Does one-dimensional (1D) adatom and cluster diffusion of Pt on the Pt(110)-(1 × 2) surface lead to 1D ripening?

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Abstract. The technique of scanning tunnelling microscopy (STM) uniquely allows dynamic processes on surfaces to be followed directly in real space and at atomic resolution. Results for the surface diffusion of Pt adatoms and clusters on the anisotropic, missing row reconstructed Pt(110)-(1 × 2) surface are briefly reviewed. Mass transport in this system is entirely one-dimensional (1D) since, at low adatom coverage, atoms and clusters are confined to the missing row troughs. In this paper, we therefore address the question if Pt/Pt(110)-(1 × 2) is a 1D model system to study late stage growth phenomena such as island ripening? From STM measurements, we quantify the morphology changes resulting from annealing a surface configuration with small 1D Pt islands in the missing row troughs to temperatures in the interval 369–395 K. Interestingly, the resulting increase in island sizes (ripening) cannot be accounted for by the known island and adatom mobilities within a 1D model. An explanation is provided from dynamic, time-resolved ‘STM-movies’ that directly reveal two novel island-mediated mechanisms for inter-trough mass transport which cause the Pt/Pt(110)-(1 × 2) system not to be purely 1D at the higher surface coverage used in the annealing experiments.
1. Introduction

Growth of crystals and thin films by vapour deposition of atoms onto solid surfaces is one of the most fundamental areas of surface science and much effort has been directed at elucidating the mechanisms of the underlying microscopic processes such as diffusion of adatoms on surfaces, nucleation and growth of adatom clusters, and cluster decay and ripening [1]–[4].

Over the last two decades, the technique of scanning tunnelling microscopy (STM), and other related local probe microscopies, has revolutionized the field of surface science by providing direct real space images of surfaces at high (atomic) resolution [5]. By STM it is uniquely possible to, on one hand, zoom in on atomic scale details of the growth process and, on the other hand, obtain information on the surface morphology at larger length scales. STM can be applied both in a static mode where surface morphologies resulting from specific growth conditions are studied ‘after-the-fact’ [3, 4] and in a dynamic mode where microscopic processes and changes to the surface morphology are followed directly by acquiring a series of consecutive STM images. From a quantitative analysis of the resulting time-lapse ‘STM-movies’, information on rates for different microscopic processes can be deduced. If experiments are furthermore performed at a range of temperatures, Arrhenius analysis of the deduced rates becomes possible, allowing activation energies and prefactors to be extracted. Surface diffusion, the most fundamental process involved in solid-on-solid growth, has been investigated by this approach in a multitude of different systems, including self-diffusion of metal [6, 7] and semiconductor [8] adatoms and clusters [9], vacancy diffusion on metals [10], oxides [11] and semiconductors [12], adatom hetero-diffusion [13, 14], diffusion of small gaseous adsorbates on metal [15, 16] and oxide surfaces [17], and diffusion of larger organic molecules [18, 19].

In the following, we briefly review results from our own group on dynamic processes for Pt adatoms on the Pt(110) surface, obtained by dynamic, time-resolved, variable temperature STM [6], [20]–[22]. This review also sets the stage for the bulk part of this contribution which concerns new results for late-stage growth and ripening in this homo-epitaxial system.

As illustrated in figure 1, the clean Pt(110) surface forms a so-called (1×2) missing-row reconstruction in which every second close-packed atomic row of the idealized bulk truncation is removed, resulting in a highly anisotropic surface morphology with one-dimensional (1D) troughs separated by close-packed rows of Pt atoms [23]. The sidewalls of the troughs are (111) micro-facets. An STM-image of the Pt(110)-(1×2) surface after a sub-monolayer amount of Pt has been deposited is shown in figure 2. The close-packed Pt rows separating the missing row troughs are imaged with atomic resolution. Deposited Pt atoms are found in the 1D troughs, both as single adatoms and in the form of dimers (two-adatom clusters) and islands (longer chains of agglomerated adatoms).

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Figure 1. Structure of the Pt(110)-(1 × 2) missing row reconstructed surface. An individual adatom and a four-adatom long island is shown in the lower and upper missing row trough, respectively. The rectangles to the left and right indicate (1 × 2) and (1 × 1) unit cells, respectively.

Figure 2. Atomically resolved STM image of the Pt(110)-(1 × 2) missing row reconstructed surface obtained after deposition of a sub-monolayer amount of Pt (230 × 244 Å²). The deposited Pt atoms can be seen in the troughs as individual adatoms, dimers and larger 1D islands.

From STM-movies, typically containing several hundred consecutive images acquired with a time separation of 1–10 s, we have investigated the dynamics of Pt adatoms and adclusters on this surface in the temperature interval 300–380 K, and in the shown low-coverage regime (for an example, see movie 1 in the supplementary material). We find that the individual Pt
adatoms are confined to the missing row troughs and have never observed them to traverse into neighbouring channels. The mass transport on the surface from adatom diffusion is thus entirely 1D. Detailed statistical analysis of the distribution over observed image-to-image displacements of individual adatoms have shown that it cannot be accounted for by simple atomic hops between nearest-neighbour sites [6]. Theoretical modelling has later ascribed this to a preferred adatom diffusion path involving meta-stable sites on one of the (111) side-walls of the troughs, rather than directly along the trough as one might intuitively expect [24]–[26]. This facet walk occurs on a time-scale too short to make it directly observable by STM. However, since it will sometimes result in comparatively fast transitions between non-nearest-neighbour sites along the trough, the RMS displacement for the atomic transitions between stable adsorption sites will be slightly larger than one lattice spacing which is indeed observed in the experiments [19]. Direct STM evidence for the facet mechanism was obtained by studying the 1D Pt clusters situated in the troughs. For these clusters, the facet mechanism leads to a special ‘leap-frog’ diffusion mode in which an end atom of the cluster moves, via the facet path, up onto the sites formed by the remainder of the cluster, and one of the adjacent close-packed rows diffuses along the cluster and drops down again at the other end, leading to a net displacement of the cluster by one lattice spacing along the trough [20, 27]. The lifetime of the adatom on top of the cluster is sufficient to make it directly observable in STM images. In addition to diffusion of adatoms and clusters, the decay of dimers and larger 1D islands was investigated from the STM movies [21]. Whereas dimers dissociate quite readily, the inter-atomic binding is substantially increased for the longer adatom chains for which dissociation events were not statistically significant at the investigated temperatures. The rates for all these microscopic processes, adatom and cluster diffusion, and dimer and cluster decay, were quantified from the STM movies [6, 20, 21]. Adatom site-to-site transitions occur with the highest rate (1–0.001 transitions per second depending on the temperature in the interval 380–300 K), whereas cluster diffusion occurs with a rate approximately 2 orders of magnitude lower. Dimers dissociate with a rate intermediate between adatom and cluster diffusion, whereas detachment of adatoms from larger clusters occurs with a considerably lower rate, at least 1–2 orders of magnitude lower than the rate for the cluster displacements. (See [25] for a critical theoretical analysis of the ordering of activation energy barriers for some of these processes.)

In summary, these previous investigations have shown the Pt/Pt(110)-(1×2) system to exhibit entirely 1D mass-transport, along the missing row troughs by adatom and cluster diffusion. The work described in the present contribution was initiated in order to investigate coarsening in this apparently 1D system. This is briefly motivated in the following.

Growth of thin films by vapour deposition typically results in non-equilibrated structures determined by kinetic factors of the growth process [3]. The eventual approach to thermodynamic equilibrium involves coarsening, i.e., an increase in the length-scale of the growth features [28]. The typical mechanism behind this process is Ostwald ripening where larger islands grow at the expense of smaller ones. Thermodynamically, Ostwald ripening is driven by the Gibbs–Thomson effect according to which the vapour pressure outside a curved gas-surface interface is larger for smaller curvature [28, 29]. Microscopically, Ostwald ripening implies that atoms detach from smaller islands and meander about on the surface and attach to larger ones [30]. An alternative channel to coarsening, which can sometimes dominate over Ostwald ripening, is coalescence of diffusing adatom islands [31].

Coarsening has been studied for 3D [28] and 2D [30, 31] islands on surfaces. A situation with 1D chain-like islands is interesting since for 1D islands above a certain size the atoms at the ends of the islands will detach with a rate that is independent of the island-size (provided
there are no long-range effects). Koh and Ehrlich have recently studied such a case without a Gibbs–Thomson driving force and introduced a process referred to as stochastic ripening [32]. A system with 1D islands but 2D adatom diffusion between the islands, as studied in [32], differs from a truly 1D system, for which new and interesting effects may occur. For instance, the lattice gas between islands will interact more strongly for a truly 1D system since diffusing adatoms will not be able to pass each other. This could be imagined to result, e.g., in an enhanced probability for nucleation of new clusters during the ripening process compared to the 2D case.

In our experiments, we have recorded the morphology changes occurring when a surface with 1D Pt islands situated in the missing row troughs of the Pt(110)-(1 × 2) surface is thermally annealed. From a quantitative analysis of the resulting increase in island sizes, we found the interesting result: the known mobility of islands and adatoms is far from sufficient to account for the observed coarsening within a purely 1D model. Based on direct observations of the surface dynamics by time-lapsed STM, we conclude that the origin of this apparent discrepancy is that the Pt/Pt(110)-(1 × 2) system cannot be treated as purely 1D at the higher adatom coverage used in the annealing experiments. We found two different mechanisms for cross-channel mass transport both of which result from interactions between islands situated in neighbouring missing-row troughs. These novel mechanisms enable a much more efficient coarsening than expected in a purely 1D system.

2. Experimental procedure

The experiments were performed in an ultra-high-vacuum (UHV) chamber with a base pressure around 5 × 10⁻¹¹ mbar equipped with a home-built variable temperature UHV-STM as well as standard facilities for sample preparation and characterization (for details on the Aarhus STM, see [33]–[35] as well as www.specs.de). The Pt(1 1 0) single crystal was sputter-cleaned with 1.5 kV Ne ion bombardment followed by annealing to 980 K with a ramp of 2 K s⁻¹. This resulted in a clean surface and it was in its (1 × 2) reconstructed state as judged by Auger electron spectroscopy (AES), low-energy electron diffraction (LEED) and STM. Pt adatoms were deposited onto the surface by resistive heating of a thoroughly outgassed 0.4 mm diameter 99.995% pure Pt wire, wound to a small coil to ensure homogeneous heating. There was no significant pressure rise in the chamber during deposition.

To prepare the initial configuration of islands, Pt was deposited onto the (1 × 2) surface at a sample temperature of 344 ± 2 K (deposition time 5 min). The resulting Pt adatom concentration corresponds to 27 ± 4% of the sites available in the missing-row troughs (i.e. an absolute coverage of 0.13 monolayers). After switching off the deposition flux, the sample was kept at the deposition temperature for another 10 min to allow the deposited adatoms to incorporate into 1D islands. Subsequently, the sample temperature was increased, within less than 2 min, to the final annealing temperature. Care was taken that the temperature did not increase above its final value. Annealing was carried out for 10 min after which the sample was cooled, within less than 1 min, to temperatures below 270 K where the adatom mobility and other dynamic processes are completely frozen out. During the annealing period, the temperature was maintained constant to within ±1 K and a total of 5 annealing temperatures in the temperature interval between 369 and 395 K were investigated. STM imaging was performed at temperatures in the interval from 230 to 270 K. The sample was not imaged prior to the annealing in order to avoid unnecessary temperature transients. From the acquired STM images, between 100 and 300 adatom islands were analysed for each annealing temperature and for the initial configuration.
3. Results and discussion

The deposition of Pt adatoms onto the surface and the subsequent annealing leads to the formation of adatom islands as shown in the STM images of figures 3(a)–(d), obtained for successively higher annealing temperatures. Two types of adatom islands can be found: (i) islands consisting of adatom chains situated entirely within the troughs, and (ii) islands where part of the adatoms are situated in the troughs and part of the adatoms occupy adsorption sites formed by these underlying atoms and one of the surrounding close-packed rows (see figure 3(e) for a schematic model). These latter islands, which are depicted brightest in the STM images, will be referred to as covered or bi-layer islands in the following.

Figure 3. STM-images showing the changes to the island morphology on the Pt(110)-(1 × 2) surface that occur upon annealing. The images (a)–(d) were obtained after annealing to 373, 379, 384 and 395 K respectively. Note that the area imaged in (a) (153 × 163 Å²) is smaller than that in (b)–(d) (230 × 244 Å²). (e) Ball-models illustrating the restructuring of the islands.
Although geometrically there is room for two adatom chains on top of each of the surface patches formed by an in-trough adatom island with two surrounding close-packed rows, such configurations are not observed. It is furthermore characteristic that the bi-layer islands are predominantly covered completely along their length axis; the fraction being only partially covered is below 5% for all temperatures.

Size-distributions for islands in the starting configuration as well as those obtained after annealing to 379 and 395 K, respectively are shown in figures 4(a)–(c) (note that the abscissa has been normalized to the average island size, $s_{av}$, as indicated on the plots). The contributions from islands that are not covered and from bi-layer islands are shown in black and grey, respectively. With increasing annealing temperature, an increasing fraction of the islands become covered. The islands that are not covered are the smaller ones. Figure 5 shows the average island-size for bi-layer islands that are covered completely and for islands that are not covered in the second layer, respectively, as a function of annealing temperature. While the average size of the bi-layer islands increases with increasing temperature, the islands that are not covered remain to have roughly the same size.

The starting configuration (before the final annealing step) consists of comparatively small islands of mean size $\sim 6–7$ atoms which are exclusively of type (i), i.e. not covered. The bi-layer islands are therefore not a growth phenomenon resulting from deposition on top of already existing islands, but must stem from material transport from the troughs and up on top of the adatom chains during the annealing period where no direct Pt deposition flux is present.

In the following discussion, we focus on (i) the change in island morphology, and (ii) the increase in the island sizes. Firstly we consider the changes from an energy perspective and secondly from a kinetic perspective.

The coarsening observed upon annealing is an approach towards thermodynamic equilibrium since the increase in island sizes reduces the number of unsaturated bonds at the ends of the islands. A thermodynamic driving force for the change of island morphology into bi-layer islands can also be identified: an island situated entirely within a missing row trough creates a patch of unreconstructed $\left(1 \times 1\right)$ surface. However, the equilibrium structure of the Pt(110) surface is the $\left(1 \times 2\right)$ missing row reconstruction. A repulsive interaction must therefore exist between the atoms of the island and the surrounding close-packed rows (as is also supported by the absence of bi-layer islands covered by two adjacent adatom chains). By moving adatoms from the missing-row trough up on top of the island, the adatoms being moved minimize these repulsive interactions while keeping the same coordination to atoms below. (A transformation of the $\left(1 \times 1\right)$ surface somewhat similar to the island restructuring reported here has been observed in an early field ion microscopy (FIM) study [36].)

Although the observed coarsening and changes of island morphology seem reasonable from a thermodynamic viewpoint, the question of the underlying kinetic mechanisms still remains. In this respect, the formation of bi-layer islands is surprising since the necessary ascending motion of adatoms over a step-edge is a microscopic process normally not considered in growth models [37]. However, from our earlier observations of cluster mobility by the leap-frog mechanism [20] we know that in the present case it is indeed possible for atoms at the ends of the islands to move up on top of the adatom chains situated in the troughs. While island mobility results from such atoms diffusing over the islands, the formation of the bi-layer islands most probably results from such atoms nucleating and growing into the adatom chains found on top of these islands. The observation that the bi-layer islands are typically covered completely could result from such a mechanism: nucleation would require the more or less likely event that two (or more) atoms
Figure 4. Island size distributions derived from the annealing experiments. (a) Shows the start-configuration whereas (b) and (c) depict the distributions obtained after annealing to 379 and 395 K, respectively. The contribution from bi-layer islands is shown in grey, whereas the contribution from islands that are not covered is shown in black. The island sizes have been scaled by the average island size $S_{av}$ (averaging over both covered and not covered islands), and the distributions are normalized to unit area (see [40]).

simultaneously moved on top of an island. Once a nucleus was formed, the second layer chain would quickly grow to cover the whole island.

We now turn to discuss the coarsening. In traditional Ostwald ripening, the redistribution of mass between islands is mediated by adatoms that detach, migrate over the surface and attach to other islands [28]. While such a process may occur to some extent in the present case, our earlier observation [20, 21] that, for the present system, the rate for island displacement is considerably
Figure 5. Mean size of islands that are completely covered and those that are not covered, respectively, as a function of annealing temperature. The data point at 344 K represent the starting configuration for the annealing experiments. For bi-layer islands, the sizes include both the atoms in the troughs as well as those in the second layer.

larger than the rate by which adatoms detach from the islands and diffuse out into the troughs, means that coalescence of migrating islands must necessarily play the dominating role in the coarsening process. Interestingly, however, it is not possible to account quantitatively for the observed coarsening solely by invoking 1D island diffusion as the following crude estimate clearly shows: in the starting configuration for the annealing experiments approximately 25% of the adsorption sites available in the missing row troughs are occupied by adatoms and the islands have a mean size of ∼7 atoms. The mean island–island separation along the troughs is thus ∼21 nearest-neighbour sites. Following annealing for 10 min at 379 K, the mean island size is doubled to ∼14 atoms (see figure 4(b)). However, the determined island displacement rate at this temperature is only ∼0.01 s$^{-1}$ [20], so during the annealing period of 10 min, an island only makes on average ∼6 displacements (and the number of times an atom leaves an island is considerably smaller). Clearly, this island mobility is not sufficient to explain the observed degree of coarsening. We estimate that the island displacement rate necessary to account for the observed coarsening within a completely 1D model is 1–2 orders of magnitude larger than that observed experimentally.

The origin of this apparent discrepancy has been identified as the surface adatom coverage. Our initial dynamic STM experiments were carried out at low adatom coverages of a few percentage in order to minimize interactions between adatoms. In the present ‘quench-and-look’ experiments, however, the coverage is considerably higher with adatoms in ∼25% of the sites in the missing-row troughs. We have therefore directly monitored the surface dynamics by STM also at higher adatom coverages. From the acquired data two novel channels for mass transport across the close-packed rows have been identified. Both mechanisms require nearby islands in adjacent troughs and therefore become significant only at higher adatom coverage.
The first channel is illustrated in figure 6, showing a series of time-lapsed STM-images depicting two adatom islands situated in adjacent troughs. While the total number of atoms within the islands stays constant during the sequence, the sizes of the individual islands fluctuate as clarified in the schematic insets. It is thus apparently possible for the two islands to exchange atoms. The atomistic mechanism enabling this mass transport is most likely the same promotion of end-atoms on top of the islands that is involved in the island mobility by the leap-frog mechanism and the morphology change into bi-layer islands. The two adjacent islands form a small patch of (1 × 1) surface. The self-diffusion on unreconstructed Pt(110) is known to be 2D occurring with approximately equal rates along and perpendicular to the close-packed direction [36, 38, 39]. Therefore, once an atom has moved up on top of the (1 × 1) patch, it will be able to cross the close-packed row and descend in the adjacent trough, enabling the observed exchange of atoms between the two islands.

The second channel for inter-trough mass transport involves the movement of the close-packed row between two adjacent islands as depicted in figures 7(a)–(c) showing three consecutive frames from an STM movie. From image (a) to (b) a number of atoms from the close-packed row above island ‘1’ displace into the upper adjacent trough augmenting island ‘2’, and from (b) to (c) the atoms of island ‘1’ shift into the positions formerly filled by this close-packed row. The net result of these processes is that island ‘1’ moves to an adjacent trough and joins island ‘2’. Although the image sequence shows the intermediate state with atoms displaced out of the close-packed row, the individual atomic steps leading to the displacement are too fast for us to resolve by STM. With reference to figure 7(d) we speculate that the observed process occurs because (i) the atoms shown with heavy outline in the centremost close-packed row are raised in energy due to the repulsion from the adjacent island, and (ii) the energy barrier for
Figure 7. STM images and ball model illustrating the channel for inter-trough mass transport that involves a shift of the close-packed row into an adjacent trough. See text for details.

Irrespective of the atomistic origins of the observed cross-channel adatom motion, it directly shows that the Pt/Pt(110)-(1 × 2) system cannot be treated as purely 1D at the higher adatom coverage used in the annealing experiments. In the starting configuration for these experiments, an island is likely to have a neighbour very close if not only islands in the same but also the two adjacent troughs are considered. With the observed mechanisms enabling such adjacent islands to merge together comparatively fast, it becomes possible to account for the coarsening observed in the annealing experiments.

4. Conclusion

In summary, we have characterized morphology changes that occur to 1D Pt islands situated in the missing row troughs of the Pt(110)-(1 × 2) surface when the surface is annealed. We find a coarsening in the sizes of the islands and they furthermore restructure into a two-layer morphology. A qualitative explanation for the island restructuring is proposed in terms...
of nucleation and growth of adatoms that have moved up on top of the islands, which is known to be possible from our earlier results for the diffusion of 1D clusters by the so-called leap-frog mechanism. Most interestingly, we find that the observed increase in island sizes cannot be accounted for by the known rates for island diffusion and decay within a completely 1D ripening model, as would otherwise have been expected from our earlier measurements performed at low adatom coverage. We have resolved this apparent discrepancy by monitoring the surface dynamics directly at higher coverage, identifying two novel mechanisms for inter-trough mass transport. These results show that the Pt/Pt(110)-(1 × 2) system cannot, in general, be viewed as a 1D model system to study late stage growth phenomena, such as ripening.

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