene arm. Periodic patterns of nano-opening in graphene or high aspect ratio bars have also been obtained. We show specifically, in cantilever configuration (Fig. 2), that we are able to release the native stress existing in epitaxial graphene by 3 orders of magnitude. In order to surpass the limit of structure collapsing in thin film materials, we have created ultrathin film hybrid structures of suspended graphene-SiC (<10 nm thickness) which remain stable, even for very peculiar patterning like zig-zag spring anchoring.

In this paper, we implement not only nanostructures in large graphene membrane but also we modify the intrinsic properties of the membrane like strain and thermal conductivity. We have seen that the physical description of our system is also reduced to

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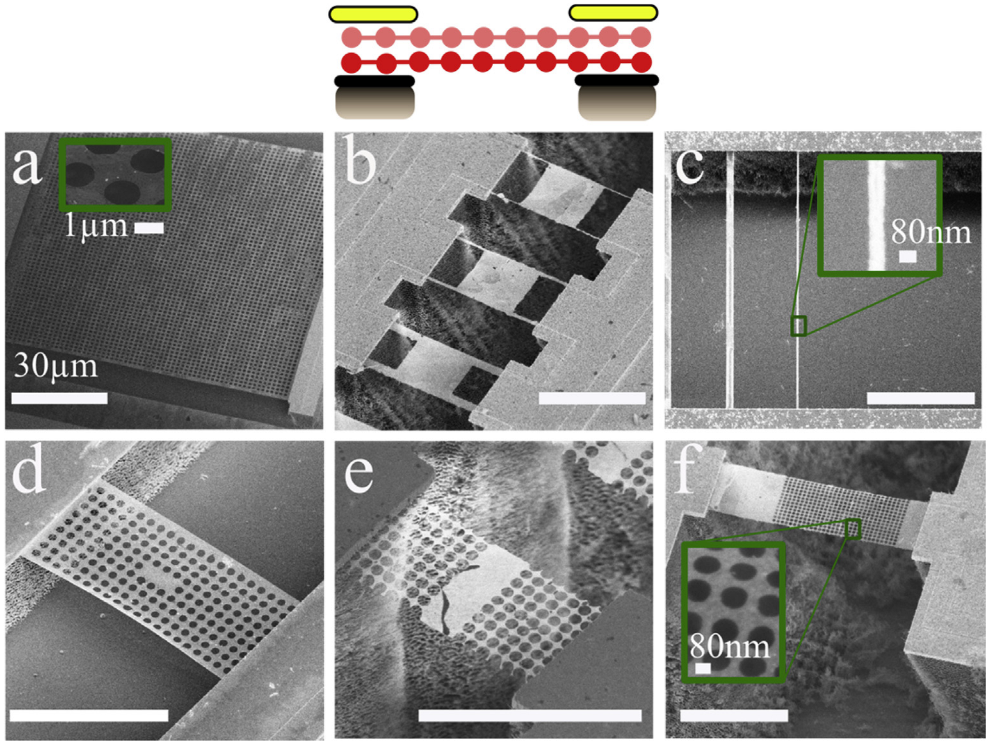

Fig. 1. Graphene nanostructuring: At the top, a schematic of a suspended graphene bilayer clamped at both side with gold and SiC. a-f) SEM images of suspended graphene structures with different geometries from 20 nm up to 200 μm (at low voltage, 1kVolt, in order to reduce the membrane damage and improve the contrast). **d** represents the possibility of a phononic cavity-like made in a graphene membrane under the Raman laser set-up. **c,d,e,f** are example of holes and thin bar made within clean graphene membrane. The porosity for the "holes area" of structure **e** is around 75%. **a** shows membrane with 100 μm side length and 1 μm hole patterns. All scales bar are 5 μm unless specified. (A colour version of this figure can be viewed online.)

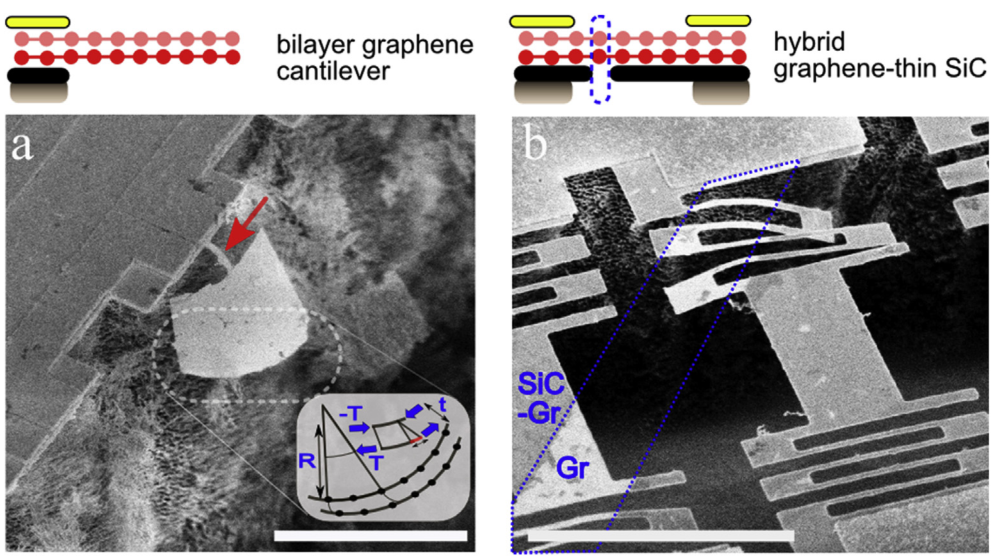

Fig. 2. Mechanical stability with extreme nanostructuring. a) An example of a released membrane at the limit of free-end cantilever shape structure for few layer graphene maintains by a tiny arm (black arrow). In inset a schematic of a curve graphene multilayer, represents the central plate curvature. We show the external layer extension (red portion) due to curvature R and internal shear layer stress T. **b)** Hybrid structures of very thin SiC-graphene can be realized when the etching of SiC is stopped before the complete release of graphene. These membranes are very robust against nanostructuring as seen in the images with a zig zag spring for anchoring. For example the white region (inside the blue dash line) is graphene only and does not fully sustain the nanostructuring. Scales bar are 5 μm. (A colour version of this figure can be viewed online.)

nanoscale considerations. We have realized a series of experiments with μ-Raman spectroscopy in order to extract thermal transport, strain and doping properties, and also correlate them at the nanoscale. Raman spectroscopy is largely used for nanomaterial, like graphene, because it is a versatile tool for strain, thermal transport, doping [12,13]. This technique introduces the Stokes or Anti Stokes processes including phonons, photons and electron-holes pair diffusion processes. The well-known G, 2D and D phonons resonances, characteristic of carbon-carbon sp^2 binding, the valley degeneracy of K–K′ Dirac cones and defects have been