

Supporting Information

Dynamic repulsive interaction enables an asymmetric electron-phonon coupling for improving Raman scattering

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Enhancement factor (EF) calculation method

The (enhancement factor) EF is calculated according to the following formula:

$$EF = \frac{I_{SERS} / N_{SERS}}{I_{norm} / N_{norm}} \quad \backslash *$$

MERGEFORMAT (1)

where

$$N_{SERS} = \frac{CVN_A A_{Raman}}{A_{sub}} \quad \backslash *$$

MERGEFORMAT (2)

$$N_{norm} = \frac{\rho h N_A A_{Raman}}{M} \quad \backslash *$$

MERGEFORMAT (3)

Thus

$$\frac{N_{norm}}{N_{SERS}} = \frac{\rho h A_{sub}}{CVM} \quad \backslash *$$

MERGEFORMAT (4)

$$EF = \frac{I_{SERS}}{I_{norm}} \times \frac{\rho h A_{sub}}{CVM} \quad \backslash *$$

MERGEFORMAT (5)

Table 1 Parameters and values in equations above.

Symbol	Meaning	Value
EF	Enhancement factor	2.15×10^8 (611 cm^{-1}) 5.59×10^7 (772 cm^{-1})
I_{SERS}	Intensity of selected peak in SERS spectra	29.10 (611 cm^{-1} , 1×10^{-12} mol/L) 14.08 (772 cm^{-1} , 1×10^{-11} mol/L)
N_{SERS}	Probe molecules number illuminated by laser in SERS spectra	(calculated)
I_{norm}	Intensity of selected peak in normal Raman spectra	6767.46 (611 cm^{-1} , original value) 3426.66 (772 cm^{-1} , original value)
N_{norm}	Probe molecules number illuminated by laser in normal Raman spectra	(calculated)

C	Molar concentration of probe molecule	1×10^{-12} mol/L
V	Volume of probe molecule solution droplet	10 μ L
N_A	Avogadro constant	6.02×10^{23}
A_{Raman}	Laser spot area	1 μ m (diameter)
A_{sub}	Area covered by probe molecule solution	$\sim \pi/4$ cm ²
ρ	Mass density of probe molecule	0.79 g/cm ³
h	Penetration depth of laser	~ 21 μ m
M	Molar molecular weight of probe molecule	479 g/mol

For the acquisition of I_{norm} , R6G powder was dissolved into 1 mol/L and dripped to silicon wafer to conduct normal Raman test when completed dried. All spectra and intensity values acquired in the same circumstance were averaged (take the median) by at least 10 instances to minimize inevitable testing fluctuation. **Fig. S1** presents the contrast of different results of R6G powder and completely dried solution with significant intensity deviation. Comparing to the powder, dried solution possesses tightly converged molecules that contributed to the signal. Additionally, powder will scatter most of the incident photons as well as Raman photons.

Besides, the probe molecules adsorbed on substrate and silicon wafer were considered to be uniform with surface number density ρ_s based on following formula:

$$\rho_s = \frac{CVN_A}{A_{sub}} \quad |*$$

MERGEFORMAT (6)

The ρ_s under 1×10^{-12} mol/L would be 7.67×10^{10} molecules/m².

Polarized Raman test method

Polarized Raman testing was facilitated by instruments integrated with ‘half-wave plate’ (retarder). The polarization angle of laser could be adjusted by rotating half-wave plate. Samples were placed on platform and not rotated manually. Raman signals were acquired under certain rotation angle with increment of 15 degree, according to **Fig. 4(d)**. Notably, the polarization angle of transmitted laser will rotate 2θ degree if half-wave plate was rotated θ degree, leading to the 4-lobed shape of polarized diagram in **Fig. 4(d)**.

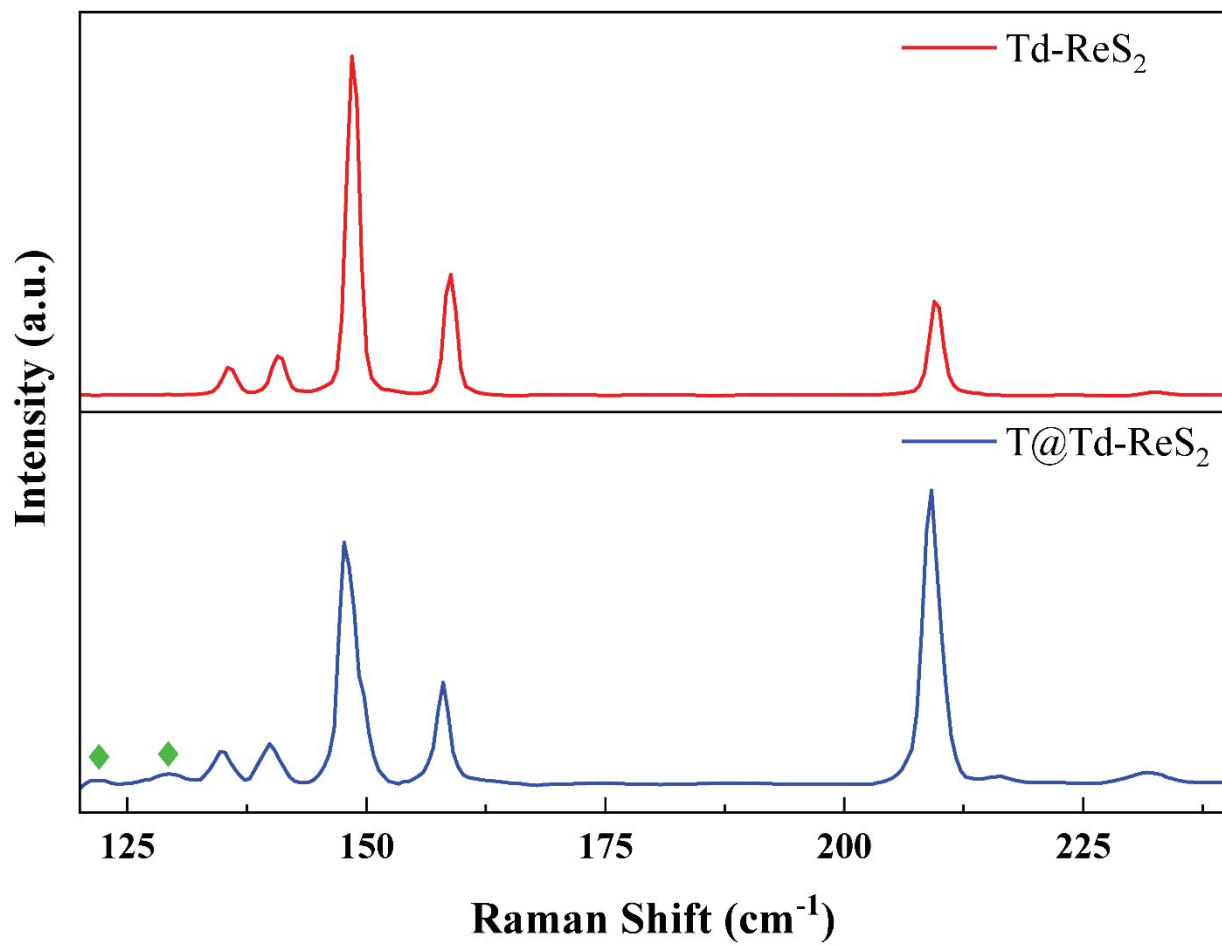


Fig. S1 Raman signal of Td-ReS₂ and T@ Td-ReS₂, where green rhombus represents the newly introduced Raman peak on 122 cm⁻¹ and 129 cm⁻¹.

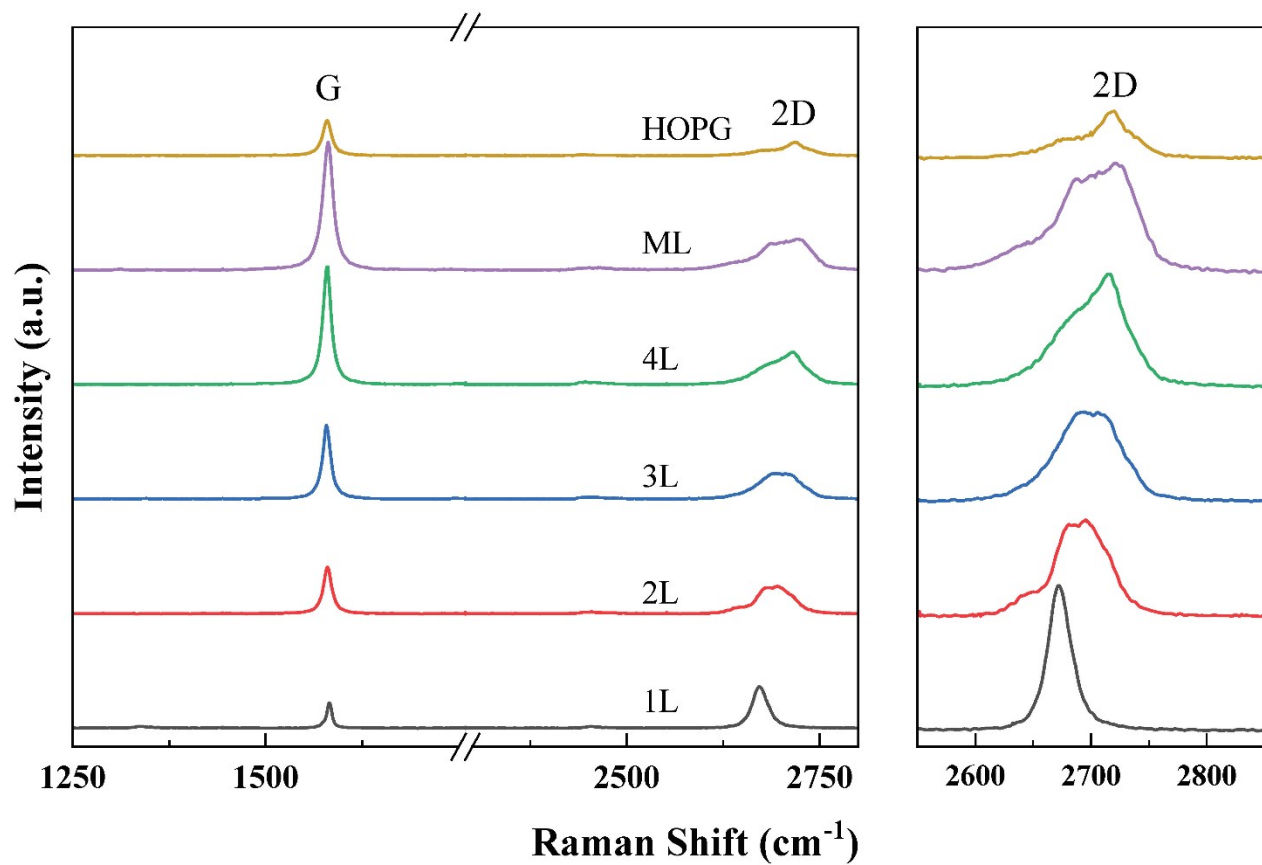


Fig. S2 Raman signal of 1-4 layer, multilayer graphene and HOPG.

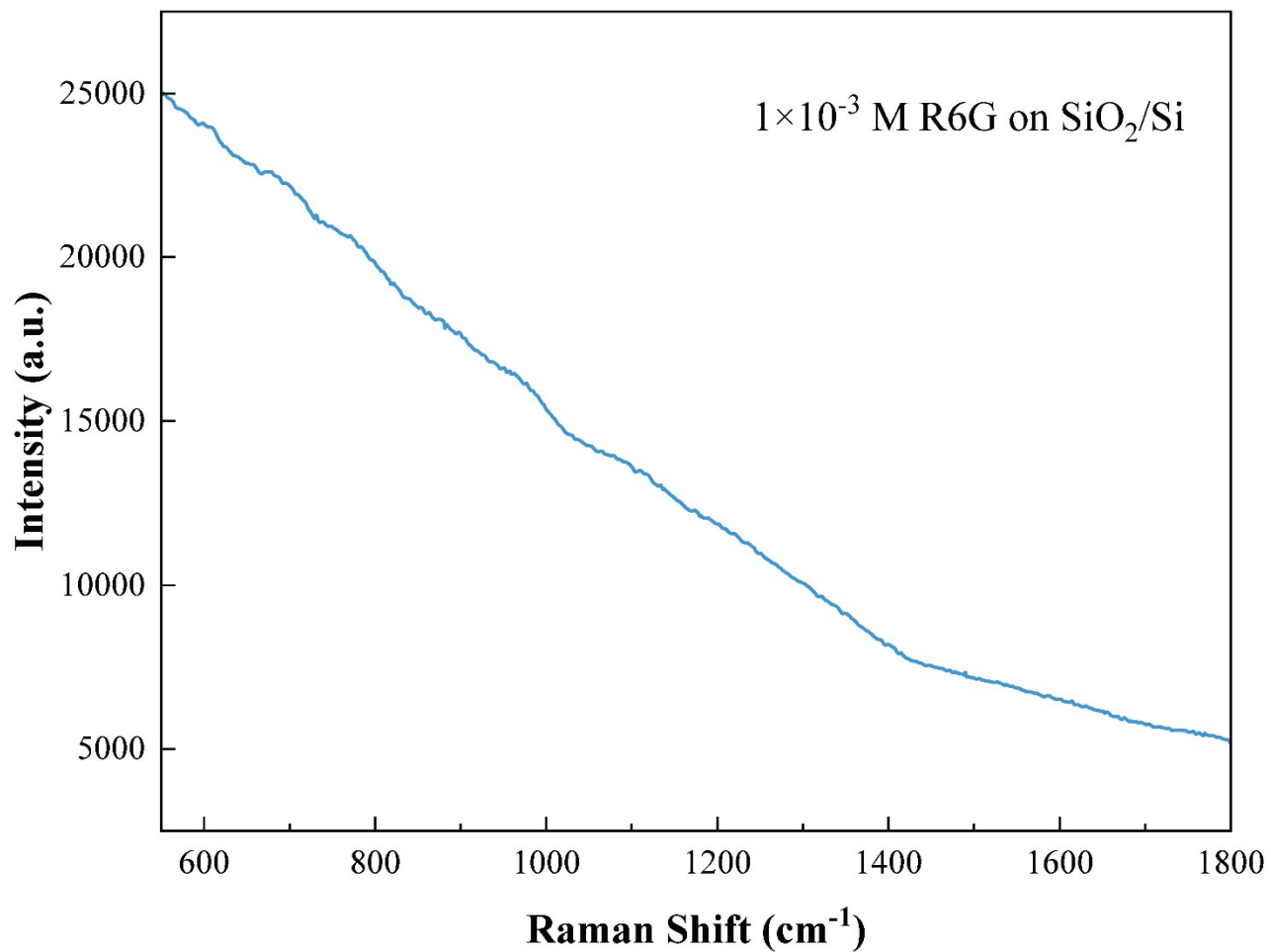


Fig. S3 Intensive fluorescence background of 1×10^{-3} M R6G solution.

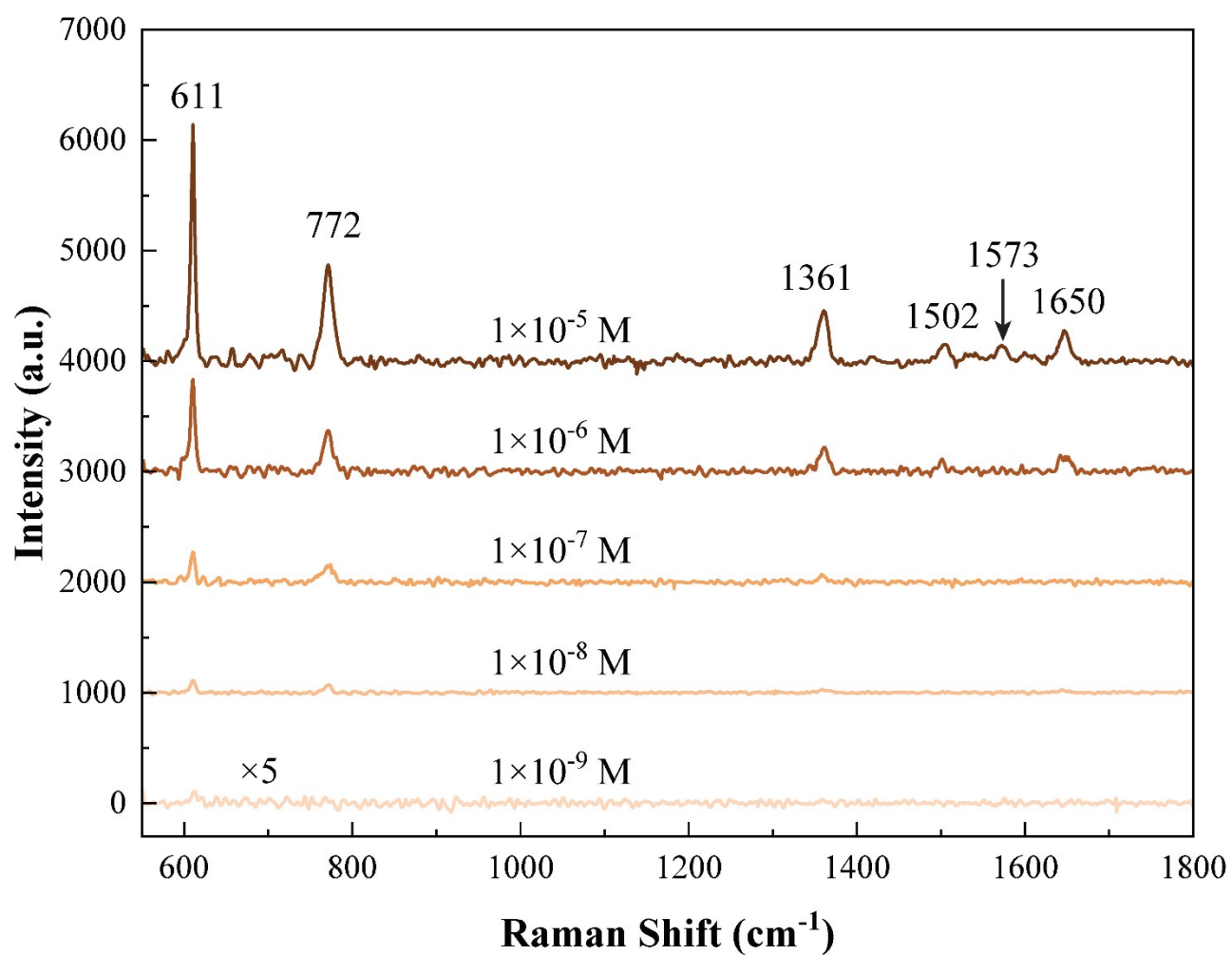


Fig. S4 SERS spectra of R6G on T@Td-ReS₂ as control experiment with LOD of 1×10⁻⁹ M.

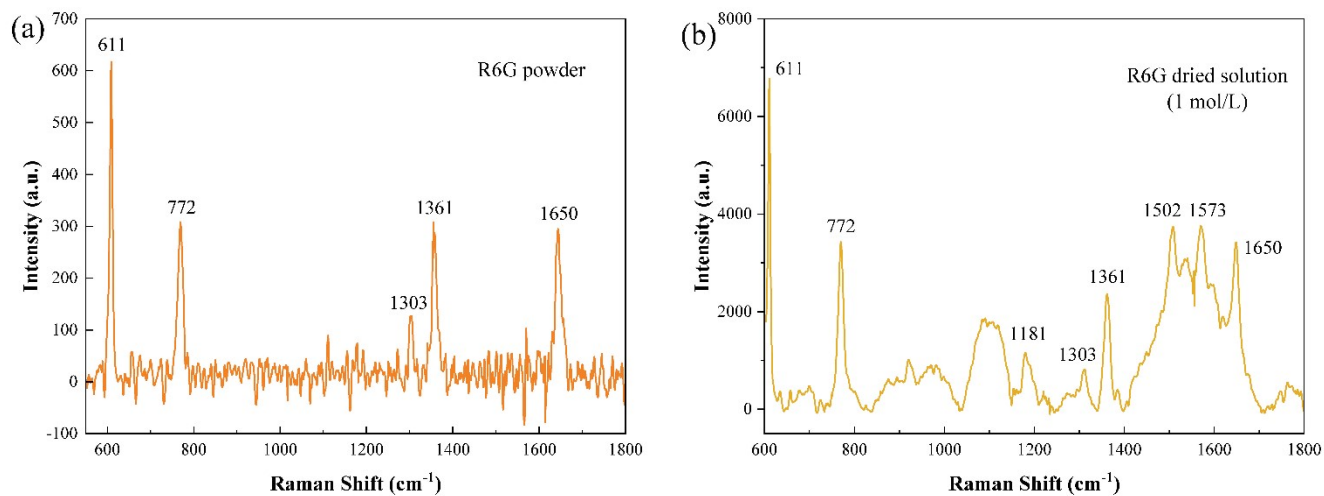


Fig. S5 Raman signal of (a) R6G powder and (b) 1 M dried R6G solution. The EFs were calculated based on the intensity of 611 cm⁻¹ and 772 cm⁻¹ in (b).

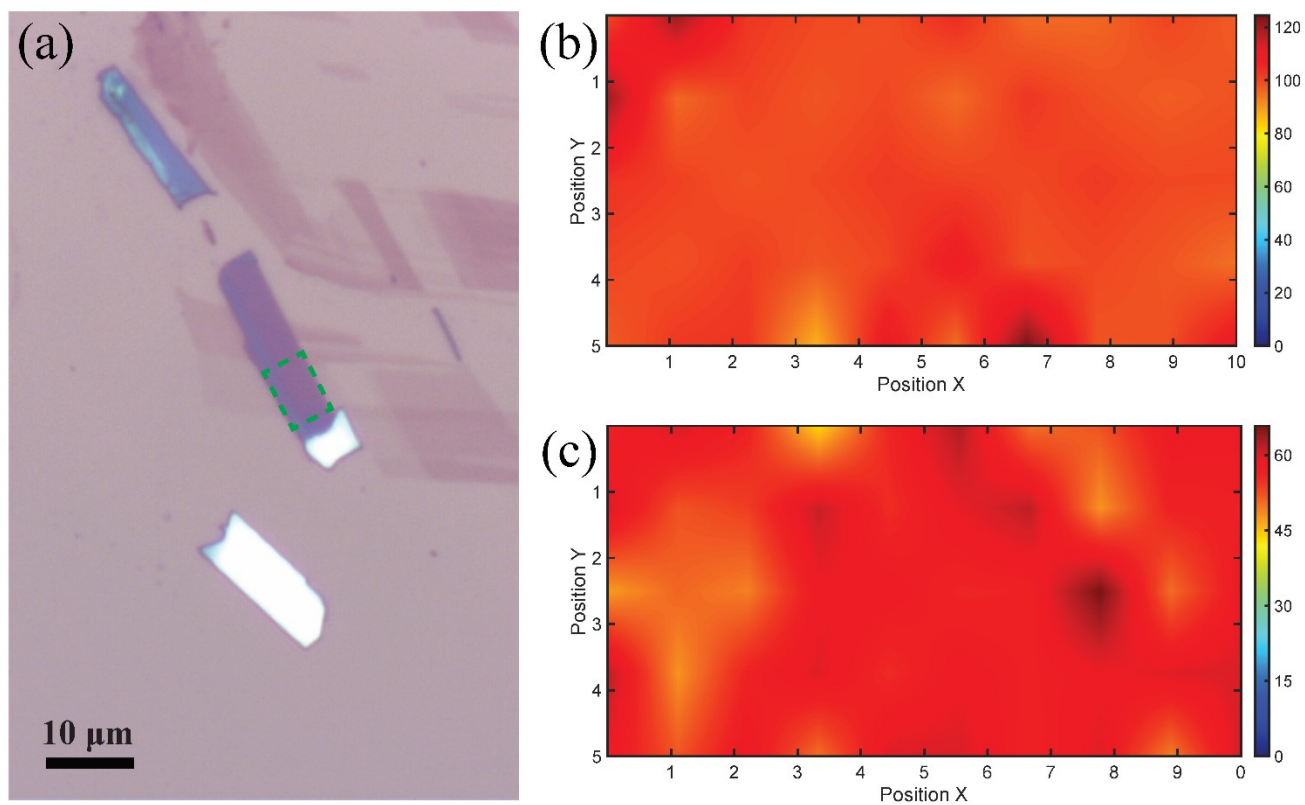


Fig. S6 (a) Optical microscopic image of the selected mapping area. Heatmap of (b) 611 cm^{-1} and 772 cm^{-1} .

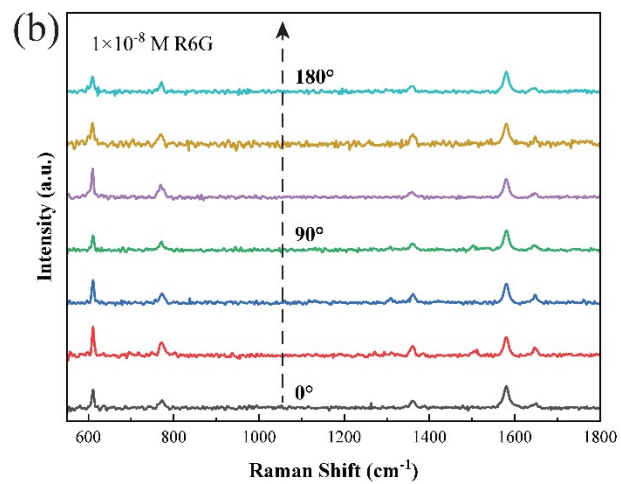
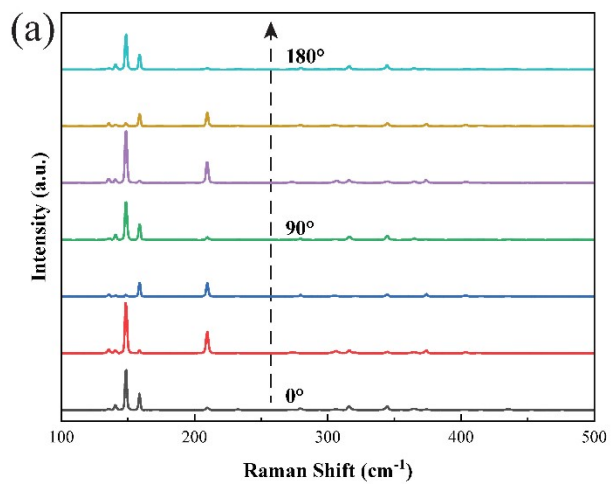


Fig. S7 Angle-resolved Raman spectra of (a) ReS_2 and (b) adsorbed R6G with concentration of $1 \times 10^{-8} \text{ M}$

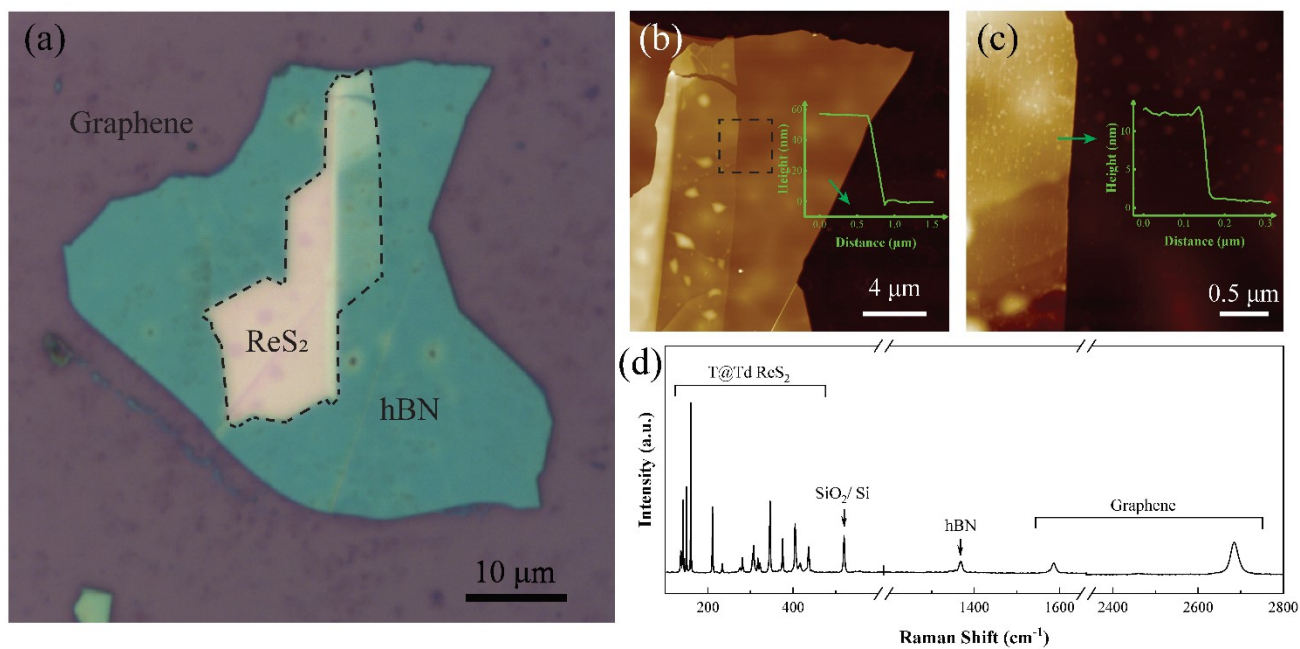


Fig. S8 (a) Optical microscopic image of T@Td-ReS₂/hBN/G heterostructure. (b) AFM image of heterostructure presented in (a). Blue arrow indicates the cross section direction. (c) Zoomed AFM image of area marked by dashed-line box in (b) and (d) corresponding cross section.

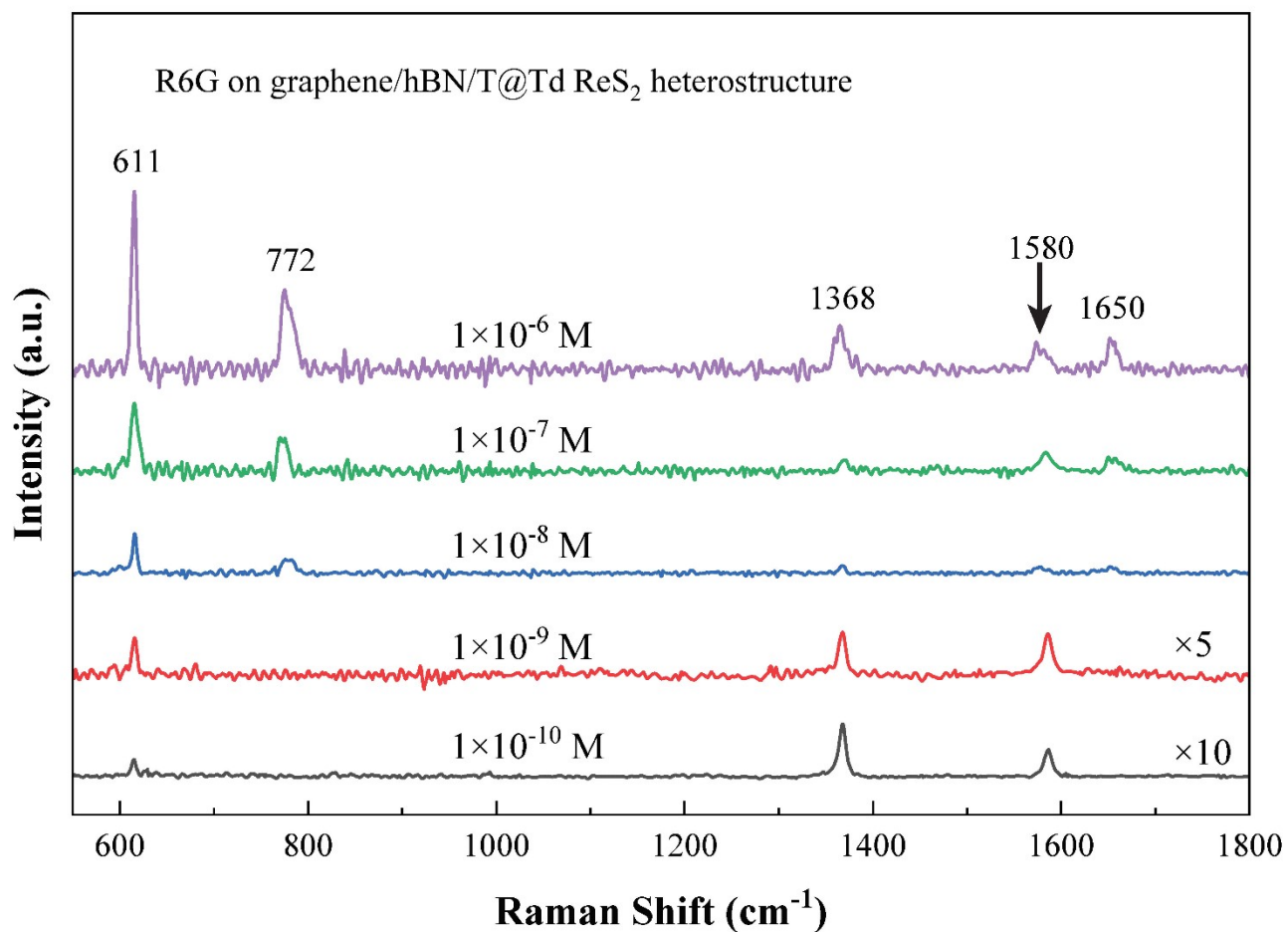


Fig. S9 SERS spectra of R6G molecules adsorbed on T@Td-ReS₂/hBN/G with LOD down to 1×10^{-10} M, where the peak located at ~ 1368 cm⁻¹ and ~ 1580 cm⁻¹ are attributed to hBN and graphene, respectively.

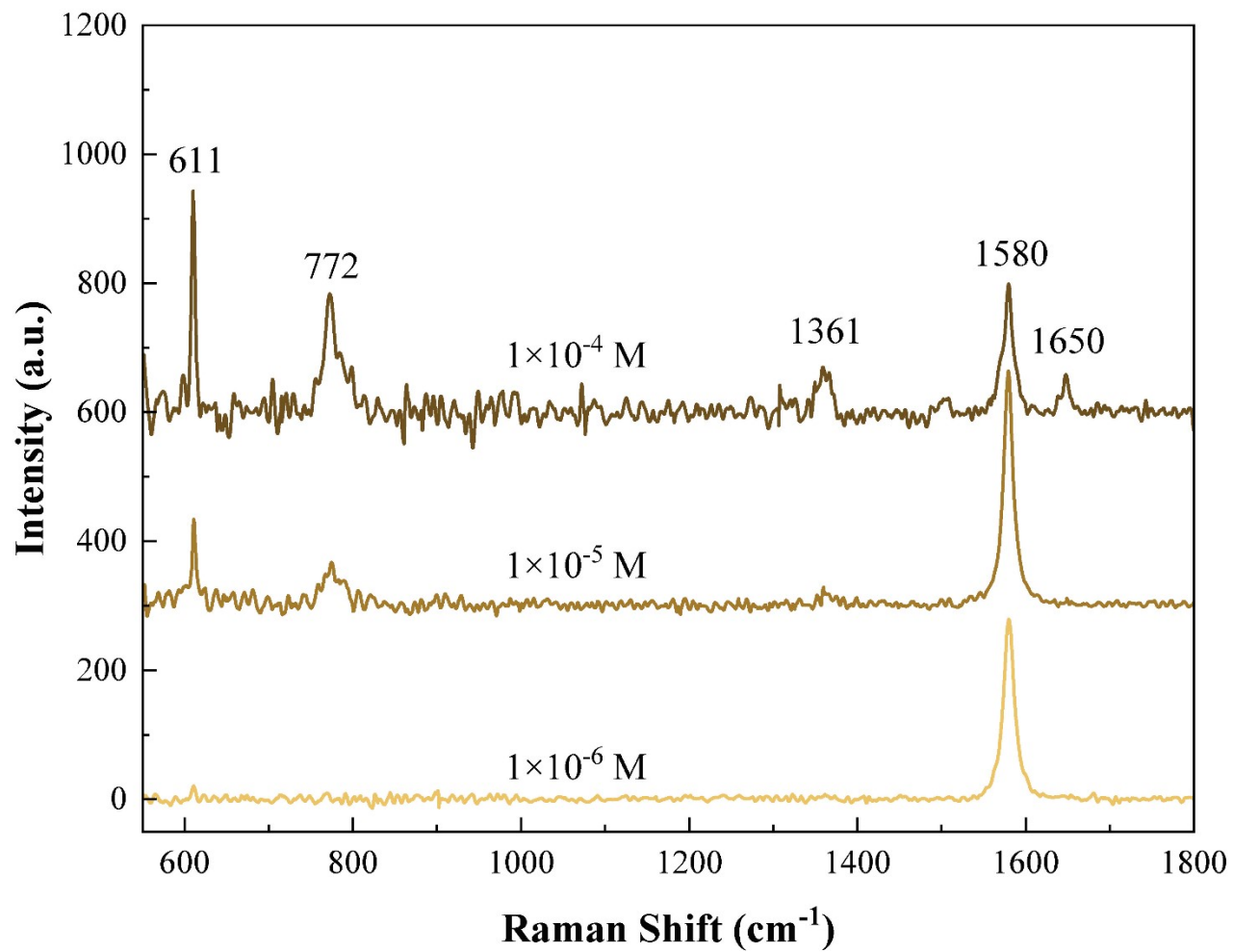


Fig. S10 SERS spectra of R6G on G/T@Td-ReS₂ excited by 633 nm laser with LOD of 1 × 10⁻⁶ M.