

Development of X-ray Emission Spectroscopy as a Tool in Catalysis

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The electronic states that give rise to the edge of an X-ray absorption spectrum are often resonantly excited states with a finite lifetime. The energy released in the decay process is carried either by an electron promoted into the continuum (resonant Auger effect) or by a photon (resonant X-ray scattering or RXS). Depending on the core level probed, transitions to different orbitals occurs with differing probabilities. The X-ray emission spectrum (XES) of a sample can be measured by scanning the energy of the fluorescence signal with a secondary spectrometer that is based on Bragg optics. In this case, the instrumental energy broadening is below the spectral line broadening due to the core hole lifetime. It is now possible to scan either the emission energy at fixed incident energy or, alternatively, the intensity of a single emission line as a function of incident energy. In the latter case, the core hole broadening observed in a 'normal' transmission or fluorescence detected absorption spectrum is replaced by a modulated lifetime broadening that is considerably sharper than the broadening due to the core hole life time. The pre-edge features present in the XANES of 3d transition metals become better separated from the main edge and are thus much more accessible to a detailed analysis. An even more dramatic sharpening effect is observed in the XANES of (4f and) 5d transition metals when this technique is applied. High energy resolution XANES data have been obtained on Au and Pt nanoparticles and provided detailed insights on reactant adsorption sites as well as on reaction mechanisms, as will be shown in this presentation. An outlook into the further planned developments of RXS for applications in homogeneous and heterogeneous catalysis will be presented.