

From Crystal Structures to Rotational Excitations of Bound H₂ by Computer Simulations

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The low energy rotational excitations of hydrogen molecules bound on or in a solid are highly sensitive to the details of its interactions with the host material, and as such of great importance in areas such as hydrogen storage, proton transfer, catalysis of wide range of chemical reactions, etc. The most sensitive experimental probes of hydrogen, NMR and inelastic and quasielastic neutron scattering (INS) experiments provide useful data, but are difficult to interpret in great detail without theory. Simulations are challenging, owing to the need to treat quantum dynamical (QD) effects accurately, and the necessity to derive an accurate potential energy surface (PES) from crystallographic data for calculation of the rotational energy levels. Hydrogen molecules, however, were typically located by simulated annealing in our simulation. We have carried out an extensive series of 2D quantum rotation computational studies on H₂ in a number of MOF's employing a general purpose materials sorption potential along with explicit many-body polarization interactions, and more recently, for coordinated hydrogen in a metal complex using DFT to generate the PES. The results of our systematic spectroscopic and computational studies of H₂ in a large number of porous materials are in remarkable agreement, and provide detailed information on the effectiveness of hydrogen binding at different types of sites, and thereby could give direction for efforts in the synthesis of new materials with improved hydrogen interactions with hybrid porous materials.