Stacking at the bottom

Weak interactions in layered graphene derivatives - a high level *ab-initio* study

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In Short

- Evaluating the properties of nanomaterials by stacking.
- Interaction of 2D materials with liquids and gases.
- Evaluating different methods/basis sets for *ab-initio* multi-scale modeling of materials.

Research into what are collectively called 2D-materials, i.e. thin film structures of only a few atomic layers, has come a remarkably long way since its (very recent) beginnings. Initial interest was sparked by the seminal paper by Geim and Novoselov[1] in which they reported the first experimental creation of graphene, a single-layered, fully π-conjugated carbon structure. In the wake of this, a lot of interest has been focused on the properties of this remarkable material, its high electric and thermal conductivity as well as tensile strength[2]. Though, while possessing many of the properties desirable for technological applications, pristine graphene suffers from a major deficiency. This deficiency lies in it being a semi-metal/zero-band gap semiconductor, which makes applications as a channel material in future transistor applications difficult.

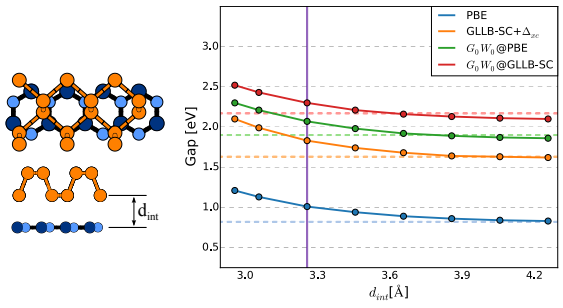

***Figure 1:** The left-hand figure shows an atomic model of the phosphorene/BN bilayer system as well as the definition of the interlayer distance used. The right-hand image shows the evolution of the electronic band gap as a function of the interlayer spacing for the same system.*

Driven by the excitement sparked by the discovery of graphene, the spotlight was shone on many other examples of 2D atomic structures [3], some that had been well-known for other applications such as hexagonal boron nitride or molybdenum disulfide, others that were synthesized for the first time. Chemical modifications of graphene itself take a special place amongst this latter category, the most notable examples being its fully hydrogenated (graphane), fully fluorinated (fluorographene) as well as fully chlorinated form (chlorographene).

Over the last years considerable attention was dedicated to the study of these materials, focusing in particular on the possibility of modifying their properties by combining them into vertical multilayerd superstructures as layed out in [4].

The aim of our project is to apply high-level theoretical methods to the investigation of these structures to gain insights which might help to guide experimental research. Additionally, we want to provide crucial insights from careful investigation of the individual factors governing the properties of these fascinating materials. One work on the influence of the electric field on layered structures has already been published in the scientific literature [5].

An example of the systems studied during the first part of our project is shown in figure 1. This particular work, which is in the course of being submitted for publication [6], investigates the phosphorene (a 2D form of phosphorus) band gap in contact with hexagonal boron nitride. As phosphorene is highly susceptible to degradation by oxygen under ambient conditions with boron nitride being proposed as a protective material, its influence on the properties of the potential next-generation semi-conductor is obviously of great interest.

As a second example, figure 2 shows work studying the properties of some heavy halogen functionalized graphene derivatives. These materials which were first studied in detail by us, were found to demonstrate a number of fascinating properties, not least the presence of an optical gap very close to the maximum efficiency limit of solar cells. This, combined with their predicted high stability, makes them excellent candidates for future solar cell applications. Again, a manuscript summarizing our results is in preparation and will be submitted for publication shortly [7].

During the second part of our project, we now wish to extend upon these existing results by expanding the number of 2D materials investigated. Great focus will lie on the properties of molybdenum disulfide concerning both its interactions with other 2D materials and with water and hydrogen sulfide. This research will be performed in close collaborations with our experimental partners from the groups of Prof. Dr. Kyrill Bolotin and Prof. Dr. Katharina Franke at FU Berlin, physics department. Using different computational approaches ranging

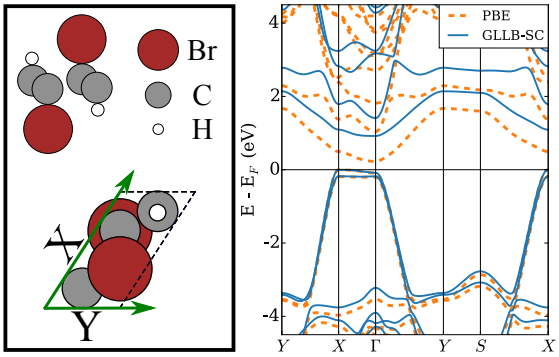

Figure 2: *The left-hand figure shows an atomic model of one of the system whose electronic band structure is shown on the left-hand site at both the GLLB-SC and PBE level.*

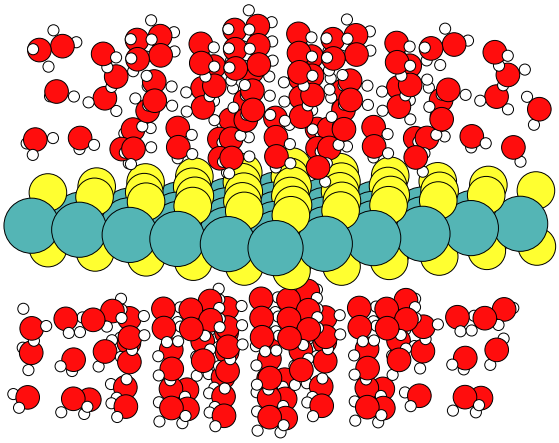

Figure 3: *MoS_2 monolayer in a cell of liquid water.*

from fast LCAO+vdW-DFT to highly accurate GW calculations, we will investigate the interaction of both gaseous and liquid water and hydrogen sulfide with MoS_2 which we hope can lead the way to applications ranging from highly-sensitive gas sensing to the photocatalytic splitting of water.

Given the great success of our earlier studies as well as the exciting prospect of a close collaboration with excellent experimental groups located at our same university, we hope that the second part of our project will match up to and surpass its first iteration.

WWW

http://www.bcp.fu-berlin.de/chemie/

More Information

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