

Enhanced Photoresponse in Curled Graphene Ribbons

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1. Device fabrication and SEM imaging

We patterned 5- μm -wide trenches on 170- μm -thick transparent fused silica substrates using photolithography via a GCA Autosteper. 5- μm -deep trenches were etched via an Oxford 80 RIE. Source and drain electrodes were patterned in the same way and deposited with 5 nm of Ti and 40 nm of Pt via an e-beam evaporator. Single layer graphene was grown on copper foils at 950 °C in the presence of 35 sccm of methane and 8 sccm of hydrogen via chemical vapor deposition. Then a standard wet transfer process was applied and a poly-methyl methacrylate (PMMA) layer was spin-coated on top of the copper foil to hold the graphene film. The PMMA-graphene film was separated from the copper foil through the wet etching in iron chloride solution, and then transferred onto the pre-patterned chip after cleaning in multiple DI water baths.

Thermal annealing process has been applied to eliminate the PMMA layer and to make CGRs. The sample was heated in the furnace for 30 min with the presence of 200 sccm argon under 420 °C at one atmosphere, which led to a complete evaporation of the PMMA^{1, 2}. During the evaporation, the graphene that adhered to the substrate would be hold still due to the van der Waals interaction, while the suspended part was more affected and formed the curled structures.

2. MD Simulations of CGRs

Classical MD simulations were performed using the LAMMPS package³. A bond-order potential (LCBOP)⁴ was used to model the atomic interactions and included intrinsic long-range order interaction between the layers. The simulations in Figure 1b were performed using microcanonical (NVE) ensemble on a 1- μm -long and 0.1- μm -wide graphene ribbon containing 3,840,000 C atoms. During the system equilibration before dynamic deformation the simulation box dimensions parallel to the graphene surface were fixed to correspond to the equilibrium lattice constant of unstrained graphene at T=300K. The structural evolution during temperature annealing was simulated to mimic desorption of PMMA molecules. Groups of atoms were randomly selected in the graphene nanoribbon and given

random momenta at the initial time (Fig. S2 *top*). To accelerate the structural deformation, the momentum value was chosen to be equivalent to heating up to ~ 2500 K, while the total momentum remained zero. Average temperature remained close to 400 °C, similar to the experimental value. Boundary conditions for the larger dimension kept the ends flat with a restoring elastic force $F = kx$, according to the experimental conditions. Other dimensions had no restraints. After 75 ps, graphene nanoribbon formed wrinkles and developed a multilayer-like structure in the middle (Fig. S2 *middle*). Within 150 ps after initial momenta were given, the ribbon curled and locally formed multi-layers, resulting in a quasi-one-dimensional structure in the middle (Fig. S2 *bottom*). Several sets of atomic groups and momenta, namely 4 different simulations, were picked to explore the nanoribbon structural evolution. While the tendencies in structural deformations during all the simulations remained the same, the time required for the formation of the curled region in the middle of the nanoribbon varied.

3. Supplemental Figures

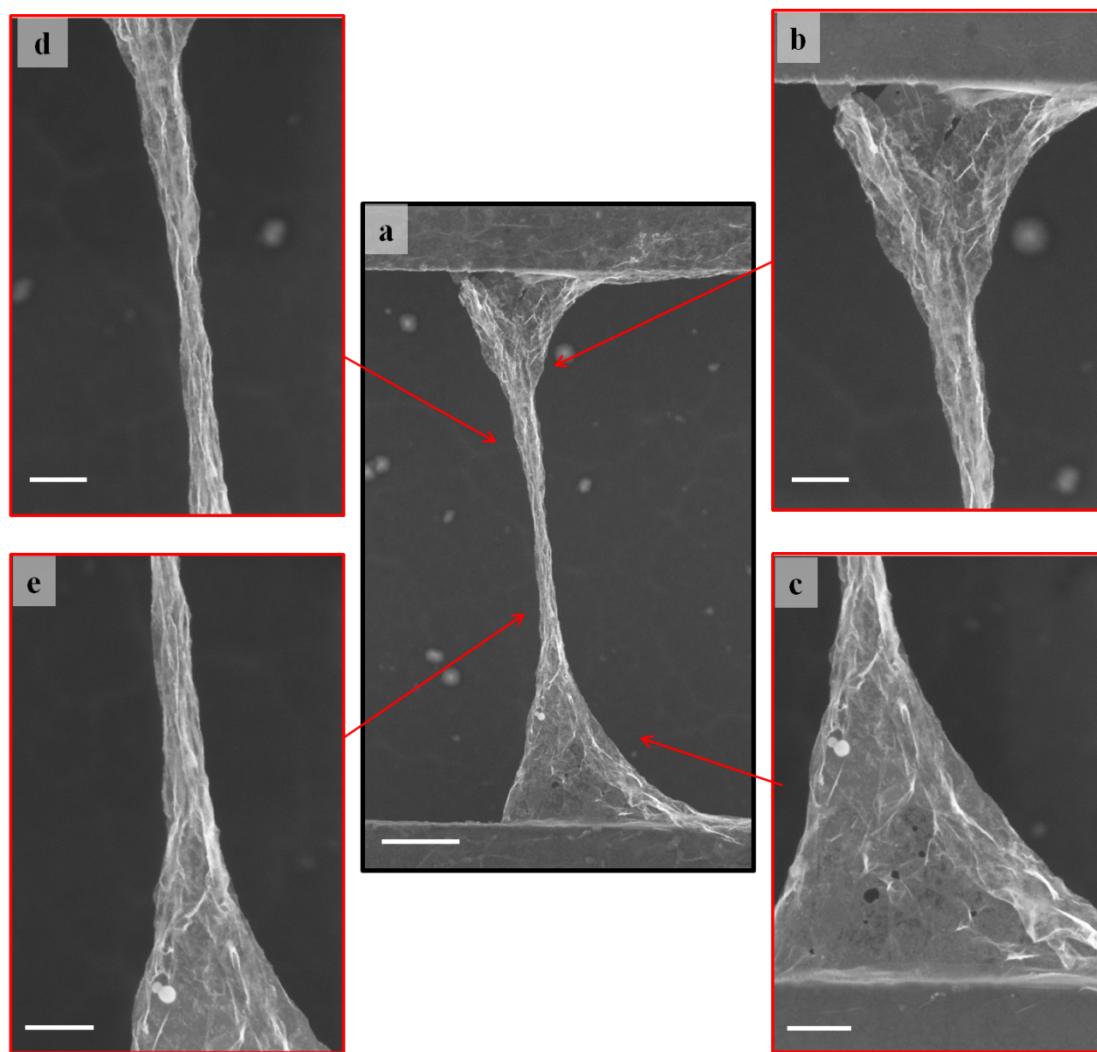


Figure S1. SEM images of a CGR. **a.** SEM image of the CGR device. The close-up images of different regions: **b.** top; **c.** bottom; **d.** top-to-mid; and **e.** mid-to-bottom. The scale bars are 1 μm (a) and 400 nm (b-e), respectively.

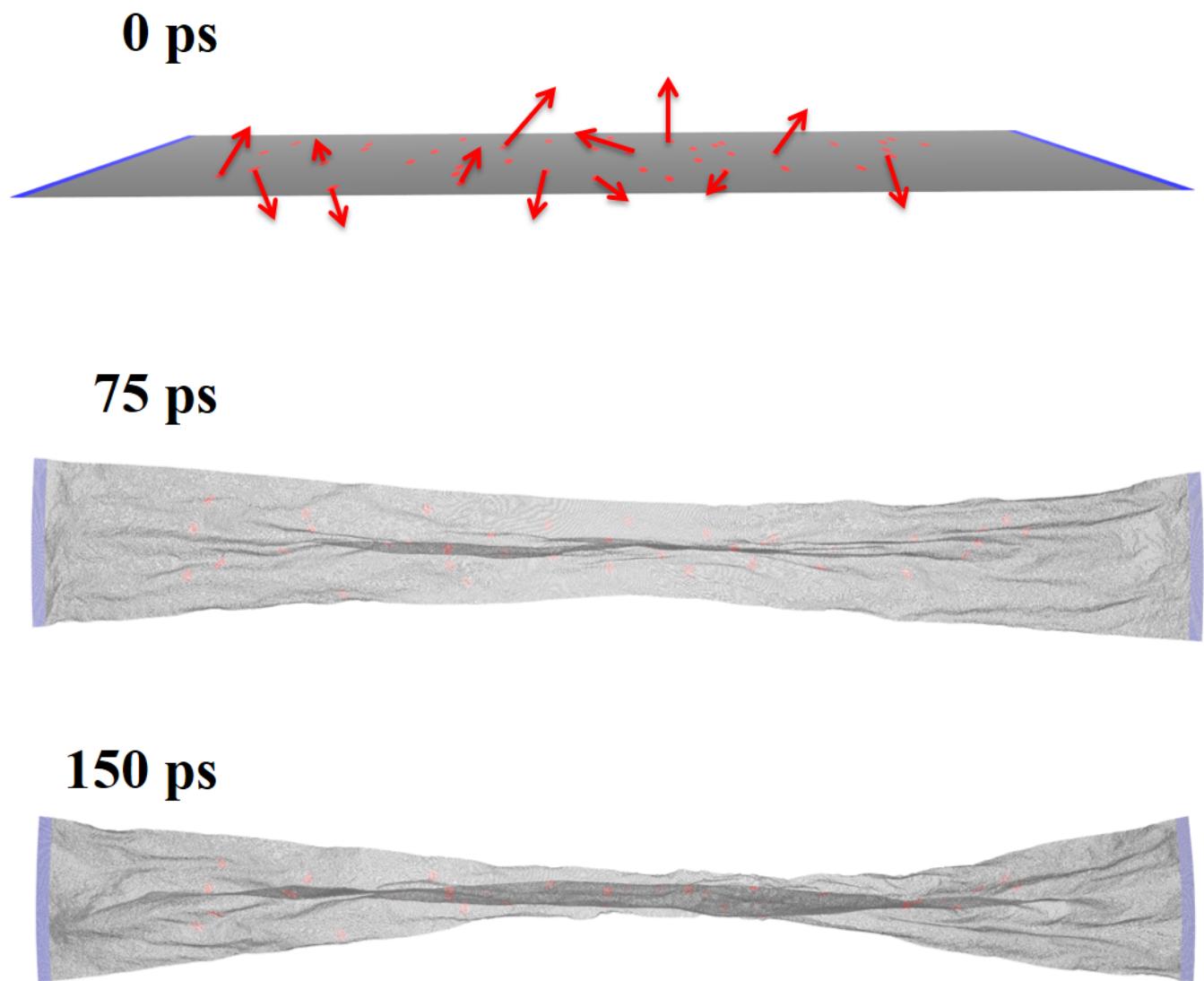


Figure S2. MD simulations of CGRs. MD simulation cell with a restoring elastic force $F = kx$ at each end and random momenta given to randomly selected regions along the flat ribbon (*top*) and the simulated CGR structure after different relaxation times: initial folding after 75 ps (*middle*) and curled structure forming after 150 ps (*bottom*).

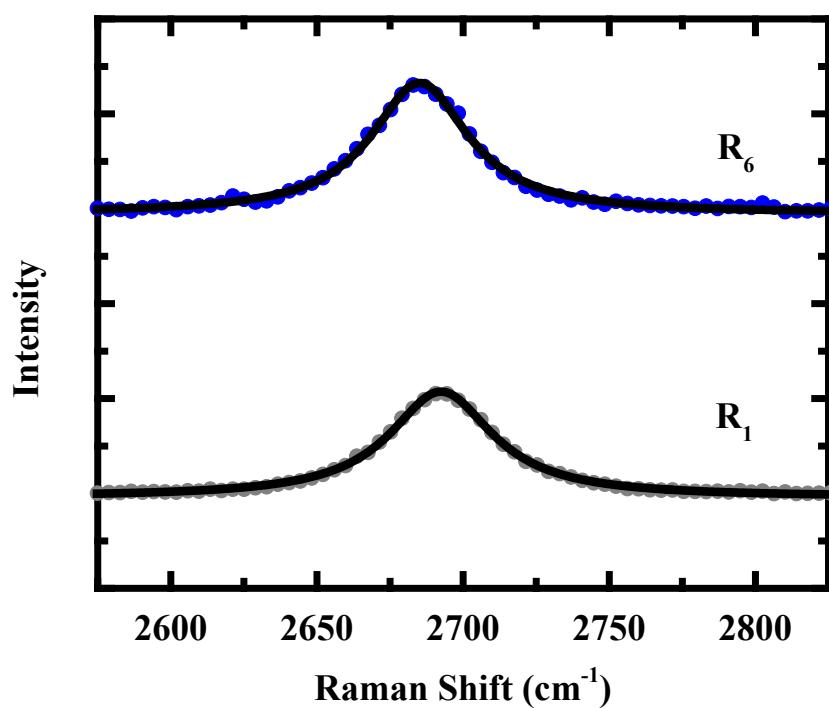


Figure S3. Raman spectra of a CGR. Raman spectra and their Lorentzian fit for regions R_1 and R_6 of the CGR in Figure 1.

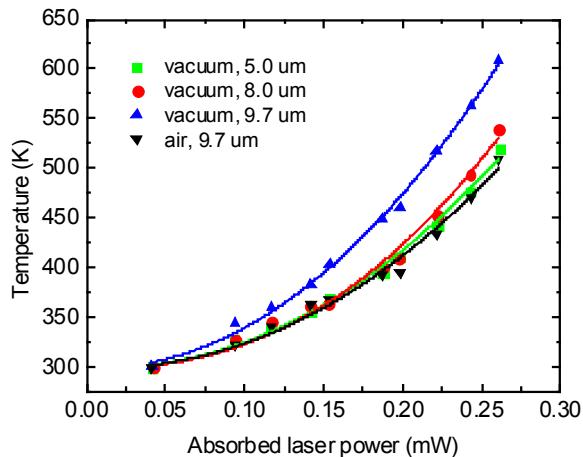


Figure S4. Temperature dependence on incident laser power of suspended graphene. Temperature is measured by Raman 2D peak shift versus absorbed laser power⁵. The temperature dependence is different for graphene of different dimensions or in different environments. The fittings (solid lines) reveal the temperature is proportional to P_L^β , where β is between 2.1 to 2.3. For suspended graphene of 9.7 μm diameter in the air (black line), $\beta = 2.1$. Here we assume room temperature is 298 K.

4. Supplemental References

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