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Shedding light on the nanoworld: diffusion-limited nanostructures from laser-focused atom deposition

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Abstract

We study the role of surface diffusion in the fabrication of nanostructures by laser-focused ("cold") atom lithography, using kinetic Monte Carlo simulations of a growth model that accounts for spontaneous organisation of 3D islands. Contrary to intuition, we find that the lateral size of the nanostructures is barely affected by surface diffusion of deposited adatoms, even when the adatom diffusion length exceeds the standing-wave field wavelength. ***To cite this article: F. Nita, A. Pimpinelli, C. R. Physique 5 (2004).***

Résumé

De la lumière sur le nanomonde : nanostructures limitées par la diffusion obtenues par dépôt d'atomes focalisés par laser. Le rôle de la diffusion de surface dans la fabrication de nanostructures par lithographie d'atomes focalisés par laser (atomes « froids »), est étudiée en utilisant des simulations Monte Carlo cinétiques d'un modèle de croissance qui tient compte de la formation spontanée d'îlots 3D. Contrairement à l'intuition, il s'avère que la taille latérale des nanostructures est peu sensible à la diffusion de surface des adatomes déposés, même lorsque la longueur de diffusion des adatomes dépasse la longueur d'onde de l'onde stationnaire des faisceaux laser. ***Pour citer cet article : F. Nita, A. Pimpinelli, C. R. Physique 5 (2004).***

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Mots-clés : Optique atomique ; Lithographie atomique ; Dépôt d'atomes focalisés par laser ; Nanostructures ; Nanofabrication

1. Introduction

There is a continued interest in the understanding, description and control of structures at the nanometer scales [1].

In particular, many kinds of atoms deposited on a foreign crystalline substrate, such as Ge over Si, are known to self-organize, that is to spontaneously aggregate into three dimensional nano-islands of very similar sizes (narrow island-size distribution), randomly distributed on the substrate [2]. Controlling the spatial distribution of the aggregates is a major technological problem.

On the other hand, atomic physicists have developed techniques based on the interaction between a free atom and laser light. These techniques, awarded with the 1997 Nobel Prize to C. Cohen-Tannoudji, S. Chu and W.D. Phillips, exploit the pressure exerted by radiation over matter for slowing down (i.e., *cooling*) atoms in a vapour, or in an atomic beam [3]. Such 'cold atoms' can be trapped by an appropriate arrangement of lasers, and then positioned at will, with high precision, in space [4–6].

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Such high precision in spatially positioning atoms have been employed to arrange 'cold atoms' from a beam onto a substrate according to a predetermined pattern, 'drawn' with the laser beams. To be able to use cold atoms to build regularly distributed nanostructures, the effect of surface diffusion after deposition has to be studied.

A first step in the investigation of the role of surface diffusion in cold atom deposition has been taken by McClelland and coworkers [7]. They have considered three different models, all of them defined on a one dimensional lattice, and have simulated the growth of a single nanostructure. The models were: (i) an atomic deposition model where atoms are deposited over preexisting columns (SOS condition), and only free and singly bonded atoms are allowed to diffuse, the diffusion rates obeying a thermally activated law; (ii) a ballistic deposition model where atoms stick where they touch the growing aggregate, even on the sides; (iii) a ballistic deposition model augmented with a relaxation rule that allows the deposited atoms to minimize their bonding energy. Both latter models are athermal by construction.

As the authors themselves remark, all three models are quite unsatisfactory, either a posteriori for their outcome (model (i)) or a priori (models (ii) and (iii)). Indeed, model (i) is not able to reproduce the experimental profile, while models (ii) and (iii) are not appropriate for describing deposition in molecular beam epitaxy (MBE) or similar growth techniques, even though the behaviour of the surface profile in model (iii) qualitatively resembles the experimental observations.

In the present work, we tackle the problem from the perspective of deposition, diffusion and aggregation models designed for MBE, accounting for the full two dimensional character of the process, and building in the essential ingredient of a driving force to spontaneous formation of three dimensional aggregates (islands).

2. Spontaneous formation of nanostructures

The self-assembly of deposited nanostructures is a complicated process, which is not yet fully understood [1]. In fact, when atoms of species A are deposited on a substrate B they have to fit into a crystal lattice that has, in general, a different lattice constant from their own. Elastic strains result, that affect adatom diffusion, as well as adatom attachment/detachment to clusters. Elastic effects are long-ranged, and if it is easy to take them into account (within linear elasticity) in a calculation that minimizes the total energy of the systems, in order to decide if three dimensional islands are more favourable compared to two dimensional ones, it is on the contrary quite hard to account for them in numerical calculations, such as kinetic Monte Carlo simulations, that would allow the investigation of the kinetics of the formation of three dimensional islands during deposition and growth [8,9].

For this reason, we will not attempt to make a model that quantitatively describes the formation of quantum dots. We will rather mimic very roughly one of the effects of strain on adatom diffusion, namely the increase of the diffusion barrier from the center of an island to its edge, as seen by a diffusing adatom on top of it. This effect is reproduced here qualitatively by introducing an addititional potential energy barrier for adatoms coming to the step edge, and hindering their descent to the lower level. Even though the physical origin is assumed here to be the elastic strain, this barrier is the analog of the Ehrlich–Schwoebel barrier in homoepitaxy [10].

The present model reproduces two essential features of quantum dots formation: first, it give rise to three dimensional islands (e.g., pyramids) of rather uniform size; second, the resulting islands form at an average distance ℓ_s that is principally determined by surface diffusion.

3. Simple models for cold atom deposition

As we have already mentioned in the first section of this paper, light can be used to deflect a neutral atomic beam during deposition onto a substrate [4–7]. So, if a laser beam is directed across the surface on a substrate and retroflected, a standing wave region will be created in the proximity of the surface. Also, if the wavelength corresponds to a resonance in the atom there is an induced electric dipole moment. This electric dipole moment will interact with the radiation field, its source in fact, and the force corresponding of this interaction is proportional to the gradient of the light intensity. The force is directed along the gradient of the light intensity so the atoms are pushed in the same direction. In conclusion it is possible to say that this standing wave region acts like an array of cylindrical lenses for the incoming atoms [11]. The distances between these lenses is $\lambda/2$ where λ is the optical wave length.

When it was supposed that the standing wave region can be assimilated with an array of cylindrical lenses it was taken into account the presence of only one laser radiation that is directed across the surface on the substrate. In this case, on the surface of the substrate, it can obtain a regular array of lines with a highly uniform height and width.

Experimentally it was established that the aspect of the nanoscale features depends on the growth conditions. So the height depends on the atomic beam intensity (deposition rate) and the deposition time. The width is a function of the atomic beam

collimation and the standing wave intensity and profile. By variation of these parameters one can obtain a desired nanoscale feature [12].

To examine the role played by each of these parameters we have performed Monte Carlo simulations. To modulate the deposition flux we used two shape functions:

$$f_1(y) = c + \frac{2\pi}{\lambda}\frac{\cos\frac{2\pi}{\lambda}y + 1}{\sin\frac{2\pi}{\lambda}L_y + \frac{2\pi}{\lambda}L_y}, \tag{1}$$

$$f_2(y) = c + \exp\left[\frac{-1}{|\cos\frac{2\pi}{\lambda}y|^{2k}}\right], \tag{2}$$

where λ is the wavelength and L_y is the dimension of the system in the y direction. The dimensions of the system are given in lattice parameter units, and the values of constants c and k were chosen to 0.03 and 5 respectively.

The deposition flux is spatially modulated by function (1) or (2). The modulation so obtained is one-directional, the flux being still uniform in the x direction. However, surface diffusion is two-dimensional, so that our model can also be used to study bi-directional flux profiles, and anisotropic substrates on which diffusion is different in the x and y directions. We will report on such investigations elsewhere.

Here we use a simple, isotropic diffusion model that accounts for spontaneous organization of 3D islands. The model is an SOS model on a simple cubic lattice, with periodic boundary conditions.

With respect to the interaction between an atom and its neighbours, we have taken into account only the interaction between nearest neighbours. Considering that the interaction energy between two such atoms is ε, for an atom the total interaction energy between it and its surrounding will be proportional to the number n of its neighbours, $-n\varepsilon$.

The diffusion process can be always described as a succession of elementary diffusion processes, an elementary diffusion process corresponding to a movement of an atom between two nearest neighbour sites. We have considered for the activation barrier a model in which the jump frequency depends on the energy in initial state, $-n_i\varepsilon$, according to

$$f = \nu\exp\left(-\frac{\varepsilon_d - n_i\varepsilon}{k_B T}\right), \tag{3}$$

where ν is the attempt frequency, k_B and T are the Boltzmann constant and the temperature; ε_d represents the activation barrier for an isolated atom ($n_i = 0$). For interlayer diffusion the activation energy becomes

$$\varepsilon_d + E_b - n_i\varepsilon, \tag{4}$$

where ε and ε_d have the same meaning as above and E_b is Schwoebel barrier.

Obviously the model described by the jump frequency given by Eq. (3) obeys detailed balance if there is no restriction on n_i values.

However, at low temperatures the motion of atoms with $n_i \geqslant 3$ is very unlikely. We used then a simplified model called Limited Diffusion Model (LDM), whose jump frequency is given by relation 3 where n_i has only three possible values: 0, 1, 2.

In other words we have taken into account the diffusion for isolated atom, and for the atoms that have one or two nearest neighbours. The diffusion for the atoms that have more than two nearest neighbours is forbidden. This model does not obey detailed balance, and cannot be valid at high temperature, nor too close to equilibrium. On the other hand, our choice is dictated by computational convenience, and nothing prevents in principle to relax our assumptions and to obey detailed balance.

Using LDM, simulations for different wavelengths of standing wave field and also for two different temperatures (100 K and 700 K) were made. The deposition rate was the same for all simulations: 1 monolayer/second (ML/sec). The values for ν, ε and ε_d were always the same: 10^{14} s^{-1}, 0.5 eV and 1 eV respectively. The simulations were performed for several values of the Schwoebel barrier, $E_b = 0$, $E_b = 1$ eV and $E_b = 2$ eV.

4. Results of kinetic Monte Carlo simulations: discussion

After completion of each layer the average surface profile along the direction of the light pattern, y, was computed.

At low temperature (100 K) we have performed simulations for only one wavelength of the modulation functions $\lambda = 500$ (we recall that all the distances are measured in lattice parameters). There are no differences between the results corresponding to different values of Schwoebel barrier and the average surface profile coincides with the corresponding flux profile. Indeed, at such low temperature the morphology of a surface is mainly determined by deposition, not by diffusion.

At high temperatures (700 K) the results are strongly dependent on the value of Schwoebel barrier. For all λ and $E_b = 0$, in the case of the f_1 deposition function, after deposition of 100 ML the surface flattens out completely (Fig. 1(a)).

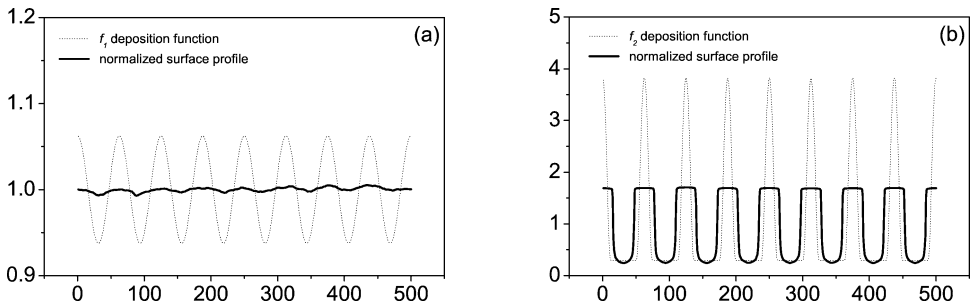

Fig. 1. The normalized surface profiles for LDM diffusion model after the deposition of 100 ML at 700 K in the case of (a) the f_1 function and (b) the f_2 function ($\lambda = 62.5$) and for $E_b = 0$.

Fig. 1. Les profiles normalisés de la surface pour les modèles de diffusion LDM, après dépôt de 100 MC à 700 K, dans le cas de la fonction (a) f_1 et de la fonction (b) f_2 ($\lambda = 62{,}5$) et pour $E_b = 0$.

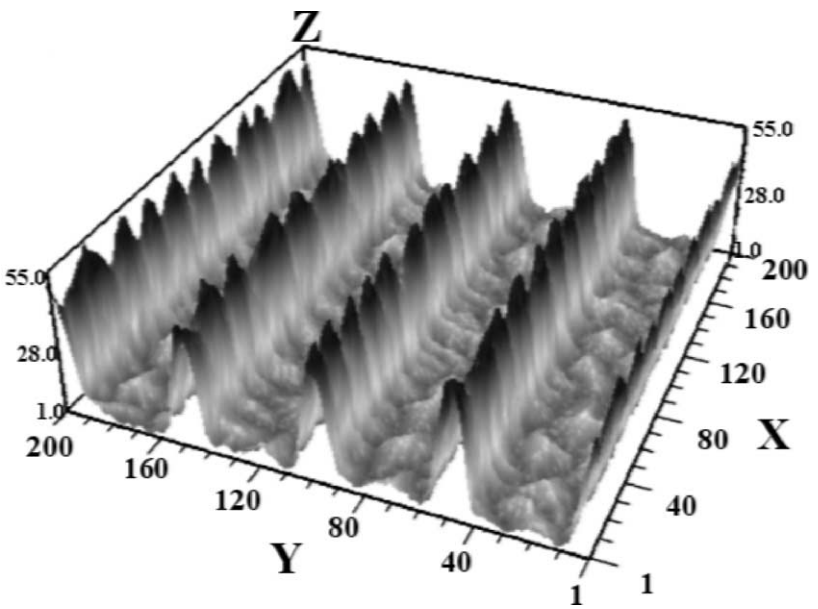

Fig. 2. The surface obtained after the deposition of 10 ML at 700 K for LDM diffusion model, in the case of the f_2 deposition function ($\lambda = 62.5$) and for $E_b = 2$ eV.

Fig. 2. La surface obtenue après dépôt de 10 MC à 700 K, pour le modèle de diffusion LDM, dans le cas de la fonction de dépôt f_2 ($\lambda = 62{,}5$) et pour $E_b = 2$ eV.

For the narrower f_2 deposition function for all λ and for $E_b = 0$ surface diffusion again acts so to flatten and widen the surface profile compared to the shape of the deposition flux (Fig. 1(b)).

Fig. 2 shows the surface that we have obtained for $E_b = 2$ eV after the deposition of 10 ML in the case of the f_2 deposition function and for $\lambda = 62.5$. The morphology of the simulated surfaces obtained using f_2 deposition function, is strikingly similar to the morphology of the real surfaces obtained by experiment. In particular, the average profile in the y direction reproduces well the profile of the as-deposited material. Also, the 'grain' structure of the ridges in the x direction, reproduces well the morphologies observed in real experiments with chromium on chromium [11].

5. Conclusions

Because the shape of the nanoscale features depends on the growth conditions, to assess the role played by each experimental parameter we have performed Monte Carlo simulations using a SOS growth model and two different deposition functions.

In qualitative agreement with experimental data we have observed that the feature width is a function of the standing wave profile, and also that the normalized height depends on the deposition time.

At low temperature there are no differences between the results corresponding to different values of Schwoebel barrier. This result is expected because at such temperatures the morphology of a surface is mainly influenced by the deposition parameters rather than by the diffusion parameters. At high temperatures the results are dependent on the Schwoebel barrier. This is also expected because at high temperatures the diffusion process is the most important process that determines the morphology of a surface.

Our results show that Kinetic Monte Carlo simulations of simple growth model can be used to reproduce a number of features observed in real cold atom deposition. Such simple models may then be quite helpful in predicting of more complex features, or the outcome of cold atom deposition on anisotropic, or pre-patterned surfaces, or in the presence of several deposited atomic species. We are now pursuing our investigations in these directions.

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