

Monolayer MoS₂ Strained to 1.3% With a Microelectromechanical System

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Abstract—We report on a modified transfer technique for atomically thin materials integrated into microelectromechanical systems (MEMS) for studying strain physics and creating strain-based devices. Our method tolerates the non-planar structures and fragility of MEMS while still providing precise positioning and crack-free transfer of flakes. Furthermore, our method used the transfer polymer to anchor the 2D crystal to the MEMS, which reduces the fabrication time and increases the yield, and allowed us to exploit the strong mechanical coupling between the 2D crystal and polymer to strain the atomically thin system. We successfully strained single atomic layers of molybdenum disulfide (MoS₂) with MEMS devices for the first time and achieved greater than 1.3% strain, marking a major milestone for incorporating 2D materials with MEMS. We used the established strain response of MoS₂ Raman and photoluminescence spectra to deduce the strain in our crystals and provide a consistency check. We found good comparison between our experiment and the literature. [2018-0144]

Index Terms—MEMS, monolayer MoS₂, strain, Raman, photoluminescence.

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This paper has supplementary downloadable material available at <http://ieeexplore.ieee.org>, provided by the authors. This includes discussion of strain achieved in prior work combining 2D materials with MEMS, details regarding the repair of device M26, displacement versus power characterization of our devices, derivation of substrate dependent effective Poisson's ratio, results assuming different Poisson's ratio values, and FEA simulation of strain inhomogeneity.

Color versions of one or more of the figures in this paper are available online at <http://ieeexplore.ieee.org>.

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I. INTRODUCTION

TWO dimensional (2D) materials can withstand an order of magnitude more strain than their bulk counterparts, which results in dramatic changes to electrical [1], thermal [2] and optical properties [3], [4]. Ideally, we would be able to precisely control the strain field in these systems to study in detail the effect of strain and to create new strain-based devices. However, current techniques offer limited control over the strain field, and require bulky pressure chambers [3], [5] or large beam bending equipment [6], [7] incompatible with most applications. Here we demonstrate that MEMS can be used to dynamically strain atomically thin materials, which provides a method for straining 2D materials that can be incorporated in technologically relevant devices.

Previous experiments have used MEMS to strain nanomaterials such as nanotubes [8], [9], trilayer graphene [10], and monolayer graphene [11], [12]. Similar to the nanotube and trilayer graphene experiments, we adopt thermally isolated chevron actuators. A colorized SEM image of one of our devices is shown in Fig. 1. Our devices are fabricated using MEMSCAP's PolyMUMPS [13] process which has three polysilicon layers. The first layer we use for grounding, and the second (2 μm thick) and third (1.5 μm thick) are combined to make rigid, double thick structures (3.5 μm thick). While 2D materials are atomically thin, it still requires significant force to strain them, as they are very stiff. Graphene, for example, is the stiffest material ever measured [14]. For this reason chevron actuators are ideal for straining our 2D crystals because they are capable of large pull forces [15]. The actuator in Fig. 1 is located on the right side. These actuators create motion through the mechanical amplification of thermal expansion caused by Joule heating of the beams (3 μm wide). To minimize heating the sample stage with the actuator, we thermally isolate the actuator with long, thin thermal relief tethers (2.5 μm wide) that have a large thermal impedance. Further, we place many soft heat sink springs (3 μm wide) in parallel near the sample stage to create a low thermal impedance between the sample stage and the MEMS die. This geometry creates effectively a thermal resistor divider circuit dramatically, cutting down on the heat that reaches our atomically thin samples [16]. The samples are placed on the stage on the left side of Fig. 1, shown in the inset. The stage has an anchor side that is secured to the MEMS die on the left, and a shuttle side that is connected to the actuator to

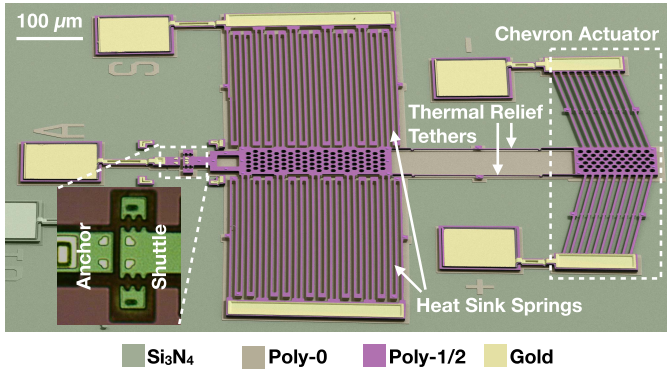


Fig. 1. Colorized SEM image of a typical MEMS device used for straining 2D materials. The atomically thin crystal is placed on the left side of the MEMS across a $3 \mu\text{m}$ gap between the anchor and shuttle shown in the inset optical image. The shuttle is straddled by verniers that allow precise optical measurement of the shuttle's displacement. To the right of the sample stage there are a series of very soft springs and long thin tethers which together isolate the sample stage from the heat generated in the Chevron actuator at the far right end of the MEMS.

stretch the 2D crystal on the right. The two sample stages are separated by a gap that is nominally $3 \mu\text{m}$.

The most significant challenge in straining 2D materials with MEMS is anchoring the 2D crystal to the substrate. Crystals capable of forming 2D systems have strong in-plane bonds, but weak, van der Waals, out-of-plane bonds which is why it is a straight forward technique to obtain single layers from exfoliation of bulk crystals. This is also why many crystals that have become standard 2D materials are also known as good lubricants, graphene and molybdenum disulfide (MoS_2) for example. A number of strategies have been used to improve the bonding between atomically thin flakes and various substrates. Gold has been used with graphene in experiments stretching Kirigami structures [17], and photoresists have been used with graphene resonators to reduce damping caused by slipping between the membrane and substrate [12], [18], [19]. However, in the majority of these examples the strain within the crystal structure is small, and hence the required anchor force is small. For example in the Kirigami structures the deformation is macroscopic and there is actually little strain within the material. Most applicable to our work is the work by Garza *et al.* [10] who anchored trilayer graphene using a femto-pipette to epoxy the 2D material to the MEMS. While we find this work [10] to be a valuable reference for many important concepts regarding the use of MEMS to strain 2D materials, a close examination of the data shows that appreciable strain was not achieved in that experiment. For further details see supplementary material S1. We use a polymer to place the atomically thin flake onto our MEMS device. Instead of removing the polymer to obtain a clean sample, we leave the polymer coating the flake and sample stage region of the MEMS providing a strong mechanical coupling between the two. The obvious draw back of this method is that the atomically thin crystal remains coated in polymer during actuation, but this is a suitable compromise at this time to make progress in the incorporation of 2D materials with MEMS.

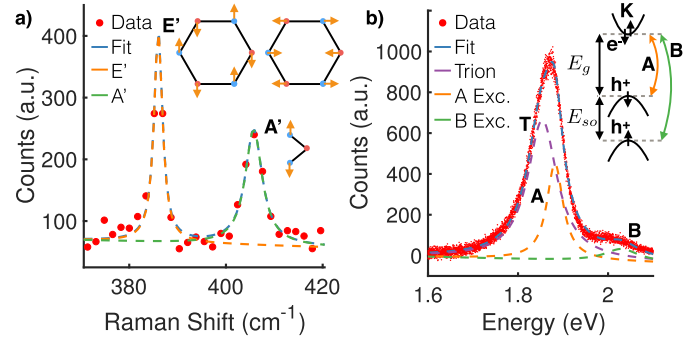


Fig. 2. **a)** Raman spectrum: Next to each peak in the spectrum is a diagram depicting the motions of the atoms in the corresponding phonon mode. The red and blue circles are molybdenum and sulfur atoms. **b)** PL spectrum: The trion, A exciton, and B exciton components are broken out individually. Inset is a diagram of the band structure near the K point of the BZ, which shows the spin-orbit splitting of the valence band responsible for the separation between the A and B excitons.

An additional difficulty with incorporating atomically thin crystals with MEMS are the fragile, non-planar MEMS structures. The vast majority of transfer methods in the literature are targeted at placing 2D materials on flat substrates. With MEMS we have to gracefully handle steps on the substrate that are several micrometers high. Further, we need to release our MEMS devices prior to transfer to avoid exposing the anchoring polymer and 2D crystal to hydrofluoric acid (HF), which means that we are transferring our flakes onto very fragile structures. Not only is there the possibility of the HF degrading the transfer polymer anchoring the flakes, but there is also the possibility that the HF would etch the thin native oxide layer between the transfer polymer and poly-silicon structure greatly reducing the mechanical coupling between the two. Further, since the transfer polymer covers a large portion of the sample stage, an extended HF exposure would be necessary to release the MEMS making the native oxide etch even more likely. To circumvent these issues we have developed a technique for transferring 2D materials onto MEMS utilizing a specially designed microstructure which facilitates a gentle, non-planar compliant transfer after HF release.

In the experiments presented below we focus exclusively on monolayer MoS_2 for our samples. MoS_2 is a direct-gap semiconductor [20] with two inequivalent valleys with opposite spins [21]. These properties make MoS_2 an interesting material for building nano-electronic devices such as transistors [22] and phototransistors [23]. The two valleys can be coherently optically addressed [24]–[26], which may be useful in novel applications such as valleytronics [25] and spintronics [27]. For these reasons there is an extensive body of research on MoS_2 including the strain response of the Raman and photoluminescence (PL) spectra [3], [4], [6], [7]. We will rely on this literature to determine the strain in our 2D crystals from the Raman and PL spectra we measure, proving that we are able to strain atomically thin flakes with MEMS.

Fig. 2 shows typical unstrained Raman and PL spectra for monolayer MoS_2 . Next to each peak in the Raman spectrum in Fig. 2a is a diagram depicting the atomic displacements of the corresponding phonon mode. Of interest to us are

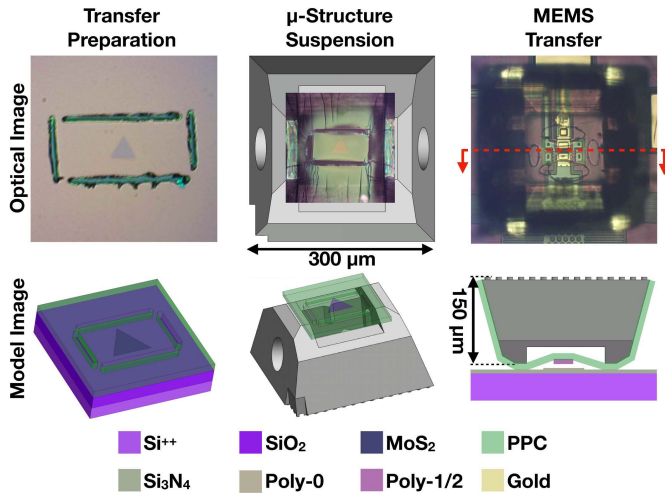


Fig. 3. The top row contains optical images of actual samples and devices, while the bottom row contains model images that provide a schematic view of the fabrication method. Each column corresponds with a step in the fabrication process, and all steps except the CVD growth of the MoS₂ are displayed here. The microstructure shown in the last two columns has a vertical hole running through it, which is how we see the MEMS through the structure in the last column.

the degenerate in-plane E' modes with an unstrained energy of 385 cm^{-1} , and the out-of-plane A' mode with an unstrained energy of 405 cm^{-1} [6], [7], [28]. Corresponding with the honeycomb crystal lattice, MoS₂ has a hexagonal Brillouin Zone (BZ), and like graphene the low energy electronic states occur at the K and K' points in the corners of the BZ where the band gap is at its minimum. The band structure near the K point is shown in the inset of Fig. 2b, and the structure is identical at the K' point but with spins flipped due to time-reversal symmetry [21]. Notably the valence band is split by spin-orbit coupling which results in two exciton peaks in the PL spectrum [20]. The A and B exciton peaks correspond with the upper and lower valence bands and have unstrained energies of 1.89 eV and 2.03 eV. A third peak makes a considerable contribution to the PL spectrum and corresponds with a trion, a bound state of two electrons and hole [29], [30]. The contributions for each of these components of the PL are shown in Fig. 2.

II. METHODS

A. Device Fabrication

An overview of our device fabrication method is shown in Fig. 3, and has 4 steps (3 shown in the figure): sample growth, transfer preparation, microstructure suspension, and MEMS transfer. **Sample Growth** Our monolayer MoS₂ crystals are grown via Chemical Vapor Deposition (CVD); the details of which are provided in a previous publication [3] which demonstrates the high quality and strength of our MoS₂ films and characterizes the Raman and PL strain response using pressurized micro-chambers. Importantly the growth is done on a degenerately doped silicon substrate with 285 nm of oxide, which allows us to identify isolated monolayer flakes without cracks of suitable size, $\sim 60 \mu\text{m}$ on a side.

1) *Transfer Preparation*: Our method begins like most methods for transferring atomically thin flakes [31]–[35]; we spin a transfer polymer onto the MoS₂ film which enables us to pull the flake free of its substrate and move it onto a new substrate. In our case we use Poly(propylene carbonate) (PPC), because we find it to be less brittle than the more commonly used Poly(methyl methacrylate) (PMMA). Next we deviate from the standard methods in two ways: 1) We make “strain relief” cuts in the transfer polymer using a probe in a micro-manipulator. These cuts allow us in the next step to tightly stretch the polymer over a microstructure without creating cracks in the sample. The cuts are achieved by carefully pressing the probe tip into the PPC and dragging the tip across the surface slowly to account for the viscoelastic flow of the PPC. We have found that the size of the probe tip is critical to making good cuts, and use GGB Industries T-4-60 Tungsten probe tips, which provide enough rigidity to puncture the PPC while still offering a fine enough tip ($< 3 \mu\text{m}$) to make precise cuts. 2) We are able to release the transfer polymer and flake from the growth substrate with a simple deionized water bath. Notably this is safer and cleaner than typical methods that use hydrofluoric (HF) acid or other chemicals to etch the substrate away from the flake.

2) *Microstructure Suspension*: Now that the MoS₂ is freely suspended on PPC we can place it on a microstructure designed specifically for use with our MEMS. The microstructure is fabricated via Direct Laser Writing (DLW), a high-resolution (sub-micrometer) 3D printing technique. Additional details of the 3D printing technique and mechanical properties of the microstructure material can be found in [36]. The transfer of the flake and PPC onto the microstructure is accomplished using a micromanipulator for positioning, and temperature controlled stage to slightly heat the microstructure ($\sim 35^\circ\text{C}$). The heat improves adhesion and reduces strain on the polymer and flake. The optical image in Fig. 3 clearly shows cracks in the PPC, but the cracks stop at the strain relief cuts leaving the sample pristine. Note that the PPC is mounted on the microstructure with the flake on the side of the PPC facing away from the microstructure. This means that the flake will be in direct contact with the MEMS structure when it is transferred onto the MEMS in the next step, which facilitates electrical connectivity between the flake and MEMS. We have observed the conductivity between the anchor and shuttle increase from zero, pre-transfer, to a finite value after transferring a graphene flake using the method presented in this paper, demonstrating electrical connectivity.

3) *MEMS Transfer*: Finally, the flake and PPC are transferred onto the MEMS. Prior to transfer the MEMS is released in HF, functionality is electrically tested using a probe station, and a thorough cleaning via oxygen plasma is done. Similar to the transfer onto the microstructure a micromanipulator is used to position the sample over the MEMS, and the MEMS is heated. This time the stage is heated to $\sim 75^\circ\text{C}$ in order to heat the PPC through its glass transition temperature ensuring that the PPC melts onto the MEMS structure. The temperature controlled stage also allows us to achieving a gentle transfer by using the thermal expansion of the stage to bring the sample in contact with the MEMS. The last step is to heat the MEMS

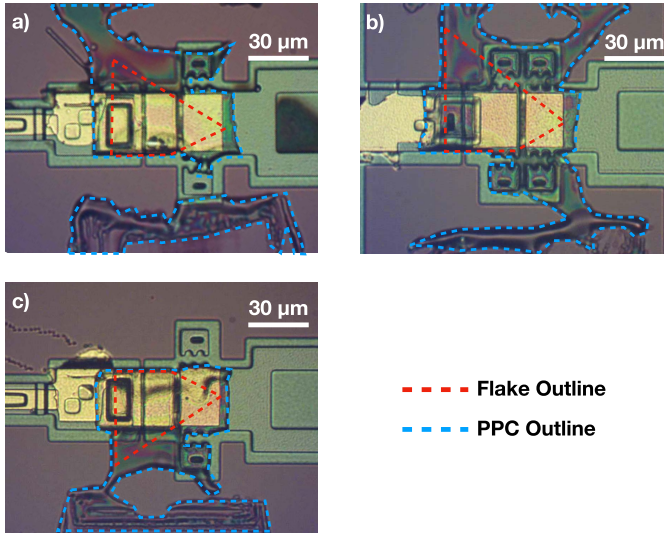


Fig. 4. Optical images of three devices which showed strain response. The blue dashed line outlines the regions covered in PPC, and the red dashed line outlines the MoS₂ flakes on the devices. a) Device M24 b) Device M25 (Force Meter) c) Device M26. Note that the flake for each device began as a nearly perfect equilateral triangle, and that the missing corners in the images result from freely hanging PPC rolling up during the transfer.

to 90°C on a hotplate for 10 minutes. This allows the PPC to fully melt enabling the 2D material to fully conform to the MEMS substrate. Finally, the MEMS is mounted in an integrated circuit socket and ball bonded.

While the techniques described above require some non-standard methods, we have found that the **Transfer Preparation** and **Microstructure Suspension** steps can be completed, with practice, with nearly 100% yield. However, the **MEMS Transfer** step has a roughly 75% yield despite significant optimization effort. The primary remaining failure mode is that the flake and PPC do not make contact with the MEMS even when the microstructure is in contact and even compressed against the MEMS substrate. We suspect that this occurs because the PPC is not stretched tightly across the microstructure leading to the PPC bowing into the microstructure away from the MEMS substrate. We suspect that slight over heating of the PPC during the transfer onto the microstructure is the root cause, but tests have been inconclusive thus far.

Fig. 4a-c show optical images of three devices we fabricated using our method. We will refer to these devices and the datasets collected from them as M24, M25, and M26 throughout the text. Note that M25 has a different sample anchor stage from M24 and M26. The anchor on M25 is mounted on springs to measure the stress in the 2D crystal. Common to all of these devices is a coating of PPC, which has been outlined in blue in the optical images. While the PPC does provide a strong mechanical coupling between the flake and the MEMS, it is a viscoelastic polymer, which damps the actuation of the sample stage shuttle. This introduces some subtleties in interpreting our data as discussed below. From these three devices we collected four datasets. The fourth dataset comes from a second experiment with device M26 collected after repairing the device. The first experiment

on M26 ended prematurely when the thermal relief tethers fractured. We repaired the breaks in the tethers by UV gluing scavenged MEMS parts over the breaks in the tethers as described and shown in supporting material S2. We will refer to the fourth dataset, collected after repairing M26, as M26 v2.

B. Measurements

Fig. 5 shows an example data set consisting of Raman and PL spectra measurements made on M26 v2 under actuation. Included in Fig. 5 are the various peak positions versus strain and the corresponding slopes giving the rate at which strain changes the peak position. These slopes are tabulated below for each data set in Table IV and Table V for Raman and PL data respectively.

The Raman and PL measurements were made using a Renishaw spectrometer with an 1800 line per mm grating. The MoS₂ films were excited with an Argon ion laser with wavelength 514.5 nm with a beam waist of $\sim 1 \mu\text{m}$. Considering the fragility and low thermal dissipation in our suspended samples, power was kept below 20 μW to avoid heating and sample damage. Heat from the Chevron actuator can significantly elevate the temperature of the entire die if a good path for thermal dissipation is not established. To prevent this our dies are mounted with silver epoxy to a copper plate which can be cooled with a thermal electric cooler (TEC) as needed. The copper plate has an internal platinum resistive thermal device for monitoring the temperature, and we periodically check the temperature of the sample anchor and shuttle stages using Raman thermometry [10], [37], [38]. For our actuators we are able to sufficiently strain the samples with less than 250 mW of power, and we find that as long as the die has a low thermal impedance to the copper plate, the TEC is not necessary.

Since friction between the MoS₂ film and MEMS is low, it is important that we are able to detect strain at its earliest onset. We achieve this by continuously monitoring the A exciton peak, the strongest peak, while slowly increasing the power to the actuator in steps which will increase the strain by less than 0.5%. The step size is determined by a precise measurement of the size of the suspended portion of the sample, always underestimating for safety, and relying on our calibrated displacement versus power curve for our devices (see supporting material S3). As soon as a shift is noticed in the peak position, the power is held constant while taking PL and Raman measurements. Because of the viscoelastic behavior of our polymer anchor, in most cases it was possible to simply wait ~ 10 minutes between measurements for the peak positions to shift further. The wait-measure cycle was repeated until the peak either stopped shifting, in which case more power was applied, or the peak relaxed marking either a major slip between the PPC and MEMS or in several cases the thermal relief tethers breaking. While in some cases it was possible to get further strain response from the sample, all datasets we have analyzed here are monotonic in A exciton peak shift.

Given the viscoelastic behavior of our devices the amount of time spent measuring spectra becomes an important trade-off

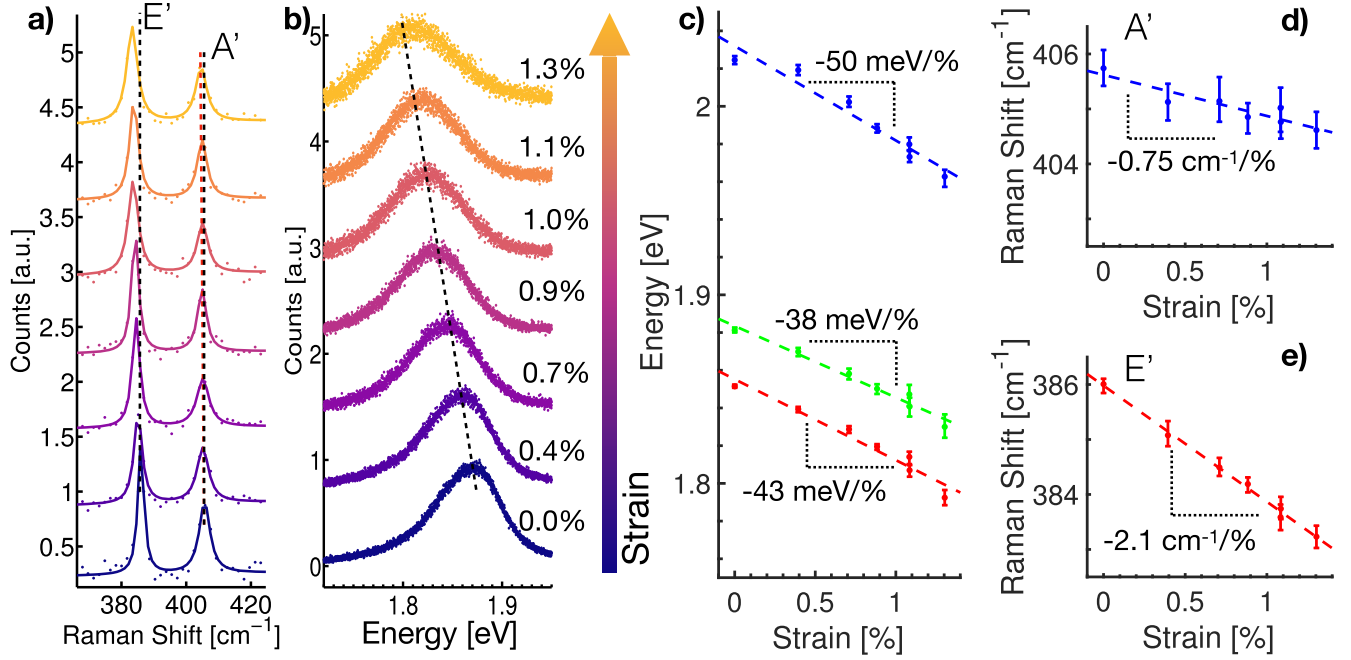


Fig. 5. Raman and PL spectra of sample M26 v2 as a function of strain. Values for the strain are extracted from the peak positions as described in the text. **a)** Raman Spectra with increasing strain, dashed line indicates the unstrained peak positions. **b)** PL Spectra, dashed line provides a guide to the eye how the peak shifts with strain. **c)** Trion peak position versus strain in red, A exciton peak position versus strain in green, and B exciton peak position versus strain in blue. **d)** A' phonon energy versus strain **e)** E' phonon energy versus strain.

between collecting high quality data (long time) and strain resolution (short time). We found that at a minimum, we needed to collect Raman data for 135 s to have adequate statistics for our analysis. The PL has a much stronger signal, only requiring 10 s, but because of the large spectral range of the measurement the grating must be rotated during the acquisition so the measurement takes ~ 3 min. A lower resolution grating could not be used to shorten this time, since the Raman features are narrow, only ~ 10 data points per peak. The measurement time results in a small time delay between the measurement of each of the PL peaks, and a much larger time delay between when the Raman and PL data are collected. The implications of these time delays on our data is discussed in the sections below.

III. DETERMINING STRAIN FROM PEAK POSITIONS

Strain changes the electron and phonon band structures, which we measure as shifts in the Raman and PL peak positions. Group theory places strong restrictions on the functional dependence of peak positions on the strain tensor. Further, since the strain is small, we limit our analysis to first order in strain. For our purposes there are only two point group representations of the crystal symmetry that are of interest, A' and E' . A' is the trivial representation and must be rotationally invariant. The only first order rotational invariant of the strain tensor is the trace, $\epsilon_{xx} + \epsilon_{yy}$, which is also called the hydrostatic strain since it is the strain that is experienced when a material is compressed on all sides equally as is the case when compressing with a fluid. Hence peaks that transform under the A' representation must change

under uniaxial strain according to the formula

$$\omega_{A'} = \omega_{0A'}[1 - \gamma_{A'}(1 - \nu)\epsilon] \quad (1)$$

where $\omega_{0A'}$ is the zero strain energy, $\gamma_{A'}$ is the Grüneisen parameter, ν is the Poisson's ratio, and ϵ is the magnitude of the uniaxial strain. Both the Raman A' peak and the PL peaks shift under strain according to (1), and the above notation for the A' phonon is used when referring to the A exciton peak except replacing ω with E and A' with A .

The E' phonon peak is a degenerate peak that shifts under uniaxial strain according to

$$\omega_{E'}^{\pm} = \omega_{0E'} \left\{ 1 - [\gamma_{E'}(1 - \nu) \mp \frac{\beta_{E'}}{2}(1 + \nu)]\epsilon \right\} \quad (2)$$

where $\omega_{0E'}$ is the zero strain energy, $\gamma_{E'}$ is the Grüneisen parameter (this term is identical with the strain term in (1)), and $\beta_{E'}$ is the shear deformation potential. The \pm in (2) denotes the lifting of the degeneracy under strain, which splits the peak into two peaks, the $+$ peak and the $-$ peak.

Table I provides a list of measured Grüneisen parameters and shear deformation potential values for the various Raman and PL peaks of MoS₂. Most experiments reported errors only for their measurement of the shift rate of the peak position with respect to strain, and not for the values of Grüneisen parameter or shear deformation potential. So most errors reported in Table I are adapted from the shift rate errors in the literature. The notable exception to this is [4], which reported error bars for $\gamma_{E'}$ and $\beta_{E'}$. However, these parameter values disagree substantially from the rest of the literature, which is consistent, so they have not been used in computing the effective values at the bottom of the table.

TABLE I
GRÜNEISEN, γ , AND SHEAR DEFORMATION POTENTIALS, β ,
FOR RAMAN AND PL PEAKS OF MoS₂

Reference	$\gamma_{E'}$	$\beta_{E'}$	$\gamma_{A'}$	γ_A
Lloyd <i>et al.</i> [3]	0.68 ± 0.1	NA	0.21 ± 0.1	2.6 ± 0.2
Rice <i>et al.</i> [6]	0.65 ± 0.1	0.34 ± 0.1	0.21 ± 0.1	NA
Wang <i>et al.</i> [7]	0.6 ± 0.1	0.3 ± 0.1	NA	NA
Conley <i>et al.</i> [4]	$1.1 \pm 0.2^*$	$0.68 \pm 0.1^*$	NA	3.7 ± 0.6
Effective Values ^a	0.64 ± 0.06	0.32 ± 0.07	0.21 ± 0.07	2.7 ± 0.2

^aEffective values and standard deviations are the maximum likelihood values and distribution standard deviation assuming the corresponding Gaussian distributions for the literature values.

*Values not used in computing effective values because of substantial disagreement with the rest of the literature.

Our objective is to use the known formulas for the strain behavior of the peaks, along with the parameters in Table I to infer the strain in our measurements from the observed peak positions. However, to do so a value for the Poisson's ratio, ν , must be provided. It is generally assumed that an atomically thin flake will inherit the Poisson's ratio of its substrate since it is assumed the two stick to each other well. We'll address this assumption more directly below, but for the time being adopt this assumption. The Poisson's ratio of PPC is not known, so is approximated from two similar polymers, Poly(bisphenol A carbonate) with $\nu = 0.41$ and Polypropylene with $\nu = 0.43$ [39]. Thus in the analysis that follows ν is assumed to have a value of 0.42. For comparison, $\nu \approx 0.27$ for monolayer MoS₂ [40]–[42].

An additional consideration that needs to be made in analyzing the data regards the degeneracy of the E' mode which is lifted under uniaxial strain. As the strain breaks the crystal symmetry, the two degenerate E' modes split into a mode that is parallel with the major strain axis, E'_- , and a mode that is perpendicular to the major strain axis, E'_+ . However, the Raman spectra in Fig. 5a does not show split E' modes. This is due to the accidental selection of only the E'_- mode in our measurement setup. The selection rules for the two modes are

$$I_- \propto \sin^2(\theta_i + \theta_s + 3\phi_\epsilon) \quad (3)$$

$$I_+ \propto \cos^2(\theta_i + \theta_s + 3\phi_\epsilon) \quad (4)$$

where ϕ_ϵ is the angle between the ZZ axis of the crystal lattice and the major strain axis, and θ_i and θ_s are the incident and scattered polarizations of light relative to the major strain axis in the Raman measurement [6], [7], [43], [44]. Fig. 6 shows an image of one of our MoS₂ films after the PPC has been removed making the edges of the flake obvious, and includes markers showing the incident and scattered polarizations of light selected in our experiment. The PPC was removed by placing the device in a 60°C acetone bath for one hour, and keeping the sample in the bath for another two days at room temperature to ensure all the PPC was dissolved. The device was then transferred to a methanol bath and critical point dried with carbon dioxide. Given it is highly preferential for CVD MoS₂ to grow with ZZ edge termination [45]–[47],

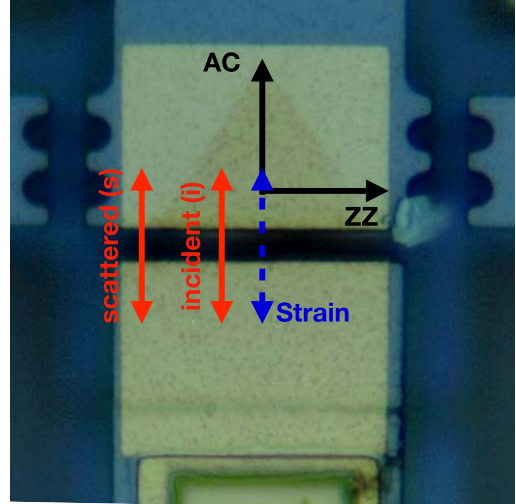


Fig. 6. Orientation of crystal and incident and scattered polarizations that lead to accidental selection of only the E'_- mode.

TABLE II
COMPOSITE STRAIN ANALYSIS FOR M26 v2 DATA SET

Step	A Exciton [%]	A' Phonon [%]	E' Phonon [%]	Composite [%]
0	-0.04 ± 0.04	-0.49 ± 0.66	0.00 ± 0.06	-0.03 ± 0.03
1	0.34 ± 0.07	0.76 ± 0.68	0.40 ± 0.10	0.37 ± 0.06
2	0.74 ± 0.10	0.72 ± 0.82	0.65 ± 0.07	0.68 ± 0.06
3	1.01 ± 0.09	1.31 ± 0.56	0.78 ± 0.06	0.86 ± 0.05
4	1.11 ± 0.19	0.98 ± 0.81	1.04 ± 0.10	1.06 ± 0.09
5	1.33 ± 0.17	1.49 ± 0.58	0.97 ± 0.09	1.06 ± 0.08
6	1.69 ± 0.21	1.79 ± 0.67	1.19 ± 0.09	1.28 ± 0.08

the edges of the flake indicated that the ZZ axis of the crystal is horizontal in Fig. 6. Since the flakes are placed on the MEMS pointing along the direction of strain, $\phi_\epsilon = 90^\circ$. The accidental scattered polarization selection results from the relative transmission efficiency of the grating in our spectrometer, which transmits the vertical polarization with $10\times$ the efficiency of the horizontal polarization, hence $\phi_s = 0^\circ$. The laser has vertical polarization so $\phi_i = 0^\circ$ as well. This combination of angles makes $I_+ \approx 0$.

To extract the strain from the Raman and PL spectra, the spectra are first fit to determine the energy position of the A exciton, E' , and A' peaks. The PL and Raman peak positions are used individually to calculate a value of strain. Then these individual strain values are combined as the inverse variance weighted mean. An example of this analysis is shown in Table II for the M26 v2 data set. The PL are fit with three Lorentzian peaks (one each for the trion, A exciton and B exciton) and a linear background. The Raman spectra are fit with two Lorentzian peaks (one each for the E' and A' modes) and a linear background. We find that the maximum strain achieved in our experiments before flake slipping or device breaking occur is $1.3 \pm 0.1\%$. Note that this strain is well below the yield strain of MoS₂, $> 6\%$ [42], so there is considerable room to improve our technique. Table III shows the maximum change in strain observed and the pre-strain in

TABLE III
MAXIMUM CHANGE IN STRAIN ACHIEVED AND PRE-STRAIN

Device	Max. Change in Strain [%]	Pre-Strain [%]
M24	0.76 ± 0.08	-0.01 ± 0.09
M25	0.63 ± 0.05	-0.05 ± 0.03
M26	0.86 ± 0.11	0.03 ± 0.06
M26 v2	1.30 ± 0.09	-0.03 ± 0.03

TABLE IV
SLOPES FOR RAMAN PEAK STRAIN RESPONSE

Device	E'_- [cm ⁻¹ /%]	A' [cm ⁻¹ /%]
M24	-1.95 ± 0.12	-0.78 ± 0.68
M25	-2.28 ± 0.43	-0.73 ± 0.52
M26	-1.45 ± 0.49	-0.75 ± 0.57
M26 v2	-2.11 ± 0.19	-0.75 ± 0.46
Lit.	-2.32 ± 0.41	-0.49 ± 0.17

TABLE V
SLOPES FOR PL PEAK STRAIN RESPONSE

Device	Trion [meV/%]	A Ex. [meV/%]	B Ex. [meV/%]
M24	-55 ± 5	-45 ± 9	-71 ± 13
M25	-84 ± 12	-81 ± 20	-29 ± 14
M26	-110 ± 22	-72 ± 25	-74 ± 21
M26 v2	-43 ± 2	-38 ± 3	-50 ± 5
Lit.	-	-30 ± 2	-

each device. Pre-strain is calculated assuming the unstrained A exciton energy is 1.88 eV, the E' phonon energy is 386 cm⁻¹, and A' phonon energy is 405.5 cm⁻¹. Table IV and Table V show the slopes for the Raman and PL peaks respectively for each dataset and the expected slopes given the literature values for the Grüneisen and shear deformation potential for each peak.

IV. STRAIN RESPONSE COMPARISON WITH LITERATURE

Since strain in our experiments is determined from literature values of the strain response of the various Raman and PL peaks, any attempt to compare the strain response we observe with literature would be circular reasoning. However, if we take ratios of the strain response of the peaks, then we eliminate strain as an independent variable, and create truly independent measures that can be compared with literature. Since there are three peaks with known strain response we can create three ratios of strain responses:

$$\frac{d\omega_{A'}}{dE_A} = \frac{\frac{d\omega_{A'}}{d\epsilon}}{\frac{dE_A}{d\epsilon}} = \frac{\omega_{0A'} \gamma_{A'}}{E_{0A} \gamma_A}, \quad (5)$$

$$\frac{dE_A}{d\omega_{E'}^-} = \frac{\frac{dE_A}{d\epsilon}}{\frac{d\omega_{E'}^-}{d\epsilon}} = \frac{E_{0A}}{\omega_{0E'} \gamma_{E'} (1 - \nu) + \frac{\beta_{E'}}{2} (1 + \nu)}, \quad (6)$$

$$\frac{d\omega_{A'}}{d\omega_{E'}^-} = \frac{\frac{d\omega_{A'}}{d\epsilon}}{\frac{d\omega_{E'}^-}{d\epsilon}} = \frac{\omega_{0A'} \gamma_{A'} (1 - \nu)}{\omega_{0E'} \gamma_{E'} (1 - \nu) + \frac{\beta_{E'}}{2} (1 + \nu)}. \quad (7)$$

Table VI contains values for the ratios computed from the experimental data and from the Grüneisen parameter and shear

TABLE VI
RELATIVE SHIFT RATES OF PEAKS

Slope Ratio	M24	M25	M26	M26 v2	Lit.
$\frac{d\omega_{A'}}{dE_A}$ [cm ⁻¹ /eV]	18 ± 6	10 ± 3	8 ± 3	20 ± 5	17 ± 22
$\frac{dE_A}{d\omega_{E'}^-}$ [meV/cm ⁻¹]	23 ± 3	36 ± 5	40 ± 8	18 ± 1	15 ± 6
$\frac{d\omega_{A'}}{d\omega_{E'}^-}$ [-]	.41 ± .15	.32 ± .12	.43 ± .17	.35 ± .10	.24 ± .34

deformation potential values from the literature. In calculating the 1 σ confidence interval for the literature values of the ratios, it was necessary to assume log normal distributions for the parameters. This is because the uncertainty is large relative to the parameter values and the parameters must be non-negative. Further, 1 σ confidence intervals were not available for all sources. In such cases we have assumed the interval to be equal to the worst reported interval for the same parameter by an alternative source. The interval for $\gamma_{A'}$ has to be completely assumed since none of the sources provide an interval. We have assumed the confidence interval to be ± 0.1 for each measurement, the largest interval for any of the Raman parameters used in the analysis. Given the non-linear functional form of the slope ratios, non-normal distribution for the parameters, and large uncertainties, the literature values for the ratios were computed using the Monte Carlo method with 10⁷ samples. Several calculations with a smaller number of samplings were done to ensure convergence of the calculation.

V. DISCUSSION

The data display clear inconsistencies. The most obvious trend is that devices M25 and M26 behave differently from M24 and M26 v2. M25 and M26 both have inconsistent slopes for the PL peaks. Not only is this different from the behavior of M24 and M26 v2, but also from experiments on biaxially strained MoS₂ [3] where all the peaks shift with roughly the same slope. This behavior is also apparent in Table VI where it results in low values for $\frac{d\omega_{A'}}{dE_A}$ and high values for $\frac{dE_A}{d\omega_{E'}^-}$. Further, $\frac{d\omega_{A'}}{dE_A}$ is arguably the most important comparison we can make with literature since it is independent of the Poisson's ratio. However, the M25 and M26 values for $\frac{d\omega_{A'}}{dE_A}$ are significantly lower than the values for M24 and M26 v2. In the case of M25 we have good reason to be suspicious of the consistency of the data because the sample stage was designed to shift in order to act as a force meter. We believe that the viscoelasticity of the anchor polymer allowed the sample stage to slowly creep, reducing the strain between the PL and Raman measurements. This hypothesis is supported by the fact that $\frac{d\omega_{A'}}{d\omega_{E'}^-}$, a comparison between two Raman peaks, is consistent with the other data sets. As for the M26 data, we suspect that one of the tethers connecting the actuator to the sample shuttle broke while acquiring the data and that the failure only came to our attention after the second tether broke. This would have caused the strain to partially deviate from uniaxial, and could have also introduced a similar reduction in strain between PL and Raman measurements.

Like the M25 data, $\frac{d\omega_{A'}}{d\omega_{E'}}$ for the M26 data is consistent with the other two data sets suggesting some slip between Raman and PL measurements. Considering the uncertainty regarding the M25 and M26 data sets we will disregard them in our discussion below.

Now we return to the peak slopes in Table IV and Table V. For the M24 and M26 v2 data the slopes are consistent for the trion, A exciton and B exciton as expected from literature [3], and the discrepancy between the data sets is not much more than two standard deviations. Similarly, the literature value for the A exciton slope is less than two standard deviations from the slopes for either data set, but does appear low. Turning to the Raman slopes, we see that while the A' slope is consistent across all data sets, there is a very large uncertainty in its value. The large uncertainty is due to the small shift in the A' peak under strain and low amplitude of the peak, which result in low confidence in the peak position and thus slope. Though the literature value for the A' slope is within a single standard deviation of all the measured slopes, our data suggest that the literature value is low. There is also good agreement between the E'_- slope of the M24, M26 v2, and literature values, and that all three slope ratios in Table VI are in good agreement between M24, M26 v2 and the literature values. This gives us good confidence in the strain values we have derived from the data.

We have assumed throughout that the MoS₂ flakes inherit the Poisson's ratio of the PPC. Here we evaluate the validity of that assumption. In other experiments that strain 2D materials on a substrate, strain is calculated in the substrate, neglecting the small perturbations caused by the atomically thin flake. Then it is assumed that there is no slipping between the flake and the substrate, so the strain in the flake must be the same as in the substrate. Hence, the flake inherits the Poisson's ratio of the substrate. However, in our case, the 2D material is much more than a perturbation to the strain distribution. For mechanical calculations, the effective Young's modulus and thickness of MoS₂ are ~ 270 GPa and 0.65 nm [40], [42], while the Young's modulus of PPC is ~ 37 MPa [48], [49] and we estimate the thickness to be no greater than 600 nm given the optical interference of comparably prepared films of PPC on silicon substrates. Thus the effective 2D Young's modulus of MoS₂ and PPC are 175 N/m and 22 N/m respectively, and it is no longer a good assumption that the substrate elastic constants alone determine the strain distribution. In the supplementary material, S4, we discuss the boundary conditions and a first-order method for estimating the effective Poisson's ratio for 2D materials adhered to substrates. Importantly, the effective Poisson's ratio depends on the thickness of the PPC substrate, and this potentially explains the discrepancies between the experimental and literature values for the Raman and PL peak slopes in Table IV and Table V. Further, since the PPC could be of slightly different thicknesses on the M24 and M26 v2 devices, the effective Poisson's ratios could be slightly different which would explain some of the device to device variation in peak slopes. To assess this possibility we performed our analysis using 0.35 and 0.27 for the Poisson's ratio, and tabulated the results in the supplementary material S5. The analysis reveals

that our results vary only slightly with Poisson's ratio, but that a smaller value for the Poisson's ratio does give marginally better agreement between our results and literature.

An additional source of error worthy of discussion is the difference in strain due to viscoelastic drift between the time of the PL and Raman measurements. We did attempt to correct for the drift in strain in the M24 dataset by measuring the peak shift versus time in the Raman and PL data, and then take ratios of the shift rates to determine values of $\frac{d\omega_{A'}}{dE_A}$, $\frac{dE_A}{d\omega_{E'}}$, and $\frac{d\omega_{A'}}{d\omega_{E'}}$. However, the changes to the slope ratios were much less than the confidence intervals, and there is significant uncertainty as to the exact time delay, so we chose not to include this correction in our analysis.

We would have tested the homogeneity of strain in our experiments had it been possible. However, once our samples began to show signs of strain, the strain continued to increase uncontrollably due to viscoelastic draft, and we had to take measurements as fast as possible in order to capture the strain progression. In lieu of such measurements we performed simulations to estimate the strain inhomogeneity caused by the expected trapezoidal shape of the suspended sample. Simulation results and details are provided in the supplementary material S6. The simulations show a highly uniform strain distribution in the middle of the sample, which demonstrates that the uniformity of the stress and strain in the middle of the sample is robust to perturbations at the edge of the sample where irregularities in the PPC and flake are most likely to occur.

Furthermore, if it wasn't for viscoelastic drift we would have performed Raman measurements sweeping the incident and/or scattered beam polarizations to confirm crystal lattice orientation and observe E' splitting. Note that the splitting would not have been well resolved because the expected split at 1.3% strain is 2.3 cm^{-1} and the full width half maximum of the Raman E' peak is $\approx 3.5 \text{ cm}^{-1}$. However, the change in peak position and effective width would be unmistakable.

VI. CONCLUSION

In conclusion, we have strained monolayer MoS₂ with a MEMS for the first time, and achieved $1.3 \pm 0.1\%$ strain. This is a major milestone in the field of 2D materials and MEMS, and marks an important advancement towards creating novel devices with 2D materials. While there is much work to be done in improving the sample quality and anchoring of the 2D material, this opens a direct path towards building novel strain based devices such as strain tunable LEDs, FETs, and even a low resolution spectrometer by adjusting the absorption spectrum. Further, the MEMS platform offers many exciting avenues for exploring physics in 2D systems by enabling strain engineering. Some obvious examples include Pseudo-magnetic field generation and exciton confinement for forming exciton condensates.

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