Strain-engineering the Schottky barrier and electrical transport on MoS$_2$

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Abstract
Strain provides an effective means to tune the electrical properties while retaining the native chemical composition of the material. Unlike three-dimensional solids, two-dimensional materials withstand higher levels of elastic strain making it easier to tune various electrical properties to suit the technology needs. In this work we explore the effect of uniaxial tensile-strain on the electrical transport properties of bi—and few-layered MoS$_2$, a promising 2D semiconductor. Raman shifts corresponding to the in-plane vibrational modes show a redshift with strain indicating a softening of the in-plane phonon modes. Photoluminescence measurements reveal a redshift in the direct and the indirect emission peaks signaling a reduction in the material bandgap. Transport measurements show a substantial enhancement in the electrical conductivity with a high piezoresistive gauge factor of $\sim 321$ superior to that for Silicon for our bi-layered device. The simulations conducted over the experimental findings reveal a substantial reduction of the Schottky barrier height at the electrical contacts in addition to the resistance of MoS$_2$. Our studies reveal that strain is an important and versatile ingredient to tune the electrical properties of 2D materials and also can be used to engineer high-efficiency electrical contacts for future device engineering.

Supplementary material for this article is available online

Keywords: strain engineering, MoS2, Schottky barrier, bandgap engineering, Piezoresistivity

(Some figures may appear in colour only in the online journal)

1. Introduction
Strain in the crystal lattice, by altering the lattice constants, orbital interaction and coupling between the neighboring atoms provides a powerful means to engineer the physical properties of a solid. Carrier mobility enhancement in strained Si on Silicon–Germanium substrates offered a vital solution for complementary metal-oxide-semiconductor (CMOS) performance improvements [1, 2]. An elastic strain of 1% is reported to induce a change of $\sim 100$ meV in the material bandgap [3]. In three-dimensional high-tensile materials such as stainless steel, only a small fraction $<1\%$ of the applied strain results in useful elastic deformation of the lattice while the rest relaxes via plastic deformations such as defects and dislocations [2, 4]. Contrary to the bulk materials, lower dimensional systems are shown to withstand higher levels of elastic strain [5, 6]. Owing to the two-dimensional nature the van der Waals (vW) materials are shown to host higher degrees of in-plane strain [7, 8]. Graphene has been reported to withstand the highest amount of strain [7] while the transition metal di-chalcogenides (TMDC) following closely [8]. Among the TMDCs, MoS$_2$, a promising microelectronics material, boasts a surprisingly high Young’s modulus and have shown to withstand much larger amounts of strain ($\sim 10\%$) [8, 9] compared to many of the high tensile materials [2]. The possibility of engineering the bandgap, carrier mobility, carrier density, effective mass etc, by the application of strain can establish MoS$_2$ as a potential microelectronic and straintronic material.

Reports on strain-tuning physical properties on TMDCs have been focussed mostly on spectroscopic studies such as Raman scattering and photoluminescence (PL) studies [10–14]. A continuous reduction of the bandgap, effective
mass and a transition from the direct to the indirect bandgap has been predicted in TMDCs [15–17]. It has been shown that the bandgap of MoS$_2$ can be continuously tuned by applying in-plane strain [11, 13, 18]. In addition, a direct to indirect bandgap transition and metal-insulator transition are also observed on MoS$_2$ [11, 13] and MoTe$_2$ [19].

Potential for device applications of any phenomena require its manifestation in electrical transport experiments. Such reports, where the effect of strain on the electrical transport properties such as the conductivity, the band gap and the metal-semiconductor contact barrier heights on TMDCs are only a handful [18–25]. Most of the studies utilize an atomic force microscope (AFM) tip [18, 19, 21, 22] to apply local strain, while transport studies on the global application of strain are only a few [23–26]. On MoS$_2$, the strain applied by the AFM tip propagates only up to a distance of $\sim$500 nm [18] laterally while an accurate study of the bandgap, the effective mass, the Schottky barrier height (SBH) at the contacts, etc, requires the application of strain uniformly across the sample. Moreover, the tip-induced modifications to the transport properties also need to be addressed. Bending the substrate in a direction normal to its plane exploiting a single or a double-ended beam geometry [23, 24] and, stretching or compressing the substrate [25] are the most common methods used to apply uniform strain on TMDCs. Piezoelectric and piezoresistive effects [23, 25] have been observed on MoS$_2$ by these methods.

MoS$_2$ forms Schottky junctions with commonly used contact materials making the realization of efficient electrical contacts challenging [27–30]. Though the SBH is predicted to depend on the work-function of the metal and the electron affinity of the semiconductor [31], the dominant Fermi-level pinning mechanism [30, 32] makes the SBH reduction utilizing lower work-function metals ineffective [28, 33]. It has been shown by density functional calculations that the in-plane strain can lower the SBH and the contact resistance mediated by bandgap reduction and improved hybridization of electronic orbitals of the MoS$_2$ and the contact metal [34]. A complete study requires addressing both the changes to the electronic properties such as the bandgap and the behavior of the electrical contacts against uniform strain, which forms the major focus of this report.

In this work the effect of uniaxial strain on the bandgap, electrical conductivity and the SBH at the electrical contacts of bi- and few-layered MoS$_2$ devices is explored. We utilize a three-point bending apparatus to exert the strain. The strain induced on to the sample is characterized using Raman spectroscopy. The modulation of the bandgap with strain is quantified using PL spectroscopy. Low-noise electrical transport measurements on the devices reveal a high piezoresistivity with a gauge factor (GF) larger than that for Silicon [35]. Our observations show both the conductivity of the material and also the SBH at the electrical contacts undergoes a substantial reduction with strain. Our studies reveal that strain is an important handle to tune the electrical properties of the material and also can be used to engineer high-efficiency electrical contacts for future device engineering.

2. Methods

Commercially procured bulk MoS$_2$ crystals (SPI supplies) are mechanically exfoliated with the help of an adhesive tape and subsequently transferred on to a 0.5 mm thick flexible substrate (polyimide, Sigma-Aldrich) using the PDMS assisted dry transfer technique [36]. A uniaxial strain is transferred onto the MoS$_2$ flake by bending the flexible substrate normal to its plane exploiting a micrometre-controlled three-point bending apparatus. A schematic drawing of the bending mechanism is shown in figure 1(a). From the thickness $2t$ and the bending radius $\rho$ of the substrate, as shown in figure 1(b), we calculate the strain induced on the substrate and the sample, $\varepsilon = t/\rho$ [11]. A plot of the strain vs the number of turns on the micrometre screw is shown in the figure 1(c). Raman and PL spectroscopy are performed using Horiba Xplora Plus spectrometer with a 532 nm excitation with 2400 lines mm$^{-1}$ grating and 600 lines mm$^{-1}$ respectively. Electrical contacts to the flakes are defined using photo-lithography followed by Cr (2 nm)/Au (30 nm) metallization.

3. Results & discussion

Tensile strain results in the softening of the lattice spring constants and in-turn the crystal phonon modes. Raman spectroscopy is an effective tool to study the phonon modes and the variation in the Raman shifts has been used to study and characterize strain in MoS$_2$ [10, 12, 37]. Here, we utilize the Raman spectroscopy to identify and characterize the strain exerted on to the MoS$_2$ flake from the bending apparatus. Raman spectra representing the prominent vibrational modes, E$_{1g}^{\text{2}}$ and A$_{1g}$, for the mono- and the bi-layer MoS$_2$ flakes for various percentage of applied tensile strain are shown in figures 1(d) and (e) respectively. The inset to figure 1(d) illustrates the vibrational modes; the E$_{1g}^{\text{2}}$ mode $\sim$380 cm$^{-1}$ corresponds to the in-plane vibrational mode while the A$_{1g}$ mode $\sim$400 cm$^{-1}$ corresponds to the out-of-plane vibrational mode. Raman spectra is also used to probe the number of layers in the MoS$_2$ flake in the few-layer (<4 layers) limit [11, 38–40]. In this work the primary estimation of the thickness of MoS$_2$ flake is performed using optical contrast [13, 41] and the Raman spectra prior to the application of strain on to the sample [red open-circles in figures 1(d) and (e)] is used to confirm the number of layers. We obtain a shift of $\sim$19.2 cm$^{-1}$ and $\sim$22.03 cm$^{-1}$ for the unstrained mono- and bi-layer samples respectively; these values are consistent with the reported shifts for the mono- and the bi-layer MoS$_2$ samples on similar substrates [11, 38, 39]. (See supporting information S1 (stacks.iop.org/Nano/31/275703/mmedia)). The optical images of the corresponding MoS$_2$ flakes are shown in supporting information S2.

From figures 1(d) and (e), we observe a redshift in the E$_{1g}^{\text{2}}$ peak corresponding to the in-plane vibrations of the Mo and S atoms indicating a softening of the mode. Figures 1(f) and (g) show the variation in the Raman shifts for both the E$_{1g}^{\text{2}}$ and the A$_{1g}$ modes for the mono- and the bi-layer samples as a function...
Figure 1. (a) Schematic representation of the bending apparatus. A flexible polyimide sheet of width 2 cm and thickness 0.5 mm is clamped between two knife edges with a distance of 4.5 cm between them. The centre of the substrate is deflected to apply the strain using a calibrated micrometre screw. (b) Geometrical representation of the strained substrate with thickness. The yellow arc represents the substrate with thickness 2t, length l and ρ is the radius of curvature. The convex side of the bend substrate provides tensile strain. The Radius of Curvature ρ is calculated using the formula $(ρ − t)^2 = ρ^2 + (l/2)^2$. (c) Plot of strain vs number of turns. (d) Raman spectra for the monolayer MoS$_2$ sample for various levels of applied strain levels. Inset: illustration of the $E_{12g}$ and $A_{1g}$ vibrational modes. (e) Raman spectra of the bilayer MoS$_2$ sample for various strains. (f) & (g) Variation in the Raman shifts the $E_{12g}$ and the $A_{1g}$, for the monolayer and the bi-layer samples respectively as function of strain.

Strain has shown to have a profound effect on the bandgap of TMDC materials and, PL spectroscopy has been used monitor and characterize the strain induced bandgap variation in these systems [11, 13, 39]. In this session, we characterize the strain induced bandgap change on our mono- and the bilayer samples using PL spectroscopy. The results thus obtained are used to get more insights into and analyse the electrical transport data presented in the subsequent session. Figures 2(a) and (b) show representative PL spectra taken as a function of applied tensile strain on mono- and bi-layer MoS$_2$ samples respectively. Optical images of the corresponding mono- and bi-layer samples are given in the supporting information S2. The monolayer MoS$_2$ is characterized by a single PL emission peak at $\sim$1.84 eV corresponding to the direct inter-band transition at the K—K point in the Brillouin zone [42]. In contrast, the bilayer MoS$_2$
flake exhibits two peaks; the A peak at $\sim 1.84$ eV corresponding to the direct transition at K—K point while the I-peak at $\sim 1.56$ eV is due to the indirect transition at the K—I point in the Brillouin zone [43]. We observe a red-shift in the PL peaks of both the mono- and the bi-layer samples signalling a reduction in the bandgap with strain. The bandgap vs applied strain extracted from the PL spectra for the mono- and the bi-layer samples are shown in figures 2(c) and (d) respectively. The PL peak for the monolayer shows a red-shift of $\sim 78 \pm 4$ meV/% strain. For the bilayer, the A and the I peak show red-shifts of $\sim 34 \pm 3$ meV/% strain and $\sim 155 \pm 11$ meV/% strain respectively. Our observation of strain-induced reduction in the bandgap is consistent with the existing theoretical and experimental reports [11–13, 39] confirming reduction in the MoS$_2$ bandgap and an effective transfer of strain on to the MoS$_2$ samples from the bending apparatus.

Any variation in the electrical properties such as the bandgap, the effective mass and the carrier concentration need to reflect in the electrical transport characteristics of the material. Here, we investigate the influence of strain on the electrical transport characteristics using a bi-layer MoS$_2$ sample. Transport characterization conducted on a few-layered sample exhibiting similar behaviour is shown in the supporting information S3. Our monolayer MoS$_2$ samples on the polyimide substrate showed insulating behaviour. Surface roughness of substrate are shown to play a crucial role in reducing the conductivity of MoS$_2$ [44, 45] and, we believe that the substantial RMS surface roughness of the substrate, $\sim 2–4$ nm (see supporting information S4) made the samples nonconducting. Transport characterization conducted on a few-layered sample exhibiting similar behaviour is shown in the supporting information S3. Figure 3(a), open circles, show I–V characteristics of the bi-layer MoS$_2$ device for various percentage of applied strain. The inset shows the optical image of the device. The I–V characteristics show an enhancement in the current through the device with strain. The resistance of the device extracted from the linear low-bias regime of the I–V characteristics, shown in red-circles in figure 3(b), shows a substantial reduction with strain. For the strain levels applied in this experiment, the bandgap change induced resistance variation dominates and the contribution from other effects such as the variation in the effective-mass are negligible [17, 46]. For a semiconductor, the bandgap variation has an exponential influence on the carrier concentration and in turn the resistance [47, 48]. We estimate the change in the bandgap with strain, $\frac{\partial E_g}{\partial \varepsilon}$, from the variation of the resistance with strain, shown in figure 3(b) red-circles, using the relation [48] $R = R_0 \exp \left( -\frac{\partial E_g}{\partial \varepsilon} \frac{e^2}{2\pi \hbar} \varepsilon \right)$. The $\frac{\partial E_g}{\partial \varepsilon}$ $\sim 195$ m eV/%-strain thus obtained is considerably higher than $\sim 155$ meV/%-strain, that we extracted from the PL spectroscopy for our bi-layer sample, and also exceeds those reported elsewhere [11, 18]. Unlike the PL spectroscopy, the measurements discussed here probes not only the material but also the behaviour of the electrical contacts.

Metal-semiconductor contacts seldom yield Ohmic behaviour and most of the commonly used metals yield Schottky contacts with MoS$_2$ [49]. The nature and behaviour of the contacts depend on the metal work-function, electron affinity of the semiconductor and also on the nature and density of the interface states [31].
Figure 3. I–V characteristics. (a) Representative I–V characteristics for the bilayer MoS$_2$ device vs strain. Inset—optical image of the device. (b) Red open-circles show resistance vs strain extracted from the low-bias regime of the I–V characteristics and red-line shows an exponential fit giving $\frac{\partial E_g}{\partial \varepsilon} \sim 195$ m eV/% strain. The inset shows the circuit model used for the device simulation. Blue open-squares represent the resistance extracted from the simulation for different strains. (c) Change in $\Delta R/R_0$ (red open-triangles) and the bandgap $\Delta E_g$ (blue open-circles) with strain. (d) Change in SBH the source and the drain contacts, $\varphi$, with strain. (e) Band-diagram illustrating the prominent effects of strain on transport in the device. Strain causes a reduction in the bandgap resulting in (i) more carriers and reduction in the material resistance and (ii) the SBH—facilitating an enhancement in the transport.

The I–V characteristics shown in figure 3(a) have two regimes; (i) a nearly linear low-bias regime and, (ii) a current-saturation regime at higher source-drain bias. This is atypical of a device with ohmic contacts and, the presence of a saturation region suggests the formation of potential barriers at the source and the drain contacts [50, 51]. The saturation current has an exponential dependence on the barrier [31, 47]. The rise in both the low-bias device conductance and the saturation current suggest that not only the device conductance but also the potential barriers formed at the electrical contact regions are modified by the strain.

To segregate the behaviour of the contacts and the material against strain, we analyse the data with the circuit model [22, 50, 52] shown in the inset to figure 3(b). The model consists of two Schottky diodes connected back-to-back representing the electrical contacts while the MoS$_2$ flake is modelled as a variable resistor connected between the diodes. Both the transport through the diodes and the resistor are influenced by the applied strain.

The I–V characteristics of the Schottky diode representing contacts is given by [47]

$$I_i(V_i) = I_{0,i} \exp \left( \frac{qV_i}{\varepsilon_i k_B T} \right) \left[ 1 - \exp \left( \frac{qV_i}{k_B T} \right) \right]$$

where, $I_{0,i}$ is the reverse saturation current, $A_e$ is the contact area of the metal—semiconductor junction, $A^*$ is the Richardson’s constant, $V_i$ is the voltage drop across each junction, $\varepsilon_i$ is the ideality factor, $q$ is the electronic charge, $k_B$ the Boltzmann constant and, $T$ is the temperature. The area of contact for the electrodes, $\sim$400 $\mu$m$^2$, are determined from the optical image of the device. The initial guess to the resistance of the MoS$_2$ flake for each strain level is obtained from the linear part of the low-bias I–V characteristics. The initial guess to the $I_{0,i}$ are obtained from the current at the saturation region of the I–V characteristics. From the fit to the I–V traces corresponding to different strain values we extract the reverse saturation current, the ideality factor and the resistance of the MoS$_2$ flake.
From the reverse saturation curve we extract the refined SBH using the relation \([22, 47]\), \(\phi_1 = \left(\frac{h}{e}\right)\ln\left(\frac{\Delta \phi_{SBH}(c)}{k_B T}\right)\). More details on the fitting procedure is given in the supporting information S5.

Cr/Au contacts on MoS\(_2\) has been shown to exhibit Schottky behaviour and, the carrier injection across the barrier is governed by thermionic emission mechanism at room temperature [29, 47, 50]. Considering this, we have restricted the ideality factor \(n_i\) of the junctions in the vicinity of 1. The goodness-of-fit is assessed by inspecting the coefficient-of-determination \(R^2\) vs the \(n_i\) and reverse saturation current \(I_{03}\) as shown in the supporting information S6 and, we obtain a refined \(n_i\) of 1.04, 1.01 for the source and the drain barriers. The fits we obtained, shown in solid lines in figure 3(a), exhibit excellent agreement with the experimental I–V characteristics. The resistance of the MoS\(_2\) as a function of strain, extracted from the fit is shown in the blue-squares in figure 3(b). Assuming an exponential dependence of resistance to the strain \([18, 48]\) we obtain \(\frac{\Delta R}{R_0} \sim 147\) meV/%-strain for the bi-layer sample, agreeing well with the PL data shown in figure 2(d) and also concurs with other spectroscopic studies [11]. The variation in the device bandgap with strain extracted from the simulated resistance values is shown in blue open-circles and, the change in device resistance \(\Delta R\) vs strain is shown in red open-triangles in figure 3(c), where \(R_0\) is the resistance of the unstrained sample and \(\Delta R\) is the change in resistance. From the resistance change we extract a piezoresistive gauge factor \(GF = \frac{\Delta R}{R_0} / \phi \sim 321\) for the bi-layer device where \(\phi\) is the strain on the sample. The GF for our bi-layer sample is substantially larger than that reported for Silicon [35].

Figure 3(d) shows the reduction in the SBH \(\phi\) obtained from the fit for the source and the drain barriers. The bilayer sample shows a reduction of \(~55\) meV/% strain in SBH. The few-layer sample shown in supporting information S3 shows a reduction of 40 meV/% strain. We infer that the major contribution to the SBH is the strain-induced bandgap reduction. The tensile strain results in lowering the conduction band minima at the \(\Gamma\)-point of the Brillouin zone, which will result in the reduction of the effective Schottky barrier at the source and the drain contacts [11, 34]. Figure 3(e) illustrates the two dominant mechanisms contributing to the conductance modification (i) the bandgap induced change in the material resistance via carrier concentration enhancement (ii) the SBH reduction at the contacts, both will result in the enhancement of device conductance. Apart from the bandgap-change induced reduction in SBH, the contact-metal layer can also make better hybridization with the MoS\(_2\) orbitals as a result of the reduction in the interlayer distance and weakening of the \(dz^2\) orbitals [34]. The actual SBH in MoS\(_2\)-metal contacts are reported to be different from those predicted by the Schottky-Mott rule [31] and is mostly decided by Fermi-level pinning mechanisms [30, 32]. In this scenario the ability to tune the SBH continuously by strain and engineer high efficiency electrical contacts will provide a major boost to the device engineering on MoS\(_2\).

### 4. Conclusions

Strain can play a lead role in tuning the electrical properties of MoS\(_2\) and other 2D materials for device engineering. There are only a few studies on probing the transport properties under the action of uniform strain across the sample. This manuscript is devoted to understand the effect of uniaxial in-plane tensile-strain on the electrical properties of MoS\(_2\) samples. A three-point bending apparatus is used to induce uniform strain across the sample. Raman spectroscopy is used to infer and calibrate the strain on the lattice. The in-plane Raman modes showed a red-shift signaling softening of the in-plane phonon modes while the out-of-plane Raman modes remained relatively unaffected by the strain. A red-shift in the PL emission peaks corresponding to both the direct and indirect transitions is observed signaling a reduction in the bandgap with strain. Electrical transport studies with two-probe geometry are conducted on bi- and few-layered samples. Modulation of electrical conductivity and SBH at the electrical contacts against strain are the main themes of the manuscript. We observed a strong enhancement in the device conductance with strain for both bi- and few-layered devices. Our bi-layer device exhibited a strong piezoresitive a GF \(~321\) superior to that of Silicon [35] while the few-layered device exhibited a GF \(~48\). In addition to the reduction in the flake resistance, we find a reduction in the source and the drain Schottky barriers heights. The tensile strain results in a continuous lowering of the conduction-band minima at the \(\Gamma\)-point of the Brillouin zone inducing a corresponding lowering of the Schottky barrier height [11, 34]. Major contribution to the MoS\(_2\) density of states at Fermi-level arise from the \(d\)-orbital of Mo atoms [34]. The increased overlap of the metal atom orbitals as a result of strain with these \(d\)-orbitals can also play a role in the reduction of the contact barrier heights. Resistance change against strain has a substantial contribution from the Schottky barrier modulation also. This effect can be used for engineering high-efficient, nanoscale strain sensors utilizing this effect. 2D systems offer a versatile platform to apply substantial amount of strain in a reversible manner. Mobility and conductivity enhancements, bandgap and SBH reductions with strain can bring a new perspective in device engineering on MoS\(_2\), making it a potential platform for future microelectronics and straintronics technologies.

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Author contributions

MT conceived the problem. APJ and AT prepared and measured the devices and analyzed the data. MT and APJ prepared the manuscript.

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