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Stochastic inhomogeneous structures in nonequilibrium systems

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It is shown that stable inhomogeneous states of a complicated type, namely stochastically inhomogeneous structures (SIS), can arise spontaneously in kinetic phase transitions in homogeneous nonequilibrium systems, i.e., at the points when their homogeneous state becomes unstable. The phase portrait of the SIS has even in the one-dimensional case a set of nonisolated limit cycles, at some point of which a phase trajectory can go over randomly from one cycle to another. The onset of SIS is established by analyzing a system of two nonlinear differential equations of the diffusion type, which describe a definite class of nonequilibrium systems. The latter, in particular, include an electron-hole plasma uniformly heated by electromagnetic radiation, a weakly ionized gas plasma, nonequilibrium superconductors, as well as a number of important chemical and biological systems whose properties are determined by autocatalytic reactions. Static as well as traveling SIS can be excited in the systems under consideration also by a finite inhomogeneous perturbation. Methods are developed for a self-consistent qualitative derivation of one-dimensional and radially symmetrical stationary solutions and of the analysis of their stability. The form and velocity of the traveling one-dimensional SIS are found. General requirements on the form of two- and three-dimensional SIS are formulated on the basis of the stability analysis. It is shown that under the same conditions there exist in the system a number of different SIS, and the distinguishing features of the evolution of their instability with changing state of the system are analyzed. Explanations are offered for the experimental data and for the results of numerical investigations of a number of systems in which spatial dissipative structures arise.

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1. INTRODUCTION

The onset of a new structure in a system under thermodynamic equilibrium in the course of a phase transition can be regarded as the result of the growth of the order-parameter fluctuations on going through the point at which the initial phase loses stability. There can also be produced in this case spatially inhomogeneous distributions of the order parameter, for example multidomain states, noncommensurate phases,^{1,2} or a substructure of heterophase alloys.³ In a certain sense, similar phenomena occur in nonequilibrium homogeneous systems when their homogeneous state becomes unstable. Such a kinetic phase transition, however, leads to more varied effects: homogeneous and inhomogeneous oscillations can arise in nonequilibrium systems, traveling pulses and nonlinear waves are excited, or complicated stable inhomogeneous structures (IS) can be formed.⁴⁻¹⁷

In contrast to a true phase transition, in which new coherent states are established as a result of effective long-range interaction, in the here considered nonequilibrium systems the IS are produced as a result of diffusion processes.^{6,9,12,14-17} The spontaneous formation of IS as a result of diffusion instability can be described in unified fashion for a large class of physi-

cal, chemical, and biological systems whose properties depend on two parameters θ and η that differ in their spatial dispersion.¹⁴⁻¹⁷ The layering in such systems is due to the spatial decoupling of the "rapidly varying" parameter θ from the "slowly varying" parameter η when the system is unstable to θ at constant η . A nonlinear theory of one-dimensional IS (layers) for this class of systems was developed in preceding papers,^{16,17} in which principal attention was paid to spatially periodic structures, as well as to IS in the form of single layers.

The properties of the considered systems are described by two nonlinear equations of the diffusion type:

$$\tau_\theta \frac{\partial \theta}{\partial t} = L^2 \Delta \theta - q(\theta, \eta, A, \dots, G), \quad (1)$$

$$\tau_\eta \frac{\partial \eta}{\partial t} = L^2 \Delta \eta - Q(\eta, \theta, A, \dots, G) \quad (2)$$

with cyclic boundary conditions or with boundary conditions

$$\mathbf{n} \nabla \theta|_S = \mathbf{n} \nabla \eta|_S = 0, \quad (3)$$

corresponding to the absence of fluxes through the surface S of the system. These are the basic equations for the study of IS and of the propagation of perturbations in biological systems.⁶⁻¹² They correspond, in

particular, to the models of Turing^{6,7,9} and Fitz-Hugh and Nagumo.^{11,12} They also describe in greatest detail the experimentally investigated autocatalytic reaction of Belousov and Zhabotinskii.^{8,12} In the latter, θ and η are the concentrations of the intermediate products of the reactions; l , L and τ_θ , τ_η are the corresponding diffusion lengths and relaxation times, while

$$q = C\theta^2 + \eta(D\theta - E)(F\theta + G)^{-1} - \theta(1 - \eta); \quad Q = B\eta - \theta(1 - A\eta), \quad (4)$$

where A , B , C , D , F , and G are the kinetic coefficients and certain constant that characterize the rates of the reactions and the concentrations of the initial and final products.^{8,12}

Also reducible to the system (1), (2) are in essence the equations that describe the current distribution in Joule-heated semiconductor devices,^{14,18} as well as the equations describing a gas-discharge or an electron-hole plasma heated by an electric field or by electromagnetic radiation.^{13,15-17,19} In the case of the electron-hole plasma, the system (1), (2) consists of the balance equations for the particle numbers and energies, with θ a certain effective temperature of the electron-hole plasma.¹⁶ Equations (1) and (2) describe also layering in quasi-equilibrium systems, particularly ferroelectric semiconductors,²⁰ magnetic semiconductors,²¹ and nonequilibrium superconductors.²²

The problem of finding inhomogeneous stationary states of the system (1), (2) and of investigating their stability is therefore quite general, and its solution can reveal the general properties of a rather large class of nonequilibrium systems.

2. CONDITIONS FOR SPONTANEOUS FORMATION OF IS

The requirements on the form of the zero-isoclines of Eqs. (1), at which the IS are spontaneously produced, can be formulated on the basis of a linear analysis of the stability of the homogeneous states. It follows from this analysis that stability is lost if one of the following conditions is satisfied¹⁴:

$$\tau_\eta(q_0' + k^2 l^2) + \tau_\theta(\theta_n' + k^2 L^2) < 0, \quad (5)$$

$$k^2 l^2 L^2 + k^2 l^2 Q_n' + k^2 L^2 q_0' + Q_n' q_0' - Q_0' q_n' < 0. \quad (6)$$

From (5) and (6) it follows that at $L \gg l$ layering arises in such systems relative to

$$k = k_0 = (lL)^{-1/2} (Q_n' q_0' - Q_0' q_n')^{1/2}, \quad (7)$$

where

$$q_0' < -[(lL)^2 Q_n' + 2(lL)(Q_n' q_0' - Q_0' q_n')^{1/2}], \quad (8)$$

and $Q_n' > 0$ and $q_n' Q_0' < q_0' Q_n' < 0$.¹⁴ When $Q_0' > 0$ and $q_n' < 0$, then $d\eta/d\theta < 0$ [region II, Fig. 1(a)] for both zero-isoclines in the stability region of the homogeneous state. At some points θ_1^0 and θ_2^0 , where the q_0' vanish, the derivative $d\eta/d\theta$ reverses sign, i.e., the zero-isocline $q(\eta, \theta) = 0$ has an N-shaped form [Fig. 1(a)].

Inasmuch as in the general case the sign of Q_n' or Q_0' is not connected with the sign of q_0' , the condition $d\eta/d\theta < 0$ can be preserved on the zero-isocline $Q(\eta, \theta) = 0$ also outside the region $\theta_1^0 \leq \theta \leq \theta_2^0$ [Fig. 1(a), curves a and c]. Similar arguments for the case $Q_0' < 0$ and q_n'

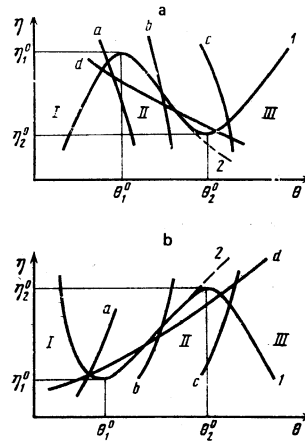


FIG. 1. The form of possible (a) and (b) zero-isoclines of Eqs. (1) and (2). Curves 1, 2 and a, b, c, d illustrate some of the possible cases realized at different values of the external parameters A , B , ... (for a supercooled system the corresponding values of θ for the homogenous state lie in region I, $\theta_{\text{hom}} \lesssim \theta_1^0$ —case a; for the heated region— $\theta_1^0 \lesssim \theta_{\text{hom}} \lesssim \theta_2^0$, i.e., θ_{hom} lies in region II—case b; for the supercooled region, $\theta_{\text{hom}} \gtrsim \theta_2^0$ —region III—case c; case d corresponds to equilibrium between the supercooling and superheating phases).

> 0 lead to the zero-isocline form shown in Fig. 1(b). The formulated conditions on the form of the zero-isocline are satisfied for all the systems discussed in the Introduction. In particular, the zero-isoclines corresponding to a heated electron-hole plasma^{16,23} and to the Belousov-Zhabotinskii reaction (4) take the form shown in Fig. 1(b).

3. ONE-DIMENSIONAL STOCHASTIC INHOMOGENEOUS STRUCTURES

In the one-dimensional case Eqs. (1) and (2) take the form

$$\frac{d^2 \eta}{dx^2} - Q(\eta, \theta) = \eta_{xx}'' + \frac{dU_\eta(\eta, \theta(\eta))}{d\eta} = 0, \quad (9)$$

$$\frac{\varepsilon^2 d^2 \theta}{dx^2} - q(\theta, \eta) = \varepsilon^2 \theta_{xx}'' + \frac{dU_\theta(\theta, \eta(\theta))}{d\theta} = 0, \quad (10)$$

where the length is measured in units of L , and $\varepsilon = l/L \ll 1$. It is seen from (9) and (10) that the stationary inhomogeneous solutions $\eta(x)$ and $\theta(x)$, i.e., the IS, are best regarded as one-dimensional trajectories of two interacting particles moving with coordinates η and θ and a "time" x in the respective potentials $U_\eta[\eta, \theta(\eta)]$ and $U_\theta[\theta, \eta(\theta)]$. For a consistent construction of the potentials U_η and U_θ and of the solutions $\eta(x)$ and $\theta(x)$, we develop a certain iteration procedure, in which we use as the zeroth approximation the form of U_η and U_θ and of $\eta(x)$ and $\theta(x)$ in the regions of the slowly and rapidly varying distributions, respectively.^{16,17}

Corresponding to the slowly varying distributions are the solutions (9) and (10) at $\varepsilon = 0$.^{16,17} It is seen from Fig. 1 that the $\eta(\theta)$ dependence given by $q(\theta, \eta) = 0$ is not single-valued, therefore the potential U_η (9) has three independent branches, I, II, and III [Fig. 2(a)]. The extremum of U_η corresponds to that branch on which is located the point of intersection of the zero-isoclines of Eqs. (9) and (10) (Figs. 1 and 2). From

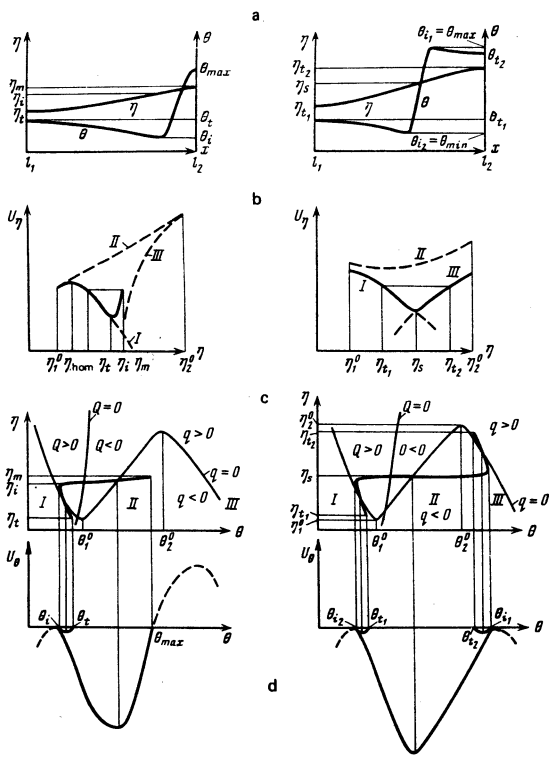


FIG. 2. Forms of certain elementary IS (a) and method of their construction. (The figures on the left correspond to a distribution in the form of a hot layer at the boundary of the segment, and on the right—in the form of a broad layer at the boundary of the segment). The forms of the potentials U_η (b) U_θ (d) (the dashed curves represent the potentials in the potentials in the approximation of slowly varying and rapidly varying distributions). Form of the zero-isocline (c) and the true plots of $\eta(\theta)$ (thick curves) for the corresponding IS. In this figure, as in all the following ones, the IS are constructed for the case of the zero-isoclines of Fig. 1b. The construction of the case of Fig. 1a is similar, with the variation of η and θ in the region of the slowly varying distributions, in contrast to those shown in Fig. 2–5, is not in counterphase but in phase.¹⁶

the condition $Q'_\eta > 0$ it follows in this case that U_η has a minimum when the point of intersection of the zero-isoclines lies on the unstable section (branch II, Fig. 1). However, all the slowly varying distributions corresponding to branch II of the potential U_η are unstable.¹⁵⁻¹⁷

The rapidly varying distributions correspond to solutions of Eqs. (9) and (10) at $\eta = \eta_0 = \text{const}$.^{16,17} In the range of values $\eta_1^0 < \eta_0 < \eta_2^0$ the potential U_θ takes the form of a potential well, i. e., $\theta(x)$ can have periodic solutions with a small characteristic length of the order of l . However, all the rapidly varying distributions, in the form of two and more oscillations, are unstable.¹⁶ Stable solutions can be combinations of smooth distributions corresponding to branches I and III of the potential U_η , with a single oscillation or half-oscillation of the rapidly varying distribution.^{16,17} Rapidly varying distributions can change into slowly varying ones only near saddle points of the potentials U_θ [Fig. 2(b)], when the variation of $\theta(x)$ becomes increasingly smoother as these points are approached.

We shall first describe the method of consistently constructing $\theta(x)$ and $\eta(x)$ and of the potentials U_η and U_θ in the entire range of variation of θ and η for a relatively short sample of length $l_x = l_1 - l_2 \leq L$, on the boundaries of which $\theta'_x = \eta'_x = 0$. It follows from (9) and (10) that the solutions should satisfy the integral conditions

$$\int_{\eta_1}^{\eta_m} Q(\eta, \theta(\eta)) d\eta = \int_{l_1}^{l_2} Q dx = 0; \quad \int_{\theta_1}^{\theta_m} q(\theta, \eta(\theta)) d\theta = \int_{l_1}^{l_2} q dx = 0, \quad (11)$$

where η_1 , θ_1 and η_m , θ_m are the values of η and θ at the extremal points (at the boundaries of the sample), corresponding respectively to the smooth and abrupt distributions [Fig. 2(a) on the left].

3.1. In a “supercooled” system (Fig. 1) of small size ($l_x < L$) the conditions (11) are most easily satisfied by a distribution of $\theta(x)$ in the form of “narrow” hot layer at the sample boundary (Fig. 2). The relative change of η in the region of the slowly varying distribution is

$$\Delta\eta = \eta_1 - \eta_2 \approx l_x/L,$$

and in the region of the rapidly varying distribution

$$\eta_m - \eta_1 \approx l/L.$$

It follows then from (11) that in the region of the rapidly varying distribution

$$Q \approx -e^{-l_x/L}.$$

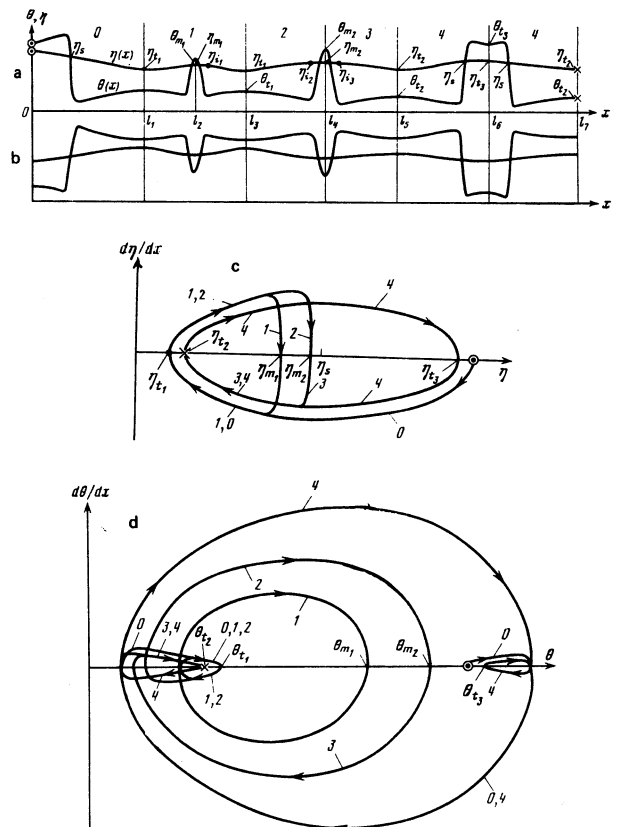


FIG. 3. Form of certain one-dimensional SIS (a, b) and fragment of the phase portrait (c, d) corresponding to Fig. 3a. (The sections of the phase trajectories and of the corresponding distributions are designated by the same numbers in Fig. 3a, 3b, and 3c).

Inasmuch as $dU_\eta/d\eta = -Q$, it follows that near a certain $\eta = \eta_i$ [Fig. 2(b)] the potential U_η moves sharply upward from the branch I of the potential U_η , which corresponds to the slowly varying distributions, and forms a steep wall, i. e., it is transformed into a potential well. The distribution $\eta(x)$ corresponds to finite motion of the particle in such a well.

In the region of slowly varying distributions it is possible to the form of the zero-isocline $q = 0$ [Fig. 2(c)], and of $\eta(x)$ to reconstruct $\theta(x)$ [Fig. 2(a)]. Thus, near $\eta = \eta_i$, where $\theta'_x = 0$, the value of $\theta(x)$ decreases with a characteristic length of the order of unity (L), and therefore $\theta''_x \approx -1$, i. e., $q \approx -\varepsilon^2$ according to (10). In the region where the smooth and abrupt distributions are joined, θ'_x should again go through zero [Fig. 2(a)], i. e., at a certain point of the smooth distribution $q = -\varepsilon^2 \theta''_x = 0$, and consequently the true plot of $\eta(\theta)$ intersects the plot of the local relation $q(\theta, \eta) = 0$ [Fig. 2(c)]. Since $dU_\theta/d\theta = -q$, the potential U_θ that describes the slow variation of $\theta(x)$ takes the form of a shallow (of the order of ε^2) potential well. From the condition of continuity of θ''_x , this well can be joined together with the U_θ well corresponding to the rapidly varying distribution only near the saddle point, where $dU_\theta/d\theta \rightarrow 0$ [Fig. 2(d)].

In a "heated" system (Fig. 1) the conditions (11) are satisfied as a rule by a broad layer at the sample boundary. Such a solution corresponds to a transition from motion in branch I of the potential U_η [Fig. 2(b)] to motion in branch III of the potential U_η through a rapidly varying distribution in the form of a half-oscillation near $\eta_0 = \eta_s$, at which the extrema of U_θ coincide [Fig. 2(c)]. Since motion in branch I of the potential U_η corresponds to $Q > 0$, and in branch III to $Q < 0$, it follows that in the region of the rapidly varying distribution Q reverses sign, and U_η has a minimum near $\eta \approx \eta_s$ [Fig. 2(b)].

In a small-size "superheated" system (Fig. 1), the conditions (11) are easiest to satisfy by solutions corresponding to a distribution in the form of a "cold" layer at the sample boundary.^{16,17} A self-consistent construction of the potentials and solutions is similar in this case to the case of a narrow "hot" layer, the only difference being that we use branch III rather than I of the potential U_η .

3.2. In long samples ($l_x \gg L$), periodic IS can be constructed by successive mirror reflections of one of the types of the elementary distributions [Fig. 2(a)]. In addition, in a supercooled system there are realized distributions in the form of a single hot layer or several layers far from one another. Corresponding to such a distribution is a trajectory of a "particle" in the potential U_η . This trajectory becomes closed at the saddle point $\eta_t = \eta_{hom}$ [Fig. 2(b)], and when this point is approached $\eta(x)$ and $\theta(x)$ tend to homogeneous values. Similarly, in a superheated system there can exist a single cold layer or several widely spaced layers. In a heated system, a solution in the form of a single layer is unstable, inasmuch as it goes over on the periphery into an unstable homogeneous state.

3.3. A distribution in the form of a hot layer [from l_1 to l_3 on Fig. 3(a)] can be joined at the point $x = l_3$, where $\eta = \eta_{t1}$ and $\theta = \theta_{t1}$, to a distribution of similar form but with different values η_{m2} and θ_{m2} at a certain point l_4 [Fig. 3(a)]. In fact, it follows from the construction method (Fig. 2) that if the dimension (the length $l_1 - l_2$) of a short sample is varied, it is possible to construct a set of potentials U_η and U_θ , and consequently a set of solutions that differ in the values of η_t and η_m on the sample boundary and of η_i in the transition region (Fig. 2). Naturally, one can choose from among this set various distributions with close values of η_i but with different values of η_t and η_m . This situation can be represented as a branching of the potential U_η near $\eta \approx \eta_i$ [Fig. 4(a)]. This branching that takes place at different values of η_i , therefore among the set of potentials U_θ corresponding to different values of η_i it is possible to choose those corresponding to motion of a particle with one and the same value of η_{t1} at the turning point [Fig. 4(a)]. The potentials U_θ corresponding to these distributions take the form of Fig. 4(b). This makes it possible to join together at the point l_3 [Fig. 3(a)] solutions in the form of narrow hot layers of different amplitude.¹⁶ This joining of different solutions in the region of the smooth distributions at $\eta = \eta_t$ and $\theta = \theta_t$ will be called a junction of type 1.

A junction of type 1 can be produced between a narrow hot layer and a broad one [at the points $x = l_1$ and $x = l_5$, Fig. 3(a)]. The potentials U_η and U_θ corresponding to this case are shown in Figs. 4(c) and 4(d), respectively. Similarly, a type-1 junction (on the branch III of the potential U_η) is produced between distributions

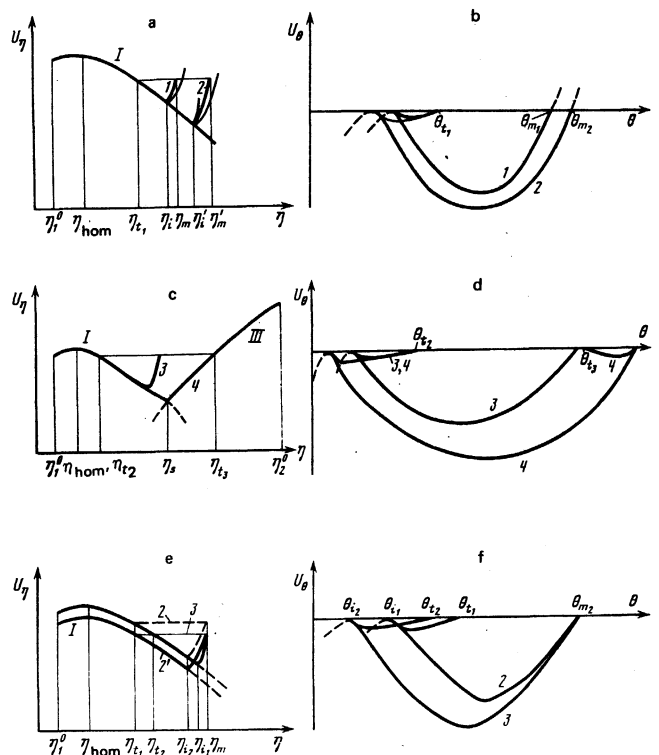


FIG. 4. Illustrating the method of constructing unperiodic and nonsymmetrical IS.

in the forms of narrow cold layers with different amplitudes and different periods, on the one hand, and broad layers [Fig. 3(b)].

3.4. It is possible to join together different elementary distributions not only in the region of slowly varying distributions (of type 1 at η_t and θ_t , Fig. 3) but also in the region of rapidly varying distributions (of type 2 at points η_m and θ_m , Fig. 3). This type of joining [at the point $x=l_4$, Figs. 3(a) and 3(b)] leads to a distribution in the form of an asymmetrical hot (cold) layer.

Among the many potentials U_η and U_θ , there can exist some corresponding to elementary distributions with identical values of η_m and θ_m but with different values of η_t and θ_t [potentials 2 and 3 in Figs. 4(e) and 4(f)]. By shifting the potentials U_η [in the form of the dashed curve 2 on Fig. 4(e)] down by a certain amount [curve 2', Fig. 4(e)] in such a way that the values of U_η for the different trajectories coincide at the point $\eta=\eta_m$, we obtain a certain potential U_η [solid curve on Fig. 4(e)], in which the motion of the particle corresponds to a distribution of the form of an asymmetrical hot layer [$l_3 \leq x \leq l_5$, Fig. 3(a)]. A distribution in the form of an asymmetrical cold layer [$l_3 \leq x \leq l_5$, Fig. 3(b)] is constructed in similar fashion. A type 2 juncture is also possible between different broad layers.

3.5. Joining together the solutions in the form of elementary distributions in accordance with type 1 or 2 at various points, it is possible to construct stochastically inhomogeneous structures (SIS) of complicated form (Fig. 3). The corresponding phase trajectory on the plane η and θ does not have isolated limit cycles (homoclinic trajectories) typical of ordinary dynamic systems. The situation here recalls that realized in generators of stochastic oscillations, for which the presence of strange attractors is typical.²⁴ In the considered systems, however, the "random" variation of θ and η is not with respect to time, but with respect to the coordinate x . This difference is significant, since l_x is finite, and definite conditions are specified at the boundaries of the system. Thus, SIS are realized at $l_x \gg L$ and for definite parameters of the system (for a definite form of the zero-isoclines and for a definite point of their intersection). The transition from one limit cycle to another takes place in random fashion in the region of the slowly (type 1) or rapidly (type 2) varying distribution at points where $\theta'_x=0$ (accurate to ϵ).

4. CENTRALLY SYMMETRICAL IS

Centrally symmetrical stationary distributions in the two- and three-dimensional cases satisfy, according to (1) and (2), the equations

$$\begin{aligned} \frac{d^2\eta}{d\rho^2} + \frac{2^s}{\rho} \frac{d\eta}{d\rho} + \frac{dU_\eta}{d\eta} &= 0; \\ \epsilon^2 \frac{d^2\theta}{d\rho^2} + \epsilon^2 \frac{2^s}{\rho} \frac{d\theta}{d\rho} + \frac{dU_\theta}{d\theta} &= 0, \end{aligned} \quad (12)$$

where $s=1$ (or zero) for spherically (cylindrically) symmetrical IS. The solutions of the system (12) can also be formally represented as an aggregate of elementary one-dimensional trajectories of two interact-

ing particles, but in contrast to (9) and (10), the particles move in the presence of friction forces of constant sign, which decrease with increasing ρ . We therefore have here in place of (11)

$$\begin{aligned} \Delta U_\eta &= \int_{\eta_1}^{\eta_2} Q d\eta = \int_{R_1}^{R_2} \left(\frac{d\eta}{d\rho} \right)^2 \frac{2^s}{\rho} d\rho > 0; \\ \Delta U_\theta &= \int_{\theta_1}^{\theta_2} q d\theta = \epsilon^2 \int_{R_1}^{R_2} \left(\frac{d\theta}{d\rho} \right)^2 \frac{2^s}{\rho} d\rho > 0. \end{aligned} \quad (13)$$

Thus, in contradiction to the statements of Fife²⁵ and of Nicolis and Prigogine (Ref. 7, formula 8.85), the condition (11) is not general. The finite curvature of the surface that separates the "phases" in the IS leads to the onset of a "surface-tension force" that causes violation of the "Maxwell rule" equivalent to conditions (11).

In a large-radius heated system (Fig. 1), solutions may be realized in the form of spherical (cylindrical) layers²⁶ of various thicknesses. Since $\Delta U_\theta > 0$ (13), on going from branch III to branch I of the potential U_η [trajectory of particle 1, Fig. 5(b)] the potential U_θ should take the form of curve 1 of Fig. 5(c). Such a potential is realized at $\eta_1 < \eta_s$. At the turning point $\eta=\eta_{t1}$ [$\rho=R_1$, Figs. 5(a) and 5(b)], a transition takes place from the first to the second trajectory in the po-

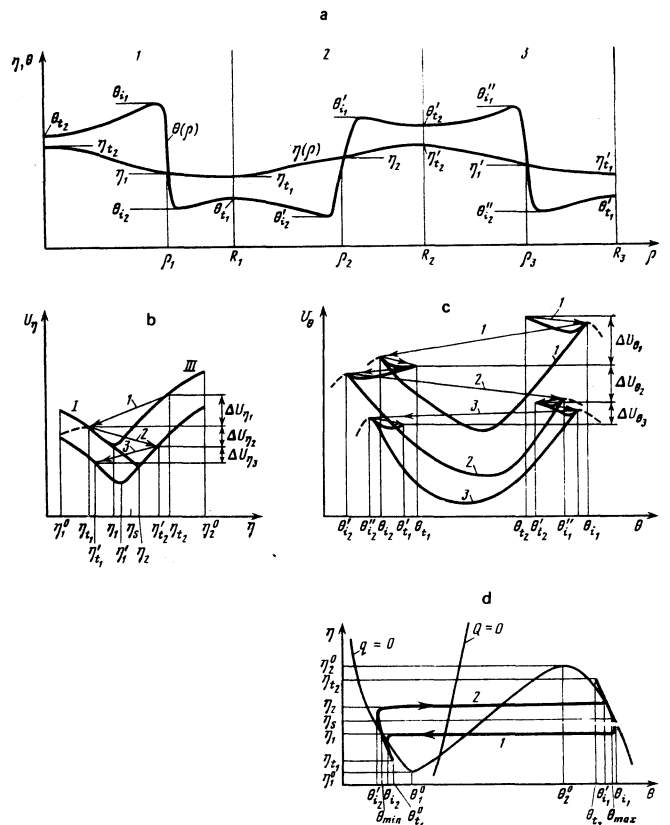


FIG. 5. IS in the form of hot spherical (cylindrical) layers (a) and method of their construction: forms of the potentials U_η (b) and U_θ (c) with the corresponding trajectories of the particles in them, d—form of the zero-isoclines $Q=0$ and of the true plots of $\eta(\theta)$ (thick curves). (The IS sections 1, 2, and 3 on Fig. 5a correspond to the curves with the same numbers on Figs. 5b, 5c, and 5d).

tential U_n , whose minimum is located at $\eta_2 > \eta_s$ [Fig. 5(b)]. Actually the transition from branch I to III [Fig. 5(d)] can take place from the potential U_θ takes the form of curve 2 of Fig. 5(c). At $R_2 \leq \rho \leq R_3$ [Fig. 5(a)], $\eta(\rho)$ and $\theta(\rho)$ correspond to the third trajectory in the potential U_n , whose minimum corresponds to $\eta_1 > \eta'_1 > \eta_s$, since ΔU_n and ΔU_θ (13) decrease with increasing ρ .

Thus, the larger the curvature of the spherical (cylindrical) layer, the more distorted it is compared with the symmetrical broad layer in the one-dimensional case. On the other hand, with increasing ρ , the "friction force" (13) tends to zero, and the spherical layers become more symmetrical. Owing to the finite curvature, a narrow spherical (cylindrical) layer is also distorted. Just as in the one-dimensional case, the combination of broad spherical (cylindrical) with one another and with narrow layers can be quite complicated. An SIS is easier to realize in the form of a sequence of different hot layers when θ_{hom} lies near θ_1^0 , and in the form of cold layers near θ_2^0 (Fig. 1).

In a supercooled system (Fig. 1), solutions exist in the form of a single "hot drop" (or "spot"). The latter is possible when the trajectory 1 terminates at the point $\eta_{i1} = \eta_{\text{hom}}$ corresponding to an extremum of U_n [dashed curve in Fig. 5(b)]. An IS in the form of a single drop can be produced either by a finite external perturbation or when a more complicated IS becomes unstable when one of the external parameters A, B, \dots changes. Similarly, in a superheated system (Fig. 1) of large size it is possible to excite a stable (Sec. 6) single "cold drop" (or spot) of small as well as large radius. We note that the greater the difference between θ_1^0 and θ_2^0 (Fig. 1), the easier it is to realize the conditions (13) for the onset of a small-radius drop (or spot).

5. TRAVELING SIS

States in the form of SIS that move without attenuation can be realized in the systems under consideration. We analyze this for the one-dimensional case. Changing over in (1) and (2) to the variable $x - vt$, we obtain

$$\begin{aligned} \frac{d^2 \eta}{dx^2} + v \frac{d\eta}{dx} + \frac{dU_n}{d\eta} &= 0; \\ \varepsilon^2 \frac{d^2 \theta}{dx^2} + \alpha v \frac{d\theta}{dx} + \frac{dU_\theta}{d\theta} &= 0, \end{aligned} \quad (14)$$

where time is measured in units of τ_n ($\alpha = \tau_\theta / \tau_n$), and the velocity in units of L / τ_n . Equations (14) are of the same form as (12) except that the friction forces of constant sign are proportional in them to the constant velocity v and not to ρ^{-1} . Therefore, the principle of construction of traveling broad and narrow layers, and also of their various combinations, is similar to that described in Sec. 4. Moving SIS are distorted relative to the static ones, and the distortion is larger the higher the velocity. Multiplying Eqs. (14) by η'_x and θ'_x respectively and integrating over each i th elementary segment on whose end points $\eta'_x = \theta'_x = 0$, we obtain the conditions for the determination of the velocity

$$v = \Delta U_{\eta_i} \left[\int \left(\frac{d\eta}{dx} \right)^2 dx \right]^{-1} = \Delta U_{\theta_i} \alpha^{-1} \left[\int \left(\frac{d\theta}{dx} \right)^2 dx \right]^{-1}, \quad (15)$$

where ΔU_{η_i} and ΔU_{θ_i} are the decreases of the potential

energies of the particles for each elementary trajectory [Figs. 5(b) and 5(c)]. The change ΔU_θ or ΔU_n for a broad layer can be larger than for a narrow one, so that broad layers can move with larger velocity than narrow ones.

6. STABILITY OF IS

6.1. The form of the realized SIS can be deduced in the general case only from an analysis of their stability. Linearizing Eqs. (1) and (2) with respect to

$$\delta\theta = \delta\theta(x) e^{-t}, \quad \delta\eta = \delta\eta(x) e^{-t},$$

we obtain¹⁾

$$(\hat{H}_n - \beta\gamma) \delta\eta = -\varepsilon^2 Q'_\theta \delta\theta, \quad \hat{H}_n = -\Delta + V_n, \quad V_n = \varepsilon^2 Q'_n(\eta(x), \theta(x)), \quad (16)$$

$$(\hat{H}_\theta - \gamma) \delta\theta = -q'_n \delta\eta, \quad \hat{H}_\theta = -\Delta + V_\theta, \quad V_\theta = q'_\theta(\theta(x), \eta(x)). \quad (17)$$

In these equations and hereafter the length is measured in units of l and the time in units of τ_θ ; $\beta = (\tau_n / \tau_\theta)(l^2 / L^2) \equiv \alpha^{-1} \varepsilon^2$.

We expand $\delta\eta$ and $\delta\theta$ in series in the eigenfunctions $\delta\eta_i$ and $\delta\theta_n$ of the operators \hat{H}_n and \hat{H}_θ and, by suitable transformations, obtain

$$\det \left[(\lambda_n - \gamma) \delta_{nm} + \sum_{i=0}^{\infty} P_{inm} \right] = 0, \quad (18)$$

$$P_{inm} = -\langle q'_n \delta\eta_i \delta\theta_m \rangle \langle Q'_\theta \delta\theta_n \delta\eta_i \rangle (\mu_i - \alpha^{-1} \gamma)^{-1}, \quad (19)$$

where the symbol $\langle \dots \rangle$ denotes averaging of the function over the volume of the system.

Some conclusions concerning the eigenvalues λ_n and $\mu_i = \varepsilon^2 \mu_i$ of the operators \hat{H}_θ and \hat{H}_n respectively can be drawn even from the linear theory of stability (Sec. 2). It follows from (2) and (6) that if $q \neq f(\eta)$, then the fluctuations $\delta\theta$ increase when $q'_\theta < 0$. The more stringent instability condition (8) is due to the fact that $\delta\theta$ causes perturbations $\delta\eta$ that are damped. In fact, owing to $V_n = \varepsilon^2 Q'_n > 0$ (16), all the $\mu_i > 0$ (Ref. 16) and the change $\delta\eta$ causes damping of the development of the fluctuations $\delta\theta$. On the other hand, the presence of instability in $\delta\theta$ denotes that some of the λ_n should be negative, i. e., $V_\theta = q'_\theta < 0$ in a certain region. Inasmuch as on the branches I and III of the $\eta(\theta)$ zero-isocline $q = 0$ (Fig. 1) we have $q'_\theta > 0$, and on branch II we have $q'_\theta < 0$, (see Sec. 2), it follows that in the IS regions corresponding to slowly varying distributions $V_\theta > 0$, and only in a certain part of the region of the rapidly varying distributions do we have $V_\theta < 0$ [near the point of intersection of the zero-isocline $q = 0$ with the true plot of $\eta(\theta)$, shown for the one-dimensional case in Fig. 2(c)].

Thus, the potential V_θ corresponding to stable IS, i. e., to successive combinations of slowly and rapidly varying distributions, constitutes in the general case a series of narrow (at least in one of the directions) potential wells with sufficiently large distances between them. Consequently, $\delta\theta_n$ with $\lambda_n < 0$ are strongly localized in the regions of the rapidly varying distributions, where $V_\theta < 0$ (for the one-dimensional case, see Fig. 8 of Ref. 16).

We emphasize that IS describable by only one of the equations (1) or (2) are unstable (Refs. 4 and 7).²⁾ In particular, at constant η , any inhomogeneous station-

ary solution of equation (1) is unstable. Indeed, at $\delta\eta=0$ the spectrum of the values of γ coincides according to (17) with the eigenvalues λ_n of the operator \hat{H}_0 . Applying the operator ∇ to Eq. (1) for the stationary states, and then multiplying by the unit vector \mathbf{n} at $\eta=\text{const}$, we find that $\delta\theta \propto \mathbf{n} \cdot \nabla\theta(\mathbf{r})$ (for cyclic boundary conditions) is the eigenfunction $\delta\theta_k$ corresponding to the eigenvalue³⁾ $\lambda_k=0$. Any inhomogeneous solution $\theta(\mathbf{r})$ under cyclic boundary conditions has not less than one extremum, and consequently $\mathbf{n} \cdot \nabla\theta(\mathbf{r})$ has not less than one node. This means that $\delta\theta_k = \mathbf{n} \cdot \nabla\theta(\mathbf{r})$ is not a function of the ground state of the operator \hat{H}_0 , and, at any rate, $\lambda_n < 0$ for $n < k$.²⁸ This conclusion remains in force under the boundary conditions (3), as well as for a monotonic distribution for which $\lambda_0 < 0$ by virtue of the fact that $\nabla[\nabla\theta(\mathbf{r})]$ is not zero on the boundary of the sample.¹⁶

6.2. We illustrate first the damping character of the variation of η for a system whose size is less than L . In such a system, the $\delta\eta_k(\mathbf{r})$ are strongly damped as a result of the large diffusion fluxes they cause, i. e., μ_k with $k \geq 1$ greatly exceed μ_0 .¹⁶ This allows us to confine ourselves to the first terms in the sums of (18):

$$\left(1 + \sum_{n=0}^{\infty} P_{0nn} (\lambda_n - \gamma)^{-1}\right) \prod_n (\lambda_n - \gamma) = 0. \quad (20)$$

Since $\delta\eta_0$ is a weak function of the coordinate, we have according to (19)

$$P_{0nn} = a_n \mu_0 (\mu_0 - \alpha^{-1} \gamma)^{-1}, \quad a_n = -\langle q_n' \delta\theta_n \rangle \langle Q_0' \delta\theta_n \rangle \mu_0^{-1}. \quad (21)$$

In contrast to an electron-hole plasma,^{16,17} in the general case P_{0nn} (21) depends on γ and on $\alpha = \tau_\theta / \tau_\eta$. We shall show that in the well V_0 corresponding to a broad layer at the sample boundary [Fig. 2(a)], only $\lambda_0 < 0$ and $|\lambda_0| \ll 1$, in contrast to a narrow layer, for which $\lambda_0 \approx -1$.¹⁶ Differentiating Eq. (10) with respect to x , we obtain

$$\hat{H}_0 \frac{d\theta}{dx} + q_n \frac{d\eta}{d\theta} \frac{d\theta}{dx} = 0. \quad (22)$$

Multiplying (22) from the left by $\delta\theta_0$ and averaging over the sample, we obtain²⁸

$$\lambda_0 = \left[-\langle q_n' \frac{d\eta}{d\theta} \frac{d\theta}{dx} \delta\theta_0 \rangle + \left(\delta\theta_0 \frac{d^2\theta}{dx^2} \Big|_l \right) \left[\langle \delta\theta_0 \frac{d\theta}{dx} \rangle \right]^{-1} \right]^{-1} \approx -\varepsilon \frac{L_x}{L}. \quad (23)$$

If it is recognized that θ_x' and $\delta\theta_0$ attenuate exponentially outside the region of the abrupt distribution, where $d\eta/d\theta \approx \varepsilon(L_x/L)$ (see Sec. 3), then we obtain an estimate for λ_0 (23). The depth and width of the potential well V_0 is of the order of infinity, therefore $|\lambda_0 - \lambda_1| \approx 1$ (see in particular, Refs. 16, 17, and 29). Since $\lambda_0 > -\varepsilon$, it follows that $\lambda_1 \approx 1$, i. e., the spectrum of λ_n contains only one small negative value.

We shall show now that this conclusion remains in force also for radially symmetrical fluctuations in the case of a single spot or drop (Sec. 4), at least one with a large radius $\rho_1 \gg l$. Differentiating the second equation of (12) with respect to ρ , we have

$$\begin{aligned} \hat{H}_0 \frac{d\theta}{d\rho} + \left(q_n' \frac{d\eta}{d\theta} + \frac{2'}{\rho^2} \right) \frac{d\theta}{d\rho} &= 0; \\ \hat{H}_0 &= -\frac{d^2}{d\rho^2} - \frac{2'}{\rho} \frac{d}{d\rho} + V_0. \end{aligned} \quad (24)$$

Then, in analogy with the derivation of (23), we find at $\rho_1 \gg l$ that

$$\begin{aligned} \lambda_0 &= \left[-\langle \delta\theta_0 \frac{d\theta}{d\rho} \left(\frac{2'}{\rho^2} + q_n' \frac{d\eta}{d\theta} \right) \rangle + \delta\theta_0 \frac{d^2\theta}{d\rho^2} \Big|_{\rho_1} \right] \left[\langle \delta\theta_0 \frac{d\theta}{d\rho} \rangle \right]^{-1} \\ &\approx -\varepsilon \frac{R_1}{L} + \frac{l^2}{\rho_1^2}, \end{aligned} \quad (25)$$

where R_1 is the radius of the entire system ($l \ll R_1 < L$). The estimate in (25) is valid at $\rho_1 \approx l$, while in the case of a small-radius drop ($\rho_1 \approx l$) we have according to (25), $\lambda_0 \approx -1$, just as for a narrow layer.¹⁶

Thus, in the λ_n spectrum of the considered IS, only $\lambda_0 < 0$, and $P_{000} \neq 0$ (21), so that it follows from (20) that the stability condition reduces to the absence of zeros of the complex function

$$f(\omega) = 1 + \sum_{n=0}^{\infty} \frac{a_n [(\lambda_n - \alpha^{-1} \mu_0^{-1} \omega^2) + i\omega(1 + \alpha^{-1} \mu_0^{-1} \lambda_n)]}{(\lambda_n^2 + \omega^2)(1 + \alpha^{-2} \mu_0^{-2} \omega^2)} \quad (26)$$

in the upper half-plane of $\omega = -i\gamma$. According to the argument principle, the number of zeros n in the upper half-plane of ω is $p + (2\pi)^{-1} \arg f(\omega)$. Since only $\lambda_0 < 0$, the function $f(\omega)$ has only one pole p . The change of $\arg f(\omega)$ on going around the upper ω half-plane is uniquely determined when

$$\sum_{n=0}^{\infty} a_n (1 + \alpha^{-1} \mu_0^{-1} \lambda_n) > 0. \quad (27)$$

In this case $\omega \text{Im} f(\omega) > 0$ and at

$$D = 1 + \sum_{n=0}^{\infty} \frac{a_n}{\lambda_n} < 0 \quad (28)$$

we have $\arg f(\omega) = -2\pi$, since $f(\pm 0) = D$ and $f(\pm \infty) = 1$. Consequently, $n = p - 1 = 0$, i. e., $f(\omega) = 0$ has no solutions in the upper half-plane, and the considered distributions are stable. On the other hand if $D > 0$, then $\arg f(\omega) = 0$ and $n = p = 1$, i. e., the IS is unstable.

Following the preceding studies,^{16,17} it can be shown that

$$\frac{d\langle \eta \rangle}{dA} = D^{-1} \mu_0^{-1} \left[-\langle Q_A' \rangle + \sum_{n=0}^{\infty} \langle Q_0' \delta\theta_n \rangle \langle q_n' \delta\theta_n \rangle \lambda_n^{-1} \right], \quad \mu_0 = \langle Q_n' \rangle. \quad (29)$$

Thus, the points at which $d\langle \eta \rangle / dA = \infty$ determine the limits of the interval of variation of the parameter A , within which the stability condition (28) is satisfied. The dependences of $\langle \eta \rangle$ on A in the one-dimensional case were obtained for certain systems earlier.^{16,17,30} On the basis of the analysis developed in these papers, it is easy to construct the dependence of $\langle \eta \rangle$ on any of the parameters in various systems for the case of periodic two- and three-dimensional IS. When the state of the system depends on several parameters A, B, C, \dots (1), (2), then, analyzing the dependence of $\langle \eta \rangle$ on them, we can establish that region of their variation in which (29) is not satisfied, and consequently the given IS is certainly unstable.

Since the IS consists of sections close to slowly varying distributions, for which $q_n' Q_0' < 0$, and rapidly vary-

ing distributions, for which $q_n'Q_n' < 0$ at least near the points of intersection of the zero isocline $q = 0$ with the true plot of $\eta(\theta)$, it is natural to assume that all $a_n \geq 0$ (21). At $a_n \geq 0$, the condition (27) is certainly satisfied if

$$\lambda_0 > -\alpha\mu_0. \quad (30)$$

According to (16) and (17),

$$\lambda_0 \approx \langle q_n' \delta \theta_n^2 \rangle, \quad \mu_0 \approx \langle Q_n' \delta \eta_n^2 \rangle,$$

i. e., the violation of condition (30) correlates with the satisfaction of the condition (5) and is connected with the frequency mismatch of the parameters η and θ (Ref. 14): at $\alpha = \tau_\theta / \tau_\eta \ll 1$, the variation of η is incapable of following the growing fluctuation $\delta \theta_0$. If it is recognized that in (27) all the terms, starting with $n=1$, are larger than zero, then we arrive at the conclusion that the IS are more stable than the homogeneous state. This is connected with the stabilizing influence of the regions of the smooth distributions, for which $\lambda_n > 0$. Moreover, inasmuch as for broad layers and radially symmetrical distributions of large radius we have $|\lambda_0| \ll 1$, the condition (30) can be satisfied for them even at $\alpha \ll 1$. Thus, even if (5) is satisfied, stable homogeneous states can be excited in the system by finite external perturbations. This conclusion is confirmed by computer experiments.^{31,32}

It follows from an analysis of (20) and (26) that any IS region to which a single well V_θ of arbitrary shape corresponds is unstable if more than one of the eigenvalues in the spectrum of V_θ is negative. The only exceptions are those IS regions for which $\lambda_n > -\varepsilon$. In the analysis of their stability it is necessary to take into account the $\delta \eta_k$ with $k > 0$, which are not taken into account in the derivation of (20) and (29).

6.3. We consider now the stability of a single layer, i. e., of a distribution that is inhomogeneous along the x axis, in a three-dimensional system that is small only along the x axis ($l_x < L$). Linearizing Eqs. (1) and (2) with respect to

$$\begin{aligned} \delta \theta_{n_1, n_2, n_3} &= \delta \theta_{n_1}(x) \cos(k_{n_2} y) \cos(k_{n_3} z) e^{-\eta}, \\ \delta \eta_{k_1, k_2, k_3} &= \delta \eta_{k_1}(x) \cos(k_{k_2} y) \cos(k_{k_3} z) e^{-\eta}, \\ k_{n_2} &= \pi n_2 / l_y, \quad k_{n_3} = \pi n_3 / l_z, \end{aligned}$$

we arrive at the system (16) and (17), in which the operator Δ is replaced by $d^2/dx^2 - k_{n_2}^2 - k_{n_3}^2$. As a result, the eigenvalues of the operators \hat{H}_θ and \hat{H}_η take on the form

$$\lambda_{n_1, n_2, n_3} = \lambda_{n_1} + k_{n_2}^2 + k_{n_3}^2, \quad \mu_{k_1, k_2, k_3} = \mu_{k_1} + k_{k_2}^2 + k_{k_3}^2, \quad k_{n, m}^2 = k_n^2 + k_m^2,$$

where λ_{n_1} and μ_{k_1} are the eigenvalues for the one-dimensional case.

When the dimension of the system in the direction of the x axis is small ($l_x < L$) then, as in Sec. 6.2 above, all the inhomogeneous perturbations $\delta \eta_{k_1}(x)$, starting with $l_1 \geq 1$, can be neglected. We then arrive likewise at (20) and (21), in which λ_n is replaced by $\lambda_{n_1} + k^2$, and

$$P_{0n, n_1} = \frac{a_{n_1}(\mu_0 + k^2 e^{-2})}{\mu_0 + k^2 e^{-2} - \alpha^{-1} \gamma}, \quad a_{n_1} = -\frac{\langle q_n' \delta \theta_n \rangle \langle Q_n' \delta \theta_n \rangle}{\mu_0 + k^2 e^{-2}}. \quad (31)$$

The symbol $\langle \dots \rangle$ denotes averaging only along the x axis. At fixed k , there can be only one negative eigen-

value $\lambda_0 + k^2$, and therefore the stability condition reduces to (28) with the corresponding λ_n and a_n (31). Since the functions $\delta \theta_{n_1}(x)$ with $n_1 > 1$ oscillate and are smeared out over the entire sample along the x axis, their contribution to the sum of the condition (28) turns out to be negligible.^{16,17} This is all the more valid in the case of a broad layer, for which, as established earlier, $|\lambda_0| \ll \lambda_1$. Taking this remark into account in (31), we can rewrite the stability criterion (28) for $\lambda_0 + k^2 < 0$ in the form

$$1 - \langle q_n' \delta \theta_n \rangle \langle Q_n' \delta \theta_n \rangle (\mu_0 + k^2 e^{-2})^{-1} (\lambda_0 + k^2)^{-1} < 0. \quad (32)$$

According to (32), the stability is violated with respect to fluctuations with k directed perpendicular to the x axis and close in absolute value to

$$k_0 = e^{1/2} [\lambda_0 \mu_0 - \langle q_n' \delta \theta_n \rangle \langle Q_n' \delta \theta_n \rangle]^{1/2}, \quad (33)$$

when

$$\lambda_0 < -\{e^{1/2} \mu_0 + 2e[\lambda_0 \mu_0 - \langle q_n' \delta \theta_n \rangle \langle Q_n' \delta \theta_n \rangle]^{1/2}\}. \quad (34)$$

Since $\lambda_0 \approx \langle q_n' \delta \theta_n^2 \rangle$, and $\mu_0 \approx \langle Q_n' \delta \eta_n^2 \rangle$, the instability criterion (34) can in fact be obtained from (8), by using it for a structure region with dimension $\delta x \approx l$. In the region of rapidly varying distribution we have $q' < 0$, and this region determines the stability of the IS. A broad layer corresponds to $\lambda_0 \approx -\varepsilon l_x / L$ (25), and therefore, according to (34), it is stable in samples with $l_x < L$ and with arbitrary values of l_y and l_z . This conclusion explains the results of the computer experiment.^{31,32} A narrow layer, on the other hand, corresponds to $\lambda_0 \approx -1$, and it is unstable to fluctuations $\delta \theta(y, z)$ that tend to break up the distribution along the axes y and z .

In a sample of radius $R_1 < L$, a distribution in the form of a single drop (or spot) (see Sec. 4) of large radius $\rho_1 \ll l$ corresponds to a potential well V_θ with a spectrum that contains besides λ_0 (25) also other $\lambda_j^0 < 0$. The latter correspond to fluctuations that depend on the angles φ and α :

$$\delta \theta_j^0 = \delta \theta_j^0(\rho) P_j^m(\cos \alpha) e^{im\varphi}.$$

Linearizing Eqs. (1) and (2) with respect to the fluctuations $\delta \theta$ and $\delta \eta$ in a spherical coordinate system, we arrive at (16) and (17), in which

$$\hat{H}_\eta = \hat{H}_\eta^j = -\frac{d^2}{d\rho^2} - \frac{2}{\rho} \frac{d}{d\rho} + V_\eta + j(1+j)\rho^{-2}, \quad (35)$$

$$\hat{H}_\theta = \hat{H}_\theta^j = -\frac{d^2}{d\rho^2} - \frac{2}{\rho} \frac{d}{d\rho} + V_\theta + j(1+j)\rho^{-2}. \quad (36)$$

The principal eigenvalues of the operators \hat{H}_η^j and \hat{H}_θ^j corresponding to $j=0$, i. e., the eigenfluctuations $\delta \eta_0^0$ and $\delta \theta_0^0$, are equal respectively to μ_0 and λ_0 (25). Recognizing that $\delta \theta_j^0(\rho)$ have no nodes with respect to ρ and are localized in a region of the order of unity (l) of the well V_θ , we obtain, in analogy with the derivation of (23),

$$\lambda_j^0 = \lambda_0 + j(1+j)(l/\rho_1)^2, \quad \mu_j^0 \approx \mu_0 + j(1+j)e^{-2}(l/\rho_1)^2. \quad (37)$$

Carrying out the transformation in the same approximations as in the derivation of (32), we find that the stability condition for the considered case reduces to (32), in which the spectra of a single layer $\mu_0 + k^2 e^{-2}$ and $\lambda_0 + k^2$ are replaced by μ_j^0 and λ_j^0 . It follows from

this criterion that the most critical are the fluctuations with $j_0 \approx k_0 \rho_1$, and the drop is unstable if the condition (34) is satisfied. This generalization of the criterion (34) is natural, since the considered case is in fact equivalent to an analysis of the stability of the single layer with $l_j = 2\pi \rho_1$. Inasmuch as for a large drop ($L \gg \rho_1 \gg l$) we have $\lambda_0 \approx -R_1/L$ (25), it is stable according to (34), at least at $R_1 < L$. It follows also from this that a broad spherical layer (Fig. 5) is stable to radially non-symmetrical fluctuations.

On the other hand, a narrow layer is unstable, since in its case $\lambda_0^0 \approx -1$, and in accordance with (37) there are many $\lambda_j^0 \approx -1$. A drop (or spot) of small size ($\rho_1 \approx l$) corresponds to $\lambda_0^0 \approx -1$ (25). The next values of λ_j^0 , according to (37), are increased by an amount $\approx j(1+j)$, and can therefore be positive. The damped character of the fluctuations $\delta\theta_j^0$ with $j > 0$ is obvious and is connected with the strong diffusion spreading of the inhomogeneous changes in the region of the drop of radius l . For the same reason, $\lambda_1 > 0$.

Thus, the drop or spot is stable to radially non-symmetrical fluctuations. The IS in systems (e.g., in the Turing model) that have only one stable branch on the zero-isocline $q=0$ (curve 2, Fig. 1) cannot contain regions in the form of broad layers or drops with $\rho \gg l$. Therefore the stable IS in them take the form of one or several drops (spots) of small radius. On the other hand, stability with respect to a radially symmetrical fluctuation $\delta\theta_0^2$, which corresponds to $\lambda_0 \approx -1$, is given by the condition (28). The latter is satisfied at approximately the same external parameters A, B, \dots , at which a narrow layer is stable in the one-dimensional case.¹⁶ Generalizing these results, we can state that any IS region that corresponds in any of the cross sections to a narrow layer is unstable if in one of the directions the region of the rapidly varying distribution greatly exceeds l , inasmuch as this region contains, besides $\lambda_0 \approx -1$, also other $\lambda_n < -(\approx -1)$.

6.4 The IS corresponding to single wells V_0 in which only $\lambda_0 < 0$ and which are separated by sufficiently large distances, are stable. We shall illustrate this using as an example a spatially periodic IS with period R_0 . At $R_0 \gg 1(l)$, the proper wave function $\delta\theta_0(\mathbf{r} - \mathbf{r}_i)$ of an isolated i th well overlaps weakly its neighbor, and therefore the level λ_0 of this well splits into a band:

$$\lambda_0^* = \lambda_0 + \Delta\lambda_0 + 2\Delta\lambda \sum_{j=1}^3 \cos(k_n R_0),$$

whose width is

$$\Delta\lambda \propto \exp(-R_0); \quad \Delta\lambda_0 \propto \exp(-R_0); \quad k_{n_j} = 2\pi n_j / R_0 N_j;$$

$$k_n^2 = k_{n_1}^2 + k_{n_2}^2 + k_{n_3}^2; \quad n_j = 0, \pm 1, \pm 2, \dots, \pm(N_j - 1)/2, \quad N_j/2;$$

N_1, N_2, N_3 is the number of wells (drops) in the considered IS, and the proper wave function $\delta\theta_n$ of the problem can be represented as a superposition of $\delta\theta_0(\mathbf{r} - \mathbf{r}_i)$.

When $R_0 \leq L$, the function $\eta(\mathbf{r})$, and consequently also the potential $V_n(\mathbf{r})$ changes little, with the exception of narrow regions (of the order of l). Therefore the potential V_n can be regarded as constant, and

$$\mu_i = \langle Q_n' \delta\eta_0^2 \rangle + k_i^2 e^{-2}, \quad \delta\eta_n \propto \exp(ik_i r). \quad (38)$$

Substituting the wave functions $\delta\theta_n$ and $\delta\eta_i$ and the eigenvalues λ_0 and μ_i in (19), we obtain from (18), after suitable transformation,¹⁶ the stability criterion

$$\lambda_0 + \Delta\lambda_0 + 2\Delta\lambda \sum_{j=1}^3 \cos(k_n R_0) + a_0 \mu_0 (\mu_0 + k_n^2 e^{-2})^{-1} > 0, \quad (39)$$

which coincides for the one-dimensional case with the criterion that follows from Eq. (51) of Ref. 16.

The condition (39) follows in fact from (32) if $\lambda_0 + k^2$ in the latter is replaced by

$$\lambda_0 + \Delta\lambda_0 + 2\Delta\lambda \sum_{j=1}^3 \cos(k_n R_0).$$

In the derivation of (30) it was noted that $a_n \geq 0$ and $a_0 \approx 1$.^{16,17} Therefore, when

$$\max k_n^2 e^{-2} = 3\pi^2 (L/R_0)^2 \leq 1.$$

The condition (39) is satisfied even in the case of small drops, for which $\lambda_0 \approx -1$, let alone drops of large radius, which are stable at short distances from one another, inasmuch as $\lambda_0 > -\epsilon$ for these drops. The most dangerous are the fluctuations $\delta\theta_n$ that tend to increase the size of the drop or the value of θ in the drop at the expense of decreasing these parameters in the neighboring drops. Therefore a distribution in the form of several drops becomes unstable, without reaching the points where $d\eta/dA = \infty$, and the more drops the less stable they are to supercooling or superheating. In a heated system, R_0 cannot be too large. With increasing R_0 , the region of slowly varying distribution between drops comes ever closer to a homogeneous unstable state, and when θ lands in the interval (θ_1^0, θ_2^0) (Fig. 1), the IS becomes unstable. On the contrary, in a supercooled or superheated system, the drops can be arbitrarily far from one another, inasmuch as the homogeneous state is stable.

We note that the larger the differences between θ in the drop and outside the drop, the closer the small-radius drops can be located to one another. The reason is that with increasing contrast of the drop the potential V_n becomes steeper¹⁶ and acquires the shape of an ever deeper potential well. In such a well V_n , the eigenfunction of the ground state $\delta\eta_0(\mathbf{r} - \mathbf{r}_i)$ turns out to be localized and therefore produces practically independent damping of the growing fluctuation $\delta\theta_0(\mathbf{r} - \mathbf{r}_i)$ in each of the wells V_0 . The described situation is easier to realize at not too small values of ϵ , as is confirmed also by computer experiments.^{31,32}

Thus, a periodic IS is stable if the fragments of which it is made up satisfy the conditions formulated in Secs. 6.2 and 6.3, and the shortest-wavelength anti-binding combination of the growing fluctuations $\delta\theta_0(\mathbf{r} - \mathbf{r}_i)$, which describe the "transfer" of θ between neighboring fragments, is followed-up by the damping change $\delta\eta_{N_j/2}$ of η . This conclusion pertains also to SIS, which should be more stable to transfer of θ between neighboring different fragments. In fact, since the SIS correspond to different wells V_{θ_i} the distances between which are much larger than l (Sec. 6.1), it follows that not only λ_{n_i} corresponding to them are changed little by allowance for their overlap. In addition, in the SIS a small-drop radius can be located closer to the wall that

separates the superheated and supercooled regions of the system than to another small-radius drop. The reason is that the growing fluctuation $\delta\theta_{0_i}$ corresponding to such a wall is easier to attenuate by a smaller change $\delta\eta$ than the fluctuation $\delta\theta_{0_i}$ corresponding to a small-radius drop (Sec. 6.2).

When the system parameters A, B, \dots are changed, the SIS becomes unstable in one of its fragments either because the conditions formulated in Secs. 6.2 and 6.3 for the stability of this fragment are violated, or else as a result of transfer of θ between some neighboring fragments. These fragments are less stable the closer the locations of the potentials V_{θ_i} corresponding to them, and the fragments become closer in form to a small-radius drop.

6.5. To investigate the stability of traveling IS (Sec. 5), we linearize Eqs. (1) and (2) with respect to small deviations

$$\delta\theta(x, t) = \delta\theta(x)e^{-t}, \quad \delta\eta(x, t) = \delta\eta(x)e^{-t}$$

from the stationary self-similar solution of Eqs. (14). As a result we obtain the system (16) and (17), in which

$$\hat{H}_\theta = -\frac{d^2}{dx^2} - v\frac{d}{dx} + V_\theta; \quad \hat{H}_\eta = -\frac{d^2}{dx^2} - v\beta\frac{d}{dx} + V_\eta. \quad (40)$$

Here v is measured in units of l/τ_θ . The problem of finding the spectra λ_n and $\tilde{\mu}_l$ and the eigenfunctions $\delta\theta_n$ and $\delta\eta_l$ of these operators under the considered boundary conditions is self-adjoint.²⁷ Therefore an investigation of the stability of traveling IS is in fact similar to the investigation described above. The spectrum γ satisfies the condition (18).

In the case of traveling IS, to study the spectra of the operators \hat{H}_θ and \hat{H}_η , one can use also a quantum-mechanical analogy, by rewriting the corresponding equations in the normal Liouville form. It is then easy to verify that the spectra λ_n and $\tilde{\mu}_l$ of the traveling IS have the same singularities as those of the static IS, so that the results on the stability of the static one-dimensional IS remain qualitatively valid also for traveling IS.

7. CONCLUSION

7.1 In a one-dimensional system, under the same conditions (parameters A, B, \dots) there exist a whole set of different elementary distributions [fragments, Fig. 2(a)], on the boundaries of which $\eta'_x = \theta'_x = 0$, and the values η and θ and of their second derivatives at one of the boundaries coincide (accurate to ε), but can have jointly different values (Fig. 3). As a result of this, not only different periodic structures, but also different stochastically inhomogeneous structures (SIS) can arise in the system, in the sense that different such fragments can be randomly disposed over the length of the system.

Each elementary fragment satisfies the integral relations (11) and the stability condition. Thus, in a supercooled system (Fig. 1), IS are possible in the form of different single hot narrow or broad layers, as well as their combinations, which can go over into a homogeneous stable state on some bounded segment.

In the case of "two-phase equilibrium" (case d, Fig. 1), the SIS can contain simultaneously sections corresponding to broad and narrow layers that are hot as well as cold. Sections of the IS in the form of one or several layers can then go over on the periphery into one of the stable homogeneous states. It is possible to excite SIS in such a system only by finite inhomogeneous perturbations.

The homogeneous state of the heated system (Fig. 1) is unstable. Therefore SIS are spontaneously produced in them and acquire nowhere a homogeneous distribution; as an alternative, homogeneous oscillation can arise at $\tau_0 \ll \tau_n$.^{6-9,14} In the latter case, nevertheless, it is possible to excite stable SIS (Sec. 6.2). If an SIS is excited on some section of the system by inhomogeneous excitation, then the structure either vanishes or leads to the appearance of an SIS in the entire system. Such a self-adjustment of the IS was observed both in experiment^{3,12} and in a computer analysis.³²

7.2 In two- and three-dimensional systems of sufficiently large size, one-dimensional distributions in the form of broad layers are stable (Sec. 6.3). There can also exist in them IS that are radically symmetrical or nearly so, in the form of different sets of spherical or cylindrical broad layers [Fig. 5(a)]. The latter were observed in two-dimensional systems.^{3,12} Narrow layers, on the other hand, are unstable to breakup into smaller regions.

7.3. In the general case, two- and three-dimensional IS can have an exceedingly complicated form, as confirmed also by numerical investigations.³² From an analysis of their stability (Sec. 6) it follows that the regions of smooth variation of $\eta(\mathbf{r})$ and $\theta(\mathbf{r})$ correspond to the supercooled or superheated phases of the system. The heated unstable state is located only in narrow (with dimension of the order of l) surface layers between the regions of the smooth changes of $\eta(\mathbf{r})$ and $\theta(\mathbf{r})$. Any SIS fragment containing a heated-phase region surrounded on all sides by a region of one of the stable phases is unstable if the dimension of the heated region exceeds greatly l in at least one of the sections. Heated regions close in shape to the hot or cold small-size drop (Sec. 6.3) and located at distances substantially larger than l from one another and from the "heterophase" surface layer, can be stable (Sec. 6.4).

In any section, the SIS is similar to one of the stable distributions for the one-dimensional case. However, owing to the different curvatures of the neighboring elements (Sec. 4) and to the greater possibility of satisfying the integral conditions, these distributions can be even more varied than in the one-dimensional case.

7.4. The IS of the type given here is stable only in a definite range of variation of the parameters A, B, \dots , and becomes unstable on the boundary of this range. The development of the instability varies in character in periodic IS and SIS. For the former, a simultaneous restructuring of the entire IS, leading to a change in the number of layers,^{16,17} drops, or spots present in it, is more probable. In the SIS, stability is lost in one or several less stable fragments (Sec. 6), in which in-

homogeneous oscillations ("local swirling") can arise, or else the unstable fragments act in a certain sense as leading centers^{8,12} from which the restructuring of the entire SIS begins.

7.5. The spontaneous appearance of stable IS when the parameters of the system are changed confirms the idea advanced in Refs. 6 and 7, that self-organization is possible in nonequilibrium homogeneous systems. A true self-organization will occur in systems in which at least one of the parameters (A) varies autonomously during the course of the nonequilibrium process. As a result, IS of more and more complicated types will spontaneously occur in the system.

7.6. The form of the SIS for given parameters A, B, \dots is restricted by integral relations, by the character of the zero-isoclines, and by the stability conditions. As a result, if an inhomogeneous fragment is produced in some region of the heated system, this perturbation will either attenuate or else, as a result of self-adjustment to this fragment, one of the SIS containing a region close to the given fragment will be reconstructed. In this sense, the systems considered have associative memory: specifications of the parameters A, B, \dots means storage of an entire set of definite patterns, which can be reconstructed with certain probability by means of one of their fragments.

7.7. In the considered dissipative systems it is possible to excite various SIS traveling with different velocities without damping: in a supercooled system, for example, in the form of a single stable or several widely spaced (including also different ones) hot layers; in a superheated system—in the form of cold layers; in a heated system—in the form of a traveling sequence of layers with distances between them not greatly exceeding L . The higher the speed of the IS, the more distorted they are compared with the static ones (Sec. 5). Traveling IS can be excited also at $l \gg L$, but $\tau_\theta \ll \tau_n$, when there are no static IS. In this case the results of the present paper go over into those obtained in the study of the propagation of a pulse in a nerve fiber.¹¹

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¹⁾ In particular cases, ^{16, 17} certain functions of $\eta(\mathbf{r})$ and $\theta(\mathbf{r})$ can precede γ . However, when they are positive, this is of no importance, since these equations can be reduced to the form (16) and (17) by transformation into the normal Liouville form.²⁷

²⁾ The stability of single strings and domains in semiconductors with non-single-valued current-voltage characteristics is connected with the equations for the external circuit,^{4, 5} the role of which in the systems considered here is played by Eq. (2) for each region of size $\leq L$.

³⁾ This conclusion pertains also to the system (16) and (17), i.e., $\gamma = 0$ corresponds to the fluctuations $\delta\theta \sim \mathbf{n} \cdot \nabla\theta(\mathbf{r})$ and $\delta\eta \sim \mathbf{n} \cdot \nabla\eta(\mathbf{r})$.

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