The puzzle of H$_2$ on Si(100):\\ a quantum Monte Carlo study

Table of Contents

The puzzle of H_2 on Si(100): a quantum Monte Carlo study

Claudia Filippi
Department of Physics, National University of Ireland, Cork, Ireland

The interaction of hydrogen with the silicon surface poses several interesting problems. The desorption of H_2 from Si(100) follows first−order kinetics, posing definite constraints on possible desorption mechanisms. The sticking coefficient of hydrogen on silicon is very small but no substantial adsorption barrier has been observed in desorption experiments. Theoretically, various attempts to explain these experimental findings have lead to no definite conclusion: density functional theory (DFT) calculations on extended slab geometries and highly correlated quantum chemistry calculations on small model clusters lead to completely different desorption mechanisms, each with limited agreement with experiments.

Using quantum Monte Carlo techniques, we compute accurate barriers for adsorption/desorption of molecular hydrogen on Si(100), for large model clusters of the Si surface. We show that the mechanism preferred in slab−DFT studies does not explain the experimental findings: the agreement between DFT calculations and experiments on the desorption barrier was fortuitous and simply due to the use of approximate exchange−correlation DFT functionals. Future calculations on different desorption mechanisms will be discussed.

In collaboration with Sorcha Healy (NUI, Cork), Peter Kratzer and Matthias Scheffler (FHI, Berlin), Eckhard Pelhke (University of Essen).
