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Visualization of steps and surface reconstructions in Helium Ion Microscopy with atomic precision

Gregor Hlawacek *[,1], Maciej Jankowski [2,3], Herbert Wormeester [4], Raoul van Gastel [4], Harold J.W. Zandvliet [4], Bene Poelsema [4]

Physics of Interfaces and Nanomaterials, University of Twente, PO Box 217, 7500 AE Enschede, The Netherlands

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ABSTRACT

Helium Ion Microscopy is known for its surface sensitivity and high lateral resolution. Here, we present results of a Helium Ion Microscopy based investigation of a surface confined alloy of Ag on Pt(111). Based on a change of the work function of 25 meV across the atomically flat terraces we can distinguish Pt rich from Pt poor areas and visualize the single atomic layer high steps between the terraces. Furthermore, dechanneling contrast has been utilized to measure the periodicity of the hcp/fcc pattern formed in the 2–3 layers thick Ag/Pt alloy film. A periodicity of 6.65 nm along the $\langle\bar{1}12\rangle$ surface direction has been measured. In terms of crystallography a hcp domain is obtained through a lateral displacement of a part of the outermost layer by $1/\sqrt{3}$ of a nearest neighbor spacing along $\langle\bar{1}12\rangle$. This periodicity is measured with atomic precision: coincidence between the Ag and the Pt lattices is observed for 23 Ag atoms on 24 Pt atoms. The findings are perfectly in line with results obtained with Low Energy Electron Microscopy and Phase Contrast Atomic Force Microscopy.

1. Introduction

Helium Ion Microscopy (HIM) [1,2] has become a powerful imaging tool with very high lateral resolution and surface sensitivity. With a lateral resolution better than 0.5 nm [3] one would hope that also information on the lattice structure of a sample surface can be obtained using this method. So far this has not been demonstrated. However, results on several atoms high steps and surface termination of Ti_3SiC_2 [4] show that this goal is not out of reach. In addition, theoretical reports indicate that imaging of the atomic structure should in principle be possible for very thin layers [5]. In a real experiment one would have to care about vacuum levels to minimize hydrocarbon contamination and—in particular on thick samples—damage by the recoiling substrate atoms as well as the implanted Helium. So far this has prevented the observation of features related to the atomic structure of the sample surface.

Here, we present—to the best of our knowledge—the first observation of single atom high steps with a Helium Ion Microscope. In addition we will demonstrate that under specific conditions the HIM is able to distinguish areas in which a small number of atoms have been moved from the bulk lattice position by a fraction of an interatomic spacing.

Since its introduction by Ward et al. [6] Helium Ion Microscopy (HIM) has become an important microscopy technique providing high resolution images of sample surfaces. This is true for conducting as well as insulating materials. The present work is based on the well known image formation mechanisms in HIM which utilize secondary electrons excited by the primary ion and ejected from the sample surface [7–9]. It relies on the high surface sensitivity [10] of the tool and the fact that channeling can be exploited to enhance the imaging of thin surface layers [11].

Low Energy Electron Microscopy (LEEM) [12] and Atomic Force Microscopy (AFM)—in particular phase contrast AFM [13–15]—have been used to benchmark our findings.

The Ag/Pt(111) system is a representative example of a surface confined alloy, which is widely studied in the field of surface

* Corresponding author. Present address: Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, Bautzner Landstr. 400, 01328 Dresden, Germany

E-mail address: g.hlawacek@hzdr.de (G. Hlawacek).

[1] Acquired and analyzed all HIM based data, AFM analysis, and wrote the manuscript.

[2] Present address: ID03 Surface Diffraction Beamline, ESRF – The European Synchrotron, 71 Avenue des Martyrs, 38400 Grenoble, France.

[3] Acquired and analyzed all LEEM based data.

[4] Helped in interpreting the results and ensured financial support.

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science [16–21]. Deposition under UHV conditions of a 2–3 layers[5] thick Ag film on Pt(111) at room temperature followed by an annealing step above 550 K results in irreversible changes of the surface morphology [16]. These changes were identified by Brune et al. [22] using Scanning Tunneling Microscopy (STM) as the formation of a well-ordered periodic dislocation network, formed through material intermixing in the first two layers of the deposited film [23,24]. The symmetry, periodicity and level of ordering of the network depends on the magnitude of material intermixing [25] which is controlled mainly by the substrate temperature during deposition. The structural model of the network formed at 800 K was revised recently by Aït-Mansour et al. [24]. According to their model, the deposition of a 2 layer thick Ag film on Pt(111) followed by annealing at 800 K leads to the exchange of atoms between Pt(111) and the deposited Ag layer. The Pt interface layer contains Ag inclusions, whereas the expelled Pt atoms from the substrate form inclusions in the top layer of the alloy. The mixing of the atoms is limited only to fcc-stacking sites. Stacking faults are present at the Pt interface layer and the formed network has a 3-fold symmetry. Further deposition of Ag leads to the growth of a third layer which was reported to be purely silver and hexagonal in structure [24]. The formed dislocation network at the alloy-Pt(111) interface causes periodical undulations of the third and subsequent Ag layers [25].

2. Experimental

Helium Ion Microscopy has been performed in an ultra high vacuum (UHV) HIM [26,27] Orion$^+$ from Carl Zeiss Microscopy. The system has a base pressure of 2×10^{-9} mbar. This pressure is reached thanks to a stainless steel sample chamber with Conflat type flanges, a modified pumping and load lock strategy, a 5000 l/s titanium sublimation pump and a differentially pumped door gasket. The system is equipped with a standard Everhardt Thornley Detector to record secondary electron (SE) based images. In addition, detectors to count back scattered helium (BSHe) [11,28] measure their energy [29,30] and collect photons to enable ionoluminescence [31,32] studies of materials are present. The presented images were recorded using a sample tilt of 35° to exploit channeling into the underlying bulk crystal. This is necessary to maximize the surface contrast [10].

The samples were prepared and initially characterized in a Low Energy Electron Microscope (LEEM) Elmitec III. The used Pt(111) crystal had a miscut angle of less than 0.1°. Surface cleaning was done by prolonged repetitive cycles of argon ion bombardment, annealing in 2×10^{-7} mbar of oxygen at 800 K, and subsequent flashing to 1300 K.

High purity silver (99.995%) was deposited from a molybdenum crucible mounted in an electron beam evaporator (Omicron EFM-3). The growth of the silver layers was tracked in situ and real time using bright-field mode [33,34] in LEEM.

Venting and pumping of the vacuum systems has been timed to minimize contamination of the sample surface while transporting it from the LEEM to the HIM vacuum chamber. However, adsorption of hydrocarbons on the film surface is most likely and could not be prevented. Since oxidation is strongly suppressed by the presence of silver [21] oxygen is considered a lesser problem. The sample has been imaged in the LEEM after the HIM analysis has been completed. Comparing the results to data recorded before the sample transport showed no significant changes relevant for the present study.

[5] Coverage is given in monolayer equivalents based on the Pt(111) surface unit cell.

The AFM measurements were done under ambient conditions with an Agilent 5100 AFM employing amplitude-modulation to record the topography. A MikroMasch Al back-coated NSC35 Si_3N_4 cantilever with a tip radius of 8 nm was used in these measurements. The resonance frequency of this cantilever type is 205 kHz and the nominal spring constant is 8.9 N/m. For the measurements an amplitude set-point of 90% was used and the oscillation amplitude was in the range between 30 nm and 40 nm.

3. Results

3.1. Surface mounds and HIM sample alignment

In Fig. 1 low magnification images of the Ag/Pt(111) sample surface recorded by LEEM and HIM are shown. The clean Pt(111) is occasionally decorated by mounds which sometimes originate from screw dislocations. These mounds are present on the clean Pt (111) surface and are neither formed nor affected by the subsequent Ag deposition. Representative examples of such surface mounds are presented in Fig. 1. These mounds occur after annealing and a relatively swift cooling due to surface-bulk mass exchange [35]. The dark lines (a few marked by yellow arrows) in Fig. 1(A) and (B) are steps separating atomically flat terraces. Mounds and step bunches are typically accompanied by wide terraces like in the top half of the image in Fig. 1(A): Step migration during the annealing stage is suppressed by the mounds giving rise to step bunching and, at the same time, to wider terraces away from the obstruction caused by the mound. The mounds are threefold symmetric as it becomes evident by looking at the relative lengths of the edge segments: opposite sides have different lengths, in other words mirror symmetry is absent. Moreover, adjacent facets of the mound have different brightnesses as is seen clearly in Fig. 1(A) and (B). Unfortunately, field distortions [36] prevent a full structural characterization of the mounds by LEEM. Still we can extract important information. Steps along $\langle\bar{1}10\rangle$ azimuthal directions on fcc (111) surfaces are different. They either have (111) or (001) type microfacets of which the former are energetically favored [37]. The longer mound edges on opposing sides are apparently energetically favored and from that fact we can derive that the brighter sides of the mounds are of the $[\bar{1}10]$–(111) (oriented microfacets) type, while the darker sides in between are of the $[\bar{1}10]$–(100) type. The threefold symmetry allows us to determine fully the orientation of the mounds in a field free situation such as in HIM. The curved dark line on top of the mound represents a monatomic step (marked by yellow arrows in Fig. 1(B)) as we conclude from the clear correspondence with the data in Ref. [35] and references therein.

The appearance of such mounds in HIM can be seen from Fig. 1 (C) and (D). A low magnification image of a mound is presented in Fig. 1(C). Note that there is no defined correspondence of the azimuth directions in the LEEM and the HIM data. A higher magnification of the mound sidewall is shown in Fig. 1(D). Using HIM images as the one presented in Fig. 1(C) and (D) the orientation of the sample with respect to the beam can be determined. While in the center right part of Fig. 1(D) straight step bunches are visible (terminated by {001} facets), irregular and curved step bunches are visible at the top and bottom of the image. The straight step bunches are preferential oriented parallel to the $\langle\bar{1}10\rangle$ directions of the Pt(111) sample surface. Using these step bunches the sample has been aligned with the He^+ beam parallel to the $[\bar{1}\bar{1}0]$ direction and parallel to the Pt(001) plane. The direction of the arrows in Fig. 1(C) corresponds to the surface projection ($\langle\bar{1}\bar{1}2\rangle$) of the surface normals of the less abundant {001} facets. Obviously the dominant {111} oriented step facets are situated just in between. In this channeling condition the