1.3.10 Strain engineering in two-dimensional materials

Two-dimensional (2D) materials constitute a large family of one-atom-thick crystals offering a broad range of transport and optoelectronic properties. A key peculiarity of 2D materials is that the electron system is readily accessible at the surface of the crystal and can be easily tuned by the proximity of different materials as well as by other perturbations including, in particular, local mechanical deformations that can have a profound influence on the electron-system behavior.

The control of mechanical deformations in 2D materials, such as graphene, transition metal dichalcogenides, or even their Van der Waals heterostructures, opens exciting perspectives for the engineering of the electronic states and of the optoelectronic response. These include the possibility to create/tune the local bandgap, the creation of gauge field and strain-induced quantization and in general the modulation of the band dispersion and electron properties. While many of these concepts have been ideally validated in artificial-lattice studies, their actual implementation using true atomic lattices remains today largely elusive. In the past few years, we have developed a set of methods to controllably induce and measure strain in 2D crystals using different actuation strategies.

1. Measurement of local strain. The precise quantification of strain is an obvious key prerequisite to strain engineering, but it is also very important in view of the application of 2D crystals in the context of nanomechanical devices, such as nanoresonators (see Fig.1a). The current fabrication procedures are indeed typically based on delicate transfer schemes and thus prone to the creation of uncontrolled strain profiles in suspended membranes, which can have a detrimental/unknown effect on the final device behavior. The most common method to map strain in graphene and other 2D materials is micro Raman spectroscopy, whose resolution in the far field is limited by diffraction (typical laser excitation has a wavelength of 532nm). Near-field techniques can go beyond this limit and we have developed a method to precisely quantify the local mechanical properties by atomic force microscopy, thanks to a new procedure to deconvolve the complex combination of effects that determine the mechanical response of the membrane [4].



Figure 1. (a) Atomic force microscope (AFM) topography of a suspended graphene membrane. (b) Strain profile caused by fabrication, obtained based on AFM force spectroscopy. (c) Strain profile of the same membrane, obtained using conventional micro Raman. From [4].

2. Deterministic creation of custom strain profiles. A well-known method to strain a 2D material consists in creating a circular membrane and subject it to a

differential pressure, creating an approximately homogeneous strain profile. A first method we explored to induce custom strain profiles on graphene thus focused on the study of pressurized membranes with different shapes, since the shape of the membrane boundary has a direct effect on the induced strain profile. In particular, we have demonstrated how the application of a differential pressure to an elliptical membrane can be exploited to induce an anisotropic strain, depending on the orientation of the major and minor axes of the ellipse [6]. This simple approach has a limited flexibility with respect to the possibility to create true local strain actuators. For this reason, we demonstrated a novel method based polymeric actuators that can be nanopatterned with a great freedom by electron-beam nanolithography [2], see Fig.2a-c. In particular, strong local anisotropic strain could be demonstrated, as visible in Fig.2d and 2e, and the deterministic creation of nanowrinkles was reported in the suspended membrane. The method provides an ideal platform to induce custom strain profiles, which would be very challenging to obtain using more conventional techniques reported in the literature, and it can be easily combined with a variety of imaging and spectroscopic techniques including in particular scanning electron microscopy, atomic force microscopy and micro Raman and micro photoluminescence [3, 4]. All these techniques are in fact crucial to measure and understand the mechanical configuration of the material.



Figure 2. (a) Sketch of a suspended graphene membrane structure, including polymeric layers suitable for the creation of local strain actuators. (b) Optical image of one of the studied devices: the dark circular region corresponds to the suspended part of the membrane; shaded rectangles are the polymeric actuators. (c) Raman G-peak at position A and B of the membrane, see (b): no strain is detected in B while a strongly anisotropic strain is detected in the pulled region A. (d) Polarization-dependent measurements in position A highlights the oscillatory behavior of the amplitudes of the G+ and G- peaks; the effect can be used to directly determine the orientation of the crystal axes in the membrane. From [2].

3. Multilayer systems. A unique option of 2D materials is that they can be relatively easily stacked on different substrates [1] or in VdW heterostructures [4]. VdW systems are particularly promising in the context of the control of mechanical deformations, since the friction between the stacked layers can be very low. In this case, in our activities we studied the deformation of WS₂ on top

of graphene, using its impact on the position of the local photoluminescence (PL) response of the dichalcogenide monolayer as a probe of the local induced strain, see Fig. 3. The same approach can be applied to the many possible combinations of 2D materials that are currently studied within the scientific community and we expect it to be relevant for the creation of deterministic wrinkle structures and to control the relative mechanical configuration of the stacked layers, which is known to have a strong influence on the electronic states.



Figure 3. (a) Scanning electron micrograph of one of the WS2-graphene heterostructures obtained by chemical vapor deposition of WS2 on top of bilayer graphene obtained by thermal decomposition of SiC. (b) Typical device structure, including two polymeric actuators (micrometric artificial muscles, MAMs) pulling the WS2 in the horizontal direction; in overlay, averaged shift of the photoluminescence (PL) position. (c) Comparison between the PL of the pristine WS2 crystal and after the strain actuation. From [3].

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