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SIMPLE MODELS FOR NANOCRYSTAL GROWTH

Pablo Jensen
Laboratoire de Physique de la Matière Condensée et des Nanostructures,
CNRS and Université Claude Bernard Lyon-1, 69622 Villeurbanne Cédex, France

1. Introduction

Growth of new materials with tailored properties is one of the most active research directions for physicists. As pointed out by Silvan Schweber in his brilliant analysis of the evolution of physics after World War II [1] "An important transformation has taken place in physics: As had previously happened in chemistry, an ever larger fraction of the efforts in the field were being devoted to the study of novelty rather than to the elucidation of fundamental laws and interactions [. . .] The successes of quantum mechanics at the atomic level immediately made it clear to the more perspicacious physicists that the laws behind the phenomena had been apprehended, that they could therefore control the behavior of simple macroscopic systems and, more importantly, that they could create new structures, new objects and new phenomena [. . .] Condensed matter physics has indeed become the study of systems that have never before existed. Phenomena such as superconductivity are genuine novelties in the universe."

Among these new materials, those obtained as thin films are of outstanding importance. Indeed, the possibility of growing thin films with desired properties is at the heart of the electronics technological revolution (for a nice introduction to the history of that revolution, see Ref. [2]). Thin film technology combines the three precious advantages of miniaturization, assembly line production (leading to low cost materials) and growth flexibility (depositing successively different materials to grow complex devices). Recently, the search for smaller and smaller devices lead to the new field of nanostructure growth, where one tries to obtain structures containing a few hundred atoms. As a consequence, an impressive quantity of deposition techniques have been developed to grow carefully controlled thin films and nanostructures from atomic deposition [3].

S. Yip (ed.),
Handbook of Materials Modeling, 1769–1785.

While most of these techniques are complex and keyed to specific applications, Molecular Beam Epitaxy (MBE) [4] has received much attention from physicists [5], mainly because of its (relative) simplicity. A younger technique, which seems promising to grow nanostructured materials with tailored properties is *cluster* deposition. Here, instead of atoms, one uses a beam of *preformed* large "molecules" containing typically 10–2000 atoms, the clusters. This technique has been shown to produce films with properties different from those obtained by the usual atomic beams. It is reviewed in Ref. [6] and will be considered no further in this short chapter.

Due to the technological impetus, a tremendous amount of both experimental and theoretical work has been carried out in this field, and it is impossible to summarize every aspect of it here. I will therefore concentrate on simple models adapted to understand the first stages of growth (the submonolayer regime).

2. Nanostructures: Why and How

As argued in the Introduction, the miniaturization logic naturally leads to trying to grow devices at the nanometer scale. This domain is very fashionable nowadays and the interested reader can find several information sources: for a simple and enjoyable introduction to the progressive miniaturization of electronics devices, see Ref. [7]. For more technical discussions, see for example Refs. [8, 9] and the journals entirely devoted to this field [10]. The reader is also referred to the enormous number of World Wide Web pages, especially those quoted in Ref. [13].

Besides the obvious advantages of miniaturization (for device speed and density on a chip), it has been argued [9] that the (magnetic, optical and mechanical) properties of nanostructured films can be intrinsically different from their macrocrystalline counterparts. For example, recent studies of the mechanical deformation properties of nanocrystalline copper [11] have shown that high strain can be reached before the appearance of plastic deformation, thanks to the high density of grain boundaries. Nanoparticles are also interesting as model catalysts [12].

The usual technology to grow thin films is deposition of atoms from a vapor onto a flat substrate. This technique was mainly used to grow relatively thick films (thickness larger than 100 nm typically). Recent developments with MBE allowed to control the growth at the atomic level, and for several materials it is possible to grow atomically flat surfaces over many micrometers. The same is true for interfaces in multilayer films, which are interesting for applications in electronics and magnetism. I refer the reader interested in the techniques and applications of atom deposition to several reviews [3].

I will focus here on a particular direction: the control of the *submonolayer* regime, i.e., before deposition of a single monolayer. There are two main interests: from the fundamental point of view, this regime allows a clearer determination of the atomic processes present during growth (the "elementary" processes to be described below). The models presented in this paper are useful in this regime and have allowed to understand and quantify many aspects of this regime of growth. One can also justify the study of the formation of the first layer since it is a template for the subsequent growth of the film [14, 15]. To grow a periodic array of nanometer islands of well-defined sizes, a promising direction seems to be the growth of strained islands by heteroepitaxy, stress being an ordering force which can lead to order [16]. However, growth in presence of elastic forces is beyond the capabilities of the present models which only take into account some of their effects (see below and Ref. [17]). Therefore I will not discuss this important subfield further.

There are already many good reviews on atomic deposition with different emphasis: for a simple introduction, see Refs. [5], for more technical presentations, see Refs. [18–21]. One can find also a comprehensive compilation of measurements and analysis of atomic diffusion [22] or one more specific to metal surfaces [23] or to metal atoms deposited on amorphous substrates [24] or on oxides [25].

3. Models of Atom Deposition

3.1. Introduction to Kinetic Monte Carlo Simulations

Given an experimental system, how can one predict the growth characteristics for a given set of parameters (substrate temperature, incoming flux of particles . . .)?

3.1.1. A bad idea: molecular dynamics simulations

A first idea – the "brute-force" approach – would be to run a molecular dynamics simulation (see Ref. [26]). It should be clear however that such an approach is bound to fail since the calculation time is far too large. The problem is that there is an intrinsically large time scale in the growth problem: the mean time needed to fill a significant fraction of the substrate with the incident atoms. An estimate of this time is fixed by t_{ML}, the time needed to fill a monolayer: $t_{\mathrm{ML}} \simeq 1/F$ where F is the atom flux expressed in monolayers per second (ML/s). Typically, the experimental values of the flux are lower than $1\,\mathrm{ML/s}$, leading to $t_{\mathrm{ML}} \geq 1\,\mathrm{s}$. Therefore, there is a time span of about 13 decades between the typical vibration time of an atom (approximately given by the Debye frequency 10^{-13} s, the lower time scale for the simulations) and t_{ML}, rendering hopeless any "brute-force" approach.

3.1.2. Choosing clever elementary processes

To reduce this time span, a more clever approach is needed. The idea is to "coarsen" the description by using elementary processes such as those sketched in Fig. 1. This idea is similar to the usual renormalization technique, but here one hides the shortest times (as one hides the highest energies) in "effective" parameters (see [1, 27] for a simple discussion on this point). For a discussion of the most relevant elementary processes for atomic deposition, see below and [18]. The rates of the different processes could in principle be calculated using the empirical or *ab initio* potentials, or be taken as parameters in the analysis. However, given the high number of possible processes it is more convenient to choose only some of them in the analysis. The advantage of this approach is that using a limited number of elementary processes allows to understand in detail their respective role in determining the growth characteristics. Moreover, a model with too many parameters can reproduce almost any experiment and it is dubious that meaningful comparisons can be obtained.

The drawback of the "elementary processes" approach is that before interpreting an experiment in the framework of one of these models one has to be sure that no other process than those chosen is present, for otherwise the interpretation could be meaningless. The case-in-point example for warning against a too rapid interpretation of experiments by elementary processes is

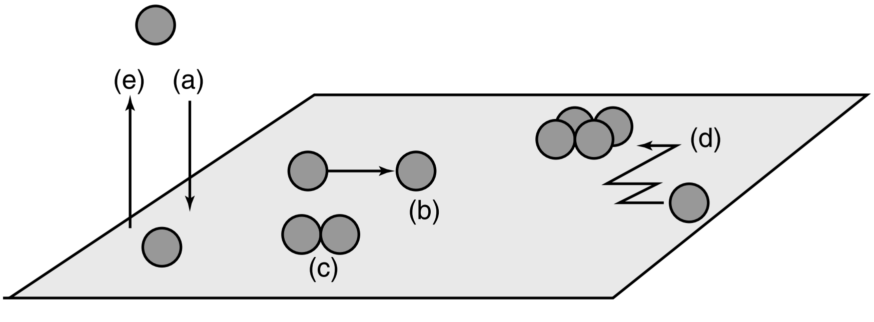

Figure 1. Main elementary processes considered in this paper for the growth of films by atom deposition. (a) adsorption of a atom by deposition; (b) and (d) diffusion of the isolated atoms on the substrate; (c) formation of an island of two monomers by juxtaposition of two monomers (nucleation) (d) growth of a supported island by incorporation of a diffusing atom (e) evaporation of an adsorbed atom.

the growth in the Pt/Pt(111) system. The initial experimental observations by the Comsa group had been thoroughly interpreted with a variety of elementary processes, only to discover, after several years, that the experimental results were determined by an unexpected process, not included in any of the simulations: contamination by CO adsorbates... See the full story in Ref. [28].

A simple physical rationale for choosing only a limited set of parameters is the following (see Fig. 2). For any given system, there will be a "hierarchy" of time scales, and the relevant ones for a growth experiment are those much lower than $t_{\mathrm{ML}} \simeq 1/F$. The others are too slow to act and can be neglected. The problem is that which processes are relevant or not depends on the precise system under study. For example, for typical metal on metal systems, the evaporation time is larger than the time needed to break a single bond. Thus, evaporation can be neglected in the analysis even at high temperatures where atoms can detach from islands. For metal atoms deposited on some insulating surfaces, the contrary might be true: since the bond between an adatom and the substrate may be weaker than the bond between two metal adatoms,

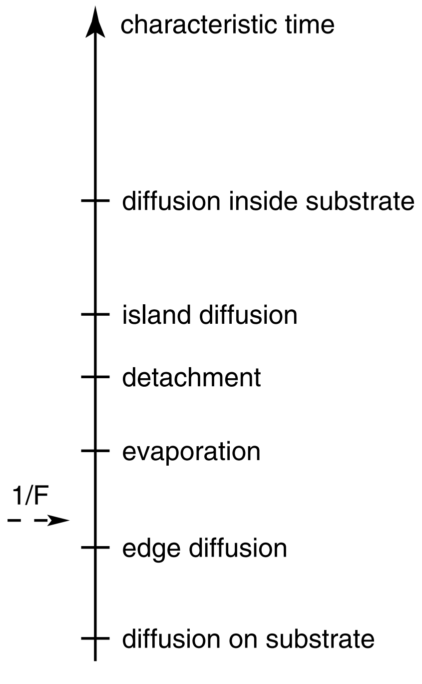

Figure 2. Time scales of some elementary processes considered in this paper for the growth of films by atomic deposition. The relevant processes are those whose timescale are smaller than the deposition time scale shown by the arrow in the left. In this case, models including only atom diffusion on the substrate and along the island (or step) edges are appropriate.

evaporation from the substrate occurs even at low temperatures for which islands are still stable and there is no adatom detachment.

3.1.3. Combining the elementary processes: Kinetic Monte Carlo

Now, given a set of elementary processes, there are two possibilities to predict the growth. The oldest is to write "rate-equations" which describe in a mean-field way the effect of these processes on the number of isolated atoms (called monomers) and islands of a given size. The first author to attempt such an approach for growth was Zinsmeister [29] in 1966, but the general approach is similar to the old rate-equations, first used by Smoluchovsky for particle aggregation [30]. Recently, Bales and Chrzan [35] have developed a more sophisticated self-consistent rate-equations approach which gives better results and allows to justify many of the approximations made in the past. However, these analytical approaches are mean-field in nature and cannot reproduce all the characteristics of the growth. Two known examples are the island morphology and the island size distribution (see [35] and also recent developments to improve the mean-field approach in [31].

There is an alternative approach to predict the growth: Kinetic Monte Carlo (KMC) simulations. Here one simply implements the processes chosen in a computer program with their respective rates and lets the computer simulate the growth. KMC simulations are an exact way to reproduce the growth, in the sense that they avoid any mean-field approximation. Given the calculation speed of present-day computers, systems containing up to 4000×4000 lattice sites can be simulated in a reasonable time (a few hours), which limits the finite size effects usually observed in this kind of simulation. Let me now discuss in some detail the way KMC simulations are implemented to reproduce the growth, once a set of processes has been defined, with their respective rates ν_{pro} taking arbitrary values or being derived from known potentials.

There are two main points to discuss here: the physical correctness of the dynamics and the calculation speed. Concerning the first point, it should be noted that, originally [32], Monte Carlo simulations aimed at the description of the *equations of state* of a system. Then, the MC method performs a "time" averaging of a model with (often artificial) stochastic kinetics: time plays the role of a label characterizing the sequential order of states, and need not be related to the physical times. One should be cautious therefore on the precise Monte Carlo scheme used for the simulation when attempting at describing the kinetics of a system, as in KMC simulations. Note that the KMC approach is fundamentally different from the usual Monte Carlo algorithm, where one looks for the equilibrium properties of a system, using the energy differences of the different configurations. Instead, in KMC, one is interested in the kinetics, using the different energy *barriers* for the transitions between the different configurations.

Let me address now the important problem of calculation speed. One could naively think of choosing a time interval Δt smaller than all the relevant times in the problem, and then repeat the following procedure:

1. choose one atom randomly
2. choose randomly one of the possible processes for this atom
3. calculate the probability p_{pro} of this process happening during the time interval $\Delta t \, (p_{\text{pro}} = \nu_{\text{pro}} \Delta t)$
4. throw a random number p_r and compare it with p_{pro}: if $p_{\text{pro}} < p_r$ perform the process, if not go to the next step
5. increase the time by Δt and go to (1)

This procedure leads to the correct kinetic evolution of the system but might be extremely slow if there is a large range of probabilities p_{pro} for the different processes (and therefore some $p_{\text{pro}} \ll 1$). The reason is that a significant fraction of the loops leads to rejected moves, i.e., to no evolution at all of the system.

Instead, Bortz *et al.* [33] have proposed a clever approach to eliminate *all* the rejected moves and thus reduce dramatically the computational times. The point is to choose not the atoms but the *processes*, according to their respective rate ν_{pro} and the number of possible ways of performing this process (called Ω_{pro}). This procedure can be represented schematically as follows:

1. update the list of the possible ways of performing every possible process Ω_{pro}
2. randomly select one of the process, *weighting* the probability of selection by the process rate ν_{pro} and Ω_{pro}: $p_{\text{pro}} = (\nu_{\text{pro}}\Omega_{\text{pro}}) \Big/ \left(\sum_{\text{processes}} \Omega_{\text{pro}}\nu_{\text{pro}}\right)$
3. randomly select a atom for performing this process
4. move the atom
5. increase the time by $\mathrm{d}t = \left(\sum_{\text{processes}} \Omega_{\text{pro}}\nu_{\text{pro}}\right)^{-1}$
6. goto (1)

This procedure implies a less intuitive increment of time, and one has to create (and update) a list of all the Ω_{pro} constantly, but the acceleration of the calculations is worth the effort.

A serious limitation of KMC approaches is that one has to assume a finite number of local environments to obtain a finite number of parameters. This confines KMC approaches to regular lattices, thus preventing a rigorous consideration of elastic relaxation, stress effects ... everything that affects not only the *number* of first or second nearest neighbors but also their precise position. Indeed, considering the precise position as in MD simulations introduces a *continuous* variable and leads to an infinite number of possible configurations or processes. Stress effects can be introduced approximately in KMC simulations [17] by allowing a variation of the bonding energy of an atom to an island as a function of the island size (the stress depending on the size), but it is unclear how meaningful these approaches are.

3.2. Basic Ingredients of the Growth

What is likely to occur when atoms are deposited on a surface? I will analyze in detail the following elementary processes: deposition, diffusion and evaporation of the atoms and their interaction on the surface (Fig. 1). The influence of surface defects which could act as traps for the atoms is also addressed.

The first ingredient of the growth, *deposition*, is quantified by the flux F, i.e., the number of atoms that are deposited on the surface per unit area and unit time. The flux is usually uniform in time, but in some experimental situations it can be pulsed, i.e., change from a constant value to 0 over a given period. Chopping the flux can affect the growth of the film significantly [36].

The second ingredient is the *diffusion* of the atoms which have reached the substrate. I assume that the diffusion is brownian, i.e., the atom undergoes a random walk on the substrate. To quantify the diffusion, one can use both the usual diffusion coefficient D or the diffusion time τ, i.e., the time needed by an atom to move by one diameter. These two quantities are connected by $D \sim d^2/(4\tau)$ where d is the hop length. The diffusion is here supposed to occur on a perfect substrate. Real surfaces always present some defects such as steps [37], vacancies or adsorbed chemical impurities. The presence of these defects on the surface can significantly alter the diffusion of the atoms and therefore the growth of the film.

A third process which could be present in growth is *re-evaporation* of the atoms from the substrate after a time τ_e. It is useful to define $X_S = \sqrt{D\tau_e}$ the mean diffusion length on the substrate before desorption.

The last simple process I will consider is the *interaction* between atoms. The simplest case is when (a) atoms ignore each other as long as they are not immediate neighbors (b) atoms attach irreversibly upon contact.

Point (a), commonplace in all simulations until recently, has been challenged by precise calculations of the potential felt by an atom approaching another atom or an island [38]. It has been shown that, for some systems, past the short range, a repulsive ring is formed around the adatoms (Fig. 3). The magnitude of the repulsion can be comparable to the diffusion barrier. Therefore, not taking this repulsive effect into account can lead to island densities much larger than experimentally observed. It remains to be seen how general this repulsive ring is.

Point (b) is not correct at high temperatures, because atom-atom bonds can be broken. This situation is discussed in Section 4.2.

The usual game for theoreticians is to combine these elementary processes and predict the growth of the film. However, experimentalists are interested in

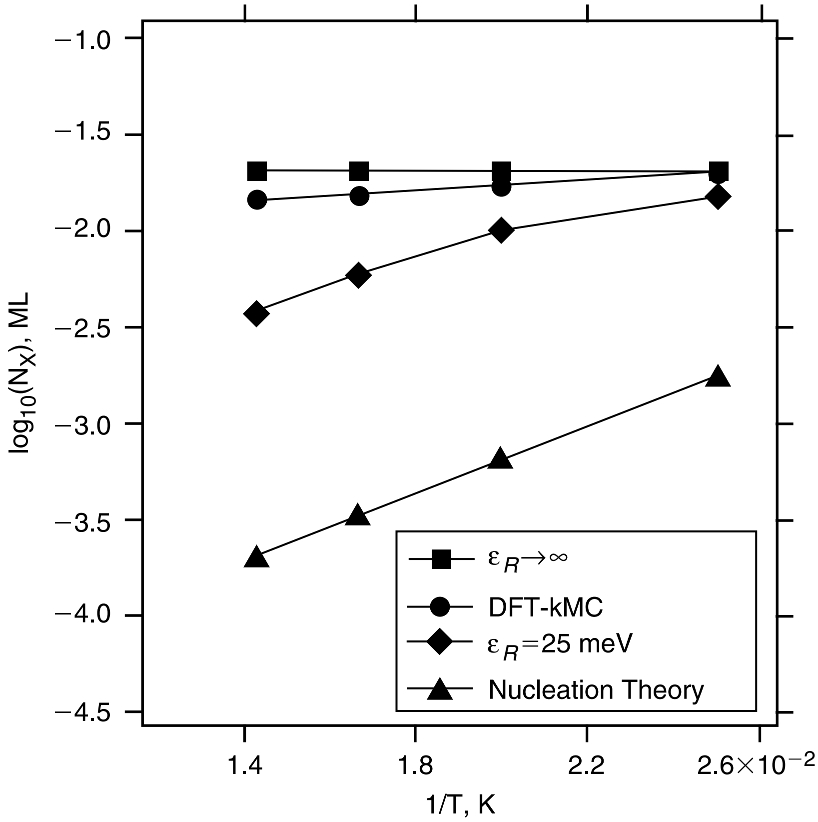

Figure 3. Arrhenius plot of the island density as a function of temperature from an impermeable repulsive ring (squares), a KMC model including the repulsion (circles), a simplified KMC model including a repulsive ring with 25 meV (diamonds), and nucleation theory (not including the repulsion efect (triangles). After Ref. [38].

the reverse strategy: from (a set of) experimental results, they wish to understand which elementary processes are actually present in their growth experiments and what are the magnitudes of each of them (this is what physicists call “understanding a phenomenon”). The problem, of course, is that with so many processes, many combinations will reproduce the same experiments. Then, some clever guesses are needed to first identify which processes are present. I gave several hints in a previous review [6] and will not address this question in detail here.

4. Predicting Growth with Computer Simulations

“Classical” studies [19] have focused on the evolution of the concentration of islands on the surface as a function of time, and especially on the saturation island density, i.e., the maximum of the island density observed before reaching a continuous film. The reason is of course the double possibility to calculate it from rate-equations and to measure it experimentally by conventional microscopy. I will show other interesting quantities such as island size distributions which are measurable experimentally and have been recently calculated by computer simulations [40, 41].

Since I am only interested in the *sub*monolayer regime, there is no need to take into account atoms falling on preexisting islands, except for the asymptotic case of strong evaporation discussed in Ref. [40]. Most metal on metal growth corresponds to this case, while metal on insulating surfaces grows by forming 3d islands (this is called the Wolmer–Weber growth mode, see for example Refs. [42]).

4.1. Two Dimensional Growth: Irreversible Aggregation

I first study the formation of the islands in the limiting case of irreversible aggregation, for two growth hypothesis: negligible or important evaporation.

4.1.1. Complete condensation

Let me start with the simplest case where only diffusion takes place on a perfect substrate (no evaporation). Figure 4a shows the evolution of the monomer (i.e., isolated atoms) and island densities as a function of deposition time.

We see that the monomer density rapidly grows, leading to a rapid increase of island density by monomer-monomer encounter on the surface. This goes on until the islands occupy a significant fraction of the surface, roughly 0.1%. Then, islands capture efficiently the monomers, whose density decreases. As a consequence, it becomes less probable to create more islands, and their number increases more slowly. When the coverage reaches a value close to 15%, coalescence starts to decrease the number of islands. The maximum number of islands at saturation N_{sat} is thus reached for coverages around 15%. Concerning the dependence of N_{sat} as a function of the model parameters, it has been shown that the maximum number of islands per unit area formed on the surface scales as $N_{\text{sat}} \simeq (F/D)^{1/3}$[19]. Simulations [6, 35, 39] and theoretical analysis [34] have shown (Fig. 6) that the precise relation is $N_{\text{sat}} = 0.53(F\tau)^{0.36}$ for the *ramified* islands produced by pure juxtaposition. This relation is very important since it allows, from an experimental measure of N_{sat}, to determine the value of τ (F is generally known), *provided* one knows that the simple hypothesis made are appropriate for the experiments.

To show that this limiting case is not only of theoretical interest, let me show an experimental example. Thanks to a technological innovation, a scanning

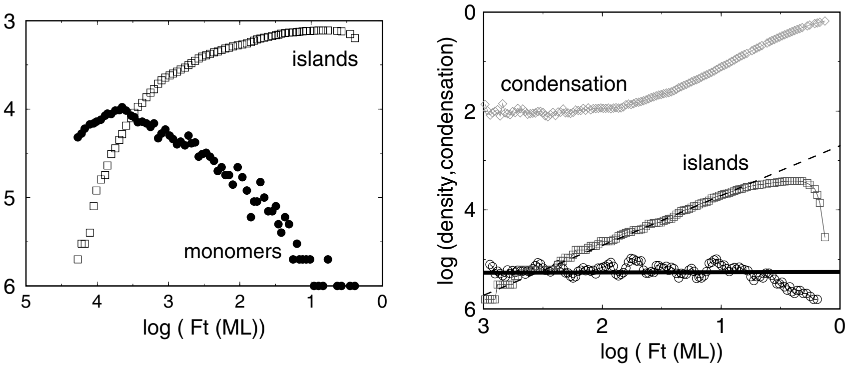

Figure 4. Evolution of the monomer and island densities as a function of the thickness (in monolayers), for islands formed by irreversible aggregation: (a) complete condensation, $F = 10^{-8}$, $\tau_e = 10^{10}$, $\tau = 1$ (leading to $X_S = 10^5$ and $\ell_{CC} = 22$) (b) important evaporation, $F = 10^{-8}$, $\tau_e = 600$ ($\tau = 1$) ($X_S = 25$ and $\ell_{CC} = 22$). ℓ_{CC} represents the mean island separation at saturation for the given fluxes when there is no evaporation [40]. The length units correspond to the atomic diameter. In (b) the "condensation" curve represents the total number of particles actually present on the surface divided by the total number of particles sent on the surface (Ft). It would be 1 for the complete condensation case, neglecting the monomers that are deposited on top of the islands. The solid line represents the constant value expected for the monomer concentration (equal to $F\tau_e$).

tunneling microscope operating a very low temperatures, a group in Lausanne University could observe, for the first time, the beginning of the growth of a film at the atomic scale [43]. Working at very low temperatures (50 K) is essential to "hide" many elementary processes (which cannot be thermally excited) and render the growth simple enough, so that the naive models of theoreticians can be relevant (for an introduction to the strategies used by physicists to understand nature, see [44]). Figure 5 shows that simple models as the ones presented in this paragraph are able, in these conditions, to reproduce in detail the experimental results.

4.1.2. Evaporation

What happens when evaporation is also included? Figure 4b shows that now the monomer density becomes roughly a *constant*, since it is now mainly determined by the balancing of deposition and evaporation. As expected, the constant concentration equals $F\tau_e$ (solid line). The number of islands increases linearly with time, since the island creation rate is given by the probability of atom-atom encounter, which is roughly proportional to the square atom concentration. We also notice that only a small fraction $(1/100)$ of the monomers do effectively remain on the substrate, as shown by the low condensation

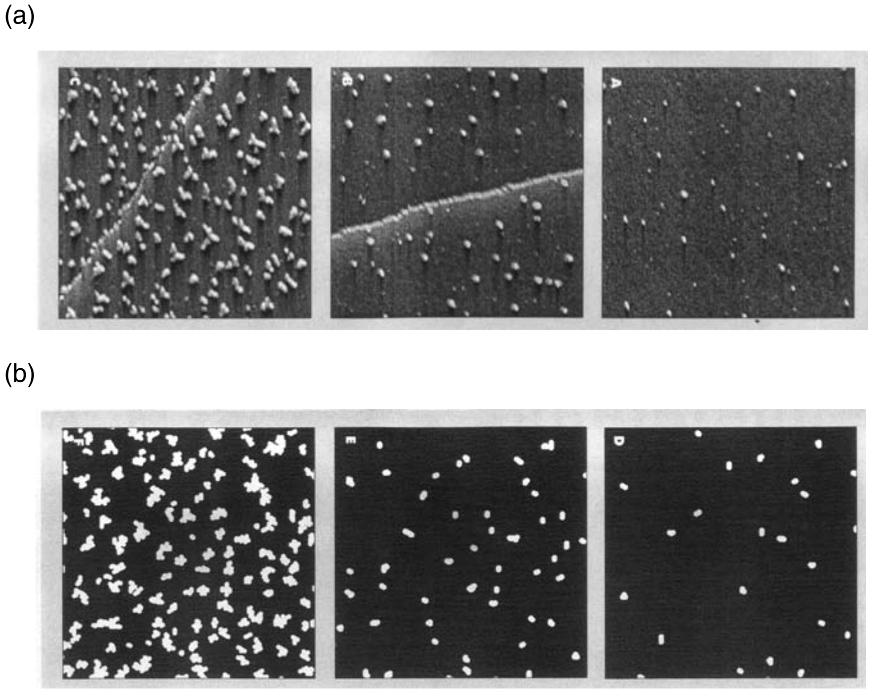

Figure 5. Comparison of the morphologies of experimental (silver atoms deposited on platinium, a–c) and predicted with KMC models (d–f) submonolayer films of different thicknesses (see text). These figures show a small portion of the surface, 160 atomic diameters wide. To adjust the experimental results, we had to take the following rates for the elementary processes: a diffusion hop every 2 ms, thirty atoms being deposited every second on this square.

coefficient value at early times. This can be understood by noting that the islands grow by capturing only the monomers that are deposited within their "capture zone" (comprised between two circles of radius R and $R + X_S$). The other monomers evaporate before reaching the islands. As in the case of complete condensation, when the islands occupy a significant fraction of the surface, they capture rapidly the monomers. This has two effects: the monomer density starts to decrease, and the condensation coefficient starts to increase. Shortly after, the island density saturates and starts to decrease because of island-island coalescence. Figure 6 shows the evolution of the maximum island density in the presence of evaporation. A detailed analysis of the effect of monomer evaporation on the growth is given in Ref. [40], where is also discussed the regime of "direct impingement" which arises when $X_S \leq 1$: islands are formed by *direct impingement* of incident atoms as first neighbors of adatoms, and grow by direct impingement of adatoms on the island boundary. An experimental observation of the evaporation regime can be found in Ref. [45].

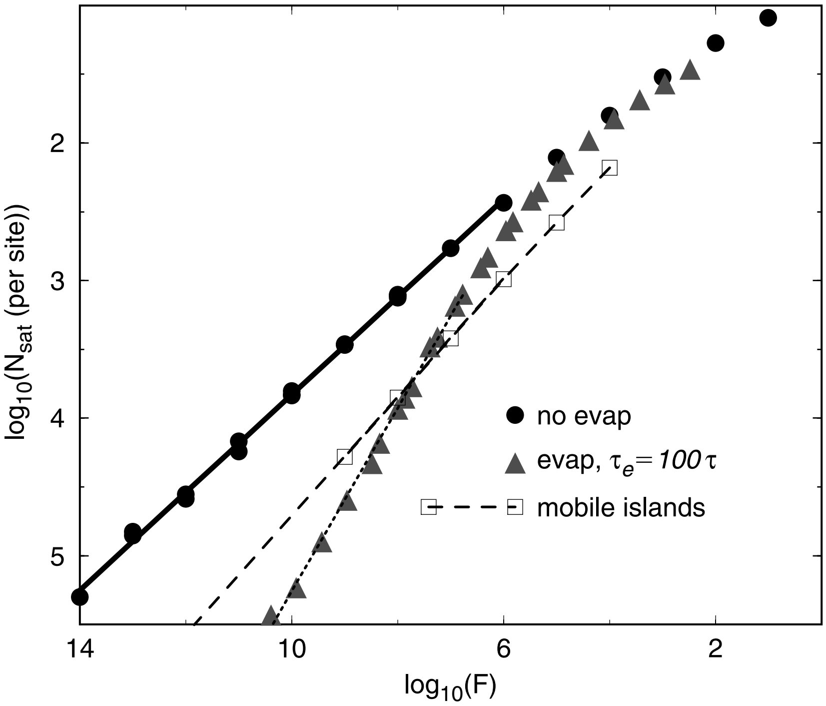

Figure 6. Saturation island density as a function of the flux for different growth hypothesis indicated on the figure, always in the case of island growth by irreversible aggregation. *"no evap"* (circles) means complete condensation. Triangles show the densities obtained if $\tau_e = 100\tau$. In the preceding cases, islands are supposed to be immobile. This hypothesis is relaxed for the last set of data, *"mobile islands"* (squares) , where island mobility is supposed to decrease as the inverse island size [39] (there is no evaporation). The dashed line is an extrapolation of the data for the low normalized fluxes. Fits of the different curves in the low-flux region give: *"no evap"* (solid line): $N_{\mathrm{sat}} = 0.53(F\tau)^{0.36}$; *"evap"*(dotted line): $N_{\mathrm{sat}} = 0.26F^{0.67}\tau^{-1/3}\tau_e$ (for the τ and τ_e exponents, see [40]) and *"mobile islands"* (dashed line): $N_{\mathrm{sat}} = 0.33(F\tau)^{0.42}$.

4.2. Reversible Aggregation

Previous results were obtained by assuming that atom-atom aggregation is irreversible. It is physically clear that at high temperatures atoms can detach from islands, and this has to be included in the models. The rate-equations approach [19] introduce a critical size i^* defined as follows: islands containing up to i^* atoms decay, while larger islands are stable. This means that only the concentration of sub-critical islands is in equilibrium with a gas of monomers. The concept of critical size was adopted for practical reasons (it simplifies the mathematical treatment) even if the macroscopic thermodynamical notions implicitly employed are difficult to justify for such small systems [18]. A more satisfactory approach was developed with the help of KMC simulations [41, 46]: instead of defining arbitrarily a critical size, one uses binding energies for atoms and studies which islands grow and decay. KMC simulations have shown that the morphology of the submonolayer films change dramatically from ramified to compact islands as the ratio of bond energy to substrate temperature is varied (Fig. 7) and that the critical size is ill defined, the control parameter being the ratio of the dimer dissociation rate to the rate of adatom capture by dimers [41, 46]:

$$\lambda = \frac{N_2/\tau_1}{D\rho N_2} \tag{1}$$

where τ_1 is the mean time for a dimer to dissociate, D is the diffusion constant for monomers and ρ, N_2 represent the densities of adatoms and dimers respectively. The case $\lambda \sim 0$ represents irreversible aggregation whereas large λ values mean that islands can dissociate easily.

In the case of reversible atomic aggregation, the scope is to determine the aggregation parameter λ (defined in Eq. 1). This can be done in several ways [41]:

1. By studying the flux dependence of N_{sat}: the exponent depends on λ;
2. By measuring the island size distribution which also uniquely depends on λ;
3. By measuring the nucleation rate and studying its dependence on the incident flux.

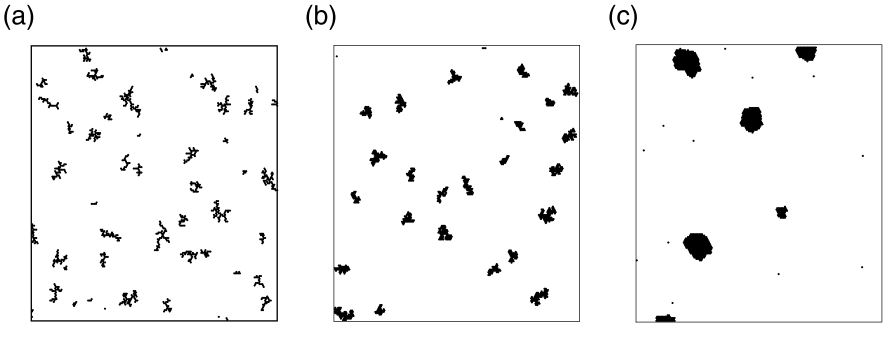

Figure 7. Morphology of the films obtained with reversible aggregation for atomic deposition with different atom-atom bond energies. The temperature is fixed to 400 K, the activation energy for diffusion of isolated atoms to 0.45 eV, the flux to 1 ML/s and the thickness to 0.03 ML. The bond energies are: (a) 0.5 eV, (b) 0.2 eV and (c) 0.1 eV.

Once λ has been found, it is in principle possible to extract the microscopic parameters, even if in practice uncertainties remain because of the limited amount of experimental data generally available and the high number of fit parameters (for examples of such fits see [15, 41, 46].

5. Conclusion

Modeling crystal growth is a rapidly evolving field. This is due to rapid developments in the experimental side: near-field microscopy (for example, scanning tunneling microscopy), control of the growth conditions (low temperature, vacuum). Thanks to all these improvements, experiments can be carried out on "theoretical" surfaces, namely carefully controlled surfaces similar to those that theoreticians can study. On the theoretical side, better algorithms to combine the different growth ingredients have been developed, and we now have better methods to predict atom-atom interaction (mainly the *ab initio* approach). For a recent informal review, see [47]. Many challenges remain, however: predicting, from atomistic level simulations, the behavior of the system on a macroscopic scale, which is difficult mainly when several intermediate scales are relevant (for example if elastic interactions are important); predicting, from precise simulations carried out over static configurations or, at best, nanoseconds, the behavior of a system over seconds or hours. These are not challenges only for surface science but also for physics in general (modeling of brittle or ductile fracture, ageing phenomena...), which leaves some hope that other fields will help us solving our problems!

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