Supplementary Information

Layer-number-dependent Photoswitchability in 2D MoS_2-Diarylethene Hybrids

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1. AFM Imaging of MoS_2-DAE Samples on SiO_2/Si Substrates

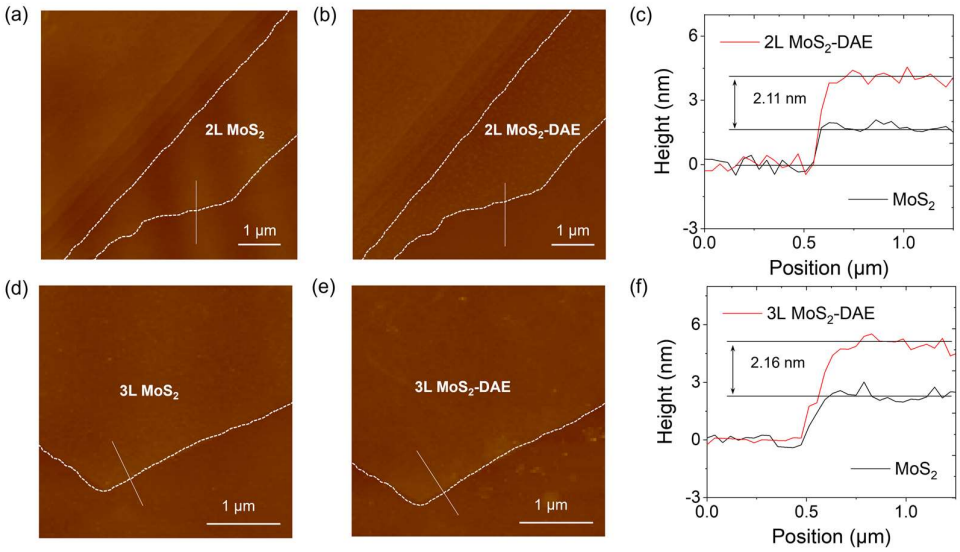

Figure S1. AFM images of (a) 2L MoS_2 flake and (b) 2L MoS_2-DAE hybrid are shown, with the corresponding height differences depicted in (c). The thickness of the 2L MoS2 flake is ~1.60 nm, while the DAE measures ~2.11 nm. Additionally, AFM images of (d) 3L MoS_2 flake and (e) 3L MoS_2-DAE hybrid are provided. The thickness of the 3L MoS_2 flake is ~2.28 nm, and the DAE measures ~2.16 nm. These samples were used for PL measurements presented in Figure 2e-f.

2. PL Measurements of Additional MoS_2-DAE Hybrid Samples

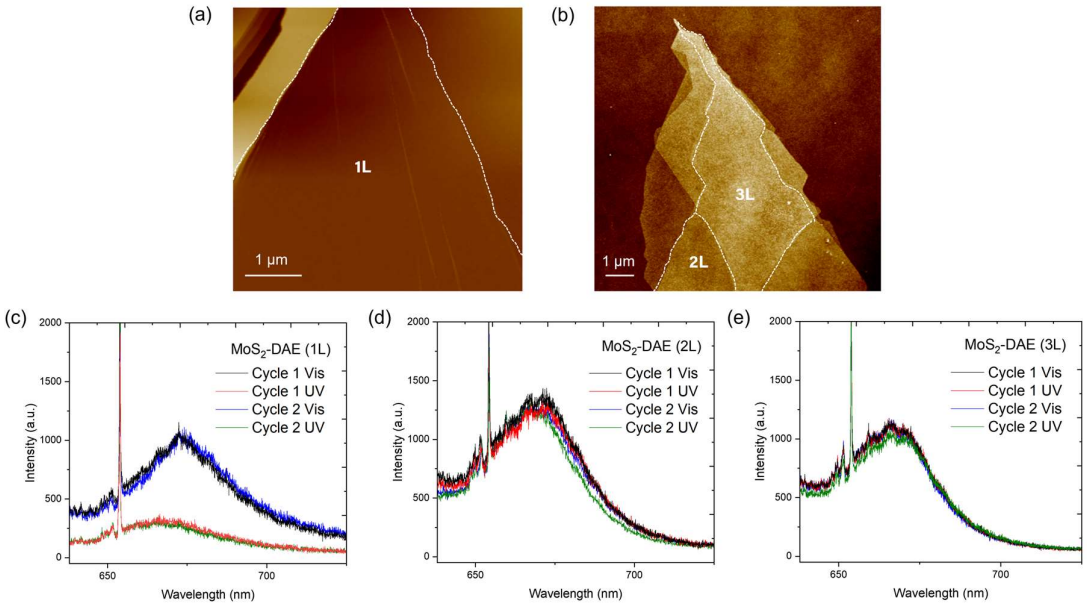

Figure S2. AFM image of MoS_2 flakes featuring (a) 1L, (b) 2L, and 3L, with the boundaries highlighted by dotted lines. The photo-modulation processes were repeated twice on the 1L to 3L MoS_2-DAE system. (c) The 1L MoS_2 exhibits significant PL quenching after UV irradiation, with the signal fully recovering under visible light. In contrast, the hybrids with (d) 2L and (e) 3L MoS_2 show no notable PL quenching.

3. Control Experiments with Monolayer MoS_2

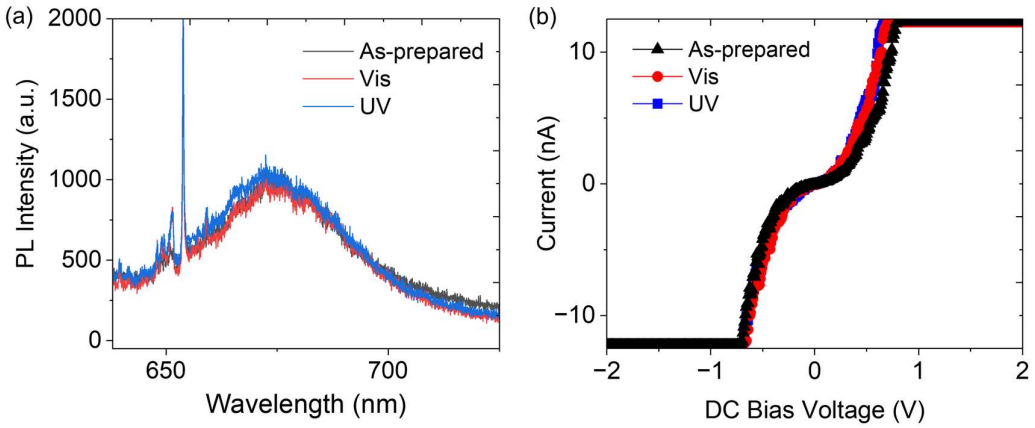

Figure S3. Control experiments were conducted with 1L MoS_2 (without a DAE layer) to confirm the effects of visible and UV light irradiation, which also indicates minimal substrate effects on charge transfer interactions. (a) The PL emission of as-prepared monolayer MoS_2 is consistent with those after exposure to visible and UV irradiation, showing no significant differences. (b) The C-AFM results also indicate that the variations between each case are minimal. These results indicate that optical modulation in the 1L MoS_2-DAE system arises from the unique hybrid interaction between DAE and 1L MoS_2.

4. AFM Imaging of MoS_2 Samples on ITO Substrates

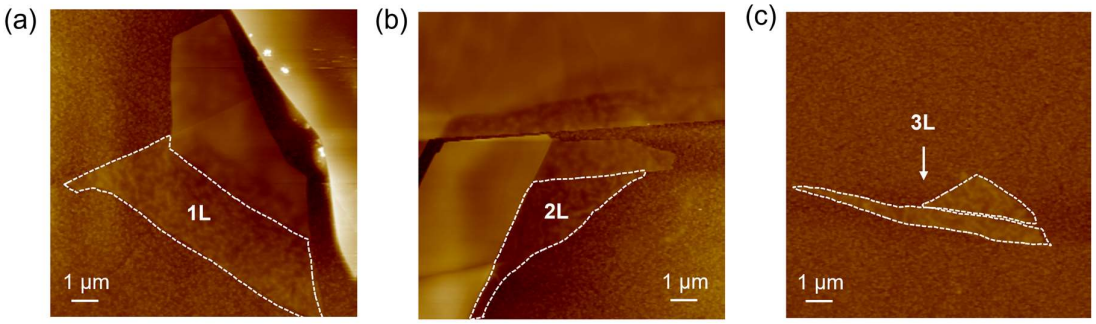

Figure S4. AFM images of (a) 1L MoS_2, (b) 2L MoS_2, and (c) 3L MoS_2 flakes fabricated on ITO substrates. These samples were used to measure C-AFM data presented in Figure 4.

5. C-AFM Measurements of Additional MoS_2-DAE Hybrid Samples

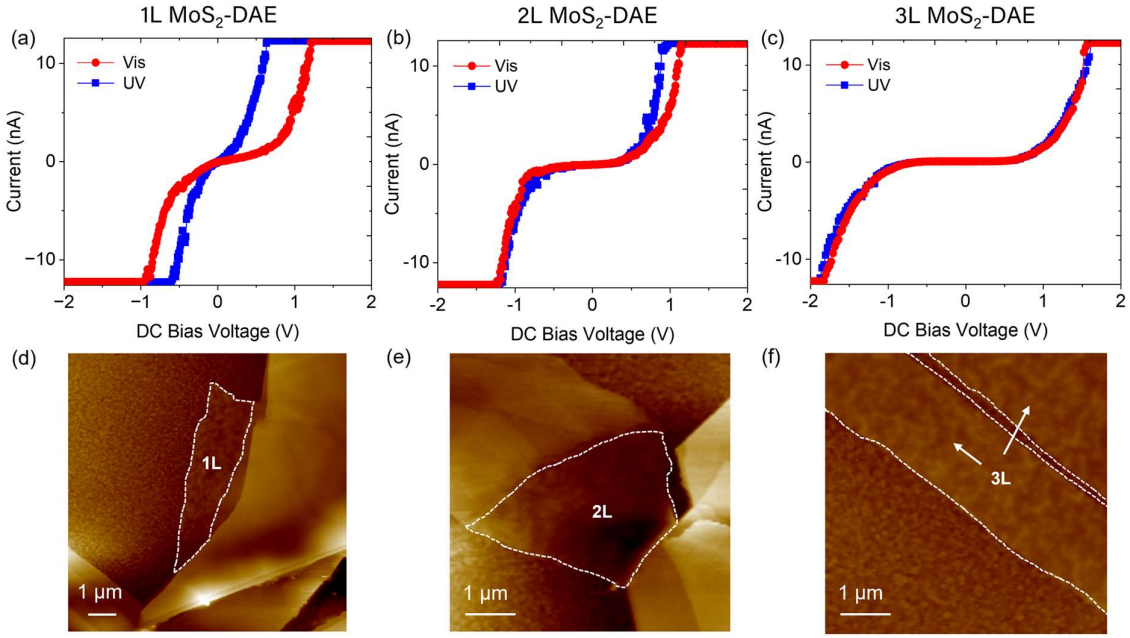

Figure S5. Additional C-AFM measurements were conducted on MoS_2-DAE hybrid samples under both visible and UV light irradiation. These measurements were performed on (a) 1L MoS_2, as illustrated in the AFM image (d); (b) 2L MoS_2, as depicted in the AFM image (e); and (c) 3L MoS_2, as shown in the AFM image (f).

6. DFT Calculations

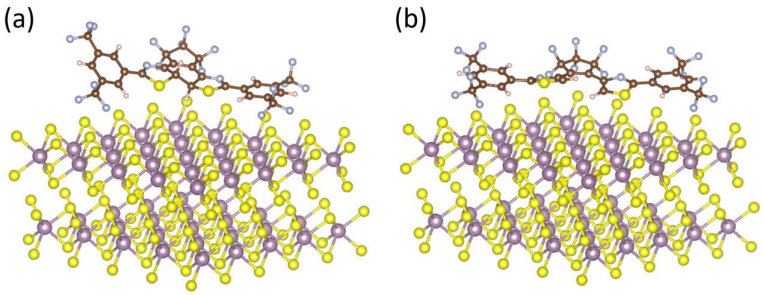

Figure S6. Representative relaxed hybrid structures. (a) Closed- and (b) Open-form DAE on 2L MoS_2-DAE. The DAE conformation and interaction at the hybrid heterointerface are consistent with those between DAE and 1L and 3L MoS_2.