**Supplementary Information for**

**Performance of Monolayer Graphene Nanomechanical Resonators with Electrical Readout**

Changyao Chen, Sami Rosenblatt, Kirill I. Bolotin, William Kalb, Ioannis Kymissis, Horst L. Stormer, Philip Kim, Tony F. Heinz, James Hone*

*e-mail: jh2228@columbia.edu

**A. Sample identification**

Graphene flakes are deposited by mechanical exfoliation and patterned by electron beam lithography. After electrode deposition and SiO₂ removal, samples are transferred and dried with a critical point dryer (model 13200J-AB from SPI-DRY) to avoid surface tension during the phase transition that causes suspended graphene to collapse. Samples are checked by atomic force microscopy (AFM) to verify suspension. Raman spectroscopy is then performed on the suspended region; the width of the 2D mode identifies the number of layers (Fig. S1). This mode shows no difference before and after graphene suspension.

**B. Electrical measurement system**

Samples are wire bonded and mounted onto a chip carrier. Coaxial cables with a characteristic impedance of 50 Ω are soldered directly to the chip carrier. Each input (drain and gate) is terminated with a matching load consisting of a 50Ω resistor in series with a 100 nF
capacitor to ground (Fig. S2a). This load allows for simultaneous DC and RF biasing (realized by means of a wideband bias tee, ZFBT-4R2GW+ from Minicircuits) while minimizing RF reflections. Each output (source) is terminated with a 100 nF capacitor to ground, which not only is used as a low-pass filter with corner frequency below 30 kHz, but also serves the purpose of anchoring the RF ground to eliminate ringing in the output; in the absence of the capacitor, the mixing current undergoes large fluctuations as a function of frequency, as the output cable becomes a quarter-wave transformer with variable impedance. The capacitance between the contact pads (120 μm × 120 μm, with 300 nm SiO₂ dielectric) and the underlying silicon gives a cutoff frequency of ~1 GHz. The frequency mixer used to generate the sidebands can be applied either to the drain or gate side with equal results. No sideband rejection is employed, effectively doubling the amount of current generated by one sideband at the output. Power control is obtained by a combination of fixed attenuators and a digital step attenuator (model ZX-76 from Minicircuits).

Due to the phase difference between the mechanical oscillation and the electrical signal, we usually see asymmetric mixed-down current lineshapes instead of the expected Lorentzian peak¹. We correct the phase of the electrical signal by simply changing the length of the coaxial transmission line (BNC cable) to either drain or gate, until a Lorentzian resonance lineshape is observed (Fig. S2b). In the 10-100 MHz regime, this correction technique is possible because a quarter wavelength is of the order of a few feet (at larger frequencies, commercial delay lines may be used).

C. Background mixing current and Charge Neutrality Point (CNP)
The mixing current has two contributions: a background current $I_{BG}$ which exists even in non-suspended samples and a resonant term $I_{peak}$ which is proportional to the vibration amplitude. The background mixed-down current is given by:

$$I_{BG} = \frac{dG}{dV_g} \delta V_{sd} \delta V_g$$  \hspace{1cm} (S1)

where $G$ is the conductance, $V_g$ is the DC gate voltage, and $\delta V_{sd}$ and $\delta V_g$ are the RF voltages applied to the drain and gate, respectively. Excellent agreement is observed between the mixed-down current and the expected value from the above equation at low frequencies (compared to the resonance but still significantly larger than the modulation frequency $\Delta f$), as seen in Figs. S2c and S2d. The measured mixing current at the CNP is zero as expected. However, we often find it located beyond +/- 10 V for as-fabricated devices, resulting in a nearly constant slope $(dG/dV_g)$. Joule heating typically brings the CNP to the vicinity of $V_g = 0$, and additional mass deposition drives it away, due to charge doping induced by the evaporated material.

**D. Vibration amplitude estimation and nonlinear drive**

In the harmonic regime, the vibration amplitude has a Lorentzian lineshape. Assuming phase correction has been applied, the mixed-down current will follow the vibration, and the maximum amplitude of vibration will be given by:

$$\delta z_{max} = \frac{1}{2} Z_0 I_{peak} / (V_g \delta V_{sd} \frac{dG}{dV_g})$$  \hspace{1cm} (S2)
where $I_{\text{peak}}$ is the current on resonance (excluding background) obtained from the Lorentzian fit, $\delta V_{sd}$ is the RF drain voltage, $V_g$ is the DC gate voltage, and $Z_0$ is the distance between the graphene plane and the substrate\(^1\). The factor of 1/2 comes from the use of two sidebands ($f + \Delta f$ and $f - \Delta f$) generated by the mixer in our setup.

If the sheet is driven beyond the harmonic regime, the vibration amplitude initially tops off and the current peak broadens. It then becomes hard to fit the peak with a simple Lorentzian lineshape and estimate the vibration amplitudes with precision, but it is clear that the quality factor worsens and that the oscillation stops increasing. Experimentally, we accomplish nonlinear studies by applying low power to the mixer input while applying large power to the other input. Given that commercial mixers may allow as much of ~ 20 dB at the main frequency, care must be taken when trying to measure the response to a large signal: if the amount of direct power appearing at the mixer exceeds that applied by the other input at the main frequency, then the mixer will drive both the oscillation and the electrical response. Because of the potential difference between drain and gate, large $\delta V_{sd}$ may also effectively drive the suspended graphene into resonance when $\delta V_g$ is small. With large $\delta V_{sd}$, we observe nonlinear lineshapes and hysteresis of the frequency sweep (Fig. S3).

E. Assignment of gold and graphene resonances

The 2D structure of graphene determines that its mechanical resonant frequency is highly sensitive to tension, which results in high tunability with external tension, as described by Eq. (1) in main text. In contrast, the resonant frequency of the suspended gold beam is not expected to
be changed by relatively small external tension due to the large bending rigidity, and is given by (doubly clamped beam)\(^2\):

\[
f_{\text{gold}} = 1.03 \frac{t}{L_{\text{gold}}^2} \sqrt{\frac{E}{\rho}}
\]

(S3)

where \(t\) is the thickness of the suspended metal beam (typical 80 nm Au on top of 1 nm Cr), \(L_{\text{gold}}\) is the length of the suspended section of the gold electrode (slightly larger than width of graphene because of the isotropic etching of SiO\(_2\)), and \(E\) and \(\rho\) are the Young’s modulus and density of bulk gold. Figure S4a shows the measured gold resonances as a function of \(t/L_{\text{gold}}^2\).

There is good agreement with Eq. (S3). We attribute the deviations from the theoretical prediction to the random built-in tension generated during the fabrication process. Figure S4b shows the measured resonant frequency of a series of 6 devices of different length fabricated from the same graphene flake, measured at high \(V_g\) to minimize the effect of uncontrolled built-in tension. The frequency varies roughly as \(1/L_{\text{graphene}}\) in agreement with Eq. (1). This scaling also shows that resonances in the GHz regime should be achievable for device lengths in the ~100 nm range.

**F. Continuum mechanics model**

Because the bending stiffness of the monolayer graphene is negligible\(^3\), the graphene resonator can be treated as a doubly clamped membrane. Furthermore, we simplify our analysis to a 1D string with length \(L\) (Fig. S5). The bonding between graphene and electrode provides good adhesion\(^4\), and both AFM and SEM images after measurement confirm that the graphene sheets do not collapse. The electrostatic force between the sheet and the back gate is given by
\[ F = \frac{1}{2} \frac{\partial C_g}{\partial z} V_g^2, \]
where \( C_g \) is capacitance between the gate and the graphene (we assume a parallel-plate capacitor with spacing of 300 nm\(^5\)) and \( \frac{\partial C_g}{\partial z} \) is the spatial derivative. If we approximate the shape of the graphene under the electrostatic force as a circular arc with radius \( r \), then the total tension in the string approximation can be treated as \( T = T_0 + T_e = F / r L \), where the tension \( T \) includes the initial tension \( T_0 \) and the external tension \( T_e \) induced by the electrostatic force (Fig. S5). The external tension \( T_e \) is a result of elastic elongation of the string\(^3\), and therefore \( T_e = wEt\Delta L / L \), where \( w \) and \( t \) are the width and thickness of the device, \( E \) is the Young’s modulus. When \( \Delta L \ll L \), we have \( T_e = wEtL^2 / 24r^2 \), and finally
\[ T_e(T_e + T_0)^2 = wEtF^2 / 24, \]
yielding
\[ T = T_0 + \sqrt{\frac{a + \frac{2}{27} T_0^3 + \sqrt{\frac{a^2 + \frac{4}{27} aT_0^3}{2}}}{2}} + \sqrt{\frac{a + \frac{2}{27} T_0^3 - \sqrt{\frac{a^2 + \frac{4}{27} aT_0^3}{2}}}{2}}, \]  
(S4)

where \( a = \frac{1}{48} wEt(\frac{\partial C_g}{\partial z})^2 V_g^4 \). Combined with equation (1) from the main text, we have an analytic expression to predict the resonant frequency of a graphene resonator under electrostatic force. Here, we corrected the offset of \( V_g \) due to the trapped charge. When \( T_e \gg T_0 \) (at large \( V_g \)), the extra tension varies as \( V_g^{4/3} \), so that the frequency varies as \( V_g^{2/3} \). Fits to the measured data using this model with mass density and initial strain as free parameters provide excellent agreement with the experimental data.
When there is resist residue (Polymethyl methacrylate, PMMA) or pentacene on top of the graphene, we can treat the combination as a multilayer stack made from uniform films. Its equivalent two-dimensional elastic stiffness is

\[
\overline{Et} = \sum_{m=1}^{n} \frac{E_m t_m}{t_m}
\]  

where \( \overline{E_m} \) and \( t_m \) are the elastic modulus and thickness of \( m^{th} \) layer, respectively. Since the Young’s modulus of graphene\(^7\) is three orders of magnitude larger than that of pentacene or PMMA (order of GPa), the equivalent elastic stiffness is dominated by the contribution coming from the graphene. The measured two-dimensional elastic stiffness of graphene (342 N/m) gives an effective Young’s modulus of 1.02 TPa when a thickness of 0.335 nm is used.

G. Thermal expansion coefficient of graphene resonator

When the operating temperature is lowered, we observe consistent increase of all resonant frequencies, as well as decrease of tunability over the same \( V_g \) range (Fig. 6a). The upshift of resonant frequency at small \( V_g \) implies an increase of built-in tension across the suspended graphene sheet, in turn minimizing the effect from external tension induced by \( V_g \). Degradation of the tunability is therefore expected. This increase of tension comes from the isotropic contraction of suspended metal contacts (Fig. S6a), and is reversible upon thermal cycling. Although the contraction of the metal beam is not uniform due to the clamping to the oxide, we can consider it as equivalent to uniform shrinkage along the graphene resonator direction within this small deformation range. Since the suspended graphene is only anchored to
metal contacts, which act like doubly clamped beams on the substrate, we are able to extrapolate the thermal expansion coefficient of the graphene resonator using the thermal expansion coefficient of gold (bulk)$^8$ and silicon$^9$.

### H. Mass sensitivity

The minimum resolvable mass is determined as:

$$
\delta m = \frac{\partial M}{\partial f} \delta f \times 10^{-DR/20} \approx -\frac{2M_{\text{eff}}}{Q} \times 10^{-DR/20}
$$

where $M = 7.4 \times 10^{-16} \text{g}$ is the mass of the graphene per $\mu\text{m}^2$, $Q \approx 14000$ is the quality factor, $DR = 40 \text{dB}$ is the dynamic range at $T = 5K$, thus this yields the result of $1 \text{ zg}/\mu\text{m}^2$.

The mass sensitivity $S_{m}^{1/2}$ is given by:

$$
S_{m}^{1/2} = \frac{\partial M_{\text{eff}}}{\partial f_0} \frac{\partial f}{\partial I} S_{n}^{1/2}
$$

where $M_{\text{eff}} = 1.5 \times 10^{-15} \text{g}$ is the mass of the resonator, and $(\frac{\partial I}{\partial f}) \approx (\frac{\partial I}{\partial f_0}) = 1.24 \times 10^{-16} \text{ A/Hz}$ is an approximation of current change when the device is swept through the resonance. From Fig 6b, $(\frac{\partial I}{\partial f})$ is the slope of the response function. $S_{n}^{1/2} = 4.12 \times 10^{-15} \text{ A/}\sqrt{\text{Hz}}$ is the current noise spectral density of the measurement. Overall, the resulting mass sensitivity is $7.6 \times 10^{-22} \text{ g}/\sqrt{\text{Hz}}$. 
Figure S1. **a**, Optical microscope image of suspended graphene. **b**, AFM image of the same sample in **a**. **c**, Raman spectrum of the same sample in **a**, showing the lineshape of 2D peak (green dots) with Lorentzian fit (red line). **d**, Height profile corresponding to the white line.
Figure S2. **a**, Electrical circuit setup with matching loads at each connection. **b**, Resonant peak with different lengths of transmission line (Pomona 2249-C, cable length added to drain side is 2.5 feet), showing different line shape due to phase change. Amplitudes (peak values):

\[ \delta V_g = 14 mV, \delta V_{sd} = 100 mV. \]

The calculated phase change at 67 MHz is 170°. **c**, Gate sweep with \( V_{sd} = 10 mV \), CNP is close to \( V_g = -3 V \). **d**, Mixing current measured from the same device shown in **c**, operated at 1 MHz, \( \delta V_{sd} = 5 mV, \delta V_g = 100 mV \). Black points show predicted values of mixing current according to equation (S1). All mixing currents are recorded in rms values.
Figure S3. Nonlinear behavior with large drive power, bistable oscillation is observed when frequency is swept up and down. Mixing current is recorded in rms values.
Figure S4. **a**, Observed gate-independent gold resonances, assigned to vibration of the gold electrodes, scale as \( t / L_{\text{gold}}^2 \), consistent with beam theory. The dotted line shows the expected value using \( \sqrt{E/\rho} = 2400 \text{ m/s} \) for gold. **b**, The maximum resonant frequency (at large \( V_g \)) scales inversely with length for devices made from same flake of graphene, consistent with a membrane model.
Figure S5. Continuum mechanics model for graphene resonator, simplified to 1D case. L is the length between the two metal clamps, $L + \Delta L$ is length of graphene under strain, F is the electrostatic force, T is the longitudinal tension, r is the radius of curvature.
Figure S6. a, Deformation of the suspended metal contacts due to temperature change.

\[ \Delta T = -100K \]

b, AFM image of the same device at room temperature, scale bar: 1 \( \mu \)m.

References