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Abstract. We study the optimal conditions for nanoisland growth in ion beam assisted deposition (IBAD). This situation occurs when adatom islands remain small enough to prevent the onset of three-dimensional growth, while at the same time preventing ion-induced surface erosion. We develop a rate equation model, which includes continuous deposition of adatoms and creation of vacancies, recombination of vacancies at adatom island edges and recombination of adatoms at vacancy island edges. Using this model we demonstrate that at onset between the rough and smooth layer-by-layer growth regimes there is a simple exponential relation between the largest size of the vacancy islands and the external control parameters of the growth.

1. Introduction

Ion beam assisted deposition (IBAD) of thin films is a technique where energetic ion beams with energies ranging from a few tens of eV up to a few keV are used to facilitate smooth epitaxial growth. In IBAD the impacts of the energetic ions can prevent the onset of three-dimensional (3D) growth mode and the surface can be kept in two-dimensional (2D) layer-by-layer growth mode, and thus layers with improved smoothness can be obtained [1, 2].

In the present work, we develop a rate equation model for IBAD, where the reaction kinetic rates serve as the primary parameters instead of the ion energy and mass [3]. It remains an independent task to relate the kinetic reaction rates to the parameters describing the bombardment in various special cases, which can be done either through simulations or analytically. To this end, we propose here a simplified description of submonolayer surface growth in cases where island and vacancy island growth takes place simultaneously, and find the transitional stage of growth where vacancy islands begins to be filled and eventually shrink. This borderline result gives us the limiting case, where IBAD is still applicable without causing surface erosion or destruction. The growth problem is described by using the well-established reaction kinetic model approach [3, 4], which is solved by an efficient simulation method [4–6]. The model includes the rates of all of the physically relevant events for the present case of ion-assisted submonolayer growth. The rate equation model allows us to obtain detailed information about the adatom and vacancy island size distribution during growth. As our main result, we obtain a general description of the critical coverage where the transition from growing vacancy island to decaying vacancy islands takes place.
2. Growth Model
The basic atomistic processes during growth considered here all influence the time evolution of the adatom and vacancy island number densities \( n_s \) and \( n_s' \), respectively, which are the fundamental variables in the Reaction Kinetic Model (RKM) given by a two infinite sets of equations:

\[
\begin{align*}
    n_s &= f_d + f_a + f_f + f_r \\
    n_s' &= f_d' + f_a' + f_f' + f_r'.
\end{align*}
\]

For each quantity, the rate of change of the distribution equals the total fluxes \( f_i \) due to the basic atomistic processes. These fluxes are functions of the density variables and include \( f_d \) for deposition of adatoms on the surface, \( f_a \) for attachment of adatoms to islands, \( f_f \) for detachment of adatoms from islands, and \( f_r \) for interlayer recombination. In the second equation, the primes denote the corresponding fluxes for vacancies. In addition, the flux of deposited adatoms given by \( \Phi \) monolayers per second (ML/s) and the flux of the ion bombardment set up the two other two important experimental control parameters. Additionally of these processes, we take into account here only those including single adatom or vacancy, thus excluding island break-up and island-island coalescence. The individual terms are explained in detail in the following.

The rate of deposition \( \Phi \) is expressed in ML/s. The rate of deposition for islands is given by \( f_d = \Phi \delta_{1,s} \). The term \( \delta_{1,s} \) explicitly limits the deposition to single adatoms only. Similarly, for the vacancy creation \( f_d' = \xi \Phi \delta_{1,s} \), where the prefactor \( \xi \) expresses the relative strength of the two flux terms and acts as an externally controllable variable.

The aggregation of islands is driven by adatom diffusion, and in IBAD in addition to this also island diffusion influences the aggregation rates [3, 7, 8]. In the case of a metallic surfaces the diffusion coefficient for islands of size \( s \) can be taken to follow a simple power law [9] \( D_s = K_0 s^{-\mu} \). A good choice for the exponent is \( \mu = 2 \), which is taken as a fixed model parameter in the following. The prefactor \( K_0 \) gives the scale for the aggregation relative to the deposition. A dimensionless parameter \( R = K_0/\Phi \) is introduced to fix the relative strength of aggregation and deposition, and also acts as an external variable. The aggregation rate, as described by the aggregation kernel for islands of sizes \( i \) and \( j \) can be then written as \( K_{ij} = (D_i + D_j)(\delta_{ii} + \delta_{ij} - \delta_{ii} \delta_{ij}) \), which allows island-adatom processes only. Thus, the flux \( f_a \) due to aggregation for islands of size \( s \) is given by

\[
f_a = \frac{1}{2} \sum_{i+j=s} K_{ij} n_in_j - \sum_{i=1}^{\infty} K_{is} n_in_s.
\]

The flux \( f_a' \) due to aggregation for vacancies is defined similarly.

Recombination takes place between islands and vacancies. Here it will be limited to the cases where a single adatom falls into a large vacancy island, and where a single vacancy gets filled from a larger island. Adatom-adatom and vacancy-vacancy dimer formations are also included.

The recombination kernel is of the same form as the aggregation kernel, but two more parameters need to be introduced here. The flux corresponding to the recombination can be now written as \( f_a = \gamma (\frac{1}{2} \sum_{i+j=s} K_{ij} n_in_j - \sum_{i=1}^{\infty} K_{is} n_in_s) \), for adatoms, and similarly for vacancies. The relative strength of recombination is controlled through \( \gamma \).

The detachment of single adatoms from islands is the only relevant mechanism of island breakup for small metallic clusters [4]. The probability of a single adatom to detach from an island is proportional to the length of the perimeter of a compact island, and therefore detachment kernel can be given by \( F_{ij} = F_0(i+j)^\alpha (\delta_{ii} + \delta_{ij}) \), where the parameter \( \alpha \) is fixed to \( \alpha = 1/2 \) in the following to reflect the assumption of detachment from the edge of compact islands. The prefactor \( F_0 \) gives the scale of detachment relative to aggregation. In simulations, the finite number of lattice sites is included into this parameter, and the notation is changed to
Figure 1. (a) The island size distribution function for different cases: no recombination and equal deposition of adatoms and creation of vacancies (solid discs), added recombination ($\gamma = 0.6$) (solid squares), and added strong recombination ($\gamma = 1.0$). The upper dotted line with solid triangles shows the island size distribution function and the lower one the vacancy size distribution function. For all shown cases $\theta = 0.25$. (b) The mean island size $\bar{s} = \left[ \sum_{i}^{\infty} s^2 n_s(\theta) \right]/\theta$ as a function of coverage $\theta$ is plotted for the cases in (a) (see legend for details). The case shown uses $(R, \kappa) = (10^5, 10^{-3})$.

$F_0$ to reflect their difference. An additional externally controlled parameter $\kappa = F_0/K_0$ is then introduced. With this parameterization the adatom flux due to detachment is

$$f_f = -\frac{1}{2} \sum_{i+j=s} F_{i,j} n_{i+j} + \sum_{i=1}^{\infty} F_{i,s} n_{i+s},$$

and similarly for the vacancies.

3. Results

We consider here case, where the two central control parameters $(R, \kappa) = (10^5, 10^{-3})$, where the product $\kappa R = 100$.

In Fig. 1(a) we show the adatom and vacancy island size distributions as a function of coverage for different recombination rates $\gamma$ at the maximum simulated coverage of $\theta = 0.25$. The results demonstrate the basic qualitative behaviour: steady growth of islands ($\gamma = 0.0$), slowed down growth ($\gamma = 0.6$) and terminated growth ($\gamma = 1.0$). In the last case the stationary island size is reached and the growth rates of vacancy and adatom islands are identical.

In Fig. 1(b) we show the mean size of the vacancy islands when the recombination rate $\gamma$ changes. In this case the maximum size is reached at different stages of growth, and the coverage $\hat{\theta}$ where this happens is defined as the turning point (or the point of the inflection) of the curve. The quantity $\hat{\theta}$ at the turning point gets smaller as $\gamma$ grows, and when $\gamma \to 1$, $\hat{\theta}$ attains values between 0.05 and 0.01, for $\xi$ between 0.8 and 0.2, respectively.

Fig. 2 shows the dependence of the maximum island size as a function of $\gamma$ for different values of $\xi$ varying from 0.2 to 1.0. It is interesting to note that the different curves representing the behaviour of the maximum island size can be collapsed to a single curve by using the fitting formula $\hat{s}_{\text{max}} = S_0 \exp(-\Gamma_k \gamma)$, where the prefactor can be fitted with a linear dependence on $\xi$ as $S_0 \propto \xi + \xi_0$, and where $\xi_0$ depends on $(R, \kappa)$. Similarly, we find that the coverage $\hat{\theta}$ at the maximum mean vacancy island size can be fitted by using the expression $\hat{\theta} = \Theta_0 \exp(-\Gamma_\theta \gamma)$. 

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4. Discussion and conclusions
In this work we have described a rate equation model for submonolayer growth in a situation where the flux of adatoms coming on the surface creates surface vacancies. The surface diffusion of these adatoms and single vacancies leads to island formation for both. The recombination process between adatoms and surface vacancy islands, and also between single surface vacancies and adatom islands, have been included in the model. We determined optimum conditions for submonolayer nanoisland growth in IBAD. This situation, which is characterized by the onset of smooth layer-by-layer growth, requires knowledge of the borderline case, where vacancy islands begin to be filled.

Before this borderline case is reached, there is steady growth of adatom and vacancy islands. However, the smaller the critical coverage below which this region of steady growth takes place, the sooner the vacancy islands start to fill and shrink away, and the more advantageous IBAD is in maintaining the desired layer-by-layer growth. Therefore, knowledge obtained in the present study about the critical size and the maximum island size and how they depend on the values of the control parameters, should be useful in tailoring growth through ion-assisted deposition.

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