# Structure and evolution of the nucleus of a new phase in first-order phase transitions

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We use a generalized Ginzburg–Landau equation which describes the evolution of the order-parameter field to study the nucleus-forming process in first-order phase transitions in two- and three-dimensional systems. We establish that even in a system with isotropic symmetry the critical nucleus is in the general case not spherical and that the size of a typical nucleus is determined by the scale on which the interaction which is responsible for the formation of the ordered phase decreases. We study the relaxation of an arbitrary fluctuation field and show that spherical nuclei with a size which is much larger than the wall thickness (which are assumed to be the critical ones in a simple phenomenological theory) are appreciably supercritical and are formed only at a relatively late stage of the evolution.

## INTRODUCTION

It is well known that the transition of a metastable phase into a stable one proceeds via a fluctuating onset and subsequent growth of a nucleus of a new phase in a homogeneous medium. One of the key problems in the kinetics of first-order phase transitions (PT1) is the problem of the appearance and the structure of a critical nucleus—a large-scale fluctuation of the order parameter initiating the transition of the whole of the distributed system from a metastable into an absolutely stable state.[1–3]

We consider in the present paper the evolution of arbitrary fluctuations of the order-parameter field in an isotropic $d$-dimensional space and the nucleus formation in a metastable medium, essentially leaving aside the kinetics of coalescence processes. The latter may be decisive only in the concluding stages of the evolution of a system undergoing a phase transformation when the metastability of the "matrix" in which the nucleus grows becomes very weak.[4,5]

In the framework of a phenomenological theory, the evolution of the order parameter in nonequilibrium systems is usually described by a generalized Ginzburg–Landau equation.[6] This is a nonlinear diffusion-type equation which in the general case has the form

$$\varphi_t = -\gamma \frac{\delta \mathscr{F}}{\delta \varphi}, \tag{1}$$

where $\mathscr{F}$ is the Ginzburg–Landau functional of the system considered, $\gamma$ is a positive kinetic coefficient, $\varphi(x,t)$ is the order-parameter field, and $\varphi_t$ is its time derivative. In the linear approximation this equation describes the relaxation of the order parameter to the equilibrium position, and it first appeared in a well known paper by Landau and Khalatnikov.[7] Later on it was used to describe the motion of the boundary between a solid and a liquid phase in the theory of crystal growth,[8] and as the soliton aspects of the theory of nonlinear equations were developed it was very successfully exploited in one-dimensional models for studying localized excitations near a phase-transition point and the dynamics of a plane phase-transformation front (see, e.g., Refs. 9 to 15).

Following the above-mentioned papers, we assume that the evolution of the order parameter is completely determined by Eq. (1), i.e., the framework of the model (1) will be a natural constraint on the applicability of the results. The true physical picture of the processes in PT1 may be much more elaborate because in actual systems a PT is accompanied as a rule by the appearance of inhomogeneous long-range fields. It is well known that the latter determine to a considerable extent even the very possibility of decay of a system into coexisting phases.[16,17]

We use the model (1) to consider in the present paper the kinetics of nucleus formation and the subsequent growth of domains of the new phase in two- and three-dimensional systems with a polynomial Ginzburg–Landau functional. The basic numerical and theoretical calculations are carried out for two kinds of expansion of the free energy density which allow us to describe PT1:

a) a $\varphi^4$ model including a third-order term;

b) a free-energy expansion containing even powers of the order parameter up to $\varphi^6$.

In the first sections of the paper we concentrate mainly on the structure and the stability of an isolated critical nucleus of the energetically favourable phase inside the metastable one. We consider the growth of an isolated domain of the new phase and find the characteristic velocities of the domain wall motion.

In subsequent sections we use numerical experiments to study the evolution of arbitrary mesoscopic inhomogeneities in the order parameter field. Such inhomogeneities arise as the result of the normal thermal fluctuations in the disordered phase in the vicinity of the transition point. They are just the ones which determine the structure of the critical configuration of the order parameter field which initiates the phase transition and which as a rule is not spherical. We discuss the interesting aspects of the problem connected with the change in the effective dimensionality of the inhomogeneous order parameter distribution in the

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various stages of the evolution of the system. We demonstrate the important role of stationary states which manifest themselves as metastable attractors in the process of proceeding to an absolutely stable state.

## 1. STRUCTURE OF THE ISOTROPIC CRITICAL NUCLEUS. STATICS

We first discuss the problem, traditional in the kinetics of phase transformations, of the shape and the structure of the critical nucleus of the new phase.[18] In the static case the critical distribution of the order parameter density $\varphi(\mathbf{r})$ must correspond to an extremum (saddle-point) of the nonequilibrium Ginzburg–Landau functional in the form:[1–3]

$$\mathscr{F}[\varphi]=\int d^d r[\tfrac{1}{2}(\nabla\varphi)^2+F(\varphi)], \tag{2}$$

where $d$ is the dimensionality of the space. In other words, the required distribution $\varphi(\mathbf{r})$ must be a static unstable solution of Eq. (1).

The local energy density $F(\varphi)$ is in the general case an arbitrary function of the order parameter and is invariant under the paraphase symmetry group of the system studied. For simplicity we restrict ourselves in what follows to a scalar order parameter and to two types of expansion of the free energy, respectively:

$$\text{a) } F(\varphi)=\tfrac{1}{2}\tau\varphi^2-\tfrac{2}{3}a\varphi^3+\tfrac{1}{4}b\varphi^4, \tag{3}$$

$$\text{b) } F(\varphi)=\tfrac{1}{2}\tau\varphi^2-\tfrac{1}{2}a\varphi^4+\tfrac{1}{6}b\varphi^6. \tag{4}$$

These simple expansions have in fact a very wide applicability since for various physical systems one can construct near the critical point a scalar combination of the components of the order parameter (which are properly ordered in the transition) and reduce the problem to a study of the standard catastrophes described by the expansions (3) and (4).[19] Moreover, the expansions (3) and (4) describe rather well the localized excitations in the vicinity of phase transitions in actual physical systems, for instance, in binary mixtures, uniaxial magnetics, martensites, and so on (see, for instance, Ref. 20 and also Ref. 21 and the literature cited in those references).

The function $F(\varphi)$ must have a metastable minimum at $\varphi=0$ and be energetically favorable for $\varphi=\varphi_0\neq 0$, if all constants in Eqs. (3) and (4) are positive and $a^2>\tau b$, where

$$\text{a) } \varphi_0=\frac{1}{2b}\,[a+(a^2-\tau b)^{1/2}],$$

$$\text{b) } \varphi_0^2=\frac{1}{b}\,[a+(a^2-\tau b)^{1/2}],$$

and it is thus suitable to describe the behavior of a system between binodal and spinodal supercooling.

One usually assumes that in isotropic space the critical nucleus is spherical. In the last section we shall discuss in detail the legitimacy of this assumption. But for the present we follow the conventional approach and give a number of results corresponding to an isotropic static distribution of the order parameter. The structure of the critical nucleus with its center at a point $\mathbf{r}_0$ is in that case determined by the solution of the equation

$$\varphi_{rr}+\frac{d-1}{r}\,\varphi_r-F_\varphi=0 \tag{5}$$

with the boundary conditions

$$\varphi_r=0 \text{ as } \mathbf{r}\to\mathbf{r}_0 \text{ and } \varphi\to 0 \text{ as } r\to\infty. \tag{6}$$

We note that the static problem (5), (6) with boundary conditions at infinity can in fact describe the shape of the critical nucleus only in the initial stages of the evolution of the system when the nucleus of the energetically favorable phase is rather small.

Taking into account the experience[3,9–14] gained by studying Eq. (5) in the one-dimensional case ($d=1$), we can expect that for arbitrary $d$ the required localized solution must also be a special integral ("separatrix") separating two different kinds of solution. The presence of the term $[(d-1)/r]\varphi_r$ in Eq. (5) makes it impossible to use the analytically convenient phase portrait method since the phase trajectories may intersect in it. However, bearing in mind condition (6) this can be done formally to find a solution which is the separatrix for a special class of equilibrium distributions $\varphi(r)$ which satisfy the restriction (6).

The results of a numerical solution of Eq. (5) for $d=3$ and a detailed discussion of them are given in a previous paper by the present authors[22] where we also give the actual values of the parameters on the right-hand side of (5) which are used in the calculations and which determine the position of the fixed points in the phase portrait.

We note that qualitatively the form of $\tilde{\varphi}(r)$ for $d=3$ resembles the known analytical solutions for $d=1$. However, there is also an essential difference. For $d=1$ Eq. (5) reduces to the form $(\varphi r)^2=2F(\varphi)+\text{const}$ and the quantity $\tilde{\varphi}(0)$ is exactly the same as the value of $\varphi$ for which $F(\varphi)$ vanishes, which has physically no special meaning. For $d=3$ the value $\tilde{\varphi}(0)$ lies considerably higher than for $d=1$. The numerical solution gives $\tilde{\varphi}(0)$ practically the same as the equilibrium $\varphi_0$ for uniform ordering which one would intuitively expect for an actual nucleus. One should, however, bear in mind that here we are dealing only with a natural physical interpretation of a spherically symmetric localized excitation as the critical nucleus of a new phase with an equilibrium value $\varphi=\varphi_0$.[1)] We shall show in what follows that in kinetics the noted fact that $\tilde{\varphi}(0)$ is close to $\varphi_0$ is not decisive, and a spherical distribution of the order parameter is practically never realized as a critical nucleus.

We have shown earlier that the $\tilde{\varphi}(r)$ profiles simulate rather closely "plane walls" which are described by functions of the form $\varphi_0\{1-\tanh[(r-R)/a]\}/2$.[22] The quantities $R$ and $a$ which determine the effective radius of the nucleus and the effective width of its wall, being chosen from the best approximation to the solution $\tilde{\varphi}(r)$, at the same time also give a good approximation to the saddle point of the function $\mathscr{F}(a,R)=\mathscr{F}[\varphi(r;a,R)]$, which corresponds to the functional (2) in the above indicated class of functions $\varphi(r;a,R)$.

We note again the results which in our view are the most important ones that follow from the simple analysis carried out above. The critical nucleus of the new phase is described by a static solution of Eq. (1). This solution is a saddle point for the Ginzburg–Landau functional and is the only one in the case of an isotropic distribution of the parameter $\varphi$. Its structure is such that the thickness of the wall where the transition of the inhomogeneous $\varphi(r)$ field from one equilibrium value to another one takes place is comparable in magnitude to the effective radius of the nucleus ($a \approx R$).

## 2. PERTURBATION OF THE ISOTROPIC CRITICAL NUCLEUS AND GROWTH OF AN ISOLATED DOMAIN OF THE NEW PHASE

The stability of the static solution is determined by the second variation of the functional $\mathscr{F}[\varphi]$ for small deviations $\varphi$ from $\tilde{\varphi}(\mathbf{r})$ $(\tilde{\varphi}(\mathbf{r}) \to \tilde{\varphi}(\mathbf{r}) + \varphi)$:

$$\delta^2 \mathscr{F}[\varphi] = \frac{1}{2} \int d^d r [(\nabla\varphi)^2 + F_{\varphi\varphi}(\tilde{\varphi}(\mathbf{r}))\varphi^2], \qquad (7)$$

and the evolution of small perturbations $\varphi$ on the background of a stationary solution $\tilde{\varphi}(r)$ is described by the linearized Eq. (1):

$$\frac{1}{\gamma}\varphi_t = -\Delta\varphi + F_{\varphi\varphi}[\tilde{\varphi}(\mathbf{r})]\varphi. \qquad (8)$$

Solving Eq. (8) by standard methods we arrive at an eigenvalue problem,

$$\Delta\varphi - \{\lambda + F_{\varphi\varphi}[\tilde{\varphi}(\mathbf{r})]\}\varphi = 0, \qquad (9)$$

with an effective potential $U(r) = F_{\varphi\varphi}[\tilde{\varphi}(r)]$ determined both by the structure of the function $F(\varphi)$ and by the spatial dispersion of the actual static solution $\tilde{\varphi}(r)$. We study this problem for the dimensionalities $d=1, 2, 3$ which are physically of most interest.

We note first of all that the perturbations $\varphi$ in principle do not need to conserve the isotropy of the static solution even in the case where isotropy occurred for the initial $\tilde{\varphi}(r)$ function.[2)] It will, however, be shown in the last section that a spherical distribution of a nonequilibrium $\varphi$ field is an attractor in isotropic space and, therefore, stable against anisotropic deviations. Taking this into account, and also for clarity, we restrict ourselves in the present section to the isotropic variant of Eq. (9) which is sometimes called the Jacobi equation:[23]

$$\varphi_{rr} + \frac{d-1}{r}\varphi_r - \{\lambda + F_{\varphi\varphi}[\tilde{\varphi}(r)]\}\varphi = 0. \qquad (10)$$

The standard substitution

$$\varphi = \tilde{\varphi} \exp \int \alpha(r) dr, \quad \text{with } \alpha(r) = -(d-1)/(2r),$$

reduces[10] to a quasi-one-dimensional Schrödinger equation with an effective potential

$$U_{\text{eff}} = U - \frac{(d-1)(3-d)}{4r^2}. \qquad (11)$$

For $d=3$ and $d=1$ this potential is as before the same as the corresponding function $F_{\varphi\varphi}[\tilde{\varphi}(r)]$. Recall that if $d=1$ the maximum of the function $\tilde{\varphi}(r)$ is for $r=0$ the same as the point $\varphi_1$ where $F(\varphi_1)=0$. For the expansions (3) and (4) we have, respectively:

$$\text{a)}\ \varphi_1 = \frac{4a - (16a^2 - 18\tau b)^{1/2}}{3b},$$

$$\text{b)}\ \varphi_1^2 = \frac{3a - (9a^2 - 12\tau b)^{1/2}}{2b}.$$

One can easily show by using these expressions that $F_{\varphi\varphi}(\varphi_1) < 0$ in the metastability region. On the other hand, it is clear that as $\varphi \to 0$ the quantity $F_{\varphi\varphi}(\varphi)$ becomes positive. On the whole $U_{\text{eff}}(r)$ has the form shown in Fig. 1a. For $d=3$ the solution $\tilde{\varphi}(r)$ starts from the vicinity of the minimum at $\varphi = \varphi_0$ where $F_{\varphi\varphi}(\varphi) > 0$. Later on as the quantity $\varphi$ decreases the sign of $F_{\varphi\varphi}(\varphi)$ is reversed, the effective potential goes through a minimum and again becomes positive as $\varphi$ approaches the stable value $\varphi=0$. We show in Fig. 1b a distribution of $U_{\text{eff}}(r)$ for the case $d=3$ which is typical of such a structure of the nucleus.

The qualitative difference of the potentials $U_{\text{eff}}$ for $d=1$ and $d=3$ is clear. A calculation of the spectrum gives for each discrete eigenvalue $(-\lambda) < 0$ (i.e., $\lambda > 0$ which corresponds to the required instability) lying inside the potential well. However, the nature of the localization of the corresponding eigenfunctions is different. For $d=3$ the eigenfunction with $\lambda > 0$ is zero at the center of the nucleus and mainly localized in a spherical zone of finite width. This dictates the character of the evolution of the distribution shown in Fig. 2, which agrees splendidly with the character expected in the simple phenomenological approach. For $d=1$ the analogous eigenfunction is mainly localized in the neighborhood of zero and the evolution of the $\varphi$ distribution has the form shown in Fig. 3. Its distinguishing feature is that initially the $\varphi$ density increases in amplitude near $r=0$, followed already by expansion of the domain of the new phase as a whole.[3)] It will become clear in what follows that this peculiarity in the growth of low-dimensional nuclei plays a very important role in the kinetics of phase transformations.

For $1 < d < 3$ the effective potential contains a correction $\propto 1/r^2$ and already even qualitatively it does not repeat the behavior of the function $F_{\varphi\varphi}[\tilde{\varphi}(r)]$. We show in Fig. 1c the function $U_{\text{eff}}(r)$, found numerically, for the $d=2$ case. In the same figure we indicate the position of the $-\lambda < 0$ level in this potential. It is clear that the dimensionality $d=2$ corresponds to an "intermediate localization" of the corresponding eigenfunction. As a result there occurs a simultaneous increase in the amplitude of $\varphi$ and an expansion of the nucleus.

In later stages of the evolution the radius of the nucleus becomes rather large and, as we noted already, its boundary can be considered as being practically a planar formation. Using this, it is useful for what follows to reproduce a few results obtained analytically for $d=1$. Bearing in mind that qualitatively they are similar for both models considered, we give in what follows for the sake of

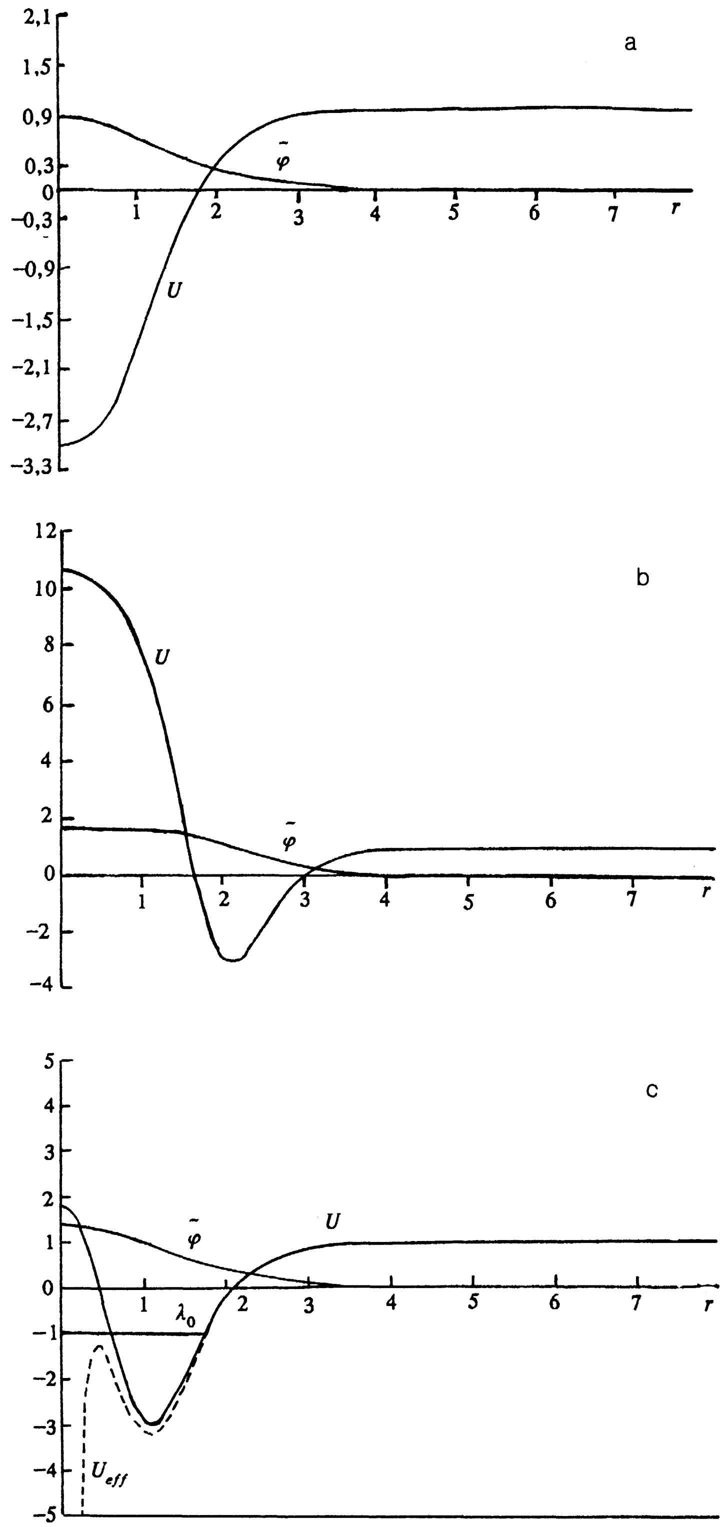



FIG. 1. The $U_{\text{eff}}$ potential and the $\tilde{\varphi}(r)$ profile corresponding to it: a) for $d=1$; b) for $d=3$; c) for $d=2$.

brevity the results only of the analysis of the $\varphi^6$ model. For $d=1$ and a suitable choice of the time normalization we can write Eq. (1) in the form:

$$\varphi_t=-\varphi_{rr}+\tau\varphi[1-(\varphi/\varphi_+)^2][1-(\varphi/\varphi_-)^2], \qquad (12)$$

where $\varphi_+$ and $\varphi_-$ determine the positions of the zeroes of the derivative $F_\varphi$ together with $\varphi=0$:

$$\varphi_\pm^2=[a\pm(a^2-\tau b)^{1/2}]/b. \qquad (13)$$

In the binodal point $\tau=\tau_0$ the zeroes $\varphi_{1,2}^2=[a\pm(a^2-4\tau b/3)^{1/2}]/b$ of the functon $F(\varphi)$, degenerate into a single point $\varphi_{1,2}^2=3a/2b$ which in this case is the same as $\varphi_+^2$ so that $F(\varphi)$ takes the form

$$F(\varphi)=\tfrac{1}{2}\tau\varphi^2[1-(\varphi/\varphi_+)^2]^2. \qquad (14)$$

In this case Eq. (12) has a static solution (a wall separat-

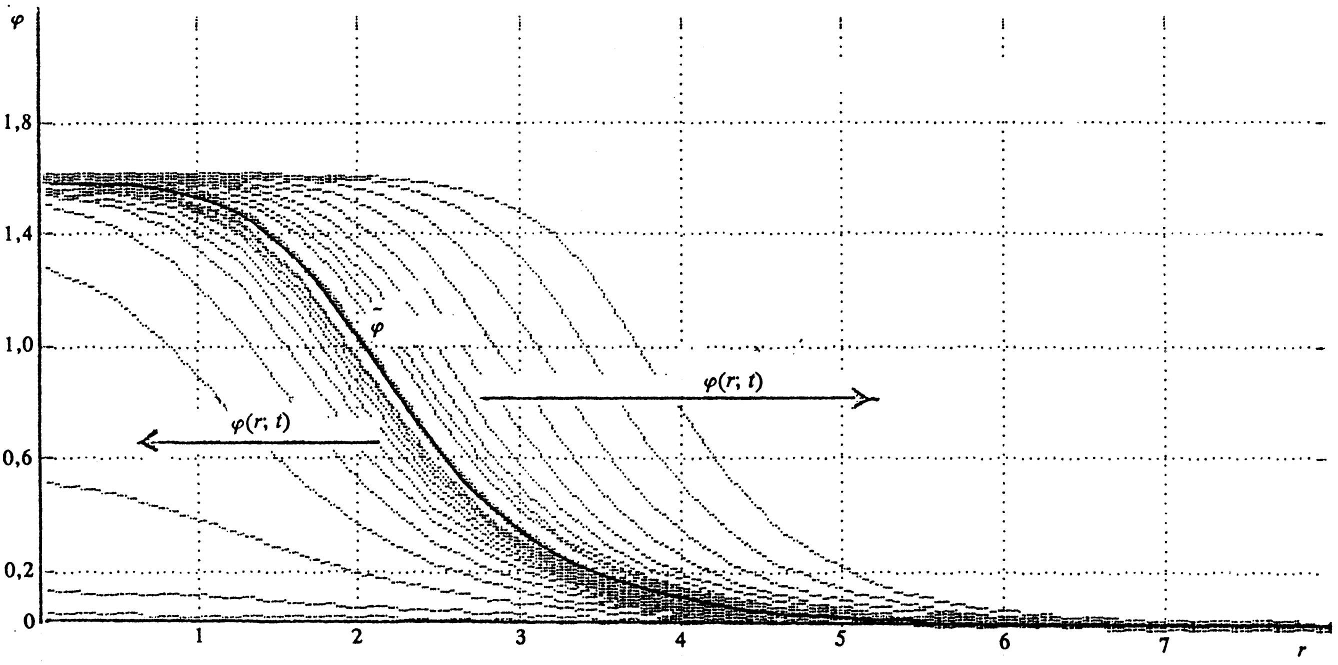



FIG. 2. Evolution of small perturbations of the $\varphi$ distribution for $d=3$. The heavy line identifies the $\tilde{\varphi}(r)$ profile.

ing the energetically equivalent phases with $\varphi=0$ and $|\varphi|=\varphi_+$) so that $\varphi_r^2=2F$ and correspondingly

$$\left(\frac{\varphi_-}{\varphi_+}\right)^2=\frac{1}{1+\exp(kr)}, \quad \text{with } k=2\tau_0^{1/2}. \tag{15}$$

The derivative $\varphi_r\propto\varphi[1-(\varphi/\varphi_+)^2]$ vanishes as $r\to\pm\infty$ inside each of the phases. It is essential that the factor $\varphi[1-(\varphi/\varphi_+)^2]$ also remains in Eq. (14) which describes $\varphi(r,t)$ in the region between the binodal and the spinodal

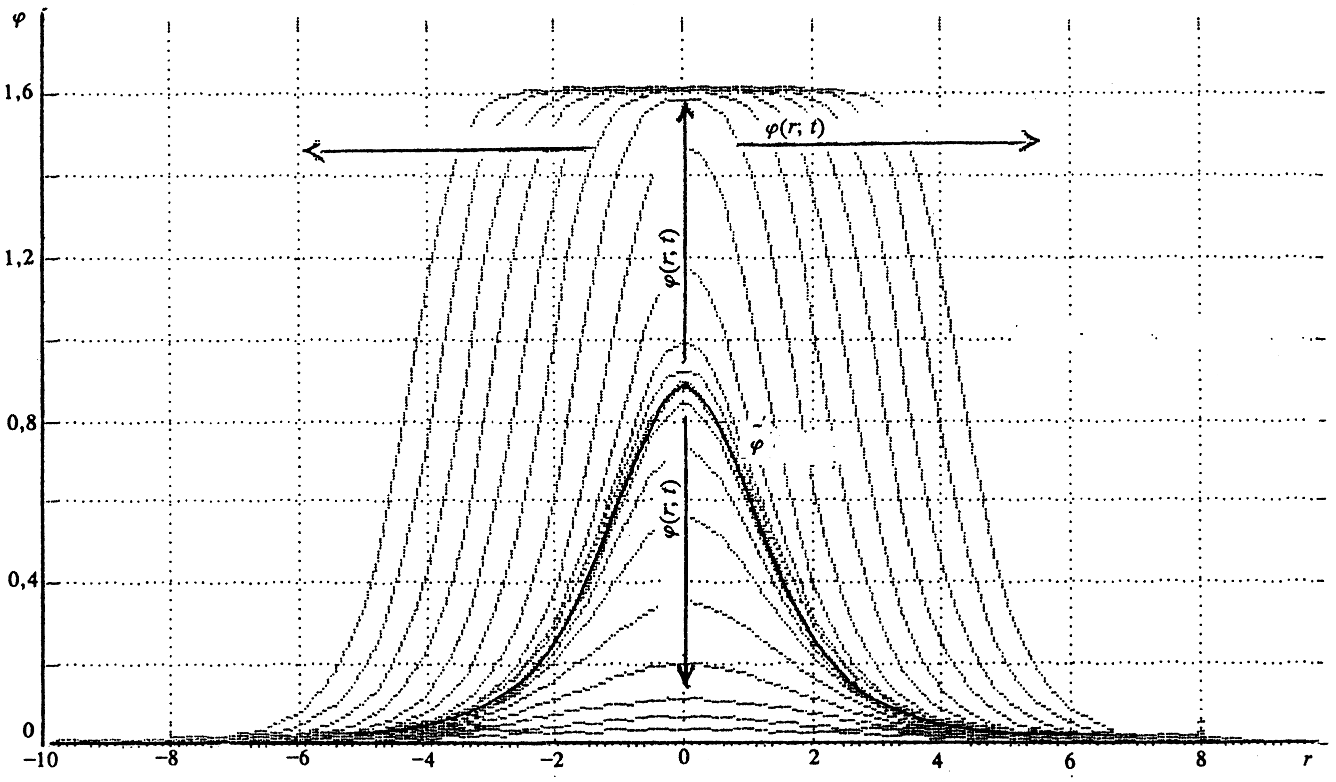



FIG. 3. The same as in Fig. 2 for the $d=1$ case.

supercooling. This formal fact enables us to look for a stationary solution of Eq. (12) in the form (15) which automatically satisfies the condition $\varphi_r \propto \varphi[1-(\varphi/\varphi_+)^2]$, with the substitution $r \to r - vt$, where

$$v=\frac{2}{k}(\tau-k^2/4), \quad 3k^2/4=\tau(\varphi_+/\varphi_-)^2. \tag{16}$$

As $\tau \to \tau_0$ we have $(\varphi_+/\varphi_-) \to 3$, $k^2 \to 4\tau$ and, hence, $v \to 0$. For $d \neq 1$ Eq. (1) has the form

$$\varphi_t=-\varphi_{rr}-\frac{d-1}{r}\varphi_r+F_\varphi. \tag{17}$$

We now use the fact that as $R \to \infty$ the front of the domain becomes practically plane. Its velocity should then tend to a constant value equal to the one-dimensional velocity (16) of the plane front. The relative increase of $R$ per unit time decreases and for an estimate we can use the approximation $(d-1)/r \to (d-1)/R$. We find $v_{\text{eff}}=v-(d-1)/R$, which agrees with the assumption we used that $v$ tends to a constant as $R \to \infty$. The numerical solution of Eq. (17) for $d=3$ confirms that the velocity of the boundary is "attracted" to the velocity of the plane front following a $1/R$ law. This makes the analytical results for one-dimensional excitations applicable to the subsequent stage of the evolution. We demonstrate in what follows that such excitations are attractors for the evolution of arbitrary fluctuations.

## 3. EVOLUTION OF ARBITRARY LARGE-SCALE FLUCTUATIONS AND EMERGENCE OF A NUCLEUS

So far we have bypassed the problem of the onset of (spherical) critical nuclei. Yet it is rather obvious that even in an isotropic medium an arbitrary field of order-parameter fluctuations is isotropic only in the mean. There arise a number of obvious problems in this connection. First of all, how can an isolated spherical excitation with an amplitude $\varphi$ close to the equilibrium value $\varphi_0$, which is separated from other such excitations by a "vacuum of fluctuations" in which $\varphi \approx 0$, arise through fluctuations? Could a critical nucleus have a different shape (and what shape)? What happens if expanding (spherical) domains of the new phase (amongst them domains of opposite sign) collide? We attempt to give in the present section an answer to these problems.

First of all we forgo the assumption of a necessarily spherical shape of the critical nucleus. We turn again to Figs. 2 and 3 describing the perturbation of a nucleus for $d=3$ and $d=1$ and consider the standard scenario of a phase transition. Let there be some "noise" of the fluctuations in the ordered phase $\varphi(\mathbf{r})$ in a three-dimensional system and let the temperature of this system be lowered from the paraphase in the metastability region.

At some temperature the amplitude of the separate excitations $\varphi(\mathbf{r})$ becomes sufficient that they turn out to be critical nuclei. We note that in the most general case the maximum $\varphi(\mathbf{r})$ density decreases differently in different directions of $\mathbf{r}$. Moreover, one can always find amongst the maxima such for which the velocity along one (or two) directions is considerably smaller than along other directions. Such density splashes can be interpreted as quasi-low-dimensional structures evolving in accordance with the one- or two-dimensional Eq. (1). This is most obvious when the expected radius of the critical nucleus is small and the shape fluctuations appreciable. To study the problem in this case we must turn to a more general form of the equation for $\varphi(\mathbf{r},t)$:

$$\frac{1}{\gamma}\varphi_t=-\Delta\varphi+F(\varphi), \tag{18}$$

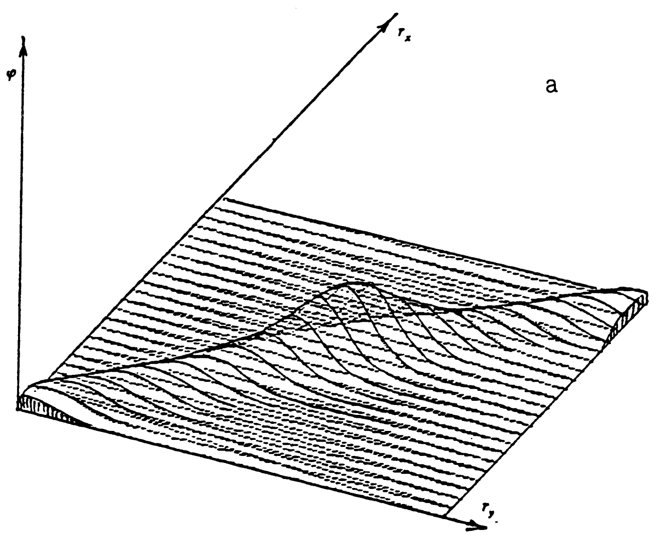
where $F(\varphi)$ is defined by Eq. (4).

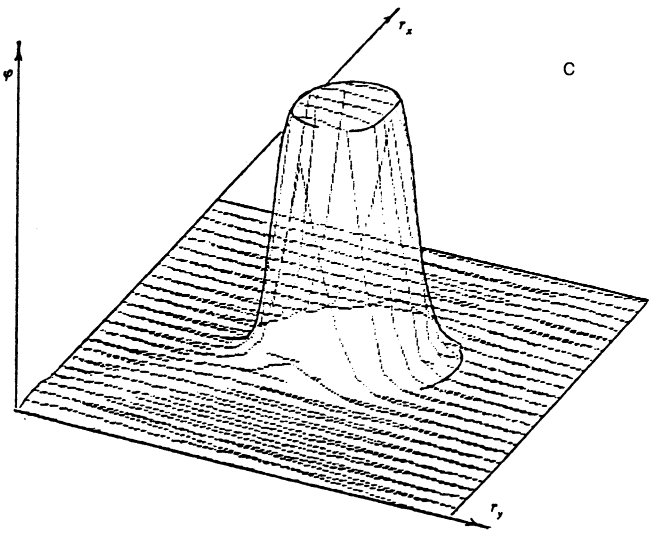
However, for one and the same amplitude $\varphi$ (which may be much smaller than $\varphi_0$!) and one and the same temperature (the latter determines the parameters of the potential $F$) the scenarios for the evolution of three-dimensional and low-dimensional nuclei (see Figs. 2 and 3) may be radically different. In fact, whereas for $\varphi \ll \varphi_0$ a spherical nucleus practically always collapses, a low-dimensional nucleus may, in contrast, start to grow. It is important here that this growth is primarily due to the growth of the amplitude $\varphi(\mathbf{r})$ on the background is the average one. As a result, when the temperature is lowered from the paraphase the spherical structures are in reality by far not the first to become critical.

We now turn to Eq. (17) from which it is clear that the velocity of the front of the excitation is the higher the larger its radius of curvature. One can clearly also use the concept of a local curvature for an anisotropic excitation described by Eq. (18). It is then rather obvious that convex parts of the surface will have a somewhat lower velocity than other parts. As a result the expanding surface of a low-dimensional nucleus must so to speak "overtake" its parts which are most convex in front. The nucleus must then gradually become isotropic. This process occurs indeed. We show in Fig. 4 a number of stages through which a supercritical low-dimensional nucleus passes. After being made isotropic, the nucleus becomes three-dimensional. However, the density $\varphi$ inside it is at that moment already close to $\varphi_0$ and it is also already supercritical.

So far we have used the idea of an isolated critical nucleus. Apparently in a uniform medium it would be more realistic to consider the problem of an arbitrary critical fluctuation density $\varphi(\mathbf{r})$. At present it is impossible to solve this problem analytically. A numerical experiment, however, gives the following results.

We show in Fig. 5 the relaxation of a typical $\varphi(\mathbf{r})$ distribution. One sees easily that the short-wavelength fluctuations which fill the space between the growing domains of the new phase are rapidly damped, whereas the remaining domains become isotropic. In the growth process the domains described here become universal. Figure 6 illustrates the process of the attraction of the $\varphi(\mathbf{r})$ distribution to the attractor structure in one-dimensional space. A similar process also occurs for $d \neq 1$. The attracting surface is then obtained by rotating the curve shown in Fig. 6 around the horizontal axis. One can check immediately that this curve is the same as function (15) (or,



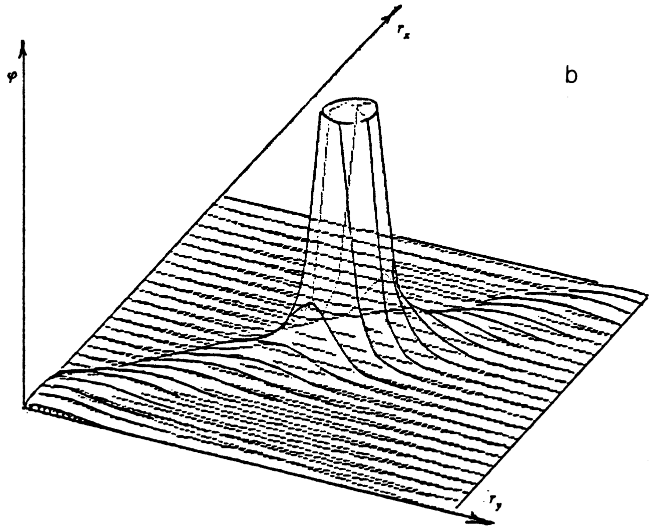




FIG. 4. Stages passed through by a supercritical low-dimensional nucleus during the evolution process: a) bare relief of the $\varphi(\mathbf{r})$ density; b) growth of the amplitude of the maximum of $\varphi(\mathbf{r})$ up to its equilibrium value; c) isotropization of a domain of the new phase.

$\varphi_r \propto \varphi[1-(\varphi/\varphi_+)^2])$ for $r \to r - vt$, which describes the motion of the wall between the two phases with $\varphi=0$ and $\varphi=\varphi_0$, respectively.

The attractor nature of this solution has a deep physical meaning. Indeed, in correspondence with the general principles of nonequilibrium thermodynamics[25–27] dissipation reaches a minimum in the stationary solution. In the present case the flow of energy from the system has the form:

$$\frac{d\mathscr{F}}{dt} = -\int_{-\infty}^{\infty} \frac{\delta\mathscr{F}}{\delta\varphi} \bar{\varphi} dx = -\int_{-\infty}^{\infty} \bar{\varphi}^2 dx = -Q, \quad (19)$$

and its change with time is correspondingly:

$$\frac{dQ}{dt} = -\frac{d}{dt} \int_{-\infty}^{\infty} \dot{\varphi}^2 dx = \int_{-\infty}^{\infty} \frac{\partial \dot{\varphi}^2}{\partial t} dx. \quad (20)$$

For a localized stationary solution of the form $\varphi=\varphi(x-vt)$ we have $\partial\varphi/\partial t = -v\partial\varphi/\partial x$ and Eq. (20) gives

$$\frac{dQ}{dt} = v \int_{-\infty}^{\infty} \frac{\partial \dot{\varphi}^2}{\partial x} dx = v\dot{\varphi}^2 \Big|_{-\infty}^{\infty} = 0. \quad (21)$$

In other words the minimum dissipation is reached on the stationary trajectory with the asymptotes $\varphi=0$ and $\varphi=\varphi_0$. We note in passing that in the general case one can in turn use the lowering of the dissipation on the stationary trajectories for a numerical search for all such solutions. Indeed, in the phase portrait the stationary solutions are fixed and thanks to the lowering of the dissipation they are attractors. Using a rather large random initial bulk $\varphi(\mathbf{r})$ one can observe all such attractors which are admitted by the system studied.

Figure 5 illustrates also collision processes and adhesion of separate (spherical) expanding nuclei—domains of the new phase. Additionally we show in Fig. 7 the emergence of a domain wall when domains with a different sign collide.

This is just as typical for the $\varphi^6$ model as a collision of the same kind of domains of the new phase since the formation of nuclei of either sign is equally probable. The idealized situation shown in Fig. 7 illustrates this process

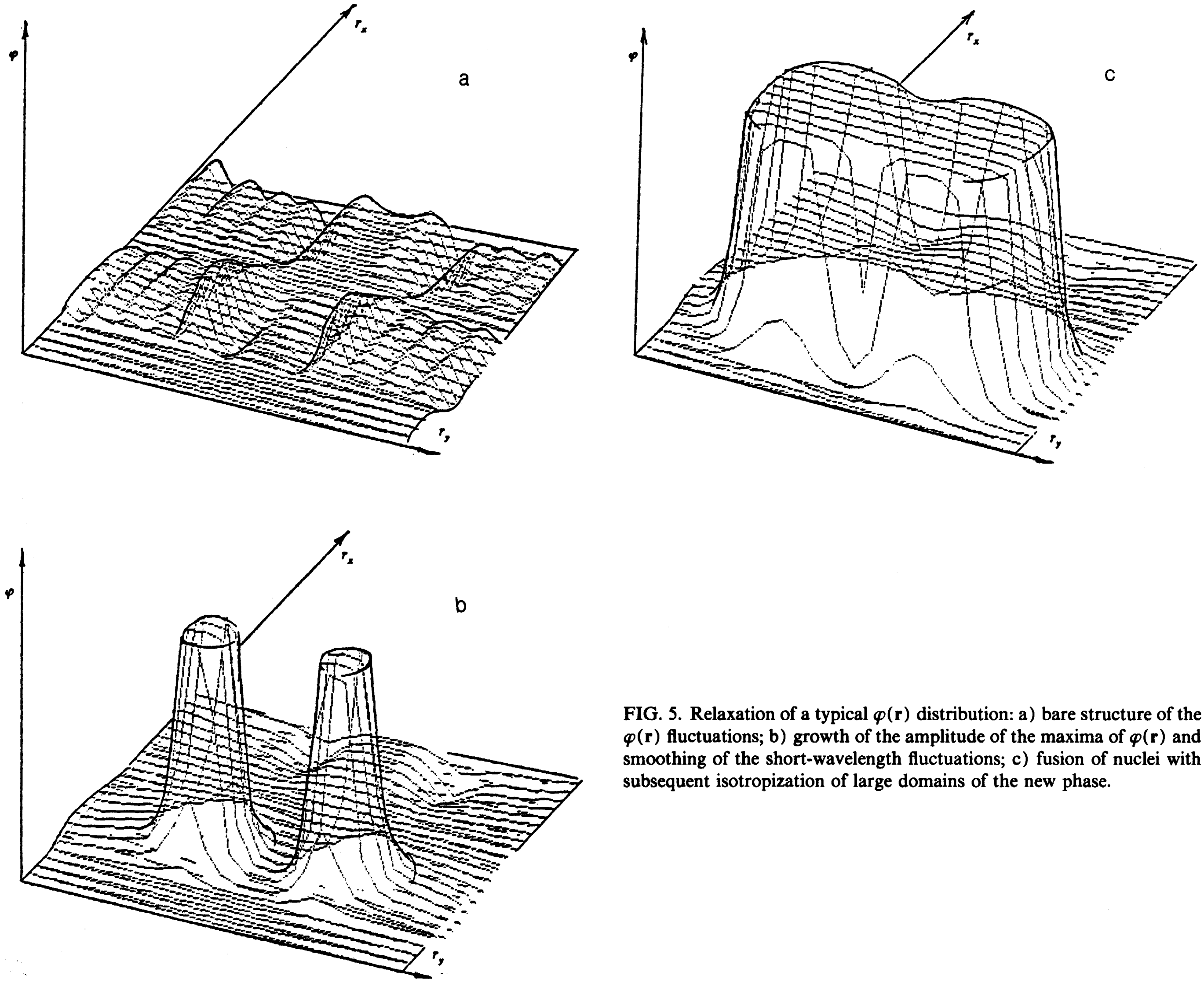



FIG. 5. Relaxation of a typical $\varphi(\mathbf{r})$ distribution: a) bare structure of the $\varphi(\mathbf{r})$ fluctuations; b) growth of the amplitude of the maxima of $\varphi(\mathbf{r})$ and smoothing of the short-wavelength fluctuations; c) fusion of nuclei with subsequent isotropization of large domains of the new phase.

(consistently observed during the numerical simulation) only "in its pure form."

Formally the system leaves in this process the metastable stationary (but not static!) $d$-dimensional attractor and is attracted to a topologically nonremovable static attractor of lower dimensionality. Physically this means that the dissipation reduces the phase volume occupied by the system, including a reduction it through a constant lowering of its dimensionality (down to $d=0$ for uniform ordering or $d=1$ for static domain walls).

In this connection it is interesting to note that the frequently used simple analytical expressions which are obtained here and in the above-mentioned papers not only turn out to be applicable to quasi-one-dimensional systems, which are in reality extremely rare, but also describe the regular stages of the evolution of normal three-dimensional systems.

The described relaxation process is slowed down by at least two factors. First of all there is the nonremovable noise of the fluctuations in the $\varphi(\mathbf{r},t)$ field at non-zero temperatures. A corresponding term was added in Eq. (1) and led, on the one hand, to automatic appearance of a nucleus without any specification of the $\varphi(\mathbf{r},0)$ distribution and, on the other hand, to a "blurring" of the idealized picture of the relaxation given above. Secondly there are the dynamic oscillations of the density $\varphi$. This last factor was also taken into account by a modification of Eq. (1)

$$\frac{1}{c^2}\varphi_{tt}+\frac{1}{\gamma}\varphi_t=-\frac{\delta\mathcal{F}}{\delta\varphi}, \tag{22}$$

where $c$ is a characteristic velocity of the sound excitations in the system. The presence of the $\varphi_{tt}$ term affects especially the behavior of the high-frequency (short-wavelength) modes. Whereas before they were damped faster than the other modes, now such oscillations are preserved for rather a long time, just as the noise, somewhat blurring the idealized relaxation picture. The shape of the stationary attractor is also slightly deformed.

Notwithstanding the fact that the pictures given in the present paper correspond to the $\varphi^6$ model, the correspond-

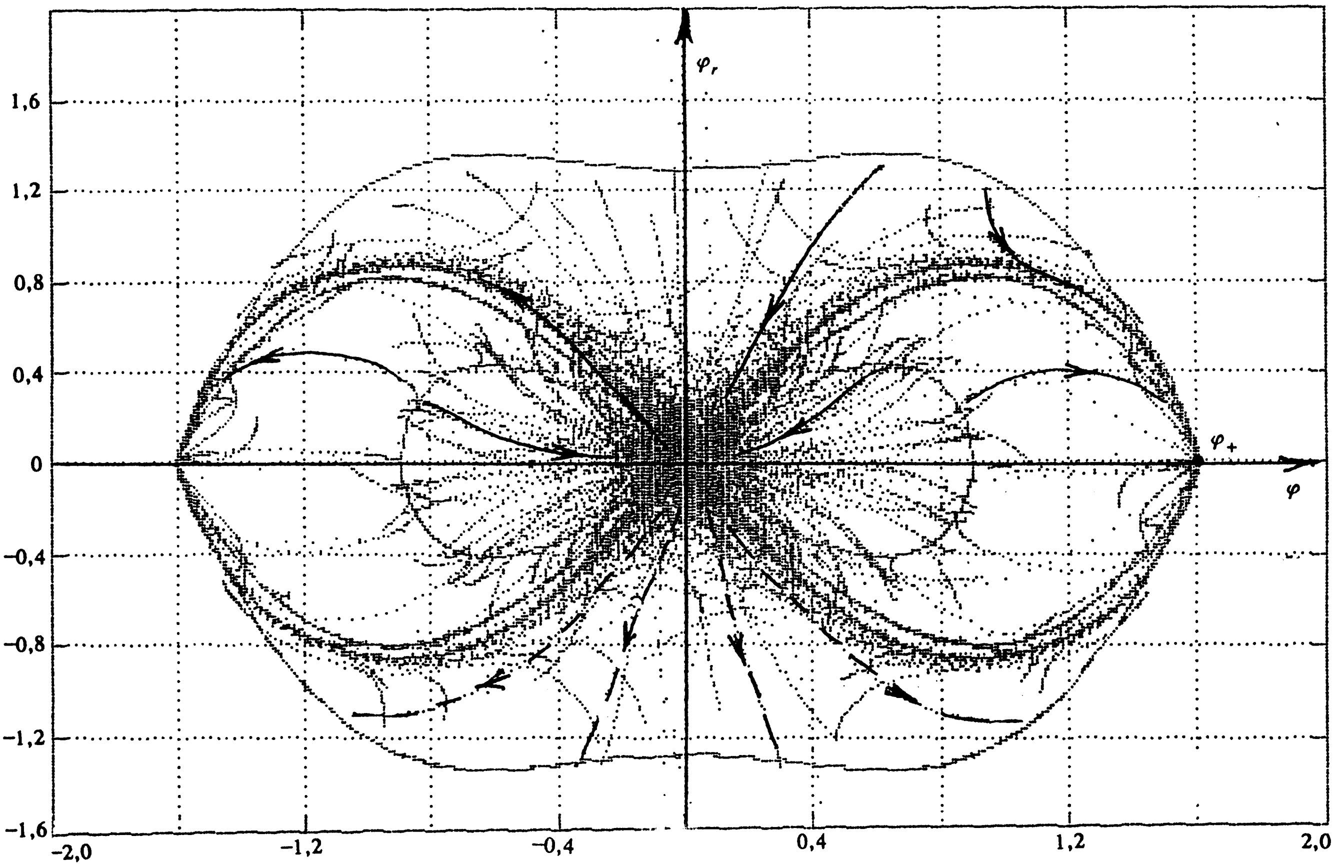



FIG. 6. Attraction of the $\varphi(\mathbf{r})$ distribution to an attractor. The solid arrows show typical directions in which the phase points of the initial distribution develop; the dashes show the motion of the phase points in the concluding stage of the evolution when domain boundaries are formed.

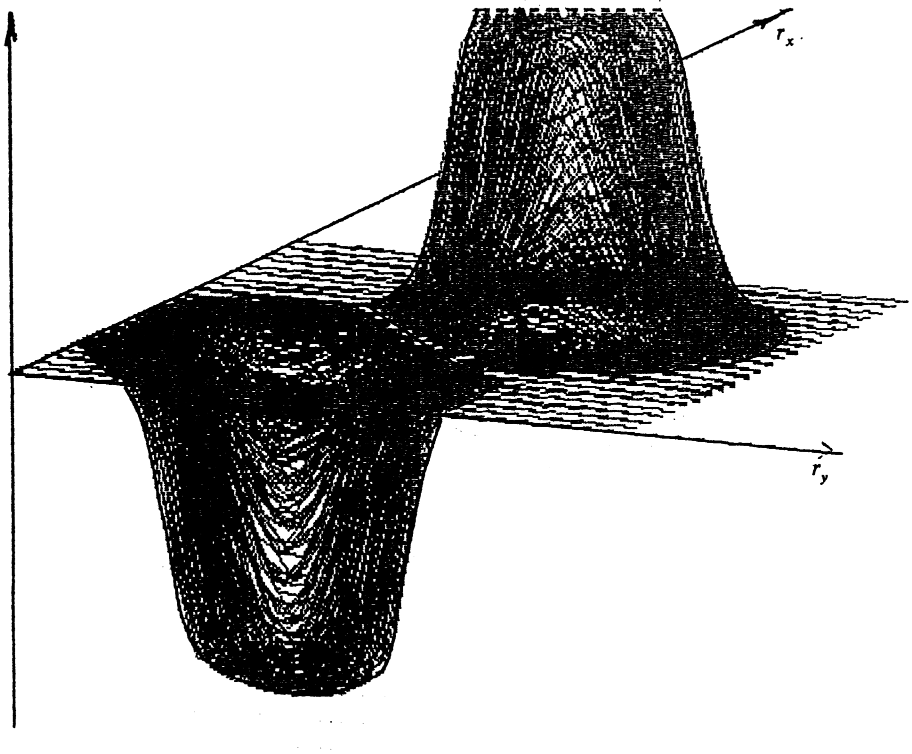



FIG. 7. Formation of a domain wall when nuclei with different signs collide (the heavy straight line identifies its intersection with the $\varphi=0$ plane).

ing calculations were also carried out for the model described by Eq. (3) and all conclusions reached above apply to it to the same degree.

Finally, a situation is possible when the change in the magnitude of $\varphi$ in a first-order phase transition is so large that it is no longer possible to restrict oneself to one of the standard catastrophes (3) or (4). The simplest generalization is the case when the function $F(\varphi)$ is an arbitrary polynomial which has a minimum at two or more points. The derivative of the function $F(\varphi)$ can in that case be written in the form

$$F_\varphi(\varphi)=\varphi(1-\varphi/\varphi_+)\Phi(\varphi), \tag{23}$$

where $\varphi_+$ determines the position of the $F(\varphi)$ minimum closest to zero; $\Phi(\varphi_+)$ and $\Phi(0)$ are constants. Using (23), Eq. (14) takes the form

$$\varphi_t=-\varphi_{rr}+\tau\varphi(1-\varphi/\varphi_+)\Phi(\varphi), \tag{24}$$

or for a stationary solution

$$\varphi_r(d\varphi_r/d\varphi-v)=\varphi(1-\varphi/\varphi_+)\Phi(\varphi). \tag{25}$$

Equation (25) has a solution such that $\varphi_r(0)=\varphi_r(\varphi_+)=0$:

$$\varphi_r=\varphi(1-\varphi/\varphi_+)A(\varphi), \tag{26}$$

where the function $A(\varphi)$ is the solution of the equation

$$(1-2\varphi/\varphi_+)A^2(\varphi)+\tfrac{1}{2}\varphi(1-\varphi/\varphi_+)dA^2/d\varphi-vA=\Phi(\varphi). \tag{27}$$

It is clear that as a solution of Eq. (27) the function $A(\varphi)$ must satisfy the conditions

$$A(0)[A(0)-v]=\Phi(0),$$

$$A(\varphi_+)[A(\varphi_+)+v]+\Phi(\varphi_+)=0.$$

One of these conditions fixes the velocity $v$ of the boundary and the second one guarantees the uniqueness of the required solution. For the potentials considered by us above the function $A(\varphi)$ is equal to const $\cdot\,(1+\varphi/\varphi_+)$ for the $\varphi^6$ model and a constant for the $\varphi^4$ model.

The numerical experiments carried out for various, including nonpolynomial, functions $F(\varphi)$ corroborate the fact that there exists a stationary attractor $\varphi_r=\varphi_r(\varphi)$ which corresponds in the phase plane to a curve connecting the points (0,0) and $(\varphi_+,0)$.

One of the authors (T.K.S.) thanks S. Leble for fruitful discussions.

[1)] Strictly speaking the exact equality $\tilde{\varphi}(0)=\varphi_0$ is inadmissible since $\varphi_0$ is a fixed point of Eq. (5). However, the numerical difference is $|\tilde{\varphi}(0)-\varphi_0|\leqslant 10^{-3}\varphi_0$ for $d=3$ and is hardly changed for $d>3$. In any case this difference is very small and unimportant for the physical picture.

[2)] Traditionally the stability of the critical nucleus is analyzed only in the class of isotropic solutions, i.e., a stability necessarily with respect to the expansion or collapse of its wall as a whole. Strictly speaking, however, the deviation of the nucleus from sphericity requires an independent analysis, especially in the context of the problem of the possibility of a spontaneous appearance of a static isotropic distribution of the $\varphi$ field.

[3)] We emphasize again that Fig. 3 demonstrates the evolution of a single nonstationary solution. The outwardly similar Fig. 3 of Ref. 24 shows separate stationary solutions of a purely dynamic equation (so that each curve corresponds to a fixed value of the velocity $v$ in the variable $x-vt$) which is known to describe satisfactorily the martensitic transformations studied in Ref. 24.

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