Ab-initio Studies of the Adsorption of H on the Ir(100)-1x1 and -5x1-hex Surface

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The ground state of the Ir(100) surface is the 5x1 quasi-hexagonally reconstructed phase Ir(199)-5x1-hex. This is known to reconstruct to another 5x1 phase when exposed to hydrogen. It consists of atomically thin Ir wires arranged in, on average, 5-fold long-range order on the unreconstructed substrate [1]. Besides these stable 5x1 phases, Ir is known to have a metastable 1x1 phase which can be observed up to around 480°C [2,3].

As a first part of our studies, the adsorption of H on this metastable 1x1 phase was elucidated via first-principle calculations using density functional theory (DFT) via the Vienna Ab-initio Simulation Package (VASP [4]). These calculations yield total energies as well as vibrational and electronic properties. The results show that more than 1eV per H atom is necessary to occupy either tetrahedron or octahedron sites below the surface, i.e. the presence of subsurface hydrogen is very unlikely which is in line with experimental thermal desorption spectra. As a consequence, we focus on the site- and concentration dependence of the adsorption energy of H on the Ir(100) top layer. Our DFT calculations (which include vibrational zero-point energies) indicate that for all considered H-concentrations (0.25 – 2 monolayers) the bridge position is the energetically favoured adsorption site. At 1 monolayer coverage an adsorption energy of 610 meV per H-atom results. The energetic hierarchy of the H sites considered is bridge, top and hollow position with the latter being less favoured by 160 meV and 353 meV, respectively (Fig. 1).

These unexpected results for the adsorption site are discussed with respect to the electronic properties of the Ir(100)-1x1 surface.

As a second part we focused on the 5x1 phase, in particular on the H induced surface reconstructuring. The parameters of the reconstructed 5x1-hex phase resulting from DFT calculations agree well with experimental data. Analogous to the 1x1 phase, favoured adsorption sites of H will be discussed along with electronic properties.

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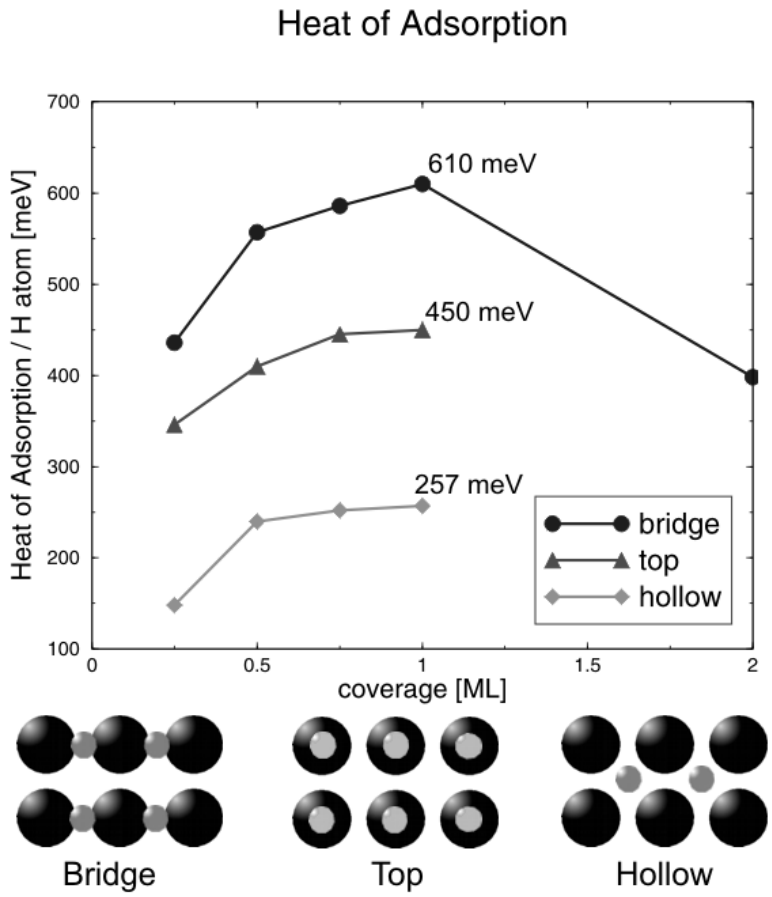

Figure 1: Heat of adsorption for considered adsorption sites