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Mean field approach for describing thin film morphology

M Tomellini¹ and M Fanfoni²

¹ Dipartimento di Scienze e Tecnologie Chimiche, Università di Roma Tor Vergata, Via della Ricerca Scientifica, 00133 Roma, Italy

² Dipartimento di Fisica, Università di Roma Tor Vergata, Via della Ricerca Scientifica, 00133 Roma, Italy

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Abstract

A mean field approach is employed for determining the kinetics of two main quantities for film morphology, namely the number of islands and the film perimeter. These two quantities have been computed for both coalescence and impingement growth regimes. For each case the growth of two- and three-dimensional islands has been considered. In addition, the effect of the spatial correlation among nuclei has been taken into account on the grounds of the hard core interaction. Analytic formulae have been derived for each case.

1. Introduction

Vapour condensation on solid substrates takes place thanks to the occurrence of microscopic processes, such as adsorption, surface diffusion and monomer aggregation, which usually occurs under non-equilibrium conditions [1]. Ultimately, the final morphology of a film stems from the interplay among the kinetics of those processes. Several physical quantities are used for characterizing the film morphology [2], such as the number density of islands $N(\Theta)$, the fraction of substrate surface covered by the film Θ , the perimeter of the film $\Gamma(\Theta)$, the surface width $w(\Theta)$, and the height–height correlation function $G(r, \Theta)$, r being the relative distance of two points on the substrate surface. All these quantities are experimentally accessible by means of an appropriate microscopy. On the theoretical side several methods are employed for modelling the film growth [2]: scaling analysis, rate equations, theory of stochastic processes and kinetic Monte Carlo (KMC). Recently, the interplay between rate equations and the theory of stochastic processes of dots has been discussed in [3] in the framework of the mean field (MF) hypothesis.

One of the first articles where the mean field approach to film growth was applied dates back to 1971 [4]. The author, Vincent, evaluated the number density of islands in a coalescence regime [1, 5]. His approach was subsequently improved by Briscoe and Galvin [6], who modelled the density of islands and the fractional surface coverage in film growth governed by the coalescence mechanism by means of kinetic equations and statistical arguments. In this context, mean field is meant as the possibility to treat the kinetics employing a couple

of parameters, i.e. the average island dimension and the relative standard deviation of the island distribution. Recently, as far as the island density behaviour is concerned, by exploiting the statistical mechanics of rigid discs, we have shown [7] that the island density in a two-dimensional (2D) coalescence mechanism, within the binary collision approximation, scales as

$$n(\Theta) = \frac{N(\Theta)}{N(0)} = (1 - \Theta)^{2a} e^{-\frac{2(1-a)\Theta}{1-\Theta}}, \quad (1)$$

where $a \approx 1/2$. Equation (1) in the limit $\Theta \rightarrow 0$ leads to the well-known result $n(\Theta) \simeq 1 - 2\Theta$ [8, 9]. However, the film growth can also proceed in such a way that redistribution of matter among islands does not take place after collision. This mechanism is referred to as impingement. With an unexpected outcome, as shown in [9], the behaviour of $n(\Theta)$ is found to be the same for both coalescence and impingement although the evolution of Θ is completely different for the two mechanisms.

In the case of nucleation and growth ruled by impingement the Θ kinetics is given by the Kolmogorov–Johnson–Mehl–Avrami (KJMA) formula, which holds for Poissonian nucleation [3]. Besides, on the basis of the KJMA approach other quantities representative of the film morphology have been computed analytically: these are the film roughness and total perimeter of the film $\Gamma(\Theta)$ [10–12]. The latter is the length (per unit substrate area) of the border of the projection of the islands on the substrate. This approach has been extensively employed for describing the growth of diamond film at a deformed silicon substrate, where the nucleation was simultaneous and the growth governed by impingement [13, 14]. On the other hand, to the best of our knowledge, analytical modelling based on the MF theory of the film perimeters for 2D and 3D growths, in the coalescence regime, are still lacking. As outlined below, providing such a model is one of the purposes of this paper.

In the impingement case the mean field approach allowed us to evaluate $n(\Theta)$ and $\Theta(t)$ for a non-random arrangement of the nucleation centres, t being the running time. The computation was performed in the specific case of hard core interaction and successfully compared with Monte Carlo simulations. The latter were performed on a 300×300 square lattice with 90 nucleation centres. The clusters were allowed to grow linearly with time and cyclic boundary conditions were taken into account. The uncertainty of the simulated $n(\Theta)$ is within the size of the open symbol and, when correlation was included, $\Theta^* = 0.8$ was used (figure 5, see section 2.2 for the definition of Θ^*). Even under these circumstances an MF theory providing analytic solutions to the kinetics of the perimeter is lacking both for coalescence and impingement mechanisms.

The purpose of the present contribution is to present a comprehensive MF analysis of 2D and 3D film morphologies, as driven by impingement and coalescence mechanisms, in terms of film *perimeter* and *island density* kinetic quantities. The study is also extended to tackle both the cases of random and non-random distribution of nucleation centres. The paper is organized as follows. Section 2.1 is devoted to the computation of the perimeter in the 2D and 3D cases for a Poissonian distribution of nuclei. MF computations are also derived for $n(\Theta)$, $\Theta(t)$ and $\Gamma(\Theta)$ quantities. In section 2.2 the MF approach is extended to treat the correlated nucleation case in the framework of the hard core interaction.

2. Results and discussion

2.1. Random distribution of nuclei

2.1.1. 2D island growth. We will deal with the simultaneous nucleation case, which is that all nuclei start growing at $t = 0$, t being the running time. Nuclei have a circular shape and their

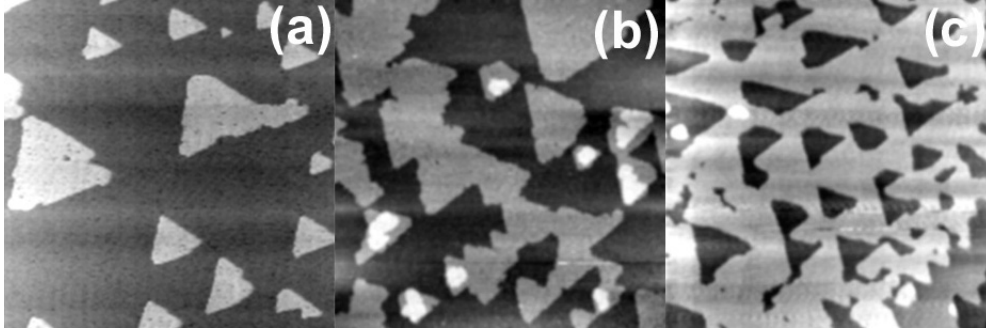


Figure 1. STM images of a Si(111) surface after a Ge deposition (L) at 500 °C: (a) $(200 \times 200) \text{ nm}^2$, $L = 0.8 \text{ ML}$; (b) $(200 \times 200) \text{ nm}^2$, $L = 1.5 \text{ ML}$; (c) $(770 \times 770) \text{ nm}^2$, $L = 2.0 \text{ ML}$. The pictures show that the growth mechanism is basically impingement and that nucleation is not simultaneous. In panel (c) the coverage is beyond the percolation threshold that, for triangles, is about $\Theta \simeq 0.5$ [9].

size increases according to an *a priori* microscopic growth law that is the same for all nuclei. Islands result by the collision of more nuclei. In the impingement case (figure 1) the nuclei maintain their individuality within the islands, whereas in the coalescence regime the island shape is conserved during the collision process.

It is well known that in the former case $\Theta = 1 - E_v(R)$, where $E_v(R)$ is the probability of finding a region of area πR^2 centred at some arbitrary point empty of nucleus centres, $R \equiv R(t)$ is the radius of the nucleus at time t and for a Poissonian process $E_v(R) = e^{-\pi N_0 R^2} = e^{-\Theta_e}$, where N_0 is the density of nuclei. The perimeter is promptly obtained through $\Gamma = -\frac{\partial E_v}{\partial R} = -\frac{\partial \Theta_e}{\partial R} \frac{\partial E_v}{\partial \Theta_e}$ and can be recast as a function of Θ as [12]

$$\Gamma(\Theta) = 2\sqrt{\pi N_0}(1 - \Theta) \left[\ln \frac{1}{1 - \Theta} \right]^{1/2}. \quad (2)$$

As far as the impingement process is concerned, it is worth recalling that the $\Gamma(\Theta)$ kinetics is the same for both 2D and 3D nuclei provided the shape of the projection of the nucleus on the substrate is the same. A similar consideration holds for the $n(\Theta)$ behaviour. Besides, equation (2) has been applied to experimental data to derive the number of initial islands in the limit of simultaneous nucleation [13, 15] or exploited to determine the energetics of homoepitaxy of 2D systems [16].

For island growth with coalescence the mean field approach is based on the concept of average island, namely, at any given coverage (time) all islands have the same shape and appropriate size (discs in the case under discussion). The expressions for the surface coverage and the film perimeter are $\Theta = N_0 n(\Theta) \pi D_{(2)}^2 / 4$ and $\Gamma(\Theta) = N_0 n(\Theta) \pi D_{(1)}$ respectively, where $D_{(k)} = (\sum_i D_i^k / N)^{1/k}$ is the mean diameter on the k -basis [6]. This is a proper way for estimating the averages of k -dimensional quantities. In fact $D_{(1)}$ is the average obtained from the total perimeter while $D_{(2)}$ is from the total coverage. It goes without saying that, in general, $D_{(1)} \neq D_{(2)}$. Briscoe and Galvin showed that for a relatively narrow distribution the ratio between the various means can be approximated as

$$\frac{D_{(k+b)}}{D_{(k)}} = m^b, \quad (3)$$

where b is a real number and m is in the range 1.00–1.05. The MF expression of the film perimeter therefore reads

$$\Gamma(\Theta) = \frac{2}{m} \sqrt{N_0 \pi n(\Theta) \Theta}, \quad (4)$$

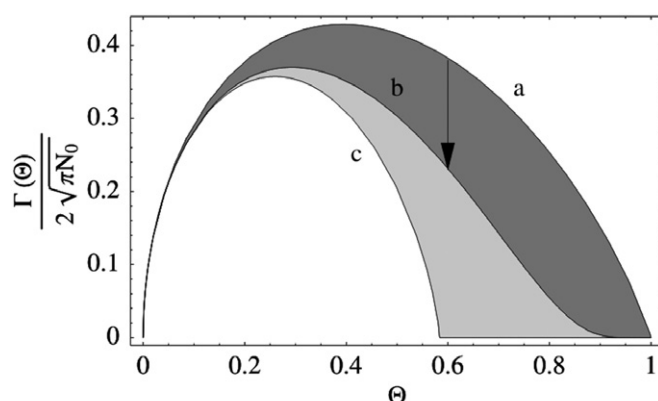


Figure 2. Film perimeter as a function of coverage for 2D and 3D islands: random nucleation case. (a) Impingement; (b) coalescence of 2D islands; (c) coalescence of 3D islands. The grey zone between the (a) and (b) curves accounts for the growth mechanism intermediate between impingement and coalescence.

where use was made of equation (3). According to equation (4), the coverage dependence of the perimeter is related to the kinetics of the island density.

Equation (4) also holds for 3D island growth provided the appropriate $n(\Theta)$ function is employed (see section 2.1.2).

The perimeter of the 2D film is obtained by plugging equation (1) into equation (4) as

$$\Gamma(\Theta) = \frac{2}{m} \sqrt{N_0 \pi} [\Theta(1 - \Theta)]^{1/2} e^{-\frac{\Theta}{2(1-\Theta)}}. \quad (5)$$

The behaviour of the normalized perimeters for the coalescence and the impingement cases is shown in figure 2 for $m = 1$. As is apparent, at any coverage value the growth ruled by the coalescence mechanism exhibits a lower perimeter value. For instance at $\Theta = 0.5$ the normalized difference is equal to 0.1129. Because of the universality of the $n(\Theta)$ function with respect to the mechanism of island collision [7, 9] in the partial coalescence case the $\Gamma(\Theta)$ curve is expected to fall in the shaded area of figure 2.

2.1.2. 3D island growth. In this subsection we determine the kinetics of the film perimeter for hemispherical islands by employing equation (4). To this end it is necessary to evaluate $n(\Theta)$ for the 3D case resorting to the binary-collision model [7] according to which

$$\frac{dn}{n} \equiv \frac{dN}{N} = -\frac{1}{2} H_p(\sigma) d\sigma, \quad (6)$$

where σ is the disc diameter and $H_p(\sigma) d\sigma$ is the probability that, given a disc at some arbitrary position, the centre of the nearest disc lies between σ and $\sigma + d\sigma$. In this modelling the hard disc coverage is equal to

$$\Theta = N\pi\sigma^2/4, \quad (7)$$

which implies $\sigma \equiv D_{(2)}$. To solve equation (6) for $n(\Theta)$ it is necessary to relate the increments of the disc diameter and of the surface coverage. This is done by making use of the *intrinsic* growth law of the island as introduced by Briscoe and Galvin in [6]. The changing rate of the surface coverage can be expressed according to

$$d\Theta = \frac{N\pi\sigma}{2} d\sigma + \frac{\pi\sigma^2}{4} dN, \quad (8)$$

as obtained by carrying out the derivation of equation (7). Although simple, the meaning of this equation is a little subtle. In fact, in equation (8) the increment $d\sigma$, in addition to the obvious variation due to the intrinsic growth, takes into account the variation induced by the events of coalescence. Alternatively, the coverage kinetics can be rewritten by decoupling the contributions due to the intrinsic growth of the island and to the coalescence process [6] as

$$d\Theta = \frac{N\pi\sigma}{2} (d\sigma)_I + \frac{\kappa\pi\sigma^2}{4} dN, \tag{9}$$

where $(d\sigma)_I$ is the intrinsic growth rate of the average island and the second term of the equation accounts for the coverage change caused by the pairwise coalescence events. In particular, $\kappa = 2 - 2^{2/3} \approx 0.41$ if both mass and shape are conserved in the collision. As far as equation (6) is concerned, we note that in this expression $d\sigma$ has to be identified with $(d\sigma)_I$; this is because the diameter increment is intended just before the collision event takes place. From equation (9) we get

$$(d\sigma)_I = (N\pi\sigma/2)^{-1} (d\Theta - \kappa\pi\sigma^2 dN/4). \tag{10}$$

Moreover from [17]

$$H_p(\sigma) = \frac{8\Theta(1-a\Theta)}{\sigma(1-\Theta)^2}, \tag{11}$$

which, together with equation (10) allows us to rewrite equation (6) as

$$\frac{dn}{n} = -\frac{2(1-a\Theta)}{(1-\Theta)^2 - 2\kappa\Theta(1-a\Theta)} d\Theta. \tag{12}$$

It is worth noting that for 2D growth the island area is conserved in coalescence, thus $\kappa = 0$ and equation (1) is the solution of equation (12) [7]. For $a = 1/2$ the solution of equation (12) reads

$$n(\Theta) = \left[\frac{(1 - \frac{\Theta}{1-\lambda})^{1+\lambda}}{(1 - \frac{\Theta}{1+\lambda})^{1-\lambda}} \right]^{\frac{(1-\lambda^2)}{2\lambda}}, \tag{13}$$

where $\lambda = \sqrt{\frac{\kappa}{\kappa+1}}$. Equation (13) holds for $\Theta < \Theta_c = 1 - \lambda$, where Θ_c is one of the roots of the denominator of equation (12). The validity of the modelling has been tested by means of Monte Carlo (MC) simulation of 3D growth of hemispherical islands. The result is shown in figure 3, where the best fit of equation (13) to the MC output is obtained for $\Theta_c = 0.58$, that is $\kappa = 0.21$. This value has to be considered as the average κ value over the entire kinetics. The evaluation of the film perimeter is eventually performed by inserting equation (13) in equation (4). Its behaviour is displayed in figure 2, where it can be compared with the curve representative of the impingement mechanism which, in the Θ domain, holds for 3D film growth, too. However, a more revealing comparison between impingement and coalescence is obtained by expressing the perimeter as a function of film thickness $h = Ft/\rho$, F being the flux of the condensing atoms and ρ the density of the film. To this end it is necessary to evaluate $\Theta \equiv \Theta(h)$. As regards the impingement mechanism, this is computed through the KJMA theory according to $\Theta = 1 - e^{-\pi N_0 R^2}$, where the nucleus growth is assumed to proceed by direct capture of gas monomer at the nucleus surface. Under these circumstances, by denoting with s the area of the gas–nucleus interface (of a generic nucleus) at any time of the kinetics, we get $sF dt = s\rho dR$, that is $R(t) = Ft/\rho = h$. Accordingly, the KJMA kinetics reads $\Theta(\eta) = 1 - e^{-\pi\eta^2}$, with $\eta = h\sqrt{N_0}$. On the other hand, since in the coalescence case $\Theta = N\pi D_{(2)}^2/4$ and $h = N\pi D_{(3)}^3/12$, by using equation (3) we end up with the relationship

$$\xi(\eta) = \Theta^{-3}n(\Theta), \tag{14}$$

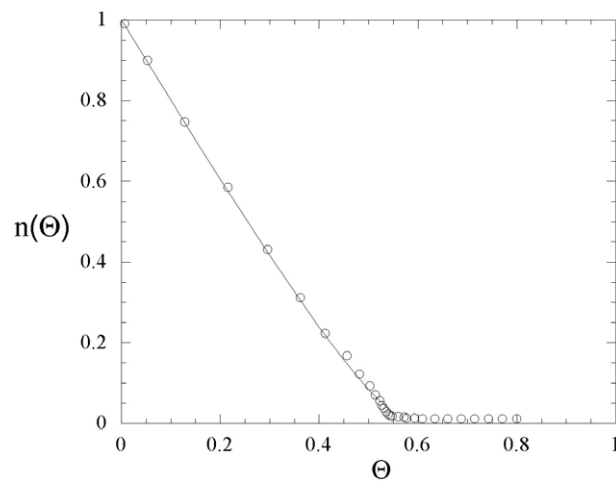


Figure 3. Normalized number of islands for 3D island growth in coalescence regime as a function of coverage.

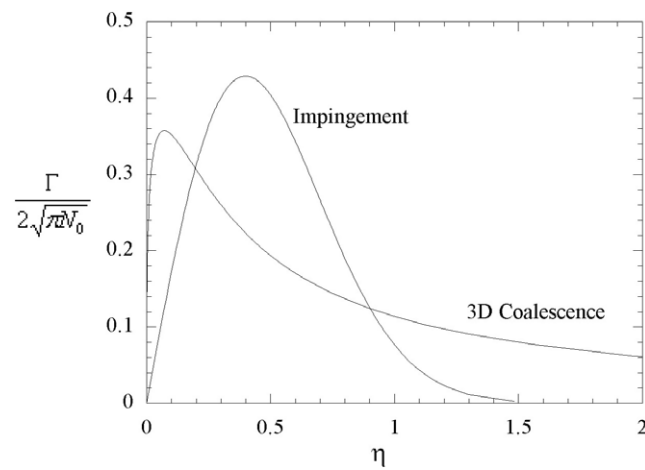


Figure 4. Film perimeter for 3D island growth as a function of the film thickness, $\eta = \frac{Et}{\rho} \sqrt{N_0}$, for impingement and coalescence regimes.

where $\xi(\eta) = 4m^6(9\pi\eta^2)^{-1}$ and $n(\Theta)$ is given by equation (13). The comparison between the film perimeters in the thickness domain is shown in figure 4, where the kinetics is reported as a function of η for $m = 1$.

For completeness it must be said that the two mechanisms, with respect to the growth law, have been treated in a slightly different manner. In fact, in the impingement case we did not consider the contribution to island growth arising from the fraction of adatoms coming from the uncovered substrate. The inclusion of this contribution determines, at most, a variation of 15% in the $\Theta(\eta)$ kinetics. Nevertheless the difference shown in figure 4 between the two mechanisms remains striking, because of the jamming point characteristic of coalescence. The jamming point, namely the asymptotic value of the surface coverage, is due to the balance between the reduction of covered area, per pair of coalescence events, and the intrinsic growth. It goes without saying that this argument makes sense for a statistical ensemble of islands. A

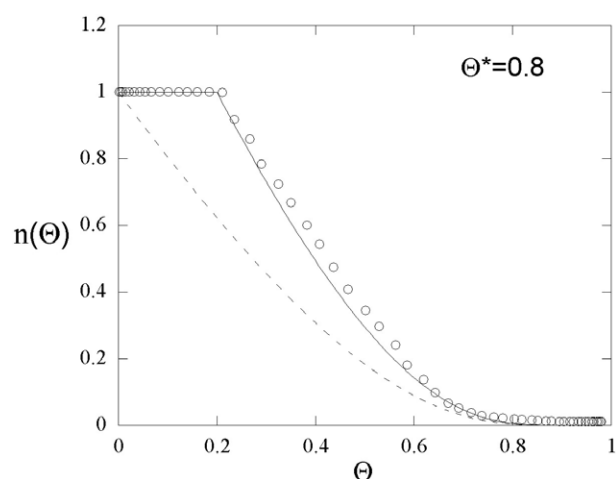


Figure 5. Normalized number of islands for 2D island growth in the coalescence regime as a function of coverage. The broken line refers to random nucleation. Open circles are the Monte Carlo output for hard core correlated nucleation. The degree of correlation is given by the quantity Θ^* defined in the text. The full line is the curve obtained from equation (15).

similar comment also applies to the results displayed in figure 2 for the 3D island where, at the jamming point ($\Theta \simeq 0.6$), the film perimeter is different from zero. However, once normalized to the square root of the nucleation density, this quantity is in fact negligible on the linear scale.

2.2. Spatially correlated nuclei: hard core model

Insofar as equation (4) also holds for a non-random distribution of nucleation centres, the perimeter evolution rests on the determination of $n(\Theta)$ both in two dimensions and three dimensions. Let us now consider the growth of spatially correlated nucleation centres according to the hard core interaction [18], i.e. the distance between two nuclei not being less than, say, σ_{hc} . As a consequence $n(\Theta) = 1$ as long as $\Theta \leq \pi N_0 \sigma_{\text{hc}}^2 / 4 = \Theta^* / 4$. In the framework of the MF approach one also observes that for $\Theta > \Theta^* / 4$ the system can be thought of as a random arrangement of hard discs for which the kinetics equation (6) applies. Integration of equation (12) at $\Theta > \Theta^* / 4$ gives rise to

$$n(\Theta)|_{\Theta > \Theta^*/4} = \left(\frac{n(\Theta)}{n(\Theta^*/4)} \right), \quad (15)$$

where $n(\Theta)$ is given by the solution of equation (12). Equation (15) holds for both 2D and 3D growth with coalescence. In fact, as reported above, the 2D case is recovered by setting $\kappa = 0$ in equation (12).

2.2.1. 2D island growth. In the 2D case, the validity of the MF model has been tested through MC simulation. In particular, the analytical solution, equation (15), has been compared to MC simulation of growth with impingement at $\Theta^* = 0.8$. This is justified by assuming the same kinetics for both growths even in the non-random case [9]. The results reported in figure 5 show that the agreement between the two kinetics is satisfactory. The kinetics of the film perimeter is evaluated through equation (4) as displayed in figure 6.

The next step is to evaluate the film perimeter for growths governed by impingement of spatially correlated nuclei. Very recently we have shown that in the case of impingement and

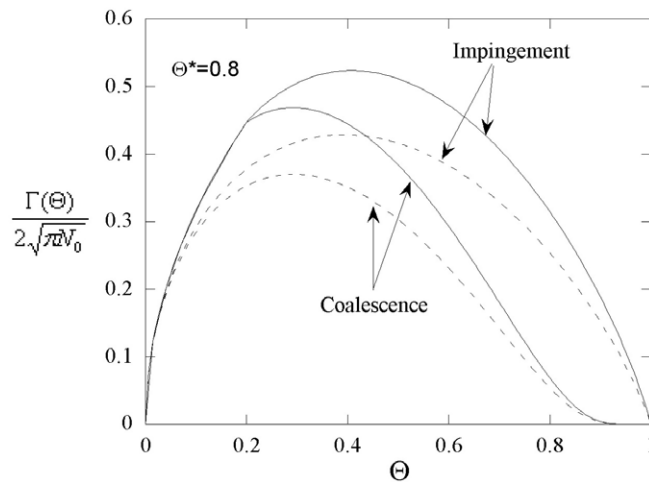


Figure 6. Comparison between correlated (full line) and uncorrelated (broken line) nucleations for both 2D coalescence and impingement regimes. The degree of correlation is given by the quantity Θ^* defined in the text.

hard core correlation [7]

$$E_v(\Theta_e; \Theta^*) = (1 - \Theta_e) \chi_{[0, \frac{\Theta_e^*}{4}]}(\Theta_e) + \left(1 - \frac{\Theta^*}{4}\right) e^{-\frac{\Theta^*}{4(1-\Theta^*/4)^2} \left[4 \frac{\Theta_e}{\Theta^*} - \sqrt{\Theta^* \Theta_e} + \frac{\Theta^*}{2} - 1\right]} \chi_{[\frac{\Theta_e^*}{4}, \infty)}(\Theta_e) \quad (16)$$

where $\Theta_e = N_0 \pi R^2$ is the so-called extended surface and the indicator function is defined as $\chi_A(x) = 1$ for $x \in A$ and it is equal to zero for $x \notin A$. Thus the kinetics of the perimeter can be computed as ($a = 1/2$)

$$\Gamma(\Theta) = -2\sqrt{\pi N_0 \Theta_e} \frac{\partial E_v}{\partial \Theta_e} = 2\sqrt{\pi N_0} \left\{ \sqrt{\Theta_e} \chi_{[0, \frac{\Theta_e^*}{4}]}(\Theta_e) + \frac{(1 - \Theta)}{(1 - \Theta^*/4)^2} \left(\sqrt{\Theta_e(\Theta)} - \frac{\Theta^*}{8} \sqrt{\Theta^*} \right) \chi_{[\frac{\Theta_e^*}{4}, \infty)}(\Theta_e) \right\} \quad (17)$$

where

$$\Theta_e(\Theta) = \frac{\Theta^*}{64} \left\{ \Theta^* + \left[(4 - \Theta^*)^2 \left(1 - \frac{4}{\Theta^*} \ln \frac{1 - \Theta}{1 - \Theta^*/4} \right) \right]^{1/2} \right\}^2 \quad (18)$$

as computed from the equation $E_v(\Theta_e; \Theta^*) = 1 - \Theta$. The behaviour of equation (17) is also displayed in figure 6.

2.2.2. 3D island growth. The application of the MF approach to 3D coalescence of correlated nuclei can be performed, using equations (15) and (13). The kinetics of the island density is shown in figure 7 for $\Theta^* = 0.8$ and it is compared to that of the random case. The corresponding perimeters are reported in figure 8 as a function of coverage. According to figure 8 one observes that the impact of the correlation among nuclei on film perimeter is important almost in the entire range of coverage. On the other hand, as far as the fractional coverage is concerned, the arrangement of nuclei seems to play a less important role as witnessed by figure 9, which displays the behaviour of the coverage as a function of film

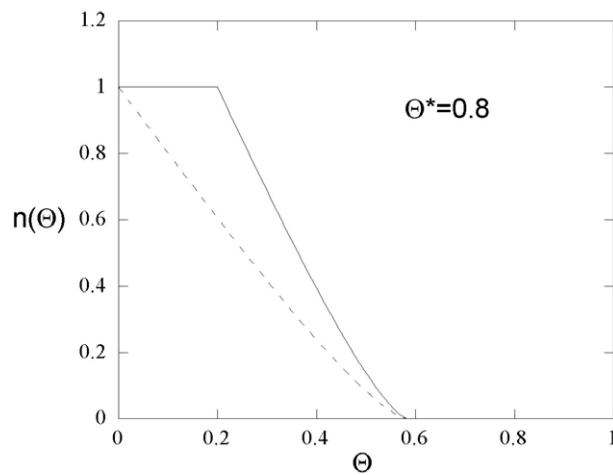


Figure 7. Normalized number of islands for 3D island growth in the coalescence regime as a function of coverage. The broken line refers to random nucleation. The degree of correlation is given by the quantity Θ^* defined in the text. The full line is the curve obtained from equation (15).

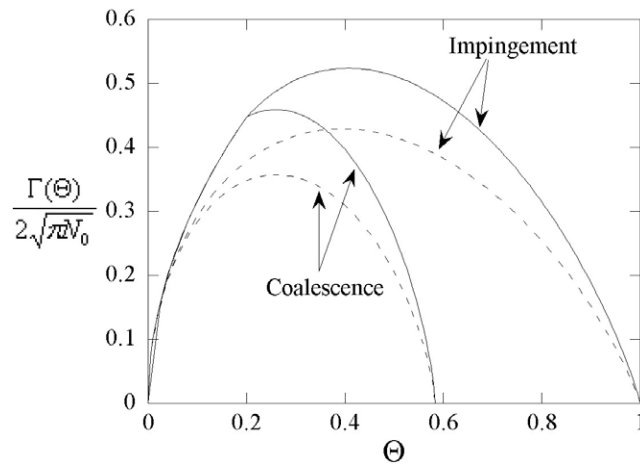


Figure 8. Comparison between correlated (full line) and uncorrelated (broken line) nucleations for both 3D coalescence and impingement regimes. The degree of correlation is given by the quantity Θ^* defined in the text.

thickness. The $\Theta(\eta)$ curves (random ($\Theta^* = 0$) and correlated ($\Theta^* = 0.8$) nucleation) have been computed through equations (13)–(15), where $n(\Theta)|_{\Theta < \Theta^*/4} = 1$.

The morphology of the film growth ruled by the impingement has also been investigated in comparison to the coalescence growth mode. The results are shown in figures 8 and 9 for the perimeter and the fraction of coverage, respectively. It is worth pointing out that unlike the impingement case, in the coalescence regime, due to the 3D island shape, the closure of the film is never achieved whatever the thickness of the film.

The MF analysis indicates a dramatic impact of the growth morphology on the film perimeter. In this context, we speculate that such a property of the system can be exploited to measure physical quantities linked to the perimeter. Let us assume that, at a given coverage

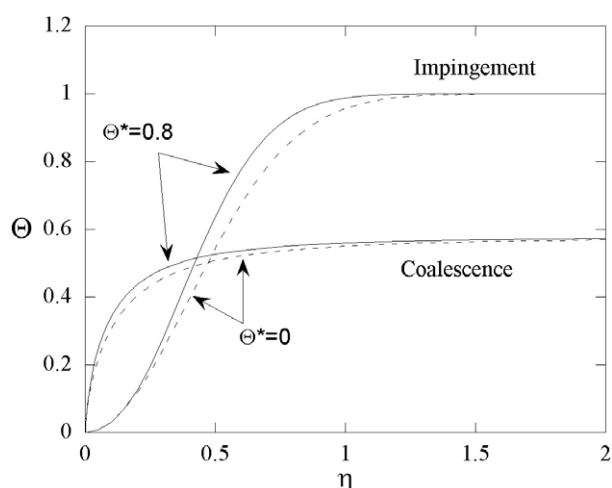


Figure 9. Comparison between correlated (full line) and uncorrelated (broken line) kinetics of surface coverage, for both 3D coalescence and impingement regimes, as a function of the film thickness, $\eta = \frac{Ft}{\rho} \sqrt{N_0}$. The degree of correlation is given by the quantity Θ^* defined in the text.

value, and for a given correlation degree, the system can undergo a transition from a metastable to a stable morphology. For example, we think of the case where a temperature increase (which implies a larger mobility of the atoms) triggers an impingement–coalescence transition. By assuming that the transition only affects the shape of the islands, island number being constant, for 2D islands the film perimeter changes along the arrow of figure 2. The grey zone between the two curves accounts for the growth mechanism intermediate between impingement and coalescence. The energetics of this process reads $\Delta E_{2D,2D} = \gamma_{c,v} h \Delta \Gamma(\Theta)$, where $\gamma_{c,v}$ is the surface energy of the island–vacuum interface, and h is the island thickness. For $\Delta \Gamma(\Theta)/2\sqrt{N_0\pi} = -0.1$ (that is $\Theta \approx 0.6$), $N_0 \approx (10^{10}–10^{12}) \text{ cm}^{-2}$, $\gamma_{c,v} \approx 10^{-4} \text{ J cm}^{-2}$ and $h = 10^{-7} \text{ cm}$ one gets $\Delta E_{2D,2D} \approx -(100–10) \mu\text{J cm}^{-2}$, figures that are within the resolution of a microcalorimetric measurement [19]. The transition towards a stable state is witnessed by the exothermicity of the process. In principle the knowledge of $\Delta E_{2D,2D}$ allows one to estimate $\gamma_{c,v}$. In addition, thanks to the very high heating rate available with this technique, it could also be possible to measure the transition kinetics under non-isothermal conditions. By assuming the relaxation process towards equilibrium ruled by atom mobility at the island periphery, the kinetics reads $\frac{d\Gamma}{dt} = -K_{\Gamma}(\Gamma - \Gamma_e)$, K_{Γ} being the rate coefficient, and $\frac{dE_{2D,2D}}{dt} = \gamma_{c,v} h \frac{d\Gamma}{dt}$. By inserting the $\frac{d\Gamma}{dt}$ expression as computed by integrating the rate equation for Γ , we end up with $\frac{dE_{2D,2D}}{dt} = -\gamma_{c,v} h K_{\Gamma}(\Gamma_0 - \Gamma_e) \exp(-\int_{T_0}^T (K_{\Gamma}/\dot{T}') dT')$, where $\Gamma_0(\Gamma_e)$ is the initial (equilibrium) perimeter, $T(t)$ is the temperature and T_0 is the initial temperature.

In the case of a 2D \rightarrow 3D-impingement \rightarrow coalescence transition involving hemispherical islands, it is possible to show that $\Delta E_{2D,3D} = \gamma_{c,v}(3\Theta_e - 2\Theta_i) - \beta(\Theta_e - \Theta_i)$, where β is the adhesion work, Θ_i is the fractional coverage of the 2D film and $\Theta_e = (3h/2)^{2/3}(\pi N_0 n_{2D}(\Theta_i))^{1/3} \Theta_i^{2/3}$ is the coverage at equilibrium. Figure 10 shows the $\Delta E_{2D,3D}$ versus Θ_i function for $N_0 \approx 10^{12} \text{ cm}^{-2}$, $\gamma_{c,v} \approx 10^{-4} \text{ J cm}^{-2}$, $h = 10^{-7} \text{ cm}$ and several values of $\beta/\gamma_{c,v}$ that, in order to ensure stable 3D islands, ought to be lower than 2 [20]. The figure shows that even in this case the energetics of the process could be exploited for measuring the adhesion work provided $\gamma_{c,v}$ is known and the fractional coverage is sufficiently large. In figure 10 the typical experimental resolution ($10^{-5} \text{ J cm}^{-2}$) is indicated by a horizontal line.

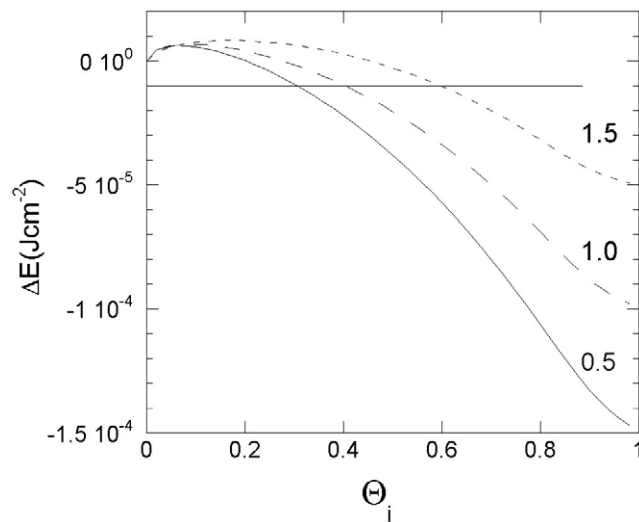


Figure 10. Energetics of the 2D \rightarrow 3D transition as a function of the 2D film coverage (Θ_i). The curves are plotted for different figures of the dimensionless ratio between the adhesion work and the surface energy, $\beta/\gamma_{c,v}$.

In summary, we have shown that by means of a MF approach two important quantities characterizing the morphology of a thin film, namely the island density and the total cluster perimeter, can be attained in analytic form. In doing this the two extreme growth modes, impingement and coalescence, have been taken into account, distinguishing also between 2D and 3D regimes, for both random and correlated arrangement of clusters.

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References

- [1] Zinke-Allmann M 1999 *Thin Solid Films* **346** 1
- [2] Barabasi A-L and Stanley H E 1995 *Fractal Concepts in Surface Growth* (Cambridge: Cambridge University Press)
- [3] Fanfoni M and Tomellini M 2005 *J. Phys.:Condens. Matter* **17** R571
- [4] Vincent R 1971 *Proc. R. Soc. A* **321** 53
- [5] Jensen P 1999 *Rev. Mod. Phys.* **71** 1695
- [6] Briscoe B J and Galvin K P 1991 *Phys. Rev. A* **43** 1906
- [7] Tomellini M and Fanfoni M 2005 *Phys. Rev. B* **72** 155407
- [8] Stowell M J and Hutchinson T E 1971 *Thin Solid Films* **8** 41
- [9] Fanfoni M, Tomellini M and Volpe M 2001 *Phys. Rev. B* **64** 075409
- [10] Trofimov V I and Park H S 2003 *Appl. Surf. Sci.* **219** 93
- [11] Pacchiarotti B, Fanfoni M and Tomellini M 2005 *Physica A* **358** 379
- [12] Tomellini M and Fanfoni M 1996 *Surf. Sci.* **349** L191
- [13] Polini R, Tomellini M, Fanfoni M and Le Normande F 1997 *Surf. Sci.* **373** 230
- [14] Fanfoni M, Polini R, Sessa V, Tomellini M and Volpe M 1999 *Appl. Surf. Sci.* **152** 126
- [15] Motta N, Sgarlata A, Calarco R, Nguyen Q, CastroCal J, Patella F, Balzarotti A and De Crescenzi M 1998 *Surf. Sci.* **406** 254

-
- [16] Markov I 1997 *Phys. Rev. B* **56** 12544
 - [17] Torquato S, Lu B and Rubinstein J 1990 *Phys. Rev. A* **41** 2059
 - [18] Hermann H, Mattern N, Roth S and Uebele P 1997 *Phys. Rev. B* **56** 13888
 - [19] Allen L H and Lai S L 1998 *Microscale Thermophys. Eng.* **2** 11
 - [20] Budevski E, Staikov G and Lorenz W J 1996 *Electrochemical Phase Formation and Growth* (New York: VCH) chapter 4