

Quantum stabilization of atomically flat metal nanofilms

F. Calleja¹, J.J. Hinarejos¹, M.G.C.Passeggi², J. Camarero¹, A.L. Vázquez de Parga¹ and R. Miranda¹

¹Departamento de Física de la Materia Condensada,
Universidad Autónoma de Madrid. Cantoblanco. 28049 Madrid, Spain

²Grupo de Física de Superficies, INTEC, Güemes 3450, 3000, Santa Fe, Argentina

The search for methods to produce structures highly organized at the atomic scale during epitaxy is an active research area since it holds the promise for basic discoveries and novel technological applications. Along with the well known kinetic and thermodynamic parameters, it was recently found that Quantum Size Effects can play a crucial role during epitaxial growth [1-3]. For instance, metallic islands of uniform height with steep edges and flat tops, can be grown on both semiconductor [2] and metal surfaces [3,4]. In both cases, potential barriers at the interfaces confine the electrons within the metal overlayers in the direction perpendicular to the layers.

The energy associated to these Quantum Well States (QWS) might be large enough to stabilize preferentially certain shapes. We reported previously that the equilibrium height distribution of Pb nanoislands grown at elevated temperature on Cu(111), as obtained from STM images, showed "magic" heights, i.e. certain heights appeared much more frequently than others [3]. The QWS dictate the equilibrium shape of Pb nanocrystals by eliminating from the height distribution those nanodots whose heights would have a QWS at the Fermi level [3]. This effect is now used to stabilize at 300 K films of Pb with specific heights, which were deposited at low temperatures as flat films.

The experiments have been performed in a UHV system equipped with LEED/AES and a variable temperature Scanning Tunneling Microscope (STM). The Cu(111) surface was cleaned by standard methods including ion sputtering and annealing. After cleaning, the sample displayed a sharp LEED pattern and atomically resolved high-quality STM images. Pb was evaporated from a Knudsen cell with the sample placed in the STM stage. The flux of Pb atoms, i.e. the deposition rate, was kept constant and the temperature of the Cu(111) substrate during and after the deposition was varied from 70 K up to 300 K. This *in-situ* experiment allows the direct observation of structural transformations on an atomic scale, as well as control and elucidation of the dynamic behavior as a function of sample temperature.

Pb grows on Cu(111) at 300K in the Stranski-Krastanov mode of growth. After the completion of a single compact monolayer, 3D (111)-oriented Pb islands grow on the surface. Fig 1 shows a representative image of a Pb film deposited at 300 K. The surface is covered with 3D islands, most of them of similar height. Only the islands nucleated at step bunches are significantly higher. The inset shows the atomically resolved (4x4) wetting monolayer of Pb that exists between the islands. Tunneling Spectroscopy performed by placing the STM tip on top of islands of different heights show the QWS corresponding to each thickness [3] as can be seen in Fig 2.

Growth of Pb at 70 K, on the contrary, occurs layer-by-layer. Films of Pb of different thicknesses have been deposited and imaged at 65 K. STM at variable temperature is used to follow the evolution of the films morphology during annealing. The upper panel of Figure 3 shows the morphology of a film containing 7.8 ML as deposited at 70 K. The 7th layer is complete and a large fraction of the surface is covered with dendritic islands of the 8th layer. On some of the largest dendritic islands small nuclei of layer number 9 can also be seen.

Calculations of the total energy [5] of Pb films on Cu(111) as a function of the thickness of Pb, identify the magic heights in agreement with the experimentally observed values for nanodots [3]. Since the calculation was carried out for laterally extended films we explore now its validity to predict the relative stability of the different thicknesses. We find that each Pb layer becomes unstable at a different temperature. The layers with a non magic height (i.e. 7 ML), which are atomically flat at low temperatures because they are trapped by kinetic constraints, decompose into more stable heights (i.e. 6 and 8 ML-high islands). The layers whose thickness correspond to magic heights are more stable. In the case of some particularly stable thickness, such as 8 ML, an atomically flat film of Pb could be stabilized up to room temperature, as shown in the lower panel of Fig 3. Thus, quantum size effects have been used to stabilize at 300 K flat films of Pb that, when deposited directly at 300 K would produce 3D islands.

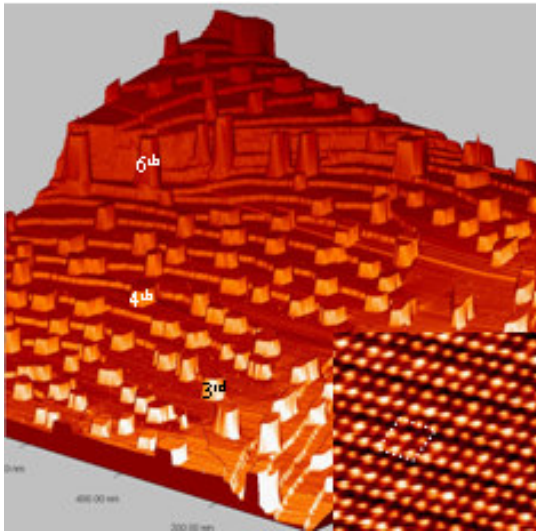


Figure 1 1 μm x 1 μm STM image of Pb nanocrystals grown at 300 K on Cu(111). The total amount deposited is 2 ML. The inset shows the (4x4) monolayer of Pb that wets uniformly the Cu surface.

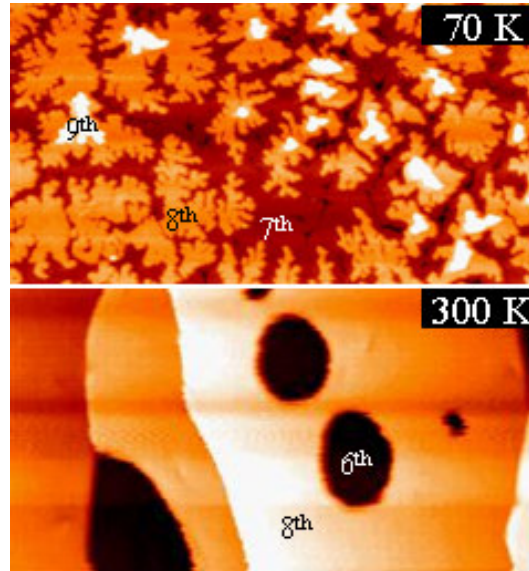


Figure 3 *Top*: 200 nm x 100 nm STM image of 7.8 ML Pb grown on Cu(111) at 70 K. *Bottom*: 200 nm x 100 nm STM image after heat the sample at 300 K.

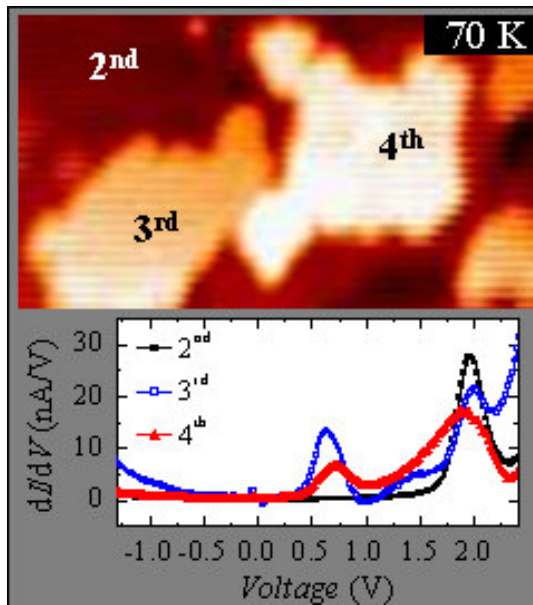


Figure 2 *Top*: 200 nm x 100 nm STM spectroscopic image of a 3.3 ML thick Pb film grown on Cu(111) at 70 K. The dc voltage is 0.6 V. *Bottom*: Local tunnelling conductance versus simple voltage recorded at constant height above the areas with different Pb layer height in the image above.

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