A direct comparison of CVD-grown and exfoliated MoS₂ using optical spectroscopy

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Abstract
MoS₂ is a highly interesting material, which exhibits a crossover from an indirect band gap in the bulk crystal to a direct gap for single layers. Here, we perform a direct comparison between large-area MoS₂ films grown by chemical vapor deposition (CVD) and MoS₂ flakes prepared by mechanical exfoliation from mineral bulk crystal. Raman spectroscopy measurements show differences between the in-plane and out-of-plane phonon mode positions in CVD-grown and exfoliated MoS₂. Photoluminescence (PL) mapping reveals large regions in the CVD-grown films that emit strong PL at room-temperature, and low-temperature PL scans demonstrate a large spectral shift of the A exciton emission as a function of position. Polarization-resolved PL measurements under near-resonant excitation conditions show a strong circular polarization of the PL, corresponding to a valley polarization.

Keywords: MoS₂, Raman spectroscopy, photoluminescence, excitons

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

1. Introduction
In recent years, two-dimensional crystal structures [1] have attracted a lot of attention. Many layered crystals can be mechanically exfoliated to yield flakes of single-layer thickness, just as graphene can be prepared from graphite. Transition metal dichalcogenides such as MoS₂ and WS₂ are currently studied by many groups, as they show drastic changes of the band structure with reduction of the number of layers. For MoS₂, a transition from indirect to direct band gap was theoretically predicted [2] and experimentally observed [3, 4], making the material interesting for possible opto-electronic applications. The band structure of monolayer MoS₂ allows for optical generation and detection of a valley polarization [5]. Large excitonic effects are predicted due to the two-dimensional crystal structure [6]. Additionally, a number of studies demonstrated transistor [7] and memory devices [8] with attractive properties. The majority of experimental studies have been performed on MoS₂ flakes exfoliated from mineral bulk crystal. While this material is readily available for fundamental research and even allows for preparation of prototypical integrated circuits [9], scalable application of atomically thin MoS₂ will require wafer-scale deposition of films with well-controlled properties. A number of different methods for preparation of large-area MoS₂ films have been developed in recent years. It was demonstrated that chemical exfoliation of bulk MoS₂, in combination with mild annealing, can yield thin films which show pronounced photoluminescence (PL) [10]. Another approach is based on the sulfurization of thin metallic Mo films in nitrogen [11] or Argon [12] atmosphere. Chemical vapor deposition (CVD) and physical vapor deposition of MoS₂ was reported for different substrates, including graphene [13], copper [14] and sapphire [15].

Here, we report on optical spectroscopy of large-area MoS₂ films grown by CVD on SiO₂. We utilize Raman spectroscopy, as well as PL spectroscopy at room temperature and low temperatures to directly compare the properties of these films to MoS₂ flakes prepared by mechanical
exfoliation. PL mapping reveals large regions in the CVD-grown films that emit strong PL at room-temperature, and low-temperature PL scans demonstrate a large spectral shift of the A exciton emission peak as a function of position. A strong circular polarization of the PL is observed under near-resonant excitation conditions at low temperature, indicating an optically oriented valley polarization. We find pronounced differences between the in-plane and out-of-plane phonon mode positions and linewidths in CVD-grown and exfoliated MoS$_2$, and a clear correlation between the Raman and PL spectra in the CVD-grown films.

2. Methods

2.1. Sample preparation and optical characterization

The synthesis processes for the MoS$_2$ growth is as follows: We use two alumina crucibles (Aldrich Z561738, 70 mm × 14 mm × 10 mm), one containing 0.55 g of MoO$_3$ (Aldrich 99.5%) and one containing 0.70 g of sulfur powder (Alfa 99.5%) as our Mo and S sources, respectively. These crucibles are placed in a quartz process tube (2 inch diameter), which is inserted into a three zone tube furnace (Mellen TT12), in which only the center zone is heated. A flow of nitrogen gas (99.999%) is used to purge the tube (5.0 SCFH) for 15 min after which the flow is reduced (0.5 SCFH). The crucible containing MoO$_3$ is placed at the center of the heated zone with the substrate placed across the middle of the crucible. The crucible containing sulfur is placed 30 cm away from the crucible substrate placed across the middle of the crucible. The crucible containing sulfur is heated to 700 °C in 43 min, as measured by a type-K thermocouple at the central heated zone and is held at this temperature for 10 min. Subsequently, the furnace is turned off and allowed to cool naturally to 500 °C. Compressed air is then used to rapidly cool the furnace to room temperature. This procedure yields samples as shown in figure 1(a), where a deposition of material is clearly visible on a 10 mm wide strip along the center of the substrate. Optical microscopy reveals regions of uniform apparent color, indicating a constant MoS$_2$ layer thickness, which are about 300 µm wide, at the edges of this strip, see figure 1(b). The long axis of these regions is aligned with the nitrogen flow direction during growth. We will show below that a pronounced PL emission corresponding to single-layer MoS$_2$ is observed in these regions. At the edge of these regions, the continuous MoS$_2$ layer transitions to a disordered array of individual islands, which are mostly triangular in shape and show the same apparent color (figure 1(c)).

The mineral MoS$_2$ flakes used for comparison are prepared using the mechanical exfoliation method well-established for graphene. A p-doped silicon wafer with 300 nm SiO$_2$ layer and lithographically defined metal markers is used as a substrate. An optical microscope is used to identify flakes that contain single or few layer-regions based on the optical contrast [7, 16], the thickness of individual exfoliated flakes is determined by Raman spectroscopy based on the frequency difference of the characteristic Raman modes E$_{2g}$ and A$_{1g}$ [17, 18]. Mechanical exfoliation yields a small number of single- and few-layer MoS$_2$ flakes with a size of several micrometers. Figure 1(d) shows a typical optical microscope image of flakes prepared by mechanical exfoliation, the larger-magnification image in figure 1(e) contains a flake with a single-layer region.

2.2. Optical spectroscopy

Raman spectroscopy measurements are performed at room temperature. For this, we utilize a microscope setup, in which a 532 nm cw laser is coupled either into a 100 × (1 µm laser spot size) or a 20 × (3 µm laser spot size) microscope objective, which also collects the scattered light in backscattering geometry. The scattered light is recorded using a grating spectrometer equipped with a cooled charge-coupled device sensor. A longpass filter is placed at the entrance of the spectrometer to suppress elastically scattered laser light. The sample is mounted on a motorized XY table and scanned under the microscope. Room-temperature PL measurements are performed using the same setup. For low-temperature PL measurements, the samples are mounted in vacuum on the cold finger of a small He-flow cryostat, which can also be scanned under the microscope. In PL and Raman scanning experiments, full spectra are collected for sample positions defined on a square lattice. For all the scans performed on the CVD-grown sample, the 20x microscope objective was...
utilized. An excitation density of about 170 kW cm\(^{-2}\) is used in the Raman and PL experiments, unless specified otherwise. In order to extract information from these spectra, an automated fitting routine is employed, which yields the integrated intensity, spectral position and full width at half maximum (FWHM) of the characteristic PL and Raman spectral features. To study valley polarization effects, near-resonant excitation is employed in the PL setup. For this, a tunable, frequency-doubled pulsed fiber laser system is utilized. Its center wavelength is tuned to 633 nm. This laser is circularly polarized by a quarter-wave plate and coupled into the microscope system. A longpass filter is utilized to suppress scattered laser light, and the circular polarization of the PL is analyzed using a second quarter-wave plate and a linear polarizer placed in front of the spectrometer.

### 3. Results and discussion

#### 3.1. Photoluminescence

First, we discuss the PL spectra of the CVD-grown films and compare them to those of exfoliated MoS\(_2\). Pronounced PL is observed only on single-layer MoS\(_2\) flakes; PL yield for two and more layers of MoS\(_2\) is significantly reduced and red-shifted with increasing number of layers [3, 4]. The characteristic PL emission observed in single-layer MoS\(_2\) at room-temperature stems from the A exciton peak, which corresponds to a direct transition from the conduction band minimum to the uppermost valence band maximum at the K points in the Brillouin zone.

Figure 2\((a)\) shows a false color plot of the A exciton PL intensity for the whole CVD-grown sample. The corresponding scanning PL measurement was performed using a step size of 250 \(\mu\)m. We note that strong PL, with an integrated intensity comparable to that of exfoliated flakes, is observed in two strip-like regions on the sample, each of which is about 8 mm long. The width of these strips cannot be discerned directly due to the low resolution of the large-area scan shown in figure 2\((a)\). To study these regions in more detail, high-resolution scans were performed in the area denoted by the orange square in figure 2\((a)\). False color plots created from such a scan, with a scan region of 2 mm and a step size of 50 \(\mu\)m, are shown in figure 2\((b)\). This scan was performed at liquid-helium temperature. From this scan, we can determine the width of the region showing strong PL to be on the order of 200 \(\mu\)m, in agreement with the width of the regions showing homogeneous contrast observed in the optical microscopy. We also note that the increase of the PL intensity is gradual at the upper boundary of this region (corresponding to the transition from a monolayer covered with additional material to a bare monolayer of MoS\(_2\)), while it drops sharply at the bottom (corresponding to the transition from continuous monolayer to isolated islands and bare substrate). This behavior is also seen in the higher-resolution line scan of the PL intensity shown in figure 2\((c)\). Thus, the areas of the CVD-grown samples which show PL comparable to single-layer MoS\(_2\) flakes exceed the typical size of exfoliated flakes by several orders of magnitude, and allow for optical spectroscopy experiments without the use of high-resolution microscope objectives.

Now, we investigate the PL spectra in more detail. The PL data obtained in the scan shown in figure 2\((b)\) allows us to extract the spectral position of the A exciton peak as a function of position on the sample. This is shown in figure 3\((a)\) as a false color plot. We clearly see that the peak position shifts by more than 30 meV in the region which emits strong PL. This shift is pronounced in the direction perpendicular to the long axis of the region, while the peak position is nearly constant along the long axis. Individual spectra measured at different positions of the scan region are shown in figure 3\((b)\), the positions P1–P3 are separated by 100 \(\mu\)m along the y direction. The vertical lines in figure 3\((b)\) serve as a guide to the eye to mark maximum and minimum A exciton peak positions. The spectral shift indicates that the growth conditions, and the corresponding microscopic properties of the MoS\(_2\) layer, change along the y direction. Three effects may contribute to this large spectral shift. It was shown theoretically by several groups that the band gap of MoS\(_2\) can be strongly influenced by the application of strain, where biaxial compressive strain increases the band gap, while biaxial tensile strain decreases the band gap. The calculated values for band gap shift as a function of biaxial strain range from 105 to 190 meV per cent [19–22]. A variation of the local biaxial strain can be generated during the growth process, which yields a temperature gradient across the substrate as the furnace is...
switched off. The observed shift of about 30 meV would correspond to a change of the local biaxial strain between 0.15% and 0.3%.

Additionally, it is very likely that there is a gradient of the Mo and S concentration across the substrate during growth, which may lead to local changes of the film composition. For example, a variation of the concentration of S vacancies would change the local carrier density. In n-doped MoS₂ layers, both, negatively charged excitons (trions) and neutral A excitons can be observed, with a binding energy of about 20 meV for the trions [23]. These two emission peaks typically merge due to their large linewidths, and the peak position of the resulting PL emission then depends on the relative contributions by excitons and trions. Transport measurements on our CVD-grown films indicate n-type conductivity [24], so that the observed PL emission may stem from a superposition of neutral and charged excitons, with a gradual change of the spectral weight due to changing carrier concentration. There may also be a locally varying deposition of amorphous S or Mo during growth, which would change the dielectric environment and modify the Coulomb screening. Recent calculations have yielded a very large binding energy of 0.85 eV for the A exciton in MoS₂ [6]. The exciton binding energy is proportional to $\epsilon^{-2}$, therefore a change of the effective dielectric constant $\epsilon$ by 2% would account for a spectral shift of the A exciton emission of 30 meV.

In addition to the shift of the A exciton peak position, we also note that the FWHM of the peaks changes as a function of position, with values between 80 and 60 meV. Direct comparison of these PL spectra with a typical spectrum obtained from a single-layer MoS₂ flake at liquid-helium-temperatures demonstrates that the exfoliated MoS₂ has a significantly lower FWHM of about 37 meV. This indicates a larger inhomogeneous broadening of the A exciton transition in the CVD-grown sample due to local disorder, which cannot be resolved with the spatial resolution obtained in the PL experiment. In exfoliated MoS₂ flakes at low temperatures, we also observe a second, lower-energy PL peak (S exciton), which is associated with localized excitons bound to surface adsorbates [25, 26]. This peak is also observed in some areas of the CVD-grown sample, e.g., at position P3 and weakly at position P2, yet absent on the majority of the regions emitting strong PL. A complete suppression of this S exciton peak was previously observed in experiments on oxide-covered MoS₂ flakes [26], its absence on most of the CVD-grown sample may therefore indicate partial coverage of the CVD monolayer with additional material.

Next, we discuss the temperature dependence of the PL emission. Figure 4(a) shows a series of PL spectra measured on the CVD-grown sample for different sample temperatures. In order to avoid thermal drift of the sample position during changes of the temperature, a two-dimensional PL scan was performed at each temperature, and the spectra shown in figure 4(a) were created from these scans by averaging data in a 300 μm wide region. We note that the maximum of the A exciton emission redshifts by 35 meV in the temperature range from 4 K to 300 K, as shown in figure 4(b). Additionally, the spectral width of the A exciton emission increases with increasing temperature. By contrast, in exfoliated MoS₂ flakes, we observe a spectral redshift of the A exciton peak by 72 meV in the same temperature range. The redshift indicates a temperature-induced reduction of the band gap due to thermal expansion of the crystal lattice. The fact that the redshift is far less pronounced in the CVD-grown sample shows that the CVD-grown MoS₂ film strongly adheres to the SiO₂.
excitation of electron–hole pairs either at the K leads to optical selection rules which allow for valley-selective points in the Brillouin zone [5], depending on the photon substrate, which has a very small thermal expansion coefficient measured as a function of excitation density at room temperature. Figure 5(a) shows two PL spectra collected for co- and contra-circular excitation/detection helicities. The star symbols indicate the circular polarization degree of the PL as a function of energy.

Figure 5. (a) Normalized PL spectra of the CVD-grown sample measured as a function of excitation density at room temperature. (b) Polarization-resolved PL spectra measured at liquid-helium temperature under near-resonant, circularly-polarized excitation, for co- and contra-circular excitation/detection helicities. The star symbols indicate the circular polarization degree of the PL as a function of energy.

Excitation-density-dependent PL measurements of the CVD-grown sample performed at room temperature (see figure 5(a)) using the 532 nm laser show no significant changes of the spectral shape, A exciton peak position or linewidth from 2 kW cm$^{-2}$ up to about 100 kW cm$^{-2}$. For further increases of the excitation density, we observe the emergence of a second, higher-energy, PL peak, which becomes more pronounced as the excitation density is increased further. This peak corresponds to the B exciton (direct transition from the conduction band minimum to the lower valence band maximum at the K points in the Brillouin zone.) The emergence of the B exciton peak at high excitation densities is due to state-filling effects: the nonresonant excitation leads to the formation of both A and B excitons. For low excitation densities, however, most of the holes can relax to the upper valence band before photocarrier recombination, so that only A exciton PL emission is observed. At high excitation densities, the occupation of the upper valence band reduces the available states for this relaxation process, so that B exciton PL emission emerges. A similar behavior was recently observed also in exfoliated monolayer MoS$_2$ flakes [29].

Finally, we discuss the circular polarization of the PL of the CVD-grown sample at low temperatures. In monolayer MoS$_2$, the lack of an inversion center in the crystal structure leads to optical selection rules which allow for valley-selective excitation of electron–hole pairs either at the K$^+$ or the K$^−$ points in the Brillouin zone [5], depending on the photon helicity. If this optically generated valley polarization persists on the timescale of the photocarrier recombination time, it is directly observable in the circular polarization degree of the PL [30–33]. Figure 5(b) shows two PL spectra collected for co- and contra-circular excitation and detection under near-resonant excitation using the frequency-doubled pulsed fiber laser system. In this experiment, the high-energy part of the A exciton emission is blocked by the longpass filter which is used to suppress the excitation laser. For co-circular excitation and detection, we note a significantly larger PL intensity than for contra-circular excitation and detection. From the two spectra, the circular polarization degree of the PL is calculated by dividing the difference of the PL intensities by their sum. For this, the PL intensities are spectrally averaged in 15 meV wide windows to reduce the noise. The circular polarization degree is plotted as a function of PL energy in figure 5(b). We find a near-constant value of more than 40% in the entire spectral range of the A exciton emission that is observable. This value is comparable to values we observe in exfoliated monolayer MoS$_2$ flakes under similar experimental conditions (not shown). Recently, it was demonstrated that the circular polarization degree strongly depends on the excess energy provided by the excitation laser [34]. The large circular polarization degree of the PL we observe is a clear indication that photocarrier excitation and recombination in the CVD-grown films follow the same selection rules as in mineral MoS$_2$. Therefore, these films will be highly useful for future studies of coupled spin and valley physics in monolayer MoS$_2$.

3.2. Raman spectroscopy

Now, we discuss the Raman spectra of the CVD-grown films and compare them to those of exfoliated MoS$_2$. For MoS$_2$ prepared by mechanical exfoliation, Raman spectroscopy is an excellent tool to map the number of layers in individual flakes. The most prominent Raman modes that can be observed in MoS$_2$ under nonresonant excitation are the E$_{2g}$ mode, which corresponds to an in-plane optical vibration of Mo and S atoms, and the A$_{1g}$, which is an out-of-plane optical vibration of the S atoms. The frequencies of both modes depend on the number of layers: while the A$_{1g}$ mode stiffens with additional layers, the E$_{2g}$ mode anomalously softens with additional layers [17]. This softening is attributed to enhanced dielectric screening [18]. Thus, the difference $\omega_{A_{1g}} - \omega_{E_{2g}}$ between the two modes can be used as a fingerprint for the number of layers. This technique is well-suited to differentiate between monolayers and flakes of up to four layers, while for thicker layers the mode positions approach the bulk limit. Thicker flakes can be differentiated by analyzing the frequency of the Raman shear mode, which also shows a strong dependency on the number of layers [35–37]. Recently, the effects of strain and carrier density on the Raman modes of MoS$_2$ were studied by several groups. Hui et al. [38] observed a blueshift of E$_{2g}$ and A$_{1g}$ modes for biaxial compressive strain, while calculations show a redshift of both phonon modes with biaxial tensile strain [39]. In a top-gated MoS$_2$ transistor device, a clear dependency of the mode positions and amplitudes on the carrier density was observed [40]. With increasing carrier density, the A$_{1g}$ mode softens, while the E$_{2g}$ mode remains unaffected. Additionally, the relative intensity of the Raman modes changes. For low carrier density, the A$_{1g}$ mode is more intense than the E$_{2g}$ mode, for high carrier density, the intensity ratio reverses.

In order to determine the information that can be extracted from Raman spectra on the CVD-grown films, we performed...
The vertical lines mark the positions of A1 in the same positions of the CVD-grown film as shown in (a). They serve as guide to the eye. For each spatial position, both, Raman and PL spectra were measured at room temperature. Figure 7.

Figure 6. (a) False color plot of the A exciton PL intensity as a function of position on the CVD-grown sample measured at room temperature. (b) False color plot of the frequency difference of the A1g and E1g Raman modes as a function of position for the same scan area as shown in (a).

We now discuss the Raman spectra in more detail. Figure 7(a) shows four Raman spectra collected at different positions of the CVD-grown film and on exfoliated monolayer (ML) and bulk-like flakes. All spectra are normalized to the amplitude of the A1g mode. The vertical lines mark the positions of A1g and E1g in an exfoliated monolayer and serve as guide to the eye. (b) PL spectra measured on the same positions of the CVD-grown film as shown in (a).

In scanning Raman experiments, we observe a substantial shift of the A exciton peak as a function of strain [38, 39]. A variation of biaxial strain in the CVD-grown film from position Y1 to Y4 cannot explain the observed frequency difference change, as both E- and A-modes were shown to shift in the same direction as a function of strain [38, 39]. From the different evolution of ωDiff and PL intensity we observe in the CVD-grown film, we infer that the carrier density in the CVD-grown film is lower than that observed in exfoliated flakes, the opposite is observed. From this observation, we can infer that the carrier density in the CVD-grown film may be significantly smaller than in the exfoliated flake [40]. Additionally, we note that the linewidths for both Raman modes in the CVD-grown film are larger than in the exfoliated flake, and the E1g mode is asymmetric. The mode positions for the CVD-grown film are in between the values observed for monolayer and bulk-like exfoliated flakes. A small increase of the frequency difference ωDiff (less than 0.5 cm⁻¹) is observed for the transition from isolated islands (Y1) to the film region showing the largest PL signal (Y2). The regions which emit less PL show further increase of the frequency differences, with position Y4 having a value of ωDiff = 25 cm⁻¹, comparable to bulk-like exfoliated MoS2. For exfoliated MoS2, however, a drastic reduction of the PL emission intensity, accompanied by a large redshift of the PL peak position, is observed for the change from monolayer to bilayer [3, 4] and thicker flakes. A variation of biaxial strain in the CVD-grown film from position Y1 to Y4 cannot explain the observed frequency difference change, as both E- and A-modes were shown to shift in the same direction as a function of strain [38, 39]. From the different evolution of ωDiff and PL intensity we observe in the CVD-grown film, we infer that the carrier density in the CVD-grown film is lower than that observed in exfoliated flakes, the opposite is observed. From this observation, we can infer that the carrier density in the CVD-grown film is lower than that observed in exfoliated MoS2. Some regions of the CVD-grown film also show PL emission associated with excitons bound to surface adsorbates. Temperature-dependent PL measurements reveal a small redshift of the A exciton peak, indicating that the CVD-grown MoS2 adheres strongly to the SiO2 substrate. In low-temperature PL experiments under near-resonant excitation, we observe a large circular polarization degree of the PL, commensurate with an optically induced valley polarization.

In conclusion, we have used Raman and PL spectroscopy to directly compare CVD-grown MoS2 to exfoliated flakes. The CVD process used to prepare our films yields large areas which show PL comparable to monolayer MoS2 flakes. In low-temperature PL measurements, we observe a spectral shift of the A exciton peak as a function of position. The linewidth of the A exciton is larger than that observed in exfoliated MoS2. Some regions of the CVD-grown film also show PL emission associated with excitons bound to surface adsorbates. Temperature-dependent PL measurements reveal a small redshift of the A exciton peak, indicating that the CVD-grown MoS2 adheres strongly to the SiO2 substrate. In low-temperature PL experiments under near-resonant excitation, we observe a large circular polarization degree of the PL, commensurate with an optically induced valley polarization.

In scanning Raman experiments, we observe a substantial shift of the characteristic Raman modes of MoS2 in the region of the CVD-grown film which emits strong PL. This behavior differs from the Raman spectra observed in exfoliated MoS2, where optical microscope, while Y2–Y4 are positions on different parts of the monolayer-like region. Let us first compare the Raman spectra to those collected from exfoliated flakes. All spectra shown in figure 7(a) are normalized to the A1g mode amplitude. We clearly see that in the CVD-grown sample, the E1g amplitude is smaller than the A1g, while in the exfoliated flakes, the opposite is observed. From this observation, we can infer that the carrier density in the CVD-grown film may be significantly smaller than in the exfoliated flake [40]. Additionally, we note that the linewidths for both Raman modes in the CVD-grown film are larger than in the exfoliated flake, and the E1g mode is asymmetric. The mode positions for the CVD-grown film are in between the values observed for monolayer and bulk-like exfoliated flakes. A small increase of the frequency difference ωDiff (less than 0.5 cm⁻¹) is observed for the transition from isolated islands (Y1) to the film region showing the largest PL signal (Y2). The regions which emit less PL show further increase of the frequency differences, with position Y4 having a value of ωDiff = 25 cm⁻¹, comparable to bulk-like exfoliated MoS2. For exfoliated MoS2, however, a drastic reduction of the PL emission intensity, accompanied by a large redshift of the PL peak position, is observed for the change from monolayer to bilayer [3, 4] and thicker flakes. A variation of biaxial strain in the CVD-grown film from position Y1 to Y4 cannot explain the observed frequency difference change, as both E- and A-modes were shown to shift in the same direction as a function of strain [38, 39]. From the different evolution of ωDiff and PL intensity we observe in the CVD-grown film, we infer that the carrier density in the CVD-grown film is lower than that observed in exfoliated flakes, the opposite is observed. From this observation, we can infer that the carrier density in the CVD-grown film is lower than that observed in exfoliated MoS2. Some regions of the CVD-grown film also show PL emission associated with excitons bound to surface adsorbates. Temperature-dependent PL measurements reveal a small redshift of the A exciton peak, indicating that the CVD-grown MoS2 adheres strongly to the SiO2 substrate. In low-temperature PL experiments under near-resonant excitation, we observe a large circular polarization degree of the PL, commensurate with an optically induced valley polarization.

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similar shifts only occur for an increasing number of layers and a corresponding suppression of the PL due to changes in the band structure. Our observations indicate that the CVD-grown MoS$_2$ material is well-suited for future studies of exciton physics and valley effects. Further optimization of the growth process should yield even larger, homogeneous MoS$_2$ regions and may pave the way for device applications.

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