Optical identification of layered MoS$_2$ via characteristic matrix method

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Supplementary Information (S1)

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.)

<table>
<thead>
<tr>
<th>Sample (a)</th>
<th>MoS$_2$ layer number</th>
<th>$I_{exp}$ (a.u.)</th>
<th>$I_{exp}(\text{MoS}<em>2)-I</em>{exp}(\text{substrate})$</th>
<th>$C_{exp}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>142.0</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>1</td>
<td>127.0</td>
<td>-15</td>
<td>-0.1056</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>117.5</td>
<td>-24.5</td>
<td>-0.2085</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>151.0</td>
<td>-40.5</td>
<td>-0.2115</td>
<td></td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>Sample (b)</th>
<th>MoS$_2$ layer number</th>
<th>$I_{exp}$ (a.u.)</th>
<th>$I_{exp}(\text{MoS}<em>2)-I</em>{exp}(\text{substrate})$</th>
<th>$C_{exp}$</th>
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</thead>
<tbody>
<tr>
<td>1</td>
<td>181.5</td>
<td>-10</td>
<td>-0.0522</td>
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</tr>
<tr>
<td>2</td>
<td>162.0</td>
<td>-29.5</td>
<td>-0.1540</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>146.0</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sample (c)</th>
<th>MoS$_2$ layer number</th>
<th>$I_{exp}$ (a.u.)</th>
<th>$I_{exp}(\text{MoS}<em>2)-I</em>{exp}(\text{substrate})$</th>
<th>$C_{exp}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>x</td>
<td>140.5</td>
<td>-5.5</td>
<td>-0.0377</td>
<td></td>
</tr>
<tr>
<td>y</td>
<td>135.0</td>
<td>-11</td>
<td>-0.0753</td>
<td></td>
</tr>
</tbody>
</table>

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 \, \mu m 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 C_{theor} and C_{exp} 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$_2$.

\[
M_{1L,MoS2} = \begin{bmatrix} A & B \\ C & D \end{bmatrix},
\]

where:

\[
A = \cos(\delta_{MoS2}),
\]
\[
B = j \cdot \sin(\delta_{MoS2}) / \eta_{MoS2},
\]
\[
C = j \cdot \sin(\delta_{MoS2}) \cdot \eta_{MoS2},
\]
\[
D = \cos(\delta_{MoS2}),
\]

\[
\delta_{MoS2} = 2\pi \cdot N_{MoS2} \cdot d_{MoS2}
\]
\[
\eta_{MoS2} = \sqrt{\frac{\varepsilon_0}{\mu_0}} \cdot N_{MoS2}
\]

$N_{MoS2}$ is the wavelength-dependent complex refractive index of MoS$_2$ (Fig. S2 (a)). For monolayer, $N_{1L} = n_{1L} - jk_{1L}$; for bulk, $N_{bulk} = n_{bulk} - jk_{bulk}$. $\varepsilon_0$ is the permittivity of vacuum (8.854187817e-12) and $\mu_0$ 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$_2$:

\[
M = M_{1L,MoS2}
\]

For 2L MoS$_2$:

\[
M = M_{1L,MoS2} \cdot M_{1L,MoS2}^2
\]

For 3L MoS$_2$:

\[
M = M_{1L,MoS2}^3
\]

\[
\vdots
\]

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:

\[
M = M_{1L,MoS2} \cdot M_G \cdot M_{SiO2}
\]

$M_G$ 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:

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

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

The reflection coefficient of film:

\[
r = \frac{(A + B \cdot \eta_G) \cdot \eta_0 - (C + D \cdot \eta_G)}{(A + B \cdot \eta_G) \cdot \eta_0 + (C + D \cdot \eta_G)}
\]

The reflectivity of film:

\[
R = r \cdot r^* \quad (S14)
\]
\[ r_0 = \frac{N_0 - N_G}{N_0 + N_G} \]  
\[ R_0 = r_0 \cdot r_0^* \]  
\[ \eta_0 = \frac{\varepsilon_0}{\mu_0} \cdot N_0 \]  
\[ \eta_G = \frac{\varepsilon_0}{\mu_0} \cdot N_G \]  
\[ \text{Contrast} = \frac{R - R_0}{R_0} \]

The reflectivity of substrate:

\[ R_0 = r_0 \cdot r_0^* \]  
\[ \eta_0 = \frac{\varepsilon_0}{\mu_0} \cdot N_0 \]  
\[ \eta_G = \frac{\varepsilon_0}{\mu_0} \cdot N_G \]

Optical contrast:

\[ \text{Contrast} = \frac{R - R_0}{R_0} \]

\( N_0 \) 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 \((\text{Fig. S2 (b)})\).

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 \((\text{Fig. 3 (c)})\). Because the function of \( S(\lambda) \) and \( R(\lambda) \) were unknown, we used the numerical integration \((S20)\) to carry out the calculation.

The reflectivity under continuous spectrum:

\[ R = \frac{\sum_{i=1}^{n} S(\lambda_i) \cdot R(\lambda_i) \cdot \Delta\lambda}{\sum_{i=1}^{n} S(\lambda_i) \cdot \Delta\lambda} \]

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

\[ \text{Contrast} = \frac{R - R_0}{R_0} \]

Reference