Charge neutral MoS2 field effect transistors through oxygen plasma treatment
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Lithographically fabricated MoS₂ field effect transistors suffer from several critical imperfections, including low sub-threshold swings, large turn-on gate voltages ($V_T$), and wide device-to-device variability. The large magnitude and variability of $V_T$ stems from unclean interfaces, trapped charges in the underlying substrate, and sulfur vacancies created during the mechanical exfoliation process. In this study, we demonstrate a simple and reliable oxygen plasma treatment, which mitigates the effects of unintentional doping created by surface defect sites, such as S vacancies, and surface contamination. This plasma treatment restores charge neutrality to the MoS₂ and shifts the threshold turn-on voltage towards 0 V. Out of the 10 devices measured, all exhibit a shift of the FET turn-on voltage from an average of $-18$ V to $-2$ V. The oxygen plasma treatment passivates these defects, which reduces surface scattering, causing increased mobility and improved sub-threshold swing. For as-prepared devices with low mobilities (~0.01 cm²/V s), we observe up to a 190-fold increase in mobility after exposure to the oxygen plasma. Perhaps the most important aspect of this oxygen plasma treatment is that it reduces the device-to-device variability, which is a crucial factor in realizing any practical application of these devices. Currently, lithographically fabricated MoS₂ transistors suffer from two imperfections detrimental to low power CMOS applications: (1) the large turn-on gate voltages ($V_T$) needed to switch the transistor state and (2) severely degraded sub-threshold swing. The origin of both these imperfections can be traced back to the high density of interface trap charges and substrate interactions. In particular, the large turn-on voltages in MoS₂-based FETs arise due to sulphur vacancies, which are known to inherently n-dope the MoS₂ channel, moving it away from charge neutrality.

In the work presented here, we show that the controlled exposure to a downstream (remotely generated) oxygen plasma dramatically improves the measured transport ($I$–$V$) characteristics. The turn-on voltage ($V_T$) moves closer to zero gate voltage due to screening of charged impurities, and a reduction of the sub-threshold voltage swing is observed. Typically, devices fabricated in this study exhibit carrier mobilities (~1–10 cm²/V s) near typically reported values in literature. On occasion however, a fabricated device may yield rather low mobility. In such rare cases, where the device mobility is remarkably low (~0.01 cm²/V s), likely limited by impurity scattering, we see a dramatic (up to

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190-fold) improvement in carrier mobilities. These lower mobility, as-fabricated devices tend to have a layer thickness <5 layers, making them more susceptible to scattering from surface impurities. After plasma treatment, these defective devices are also restored to moderate device performance.

**EXPERIMENTAL DETAILS**

We mechanically exfoliate MoS$_2$ onto a pre-cleaned Si/SiO$_2$ wafer. Few-layer (3–15 layers) flakes are located using gated MoS$_2$ FET device. (c) A log-linear plot of the source-drain current as a function of the applied gate voltage.

FIG. 1. (a) Schematic diagram and (b) optical microscope image of a back-gated MoS$_2$ FET device. (c) A log-linear plot of the source-drain current as a function of the applied gate voltage.

In Figure 1(c), the source-drain current ($I_D$) is plotted on a log scale as a function of the applied gate voltage $V_G$. The device shows n-type conductance at positive applied gate voltages, and is turned off at negative gate voltages. The devices also exhibit a linear dependence of current on the applied source-drain bias voltages (as shown in Figure S1 of the supplementary material), which indicates that Ohmic contacts are made with the metal contacts. This particular device shows a shift of the turn-on voltage from $-19.4$ V to $-0.9$ V after exposure to the remote oxygen plasma. For the 10 FET devices measured in this study, we observe a shift in the turn-on voltage from an average of $-18.5$ V to $-2$ V, as summarized in Table I. As mentioned before, the large magnitude of the turn-on voltage arises due to the abundance of sulphur vacancies in mechanically exfoliated MoS$_2$, and this has been reported previously by various groups (see Table S1). The plasma is comprised of charged ions and neutral O radicals.

**RESULTS AND DISCUSSIONS**

### Table I. The FET carrier mobilities ($\mu$) and turn-on voltages ($V_{T}$) for 10 MoS$_2$ FET devices before and after remote oxygen plasma treatment with layer thickness more than five layers.

<table>
<thead>
<tr>
<th>Device</th>
<th>$V_{T}$ (Pre) (V)</th>
<th>$V_{T}$ (Post) (V)</th>
<th>$\Delta V_T$ (V)</th>
<th>$\mu_{pre}$ (cm$^2$/V s)</th>
<th>$\mu_{post}$ (cm$^2$/V s)</th>
<th>$\mu_{post}/\mu_{pre}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$-14$</td>
<td>$-3$</td>
<td>$11$</td>
<td>$51.4$</td>
<td>$21.79$</td>
<td>0.4</td>
</tr>
<tr>
<td>2</td>
<td>$-20$</td>
<td>$6$</td>
<td>$26$</td>
<td>$22.1$</td>
<td>$5.72$</td>
<td>0.3</td>
</tr>
<tr>
<td>3</td>
<td>$-16$</td>
<td>$-12$</td>
<td>$4$</td>
<td>$45.5$</td>
<td>$24.69$</td>
<td>0.5</td>
</tr>
<tr>
<td>4</td>
<td>$-17$</td>
<td>$0.5$</td>
<td>$17.5$</td>
<td>$29.6$</td>
<td>$5.51$</td>
<td>0.2</td>
</tr>
<tr>
<td>5</td>
<td>$-19.5$</td>
<td>$-6$</td>
<td>$13.5$</td>
<td>$38.9$</td>
<td>$13.38$</td>
<td>0.3</td>
</tr>
<tr>
<td>6</td>
<td>$-18$</td>
<td>$2$</td>
<td>$20$</td>
<td>$19.8$</td>
<td>$16.64$</td>
<td>0.8</td>
</tr>
<tr>
<td>7</td>
<td>$-18$</td>
<td>$-2$</td>
<td>$16$</td>
<td>$25.4$</td>
<td>$21.00$</td>
<td>0.8</td>
</tr>
<tr>
<td>8</td>
<td>$-22$</td>
<td>$-6$</td>
<td>$16$</td>
<td>$26.6$</td>
<td>$12.50$</td>
<td>0.5</td>
</tr>
<tr>
<td>9</td>
<td>$-20$</td>
<td>$-3$</td>
<td>$17$</td>
<td>$25.5$</td>
<td>$15.65$</td>
<td>0.6</td>
</tr>
<tr>
<td>10</td>
<td>$-15$</td>
<td>$0$</td>
<td>$15$</td>
<td>$18.0$</td>
<td>$14.31$</td>
<td>0.8</td>
</tr>
<tr>
<td></td>
<td>$-17.95$</td>
<td>$-2.35$</td>
<td>$15.6$</td>
<td>$30.3$</td>
<td>$15.1$</td>
<td>0.53</td>
</tr>
</tbody>
</table>

### Table II. The FET carrier mobilities ($\mu$) and turn-on voltages ($V_{T}$) for 3 MoS$_2$ FET devices before and after remote oxygen plasma treatment with layer thickness less than five layers.

<table>
<thead>
<tr>
<th>Device</th>
<th>$V_{T}$ (Pre) (V)</th>
<th>$V_{T}$ (Post) (V)</th>
<th>$\Delta V_T$ (V)</th>
<th>$\mu_{pre}$ (cm$^2$/V s)</th>
<th>$\mu_{post}$ (cm$^2$/V s)</th>
<th>$\mu_{post}/\mu_{pre}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$-20$</td>
<td>$-4$</td>
<td>$16$</td>
<td>$0.017$</td>
<td>$3.21$</td>
<td>192</td>
</tr>
<tr>
<td>2</td>
<td>$-19$</td>
<td>$4$</td>
<td>$23$</td>
<td>$0.001$</td>
<td>$0.02$</td>
<td>15.0</td>
</tr>
<tr>
<td>3</td>
<td>$-18$</td>
<td>$1.5$</td>
<td>$19.5$</td>
<td>$0.002$</td>
<td>$0.03$</td>
<td>18.4</td>
</tr>
<tr>
<td>Avg</td>
<td>$-19$</td>
<td>$0.5$</td>
<td>$19.5$</td>
<td>$0.007$</td>
<td>$1.08$</td>
<td>75.1</td>
</tr>
</tbody>
</table>
radicals that bind readily to S-vacancies and form covalent bonds with the Mo atoms, since O has the same valence number as S, thus mitigating the doping effects associated with the vacancy. In fact, O has a slightly higher electronegativity than S and will bind more strongly than a corresponding S atom.

It should be noted that the results obtained here using a remotely generated oxygen plasma stand in contrast to those obtained utilizing a conventional local O2 plasma. Islam et al. reported the destructive effects of oxygen plasma exposure to the surface of MoS2, showing a severe degradation of the FET mobility after just a few seconds of exposure to the locally generated oxygen plasma.20 In contrast, using the gentler, remotely generated O2 plasma, we typically observe only a slight degradation of carrier mobility, despite using considerably longer exposure times (approximately 3 min). However, sample degradation is a more significant factor for monolayer and bilayer flakes.

Our initial intent was to use this oxygen plasma to selectively remove the surface residue created during the lithographic process. The AFM images in Figure 2 show an exfoliated MoS2 flake before and after oxygen plasma treatment. Before plasma treatment, this flake exhibits substantial surface residues left over from the lithographic processes (i.e., ethyl lactate-6, PMMA 950 C2, acetone, isopropanol). It is also possible that the tape residue from the mechanical exfoliation process also contributes to this unwanted surface contamination on the MoS2 flake. After plasma cleaning, a majority of this residue is removed, without damaging the MoS2 flake itself.

Also evident from the $I$–$V$ characteristics of Figure 1(c), is the reduction of the sub-threshold gate-voltage swing ($S$) after treatment with oxygen plasma. The sub-threshold swing is defined as the change in gate voltage required to reduce the current by one decade, i.e., $S = \log(10) \cdot \left( \frac{dV_G}{d(\log(ID))} \right)$, where the derivative $dV_G/d(\log(ID))$ is taken at the onset of current. In conventional FETs, sub-threshold transport is dominated by carrier diffusion, and the sub-threshold swing is largely determined by the MOS capacitance, given by the expression $S = \log(10) \cdot \left( \frac{dV_G}{d(\log(ID))} \right) \approx \frac{q}{C} \log(10) \left( 1 + \frac{C + C_D}{C + C_{it}} \right)$. Here, $C$ is the oxide (back-gate) capacitance, $C_D$ is the depletion layer capacitance, and $C_{it}$ is the capacitance associated with the interface trap charges. From the data in Figure 1, we obtain values of 5.26 V/dec and 3.05 V/dec for the sub-threshold swing before and after oxygen plasma treatment, respectively. This reduction in sub-threshold swing too may be understood as a consequence of the reduction in interfacial charge density, arising primarily due to Sulfur vacancies. The two-fold improvement in sub-threshold voltage swing is important for efficient device operation, as it enables efficient switching the FET state (on/off) at low voltages, and hence minimizes power dissipation.
The field effect mobility of our devices is obtained from the transconductance data shown in Figure 3 using the expression $\mu = \frac{dI_d}{dV_g} \frac{L}{W C_{24}}$, where $W$ and $L$ are the channel width and length, respectively, $C$ is the back gate capacitance, $V_g$ is the gate voltage, $V_{DS}$ is the bias (source-drain) voltage, and $I_d$ is the current flowing through the channel. Here, the derivative $dI_d/dV_g$ is taken at the steepest point on the $I$–$V$ curve from Figure 3. Although this observed device mobility can depend on various factors, such as contact resistance, and surface interactions, looking at Table I, we observe two distinct trends in the change in mobility due to plasma treatment. For devices in Table I, showing moderate carrier mobilities (~10 to 100 cm$^2$/V s), the mobility is typically slightly reduced (by approximately a factor of 2) due to the plasma treatment, indicating that exposure to oxygen plasma does introduce some disorder in the MoS$_2$ lattice. However, devices with relatively poor carrier mobilities (~0.001 cm$^2$/V s) in Table II show significantly enhanced carrier mobilities after plasma treatment. These devices also happened to be fabricated using thinner flakes of MoS$_2$, where surface scattering is expected to play a more significant role. The low mobilities in these devices also indicate they suffer from strong extrinsic scattering effects, such as the presence of tape residue on the MoS$_2$ surface, rendering them useless. On average, we observe a 75-fold improvement in device mobility for such “damaged” devices, as summarized in Table II. We attribute this dichotomy in trends to the competing effects of oxygen plasma exposure on the MoS$_2$ surface. While removal of surface impurities from the surface of “damaged” flakes reduces impurity scattering and improves mobility, the direct bombardment of the MoS$_2$ flake by plasma creates disorder, and degrades the carrier mobility in thicker flakes where surface effects are not the dominant scattering mechanism. Further, the increase in the “off-state” current, as seen in Figure 1(c), is also likely a consequence of an increased density of electronic states within the MoS$_2$ band gap. Here, the use of a remotely generated plasma minimizes the latter effect, allowing the removal of impurities, without significantly increasing the defect density. However, the use of longer etching times (over 4 min), or larger RF powers, typically does damage the MoS$_2$ flake itself, with thinner flakes being more susceptible to degradation.

While the same processing steps were used to fabricate all of the devices in this study, we found that the lowest mobility transistors ($\mu < 0.1$ cm$^2$/V s) show signs of organic residue on the surface, as shown in the AFM image in Figure 2. This indicates that the mobility in these cases is not limited by intrinsic processes (such as defect or phonon scattering in the MoS$_2$), but rather scattering from the extrinsic contaminant species. Thus, these devices show enhanced carrier mobilities due to the removal of surface residue through plasma treatment. It is known that MoS$_2$ transistors are also afflicted by surface contaminants and charged impurities, which dope the material n-type$^{19}$ and move the turn-on voltage away from zero applied gate voltage. The oxygen plasma consists of negatively charged ions of oxygen and nitrogen, as well as charge neutral free radicals. We believe that these ions and radicals from the plasma bind preferentially to the S-vacancy sites in MoS$_2$, mitigating the doping effects associated with the vacancy. This reduces the net charge on the defect sites, thereby reducing their effect on the turn-on voltage. The optimal etch time of 2–3 min corresponds to the amount of time needed to “saturate” all the charged impurity sites. Any further increase in exposure time does not lower the turn-on voltage.

For the particular device shown in Figure 3(a), which was seemingly damaged upon fabrication, $W = L = 4$ µm and $C = 11.5$ nF/cm$^2$ (for a 300 nm SiO$_2$ back gate). The estimated mobility of the as-fabricated FET device is 0.017 cm$^2$/V s, which is considerably lower than the typically reported values in literature. However, after treatment with the remote oxygen plasma, the carrier mobility is found to increase to 3.21 cm$^2$/V s, which is comparable to typical values reported in literature.$^4$ While most reports in the literature focus on clean FET devices showing higher mobility, our work demonstrates a method to improve even the very worst fabricated devices. In contrast, Figure 3(b) shows the $I$–$V_g$ characteristics of another MoS$_2$ device not limited by extrinsic scattering mechanisms. Here, the carrier mobility drops by 16% from 19.8 cm$^2$/V s to 16.6 cm$^2$/V s after the 3 min oxygen plasma exposure.

Interestingly, we observe that exposure to oxygen plasma is also accompanied by an increase in the hysteresis of the $I$–$V_g$ characteristics, as shown in Figure S3 of the supplementary material. Previous studies on organic transistors,$^{22}$ inorganic semiconductors,$^{23}$ as well as carbon nanotubes$^{24}$ have shown that hysteresis is a consequence of a dynamic gating of the channel due to mobile surface charges, which may move upon the application of an external field, thereby changing the charge neutrality point. The increased hysteresis is similarly attributed to the dynamic gating caused by adsorption of oxygen plasma species onto the MoS$_2$ surface. As reported in our previous work, this oxygen plasma treatment improved the luminescence efficiency of few-layer MoS$_2$ by up to 20-fold due to an indirect-to-direct bandgap transition.$^{17,25,26}$ In addition, the photoluminescence linewidth becomes substantially narrower after plasma treatment, as shown in Figure 4. The photoluminescence spectra allow one to determine the relative lifetime of the excitons, whereas transport measurements give a relative measure of the free carrier lifetimes. In transition metal dichalcogenides, there is a dramatic change in the band structure of the materials, as layer thickness is increased above one monolayer. While the most pronounced aspect of this change is the transition to an indirect gap semiconductor in multilayer TMDCs, this change also has a bearing on the selection rules for allowed intervalley scattering processes in monolayer MoS$_2$. Hence, the PL spectral linewidth is found to be narrower in monolayer MoS$_2$ than in multilayer MoS$_2$. Since the exposure to oxygen plasma is also shown to decouple the individual layers, leading to a monolayer-like bandstructure, a similar effect is seen in our spectra. This direct gap transition is highly desirable for applications in optoelectronic devices and, coupled with the improved FET device performance, could pave the way for next generation TMDC based devices. It should be noted that the chemical reaction...
induced by the O-plasma treatment is permanent and stable, and these devices do not revert to their original \( I-V \) characteristics over time. The plasma treatment, however, does result in an interlayer decoupling, and some delamination of the MoS\(_2\) is observed when stored under ambient conditions over the span of a few weeks. Therefore, some strategy for hermetic sealing will have to be implemented in order to overcome this instability in practical device applications.

**CONCLUSIONS**

In summary, we demonstrate a reliable and scalable method using an oxygen plasma treatment to dramatically reduce the turn-on voltage required for switching operation in MoS\(_2\) transistors and simultaneously improve the subthreshold swing of these devices; both are key parameters for enabling low-power electronic devices and sensing applications. The key novelty in this work lies in bringing MoS\(_2\) FETs close to charge neutrality, presumably through the passivation of charged Sulfur vacancies, which typically \( n \)-dope exfoliated MoS\(_2\).

Estimating the relative importance of various scattering mechanisms (e.g., surface impurities, defects, and phonons) requires a more detailed temperature dependent study of the transport characteristics. However, in our work we observe the following trends:

1. Thin MoS\(_2\) flakes (1–2 layers) are more easily damaged by the oxygen plasma treatment. It is possible to still achieve improved FET performance with the oxygen plasma treatment, but the exposure time and pressure need to be controlled more carefully.

2. Devices with thicker flakes (3–5 layers) are more robust to oxygen plasma exposure, and yet are significantly impacted by the presence of surface impurities. These are the devices that yield both turn-on gate voltages closer to 0 V, as well as increase in carrier mobility, especially in cases where the device performance is limited by extrinsic scattering mechanisms.

3. Thick MoS\(_2\) flakes (>5 layers) also exhibit turn-on gate voltages far from 0 V. However, their device mobilities are less prone to scattering from surface impurities. Hence, after exposure to the remote oxygen plasma, we see a shift of the charge neutrality point toward zero applied gate voltage, but moderate reduction of carrier mobility is also observed.

Lithographically fabricated FETs also suffer from non-uniformities and huge variability in device-to-device performance, which is greatly reduced upon exposure to the remote oxygen plasma. Our results also shed light on the role of defect and impurity scattering mechanisms limiting device mobilities in MoS\(_2\) transistors. While this result is not meant to compete with other methodologies used to make high mobility FET devices, such as using suspended MoS\(_2\) or MoS\(_2\) sandwiched between two flakes of boron nitride, it provides a scalable route for creating moderate-mobility MoS\(_2\) FETs with lower subthreshold swing and turn-on voltages, while significantly reducing the device-to-device variability.

**SUPPLEMENTARY MATERIAL**

See supplementary material for additional data regarding the influence of the metal-semiconductor contact, as well as a comparison with results from existing literature.

**ACKNOWLEDGMENTS**

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FIG. 4. Enhanced photoluminescence emission from a four layer thick MoS\(_2\) after exposure to remotely generated oxygen plasma.