Supporting Information

Energy Transport State Resolved Raman for Probing Interface Energy Transport and Hot Carrier Diffusion in Few-Layered MoS$_2$

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S1. Physical model details for picosecond Raman

In the pico-second Raman (zero-transport state), we apply a ps laser (repetition rate is 48.2 MHz, the pulse duration is 13 ps) under 50× (NA=0.5) objective with $r_0$ as 0.531 μm (1/e peak value) with the same wavelength as the CW laser to focus on the sample to do localized heating and Raman experiment. Using the same wavelength for the two laser sources ensures that the samples have the same laser absorption level in two heating states. In the same way, we obtain the RSC for this ps laser heating case: $\chi_{ps}=\omega\delta P$. The collected Raman signal reflects the Raman signal-weighted temperature rise during ps laser heating. Within this very short heating time (13
ps), the thermal diffusion length ($L_t$) for MoS$_2$ and c-Si is only around 38 nm and 68 nm, respectively. ($L_t = 2\sqrt{\alpha_k t_0}$, $\alpha_k=2.75\times10^{-5}$ m$^2$/s for MoS$_2$ and $8.92\times10^{-5}$ m$^2$/s for Si, is the in-plane thermal diffusivity; $t_0=13$ ps, is the ps laser pulse width). They are much smaller than the laser spot size ($r_0=0.53$ μm). So we could neglect the heat conduction from the heating region.

Besides, the thermal relaxation time of the MoS$_2$ nanosheets for our interface structure ($R$ is in the order of $10^{-7}$ K·m$^2$/W) is estimated to be around 1.9 ns ($=\delta z\cdot \rho c_p R$. $\delta z$ takes 10 nm, is the thickness of MoS$_2$ nanosheets; $\rho c_p=1.89$ MJ/m$^3$K, is the volumetric heat capacity, $R$ takes $10^{-7}$ K·m$^2$/W, is the interface thermal resistance.) which is much longer than the heating time (13 ps) and much shorter than the laser cooling time (20.8 ns). Therefore, during ps laser heating, the energy absorbed in MoS$_2$ is used to heat up the sample, but is not transferred out of the heating region to other MoS$_2$ region or the substrate. Also the sample will be fully cooled to the ambient temperature during the pulse interval. This ensures the RSC measured during ps laser heating has no DC accumulation effect from previous pulses. In summary, the RSC ($\chi_{ps}$) is mainly coming from the volumetric heat capacity of the sample ($\rho c_p$). $D$ and $R$ have an extremely limited contribution to the temperature rise so that we could use this heating state to distinguish the roles of $\rho c_p$ and ($R, D$).

While evaluating the above different states, we do not consider the heat loss to the environment during the Raman measurement in atmospheric condition. This is because, as has been studied for the supported 2D material, the heat loss through radiation or natural convection is negligible compared to the interfacial heat flux. In our steady-state experiment, the measured RSC is proportional to the temperature rise of the two materials, which is a Raman/laser intensity weighted average over the laser spot size as
For the zero-transport state, the measured temperature rise includes both time and space averaged over the pulse width and heating domain as

\[
\overline{T}_{\text{ps}} = \int_0^{\tau_p} \int_0^{V_v} I_a(r,t)T(r,t)dv \int_0^{V_v} I_a(r)dv .
\]  

(S2)

S2. Data Reduction details

When a laser beam irradiates the sample surface, multiple reflections happen at the interface between MoS₂ and c-Si as shown in Figure 6(b). Based on the optical properties of these two materials and the Transfer Matrix Method (TMM),² we could determine the transmitted power at the top surface \( I_{01} \), the reflected power at the bottom surface \( I_{02} \) of MoS₂ and the transmitted power in c-Si top surface \( I_{03} \). Our method could eliminate the errors from the local laser absorption evaluation and temperature coefficient calibration. We have following detailed explanations.

The temperature coefficient \( \chi_T \) for MoS₂ and c-Si is unknown in our work. For steady-state Raman, the temperature rise (K/mW) of MoS₂ is from two parts: \( \Delta T_1 \) by the direct laser energy absorption and \( \Delta T_2 \) by the thermal energy absorption in c-Si. So we express the temperature rise by RSC as

\[
\chi_{\text{CW, MoS}_2} = (\Delta T_1 + \Delta T_2) \times \chi_{T, \text{MoS}_2} .
\]  

(S3)

We have \( \Delta T_1 \propto (I_{01} + I_{02}) \) and \( \Delta T_2 \propto I_{03} \). Similar to MoS₂, the temperature rise (K/mW) of c-Si
is also from two parts: $\Delta T_3$ by thermal energy absorption in MoS$_2$ and $\Delta T_4$ by the direct laser energy absorption in c-Si. It could be represented by RSC as

$$\chi_{CW, Si} = (\Delta T_3 + \Delta T_4) \times \chi_{T, Si},$$  \hspace{1cm} (S4)

We also have $\Delta T_3 \propto (I_{01} + I_{02})$ and $\Delta T_4 \propto I_{03}$.

For the zero-transport state ps laser Raman, it is simpler. Only laser absorption happens (the fast thermalization process), and there is no heat conduction. So we could express the temperature rise (K/mW) of MoS$_2$ and c-Si by RSC as

$$\chi_{ps, MoS} = \Delta T_5 \times \chi_{T, MoS_2},$$  \hspace{1cm} (S5)

$$\chi_{ps, Si} = \Delta T_6 \times \chi_{T, Si},$$  \hspace{1cm} (S6)

where $\Delta T_5 \propto (I_{01} + I_{02})$ and $\Delta T_6 \propto I_{03}$.

From the 3D simulation, for the steady-state CW laser heating, we could get $\Delta T_1$ and $\Delta T_3$ by setting zero absorption in c-Si, $\Delta T_2$ and $\Delta T_4$ by setting zero absorption in MoS$_2$. From zero-transport state ps laser heating, we could directly get $\Delta T_5$ and $\Delta T_6$ by only considering the laser absorption.

From Eqs. (S3) and (S5), we express the normalized RSC of MoS$_2$ measured in the experiment as

$$\Theta_{MoS_2} = \frac{\chi_{CW, MoS_2}}{\chi_{ps, MoS_2}} = \frac{\Delta T_1}{\Delta T_5} + \frac{\Delta T_2}{\Delta T_5},$$  \hspace{1cm} (S7)
From Eqs. (S4) and (S6), we express the normalized RSC of c-Si as

\[
\Theta_{\text{Si}} = \frac{\chi_{\text{CW, Si}}}{\chi_{\text{ps, Si}}} = \frac{\Delta T_3}{\Delta T_6} + \frac{\Delta T_4}{\Delta T_6}.
\] (S8)

The temperature rises \(\Delta T_1, \Delta T_2, \Delta T_3, \Delta T_5, \) and \(\Delta T_6\) are only related to the laser absorption in two materials. Additionally, we have \((\Delta T_2/\Delta T_3) \propto [I_{03}/(I_{01} + I_{02})]\) and \((\Delta T_3/\Delta T_6) \propto [(I_{01} + I_{02})/I_{03}]\). To eliminate the laser absorption effect, we multiply these two temperature rise ratios from Eqs. (S7) and (S8) as

\[
\Theta_{\text{MoS}_2} = \frac{\Delta T_1}{\Delta T_5} + \frac{(\Delta T_2/\Delta T_3) \times (\Delta T_4/\Delta T_6)}{\Theta_{\text{Si}} - \Delta T_4/\Delta T_6}.
\] (S9)

In Eq. (S9), term \(\Delta T_1/\Delta T_2\) is not affected by laser absorption rate because both \(\Delta T_1\) and \(\Delta T_2\) are proportional to \((I_{01}+I_{02})\). Similarly, term \(\Delta T_4/\Delta T_6\) also has nothing to do with laser absorption rate because both \(\Delta T_4\) and \(\Delta T_6\) are proportional to \(I_{03}\). So the normalized RSC value is now only a function of two parameters: \(\Theta(R, D)\), the effect from laser absorption evaluation is successfully ruled out.

**S3. ET-Raman experiment results summary**

Yim *et al.* have studied the thickness dependent bandgap of MoS\(_2\) nanosheets.\(^3\) The change in the band structure with layer number is due to quantum confinement and the resulting change in hybridization between \(pz\) orbitals and S atoms and \(d\) orbitals on Mo atom.\(^4\) We fit the \(E_g\) values for different thickness samples by an exponential function
\[ E_g (eV) = 0.5836 \times \exp(-\text{thickness}/3.525) + 1.29 \] and extract the \( E_g \) values for our samples as shown in Table S1.

**Table S1.** Summary of Raman experiment results of seven MoS\(_2\) samples. The thickness dependent bandgap is extracted from Yim et al.’s work.\(^3\) The steady-state Raman shift power coefficient (RSC) values under 20\(\times\) and 100\(\times\) objective with CW laser (\(\chi_{\text{CW1}}\) and \(\chi_{\text{CW2}}\)) for \(E_{2g}^1\) mode of MoS\(_2\) and c-Si. The zero-transport state RSC values under 50\(\times\) objective with ps laser (\(\chi_{\text{ps}}\)). Also the normalized RSC (\(\Theta_1\) and \(\Theta_2\)).

<table>
<thead>
<tr>
<th>Sample thickness</th>
<th>Band gap</th>
<th>Raman mode</th>
<th>(\chi_{\text{CW1}}) (cm(^{-1})/mW)</th>
<th>(\chi_{\text{CW2}}) (cm(^{-1})/mW)</th>
<th>(\chi_{\text{ps}}) (cm(^{-1})/mW)</th>
<th>(\Theta_1)</th>
<th>(\Theta_2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.6 nm 1.38</td>
<td></td>
<td>(E_{2g}^1) of MoS(_2)</td>
<td>-(0.026(\pm)1.1(\times)10(^{-3}))</td>
<td>-(0.150(\pm)6.5(\times)10(^{-3}))</td>
<td>-(0.181(\pm)6.3(\times)10(^{-3}))</td>
<td>-(0.145(\pm)8.1(\times)10(^{-3}))</td>
<td>-(0.829(\pm)4.6(\times)10(^{-2}))</td>
</tr>
<tr>
<td></td>
<td></td>
<td>c-Si</td>
<td>-(0.008(\pm)2.8(\times)10(^{-4}))</td>
<td>-(0.029(\pm)1.8(\times)10(^{-4}))</td>
<td>-(0.057(\pm)1.8(\times)10(^{-4}))</td>
<td>-(0.136(\pm)6.6(\times)10(^{-3}))</td>
<td>-(0.513(\pm)3.5(\times)10(^{-2}))</td>
</tr>
<tr>
<td>7.8 nm 1.35</td>
<td></td>
<td>(E_{2g}^1) of MoS(_2)</td>
<td>-(0.034(\pm)1.5(\times)10(^{-3}))</td>
<td>-(0.171(\pm)9.5(\times)10(^{-3}))</td>
<td>-(0.203(\pm)8.5(\times)10(^{-3}))</td>
<td>-(0.169(\pm)1.0(\times)10(^{-2}))</td>
<td>-(0.841(\pm)5.9(\times)10(^{-2}))</td>
</tr>
<tr>
<td></td>
<td></td>
<td>c-Si</td>
<td>-(0.008(\pm)3.6(\times)10(^{-4}))</td>
<td>-(0.027(\pm)1.5(\times)10(^{-4}))</td>
<td>-(0.051(\pm)2.2(\times)10(^{-4}))</td>
<td>-(0.151(\pm)9.6(\times)10(^{-3}))</td>
<td>-(0.531(\pm)3.8(\times)10(^{-2}))</td>
</tr>
<tr>
<td>9.6 nm 1.33</td>
<td></td>
<td>(E_{2g}^1) of MoS(_2)</td>
<td>-(0.030(\pm)1.0(\times)10(^{-3}))</td>
<td>-(0.158(\pm)6.4(\times)10(^{-3}))</td>
<td>-(0.193(\pm)6.4(\times)10(^{-3}))</td>
<td>-(0.157(\pm)7.4(\times)10(^{-3}))</td>
<td>-(0.818(\pm)4.3(\times)10(^{-3}))</td>
</tr>
<tr>
<td></td>
<td></td>
<td>c-Si</td>
<td>-(0.009(\pm)1.1(\times)10(^{-3}))</td>
<td>-(0.027(\pm)1.2(\times)10(^{-3}))</td>
<td>-(0.050(\pm)1.2(\times)10(^{-3}))</td>
<td>-(0.166(\pm)4.2(\times)10(^{-3}))</td>
<td>-(0.523(\pm)2.6(\times)10(^{-2}))</td>
</tr>
<tr>
<td>12.0 nm 1.31</td>
<td></td>
<td>(E_{2g}^1) of MoS(_2)</td>
<td>-(0.040(\pm)1.5(\times)10(^{-3}))</td>
<td>-(0.187(\pm)6.6(\times)10(^{-3}))</td>
<td>-(0.197(\pm)7.9(\times)10(^{-3}))</td>
<td>-(0.204(\pm)1.1(\times)10(^{-2}))</td>
<td>-(0.947(\pm)5.1(\times)10(^{-2}))</td>
</tr>
<tr>
<td></td>
<td></td>
<td>c-Si</td>
<td>-(0.009(\pm)2.8(\times)10(^{-3}))</td>
<td>-(0.029(\pm)6.2(\times)10(^{-3}))</td>
<td>-(0.056(\pm)2.9(\times)10(^{-3}))</td>
<td>-(0.157(\pm)9.6(\times)10(^{-3}))</td>
<td>-(0.517(\pm)2.9(\times)10(^{-2}))</td>
</tr>
<tr>
<td>13.2 nm 1.30</td>
<td></td>
<td>(E_{2g}^1) of MoS(_2)</td>
<td>-(0.033(\pm)1.3(\times)10(^{-3}))</td>
<td>-(0.175(\pm)8.3(\times)10(^{-3}))</td>
<td>-(0.187(\pm)6.0(\times)10(^{-3}))</td>
<td>-(0.175(\pm)9.1(\times)10(^{-3}))</td>
<td>-(0.935(\pm)5.4(\times)10(^{-3}))</td>
</tr>
<tr>
<td></td>
<td></td>
<td>c-Si</td>
<td>-(0.008(\pm)3.8(\times)10(^{-4}))</td>
<td>-(0.025(\pm)1.0(\times)10(^{-3}))</td>
<td>-(0.059(\pm)3.2(\times)10(^{-3}))</td>
<td>-(0.141(\pm)9.9(\times)10(^{-3}))</td>
<td>-(0.430(\pm)2.9(\times)10(^{-2}))</td>
</tr>
<tr>
<td>15.6 nm 1.30</td>
<td></td>
<td>(E_{2g}^1) of MoS(_2)</td>
<td>-(0.040(\pm)1.1(\times)10(^{-3}))</td>
<td>-(0.189(\pm)7.7(\times)10(^{-3}))</td>
<td>-(0.201(\pm)5.6(\times)10(^{-3}))</td>
<td>-(0.197(\pm)7.9(\times)10(^{-3}))</td>
<td>-(0.940(\pm)4.6(\times)10(^{-3}))</td>
</tr>
<tr>
<td></td>
<td></td>
<td>c-Si</td>
<td>-(0.008(\pm)5.3(\times)10(^{-4}))</td>
<td>-(0.026(\pm)8.7(\times)10(^{-4}))</td>
<td>-(0.050(\pm)2.7(\times)10(^{-3}))</td>
<td>-(0.164(\pm)1.4(\times)10(^{-3}))</td>
<td>-(0.514(\pm)3.2(\times)10(^{-2}))</td>
</tr>
<tr>
<td>17.4 nm 1.30</td>
<td></td>
<td>(E_{2g}^1) of MoS(_2)</td>
<td>-(0.045(\pm)2.2(\times)10(^{-3}))</td>
<td>-(0.217(\pm)9.4(\times)10(^{-3}))</td>
<td>-(0.210(\pm)4.5(\times)10(^{-3}))</td>
<td>-(0.212(\pm)1.1(\times)10(^{-2}))</td>
<td>-(1.03(\pm)5.0(\times)10(^{-2}))</td>
</tr>
<tr>
<td></td>
<td></td>
<td>c-Si</td>
<td>-(0.006(\pm)1.7(\times)10(^{-4}))</td>
<td>-(0.021(\pm)7.8(\times)10(^{-4}))</td>
<td>-(0.054(\pm)1.2(\times)10(^{-3}))</td>
<td>-(0.114(\pm)4.0(\times)10(^{-3}))</td>
<td>-(0.386(\pm)1.7(\times)10(^{-2}))</td>
</tr>
</tbody>
</table>
S4. Laser pulse accumulation effect from substrate

For the pulsed laser (ps laser) heating, the temperature rise of the sample is from a single pulse and the steady-state accumulation of the heat. In our experiment, the time \( t_c \) between pulses from ps laser is around 20.8 ns as shown in Fig. 1(e). Moreover, compared to SiO\(_2\) substrate, c-Si has a larger thermal conductivity and therefore could dissipate the heat more efficiently.\(^5\) Besides, the thermal diffusion length \( L_t \) in c-Si within one repetition period could be estimated as

\[
L_t = 2\sqrt{\alpha_k t_c} = 2.71 \, \mu m,
\]

where \( \alpha_k \) is the thermal diffusivity of c-Si. Scaling this result to the three dimensions, the heat will outspread over a hemisphere of radius \( L_t \) after a \( t_c \) has elapsed. The temperature of the sample will be cooled down to

\[
\Delta T = \frac{r_0^2 (\text{ps laser, } 50 \times \pi \times L_{\text{thickness}})}{(2/3 \times \pi \times L_t^3)}
\]

of its original. For the 6.6 nm thick MoS\(_2\) sample, \( \Delta T = 15.8 \, \text{K/mW} \). In our ps laser Raman experiment, the highest temperature of MoS\(_2\) is heated to

\[
\Delta T_{6.6\,\text{nm}} = 15.8 \, \text{K/mW} \times 6.93 \, \text{mW} = 109 \, \text{K}
\]

It will be cooled down to only around 0.02 K before the next pulse arrives. As a result, we will not consider this pulse accumulation effect from substrate in this work.
S5. The effect of optical properties

Table S2. The study results for the effect of the optical properties of MoS2 on final normalized RSC values. By varying the complex index of MoS2, based on the calculated temperature rise $\Delta T_i$ to $\Delta T_6$, $\Theta(\text{MoS}_2)$ has a variance of less than one thousandth of that based on the original complex index (4.4-1.1i).

<table>
<thead>
<tr>
<th>Objective lens</th>
<th>Complex index</th>
<th>$\Delta T_1$ (K)</th>
<th>$\Delta T_2$ (K)</th>
<th>$\Delta T_3$ (K)</th>
<th>$\Delta T_4$ (K)</th>
<th>$\Delta T_5$ (K)</th>
<th>$\Delta T_6$ (K)</th>
<th>$\Theta(\text{MoS}_2)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>20×</td>
<td>4.4-1.1i</td>
<td>1.95</td>
<td>0.168</td>
<td>0.124</td>
<td>0.162</td>
<td>17.24</td>
<td>0.629</td>
<td>0.097</td>
</tr>
<tr>
<td></td>
<td>2.2-1.1i</td>
<td>2.30</td>
<td>0.203</td>
<td>0.146</td>
<td>0.197</td>
<td>20.30</td>
<td>0.762</td>
<td>+0.11%</td>
</tr>
<tr>
<td></td>
<td>5.5-1.1i</td>
<td>1.80</td>
<td>0.143</td>
<td>0.114</td>
<td>0.139</td>
<td>15.87</td>
<td>0.538</td>
<td>+0.05%</td>
</tr>
<tr>
<td></td>
<td>4.4-0.55i</td>
<td>2.08</td>
<td>0.193</td>
<td>0.132</td>
<td>0.187</td>
<td>18.43</td>
<td>0.723</td>
<td>-0.21%</td>
</tr>
<tr>
<td></td>
<td>4.4-2.2i</td>
<td>1.70</td>
<td>0.129</td>
<td>0.108</td>
<td>0.125</td>
<td>15.05</td>
<td>0.484</td>
<td>-0.02%</td>
</tr>
<tr>
<td>100×</td>
<td>4.4-1.1i</td>
<td>11.80</td>
<td>0.335</td>
<td>0.242</td>
<td>0.358</td>
<td>17.24</td>
<td>0.629</td>
<td>0.550</td>
</tr>
<tr>
<td></td>
<td>2.2-1.1i</td>
<td>13.89</td>
<td>0.406</td>
<td>0.285</td>
<td>0.433</td>
<td>20.31</td>
<td>0.762</td>
<td>+0.21%</td>
</tr>
<tr>
<td></td>
<td>5.5-1.1i</td>
<td>10.86</td>
<td>0.287</td>
<td>0.223</td>
<td>0.306</td>
<td>15.87</td>
<td>0.538</td>
<td>+0.50%</td>
</tr>
<tr>
<td></td>
<td>4.4-0.55i</td>
<td>12.61</td>
<td>0.385</td>
<td>0.259</td>
<td>0.411</td>
<td>18.43</td>
<td>0.723</td>
<td>-0.13%</td>
</tr>
<tr>
<td></td>
<td>4.4-2.2i</td>
<td>10.29</td>
<td>0.258</td>
<td>0.211</td>
<td>0.275</td>
<td>15.05</td>
<td>0.484</td>
<td>-0.10%</td>
</tr>
</tbody>
</table>
S6. Sensitivity of $D$ and $R$ to MoS$_2$ in-plane thermal conductivity

To study how $D$ and $R$ value is sensitive to the MoS$_2$ in-plane thermal conductivity ($k_\parallel$), we perform the 3D modeling by varying the $k_\parallel$ value. Without changing other parameters, we calculate the normalized RSC values to extract $D$ and $R$ as shown in Table S3. While increasing $k_\parallel$ by 10%, $R$ is not sensitive to that. This is because $D$ and $k_\parallel$ only contribute to the in-plane thermal energy distribution. However, $D$ is very sensitive to $k_\parallel$ and decreases by 16.6%. During the diffusion process, the hot carrier only carries ~60% ($E_g/E$) of the photon energy and the phonon could carry the total photon energy. To keep the same temperature drop, $D$ is supposed to change more.

Table S3. The study results for the sensitivity of $D$ and $R$ to MoS$_2$ in-plane thermal conductivity ($k_\parallel$). By only varying $k_\parallel$ of MoS$_2$, based on the calculated temperature rise $\Delta T_1$ to $\Delta T_6$, $D$ is sensitive to the $k_\parallel$ value.

<table>
<thead>
<tr>
<th>Thermal conductivity $k_\parallel$ (W/m·K)</th>
<th>$D$ (cm$^2$/s)</th>
<th>$R$ (10$^{-7}$ K·m$^2$/W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>52</td>
<td>$0.637^{+0.193}_{-0.154}$</td>
<td>$1.75^{+0.08}_{-0.08}$</td>
</tr>
<tr>
<td>57.2 (+10%)</td>
<td>$0.531^{+0.163}_{-0.134}$ (-16.6%)</td>
<td>$1.75^{+0.08}_{-0.08}$</td>
</tr>
<tr>
<td>46.8 (-10%)</td>
<td>$0.771^{+0.206}_{-0.169}$ (+21.0%)</td>
<td>$1.76^{+0.08}_{-0.08}$</td>
</tr>
</tbody>
</table>
S7. Interference Induced Raman Enhancement

The multiple reflections of the incident laser beam and Raman signal within the supported nanosheets and the spacing have been studied in previous work. The net absorption factor \( F_{ab} \) is given by

\[
F_{ab} = t_1 \left( 1 + r_1 r_5 e^{2i\beta_5} e^{-i\beta_1} + (r_2 + r_5 e^{2i\beta_5}) e^{i(2\beta_1 \cdot \beta_2)} \right) \frac{1}{1 + r_2 r_3 e^{2i\beta_3} + (r_2 + r_5 e^{2i\beta_5}) r_3 e^{2i\beta_3}},
\]

(S10)

where

\[
t_1 = 2 \tilde{n}_0 / (\tilde{n}_0 + \tilde{n}_1), \quad r_1 = (\tilde{n}_0 - \tilde{n}_1) / (\tilde{n}_0 + \tilde{n}_1), \quad r_2 = (\tilde{n}_1 - \tilde{n}_2) / (\tilde{n}_1 + \tilde{n}_2), \quad \text{and} \quad r_3 = (\tilde{n}_2 - \tilde{n}_3) / (\tilde{n}_2 + \tilde{n}_3)
\]

are Fresnel transmittance and reflection coefficients for the interface involving air (0), MoS\(_2\) nanosheets (1), air (2), and Si (3). \( \tilde{n}_0, \tilde{n}_1, \tilde{n}_2, \) and \( \tilde{n}_3 \) are the refractive indices for air, MoS\(_2\) nanosheets, air, and c-Si, respectively. \( \beta_1 = 2\pi x \tilde{n}_1 / \lambda, \beta_2 = 2\pi d_1 \tilde{n}_1 / \lambda \) and \( \beta_2 = 2\pi d_2 \tilde{n}_2 / \lambda \), where \( x \) is the depth of the point where interaction occurs, \( \lambda \) is the wavelength of the incident laser (532 nm), \( d_1 \) and \( d_2 \) are the thickness of MoS\(_2\) nanosheets and c-Si, respectively. We assume there is no spacing between MoS\(_2\) nanosheets and the c-Si substrate. The Raman intensity variation with the MoS\(_2\) thickness will be compared with the experimental results to evaluate whether there is an interface spacing.

Similarly, the net scattering factor \( F_{sc} \) is described by

\[
F_{sc} = t'_1 \left( 1 + r_1 r_5 e^{2i\beta_5} e^{-i\beta_1} + (r_2 + r_5 e^{2i\beta_5}) e^{i(2\beta_1 \cdot \beta_2)} \right) \frac{1}{1 + r_2 r_3 e^{2i\beta_3} + (r_2 + r_5 e^{2i\beta_5}) r_3 e^{2i\beta_3}},
\]

(S11)

where \( t'_1 = 2 \tilde{n}_1 / (\tilde{n}_0 + \tilde{n}_1) \) and \( \lambda \) is the wavelength of the \( E_{2g}^1 \) mode of MoS\(_2\). Then we could express the theoretical Raman intensity \( F \) of MoS\(_2\) as
In the calculation, the refractive index of different thick MoS₂ nanosheets is extracted from Yim et al.’s work. The refractive indices of c-Si are 4.15 + 0.05i and 3.99 + 0.33i for incident laser and Raman scattering, respectively.
References


