PROBING REACTION DYNAMICS AT METAL SURFACES WITH H_2 DIFFRACTION

P. Nieto, D. Barredo, G. Laurent and Daniel Farías
Departamento de Física de la Materia Condensada
Universidad Autónoma de Madrid, 28049 Madrid, Spain

Studies of elementary collision processes of H_2 with metal surfaces can provide benchmark tests[1] of theoretical methods that are increasingly used to aid the design of new heterogeneous catalysts. Molecular beam and associative desorption experiments have been carried out to understand the main factors that govern H_2 dissociation at the surface. In addition, vibrationally inelastic and rotationally inelastic scattering experiments have provided useful information on how certain features of the potential energy surface (PES) control the experimental observations.

A different point of view is provided by diffraction experiments. H_2 diffraction from metal surfaces is more complex than He diffraction, since the PES is six-dimensional and the coupling with the dissociative adsorption channels comes into play[2]. Thus, H_2 diffraction is a very a promising technique to gauge the molecule-surface PES and dynamics. We have recently shown that this is possible by performing H_2 diffraction experiments on reactive Pd(111) and non reactive NiAl(110) surfaces at 70-150 meV. By comparing with six-dimensional quantum dynamics and classical trajectory calculations we showed for the first time that accurate diffraction patterns can be obtained from state-of-the-art PES based on density functional theory[3]. Once the PESs are validated, they can be used to study in detail the relationship between the trajectories followed by the H_2 molecules and the different channels involved in reactivity, like direct dissociation and dynamic trapping.

Finally, I will address the problem of the validity of the Born-Oppenheimer approximation for molecule-metal surface reactions, which has been recently questioned due to the possibility of electron-hole pair excitations[4]. We have performed experiments and six-dimensional quantum dynamics calculations on the scattering of molecular hydrogen from Pt(111), obtaining absolute diffraction probabilities. The comparison for in-plane and out-of-plane scattering, and results for dissociative chemisorption in the same system, show that for hydrogen-metal systems, reaction and diffractive scattering can be accurately described using the Born-Oppenheimer approximation[5].

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