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 Building 99, Seminar Room I+II (EG)

G. Barratt Park

Institut für Physikalische Chemie, Universität Göttingen
Max-Planck-Institut für biophysikalische Chemie

Mode-specific and axis-specific mechanisms for energy exchange between polyatomic molecules and surfaces

The exchange of energy between molecules and surfaces plays a crucial role in heterogeneous catalysis. Molecular beam surface scattering experiments have substantially enhanced our understanding of energy exchange mechanisms. However, to date, almost all quantum state-resolved scattering experiments have focused on atoms or diatomic molecules and almost nothing is known about how the added degrees of freedom in polyatomic molecules influence dynamics at surfaces. I will discuss two results from ongoing experiments on formaldehyde scattering.

1. The conversion of translation to rotational motion often plays a major role in the trapping of small molecules at surfaces, a crucial first step for surface chemistry. We employ a new rotationally-resolved 1+1' resonance-enhanced multiphoton ionization (REMPI) scheme to measure the rotational distribution of formaldehyde molecules directly scattered from the Au(111) surface. The results indicate a pronounced propensity to excite *a*-axis rotation (twirling) rather than *b*- or *c*-axis rotation (tumbling or cartwheeling), and are consistent with a sterically induced rotational rainbow scattering model. The results suggest that *a*-axis rotation profoundly affects trapping probability at high incidence energies (>0.6 eV)

2. In recent decades, molecular beam scattering experiments from metal surfaces have demonstrated the importance of non-adiabatic coupling between electron hole pair excitation and molecular vibration (eHP-V coupling). Research on diatomic molecules suggests that the eHP-V mechanism involves transient electron transfer from the surface into the molecular LUMO. To examine this effect in polyatomic molecules, we have scattered formaldehyde, prepared in different vibrational levels of the metastable \tilde{a}^3A_2 electronic state from a low-work function Cs-covered surface and made quantitative measurement of the exo-electrons emitted from the surface. Preliminary results indicate that the efficiency of exo-electron generation from ν_4 (out-of-plane wag) is significantly greater than that of ν_2 (CO stretch), which is consistent with expectations for Franck-Condon overlap between the neutral \tilde{a}^3A_2 state and the ground electronic state of the formaldehyde anion.

