ON THE ROLE OF KINKS AND STRAIN IN HETEROEPITAXIAL GROWTH: AN STM STUDY

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Interlayer diffusion of Co over steps of vacancy islands on the Pt(111) surface as studied by scanning tunneling microscopy is presented. It is demonstrated that Co atoms descend Pt steps by an exchange diffusion process at the step edge with the Pt atoms. Further, the exchange diffusion process is observed to occur at the corners (kinks) of the vacancy islands. The importance of kinks concerning whether the growth mode of a heteropitaxial film is two-dimensional or three-dimensional is demonstrated for the case of thin Co films on Pt(111). We argue that the strain in the Co film is to a large extent responsible for the kink formation.

1. Introduction

Information on the structure, morphology and growth mode of thin metal films on metal substrates is essential for understanding magnetic and/or electronic properties observed at the interface. Moreover, if certain magnetic or electronic properties are desired at the interface, a deeper understanding of the atomic scale processes governing the thin film growth is crucial.

Interlayer diffusion across steps is a decisive process as to whether a film will exhibit a layer-by-layer (two-dimensional, 2D) or an island growth (three-dimensional, 3D). Thus, this process is of fundamental importance, and has accordingly been studied intensively in the recent past in an effort to understand the energy barrier [Ehrlich–Schwoebel (ES) barrier] of the interlayer transport mechanism. Interlayer diffusion is described in Fig. 1(a); the atom on the upper terrace may descend the step by either a hopping process or an exchange process (concerted substitution). Further, on fcc(111) surfaces, two different kinds of straight steps exist: the so-called A and B steps [Fig. 1(b)], with (100) and (111) microfacets, respectively, which, since they may exhibit different ES barriers, are important. In addition, there is a growing consensus that step features such as corners and kinks [Fig. 1(c)], which are geometrically similar, may play a crucial role in interlayer diffusion and thus the growth mode.

Previously, investigations of the atomic scale processes in interlayer diffusion have been limited to...
computational studies\textsuperscript{1–6} and field ion microscopy.\textsuperscript{7–10} This paper will demonstrate that scanning tunneling microscopy, using chemical contrast, offers a unique ability to directly study interlayer diffusion on the atomic scale. We will use the Co/Pt(111) thin film interface system, which is an interesting system because of its particular magnetic properties at the interface,\textsuperscript{11} to illustrate the atomic scale findings.

2. Results and Discussion

2.1. Interlayer diffusion on the atomic scale

For experimental details, see Refs. 12–14. In order to study interlayer diffusion on the atomic scale, we created vacancy islands on the clean Pt(111) surface by sputtering with 2 keV Ar\textsuperscript{+} with an Ar\textsuperscript{+} ion dose of $4 \times 10^{13}$/cm\textsuperscript{2} at a sample temperature of approximately 650 K.

Figure 2(a) is an STM image of the Pt(111) surface after such a procedure, the inset showing a one-atom-deep vacancy island with atomic resolution displaying the same features as found previously,\textsuperscript{15–16} the longer sides being B steps.

Evaporation of 0.04 monolayers (ML) of Co results in the appearance of the vacancy islands as shown in Figs. 2(b) and 2(c). As in our previous studies of Co/Pt(111),\textsuperscript{12} due to chemical contrast, Co atoms appear darker than Pt. It may be seen that two different types of Co atoms exist at or close to the steps of the vacancy islands. One type is situated at the step edges (type 1) while the second type of Co is situated behind the first row of Pt atoms on the upper terrace (type 2), as indicated by the white and black arrows, respectively. Previously\textsuperscript{13} we have shown that it is possible to relate the number of type 1 atoms to the number of Co atoms that should have adsorbed within the vacancy island and then have diffused to the step of the vacancy island. If diffusion over the step edge would occur, the number of type 1 Co atoms should exceed the number that is actually observed.\textsuperscript{13} Thus, we conclude that Co diffusion over the step edge does not occur. In the case of the type 2 Co atoms, since these atoms are situated behind the first row of Pt atoms, only interlayer diffusion by an exchange diffusion process may explain this appearance. Hence, the type 2 Co atoms originate from Co atoms that have adsorbed on the upper terrace and later diffused to the step edge of the vacancy island where an exchange diffusion process with the Pt atoms at the step edge has occurred. These two observations lead to the conclusion that interlayer transport on Pt(111) occurs only by an exchange diffusion process and not by diffusion over the Pt step.\textsuperscript{13}

Fig. 2. (a) Vacancy islands on Pt(111) induced by sputtering at elevated temperature (100×100 nm\textsuperscript{2}). The inset shows a vacancy island with atomic resolution, with the A and the B step indicated. (b) Vacancy islands after deposition of 0.04 ML of Co (5×10 nm\textsuperscript{2}). The black and white arrows indicate Co atoms behind and at the step edge, respectively. (c) As in (b) but with a larger vacancy island (7×10 nm\textsuperscript{2}).
Additional information on the interlayer diffusion may be gained by counting the number of type 2 atoms with respect to the A and the B step. No preference for descent is found at any of these steps; in contrast, the number of type 2 atoms is found to be constant irrespective of the size of the vacancy islands (as opposed to the length of the step edges). This behavior may be seen in Figs. 2(b) and 2(c), and leads to the conclusion that the exchange diffusion process at steps in the Co/Pt(111) system does not occur at straight steps but rather at the corners of the vacancy islands, since the number of corners is constant (six).

The fact that an exchange process at a kink or a corner is a favored process is not new. For instance, it has been found that homoepitaxy of Pt on Pt(111) is 2D at high and low temperatures, but 3D at intermediate temperatures.\textsuperscript{17,18} One explanation for this so-called re-entrant 2D growth is an irregular island shape at lower temperatures. Irregular islands would increase the density of kinks or concave corners, thus facilitating the descent of adatoms.\textsuperscript{4,5,19} Further, we have performed a large number of embedded atom method (EAM) calculations which confirm the above experimental observations.\textsuperscript{6} Below we will further illustrate the importance of kinks for the growth mode.

### 2.2. Co on Pt(111): kinks and the growth mode

In Fig. 3 a set of STM images is shown from Co coverages on Pt(111) ranging from 2.2 to 10 ML. Three observations can be made directly from these images: (1) only a few terrace levels are observed at low coverages as compared to higher coverages; (2) the steps are much rougher at lower coverages; (3) at higher coverages, triangular terraces are stacked on top of each other facing the same direction.

The first observation implies that the growth is 2D at lower coverage. Indeed it was shown that 2D growth prevails up to approximately 3 ML, where a transition to 3D growth occurs until approximately

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig3.png}
\caption{STM images (300 × 300 nm\textsuperscript{2}) showing the development of the growth and morphology of thin Co films on Pt(111). (a) STM image from a 2.2-ML-thick Co film on the Pt(111) surface; (b) STM image from a 3.5-ML-thick Co film on the Pt(111) surface; (c) STM image from a 5-ML-thick Co film on the Pt(111) surface; (d) STM image from a 10-ML-thick Co film on the Pt(111) surface.}
\end{figure}

Fig. 4. (a) The adisland shapes expected if the formation process or growth speed is equivalent or nonequivalent for the two facets. (b) A and B step sequence for hcp and fcc. Rectangles and triangles refer to A and B steps, respectively. (c) Hcp and fcc leads to triangles stacked facing the opposite or the same direction, respectively.\textsuperscript{21}
At this latter coverage, the terrace areas could be shown to follow a Poisson distribution, in agreement with a 3D growth mode with no interlayer diffusion.

The second observation implies the presence of a large number of kinks and corners at low coverages. The 2D growth now finds its natural explanation; kinks and corners are favorable sites for interlayer diffusion, as shown above, promoting 2D growth.

The third observation is related to the stacking sequence of the Co film (see Fig. 3; 5 ML and above) when the growth is 3D. The observation of triangular terraces may be explained by the fact that either the A or the B step is preferred energetically or has a different growth speed. The fact that the triangles are stacked facing the same direction demonstrates that the stacking of the Co layers is predominantly fcc (Fig. 4). This conclusion is confirmed by the existence of inclined stacking faults in the Co film\(^{14}\) with a height of 1/3 or 2/3 of the monoatomic step, which may only exist in stacked layers with fcc stacking. By the use of the directions of these stacking faults, it could also be shown that the longer side of the triangles is an A step.

Returning to the lower coverages, we believe that an important reason for the large number of kinks and corners is the large tensile stress induced by the lattice mismatch (9.4\%) between Co and Pt. The stress and the resulting strain at the interface is visualized in Fig. 5; the stress is partially relieved by a dislocation network in the first Co layer (for details, see Ref. 14) and in the second layer by a moiré structure.\(^{14,20}\) The out-of-plane strain modulation of the moiré structure, which is due to the fact that Co atoms in on-top sites of Pt appear higher than Co atoms in hollow sites, may be observed up to the 11th level of the stacked triangles (see Fig. 6). The in-plane strain is closely related to the out-of-plane strain (albeit more difficult to estimate), since the Co atoms try to avoid on-top sites and are thus more compressed in these areas as compared to in the hollow sites where they try to follow the substrate lattice, in accordance with a Frenkel–Kontorova model. At lower coverages the strain is high, and as a result several important phenomena, such as surface diffusion, nucleation,\(^{21}\) diffusion along steps and around island corners or kinks will be affected, leading to smaller island size at lower coverages [see Fig. 3(a)] and influencing their shapes. However, the detailed process leading to the formation of kinks in a strained system cannot be determined without simulations or calculations.

As the Co coverage is increased, the steps become straighter. The reason for this is the decreasing strain, which again influences the phenomena mentioned above. In addition, we also suggest that step–step interaction becomes more important at higher coverages, which further contributes to

Fig. 5. STM image (40 × 50 nm\(^2\)) of 0.8 ML of Co. The inset to the upper left shows a dislocation network of the first Co layer as well as the Pt(111) surface with a step edge (10 × 15 nm\(^2\)). The inset to the upper right shows the moiré structure which is formed in the second Co layer (10 × 10 nm\(^2\); this inset is from a 3.5-ML-thick Co film).

Fig. 6. The corrugation of the moiré structure as a function of the terrace level.
straighter steps. Straighter steps should prohibit interlayer diffusion and as a result we observe 3D growth. However, if the Co film is thick enough to accommodate the whole misfit, one would expect from classical growth theory that the system should be equivalent to a homoepitaxial growth mode leading to a complete wetting of the surface, allowing layer-by-layer growth again. At coverages as high as 15 ML we still do not observe layer-by-layer growth; this implies either that the misfit is not accommodated to allow a complete wetting, or that the system is disturbed by stacking fault defects serving as nucleation centers of 3D island. Interestingly, the latter situation would lead to 3D growth without any chance to recover to 2D growth, since no interlayer diffusion occurs due to the straight steps.

3. Conclusion

In conclusion, we have shown, by the use of STM, that kinks and corners are favorable sites for interlayer diffusion. We have illustrated the importance of this finding by studying the growth of thin films of Co on Pt(111) at RT. The induced stress at the interface by the lattice mismatch between Co and Pt has been put forward as a reason for the growth morphology.

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