Surface Science 606 (2012) 702–710

Contents lists available at SciVerse ScienceDirect

Surface Science

journal homepage: www.elsevier.com/locate/susc

Nucleation and growth mechanisms of Fe on Au(111) in the sub-monolayer regime

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ARTICLE INFO

Article history:
Received 7 October 2011
Accepted 12 December 2011
Available online 19 December 2011

Keywords:
Scanning Tunneling Microscopy
Fe/Au(111)
Cluster nucleation and growth
Nanoislands
Rate equation models

ABSTRACT

The growth of Fe on Au(111) at 300 K in the sub-monolayer regime has been investigated using scanning tunneling microscopy, focusing on the mechanisms of nucleation, coalescence and interlayer diffusion. Below a coverage of 0.1 ML, Fe growth proceeds in a well-ordered fashion producing regular arrays of islands, while approaching the island coalescence threshold (above 0.35–0.4 ML), we observed a consistent increasing of random island nucleation. These observations have been interpreted through rate equation models for the island densities in the presence of preferred nucleation sites. The evolution of the second layer fraction has also been interpreted in a rate equation scheme. Our results show that the ordered to random growth transition can be explained by including in the model bond breaking mechanisms due to finite Fe–Fe bond energy. A moderate interlayer diffusion has been inferred from data analysis between the second and the first layer, which has been used to estimate the energy barrier of the adatoms descending process.

1. Introduction

The possibility of growing ordered arrays of nanometer sized clusters supported on surfaces is of particular interest for a wide range of scientific and technological fields [1]. In the field of data storage technology, self-assembled arrays of molecules and clusters containing magnetic elements are currently under intensive investigation since they represent model systems for investigating magnetic properties at the nanoscale and thus engineering novel materials to overcome the actual limit of information density [2]. With the aim of obtaining a dense and well-ordered ensemble of clusters, it is important to understand which mechanisms lead to the formation of cluster arrays and, more important, which mechanisms could destroy an eventual ordered pattern. Moreover, as the magnetic stability increases with the clusters size, it is important to understand how to increase the cluster volume without increasing their lateral dimension over the threshold of coalescence, e.g. by favoring a vertical growth instead of a layer-by-layer one.

The Au(111) surface represents an interesting and peculiar highly polarizable substrate for growing magnetic clusters and thin films. Due to its herringbone reconstruction [3], it favors cluster growth in a well-ordered fashion [4] and it represents an interesting case of study for cluster growth and organization. It was shown that Ni [4], Co [5] and Fe [6,7] start to grow in regular arrays of clusters when the coverage is below 0.3–0.6 ML. The strain induced by the herringbone reconstruction plays also a fundamental role in determining the evolution of the growth when clusters start to coalesce and to form a continuous film. Depending on the deposited materials, different growth regimes were observed with a consequent influence on the associated magnetic properties. In particular, Fe clusters on Au(111) show a strong interplay between electronic, magnetic and structural properties [8–11]. It was found that, in the sub-monolayer regime, Fe on Au (111) grows in an epitaxial fcc phase nucleating in well ordered arrays of monolayer islands of triangular or diamond-like shapes [6,7]. However, further deposition of Fe leads to an increase of the islands size, to the nucleation of new islands in a random fashion and to the coalescence of the grown islands. Moreover, a complete layer-by-layer growth is not favored when the islands extend outside the nucleation sites and the second layer starts growing well before the first is completed [6,7,12,13]. Magnetic properties closely follow the structural changes of Fe islands and ultra-thin films [10,11,14–16]. For Fe coverages below 0.3 ML, the magnetization of Fe islands was found to be in-plane [10] while it was observed to turn into the out-of-plane direction for Fe coverages in the 0.4–2.8 ML range [11]. It follows that investigating the Fe/Au(111) morphology evolution is crucial to shed light on the mechanisms which drive the changes in the physical properties. Although a lot of effort has been put in understanding the mechanisms leading to ordered nucleation on Au(111) in the first stage of growth, it is still not clear how to interpret the morphology evolution of Fe islands at further coverages, which is indeed related with the magnetic transitions of the system.

In particular, three relevant points have still to be faced: a) a precise identification of the coalescence threshold among Fe islands, b) an explanation for the transition between ordered to random nucleation

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doi:10.1016/j.susc.2011.12.008

and c) an estimation of the interlayer diffusion which leads the successive layers evolution.

In this paper, we report on a Scanning Tunneling Microscopy (STM) investigation of the growth mechanisms for the Fe on Au(111) surface occurring in the sub-monolayer regime. Through a statistical analysis of the morphological properties of the system, we identified the coalescence threshold among Fe islands. The presence of on-elbows and out-of-elbows islands has been quantified as a function of Fe coverage and interpreted through the use of Rate Equation (RE) models. This well established approach provides a good description of average quantities such as the island density and average size, as already proved for a large number of physical situations [17,18], and is also applicable to nucleations at point defects [19] and in the presence of a dense net of point "traps," such as for the Au(111) vicinal surfaces [20–22]. We apply a RE model also for interpreting the second layer growth evolution and obtain an estimation of the Schwoebel barrier for the interlayer descending mechanism.

2. Experimental details

An Omicron VT-SPM apparatus was used for preparing and measuring the surface. The Au(111) surface (a commercial substrate of evaporated gold on mica) was prepared in UHV by 15 min Ar^+ sputtering at 1 keV at a sample temperature of 800 K, which was kept for at least 10 min after the sputtering. After several cycles, a clean reconstructed surface was obtained with a low level of contaminations. Fe was deposited on a clean Au(111) surface at room temperature (RT) in UHV (5×10^{-11} mbar) by means of an electron-beam evaporation source (Fe wire purity 99.99%). The deposition rate (about 0.2 ML/min) was controlled by monitoring the ion flux, which is proportional to the flux of evaporated atoms. Morphological properties were studied by means of constant current STM images taken with the sample and the tip kept at room temperature (RT). Coverage ranging from 0.07 ML up to 0.7 ML was estimated by a software analysis of the STM images. In the text, we refer to coverage as the total amount of deposited Fe considering both the contribution of the first and the second Fe layers.

3. Results

The Au (111) surface shows a uniaxial reconstruction along the $<1\bar{1}0>$ directions, alternating fcc and hcp regions separated by discommensuration lines (DLs) running along the $<11\bar{2}>$ directions [3] (see Fig. 1a). Atoms surrounding those lines produce a peculiar corrugation of the surface called ridge. At larger scales, the uniaxial deformation is however not favored and the surface forms domains with a rotation of ±120 degrees (Fig. 1b).

The ridges of the superstructure are forced to bend at the domain boundaries, forming a periodic zig-zag structure with a period of about 15 nm. As a consequence, characteristic sites called elbows are formed where ridges match the boundaries. Depending on the boundaries, two kinds of elbows are formed, usually named as bulged and pinched. Both the superstructure and the long range order induce an inhomogeneous compressive strain of the surface, which is larger at ridges and elbows and smaller in fcc and hcp regions. The interatomic distances are different in the different regions as well, varying form 2.86 Å in the fcc or hcp regions to 2.72–2.75 Å in the ridges and to 2.65–2.68 Å at the elbows [23]. The separation between two adjacent elbows is minimum along the $<11\bar{2}>$ directions and is about 7.3 nm [6], while it is larger along the $<1\bar{1}0>$ directions (typically around 15 nm). The elbows of the reconstruction represent peculiar sites for nucleating metal clusters in well ordered arrays [24]. Such peculiar growth is intimately connected to the non-uniform strain of the herringbone reconstruction that drives the diffusion of deposited adatoms on the surface [25]. For low coverages, distances between clusters follow the long range order of the reconstruction. As the coverage of the deposited metals increases, different behaviors have been observed depending on the material. As an example, Pd and Pt start to distort the original herringbone reconstruction removing part of the accumulated stress by intermixing with the surface [26] or changing the position of DLs with respect to native position [27,28]. Concerning the Fe/Au(111) system, it was instead shown that Fe continues the growth after the initial nucleation without significantly distorting the underneath Au reconstruction. As a consequence, islands grow in size and, due to the smaller separation along the $<11\bar{2}>$ directions, they start coalescing in a quasi 1D fashion [6,7,29].

This scenario is well confirmed by our STM images shown in Fig. 2. Four representative coverages are shown. Similarly to previous findings [6,7], at a coverage of 0.1 ML (Fig. 2a), Fe islands nucleate preferentially at the elbows and an ordered growth is observed. Bulged elbows favor triangular-shaped islands while pinched elbows are covered by diamond-shaped ones [7,10,13]. At this coverage, almost all the Fe atoms are accommodated in monolayer islands. However, already at 0.2 ML (Fig. 2b) a few islands grow out of the elbows positions, indicating that the nucleation at the elbows does not represent the only possibility for Fe to nucleate. The situation is significantly different for a coverage of 0.4 ML (Fig. 2c). A certain number of Fe islands have

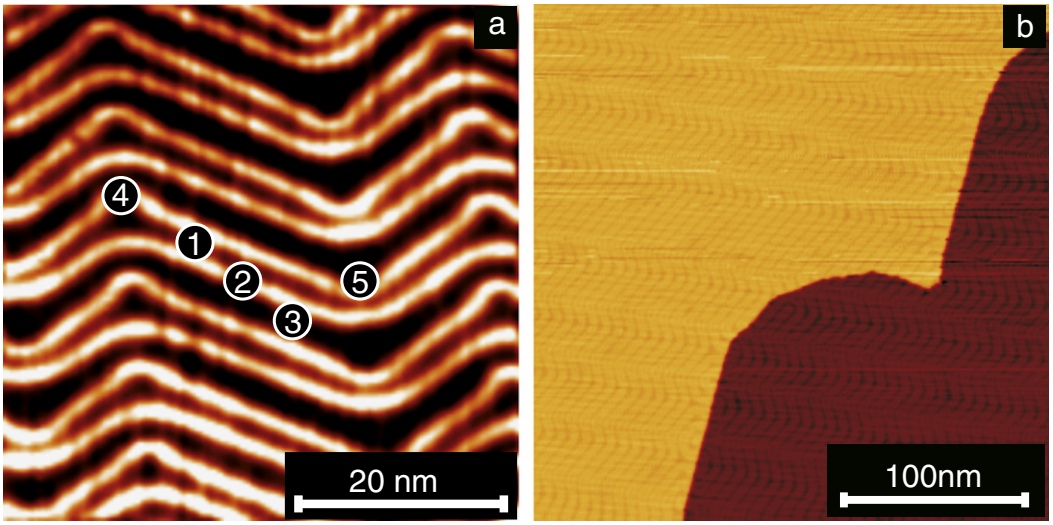

Fig. 1. a) and b): STM images of the herringbone reconstruction of Au(111) ($V_b=-1$ V and $I_s=1$ nA). In picture a), the different regions of the reconstruction are labeled as (1) hcp region, (2) discommensuration line (DL), (3) fcc region, (4) bulged elbow and (5) pinched elbow.