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Quantifying photoluminescence variability in monolayer molybdenum disulfide films grown by chemical vapour deposition

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## Abstract

Monolayer molybdenum disulfide (MoS<sub>2</sub>) is a promising candidate for inclusion in optoelectronic technologies, owing to its two-dimensional (2D) nature and resultant novel photoluminescence (PL). Chemical vapour deposition (CVD) is an important method for the preparation of large-area films of monolayer MoS<sub>2</sub>. The PL character of as-prepared monolayer MoS<sub>2</sub> must be well understood to facilitate detailed evaluation of any process-induced effects during device fabrication. We comparatively explore the PL emission from four different commercially available CVD-grown MoS<sub>2</sub> monolayer films. We characterize the samples via Raman and PL spectroscopy, using both single-spot and mapping techniques, while atomic force microscopy (AFM) is applied to map the surface structure. Via multipeak fitting, we decompose the PL spectra into constituent exciton and trion contributions, enabling an assessment of the quality of the MoS<sub>2</sub> monolayers. We find that the PL character varies significantly from sample to sample. We also reveal substantial inhomogeneity of the PL signal across each individual MoS<sub>2</sub> film. We attribute the PL variation to non-uniform MoS<sub>2</sub> film morphologies that result from the nucleation and coalescence processes during the CVD film development. Understanding the large variability in starting PL behaviour is vital to optimize the optoelectronic properties for MoS<sub>2</sub>-based devices.

# 1. Introduction

Two-dimensional (2D) materials have been the subject of significant research interest, sparked by the successful isolation of graphene in 2004 [1]. Graphene's characteristic lack of an intrinsic bandgap excludes its application in semiconducting electronic devices: transition metal dichalcogenides (TMDCs) have emerged as exciting 2D alternatives [2]. TMDCs assume an MX<sub>2</sub> stoichiometry, with a monolayer comprising hexagonally packed M atoms sandwiched between X atoms [3]. Strong in-plane covalent bonds exist between the M and X atoms, with comparatively weak van der Waals interactions holding the X-M-X layers together [4]. The weak inter-layer forces enable the facile isolation of individual monolayers, commonly achieved via simple mechanical exfoliation techniques. TMDCs exhibit exceptional physical and chemical properties at single-layer thickness, originating from reduced dimensionality and quantum confinement effects [5]. Bulk MoS<sub>2</sub>, a prototypical TMDC, possesses an indirect bandgap of ~1.2 eV [6], yet monolayer MoS<sub>2</sub> has a direct bandgap of ~1.8 eV [7, 8]. The direct bandgap dominates the optical properties of monolayer MoS<sub>2</sub> demands the scalable growth of high-quality, continuous single-layer films. As such, chemical vapour deposition (CVD) methods have enabled the synthesis of large-area monolayer MoS<sub>2</sub> [17–22] and MoS<sub>2</sub> nanotube heterostructures [23].

With a direct bandgap, monolayer MoS<sub>2</sub> emits a relatively strong photoluminescence (PL) signal, arising from the radiative relaxation of excitonic states [8]. The PL spectrum from CVD-monolayer MoS<sub>2</sub>, namely the



absolute intensity and energy position, is known to differ from that of mechanically exfoliated single-layer  $MoS_2$ and has been reported to vary widely from sample to sample [24–27]. However, a rigorous comparison of the PL properties from different commercially available CVD-grown single-layer  $MoS_2$  films has not been published in the literature. Moreover, the PL character of  $MoS_2$  is known to be sensitive to external perturbation, and the impact of any treatment on the optical properties of monolayer  $MoS_2$  is often quantified by relative changes in the absolute PL signal strength and shifts in the energy position of the spectrum [28–30]. To assess the true effect of an external treatment on the PL behaviour of monolayer  $MoS_2$ , the exact single-spot PL measurement location must be identified before and after treatment. With no such practice described in the literature, we cannot conclude that this is a commonly employed procedure. It is important to ascertain whether any apparent alteration of the PL emission is an effect of treatment or a result of a nonuniform PL signal across the  $MoS_2$ sample. In this work, we investigate the room temperature PL emission from several untreated commercially available large-area  $MoS_2$  monolayer films synthesised via chemical vapour deposition (CVD). We find significant sample-to-sample variation of the PL signal. We also detect sizeable inhomogeneity of the PL intensity across each nominally uniform  $MoS_2$  film, which we associate with variation in the CVD-grown  $MoS_2$ film morphology.

# 2. Experimental details

#### 2.1. Materials

Four 1 cm  $\times$  1 cm MoS<sub>2</sub> commercially available monolayer films (>99%) grown via CVD on SiO<sub>2</sub>/Si were acquired from two different suppliers, with two films obtained from each manufacturer. All films arrived in vacuum-sealed packaging and were stored in a desiccator following cleaving to minimise any ageing effects from exposure to an ambient environment. The four films are referred to as films X1, X2, Y1, and Y2. Each film was cleaved into nine smaller samples of approximately equal size, as illustrated schematically in figure 1.

#### 2.2. Raman and PL spectroscopy

Single-spot Raman and PL measurements were acquired from two random locations on each cleaved  $MoS_2$  sample, yielding 18 Raman and 18 PL spectra for each film. Film Y1 was an exception to this, where only one measurement site was used per sample, giving 9 Raman and 9 PL spectra. An average PL spectrum was then calculated for each of the four films. All data were collected at room temperature using a Renishaw inVia Reflex Raman microscope with a 532 nm excitation laser at 0.1% of maximum power (~0.18  $\mu$ W). Spectra were obtained in standard confocal mode using a 50× Leica objective lens with a numerical aperture of 0.75 and a grating with 1800 lines/mm. This optical configuration was also used to obtain optical micrographs of some regions of the films. The acquired single-spot Raman data were the sum of 4 accumulations of 5 s each, with the corresponding PL data the sum of 4 accumulations of 10 s each. PL mapping data were also recorded for one sample cleaved from film X2. Obtained in static mode with a step size of 0.5  $\mu$ m, each PL map was centred around 1.85 eV to yield an energy range of 1.78 eV–1.93 eV and capture the position of maximum absolute

intensity. The spectrum at each point was fitted with a single Lorentzian curve, with the peak intensity extracted and mapped as a function of position. All data were collected using the Renishaw WiRE 3.1 software package and any cosmic-ray features were removed from the spectra where necessary.

#### 2.3. Atomic force microscopy

Surface morphologies were visualised via atomic force microscopy (AFM) using a Bruker Dimension Icon in the PeakForce Tapping Mode with a ScanAsystAir tip (with a nominal tip length of 115  $\mu$ m, a tip radius of 2 nm and a spring constant of 0.4 Nm<sup>-1</sup>) [31]. The images were obtained at a scan rate of 0.5 Hz, with 256 lines per scan to achieve a suitable resolution. Image processing and analysis were performed in the Gwyddion 2.60 software package [32].

## 3. Results and discussion

We first discuss the expected morphology of a  $MoS_2$  monolayer film synthesised via CVD on a  $SiO_2/Si$  substrate. As outlined in figure 2(a), the CVD-growth of a  $MoS_2$  monolayer film on  $SiO_2/Si$  nucleates at defect sites on the substrate surface, with randomly orientated triangular domains of MoS<sub>2</sub> forming. These triangular regions coalesce to form a large-area continuous monolayer film with inter-domain grain boundaries [19, 33, 34]. Discontinuous regions and isolated domains will exist where complete monolayer closure is not achieved. This is evident in the films used in this study, as exemplified in figure 2(b) by the optical micrograph taken from a random region of film X1. While invisible to optical microscopy techniques, AFM mapping offers facile visualisation of inter-domain grain boundaries, as illustrated in figure 2(c). We utilise Raman spectroscopy and AFM to distinguish regions of continuous  $MoS_2$  monolayer film from voids that reveal the underlying  $SiO_2/Si$ substrate. As shown in figures 2(d) and (e), single-spot Raman measurements were taken at two locations on a sample cleaved from film X1 and are displayed in figure 2(f). Monolayer MoS<sub>2</sub> has a distinct Raman signature that comprises two characteristic vibrations: an in-plane  $E_{2g}^1$  mode at ~384 cm<sup>-1</sup> and an out-of-plane  $A_g^1$  feature at ~403 cm<sup>-1</sup> [35]. The separation between these two peaks,  $\Delta k$ , is a robust indicator of monolayer thickness and is typically ~18–21 cm<sup>-1</sup> [36, 37]. Hence, with no Raman modes observed from the triangular region in this wavenumber range, this feature is likely to be a void in the surrounding continuous monolayer MoS<sub>2</sub> film. To confirm this triangular area as the exposed SiO<sub>2</sub>/Si substrate, a height profile traced along its boundaries with the expected MoS<sub>2</sub> film was extracted from AFM image data, as shown in figure 2(g). From the obtained height trace, plotted in figure 2(h), a step of ~0.7 nm was estimated, in excellent agreement with the reported thickness of a MoS<sub>2</sub> monolayer [2, 8, 38]. All four sample types studied exhibit broadly similar characteristics to film X1.

Next, we illustrate the spatial non-uniformity of the PL signal across a CVD-grown MoS<sub>2</sub> monolayer film via PL mapping. In figure 3, we choose to compare two distinctly different regions of the same sample (X2). We first consider a region of continuous monolayer MoS<sub>2</sub> film for which the morphology is shown by the AFM image in figure 3(a), with the corresponding PL map presented in figure 3(b). It is evident that the maximum PL intensity varies considerably, even over a 12  $\mu$ m  $\times$  12  $\mu$ m area of relative structural uniformity. Some surface contamination is apparent, indicated by the presence of bright spots in the AFM image. However, with no corresponding features in the PL map, such contaminants are not thought to contribute significantly to the varying PL signal. To demonstrate the variation in the strength of the PL emission further, we extract two singlespot spectra from the PL mapping data, as plotted in figure 3(c). We propose that the spatial variation of the PL signal observed across the nominally uniform MoS<sub>2</sub> monolayer films studied here can be attributed to variations in the film morphology that result from the CVD synthesis [39]. Secondly, we consider a region of the same sample in which there remain triangular domains, which is expected when the coalescence of triangular domains has not happened completely in the previously introduced growth model (figure 1(a)). An AFM map of the region studied is shown in figure 3(d) and the resulting maximum PL intensity map is shown in figure 3(e). Spectra for two spots are shown in figure 3(f). Triangular domains of CVD-grown monolayer MoS<sub>2</sub> are known to exhibit nonuniform PL character, where the PL signal weakens and redshifts close to the domain edge [27, 40]. Since CVD-MoS<sub>2</sub> monolayer films originate from such domains, some spatial variation in the PL emission across the coalesced film can be anticipated, with a depleted PL intensity measured at structural defect sites and grain boundaries [27]. These expectations are in excellent agreement with our PL mapping measurements, where clear spatial inhomogeneity of the PL signal strength was observed across a MoS<sub>2</sub> monolayer film. An important conclusion from the data in figure 3 is that there are substantial variations in PL emission from a single MoS<sub>2</sub> film. The variations occur across a region of fully coalesced domains, as well as in regions in which complete coalescence has not occurred.

We now turn our attention to variations in PL emission from sample-to-sample. The average PL spectrum for each of our four distinct MoS<sub>2</sub> monolayer films is displayed in figure 4, with the corresponding range of PL

3



**Figure 2.** (a) Schematic representation of the CVD-growth of a  $MoS_2$  monolayer film on  $SiO_2/Si$ . The black arrows indicate the development of the film growth process. (b) Representative optical micrograph of a large-area region. (c) Representative topographic AFM image of a different area. (d) Optical micrograph of a third region. The red and blue spots highlight the locations where single-spot Raman and PL spectra were measured. (e) Topographic AFM image of the region displayed in (d). (f) Single-spot Raman spectra taken from two locations marked with blue and red dots in the region shown in (d) and (e). Where an appreciable Raman spectrum was obtained, the characteristic Raman peak separation is highlighted. (g) Enlarged topographic AFM image of the region highlighted by the black dashed square in (e). (h) Height profile measured along the magenta arrow in (g). All images in this figure are taken from regions cleaved from film X1.

spectra included in each mean calculation indicated by the shaded regions. As will be confirmed later, the measurements arose from areas known to be MoS<sub>2</sub> monolayers.

From film to film, we observe significant variation in the MoS<sub>2</sub> PL character. The intensity of the average PL signal from films X2 and Y2 is much greater than that emitted from films X1 and Y1, with an observed difference in maximum absolute intensity of up to an order of magnitude under identical PL exposure conditions. It is also clear that the PL emission differs between samples cleaved from the same film, with films X2 and Y2 both seen to exhibit large ranges of maximum PL intensities compared to their sister samples X1 and Y2 respectively. In addition to differences in the absolute PL signal strength, we found significant variation in the peak position. The average peak intensity obtained for each film occurred at an energy between 1.84 and 1.86 eV, while the full

4



range of peak energies extracted from the samples studied here spanned 1.83–1.89 eV. As plotted in figure 5, Raman spectra recorded at the same positions on each film show consistent  $E_{2g}^1$  to  $A_g^1$  peak separation,  $\Delta k$ , in the expected 18–22 cm<sup>-1</sup> range, indicating monolayer thickness in each case [36, 37]. Hence, we can exclude differing layer thicknesses or the presence of multilayer regions as the origin of the observed variation in PL response.

The overall PL spectrum from as-prepared monolayer  $MoS_2$  is widely reported to be the superposition of three individual contributions: A and B exciton transitions, and a trion (A<sup>-</sup>) emission [41]. The excitonic features are associated with transitions between the conduction band minimum (CBM) and the spin-orbit split valence band maximum (VBM) at the K-point [42]. The negatively charged trion, a three-body quasiparticle comprising an exciton and an additional electron, results from a native excess of electrons in monolayer MoS<sub>2</sub>. The relative ratios of the intensities of the constituent PL features can reveal insight into the respective densities of charge carriers and defect states in a monolayer MoS<sub>2</sub> sample. The ratio of the intensities of the trion and A exciton contributions, A<sup>-</sup>/A, can be related to the excess electron concentration in monolayer MoS<sub>2</sub>, since trion formation is encouraged by a greater abundance of electrons [43]. McCreary et al suggested that the ratio of the intensity of the B excitonic peak to that of the A exciton, B/A, could be an indicator of the quality of a MoS<sub>2</sub> monolayer [25]. The association of this quantity to the defect state density can be rationalised in terms of the differing dynamics of the two neutral excitons. Since the A excitonic complex is much longer-lived than the B exciton, the A exciton dominates the PL signal in untreated MoS<sub>2</sub>, yielding a small B/A ratio. Defect states can facilitate the non-radiative relaxation of both A and B excitons. Any defect-induced attenuation of the B excitonic signal will be significantly less than that of the A exciton, due to its much shorter lifetime. Therefore, a larger B/A value implies a higher defect state density and lower-quality sample. It is important to note that in McCreary et al's analysis of the excitonic ratio, the overall MoS<sub>2</sub> PL spectrum is treated as the superposition of two Lorentzian curves, A and B, with any trionic contribution accounted for by the A peak. Here, since we employ a three-peak deconvolution, we define our exciton ratio as  $B/(A+A^{-})$ .

To reveal the constituent spectral components, we performed a multipeak Lorentzian fit of each PL signal obtained in this study. The energies of the individual exciton and trion peaks were constrained within approximate initial estimates, with their linewidths allowed to vary freely. A representative deconvoluted PL spectrum is shown in figure 6(a) and outlines the typical trion and exciton emissions. The  $B/(A+A^-)$  and  $A^-/A$  ratios were determined for each individual sample cleaved from the four  $MoS_2$  monolayer films and comparisons of these values are plotted in figures 6(b) and (c), respectively, with the mean averages highlighted by the black dashed lines. Each  $MoS_2$  monolayer film was found to possess a different mean  $B/(A+A^-)$  ratio, within a range of 0.29 to 0.65. It can therefore be deduced that the  $MoS_2$  monolayer quality varies from film to film. Additionally, a wide range of  $B/(A+A^-)$  values was observed between samples cleaved from the same film.



**Figure 4.** Average PL spectra for monolayer  $MoS_2$  films (a) X1, (b) X2, (c) Y1 and (d) Y2. The solid line corresponds to the average PL spectra, whilst the shaded area corresponds to the range of PL spectra measured. For films X1, X2 and Y2, 18 points on each film were measured, with 9 measurements recorded from film Y2.



**Figure 5.** Comparison of the (a) position of the  $E_{lg}^1$  Raman peak, (b) position of the  $A_g^1$  Raman peak and (c) the  $E_{lg}^1$  to  $A_g^1$  Raman peak separation ( $\Delta k$ ) for each monolayer MoS<sub>2</sub> sample cleaved from the four films X1, X2, Y1 and Y2. The black dashed lines indicate the mean values, with the error bars representative of the standard deviation associated with each dataset.

To further examine the extent of variability in the  $B/(A+A^-)$  ratio across each  $MoS_2$  monolayer film, we calculated the standard deviation in the  $B/(A+A^-)$  values,  $\sigma_{B/(A+A^-)}$ . From this, we determined four coefficients of variation for each average  $B/(A+A^-)$  ratio,  $CV_{B/(A+A^-)}$ , defined as the ratio of the standard deviation to the mean [44]. A larger  $CV_{B/(A+A^-)}$  indicates a greater degree of variation. The  $CV_{B/(A+A^-)}$  values associated with each film are detailed in table 1. It is evident that the level of variation in the  $B/(A+A^-)$  ratio is different for each of the four  $MoS_2$  monolayer films, with films X1 and X2 displaying the most variable  $B/(A+A^-)$  values. The significant  $CV_{B/(A+A^-)}$  suggests an inhomogeneous distribution of defect states across each film. From the  $A^-/A$ 



**Figure 6.** (a) Representative deconvoluted PL spectrum taken from a single site on a sample cleaved from film X2, showing the excitonic (A and B) and trionic (A<sup>-</sup>) spectral components. (b) Comparison of the  $B/(A+A^-)$  ratios calculated for each monolayer  $MoS_2$  sample cleaved from the four films X1, X2, Y1 and Y2. (c) Comparison of the average A<sup>-</sup>/A ratios calculated for each monolayer  $MoS_2$  sample cleaved from the four films X1, X2, Y1 and Y2. (d) Comparison of the average FWHM of the dominant PL peak calculated for each monolayer  $MoS_2$  sample cleaved from the four films X1, X2, Y1 and Y2. (d) Comparison of the average FWHM of the dominant PL peak calculated for each monolayer  $MoS_2$  sample cleaved from the four films X1, X2, Y1 and Y2. (d) Comparison of the average FWHM of the dominant PL peak calculated for each monolayer  $MoS_2$  sample cleaved from the four films X1, X2, Y1 and Y2. (d) Comparison of the average FWHM of the dominant PL peak calculated for each monolayer  $MoS_2$  sample cleaved from the four films X1, X2, Y1 and Y2. In (b), (c) and (d), the black dashed lines indicate the mean values, with the error bars representative of the standard deviation associated with each dataset.

 $\label{eq:table_standard} \mbox{Table 1. Mean values, standard deviations, and coefficients of variation for the $B/(A+A^-)$ and $A^-/A$ ratios of the four $MoS_2$ monolayer films studied here.}$ 

Film	Mean B/(A+A <sup>-</sup> )	$\sigma_{\rm B/(A+A)}$	$CV_{\rm B/(A+A^-})$	Mean A <sup>-</sup> /A	$\sigma_{\rm A}{/{\rm A}}$	CV <sub>A</sub> -/A
X1	0.18	0.076	0.42	1.77	1.66	0.94
X2	0.15	0.089	0.59	2.89	3.68	1.27
Y1	0.17	0.027	0.16	2.31	2.40	1.04
Y2	0.05	0.004	0.07	2.16	3.33	1.54

ratio, we elucidate information on the carrier densities in the four  $MoS_2$  films. With a mean  $A^-/A$  ratio > 1, each film is concluded to possess an excess of electrons. The  $A^-/A$  ratio can be related to the overall absolute PL intensity, with a higher ratio thought to correspond to a weaker PL signal since trions are more susceptible than neutral excitons to relaxation *via* non-radiative processes [41, 45]. As in our evaluation of the variation in each  $B/(A+A^-)$  ratio, we apply a similar analysis to the  $A^-/A$  ratio and the corresponding  $CV_{\overline{A}/A}$  values are also shown in table 1. Like the  $B/(A+A^-)$  ratio, a significant coefficient of variation was calculated for each  $A^-/A$  intensity ratio, further supporting the proposal of a spatially varying PL character across each monolayer film. With variable  $A^-/A$  ratios, it can be deduced that the charge carrier density varies across all the distinct  $MoS_2$  monolayer films studied here. Hence, *via* assessment of the constituent PL peak ratios, we have revealed both the

Table 2. Mean values, standard deviations, and coefficients of variation for the FWHM of the A exciton emission from the four  $MoS_2$  monolayer films studied here.

Film	Mean FWHM (meV)	$\sigma_{\rm FWHM}$ (meV)	CV <sub>FWHM</sub>			
X1	84	85	1.02			
X2	87	19	0.22			
Y1	109	9	0.08			
Y2	74	20	0.28			

carrier- and defect-state densities to vary widely. This variation is evident not only from film to film but also between samples cleaved from the same supposedly uniform MoS<sub>2</sub> film.

In addition to the excitonic intensity ratio, the linewidths of the constituent features in a  $MoS_2$  PL spectrum can be used to assess CVD-MoS<sub>2</sub> quality, since defects are known to broaden the PL signal from monolayer TMDCs [46]. To perform line shape analysis, we calculate the mean full width at half maximum (FWHM) of the dominant PL peak for each monolayer MoS<sub>2</sub> film studied here. A comparison of the four mean FWHM values is presented in figure 6(d). As in the preceding discussion of the A<sup>-</sup>/A and B/(A+A<sup>-</sup>) ratios, we computed the standard deviation,  $\sigma_{FWHM}$ , and corresponding coefficient of variation,  $CV_{FWHM}$ , and these are given in table 2. We find considerable film-to-film variation in the width of the major MoS<sub>2</sub> PL peak, confirming the varying sample quality of the MoS<sub>2</sub> films studied here. It can be seen in figure 6(d) that there is a wide range of possible linewidths associated with each mean FWHM and this serves as further evidence of non-uniformity of the PL character across each monolayer film. This is further exemplified by the coefficients of variation, with some variation in the FWHM determined across each MoS<sub>2</sub> film. The degree of this variation differs from film to film, with relatively minor variation in the width of the PL peak calculated for samples cleaved from film Y1 but more substantial variation in width calculated for films X1, X2 and Y2.

We have demonstrated the ability of PL mapping to reveal the varying PL intensity across MoS<sub>2</sub> monolayer films. PL mapping is therefore an important tool for assessment of any treatment-induced modification of the PL efficient and doping level of CVD-monolayer MoS2. The impact of an external treatment of monolayer MoS2 on the PL character is commonly reported in terms of a relative enhancement of the absolute PL signal strength, determined via pre- and post-treatment single-site measurements [47-50]. Determination of the true enhancement of the absolute MoS<sub>2</sub> PL intensity demands precise measurements at the same location before and after treatment. We cannot assume that this practice is commonplace in the literature, since is not typically explicitly stated. Some studies seek to overcome this problem by averaging the MoS<sub>2</sub> PL signal over several measurement spots. However, our PL mapping results have shown the PL emission to vary significantly across a given MoS<sub>2</sub> sample, so extreme PL intensities will skew any average and yield inaccurate enhancement values. PL enhancement factors have also been quoted as an integrated area-corrected number [51, 52], yet this approach can generate inflated enhancements that further confuse the true value. As such, the spatial variation in the PL behaviour of MoS<sub>2</sub> monolayer films revealed here limits the reliability of enhancement factors as a metric for the performance of treated MoS<sub>2</sub>. PL mapping eradicates the issues that arise from a spatially variable PL emission, offering visualisation of the PL intensity over a specified region of a given MoS<sub>2</sub> sample and enabling facile evaluation of the result of any treatment. Moreover, PL mapping can be used in conjunction with other imaging techniques, namely topological AFM, to infer the PL response from different features in the film morphology. The value of PL mapping methods for an accurate description of treatment-modified PL signals from MoS<sub>2</sub> monolayer films is clear.

## 4. Conclusions

We have performed a systematic study into the PL emission from  $MoS_2$  monolayer films, finding considerable variation from region-to-region of a single sample, and even larger variation (up to an order of magnitude in absolute PL intensity maximum) between four nominally equivalent commercial sample types. Differing  $B/(A+A^-)$  ratios for each  $MoS_2$  film indicate varying degrees of sample quality. We also found significant spatial variation of the PL intensity across each nominally continuous  $MoS_2$  film. This was ascribed to the non-uniform  $MoS_2$  film morphology that results from the CVD growth process. This work provides valuable insight into the PL character of commercially available CVD-grown  $MoS_2$  monolayer films, highlighting sizable variation in the PL behaviour of  $MoS_2$  monolayer films that must be considered prior to any treatment or device performance evaluation.

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## Data availability statement

The data that support the findings of this study are openly available at the following URL: https://wrap.warwick. ac.uk/182082. Requests for additional data should be made to the correspondence authors.

#### **Conflict of interest**

The authors have no conflicts to disclose.

#### **CRediT** authorship contribution statement

B F M Healy: conceptualization, formal analysis, investigation, methodology, writing the original draft, review and editing. S L Pain: conceptualization, investigation, formal analysis (supporting), draft editing. J Lloyd-Hughes: formal analysis, draft editing. N E Grant: conceptualization, supervision, draft editing. J D Murphy: conceptualization, funding acquisition, project administration, resources, supervision, draft editing.

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