Tuned Mullins-Sekerka instability: Exact results

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Mullins-Sekerka's instability at 3D self-similar growth of a spherical seed crystal in an undercooled fluid is discussed. The exact solution of the linearized stability problem is obtained. It is quite different from the conventional results of the quasisteady approximation. The instability occurs much weaker, so that instead of exponential growth in time, unstable modes exhibit just power-law-growth. The relative growth rates of different modes vary in time and depend on their initial amplitudes. It allows control over the growth of each mode individually and tailoring the instability, to obtain a desired shape of the growing crystal at a given time.

I. INTRODUCTION

Exact solutions play a unique role in physics. The point is that often important new features of a problem are observed in a range of parameters, lying beyond the applicability conditions of various approximate approaches, so that the exact solution is the only way to detect and study these features. In this paper, we present a new class of exact solutions of a linear stability problem describing the Mullins-Sekerka instability of self-similar spherically symmetric growth of a spherical seed.

Discovered more than 50 years ago [1,2] the Mullins-Sekerka instability is still studied extensively (see, e.g., Ref. [3]). The manifestation of the instability has been observed far beyond the initial narrow framework of crystal growth. It may control tumor formation [4], supramolecular networks [5], formation of electron-hole drops in semiconductors [6,7], ripples in graphene [8], etc.

In the present paper, we show that the self-similarity together with extension of the problem beyond the commonly utilized quasisteady approximation [9] changes the instability qualitatively, so that exponential growth of unstable modes is replaced by the one controlled by power-laws. While a characteristic time of the exponential instability (inverse increment) is determined by the intrinsic properties of the corresponding stability problem, the power-law-controlled instability does not possess any intrinsic characteristic time at all. For each unstable mode a characteristic time to grow is determined by the initial amplitude of this mode. Moreover, the relative rates of growth of different modes vary in time and also depend on their initial amplitudes. It provides a new, unique opportunity controlling the instability and even tailoring it to obtain a crystal with a desired shape, which may be important for numerous applications.

II. PROBLEM FORMULATION

The conventional Mullins-Sekerka instability [1,2] arises either at diffusion-controlled growth of a solid phase into a supersaturated solution, or at solidification of an undercooled melt. The theory of the phenomenon is practically identical in

both the cases. For definiteness in what follows we consider the solidification of a melt.

The physical grounds for the instability have been pointed out in the pioneer paper of Mullins and Sekerka [1]. Solidification results in a latent heat release. The released heat is transferred from the solid-liquid interface to the undercooled melt by heat diffusion. The larger the temperature gradient in the vicinity of the interface the better the heat transfer and hence the larger the solidification rate. If a bulge arises at the interface, it results in a sharpening of the temperature gradient in the vicinity of the bulge. Therefore, the interface velocity at the tip of the bulge becomes larger than that at the rest of the interface. It creates positive feedback, resulting in the instability; see Fig. 1.

III. MODEL

To simplify the problem we neglect the difference in densities of the solid and liquid phases. In this case the solidification does not induce any mass transfer, being controlled by the heat transfer entirely. We also neglect anisotropy of the solid phase, so that a spherically symmetric solution to the problem is admitted. Finally, we suppose that the solidification temperature T_s is a material constant T_{s0} (the Stefan boundary condition).

Note, actually, T_s is not a constant. It depends on the interface curvature through the capillary effect [1]. Owing to this dependence, short-wavelength perturbations to the interface are stabilized. As a result, a certain characteristic scale for the most rapidly growing perturbations comes into being. It plays the key role in the pattern selection processes, describing, e.g., the dendritic structure of crystals [9]. The neglect of the capillary effect removes the characteristic scale for the most "dangerous" perturbations. Then, it seems the assumption $T_s = $ const makes the problem ill-posed.

In this connection, it is relevant to mention a series of mathematically elegant publications devoted to exact nonlinear solutions of the so-called 2D Laplacian growth, closely related to the problem in question; see, e.g., Ref. [10] and references therein. In the absence of the capillary effect most of these solutions show finite-time singularities via the formation of cusps [11–13]. The cusps make the exact solutions physically meaningless.
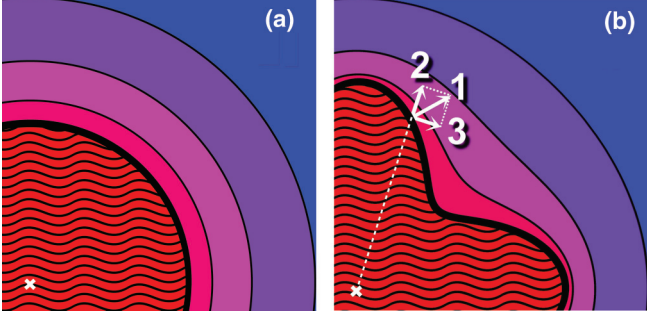
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FIG. 1. (Color online) A temperature profile in an undercooled melt in the vicinity of a growing crystal (schematically). The solid phase is indicated by filling with a wavy pattern. The solid-liquid interface is shown with a thick line. Thin lines designate the sequential isotherms with fixed temperature step ΔT. The unperturbed spherically symmetric crystal (a) and fourfold deformed one (b). The deformation gives rise to an increase in the modulus of the temperature gradient at convex parts of the interface and in decrease of it at concave ones (note the corresponding changes of the distances between the sequential isotherms). Moreover, perturbed heat flow (1) in addition to radial (2) gets an azimuthal component (3) directed from the convex to concave parts of the interface; see arrows in (b). Both effects enhance the heat transfer from the bulges and suppresses the one from the concave parts.

Fortunately in our case the situation is not so dramatic. If a system exhibits an instability, growth of the amplitudes of the unstable modes sooner, or later drives it beyond the applicability conditions of the corresponding linear stability problem. Thus, any linear stability problem always has a finite lifetime. For the problem in question the neglect of the capillary effect imposes certain constraints on applicability of the results of the subsequent analysis. Since in the present paper we are restricted by the framework of the linear stability analysis solely, to justify the fixed-temperature boundary condition the constraints should hold until the linear stability problem itself becomes invalid. The corresponding quantitative conditions will be discussed later on.

IV. UNPERTURBED SOLUTION

To begin with, we consider spherically symmetric growth of a seed crystal. It is convenient to introduce dimensionless temperature $\theta = (T - T_\infty)C/L$. Here C stand for the specific heat of the melt; L is the solidification latent heat and T_∞ designates the initial temperature of the undercooled melt ($T_\infty < T_{s0}$). Under the assumptions made the solidification process is described by the following boundary-value problem with a moving boundary [14]:

$$\frac{\partial \theta}{\partial t} = \chi \frac{1}{r^2} \frac{\partial}{\partial r}\left(r^2 \frac{\partial \theta}{\partial r}\right), \tag{1}$$

$$\theta|_R = \theta_s = \text{const} > 0, \tag{2}$$

$$\chi \frac{\partial \theta}{\partial r}\bigg|_R = -\frac{dR}{dt}, \tag{3}$$

$$\theta \to 0 \text{ at } r \to \infty, \tag{4}$$

$$\theta(0, r) = 0. \tag{5}$$

Here χ stands for the heat diffusivity of the melt, $\theta_s = (T_{s0} - T_\infty)C/L$, and $R(t)$ is the radius of the growing crystal. Note, function $R(t)$ is not known *a priori*. It should be defined as a result of the problem solution. Thus, in contrast to the conventional heat transfer problems, where a temperature profile is a single unknown function, now we have two unknown functions: $\theta(r,t)$ and $R(t)$. Because of that the problem in question requires an extra boundary condition. The condition does not follow from the energy conservation law, but should be derived from the solidification kinetics. In our case this condition is Eq. (2), stipulated for the temperature at the interface to be equal to T_{s0}. Let us stress that Eq. (2) means that the temperature inside the solid phase is $T_{s0} = \text{const}$; i.e., there is no heat transfer at $r < R$.

The only dimensional constant in Eqs. (1)–(5) is χ. It does not allow to build up dimensionless spatial and temporal variables separately. The only way to introduce a dimensionless variable in this case is to consider ratio $r/\sqrt{\chi t}$. Therefore, according to the general principles of dimensional analysis, instead of being a function of two independent variables, r and t, the solution to the problem should be a function of just a single variable proportional to this ratio, e.g.,

$$\xi = \frac{r}{2\sqrt{\chi t}}. \tag{6}$$

The corresponding ordinary differential equation is integrated easily. Its solution, satisfying the specified initial and boundary conditions, is as follows:

$$\theta = \theta_0(\xi) \equiv 2\alpha^3 \exp(\alpha^2) \int_\xi^\infty \frac{\exp(-\eta^2)}{\eta^2} d\eta, \tag{7}$$

$$R = R_0(t) \equiv 2\alpha \sqrt{\chi t}, \tag{8}$$

where α is a root of transcendental equation:

$$2\alpha^3 \exp(\alpha^2) \int_\alpha^\infty \frac{\exp(-\eta^2)}{\eta^2} d\eta = \theta_s. \tag{9}$$

Though a formal solution of Eq. (9) exists at any $\theta_s < 1$, physically meaningful are only small (relative to L/C) undercooling, when $\theta_s \ll 1$. In this case, Eq. (9) admits a simple approximate solution:

$$\alpha \simeq \sqrt{\theta_s/2}. \tag{10}$$

V. STABILITY PROBLEM: QUALITATIVE ANALYSIS

Owing to the Mullins-Sekerka instability the obtained spherically symmetrical solution should be unstable against perturbations breaking the symmetry. To get some hints as to how to attack the corresponding stability problem, let us remember certain general issues of the stability analysis. Usually, an arbitrary small perturbation is expanded in terms of the eigenfunctions of the unperturbed problem. Then, the evolution of every separate mode in the linearized stability problem may be studied independently. For a steady unperturbed solution the temporal dependence of a given nth mode is supposed to have the form $\exp(\gamma_n t)$, where the specific dispersion law (the dependence γ_n on n) is defined by the solvability conditions. For more details, see, e.g.,Refs. [15,16].

If the unperturbed solution is time-dependent, but its characteristic temporal scale is small relative to the one for the instability to arise, a quasisteady approximation may be employed. In this case γ_n becomes time-dependent too and $\exp(\gamma_n t)$ transforms into $\exp[\int \gamma_n(t) dt]$.

Let us see how it affects instability of blowing up solutions arising in self-similar problems, which do not allow us to build from their constants a quantity with dimension of time. In these cases the only possibility to make the dimension of γ_n equal to $[s^{-1}]$ is to suppose that $\gamma_n = \mu_n/t$, where μ_n is a dimensionless constant. Then, $\int \gamma_n(t) dt = \int (\mu_n/t) dt = \mu_n \log t$, and instead of the exponential growth we obtain a much slower one, controlled by the power law: t^{μ_n}, that is the instability is partly stabilized.

The physical grounds for the stabilization are related to the fact that usually the instability rate is controlled by the characteristic values of the gradients of the corresponding variables (see the arguments, explaining the Mullins-Sekerka instability presented above). For blowing up solutions the characteristic values of the gradients decrease with the course of time and hence the instability rate should decrease too. Thus, the hint is that the instability should be controlled by a power law [17].

VI. STABILITY PROBLEM: RIGOROUS ANALYSIS

To proceed with the rigorous analysis, we have to generalize the boundary-value problem Eqs. (1)–(5) to the case of a non-spherically symmetric temperature field. The generalization reads [18]

$$\frac{\partial \theta}{\partial t} = \chi \Delta \theta, \tag{11}$$

$$\theta|_{R(\vartheta,\varphi,t)} = \theta_s = \text{const} > 0, \tag{12}$$

$$\chi(\mathbf{n} \cdot \nabla \theta)|_{R(\vartheta,\varphi,t)} = -\left(\mathbf{n} \cdot \frac{\partial \mathbf{R}}{\partial t}\right) \tag{13}$$

$$\theta \to 0 \quad \text{at} \quad r \to \infty, \tag{14}$$

$$\theta(0, r) = 0, \tag{15}$$

where $R(\vartheta,\varphi,t)$ is an unknown function, describing the position of the interface according to equation $r = R(\vartheta,\varphi,t)$ and $\mathbf{n}$ is the normal vector.

To analyze the linear stability of solution Eqs. (7) and (8) we perturb it with arbitrary smooth small perturbations: $\theta = \theta_0 + \epsilon \theta_1$, $R = R_0 + \epsilon R_1$, where ϵ is a small constant. Then, it is convenient to project the boundary conditions, imposed at the perturbed interface, to the unperturbed boundary $r = R_0(t)$, expanding them in powers of small ϵR_1 and taking into account terms of the zeroth and first orders in ϵ only [18,19]. In this case, Eq. (12) transforms as follows:

$$\theta|_{R_0+\epsilon R_1} \simeq \theta_0|_{R_0} + \left.\frac{\partial \theta_0}{\partial r}\right|_{R_0} \epsilon R_1 + \epsilon \theta_1|_{R_0} = \theta_s.$$

Next, bearing in mind that $\theta_0|_{R_0} = \theta_s$, see Eq. (2), and that $\partial \theta_0/\partial r|_{R_0} = -d R_0/\chi dt = -\alpha/\sqrt{\chi t}$, according to Eqs. (3)

and (8), we eventually obtain

$$\theta_1|_{R_0} = \frac{\alpha R_1}{\sqrt{\chi t}}. \tag{16}$$

Treating Eq. (13) in an analogous manner, it should be taken into account that $n_r = O(1)$ and $n_\vartheta \sim n_\varphi = O(\epsilon)$. The treatment yields

$$\chi R_1 \left.\frac{\partial^2 \theta_0}{\partial r^2}\right|_{R_0} + \chi \left.\frac{\partial \theta_1}{\partial r}\right|_{R_0} + \frac{\partial R_1}{\partial t} = 0. \tag{17}$$

Note, that $\partial^2 \theta_0/\partial r^2$, entering into this condition, may be expressed in terms of $d\theta_0/d\xi$, according to Eq. (1), and that $(d\theta_0/d\xi)_{R_0} = -2\alpha$; see Eqs. (3), (6), and (8).

In the subsequent analysis it is convenient to transfer from r to ξ. Then, in accord with what has been said above, the eigenfunctions of the linearized stability problem should have the form [20]

$$\theta_1 = t^\mu f_\ell(\xi) Y_\ell^m(\vartheta,\varphi), \tag{18}$$

$$R_1 = A t^\zeta Y_\ell^m(\vartheta,\varphi), \tag{19}$$

where A is a constant and $Y_\ell^m(\vartheta,\varphi)$ stand for the Laplace spherical harmonics.

Next, Eqs. (16) and (17) yield

$$\zeta = \mu + \tfrac{1}{2}, \tag{20}$$

Thus, the stability problem is reduced to a boundary-value problem for $f_\ell(\xi)$. It is convenient to join two boundary conditions Eqs. (16) and (17), dividing one by the other. Finally, the problem reads as follows:

$$\frac{d^2 f_\ell}{d\xi^2} + \left(\xi + \frac{1}{\xi}\right)\frac{df_\ell}{d\xi} - \left[4\mu + \frac{\ell(\ell+1)}{\xi^2}\right]f_\ell = 0, \tag{21}$$

$$\left.\frac{d}{d\xi}(\log f_\ell)\right|_\alpha = -\frac{2(\mu + \alpha^2) + 3}{\alpha}, \tag{22}$$

$$f_\ell \to 0 \quad \text{at} \quad \xi \to \infty. \tag{23}$$

It is worth mentioning that Eqs. (21)–(23) do not depend on m, that is each eigenvalue μ_ℓ has $(2\ell + 1)$-fold degeneracy.

Equation (21) is exactly integrable. The integral, satisfying boundary condition Eq. (23), is

$$f(\xi) = \xi^{-3/2} \exp\left(-\frac{\xi^2}{2}\right) W_{\lambda,\nu}(\xi^2), \tag{24}$$

where $W_{\lambda,\nu}(z)$ stands for the Whittaker function [21] and

$$\nu = \frac{2\ell + 1}{4}, \quad \lambda = -\left(\mu + \frac{3}{4}\right). \tag{25}$$

Among various presentations of $W_{\lambda,\nu}(z)$ we select the following [21]:

$$W_{\lambda,\nu}(z) = B z^\lambda e^{-z/2} \int_0^\infty x^{\nu-\lambda-\frac{1}{2}} e^{-x}\left(1 + \frac{x}{z}\right)^{\nu+\lambda-\frac{1}{2}} dx. \tag{26}$$

Here B is a constant.

The only remaining condition to be satisfied is Eq. (22). Substitution of Eqs. (24)–(26) into Eq. (22) yields

$$\frac{(\lambda + \nu - \frac{1}{2}) \int_0^\infty \frac{x^{\nu - \lambda + \frac{1}{2}}}{\alpha^3} (1 + \frac{x}{\alpha^2})^{\lambda + \nu - \frac{3}{2}} e^{-x} dx}{\int_0^\infty x^{\nu - \lambda - \frac{1}{2}} (1 + \frac{x}{\alpha^2})^{\lambda + \nu - \frac{1}{2}} e^{-x} dx} = 0. \quad (27)$$

Note, that the integrands in Eq. (27) both are nonnegative and the integrals converge at $\mu > -(\ell + 3)/2$, that is for all unstable modes, if any. Thus, the integrals in the left-hand-side of Eq. (27) are certain finite-positive quantities. In this case, Eq. (27) holds if and only if the prefactor in the numerator of its left-hand-side vanishes: $\lambda + \nu - \frac{1}{2} = 0$. It yields the dispersion relation

$$\mu_\ell = \frac{\ell}{2} - 1; \quad \zeta_\ell = \frac{\ell - 1}{2}; \quad (28)$$

see Eqs. (20) and (25). It is important to stress that exponents μ_ℓ and ζ_ℓ depend on ℓ solely and *do not depend on the undercooling rate* θ_s.

VII. DISCUSSION OF THE RESULTS

The temporal evolution of the perturbations to the interface is controlled by exponent ζ_ℓ; see Eqs. (19) and (28). The perturbations with $\ell = 0$, which do not break the spherical symmetry of Eqs. (7) and (8), decay. It is an obvious consequence of the uniqueness of the solution of Eqs. (1)–(5) given by Eqs. (7) and (8). Perturbations with $\ell = 1$ are time-independent. It follows from the translational symmetry of the problem, because in the linear approximation these perturbations correspond just to a shift of the center of the coordinate frame for solution Eqs. (7) and (8). Perturbations with $\ell = 2$ grow as $\sqrt{t}$, that is with the same rate as $R_0(t)$ does. It occurs because in the linear approximation such perturbations transform the spherically symmetric solution Eqs. (7) and (8) into ellipsoidal. On the other hand, the problem in question admits an exact solution with the ellipsoidal symmetry analogous to Eqs. (7) and (8) [22]. These *rigorous* results confirm the ones of Ref. [1] obtained in a quasisteady approximation.

According to Eq. (28) at $t \to \infty$ all other modes grow faster than the unperturbed solution and therefore should be regarded as unstable. The exponent ζ_ℓ, i.e., the instability rate, increases linearly with an increase in ℓ. Meanwhile, actually, as it already has been pointed out, perturbations with large enough ℓ are stabilized owing to the capillary effect [1]. The obtained unlimited increase of ζ_ℓ at large ℓ is an apparent defect of our model related to the employment of the plain Stefan boundary condition [Eqs. (2) and (12)] with the fixed solidification temperature T_{s0}.

Thus, the model in question should have a limited range of validity. To find the corresponding applicability conditions, let us modify Eqs. (2) and (12), taking into account the mentioned dependence of the solidification temperature on the interface curvature $\mathcal{K}$. Expanding this dependence in powers of small $\mathcal{K}$ and dropping higher order terms, we obtain $T_s \simeq T_{s0}(1 - \Gamma\mathcal{K})$, where Γ is the capillary constant.

Utilization of this modified expression for the solidification temperature contributes to the problem a constant with dimension of length and hence breaks the self-similarity. Therefore, the modification of the boundary condition affects both the

unperturbed spherically symmetric problem and the problem of its stability. For the former it is seen straightforwardly that the capillary correction to T_{s0} is negligible provided $R_0 \gg R_c$, where $R_c = 2\Gamma T_{s0}/(T_{s0} - T_\infty)$ is the radius of a critical nucleus at the given undercooling. For the later the question is a bit more tricky.

Taking into account that for a slightly perturbed sphere the mean curvature may be presented as [1]

$$\mathcal{K} \simeq \frac{2}{R_0} \left(1 - \epsilon \frac{R_1}{R_0}\right) - \epsilon \frac{\Delta_{\theta,\varphi} R_1}{R_0^2},$$

where $\Delta_{\theta,\varphi}$ stands for the angular part of the Laplacian expressed in spherical coordinates, and that $\Delta_{\theta,\varphi} Y_\ell^m = -\ell(\ell + 1) Y_\ell^m$, the modified boundary condition Eq. (16) reads

$$\theta_1|_{R_0} = \left[\frac{\alpha}{\sqrt{\chi t}} - \Gamma\theta_s \frac{(\ell + 2)(\ell - 1)}{R_0^2}\right] R_1. \quad (29)$$

Our model is physically adequate provided the second term in square brackets in Eq. (29) is small relative to the first, which allows reducing Eq. (29) to Eq. (16). Bearing in mind Eqs. (8) and (10), this condition may be written as follows:

$$(\ell + 2)(\ell - 1) \simeq \ell^2 \ll \frac{R_0(t)}{\Gamma}; \quad (30)$$

see Eq. (8). Since ℓ is not bounded from above this condition inevitably is violated at $\ell = O(R_0/\Gamma)$. The evolution of such perturbations is not described by the developed theory. These perturbations are stabilized [partly at $\ell = O(R_0/\Gamma)$, or completely at $\ell \gg R_0/\Gamma$] owing to the capillary effect [1]. However, due to inequalities $\Gamma \ll R_c \ll R_0(t)$ for the problem in question the stabilization occurs at very large values of ℓ, which often may correspond to the scales lying below the one required for applicability of the developed macroscopic approach itself, especially at manifestations of the Mullins-Sekerka instability in unconventional problems, such as those discussed in Refs. [4–8]. Note also, while the condition $R_0(t) \gg R_c$ depends on the rate of undercooling $T_{s0} - T_\infty$, Eq. (30) does not.
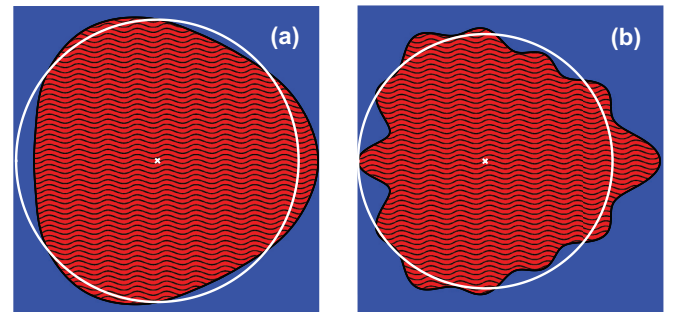

FIG. 2. (Color online) Example, illustrating the opportunity of shape-control. Temporal evolution of the cross-section of a growing crystal by an equatorial plane passing through z axis. The spherical seed is perturbed by two eigenmodes with the same amplitudes; ℓ equals 3 and 12, respectively: $\frac{R}{2\sqrt{\chi t}} = 0.1 + 0.01\sqrt{\frac{t}{\tau}} Y_3^0 + 0.01(\frac{t}{\tau})^5 Y_{12}^0$. The solid phase is indicated with a wavy pattern. (a) At $t = 0.5\tau$ the shape is determined by the smoother perturbation with $\ell = 3$. (b) At $t = \tau$ it is strongly affected by the perturbation with $\ell = 12$.

Applying the results obtained to describe the evolution of an actual small but finite perturbation, we have to specify its initial amplitude. The amplitude provides the characteristic spatial Λ and temporal $\tau = \Lambda^2/\chi$ scales. The specific choice of these scales depends on the type of the initial perturbation (induced, spontaneous, etc.). Discussion of this issue lies beyond the scope of the present paper. However, just the fact that τ exists, together with the obtained power-law growth of the unstable modes results in the conclusion that at $t \ll \tau$ perturbations with small ℓ play the dominant role, while at $t \gg \tau$ the case is opposite; see Fig. 2. It should be stressed that Λ and τ may depend on ℓ, being different for different modes. It provides additional opportunities to control and tailor the instability.

VIII. CONCLUSION

The exact solution for the linearized stability problem of self-similar crystal growth has been obtained. The solution exhibits qualitative differences with respect to the conventional approximate results. Specifically, instead of exponential growth of unstable modes it yields a much slower one, controlled by power laws. The exponents of these laws have been obtained in the explicit form; see Eq. (28). The characteristic time for each unstable mode is defined by its initial amplitude: $\tau_\ell = \Lambda_\ell^2/\chi$. The applicability conditions for the developed analysis read $R_0(t) \gg R_c$; $\ell^2 \ll R_0(t)/\Gamma$. These results shed new light on the old important phenomenon and provide a new opportunity to control the shape of the growing seed by tuning initial amplitudes of different unstable modes.

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