

**Supporting Information**

**Thermal Conductance between Water and nm-thick WS<sub>2</sub>: Extremely Localized Probing  
Using Nanosecond Energy Transport State-Resolved Raman**

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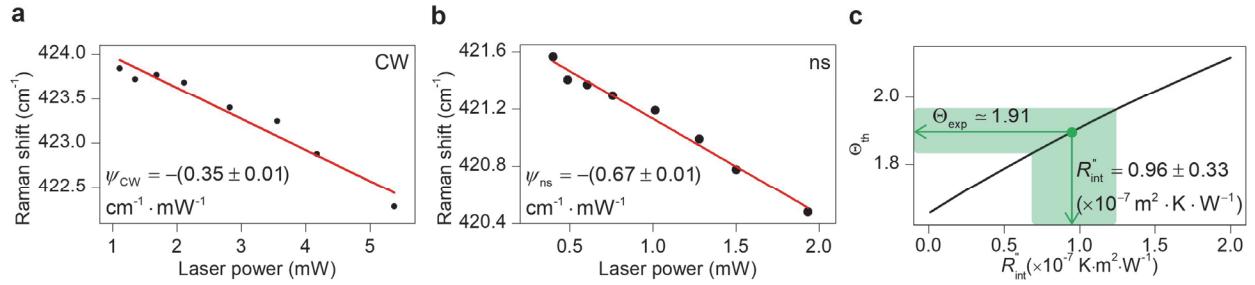
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## S1. Interfacial thermal conductance ( $G_{\text{int}}$ ) characterization of water-WS<sub>2</sub> nm-film using A<sub>1g</sub>

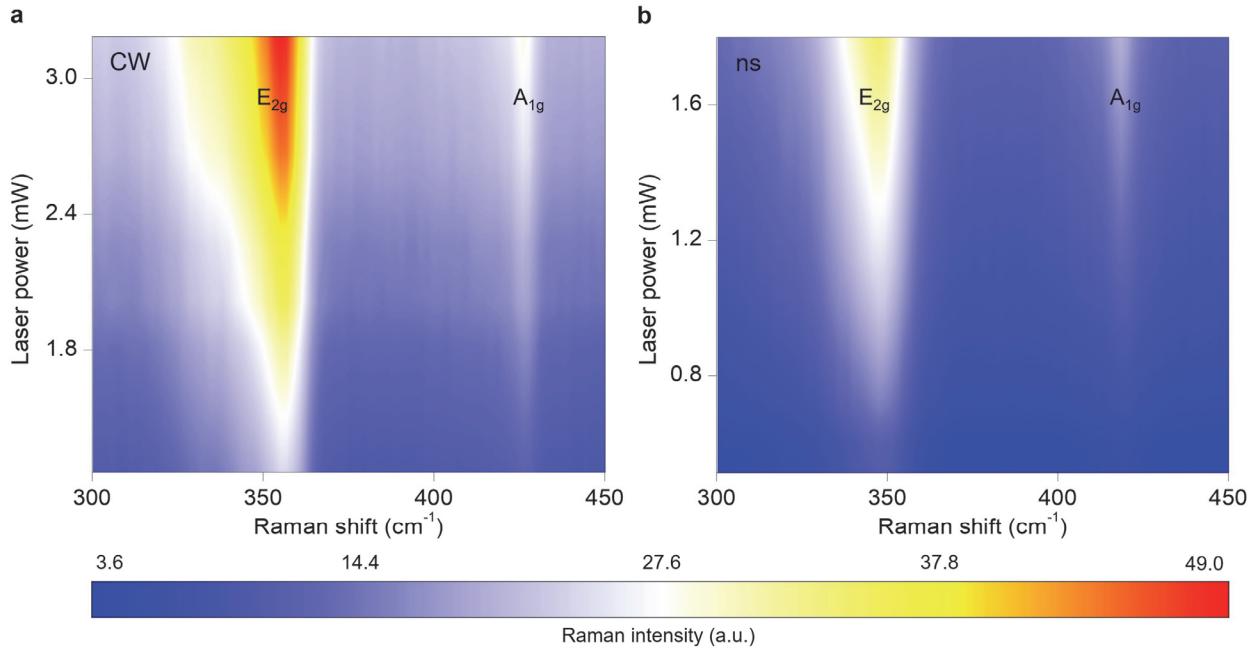
### Raman mode

Another suspended WS<sub>2</sub> film, called Sample 4, is prepared over the 10  $\mu\text{m}$  circular hole, and is used to find  $R''_{\text{int}}$  and  $G_{\text{int}}$  by studying the A<sub>1g</sub> peak of WS<sub>2</sub> sample. Similar sample characterization process is conducted for this sample, and its thickness ( $t$ ) and roughness ( $R_q$ ) is measured as 65 nm and 7.09 nm, respectively. Based on this, roughness to thickness ratio ( $[R_q/t] \times 100$ ) is  $\sim 11$  percent. Also, CW and ns laser spot radii are measured as 2.16  $\mu\text{m}$  and 1.59  $\mu\text{m}$ , respectively. These two values are used in our numerical calculation. Laser power range under CW heating state is 1.11-5.37 mW, and is 0.40-1.93 mW for ns case. Using similar Lorentzian peak fitting, Raman shift of A<sub>1g</sub> peak for this sample at various laser powers is determined. This is shown in Figure S1(a) & (b) for CW and ns cases, respectively. Using these two figures, RSC values of CW ( $\psi_{\text{CW}}$ ) and ns ( $\psi_{\text{ns}}$ ) modes are found as:  $-(0.35 \pm 0.01) \text{ cm}^{-1}\cdot\text{mW}^{-1}$  and  $-(0.67 \pm 0.01) \text{ cm}^{-1}\cdot\text{mW}^{-1}$ , respectively. Again, experimental normalized RSC ( $\Theta_{\text{exp}}$ ) is calculated as:  $\Theta_{\text{exp}} = \psi_{\text{ns}}/\psi_{\text{CW}}$ , and is equal to  $1.91 \pm 0.07$ . Similar 3D numerical calculation is conducted using this 65 nm sample, and considering the abovementioned laser spot radii, and  $R''_{\text{int}}$  is determined, as shown in Figure S1(c). Finally,  $G_{\text{int}}$  is determined to be  $10.4 \pm 2.65 \text{ MW}\cdot\text{m}^{-2}\cdot\text{K}^{-1}$ . This result is in very good agreement with the results that are reported in Table 4 of the paper, and shows that our nET-Raman technique is capable of measuring  $G_{\text{int}}$  using both E<sub>2g</sub> and A<sub>1g</sub> Raman modes.

Additionally, Figure S2 shows the 2D Raman intensity contour of Sample 3 under CW and ns states. It can be seen from both contours that both Raman peaks are red-shifted with increased laser power. Also, these contours indicate that Raman intensity of E<sub>2g</sub> peak is normally higher than that of A<sub>1g</sub> peak, which makes fitting E<sub>2g</sub> peak more reliable and accurate.



**Figure S1.** Raman shift power coefficient ( $\psi$ ) corresponding to  $A_{1g}$  peak of WS<sub>2</sub> nm-film under (a) CW and (b) ns laser of Sample 4. (c) Measured  $R''_{\text{int}}$  and its uncertainty of Sample 4.



**Figure S2.** 2D contour of Raman intensity against laser power and Raman shift for (a) CW and (b) ns heating states. These contours show the Raman shift ( $\omega$ ) and Raman intensity ( $I$ ) variation of E<sub>2g</sub> and A<sub>1g</sub> modes of WS<sub>2</sub>. Generally,  $I$  is higher at higher laser powers ( $P$ ), and  $\omega$  redshifts with increased  $P$ . Note that these 2D contours are related to Sample 3.