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## SUSPENDED GRAPHENE BEAMS WITH TUNABLE GAP FOR SQUEEZE-FILM PRESSURE SENSING

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### ABSTRACT

We present suspended graphene pressure sensors fabricated using an innovative surface micro-machining process. The great advantage of this process is that the molybdenum (Mo) catalyst layer for multi-layer graphene chemical vapor deposition (CVD) is also used as a sacrificial layer to suspend the graphene. This method allows for accurate control of the gap size under the beam by simply varying the catalyst thickness. Furthermore, the need for transfer of the graphene layer is eliminated. Using this method, wafer-scale graphene squeeze-film pressure sensors are fabricated and characterized.

### KEYWORDS

Graphene, suspended beam, resonator, squeeze-film effect, pressure sensor

### INTRODUCTION

The mechanical properties of graphene are extraordinary: its Young's modulus is the highest known (~1 TPa), and of all impermeable membranes it has the lowest mass density and bending rigidity [1, 2]. Combined with its nm-scale thickness, these properties make graphene a very attractive material for nanomechanical pressure sensors. Potentially, graphene-based pressure sensors can have a significantly higher responsivity and smaller size than their Si counterparts [3, 4].

Most conventional sensors measure pressure by detecting the deflection of a membrane that seals a cavity which is kept at a stable pressure. However, state-of-the-art graphene sealed cavities show leak time-constants in the order of hours [4], and currently it is uncertain if their hermeticity can ever be increased to provide a stable pressure for the entire sensor lifetime, which is on the order of years. To circumvent this problem, recently graphene-based squeeze film pressure sensors have been demonstrated, which rely on a change in resonance frequency due to the squeeze film effect [3].

The main challenge remains the fabrication of suspended graphene membranes with a controllable gap. Until now, graphene had to be manually transferred over a cavity, while preventing stiction during transfer. This required relatively deep cavities. Moreover, such a manual process is time-consuming, not suitable for mass-production, and results in a substantial yield loss [5].

Previously, we demonstrated a transfer-free graphene fabrication process, which was used to fabricate graphene chemistors on wafer-scale [6]. In this work, we extend this fabrication method to allow for the wafer-scale fabrication of suspended graphene beams with a

controllable nanometer gap-size. Using a laser interferometry setup, we show that the mechanical resonance frequency of the graphene beam is pressure dependent due to the squeeze-film effect.

### EXPERIMENTAL

In our previous work, it was found that Mo thin-films can be used as catalyst for the chemical vapor deposition (CVD) of graphene [7]. The advantage of Mo over traditional catalyst like Ni and Cu is that multi-layer growth is more uniform and that it has a significantly higher melting temperature. The latter prevents the deformation of the patterned catalyst, as growth is performed close to 1000 °C. We discovered that by pre-patterning the Mo before graphene CVD, followed by wet etching of the catalyst after CVD, graphene overgrowth along the edges of the catalyst would assure it anchors to the SiO<sub>2</sub> underneath the Mo and adheres to the SiO<sub>2</sub> at the exact same place upon drying [6].

We start with a pre-patterned layer of Mo (Fig. 1a). As it will be shown later, the thickness of the Mo layer will directly define the gap size between the graphene and the SiO<sub>2</sub>, which can therefore be tuned easily. Graphene is deposited at 980 °C using CH<sub>4</sub>/H<sub>2</sub>/Ar (25/40/960 sccm) at 25 mbar for 5 min in an AIXTRON Blackmagic (Fig. 1b).

Following the graphene deposition, the metal contacts are defined by coating the wafers with negative resist, followed by Cr/Au (10/100 nm) evaporation and lift-off (Fig. 1c). These contacts will act as an electrical contact and, at the same time, support the graphene beam. To release the structures, the Mo catalyst is used as a sacrificial layer. It is etched in H<sub>2</sub>O<sub>2</sub>, after which the sample is first rinsed in DI water and then in isopropanol. Using critical point drying (CPD) with CO<sub>2</sub> the samples are dried (Fig. 1d).

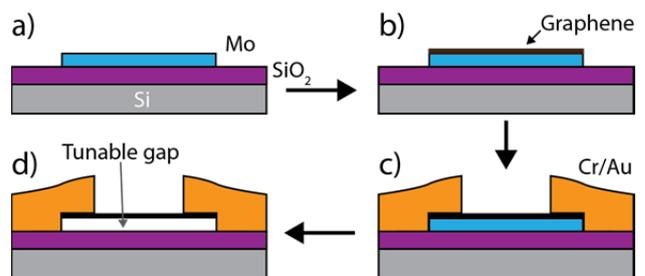


Figure 1: Fabrication process for suspended graphene beams: a) PVD deposition and dry etching of Mo catalyst with the desired gap thickness on 100 nm SiO<sub>2</sub>; b) CVD graphene growth on Mo; c) Cr/Au (10/100 nm) deposition and lift-off; d) wet etching of Mo in H<sub>2</sub>O<sub>2</sub>, and CPD for release.

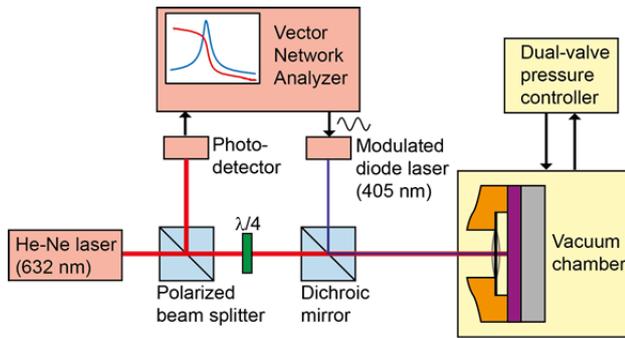


Figure 1: Optical setup used to detect the mechanical motion of the suspended graphene beam mounted in a pressure controlled chamber.

Samples are characterized using a Renishaw inVia Raman spectroscope with 514 nm laser, FEI Nova scanning electron microscope (SEM), NT-MDT nTegra Aura atomic-force microscope (AFM), and Keysight B2900A source-measurement unit (SMU).

To measure the mechanical motion of the suspended graphene beam, a laser interferometry setup as shown in Fig. 2 was used. For more details on the setup, the reader is referred to the work of Cartamil-Bueno et al. [8]. In short, a HeNe laser is used to detect the motion by interference between the graphene beam and the SiO<sub>2</sub> substrate, which are mounted in a pressure controlled chamber. The modulated diode laser provides optothermal actuation to the beam. A vector network analyzer is used to detect the motion of the beam.

## RESULTS AND DISCUSSION

A SEM image taken from a device with a 200 nm gap after release and CPD can be found in Fig. 3. The dimensions of the suspended beam are nominally 4 by 4 μm. A clear gap can be observed underneath the graphene beam. The graphene layer follows the roughness of the original Mo layer. Furthermore, small pinholes (black spots) can be observed. These pinholes can be removed by tuning of the CVD recipe

To demonstrate that the gap between the graphene and the SiO<sub>2</sub> can be modified using the Mo catalyst thickness, devices with 100 nm and 200 nm of Mo were fabricated and released. SEM close-ups of the gaps for the 100 and 200 nm samples are reported in Fig. 4a and 4b, respectively. As the SiO<sub>2</sub> layer is smooth, the exact location directly underneath the beam cannot be determined. The shadow due to electron shielding was used as a reference for the gap size. Due to this, the measured height of the gap will be different from the actual height.

In order to confirm that the gap size is indeed the thickness of the Mo catalyst, AFM was performed. In Fig. 5 a close-up of a suspended beam of graphene deposited on 200 nm thick Mo layer after release is shown. The average height of the graphene membrane was found to be  $180 \pm 20$  nm. For a membrane deposited on 100 nm Mo after release, the measured height was  $110 \pm 30$  nm.

The release of the graphene beam is performed using H<sub>2</sub>O<sub>2</sub> for etching and supercritical CO<sub>2</sub> for drying. To confirm that graphene is not damaged by these processes steps, Raman spectroscopy was performed on the

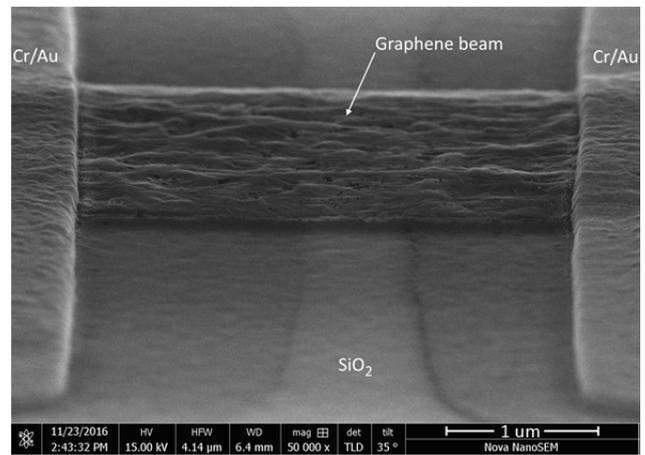


Figure 2: SEM image of the multi-layer graphene beam with Cr/Au contacts after 200 nm Mo etching and CPD. The roughness is imprinted in the graphene by the Mo catalyst during deposition. Sample tilt is 80°.

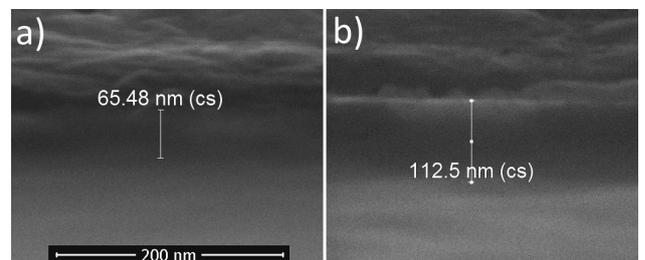


Figure 3: Close-up of the gaps for devices with 100 nm (a) and 200 nm Mo (b). Sample tilt is 80°. The measured height is the distance between the membrane and the shadow caused by electron shielding, the actual gapsize is likely close to that of the original Mo thickness.

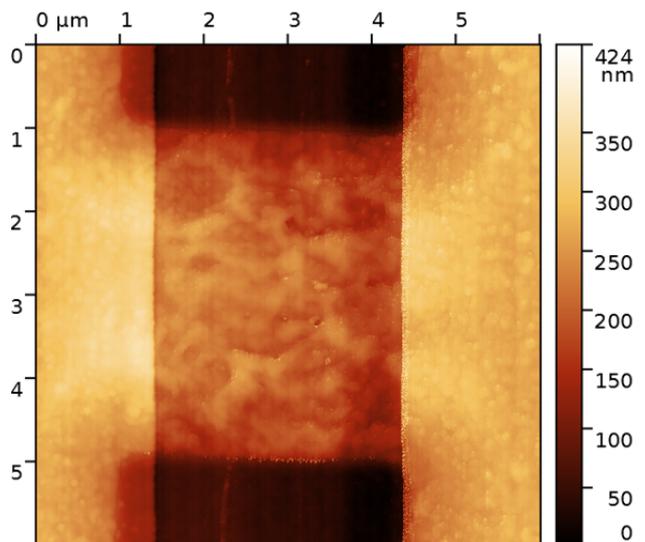


Figure 4: Atomic force microscope close-up of a suspended graphene beam with 200 nm gap. The average distance between the oxide (black) and graphene beam (orange) is  $180 \pm 20$  nm.

graphene beam before and after release. A typical Raman spectrum of graphene has a G-band at  $1580 \text{ cm}^{-1}$ , which is related to the sp<sup>2</sup> C-C bond, and a 2D band at  $2700 \text{ cm}^{-1}$ . The latter is related to the number of layers of the graphene layer; a narrow and intense peak (with respect to

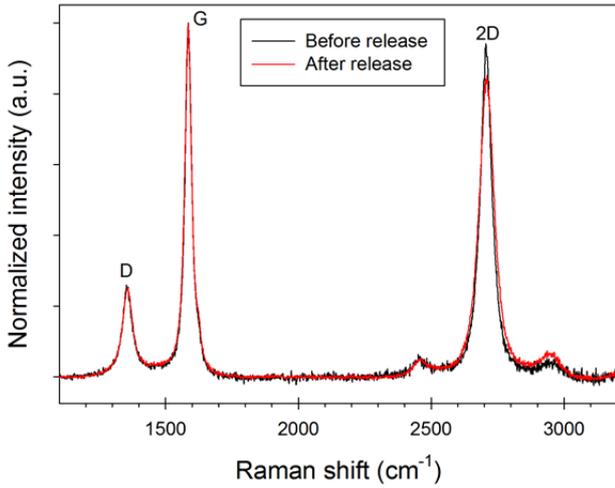


Figure 5: Raman data ( $\lambda = 514 \text{ nm}$ ) of the graphene layer before and after release using  $\text{H}_2\text{O}_2$  and CPD. The similarity in the ratios and widths of the peaks, especially the defect related D-peak, indicate that the graphene is not modified by the release.

the G-band) indicates a mono-layer [9], while for multi-layers the ratio becomes comparable or lower than the G-band, and the peak width increases. Finally, a defect related D-band can be present around  $1350 \text{ cm}^{-1}$ .

As can be observed from the Raman spectra (see Fig. 6), the graphene layer is multi-layer graphene. Previously, the thickness of the graphene was determined to be  $\sim 8 \text{ nm}$  using UV-Vis measurements [6]. The D-band originates from the CVD graphene deposition process and is not caused by the post-processing [6]. Finally, judging from the similarity of the spectra before and after release, it can be concluded that the release process does not affect the graphene quality significantly.

Two-point probe I-V measurements were performed on devices with a gap of  $100 \text{ nm}$  in order to determine the resistivity of the graphene beams and the yield. The devices for which the beam was absent, or where the resistance deviates significantly (i.e. 6 times the standard deviation) from the average, are considered defective. Fig. 7 displays the I-V characteristics of 15 measured devices with the same dimensions. The small non-linearity in the characteristics likely originates from the graphene-Cr/Au contacts.

A total of 60 devices were measured, of which only one was found to have an open connection. This results in a yield of 98 %. The average resistance of the working devices was found to be  $4.09 \pm 0.58 \text{ k}\Omega$ .

Using the setup depicted in Fig. 2, the mechanical frequency response of the suspended membrane with different gap sizes were determined under vacuum conditions. The results for two devices are shown in Fig. 8. Clear resonance peaks can be observed for both devices. For the 200-nm gap beam, a split-mode second order overtone can be observed.

Squeeze-film pressure sensors have an open venting channel underneath the membrane that is connected to the ambient. When the gas underneath the membrane is being compressed at a high enough frequency, the gas cannot escape because of the viscous forces. This compression will add stiffness to the

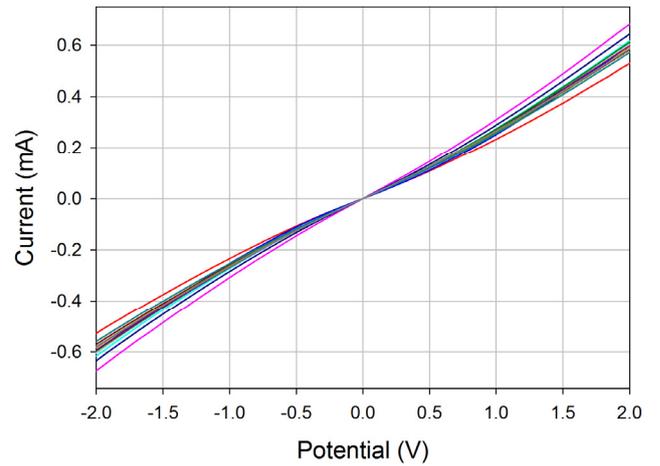


Figure 6: I-V curves of 15 suspended graphene beams measured by 2-point measurements;  $R_{\text{avg}} = 4.09 \pm 0.58 \text{ k}\Omega$ . For the total of 60 electrically measured devices with  $100 \text{ nm}$  gap, the yield was 98 %.

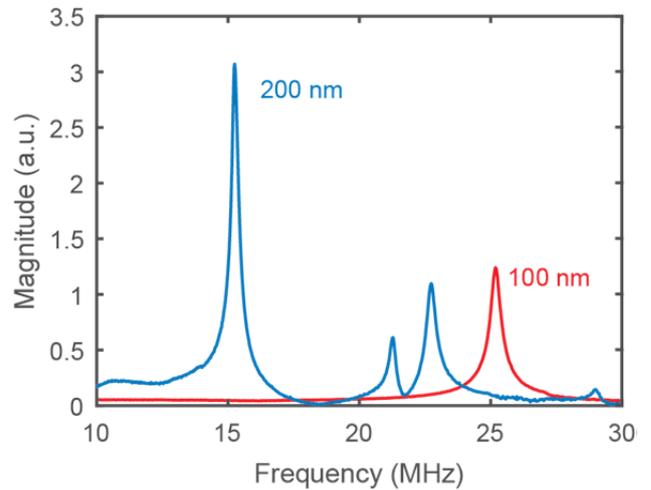


Figure 7: Mechanical frequency response function measured in vacuum for the  $100 \text{ nm}$  (red) and  $200 \text{ nm}$  (blue) gap device.

membrane, which will be a function of pressure. The observed resonant frequencies of our devices are sufficiently high to make sure the device operates in this regime [3]. The resonance will be a function of pressure according to:

$$f_{\text{res}}^2 = f_0^2 + \frac{p_{\text{amb}}}{4\pi^2 g_0 \rho h} \quad (1)$$

Here,  $f_{\text{res}}$  is the pressure dependent resonant frequency,  $f_0$  the resonance frequency in vacuum,  $p_{\text{amb}}$  the ambient pressure,  $g_0$  the gap height, and  $\rho h$  the mass per unit square [3]. As graphene has a low mass per unit square and our process allows for a small gap size, a high responsivity vs. pressure can be obtained.

Figs. 9 and 10 show the pressure dependency of the resonant frequencies of our devices. As expected from eq. (1) the resonance frequency increases with pressure, while the Q-factor decreases due to viscous forces. At low pressures (10-100 mbar) the response of the  $100\text{-nm}$  gap device is  $310 \text{ Hz/mbar}$ , while at high pressures it is  $165 \text{ Hz/mbar}$ .

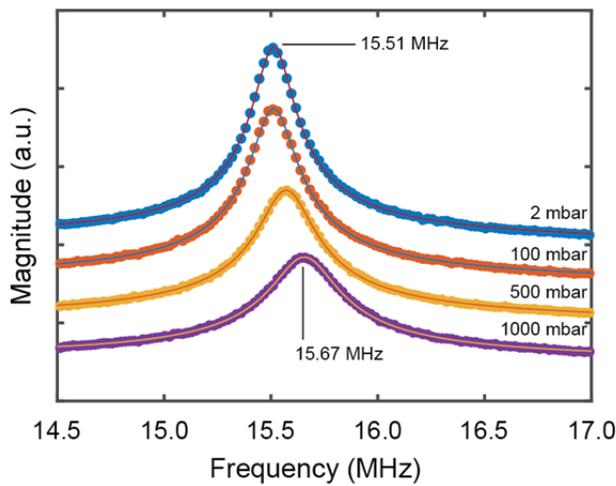


Figure 8: Measured frequency responses of a 200-nm gap device at different pressures. A harmonic oscillator model is fitted to extract their resonance frequency and  $Q$ -factor.

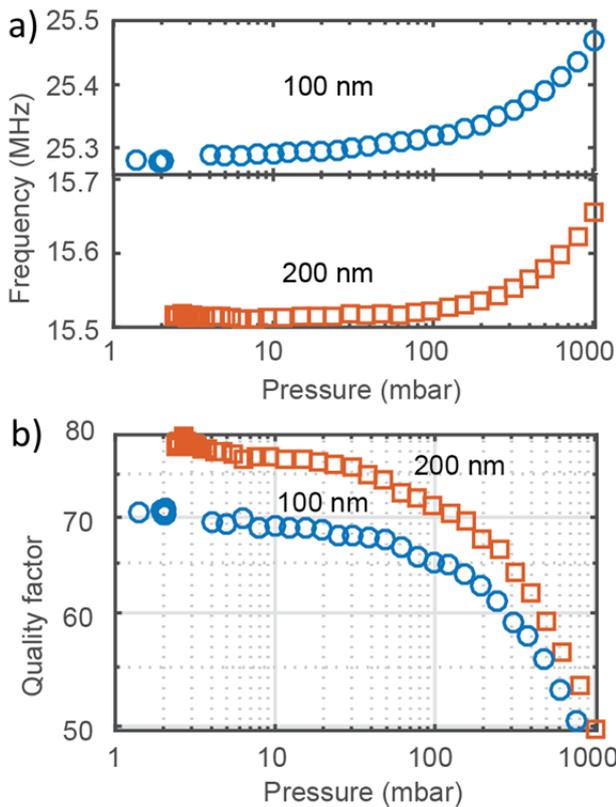


Figure 9: a) Resonance frequency of the fundamental mode of both devices as function of pressure. Due to the squeeze-film effect the resonance frequency increases. b)  $Q$ -factor of the resonance as function of pressure. The decrease is due to the viscous forces.

Compared to previous results reported in the literature obtained on manually transferred graphene [3], the shift in frequency was found to be lower (9 kHz/mbar at low pressure, and 1 kHz/mbar at high pressure), while our  $Q$ -factors at high pressure are 5 times higher. The results obtained are similar to that of the best performing MEMS-based squeeze-film pressure sensors, while the area of the suspended membrane is 25 times smaller for our device [10]. Our first simulations are indicating that

the reduction in responsivity compared to transferred exfoliated graphene could be related to the pinholes in the CVD graphene, as observed in Fig. 3. We recently optimized the graphene CVD conditions to reduce the number of pinholes significantly.

## CONCLUSIONS

We demonstrated a wafer-scale transfer-free process to realize graphene beams suspended on accurate nano-sized gaps. As the catalyst on which the graphene is deposited also acts as the sacrificial layer to release the graphene beam, the gap size can be easily adjusted, depending on the specific application. The process did not change the graphene layer quality. Using this process, suspended graphene beams with a high yield (98 %) were fabricated. It was shown that the resonance frequency is dependent on pressure due to the squeeze-film effect. The obtained response is similar to the best performing MEMS devices, while our device is 25 times smaller. This demonstrates the suitability of our method for the scalable fabrication of graphene-based sensing devices and resonators.

## ACKNOWLEDGEMENTS

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