Band alignment and interlayer hybridisation in transition metal dichalcogenide/hexagonal boron nitride heterostructures

Samuel J Magorrian
A. J. Graham, N. Yeung, F. Ferreira, P. V. Nguyen, A. Barinov, V. I. Fal'ko, N. R. Wilson, N. D. M. Hine
Department of Physics, University of Warwick, Coventry, United Kingdom
samuel.magorrian@warwick.ac.uk

In van der Waals heterostructures, the relative alignment of bands between layers, and the resulting band hybridisation, are key factors in determining a range of electronic properties. This work[1] examines these effects for heterostructures of transition metal dichalcogenides (TMDs) and hexagonal boron nitride (hBN), an ubiquitous combination given the role of hBN as an encapsulating material. We compare results of linear-scaling density functional theory (DFT) calculations using large low-strain supercells with experimental angle-resolved photoemission spectroscopy (ARPES) results. We explore the hybridisation between the valence states of the TMD and hBN layers, and show that it introduces avoided crossings between the TMD and hBN bands, with umklapp processes opening 'ghost'[2] avoided crossings in individual bands. Comparison between DFT and ARPES spectra for the $MoSe_2$/hBN heterostructure (Fig. 1) shows that the valence bands of $MoSe_2$ and hBN are significantly further separated in energy in experiment as compared to DFT. We then show that a novel scissor operator can be applied to the hBN valence states in the DFT calculations, to correct the band alignment and enable quantitative comparison to ARPES, explaining 'ghost' avoided crossings and other features in the ARPES spectra.

References

[1] S. J. Magorrian *et al.*, 2D Materials 9 (2022) 045036
[2] A. J. Graham *et al.*, 2D Materials 8 (2020) 015016

Figures

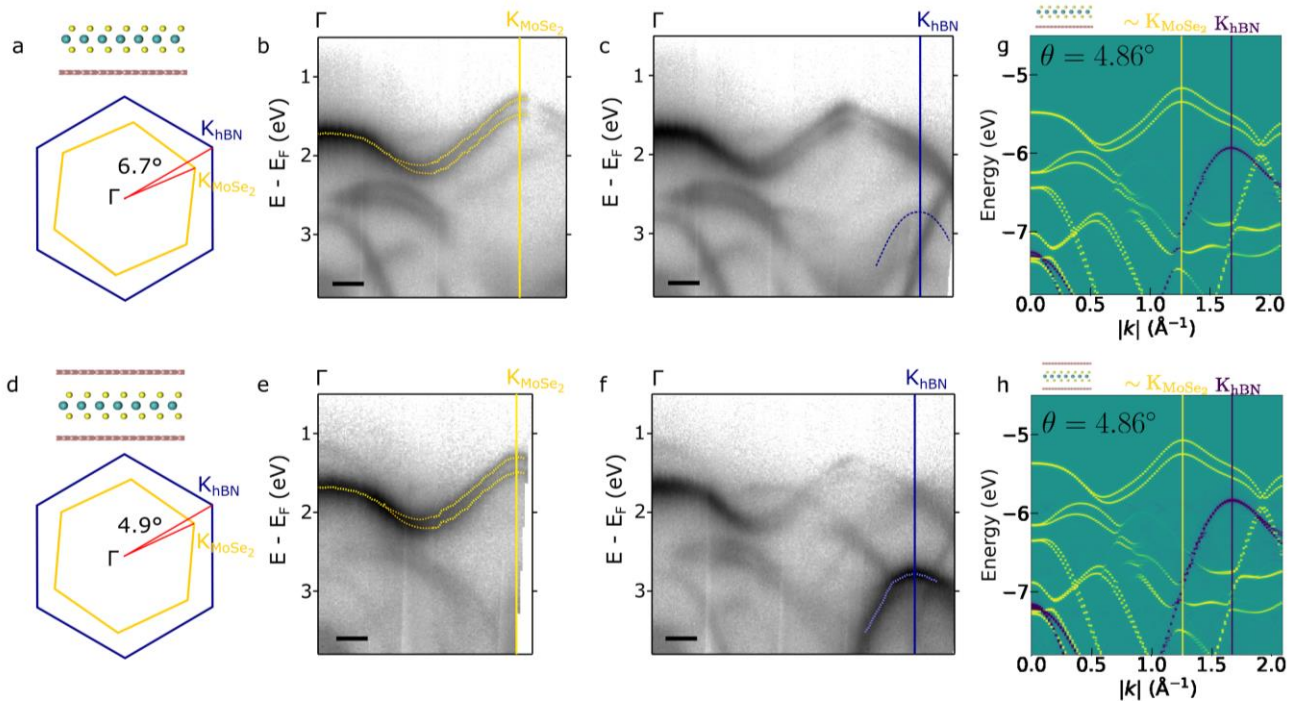

Figure 1: Comparison of ARPES and unfolded DFT spectra (with no scissor correction applied at this stage) for $MoSe_2$/hBN heterostructures. (a) Schematics of $MoSe_2$/hBN heterostructure and of $MoSe_2$ and hBN Brillouin zones. (b) and (c) slices from ARPES spectra along K ($MoSe_2$) and K (hBN) directions, respectively. (c)-(f) as (a)-(c), but for a region of the heterostructure with hBN also on top of the $MoSe_2$. These are shown with LS-DFT spectra for $MoSe_2$ nearly aligned with hBN, for monolayer hBN on one side (g) and both sides (h) of $MoSe_2$.