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## Exfoliation and transfer of millimetre-sized MoS<sub>2</sub> flakes on arbitrary substrates

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Two-dimensional (2D) materials have the potential to strongly and sustainably influence technological development in the fields of optoelectronics, energy production and management, catalysis and more. One limiting factor that presently prevents the full exploitation of these materials is, however, the difficulty of obtaining large-scale, high-quality 2D samples on arbitrary substrates. In this work, we introduce a significant generalization of previously reported gold-assisted exfoliation techniques for TMDCs, marking a step forward towards the fabrication of macroscopic 2D material samples on arbitrary substrates. We achieved the successful production of millimetre-sized monolayer MoS<sub>2</sub> onto silica, PDMS, and both thermal and native oxidized silicon wafers. Moreover, our method simplifies previously reported gold-assisted exfoliation methods by removing substrate functionalization procedures and complex steps to achieve a reliable and reproducible procedure. The crystal quality of the monolayers was probed using XPS, Raman and photoluminescence spectroscopies, revealing a negligible presence of contaminants and defects in the samples. Furthermore, using imaging ellipsometry, we could investigate, on the millimetre scale, the sample morphology and the selectivity of the exfoliation process to produce single layer MoS<sub>2</sub> flakes. Finally, we further extended the capability of our exfoliation method by enabling the seamless transfer of large-area samples from PDMS to advanced substrates, unlocking new possibilities for large-scale 2D device fabrication.

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## Introduction

Ever since the first isolation of graphene,<sup>1</sup> 2-dimensional (2D) materials have become almost ubiquitous in condensed matter physics due to their amazing physical properties and ease of fabrication.<sup>2</sup> Among the large family of 2D materials, group-VI transition metal dichalcogenides (TMDCs) have drawn particular interest due to their semiconducting nature, excellent optoelectronic properties and widespread availability.<sup>3</sup> Their 2D character indeed promotes the appearance of unique properties, among which are the indirect-to-direct band gap transition when going from bulk to monolayer,<sup>4</sup> very high exciton binding energies<sup>5</sup> and spin-valley coupling in the electronic structure.<sup>6</sup> A wide range of possible applications arise from these outstanding properties, laying the ground for a possible technological revolution in fields such as electronics, optics, and computing.<sup>7,8</sup>

One of the major obstacles for the full exploitation of 2D systems has always been the lack of a scalable and reliable method for the deterministic production of spatially extended and high-quality monolayers. The most widely employed fabrication methods, such as the mechanical Scotch tape method,<sup>1</sup> liquid phase exfoliation,<sup>9</sup> and chemical vapor deposition,<sup>10</sup> all exhibit some intrinsic drawbacks in terms of either yield, scalability, or sample quality. Recent studies have shown great progress in this direction, reporting for example the wafer-scale growth of 2D materials, albeit with multiple translational grain boundaries;<sup>11–13</sup> a recently reported method based on 2D Czochochalski growth also promises to address these issues by producing almost defect-free extended monolayers,<sup>14</sup> but the true scalability and universality of these methods have yet to be fully demonstrated.

In general terms, however, the conceptual and practical simplicity of the mechanical exfoliation method remains unparalleled; for this reason, great curiosity surrounded the report of the breakthrough achievement of extended TMDC monolayer fabrication obtained by introducing metallic layers to complement the conventional Scotch-tape method.<sup>15–22</sup> The rationale behind this innovative approach is to perform the mechanical exfoliation of the TMDC from the parent crystal not directly by tape, but by means of a metallic layer, whose

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adhesion to the TMDC surface is stronger than the TMDC's interlayer adhesion.<sup>15</sup> Experiments showed that large monolayer (1L) TMDC areas could be peeled from the parent crystal in this way, significantly improving with respect to the original metal-free version.

Following early implementations,<sup>16,17,23</sup> metal-assisted exfoliation has been gradually improved and refined,<sup>19,20,24</sup> to the point of producing high-quality millimetre-sized monolayer TMDCs on selected substrates. The interaction between gold surfaces and TMDCs has been described in various ways.<sup>24</sup> Some studies characterize it as 'covalent-like quasi-bonding'<sup>25,26</sup> or 'mixed vdW-covalent',<sup>27</sup> while others refer to it as physisorption/chemisorption<sup>28</sup> or simply as a 'strong' interaction.<sup>15,29–32</sup> The roles of strain<sup>33,34</sup> and electrostatics<sup>35</sup> have been investigated. Beyond gold, several other metals have been shown to effectively exfoliate a variety of 2D materials.<sup>36–38</sup> Nevertheless, gold remains the primary focus of most studies, as it is resistant to oxidation and can be readily grown.<sup>27</sup>

Liu *et al.*, in particular, perfected gold-mediated exfoliation to the point of exfoliating large-area 1L samples onto non-metallic substrates,<sup>39</sup> seemingly addressing the issues and limitations that were present in early studies. A crucial aspect of their procedure was the exploitation of a thermal release tape (TRT) as a vector to mechanically sustain the sacrificial Au layer during the exfoliation of the TMDC and the transfer to the target substrate. Upon pressing TRT/Au/TMDC on a substrate of choice, the TRT was released by heating the system, and the sacrificial layer was etched, leaving the TMDC 1L behind. A later study by Petrini *et al.*,<sup>40</sup> however, despite strictly adhering to Liu's recipe, reported systematic cracking and wrinkling of the metal layer due to heat-induced stress of the TRT polymer, resorting to functionalization of the target substrate with (3-aminopropyl)triethoxysilane (APTES) in order to increase the adhesive force between the substrate and the TMDCs and circumvent the issue.<sup>40</sup> The procedure, albeit introducing an extra fabrication step, ensured improved mechanical stability of the TMDC/metal stack and successfully yielded large-area monolayers of TMDC, incidentally promoting stronger adhesion to the target substrate, a factor that may be beneficial in subsequent processes that require sample sonication<sup>41</sup> or detrimental when further transfer is required.

In order to be able to fabricate large-area 1L flakes of TMDCs on a large variety of substrates and leave open the possibility to further transfer them onto other systems, we developed an improved version of previously reported sacrificial gold layer-assisted exfoliation methods<sup>39,40</sup> that significantly extends the range of applicability of large-scale exfoliation of TMDCs. In detail, the surface functionalization of the target substrate was replaced with a simple reversible mechanical constraint, a rigid PDMS layer, introduced during the critical TRT-release stage, thereby altogether avoiding the need for intermediate additional chemical processes and additional fabrication steps that might compromise the sample quality. With this approach, we demonstrated the possibility of achieving large-area TMDC 1Ls on substrates as diverse as Si wafers with native (2 nm) and

thermal (300 nm) oxide, bulk fused silica and polydimethylsiloxane (PDMS). Moreover, by exploiting the successful exfoliation on PDMS, which distinguishes our method from nearly all previously reported metal-assisted exfoliation methods, we demonstrated that large-area 1L TMDC flakes could be further deterministically transferred onto a wide variety of other substrates with microscopic control over crystal positioning, exploiting well-established PDMS-based dry-transfer methods.<sup>42,43</sup>

## Exfoliation of large-area monolayer MoS<sub>2</sub>

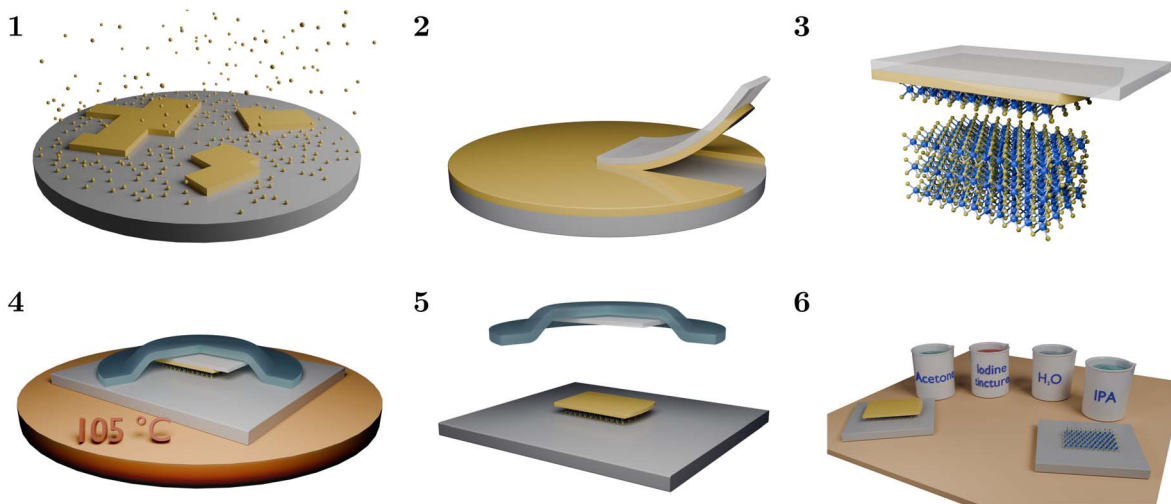
In this work, we focused on a natural crystal of MoS<sub>2</sub> as the parent bulk material for exfoliation, since its generalization to the many other 2D materials that are compatible with gold-assisted exfoliation is straightforward.<sup>25,39</sup> Since substrate compatibility was reported to be a delicate issue within existing gold-assisted exfoliation reports,<sup>40</sup> we chose to address different target substrates, namely: (i) p-type silicon (resistivity ~20 Ω cm) with native SiO<sub>2</sub> oxide (SiO<sub>2</sub>(nat)/Si); (ii) n-type silicon (resistivity ~20 Ω cm) with a thermally grown SiO<sub>2</sub> layer (SiO<sub>2</sub>(285 nm)/Si); (iii) fused silica; (iv) polydimethylsiloxane (Sylgard 184 with a 10:1 cross-linker/curing agent ratio), chosen as representative of different domains of future applications in the fields of electronics, optics and flexible devices. The choice of the doping type for the silicon wafers was mainly driven by the availability of the silicon wafers.

Fig. 1 shows the different steps of the exfoliation process. (1) A 150 nm-thick gold film was deposited *via* sputtering onto a Si wafer previously cleaned in oxygen plasma for 5 minutes at 50 W in order to remove contaminants on the surface; no adhesion layer was deposited in order to ensure easy peeling in the subsequent step. (2) A Nitto Denko Revalpha RA-95LS(N) thermal release tape (TRT) was then placed directly on the gold coated wafer and used to pick up a centimetre-sized piece of the gold film from the silicon wafer. (3) The TRT-supported gold film was pressed onto a freshly cleaved bulk MoS<sub>2</sub> crystal. This step was performed in the shortest time possible, in order to prevent the contamination of the gold surface and the MoS<sub>2</sub> crystal from ambient air.<sup>44</sup> The TRT/Au stack was then peeled off the bulk MoS<sub>2</sub>, stripping a monolayer MoS<sub>2</sub> in the process, and pressed onto the target substrate of choice.

At this stage, in order to release the sample, the TRT needs to be heated above 105 °C; normally, this results in the TRT bending away from the underlying substrate, lifting the thin MoS<sub>2</sub>/Au layer and preventing its transfer.<sup>40</sup>

In this work, we prevented the detrimental TRT buckling by (4) fully covering the MoS<sub>2</sub>/Au/TRT system by means of an adhesive rigid layer, thereby providing a mechanical constraint for the TRT and allowing it to lose adhesion while simultaneously maintaining the Au/MoS<sub>2</sub> assembly firmly in contact with the substrate. For our experiments, the constraint layer consisted of 1.5 mm-thick PDMS, which ensured surface cleanliness and prevented sample contamination. The PDMS





**Fig. 1** Step-by-step schematic representation of the exfoliation process. (1) A 150 nm thick gold film is deposited *via* sputtering onto a silicon wafer. (2) Part of the gold layer is peeled off from the wafer by means of the TRT. (3) The gold/TRT stack is exploited to exfoliate a MoS<sub>2</sub> monolayer from the bulk crystal. (4) The MoS<sub>2</sub>/Au/TRT system is laid on the desired substrate, pressed with a PDMS layer (light blue) and heated to promote the detachment of the TRT. (5) The MoS<sub>2</sub>/Au stack is detached from the TRT. (6) The gold layer is etched by sequentially dipping the substrate/MoS<sub>2</sub>/Au system in acetone, iodine tincture, H<sub>2</sub>O and IPA.

was prepared using Sylgard 184 with a 10 : 1 cross-linker/curing agent ratio and cured for 24 hours at 60 °C. The PDMS was placed directly on the MoS<sub>2</sub>/Au/TRT stack upon cutting a large enough region to fully cover the sample. (5) The PDMS/TRT stack was then removed, leaving the Au/MoS<sub>2</sub>/substrate system behind, and (6) the gold layer was dissolved by standard Au-etching procedures: the sample was first washed in acetone for a few minutes to remove any organic contamination from the tape, then the gold layer was etched using iodine tincture (1 : 4 : 40 I<sub>2</sub> : KI : H<sub>2</sub>O) and washed in DI water, in IPA, and finally dried using dry nitrogen.

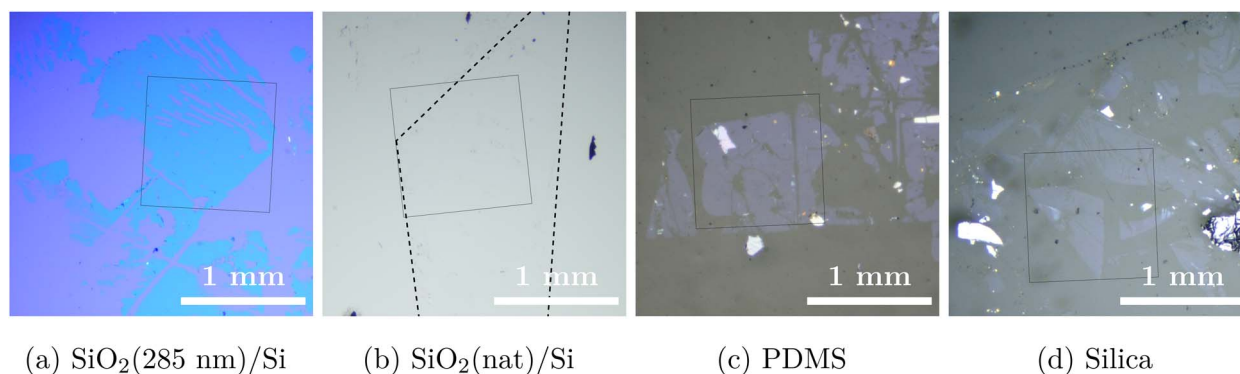
Steps (2) to (4) were performed under a controlled nitrogen atmosphere in order to minimize the humidity level during the exfoliation procedure, effectively reducing airborne contamination of the Au/MoS<sub>2</sub> interface and consequently improving the exfoliation throughput.<sup>44–46</sup>

In SI, Fig. S1 shows photos of the various steps of the exfoliation procedure.

## Imaging ellipsometry of large-scale monolayers

In Fig. 2, we show optical microscopy images of one representative sample for each substrate type. The samples on SiO<sub>2</sub>(285 nm)/Si, fused silica and PDMS exhibit an optical contrast sufficiently high to be located under a normal optical microscope, while the flakes exfoliated on Si wafers with native oxide are not readily observable, as indeed expected.

In order to overcome this complication and, above all, to quantitatively assess the sample morphology, we performed imaging spectroscopic ellipsometry (iSE) measurements, which allow probing of the optical properties of 2D materials<sup>47–49</sup> and ready detection of sample thickness variations with sub-nanometre precision, unambiguously identifying the number of exfoliated layers<sup>50</sup> of TMDC systems. Moreover, in contrast with atomic force microscopy (AFM), which has proved to be inefficient in measuring the thickness of monolayer



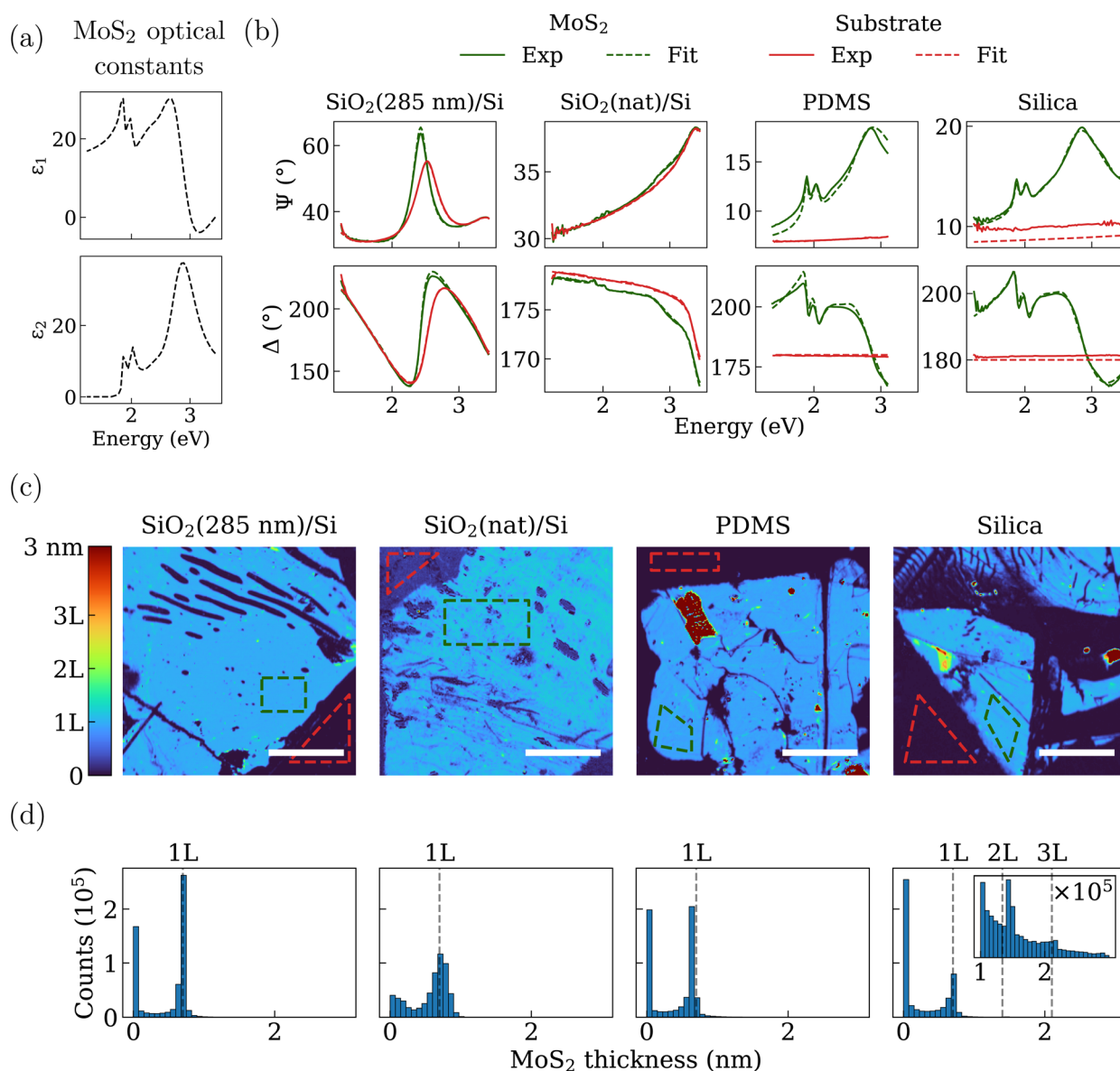
**Fig. 2** Optical microscopy of the samples exfoliated on different substrates, as indicated in the panel caption. (a) 285 nm SiO<sub>2</sub>/Si; (b) 2 nm SiO<sub>2</sub>/Si; (c) PDMS; (d) fused silica. The black rectangles indicate the area where imaging ellipsometry measurements were performed. In panel (b), where optical contrast is negligible, a black dashed line marks the approximate outline of the sample.



samples,<sup>51,52</sup> iSE thickness measurements are unaffected by the eventual presence of contaminants and by the interaction with the substrate that may lead to anomalous thickness values using AFM. Instead, the latter was used to image the surface topography and acquire information on the surface morphology of the samples. Fig. S4 displays the AFM map of a sample on SiO<sub>2</sub>(nat)/Si, showing the absence of contaminants on the MoS<sub>2</sub> surface, beside small unavoidable dust particles. Hyperspectral iSE measurements were performed by mapping the ellipsometric angles  $\Psi(\lambda)$  and  $\Delta(\lambda)$  at 129 different wavelengths spanning the near-UV, visible and near-infrared ranges, as

detailed in the Methods section. A few representative maps are reported in Fig. S2 and S3.

The experimental data were subsequently analysed by means of a layer-stack model that included, bottom to top, a semi-infinite substrate, a SiO<sub>2</sub> layer (where applicable) and a MoS<sub>2</sub> layer with a parameterized complex dielectric function adapted from our previous work<sup>53</sup> (Fig. 3a). For the latter, the tabulated optical constants can be found in the SI in Table S1. The full description of the model used for each system can be found in Fig. S5. Continuous lines in Fig. 3b show the values of  $\Psi$  and  $\Delta$  as a function of the energy of the incoming light, averaged over the homogeneous regions highlighted by the dashed polygons



**Fig. 3** (a) Optical constants of the MoS<sub>2</sub> used in the modelling.<sup>53</sup> (b) Experimental data (solid lines) and best fits (dashed lines) of the ellipsometric angles  $\Psi(\lambda)$  and  $\Delta(\lambda)$  averaged over the dashed-line regions of (c). The green lines correspond to MoS<sub>2</sub>-covered regions, whereas the dark-red ones indicate the respective substrates. (c) MoS<sub>2</sub> thickness maps extracted from the analysis of the imaging ellipsometric measurements. Light blue areas correspond to exfoliated MoS<sub>2</sub> areas. Dark red regions in the samples on PDMS and silica indicate the presence of thick multilayer regions. The scale bar is 300  $\mu$ m for all the images. (d) MoS<sub>2</sub>-thickness histograms corresponding to the maps in (c). The peaks at 0 nm and  $\sim$ 0.7 nm correspond to the substrate and the monolayer MoS<sub>2</sub> (1L), respectively. Some small bilayer  $\sim$ 1.4 nm (2L) and trilayer  $\sim$ 2.1 nm (3L) regions are also observed in the sample on silica as shown in the inset.



in Fig. 3c. The bare-substrate spectra are reported as the dark-red lines, whereas the MoS<sub>2</sub>-covered areas are reported as the green spectra. The differences between the spectra collected on different substrates are readily understandable based on the respective differences in dielectric function and morphology. In all the MoS<sub>2</sub>-covered spectra, variations of different magnitudes with respect to the spectra on the bare substrate are observed, whose extent and spectral shape depended upon the dielectric mismatch of MoS<sub>2</sub> and the substrate material. The optical model parameters were optimized to best fit the experimental data, resulting in good agreement between models and data. The resulting fit curves are represented by dashed lines in Fig. 3b for each system.

Using these models, we performed pixel-by-pixel fitting of the iSE maps, leaving the MoS<sub>2</sub> thickness as the only free-fit parameter. The resulting thickness maps are shown in Fig. 3c, and the corresponding thickness histograms are displayed in Fig. 3d. The thickness maps clearly show that, beside some inhomogeneous areas, all the substrates under consideration feature large-area uniform monolayer MoS<sub>2</sub> regions, with lateral dimensions ranging from hundreds of microns to mm. Small bilayer, trilayer and bulk regions are present on the silica and PDMS substrates, but overall it is apparent that the method is able to produce large area monolayer MoS<sub>2</sub> samples on different substrates. iSE, besides confirming the monolayer nature of the exfoliated samples on millimetre-sized regions, allowed enhancement of the visibility compared to the optical images in Fig. 2 enhancing the contrast between the MoS<sub>2</sub> and the substrate on all the substrates. This is of particular interest for SiO<sub>2</sub>(nat)/Si, PDMS and silica, where the flakes are invisible or barely visible.

## Structural, chemical and photonic properties

Once we had ascertained the presence of large-area MoS<sub>2</sub> flakes, we proceeded to assess their structural, chemical and photonic characteristics, by means of X-ray photoemission spectroscopy (XPS), Raman spectroscopy and photoluminescence (PL).

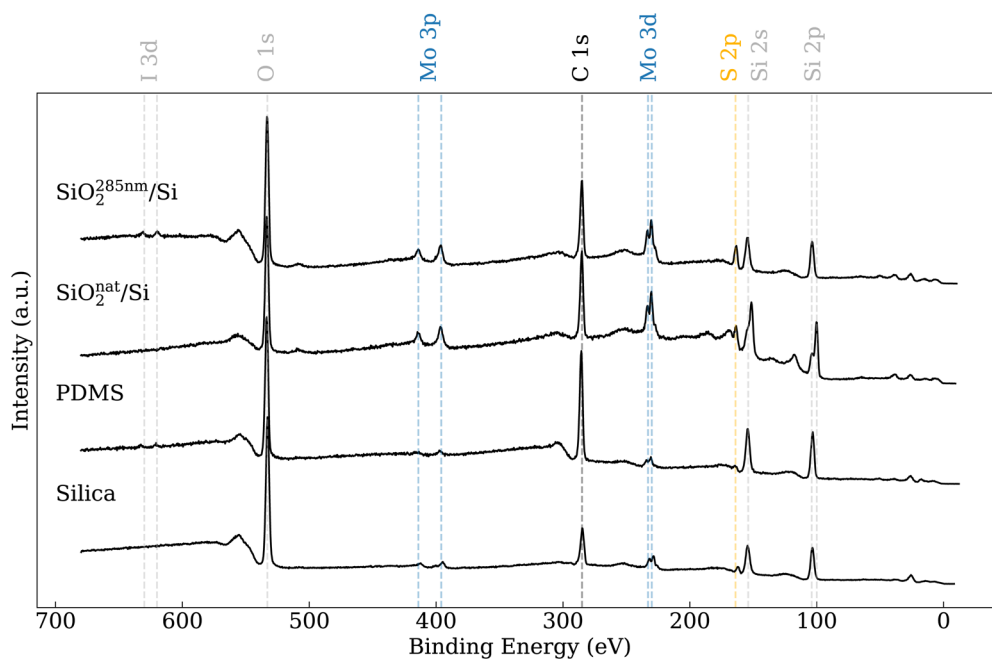
Fig. 4a shows the XPS survey spectra of large-area MoS<sub>2</sub> over the four different substrates. The S/Mo ratio was extracted from the spectra in Fig. 4a obtaining a ratio of  $1.9 \pm 0.1$  and  $2.0 \pm 0.1$  for the samples on SiO<sub>2</sub>(nat)/Si and SiO<sub>2</sub>(285 nm)/Si, respectively (please see the SI for more details). All the spectra exhibit prominent Si, S, Mo, C, and O peaks originating from MoS<sub>2</sub> (Mo and S), the substrates (Si and O, and C in the case of PDMS) and adventitious contamination (C). Traces of iodine due to the etching solution can be found on the SiO<sub>2</sub>(285 nm)/Si sample and on the PDMS sample. In particular, PDMS substrates undergoing the etching procedure turn slightly yellow after being in contact with the etchant, suggesting a reaction with PDMS that, however, does not alter its viscoelastic properties. The Si 2p peaks of the SiO<sub>2</sub>(nat.)/Si sample are split into low-BE components (pure Si) and high-BE components (SiO<sub>2</sub>).

Fig. 4b shows high resolution (HR) XPS spectra of the four samples, performed in the energy ranges corresponding to the

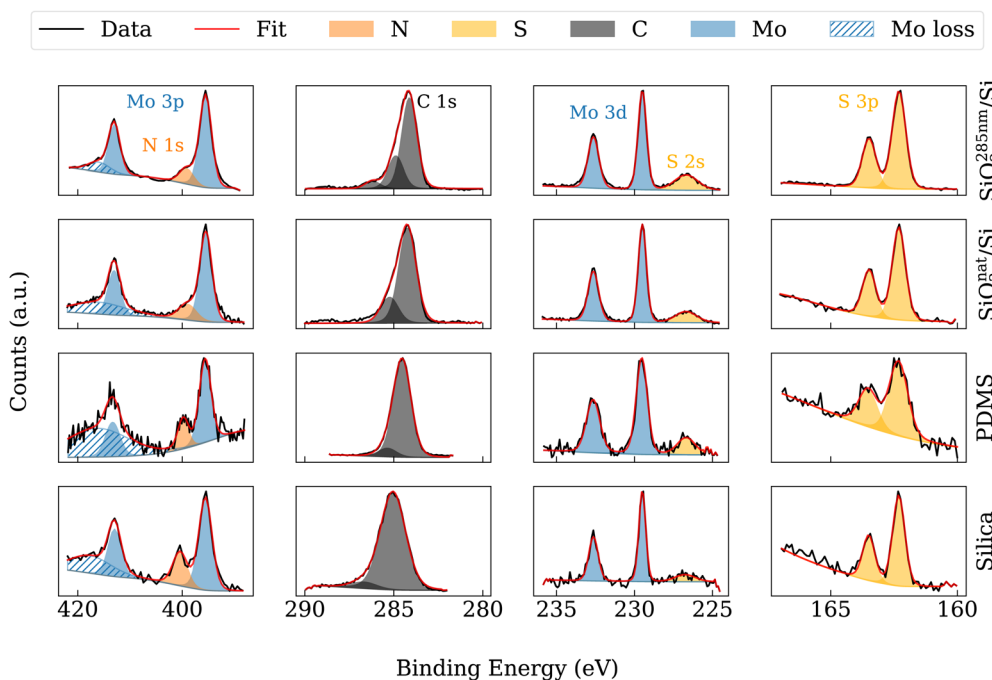
Mo 3p/N 1s, Mo 3d/S 2s, S 2p, and C 1s peaks. The high-resolution spectra were deconvoluted into their chemically shifted subcomponents as detailed in the Methods section. In general, the XPS spectra in corresponding energy regions share similar structures. The binding energies (BEs) of the Mo 3p, Mo 3d, N 1s, S 2s and S 2p peaks are all reported in Table 1. The binding energy was referenced to Mo 3d<sub>5/2</sub> at 229.5 eV, since C 1s referencing is not appropriate for comparing data across these substrates.<sup>54</sup> From the deconvolution procedure, we can observe that the Mo 3d<sub>5/2</sub>/3d<sub>3/2</sub> peak is composed of a single doublet, unambiguously indicating that only the Mo(IV) states involved in MoS<sub>2</sub> are present, and justifying the energy referencing. The broad feature centred around 416 eV BE (hatched area) can be attributed to plasmon losses. Similarly, the absence of further components near the S 2p<sub>3/2</sub>/2p<sub>1/2</sub> doublet confirms that sulphur is not involved in chemical bonds other than MoS<sub>2</sub>. The larger FWHM of the peaks (especially the S 2p) for the PDMS sample is ascribed to the superposition of contributions from monolayer and few-layer areas, given that the XPS measuring spot has a few hundred microns of lateral dimension. The N 1s peak around 399 eV BE verifies the presence of very small quantities of nitrogen that can, however, be observed in the bare substrates. Fig. S6 shows the XPS survey spectrum of each substrate prior to the deposition of MoS<sub>2</sub>.

Raman and PL measurements can be exploited to gain insights into the presence of defects,<sup>55,56</sup> strain<sup>57,58</sup> and doping<sup>59–61</sup> in monolayer flakes. Fig. 5 shows representative Raman and PL spectra of the samples on each substrate. The in-plane E<sub>2g</sub> and the out-of-plane A<sub>1g</sub> Raman mode frequencies are compatible with the typical values found in the literature. The frequency difference between the two modes was found at values ranging from  $\sim 18.6 \text{ cm}^{-1}$  to  $\sim 19.5 \text{ cm}^{-1}$  for all the samples, fully compatible with monolayer MoS<sub>2</sub> samples. PL spectra, reported in Fig. 5b, show clear differences depending on the target substrates, in particular with regard to the relative spectral weights of neutral exciton, trion and B-exciton contributions. When looking at the A<sup>0</sup> exciton ( $\sim 1.9 \text{ eV}$ ) and the A<sup>-</sup> trion ( $\sim 1.85 \text{ eV}$ ), we observe a strong variation of the peak intensity ratio  $I(A^-)/I(A^0)$  among the samples. Fig. 5c shows the mean value and deviation of the neutral-exciton/trion ratio for each substrate, with each measurement resulting from the average of over 500 points (the full distribution of the parameters is shown in Fig. S7). We can easily notice that the PDMS sample features a substantially lower  $I(A^-)/I(A^0)$  ratio when compared to the other samples, a difference compatible either with the presence of defects or with low substrate-induced doping. In the case that defects gave the larger contribution, however, the intensity ratio of the A and B excitons  $I(B)/I(A^0)$  should also increase,<sup>55</sup> but the data show that the B exciton intensity on PDMS is very weak compared to that of the A exciton, resulting in the low  $I(B)/I(A^0)$  ratio observed in Fig. 5c. As a consequence, the difference between the PL spectra of the PDMS sample and the other ones can be assigned to a lower substrate-induced doping of MoS<sub>2</sub> in the PDMS case. In contrast, the higher  $I(A^-)/I(A^0)$  ratios observed in all the remaining samples indicate higher electron doping levels, as indeed observed in MoS<sub>2</sub> samples deposited on silicon oxide





(a)



(b)

Fig. 4 (a) XPS survey spectra of the samples on the different substrates. Vertical dashed lines have been added as a guide to the eye for tracing the major XPS peaks present in the spectra. (b) High-resolution measurements performed in the Mo 3d, Mo 3p, N 1s, C 1s and S 2p energy regions. Each row corresponds to a different substrate labelled on the right side of the figure.

surfaces.<sup>62–67</sup> Detailed characterization of the substrate induced doping effect was beyond the scope of this work.

Finally, we notice that both PL and Raman spectra of native-oxide Si exhibit lower signals compared to other samples, despite being acquired under analogous experimental conditions. Since both Raman and PL spectra do not significantly

differ from those of MoS<sub>2</sub> on silica or thermal-oxide Si, we suggest that such a reduced signal might originate from dielectric screening and charge transfer effects between MoS<sub>2</sub> and the underlying Si. While, to our knowledge, there are no studies that directly involve PL and Raman measurements of MoS<sub>2</sub> on Si substrates, our hypothesis is supported by several



Table 1 Binding energies of the main XPS peaks

Substrate	S 2p <sub>3/2</sub>	S 2p <sub>1/2</sub>	S 2s	Mo 3d <sub>5/2</sub>	Mo 3d <sub>3/2</sub>	Mo 3p <sub>3/2</sub>	Mo 3p <sub>1/2</sub>	N 1s
SiO <sub>2</sub> (nat)/Si	162.3	163.5	226.7	229.5	232.6	395.5	413.1	399.0
SiO <sub>2</sub> (285 nm)/Si	162.3	163.5	226.7	229.5	232.6	395.5	413.0	399.2
PDMS	162.3	163.5	226.7	229.5	232.7	395.6	413.3	399.8
Silica	162.3	163.5	226.8	229.5	232.6	395.6	413.0	400.4

studies of TMDCs on semiconducting and metallic surfaces<sup>30,68–71</sup> showing the quenching of the MoS<sub>2</sub> PL due to charge transfer and dielectric screening effects.

## Deterministic transfer from PDMS stamps

The successful exfoliation of large-area flakes on PDMS opens up interesting perspectives, since PDMS is widely employed as a vector for the deterministic transfer of TMDC flakes onto a large variety of substrates.<sup>42,43</sup> It would, therefore, be extremely appealing to replicate such procedures with the very large flakes that we fabricate, thereby extending the range of possible target substrates to systems not currently compatible with the above-reported exfoliation technique, like noble-metal films. Furthermore, several applications of TMDCs, such as vdW heterostructures for electronics or optoelectronics, require deterministic transfer approaches, which are intrinsically not guaranteed by the exfoliation process alone. In this section, we demonstrate the deterministic transfer of MoS<sub>2</sub> on a given substrate, adapting a common all-dry-transfer method,<sup>42</sup> performing a successful transfer on SiO<sub>2</sub>(nat)/Si of our sample exfoliated on PDMS (Fig. 2c).

In Fig. 6, we show a binary intensity map of the sample before and after the transfer, constructed by identifying the MoS<sub>2</sub> regions in the corresponding optical images (Fig. S8). We can observe that the original sample (Fig. 6a) is almost entirely transferred onto the target substrate (Fig. 6b). Partial fragmentation is observed in the localized region of the sample; however, large uniform areas are still present after the sample is transferred.

The sample was then characterized by Raman and PL spectroscopy in order to assess its properties after the transfer. In Fig. 7, we compare the spectra acquired before and after performing the transfer. We can observe that the spectroscopic features of MoS<sub>2</sub> are hardly affected by the sample transfer. We observe, however, a significant reduction in both the Raman and PL intensities after the transfer, comparable to what is observed with the sample directly produced on SiO<sub>2</sub>(nat)/Si in Fig. 2b. With a similar argument to that provided in the previous section, we can trace back this quenching effect to the interaction of the MoS<sub>2</sub> sample with the underlying silicon substrate, rather than being an intrinsic feature of the transferred MoS<sub>2</sub>. Finally, we observe that the transferred sample features a lower  $I(A^-)/I(A^0)$  ratio compared to the one observed using the same substrate in Fig. 5b. This can indicate that the interaction between the substrate and MoS<sub>2</sub> is partially reduced

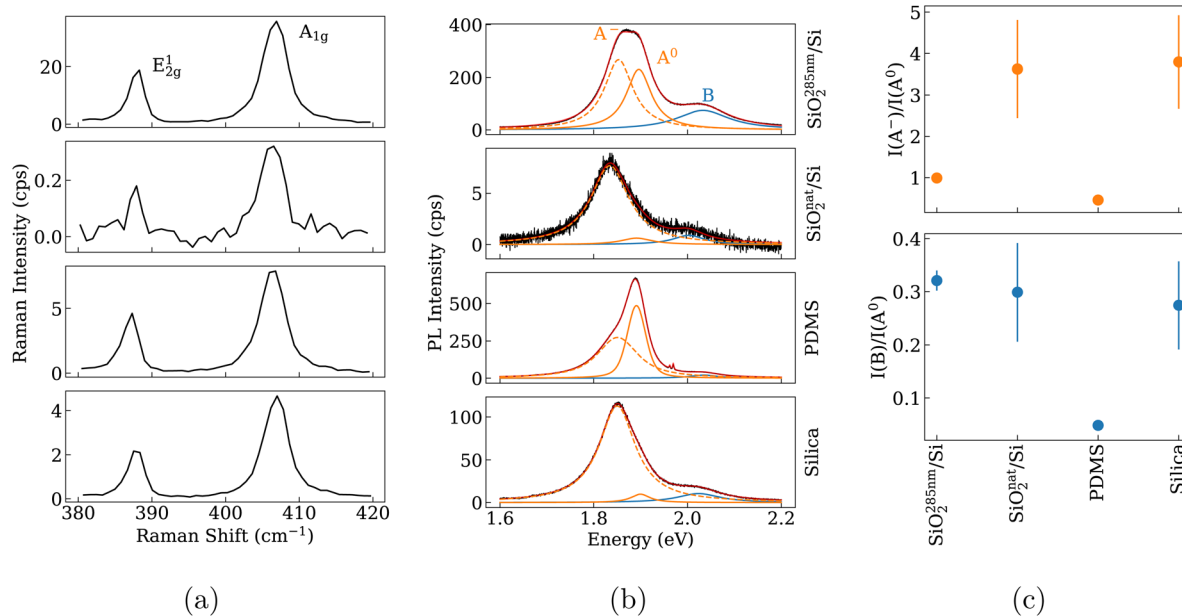


Fig. 5 (a) Raman and (b) photoluminescence spectra. The black line shows the background-subtracted experimental data. In the PL plots, the orange solid and dashed lines and the blue solid line indicate the fitted  $A^0$  exciton,  $A^-$  trion and B exciton peaks, respectively. The solid red line shows the complete fitting function. (c)  $I(B)/I(A^0)$  (bottom) and  $I(A^-)/I(A^0)$  (top) intensity ratios extracted from the distributions in Fig. S7.



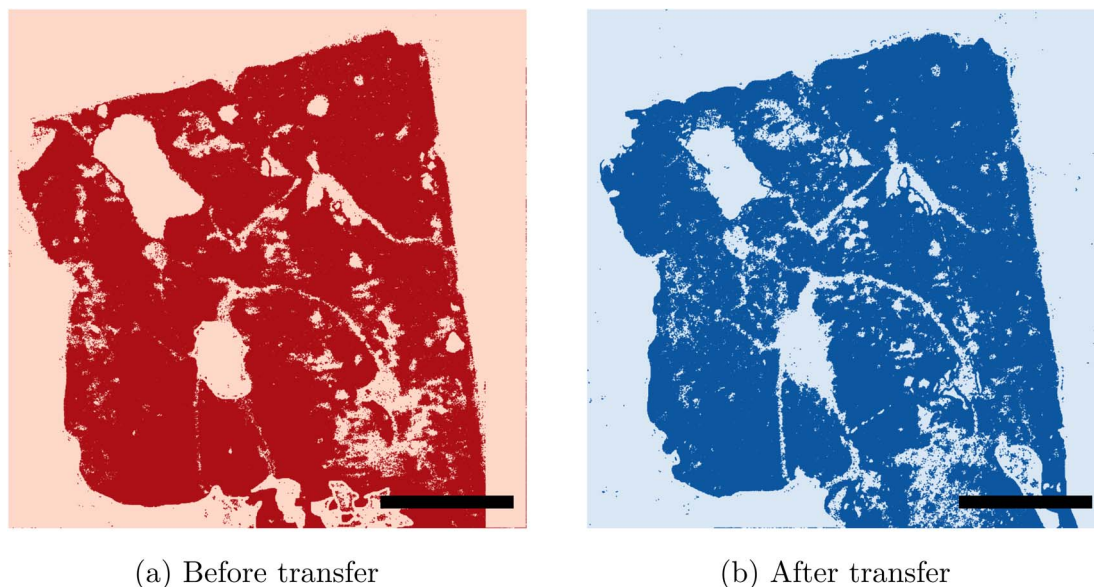


Fig. 6 Binary maps extracted from the optical images in Fig. S8 of the sample exfoliated on PDMS before (a) and after (b) its transfer on a  $\text{SiO}_2(\text{nat})/\text{Si}$  substrate. The darker colours in both images correspond to areas identified as  $\text{MoS}_2$ . The scale bar is 250  $\mu\text{m}$ .

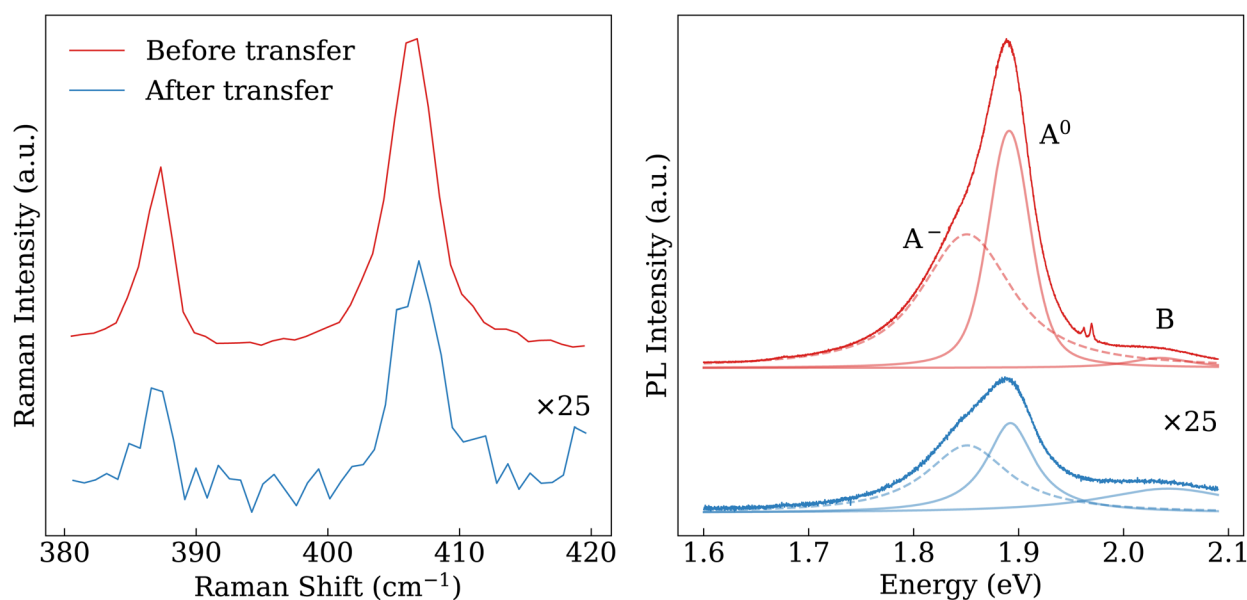


Fig. 7 Characteristic Raman (left) and PL (right) spectra of the transferred sample before and after the transfer. The intensity of the spectra of the transferred sample was multiplied by a factor 25.

due to contaminants associated with the PDMS transfer procedure.<sup>52,72</sup>

Several other transfer attempts were performed on different substrates, to demonstrate the potential of combining our large-area exfoliation method with the PDMS dry-transfer technique. In the SI, we report two representative cases of flakes transferred onto noteworthy substrates such as  $\text{Si}_3\text{N}_4$  ultra-thin membrane used for TEM measurement (Fig. S9) and optical microcavities nanofabricated on a silver film on  $\text{CaF}_2$  (Fig. S10).

## Conclusions

In the present work, we report a groundbreaking improvement of the previously reported gold-assisted mechanical exfoliation of bulk  $\text{MoS}_2$ . This method allowed the production of millimeter-sized flakes on four different substrates, representative of various applications in electronics and optoelectronics. In particular, the modification of one of the exfoliation steps, introducing a PDMS layer mechanically constraining the TRT during the release phase, allowed an increase in the reliability of the exfoliation and enabled its generalization to different



substrates. The key advantages of our method are its simplicity, scalability, reliability, cost-effectiveness and quickness. Uniquely, the exfoliation can be performed on a wide range of target substrates without modifying the exfoliation procedure. Notably, the gold-assisted exfoliation of large MoS<sub>2</sub> samples on PDMS has been demonstrated. PDMS is a resourceful substrate material that enabled us to perform further deterministic transfers of the freshly exfoliated, millimeter-sized samples onto desired substrates with micrometric control over the positioning of the 2D material.

Imaging ellipsometry has been employed to assess the local MoS<sub>2</sub> thickness over large areas, thus enabling a fast and reliable determination of the monolayer nature of our exfoliated flakes. Moreover, our samples were characterized by Raman and photoluminescence spectroscopy in order to grade the quality and uniformity of the exfoliated samples. We observed substrate-dependent variations of the spectral features of the MoS<sub>2</sub> samples, which were identified as due to substrate–MoS<sub>2</sub> interactions. Overall, a high photoluminescence yield is observed, indicating high-quality monolayer samples. A Raman and photoluminescence quenching effect for the sample on SiO<sub>2</sub>(nat)/Si was traced back to substrate/MoS<sub>2</sub> interactions, as well as intensity and PL peak shifts between different substrates.

The presented methodology represents a major breakthrough in the production of 2D materials. We believe that its flexibility and scalability will drive major advancements in the fabrication of macroscopic 2D devices, ultimately enabling the complete exploitation of the exceptional properties of 2D materials.

## Methods

Imaging Spectroscopic Ellipsometry (iSE) measurements were performed by means of a Park Systems Accurion EP4 imaging ellipsometer equipped with a laser-stabilized Xenon lamp and a monochromator. We acquired all our spectra in the 360 nm to 1000 nm range at an angle of incidence of 50° in rotating compensator mode. A 5× objective was used to focus the collected light to a CCD detector, simultaneously acquiring the whole probed area. Knife-edge illumination was used to block backside reflections and accurately measure the ellipsometric quantities on double-polished transparent substrates (PDMS and silica).

XPS spectra were acquired using a Physical Electronics PHI 5600 photoelectron spectrometer, equipped with a monochromatized Al K<sub>α</sub> source and with an electron flood gun to reduce surface charging. XPS data analysis was performed using CasaXPS software. The spectra were fitted using the symmetric Voigt-like LA(1.53, 243) line shape in CasaXPS. The S 2p, Mo 3p, and Mo 3d doublets were fitted while keeping the p<sub>3/2</sub>:p<sub>1/2</sub> and d<sub>5/2</sub>:d<sub>3/2</sub> area ratios fixed at 2 : 1 and 3 : 2, respectively. The S 2p spin-orbit peaks were also constrained to have the same full-width at half-maximum (FWHM). The Mo 3p and Mo 3d spin-orbit peaks were allowed to have different FWHMs to better reproduce the experimental data. The need for a non-identical FWHM to fit the Mo doublets is due to Coster–Kronig

broadening, which has been observed on similar compounds.<sup>73–75</sup> The C 1s peak has been fitted with two components with the same FWHM, except for the sample on SiO<sub>2</sub>(285 nm)/Si, where the spectral lineshape clearly suggests a third component at higher binding energies, which can be attributed to additional contamination.

Micro-Raman and photoluminescence spectroscopy were performed using a Jasco NRS-4100 Raman spectrometer. The probing laser was a 532 nm laser filtered to have a power of ~100 μW on the sample through a 100×, 0.9 NA objective. A 2400 grooves/mm grating was used to disperse scattered light for the Raman measurements; a 900 grooves/mm grating was used for photoluminescence measurements.

## Conflicts of interest

There are no conflicts to declare.

## Data availability

The datasets analysed during the current study are available at <https://doi.org/10.5281/zenodo.17602072>.

Supplementary information (SI): (S1) photos of the exfoliation steps; (S2) samples of Δ values acquired with the imaging ellipsometer; (S3) samples of Ψ values acquired with the imaging ellipsometer; (S4) AFM imaging of a sample on SiO<sub>2</sub>(nat)/Si; (S5) graphical representation of the models used for the fitting of the ellipsometric quantities Ψ and Δ; (S6) XPS quantitative analysis; (S7) distributions of I(B)/I(A<sup>0</sup>) and I(A<sup>-</sup>)/I(A<sup>0</sup>) from PL mapping; (S8) optical images of the MoS<sub>2</sub> sample on PDMS before and after performing its transfer onto a SiO<sub>2</sub>(nat)/Si substrate; (S9) optical images of the transfer of a MoS<sub>2</sub> monolayer onto a Si<sub>3</sub>N<sub>4</sub> TEM grid; (S10) optical images of the transfer onto a silver-coated, nanofabricated CaF<sub>2</sub> substrate; tabulated optical constants of monolayer MoS<sub>2</sub>. See DOI: <https://doi.org/10.1039/d5na00919g>.

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