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Fluctuating field near spinodal

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Abstract

Phase ordering kinetics with a non-conserved scalar order parameter is studied near a lability boundary of a first-order phase transition. A large-scale structure of this field is found to appear. Under some conditions it is analogous to a fractal structure in the critical region of a second-order transition. The chains and localized groups of new phase nuclei are generated by the maxima of the order parameter density. The results are compared with fluctuation inducing of continuous phase transitions predicted in the framework of the renormalization group (RG) approach. © 1998 Published by Elsevier Science B.V.

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1. Fluctuations and nucleation

One of the principal problems of phase transition theory is the structure of critical nuclei. Traditionally it was defined as spherical with some diameter determined by the balance between the energy of the ordered phase and the surface energy [1–3]. This description is quite appropriate for analytical thermodynamics based on a transformation of the initial dynamical problem to the study of collective continuous fields. Modern computer techniques make it possible, in principle, to study the many-body dynamical problem directly [4–8]. Recently, direct computer simulations of the phase separation and spinodal decomposition were made [9–12]; long dipolar chains were obtained numerically [13–15], and the crystallization of molecular liquids [16,17] and the crystallization of the vortices in 2D turbulence [18–20] were simulated.

Nontrivial results of dynamic simulations stimulate the study of analog structures based on continuous field analysis. Some analytical estimations can be performed on this intermediate level. Numerical simulations give a better understanding of the phenomena. In this Letter we concentrate on the phase ordering kinetics with non-conserved order parameter that is described by the generalized Ginzburg–Landau equation [21–23],

$$\partial\varphi(\boldsymbol{r},t)/\partial t = -\gamma(\delta H[\varphi(\boldsymbol{r},t)]/\delta\varphi), \qquad (1)$$

where H is the Ginzburg–Landau functional; γ is a positive kinetic coefficient, $\varphi(\boldsymbol{r},t)$ is the order parameter field. In recent works [24–29] the model (1) was used to study nucleation and growth of the new phase domains in different systems with the following general Ginzburg–Landau functional,

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$$H[\varphi(\boldsymbol{r})] = \int \mathrm{d}^d\boldsymbol{r}\,[(\nabla\varphi(\boldsymbol{r}))^2/2 + f(\varphi(\boldsymbol{r}))]$$
$$= \int \mathrm{d}^d\boldsymbol{r}\,F(\varphi(\boldsymbol{r})). \qquad (2)$$

The local free energy density for different problems (for example, for binary mixtures, axial magnets, martensites, etc. [4–8,30–32]) may be reduced to two standard catastrophes [33],

$$f(\varphi(\boldsymbol{r})) = \tau\varphi(\boldsymbol{r})^2/2 - 2\alpha\varphi(\boldsymbol{r})^3/3 + \beta\varphi(\boldsymbol{r})^4/4$$

and

$$f(\varphi(\boldsymbol{r})) = \tau\varphi(\boldsymbol{r})^2/2 - \alpha\varphi(\boldsymbol{r})^4/4 + \beta\varphi(\boldsymbol{r})^6/6$$

for the unique (scalar) combination $\varphi(\boldsymbol{r})$ of the order parameter components. At positive constants τ, α, β, and $\tau\beta < \alpha^2$ any function $f(\varphi(\boldsymbol{r}))$ has a metastable minimum at $\varphi(\boldsymbol{r}) = \varphi_0 = 0$ and a global one at $\varphi_+ = [\alpha + (\alpha^2 - \tau\beta)^{1/2}]/2\beta$ and $\varphi_+^2 = [\alpha + (\alpha^2 - \tau\beta)^{1/2}]/\beta$, respectively.

The nuclei appear from noise, $\zeta(\boldsymbol{r}, t)$, of the fluctuations. In the kinetic approach it is generated by a random term in Eq. (1),

$$\partial\varphi(\boldsymbol{r}, t)/\partial t = -\gamma(\delta H[\varphi(\boldsymbol{r}, t)]/\delta\varphi) + \zeta(\boldsymbol{r}, t),$$
$$\langle\zeta(\boldsymbol{r}, t)\rangle = 0,$$
$$\langle\zeta(\boldsymbol{r}, t)\zeta(\boldsymbol{r}', t)\rangle = 2D\delta(\boldsymbol{r} - \boldsymbol{r}')\delta(t - t'). \qquad (3)$$

Different versions of the modified equation have been used (see Refs. [21–32,34–42]). In particular, it was found [24,25] that critical nuclei appear from low-dimensional folds of the density $\varphi(\boldsymbol{r})$. Their maxima may be essentially lower than the equilibrium order parameter $\varphi_+(\boldsymbol{r})$. In this sense, the kinetic processes at second and first-order transitions are close to one another. At $T < T_c$, in both cases a new phase grows in the form of ordered domains inside the disordered matrix [41,42].

In the critical point of a second-order transition a large-scale structure of the fluctuating field is generated [43,44]. Its description [43] is based on the free energy renormalization by the fluctuations. The effective free energy $f_{\mathrm{eff}}(\varphi(\boldsymbol{r}))$ is given by a fixed solution of the RG equation [45–47]. In lowest approximation, the local part of the renormalized functional

$$H[\varphi] = \int_q G_0^{-1}(\boldsymbol{q})|\varphi(\boldsymbol{q})|^2/2 + \int \mathrm{d}^d\boldsymbol{r}\,f_{\mathrm{eff}}(\varphi(\boldsymbol{r}))$$
$$= H_0[\varphi] + H_I[\varphi] \qquad (4)$$

can be used in the kinetic equation as an effective free energy $f_{\mathrm{eff}}(\varphi(\boldsymbol{r}, t))$,

$$\partial\varphi(\boldsymbol{r}, t)/\partial t = -\gamma[\Delta\varphi(\boldsymbol{r}, t) - \partial f_{\mathrm{eff}}(\varphi(\boldsymbol{r}, t))/\partial\varphi]$$
$$+ \zeta(\boldsymbol{r}, t). \qquad (5)$$

In the fixed point of the RG equation the local density for $f_{\mathrm{eff}}(\varphi(\boldsymbol{r}))$ satisfies the RG equation,

$$\mathrm{d}f_{\mathrm{eff}}(\varphi) - (d-2)\varphi[\partial f_{\mathrm{eff}}(\varphi)/\partial\varphi]/2$$
$$+ \partial^2 f_{\mathrm{eff}}(\varphi)/\partial\varphi^2 - [\partial f_{\mathrm{eff}}(\varphi/\partial\varphi)]^2 = 0,$$

where d is the space dimensionality [48–52]. This cannot be reduced to a finite number of terms in the expansion and has asymptotes $f_{\mathrm{eff}}(\varphi) \propto \varphi^2/2 + \mathrm{const}$ at $\varphi \to \infty$ [47], instead of what is usually supposed, $f_{\mathrm{eff}}(\varphi) \propto \varphi^4/2 + o(\varphi^2)$. This anomalous behavior leads to long-living mesoscopic excitations in the system and generates a large-scale (fractal) structure of the fluctuating field in space. This structure is reflected in fine structures of the two-point correlation function

$$G(\boldsymbol{r} - \boldsymbol{r}') = \langle\varphi(\boldsymbol{r})\varphi(\boldsymbol{r}')\rangle \qquad (6)$$

and its Fourier transform $G(\boldsymbol{q}) = \langle\varphi(\boldsymbol{q})\varphi(-\boldsymbol{q})\rangle$. In the critical point $G(\boldsymbol{q})$ has a fractal structure at small momenta [44], which can be treated as the physical reason for the anomalous dimensionality described by the Fisher critical exponent $G(q) \propto q^{2-\eta}$ in the analytical theory.

The fluctuation structure in the critical region exists at $\zeta(r, t) \neq 0$ only. It drifts with time and does not lead to an ordering phase. However, it may be treated as a stationary structure, because a universal correlation function $G(q) = \langle\varphi(q)\varphi(-q)\rangle \propto q^{2-\eta}$ appears in this state. Besides, the time evolution of the averaged functional of the probability density, $w[\varphi] = \exp(-H_I[\varphi])$, coincides with a simple scale transformation [43]. This means that a scale-invariant structure develops in time [53].

Substitution of $f(\varphi)$ by $f_{\mathrm{eff}}(\varphi)$ is equivalent to partially taking account of microscopic fluctuations by integration of the partition function over small momenta. So, a kinetic simulation starts from intermedi-

ate (mesoscopic) scales. It allows one to study numerically even fine fluctuation effects (like the generation of nonzero η [44]). A rougher effect of fluctuation inducing of discontinuous phase transitions was extracted from the kinetics directly [28] (without previous renormalization of the functional by the RG procedure). In this Letter the opposite effect of inducing of a continuous phase transition by the fluctuations near the spinodal is presented.

2. Large-scale structure near spinodal

With numerical simulation the drift of the structure is directly visible. The nuclei of the new phase appear, disappear and move in space. It corresponds to the simple picture of very large fluctuations growing in a very smooth $f_{\rm eff}(\varphi)$ energy minimum at $\varphi = 0$. Some of these pictures were presented in the figures of Ref. [43]. Analogous pictures were presented also for the critical nuclei at the first-order phase transition [24,25]. In the second case, two characteristic energies should coincide to start the nucleation. They are the barrier in the free energy determined by the difference $\tau = (T - T_c)/T_c$, and the absolute value of the temperature T which is proportional to the intensity of the noise $T \sim D$ [2]. In thermodynamics the barrier disappears at the lability boundaries [1] (so-called overcooling and overheating spinodals). In kinetics, however, the metastable state can lose its stability at a nonzero barrier. This occurs even in a pure homogeneous system (without impurities, etc.) Fluctuations by themselves produce the nucleation centers.

Fluctuation inducing of a continuous phase transition (and tricritical points) has been predicted many years ago in the framework of the RG approach [54,55]. From an analytical point of view the maximum of the free energy density $f(\varphi) = f(\varphi(\boldsymbol{r}))$ disappears due to the renormalizations coming from the higher nonlinear terms in the expansion $f(\varphi) = \tau\varphi^2/2 - \alpha\varphi^4/4 + \beta\varphi^6/6 + \ldots$ This was accounted in a loop approximation and studied in different contexts [56–60], in particular, using an effective "temperature conductivity equation": $\partial f_{\rm eff}/\partial\theta = \partial^2 f_{\rm eff}/\partial\varphi^2$, where θ is the value of the one-loop correction [57]. Using $f(\varphi) = \tau\varphi^2/2 - \alpha\varphi^4/4 + \beta\varphi^6/6 + \ldots$ as an "initial condition" one can write its general solution $f_{\rm eff}(\varphi;\theta) = 2^{-1}(\pi\theta)^{-1/2}\int d\alpha \exp[-(\alpha - \varphi)^2/2\theta] f(\varphi)$ at arbitrary θ. One can prove that the maxima and minima of the effective function $f_{\rm eff}(\varphi;\theta)$ are suppressed on increasing of the θ value.

Qualitatively, the same phenomenon occurs in the presence of odd terms in the function $f(\varphi) = \tau\varphi^2/2 - \alpha\varphi^3/3 + \beta\varphi^4/4$. However, in this case the density of the fluctuating field φ averaged over the system, $\langle\varphi\rangle$, does not equal zero. It gives a main impact to the renormalization of $f(\varphi)$. Let us estimate this impact. For a stationary process the average $\langle\varphi\rangle$ changes adiabatically slowly, $(\delta H[\varphi(\boldsymbol{r},t)]/\delta\varphi)|_{\varphi=\langle\varphi\rangle} \to 0$ [25]. One can exclude this regular part from the kinetic equation $\varphi \to \varphi + \langle\varphi\rangle$ and write the following equation for the new field,

$$\begin{aligned}\partial\varphi(\boldsymbol{r},t)/\partial t &= -\gamma(\delta H[\varphi(\boldsymbol{r},t) + \langle\varphi\rangle]/\delta\varphi) + \zeta(\boldsymbol{r},t)\\ &\approx \gamma(\Delta\varphi(\boldsymbol{r},t) - \tau_{\rm eff}\varphi + \alpha_{\rm eff}\varphi^2 - \beta_{\rm eff}\varphi^3 + \ldots)\\ &\quad + \zeta(\boldsymbol{r},t),\end{aligned} \tag{7}$$

with the effective parameters

$$\tau_{\rm eff} = \tau - 2\alpha\langle\varphi\rangle + 3\beta\langle\varphi\rangle^2, \quad \alpha_{\rm eff} = \alpha - 3\beta\langle\varphi\rangle,$$
$$\beta_{\rm eff} = \beta. \tag{8}$$

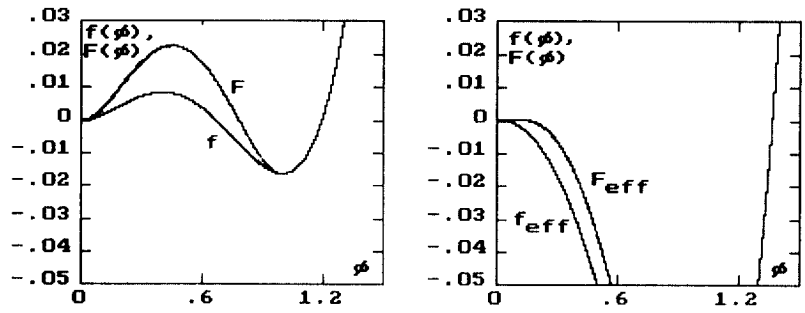
The two parameters $\tau_{\rm eff}$ and $\alpha_{\rm eff}$ have opposite dependences depending on the $\langle\varphi\rangle$ value. However, at $\beta_{\rm eff} = \beta = {\rm const} \equiv 1$ the qualitative structure of the trial function $f(\varphi)$ is determined by the unique parameter τ. At appropriate normalization of the order parameter one has $f(\varphi) = \tau\varphi^2/2 - (\tau + 1)\varphi^3/3 + \varphi^4/4$. This corresponds to the equation

$$\begin{aligned}\partial\varphi(\boldsymbol{r},t)/\partial t &= \gamma(\Delta\varphi(\boldsymbol{r},t) - \varphi(\varphi - \tau)(\varphi - 1))\\ &\quad + \zeta(\boldsymbol{r},t).\end{aligned} \tag{9}$$

Using the notations $\varphi_2\varphi_1 = \tau_{\rm eff}$ and $\varphi_1 + \varphi_2 = \alpha_{\rm eff}$, one can rewrite Eq. (7) in the same form,

$$\begin{aligned}\partial\varphi(\boldsymbol{r},t)/\partial t &\approx \gamma(\Delta\varphi(\boldsymbol{r},t) - \varphi_2^2\varphi(\varphi - \varphi_1/\varphi_2)(\varphi - 1))\\ &\quad + \zeta(\boldsymbol{r},t).\end{aligned} \tag{10}$$

The factor φ_2^2 can be excluded from this equation by a renormalization of the coordinates and time. As a result, the relation φ_1/φ_2 plays the same role as the



(a) (b)

Fig. 1. Local density of free energy. (a) Trial free energy $f(\varphi)$; (b) the same energy renormalized by the numerically found fluctuation corrections $f_{\rm eff}(\varphi)$. Complete energy densities $F(\varphi(r)) = (\nabla\varphi(r))^2/2 + f(\varphi(r))$ and $F_{\rm eff}(\varphi(r)) = (\nabla\varphi(r))^2/2 + f_{\rm eff}(\varphi(r))$ are shown also in both figures.

value τ in Eq. (7). For the lowest nontrivial order (up to $o(\langle\varphi\rangle)$ terms) one has

$$\varphi_1/\varphi_2 = [\alpha_{\rm eff} - (\alpha_{\rm eff}^2 - 4\tau_{\rm eff})^{1/2}]/[\alpha_{\rm eff} + (\alpha_{\rm eff}^2 - 4\tau_{\rm eff})^{1/2}] \approx \tau - 2\langle\varphi\rangle + o(\langle\varphi\rangle^2). \quad (11)$$

When the system goes to a spinodal, the value $\langle\varphi\rangle$ increases and the positive value of φ_1/φ_2 goes to zero. The maximum between stable and metastable minima disappears. This can be treated as a qualitative manifestation of fluctuation induced criticality in the system.

Eq. (11) gives an estimate only. This renormalization should appear automatically in numerical calculations with a trial form of the free energy and a sufficient volume of data (a numerical grid with 512×512 points is used actually). In its turn, it is interesting to check the above analytical estimates by substitution of the numerically found $\langle\varphi\rangle$ value in the formulae.

The results of the calculations are summarized in Figs. 1–5. In Fig. 1 the local density of the trial free energy (a) and the same energy renormalized by the numerically found fluctuation corrections (b) are presented.

The noise intensity $\langle\zeta(r,t)\zeta(r',t)\rangle = 2D\delta(r - r')\delta(t-t')$ for this figure is taken to suppress the maximum between global and local minima of $f_{\rm eff}(\varphi)$ for the first time. This noise is close to critical ($D \approx D_c$) for the nucleation. At the same noise the total energy density $F(\varphi(r)) = (\nabla\varphi(r))^2/2 + f(\varphi(r))$ has a very small but nonzero maximum. This is quite visible in both Fig. 1a and Fig. 1b. This maximum comes from a gradient term $(\nabla\varphi(r))^2/2$ and exists due to a nonuniform distribution of $\varphi(r)$ in the critical nuclei [25]. As a result, the nucleation does not start at the noise suppression maximum of the local $f_{\rm eff}(\varphi)$.

The space distribution of the order parameter density $\varphi(r)$ is given by the structure of the correlation function $G(r) = \langle\varphi(r)\varphi(0)\rangle$. It is shown in Figs. 2a–c. Two structures of $G(r)$ are presented. First the function is shown for $t = 0$ in Fig. 2a. It appears just with the "switch on" of the noise. The second plot (Fig. 2b) is presented at $t \to \infty$ and $D \approx D_c$. At $t \to \infty$ system goes to a stationary state with a large-scale fluctuation structure. The correlation function slowly decreases in this state, $G(r) \sim 1/r$. To test this behavior the function $G(r)$ is plotted in a double logarithmic scale also (Fig. 2c).

The large-scale structure of $\varphi(r)$ is nonuniform at $D \approx D_c$. The gradient energy $(\nabla\varphi(r))^2/2$ prevents a nucleation from this structure when the local barrier is suppressed completely. However, the impact from $(\nabla\varphi(r))^2/2$ decreases with expansion of the structure, $\varphi(r)$, in space. This process, in its turn, takes a time increasing with the scale of the structure. Owing to this, one can expect for infinite scaling growth of the structure at $D = D_c$ instead of standard nucleation.

Repeating the numerical experiments at different values of D one can localize the value of the critical

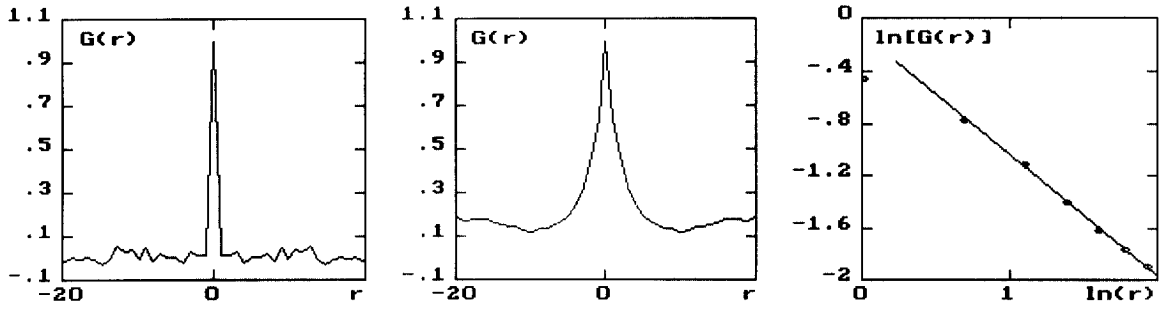


Fig. 2. Space dispersion of the correlation function $G(r) = \langle\varphi(r)\varphi(0)\rangle$. (a) At $t = 0$; (b) at $t \to \infty$. (c) The same function $G(r)$ as in b is plotted in a double logarithmic scale.

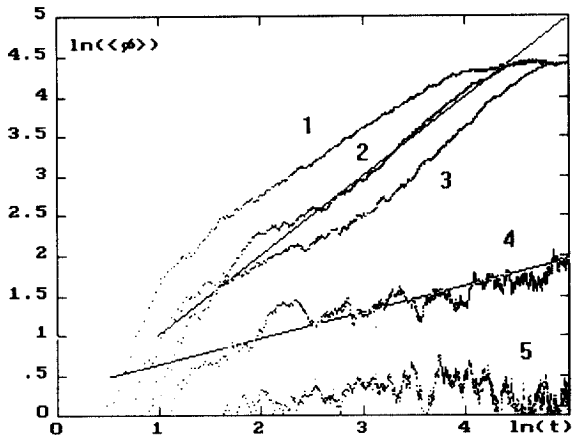


Fig. 3. The time dependences for the value $\langle\varphi\rangle$ at different amplitudes of the noise D are presented in a double logarithmic scale. Curves 1 and 2: $D \gg D_c$; curve 3: $D \geqslant D_c$; curve 4: critical regime at $D = D_c$; curve 5: saturation case $D < D_c$.

noise, $D = D_c$. At quite strong noise, $D \gg D_c$, nucleation occurs and the volume of the ordered state increases linearly with the time. This fact is illustrated by the curves 1 and 2 in Fig. 3. The time dependences for the value $\langle\varphi\rangle$ at different values of D are presented here in a double logarithmic scale. At $D \geqslant D_c$ a crossover (curve 3) to another behavior occurs. This corresponds to the formation of a large-scale fluctuation structure before nucleation.

Curve 4 corresponds to the critical regime at $D = D_c$. The scaling behavior $\langle\varphi\rangle \sim t^{1/3}$ is found for this regime. The last regime is analogous to the already mentioned development of the scale-invariant structure at the critical point. In both cases the renormalized free energy is extremely flat.

As a result domain growth occurs with the exponent $1/3$ instead of $1/2$ as expected for systems with non-conserved order parameter [61,62]. Growth here occurs through lowering of the gradient energy $(\nabla\varphi(\boldsymbol{r}))^2/2$ only. It is qualitatively similar to the systems involving a conserved order parameter where the asymptotic exponent $1/3$ is found (see, for example, Ref. [9] and references therein).

Finally, at $D < D_c$ (curve 5) saturation occurs. The beatings of the small value of $\langle\varphi\rangle$ look more important in comparison with the previous curves because the same logarithmic scale was used for all these plots. The impact of the fluctuations $\delta\varphi$ here is comparable to the value $\langle\varphi\rangle$ itself and lowers the accuracy. It is not very essential when one deals with the saturation regime. But, in principle, more averaging is needed in this case.

Fig. 4 presents the space distribution of the fluctuating field near the spinodal. The order parameter density is shown by the intensity of gray. White corresponds to the current maximum value of the order parameter

$$\varphi_{\max} \approx 0.3\varphi_+ = 0.3[\alpha + (\alpha^2 - \tau\beta)^{1/2}]/2\beta.$$

A fragment (256×512 grid points) of the total structure is shown in the plot.

When the amplitude of the noise is close to, but slightly larger than the critical one, $D \geqslant D_c$, the inten-

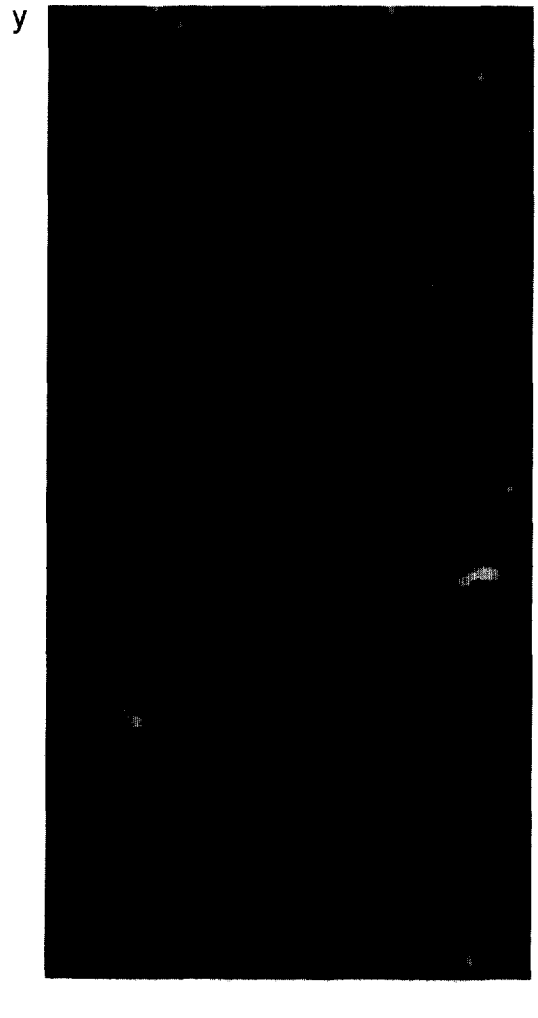


Fig. 4. Space distribution of the fluctuating field near the spinodal. The order parameter density is shown by the gray-scale. A white color corresponds to the current maximum value of the order parameter, $\varphi_{max} = 0.3\varphi_+ = 0.3[\alpha + (\alpha^2 - \tau\beta)^{1/2}]/2\beta$.

sive nucleation begins in a few places of space which have the highest current local density of the order parameter. As a rule these places are located near long (low-dimensional) density folds [25,44]. This process produces local groups and chains of the nuclei. Fig. 5 presents an example of such a group of nuclei. The gray-scale map is normalized to the equilibrium value of $\varphi = \varphi_+$. The relation between the magnitudes of the critical nuclei ($\varphi_c \approx 0.3\varphi_+$) and the basal value of $\langle\varphi\rangle$ in the fluctuation structure is seen directly from the two presentations (3D surface and

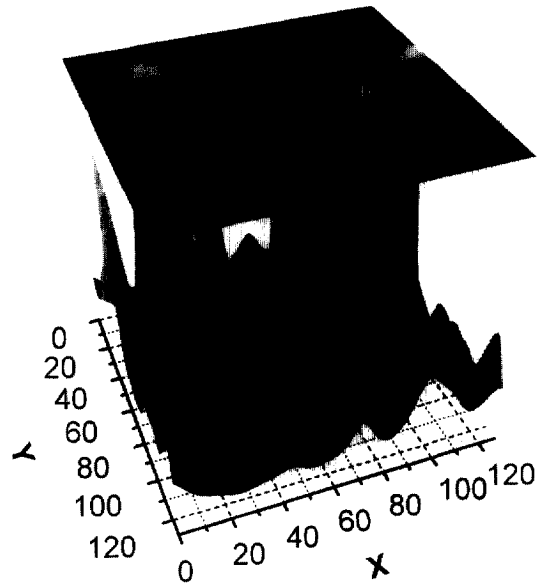


Fig. 5. An example of a local group of nuclei generated at $D \geqslant D_c$. The relation between the magnitudes of the nuclei and $\langle\varphi\rangle$ is shown by a combination of the 3D surface and the 2D map in the same figure. The gray-scale is normalized to the equilibrium value of $\varphi = \varphi_+$.

2D map, respectively) for the order parameter density $\varphi(\boldsymbol{r}) = \varphi(x, y)$.

In conclusion, we present the numerical simulations which provide evidence for the similarity between the ordering kinetics near the lability boundary of a first-order phase transition and the generation of a fractal large-scale structure of the fluctuating field in the critical region of a second-order transition. It was found also that the chains and localized groups of new phase nuclei are generated conjointly on the base of this structure from density folds of the order parameter.

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