Instability of the axial form of two-dimensional islands of a new phase in the Ginzburg–Landau theory

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We use a Ginzburg–Landau type of equation for the order parameter field to study the instability of the azimuthal form of two-dimensional islands of a new phase. We obtain instability diagrams of the angular perturbations and show that the instability occurs when the main growth mechanism is diffusional. We determine the parameter region in which the criteria obtained here are the same as the results of the phenomenological Zel'dovich–Folmer theory and of the DLA (diffusion limited aggregation) continuum model. We study the criteria for the growth of the angular perturbations in the case of a nonquasistationary growth regime. We consider the effect of the angular perturbations of the form on the value of the fractal cluster dimensionality.

1. INTRODUCTION

In experimental studies of nucleus formation processes and the further growth of a new phase on surfaces one observes azimuthally symmetric islands[1–3] as well as formations with a more complex symmetry[3,4] and also fractal clusters.[5–11] As a simple quantitative criterion for the broken axial symmetry of a cluster one normally uses the value of the exponent $\mathscr{D}$ in the relation between the number of particles in the cluster and its size R: $N \propto R^{\mathscr{D}}$. The quantity $\mathscr{D}$ is called the fractal dimension of the cluster.[5–11] The appearance of asymmetry in an island leads to the value of the index $\mathscr{D}$ deviating from the Euclidean dimensionality. In the case of the growth of fractal clusters one observes fractional and to a large degree universal values of $\mathscr{D}$. For instance, $\mathscr{D}=1.5$ to 1.9 in the case of the growth of fractal clusters in a plane.[5]

In an appreciable fraction of the experiments the growth of the clusters takes place predominantly in the plane of a surface. Such clusters are, for instance, formed during the precipitation of particle vapors from a gas phase onto a surface,[12] during the electrodeposition of metals from a mixture or a melt onto a surface,[13,14] or during the crystallization of amorphous films which are deposited on a substrate.[15,16] In what follows we shall consider just such clusters.

For the theoretical description of the phenomena of the growth of a new phase the problem arises of how to describe the growth of islands with a form which is not circular (axially symmetric), including fractal growth. As a first step it is important to determine the criteria for the form instability of circularly shaped nuclei. These problems have been studied in the framework of the Zel'dovich–Folmer theory[17–19] of first-order phase transitions and in various approaches to the dynamics of clusters using computer simulations.[6–11] In the theory of phase transitions[1–4] attempts to describe azimuthally asymmetric structures were connected with the explicit introduction of anisotropic conditions for the growth of the nuclei connected with surface tension,[3,4] anisotropies of external fields,[20,21] symmetries of the substrate,[22–25] and so on. However, it was found that the effect of symmetry breaking for the shape of the nuclei occurred also in a symmetric (isotropic) system.[26–28] It has been shown that the axial form of islands growing due to the diffusion of the adsorbed atoms is unstable. The equations obtained for the growth of the angular perturbations of the boundary of an island,

$$R(\varphi,t)=R_0(t)+\sum_{m\geqslant 2} R_m(t)\cos(m\varphi)$$

have for $R_m/R_0 \ll 1$ the form[26]

$$\frac{d}{dt}\ln R_m(t)=\rho_0 s C_0 D\frac{m-1}{R_0^2}\left\{\left(\frac{1}{R_{\rm cr}}-\frac{1}{R}\right)K_0^{-1}\left(\frac{R_0}{L}\right)^{-1}-\frac{m(m+1)}{R_0}\right\},\qquad (1)$$

where D is the diffusion coefficient, s is the area per particle in the new phase, $R_{\rm cr}=\rho_0 C_0/(\bar{C}-C_0)$ is the critical size of the new phase, $\bar{C}$ is the particle density far from the nucleus, C_0 is the equilibrium density near the linear boundary of the island, $\rho_0=\gamma s/T$, γ is the linear tension coefficient, T is the temperature, $L^2=D/\alpha$, α^{-1} is the particle desorption time, and $K_0(x)$ is a Macdonald function. The criterion for the instability of the growth, $\lambda_m>0$, with

$$\lambda_m=\frac{d}{dt}\ln\left(\frac{R_m}{R_0}\right)$$

the instability growth rate, leads to the condition

$$\frac{R_0}{R_{\rm cr}}\geqslant\frac{m(m^2-1)}{m-2}K_0\left(\frac{R_0}{L}\right),\qquad (2)$$

and when this is satisfied the harmonics with the given m are not damped. It turns out that the instability is possible only for harmonics with $m\geqslant 3$ for sufficiently large sizes of the growing islands. We note that (1) and (2) are obtained in the limit of large diffusion lengths for the particles on the surface.

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In papers on the dynamics of the growth of clusters a collection of different models is used when applied to actual physical systems. Historically the first and the most often used one is the DLA (Diffusion Limited Aggregation) model.[5–11,27–31] In it the particles wandering randomly over the lattice (lattice DLA model) or in arbitrary directions (off-lattice DLA model[29]) come in contact with a cluster and with a certain probability stick to it. The off-lattice model makes it possible to remove the possible effect of the lattice anisotropy on the properties of the cluster. A model has been developed in which not only particles, but also simple clusters can move and in which the latter subsequently stick together (Cluster–Cluster Aggregation[5–8]). Apart from models with diffusive trajectories of the particles moving to the cluster, models like the Eden ballistic model[32] have been formulated in which the particle trajectories are along straight lines which also describe fractal growth. Computer experiments indicate that the phenomenon of the growth of fractal clusters is universal and is determined by the general properties of the growth process and appears in initially isotropic symmetric models such as the off-lattice DLA model. Nonetheless in some stages of the growth the anisotropic properties of the system (lattice symmetry, anisotropy of the particles themselves, conditions at the cluster boundary simulating elastic tensions, and so on[3,4,20–25]) may show an appreciable effect leading, for instance, to a change in the fractal dimensionality.

In Refs. 27 and 28 an analytical study was carried out of the conditions for the appearance of an instability of the axial form in the framework of the DLA model. The criterion for the growth of angular perturbations of the shape of the cluster boundary obtained using the analytical DLA model[27] is the same, apart from a different notation, as the criterion (2) in the diffusive limit $R_0 \gg L$.

The studies have shown thus that for the appearance of an instability of the axially symmetric form of islands it is not necessary that the system has anisotropic properties. However, the methods[1–4,33] used to study the conditions for the shape instability of islands use, notwithstanding their physical simplicity, a number of phenomenological parameters which cannot be evaluated in the Zel'dovich–Folmer theory as well as unjustified assumptions. For instance, in the Zel'dovich–Folmer theory are introduced a surface tension coefficient, the molecular volume of the new phase, the equilibrium density of a saturated solution, and also conditions on the boundary of the island. Similar conditions and assumptions occur also in the DLA model. In essence this approach is based upon a stationary diffusion model with given boundary conditions that determine the rate of growth of the island boundary. As a result it remains unclear in how far the description of the appearance of structures with a complicated shape is determined by any particular phenomenological condition or parameter. Moreover, there are other mechanisms for the growth of clusters on a surface: diffusion of particles along the surface of the island itself, and also direct incidence of atoms from the phase above the surface onto the cluster perimeter (nondiffusive mechanism[34]). In this connection it remains unclear in how far the mechanism of the cluster growth affects their shape instability. Treatment of the shape instability effect in the framework of a more general approach would make it possible to answer those questions.

Apart from the above-mentioned Zel'dovich–Folmer theory and numerical simulation methods one also uses for the description of phase transitions an approach based upon an expansion of the free energy of the system in a series in the order parameter,[35] in which the relaxation is studied using a Ginzburg–Landau type equation for the order parameter.[35–37] Such an approach to the kinetics of a phase transition does not require additional parameters or assumptions. For second-order phase transitions in a volume or on a surface this approach made it possible to describe the growth of domains and a number of nontrivial features of the kinetics of structural transitions (see, e.g., Refs. 38 to 43).

A study of the nucleus forming process in two- and three-dimensional systems undertaken in Refs. 35 to 37 and 44 has shown that all phenomenological parameters of the Zel'dovich–Folmer theory can be expressed in terms of the expansion coefficients of the free energy and the kinetic coefficients of the order parameter equation and that all results of the Zel'dovich–Folmer theory are obtained as special cases of solutions of that equation. The order-parameter equation can be reduced to a dimensionless form. As a result the kinetics of the growth of the new phase is determined by solely two parameters: the degree of metastability h of the system and the magnitude l of the ratio of the contributions from the processes which do not and which do conserve the order parameter (when the clusters grow as the result of the adsorption of atoms by the surface these are the processes of the desorption of adatoms and their diffusive motion along the surface). All this makes it possible to assume that such an approach enables us in a natural manner (without introducing additional assumptions or parameters) to study the occurrence of a shape instability of the islands which is caused by the universal properties of the growth process.

2. ORDER-PARAMETER EQUATION

The equation describing the relaxation of the order parameter field $\xi(\mathbf{r},t)$ in the new, energetically more favorable state has the form[35–37]

$$\dot{\xi}(\mathbf{r},t) = -\hat{\mu}\delta F[\xi(\mathbf{r},t)]/\delta\xi. \tag{3}$$

Here $F[\xi(\mathbf{r},t)]$ is the free energy functional corresponding to the field $\xi(\mathbf{r},t)$, $\mathbf{r}$ is the coordinate, t is the time, and $\hat{\mu}$ is the kinetic operator which in the long-wavelength approximation has the form $\hat{\mu} = -\mu_c\nabla^2 + \mu_n$ where μ_c and μ_n are the kinetic coefficients in the case of a conserving and a nonconserving order parameter field, respectively. In the phase transitions on a surface considered here the field ξ is the deviation of the density $C(\mathbf{r},t)$ of adsorbed atoms in the new phase state from the average (quasistationary) density $\bar{C}$ of adatoms on the surface: $\xi(\mathbf{r},t) = C(\mathbf{r},t) - \bar{C}$.

The values of the kinetic coefficients μ_c and μ_n and of the free energy F in the vicinity of a phase transition point determine completely the parameters of Eq. (3), and for a description of the phase transition kinetics one does not need any free parameters or additional assumptions.[35–37] We write down the expression for the free energy in the vicinity of the phase transition point which follows from the Landau theory:

$$F[\xi]=\int d\mathbf{r}\left(\lambda\frac{\xi^2}{2}+\Omega\frac{(\nabla\xi)^2}{2}-\mathrm{B}\frac{\xi^3}{3}+\Gamma\frac{\xi^4}{4}\right). \quad (4)$$

Here λ, Ω, B, and Γ are coefficients which, in general, depend on external parameters such as the temperature and the adatom density.

When the coefficient B is different from zero the free energy (4) describes a first-order phase transition. One checks easily that the phase transition occurs for $\lambda\leqslant 2\mathrm{B}^2/9\Gamma$ and is a transition from a state with zero order parameter, $\xi=0$, to a state with $\xi=\xi_0=2\mathrm{B}/3\Gamma$.

The order-parameter equation (3) for the free energy (4) takes the form

$$\dot{\xi}(\mathbf{r},t)=(\mu_c\nabla^2-\mu_n)(\lambda\xi-\Omega\nabla^2\xi-\mathrm{B}\xi^2+\Gamma\xi^3). \quad (5)$$

This is a Ginzburg–Landau type equation which can be used to describe various phase transitions.[35–37]

The kinetic coefficients μ_c and μ_n in Eq. (3) are connected with observable quantities. Indeed, we can write Eq. (5) in the linear approximation in $\xi(\mathbf{r},t)$ describing the relaxation of small perturbations on the surface:

$$\dot{\xi}(\mathbf{r},t)=-\alpha\xi+D\nabla^2\xi,\quad \alpha\equiv\mu_n\lambda,\quad D\equiv\mu_c\lambda. \quad (6)$$

In the theory of the adsorption of atoms one uses an equation which is the same as Eq. (6),[26,34,45,46] and the quantities D and α^{-1} are the adatom diffusion coefficient (D) and the characteristic adatom lifetime due to their desorption from the surface or to their leaving the volume of the material (α^{-1}). Hence it follows that the coefficient μ_n is connected with atomic adsorption and desorption processes ($\mu_n=\alpha/\lambda$) while μ_c is connected with adatom diffusion on the surface ($\mu_c=D/\lambda$).

We rewrite Eq. (5) using dimensionless variables $\phi=2\xi/\xi_0-1$, $\boldsymbol{\rho}=\mathbf{r}/\chi$, and $\tau=t/\tau_0$:

$$\dot{\phi}(\boldsymbol{\rho},\tau)=(l^2-\nabla^2)[\nabla^2\phi+2\phi(1-\phi^2)+h(\phi+1)]. \quad (7)$$

Here $\xi_0=2\mathrm{B}/3\Gamma$ is the order parameter of the new phase at the phase transition point,

$$\chi=\xi_0(8\Omega/\Gamma)^{1/2},\quad \tau_0=\chi^4/\mu_c\Omega,\quad l^2=\chi^2\mu_n/\mu_c,$$

and the quantity $h=4(1-\lambda/\lambda_c)$ has the meaning of the degree of metastability. The dimensionless order parameter ϕ is equal in the old phase to -1 and in the new phase to $\phi=\phi_n\equiv(1+\sqrt{1+2h})/2$ ($\phi_n\approx 1+h/2$ for small degrees of metastability, $h\ll 1$).

Equation (7) contains altogether only two dimensionless parameters: h and l. The degree of metastability h determines how close one is to the phase transition point, the difference between the new and the old energy states. Near the phase transition point one may assume that the parameter h is small: $|h|\ll 1$. The parameter l in (7) determines in fact the ratio of the contributions of two different mechanisms for the growth of the islands of the new phase: direct capture and diffusive growth.[44] The parameter l varies over a broad range in different physical systems. It is just this parameter which determines the possible relaxation regimes of a system.

We note that the case of diffusive growth of the new phase is realized most often in experiments. In the Zel'dovich–Folmer theory[26] and in the DLA model[27,28] was discovered an instability of the shape of the islands also in the diffusive limit. This case, corresponding to values $l\ll 1$, is just the one which in what follows will be of the greatest interest.

3. SOLUTIONS OF THE ORDER-PARAMETER EQUATION

The order-parameter field corresponding to a critical nucleus minimizes the free-energy functional and is according to (3) a stationary solution of the order-parameter equation. For small degrees of metastability h this field can with fair accuracy be written in the form[36,44]

$$\phi_{\mathrm{cr}}(\rho)\approx\mathrm{th}(a_{\mathrm{cr}}-\rho),\quad a_{\mathrm{cr}}=2/3h,\quad h\ll 1. \quad (8)$$

The quantity a_{cr} is the critical size of an island and is determined by the degree of metastability h. It follows from the nucleus-like solution (8) that the width of the transition layer between the phases is of the order of unity in dimensionless variables and, hence, of the order of χ in dimensional variables.

We now look for nonstationary solutions of the order parameter equation (7). Using the Green function $G(\boldsymbol{\rho})=K_0(\rho l)/2\pi$ of the equation

$$(l^2-\nabla^2)G(\boldsymbol{\rho})=\delta(\boldsymbol{\rho}) \quad (9)$$

we can rewrite (7):

$$\int G(\boldsymbol{\rho}-\boldsymbol{\rho}')\dot{\phi}(\boldsymbol{\rho}',\tau)d\boldsymbol{\rho}'=\nabla^2\phi(\boldsymbol{\rho},\tau)+2\phi(1-\phi^2)+h(\phi+1). \quad (10)$$

We look for solutions of Eqs. (7) and (10) describing the formation of axially symmetric islands of the new phase, with a size R which is large as compared to the width χ of the transition layer ($a\equiv R/\chi\gg 1$) in the form of a nucleus with some corrections:[36,44]

$$\phi(\boldsymbol{\rho},\tau)=\phi_{\mathrm{cr}}(\rho-a(\tau))+w(\boldsymbol{\rho},\tau). \quad (11)$$

Here $\phi_{\mathrm{cr}}(\rho-a(\tau))$ is the solution of Eq. (7) for the critical nucleus but with a difference through the substitution $a_{\mathrm{cr}}\to a(\tau)$. To study the instability of the axial form of islands we introduce into our discussion the dependence of the size a on the azimuthal angle φ in the form

$$a(\varphi,\tau)=\sum_{m\geqslant 0}a_m(\tau)\cos(m\varphi). \quad (12)$$

The term with $m=1$ describes the shift of the nucleus as a whole and we have thus $a_1\equiv 0$. In looking for the instability conditions we restrict ourselves to small angular perturbations, i.e., $a_m/a_0\ll 1$.

It will become clear in what follows that the correction $w(\rho,\tau)$ determines the diffusion flow to the nucleus and that in the case, considered in what follows, of small l and weak degrees of metastability it is a small quantity and a slowly changing function of the coordinates:

$$|w(\rho,\tau)| \ll 1, \quad |\nabla w| \ll |w|. \tag{13}$$

Near the phase transition point ($|h| \ll 1$) the change in the size of the nucleus is usually a very slow process,[34,36,47] i.e., we may assume that the characteristic time τ_w for the establishment of a distribution of the quantity w is short compared to the characteristic time τ_a for changes in the size of the nucleus:

$$\tau_w \ll \tau_a. \tag{14}$$

We note that in two-dimensional systems the condition for quasistationarity can nevertheless be violated for small l.[44] We shall consider this case further for $l=0$.

At any finite time τ there is at large distances from the nucleus only the old phase ($\phi(\rho\to\infty,\tau)\to-1$). Since $\phi_{\rm cr}(\rho\to\infty,\tau)\to-1$, $w(\rho,\tau)$ must satisfy the condition

$$w(\rho\to\infty,\tau)=0. \tag{15}$$

Substituting the solution (11) into (10) and using the conditions (13) and (14) we get as a result an equation for the two functions $w(\boldsymbol{\rho},\tau)$ and $a(\phi,\tau)$, namely:

$$P(\boldsymbol{\rho}) = \left[\frac{1}{a} - \frac{1}{a_{\rm cr}} - \frac{a''}{a^2} - 6w(\boldsymbol{\rho},\tau)\right]\mathrm{ch}^{-2}(a-\rho) + 4w(\boldsymbol{\rho},\tau), \tag{16}$$

where

$$P(\boldsymbol{\rho},\tau) \equiv \int G(\boldsymbol{\rho}-\boldsymbol{\rho}')\dot a(\varphi',\tau)\mathrm{ch}^{-2}[a(\varphi',\tau)-\rho']d\boldsymbol{\rho}', \quad a\equiv a(\varphi,\tau),$$

$$a'' \equiv \frac{\partial^2 a(\varphi,\tau)}{\partial\varphi^2}.$$

We can show that, indeed, (16) is an equation for the function w since there is a unique relation between w and a which follows from Eq. (16) itself. Indeed, the terms in the square brackets on the right-hand side of (16) are multiplied by a function $\cosh^{-2}(a-\rho)$ which varies rapidly near the boundary of the island so that outside the boundary it drops out of the equation because it is exponentially small. We now show that in the diffusive limit, $l \ll 1$, the quantity $P(\boldsymbol{\rho},\tau)$ is a function which varies smoothly on scales of the order of $1/l$ which are larger than the scale of the transition layer of the nucleus (at least in the initial stages of the growth of the angular harmonics). One can verify this by rewriting the expression for $P(\boldsymbol{\rho},\tau)$ in the form

$$P(\boldsymbol{\rho},\tau) = \sum_{m>0} \dot a_m(\tau)P_m(\boldsymbol{\rho}), \tag{17}$$

where we have for $a_m \ll a_0$

$$P_m(\boldsymbol{\rho}) = \int G(\boldsymbol{\rho}-\boldsymbol{\rho}')\cos(m\varphi')\mathrm{ch}^{-2}(a_0-\rho')d\boldsymbol{\rho}'.$$

We shall prove the smoothness of the function $P_0(\rho)$. The integrand of $P_0(\rho)$ in (17) contains for $l\ll1$ a product of a rapidly changing function $\cosh^{-2}(a_0-\rho')$, which is nonvanishing in the vicinity of the boundary $\rho'=a_0$ of the nucleus, and the function $G(\rho,\rho')\rho'$ which varies smoothly in that neighborhood [here $G(\rho,\rho')$ is the function $G(\boldsymbol{\rho}-\boldsymbol{\rho}')$ of (17) integrated over the angles of the vector $\boldsymbol{\rho}'$]. This circumstance makes it possible to expand the function $G(\rho,\rho')\rho'$ in the variable ρ' near the maximum value $\rho'=\rho_m$ of the function $\cosh^{-2}(a_0-\rho')$ (we have here $\rho_m\approx a_0$). Restricting ourselves in (16) to the zeroth approximation for the function $G(\rho,\rho')\rho' \approx G(\rho,\rho_m)\rho_m$ we get for $P_0(\rho)$ in (17)

$$P_0(\rho) = \begin{cases} 2I_0(\rho l)K_0(a_0 l)a_0, & \rho<a_0 \\ 2K_0(\rho l)I_0(a_0 l)a_0, & \rho>a_0 \end{cases}, \tag{18}$$

where I_0 is a modified Bessel function. Hence it follows that the characteristic scale for changes in the function $P_0(\rho)$ is the quantity $1/l$. The function $P_0(\rho)$ thus changes smoothly over scales of the order of the transition layer of the nucleus in the diffusive limit, when $l\ll1$.

Thus, outside the boundary of the island Eq. (16) connects the two smoothly varying functions:

$$-P(\boldsymbol{\rho},\tau) = 4w(\boldsymbol{\rho},\tau), \tag{19}$$

and therefore this relation must also be valid on the boundary of the nucleus for $\rho\approx a$. The terms in the square brackets on the right-hand side of Eq. (16) then give us the required relation between $a(\varphi,\tau)$ and $w(\rho=a(\varphi,\tau))$:

$$w(\rho=a(\varphi,\tau)) = \frac{1}{6}\left[\frac{1}{a(\varphi,\tau)} - \frac{1}{a_{\rm cr}}\right] - \frac{1}{6}\frac{a''(\varphi,\tau)}{a^2(\varphi,\tau)}. \tag{20}$$

To find the equations for the growth of the angular perturbations we apply the operator $(l^2-\nabla^2)$ to Eq. (19). As a result we get

$$\dot a(\varphi,\tau)\mathrm{ch}^{-2}[\rho-a(\varphi,\tau)] = 4(\nabla^2-l^2)w(\boldsymbol{\rho},\tau). \tag{21}$$

Outside the boundary of the nucleus this equation is the ordinary diffusion equation for the fluxes of atoms to the growing nucleus. We integrate (21) over the transition layer where the function $\cosh^{-2}[a(\varphi,\tau)-\rho]$ is nonvanishing. We use the fact that in the initial stages of the growth of the angular perturbations we have $a_m/a_0\ll1$ and that the angles between the normal to the perturbed surface and the normal to the cylindrical surface at the same point are small: $\gamma\ll1$. This enables us to write

$$\dot a(\tau) = 2(\nabla w|_{\rho=a+0} - \nabla w|_{\rho=a-0} - 2l^2 w|_{\rho=a}). \tag{22}$$

As a result the growth rate $\dot a(\varphi,\tau)$ of the size of the nucleus is basically given by the fluxes $\partial w/\partial\rho$ of adatoms through the boundary of the island ($l\ll1$). We note that, for nonvanishing values of l, desorption and adsorption processes occur not only outside the island, but also on its surface. This leads to the appearance of adatom fluxes not only along the unshaded surface of the material, but also over the surface of the island itself.

To find explicit expressions for the fluxes we shall look for a solution of Eq. (21) outside the boundary

$$(\nabla^2-l^2)w(\rho,\tau)=0, \quad |\rho-a(\varphi,\tau)|>1 \tag{23}$$

in the form of an expansion in harmonics:

$$w(\rho,\tau)=\begin{cases} w_0\dfrac{K_0(\rho l)}{K_0(a_0 l)}+\sum\limits_{m\geq 2} w_m^{(1)}\dfrac{K_m(\rho l)}{K_m(a_0 l)}\cos(m\varphi), \\ \qquad \rho>a(\varphi,\tau) \\ w_0\dfrac{I_0(\rho l)}{I_0(a_0 l)}+\sum\limits_{m\geq 2} w_m^{(2)}\dfrac{I_m(\rho l)}{I_m(a_0 l)}\cos(m\varphi), \\ \qquad \rho<a(\varphi,\tau) \end{cases}, \tag{24}$$

where K_i and I_0 are ith order Macdonald and modified Bessel functions. The asymptotic condition (15) has been taken into account in the expansion (24). To study the stability of small perturbations of the shape of the island $a_m(\tau)$ we write down the boundary condition (20) for the function $w(\rho,\tau)$ in the approximation which is linear in a_m/a_0:

$$w(\rho=a(\varphi,\tau)) =\frac{1}{6}\left(\frac{1}{a_0}-\frac{1}{a_{\rm cr}}\right)+\sum_{m\geq 2}\frac{m^2-1}{6a_0^2}a_m\cos m\varphi. \tag{25}$$

Matching the coefficients w_0 and $w_m^{(1,2)}$ in (24) with the coefficients of the corresponding harmonics in the boundary condition (25) we get the explicit form of the fluxes: $\partial w/\partial\rho$ for $\rho=a(\varphi,\tau)\pm 0$. As a result the equations for the growth of azimuthal perturbations (and of the mean radius of the island) have in the diffusive limit $l\ll 1$ which we are studying the form

$$\dot a_0=-4w_0\left\{\frac{l}{2}\left[\frac{K_1(a_0l)}{K_0(a_0l)}+\frac{I_1(a_0l)}{I_0(a_0l)}\right]+l^2\right\}, \tag{26}$$

$$\begin{aligned}\frac{d\ln a_m}{d\tau}=&-\frac{2}{3}\frac{m^2-1}{a_0^2}\left\{\frac{l}{2}\left[\frac{K_{m+1}(a_0l)}{K_m(a_0l)}+\frac{I_{m+1}(a_0l)}{I_m(a_0l)}\right]+l^2\right\}\\ &-2l^2w_0\left\{\left[\frac{K_1(a_0l)}{K_0(a_0l)}\frac{K_{m+1}(a_0l)}{K_m(a_0l)}\right.\right.\\ &\left.-\frac{I_1(a_0l)}{I_0(a_0l)}\frac{I_{m+1}(a_0l)}{I_m(a_0l)}\right]-\frac{m+1}{a_0l}\left[\frac{K_1(a_0l)}{K_0(a_0l)}\right.\\ &\left.\left.+\frac{I_1(a_0l)}{I_0(a_0l)}\right]\right\}, \quad w_0=\frac{1}{6}\left(\frac{1}{a_0}-\frac{1}{a_c}\right).\end{aligned} \tag{27}$$

Equation (26) has been studied before, using the order parameter equation,[44] in an analysis of the growth of azimuthally symmetric islands. It is also well known in the phenomenological theory[34,45–47] and describes circular islands which are growing (for $a_0>a_{\rm cr}=2/3h$) due to fluxes of particles onto their boundary from the outside and from the inside (the first two terms, respectively) and also due to the adsorption of particles directly on the boundary of the island (last term):

$$\dot R=s_0\rho_0C_0\frac{D}{L}\left(\frac{1}{R_{\rm cr}}-\frac{1}{R}\right)\left\{\left[\frac{K_1(R/L)}{K_0(R/L)}+\frac{I_1(R/L)}{I_0(R/L)}\right]+\frac{\alpha l_0L}{D}\right\}, \tag{28}$$

where l_0 is the width of the layer in which direct capture by the perimeter of the nucleus takes place ($l_0=2\chi$ where χ is the width of the transition layer of the island).

Equation (27) describes the relaxation of the angular harmonics. For an analysis of the conditions for the appearance of a growth instability of the angular perturbations we find the instability growth rates:

$$\begin{aligned}\lambda_m=&\frac{d}{d\tau}\ln\frac{a_m}{a_0}\\ =&-\frac{2}{3}\frac{m^2-1}{a_0^2}\left\{\frac{l}{2}\left[\frac{K_{m+1}(a_0l)}{K_m(a_0l)}+\frac{I_{m+1}(a_0l)}{I_m(a_0l)}\right]+l^2\right\}\\ &-2l^2w_0\left\{\left[\frac{K_1(a_0l)}{K_0(a_0l)}\frac{K_{m+1}(a_0l)}{K_m(a_0l)}\right.\right.\\ &\left.-\frac{I_1(a_0l)}{I_0(a_0l)}\frac{I_{m+1}(a_0l)}{I_m(a_0l)}\right]-\frac{m+1}{a_0l}\left[\frac{K_1(a_0l)}{K_0(a_0l)}\right.\\ &\left.\left.+\frac{I_1(a_0l)}{I_0(a_0l)}\right]-\frac{1}{a_0}\right\}.\end{aligned} \tag{29}$$

We have obtained Eqs. (27) and (29) for arbitrary ratios of the parameters a_0 and l^{-1} in the case of large islands ($a_0\gg 1$) which are growing basically via adatom diffusion ($l\ll 1$). Let us consider the case when the diffusion length L is large compared with the radius R of the nucleus: $a_0l\ll 1$. We use the asymptotic relations for the functions $K_m(x)$ and $I_m(x)$ ($x=a_0l$).[48] In that case (27) and (29) take the form

$$\begin{aligned}\frac{d\ln a_m}{d\tau}&\approx\frac{2(m-1)}{a_0^2}\left[-\frac{w_0}{K_0(a_0l)}-\frac{m(m+1)}{3a_0}\right],\\ \frac{da_0}{d\tau}&=-\frac{2w_0}{a_0K_0(a_0l)},\\ \lambda_m&\approx\frac{2}{a_0^2}\left[-\frac{w_0(m-2)}{K_0(a_0l)}-\frac{m(m^2-1)}{3a_0}\right].\end{aligned} \tag{30}$$

In dimensional variables these equations correspond to Eqs. (1) and (2) obtained in the phenomenological theory.[26] We note that in this limit ($a_0l\ll 1$) the growth of the shape perturbations (the condition $\lambda_m=0$) can be observed only for harmonics with numbers $m\geq 3$, whereas this is not possible for $m=2$ whatever the values of the parameters.

We show in Fig. 1 the conditions for the occurrence of an instability for an arbitrary ratio of the parameters a_0 and l at $a_{\rm cr}=10^2$. The solid lines show here the relations $\lambda_m=0$ of (29). The instability regions ($\lambda_m>0$) for harmonics with numbers $m=3,...,6$ lie to the left of the $\lambda_m=0$ curves enclosing them for each corresponding harmonic. It is clear from the figure that instability is possible only for

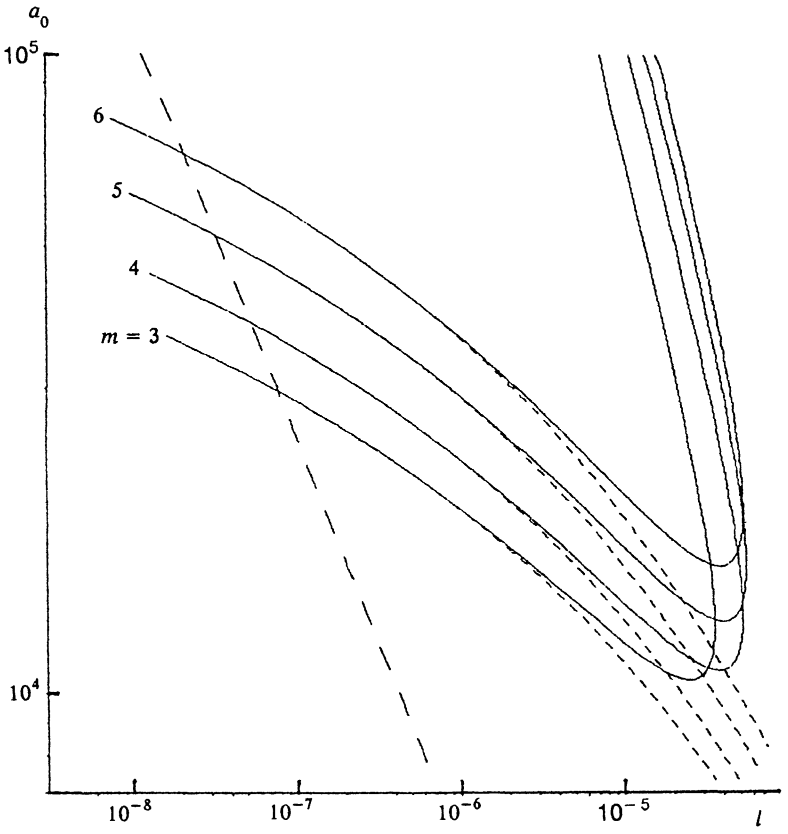

FIG. 1. Instability diagram (29), $\lambda_m=0$, for angular perturbations with the size a_0, and the parameter l parameter as variables, for harmonics with $m=3,...,6$ (numbers given at each curve) and a critical size $a_{cr}=100$. The curves with the short dashes give the solution (30) for the corresponding harmonics (in the $a_0 l \ll 1$ case). The curve with the long dashes gives the condition (31) for the violation of the quasistationary diffusive growth.

sufficiently small values of the parameter l ($l \leqslant 5.6 \cdot 10^{-5}$), i.e., for diffusion lengths L which are appreciable as compared to the width χ of the transition layer. It is necessary in that case that the nucleus have a rather large radius: $a_0 \geqslant a_0^* \approx 10^4$. The growing islands thus have initially at $a_0 < a_0^*$ an axially symmetric shape. When the size a_0^* is reached the unstable harmonic for which the condition $\lambda_m=0$ is realized starts to grow. In the $l \leqslant 3 \cdot 10^{-5}$ range the harmonic with $m=3$ is the first to start its growth [with increasing $a_0(\tau)$]. However, in the range $3 \cdot 10^{-5} \leqslant l \leqslant 5.6 \cdot 10^{-5}$ there is a region in which with increasing $a_0(\tau)$ initially either the harmonic with $m=4$ or the one with $m=5$ becomes unstable, passing respectively the third or the third and fourth harmonics. In the isotropic system considered, the growth of only one harmonic with a number $m \geqslant 6$ is not possible whatever the value of the parameter l. These harmonics ($m \geqslant 6$) can grow in this entire instability region together with other harmonics ($m=3$ to 5). By choosing the physical conditions which realize the appropriate values of l one could thus observe, in a well defined range of island sizes a_0, the growth of perturbations with symmetries of order $m=3$, $m=4$, or $m=5$, whereas other harmonics of the perturbation would be damped in time. After the instability of one of these harmonics has set in, the criteria for the instability of the other harmonics may, in general, change. One can thus determine from Fig. 1 the boundaries of the instability region and the number of the unstable harmonics on a boundary of this region.

We show in Fig. 1 also the solutions $\lambda_m=0$ for the case $a_0 l \ll 1$ of (30) (short-dashed lines for each of the corresponding harmonics) corresponding to the phenomenological growth equations (1) and (2). It is clear from the figure that the phenomenological approach corresponds to values of the parameter l in the $l \leqslant 10^{-4}$ range. The qualitative difference of the instability for $l > 3 \cdot 10^{-5}$ from the results of Ref. 26 is connected with the fact that in that parameter range for clusters which are unstable ($a_0 \geqslant a_0^*$) the flux over the surface of the island becomes important as l increases. This flux leads to vanishing of the instability, first for the harmonic with $m=3$, next for those with $m=4$ and 5, and finally for the other harmonics.

When a_{cr} decreases the obtained instability diagram shifts towards smaller sizes a and larger values of the parameter l (Fig. 2). For instance, for $a_{cr}=1$ the instability occurs already for sizes $a \sim 10^2$. A further formal decrease in a_{cr} in Eq. (29) shifts the instability region to $a \sim 1$, i.e., it is possible to realize instability in the initial stages of the cluster growth. However, the critical sizes $a_{cr} \sim 1$ are limiting cases for the analytical studies of the order parameter equation given here since the degree of metastability h becomes a quantity of the order of unity.

The physical causes for the decrease of a_{cr} are, for instance, a change in the boundary conditions, or in the linear tension coefficient, or in the sticking coefficient. A change in those quantities for clusters of one size can thus lead to a different set of unstable harmonics, i.e., to different structures. This, in turn, leads to a change in the fractal dimensionality (as will be shown below).

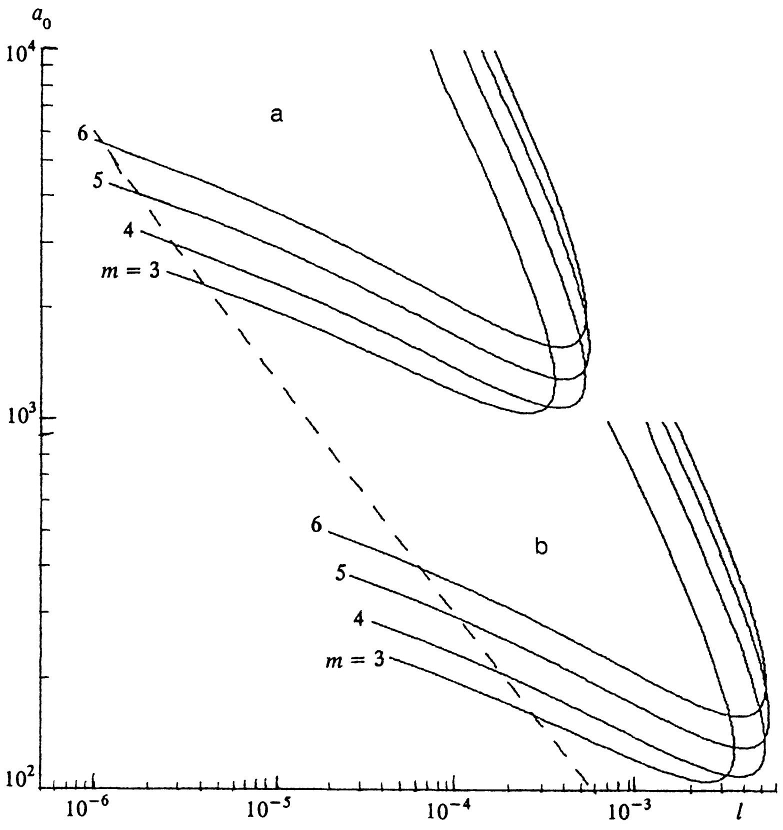

FIG. 2. Instability diagram for values of the critical size of a nucleus $a_{cr}=10$ (a) and $a_{cr}=1$ (b).

4. NONSTATIONARY GROWTH IN THE $l \to 0$ LIMIT

In the foregoing we assumed that the condition for quasistationarity of the function $w(\rho,\tau)$ of (14) was satisfied, which made it possible to drop the term $\dot{w}(\rho,\tau)$ in Eq. (16). Substituting the explicit form of the function $w(\rho,\tau)$ of (24) into that condition and also using Eq. (26) for the growth of an island we can determine more precisely the criterion for the violation of the quasistationarity of the correction to $w(\rho,\tau)$:

$$a_0^3 l^2 I_0(a_0 l) K_0(a_0 l) \approx 1. \tag{31}$$

It is clear that the quasistationarity is violated at short times, i.e., in the initial stages of the growth of the islands when their size is small. In the diffusive limit ($l \ll 1$) considered here the range of sizes and times for which the criterion (32) is satisfied is considerably broadened (Figs. 1 and 2: the dashed line in the left-hand part of the figure). One can see clearly that as $l \to 0$ the curves $\lambda_m=0$ enter the region to the left of the dashed line in which the quasistationarity condition is violated.

The case of diffusive growth of the islands (when there are no processes which do not conserve the number of particles) raises a number of problems connected with the absence of stationary solutions of the two-dimensional diffusion equation. The violation of condition (14) means that one must take into account in Eq. (21) the term $\dot{w}(\rho,\tau)$. We do this in the limiting case $l=0$ when the quasistationarity condition is certainly not realized. We then have for $w(\rho,\tau)$ the equation

$$\dot{a}(\varphi,\tau)\mathrm{ch}^{-2}[\rho-a(\varphi,\tau)]+\dot{w}(\rho,\tau)=4\nabla^2 w(\rho,\tau). \tag{32}$$

Outside the boundary it is the usual nonstationary two-dimensional diffusion equation. Inside the island this equation has a stationary solution which is independent of the coordinates. There is thus no flow inside the island, as is physically obvious since for $l=0$ there are no desorption or adsorption processes. The nonstationary solution of the two-dimensional diffusion equation can be written outside the island ($\rho > a(\varphi,\tau)$) in the form[49,50]

$$w(\rho,\tau) \approx \sum_{m \geq 2} \bar{A}_m(a(\tau))\rho^{-m}\cos(m\varphi) + \begin{Bmatrix} A_0(a(\tau))\left[1-\dfrac{\ln(\rho l)}{\ln(4\sqrt{\tau} l)}\right], & a(\tau) \leq \rho \leq 4\sqrt{\tau} \\ 0, & \rho \geq 4\sqrt{\tau} \end{Bmatrix}. \tag{33}$$

The second term is the well-known nonstationary axially symmetric solution, valid in the limit of large values of the time (τ($\sqrt{\tau}/a_0 \gg 1$). Here $A_0(a(\tau))$ is an unknown quantity determining the function $w(\rho,\tau)$ on the boundary $\rho \approx a(\varphi,\tau)$ of the nucleus. For small times ($\sqrt{\tau}/a_0 \ll 1$) one must take into account the derivative $\dot{w}(\rho,\tau)$ in the whole spatial region, including the boundary. The boundary condition for the function $w(\rho,\tau)$ therefore differs from the one for the case (20) of quasistationary diffusion. However, asymptotically one may assume for $\sqrt{\tau}/a_0 \gg 1$, when (33) is valid, the function $w(\rho,\tau)$ to be quasistationary near the boundary of the island and neglect $\dot{w}$ there. Using (20) we find the coefficients in the solution (33):

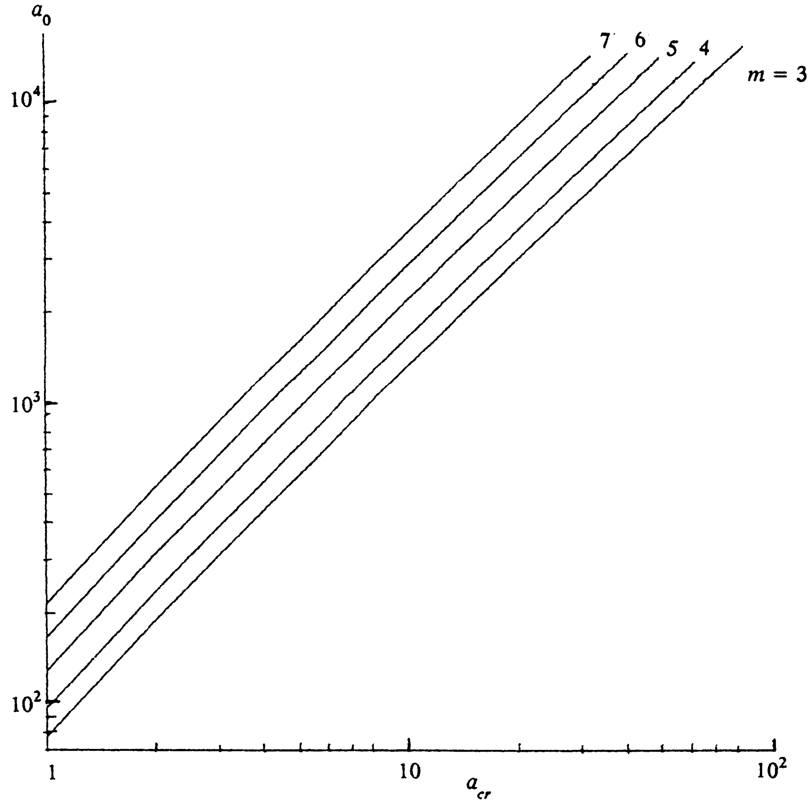

FIG. 3. Instability diagram (37), $\lambda_m=0$, for angular perturbations in the case of a nonquasistationary diffusive growth regime in the $l=0$ limit for the harmonics with $m=3,...,7$.

$$A_0(a(\tau))=\frac{\ln(4\sqrt{\tau l})}{6\ln(4\sqrt{\tau}/a_0)}\left(\frac{1}{a_0}-\frac{1}{a_{\rm cr}}\right),$$
$$A_m(a(\tau))=\frac{m^2-1}{6a_0^2}+\frac{1}{6a_0\ln(4\sqrt{\tau}/a_0)}\left(\frac{1}{a_0}-\frac{1}{a_{\rm cr}}\right), \tag{34}$$

where we have introduced the notation $A_m=\bar{A}_m(a_0^m a_m)^{-1}$. We note that the asymptotic value of $A_0(a(\tau))$ tends to w_0 of (26) as $a_0(\tau)\to l^{-1}\to\infty$, i.e.,

$$\lim_{a_0(\tau)\to\infty} A_0(a(\tau))=w_0.$$

To find the growth equations for the angular perturbations we integrate (32) over the transition layer (where we can already neglect $\dot{w}$). We use the conditions that the deviation of the shape of the island from the cylindrical one is small, similarly to what we did in the diffusive limit. The asymptotic expressions (in the limit of large time values $\sqrt{\tau}/a_0\gg 1$) for the growth rates of the harmonics in a system with a conserved order parameter ($l\to 0$) have the form

$$\dot{a}_0(\tau)=\frac{1}{3a_0\ln(4\sqrt{\tau}/a_0)}\left(\frac{1}{a_{\rm cr}}-\frac{1}{a_0}\right), \tag{35}$$

$$\frac{d}{d\tau}\ln a_m(\tau)=\frac{m-1}{3a_0^2}\left\{\frac{1}{\ln(4\sqrt{\tau}/a_0)}\left(\frac{1}{a_{\rm cr}}-\frac{1}{a_0}\right)-\frac{m(m+1)}{a_0}\right\}. \tag{36}$$

Equation (35) for the growth of a cylindrical island is known from the phenomenological theory. It is asymptotic in the long-time limit $\sqrt{\tau}/a_0\gg 1$.[49,50]

For an analysis of the instability region of the growth of the harmonics we find the growth rates of the perturbations in systems where the particle number is conserved:

$$\lambda_m=\frac{1}{3a_0^2}\left\{\frac{(m-2)}{\ln(4\sqrt{\tau}/a_0)}\left(\frac{1}{a_{\rm cr}}-\frac{1}{a_0}\right)-\frac{m(m^2-1)}{a_0}\right\}. \tag{37}$$

Neglecting the feeble dependence of the function $\ln(4\sqrt{\tau}/a_0)$ on the mean radius and the time we can write down the condition for instability of the mth harmonic ($\lambda_m\geqslant 0$):

$$a_0\geqslant(a_0)_m\equiv a_{\rm cr}\left[1+\frac{m(m^2-1)}{m-2}\ln\left(\frac{4\sqrt{\tau}}{a_0}\right)\right]. \tag{38}$$

The difference between island radii corresponding to condition (38) for the $(m+1)$st and the mth harmonic increases for large values of m approximately as $ma_{\rm cr}\times\ln(4\sqrt{\tau}/a_0)$. As a_0 increases instability sets in successively for all numbers $m\geqslant 3$.

In the $a_0\gg a_{\rm cr}$ limit and in the approximation of a weak dependence of the logarithm on its argument we can integrate the equation for the growth of the radius of an island, and the quantity a_0 is proportional to $\sqrt{\tau}$. Condition (38) takes then the form

$$a_0\geqslant(a_0)_m\equiv a_{\rm cr}\left[1+\frac{m(m^2-1)}{m-2}\ln(24a_c)\right]. \tag{39}$$

The weak time dependence drops out and there remains only a dependence on the critical size of a nucleus. It is clear that for small a_{cr} ($a_{cr} \gg 1$) the sizes for which the harmonics become unstable are also rather small (see Fig. 3).

5. COMPARISON WITH THE DLA CONTINUUM MODEL IN THE DIFFUSIVE LIMIT. FRACTAL DIMENSIONALITY

Let us compare the approach based upon the order parameter equation and the DLA model. In an analytical formulation of the DLA model the equations of that model are usually written in the form[5-11,27,28]

$$\nabla^2 u(\boldsymbol{\rho})=0, \tag{40a}$$

$$u|_i=1-\varkappa(\boldsymbol{\rho}_i), \tag{40b}$$

$$u(\rho\to\infty)=0, \tag{40c}$$

$$v_n=-\frac{(\mathbf{n},\nabla u)}{4\pi}\bigg|_i. \tag{40d}$$

The subscript i indicates here that a quantity is taken at the boundary of a cluster, $\varkappa(\boldsymbol{\rho})$ is interpreted as the curvature of the cluster boundary, and v_n is the normal rate of growth of a cluster. One checks easily that the function u of the DLA model is the analog of the field w of (11). Indeed, Eq. (40a) for the function u and condition (40c) at large distances are the same as the corresponding Eq. (23) in the diffusive limit $l\to 0$ and condition (15) for the field w. The growth rate (40c) is, apart from a numerical coefficient, the same as the rate $\dot a$ of (22) (for $l\to 0$). To compare the conditions (40b) and (25) at the cluster boundary we express (25) in terms of the curvature. In the case of small perturbations of the axial shape of an island, we have for the curvature

$$k(\varphi,\tau)=\frac{a^2+2a'^2-aa''}{(a^2+a'^2)^{3/2}}\approx\frac{a-a''}{a^2}. \tag{41}$$

Using this relation we can write condition (25) for the field w at the boundary in the form

$$w(\boldsymbol{\rho}_i,\tau)=-[1-k(\varphi,\tau)a_{cr}]/6a_{cr}. \tag{42}$$

It is clear that conditions (40b) and (42) are identical and for the quantities occurring in the DLA model and in the order parameter equation there are the relations

$$6a_{cr}w=-u,\quad a_{cr}k=\varkappa,\quad 3\dot a_0/4\pi=v_n. \tag{43}$$

Hence it follows that for $a_{cr}\sim 1$ the parameters of the order-parameter equation are essentially the same as the parameters of the DLA model.

In the course of solving the order-parameter equation we have thus studied the growth of the angular perturbations of the cluster boundary. Moreover, the main criterion in fractal growth models[5-11,27-31] is the fractal dimensionality $\mathscr{D}$. Let us study the effect of angular perturbations of the boundary on the magnitude of $\mathscr{D}$. Consider the growth of a two-dimensional cluster with one unstable harmonic m:

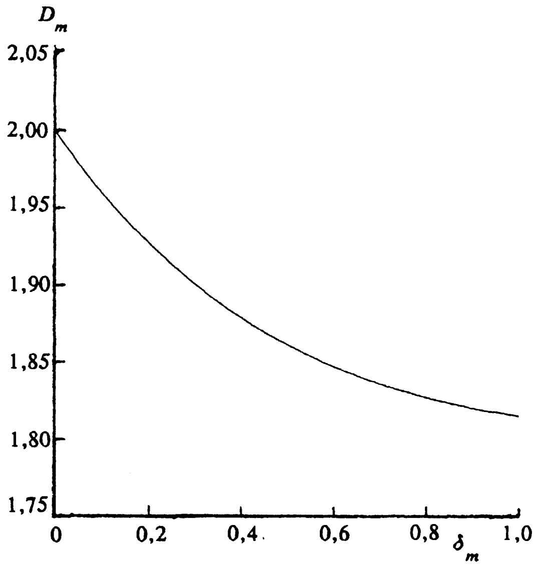

FIG. 4. The fractal dimensionality (45) as function of the amplitude ($\delta_m=a_m/a_0$) of the growing mth harmonic.

$$a(\varphi)=a_0+a_m\cos(m\varphi). \tag{44}$$

From the definition of the fractal dimensionality (see the Introduction) it follows that the area of the cluster is proportional to $R^{\mathscr{D}}$ [where R is the maximum size, which in the case (44) is equal to $R=a_0+a_m$]. As a result we get for $\mathscr{D}=D_m$:

$$D_m=2\frac{\ln[a_0(1+\delta_m^2/2)^{1/2}]}{\ln[a_0(1+\delta_m)]},\quad \delta_m=\frac{a_m}{a_0}, \tag{45}$$

We note that a cluster with a surface shape (44) or (12) is not a fractal object. Nonetheless, it is convenient to retain for the quantity (45) the term "fractal dimensionality," bearing in mind that the result of the development of the instability of similar types of cluster is a fractal. The fractal dimensionality of the cluster (45) is independent of the number m of the harmonic and determined only by the amplitudes a_0 and a_m. It is clear from Fig. 4 that the appearance of a nonspherical harmonic causes the quantity $\mathscr{D}$ to deviate from the Euclidean dimensionality $n=2$. As the ratio a_m/a_0 increases, the value of $\mathscr{D}$ decreases monotonically. The increase in the number of unstable harmonics leads to a yet larger decrease in the fractal dimensionality. One checks easily that in the case of the growth of a cluster with two unstable harmonics m and l the fractal dimensionality is $D_{m,l}<D_m,D_l$. For instance, in the initial stage of the growth the relation

$$D_{m,l}=D_m+D_l-2 \tag{46}$$

is satisfied.

A change in the structure of the unstable mode leads thus to a change in the fractal dimensionality. Moreover, we have already noted earlier that a change in the structure of an unstable mode is made possible by a change in the conditions at the cluster boundary. This agrees with the results of a simulation of fractal clusters in which a change in the boundary conditions leads to a change in the fractal dimensionality.

6. CONCLUSION

We have studied in the present paper the stability of the axial shape of two-dimensional clusters, starting from a Ginzburg–Landau type equation for the order parameter field. We have obtained criteria for the instability of harmonics and determined the size R (a_0 in dimensionless variables) of a cluster and the quantity l which is the ratio of the contributions from growth mechanisms which conserve and which do not conserve the order parameter. We have shown that shape instability occurs when the main cluster growth mechanism is diffusive (diffusion of adatoms along the surface of the material). As the processes which do not conserve the order parameter (adsorption and desorption) get stronger, the fraction of the flow along the surface of the cluster increases and this suppresses the instability. In the $a_0 l \ll 1$ limit the instability criteria obtained are the same as the results of the studies of Ref. 26 which were carried out using the phenomenological Zel'dovich–Folmer theory. In the diffusive limit $l \to 0$ the approach based on the order parameter equation is for $a_{cr} \sim 1$ analogous to the DLA model for fractal clusters. We have shown that the growth of the angular harmonics and the increase in their number leads to a lowering of the fractal dimensionality.

It follows from the results obtained that the phenomenon of an instability of a symmetric shape of a nucleus of a new phase has a universal nature in the case of a diffusive growth mechanism and does not require an additional anisotropy of the system. Such an anisotropy makes the occurrence of an asymmetry in the shape easier. We note that anisotropy may arise as the result of a phase transition when a nucleus leads to an appreciable deformation of the surrounding crystal.[51,52]

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