

O 24: Methods: Molecular simulations and statistical mechanics

Time: Tuesday 15:00–16:15

Location: SCH A01

O 24.1 Tue 15:00 SCH A01

Optimal Control of Dissipative Wave Packet Dynamics — ●ERIK ASPLUND and THORSTEN KLÜNER — Institut für Reine und Angewandte Chemie, Carl von Ossietzky Universität Oldenburg, Germany

Light interacting with atoms and molecules is not only a source of information about the atoms and molecules studied, it can also initiate charge and energy transfer processes, i.e. chemical reactions. If excitation, excited state dynamics and relaxation all happen on the same time scale, a theoretical approach is required, which allows for a faithful description not relying on a separation of time scales. This can be done within the framework of the Surrogate Hamiltonian approach [1]. In this approach, a quantum system is separated into a primary system and a bath. The primary system is then explicitly immersed in the bath which acts as a dissipative environment. A theoretical tool for the design of laser pulses to transfer an initial state to a final state is optimal control theory (OCT) [2]. Besides the traditional final-time control algorithms, there exist methods to also handle time-dependent control targets [3,4]. To gain control of a dissipative system, time-dependent OCT is combined with the Surrogate Hamiltonian method. It is shown that an effective control of a dissipative system is possible.

References: [1] Roi Baer, and Ronnie Kosloff, *J. Chem. Phys.* 106, 8862 (1997). [2] Wusheng Zhu, Jair Botina, and Herschel Rabitz, *J. Chem. Phys.* 108, 1953 (1997). [3] Yuki Yoshi Ohtsuki, Gabriel Turinici, and Herschel Rabitz, *J. Chem. Phys.* 120, 5509 (2004). [4] I. Serban, J. Werschnik, and E. K. U. Gross, *Phys. Rev. A* 71, 053810 (2005).

O 24.2 Tue 15:15 SCH A01

First-principles study of the vibrational relaxation of the H stretching on Ge(100): Isotope effects — ●SUNG SAKONG and PETER KRATZER — Fachbereich Physik, Universität Duisburg-Essen, Duisburg, Germany

The vibrational energy of covalently bonded adsorbates is dissipated to several smaller energy quanta (adsorbate vibrations and phonons) on semiconductor surfaces, when the vibrational energy is smaller than the gap. Then, electronic dissipation is precluded and the relaxation proceeds through slower channels of the coupling between vibrations and vibration-phonon interaction. Within the weak vibration-phonon coupling regime, this slow relaxation process can be described by the first-order perturbation theory using a potential energy surface obtained from first-principles calculations (*J. Chem. Phys.* **129**, 174702 (2008)). Using this framework, we analyze the vibrational relaxation of the H-Ge stretching on Ge(100) surface. Because of the small energy of Ge phonons, the downward transitions from the H stretch mode require at least two phonons to satisfy energy conservation. Most interestingly, the relaxation of the H stretching becomes up to five times faster at a heterogeneously saturated dimer using H and D, as observed in SFG experiments. The weak interaction between H and D within a dimer opens relaxation channels that require only one phonon for the transition instead of two phonons. Transitions mediated by fewer phonons corresponds to lower order expansion terms of the vibrational potential, thus these channels systematically make a larger contributions to the relaxation process and lead to a faster vibrational lifetime.

O 24.3 Tue 15:30 SCH A01

Correlation energy of two-dimensional systems: toward non-empirical and universal modeling — ●S. PITTALIS^{1,3}, E. RASANEN^{2,3}, C. PROETTO^{1,3}, and E.K.U. GROSS^{1,3} — ¹Institut fuer Theoretische Physik, Freie Universitaet Berlin, Arnimallee 14, D-14195 Berlin, Germany — ²Nanoscience Center, Department of Physics, Uni-

versity of Jyväskylä, FIN-40014 Jyväskylä, Finland — ³European Theoretical Spectroscopy Facility (ETSF)

The capability of density-functional theory to deal with the ground-state of correlated systems depends on the accuracy of functionals developed for the exchange and correlation energies. Most of the approximations developed so far have focused on three-dimensional systems. Such efforts for two-dimensional (2D) systems have been relatively scarce despite the rapidly increasing experimental and theoretical interest in 2D structures such as semiconductor heterostructures, quantum-Hall systems, graphene, and various types of quantum dots. Here we derive a correlation-energy functional for 2D systems by modeling the correlation-hole functions in such a way that they satisfy a set of exact properties. We find very encouraging results for a wide range of correlation energies covering several few-electron quantum dots with and without external magnetic field, having different spin-polarizations and current densities.

O 24.4 Tue 15:45 SCH A01

Cluster expansion formalism for adsorbate-induced surface segregation — ●TOBIAS KERSCHER and STEFAN MÜLLER — Universität Erlangen-Nürnberg, Lehrstuhl für Theoretische Physik 2, Staudtstr. 7, D-91058 Erlangen

The segregation profile of alloy surfaces can strongly depend on the presence of adsorbates. We present the development of a model cluster expansion in the framework of the computer code UNCLE [1] to address adsorption on binary alloy surfaces. In principle, we realize this by the combination of two binary cluster expansions: both occupied and unoccupied adsorbate sites as well as the individual chemical species of the alloy are considered. Density functional calculations provide the energetics for the construction of the position-dependent interactions. Supported by Deutsche Forschungsgemeinschaft

[1] D. Lerch *et al*, to be submitted to *Mod. Simul. Mater. Sci. Eng.*

[2] S. Müller *et al*, *Appl. Phys. A* **82**, 415 (2006).

O 24.5 Tue 16:00 SCH A01

Liquid drop plus shell corrections model for deformed atomic cluster on the surface — ●VERONIKA DICK¹, DORIN POENARU^{1,2}, RADU GHERGHESCU^{1,2}, ILEANA PLONSKI^{1,2}, ANDREY LYALIN¹, ANDREY SOLOV'YOV¹, and WALTER GREINER¹ — ¹Frankfurt Institute for Advanced Studies (FIAS), J.W. Goethe Universität*, Ruth-Moufang-Str. 1, 60438 Frankfurt am Main, Germany — ²Horia Hulubei National Institute of Physics and Nuclear Engineering (IFIN-HH), PO Box MG-6, 077125 Bucharest-Magurele, Romania

In the present work we adapt a simple liquid drop model (LDM) for description of stability and shape deformation of atomic clusters deposited on a surface. We investigate the role of cluster-surface interaction in the plastic deformation of the clusters deposited on a surface. We present the atomic cluster on the substrate like an oblate spheroidal cap. Analytical relationships for the deformation-dependent liquid drop model energies of oblate, prolate semi-spheroidal atomic clusters and oblate spheroidal cap have been obtained.

A superdeformed prolate hemispheroid is the most stable hemispheroidal shape within LDM. It is also the shape with maximum degeneracy of quantum states of the hemispheroidal harmonic oscillator used to compute the shell and pairing corrections. The microscopic corrections as well as total deformation energy show parabolic valley and ridges of the potential energy surfaces in the plane (deformation, number of atoms). The ground state and isomeric state deformation of clusters of various sizes depends on the interplay between the minima of LDM and shell correction energies.