Modulating Electronic Properties of Monolayer MoS₂ via Electron-Withdrawing Functional Groups of Graphene Oxide

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ABSTRACT: Modulation of the carrier concentration and electronic type of monolayer (1L) MoS₂ is highly important for applications in logic circuits, solar cells, and light-emitting diodes. Here, we demonstrate the tuning of the electronic properties of large-area 1L-MoS₂ using graphene oxide (GO). GO sheets are well-known as hole injection layers since they contain electron-withdrawing groups such as carboxyl, hydroxyl, and epoxy. The optical and electronic properties of GO-treated 1L-MoS₂ are dramatically changed. The photoluminescence intensity of GO-treated 1L-MoS₂ is increased by more than 470% compared to the pristine sample because of the increase in neutral exciton contribution. In addition, the A₁g peak in Raman spectra shifts considerably, revealing that GO treatment led to the formation of p-type doped 1L-MoS₂. Moreover, the current vs voltage (I−V) curves of GO-coated 1L-MoS₂ field effect transistors show that the electron concentration of 1L-MoS₂ is significantly lower in comparison with pristine 1L-MoS₂. Current rectification is also observed from the I−V curve of the lateral diode structure with 1L-MoS₂ and 1L-MoS₂/GO, indicating that the electronic structure of MoS₂ is significantly modulated by the electron-withdrawing functional group of GO.

KEYWORDS: monolayer MoS₂, graphene oxide, electron-withdrawing effect, photoluminescence, Raman spectroscopy
Figure 1. Sample preparation. (a) Quartz plate used as a substrate, GO coating on half the area of the quartz plate, and transfer of 1L-MoS₂ grown by CVD onto the GO-coated quartz plate. The prepared sample has 1L-MoS₂ overlapped with GO sheets and 1L-MoS₂ simultaneously. (b) PL spectrum of the 1L-MoS₂ grown by CVD. (c) Raman spectrum of the 1L-MoS₂ grown by CVD. The insets present schematics of the atomic displacements for each Raman mode. The interval between the two Raman modes is 18 cm⁻¹, which indicates 1L-MoS₂. (d) FTIR and (e) Raman spectra of the GO sheets used in this study; the GO sheets include abundant functional groups.

Figure 2. Tuning the optical properties of 1L-MoS₂ by GO coatings. (a) PL spectra measured from 1L-MoS₂ and 1L-MoS₂/GO. PL intensity and peak positions were considerably modified by contact with GO sheets. (b) Fitted PL spectra of 1L-MoS₂/GO and 1L-MoS₂. A peaks in the PL spectra were reproduced by assuming three peaks with Lorentzian functions for biexciton (XX), trion (X*), and neutral exciton (X) peaks. (c) 1L-MoS₂ and (d) 1L-MoS₂/GO with increasing excitation power. (e) Raman spectra of 1L-MoS₂ and 1L-MoS₂/GO. The A₁g mode of 1L-MoS₂/GO is blue-shifted by 1.9 cm⁻¹ compared with that of 1L-MoS₂. (f) Schematic representation of the electron-withdrawing model for tuning the electron density of 1L-MoS₂ by coating with GO.
efficiency than 1L-MoS$_2$.$^{19,20}$ Therefore, it is a challenge to find an effective approach to obtain p-type-doped 1L-MoS$_2$. Recently, the use of graphene oxide (GO) was theoretically suggested for modulating the electronic structures of MoS$_2$ by acting as a hole-injection layer; however, this idea has not yet been experimentally demonstrated.$^{21}$

In this study, we report the p-type doping of 1L-MoS$_2$ via the spin-casting of GO. It is observed that GO, with its apparent electron-withdrawing characteristics, effectively extracts electrons from a 1L-MoS$_2$ sheet with which it is in contact. Enhancement of the neutral exciton peak intensity accompanied by relative reduction of the negative trion peak in the photoluminescence (PL) spectrum and a peak shift of the Raman spectrum suggest that the optical properties of 1L-MoS$_2$ were tuned because of hole injection by the GO attachment. Moreover, the current versus voltage ($I−V$) curves of the 1L-MoS$_2$ with GO indicate the modulation of the electrical properties of 1L-MoS$_2$. We also observed current rectification in the $I−V$ curves of lateral 1L-MoS$_2$ and 1L-MoS$_2$/GO structures.

**RESULTS**

The flakes of 1L-MoS$_2$ were synthesized on a SiO$_2$ substrate using chemical vapor deposition (CVD).$^{22}$ The shape of the 1L-MoS$_2$ flakes was triangular, and the average size of the flakes was 30 μm (see the Figure S1). The detailed sample preparation procedure is depicted in Figure 1a. Hydrophilic treatment was applied to the entire surface of the quartz substrate using UV-lamp irradiation for 20 min to achieve better adhesion of the GO sheets. The GO sheets were then coated onto half of the substrate using the spin-casting method, as illustrated in Figure 1a. The average thickness of the coated GO sheets on the substrate was approximately 10 ± 4 nm (~9 ± 3 layers) (see Figure S2). Across the entire surface of the prepared template, large flakes of 1L-MoS$_2$ were transferred onto the coated GO on the SiO$_2$/Si substrate (1L-MoS$_2$/GO) using the wet-transfer method (see the Methods for details).

To examine the optical properties of the 1L-MoS$_2$, PL and Raman spectroscopy measurements were performed at room temperature. A solid-state laser with a wavelength of 532 nm was used as the excitation source, and a spectrometer equipped with three gratings (150 grooves/500 nm blaze, 600 grooves/600 nm blaze, and 1800 grooves/500 nm blaze) was utilized for the measurements. Two peaks, A (1.88 eV) and B (2.06 eV), appeared in the PL spectrum, as observed in Figure 1b, which represent the exciton transition corresponding to direct optical transitions from the near band edge.$^{23}$ In the Raman spectrum, peaks appeared at 385 (E$_{2g}$) and 403 cm$^{-1}$ (A$_{1g}$), as shown in Figure 1c. The distance between the A$_{1g}$ and E$_{2g}$ peaks ($Δ = A_{1g} − E_{2g}$) was approximately 18 cm$^{-1}$, which is characteristic of 1L-MoS$_2$.$^{24}$ To verify the presence of the functional groups of GO, Fourier-transform infrared (FTIR) spectroscopy measurements were performed at room temperature as depicted in Figure 1d. The observed functional groups are the well-known electron-withdrawing groups such as phenol, carboxyl, hydroxyl, and epoxy groups.$^{25}$ The density of negative charge in 1L-MoS$_2$ was much higher than that in GO because of the strong n-type nature of MoS$_2$, as confirmed by the zeta potential measurements (see Figure S3). This finding suggests that the electrons in 1L-MoS$_2$ can be attracted to the GO sheets. The presence of GO was verified by Raman modes of the D band (1347 cm$^{-1}$) and G band (1600 cm$^{-1}$) in the Raman spectrum, as observed in Figure 1e.

Figure 2a presents the PL spectra measured for 1L-MoS$_2$/GO (red arrow) and 1L-MoS$_2$ (black arrow). The samples for the measurement are schematically illustrated in the inset of Figure 2a. PL peaks associated with A and B excitons were observed in both samples; however, the PL intensity of 1L-MoS$_2$/GO (red curve) was greatly enhanced (by approximately 470%) compared with that of 1L-MoS$_2$ (black curve). Moreover, the peak position of the PL spectrum of 1L-MoS$_2$ was blue-shifted in the 1L-MoS$_2$/GO. The PL peak of the A excitons in 1L-MoS$_2$ appeared at 1.85 eV. However, in the PL spectrum of 1L-MoS$_2$/GO, the peak positions for the A excitons appeared at higher energy compared with those of 1L-MoS$_2$; i.e., the A peak is observed at ~1.92 eV. According to previous reports, these blue-shifted peak positions for A and B excitons could be explained by p-type doping or compressive strain of MoS$_2$ owing to lattice mismatch between the GO and MoS$_2$ layer.$^{15,26−29}$ The two peaks at approximately 2.15 eV represent Raman modes of the GO sheets, which correspond to the D and G bands.$^{30,31}$

To analyze the peak shift of the PL spectra, Lorentzian fitting and intensity normalization were applied as shown in Figure 2b. The top and bottom panels present the fitted PL spectra of 1L-MoS$_2$/GO and 1L-MoS$_2$, respectively. The PL peak for the A exciton is composed of the biexciton peak (XX) at 1.85 eV (red line), trion peak (X*) at 1.88 eV (green line), and neutral exciton peak (X) at 1.91 eV (blue line). According to a previous report, neutral excitons are converted into negative or positive trions depending on the excess carrier type.$^{32}$ Here, trion peak (X*) in the pristine MoS$_2$ is a negative trion, as reported by several studies.$^{14,15,33}$ Meanwhile, the trion peak (X*) from MoS$_2$/GO in Figure 2b can be estimated as a positive trion or negative/positive trion from the p-type doped feature of the blue-shifted PL peak.$^{32}$

The biexciton (XX) is predominant in the A exciton peak of 1L-MoS$_2$ at high excitation power.$^{32}$ The negative trion emission emergence in the as-prepared 1L-MoS$_2$ is attributed to the heavy electron (n-type) doping.$^{33}$ However, neutral exciton recombination governs the A exciton peak of 1L-MoS$_2$/GO, as observed in Figure 2b, and can be attributed to the decrease in the number of excess electrons resulting from the loss of electrons to the GO.$^{34}$ For further investigation of the optical properties of 1L-MoS$_2$ and 1L-MoS$_2$/GO, we examined the excitation-laser-power-dependent PL emission. Parts c and d of Figure 2 show the power-dependent PL intensities of 1L-MoS$_2$ and 1L-MoS$_2$/GO, respectively. Figure 2c presents the integrated PL intensity of the biexciton (XX), trion (X*), and neutral exciton (X) of 1L-MoS$_2$ with excitation powers ranging from ~0.04 to 3.2 mW (see Figure S4). In 1L-MoS$_2$, the PL intensity of the biexciton peak increased upon increasing the incident laser power, as reported for MoS$_2$.$^{32}$ and WS$_2$.$^{35}$ The biexciton (XX) intensity increased with an increase in the excitation power, which could be interpreted as an increased biexciton population induced by abundant exciton density.$^{36,37}$ However, the trion (X*) intensity decreased at high excitation powers and exhibited an opposite trend as the biexciton (XX) intensity, which is thought to have contributed to the formation of biexcitons (XX) with increasing excitation power.$^{36,37}$

The integrated PL intensity of the neutral exciton (X) peak exhibited almost negligible changes as a function of the excitation power compared with those of the biexciton (XX) and trion (X*) peaks. The saturated PL intensity of the neutral exciton (X) peak is attributed to Coulomb screening and Pauli blocking effects induced by the presence of excess free
Figure 3. Variations in the optical properties of 1L-MoS2 with different GO thicknesses: (a) PL spectra, (b) integrated PL intensity (left) and A exciton peak energy (right), (c) fwhm of the PL emission peak, (d) Raman spectra, (e) E1g mode, and (f) A1g mode.

electrons. Figure 2d presents the integrated intensity of the bieexciton (XX) and neutral exciton (X) peaks of 1L-MoS2/GO. The intensities of the neutral exciton (X), bieexciton (XX), and trion (X+) peaks increased with increasing excitation power (see Figure S5). Because the exciton-exciton annihilation and/or charge screening effect are suppressed by the reduction of excess free electrons, the intensity of the neutral exciton (X) is increased with increasing excitation power. For 1L-MoS2/GO, the PL intensity of the neutral exciton is dominant in the entire range of incident laser powers. Furthermore, similarly increased PL intensities of the bieexciton (XX) and trion (X+) are attributed to reduced excess free electrons, which result in contact with the electron-withdrawing group of GO.

In addition, we also performed confocal Raman spectroscopy on the 1L-MoS2 and 1L-MoS2/GO samples. The measured Raman spectra of 1L-MoS2 and 1L-MoS2/GO are represented as scatter plots with blue and red circles, respectively, in Figure 2e. The solid and dotted lines represent the fitted spectra obtained using the Lorentzian function. The A1g peak in the Raman spectrum of 1L-MoS2/GO was blue-shifted by ~1.9 cm$^{-1}$ compared with that of 1L-MoS2. The full-width at half-maximum (fwhm) values for the A1g modes of 1L-MoS2 and 1L-MoS2/GO were 5.5 and 3.4 cm$^{-1}$, respectively, and the Raman peak for the E2g mode of 1L-MoS2/GO was almost unchanged within spectral resolution compared with that of 1L-MoS2. A previous report indicated that the A1g phonon mode is remarkably sensitive to coupling with electrons and the bonding strength. Therefore, loss of the excess electrons in the GO-coated 1L-MoS2 yields reduced electron-phonon coupling, resulting in a narrower fwhm and stronger bonds and, thus, the blueshift. Hence, the shifted electronic properties of the 1L-MoS2 owing to the contact with the GO sheets are attributed to electron donation from the 1L-MoS2 to the GO sheets induced by the electron-withdrawing groups of the GO. The process of electron withdrawal between 1L-MoS2 and GO is schematically illustrated in Figure 2f. In other words, the considerable change in the PL/Raman spectra can be attributed to the withdrawal of electrons from the 1L-MoS2 to the GO.

To verify the effect of the GO thickness in 1L-MoS2/GO, PL and Raman spectra were obtained as a function of the GO thickness (Figure 3a). The thickness of the GO sheets was controlled by modifying the number of coating sequences performed to form the GO sheets on the substrate (see Figure S2). Since thickness fluctuations of GO sheets are considerably higher due to a simple coating process, three representative thicknesses of 10 ± 4, 15 ± 5, and 50 ± 10 nm have been applied in this study. Notably, we have used the average data points of PL and Raman spectra for each sample because of the slightly inhomogeneous distribution of optical properties of 1L-MoS2/GO. As revealed in Figure 3a, the PL peak position and spectral shape of the A exciton changed markedly as the GO thickness was varied. Figure 3b presents plots of the integrated PL intensities (left) and peak positions (right) of the A exciton measured for 1L-MoS2/GO with various GO thicknesses. As the thickness of the GO was increased, the PL intensity of 1L-MoS2/GO gradually decreased, but, still higher than that of the pristine 1L-MoS2 up to 50 nm of GO. This finding contradicts that from a previous report, which stated that PL intensity increased upon increasing the doping step. According to another previous report, as the tensile strain in 1L-MoS2 increased, the PL intensity decreased and the PL peak position was red-shifted. Using atomic force microscopy (AFM) (see Figure S6), we observed that the surface roughness and topographical fluctuation of the GO sheets increased upon increasing the thickness of the GO sheets. Therefore, the decrease in the PL intensity could be explained by higher strain of 1L-MoS2 arising from the rough surface of the coated GO. The fwhm values of PL emission in 1L-MoS2 with different GO thicknesses are presented in Figure 3c. As observed in Figure 2b, the spectral shape of PL emission in 1L-MoS2 becomes sharper after GO doping because of the affected neutral exciton peak and suppressed bieexciton peak. However, the fwhm of the PL emission in 1L-MoS2/GO slightly increased when the thickness of GO was increased, which could reflect a decrease in the doping effect (see Figure S8).

Such a reduction in the electron-withdrawing phenomenon with increasing coating thickness is supported by the Raman
spectra presented in Figure 3d. Here, the peak position and fwhm of the E\textsubscript{2g} and A\textsubscript{1g} modes in the Raman spectra were changed by varying the thickness of the coated GO sheets. The E\textsubscript{2g} mode of 1L-MoS\textsubscript{2} was red-shifted with increased fwhm by increasing the thickness of the coated GO sheets (Figure 3e). In contrast, the A\textsubscript{1g} mode of 1L-MoS\textsubscript{2} was blue-shifted with decreased fwhm through contact with the GO sheets; the amount of blueshift was almost the same with increasing thickness of the GO sheet (Figure 3f). Note that the increased fwhm of both E\textsubscript{2g} and A\textsubscript{1g} modes indicates that tensile strain of 1L-MoS\textsubscript{2} is increased.\textsuperscript{44} These findings indicate that the thickness of the coated GO sheets affects the strain of 1L-MoS\textsubscript{2} because the E\textsubscript{2g} mode is highly sensitive to the strain level.\textsuperscript{42,43} Consequently, changes in the PL and Raman spectra as a function of the GO thickness could be induced by a change in the strain effect between the 1L-MoS\textsubscript{2} and GO sheets with different thicknesses. In addition, PL and Raman data of 1L-MoS\textsubscript{2} on 1L-GO show a similar tendency as that of 1L-MoS\textsubscript{2} on multilayer GO, even though PL enhancement of 1L-MoS\textsubscript{2} on 1L-GO is lower than that of 1L-MoS\textsubscript{2} on 10 nm-thick GO, which is attributed to the lower number of functional groups of GO (see Figures S7 and S11).

To clarify the dominant reason for the dramatic changes in the optical properties of 1L-MoS\textsubscript{2} resulting from coating with GO, we performed confocal PL and Raman mapping of the samples. Figure 4a presents a confocal PL intensity image of 1L-MoS\textsubscript{2}/GO that is in accordance with Figure 2a. The inset in Figure 4a presents an optical microscopy image of the sample; the white dotted line indicates the edge of the GO region. As observed in Figure 4a, the 1L-MoS\textsubscript{2}/GO region has a greater PL intensity with higher intensity fluctuations compared with the 1L-MoS\textsubscript{2} region without GO. The higher PL intensity appearing in a broad area near the GO may be attributed to the locally strain-relaxed areas of 1L-MoS\textsubscript{2} induced by partial floating because of the slight height difference between the GO and substrate.\textsuperscript{45} Figure 4b presents PL spectra extracted from the points marked I and II in Figure 4a, which are representative positions for 1L-MoS\textsubscript{2} and 1L-MoS\textsubscript{2}/GO, respectively. Figure 4c presents a Raman intensity image for the wavenumber range of 370–420 cm\textsuperscript{-1}. The Raman scattering intensity of 1L-MoS\textsubscript{2}/GO is slightly higher than that of 1L-MoS\textsubscript{2} without GO. Figure 4d presents local Raman spectra obtained from points I and II in Figure 4c. The black and red curves represent 1L-MoS\textsubscript{2} and 1L-MoS\textsubscript{2}/GO, respectively. The E\textsubscript{2g} peak in the Raman spectrum of 1L-MoS\textsubscript{2}/GO was red-shifted by \sim 1.1 cm\textsuperscript{-1} compared with that of 1L-MoS\textsubscript{2}. The A\textsubscript{1g} peak of 1L-MoS\textsubscript{2}/GO was blue-shifted by \sim 1.5 cm\textsuperscript{-1} compared with that of 1L-MoS\textsubscript{2}. The fwhm for the A\textsubscript{1g} modes of 1L-MoS\textsubscript{2} and 1L-MoS\textsubscript{2}/GO were 11.2 and 5.3 cm\textsuperscript{-1}, respectively. The red-shift of the E\textsubscript{2g} mode reflects the effect of interfacial strain resulting in increased tensile strain of 1L-MoS\textsubscript{2}.\textsuperscript{46} The blue-shift and decreased fwhm of the A\textsubscript{1g} mode indicate p-doping of 1L-MoS\textsubscript{2} and/or an increase in van der Waals interactions at the interface between GO and 1L-MoS\textsubscript{2} (see Figure S9). To verify the presence of GO sheets under the 1L-MoS\textsubscript{2} confocal Raman measurements were conducted in the wavenumber range from 1000 to 1900 cm\textsuperscript{-1}, which includes the D and G Raman modes of GO. The confocal Raman intensity image for the D and G modes of GO is presented in Figure 4e. The white dotted line represents an edge of the underlying GO layer.

For comparison of the strain effect in 1L-MoS\textsubscript{2} GO sheets were coated on 1L-MoS\textsubscript{2} (see Figure S10). The PL and Raman enhancement with peak shift of 1L-MoS\textsubscript{2} under GO exhibit similar tendencies as those of 1L-MoS\textsubscript{2} on GO (see Figure S10). Note that 1L-MoS\textsubscript{2} under the GO exhibited higher strain than 1L-MoS\textsubscript{2} on GO because 1L-MoS\textsubscript{2} was sandwiched by GO and SiO\textsubscript{2}. Figure 4f presents the local Raman spectra at points I and II in Figure 4e. The Raman peaks at 1311 and 1570 cm\textsuperscript{-1} correspond to the D and G modes of GO, respectively. The confocal Raman spectroscopy results for GO confirm that the 1L-MoS\textsubscript{2} is placed across the edge of GO. Consequently, we can conclude that the p-doping effect dominates the enhancement of the PL intensity of 1L-MoS\textsubscript{2} with GO even though the 1L-MoS\textsubscript{2} is under tensile strain. Furthermore, to clarify that the electron withdrawing characteristic of GO affected to PL enhancement of 1L-MoS\textsubscript{2}, we functionalized GO sheets with diverse oxygen functional groups. The PL measurements revealed higher PL enhancement of 1L-MoS\textsubscript{2} upon increasing the density of functional groups in GO (see Figure S11). This result indicates that the p-doping effect of 1L-MoS\textsubscript{2}/GO mainly arose by electron withdrawal from 1L-MoS\textsubscript{2} to GO.

To confirm that a reduced electron concentration in 1L-MoS\textsubscript{2} results from coating with GO, the electrical transport
characteristics of GO-coated 1L-MoS₂ were studied using $I−V$ measurements. For the measurement, we fabricated a back-gated field effect transistor (FET) using e-beam lithography and evaporation techniques. Figure 5a presents an optical image and schematic illustration of the 1L-MoS₂ FET device and the Cr/Au (10 nm/70 nm) used as the source and drain electrodes. Figure 5b presents the source–drain current ($I_{DS}$) versus gate voltage transfer curves of 1L-MoS₂ before and after GO coating. The filled black squares and red circles represent $I−V$ curves of 1L-MoS₂ and GO-coated 1L-MoS₂, respectively. A substantially lower threshold voltage with remarkably lower current level was observed for the 1L-MoS₂ FET with GO compared with the 1L-MoS₂ FET without GO. This shows that electron concentration of 1L-MoS₂ with GO is significantly decreased and hole concentration is meaningfully increased compared with that of 1L-MoS₂ without GO, as shown in Figure 5b. Therefore, this supports the idea that the functional groups of GO induce the withdrawing the electrons from MoS₂ resulting in p-type doping.

Figure 5c shows an $I−V$ curve of the lateral 1L-MoS₂−1L-MoS₂/GO structure with voltage sweep range from −20 to +20 V. The inset is a log scale $I−V$ curve and schematic of the lateral 1L-MoS₂−1L-MoS₂/GO structure designed for the $I−V$ measurement. The distance between the anode and cathode was maintained at two metal probes maintained at 500 μm. As observed in the $I−V$ curve, a considerably lower current close to zero was observed under reverse bias; however, a dramatically increased current was observed from the threshold voltage of approximately 11 V under forward bias. This rectification of current is attributed to the relatively lower electron concentration of 1L-MoS₂/GO compared with that of 1L-MoS₂. Electrons cannot flow through this structure when reverse bias is applied because of the increased built-in potential that is formed at the interface between 1L-MoS₂ and 1L-MoS₂/GO. In contrast, electrons can easily flow through this structure under forward bias because of the decreased built-in potential. This clear current rectification phenomenon can be regarded as a typical p–n junction diode characteristic. Since the gold-coated tungsten tip was used for electrical measurement, a Schottky barrier (SB) is formed at the interface between the metal tip and 1L-MoS₂. To confirm the influence of SB on current rectification, we measured the $I−V$ curve of 1L-MoS₂−1L-MoS₂ structure (see SI, Figure S12). In the $I−V$ curve, the current rectification was not observed. This indicates that the current rectification is mostly induced by the lateral 1L-MoS₂−1L-MoS₂/GO homojunction diode structure. Consequently, the electrical transport between 1L-MoS₂/GO and 1L-MoS₂ supports the idea that the adsorption of GO modulates the electronic property of 1L-MoS₂.

CONCLUSIONS

In this work, we have demonstrated p-type doping of 1L-MoS₂ through contact with the GO sheets. Upon decreasing the thickness of the GO sheets, increased PL intensity and shifted A₁g peak positions in the Raman spectra were clearly observed and served as evidence of the tuned electronic properties of the 1L-MoS₂. The tuning of the electronic properties of 1L-MoS₂ by coating with GO can be explained by the abundant electron-withdrawing characteristics from various oxygen functional groups presented on the GO sheets. Using confocal PL/Raman spectroscopy, we observed that the p-doping effect was predominant in 1L-MoS₂ with GO through the enhancement of the PL intensity of neutral exciton and blue-shifted A₁g Raman peak. Moreover, the electrical transport characteristics of a GO/ MoS₂ FET demonstrated that a simple GO coating on 1L-MoS₂ significantly reduces the electron concentration and slightly increases the hole concentration of the 1L-MoS₂. Our technique for tuning the optical and electronic properties of 1L-MoS₂ by coating with GO sheets offers remarkable advantages for optoelectronic applications; the process is simple and low cost, and it allows large-scale fabrication and modulation of the electronic properties.

METHODS

Synthesis of Graphene Oxide. GO sheets were prepared using a modified Hummers’ method. Natural graphite was obtained from Alfa Aesar (99.999% purity, 200 mesh). First, 5 g of graphite and 350 mL of 10 M H₂SO₄ were blended. Subsequently, KMnO₄ (15 g) was slowly added over approximately 1 h and stirred continuously for 2 h in a cooled water bath. After the mixture had been strongly stirred for 3 days at room temperature, deionized (DI) water was added, and stirring proceeded for 10 min in a cooled water bath. The mixture was then stirred for an additional 2 h at room temperature after the addition of an aqueous solution of H₂O₂ (30 wt%). A diluted HCl (35 wt%) solution was then added and stirred for 30 min at room temperature. After the supernatant solution was decanted, DI water was slowly added and stirred for 30 min. Then, 1 g/L of the graphite oxide solution in water was sonicated for 1 h to exfoliate the GO sheets. To obtain dispersed GO, centrifugation at 10,000 rpm was performed for 1 h, and then the supernatant solution was decanted.

Wet Transfer of 1L-MoS₂. The wet-transfer process used in this study was conducted as follows: PMMA C4 (Micro Chem, 4 wt% in...
chlorobenzene) was coated onto large flakes of 1L-MoS2 grown on a SiO2/Si substrate to serve as a supporting layer for the transfer process. After being coated with PMMA, the sample was floated on a 1 M potassium hydroxide (KOH) solution at 80 °C for 15 min to remove the SiO2 layer. Subsequently, only large flakes of 1L-MoS2 with PMMA remained on the KOH solution. The remaining PMMA/1L-MoS2 was washed with DI water to remove any residual KOH etchant. The washed PMMA/1L-MoS2 was transferred onto the prepared GO-coated template and SiO2/Si substrate described in the main text. After the drying process was complete, the PMMA was removed using acetone.

**Device Fabrication.** The source and drain electrodes were patterned using electron-beam lithography followed by Cr/Au (10 nm/70 nm) deposition using e-beam/thermal evaporation. After the lift-off process was complete, the PMMA was removed using acetone.

**Characterization Methods.** FTIR spectra were recorded using an FTIR spectrometer (4700, JASCO). UV–vis absorption spectra were measured using a commercial spectrophotometer (V-670, JASCO). Raman and PL spectra were obtained using a multifunctional optical microscopy system (NTEGRA SPECTRA, NT-MDT). In this system, a 532 nm (2.33 eV) laser was used as the excitation source. To measure the electric potentials of GO and 1L-MoS2 in solution, zeta potential analysis (ELS-8000, Otsuka Electronics) was performed. The topography of the sample surfaces was examined using a commercial atomic force microscope in the contact mode (Anasys Instruments). Field-emission scattering electron microscopy (SEM, JSM7000F, JEOL) was performed to examine the surface morphology of the samples.

**REFERENCES**


Notes

The authors declare no competing financial interest.

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