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Quantum Monte Carlo: The application to chemically accurate reaction activation barrier evaluation on metal catalyts

To introduce the Quantum Monte Carlo approach (QMC), recap first quantum mechanics, structure, and properties. The time-dependent or stationary Schrödinger equation is used. There is currently also a hive of research activity in developing the related approaches for and heterogeneous systems. Most methods are 'deterministic', is a unique valid solution which in a few 'toy' cases, such as the H₂ molecule or the uniform electron gas model of metals, can be obtained analytically. Arbitrary accuracy for any system must then be *ab initio* full configuration interaction (full CI). This gives electron correlation and properties. These reference calculations are often approximated. QMC provides a stochastic first principles 'solution' of the Schrödinger equation. To obtain precise results, stepwise in Diffusion Monte Carlo (DMC). DMC can be made CI, with explicit and is accessible for larger and more complex systems. It is reputed to give the 'correct' answer, albeit cited with standard error. Heterogeneous systems include molecule-metal surface reactions. This theoretical prediction of catalytic activation barriers uses QMC. The QMC methodology has been used for increasingly large molecules for 30 years and 15 years for solid fermion systems. The QMC random error for such systems can be made small by long simulations and averaging techniques but any systematic contribution must be removed with the help of benchmarks compared to reliable molecular beam experiment. The breakthrough regarding QMC metal surface reaction activation barriers presented here we have shown energy values to chemical accuracy, as compared to the molecular beam results for bond dissociation of hydrogen at Cu(111) and Pt(111). These values are poorly estimated by other first principles methods, yet QMC is very satisfactory. Secondly, reactions used in an involve a key bond-dissociation step, the validated QMC approach can be extended. Recently, we have been involved in the 'hot-topic' of the water-gas shift reaction. This is carbon-monoxide reacting (conditions at a Pt(111) surface) to yield hydrogen as a and carbon-dioxide burned to recover water, so removal of carbon-dioxide is the only issue and it can be accomplished by dissolution. The forward reaction removes the toxic effluent, carbon-monoxide. This so-called 'water-gas' equilibrium can go thermodynamically 'uphill' i.e. towards hydrogen, suitable catalyst. Barrier heights are less than 20 kcal/mol. Yields are excellent since a catalytic process is selective.

Biography

Philip E Hoggan, admitted to Trinity College, Cambridge in 1978, aged 17 on his entrance examination results. After his BA in Chemistry and Mathematics, he obtained a scholarship and completed the MA in Theoretical Chemistry in 1983. He went to Nancy, France where he obtained his PhD in 1987 (age 26). In France, it was then possible to continue and obtain a DSc by research, which he defended in public (fall 1991). This was the sesame to forming his own group, which was initially an integrated theme with an FTIR catalytic intermediate group in Caen, France. The STOP Slater Type Orbital Package was software developed also in this period 1992-1998. Having already passed the National Qualification for Professors in 1993, Philip E Hoggan finally obtained the chair for theoretical chemistry in Blaise Pascal University in 1998. Since 2005, solid surfaces, metal catalyts, and semiconductors. These, he and his number of first principles methods, including Quantum Monte Carlo which is the method of choice for the catalytic work. He has a half-a-dozen international conferences, in Cambridge, Buenos Aires, two locations in Turkey as in France. He speaks 5 languages fluently.

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