Electronic Supplementary Material (ESI) for Nanoscale Horizons.

Electronic Supplementary Information

Type-I van der Waals Heterostructure Formed by MoS_2 and ReS_2 Monolayers

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1. Confirmation of Monolayer Thickness

In order to confirm the monolayer (ML) thickness of both the MoS_2 and ReS_2 layers, we utilize a combination of optical contrast, Raman, and photoluminescence (PL) measurements

One of the simplest methods to confirm the layer thickness of a sample has been to utilize optical contrast in different color channels of red, green, and blue. From previously reported studies [1,2], it has been established that ML transition metal dichalcogenides (TMDs) have a green channel contrast of about 12-13%. Figure S1(a) shows the green channel of an optical image of the heterostructure sample. The brightness of the image as a function of position along the yellow line was measured extracted from the image. From the brightness, the contrast was calculated as

$$C(x) = \frac{I_{sub} - I(x)}{I_{sub}},$$

where I_{sub} and $I(x)$ are the brightness of the substrate and brightness at position, *x*, on the yellow line, respectively. These contrast results are plotted in the inset of Fig. S1(a). It can be seen that both MLs have a similar contrast of about 12-13% range, consistent with previous studies [1,2].

In addition to the contrast measurements, photoluminescence (PL) spectroscopy was used to confirm that the MoS_2 sample is monolayer. It is well known that MoS_2 has a direct bandgap in monolayer that transitions to indirect in bilayer and thicker samples. Figure S1(b) shows the PL spectrum measured for the MoS_2 sample. The PL peak position and overall strength of the signal is consistent with that of ML MoS_2 thus confirming ML thickness of the MoS_2 layer. ReS_2 does not exhibit the direct to indirect transition and thus PL cannot be used to confirm its thickness.

Raman measurements were also used to further confirm monolayer thickness of both layers. MoS_2 is known to have a very noticeable shift in the E^1_{2g} and A_{1g} peaks that progresses from monolayer to bulk. From Li *et al*. [3], in ML, these peaks should be at 384.7 and 402.8 cm^{-1}, respectively, when excited with a 488-nm laser. These two peaks in bilayer MoS_2 are expected to be at 383.3 and 405.5 cm^{-1}, respectively, and become further separated with an increased number of layers. To account for any offsets in measurements, the difference between the E_{2g} and A_{1g} peaks can be taken to indicate thickness of MoS_2 samples.

In our data, we fit the two MoS_2 peaks with Lorentzian functions, shown in Fig. S1(c). The resulting peak positions for monolayer were found to be 385.05 ± 0.05 and 404.10 ± 0.04 cm^{-1}. The difference between our measured peaks, 19.05 cm^{-1}, is consistent with the expectation for ML. The measured results for bulk MoS_2 are also presented in Fig. S1(c), again matching the expected results.

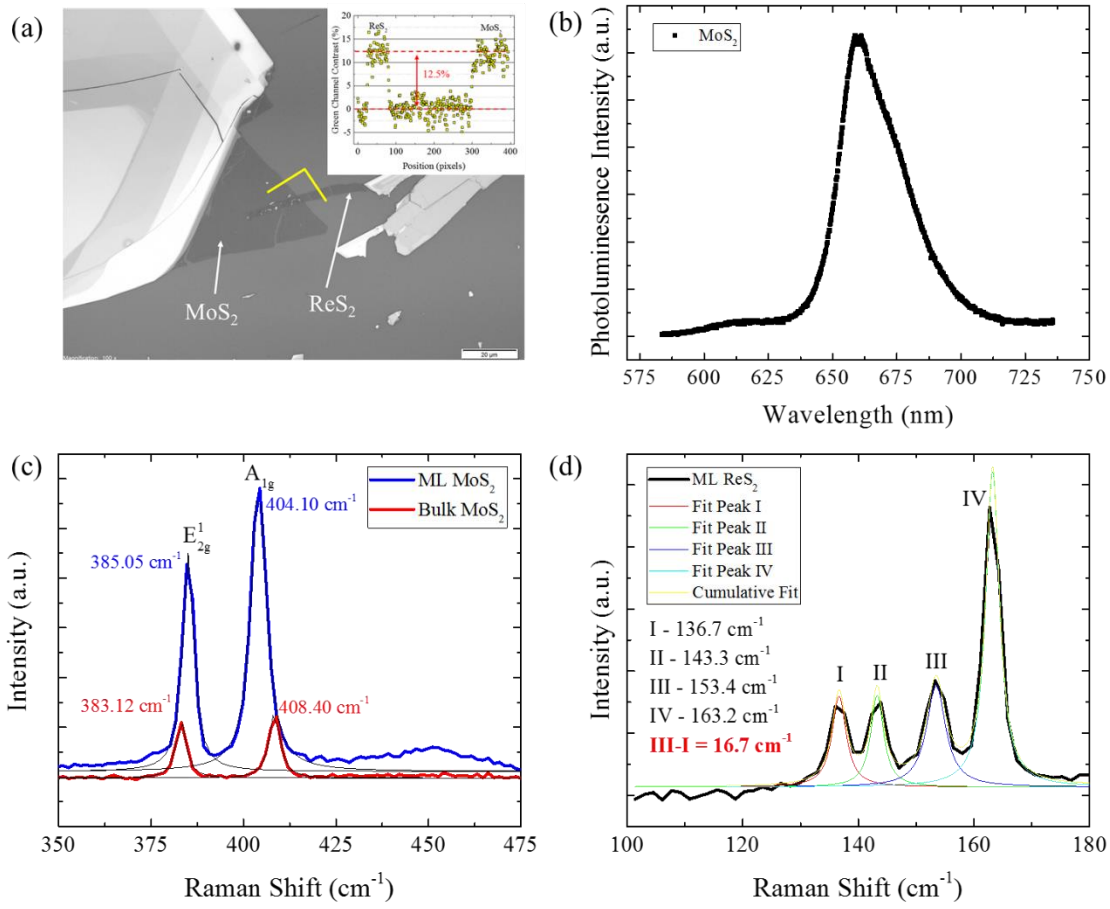

Figure S1. (a) Green channel image of the ReS_2-MoS_2 heterostructure. The inset shows the contrast of the sample along the yellow line. (b) Photoluminescence spectrum of MoS_2 sample. (c) Measured Raman spectra of ML and bulk MoS_2. (d) Measured Raman spectrum of ML ReS_2 between 100 cm^{-1} and 180 cm^{-1} (Full spectral range is shown in main text).

The Raman measurements for ReS_2 sample are presented in Fig. S1(d). As discussed in Chenet *et al* [4], four characteristic peaks in ReS_2 between 120-170 cm^{-1} can be used to identify sample thickness. These peaks are labeled I - IV in Fig. S1(d). Taking the difference between the positions of peaks I and III leads to a significant difference between monolayer and thicker ReS_2. They observed a difference of 16.8 cm^{-1} between these peaks in ML that drops to 14.9 cm^{-1} in BL and drops further in thicker samples [4]. Our result of 16.7 cm^{-1} closely matches their result for ML. Other reports on ReS_2 Raman spectra also show the presence of low frequency, shear and breathing, modes that occur in bilayer ReS_2, but are not present in monolayer [5,6].

2. Power Dependence

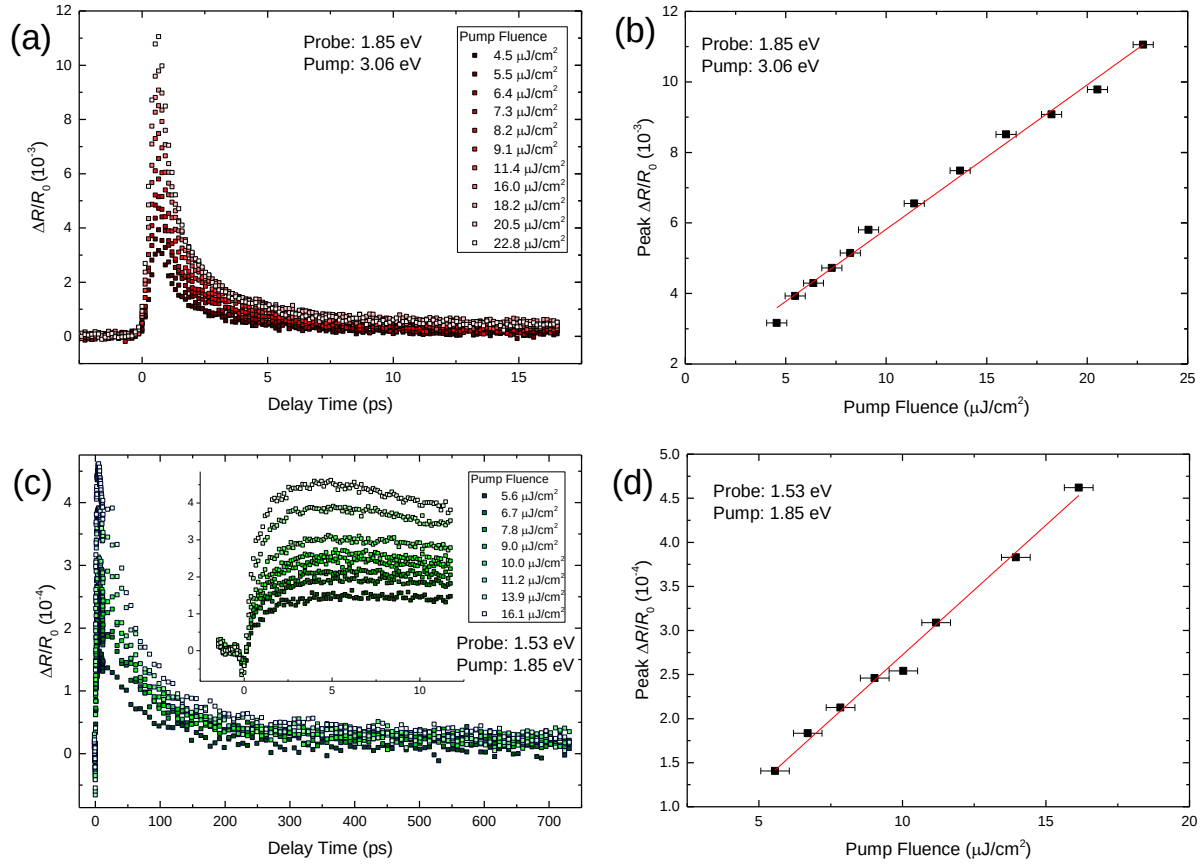

Figure S2. (a) Transient absorption measurements on the MoS_2-ReS_2 heterostructure for various peak fluences of a 3.06 eV pump, using a 1.85 eV probe (near MoS_2 A-exciton resonance). (b) Dependence of the peak $\Delta R/R_0$ signal on the pump fluence of the 3.06 eV pump. The trend line (red) is a linear fit to the data. (c) Transient absorption measurements on the heterostructure for various peak fluences of a 1.85 eV pump (near MoS_2 A-exciton resonance), probing at 1.53 eV (near ReS_2 A-exciton resonance). The inset shows the data on a shorter time scale. (d) Dependence of the peak $\Delta R/R_0$ signal on the pump fluence of the 1.85 eV pump. The trend line (red) is a linear fit to the data.

3. Computation Method:

All computations were carried out within the density functional theory (DFT) methods implemented in the Vienna *ab initio* simulation package (VASP 5.3). The generalized gradient approximation (GGA) within the form of Becke86 was adopted for exchange-correlation functional. Furthermore, the optB86b-vdW functional was used for structure optimization to account for the weak van der Waals (vdW) interactions. The interaction between the core electrons and valence electrons was modeled using the projector augmented wave (PAW) method. Energy cutoff for the plane-wave expansion was set to 500 eV. Brillouin zone sampling was performed with Monkhorst-Pack (MP) special k-point meshes. The 2D Brillouin zone integration using the Γ-center scheme was sampled with a 9 × 9 × 1 grid for transition-metal dichalcogenides. A vacuum

layer greater than 2 nm was taken so that interaction between adjacent images can be neglected. The lattice constants of ReS_2/MoS_2 heterobilayer were fixed during the structure optimization until the computed Hellmann-Feynman force on each atom was less than 0.1 eV/nm. The convergence threshold was set as 10^{-5} eV in energy for the structure optimization. For computing the band structure of the ReS_2/MoS_2 heterobilayer, the Brillouin zone of Γ-K-M-Γ was selected.

References

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