In Situ Strain Tuning in hBN-Encapsulated Graphene Electronic Devices

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ABSTRACT: Using a simple setup to bend a flexible substrate, we demonstrate deterministic and reproducible in situ strain tuning of graphene electronic devices. Central to this method is the full hBN encapsulation of graphene, which preserves the exceptional quality of pristine graphene for transport experiments. In addition, the on-substrate approach allows one to exploit strain effects in the full range of possible sample geometries and at the same time guarantees that changes in the gate capacitance remain negligible during the deformation process. We use Raman spectroscopy to spatially map the strain magnitude in devices with two different geometries and demonstrate the possibility to engineer a strain gradient, which is relevant for accessing the valley degree of freedom with pseudomagnetic fields.

KEYWORDS: hBN-encapsulated graphene, strain engineering, vdW heterostructure, Raman spectroscopy, pseudomagnetic field

The large mechanical strength of two-dimensional (2D) crystals allows one to modify their optical and electronic properties by externally induced strain fields. Graphene, one of the key examples of 2D materials, is of particular interest because of its peculiar electronic properties. A series of intriguing effects were predicted for strained graphene, such as the appearance of a scalar potential, pseudomagnetic fields, valley filtering, or superconductivity. Different methods have been introduced to generate strain in graphene. One common approach is based on suspended graphene, where strain is induced by using different microactuators or by simply bending a flexible substrate. In other approaches, graphene is not suspended and strain can be generated by bending a flexible substrate, by using highly stressed metallic pads, or by placing graphene on periodic structures. However, several challenges that need to be overcome simultaneously hamper the progress of these platforms for studying strain effects in transport experiments. First, complex fabrication usually significantly degrades the graphene quality and hinders the observation of the strain effects. In addition, the device is often limited to very basic structures, without the possibility of local gating or multiterminal devices. Second, mechanical deformations often result in changes in the gate capacitance that cannot be easily distinguished from the actual strain effects. The third challenge is that the strain should be in situ tunable and nonhysteretic to disentangle strain effects from other effects.

Here we report a straining method that meets all of the above requirements. Instead of suspending the graphene, we encapsulate the graphene with hexagonal boron-nitride (hBN) and directly strain the van der Waals (vdW) heterostructure on-substrate. This approach preserves the exceptional quality of pristine graphene, which is a significant advantage for transport experiments. We use Raman spectroscopy to demonstrate that our method is versatile and allows one to engineer various strain fields, such as strain gradients, which are important for the generation of pseudomagnetic fields. We also show that the edge contacts work reliably and can sustain strain up to ~1%. In the first low-temperature electron transport measurements, we demonstrate that our method solves the problem of an artificial gating effect due to the bending-induced change in the graphene-to-gate distance, which occurs in suspended graphene devices. Therefore, our
approach of on-substrate encapsulated graphene offers an ideal platform for studying strain effects in transport experiments.

As shown schematically in Figure 1a, we fabricate hBN-encapsulated graphene devices with edge contacts and an electrical bottom gate on a polyimide-coated phosphor bronze substrate. We use exfoliated graphene from natural graphite and usually choose ~20 nm thick hBN for the top layer and ~30 nm thick hBN for the bottom layer. Bending the substrate in a three-point bending setup, as illustrated in Figure 1b, generates a strain field in the graphene. The deformation of the substrate is determined by the displacement $\Delta z$ of the pushing-wedge relative to the mounting position. The hBN encapsulation ensures the high quality of the graphene, and the metallic contacts and the gate allow us to perform transport experiments while tuning the strain in situ. The metallic contacts are essential for generating strain in a vdW heterostructure. In the Supporting Information Figure S3, we present a comparison between devices with and without contacts. The result shows that strain cannot be induced by substrate bending in devices without contacts. On the basis of the assumption that the graphene sheet is pulled uniaxially by the contacts during the bending of the substrate, we designed devices with two different geometries in order to obtain different strain fields. This is illustrated in Figure 1c, where the rectangle (device A) is expected to result in a homogeneous strain field, while the trapezoid (device B) should exhibit a strain gradient along the y-axis, that is, perpendicular to the straining axis. An optical micrograph of two fabricated devices is shown in Figure 1d.

To characterize the strain fields for different displacements $\Delta z$, we perform Raman spectroscopy directly on a three-point bending setup at room temperature. Previous studies reported characteristic redshifts in the Raman peaks of graphene, which we now use to quantify the local strain generated in these two devices. Typical Raman spectra of the hBN peak and

![Figure 1](image1.png)

**Figure 1.** (a) Schematic cross section of our device and (b) of the three-point bending setup. The bending is described by the displacement of the pushing-wedge, $\Delta z$. (c) Illustration of the mechanism for different strain fields. The solid lines represent devices without strain while the dashed lines stand for strained devices. The arrows indicate the elongation of the device along the contacts. The magnitude of strain is shown in grayscale with black corresponding to large strain. (d) Micrograph of two typical devices with different geometries.

![Figure 2](image2.png)

**Figure 2.** (a–c) Typical Raman spectra of an encapsulated device for two different $\Delta z$ values, zoomed in to the hBN peak, the graphene G peak and 2D peak, respectively. (d–j) Spatially resolved Raman maps of $\omega_{2D}$ for device A at different $\Delta z$ values. The white dashed lines outline the device boundaries and the colored dashed boxes mark the positions of the profiles shown in Figure 3. The gray arrows show the sequence of the measurements. (k) Spatially resolved Raman maps of $\Delta \omega_{2D}$ for device A, obtained by subtracting map (d) from map (g). (l–r) Spatially resolved Raman maps of $\omega_{2D}$ for device B at different $\Delta z$ values. (s) Spatially resolved Raman maps of $\Delta \omega_{2D}$ for device B, obtained by subtracting map (l) from map (o).
the graphene G and 2D peaks are shown in Figure 2a–c, respectively, for two different Δz values. The bending of the substrate results in a redshift of all peaks, with the most prominent effect on the graphene 2D peak. For small strain values, the 2D peak can be fitted by a single Lorentzian with center frequency \( \omega_{2D} \). In the following, we use spatially resolved Raman spectroscopy to map the strain field based on the redshift of the graphene 2D peak. The same analysis for the hBN Raman peak is presented in the Supporting Information Figure S5, which shows that both hBN and graphene are strained similarly in this method.

We first focus on the investigation of the rectangular device A. In Figure 2d–f, \( \omega_{2D} \) is plotted as a function of position for device A, for a series of increasing Δz from 0 to 0.6 mm (straining) and then decreasing back to 0 mm (relaxing), as indicated by the gray arrows. With increasing Δz, \( \omega_{2D} \) shifts to lower values at all positions on the map, consistent with increasing strain everywhere in the graphene sheet. When Δz is decreased back to 0 mm, \( \omega_{2D} \) reverts back to the initial values. The mean value \( \omega_{2D} \) averaged over the whole device is plotted as a function of Δz in Figure 3a, where Δz is first increased from 0 to 0.6 mm then decreased back to 0 mm. The very symmetric V-shape reveals a linear dependence and a good reproducibility of the strain tuning in the graphene device and exhibits no significant hysteresis. This process was repeated several times (up to 8 cycles) and no degradation was observed. The corresponding average strain values (\( \bar{e} \)) are shown on the right axis, which are calculated using \( \partial \omega_{2D} / \partial e = -54 \text{ cm}^{-1} / \% \) with \( \omega_{2D} \approx 2685 \pm 4.4 \text{ cm}^{-1} \) for unstrained graphene. We note that the former value is not known very accurate and our choice is among the intermediate reported values. The latter value is obtained as the average over 10 Raman spectra measured at different positions on 3 different hBN/graphene/hBN stacks before fabrication. The strain values at Δz = 0 mm are not 0, probably due to the intrinsic strain accumulated in the device after fabrication. We obtain an average strain of up to 0.23% and a maximum strain near the contacts of 0.3% for device A at Δz = 0.6 mm. Figure 3c shows the plot of \( \omega_{2D} \) versus \( \omega_{fi} \) (center frequency of the graphene G peak) of both devices for different Δz values. The data points fall on a line of slope 2.2, which confirms strain as the origin of the redshift of the Raman peaks.

We now turn to the investigation of strain and strain gradients in the trapezoidal device B. The spatially resolved maps of \( \omega_{2D} \) for device B are plotted in Figure 2l–r for the

Figure 3. (a,b) \( \omega_{2D} \) and the corresponding strain values plotted as a function of Δz for devices A and B, respectively. (c) \( \omega_{2D} \) versus \( \omega_{fi} \) at different Δz for both devices; the gray line has a slope of 2.2. (d,e) Profiles at the center of the maps indicated in Figure 2 for Δz = 0 mm (red), 0.2 mm (orange), 0.4 mm (green), and 0.6 mm (blue). Open circles are data, solid lines are linear fit. (f) Slopes of the profiles plotted as a function of Δz. The slopes are extracted from linear fitting in (d,e). The error bars are the fitting errors. The corresponding strain gradient is shown on the right axis.
same series of $\Delta z$ as above for sample A. Also, this device shows a tunable average strain controlled by $\Delta z$. The device averaged $\omega_{2D}$ and the corresponding extracted strain values are plotted in Figure 3b as a function of $\Delta z$. At identical $\Delta z$ values, the average strain for device B is larger than that for device A due to the smaller size of device B, but shows a similar V-shape, that is, a linear, nonhysteretic dependence on $\Delta z$. These findings can also be seen directly in the Raman maps. We obtain an average strain of up to 0.38% and maximum values at the lower sample edge of 0.52% for device B at $\Delta z = 0.6$ mm. The existence of a strain gradient is visible in Figure 2m-o. At the shorter (bottom) edge of the device, $\omega_{2D}$ shows a stronger shift than that at the longer (top) edge, which matches the predicted strain pattern for a trapezoidal geometry, as illustrated in Figure 1c (see also the FEM simulations in Supporting Information Figure S6). Figure 2s shows the difference between the Raman signals at large bending (Figure 2o) and no bending (Figure 2l).

To demonstrate the deterministic generation of a strain gradient in more detail, we plot $\omega_{2D}$ for both devices in Figure 3d,e as a function of the position along the y-axis in the center of the device area, averaged over 1 $\mu$m in the x-direction, as indicated by the colored dashed boxes in Figure 2d-g,l-o. For both devices, we find a clear increase in the average strain (overall shift of the curves) and an essentially unchanged background variation with increasing $\Delta z$. In addition, for the trapezoidal geometry (device B) we find a clear linear increase in the strain when moving from the longer to the shorter sample edge.

We now take the average slope of these curves as an estimate of the large scale (nonmicroscopic) strain gradient along the y-axis. For this purpose, we plot in Figure 3f the slopes of linear fits to the data in Figure 3d (device A) and Figure 3e (device B) as a function of $\Delta z$ with the right axis showing the corresponding extracted strain gradient. The small nonzero values on the order of 1%.

The existence of a strain gradient is visible in Figure 2m and a clear increase in the average strain (overall shift of the curves) and an essentially unchanged background variation with increasing $\Delta z$. In addition, for the trapezoidal geometry (device B) we find a clear linear increase in the strain when moving from the longer to the shorter sample edge.

Figure 4. (a,b) Illustration of substrate bending for suspended and on-substrate devices, respectively. The graphene-to-gate distance changes with substrate bending for suspended devices while it remains unchanged for on-substrate devices. (c) Two-terminal differential conductance, $G$, plotted as a function of gate voltage, $V_g$, for a suspended device for different $\Delta z$ values. The inset is the micrograph of the measured device. (d) The same data as in (c) with the curves rescaled in $V_g$ with respect to $V_g=0$ V by matching the CNP of each curve with that of the curve at $\Delta z = 0$ mm. The corresponding carrier density is shown on the top axis. (e) $G$ of an on-substrate device for a similar charge carrier density range for different $\Delta z$ values. The inset shows $G$ on a larger $V_g$ range. The micrograph is the measured device. The scale bars correspond to 2 $\mu$m.

conductance, $G$, is measured as a function of the gate voltage, $V_g$, for different $\Delta z$ values using standard low-frequency lock-in techniques (see Figure 4c,e).

There are significant differences between suspended and on-substrate devices for bending experiments. For the suspended device, one can immediately find a systematic change of the curves in gate voltage with increasing $\Delta z$ (see Figure 4c), whereas such an obvious effect is absent for the on-substrate device (see Figure 4e). The effects found in the suspended device can be fully accounted for by the change in the graphene-to-gate distance when bending the substrate, as depicted in Figure 4a. This is illustrated in Figure 4d, where we plot the data of Figure 4c rescaled linearly in gate voltage for each curve with $V_g=0$ V as a fixed point. This can be understood in a simple capacitor model in which the charge induced in the graphene is given by $Q=CV_g$ with $C$ as the
effective capacitance between the graphene layer and the gate. If the capacitance is changed by a factor $\alpha$ to $\alpha C$ due to the substrate bending, the same charge $Q$ is induced at $V_g/\alpha$, which is equivalent to a rescaling in the gate voltage. This scaling factor is extracted for each curve by matching the CNP to that of the curve at $\Delta z = 0$ mm and it is linear in $\Delta z$. After rescaling, all data points fall onto the same curve, see Figure 4d. This demonstrates that the bending-induced gate effect is dominant for the suspended graphene device, which makes it very difficult to study effects due to actual strain.

This effect is absent in the on-substrate devices optimized for strain tuning. For comparison, we performed the same type of measurements also on an on-substrate device with the results shown in Figure 4e. Because the gate voltage lever arm in this device is much larger than that for the suspended device due to the shorter graphene-to-gate distance, we apply smaller gate voltages to obtain a similar carrier density range as that in the data of the suspended device (see top axes of Figure 4d,e). A electron mobility of $\sim100 000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ is extracted from a linear fit around the CNP, suggesting a high graphene quality in our device. On this gate voltage scale and also on a much larger scale (see inset of Figure 4e), all curves with different $\Delta z$ values are virtually identical, which demonstrates that there are no bending-induced changes in the carrier density or in the contact resistance in this experimental configuration. The additional conductance minimum at $V_g \approx 1.3$ V comes from a double moiré superlattice effect in encapsulated graphene.\(^{32}\)

We point out that on this scale of graphene straining we could not detect significant changes in the conductance.

In conclusion, we have successfully generated tunable and reversible strain fields in encapsulated graphene devices. Because these devices are fabricated on-substrate, that is, not suspended, it allows us to design a large variety of device geometries. As an example, we use spatially resolved Raman imaging to demonstrate that the edge contact clamping and rectangular geometry result in a fairly homogeneous straining of the graphene. In a second step, we use this design freedom to generate a strain gradient in a trapezoidal geometry. In first transport experiments, we then demonstrate another major advantage of on-substrate encapsulated devices, namely, that the bending-induced gate capacitance change can be avoided, which is crucial for studying strain effects in transport experiments. This approach is not limited to graphene but also is suitable for studying strain effects in other 2D materials and complex vdW heterostructures, for example, in MoS\(_2^{33–35}\).

Because our method is simple and intuitive, nonetheless allowing complex device structures, we expect that it will pave the way toward deterministic strain engineering and new approaches to valleytronics.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.9b01491.

Details of the fabrication and the measurement setups, comparison between devices with and without contacts, discussions of the correlation between $\omega_{2D}$ and $\omega_{C}$ analysis of the hBN Raman maps, FEM simulations, contact failure, and discussions of the suspended encapsulated device (PDF)

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**Author Contributions**

L.W. fabricated the devices, performed the measurements, and did the data analysis. S.Z. initiated the idea of hBN-encapsulation. A.B. did the FEM simulations. P.M and J.O. helped to develop the straining method. P.M., S.Z. and J.O. supported the device fabrication. A.B., P.M., and S.Z. helped with the data analysis. K.W. and T.T. provided high-quality hBN. C.S. initiated and supervised the project. L.W. and A.B. wrote the paper. All authors discussed the results and worked on the manuscript.

**Notes**

The authors declare no competing financial interest.

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