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Letter to the Editors

The concept of two mobilities in homoepitaxial growth

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Abstract

A general kinetic concept is introduced which can be used to control growth modes in homoepitaxy. Its basic idea is that during growth of a layer, the characteristic length scale associated with nucleation is deliberately varied. The power of this concept lies in the fact that it can be realized experimentally in a variety of ways and is not restricted to special systems. It helps to understand various effects reported in the literature and may serve as a guideline for future methods of growth manipulation.

Growth on a crystal surface is usually initiated by nucleation: the deposited atoms (adatoms) diffuse on the surface until they meet and form nuclei. After a short induction period, a saturation density of nuclei is reached and further deposited atoms condense at the existing nuclei which grow into two-dimensional (2D) islands. In this stage of layer growth, the surface consists of two levels separated by steps of monatomic height: the lower level is the substrate and the upper level the top of islands. It is the fate of adatoms deposited onto the *upper* level that ultimately determines how the film continues to grow. For layer-by-layer- or 2D-growth, it is a necessary condition that no nuclei are formed on top of islands before a closed layer is formed by coalescence of two-dimensional islands; adatoms deposited onto the island top have to descend onto the lower level before they can nucleate. On the other hand, if there is nucleation on top of islands before coalescence, three-dimensional structures develop and the film grows rough (multilayer- or 3D-growth).

It is the aim of this work to introduce a general concept which can be used to induce 2D-growth even under conditions which in conventional homoepitaxy would inevitably lead to 3D-growth. In order to describe this concept in a simple way, we define a nucleation length (or mean free path for nucleation) as the average distance an adatom travels before it forms a stable nucleus. We distinguish between this nucleation length in the upper level, $\bar{\lambda}$ and the nucleation length in the lower level, $\underline{\lambda}$. In a description using average quantities, $\underline{\lambda}$ is the mean separation of islands nucleated on the lower level.

We first use this notation to describe the different growth modes in conventional homoepitaxy (cf. Fig. 1). In this case the two nucleation lengths are the same: $\bar{\lambda} = \underline{\lambda}$, because by definition all levels have the same properties. The quantity that controls growth modes in homoepitaxy is an additional energy barrier for downward diffusion

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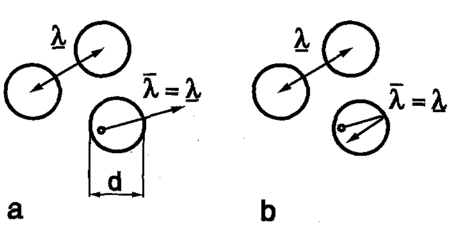

Fig. 1. Schematic sketch of a surface during monolayer growth in conventional homoepitaxy ($\bar{\lambda} = \underline{\lambda}$). Islands are assumed to be circular. (a) *No barrier* at the island edge. Before coalescence, the island size d is smaller than the island separation $\underline{\lambda}$, and hence, also smaller than $\bar{\lambda}$, so that the adatom can escape from the island before it has travelled its nucleation length $\bar{\lambda}$ (2D-growth). (b) *High barrier* at the island edge. The adatom is reflected at the island boundary. It has travelled its nucleation length $\bar{\lambda}$ before coalescence (3D-growth).

at the island edge [1–3]. In the absence of such a barrier we obtain 2D-growth: from simple geometry it follows that before coalescence the mean island size is smaller than the mean island separation $\underline{\lambda}$, so that any adatom on top of an island hits the island edge before it has travelled its nucleation length $\bar{\lambda}$, and because there is no additional barrier, it jumps onto the lower layer. Hence, there is no nucleation on top of islands before coalescence. On the other hand, if there is a significant barrier, the adatom has a high probability of being reflected at the island boundary so that it may be captured on top of the island until it has travelled its nucleation length. Again simple geometry tells us that in this case nucleation *must* take place *before* coalescence. Hence, in the case of a high barrier at the island edge, we inevitably get 3D-growth.

The concept we propose to solve this problem is to get rid of the constraint $\bar{\lambda} = \underline{\lambda}$ and – by some means or other – turn it into: $\bar{\lambda} > \underline{\lambda}$, so that the adatom on the island top hits the island boundary more often before it has travelled its nucleation length. As the barrier is high but finite the adatom eventually succeeds in jumping onto the lower level. In principle, any realization of the inequality $\bar{\lambda} > \underline{\lambda}$ may be used to improve the growth of a homoepitaxial system. However, as the nucleation length depends primarily on the mobility of adatoms we call our concept *the concept of two mobilities*.

Experimentally, different mobilities can be obtained by changing various parameters during monolayer growth. As an example we choose first the parameter substrate temperature to explain how our general concept can be turned into a recipe for growth procedures. We need two different temperatures: a low one for low mobility and a high one for high mobility. Of course, it is impossible to have two distinct temperatures in different levels of the surface *at the same time*. However, this is not a real problem, because the nucleation lengths in different levels are effective *at different times* during monolayer growth. Indeed, $\underline{\lambda}$ is effective during the nucleation period on the lower level, i.e., the first few percent of monolayer coverage. On the other hand, the probability that atoms are deposited onto and captured on 2D islands is negligibly small during deposition of the first percent of a monolayer (while the islands are still small) so that the value of $\bar{\lambda}$ is of no importance in this regime. It is only during the further growth of islands that nucleation on the upper level becomes probable and thus the size of $\bar{\lambda}$ decisive. (Conversely, during this later time, i.e., after the island density has reached saturation, $\underline{\lambda}$ is no more effective.) We can therefore realize the concept of two mobilities by increasing the temperature after the density of islands has reached saturation. In this way we keep a low mobility during nucleation on the lower level (small $\underline{\lambda}$) and a higher mobility (large $\bar{\lambda}$) when nucleation on the upper level is a danger. It has been shown that even for a system with a high barrier for interlayer diffusion like Ag/Ag(111), this recipe can be used to induce 2D-growth [4].

Of course, the temperature jump procedure has to be repeated during the growth of each layer in order to achieve layer-by-layer growth. From a practical point of view it is easier to realize a deposition rate jump (e.g., by using two evaporators with different fluxes) which has a similar effect: large rate (small $\underline{\lambda}$) during nucleation onto the lower level and small rate (large $\bar{\lambda}$) upon saturation of the density of nuclei. Markov et al. performed such modulations of both the deposition rate and the substrate temperature during epitaxial growth of Ge on Ge(111) and Si on Si(111) [5]. They obtained RHEED oscillations of improved quality compared to the conventional growth case. Whereas the interpretation in Markov's paper is mainly concerned with a better synchronization of the nucleation event, it is clear that their method is a realization of the concept of two mobilities and hence, the growth itself was improved in their experiments.

A technique often used in epitaxy to obtain better films is ion beam assisted growth [6,7]. Using our concept of two mobilities we can now understand how simultaneous ion bombardment of the growing film can reduce its roughness. Ion bombardment during deposition effectively hinders the mobility of adatoms, so that different nucleation lengths can be realized by sputtering the surface during nucleation but not during further growth of a monolayer. Using a pulsed ion beam of 600 eV Ar^+ ions during continuous deposition of Ag onto Ag(111) high quality films could indeed be grown in a layer-by-layer mode (cf. Fig. 2, curve b) [4]. The crucial point in these experiments was the use of a pulsed beam: continuous sputtering during deposition violates the principle of two mobilities because it uniformly reduces the effective mobility in any level. Indeed, growth was never improved when both deposition and sputtering were done continuously (Fig. 2, curve c).

As a final example, we note that the concept of two mobilities offers an explanation for the success of surfactant-mediated epitaxy during which the growth is improved by adding suitable adsorbates to the system. Surfactants were found to lower the mobility of adatoms [8–10] and they are not overgrown by the deposited material but always transported into the outermost layer. Especially for low concentrations of surfactants it is plausible that this transport sets in at a late stage during monolayer growth. Hence, for most of the time the surfactant atoms are only in the lower level where they reduce the nucleation length, but not in the upper level. Again, the concept of two mobilities is realized. Recent experiments by van der Vegt et al. [11,12] who induced 2D-growth of Ag on Ag(111) using Sb as a surfactant can – at least for the first monolayer – be explained along this line.

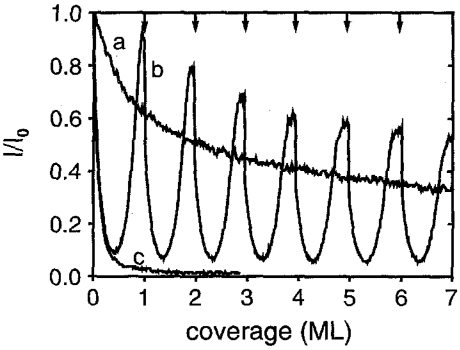

Fig. 2. Evolution of the helium specular peak height during deposition of Ag onto Ag(111) at a substrate temperature of 300 K. A monotonically decaying signal corresponds to 3D-growth, whereas for 2D-growth maxima are observed after completion of each monolayer (ML) [13]. (a) Continuous deposition at a rate of 4.7×10^{-3} ML/s without any sputtering ($\bar{\lambda} = \underline{\lambda}$): the growth mode is 3D. (b) As in (a), but additionally at the start of deposition and subsequently just after completion of each monolayer (see arrows), a short sputter pulse (600 eV Ar^+) corresponding to the removal of 0.014 ML Ag was given ($\bar{\lambda} > \underline{\lambda}$): the persistent oscillations indicate high-quality layer-by-layer growth. (c) Continuous deposition at a rate of 1.3×10^{-2} ML/s combined with continuous sputtering at a removal rate of -7.7×10^{-4} ML/s ($\bar{\lambda} = \underline{\lambda}$): the growth mode is 3D.

A general concept that can be used to understand a variety of experimental observations is certainly of help and the list of experimental examples presented here is by no means complete. But more importantly, the concept of two mobilities may serve as a guideline for future methods and open new ways of growth manipulation which can be used also for heteroepitaxial systems.

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