MAR11-2010-001106

Abstract for an Invited Paper
for the MAR11 Meeting of
the American Physical Society

Modeling the interactions of adsorbates with each other and with metal surfaces

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The interactions of molecules with metallic surfaces are fundamental to the ability of metals to catalyze reactions. One often thinks of a metal like platinum as the catalyst, but under reaction conditions the reactivity of the metal surfaces is modified by the molecules that adsorb on them. We have used quantum chemical calculations in conjunction with cluster expansions to probe the adsorption behavior of atomic adsorbates such as C, N, O, and S on late transition metal surfaces such as Rh, Ir, Pd, Pt, Cu, Ag, and Au(111). There are remarkable similarities in the adsorption behavior of these adsorbates that can be interpreted in terms of a simple adsorbate-induced surface electronic structure modification mechanism that is common to all the adsorbates and surfaces. The variations between the adsorbates and metals are readily explained in terms of the size of the metal and adsorbate orbitals and the geometry dependent overlap of these orbitals. We have constructed a new Solid State Table of these orbital radii from the quantum chemical calculations that can be used in conjunction with a simple model to rapidly estimate the electronic structure of metal and alloy surfaces with adsorbates on them.