Supplementary material for *Exfoliation and transfer of millimetre-sized MoS_2 flakes on arbitrary substrates*

Riccardo Galafassi,[*,†,‡,‖] Ermes Peci,[¶,‖] Valentina Venturino,[¶] Michele Magnozzi,[¶] Francesca Telesio,[§] Maurizio Canepa,[¶] and Francesco Bisio[†]

†*CNR-SPIN, Corso Perrone 24, 16152 Genova, Italy*

‡*RAISE Ecosystem, Genova, Italy*

¶*Optmatlab, Dipartimento di Fisica, Università degli Studi di Genova, Via Dodecaneso 33, 16146 Genova*

§*Dipartimento di Fisica, Università degli Studi di Genova, Via Dodecaneso 33, 16146 Genova*

‖*These authors contributed equally to this work*

E-mail: riccardo.galafassi@cnr.it

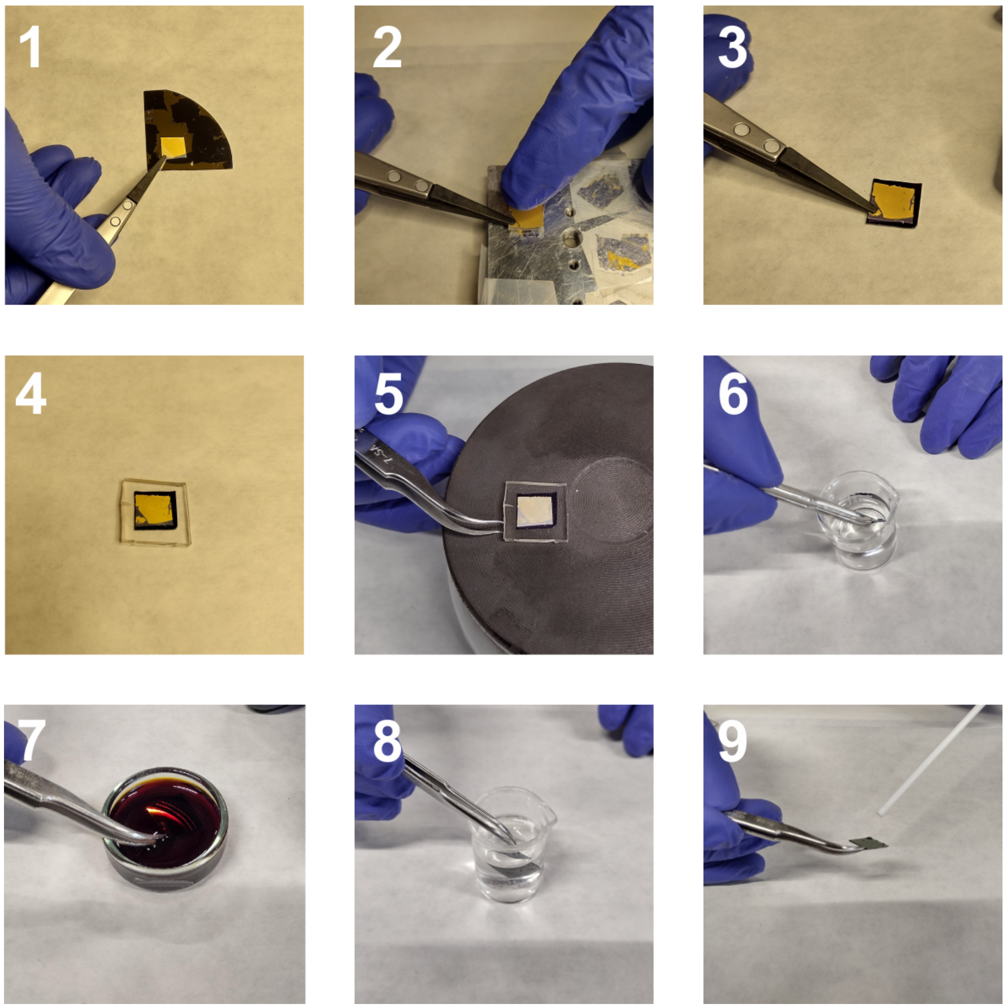


Figure S1: Exfoliation steps. 1. Gold film peel off from a silicon template using the thermal release tape. 2. TRT/Au stack in contact with a freshly cleaved bulk TMDC. 3. TRT/Au/MoS_2 stack on the substrate. 4. The stack covered by a PDMS patch. 5. Heating of the stack and release of the thermal release tape. 6. Sample wash in acetone to remove organic residues. 7. Gold film etching. 8. Sample wash in water and then in isopropyl alcohol. 9. Sample drying with nitrogen flow.

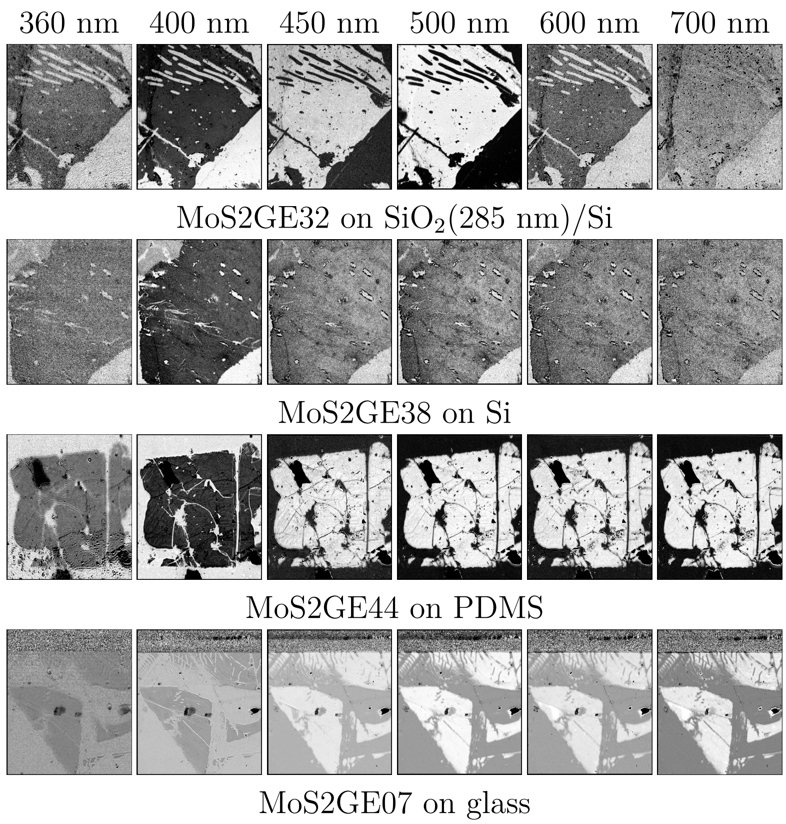


Figure S2: Samples of Δ values acquired with the imaging ellipsometer.

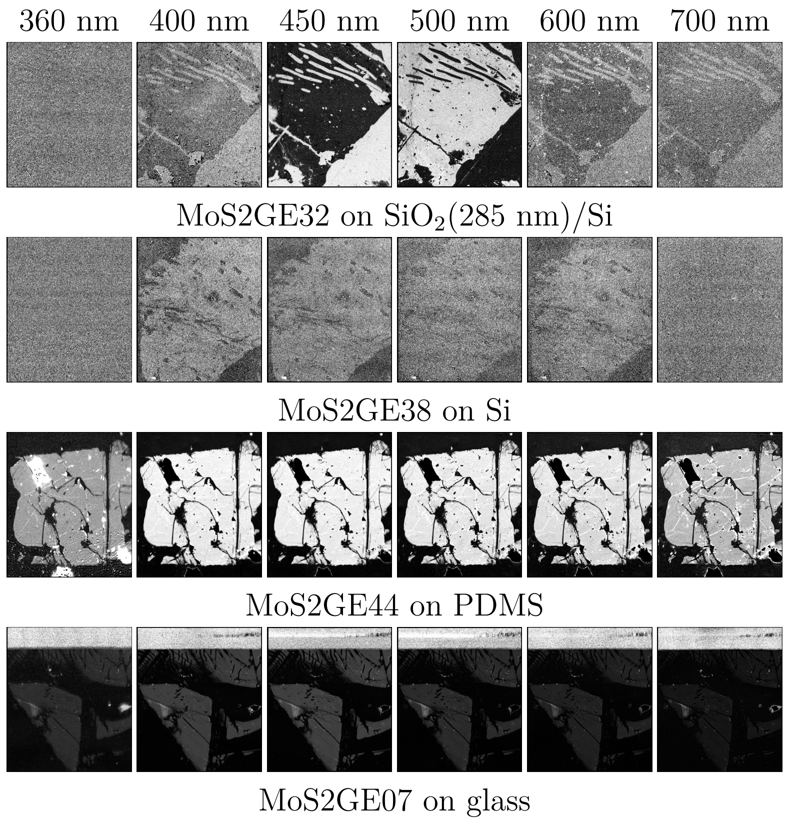


Figure S3: Samples of Ψ values acquired with the imaging ellipsometer.

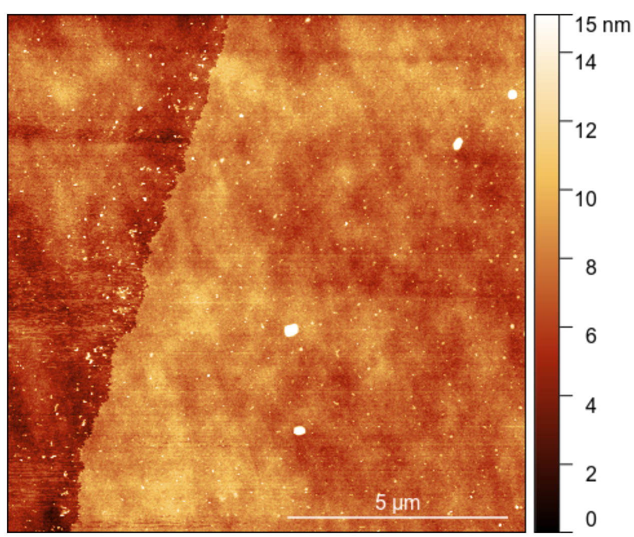


Figure S4: Atomic force microscopy imaging of a sample on SiO$_2$(nat)/Si.

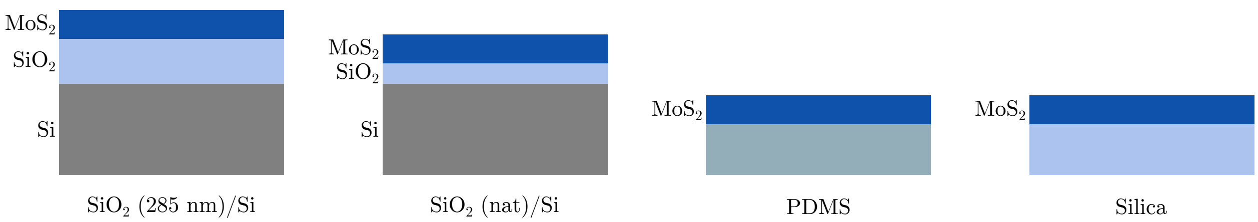


Figure S5: Graphical representation of the models used for the fitting of the ellipsometric quantities Ψ and Δ. The models included several layers representing the MoS_2 samples and substrates. For the modelling of the substrates, tabulated values were used for pure silicon (Si)[1] and SiO_2 layers[2]. The spectra of PDMS and silica were modelled using Cauchy parametric layers for which the An, Bn and Cn parameters were optimised on the spectra of the bare substrates. The MoS_2 sample was modelled with a superposition of Herzinger-Johs PSEMI and Tauc-Lorentz oscillators from a previous work[3]. The energies of the A and B excitons in the models was fitted to adapt to substrate dependent variations. In addition to the parameters modelling the layers, we fitted the MoS_2 thickness, in all cases, and the SiO_2 thickness, where applicable.

XPS quantitative analysis

In order to check the structural integrity of the MoS2 monolayer we performed a quantitative analysis of the XPS survey spectra reported in Figure 4b. We calculated the area under the S 2p and the Mo 3d peaks, after the subtraction of an appropriate background, and applied the respective sensitivity factors for the two orbitals at a photon energy corresponding to the Al Kα radiation (0.022 and 0.13 for the S 2p and the Mo 3d, respectively[4]). We obtained a ratio of S/Mo of 1.9 ± 0.1 for the sample on SiO_2(nat)/Si and of 2.0 ± 0.1 for the one on SiO_2(285 nm)/Si, fully compatible with a structurally integer system. For the remaining measurements, the low intensity of the Mo and S signal makes this kind of quantitative estimation too error prone.

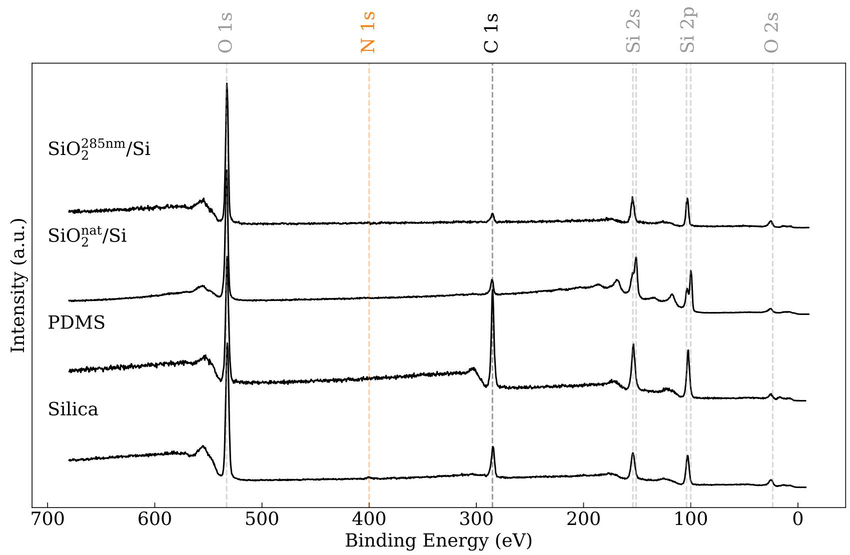
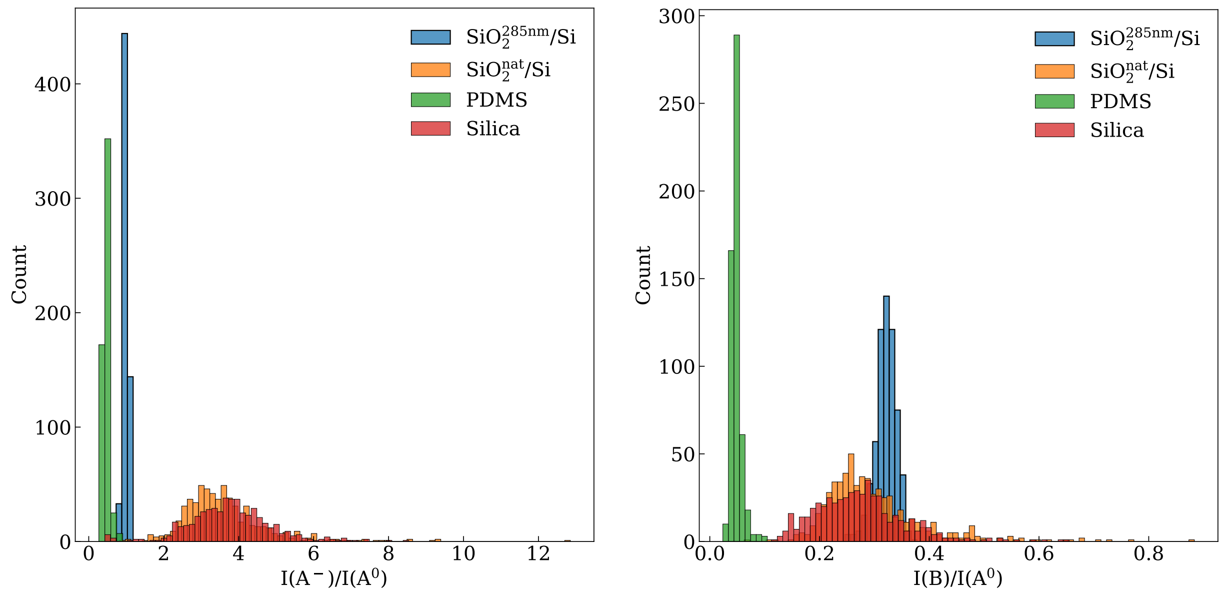
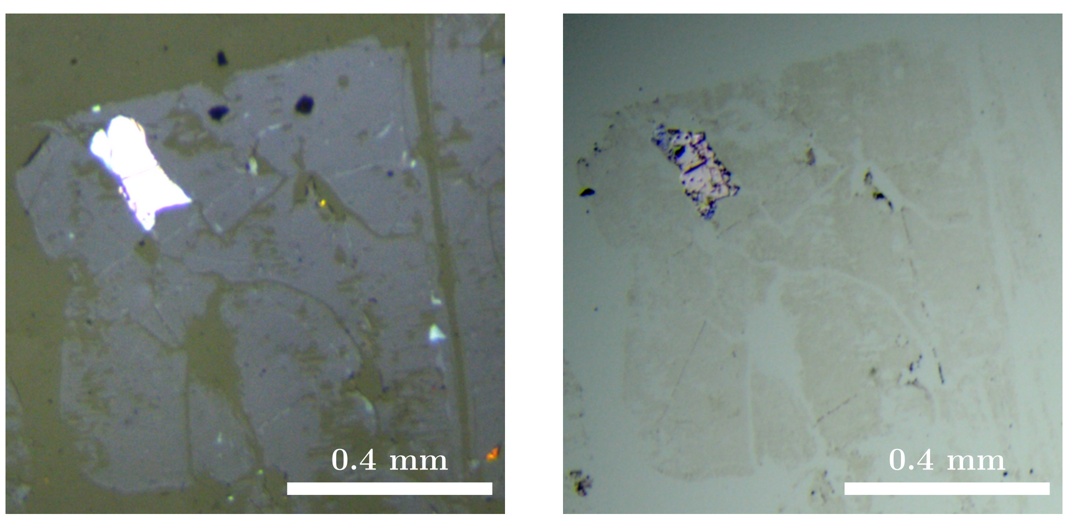


Figure S6: XPS survey spectra of the substrates used in our experiments normalised by the spectra maximum intensity value. Vertical dashed help visualize the main XPS features.



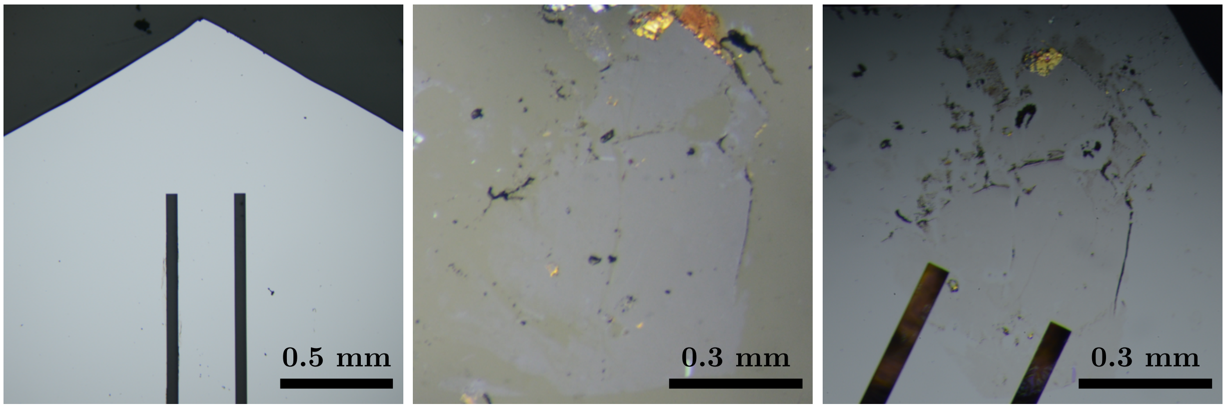
(a) (b)

Figure S7: Distributions of $I(B)/I(A^0)$ and $I(A^-)/I(A^0)$ from mapping of around 500 points.



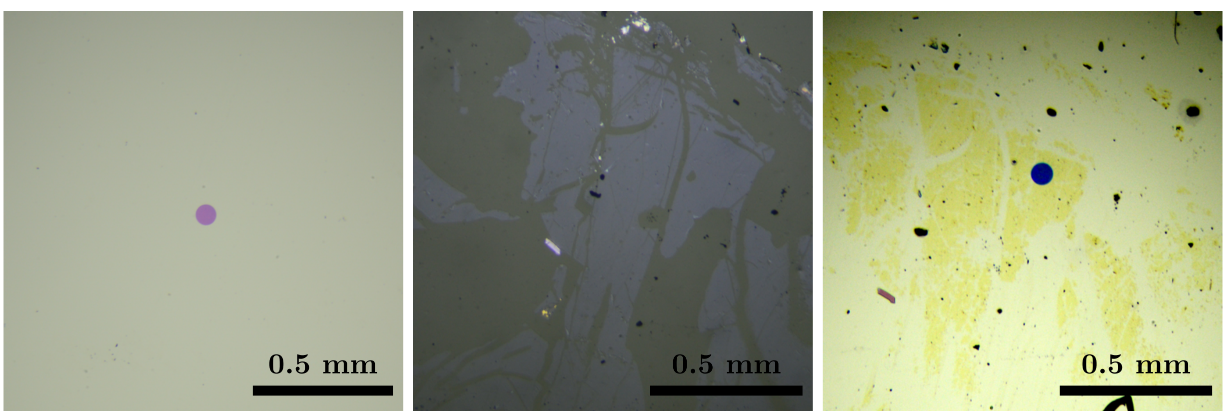
(a) (b)

Figure S8: Optical images of the MoS_2 sample on PDMS before (a) and after (b) performing its deterministic transfer on SiO_2(nat)/Si.



(a) (b) (c)

Figure S9: (a) Si_3N_4 TEM grid. (b) Original MoS_2 on PDMS. (c) MoS_2 transferred on the TEM grid.



(a) (b) (c)

Figure S10: (a) Silver coated CaF_2 substrate. At the centre (darker circle) are visibile the patterned optical microcavities. (b) Original MoS_2 on PDMS. (c) MoS_2 transferred on the patterned substrate.

Table S1: Tabulated optical constant for the MoS_2 model shown in Figure 3a.

Energy (eV)	ϵ_1	ϵ_2
1.0052	15.7611	3.7388e-08
1.0193	15.8131	-2.77919e-08
1.0338	15.8679	-2.08715e-09
1.0487	15.9259	-4.26344e-08
1.064	15.9872	-4.75251e-08
1.0798	16.0522	5.63836e-10
1.0961	16.1211	4.0188e-08
1.1129	16.1944	3.98533e-11
1.1302	16.2725	1.3251e-09
1.1481	16.3558	8.70724e-08
1.1665	16.4448	-1.21233e-09
1.1856	16.5402	3.19021e-08
1.2053	16.6426	-2.2146e-09
1.2257	16.753	6.5475e-10
1.2485	16.8818	3.758e-08
1.2584	16.9398	-1.52535e-09
1.2685	17	-4.93891e-08
1.2787	17.0625	-1.48674e-09
1.2891	17.1276	5.14795e-08
1.2997	17.1952	-9.00951e-09
1.3105	17.2657	-3.99532e-08
1.3215	17.3392	-1.03521e-08
1.3326	17.4158	-2.30929e-11
1.344	17.4959	-1.23514e-09
1.3556	17.5796	-7.82982e-11

1.3673	17.6673	-6.63194e-10
1.3793	17.7591	1.08304e-09
1.3915	17.8555	-7.82323e-10
1.4039	17.9567	1.69766e-12
1.4165	18.0633	-8.39873e-10
1.4294	18.1756	1.12025e-09
1.4425	18.2941	5.17291e-10
1.4559	18.4194	3.45319e-10
1.4695	18.5523	-5.26416e-10
1.4834	18.6934	1.80312e-10
1.4976	18.8437	7.76691e-12
1.512	19.0042	9.12672e-10
1.5267	19.1761	5.61281e-10
1.5418	19.3609	7.02997e-10
1.5571	19.5606	-7.45928e-11
1.5727	19.7773	3.76748e-10
1.5887	20.0141	2.39138e-09
1.605	20.275	6.0968e-09
1.6216	20.5657	1.06964e-06
1.6386	20.8969	0.000517594
1.656	21.2891	0.018155
1.6737	21.7193	0.0898022
1.6919	22.1624	0.182126
1.7104	22.6475	0.277068
1.7293	23.2036	0.374143
1.7487	23.873	0.473455
1.7685	24.7423	0.579947

1.7888	26.012	0.782461
1.8096	27.9273	1.50551
1.8308	30.3426	3.74028
1.8526	31.3605	9.03556
1.8749	25.6431	13.1884
1.8977	22.1149	10.3512
1.9211	22.3725	8.17636
1.9451	24.0725	7.88782
1.9697	25.135	9.5636
1.9949	24.7869	12.2951
2.0208	21.272	14.1259
2.0474	18.4641	12.3492
2.0747	17.7786	10.168
2.1028	18.4485	8.67632
2.1316	19.1495	8.09582
2.1612	19.823	7.74735
2.1917	20.5461	7.61382
2.223	21.2923	7.69472
2.2552	22.0179	7.97629
2.2884	22.681	8.42451
2.3226	23.2592	8.98477
2.3579	23.7681	9.59391
2.3942	24.2706	10.2061
2.4317	24.8692	10.8259
2.4703	25.6772	11.5422
2.5102	26.7615	12.5445
2.5514	28.0688	14.1009

2.594	29.3631	16.4818
2.6381	30.2202	19.8312
2.6836	30.1907	24.0835
2.7308	28.4881	29.1223
2.7797	24.6058	33.7185
2.8303	18.9668	36.8282
2.8828	12.4366	37.7301
2.9373	6.15841	36.2759
2.9939	1.1375	32.9609
3.0527	-2.11015	28.674
3.1139	-3.62983	24.3001
3.1775	-3.82942	20.4283
3.2438	-3.19958	17.2914
3.313	-2.12386	14.8727
3.3851	-0.817399	13.0536
3.4605	0.654344	11.7332
3.5392	2.30526	10.9185
3.6217	4.10075	10.7638
3.708	5.79801	11.4915
3.7986	6.88125	13.1611
3.8936	6.73127	15.4006
3.9936	5.03701	17.3158

References

(1) Herzinger, C. M.; Johs, B.; McGahan, W. A.; Woollam, J. A.; Paulson, W. Ellipsometric determination of optical constants for silicon and thermally grown silicon dioxide via a multi-sample, multi-wavelength, multi-angle investigation. *J. Appl. Phys.* **1998**, *83*, 3323–3336.

(2) Palik, E. *Handbook of Optical Constants of Solids: Volume 1*; 1985; Vol. 1; p 759.

(3) Peci, E.; Magnozzi, M.; Ramò, L.; Ferrera, M.; Convertino, D.; Pace, S.; Orlandini, G.; Sharma, A.; Milekhin, I.; Salvan, G.; Coletti, C.; Zahn, D. R. T.; Bisio, F.; Canepa, M. Dielectric Function of 2D Tungsten Disulfide in Homo- and Heterobilayer Stacking. *Adv. Mater. Interfaces* **2023**, *10*, 2201586.

(4) Yeh, J.; Lindau, I. Atomic subshell photoionization cross sections and asymmetry parameters: 1 Z 103. *Atomic Data and Nuclear Data Tables* **1985**, *32*, 1–155.