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The origin of faceting of ultraflat gold films epitaxially grown on mica

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Abstract

A two-step deposition process for the formation of atomically flat gold films on mica is developed. The process starts with a high deposition rate followed by a 1:100 reduced rate. Using this combination a completely wetting of mica by gold resulting in a two-dimensional growth mode and finally extremely flat gold films with large terraces are achieved. Additionally hexagonal faceting of the gold films on mica is observed at moderate temperatures which can be related to the relaxation of stress caused by different thermal coefficients of expansion of mica and gold. The stress release leads to the generation of misfit dislocations that glide along the (1 0 0) planes producing facets on the surface. Annealing experiments in a UHV-STM and thermogravimetry point to the inset of intensified hexagonal faceting due to the starting decomposition of mica at elevated temperatures.

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1. Introduction

Gold surfaces are the most suitable substrates for the formation of self-assembled monolayers due to their affinity to thiol groups of organic molecules [1]. A prerequisite for the formation of self assembled monolayers are atomically flat gold films. High quality smooth gold films can be obtained by vacuum deposition at low deposition rates onto freshly cleaved mica [2]. The influence of different parameters like deposition temperature [2–4], deposition rate [4] or annealing temperatures [5] on the morphology of gold films has been investigated in literature. In some publications, flame annealing is employed to increase the grain size and remove surface contaminants [6]. However, in most cases three-dimensional growth of gold films is obtained, while for the examination of SAMs by STM two-dimensional layer-by-layer growth

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with large atomically flat terraces is favourable. Furthermore, in some publications the films exhibit hexagonal faceting, i.e linear step edges along the three equivalent $\langle 0\,1\,\bar{1}\rangle$ directions, whereas the origin of the facets is not understood. In the following a two-step process is presented, by which extremely flat gold films with large terraces can be obtained, and a mechanism for the formation of hexagonal facets is proposed.

2. Experimental details

In this work 200 nm thick gold films are epitaxially grown by two different deposition procedures: a single step process with a constant deposition rate (0.1 nm/s) and a two-step deposition process, where a 150 nm gold film is deposited at a high rate (5 nm/s) followed by a 50 nm gold film at a considerably reduced rate (0.05 nm/s). The films are prepared in an e-gun evaporation chamber at a base pressure of 10^{-7} mbar. Prior to deposition, mica (Plano GmbH, Glimmer ULTRACLEAN, Art. Nr G250-7) is freshly cleaved and then heated in vacuum for 6 h at the deposition temperature (460 °C for the single step process, 400 °C for the two-step process). Following the deposition the films are annealed for 1 h at the deposition temperature. The films are characterised by AFM, STM (JEOL JSPM 5200, JEOL JSPM 4500), XRD and thermogravimetry. XRD reveals the (1 1 1) orientation of the film.

3. Results and discussions

The single step deposition process results in a film of many small (1–2 μm) grains with a hexagonal shape (see Fig. 1). Some grains seem to coalesce, but this process is not very pronounced and most hills are isolated from their neighbours so that deep trenches are formed between some grains. The depth of these trenches comes into the range of the overall thickness of the gold layer. These observations are in accordance with the measurement of only a very poor, if any, lateral conductivity.

Interestingly, Höpfner et al. [7], who deposited 150 nm of gold at a higher rate (1 nm/s) on mica of the same manufacture, also stated that deep holes are formed between two gold grains. However, these holes

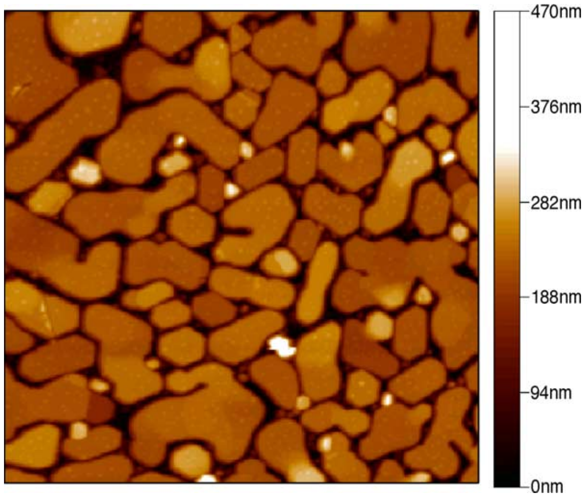

Fig. 1. AFM scan (10 μm × 10 μm) of gold on mica deposited in the single step process at 460 °C.

are not as deep as the gold layer, i.e. the gold film is continuous. This suggests that these ultraclean mica surfaces exhibit a high interface energy between mica and gold, which results in poor wetting of mica and three-dimensional growth of gold islands without lateral conductivity.

It has been shown in principle, that a two-step process can be used to increase the crystallite size of gold films on mica [7]. Using the two-step deposition process introduced here the growth of the gold film can be changed from island to two-dimensional growth due to the improved wetting forced by the higher deposition rate in the first step (see Fig. 2). The extremely low deposition rate in the second step causes a flattening of the surfaces by filling up the remaining trenches of the first step. In this two-step process Ostwald's law of stages has been applied which means that precipitation in supersaturated conditions does not lead to thermodynamically stable phases but to less stable states if the formation of solid proceeds sufficiently quickly. These less stable states convert to more stable crystalline forms by subsequent treatment in thermodynamic equilibrium conditions. The resulting gold films exhibit a very low roughness on a large surface area (0.4 nm r.m.s. on 5 μm × 5 μm). A roughness in this range has only been reached by template-stripped gold films obtained by Wagner and co-workers [9,10], which are prepared