Supporting Information

Significantly Increased Raman Enhancement Enabled by Hot-Electron-Injection-Induced Synergistic Resonances on Anisotropic ReS₂ Films

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Supplementary Methods

Calculation of the enhancement factor. The EF is calculated according to the following formula:

$$EF = \frac{I_{SERS} / N_{SERS}}{I_{norm} / N_{norm}}$$
$$N_{SERS} = \frac{CVN_AA_{Raman}}{A_{Sub}}$$
$$N_{norm} = \frac{\rho h A_{Raman} N_A}{M}$$

 I_{SERS} and I_{norm} are the intensities of the selected Raman peak in SERS spectra and normal Raman spectra, respectively. N_{SERS} and N_{norm} are the number of molecules in the testing area of SERS and normal Raman measurements, respectively. The data of bulk RhB is used as normal Raman spectrum reference. For the SERS measurement, the intensity is taken the average of 10 random spots, and the number of molecules is estimated by the second equation on the assumption that the molecules are distributed uniformly on the substrate. *C* is the molar concentration of RhB solution and *V* is the volume of the droplet (10 µL). N_A is Avogadro constant. A_{Raman} is the laser spot area. A_{Sub} is the effective area of the substrate, which is approximately $\pi/4 \ cm^2$. The confocal depth h of the laser beam is ~ 21 µm. The molecular weight *M* of RhB is 479 g mol⁻¹ and density ρ of bulk RhB is 0.79 g cm⁻³.

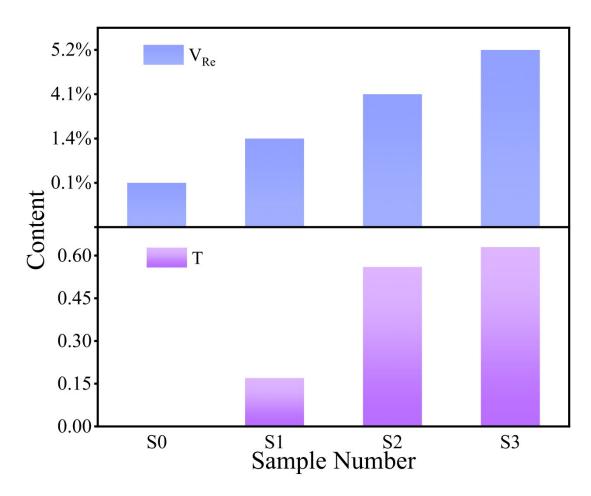


Fig. S1. The concentration of Re vacancy and T phase R6G acquired from S0 to S3.

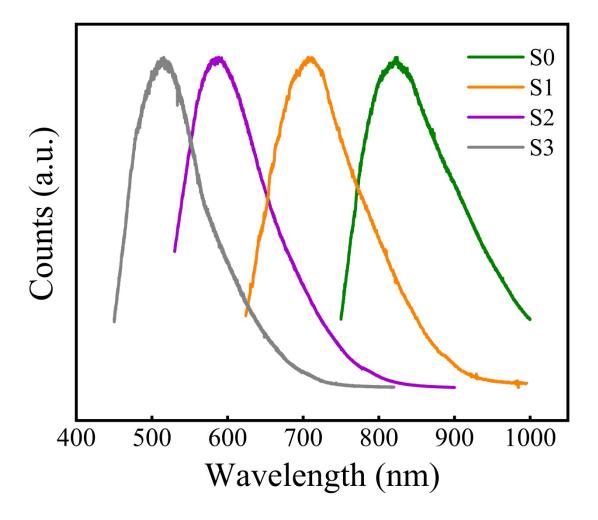


Fig. S2. PL spectra obtained from S0 to S3.

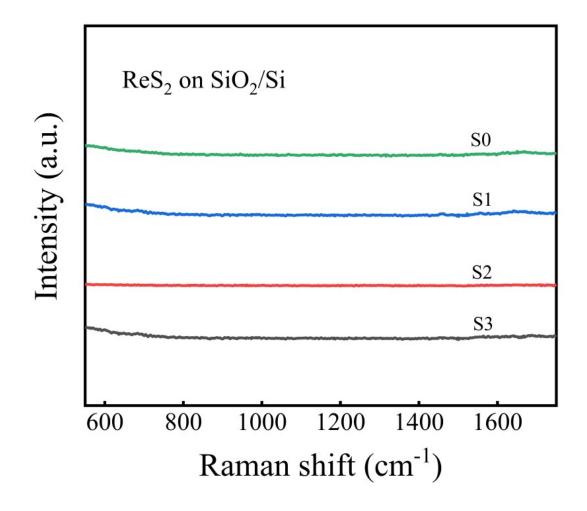


Fig. S3. The Raman spectra of bare ReS_2 of different phases in the range of 550-1750 cm⁻¹

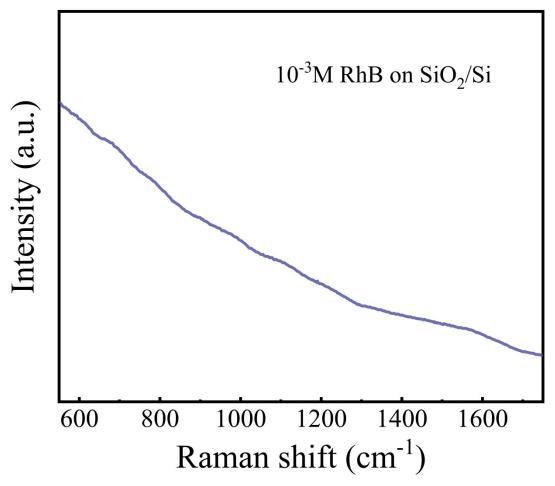


Fig. S4. Raman spectrum of RhB molecules on the SiO_2/Si substrate.

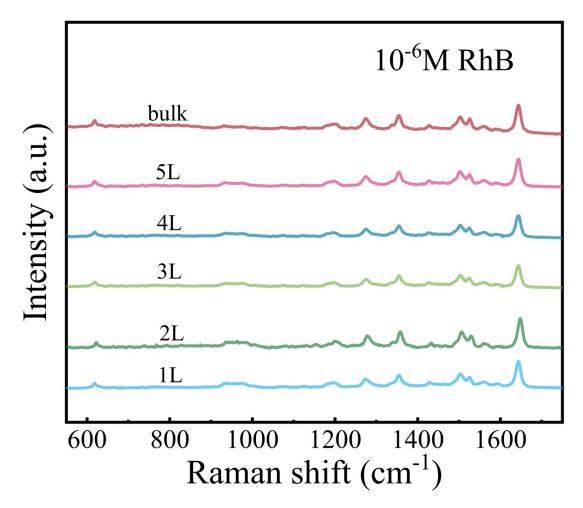


Fig. S5. Raman spectra of RhB molecules (10⁻⁶ M) on S2 varying from monolayer to

thick layers.

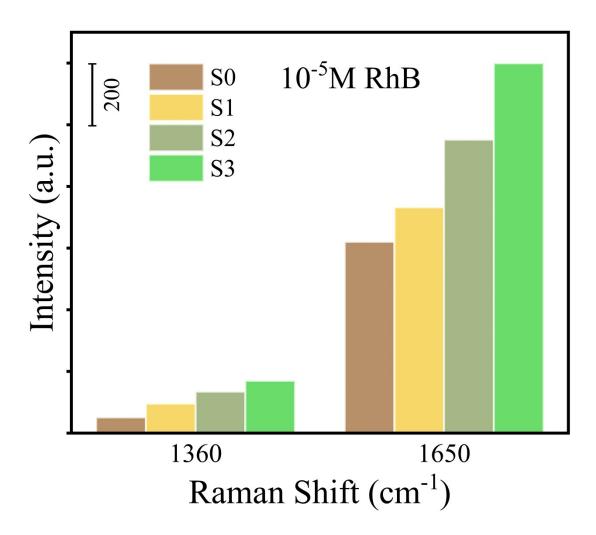


Fig. S6. SERS intensity columns derived from the enhanced Raman spectra of 10^{-5} M

RhB obtained from S0 to S3 under the 488 nm laser excitation.

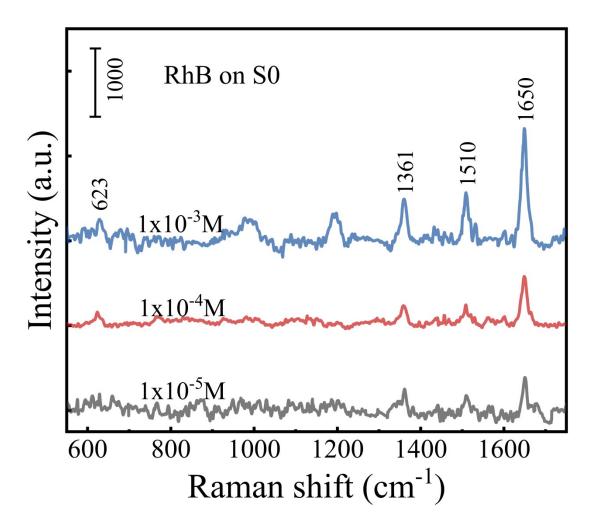


Fig. S7. SERS spectra of RhB acquired under different concentrations from 10^{-3} to

10⁻⁵ M on S0.

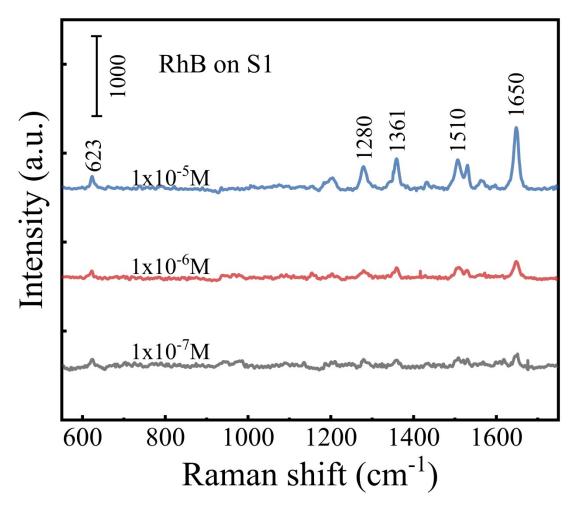


Fig. S8. SERS spectra of RhB acquired under different concentrations from 10^{-5} to

 10^{-7} M on S1.

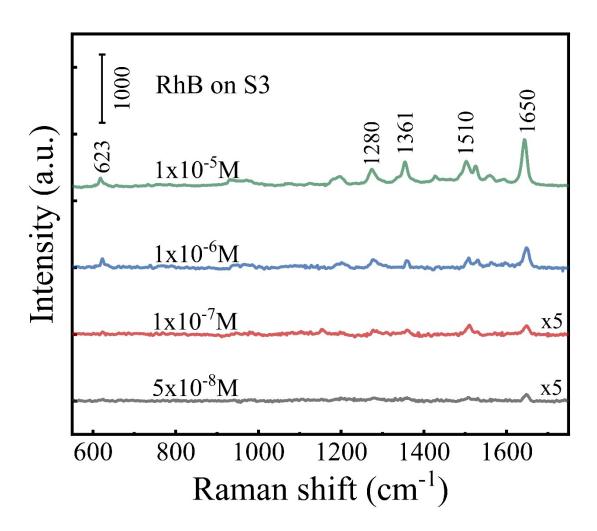


Fig. S9. SERS spectra of RhB acquired under different concentrations from 10^{-5} to

10⁻⁸ M on S3.

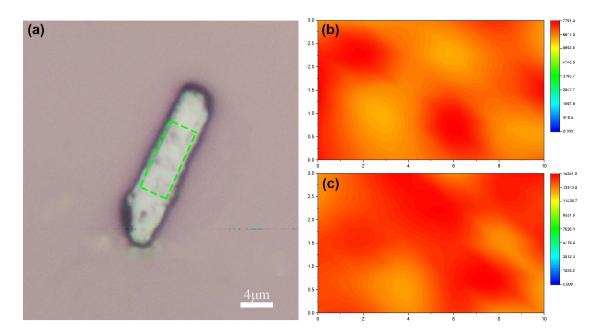


Fig. S10. (a) Optical microscopy image of T@Td ReS₂ (S2). The mapping area is marked in blue. (b, c) SERS maps from the marked area by analyzing the integrated intensity at 1360 cm⁻¹ and 1650 cm⁻¹.

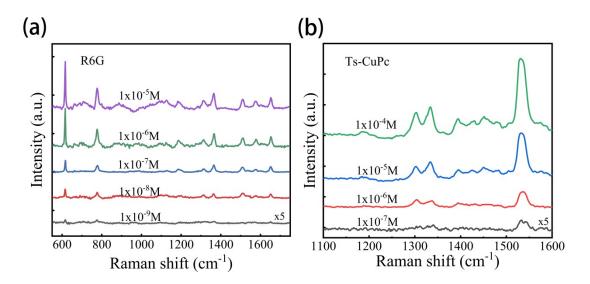


Fig.S11. (a, b) SERS spectra of R6G and CuPc molecules acquired under different concentrations from 10^{-5} to 10^{-9} M and 10^{-4} to 10^{-7} M on S2.

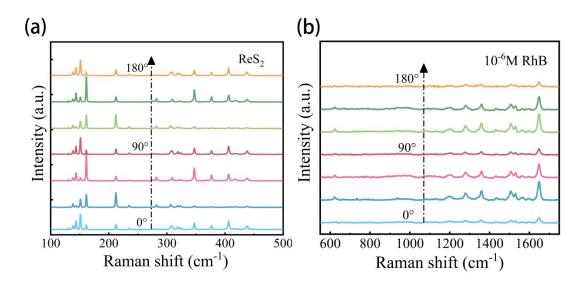


Fig. S12. (a, b) Polarized Raman spectra of ReS_2 (left) and R6G molecules on S2

(right).

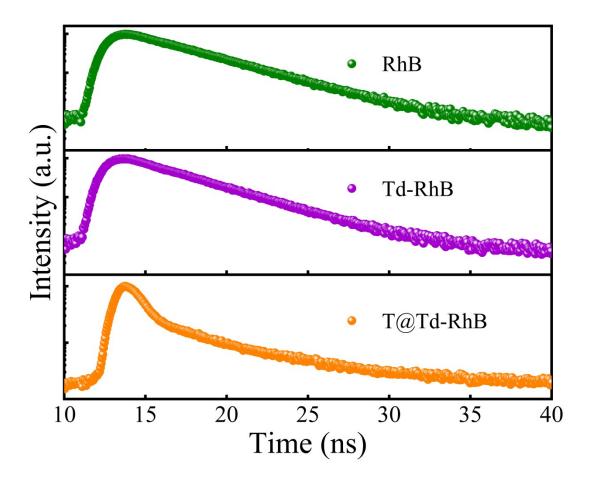


Fig. S14. Time-resolved photoluminescence spectra of pristine RhB solution, and RhB solutions mixed with Td-ReS₂ and T@Td-ReS₂ respectively.