# Supplementary materials for:

# Large scale arrays of single layer graphene resonators

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#### **Supplementary Materials**

The supplementary materials contain the following information:

- 1. A description of the graphene growth and transfer procedure.
- 2. A description of the suspended graphene fabrication procedures.
- 3. Raman measurements on CVD grown graphene

- 4. A discussion of tearing in graphene membranes
- 5. Electrical transport measurements of suspended CVD graphene membranes
- 6. A description of the AM and FM electrical mixing technique.

#### 1. Graphene Growth and Transfer:

CVD growth furnace setup:

The chemical vapor deposition is done in a 1 inch diameter low pressure, temperature controlled, gas flow furnace. Gasses include UHP Argon, Hydrogen, and Methane, with flows controlled by an automatic flow controller. Low pressure is achieved using an oil pump with a cold trap. The gas flow attachments are tightly clamped to prevent atmospheric contamination. Gas pressure should be < 10 mTorr with no gas flowing and  $\sim$  1 Torr with gas flowing.

## **Growth Procedure:**

Part 1: Preparing for growth

- 1. Use Alfa Aesar 0.025mm, 99.8% pure copper foils.
- Cut out 1.5 cm squares, notch the edge to indicate orientation, and press between glass slides to flatten.

Note: During the entire growth and transfer process, care must be taken to keep the copper foils as flat as possible. Crumpled foils lead to cracked graphene membranes and poor transfers.

- Treat the foil with the following order of solvent dips: Acetone (10 Sec), Water, Acetic Acid (10 Minutes), Water, Acetone (10 Sec), IPA (10 Sec)
- 4. Use low flow nitrogen gun to gently remove remaining IPA
- 5. Load 3-5 copper foils into CVD furnace
- 6. Pump system down to a base pressure under 10 millitorr

#### Part 3: Graphene Growth

- After base pressure is achieved, flow 6sccm of hydrogen. The pressure should rise to about 120 millitorr
- 2. Turn furnace on to reach 1000 C
- 3. Anneal foil in hydrogen at 1000 C for 10 minutes
- 4. After anneal, flow 157 sccm methane for 13 minutes. Pressure should rise to 5.5 torr
- 5. Let grow for 13 minutes
- 6. Cool slowly over 2 hours
- 7. Replace gas with 200 sccm argon for final 2 minutes, and let Argon re-pressurize the tube.

## **Graphene Transfer Procedure:**

The following description is similar to the transfer procedure developed by other groups [1-3].

- Use 8% Anisole-PMMA, spin PMMA onto one side of foil (keep track of which side) at 4000 RPM for 60 seconds. PMMA should be ~ 500 nm thick. Do not bake.
- 2. Etch graphene off of other side of copper foil using oxygen plasma.
- 3. Pour 1 M Ferric Chloride copper-etch solution.
- 4. Carefully place foil onto the surface of the copper-etch solution with PMMA side up. PMMA is hydrophilic so foil will float on acid surface
- 5. Let copper etch away completely  $\sim$  30 minutes.
- Scoop PMMA-graphene membrane into DI water. Membrane should float on the surface of water, with the PMMA side up, and the Graphene side down. Keep membrane flat to avoid cracking graphene.
- 7. Repeat 6 times into fresh DI water
- Scoop membrane out of liquid one more time with desired final substrate. The graphene should be in contact with your surface.

- 9. Let chip and membrane dry  $\sim 1$  day
- 10. Soak chip in Dichloromethane for  $\sim$  4 hours to remove PMMA.
- 11. Rinse with Acetone, then IPA
- 12. If suspended devices are desired, use a critical point dry to get chip out of solution.

## 2. Suspended Graphene Fabrication Procedure:

For Type A devices, we patterned 3 µm wide graphene ribbons on the copper foil using contact lithography and a 20 second oxygen plasma etch. We then cleaned the photoresist off the graphene by sonicating the foil in acetone for 1 minute, then soaked the foil for 10 minutes and sonicated again for 1 minute. Following the transfer procedure described above, we transferred the patterned graphene onto a PMMA membrane, then transferred the PMMA/graphene membrane onto the surface of a silicon wafer with 285 nm of oxide and a patterned array of trenches with length of 1–8 µm and depth of 285 nm. Finally we dissolved the PMMA in dichloromethane and critical point dried the chip to preserve the suspended structures.

For Type B devices, we transferred unpatterned CVD graphene on a 50 nm thick PMMA membrane onto a 200 nm thick suspended silicon nitride membrane patterned with square holes. After letting the PMMA/graphene membrane dry, we annealed the chip at 300 C in air for 2 hours. The PMMA gently bakes off the chip leaving the graphene freely suspended in a liquid-free process.

For Type C devices, we transferred un-patterned CVD graphene onto a degenerately doped silicon wafer coated with 285 nm of silicon oxide. We then patterned the deposited graphene into an array of rectangles using oxygen plasma, and cleaned the remaining photoresist of by soaking the sample in acetone for 4 hours, then annealing the sample in Argon/Hydrogen 0.8/0.2 SLM gas flow for 2 hours. We deposited 2 nm/150 nm thick Titanium/Gold electrodes on top of the patterned graphene, used

buffered hydrofluoric acid etch (BOE 6:1) to completely remove the oxide under the graphene [4, 5], and critical point dried the sample.

## 3. Sample Quality:

We verified the number of graphene layers and the sample quality using Raman Spectroscopy. Figure S1 shows the Raman Shift for graphene (a) on the copper foil directly after growth and (b) suspended between gold electrodes on a Type C device after all processing. We see an increase in the disorder of the graphene as a larger D peak after processing. The disorder is likely either due to resist contamination or at the edges of the membrane during the shaping step of the graphene.



**Figure S1**: Raman Spectra of CVD graphene a) as grown on copper foil and b) as a suspended membrane between gold electrodes.

# 4. Tearing:

Figure S2 shows the three primary modes of failure for suspended graphene membranes: (a) Partial tearing of the membrane, (b) complete tearing of the membrane, and (c) stick down on to the substrate.

Figure 2e shows that partial tears in the membrane (open circles) make very little difference to the mechanical properties of the resonators. Completely torn, and stuck down membranes are ignored in measurements.



**Figure S2**: SEM of the three primary modes of failure in Type A graphene membranes. a) Membrane with partial tears in the surface. b) Fully torn membrane. c) Stuck down membrane

# **5.** Suspended Graphene Transport Measurements:

Figure S3 shows the electrical resistance versus back gate voltage of the suspended graphene membrane shown in figure 1c. We use the equation  $\mu \sim A/C_{bg} \frac{dG}{dV_{bg}}$  to extract a lower bound on the graphene

mobility of 4000  $\text{cm}^2/\text{V}\cdot\text{s}$ .



**Figure S3**: a) DC Electrical resistance versus back-gate voltage for membrane shown in Figure 1c. b) Electrical mixing measurement setup.

## 6. Electrical Resonance Measurements:

The discussion of mixing presented here is meant to compare our mixing measurements to known techniques. For extensive derivations of the AM and FM mixing techniques for graphene and carbon nanotube resonators, see references [6-8].

As shown in Figure S3b, we apply a voltage  $V_{bg}$  to the back-gate, and a radio frequency voltage  $V_{RF}$ (t) to the drain. The gate capacitance  $C_{bg}$  causes the graphene membrane to be electrostatically attracted to the back-gate.

$$F_{bg} = \frac{1}{2} C_{bg}^{'} V_{bg}^{2} + C_{bg}^{'} V_{bg} V_{RF}(t)$$
(S1)

The static voltage tensions the graphene membrane and the RF voltage drives the sheet to resonate. By symmetry, the RF voltage can be applied either to the gate or to the drain with similar results.

To detect the motion of the resonator, we take advantage of the semi-metal properties of graphene, where the conductance of the graphene sheet  $G(V_{bg}, C_{bg})$  depends on both the applied voltage and gate capacitance. If the gate voltage changes, or the graphene moves, the conductance changes.

$$dG = \frac{\partial G}{\partial V_{bg}} dV_{bg} + \frac{\partial G}{\partial z} dz$$
(S2)

However, it is difficult to directly measure the changes in conductance due to motion at RF because the signal is small and there is a large parallel capacitance in the system. We employ two related mixing techniques to bring the signal down to low frequency. Instead of applying a pure RF signal at the drain, we apply either an amplitude-modulated signal or a frequency-modulated signal

$$V_{AM}(t) = \frac{V_{RF0}}{2} (1 + m \sin(2\pi f_{Mod} t)) \sin(2\pi f_{RF} t)$$
(S3)  
or

$$V_{FM}(t) = V_{RF0} \sin\left(2\pi \left(f_{RF} + f_{\Delta} \sin\left(2\pi f_{Mod} t\right)\right)t\right)$$
(S4)

where  $V_{RF0}$  is the drive amplitude of the resonator operating at radio frequency  $f_{RF}$ . The RF voltage is modulated at a frequency  $f_{Mod} = 1$  kHz. The amplitude of modulation is typically m = 1 for AM, and  $f_{\Delta} =$ 50 kHz for FM in our measurements. We measure the current through the graphene with a lock-in amplifier at  $f_{Mod}$ . The total current measured using AM or FM mixing is

$$I_{AM}(f_{Mod}) = \frac{1}{2} \frac{dG}{dq} \left( C_{bg} V_{RF0} + C_{bg} V_{bg} \operatorname{Re}(z^*(f_{RF})) \right) V_{RF0} \cos(2\pi f_{mod} t)$$
(S5)

or

$$I_{FM}(f_{Mod}) = \frac{1}{2} \frac{dG}{dq} C_{bg} V_{bg} V_{RF0} \frac{d\operatorname{Re}(z^*(f_{RF}))}{df_{RF}} f_{\Delta} \cos(2\pi f_{\mathrm{mod}} t)$$
(S6)

where dG/dq is the transconductance of the graphene,  $z^*(f_{RF})$  is the complex amplitude of motion, and  $\operatorname{Re}(z^*(f_{RF}))$  is the real component of the complex amplitude that is in phase with the drive force

$$\operatorname{Re}(z^*) = |z| \cos(\phi) \tag{S7}$$

There are two important observations to make about the mixing equations. First, the AM mixing current

has a background due to the pure electrical mixing in the graphene, while the FM mixing current does

not. Second, assuming a simple harmonic resonator response to drive, the AM mixing technique gives a

heartbeat shaped mixing response and the FM mixing technique gives a mode shape that is proportional

to the derivative of the AM mode shape  $d \operatorname{Re}(z^*(f_{RF}))/d f_{RF}$ . These are the mode shapes measured in

Figure 4b.

#### **References:**

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