Supporting Information

Improving the optical quality of $MoSe_2$ and WS_2 monolayers with complete *h*-BN encapsulation by high temperature annealing

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Section S1. Exfoliation method

First, a Si wafer with a 285 nm SiO_2 layer on top was diced into square shapes with side length ~1 cm. Then, in order to remove the surface contamination and improve the 2-dimensional material adhesion on the surface, oxygen plasma treatment was applied to these Si chips. The surface contamination can also be removed by sonication for 10 min in acetone and then sprayed with isopropyl alcohol (IPA) and dried with a nitrogen gun. In exfoliation, bulk crystals of the 2-dimensional material were placed on Magic Scotch tape. Then, the tape was opened and folded several times until the crystal layers were on the tape and it looked nearly transparent. The tape was then placed onto the pre-cleaned SiO_2/Si chips gently. In order to minimize the tape residue on the chips, the chip and tape needed to be annealed at 100°C for 3 min. After the chips and tape cooled down to room temperature, the tape was peeled off gently with a tweezer. Monolayers were searched for by using an optical microscope. The layers on SiO_2/Si chips with different thickness will have different optical contrast because thin film interference produced by 285 nm SiO_2 layers on Si wafers. So, the thicknesses of the layers were estimated by using an optical microscope.

Section S2. Schematic view of the Linkham THMS 600

Figure S1 is a schematic of the TMD sample placed in a Linkam cell in which the temperature and gas environment are controlled.

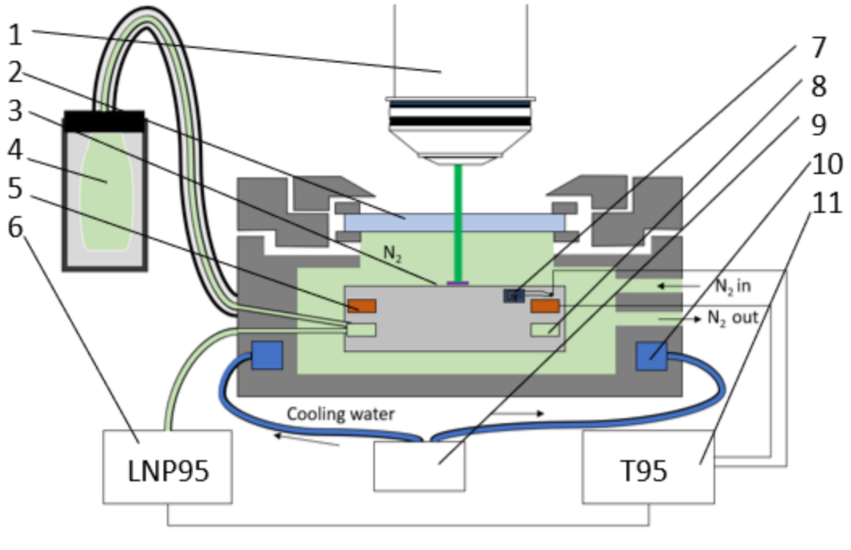

Figure S1. Schematic of a van der Waals-stacked sample placed in a Linkam cell – 1: Olympus 50× long working distance objective, 2: viewing windows glass (22-mm-diameter, 0.5-mm-thick), 3: van der Waals-stacked sample, 4: liquid nitrogen dewar, 5: heating element wire, 6: LNP95 Liquid nitrogen cooling pumping system, 7: platinum temperature sensor, 8: stainless steel cooling tube, 9: water circulator pump, 10: water cooling tube, and 11: T95 system controller.

Section S3. Experimental set up

Figure S2 is a schematic depiction of the optical apparatus where PL measurements were performed. $\lambda = 514.5$ nm cw light is produced by an argon ion laser that is focused to a ~1 μm spot size. The laser power was reduced by natural density (ND) filters to avoid high power induced damage on the sample. Plasma lines were cut off with a 532 nm low pass filter and the 514.5 nm laser light was blocked with a high pass filter. The sample was placed in a THMS600 Linkam cell where temperature and the gas environment were controlled. The PL signal was collected by a cooled spectroscopic-format CCD detector (Princeton Instruments Spec-10:400B) with a 1340 × 400 imaging array.

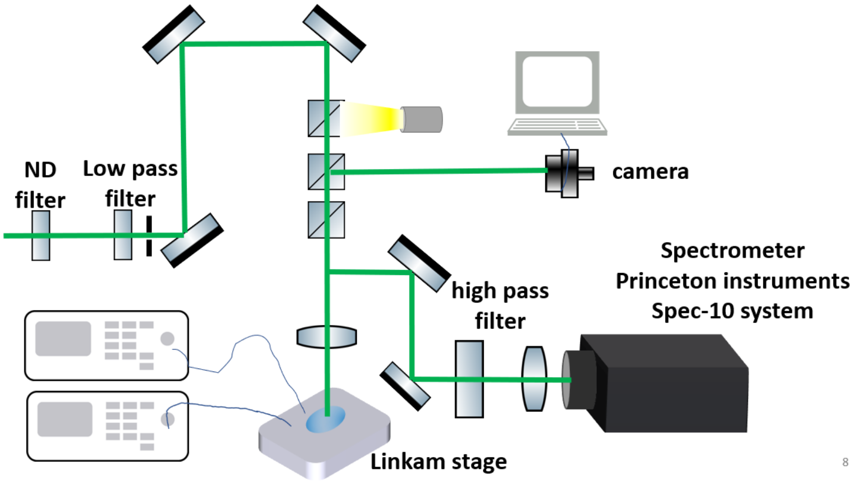

Figure S2. Optical set up for the photoluminescence measurement.

Section S4. Temperature dependence of photoluminescence

The PL spectra of the same exfoliated MoSe$_2$ ML sample encapsulated by top and bottom *h*-BN, as used in Figure 1 in the main text, were recorded as a function of temperature in the 77

K to 873 K to 77 K cycle. Figure 1d in the main text gives the PL spectra only at selected *T*. Figure S3a shows how the peak PL positions, widths and peak intensities change as a function of temperature, for PL measurements at each *T*. These PL data are consistent with the visual observation result that the $MoSe_2$ sample is well protected by the complete *h*-BN encapsulation up to 873 K.

The exciton emission energies after heating and during cooling generally track each other, but there is a red shift after the heating/cooling cycle. Figure S3b shows that the peak widths at corresponding temperatures generally are the same during heating and cooling, but they are a bit smaller during cooling. At 77 K, the exciton PL peak width is smaller than that before the heating/cooling cycle, indicating a reduction of heterogeneity and external disorder.

Figure S3c presents the peak intensities of the exciton and trion peaks as a function of temperature. The trends at corresponding temperatures during heating and cooling are similar, but the intensities after cooling are much larger, starting above 700 K, as is detailed in the main text. This is due to changes that occur during heating around 900 K, that result in the carrier concentration changes and interfacial improvements noted in the main text. During the heating cycle starting at 77 K, the trion peak appeared up to 125 K, and during the cooling step it reappeared only when the temperature had decreased to 77 K. Additionally, the ratio of the exciton and trion peak PL intensities can be an important indicator of the TMD quality. In the sandwich exfoliated $MoSe_2$ ML sample, this ratio decreases after the heating phase, indicating the possible reduction of doping and charge inhomogeneity in monolayer $MoSe_2$ (Figure S3d).

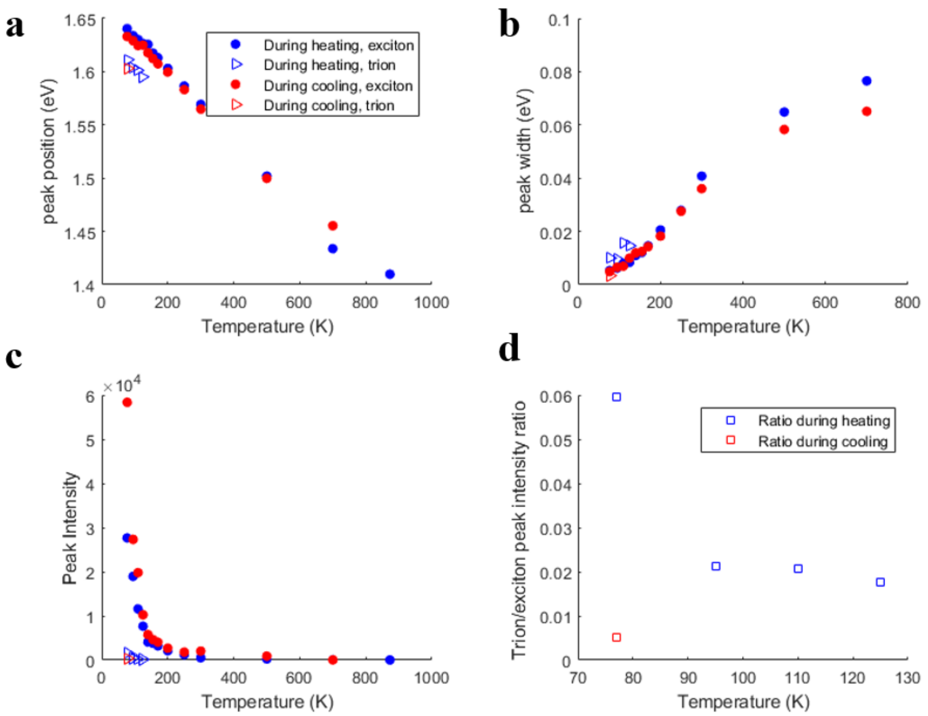

Figure S3. Temperature dependence of the PL of $MoSe_2$ ML sandwiched by *h*-BN**.** Shown are the exciton and trion peak (a) positions, (b) widths and (c) intensities vs. *T* during the 77 K to 873K to 77 K heating/cooling cycle. The plotted symbols indicated in (a), are also used in (b) and (c). (d) shows the trion/exciton peak intensity ratio at elevated temperature during heating and cooling.

Section S5. Carrier concentration calculation

The carrier density in the $MoSe_2$ ML can be described as:

$$n = \frac{CV}{e}\,, \qquad 1$$

where *V* is the gate voltage difference and C is the bottom *h*-BN capacitance per unit area, which can be expressed as:

$$C = \frac{\epsilon_r \epsilon_0}{t}\,, \qquad 2$$

where the *t* is the thickness of the bottom *h*-BN, which is 35 nm and ϵ_r is its dielectric constant. So,

$$C = \frac{\epsilon_r \epsilon_0}{t} = \frac{3.5 \times 8.85 \times 10^{-12}\ F.m^{-1}}{35 \times 10^{-9}} = 8.85 \times 10^{-4}\ F.m^{-2} \quad 3$$

$$n = \frac{CV}{e} = \frac{8.85\times10^{-4}\ F.m^{-2}\ \times 2V}{1.6\times10^{-19}C} \approx 1.1 \times 10^{12}\ cm^{-2} \quad 4$$

This concentration change is a decrease, so the negative of this number is presented in the main paper.

Section S6. Raman analysis of encapsulated monolayer WS_2

As seen in Figure S4, there were no apparent changes in the Raman spectra of the monolayer WS_2 flakes grown by CVD covered by bottom and top *h*-BN after heating, and so no apparent changes in structure.

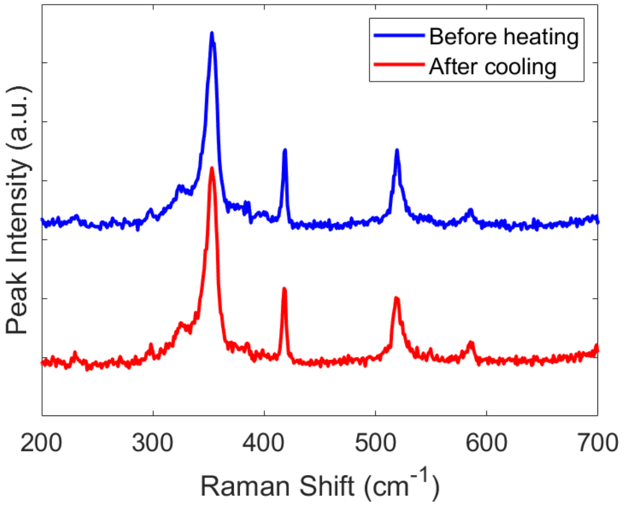

Figure S4. Raman spectra of monolayer WS_2 flakes (grown by CVD) covered by bottom and top *h*-BN on the SiO_2/Si substrate at 300 K before and after heating to 873 K in N_2.