Modelling 2D materials on a large-scale using BigDFT

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Although widely-used for simulating the electronic properties of materials, density functional theory (DFT) often struggles with computational efficiency and scalability. Order N methods offer a solution to study large and complex systems, but accuracy and reliability can be compromised in scenarios where nearsightedness is not properly imposed when defining the basis set.
The device integration of two-dimensional materials across multiple fields, from electronics and optoelectronics to energy, sensing, and beyond, demands to achieve large-scale characterization, in order to predict the performance of novel applications and the material's behavior under realistic conditions.

The BigDFT software addresses these challenges by providing a robust platform for DFT calculations with advanced features tailored for large-scale systems [1]. First, the wavelet basis set offers high accuracy and flexibility by adapting its grid in regions of low and high resolution. From this basis set, localized functions are constructed *in situ* during the self-consistency cycle. Essential for studying 2D materials and their heterostructures, the Poisson solver allows to handle surface and wire boundary conditions in a straightforward manner using the Green's function method. Finally, the quasi-observable approach yields an intuitive description of electronic states that facilitates post-processing and building QM/MM models, a crucial feature for understanding defects in low concentrations [2]. We demonstrate here the accurate treatment of strain fields, interstitial and substitutional defects inside transition metal dichalcogenides using the linear-scaling framework of BigDFT, which is illustrated in Fig.1. A novel feature is presented, where the electronic structure is described at the nanoscale using a tight-binding model. Finally, some perspectives are discussed on the *ab initio* characterization of line defects, interfaces and heterostructures of 2D materials.

References

[1] Ratcliff, Laura E., et al. The Journal of chemical physics 152.19 (2020).
[2] Dawson, William, et al. Wiley Interdisciplinary Reviews: Computational Molecular Science 12.3 (2022): e1574

Figures

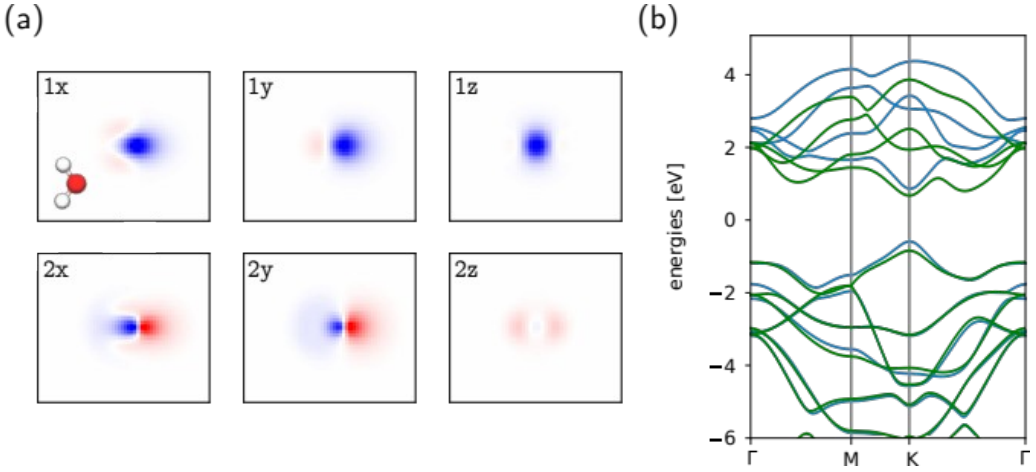

Figure 1: Linear-scaling framework of BigDFT. (a) The localized basis set adapts to the systems chemistry, as shown for the reproduction of *s*- and *p*-orbitals of a water molecule. (b) The comparison between the band structures obtained from the linear- and cubic-scaling approaches demonstrates the efficiency of the localized basis-set optimized *in situ*.