

ELECTRON TRANSPORT THROUGH SIMPLE MOLECULES FROM BAND STRUCTURE FORMALISM

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Electronic transport through simple molecules in breakjunctions constitutes a nanoelectronic paradigm which justifies extensive experimental and theoretical work in order to understand all details of the system. In particular, the existence of one almost completely transparent channel for the electronic transport through hydrogen rich platinum breakjunctions is still controversial in many aspects. Ref. [1] gives an actual review of the situation from the point of view of the mainstream. In spite of our efforts [2,3] showing that experiments are better explained if two hydrogen atoms lie transversal between Pt electrodes, most of the community still believes that a stretched hydrogen molecule lies longitudinally along the junction. Our first calculations were done using the ALACANT code [4] connecting the central part of the system which is described by any quantum chemistry method to complex Pt Bethe lattices. Although charge consistency is allowed in this process, it is clear that the scheme fails to reach the “standard” selfconsistency requirement of methods based on the density functional formalism. In this contribution, we show how to use “state of the art” CASTEP bandstructure calculations [5] to get the transport properties of nanojunctions. Actually, both the electrodes and the central part of the junction are treated on the same foot while conductance is just given by the bands appearing near the Fermi level.

CASTEP deals with crystalline cells as any bandstructure method. We take H atoms between small Pt pyramids describing the breakjunction and three slabs of bulk Pt to describe the electrodes. In this way, the whole experimental setup is considered in our approach. The price we pay for doing a really *ab initio* selfconsistent calculation is that transport occurs not through an isolated junction but through a periodic array of them. Actually, computational facilities limit the 2D size of our cell to a 4x4 Pt cluster. This gives a distance of about 1.1 nm between H atoms. Figures 1 and 2 show the 3D cells used for a transversal and dissociated hydrogen molecule and for the longitudinal just stretched one in plausible but not energy minimized positions. We end with two H atoms and 58 Pt atoms in the orthorhombic supercell. A 4x4x4 MK grid is used to describe the BZ although symmetry reduces to six or eight the number of k-points actually necessary. PBE functional is employed together with ultrasoft H and Pt pseudopotentials. The planewave basis set is defined by a 300 eV cut-off. Once selfconsistency is achieved, the bandstructure is obtained for much better grid at all irreducible 2D BZ k-points. In this way, we exactly know the number of bands at any energy, i.e., the exact number of channels at this energy. This gives an energy dependent value for the conductance G that should be averaged to get some physical –certainly statistical- meaning. Fig. 3 gives our main result, a comparison of the conductance obtained for both possible H geometries after doing running averages of G with a width of 0.5 eV. Although G of the transversal case is somewhat smaller than the experimental value of 1, once again the conductance is seemly larger than the one corresponding to the non dissociated case in agreement both with our previous works and present studies analysing the role of non-local exchange [6].

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References:

- [1] D. Djukic, K. S. Thygesen, C. Untiedt, R. H. M. Smit, K. W. Jacobsen, and J. M. van Ruitenbeek, *Phys. Rev. B*, **71** (2005) 161402(R).
 [2] Y. García, J. J. Palacios, E. San Fabián, J. A. Vergés, A. J. Pérez-Jiménez, and E. Louis, *Phys. Rev. B*, **69** (2004) 041402.
 [3] G. Chiappe, E. Louis, E. V. Anda, and J. A. Vergés, *Phys. Rev. B*, **71** (2005) R241405.
 [4] See <http://www.guirisystems.com/alacant>. ALACANT is an ab initio code for electronic quantum transport. It interfaces GAUSSIAN03 to implement the non-equilibrium Green's functions formalism.
 [5] M. D. Segall, P. J. D. Lindan, M. J. Probert, C. J. Pickard, P. J. Hasnip, S. J. Clark, M. C. Payne, *J. Phys.: Cond. Matt.* **14** (2002) 2717.
 [6] Yamila García et al. (to be published).

Figures:

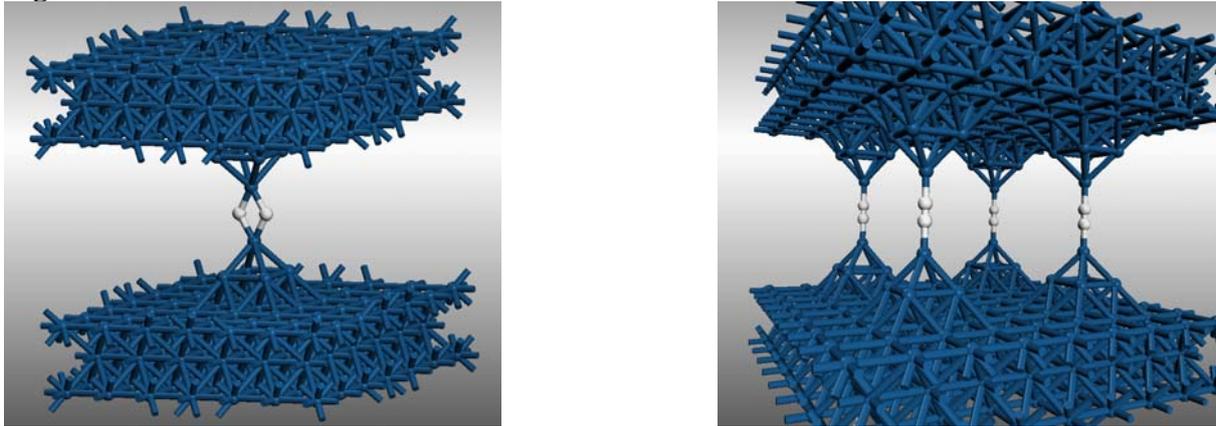


Fig. 1: Geometric structure used to get the bandstructure for the transversal dissociated H_2 molecule case.

Fig. 2: Supercell geometry used to calculate the bandstructure for a stretched hydrogen molecule in the longitudinal case.

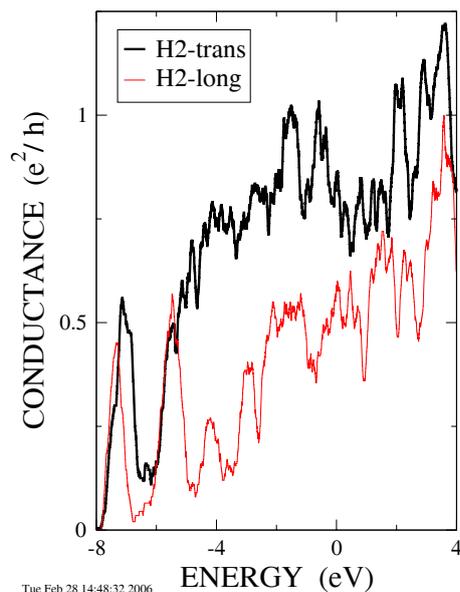


Fig. 3: Conductance as a function of the energy relative to the Fermi level for the two alternative structures studied in this work.