

Structural and electronic signatures of stacking and intralayer transitions in epitaxial bilayer WSe₂ on GaP(111)B

Meryem Bouaziz^{1,2}, Samir El Masaoudi³, Aymen Mahmoudi¹, Niels Chapuis⁴, Mathieu Silly⁵, Gilles Patriarche¹,
Iann C. Gerber³, Julien Chaste¹, Fabrice Oehler¹, Xavier Wallart⁴, Abdelkarim Ouerghi¹

¹Université Paris-Saclay, CNRS, Centre de Nanosciences et de Nanotechnologies, 91120, Palaiseau, Paris, France

²Laboratoire de Chimie Physique – Matière et Rayonnement, UMR 7614, Sorbonne Université, 4 Place Jussieu, 75005 Paris, France

³Université de Toulouse, INSA-CNRS-UPS, LPCNO, 135 Avenue de Rangueil, 31077 Toulouse, France

⁴Univ. Lille, CNRS, Centrale Lille, JUNIA ISEN, Univ. Polytechnique Hauts de France, UMR 8520-IEMN F59000 Lille France

⁵Synchrotron SOLEIL, L'Orme des Merisiers, Départementale 128, 91190 Saint-Aubin, France

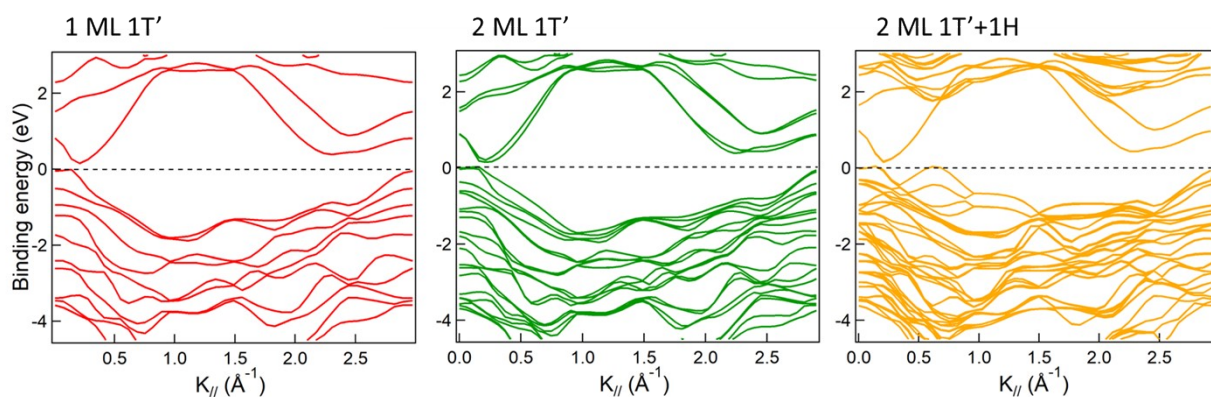


Figure S1: Comparison of the calculated electronic band structures of 1T' monolayer, 1T' bilayer, and mixed 1T'/1H bilayer WSe₂ using HSE06 calculations. The results highlight the similarity between the monolayer and bilayer 1T' dispersions, as well as the modifications induced by the presence of the underlying 1H layer. In particular, the comparison illustrates the degree of interlayer hybridization in the 1T'/1H configuration.

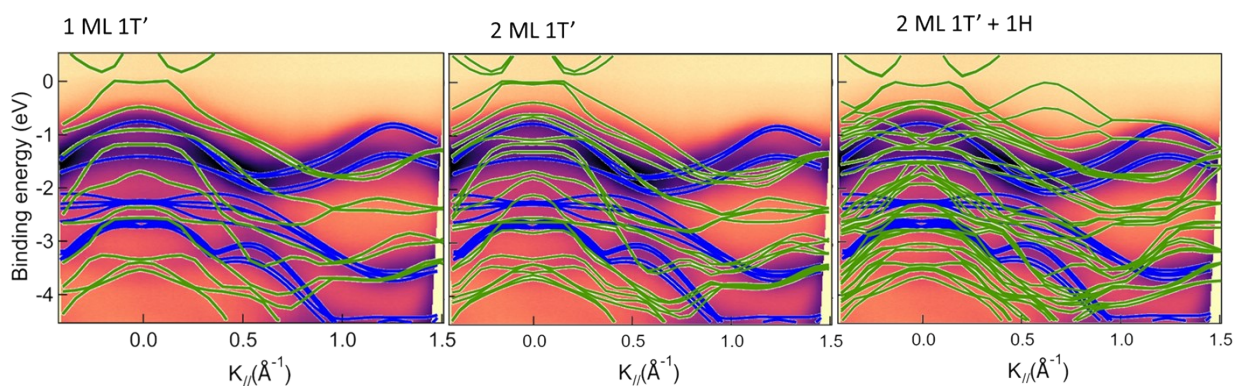


Figure S2: ARPES band dispersion of the as-grown bilayer WSe₂ compared with DFT-calculated band structures for different configurations (1T' monolayer and 1T'/1H bilayer) along the Γ -K-M directions. The Fermi level is set to zero binding energy. The comparison highlights that the experimental features are better reproduced by a monolayer-like 1T' band structure, indicating weak electronic coupling between the 1T' layer and the underlying 1H layer.