

Interactions of Terahertz Photons with Phonons of Two-dimensional van der Waals MoS₂/WSe₂/MoS₂ Heterostructures and Thermal Responses

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Section 1: The wet method corrosion prepared the large area MoS₂/WSe₂/MoS₂ heterojunction

Chemicals used: 4% PMMA anisole solution, 2 mol/L KOH solution, deionized water, acetone (or anisole).

Transfer method:

1. Place large-area MoS₂ grown on the SiO₂/Si substrate on a heating stage, and drop 4-5 drops of PMMA anisole solution using a dropper.
2. Turn on the heating stage to 100°C to solidify the PMMA into a film.
3. Immerse the solidified SiO₂/Si substrate with PMMA/MoS₂ in a 2 mol/L potassium hydroxide (KOH) solution.

4. After soaking for about 2 hours, the PMMA film will naturally separate from the substrate.
5. Use tweezers to pick up the separated PMMA film and rinse it 3-5 times in deionized water to remove any residual KOH.
6. Use a blank high-resistivity silicon substrate to lift the PMMA film from below, allowing the large-area MoS₂ to adhere to the new substrate.
7. Place the lifted substrate vertically to allow residual water to flow to one corner, and let it air dry naturally.
8. After air drying, heat the substrate with MoS₂/PMMA on an 80 °C hot plate to improve the bonding between MoS₂ and the new substrate, preventing the sample from falling off during the next steps.
9. Immerse the prepared MoS₂/PMMA/high-resistivity silicon substrate in acetone to etch away the PMMA. This process typically requires changing the acetone 3-5 times, with each soak lasting about 30 minutes.
10. After removing the PMMA, dry and package the sample for storage.

To transfer another layer of large-area WSe₂ and the top layer of large-area MoS₂, simply repeat the above wet transfer process two more times.

Section 2: the SNR for the relatively weak peaks in each Raman spectrum

Table S1. the SNR calculations of MoS₂ and WSe₂ Raman peaks

MoS ₂				WSe ₂			
Raman shift (cm ⁻¹)	I _{peak} Intensity	I _{baseline} Intensity	SNR	Raman shift (cm ⁻¹)	I _{peak} Intensity	I _{baseline} Intensity	SNR
34.83	549.59	500.824	3.93	29.03	618.74	562.56	3.71
41.66	465.733	423.156	3.43	46.76	330.266	280.762	3.27
48.47	453.3	406.86	3.74	61.76	295.98	247.68	3.19
66.17	340.3	293.094	3.80	64.45	297.927	242.52	3.66
82.45	269.13	211.949	4.61	75.32	338.866	261.077	5.14
90.59	252.81	199.287	4.31	97.01	531.906	428.105	6.87

Table S2. the SNR calculation of MoS₂/WSe₂ and MoS₂/WSe₂/MoS₂ heterostructures Raman peaks

MoS ₂ /WSe ₂				MoS ₂ /WSe ₂ /MoS ₂			
Raman shift (cm ⁻¹)	I _{peak} Intensity	I _{baseline} Intensity	SNR	Raman shift (cm ⁻¹)	I _{peak} Intensity	I _{baseline} Intensity	SNR
35.85	441.562	379.03	4.94	35.85	326.133	271.955	4.28
41.3	374.036	323.63	3.99	41.3	265.859	219.524	3.66
49.48	302.852	258.279	3.52	52.21	231.351	191.384	3.16
56.29	306.795	237.089	5.51	69.89	184.036	138.387	3.61
64.46	257.467	213.376	3.49	73.97	180.452	140.612	3.15
68.53	241.044	197.871	3.41	86.18	156.764	116.414	3.19
72.61	250.38	192.12	4.61	97.02	173.608	132.953	3.22
92.95	251.04	207.35	3.45				

Section 3: The list of specific results of phonon spectrum calculation

Table S3. The corresponding frequency values of monolayer and few-layer MoS₂ materials with different thicknesses at the Γ point.

MoS ₂	point 1	point 2	point3	point 4	point 5	point 6	point 7	point 8	point 9
1L	0	0	0	0	0	0	8.68	8.68	8.68
5L	0	0	0	0.42	0.42	0.71	8.24	8.24	8.25

MoS ₂	point 10	point 11	point 12	point 13	point 14	point 15	point 16	point 17	point 18
1L	8.68	11.75	11.751	11.75	11.78	12.34	12.34	14.3	14.3
5L	8.25	11.14	11.14	11.14	11.21	11.85	11.88	13.62	13.66

Table S4. The corresponding frequency values of monolayer and few-layer WSe₂ materials at the Γ point.

WSe ₂	point 1	point 2	point3	point 4	point 5	point 6	point 7	point 8	point 9
1L	0	0	0	0	0.04	0.04	5.14	5.14	5.14
5L	0	0	0	0.38	0.38	0.61	5.11	5.17	5.18

WSe ₂	point 10	point 11	point 12	point 13	point 14	point 15	point 16	point 17	point 18
1L	5.14	7.32	7.32	7.32	7.35	7.35	7.35	9.13	9.14
5L	5.18	7.32	7.32	7.32	7.44	7.44	7.47	9.11	9.14

Section 4: The Thermodynamic simulation and calculation model parameters and part of the calculation results

Table S5. The physical parameters of different materials for simulation modeling

Materials	geometrical shape	Area cm ²	Thickness nm	Thermal conductivity W/(m·K)	Density g/cm ³	Specific heat capacity J/(g · K)	Thermal convection coefficient (W/(m ² · K))
Top MoS ₂	quadrangle	1.49×10^{-5}	4.08	100	5.06	0.397	0.55
Middle WSe ₂	quadrangle	2.47×10^{-5}	1.86	40	9.2	4	0.33
bottom MoS ₂	pentagon	4.597×10^{-5}	3.4	100	5.06	0.397	0.18
MoS ₂ /WSe ₂	quadrangle	0.687×10^{-5}	5.26	65.793	-	2.451	1.19
MoS ₂ /WSe ₂ /MoS ₂	triangle	0.284×10^{-5}	9.34	77.396	-	1.754	2.86
SiO ₂	square	1cm×1cm	300	1.4	2.18	0.742	5-15
Si substrate	square	1cm×1cm	500 um	130	2.329	1.418	10-25

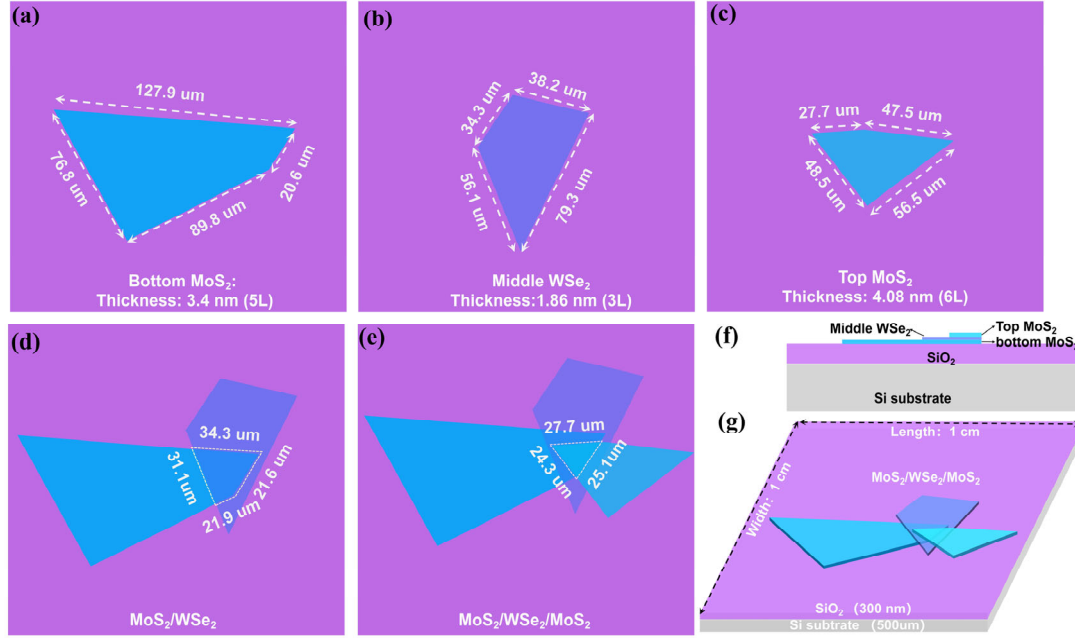


Figure S1. The geometric morphology, size, and thickness parameters of the materials. **(a)** Bottom layer MoS₂. **(b)** Middle layer WSe₂. **(c)** Top layer MoS₂. **(d)** MoS₂/WSe₂. **(e)** MoS₂/WSe₂/MoS₂. **(f)** The side view of the MoS₂/WSe₂/MoS₂ model on a large-area silicon substrate. **(g)** Top view of the model.

Figures s1a-c show the shape, size, and thickness of a single two-dimensional material. **Figure s1d** shows the shape and size of the transferred MoS₂/WSe₂ heterojunction and its overlapping region. **Figure s1e** shows the shape and size of the transferred MoS₂/WSe₂/MoS₂ heterojunction and its overlapping region. **Figure s1f and Figure s1g** show the side view and top view of the MoS₂/WSe₂/MoS₂ sample on a large area high resistance SiO₂/Si substrate (resistivity 20000 $\Omega \cdot \text{cm}$).

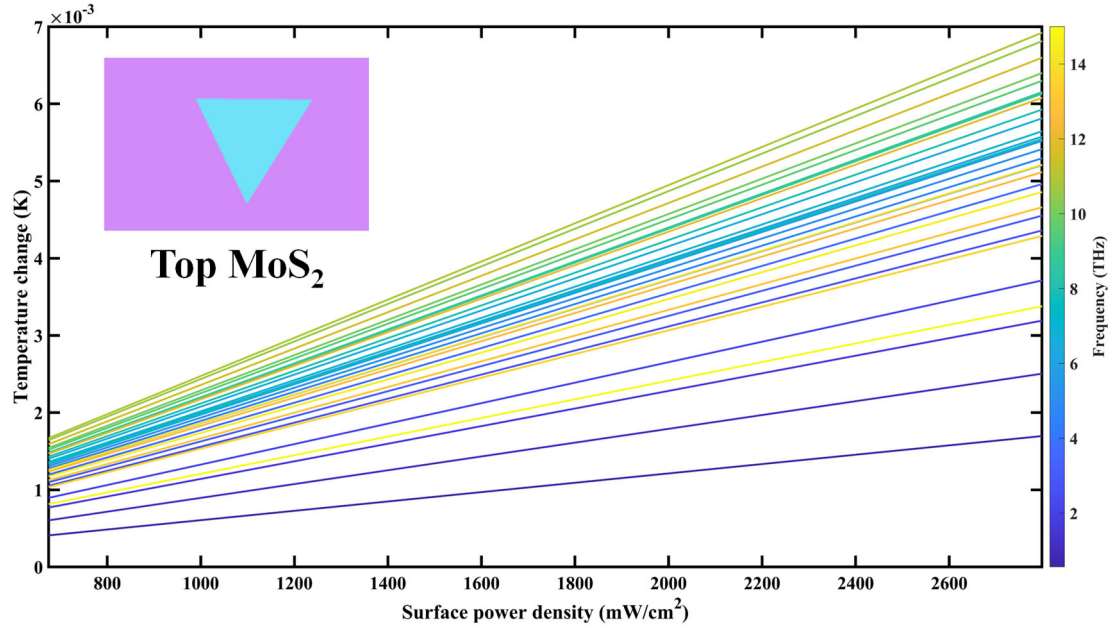


Figure S2 The temperature change ΔT of the top MoS₂ after irradiation by terahertz waves of 0.5–15 THz with THz power density variation.

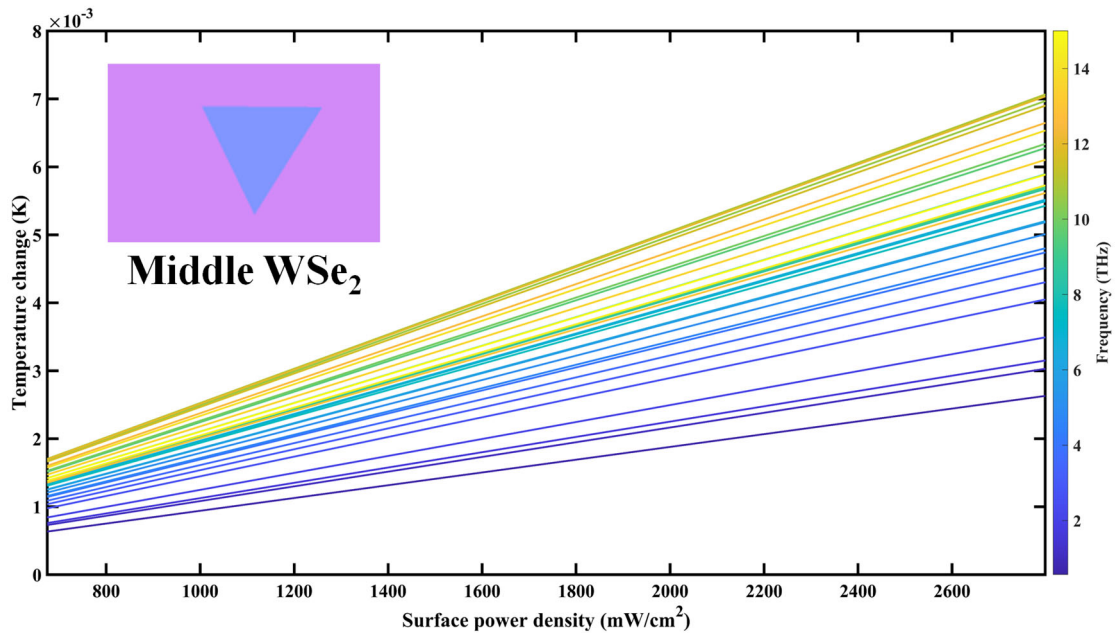


Figure S3. The temperature increases ΔT of the middle WSe₂ after irradiation by terahertz waves of 0.5–15 THz with THz power density variation.

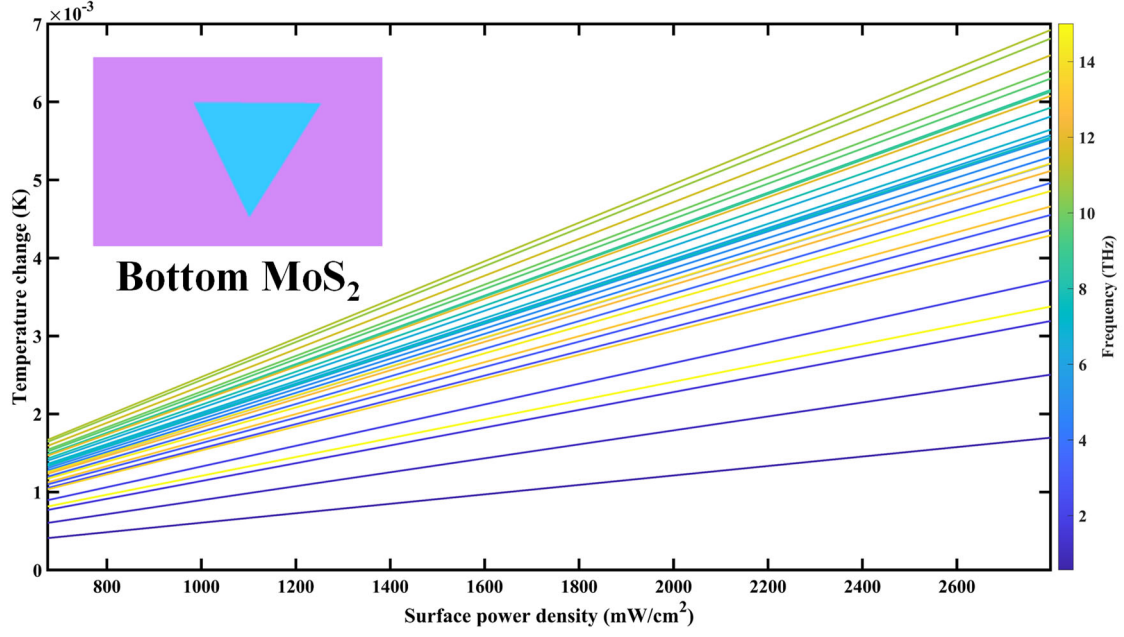


Figure S4. The temperature change ΔT of the bottom MoS_2 after irradiation by terahertz waves of 0.5–15 THz with THz power density variation.

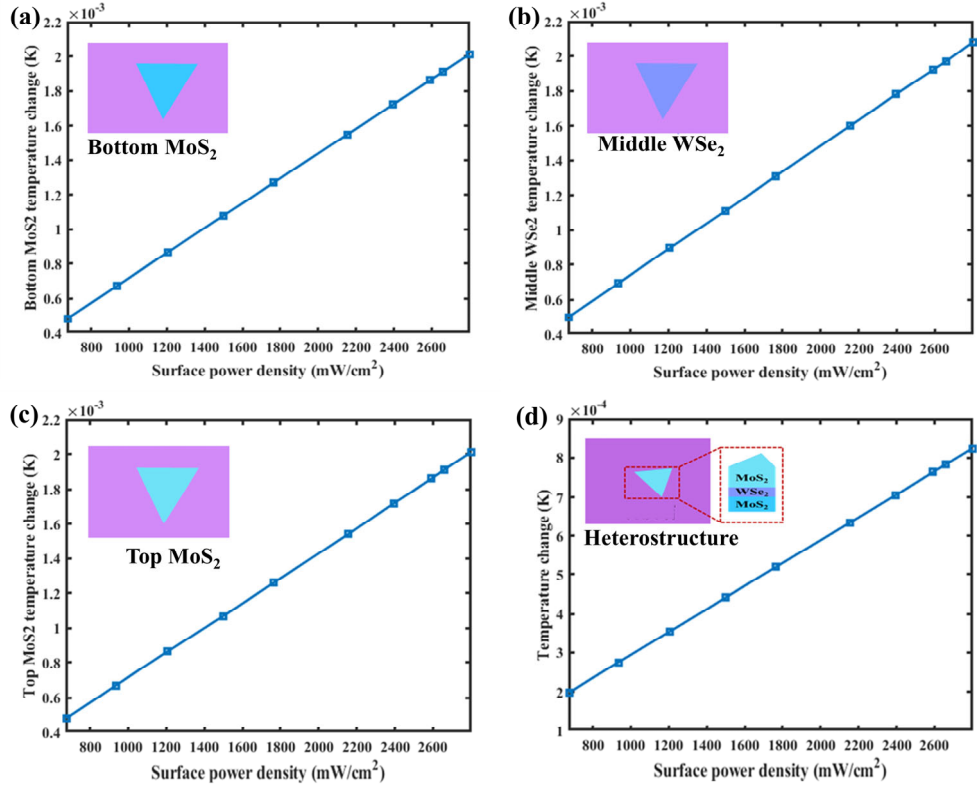


Figure S5. The simulation of temperature changes ΔT in each layer of the $\text{MoS}_2/\text{WSe}_2/\text{MoS}_2$ heterojunction after irradiation by a single terahertz pulse (pulse duration $\Delta t = 120$ s). **(a)** Bottom MoS_2 , **(b)** Middle WSe_2 , **(c)** Top MoS_2 , **(d)** $\text{MoS}_2/\text{WSe}_2/\text{MoS}_2$ heterostructure.