Seminář odd. 26
Tenkých vrstev a nanostruktur

Fyzikální ústav AVČR, Cukrovarnická 10, Praha 6

datum: 15. 11. 2018 čtvrtek

čas: 15:00

místnost: knihovna, budova A, 1.p.

TÉMA

Non-adiabatic molecular dynamics at surfaces

Peter Saalfrank

Institut für Chemie, Universität Potsdam, Potsdam, Germany

The coupling of electrons of a (metal) surface to an adsorbate, or non-adiabatic effects in general, are often neglected in previous theoretical treatments of molecular dynamics of atoms or molecules adsorbed at surfaces. However, they can have a decisive influence on various phenomena, such the vibrational relaxation of the adsorbed species, inelastic molecule-surface scattering, and femtosecond-laser induced surface photochemistry. Fully quantum, multi-dimensional dynamical approaches are unfeasible in most cases and / or rely on far-going model assumptions. Recent years have seen the emergence of powerful tools based on classical dynamics which nevertheless account for non-adiabatic effects. One such approach is Molecular Dynamics with Electronic Friction (MDEF). When coupled with the Local Density Friction Approximation (LDFA), this method allows for parameter-free, multi-dimensional molecular dynamics with adsorbate / metal electron coupling, including laser-generated, “hot electrons” in surface photochemistry. Further, explicitly nonadiabatic dynamics beyond electronic friction models can be treated by including electronically excited and the coupling between them, leading to “surface hopping” schemes when realized in a classical dynamics context.

In the present contribution, we illustrate the methodologies and apply it to a few, recent examples from our work: Using Ab Initio Molecular Dynamics with Electronic Friction (AIMDEF) method with forces andfriction coefficients computed “on the fly”, we show that vibrational relaxation of H atoms on metal (Pb) films is entirely dominated by electron-hole pairs [1]. The same method is used to study the laser-induced, hot-electron mediated associative desorption of H2 molecules and isotopic variants from a Ru surface, exhibiting interesting isotope effects [2]. The femtosecond-laser induced desorption of molecular oxygen from a Ag(110) surface is studied also with MDEF, in this case with a precomputed (DFT) potential energy surface [3]. It is shown that, depending on the adsorption site of oxygen, the reaction can either be 'electron-' or 'phonon-' mediated [2]. Similarly, for CO molecules on Ru(0001) [4] or Cu(100) [5], femtosecond-laser stimulation leads to interesting dynamical phenomena very different from purely thermal processes. As an example of “explicitly non-adiabatic dynamics”, the photoisomerization of azobenzene molecules at surfaces is treated using a surface hopping scheme [6].

[1] P. Saalfrank, J.I. Juaristi, M. Alducin, M. Blanco-Rey, R. Diez Muino, J. Chem. Phys. 141, 234702 (2015).
[2] J.I. Iuaristi, M. Alducin, P. Saalfrank, Phys. Rev. B 95, 125439 (2017).
[3] I. Lončarić, M. Alducin, P. Saalfrank, J.I. Juaristi, Phys. Rev. B 93, 014301 (2016).
[4] R. Scholz, G. Floß, P. Saalfrank, G. Füchsel, I. Lončarić, J.I. Juaristi, Phys. Rev. B 94, 165447 (2016).
[5] R. Scholz, S. Lindner, I. Lončarić, P. Saalfrank, to be published.
[6] E. Titov, G. Granucci, M. Persico, J.P. Götze, P. Saalfrank, J. Phys. Chem. Lett. 7, 3591 (2016).

odborný garant: *Ing. Pavel Jelínek, Ph.D.*