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Spectroscopy and dynamics of H/Pd(111)

We aim at investigating the motion of adsorbates on metallic surfaces with entirely quantum calculations. We want to see what quantum phenomena are likely to occur during this motion, which can be related to diffusion. The Multiconfigurational Time Dependent Hartree (MCTDH) method will be used to obtain the quantum dynamics from first principle calculations. These calculations will help us to determine the intermediate scattering function (ISF) and the dynamical structure function (DSF). In the long term we want to see how our result can be used to extract observable quantities from the ISF and the DSF such as already seen in a previous project [1,2], with the aim to correctly interpret and rationalize the experimental data.

One of the systems to be studied is hydrogen on the palladium surface Pd(111).
The vibrational spectrum study of this system reveals a tunneling splitting of the energy levels starting from the very first excited states. This study also revealed the presence of a strong Fermi resonance between a state with one quantum of vibration in the perpendicular direction to the surface and another state with two quanta of vibrations parallel to the surface. These two purely quantum phenomena are expected to have an important impact on the dynamics and the diffusion process of hydrogen on the Palladium surface.

The vibrational spectrum of the H/Pd(111) is obtained theoretically using the block improved relaxation method of the MCTDH algorithm [3]. The interaction between the atoms of the system is modelled in the Born-Oppenheimer approximation by a potential energy surface (PES) calculated by Wei Dong and his coworkers [4].

During the 26th congress of the SFP, the results of the theoretical study of the vibrational spectrum of the H/Pd(111) system will be exposed as well as the time-dependent wave packet study of this system highlighting the impact of the tunneling splitting and the Fermi resonance on the dynamics of the H/Pd(111) system. A small section on the study of the H2/Pd(111) system will also be discussed as a continuation of the previous study.

References

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