Hydrogen Incorporation Into Palladium Ultra-Thin-Films By Low Energy Electron Microscopy

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The ability of Palladium to store hydrogen is well known. Nevertheless surface science studies of the exposure of Pd(111) samples to hydrogen have found only a dense hydrogen layer betwen the last palladium-palladium layers, with additional hydrogen going into the bulk as a solid solution.[1,2] On Pd films a few atomic layers thick on a substrate which does not accept substantial quantites of hydrogen such as Ru, it should be possible to saturate completely the Pd film, and thus study the formation of PdH hydride with surface science techniques. In particular, as there is a substantial lattice mismatch betwen Pd and the beta-PdH hydride, the incorporation of H in Pd-intersticial places should induce a significant expansion of the Pd layers. Although H itself is basically invisible to LEED, the change of the Pd-Pd distances should be easily resolved[3]. In this work, we present a low energy electron microscopy study of the hydrogren exposure of thin films of Pd on Ru(0001) and W(110). We characterize and study the growth and structure of Pd thin films. LEEM is a useful technique to monitor and control the growth of thin films in real space. Furthermore, a LEEM microscope can be used to acquire selected area diffraction (SAD-LEED) to determine the structure of such film.[4]

Our experiments reveals that Pd films 2 to 6 ML thick on Ru(0001) follow the fcc sequence with the Ru in-plane lattice spacing, and present a corresponding expansion in the out-of-plane lattice spacing. Thicker films on Ru(0001) relax to a bulk-like Pd in-plane spacing. Both thick films on Ru(0001) or W(110) show a slight expansion of the last interlayer spacing, in line with reported studies on the surface of bulk Pd. We follow the dosing of atomic H on the Pd films by means of real-time LEED IV curves acquired by LEEM. Figure 1 shows low energy reflectivity curves, this technique can be used as a fingerprint for identification of surfaces changes for example for determination of adatom densities[5] or local alloy concentration[6]; aroung 22eV the first Bragg peak is located. During the dose of molecular hydrogen the position of that peak is always in the same indicating that there is no structural change. The curves only show a small change around 8-10 eV which is expected to be due to hydrogen adsorption at the surface.

On the other hand if atomic hydrogen is dosed, we can see a diplacement of the Bragg peak to lower energies (Figure2). This shift to lower energies corresponds to an expansion of the Pd layers. To quantify this expansion LEED IV fits were performed, obtaining a expansion only in the last layer, but this expansion is smaller that the one expected for a full PdH beta-hydride formation. A similar result was obtained for films with the Ru in plane lattice paramter. The expansion of the last layer is observed, as shown in Figure2 and also the feature adscribed to hydrogen absorbed on the Pd surface is reversed by heating the sample.

This research was supported by the Office of Basic Energy Sciences, Division of Materials Sciences, U.S. Department of Energy under contract No. DE-AC04-94AL8500, and the Spanish Ministry of Science and Technology Through Project No. MAT2006-13149-C02-02.

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Figures:

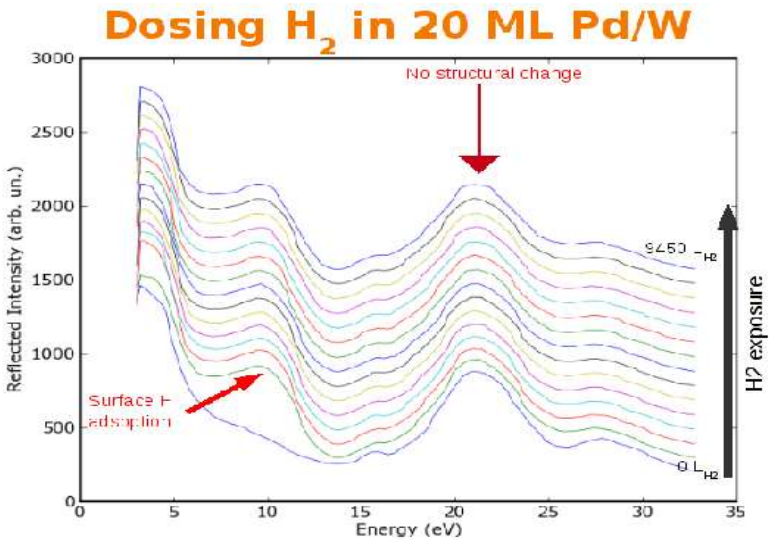

Figure1: Low energy reflectivity curves while dosing molecular hydrogen on 20 ML Pd films

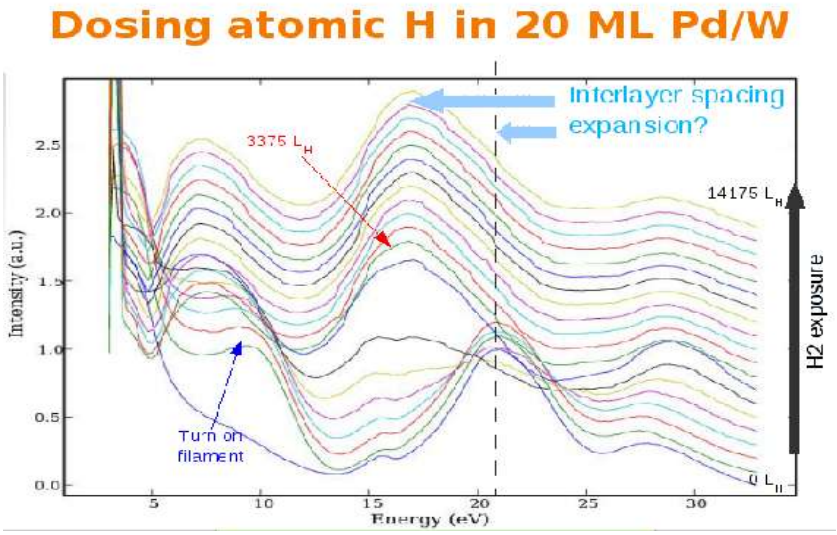

Figure2: Low energy reflectivity curves while dosing atomic hydrogen on 20 ML Pd films