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Quantum Monte Carlo Simulation of Nanoscale MgH2 Cluster Thermodynamics[1] ZHIGANG WU, Colorado School of Mines, MARK ALLENDORF, Sandia National Laboratory, JEFFREY GROSSMAN, Massachusetts Institute of Technology — We calculated the desorption energy of MgH_2 clusters using the quantum Monte Carlo (QMC) approach, which can provide desorption energies with chemical accuracy (within $\approx$ 1 kcal/mol) and therefore a valuable benchmark for such hydrogen-storage simulations. Compared with these QMC results, the widely used density-functional-theory (DFT) computations cannot reach a consistent and suitable level of accuracy across the thermodynamically tunable range for MgH_2 clusters, for a wide range of exchange-correlation functionals. Furthermore, our QMC calculations show that the DFT error depends substantially on cluster size. These results suggest that in simulating metal-hydride systems it is crucial to apply accurate methods that go beyond traditional mean-field approaches as a benchmark of their performance for a given material, and QMC is an appealing method for such a benchmark due to its high level of accuracy and favorable scaling (N^3) with number of electrons.

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