Optical identification of layered MoS_2 via characteristic matrix method

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Supplementary Information (SI)

Figures S1 (a), (b), (d) and (e) manifest that the optical contrasts are negative for 1-3L MoS_2 on SiO_2/Si in comparison to the positive contrasts of MoS_2 on quartz in the main text. Figures S1 (c) and (f) illustrate the optical contrast is affected by the gap between MoS_2 and substrate generated during the transferring process. This influence makes it difficult to identify the layer number of MoS_2 using this OM method. The optical contrasts of different layer MoS_2 in Fig. S1 have been summarized in Table S1.

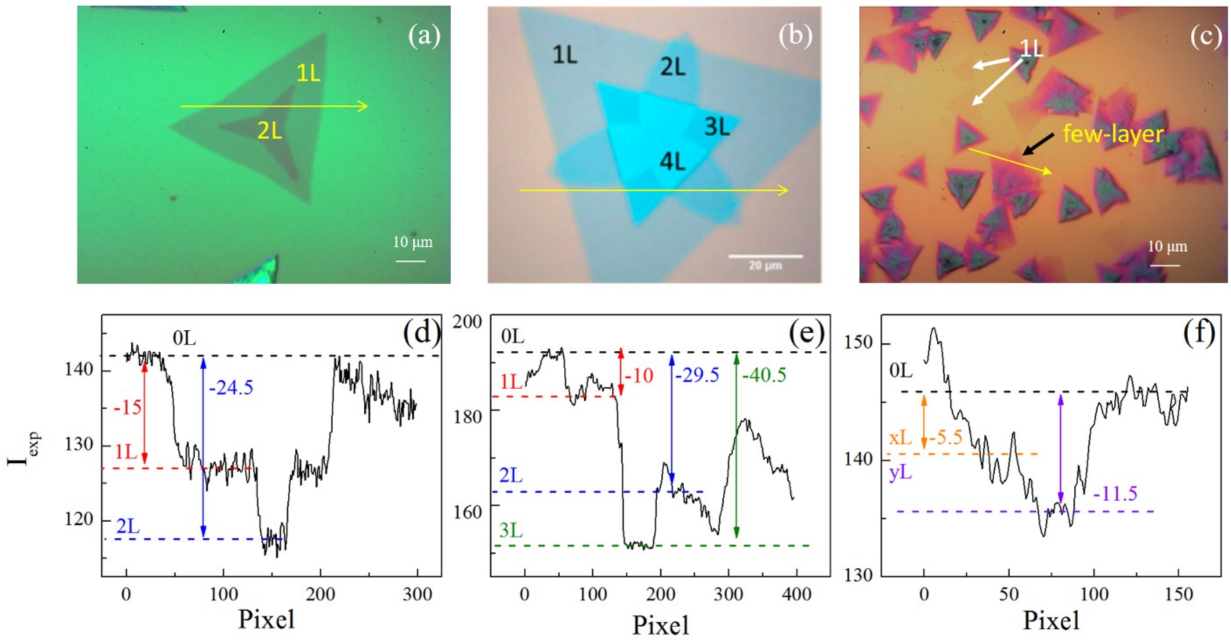

Fig. S1 (a) Optical microscopy (OM) image of CVD grown monolayer (1L) and two-layer (2L) MoS_2 on 300 nm SiO_2/Si in our experiments. (b) The copy of an OM image in Ref. [1]. (c) The CVD MoS_2 on high reflective mirror transferred from SiO_2/Si substrate. (d) - (f) are the pixels intensity curves measured by ImageJ cross the yellow arrows in (a) - (c), respectively.

Table S1. The optical contrasts of different layer MoS_2 in Fig. S1. (x and y mean the number of layers cannot be identified.)

Sample	MoS_2 layer number	I_{exp} (a.u.)	$I_{exp}(MoS_2)$-I_{exp}(substrate)	C_{exp}
(a)	0	142.0	-	-
	1	127.0	-15	-0.1056
	2	117.5	-24.5	-0.2085
(b)	0	191.5	-	-
	1	181.5	-10	-0.0522
	2	162.0	-29.5	-0.1540
	3	151.0	-40.5	-0.2115
(c)	0	146.0	-	-
	x	140.5	-5.5	-0.0377
	y	135.0	-11	-0.0753

Figure S2 (a) shows the difference of complex refractive index between 1L and bulk MoS_2 reported in Ref. [2]. After considering this difference, the calculated optical contrast of MoS_2/G/SiO_2/Si has an obvious distinction from the one in main text. As shown in Fig. S2 (c), the overall curves have dropped significantly comparing the ones in Fig. 2 (e), and the peak at ~620 nm reaches up only to ~0.42 comparing with ~0.6 for 0.3 and 0.09 μm SiO_2. The color counter plots also has an obvious distinction from Fig. 2 (f).

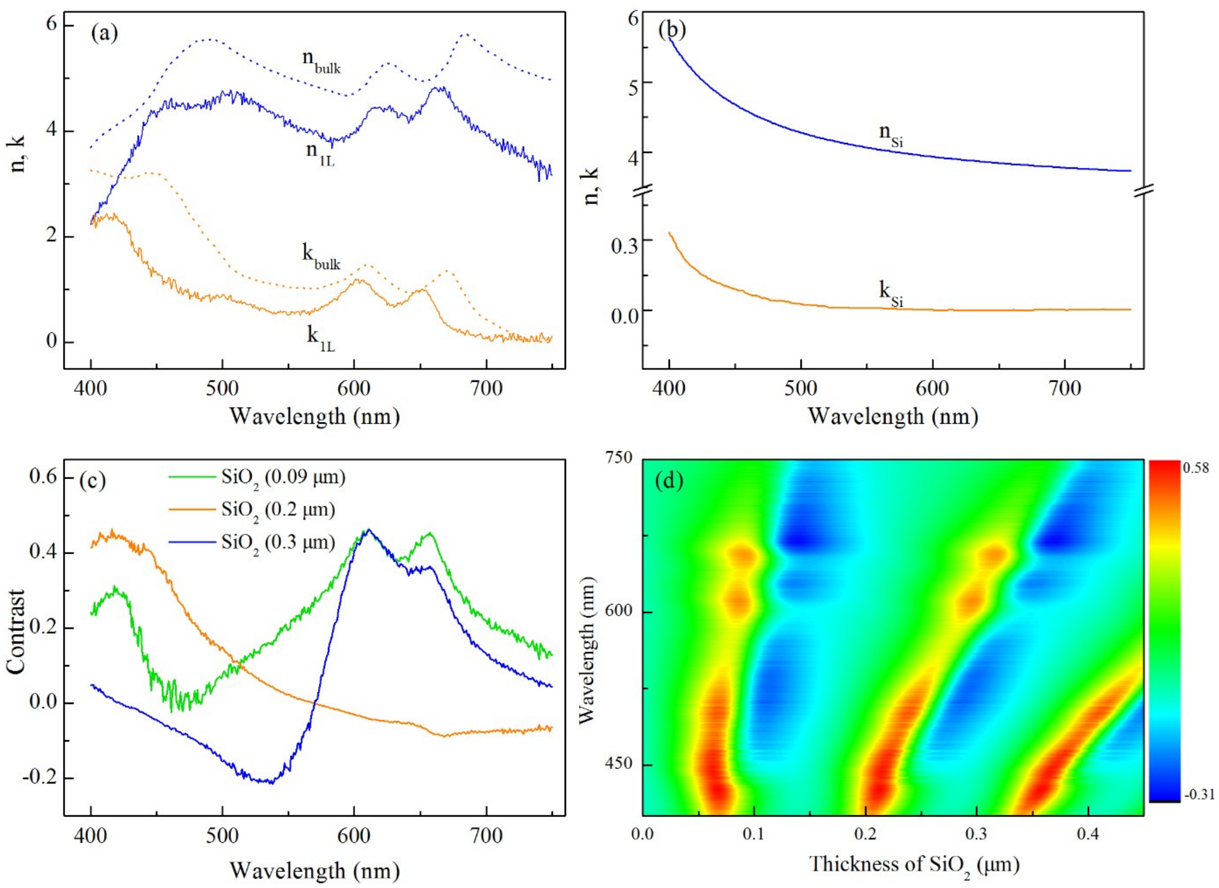

Fig. S2 (a) the real and imagery part of complex refractive index of 1L (n_{1L}, k_{1L}) and bulk (n_{bulk}, k_{bulk}) MoS_2. (b) The real (n_{Si}) and imagery part (k_{Si}) of complex refractive index of Si. (c) The calculated optical contrasts of MoS_2/G/SiO_2/Si system for 0.09, 0.2, 0.3 μm SiO_2 using the complex refractive index of 1L MoS_2. (d) Color counter plots of the contrast as a function of the thickness of SiO_2 and incident wavelength for the MoS_2/G/SiO_2/Si.

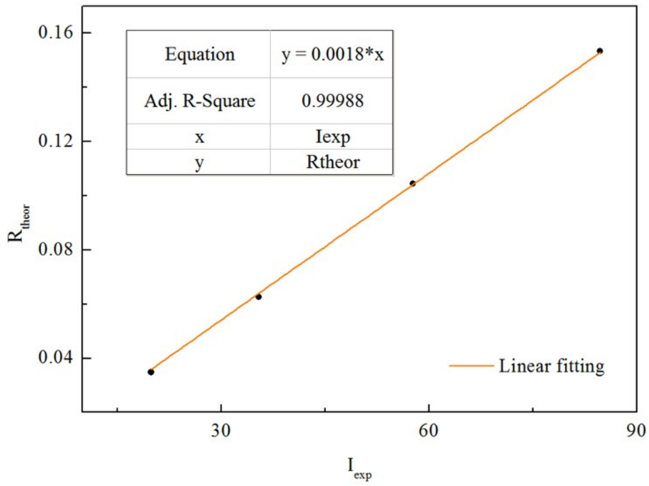

Fig. S3 The linear relationship between R_{theor} and I_{exp} calculated by using N_{bulk} as the complex refractive index of MoS_2.

Figure S4 shows the comparative results of optical contrast calculated using $N_{1L.}$ It can be seen from (c) that the linear relationship between Ctheor and Cexp deviate from the direct ratio comparing with Fig. 3 (f).

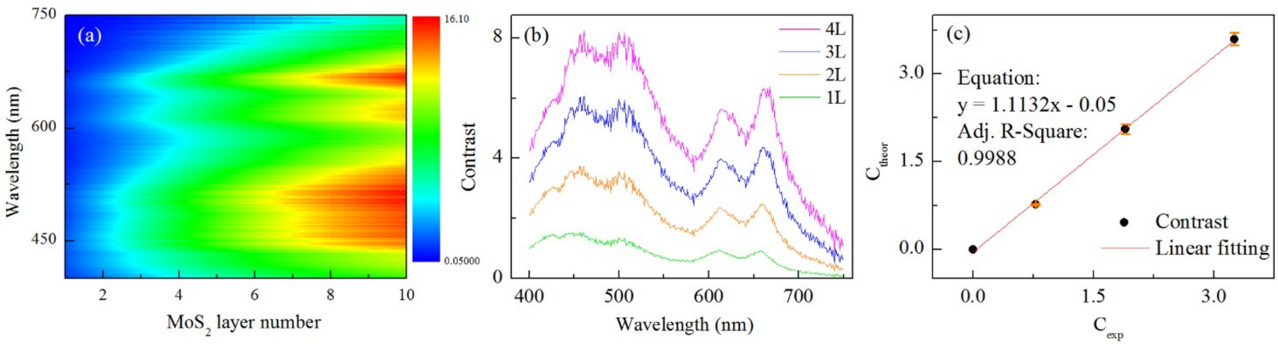

Fig. S4 Optical contrast calculation using complex refractive index of monolayer MoS_2: (a) color counter plots of the contrast as a function of the layer number of MoS_2 and the incident wavelength; (b) wavelength-dependent contrast of 1–4 layers of MoS_2 on quartz; (c) linear fitting of C_{exp} and C_{theor}.

Calculation method

In this part, we summarized the process of calculating the optical contrast of different layer MoS_2 on quartz using the characteristic method.[3]

The first step is to obtain the characteristic matrix of 1L MoS_2.

$$M_{1L\_MoS2} = \begin{bmatrix} A & B \\ C & D \end{bmatrix}, \tag{S1}$$

where:

$$A = cos(\delta_{MoS2}), \tag{S2}$$

$$B = j \cdot sin(\delta_{MoS2})/\eta_{MoS2}, \tag{S3}$$

$$C = j \cdot sin(\delta_{MoS2}) \cdot \eta_{MoS2}, \tag{S4}$$

$$D = cos(\delta_{MoS2}), \tag{S5}$$

$$\delta_{MoS2} = 2\pi \cdot N_{MoS2} \cdot d_{MoS2} \tag{S6}$$

$$\eta_{MoS2} = \sqrt{\frac{\varepsilon_0}{\mu_0}} \cdot N_{MoS2} \tag{S7}$$

N_{MoS2} is the wavelength-dependent complex refractive index of MoS_2 (Fig. S2 (a)). For monolayer, $N_{1L} = n_{1L} - jk_{1L}$; for bulk, $N_{bulk} = n_{bulk} - jk_{bulk}$. ε_0 is the permittivity of vacuum (8.854187817e-12) and μ_0 is the permeability of vacuum (1.2566370614e-6). d_{MoS2} is the thickness (0.7 nm) of monolayer MoS_2.

The second step is to obtain the characteristic matrix of film system.

For 1L MoS_2: $$M = M_{1L\_MoS2} \tag{S8}$$

For 2L MoS_2: $$M = M_{1L\_MoS2} * M_{1L_{MoS2}} = {M_{1L\_MoS2}}^2 \tag{S9}$$

For 3L MoS_2: $$M = {M_{1L\_MoS2}}^3 \tag{S10}$$

$$\vdots$$

It is easily to get the characteristic matrix of a multilayer system no matter how many layers there are. As for a heterostructure as shown in Fig. 2 (d) – MoS_2/G/SiO_2/Si, the characteristic matrix of the system can be calculated as follows:

$$M = M_{1L\_MoS2} * M_G * M_{SiO2} \tag{S11}$$

M_G and M_{SiO2} is the characteristic matrix of graphene and SiO_2, respectively, which can be obtained using S1-S5. In our calculation, the complex refractive index of graphene is (2.6-1.3j); the refractive index of SiO_2 is:

$$N_{SiO2} = \sqrt{1 + \frac{0.6961663}{1 - (0.0684043/\lambda)^2} + \frac{0.4079426}{1 - (0.1162414/\lambda)^2} + \frac{0.8974794}{1 - (9.896161/\lambda)^2}} \tag{S12}$$

The final step is to calculate the reflectivity and optical contrast.

The reflection coefficient of film: $$r = \frac{(A + B \cdot \eta_G) \cdot \eta_0 - (C + D \cdot \eta_G)}{(A + B \cdot \eta_G) \cdot \eta_0 + (C + D \cdot \eta_G)} \tag{S13}$$

The reflectivity of film: $$R = r \cdot r^* \tag{S14}$$

$$r_0 = \frac{N_0 - N_G}{N_0 + N_G} \tag{S15}$$

The reflectivity of substrate: $$R_0 = r_0 \cdot r_0{}^* \tag{S16}$$

$$\eta_0 = \sqrt{\frac{\varepsilon_0}{\mu_0}} \cdot N_0 \tag{S17}$$

$$\eta_G = \sqrt{\frac{\varepsilon_0}{\mu_0}} \cdot N_G \tag{S18}$$

Optical contrast: $$Contrast = \frac{R - R_0}{R_0} \tag{S19}$$

N_0 is the refractive index of air (N_0=1); N_G is the wavelength-dependent refractive index of the substrate. For MoS_2 on quartz, N_G is N_{SiO2} (S12); for MoS_2/G/SiO_2/Si, N_G is the wavelength-dependent complex refractive index of Si (Fig. S2 (b)).

In addition, we use the Eq. (5) in to calculate the reflectivity of the quartz and the 1-4L MoS_2 under the continuous spectrum (Fig. 3 (c)). Because the function of S (λ) and R (λ) were unknown, we used the numerical integration (S20) to carry out the calculation.

The reflectivity under continuous spectrum:

$$R = \frac{\sum_{i=1}^{n} S(\lambda_i) \cdot R(\lambda_i) \cdot \Delta\lambda}{\sum_{i=1}^{n} S(\lambda_i) \cdot \Delta\lambda} \tag{S20}$$

And the contrast under continuous spectrum can be deduced from Eq. S19.

Reference

1 I. Bilgin, F. Liu, A. Vargas, A. Winchester, M. K. L. Man, M. Upmanyu, K. M. Dani, G. Gupta, S. Talapatra, A. D. Mohite and S. Kar, *ACS Nano*, 2015, **9**, 8822-8832.

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3 M. Born, E. Wolf, Principles of optics: electromagnetic theory of propagation, interference and diffraction of light, 1964, Page 54.