Supporting Information

Observation of positive trions in α-MoO₃/MoS₂ van der Waals heterostructures

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Synthesis of MoS₂ flakes



Fig. S1: (a) Schematic of CVD setup for the synthesis of MoS_2 flakes. (b) OM image of monolayer MoS_2 flakes.

Synthesis of α-MoO₃ crystals



Fig. S2:(a) Schematic of PVD setup for the synthesis of α -MoO₃ crystals. White rectangular bars represent the free-standing crystals in the inner quartz tube side walls. (b) OM image of free-standing crystals taken on 300 nm SiO₂/Si substrate.



Fig. S3: Excitation power-dependent Raman spectra of (**a**) α -MoO₃/MoS₂ HS and (**b**) MoS₂ with 488 nm laser source. The inset in graph (a) is the magnified image of the 350-440 cm⁻¹ region, and peaks correspond to the MoS₂ characteristics E_{2g} and A_{1g} modes. The intensity of the Raman peaks increases for HS as well as MoS₂ with an increase in excitation power.



Fig. S4: (a) 632 nm laser-excited PL spectra of MoS_2 , α -MoO₃, and α -MoO₃/MoS₂ HS. As compared to the PL spectra of MoS₂, the peaks in the HS are blue-shifted. (b) The MoS₂, α -MoO₃, and HS Raman spectra with 632 nm laser excitations. The inset shows the blue shift in the HS peak with respect to the MoS₂ peaks. Also, in the case of HS peaks, positions at 406 and 416 cm⁻¹ have opposite intensity behavior in comparison to the MoS₂ peaks.



Fig. S5: Excitation power-dependent Raman spectra of (**a**) α -MoO₃/MoS₂ HS and (**b**) MoS₂ with 632 nm laser source. Insets in the graphs are the magnified view in the range 360-490 cm⁻¹. The intensity of the Raman peaks increases with the increase in excitation laser power.



Fig. S6: Excitation power-dependent PL spectra of (**a**) α -MoO₃/MoS₂ HS and (**b**) MoS₂ using a 488 nm laser source. The intensity of the PL emission peaks increases for HS as well as MoS₂ with an increase in excitation power.



Fig. S7: Excitation power-dependent PL spectra of (a) α -MoO₃/MoS₂ HS and (b) MoS₂ using a 632 nm laser source. The intensity of the PL emission peaks increases for HS as well as MoS₂ with an increase in excitation power.



Fig. S8: (a) The power-dependent PL spectra of the HS with the 488 nm laser excitation. (b-c) The variation in integrated PL intensity with the incident laser power for A and B exciton, respectively. The integrated PL intensity of the A and B exciton is fitted by the power law (I $\propto P_{\text{Laser}}^{\alpha}$) and the obtained value of the α are ~0.96 and ~1.06 for the A and B exciton, respectively.



Fig. S9: (a) The power-dependent PL spectra of the MoS₂ with the 488 nm laser excitation. (**b-c**) The variation in integrated PL intensity with the incident laser power for A and B exciton, respectively. The integrated PL intensity of the A and B exciton is fitted by the power law (I $\propto P_{\text{Laser}}^{\alpha}$) and the calculated values of the α are ~1.18 and ~0.92 for the A and B exciton, respectively.