Morphological stability of islands upon thin-film condensation

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The stability of shape of a disklike cluster growing on a substrate from supersaturated vapor of adsorbed particles is studied. The investigation is carried out by expansion in terms of cosines of a minor deflection of island shape from a disk and subsequent evaluation of expansion coefficient relations in time. In addition to diffusion of adatoms towards an island and the relation between an equilibrium concentration and curvature, some kinetic phenomena on the surface of an island, the interface energy anisotropy, and surface diffusion are taken into account. It is shown that the reevaporation process of adatoms from a substrate leads to a considerable change in the criteria of instability initiation of the island shape as compared to the three-dimensional case. The mode with number ν starts to be excited only in the case where the radius of a cluster lies within a certain interval, $R_1(\nu) < R < R_2(\nu)$. The criterion of island faceting is derived as well.

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I. INTRODUCTION

As is well known, investigation of nucleation and growth processes of thin films is of great scientific and industrial interest [1,2]. A great deal of attention is given to the examination of growth mechanisms of separate clusters on a substrate. A theory of cluster growth from vapor on a substrate is constructed in the stationary approximation in Ref. [3]. It is shown there that the growth due to absorption of adatoms from the substrate surface predominates over the immediate growth from the vapor. It was also found that when the incorporation of adatoms into an island proceeds sufficiently slowly, this process restricts the growth of clusters. The diffusion process of adsorbed atoms towards an island is a limiting one [3]. A method for considering the influence of other neighbor islands in the course of diffusion growth is proposed in Ref. [4]. A theory of nonstationary diffusion growth of an island with allowances for the motion of its boundary is advanced in Ref. [5]. It is shown there that nonstationary effects are essential when the size of an island is smaller than the diffusion mean-free-path length. The influence of nonisothermal effects on nucleation and growth of islands is studied in Ref. [6]. It is assumed in all models that surface clusters do not change their shape during the course of growth. In actual fact, numerous experiments show that the shape of clusters changes in time [1,2,7]. In some cases the shape tends to an equilibrium shape or, on the contrary, becomes unstable. The fact that the shape changes greatly influences both the film nucleation [8] and the Ostwald ripening stages [9]. In the three-dimensional case the morphological stability of a spherical particle growing from solution or melt was thoroughly studied in Ref. [10]. This growth turned out to be classical. In this current work we use the technique outlined in Ref. [10] for the case of thin-film growth and take into account a number of additional effects.

II. MORPHOLOGICAL STABILITY OF A DISK-SHAPED CLUSTER GROWING DUE TO DIFFUSION OF ADATOMS

First we analyze the most simple model. We assume that the shape of a plane cluster with height h, which grows on a substrate due to diffusion of adatoms, slightly differs from the shape of a disk with height h and radius R_0. Then, the cluster surface equation in polar coordinates (in the system connected with the cluster center) is of the form

$$R(\varphi)=R_0\left[1+\sum_{\nu=0}^{\infty}\varepsilon_\nu\cos\nu\varphi\right], \tag{1}$$

where $\varepsilon_\nu \ll 1$ are the coefficients of expansion for a cluster shape perturbation from a disk in terms of cosines; φ is the angle in polar coordinates that measured from the axis of symmetry of the cluster. The change of value ε_ν in time denotes a change in the shape of an island during the course of growth. The curvature of an island shape at an each point is equal to

$$K=\frac{1}{R}-\frac{R''}{R^2}=\frac{1}{R_0}\left[1+\sum_{\nu=0}^{\infty}(\nu^2-1)\varepsilon_\nu\cos\nu\varphi\right]. \tag{2}$$

Terms containing ε_ν powers in Eq. (2), which are higher than the first one, are dropped. Therefore, the concentration of adatoms in equilibrium with a cluster of the given form is easily found, namely,

$$n_e=n_0\exp(\gamma K)=n_0e^{\gamma/R_0}\left[1+\frac{\gamma}{R_0}\sum_{\nu=0}^{\infty}(\nu^2-1)\varepsilon_\nu\cos\nu\varphi\right], \tag{3}$$

where n_0 is the equilibrium concentration of adatoms, $\gamma=\sigma w/kT$, σ is the interface energy that is so far assumed to be isotropic, w is the volume occupied by a particle in the cluster, k is the Boltzmann constant, and T is the temperature. The distribution of adatoms around an island in the quasistationary approximation [3,6] is described by the isotropic diffusion equation,

$$D\left[\frac{\partial^2 n}{\partial r^2}+\frac{1}{r}\frac{\partial n}{\partial r}+\frac{1}{r^2}\frac{\partial^2 n}{\partial\varphi^2}\right]+J-\frac{n}{\tau}=0\ ,$$
$$n(\infty,\varphi)=J\tau\ , \tag{4}$$
$$n(R(\varphi),\varphi)=n_0 e^{\gamma/R_0}\left[1+\frac{\gamma}{R_0}\sum_{\nu=0}^{\infty}(\nu^2-1)\varepsilon_\nu\cos\nu\varphi\right]\ ,$$

where D is the isotropic diffusion coefficient, J is the flow density of atoms evaporated on a substrate, and τ is the characteristic time of adatom reevaporation. Here it is assumed that incorporation of atoms into an island occurs very rapidly; therefore, the concentration of adatoms near the cluster boundary coincides with n_e. From Eqs. (1) and (4) the distribution of adatoms over the substrate is found,

$$n(r,\varphi)=J\tau-(J\tau-n_0e^{\gamma/R_0})\frac{K_0(r/\sqrt{D\tau})}{K_0(R_0/\sqrt{D\tau})}+\sum_{\nu=0}^{\infty}\frac{K_\nu(r/\sqrt{D\tau})}{K_\nu(R_0/\sqrt{D\tau})}\left[(\nu^2-1)n_0\frac{\gamma}{R_0}-\frac{R_0}{\sqrt{D\tau}}(J\tau-n_0e^{\gamma/R_0})\frac{K_1(R_0/\sqrt{D\tau})}{K_0(R_0/\sqrt{D\tau})}\right]\varepsilon_\nu\cos\nu\varphi\ , \tag{5}$$

where K_ν is the Macdonald function of ν of the order [11]. The island growth rate, v, is mainly determined by the diffusion flow of adatoms of the island boundary [10], i.e.,

$$v=\frac{dR_0}{dt}\left[1+\sum_{\nu=0}^{\infty}\varepsilon_\nu\cos\nu\varphi\right]+R_0\sum_{\nu=0}^{\infty}\frac{d\varepsilon_\nu}{dt}\cos\nu\varphi=\frac{Dw}{h}\left[\frac{\partial n}{\partial r}\right]_{r=R(\varphi)}\ . \tag{6}$$

Substituting Eq. (5) into Eq. (6) and equating coefficients for $\cos\nu\varphi$, we finally have

$$\frac{dR_0}{dt}=\frac{Dwn_0}{h\sqrt{D\tau}}(e^{\gamma/R_c}-e^{\gamma/R_0})\frac{K_1}{K_0}\ , \tag{7}$$

$$\frac{d\varepsilon_\nu}{dt}=\frac{wn_0}{h\tau}(e^{\gamma/R_c}-e^{\gamma/R_0})\left\{\frac{K_1}{K_0}\left[(\nu-1)\frac{\sqrt{D\tau}}{R_0}+\frac{K_{\nu-1}}{K_\nu}\right]-1-\frac{\sqrt{D\tau}}{R_0}\left[(\nu^2-1)\frac{\gamma/R_0}{e^{\gamma/R_c}-e^{\gamma/R_0}}\left[\frac{K_{\nu-1}}{K_\nu}+\nu\frac{\sqrt{D\tau}}{R_0}\right]+\frac{K_1}{K_0}\right]\right\}\varepsilon_\nu\ . \tag{8}$$

Here, R_c is the critical nucleus radius determined by the term $J\tau=n_0\exp(\gamma/R_c)$ and the arguments of all the Macdonald functions are equal to $R_0/\sqrt{D\tau}$. Equation (7) is actually equivalent to the corresponding result in Ref. [3], which is derived assuming constant island shape constancy. A change in the shape is described by Eq. (8). The case where the expression in brackets with certain ν in Eq. (8) is higher than zero is important. The corresponding modes will increase while altering the cluster shape. All other modes decay. The reason for initiation of morphological instability lies in the fact that sections of the island surface, which are the most distant from the center, appear in the region of the highest concentration gradients. These high gradients exist in spite of the increase in the adatom equilibrium concentration. As a result, the island acquires a dendritic shape.

Now, we examine Eq. (8) in more detail. For the sake of simplicity we assume that $R_c\gg\gamma$ and $R_0\gg\gamma$ and make use of an asymptotic expression for $K_\nu(x)$ that is valid for rather high values of the argument x:

$$K_\nu(x)=\sqrt{\frac{\pi}{2x}}e^{-x}\left[1+\frac{\nu^2-\frac{1}{4}}{2x}+\frac{(\nu^2-\frac{1}{4})(\nu^2-\frac{9}{4})}{8x^2}+\cdots\right]\ , \tag{9}$$

Then, after transformations Eq. (8) is reduced to

$$\frac{d\varepsilon_\nu}{dt}=\frac{w\sqrt{D\tau}}{hR_0}\left[J-\frac{n_0}{\tau}\right]\left[1-\frac{R_1}{R_0}\right]\left[\frac{R_2}{R_0}-1\right]\varepsilon_\nu\ , \tag{10}$$

where $R_1(\nu)$ and $R_2(\nu)$ are equal to

$$R_1=(\nu^2-1)\sqrt{D\tau}/4-(\nu^2/2-1)R_c-\sqrt{[(\nu^2-1)\sqrt{D\tau}/4-(\nu^2/2-1)R_c]^2-(\nu^2-1)R_c\sqrt{D\tau}/2}\ , \tag{11}$$

$$R_1=(\nu^2-1)\sqrt{D\tau}/4-(\nu^2/2-1)R_c+\sqrt{[(\nu^2-1)\sqrt{D\tau}/4-(\nu^2/2-1)R_c]^2-(\nu^2-1)R_c\sqrt{D\tau}/2}\ . \tag{12}$$

In the case where $R_c/\sqrt{D\tau} \ll 1$, $R_1=R_c$, $R_2=[(\nu^2-1)/2]\sqrt{D\tau}$, it is shown in Eq. (10) that for every $\nu \geq 2$ the modes with corresponding numbers grow only within the range of $R_1(\nu) < R_0 < R_2(\nu)$ reaching their maximum value of $\varepsilon_\nu^{\max}$ for $R_0=R_2$. In order to find the time interval when the increase occurs we solve Eq. (10) assuming $R_0 \gg R_1$, $R_0=v_0 t$ $[v_0 \equiv (J-n_0/\tau)W\sqrt{D\tau}/h]$:

$$\varepsilon_\nu(t)=\varepsilon_\nu^{\max}\frac{t_\nu}{t}\exp\left[1-\frac{t_\nu}{t}\right] , \tag{13}$$

$$t_\nu=\frac{(\nu^2-1)h}{2w(J-n_0/\tau)} . \tag{14}$$

The island shape dependence in time follows from Eqs. (1) and (13), namely,

$$\begin{aligned} R(\varphi,t)&=v_0 t\left[1+\sum_{\nu=2}^{\infty}\varepsilon_\nu^{\max}\frac{t_\nu}{t}\exp\left[1-\frac{t_\nu}{t}\right]\cos\nu\varphi\right] \\ &=v_0 t+\frac{\sqrt{D\tau}}{2}\sum_{\nu=2}^{\infty}(\nu^2-1)\varepsilon_\nu^{\max}\exp\left[1-\frac{t_\nu}{t}\right]\cos\nu\varphi , \end{aligned} \tag{15}$$

We note that in this case account has been already taken of the fact that modes with numbers $\nu=0$ and $\nu=1$ are rapidly decaying and they do not contribute to Eq. (15). Obviously, at $t<t_\nu$ the mode with number ν rapidly grows, as a rule, from zero to $\varepsilon_\nu^{\max}$, and it further decays like $1/t$ ($\varepsilon_\nu=\varepsilon_\nu^{\max}/3$, when $t=0.304t_\nu$ and $t=7.08t_\nu$). According to Eq. (14), at first the mode with number $\nu=2$ decays, then the mode with number $\nu=3$, and so on. Finally, the result is the occurrence of dendrites at a certain stage of growth. The evolution of cluster shape for $h=3\times10^{-10}$ m, $w=2.7\times10^{-29}$ m^3, $n_0=10^{16}$ m^{-2}, $J=10^{19}$ m^{-2}s^{-1}, $\tau=10^{-2}$ s, $\varepsilon_3^{\max}=\varepsilon_4^{\max}=\varepsilon_6^{\max}=\varepsilon_8^{\max}=0.1$ (the remaining $\varepsilon_\nu^{\max}$ are assumed to be equal to zero) is schematically shown in Fig. 1.

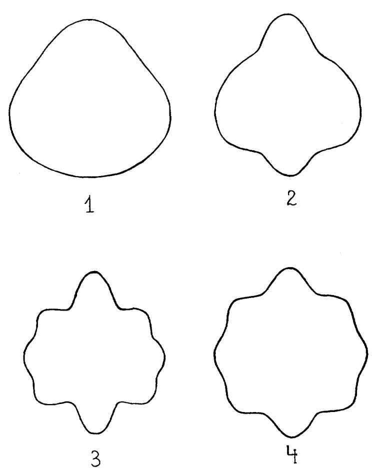

FIG. 1. Schematic representation of the evolution of the cluster shape. This case is in the diffusion regime of growth: 1, $t=3$ s; 2, $t=10$ s; 3, $t=30$ s; 4, $t=100$ s (parameter values are given in the text).

III. ADDITIONAL EFFECTS

A series of assumptions simplifying the physical pattern of the growth phenomenon was made in the above investigation. First, the interface energy σ was considered to be isotropic. This resulted in the disklike shape of a cluster as the equilibrium shape. In fact, crystalline clusters are faceted in equilibrium. Then the expansion into a series of the equilibrium shape deflection from a disk in terms of cosines gives

$$R^0(\varphi)=R_0\left[1+\sum_{\nu=2}^{\infty}\varepsilon_\nu^0\cos\nu\varphi\right] , \tag{16}$$

where ε_ν^2 are equilibrium values of ε_ν; therefore,

$$n_e=n_0 e^{\gamma/R_0}\left[1+\frac{\gamma}{R_0}\sum_{\nu=2}^{\infty}(\nu^2-1)(\varepsilon_\nu-\varepsilon_\nu^0)\cos\nu\varphi\right] . \tag{17}$$

Second, the diffusion process along the island boundary is not taken into account in the cluster growth law (6) [12]; however, the process may be of great importance for rather small R_0. Its introduction into the growth law leads to the appearance of an additional term $-(D_s\gamma w/h^2R_0^4)\nu^2(\nu^2-1)(\varepsilon_\nu-\varepsilon_\nu^0)$, where D_s is the surface diffusion coefficient.

Third, when incorporation of atoms into an island proceeds in a rather slow way, the concentration of adatoms near the cluster boundary n_b is higher than the equilibrium concentration (17) and it is determined by the following equation:

$$v=\frac{Dw}{h}\left[\frac{\partial n}{\partial r}\right]_{r=R}=\alpha D\frac{w}{h\sqrt{D\tau}}(n_b-n_0) , \tag{18}$$

where α is the dimensionless coefficient characterizing the incorporation kinetics (it should be mentioned that incorporation, similar to diffusion, is assumed to be isotropic). When $\alpha\to\infty$, $n_b\to n_e$ and when $\alpha\to0$, $n_b\to J\tau$. All three above effects result in the following change of equations for R_0 and ε_ν:

$$\frac{dR_0}{dt}=\frac{Dw}{h\sqrt{D\tau}}\frac{e^{\gamma/R_c}-e^{\gamma/R_0}}{1/\alpha+K_0/K_1} \tag{19}$$

$$\frac{d\varepsilon_\nu}{dt}=\frac{wn_0}{h\tau}(e^{\gamma/R_c}-e^{\gamma/R_0})\left\{\frac{(K_1/K_0)[(\nu-1)\sqrt{D\tau}/R_0+K_{\nu-1}/K_\nu]-1}{(K_1/K_0\alpha+1)(1+K_{\nu-1}/K_\nu\alpha+\nu\sqrt{D\tau}/R_0\alpha)}\right.$$
$$-\frac{\sqrt{D\tau}}{R_0}\left[(\nu^2-1)\frac{\gamma/R_0}{e^{\gamma/R_c}-e^{\gamma/R_0}}\frac{K_{\nu-1}/K_\nu+\nu\sqrt{D\tau}/R_0}{1+K_{\nu-1}/K_\nu\alpha+\nu\sqrt{D\tau}/R_0\alpha}+\frac{K_1/K_0}{1+K_1/K_0\alpha}\right]$$
$$\left.-\nu^2(\nu^2-1)\frac{D_s\tau}{hn_0R_0^3}\frac{\gamma/R_0}{e^{\gamma/R_c}-e^{\gamma/R_0}}\right\}\left[\varepsilon_\nu-\frac{\varepsilon_\nu^0}{1+\mathcal{H}_\nu}\right], \tag{20}$$

$$\mathcal{H}_\nu=\frac{\nu-2+(R_0/\sqrt{D\tau})(K_{\nu-1}/K_\nu-K_0/K_1)-K_{\nu-1}/K_\nu\alpha-\nu\sqrt{D\tau}/R_0\alpha}{[\nu^2(D_s/D)\sqrt{D\tau}/hn_0R_0^2](1+K_{\nu-1}/K_\nu\alpha+\nu\sqrt{D\tau}/R_0\alpha)+K_{\nu-1}/K_\nu+\nu\sqrt{D\tau}/R_0}$$
$$\times\frac{1}{\nu^2-1}\frac{R_0}{\gamma}\frac{e^{\gamma/R_c}-e^{\gamma/R_0}}{1/\alpha+K_0/K_1}. \tag{21}$$

Here, $\mathcal{H}_\nu$ are the dimensionless constants characterizing the degree of faceting of islands. Obviously, in the case when the term in brackets in Eq. (20) is less than 0, then $\varepsilon_\nu \to \varepsilon_\nu^0/(1+\mathcal{H}_\nu)$ at large time intervals. If, in addition, $|\mathcal{H}_\nu| \ll 1$, an island during the course of growth becomes faceted and approaches its equilibrium shape (16). On the contrary, in the case with a positive expression in the brackets, dendritic forms generate. The analysis shows that a mode with number ν grows only in one case, when the radius of a cluster lies within certain limits of $R_1(\nu) < R_0 < R_2(\nu)$. It is to be noted that $d\varepsilon_\nu/dt < 0$ for some initial values of ν and for all R_0. Thus, all the above three effects do not qualitatively change the morphological stability pattern; however, they greatly change it quantitatively. Equations (20) and (21) can be simplified by means of asymptotes to the Macdonald function (9) when assuming that $R_0\sqrt{D\tau}$ is rather large and γ/R_c and γ/R_0 are rather small:

$$\frac{d\varepsilon_\nu}{dt}=\frac{(\nu^2-1)\gamma wDn_0}{(1+1/\alpha)R_0^2h}\left[\frac{1}{R_c}-\frac{1}{R_0}\right]\left[\frac{1}{2(1+1/\alpha)}-\frac{R_0/\sqrt{D\tau}}{\nu^2-1}-\frac{R_c/\sqrt{D\tau}}{1-R_c/R_0}-\frac{\nu^2R_c(1+1/\alpha)D_s}{hn_0R_0^2(1-R_c/R_0)D}\right]$$
$$\times\left[\varepsilon_\nu-\frac{\varepsilon_\nu^0}{1+\mathcal{H}_\nu}\right], \tag{22}$$

$$\mathcal{H}_\nu=\left[\frac{R_0}{R_c}-1\right]\left[\frac{\sqrt{D\tau}/R_0}{2(1+1/\alpha)}-\frac{1}{\nu^2-1}\right]\left[1+\frac{\nu^2\sqrt{D\tau}D_s}{hn_0R_0^2D}\left[1+\frac{1}{\alpha}\right]\right]^{-1}. \tag{23}$$

Approximate expressions for R_1 and R_2 follow from Eq. (22) provided that $R/\sqrt{D\tau} \ll 1$,

$$R_1(\nu)\sim\nu\sqrt{2(1+1/\alpha)R_cD_s/hn_0D}, \tag{24}$$

$$R_2(\nu)\sim\frac{\nu^2-1}{2}\frac{\sqrt{D\tau}}{1+1/\alpha}. \tag{25}$$

In the case that $R_1 > R_2$ for some ν (possible only with insufficiently small $R_c/\sqrt{D\tau}$), this particular mode corresponding to ν is not excited at all. Obviously, the surface diffusion increases R_1 leaving R_2 nearly unchanged, whereas kinetic phenomena increase R_1 and decrease R_2. Faceting of clusters, as follows from Eq. (23), takes place in a rather narrow region near the critical size. Since condition $|\mathcal{H}_\nu| \ll 1$ is fulfilled at $\alpha \to \infty$ roughly for $3R_c/4 < R_0 < 3R_c/2$, in practice the faceting of clusters proceeds only at the Ostwald ripening stage, when large clusters grow due to the decay of smaller ones [9]. Decreasing α leads to expansion of the range of R_0 values, where $|\mathcal{H}_\nu| \ll 1$. Therefore, the slow incorporation of atoms into an island improves its faceting.

IV. DISCUSSION

The shape stability of a disklike cluster growing from vapor of adsorbed particles on a solid substrate was investigated. It was shown that the mode with number ν is excited only in the case where the cluster radius lies within a certain range of $R_1(\nu) < R_0 < R_2(\nu)$. This is the main difference of the shape instability of a surface cluster from an appropriate three-dimensional case of cylinder growth from solution, where the instability grows for all R_0 which are greater than a certain $R_*(\nu)$ [10,12,13]. Arrival of particles on a substrate and the reevaporation of them give rise to this difference. Now we point out one more feature that is characteristic of the instability of the growth of thin films. The mode with number $\nu=2$ may result in the distortion of a cluster shape (mode $\nu=2$ is excited only for fairly low $R_c/\sqrt{D\tau}$), whereas during the growth of a cylinder from solution this mode always decays [13]. Essential allowances are to be made for additional effects, such as surface diffusion and kinetic phenomena at the phase boundary. In particular, surface diffusion increases R_1 appreciably, whereas a delay of incorporation of atoms into an island leads to an increase in R_1 and a decrease in R_2, and on this account the first modes become, sometimes, deexcited. For instance, when $\alpha=1$, $R_c/\sqrt{D\tau}=3\times10^{-2}$, $n_0 h\sqrt{D\tau}=3\times10^{-2}$, modes $\nu=2$ and $\nu=3$ are not excited, and the mode with number $\nu=4$ appears only within the range of $0.8<R_0/\sqrt{D\tau}<1.6$. The mode growth kinetics and, hence the kinetics of distorted island shapes are described by Eqs. (19) and (20). In the simplest case of diffusion growth without allowing for diffusion along a phase boundary, the cluster shape and the characteristic time of excitation of various modes are estimated by Eqs. (14) and (15). It is shown that all modes are excited in turn and the growth of amplitudes ε_ν from 0 to $\varepsilon_\nu^{\max}$ proceeds very rapidly while decay occurs rather slowly (as $1/t$). In the case of supersaturation $J\tau/n_0-1=4$ and $h/wn_0=10^3$, the amplitude of the mode with number $\nu=3$ attains its maximum value at time interval $t_3=10^3\tau$. When an island radius is not only less than $R_1(\nu)$ for the least excited ν but is so close to R_c that $|\mathcal{H}_\nu|\ll1$, faceting of a cluster takes place during its growth. Faceting criteria $|\mathcal{H}_\nu|\ll1$ can be rather different when diffusion and incorporation anisotropies are taken into account. When controlling the morphology of clusters by means of R_c and $\sqrt{D\tau}$, depending on the arrival rate and the temperature of a substrate, we can control the film structure because it is primarily dependent on the shape of clusters at the moment of their coalescence into a solid layer. When, for instance, the preparation of a film consisting of well-faceted grains is required, it is necessary to come to the Ostwald ripening mode by decreasing J in accordance with the power-type law [9]. On the other hand, when a film of dendrite structure is to be deposited, the arrival rate and the temperature are selected in such a way that, first, the diffusion mode of growth with fairly low $R_c/\sqrt{D\tau}$ exists and, second, the island size is roughly one order of magnitude higher than the diffusion length.

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