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Surface action spectroscopy with rare gas messenger atoms

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In model catalysis and general surface science studies the vibrational characterization of surfaces is usually performed with HREELS (high resolution electron energy loss spectroscopy) or IRAS (infrared reflection absorption spectroscopy). Both methods have disadvantages: HREELS suffers from its low resolution and (in the case of ionic samples) the presence of intense multiple phonon losses, while IRAS requires normalization of the spectrum with the spectrum of a reference sample, which leads to a contamination of the spectrum with features of the reference sample. Action spectroscopy with rare gas messenger atoms [A. Fielicke, et al. Phys. Rev. Lett. 93, 023401 (2004)] is a method for the vibrational spectroscopy of gas phase clusters and does not have these disadvantages. Rare gas atoms attached to gas phase clusters may desorb when the infrared radiation illuminating the clusters is in resonance with a cluster vibration. The fragmentation rate as monitored with a mass spectrometer represents a vibrational spectrum, which can be used to draw conclusions regarding the structure. Clusters, in this case deposited on surfaces, are also very relevant in model catalysis since they greatly influence catalytic reaction paths of supported catalysts. Both cluster shape and size matter. With this topic in mind we have constructed an apparatus for the application of action spectroscopy to solid surfaces and studied V2O3(0001)/Au(111) (~10 nm thick) and a TiO2(110) single crystal surface as first test systems. The machine is situated at the free electron laser of the Fritz Haber Institute which is able to provide intense and wide-range tunable infrared radiation. For V2O3(0001)/Au(111) the well-characterized vanadyl surface vibration and other surface vibrations could be detected, while bulk states turned out to be invisible to the method. We assume that anharmonic vibrational coupling between the primary excited vibration and the rare gas vs. surface vibration is the process leading to desorption, which explains the surface sensitivity of the method as well as its insensitivity to bulk vibrations. In the case of the TiO2(110) single crystal also rare gas desorption resulting from warming of the crystal by absorbed IR radiation could be observed. This led to new understanding regarding polaritons in rutile.

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