Changes in the Photoluminescence of Monolayer and Bilayer Molybdenum Disulfide during Laser Irradiation

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ABSTRACT: Various postsynthesis processes for transition metal dichalcogenides have been attempted to control the layer number and defect concentration, on which electrical and optical properties strongly depend. In this work, we monitored changes in the photoluminescence (PL) of molybdenum disulfide (MoS2) until laser irradiation generated defects on the sample flake and completely etched it away. Higher laser power was required to etch bilayer MoS2 compared to monolayer MoS2. When the laser power was 270 μW with a full width at half-maximum of 1.8 μm on bilayer MoS2, the change in PL intensity over time showed a double maximum during laser irradiation due to a layer-by-layer etching of the flake. When the laser power was increased to 405 μW, however, both layers of bilayer MoS2 were etched all at once, which resulted in a single maximum in the change of PL intensity over time, as in the case of monolayer MoS2. The dependence of the etching pattern for bilayer MoS2 on laser power was also reflected in position changes of both exciton and trion PL peaks. The subtle changes in the PL spectra of MoS2 as a result of laser irradiation found here are discussed in terms of PL quantum efficiency, conversion between trions and excitons, mean interatomic spacing, and the screening of Coulomb interaction.

INTRODUCTION

Two-dimensional (2D) layered materials such as graphene and transition metal dichalcogenides (TMDs) are expected to be used in various incoming types of electronic and optoelectronic applications involving flexible devices.1-4 Molybdenum- and tungsten-based TMDs have band gaps that vary in thickness, resulting in peculiar electrical and optical properties distinct from those of graphene such as large current on/off ratios exceeding 106 and enhanced light–matter interaction leading to strong photoluminescence (PL) emission and photoresponsivity.5-7 Because the properties of 2D materials strongly depend on the number of layers, defects, environment, and so on, a reliable process to control the thickness and defects of 2D materials is highly sought after. Various postsynthesis processes, such as thermal annealing, high-power laser irradiation, and plasma treatment, have been shown to successfully convert thin multilayer graphene or molybdenum disulfide (MoS2) down to monolayers (1L) by thermal oxidative etching of the heated upper layers, while the bottom layer remains almost unaffected.8-14 MoS2 is a strongly interacting TMDC material that is stable under ambient conditions and has an indirect or direct band gap of 1.2–1.9 eV depending on the number of layers.15,16 On the other hand, some other experiments resulted in a different tendency: the thermal oxidative etching process in 2D materials can be easier in monolayers than in multilayers.17-19 Laser beams have been used not only as etching tools for 2D materials but also as probes to measure their physical and structural properties.20-24 Accordingly, multiple physical properties can be monitored simultaneously in real time while adjusting the defects and thickness of a 2D material using a laser beam. Oh et al. have measured the time evolution of the PL spectra of 1L-MoS2 during laser irradiation and discussed its results in terms of structural damage and associated oxygen...
adhesive. In the present work, we have monitored the PL and Raman spectra of monolayer and bilayer (2L) MoS₂ during laser irradiation until the irradiated areas of the flakes were completely etched away. Our results show that more thermal energy is required to etch 2L-MoS₂ than 1L-MoS₂, and interestingly, in the case of 2L-MoS₂ the flake was etched layer-by-layer when the laser power was not too high, while both layers of the flake were etched all at once at a sufficiently high laser power. Our results are expected to be helpful not only for the in-depth understanding and control of 2D material properties but also in the development of 2D nanodevices with specific defect concentrations, number of layers, and shapes suitable for particular applications.

**RESULTS AND DISCUSSION**

Figure 1a shows changes in the PL spectrum of 1L-MoS₂ over time while irradiating the flake with a laser power of 180 μW. Two PL peaks are observed: a main A peak between 1.8 and 1.9 eV, and a shoulder B peak over 1.9 eV (Figure S1 of the Supporting Information). The energy difference between the A and B peaks was approximately 0.15 eV, which is in good agreement with the splitting energy of the valence band of 1L-MoS₂ at the K-point by spin–orbit interaction. As the local temperature of the laser-irradiated area increased, we observed a rapid increase in PL intensity from 25 min but an abrupt decrease after 50 min. The inset in Figure 1a shows the laser-irradiated areas were completely etched away during the measurements (Figure S2 of the Supporting Information).

It is known that the A peak consists of a negatively charged trion (A°) peak and a neutral exciton (A⁺) peak since MoS₂ on SiO₂ is electron-doped due to charged impurities at the interface. The relative amounts of trions and excitons are adjustable by controlling the electron concentration: the lower the number of electrons, the lower the number of trions and the higher the number of excitons. Previous works have shown that plasma treatment, laser irradiation, etc. generate defects on MoS₂ and subsequent oxygen bonding to the defects converts trions to excitons by depleting electrons, which is accompanied by a sharp enhancement in PL intensity. In particular, Nan et al. reported that the PL intensity from defects could be greatly enhanced due to the high quantum efficiency of quasiparticles localized at the defects. Figure 1b plots the changes in intensities of A⁺, A°, and B peaks over time obtained by a deconvolution of the PL spectra into three Lorentzian curves (Figure S1 of the Supporting Information). The intensity of the A° peak was comparable with that of the A⁺ peak in the beginning. As the local temperature of the laser-irradiated area increased, PL emission was enhanced rapidly from 25 min due to the generation of defects. Here, we note that both A⁺ and A° peaks rapidly increased until 50 min, although some trions were expected to convert to excitons, which is contrary to previous results; Oh et al. reported that the A° peak rapidly increased and then gradually decreased, while the A⁺ peak continued to decrease throughout the irradiation process. Our results indicate that the PL intensities of both trions and excitons localized to the defects are greatly enhanced. As the defect concentration increased, however, the A° peak increased more than the A⁺ peak due to the conversion of some trions to excitons. After 50 min, the intensities of both A⁺ and A° peaks decreased more sharply as the remaining area of 1L-MoS₂ under laser irradiation was reduced. The weak PL spectra after 100 min were derived from the highly defected outer perimeter of the etched area, as shown in the inset of Figure 1a.
Changes in the positions and FWHMs of trion and exciton peaks during laser irradiation are shown in Figure 1c. For pristine 1L-MoS2, the position difference between A− and A° peaks, i.e., trion binding energy, was approximately 40 meV, which is comparable to previous reports.31−33 We notice the following peculiar change: the positions of the A− and A° peaks gradually blue-shifted, with the A− peak shifting more than the A° peak, thereby decreasing the energy gap between the two peaks to approximately 20 meV. On the other hand, the FWHM of each peak decreased, which is more apparent for the A° peak. In TMDCs, the position of each PL peak depends on the dielectric environment, doping, lattice parameter, and so on,28,31,32,34−36 although many works so far have discussed PL spectral changes with nearly constant positions of the A− and A° peaks.23,25,37−39 The effect of the dielectric environment was excluded in the present work since there was no change in the environment during measurements. Mak et al. reported that the A° peak red-shifted as electrons were depleted, while the A− peak slightly blue-shifted or remained almost unchanged.16 On the other hand, both A− and A° peaks have been shown to blue-shift over several tens of meV with decreasing temperature accompanying the contraction of lattice parameters.28,32,35,40 Theoretical calculation using density functional theory also showed that the energy gap at the K-point in the Brillouin zone of 1L-MoS2 increased with decreasing temperature because the valence band moved down while the position of the lowest conduction band did not change at all.32 Most of the defects generated by laser irradiation are sulfur vacancies, and oxygen bonding to sulfur vacancies minimizes deformation of the intrinsic hexagonal structure of MoS2 while reducing the electron concentration,19,25 which was reflected in the Raman spectra here (Figure S3 of the Supporting Information). Since the covalent radius of oxygen is about 60% of that of sulfur, the substitution of sulfur atoms with oxygen atoms reduces the mean interatomic spacing on the MoS2 plane. Therefore, we consider that the shifts of both A− and A° peaks by the increase of defect concentration were the result of the combination of the effects of electron depletion and lattice contraction. Our results in Figure 1c indicate that the lattice contraction effect was dominant as compared to the electron depletion effect, which is consistent with the results of Pei et al.32

The FWHMs of both A− and A° peaks became narrower with defect generation from 25 min, while trion binding energy decreased from 40 to 20 meV, which means that the screening of electron−hole or electron−electron Coulomb interactions increased. Our results are in good agreement with a report by Lin et al. in which they showed that as the dielectric constant of the surrounding material increased from 2 to 33, both A− and A° peaks blue-shifted (with the A− peak shifting more than the A° peak), and the FWHM of the A° peak narrowed from approximately 90 to 50 meV while that of the A− peak remained unchanged at approximately 75 meV.31

The Raman mode of the Si substrate under the 1L-MoS2 flake24 was also monitored during laser irradiation, but no changes in the peak position or width were observed, as shown in Figure 1d. In other words, the local temperature of the laser-irradiated substrate area remained almost unchanged,43,44 while that of the laser-irradiated MoS2 area increased above the point where it was completely decomposed. Our results indicate that the heat rapidly dissipated through the substrate,
but the MoS₂ flake and the substrate were thermally well insulated.

The effects of laser irradiation on the PL spectrum of 2L-MoS₂ are shown in Figure 2. Both A and B peaks are also observed. Despite the laser power being increased from 180 to 270 μW, the time at which the PL intensity began to increase was delayed to 180 min, indicating an enhanced tolerance to heat in 2L-MoS₂ in comparison with 1L-MoS₂. Unlike the monolayer case, here the PL spectra over time showed a double maximum during the laser etching process. The inset of Figure 2a shows that both layers (top and bottom layers) in the laser-irradiated areas were completely etched away (Figure S4 of the Supporting Information).

Changes in the intensities of A⁻, A°, and B peaks of 2L-MoS₂ over time obtained by a deconvolution of the PL spectra into Lorentzian curves (Figure S5 of the Supporting Information) are shown in Figure 2b. The intensities of the A⁻ and A° peaks were again comparable in the beginning. Then, as the defect concentration increased by laser irradiation and trions converted to excitons, the A° peak increased more than the A⁻ peak, in good agreement with the 1L-MoS₂ case. We argue that the double maximum shown in Figure 2a is the result of the top and bottom layers of the 2L-MoS₂ being sequentially etched layer-by-layer during irradiation. In other words, the first peak from 180 to 300 min is mainly due to the etching of the top layer, and the second peak from 300 to 360 min is due to the etching of the bottom layer. The Raman spectra obtained after stopping the PL measurement at 305 min were in good agreement with the typical Raman spectra of thinned 1L-MoS₂ (Figure S6 of the Supporting Information). Weak PL spectra after 360 min were from the highly defected outer perimeter of the etched area, as shown in the inset of Figure 2a. Our results show that we can remove the top layer only and leave the bottom layer of 2L-MoS₂ by adjusting the laser power and irradiation time. The larger relative intensity of the B peak to the A peak than that of the 1L-MoS₂ indicates that the flake is 2L-MoS₂, which is consistent with the Raman spectrum in Figure 2d.

The initial trion binding energy was measured to be approximately 40 meV, as shown in Figure 2c. As in the 1L-MoS₂ case, we notice that the FWHM of both A⁻ and A° peaks decreased from 180 min when the defects began to be generated. This is contradictory to a recent report by Barvat et al., which ascribed the broadening of both PL peaks of pulsed laser-deposited MoS₂ films to a relatively higher defect concentration as compared to mechanically exfoliated MoS₂ flakes. However, our results indicate that the screening of electron–hole or electron–electron Coulomb interactions has a dominant effect on the FWHM of both A° and A⁻ peaks, as discussed above.

The layer-by-layer etching of the 2L-MoS₂ by laser irradiation was also well reflected in the position changes of the exciton and trion peaks. Both A⁻ and A° peaks blue-shifted from about 180 min, hardly changed from 270 min, and then blue-shifted again from 300 min. As the A⁻ peak shifted more than the A° peak, the energy gap between the two peaks gradually decreased to approximately 20 meV at 340 min, which is equivalent to that of the highly defected 1L-MoS₂, as shown in Figure 1c. After 360 min, both peaks red-shifted and the energy gap between the two peaks increased over time since the PL signal came from highly defected 2L-MoS₂, the outer perimeter of the etched area. Consistent with the intensity changes of the PL peaks in Figure 2b, the position changes of the exciton and trion peaks in Figure 2c can be

Figure 3. Laser irradiation effects on the PL spectrum of 2L-MoS₂ at a laser power of 405 μW. (a) Changes in the PL spectrum over time under laser irradiation. Blue lines are the spectra every 30 min, and black lines are the spectra every 60 min. The inset is an optical image of the 2L-MoS₂ flake after measurements. (b) Changes in the intensities of the three PL peaks obtained by deconvoluting each PL spectrum with Lorentzian functions. The intensity of the B peak is multiplied by 5. (c) Changes in the positions and FWHMs of the exciton and trion PL peaks. (d) Changes in the PL spectrum of 2L-MoS₂ over time while irradiating with a laser of 180 μW. The inset is an optical image of the flake after measurement with the dotted circle representing the laser-irradiated area.
divided into four regions, that is, a region before defect generation up to 180 min, a region of top-layer etching between 180 and 300 min, a region of bottom-layer etching between 300 and 360 min, and a region after the complete etching of both layers.

Figure 3 shows the changes in the PL spectrum of 2L-MoS2 over time when the laser power on the flake was increased to 405 μW. The laser irradiation time required to generate defects in this case was greatly shortened, and the PL emission of excitons also increased more than that of trions as defect concentration increased. The maximum enhancement of the PL intensity during laser irradiation here was 5.5 times, a significant reduction compared to the 11 times enhancement for the 270 μW laser power, as shown in Figure 2. Moreover, no double maximum in the PL intensity over time was observed, although a small upturn appeared near 100 min, that is, considered to be due to the etching of the area corresponding to the very narrow outskirts of the laser spot with reduced power density. When the laser power was increased from 270 to 405 μW, our results show that both layers of 2L-MoS2 were etched all at once, and the maximum enhancement of the PL intensity was reduced since the flake was etched away before a sufficient defect concentration was reached. Both A− and A0 peaks continuously shifted toward higher energies over 30–150 min (with the A− peak showing a greater shift than the A0 peak) and then saturated, as shown in Figure 3b, which further supports that both layers of 2L-MoS2 were etched all at once. However, as shown in Figure 3d, no sharp enhancement in the PL intensity of 2L-MoS2 was observed even though the flake was continuously irradiated with a 180 μW laser for 7 h. That is, 2L-MoS2 was hardly affected under ambient conditions by irradiation with a laser of 180 μW, although 1L-MoS2 was completely etched away as shown in Figure 1.

## CONCLUSIONS

We have deposited monolayer and bilayer MoS2 on Si substrates covered with 300-nm-thick SiO2 and monitored the changes in their PL spectra during irradiation from a 532 nm laser until the irradiated areas were completely etched away. Higher laser power was required to etch bilayer MoS2. As laser irradiation generated defects on the MoS2 flake and etched it away, PL emission changed significantly depending on the laser power, as follows. For a laser power of 270 μW with a full width at half-maximum of 1.8 μm, the PL emission of bilayer MoS2 over time showed a double maximum due to a layer-by-layer etching of the flake. When the laser power was increased to 405 μW, however, the PL emission of bilayer MoS2 over time showed only a single maximum, as in the case of monolayer MoS2, which indicates that both layers of the flake were etched all at once. Changes in the intensities, positions, and FWHMs of trion and exciton peaks constituting the PL spectrum of MoS2 were discussed here based on changes in the quantum efficiency of PL, conversion between trions and excitons, mean interatomic spacing, and the screening of Coulomb interaction.

## EXPERIMENTAL SECTION

MoS2 flakes were deposited on Si substrates covered with thermally grown 300-nm-thick SiO2 by mechanical exfoliation of the bulk MoS2 crystal (429ML-AB, SPI Supplies). The thickness of the MoS2 flakes was estimated by optical microscopy and confirmed by micro-Raman spectroscopy. PL and Raman spectra were obtained under laboratory conditions (temperature = 21–22 °C and relative humidity = 30–40% in ambient air) in backscattering configuration using a laser line of 532 nm (LASOS, GLK 3250 TS) as an exciting light source. Other laser lines (e.g., an infrared pump laser line at 808 nm and a primary lasing line at 1064 nm) except for the 532 nm line from a diode-pumped solid-state (DPSS) laser27 were removed by placing a 532 nm band pass filter at the exit of the laser. Scattered light collected using a 50X objective lens (numerical aperture = 0.75) was analyzed using a Horiba Jobin Yvon LabRAM HR spectrometer equipped with a cooled charge-coupled device of 1024 × 256 pixels. A grating of 300 grooves/mm was used for the PL experiment, while a grating of 1800 grooves/mm was used for the Raman experiment. The full width at half-maximum (FWHM) of the laser spot on the flakes as measured by the “modified knife edge” method was approximately 1.8 μm.48 The total laser power on MoS2 flakes was measured using a power meter (Newport, 843-R) and controlled by changing the output power of the 532 nm laser. All of the laser powers mentioned in this paper are the total laser powers on the flakes, not the output power of the 532 nm laser. The PL or Raman spectra were recorded every 5 min until the MoS2 peaks almost disappeared with the laser spot fixed on the flake. The acquisition times for the PL spectra were 2 s for both 1L- and 2L-MoS2, while the acquisition times for the Raman spectra were 3 and 2 s for 1L- and 2L-MoS2, respectively.

## ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.9b04202.

Deconvolution of the PL spectra of MoS2 into three Lorentzian curves, change of the Raman spectrum of monolayer MoS2 over time during the laser irradiation, Raman spectra of MoS2 measured after PL experiments, and optical image and Raman spectra of 2L-MoS2 obtained before bottom-layer etching (PDF)

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