

## Supplementary Material

### Different types of band alignment at MoS<sub>2</sub>/(Al, Ga, In)N heterointerfaces

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The FWHM values of MoS<sub>2</sub>/InN is the highest, followed by MoS<sub>2</sub>/AlN, and the value for MoS<sub>2</sub>/GaN is the least, thus indicating that growth quality of MoS<sub>2</sub> on GaN is best among the three (Fig. S1).

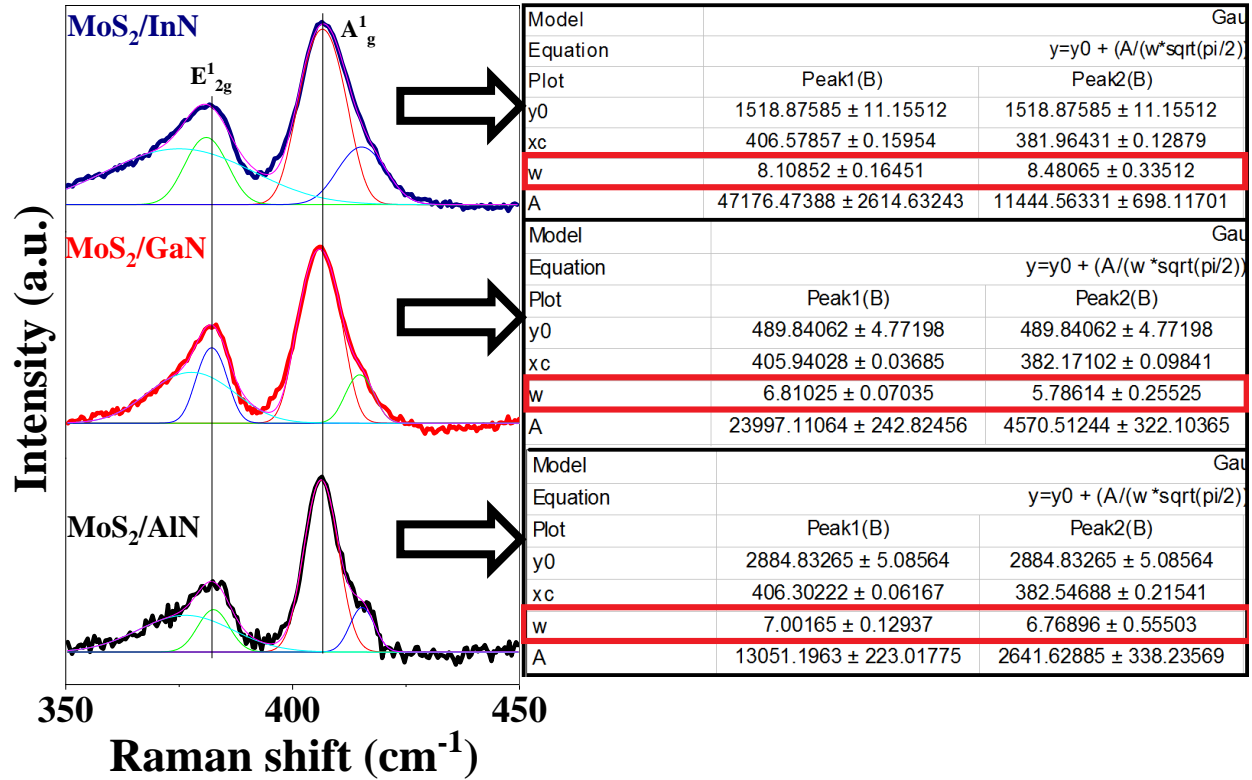


FIG. S1. Peak fitting of the two Raman modes of MoS<sub>2</sub> in different MoS<sub>2</sub>/III-nitrides. The FWHM (w) values of the corresponding spectra have been shown along (marked by a red rectangle).

The AFM images of the top surface of MoS<sub>2</sub>/(Al, Ga, In)N are shown in Fig. S2. It can be seen that the MoS<sub>2</sub> films are quite smooth, with some particulate formation in all the three samples. The root mean square (rms) surface roughness values for MoS<sub>2</sub>/AlN, MoS<sub>2</sub>/GaN, and MoS<sub>2</sub>/InN are found to be 0.571, 0.501, and 1.26 nm, respectively.

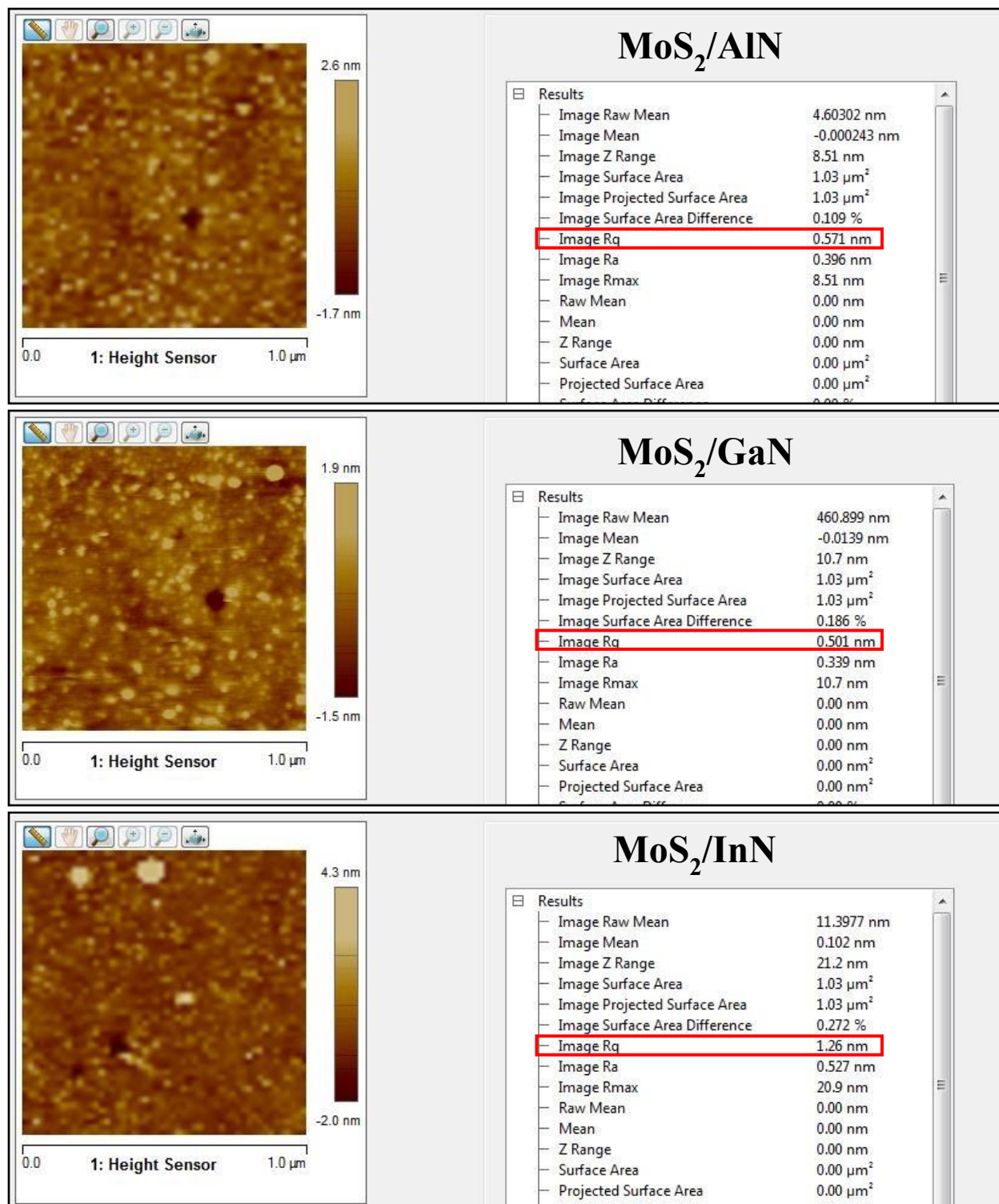


FIG. S2. AFM scans of top surface of different MoS<sub>2</sub>/III-nitrides. The RMS roughness ( $R_q$ ) values of the corresponding surfaces have been shown along (marked by a red rectangle).

The band offsets for different MoS<sub>2</sub>/III-nitrides determined by HRXPS measurements are shown in Fig. S3.

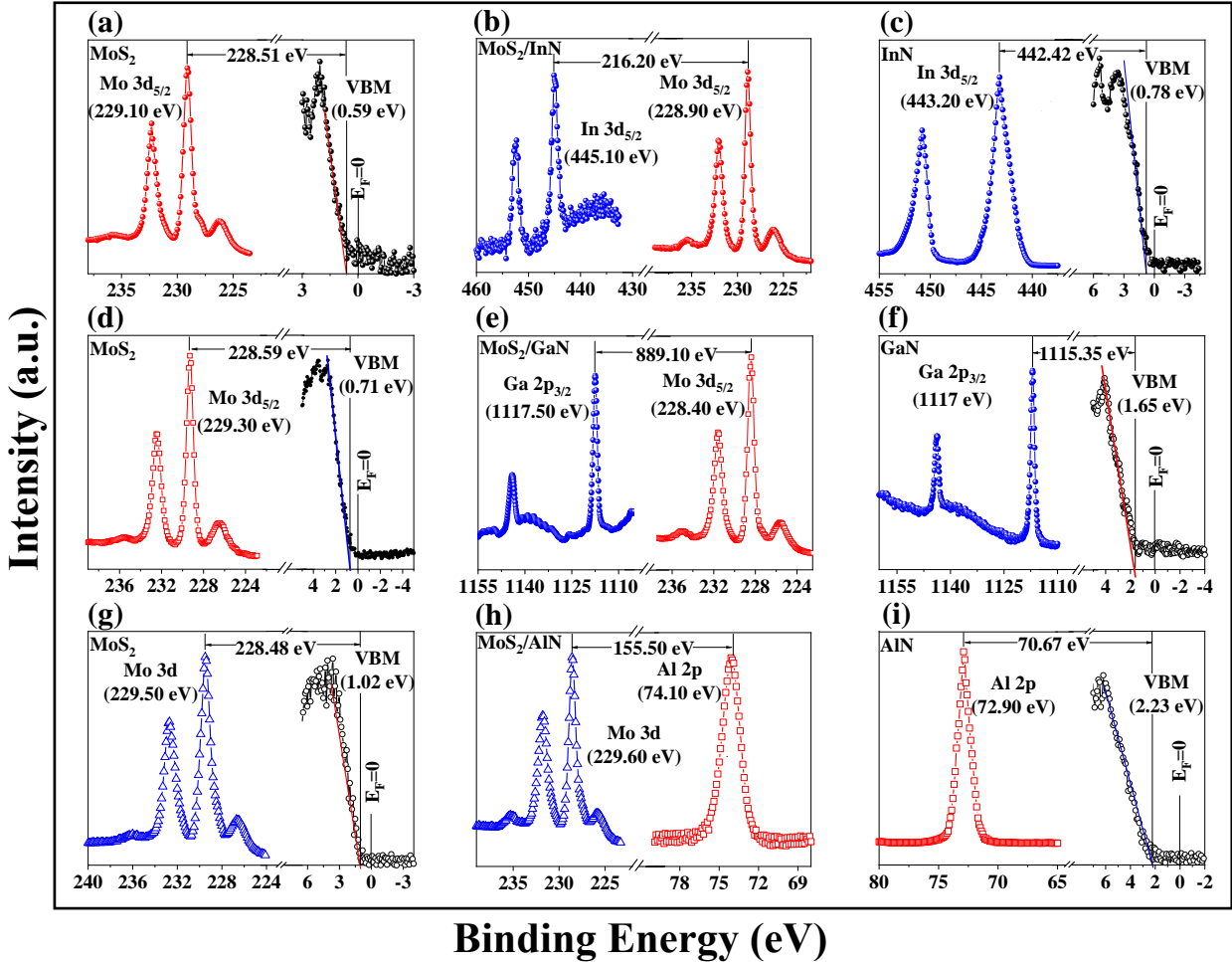


FIG. S3. For MoS<sub>2</sub>/InN: (a) Mo3d core level and valence band spectra acquired from MoS<sub>2</sub>, (b) In3d and Mo3d core level spectra of MoS<sub>2</sub>/InN heterojunction, (c) In3d core level and valence band spectra of InN epilayer; for MoS<sub>2</sub>/GaN: (d) Mo3d core level and valence band spectra acquired from MoS<sub>2</sub>, (e) Ga2p and Mo3d core level spectra of MoS<sub>2</sub>/GaN heterojunction, (f) Ga2p core level and valence band spectra of GaN epilayer; and for MoS<sub>2</sub>/AlN: (g) Mo3d core level and valence band spectra acquired from MoS<sub>2</sub>, (h) Al2p and Mo3d core level spectra of MoS<sub>2</sub>/AlN heterojunction, (i) Al2p core level and valence band spectra of AlN epilayer. Peak positions are marked in the parentheses.

**Figure S4** is a schematic of the comparison of band alignments of MoS<sub>2</sub> with different III-nitrides obtained from the above HRXPS results.

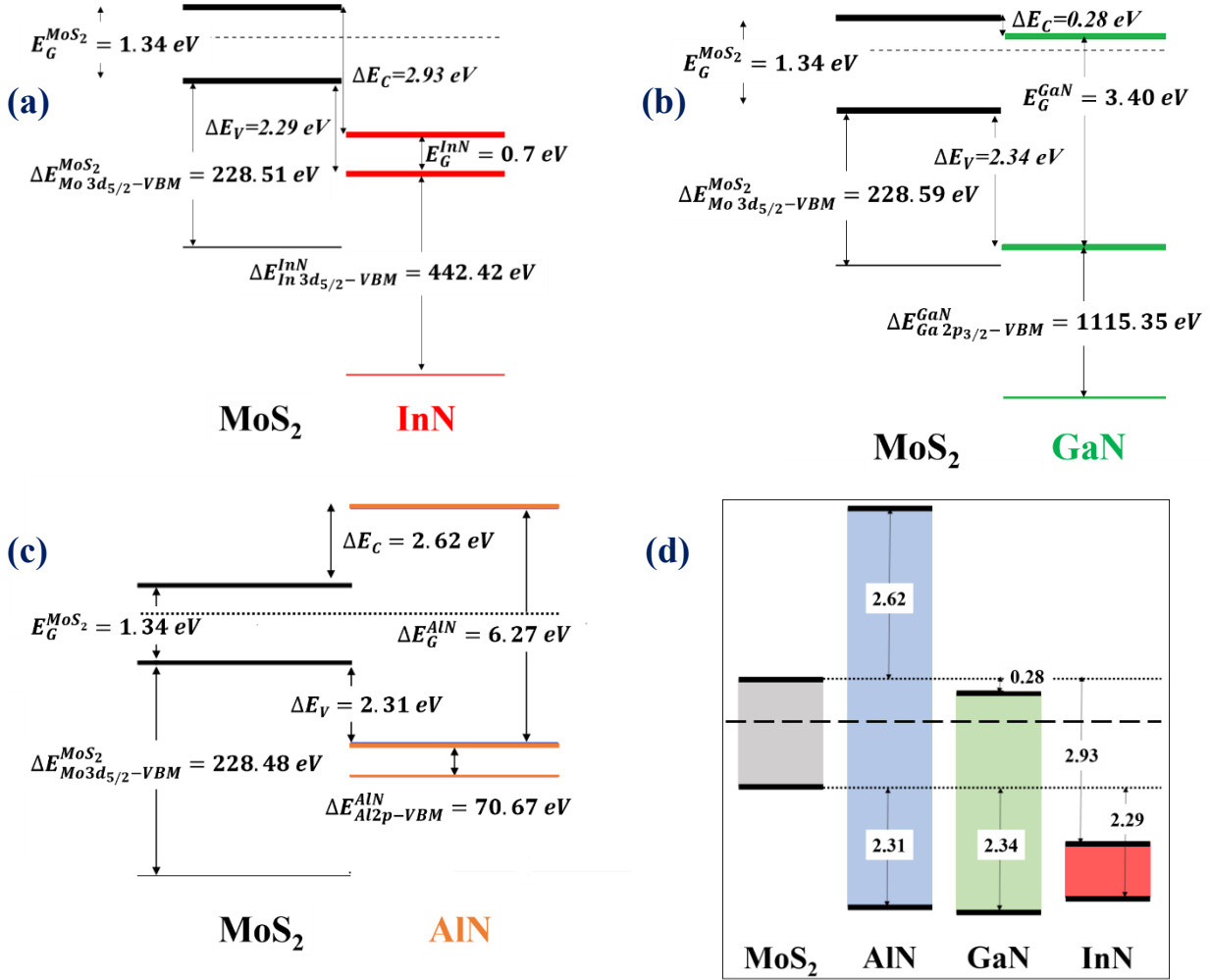


FIG. S4. Band alignment at (a) MoS<sub>2</sub>/InN, (a) MoS<sub>2</sub>/GaN, (c) MoS<sub>2</sub>/AlN, and (d) schematic of band alignments of MoS<sub>2</sub> with the III-nitride semiconductors (All energy values are in eV).