1. This relatively simple G/hBN heterosystem ("G" stands for graphene) offers four different structures, two of them are distinguished by preferable hydrogen bonding to hBN ("H on B" and "H on N" bonds are the two possibilities) and next two can be distinguished by AA *vs*. AB stacking (as shown in the figures below). From the structural TE minimization "H on B" system (when the hydrogen atoms are bonded to boron atoms, while nitrogen is involved in creating the covalent interlayer bonding with carbon atom) is 0.43 eV per 6 atom unit cell above the TE for the similar "H on N" structure. Due to large TE difference between the two systems, only "H on N" structure (with AA and AB stacking) will be discussed here in details.

For pristine (*not hydrogenated*) graphene van der Waals(VDW) bonded bilayer AA and AB stacking are practically degenerate in terms of the (very low) binding energy [2013Constantinescu] [2015Mostaani]. Similar band structure for AA and AB stacking [2011Birowska], our test simulations for such systems agree well with the published results.

Attached are (i) description of the structural models, (ii) input files for the structure plotting programs (*e.g*., it is *.xyz format for Molekel software or similar) and (iii) two input files in the Quantum Espresso (QE) format for completely hydrogenated graphene-hexagonal boron nitride (G-hBN) bilayers (or graphene-hBN 2D heterojunctions), which are the two simplest examples and represent a good starting point for calculations. Both sides of the bilayer contain 50% of H monolayer on the top and 50% on the bottom (maximum possible amount of H that can be absorbed) and they both have hexagonal symmetry as shown in the Fig. 3 in the end). Considering the preferable bonding (as dictated by the lowest total energy (TE) of the configurations) and looking at the structures side views (Figs. 1 and 2), the atomic bonding sequence in the primitive cell is H – C – C – B – N – H. The primitive cell contains six atoms, two H (one at each side), one B, one N and two C atoms, the interlayer bonding is between B and C. The two cases in Figs. 1 and 2 differ only by their stacking sequence (AA and AB), both are practically degenerate in the total energy. Both heterosystems demonstrate the direct DFT-LDA gap close to 1 eV (more details are below).

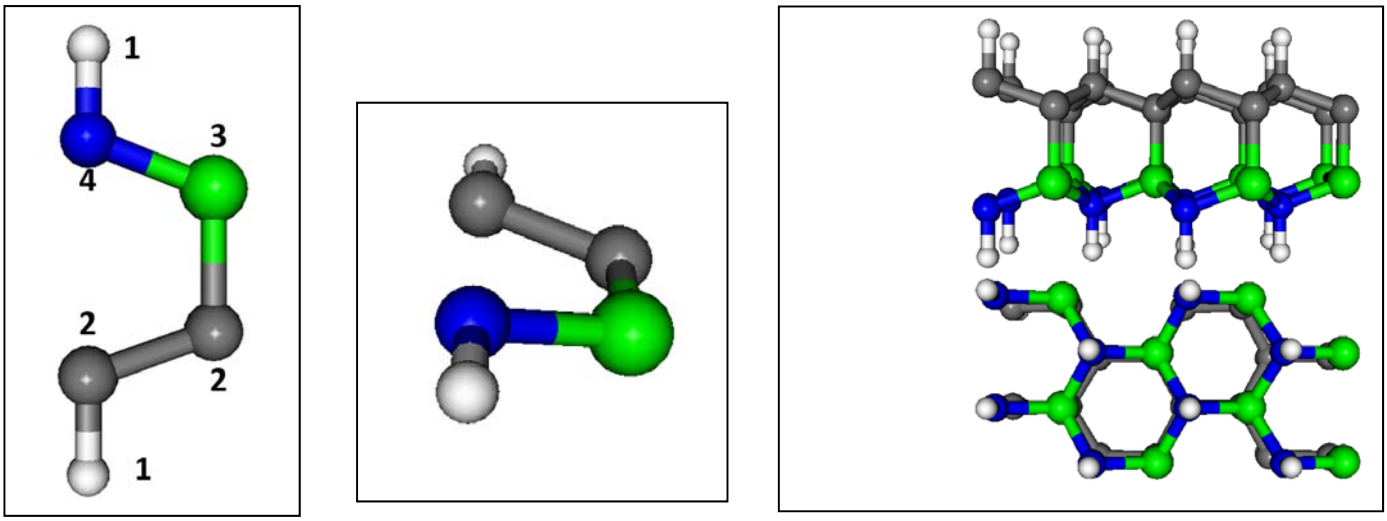

Fig. 1. 50% hydrogenated at both sides ***AA*** stacked graphene/h-BN 2D heterosystem with marginally less TE than for AB stacking (Fig. 2): side view at the primitive cell at the left), a perspective view (in the center) and periodically translated 2D crystal (at the left, upper panel is the side view and the top view is at the bottom). Atoms colour and numbers are: (1) H – white, (2) C – grey, (3) B – green, (4) N – blue.

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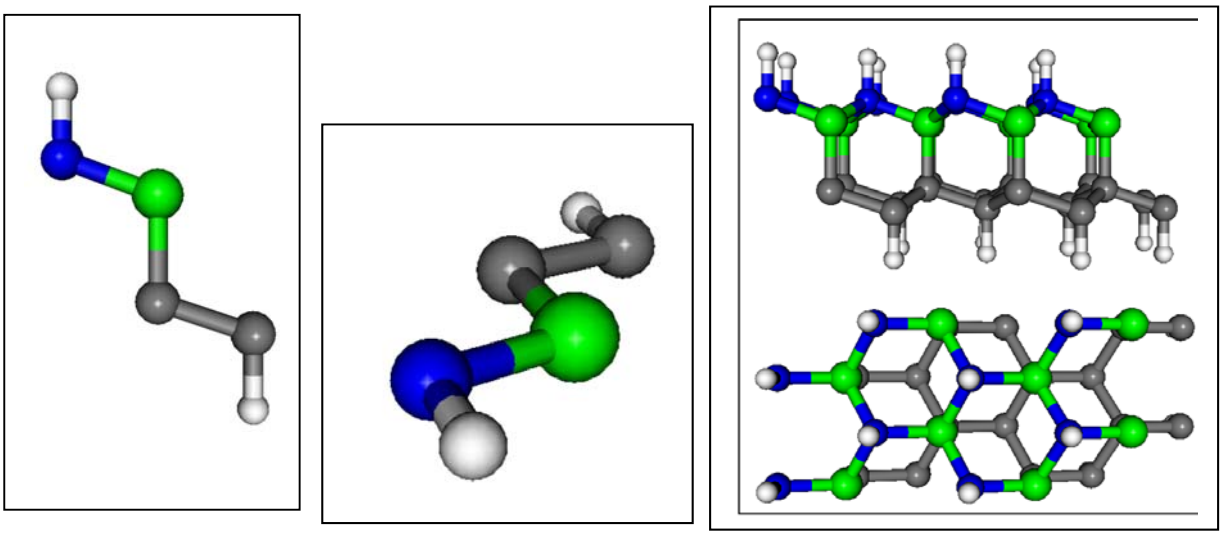


Fig. 2. 50% hydrogenated at both sides ***AB*** stacked graphene/h-BN 2D heterosystem: side view at the primitive cell at the left), a perspective view (in the center) and periodically translated 2D crystal (at the left, upper panel is the side view and the top view is at the bottom). Atoms colour and numbers are: (1) H – white, (2) C – grey, (3) B – green, (4) N – blue.

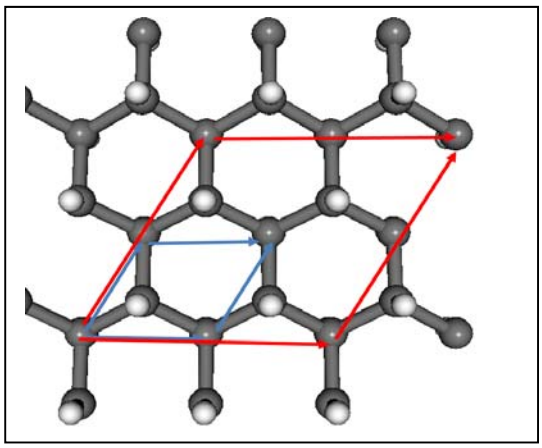


Fig. 3. Top view at the carbon terminated and 50% hydrogenated bilayer. Blue arrows show the primitive hexagonal unit sell, while the red arrows indicate 2 × 2 supercell.

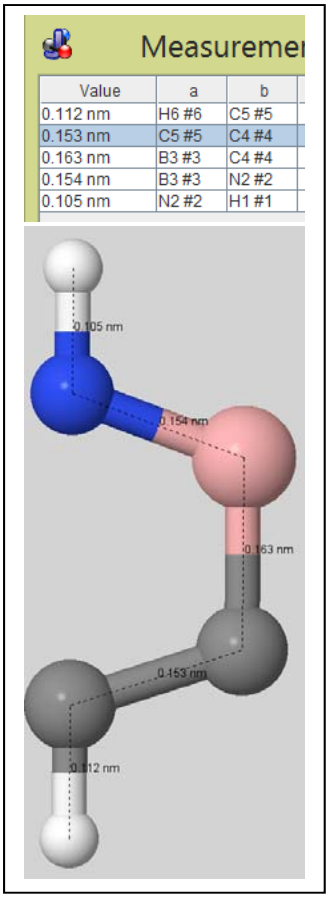
Last two weeks to confirm my old results and to more deeply consider various systems properties, I have redone a lot of atomic structure, band structure and DOS calculations for the simplest hydrogenated H-G-hBN-H bilayer (G stands for graphene, hBN is hexagonal boron nitride), collected a lot of relevant information from the literature about hBN, non-hydrogenated G-hBN systems, and prepared detailed summary of the results.

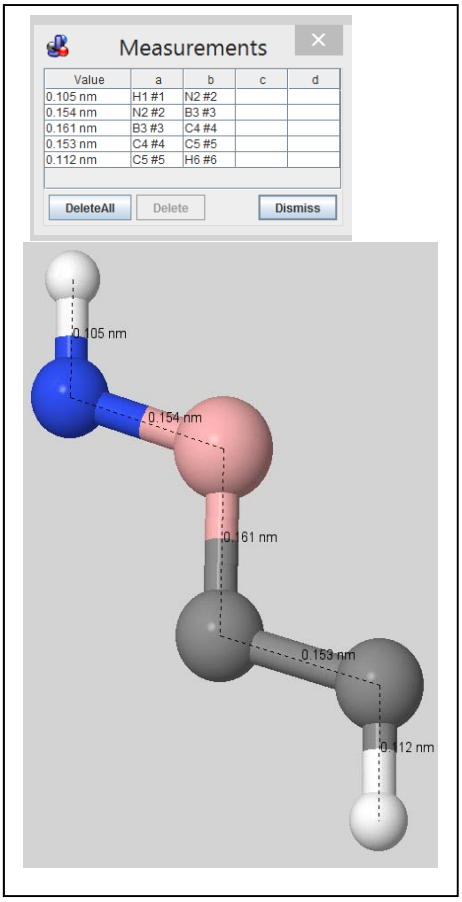
The next page shows the improved (using slightly higher bulk constant from the TE minimization) structural and LDA band structures. (We confirmed the previous results for different graphene bases systems, that from the point of view of the geometric structure LD is slightly better than GGA). As you see, AA and AB stacking are producing very similar band structure, which is direct one.

Here are improves (very slightly) structural, electron band and other results for 4.75 AU "bulk" constant, which is TE minimum for the bilayer parameters chosen.

Bond lengths B – N, C – C, and C – H are approximately the same for both structures, although C – B bonds are slightly different

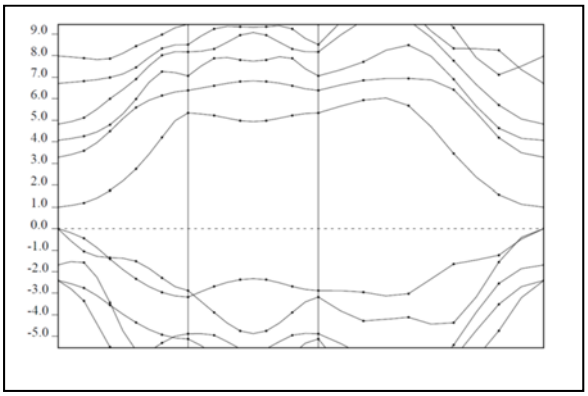
AA – staked bilayer *vs*. AB – stacked bilayer

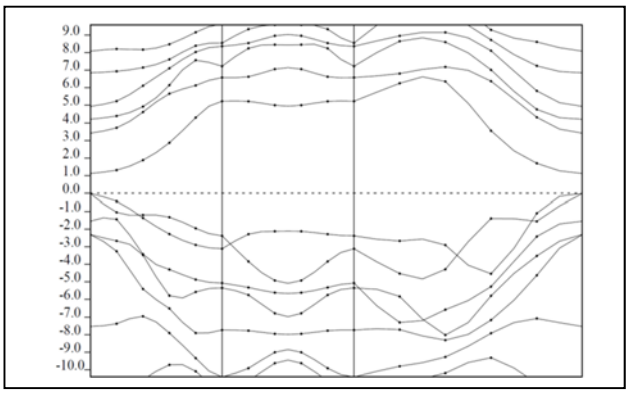




For AA stacking, ***direct*** band gap 1.0 eV

For AB stacking, ***direct*** band gap 1.15 eV





In both cases the energy gap from DOS coincides with those ones from the band structure

Now I am moving to the hydrogenated trilayer, the hydrogenated graphene “glued” to the BN substrate by hydrogenation will be the next. But we can start with the above structures.

What I’d suggest, instead of using email only, we can skype with you and talk about the systems. There are too many technical details. Let me know if this works. I am in Germany now, I believe that there is 7 hours difference. My skype id is “achkrebt”