A strain tunable single-layer MoS$_2$ photodetector

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ABSTRACT: Strain engineering, which aims to tune the bandgap of a semiconductor by the application of strain, has emerged as an interesting way to control the electrical and optical properties of two-dimensional (2D) materials. Apart from the changes in the intrinsic properties of 2D materials, the application of strain can be also used to modify the characteristics of devices based on them. In this work, we study flexible and transparent photodetectors based on single-layer MoS$_2$ under the application of biaxial strain. We find that by controlling the level of strain, we can tune the photoresponsivity (by 2-3 orders of magnitude), the response time (from <80 msec to 1.5 sec) and the spectral bandwidth (with a gauge factor of 135 meV/% or 58 nm/%) of the device.

KEYWORDS: 2D materials, MoS$_2$, strain engineering, photodetector, biaxial strain, flexible electronics, transparent substrate.
Tuning the characteristics of circuit components with an external knob is at the deep core of modern electronics. Good examples of that are the field-effect transistors whose conductance can be adjusted by means of an applied voltage to the gate electrode.\textsuperscript{1–5} The development of new tuning knobs have undoubtedly opened up possibilities to design new device concepts.\textsuperscript{6–11} Strain engineering provides a powerful route to modify the electrical and optical properties of electronic materials and thus it has the potential to become an excellent external tuning knob.\textsuperscript{12–14} Conventional strain engineering approaches, however, typically yield fixed strain levels. This has radically changed with the isolation of 2D materials which provide an excellent platform for strain engineering as they can be easily stretched and bended to a large extent in a reversible way.\textsuperscript{15–19} Moreover, optical spectroscopy techniques have demonstrated that tensile uniaxial or biaxial strain effectively decreases the bandgap in atomically thin transition metal dichalcogenides.\textsuperscript{20–26} Here we exploit the strain tunable bandgap of single-layer molybdenum disulfide (MoS\textsubscript{2}) to fabricate a photodetector device whose responsivity, response time and spectral bandwidth can be adjusted by an externally applied biaxial tensile or compressive strain. We find a remarkably large strain sensitivity of the cut-off wavelength of 58 nm/% (~135 meV/%) of strain making it possible to extend the detection spectral bandwidth with respect to pristine unstrained devices.

We fabricate single-layer MoS\textsubscript{2} photodetectors with a simple photocell geometry (Figure 1a). Single-layer MoS\textsubscript{2} is prepared by mechanical exfoliation of bulk molybdenite (see Materials and Methods for details) and the exfoliated flakes are then transferred to a Gel-Film (from Gel-Pak\textsuperscript{®}) substrate. The single-layer regions are identified by micro-reflectance spectroscopy\textsuperscript{27,28} and quantitative analysis of transmission mode optical microscopy images. The selected single-layer flakes are then transferred onto a polycarbonate (PC) substrate with pre-patterned drain-source electrodes by a dry-transfer
deterministic placement method and are annealed at 100 °C to improve the electrical contact between the flake and the electrodes. Figure 1b shows an optical microscopy image of one of the assembled single-layer MoS\(_2\) photodetector devices fabricated onto PC. Note that it has been previously demonstrated that polymeric substrates, such as PC, polyimide and polydimethylsiloxane (PDMS) can be used to integrate photonic devices. Polycarbonate has been used as substrate because of the trade-off between high Young’s modulus and large thermal expansion that allows one to biaxially stretch (or compress) the MoS\(_2\) by moderately warming up (or cooling down) the substrate while ensuring an excellent strain transfer (see Supporting Information to observe the homogeneity of the strain applied). Substrates with larger thermal expansion but lower Young’s modulus cannot effectively transduce their thermal expansion into biaxial strain, as predicted by finite elements analysis (see Supporting Information). In the case of polycarbonate (\(E = 2.5\) GPa) the calculated efficiency is larger than 80%. The mechanical model, which lacks atomistic details, gives only a coarse estimation of the strain transfer efficiency and for this reason we assume in the rest of the paper a perfect transduction of thermal expansion to biaxial strain. Note that by assuming perfect transduction, the gauge factors obtained in this work can be considered as lower bound limits. Figure 1c shows differential reflectance spectra acquired on a single-layer MoS\(_2\) device at different tensile strain levels from 0% (substrate at room temperature, \(T = 25\) °C) to 0.48% (substrate heated at \(T = 100\) °C). The A and B exciton peaks in the reflectance spectra redshift upon increasing the substrate temperature (and thus the biaxial tensile strain) indicating a narrowing of the MoS\(_2\) bandgap. Figure 1d shows the relationship between the substrate temperature increase and its biaxial expansion. We address the reader to Reference 34 for details about the measurement of thermal expansion and the calibration of the applied biaxial strain.
In order to test the spectral response of the MoS$_2$ photocell detectors we illuminate the devices with a tunable light source (Bentham TLS120Xe) to select the wavelength (with a full-width at half maximum of $\sim$10 nm) while the current across the device (biased at 10 V) is measured. The light is focused into a spot of 400 $\mu$m of diameter with a power density of 8 mW/cm$^2$. At each step during the wavelength sweep we measure both the dark and illuminated current to rule out drifts during the measurement. The responsivity $R$ of the device can be extracted from the photocurrent values as

$$R = \frac{I_{ph}}{P_{dens} \cdot A_{dev}}$$

where $P_{dens}$ is the incident light power density and $A_{dev}$ is the area of the MoS$_2$ channel. The biaxial strain applied to the device is controlled through a thermal stage (see Materials and Methods).

The responsivity spectrum measured for the unstrained device is comparable with the data reported in literature for MoS$_2$ phototransistors fabricated by electron beam lithography on SiO$_2$/Si substrates at similar illumination power density and biasing conditions (larger responsivities are reported for devices at very low illumination levels and upon much larger drain-source electric field biasing conditions).$^{35-37}$ The spectra clearly show two peak features, which are in good agreement with the B and A excitonic resonances also observed in the reflection spectra of single-layer MoS$_2$, $^{38-40}$ and an abrupt drop of the device responsivity after the A exciton peak. When the polycarbonate substrate is biaxially strained, the MoS$_2$ photocell responsivity spectrum changes significantly. In Figure 2a, the overall responsivity values increase by a factor of $\sim$100 when the strain is increased from -0.8% to 0.48% (a factor of $\sim$1000 for device #2 where the strain ranges from -1.44% to 0.48%). We attribute this effect to the applied strain as similar photocells
fabricated on SiO$_2$ (which have negligible thermal expansion) do not show this strong enhancement of the photoresponse (see Supporting Information).

To get an insight about the tensile strain induced increase of the overall responsivity we studied the response time of the devices. For MoS$_2$ photodetectors the dominant mechanism for photocurrent generation is photogating.$^{35,41-43}$ In this mechanism the responsivity is proportional to the carrier lifetime (which can be extracted from the response time of the device to pulsed illumination). While the response time to pulsed illumination for devices under compressive strain is very sharp (the response is faster than that of our experimental setup, 80 msec), tensile strained devices show slower response times $\sim$1.5 sec (Figure 2a inset) in agreement with the increase of responsivity observed in our devices. We rule out the effect of the temperature as photodetectors fabricated on SiO$_2$, with negligible thermal expansion, do not show a sizeable variation of the response time in the temperature range of 25 ºC to 100 ºC (see Supporting Information). Therefore, the increase of response time seems to be due to tensile biaxial strain although the microscopic mechanism is still unknown, and it will be subject of further study. Note that we also studied the effect of strain on the Schottky barrier height through scanning photocurrent (see Supporting Information) finding that the Schottky barrier height in our devices is very small ($\sim$14 meV for pristine unstrained devices) and they show a moderate decrease of the barrier height upon tensile straining. This mechanism also contributes to the increase of photoresponse observed for tensile stressed devices.

The excitonic features, visible in all the spectra of strained MoS$_2$, are redshifted (blueshifted) when increasing the tensile (compressive) strain value, similarly to what was observed in the differential reflectance spectra (Figure 1c), with a strain gauge factor of 31 nm/% ($\sim$94 meV/%) for exciton A (inset Figure 2b). The shift of the exciton peaks with strain in the responsivity spectra is in excellent agreement with that observed in the
differential reflectance measurements (Figure 1c and Ref. 34). The cut-off wavelength is shifted as well upon straining, as can be seen in Figure 2b. The device shows a strain gauge factor of the cut-off wavelength in the range of ~58 nm/% (~135 meV/%) of biaxial strain. Therefore, we demonstrated that applying tensile biaxial strain to the MoS$_2$ device can be an effective strategy to increase both the responsivity and the wavelength bandwidth of the photodetector (at the expense of a slower response time) while compressive strain can be exploited to yield faster photodetectors (although with a lower photoresponse and with a narrower wavelength bandwidth). This adaptable optoelectronic performance of this device can be very useful to adjust the photodetector operation to different lighting conditions, similarly to human eye adaptability (scotopic vision during the night vs. photopic vision during the daylight).  

A direct consequence of the strain induced redshift for tensile biaxial strain is that one can achieve a sizeable response for wavelengths even beyond the cut-off of pristine (unstrained) MoS$_2$. Figure 2c shows an example where the photocurrent of the device (with light of 740 nm with a power density of 5 mW/cm$^2$, applying a bias voltage of 5 V) is measured while changing the strain level in time. One can appreciate how the photocurrent at 740 nm increases substantially upon increasing the strain level of the device (from 0.16% to 0.48% of tensile biaxial strain) in a reproducible way, which can be attributed to the strain induced redshift of the device cut-off wavelength. Interestingly, the device can be strain tuned rather quickly (in ~20 seconds time scale, see Figure 2d) and this tuning time most likely is only currently limited by the thermalization time of our temperature stage and it could be further improved by employing micro-heaters located underneath the photocell device.

In order to study the reproducibility of the redshift in the responsivity spectra of single-layer MoS$_2$ photodetectors, several cycles of application/release of tensile strain were
performed in another MoS$_2$ photodetector. Figure 3 shows the device responsivity for 740 nm illumination wavelength (power density of 12 mW/cm$^2$ and bias voltage of 10 V) recorded during some of the straining cycles. We observe how the responsivity evolves from negligible values (within the experimental setup noise level) for the unstrained device towards increasingly high values for the tensile strained devices, similarly to what was displayed for few cycles in Figure 2c. The device shows a good reproducibility during the whole range of cycles (up to 40 cycles were applied).

In summary we have harnessed the strain tunability of the band structure of single-layer MoS$_2$ to fabricate photodetectors with strain actuated bandwidth. The strain in these devices can be reversibly applied through the thermal expansion (shrinkage) of their substrate material which induces tensile (compressive) biaxial stress. We find that upon tensile straining the photoresponse increases and that the excitonic features present in the spectra redshift increasing the bandwidth in agreement with previous spectroscopic works. Interestingly the spectra also develop a slowly decaying tail for long wavelengths which further increases the bandwidth. We extract a strain gauge factor for the wavelength cut-off shift of up to $\sim$58 nm/% ($\sim$135 meV/%). This remarkably large value demonstrates that 2D semiconductors hold a great promise for future straintronic devices where strain is employed as a variable tuning knob. Indeed, the possibility of strain tuning the optoelectronic performance of photodetector devices in a fast timescale opens up the possibility to fabricate artificial photonic devices that mimic the adaptability of the human eye. The photodetectors discussed in this work, which can be tuned from a fast, low responsivity and narrowband state to a slow, very sensitive and wideband one, can open the possibility of developing adaptable artificial photonic elements.

**Materials and Methods**

*Materials*
The MoS$_2$ flakes have been obtained by mechanical exfoliation with Nitto tape (SPV 224) from a bulk natural crystal (Moly Hill mine, Quebec, QC, Canada) onto a viscoelastic polydimethylsiloxane (PDMS) stamp from Gel-Pak® (Gel-Film WF x4 6.0mil).

**Device fabrication**

The MoS$_2$ flakes located on the PDMS were characterized by optical transmission microscopy (Motic BA310Met-T metallurgical microscope, equipped with a 18 megapixel digital camera AMScope MU1803 and a fiber-coupled compact spectrometer Thorlabs CCS200/M) to determine the number of layers $^{27}$. The single-layer MoS$_2$ flakes were then transferred between pre-patterned Au/Ti electrodes (fabricated by electron-beam evaporation through a metal shadow mask from Ossila®, part number E324) on polycarbonate (PC) substrate by using an all dry deterministic transfer method $^{29}$.

**Strain application**

The biaxial strain on the photodetector devices is achieved through the thermal expansion of the PC substrate, controlled by a thermal stage (Linkam HFS600-P for the measurements shown in Figure 2 and a Peltier element with an approximated consumption of 5 W to reach the highest temperature value for the rest of the measurements).

**Photocurrent spectroscopy measurements**

The current vs. voltage characteristics of the devices were measured with a Keithley 2450 source-meter unit while the devices were illuminated focusing the light coming from a fiber coupled light source into a 400 µm spot, covering the whole area of the device and providing a homogeneous power density. A Bentham TLS120Xe tunable light source was used for the measurements shown in Figure 2, a halogen lamp equipped with a VariSpec™ Liquid Crystal Tunable Filter was used for the measurement shown in Figure
S6 in the Supporting Information. The rest of the photocurrent measurements were carried out with high power fiber-coupled LED sources from Thorlabs.

ASSOCIATED CONTENT

Supporting Information

Exciton A energy maps; dependency of the transferred strain with the Young modulus; responsivity spectra and response time comparison of single-layer MoS$_2$ on PC device and single-layer MoS$_2$ on SiO$_2$/Si device; characterization of single-layer MoS$_2$ on PC device under strain; responsivity spectra for strained and unstrained single-layer MoS$_2$ device; power dependency with the strain applied; photocurrent maps and spectroscopy measurements on a single-layer MoS$_2$ device on PC at different strain levels.

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![Figure 1.](image)

**Figure 1.** (a) Schematic picture of the setup used to perform all the measurements. The MoS$_2$ photodetectors are placed on a thermal stage and illuminated from the top. (b) Optical transmission photograph of a single-layer MoS$_2$ photodetectors fabricated on polycarbonate. (c) Differential reflectance spectra measured at different temperatures and
manually translated into the vertical axis by 0.08 to facilitate the inspection. Inset: wavelength of the exciton A as a function of temperature. (d) Calibration of the polycarbonate expansion dependency on the temperature.

Figure 2. (a) Responsivity spectra of the single-layer MoS$_2$ photodetector #1 obtained by measuring under 5 different strains applied (from -0.80% to 0.48%). Each dot corresponds to the value measured under light power of 8 mW/cm$^2$ and applying a bias voltage of 10 V. Note that the responsivity values for +0.16%, -0.16%, -0.48% and -0.80% have been multiplied by 1.7, 6, 10 and 10 respectively to facilitate the comparison between the spectra. Inset: Response time for different strains applied. (b) Cut-off wavelengths extracted from the responsivity spectra of three single-layer MoS$_2$ photodetectors (#1, #2 and #3) at different strain ranges. Inset: Exciton A wavelengths extracted from the same spectra. (c) Response time to strain, being the OFF state 0.16% of strain applied and ON state 0.48% of strain applied, the cycles are measured with an applied voltage of 5 V and the 740 nm light source density power is 5 mW/cm$^2$. (d) Zoom in the third strain cycle of (c) in order to appreciate the rise and fall time of the device, estimated with the 10%-90% criterion.
Figure 3. Photodetector #4 responsivities for 740 nm LED light with light power of 12 mW/cm² and applying a bias voltage of 10 V measured for several strain cycles.
Suppporting Information for:

A strain tunable single-layer MoS$_2$ photodetector

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Figure S1. (a) Optical image of the sample to facilitate the identification of the different areas. (b-d) Exciton A energy map extracted from the differential reflectance excitonic resonance peak at 0% (b), 0.22% (c) and 0.35% (d) of strain.

Figure S2. Finite element calculation of a biaxial strain test sample consisting of a 100-μm-thick substrate and single-layer MoS$_2$ (0.7 nm thickness) on the substrate. The curve represents the maximum transferred strain in MoS$_2$ as a function of substrate’s Young’s Modulus.

Figure S3. (a) Responsivity spectra of the single-layer MoS$_2$ photodetector #4 on PC obtained at 25°C (blue), 60°C (green), 80°C (orange) and 100°C (red). These values correspond to no strain (blue), 0.22% of strain (green), 0.35% (orange) and 0.48% (red). Each dot corresponds to the value measured under light power of 12 mW/cm$^2$ and applying bias voltage of 10 V. (b) Responsivity spectra of the single-layer MoS$_2$ photodetector on SiO$_2$/Si obtained at 25°C (blue), 60°C (green), 80°C (orange) and 100°C (red). Each dot corresponds to the value measured under light power of 12 mW/cm$^2$ and applying bias voltage of 10 V. (c) Responsivity measured with a 740 nm LED light at the same conditions as (a) and (b) for photodetector 4 on PC (application of strain with the temperature) and photodetector on SiO$_2$ (no strain application with the temperature).
Figure S4. (a) Responsivity spectra of the single-layer MoS$_2$ photodetector #3 on PC obtained at 25°C (blue) and 100°C (red). Each dot corresponds to the value measured under illumination with monochromatic light with a power of 8 mW/cm$^2$ and applying bias voltage of 10 V. (b) Responsivity spectra of the single-layer MoS$_2$ photodetector on SiO$_2$/Si obtained at 25°C (blue) and 100°C (red). Each dot corresponds to the value measured under illumination with monochromatic light with a power of 8 mW/cm$^2$ and applying bias voltage of 10 V. (c) Wavelength of the exciton A for 25 °C and 100 °C, extracted from a gaussian fit. (d) Responsivity in the exciton A for 25 °C and 100 °C, extracted from a gaussian fit.
**Figure S5.** Fall response time of the photodetector on SiO$_2$/Si and photodetector #4 on PC extracted for four different temperatures, measured with 420 nm LED light at bias voltage of 10 V.

**Figure S6.** (a) Responsivity spectra of the single-layer MoS$_2$ photodetector #5 obtained for no strain (blue), 0.22% of strain (green), 0.35% (orange) and 0.48% (red). Each dot corresponds to the value measured under light power of 12 mW/cm$^2$ and applying a bias voltage of 10 V. (b) Responsivity measured with a 740 nm LED light at the same conditions as (c).
Figure S7. Responsivity spectra of the single-layer MoS₂ photodetector #6 obtained without strain (blue) and with 0.48% of strain applied (red). Each dot corresponds to the value measured under a light power of 80 μW/cm² and applying a bias voltage of 10 V.

![Graph showing responsivity spectra](image)

Figure S8. Photocurrent vs. power for different applied strain levels on the single-layer MoS₂ photodetector #3. The photocurrent is extracted from a response time measurement under a bias voltage of 10 V and illuminating with a LED source with a wavelength of 505 nm.

(a) Optical microscopy image in reflection mode of the single-layer MoS₂ photodetector #7 on PC. (b) Reflection map of the same photodetector acquired with a laser of 650 nm. (c) Current maps of the same photodetector at different temperatures. The lines are placed using the reflection maps to indicate the position of the flake and the electrodes.
Figure S10. (a), (b), (c) Current linecuts in the single-layer MoS$_2$ photodetector #7 on PC measured for different bias voltages applied (from -50 mV to 50 mV, only few selected bias voltages are displayed for clarity) in the nearby area of the electrodes at different temperatures. (d), (e), (f) Photocurrent measured in the left electrode at different bias voltages. The grey line is the linear fit of photocurrent datapoints with values above the noise level. The cross of this line with the horizontal axis represent an estimation of Schottky barrier height at each temperature.