Supporting Information

Interlayer Potential for Homogeneous Graphene and Hexagonal Boron Nitride Systems: Reparametrization for Many-Body Dispersion Effects

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As discussed in the main text, the original interlayer potential (ILP) for hexagonal boron nitride (*h*-BN)[1] was parametrized against density functional theory calculations performed using the B3LYP hybrid density functional,[2] augmented by the pair-wise dispersion correction of Tkatchenko and Scheffler (TS-vdW).[3] To demonstrate the effect of the many-body dispersion (MBD) correction scheme,[4-7] we compare in Fig. S1 the reference binding-energy curves (black) presented in the main text, obtained using the MBD corrected screened-exchange hybrid density functional of Heyd, Scuseria, and Ernzerhof (HSE),[8-11] to our previous B3LYP+TS-vdW reference data (green).[1] The inclusion of MBD effects results in smaller binding-energies and slightly increased equilibrium inter-dimer distances with respect to the TS-vdW results. These differences grow considerably with increasing contact size, thus emphasizing the importance of including MBD effects when considering interlayer interactions in large two-dimensional layered materials interfaces. Furthermore, the flexibility of the suggested ILP is clearly demonstrated, where appropriate parametrization provides good agreement with either pair-wise (blue) or MBD-based (red) reference data.

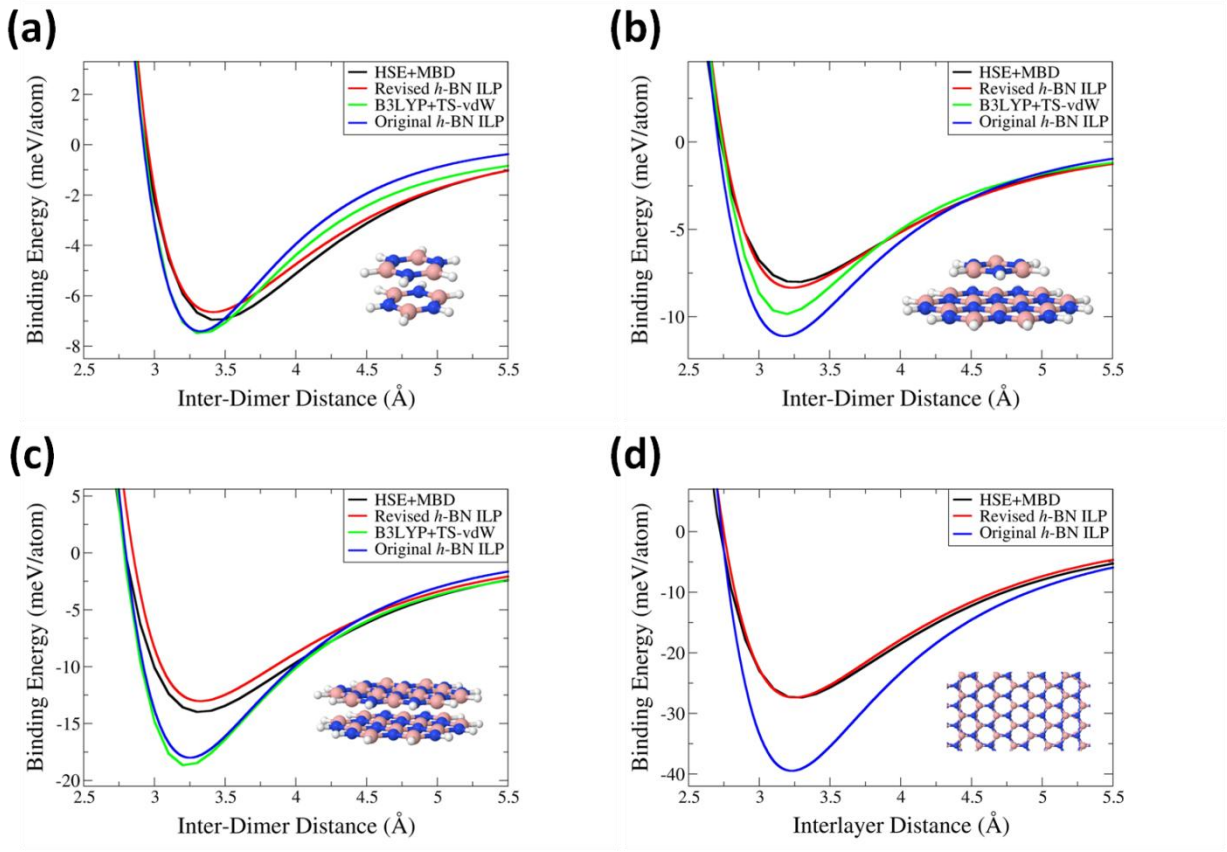

Figure S1: Binding-energy curves of (a) the borazine dimer; (b) the borazine/HBNC system; (c) the HBNC dimer; and (d) a periodic *h*-BN bilayer, all calculated using the revised *h*-BN ILP (red) parametrized against the reference HSE+MBD results (black), compared to the original *h*-BN ILP curves (blue) and their reference B3LYP+TS-vdW data (green).

References

(1) Leven, I.; Azuri, I.; Kronik, L.; Hod, O. Inter-Layer Potential for Hexagonal Boron Nitride. *J. Chem. Phys.* **2014**, *140*, 104106.
(2) Becke, A. D. Density-Functional Thermochemistry. III. The Role of Exact Exchange. *J. Chem. Phys.* **1993**, *98*, 5648-5652.
(3) Tkatchenko, A.; Scheffler, M. Accurate Molecular Van Der Waals Interactions from Ground-State Electron Density and Free-Atom Reference Data. *Phys. Rev. Lett.* **2009**, *102*, 073005.
(4) Tkatchenko, A.; DiStasio, R. A.; Car, R.; Scheffler, M. Accurate and Efficient Method for Many-Body van der Waals Interactions. *Phys. Rev. Lett.* **2012**, *108*, 236402.
(5) Ambrosetti, A.; Reilly, A. M.; DiStasio, R. A.; Tkatchenko, A. Long-Range Correlation Energy Calculated from Coupled Atomic Response Functions. *J. Chem. Phys.* **2014**, *140*, 18A508.
(6) DiStasio, R. A.; von Lilienfeld, O. A.; Tkatchenko, A. Collective Many-Body Van der Waals Interactions in Molecular Systems. *Proc. Natl. Acad. Sci.* **2012**, *109*, 14791-14795.
(7) Bučko, T.; Lebègue, S.; Gould, T.; Ángyán, J. G. Many-Body Dispersion Corrections for Periodic Systems: an Efficient Reciprocal Space Implementation. *J. Phys. Condens. Matter* **2016**, *28*, 045201.
(8) Heyd, J.; Scuseria, G. E.; Ernzerhof, M. Hybrid Functionals Based on a Screened Coulomb Potential. *J. Chem. Phys.* **2003**, *118*, 8207-8215.

(9) Heyd, J.; Scuseria, G. E. Assessment and Validation of a Screened Coulomb Hybrid Density Functional. *J. Chem. Phys.* **2004**, *120*, 7274-7280.
(10) Heyd, J.; Scuseria, G. E. Efficient Hybrid Density Functional Calculations in Solids: Assessment of the Heyd–Scuseria–Ernzerhof Screened Coulomb Hybrid Functional. *J. Chem. Phys.* **2004**, *121*, 1187-1192.
(11) Heyd, J.; Scuseria, G. E.; Ernzerhof, M. Erratum: "Hybrid Functionals Based on a Screened Coulomb Potential" [J. Chem. Phys.118, 8207 (2003)]. *J. Chem. Phys.* **2006**, *124*, 219906.