# Critical phenomena in media with breeding, decay, and diffusion

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This review is devoted to critical phenomena such as the threshold of explosive instability and kinetic transitions of the "medium populating" type in nonequilibrium systems with breeding, decay, and diffusion. A detailed analysis is made of the situation where breeding is localized within particular spatial regions (breeding centers) which arise randomly in the medium at arbitrary times and have finite lifetimes. The analogy with problems in percolation theory and second-order equilibrium phase transitions is discussed. The effect of fluctuations in external fields on competition processes in media with diffusion is examined. Diffusion in a medium with randomly distributed traps is investigated and particular attention is devoted to the contribution of statistically rare spatial configurations.

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# **1. INTRODUCTION**

There has been increasing interest recently in processes occuring in highly nonequilibrium flux-type open systems because such systems have been found to exhibit effects of *self-organization*, i.e., the formation of dissipative macroscopic structures.

Flux-type systems are coupled to an external source of energy or are connected to two thermostats at different temperatures. The result of this is that relaxation to thermal equilibrium cannot take place in such systems. Even in the steady state, a flux of energy passes through the system and is dissipated into the ambient medium.

When the energy flux is small, deviations from thermal equilibrium are also small. These deviations increase with increasing energy flux, and the system exhibits a sequence of instabilities leading to qualitative modifications of the kinetic state established in the system. Each such modification constitutes a kinetic transition<sup>1)</sup> accompanied by the establishment or complication of order in the open and highly nonequilibrium system, i.e., a reduction in its symmetry. By analogy with the corresponding phenomena in equilibrium systems, kinetic transitions. The study of these processes and of the attendant ordered structures is the concern of the modern theory of self-organization.

<sup>1)</sup>The phrase "kinetic transition" was introduced in Refs. 3 and 4.

There is a close connection between self-organization and the initiation of turbulence. L. D. Landau<sup>1</sup> was the first to point out that the transition from laminar to turbulent flow may occupy a definite range of values of the parameter characterizing the rate of supply of energy. Within this range, the flow has a regular (periodic or quasiperiodic in time) structure which is established as a result of a nonlinear limitation on the growth of the amplitudes of the first unstable modes. True turbulence, requiring statistical description, arises in the course of a successive complication of regular structures, and the attainment of exponential instability of the evolving structure against arbitrarily small perturbations.<sup>2</sup> This type of instability does not set in for any quasiperiodic state, so that the Landau mechanism cannot be used to examine the transition to the long-term turbulent state.<sup>2)</sup> However, Landau's ideas form the foundation for the approach to self-organization effects that is being developed today.

From the general point of view, self-organization is a property of open flux-type systems during the preturbulent stage, when the quasiequilibrium (laminar) state has already lost its stability, but macroscopic stochastization (transition to true turbulence) has not yet been attained. In this connection, we note the analogy between the description of the ini-

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<sup>&</sup>lt;sup>2)</sup>Substantial advances in the understanding of the properties of such a transition have been made only recently in connection with the study of strange attractors.<sup>2,6,10</sup>

tiation of turbulence in the Landau mechanism and the Landau theory of second-order equilibrium phase transitions. A detailed discussion of this analogy is given by Haken.<sup>5</sup>

The most important example of self-organization among physical systems is laser generation. The transition from noise to coherent generation as the strength of the pump increases can be interpreted as a kinetic (or nonequilibrium phase) transition.<sup>5</sup> In hydrodynamics, self-organization effects include the evolution of the regular structure of convective flows in Bénard's problem,<sup>6</sup> Taylor vortices,<sup>7</sup> and so on. Systems in which chemical reactions take place exhibit a very wide range of self-organization phenomena.<sup>8</sup> Selfwave structures involved in the Belousov-Zhabotinskiĭ reaction have been investigated in greatest detail.<sup>9</sup>

The basic mathematical models of self-organization are common to a wide class of physical, chemicl, and biological systems. They are studied by using the methods of modern theoretical physics, and are sometimes elevated to the status of an independent subject for which Haken has coined the name "synergetics" (from the Greek synergeia—working together). Synergetics is concerned with the study of critical phenomena (i.e., instabilities and kinetic transitions), nonlinear waves, and stationary structures, as well as self-stochasticity involved in cooperative behavior of highly nonequilibrium systems of various origins.

In this review, we shall examine critical phenomena such as the explosive instability and the "medium populating" kinetic transition in nonequilibrium systems with breeding, decay, and diffusion. We shall illustrate our discussion by simple models which are particularly important because of their generality. We shall consider the situation in which breeding (described by the scheme  $X + A \rightarrow 2X$ ) is localized in particular spatial regions (breeding centers) that appear randomly and independently in the medium at arbitrary times and have the same intensity, shape, and lifetime. This occurs in parallel with the homogeneous process of decay of the breeding material in the medium (the equivalent of this process is the capture of reactive particles by traps distributed within the medium).

We shall not specify the systems studied in this review to any greater extent. Breeding effects are observed in nuclear fission reactions,<sup>11</sup> in chemical chain reactions,<sup>12</sup> in autocatalysis,<sup>9</sup> in the behavior of biological materials,<sup>13</sup> and so on. Each particular case is characterized by its own specificity. However, it seems to us that it is important to concentrate our attention on typical general properties of such systems.

Breeding in a medium may be accompanied by the development of explosive instability, i.e., an unlimited rise in the amount of breeding material in the system. The threshold for explosive instability is determined by competition between breeding and decay.

When the rate of breeding or decay fluctuates, and the breeding material is capable of diffusion, the determination of the threshold for explosive instability becomes a complicated problem that is quite close to the range of problems investigated in the theory of disordered media and in percolation theory.<sup>14-16</sup>

Actually, it has been pointed out<sup>17</sup> that the transition across the explosive instability threshold may be looked upon as dynamic percolation in which there is a change in the asymptotic behavior (in time) of the mean density  $\bar{n}(t)$  of the breeding material as  $t \rightarrow \infty$ . This type of behavior is possible in the discrete "directed percolation" models that have recently been proposed.<sup>18</sup> They are discussed in Section 2.2.

Explosive instability can be suppressed by post-threshold nonlinear limiting mechanisms. A nonzero density of the breeding material is then established above the threshold in the medium, i.e., the medium becomes "populated" by this material.

The system that we shall examine is an open system that is not in equilibrium because of decay and the influx of energy into the breeding centers from outside. We are therefore entitled to interpret the population of the medium above the explosive instability threshold as a kinetic transition in the system. Analysis (see Section 2.5) shows that, in many respects (critical slowing down, correlation radius tending to infinity), this effect resembles a second-order phase transition in equilibrium systems. At the same time, fluctuations due to external agencies result in very unusual behavior.

Apart from critical phenomena in media with random breeding centers, we shall also discuss (in Section 3) competition in fluctuating media. Competition effects are an important feature of self-organizing systems. In fact, any spontaneously established macroscopic regular structure is a consequence of competition between growing unstable modes; the "surviving" mode suppresses other modes and imposes its own structure on the system. Competition effects play a special role in the evolution of biological and chemical systems.

Diffusive random walks in a medium with randomly distributed traps are discussed in Section 4.

# 2. SYSTEMS WITH RANDOM BREEDING AND DECAY

In the simplest case, such a system is described by the stochastic differential equation

$$n = -\alpha n + f(t) n, \qquad (2.1)$$

where  $\alpha$  is the constant decay rate and f(t) is the rate of breeding with given statistical characteristics, which varies randomly with time.

When the mean density  $\langle n(t) \rangle$  evaluated over the ensemble of realizations increases with time without limit, we shall say that the explosive instability threshold has been exceeded for the system (2.1). The density  $\langle n(t) \rangle$  can be calculated by direct integration of (2.1):

$$\langle n(t) \rangle = n_0 e^{-\alpha t} \left\langle \exp\left(\int_0^t f(t') dt'\right) \right\rangle.$$
 (2.2)

The last factor in this expression is directly related to an important characteristic of the random process f(t), namely, its generating functional

$$\Phi \left[ \theta \left( t \right) \right] = \left\langle \exp \left( \int_{0}^{t} f \left( t' \right) \theta \left( t' \right) dt' \right) \right\rangle.$$
(2.3)

Comparison of (2.3) with (2.2) shows that

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$$\langle n(t) \rangle = n_0 e^{-\alpha t} \Phi[1]. \tag{2.4}$$

The formula given by (2.4) is the solution of the problem of explosive instability threshold for an arbitrary randomly varying rate of breeding. For example, let us suppose that the breeding process is the superposition of independent "bursts" of length  $\tau_0$  and intensity J. The "bursts" are produced at random times:

$$f(t) = \sum_{j} J\sigma(t - t_{j}), \qquad (2.5)$$

where  $\sigma(\tau) = 1$  for  $0 < \tau < \tau_0$  and  $\sigma(\tau) = 0$  for  $\tau < 0$  and  $\tau > \tau_0$ .

If *m* is the mean numbe of "bursts" per unit time, the generating functional of the Poisson random process (2.5) has the form<sup>19</sup>

 $\Phi[\theta(t)]$ 

$$= \exp\left\{-m\int_{0}^{t} \left[1 - \exp\left(J\int_{0}^{t'} \sigma(t'-t'') \theta(t'') dt''\right)\right] dt'\right\}.$$
(2.6)

Substituting  $\theta(t) \equiv 1$  in (2.6), we find  $\Phi$  (Ref. 1) and hence the law whereby the density averaged over the ensemble varies with time:

$$\langle n(t)\rangle = n_0 \exp\left[\left(m\left(e^{J\tau_0}-1\right)-\alpha\right)t\right]. \tag{2.7}$$

The explosive instability threshold is therefore reached when the mean number of "bursts" per unit time is given by

$$m_{\rm crit} = \alpha \, (e^{J\tau_0} - 1)^{-1}. \tag{2.8}$$

When  $J\tau_0 \ll 1$ , the breeding "bursts" are weak and, as is seen from (2.8), we then have  $m_{\rm crit} = \alpha/J\tau_0$ . Such an expression for the threshold can be readily obtained by equating the mean breeding rate  $\bar{f} = mJ\tau_0$  to the decay rate  $\alpha$ .

On the other hand, in the opposite limiting case of strong "bursts,"  $(J\tau_0 \gtrsim 1)$ , we have  $m_{\rm crit} = \alpha e^{-J\tau_0}$ . This corresponds to equating the decay rate  $\alpha$  to the relative increase  $\exp(J\tau_0)$  in the amount of material per "burst," multiplied by the number *m* of "burst" per unit time.

A substantial part of our review is concerned with calculations of the explosive instability threshold for systems that generalize the model given by (2.1) to the case of distributed systems with diffusion.<sup>20-22</sup>

Let us suppose that the decay and breeding of a certain material are possible in the medium, and that the rate of decay  $\alpha$  is homogeneous in space and constant in time, whereas breeding is confined to certain specific breeding centers that arise randomly in time at random points in the medium, but have the same form, intensity, and lifetimes. The corresponding mathematical model is

$$n = -\alpha n + f(\mathbf{r}, t) n + D \Delta n, \qquad (2.9)$$

where n is the density of the material and D is its diffusion coefficient.

The fluctuating field  $f(\mathbf{r}, t)$  is the sum of identical pulses  $(\mathbf{r}_i, t_i)$  appearing at the random points  $g(\mathbf{r}, t)$ :

$$f(\mathbf{r}, t) = \sum_{j} g(\mathbf{r} - \mathbf{r}_{j}; t - t_{j}). \qquad (2.10)$$

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The mean number of pulses appearing per unit time per unit volume is constant and equal to m. The function  $g(\mathbf{r}, t)$  has the form

$$g(\mathbf{r}, t) = J\chi(\mathbf{r}) \sigma(t). \qquad (2.11)$$

The quantity J characterizes the intensity of the breeding center and the function  $\chi$  (**r**) falls rapidly to zero for  $r > r_0$ , so that  $r_0$  is the spatial dimension of an individual center;  $\chi$  (0) = 1. The lifetime of a breeding center is  $\tau_0$  and the function  $\sigma(t)$  is defined in (2.5).

In the ensuing analysis, we shall frequently use the dimensionless space-time concentration c of breeding centers, defined by

$$c = m r_0^d \tau_0, \tag{2.12}$$

where d is the dimensionality of the medium.

All breeding centers may be divided into strong and weak, depending on the relative increase in density at an individual center.

# 2.1. Strong and weak breeding centers

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The increase in density at an individual breeding center is described by

$$\dot{-n} = -D\Delta n - J\chi (\mathbf{r}) n, \quad 0 \leq t \leq \tau_0, \qquad (2.1.1)$$

which is formally identical with the Schrödinger equation with imaginary time and potential  $U = -J_{\chi}(\mathbf{r})$ . Its general solution is

$$n(\mathbf{r}, t) = \sum_{l} C_{l} e^{\lambda_{l} t} \varphi_{l}(\mathbf{r}) + \int C(\lambda) e^{\lambda t} \varphi_{\lambda}(\mathbf{r}) d\lambda, \quad (2.1.2)$$

where the sum is evaluated over the discrete and the integral over the continuous spectra of the linear operator

$$\hat{L} = D\Delta + J\chi (\mathbf{r}). \tag{2.1.3}$$

For breeding centers (J > 0), the eigenvalues  $\{\lambda_i\}$  in the discrete spectrum are positive. They correspond [see (2.1.1)] to negative energy levels of bound states in the "potential well"  $U(\mathbf{r}) = -J_{\chi}(\mathbf{r})$ .

Suppose that  $\lambda_0$  is the largest of the eigenvalues in the discrete spectrum. The breeding centers will be referred to as strong when  $\lambda_0 \tau_0 \ge 1$  and weak when  $\lambda_0 \tau_0 \le 1$ . It is clear from the general solution (2.1.2) that the increase in the amount of material at a strong center is exponentially large.<sup>3)</sup>

The eigenvalue  $\lambda_0$  can be related to the parameters Jand  $r_0$  characterizing the properties of individual centers. This can be done by exploiting the analogy with the Schrödinger equation and recalling that  $\lambda_0$  corresponds to the lowest-lying level in the potential well  $U = -J_{\chi}$ . It is well known<sup>23</sup> that the order of magnitude of the lowest-lying level in a deep well  $(J \gg D / r_0^2)$  is given by  $\lambda_0 \sim J$ . In the opposite limiting case  $(J \gg D / r_0^2)$ , which corresponds to a shallow potential well, the estimate for  $\lambda_0$  will depend on the dimen-

<sup>&</sup>lt;sup>3)</sup>We note that, since  $\lambda_0 \tau_0$  appears in the argument of the exponential, the breeding centers are strong even for  $\lambda_0 \tau_0 \approx 3 - 4$ .

sionality d of the medium. For a one-dimensional medium

$$\lambda_0 \sim J\left(\frac{Jr_0}{D}\right), \quad d=1,$$
 (2.1.4)

and for a two-dimensional medium

$$\lambda_0 \sim \frac{D}{r_0^2} \exp\left(-\frac{\nu D}{Jr_0^2}\right), \ \nu \sim 1, \quad d=2, \qquad (2.1.5)$$

whereas, for a three-dimnsional medium, a shallow potential well of this kind will not contain any discrete levels, i.e., all the eigenvalues will be  $\lambda < 0$ .

Thus, short-lived  $(\tau_0 < r_0^2 / D)$  centers turn out to be weak if  $J < J^*$ , where

$$J^* = \tau_0^{-1}, \quad d = 1, \ 2, \ 3. \tag{2.1.6}$$

If, on the other hand, the centers are long-lived  $(\tau_0 > r_0^2/D)$ , they will be weak for  $J < J^*$  where

$$J^* = \sqrt{\frac{D}{r_0^2 \tau_0}}, \qquad d = 1, \qquad (2.1.7)$$

$$J^* = \frac{D}{r_0^2} \left( \ln \frac{D\tau_0}{r_0^2} \right)^{-1}, \quad d = 2,$$
 (2.1.8)

$$J^* = \frac{D}{r_0^*}, \qquad d = 3.$$
 (2.1.9)

In the opposite limiting case  $J > J^*$ , the breeding centers are strong.

# 2.2. Explosive instability threshold for strong centers

The total increase in the amount of material at an individual strong center during its life is

$$\Delta N_1 = n_0 r_1^d e^{\lambda_0 \tau_0}, \qquad (2.2.1)$$

where  $n_0$  is the initial density (constant in space) and  $r_1$  is the localization radius of the eigenfunction  $\varphi_0(r)$  of the operator (2.1.3) corresponding to the maximum positive eigenvalue<sup>4)</sup>

$$r_{i} \equiv \left(\int \varphi_{0}\left(\mathbf{r}\right) \,\mathrm{d}\mathbf{r}\right)^{^{\prime / d}}, \qquad \int \varphi_{0}^{s}\left(\mathbf{r}\right) \,\mathrm{d}\mathbf{r} = 1. \tag{2.2.2}$$

The explosive instability threshold is determined from the condition that the rate of increase in the amount of material at the breeding centers must be equal to its rate of decay in a unit volume. If we neglect interactions between centers, this may be written in the form  $m \Delta N_1 = \alpha n_0$ , so that the critical concentration of centers is given by the following simple expression:

$$c_{\text{crit}}^{(0)} = \alpha \tau_0 \left( \frac{r_{\odot}}{r_1} \right) e^{-\lambda_0 \tau_0}. \qquad (2.2.3)$$

This expression does not take correlation effects into account. In reality, when two centers happen to be situated

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sufficiently closely to one another both in space and in time, then in addition to the independent individual increase in the amount of material at the two centers a pairwise increment arises due to the fact that the exponential increase in density on the second center does not begin from the average density  $\bar{n}$  evaluated over space at the particular instant of time, but begins from the higher value (density spot) remaining after the previous first center. There are also other possible contributions due to clusters of three, four or more centers.

For clusters consisting of two breeding centers that appear at points  $\mathbf{r}_1$  and  $\mathbf{r}_2$  at times  $t_1$  and  $t_2$ , the additional paired increase in density is

$$\Delta N_{\mathbf{i},2} (\mathbf{r}_2 - \mathbf{r}_{\mathbf{i}}, t_2 - t_{\mathbf{i}})$$
  
=  $n_0 r_1^d e^{2\lambda_0 \tau_0} \int \varphi_0 (\mathbf{r} - \mathbf{r}_2) \varphi_0 (\mathbf{r}' - \mathbf{r}_{\mathbf{i}}) G_d$   
×  $(\mathbf{r} - \mathbf{r}', t_2 - t_1 - \tau_0) d\mathbf{r} d\mathbf{r}',$  (2.2.4)

where  $G_d$  is the Green's function for the diffusion problem in space of dimensionality d:

$$G_d = (4\pi D\tau)^{-d/2} \exp\left(-\frac{\rho^3}{4D\tau}\right). \qquad (2.2.5)$$

It was assumed in the derivation of (2.2.4) that the two centers did not overlap along the time axis, i.e.,  $t_2 > t_1 + \tau_0$ .

The average contribution due to the additional increase in the amount of material on two-center clusters per unit time per unit volume is

$$\overline{\Delta N}_{\mathbf{i},2} = \frac{m}{2} \int \Delta N_{\mathbf{i},2}(\boldsymbol{\rho}, \tau) p(\boldsymbol{\rho}, \tau) \, \mathrm{d}\boldsymbol{\rho} \, \mathrm{d}\tau, \qquad (2.2.6)$$

where  $p(\rho, \tau)$  is the probability density that the second center will appear at a distance  $\rho$  after time  $\tau$  following the appearance of the first. It is well known<sup>14</sup> that, for independently appearing centers, the distribution is

$$p_{i}(\boldsymbol{\rho}, \tau) = m \exp\left(-mV_{d}\tau\right), \qquad (2.2.7)$$

where  $V_d$  is the volume of a sphere of radius  $\rho$  in a space of dimensionality *d*. The quantity  $\overline{\Delta N}_{1,2}$  must be compared with the mean increase at the individual centers.

To calculate the mean increase  $\overline{\Delta N}_{1,2}$ , we must, in general, know the explicit form of the eigenfunction  $\varphi_0(\mathbf{r})$  which, in turn, is determined by the specific shape of the breeding centers. However, in the first instance, we are interested in the limit of sufficiently low density of breeding centers, so that the centers in a two-center cluster are well separated in time. In that case, we may suppose that, during the diffusive spreading after the operation of the first center, this center leaves behind it a density spot that propagates to points at distances much greater than the initial radus  $r_1$ . When the additional paired increase is calculated, we can neglect the inhomogeneity of density in the region in which the second center subsequently appears, so that the expression for  $\overline{\Delta N}_{1,2}$  becomes much simpler:

$$\overline{\Delta N}_{1,2} = \frac{n_0}{2} m^2 r_1^{2d} e^{2\lambda_0 \tau_0} \int G_d(\rho, \tau) e^{-mV_d(\rho)\tau} d\rho \,\mathrm{d}\tau. \quad (2.2.8)$$

By evaluating this integral for media of different dimension-

<sup>&</sup>lt;sup>4)</sup>The length  $r_1$  can be estimated as follows:  $r_i \simeq r_0$  for  $J > D/r_0^2$  and  $r_i = \sqrt{D/\lambda_0}$ .

ality, we can readily show that the order of magnitude of  $\overline{\Delta N}_{1,2}$  is given by

$$\overline{\Delta N}_{1,2} \sim n_0 r_1^{2d} m e^{2\lambda_0 \tau_0} \left(\frac{m}{D}\right)^{d/(d+2)}.$$
 (2.2.9)

Thus, the additional mean increase on two-center clusters is small in comparison with the mean increase on individual centers, provided

$$\left(\frac{mr_1^{d+2}}{D}\right)^{d/(d+2)}e^{\lambda_0\tau_0}\ll 1.$$
 (2.2.10)

This condition can also readily be derived from other, simpler considerations. It is clear that mutual influence of breeding centers can be neglected if the volume v of the space-time region occupied by an enhanced-density spot due to an individual center is much smaller than the volume occupied, on average, by one center:

 $mv \ll 1.$  (2.2.11)

Let us estimate the volume v. This is approximately equal to the product of the time  $\tau^*$  necessary for the increase in density to become appreciable against the average background, by the volume of the region in space occupied by the increase in n(r, t) during this time. The characteristic volume occupied by the increase in density up to the end of the time  $\tau^*$  is determined by the diffusion length during this time, i.e., the quantity  $(D\tau^*)^{1/2}$ . In view of this, let us rewrite (2.2.11) in the form

$$m \ll \frac{1}{\tau^*} (D\tau^*)^{-d/2}.$$
 (2.2.12)

The volume v can be found approximately by considering the density at the center of the spot as a function of time. When the center ceases to exist, the mean increase in the density in its interior is

$$\delta n_0 \sim \overline{n} e^{\lambda_0 \tau_0}, \qquad (2.2.13)$$

and the radius  $r_1$  with which the spot is formed is determined by the localization radius of the eigenfunction  $\varphi_0(r)$ . As soon as the center ceases to operate, the spot begins to spread out in accordance with the diffusion law:

$$\delta n (\mathbf{r}, t) = \int G_d (\mathbf{r} - \mathbf{r}', t) \, \delta n (\mathbf{r}', 0) \, \mathrm{d}\mathbf{r}'. \qquad (2.2.14)$$

It follows that the density change at the center of the core can be approximately represented by

$$\delta n(t) \sim \frac{r_1^d \delta n_0}{(4\pi D t)^{d/2}} \sim \bar{n} \, \frac{r_1^d e^{\lambda_0 \tau_0}}{(4\pi D t)^{d/2}} \,. \tag{2.2.15}$$

The spot is appreciable against the mean background so long as  $\delta n(t) \gtrsim \bar{n}$ , and this condition eventually yields the estimate

$$\tau^* \sim \frac{D}{r_1^2} \exp\left(\frac{2\lambda_0 \tau_0}{d}\right).$$
 (2.2.16)

Substituting this in (2.2.12), we conclude that the mutual influence of breeding centers can be neglected if

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$$m \ll \frac{D}{\tau_1^{2+d}} \exp\left[-\left(1+\frac{2}{d}\right)\lambda_0 \tau_0\right]. \tag{2.2.17}$$

Simple rearrangement then readily shows that this condition is equivalent to (2.2.10).

The aim of our analysis is to determine the threshold concentration of breeding centers. Comparison of (2.2.10) with (2.2.3) shows that clusters play no appreciable role in the evolution of the explosive instability threshold, provided  $\alpha \lessdot \alpha \bigstar$  where

$$\alpha^* = \frac{D}{r_1^2} \exp\left(-\frac{2}{d} \lambda_0 \tau_0\right). \qquad (2.2.18)$$

As the decay rate  $\alpha$  increases, the contribution of two-center clusters is found to increase. The expression for the explosive instability threshold in the case of strong centers, including the correction due to paired clusters, has the following form for  $\alpha \lessdot \alpha^*$ :

$$c_{\rm crit} = c_{\rm crit}^{(0)} \left[ 1 - \gamma_d \left( \frac{\alpha}{\alpha^*} \right)^{d/(d+2)} \right], \qquad (2.2.19)$$

where

$$\gamma_{d} = \begin{cases} \frac{1}{6} \Gamma\left(\frac{1}{3}\right), & d = 1, \\ \frac{\sqrt{\pi}}{8}, & d = 2, \\ \frac{2}{5} \Gamma\left(\frac{4}{5}\right) \left(\frac{3\sqrt{2}}{5}\right)^{2/5}, & d = 3, \end{cases}$$
(2.2.20)

and  $\Gamma(x)$  is the gamma-function.

When  $\alpha \gtrsim \alpha^*$ , the threshold is substantially reduced in comparison with  $c_{crit}^{(0)}$ . Clusters consisting of a large number of breeding centers then play the main role in the determination of the threshold.

As noted in the Introduction, there is a similarity between the problem of the explosive instability threshold and problems in percolation theory. The "directed flow" model<sup>18</sup> appears to be the closest to our situation. According to the model, the medium consists of elements located at the sites of a regular lattice, which are capable of occupying a state of rest or excitation. If, at a given time, an element is in an excited state, it is found to return to its original state of rest at the next (discrete) instant of time, but it can excite each of its nearest neighbors in the lattice with a probability p. This results in a random process resembling the propagation of infection with breeding of its carriers. It is clear that, when the excitation transfer probability p is very small, the number of excited elements will decrease with time. In the opposite limiting case, when the excitation transfer probability is close to unity, the number of excited elements will rise with time without limit. The critical value of the excitation transfer probability,  $p_c$ , for which we have a finite probability of unlimited growth in the number of excited elements for a single initial excited element corresponds to the percolation threshold.

It is clear that the critical probability  $p_c$  depends on the number of nearest neighbors in the given lattice, i.e., on the coordination number z. The greater the number of elements that are in contact with the "infected" element, the higher is

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the probability that the "infection" will breed. We note that the coordination number increases with increasing dimensionality d.

In the limit of high coordination numbers, the percolation threshold can be determined in the mean-field approximation. Let us suppose that the fraction of elements in the medium that are in an excited state at the *n*th instant of time is  $f_n$ . If the number z of nearest neighbors of each element in the medium is large, we may suppose that there are  $zf_n$  excited elements among its neighbors at the *n*th instant of time. Consequently, the probability of finding the element in an excited state at the next, (n + 1)th instant of time is  $1 - (1 - p)^{zfn}$ , since we have assumed that the probabilities of excitation transfer from each of the excited neighboring elements are independent. The quantity that we have obtained gives the fraction of elements of the medium that are in the excited state at time n + 1, i.e.,

$$f_{n+1} = 1 - (1 - p)^{2f_n}. \tag{2.2.21}$$

Above the percolation threshold, the quantity  $f_n$  must tend to a nonzero finite limit:

$$\lim_{n \to \infty} f_n = f^*, \tag{2.2.22}$$

which gives the number of excited elements in the steady state, while below the percolation threshold,  $\lim_{n\to\infty} f_n = 0$ .

By examining the point transformation (2.2.21), we can readily show that its attractors can only be stationary points, with the loss of stability of a stationary point f = 0 and the creation of a new stable stationary point  $f = f^* > 0$  occuring as the probability p passes through the value

$$p_{\rm c} = 1 - \exp\left(-\frac{1}{z}\right) = \frac{1}{z} - \frac{1}{2z^2} + \dots,$$
 (2.2.23)

that determines the percolation threshold.

As the coordination number z is reduced, the meanfield approximation ceases to be valid. Fluctuation effects<sup>18</sup> then become important.

The expression given by (2.2.3) for the explosive instability threshold was obtained in the mean-field approximation for the model described by (2.9)–(2.10), since it was assumed in its derivation that the increase in the material at each of the breeding centers began from the density averaged over the volume at the given instant of time. When the rate of decay  $\alpha$  is small, this approximation is valid because the critical concentration of breeding centers is low [(2.2.10) is satisfied] and the next successive breeding center appears outside the region that has been strongly perturbed by one of the preceding centers.

As the rate of decay  $\alpha$  increases, the critical concentration of breeding centers will also increase. The first fluctuation correction to the explosive instability threshold (2.2.3), determined in the mean-field approximation, is obtained by taking into account the contribution due to two-center clusters.

As the decay rate  $\alpha$  passes through the value  $\alpha^*$ , there is a transition to strong fluctuations for which the explosive instability threshold is determined by the appearance of infi-

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nite time chains of neighboring breeding centers.

These results may be interpreted as a reduction in the effective coordination number with increasing decay rate in the medium. This interpretation is confirmed by analyses of the role of dimensionality of the medium in this problem. It is well known that the coordination number increases with increasing dimensionality d. One would therefore expect that the mean-field approximation will be valid for media with high dimensionality.

In fact, it follows from (2.2.18) that the critical decay rate  $\alpha^*$  that determines the transition from the mean field state to the highly fluctuating state increases with increasing dimensionality d of the medium. We note also that, when  $d \ge 1$ , i.e., when the dimensionality is very high,<sup>51</sup> all the centers become long-lived because the corresponding condition is  $\tau_0 \ge r_0^2/Dd$ . The quantity  $J^*$  that appears in the condition  $J \ge J^*$  which defines strong breeding centers is also found to depend on the dimensionality of the medium:  $J^* \sim dD/r_0^2$  for  $d \ge 1$ . If we fix the intensity J of centers and their size  $r_0$ , we always arrive at the limit of weak breeding centers because  $J^*$  increases with increasing dimensionality d.

#### 2.3. Explosive instability threshold for weak centers

When the explosive instability threshold is calculated in the case of weak centers  $(J < J^*)$ , we can take as the first approximation the condition that the mean breeding rate  $\overline{f}$  over the volume is equal to the decay rate  $\alpha$ . Since the mean breeding rate is

$$\overline{f} = m \int g(\rho, \tau) d\rho d\tau = cJ\zeta_1, \qquad (2.3.1)$$

where

$$\zeta_t = r_0^{-d} \int \chi(\rho) \,\mathrm{d}\rho, \qquad (2.3.2)$$

This gives us the following result for the dimensionless critical concentration of breeding centers:

$$c_{\rm crit}^{(0)} = \frac{\alpha}{\zeta_1 J} \,. \tag{2.3.3}$$

We emphasize at once that (2.3.3) will by no means always serve as a reasonable first approximation to the true explosive instability threshold. Statistical correlation effects reduce the explosive threshold, and this reduction may turn out to be very appreciable. In this section, we shall establish the limits of validity of (2.3.3) and will calculate the first corrections to it for those cases for which they are small.

It follows from (2.1) that the rate of change of the mean density evaluated over the volume is given by

$$\dot{\overline{n}} = -(\alpha - \overline{f}) \, \overline{n} + \langle \delta f \, \delta n \rangle, \qquad (2.3.4)$$

where  $\delta f = f - \bar{f}$  and  $\delta n = n - \bar{n}$ . It is thus clear that the approximate result given by (2.3.3) corresponds to the situation

<sup>&</sup>lt;sup>5)</sup>Of course, the dimensionality of the medium will not exceed three in physical problems. However, the analysis of diffusion in media of high dimensionality is not altogether purely formal. Diffusion in spaces of high dimensionality is encountered, for example, in mathematical models of biological evolution.

where correlations between density and breeding-rate fluctuations  $\delta n$  and  $\delta f$  can be neglected.

The rate of change of  $\delta n$  is described by the stochastic equation

$$\dot{\delta n} = -(\alpha - \overline{f})\,\delta n + D\Delta\delta n + \delta f(\mathbf{r}, t)\,\overline{n} + (\delta f\,\delta n - \langle \delta f\,\delta n \rangle).$$
(2.3.5)

Since, near the threshold, the mean density  $\bar{n}$  increases or decreases with time very slowly in comparison with the characteristic "microscopic times," such as the lifetime of an individual center, the mean density  $\bar{n}$  may be regarded as constant in (2.3.5).

To calculate the corrections to the explosive instability threshold in (2.3.3), we must use (2.3.5) to determine the correlator  $\langle \delta f / \delta n \rangle$  for fixed  $\bar{n}$ . The diagram technique of perturbation theory as applied to random processes in Refs. 24– 27 provide us with a consistent way of calculating this quantity. The various details of this procedure can be found by the reader in Ref. 22. Here, we confine our attention to the principal features of the application of this technique to our problem.<sup>6)</sup>

In (2.3.5), we can transform to the Fourier components of the fluctuating fields, with the result that

$$\delta n_q = G_q^0 \left[ \overline{n} \delta f_q + \int \left( \delta f_{q-q'} \delta n_{q'} - \langle \delta f_{q-q'} \delta n_{q'} \rangle \right) \mathrm{d}q' \right], \quad (2.3.6)$$

where  $q \equiv (\omega, \mathbf{k})$  and

$$G_q^0 = (-i\omega + \alpha - \overline{f} + Dk^2)^{-1}. \qquad (2.3.7)$$

The formal solution of the integral equation (2.3.6) is given by an infinite iteration series in powers of  $\delta f_q$ . Multiplying this infinite series by  $\delta f_{q'}$ , and averaging over the ensemble of fluctuations  $\delta f$ , we obtain the expression for the correlator  $\langle \delta n_q \delta f_{q'} \rangle$  and, after integration, for the quantity  $\langle \delta f(\mathbf{r}, t) \delta n(\mathbf{r}, t) \rangle$ . In graphical form, the first few terms of the infinite series for this correlator are

$$<\delta f(\mathbf{r},t)\delta n(\mathbf{r},t) > = \overline{n} \left\{ \begin{array}{c} & + \\$$

A thin solid line with an arrow represents the function  $G_q^0$  whereas broken lines with points upon them represent irreducible correlators (cumulants) of random fields  $\delta f_{q'}$ . In contrast to the Gaussian random fields, for which all the higher-order correlators degenerate to pair correlators, the Poisson random field (2.10) has nonzero cumulants of all orders. It follows that several broken lines can converge ("pair") at a given point.

A further feature of (2.3.8) is that it does not contain weakly bound diagrams that decay when one of the lines is cut. Such diagrams cancel out during the averaging procedure because of the second term in the integrand in (2.3.6).

If we define the "self energy" function  $\Sigma_q$  and the Green's function  $G_q$  in the usual way, with these two functions being related by the Dyson equation

$$\langle \delta f \, \delta n \rangle = \overline{n} \Sigma_{\mathbf{0}}, \quad G_q^{-1} = G_q^{\mathbf{0}^{-1}} - \Sigma_q^{\dagger}, \qquad (2.3.9)$$

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and if we use the function  $G_q$  to perform a partial summation of the diagrams in the series for  $\Sigma_q$ , we obtain the graphical expression

$$\Sigma_q = \bigcirc + \bigcirc + \bigcirc + \bigcirc + \cdots$$
(2.3.10)

The threshold for explosive instability is given by

$$\alpha = \overline{f} + \Sigma_0. \tag{2.3.11}$$

The quantity  $\Sigma_0$  is thus seen to detrmine the fluctuational shift of the explosive instability threshold.

Using the Dyson equation (2.3.9) and the formula given by (2.3.7), we find that the Green's function is given by

$$G_q^{-1} = -i\boldsymbol{\omega} + \boldsymbol{\alpha} - \overline{f} + Dk^2 - \Sigma_q. \qquad (2.3.12)$$

It is clear that the Green's function has a pole at q = 0.

Analysis of the contribution of different diagrams in (2.3.10) leads to the following results. All diagrams with irreducible correlators of order higher than 2 [second and fourth in (2.3.10)] are small with respect to the parameter  $J/J^*$  that characterizes the extent to which the breeding centers can be regarded as small. The more "hazardous" diagrams are those in which broken lines cross [third in (2.3.10)]. Their contributions are small in comparison with that of the first diagram provided

$$c(J\tau_0) \frac{J}{I^*} \ll 1,$$
 (2.3.13)

which restricts the concentration of breeding centers.

When the lowest-order fluctuation correction to the explosive instability threshold is taken into account (it is determined by the first of the diagrams in the series for  $\Sigma_q$ ), the expression for the critical concentration of breeding centers is

$$\boldsymbol{c}_{\text{crit}} = \boldsymbol{c}_{\text{crit}}^{0} \left( 1 - \boldsymbol{v}_{\boldsymbol{d}} \frac{J}{J^{*}} \right), \qquad (2.3.14)$$

where the numerical coefficient is given by<sup>7)</sup>

$$\mathbf{v}_{d} = \begin{cases} \frac{\zeta_{2}}{2\zeta_{1}} & \text{for} \quad r_{0} \gg l, \quad \frac{\zeta_{1}}{4} & \text{for} \quad r_{0} \ll l, \quad d = 1, \\ \frac{\zeta_{2}}{2\zeta_{1}} & \text{for} \quad r_{0} \gg l, \quad \frac{\zeta_{1}}{4\pi} & \text{for} \quad r_{0} \ll l, \quad d = 2, \\ \frac{\zeta_{2}}{2\zeta_{1}} & \text{for} \quad r_{0} \gg l, \quad \frac{\zeta_{1}}{4\pi^{2}} & \text{for} \quad r_{0} \ll l, \quad d = 3, \end{cases}$$

$$(2.3.15)$$

 $c_{\rm crit}^{(0)}$  is given by (2.3.3), and  $l = \sqrt{D\tau_0}$ .

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The lowest-order correction is sufficient if the critical concentration  $c_{\text{crit}}^{(0)}$  satisfies (2.3.13). Substitution of (2.3.14) in (2.3.13) yields

$$t\tau_0 \frac{J}{I^*} \ll 1, \tag{2.3.16}$$

which shows the restriction imposed on the decay rate  $\alpha$ .

<sup>&</sup>lt;sup>6)</sup>It would appear that these results can also be reproduced by functional methods (Furutsu-Novikov formulas<sup>28</sup>).

<sup>&</sup>lt;sup>7)</sup>The values given by (2.3.15) were obtained for the model in which it was assumed that the intensity of each individual breeding center did not remain constant within the interval  $\tau_0$  but fell off smoothly in proportion to exp  $(-t/\tau_0)$  from the instant at which the center was created.

When (2.3.13) is violated, i.e.,  $\alpha \gtrsim J^*/J\tau_0$ , the contributions of all diagrams with crossing broken lines are the same, or even greater, than the contribution of the first diagram.<sup>8)</sup> In this situation, the determination of the explosive instability threshold would require the summation of all diagrams with crossings, which presents an exceedingly difficult problem. It is, however, possible to maintain that, when  $\alpha \gtrsim J^*/J\tau_0$ , one should observe a substantial reduction in the explosive instability threshold as compared with the value of  $c_{crit}^{(0)}$ deduced from simple considerations.

We shall show in the next section that the sharp reduction in the explosive instability threshold that accompanies the reversal of the inequality given by (2.3.16) is due to the fact that the main contribution to the onset of explosive instability then begins to be provided by clusters of weak breeding centers, the behavior of which is analogous to that of an individual strong center.

# 2.4. Role of rare clusters

The condition given by (2.3.16), which ensures that the fluctuational reduction in the explosive instability threshold is small, can be obtained independently by introducing theoretical results on the spectrum of the Schrödinger equation with a stochastic potential. This approach not only elucidates the nature of the sharp reduction in the instability threshold, but also enables us to analyze differences in the fluctuational behavior of media of different dimensionality.

Let us suppose that the volume  $\Delta V$  contains an isolated cluster of weak breeding centers during an interval of time  $\Delta t$ . The increase in the amount of material within this volume is then described by

$$\boldsymbol{n} = f(\mathbf{r}, t) \boldsymbol{n} + \boldsymbol{D} \Delta \boldsymbol{n}, \qquad (2.4.1)$$

where  $f(\mathbf{r}, t)$  is the breeding rate field for the chosen cluster.

As noted in Section 2.1, this equation can be interpreted as the Schrödinger equation with imaginary time and potential  $U(\mathbf{r}, t) = -f(\mathbf{r}, t)$  if we substitute  $\hbar^2/2m \rightarrow D$ . This is so because for clusters of breeding centers  $f(\mathbf{r}, t) \ge 0$ , the potential corresponding to such clusters is a potential well that varies in time. For each given instant of time, we can calculate the spectrum of energies  $E_1(t)$  corresponding to bound states in this potential well. These energies are given by the positive eigenvalues  $\lambda_1(t) = -E_1(t)$  of the operator

$$\hat{L} = D\Delta + f(\mathbf{r}, t). \tag{2.4.2}$$

Since the time in (2.4.1) is "imaginary" as compared with the Schrödinger equation, the most rapidly growing contribution to the density  $n(\mathbf{r}, t)$  is that corresponding to the lowest-lying level in the potential well, i.e., the maximum eigenvalue  $\lambda(t) = \max \lambda_1(t)$ .

A cluster can be crudely characterized by the maximum time-averaged value of  $\lambda$  and the lifetime  $\tau$ . When  $\lambda \tau < 1$ , such a cluster remains weak and its contribution is represented by the first few terms of perturbation theory, which leads to (2.3.14). On the other hand, when  $\lambda \tau > 1$ , the increase in the material on the cluster is exponentially large.

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We note that, in the last case, the cluster must consist of a large number of breeding centers localized in a small spacetime region. The condition  $\lambda \tau > 1$  can also be written in the form  $\tau > \lambda^{-1}$ , from which it is clear that the potential  $U(\mathbf{r}, t) = -f(\mathbf{r}, t)$  for this particular cluster is a slowly-varying function of time as compared with the reciprocal "frequency"  $\lambda^{-1}$ . The adiabatic approximation<sup>23</sup> is therefore valid, and the total increase in the material on the cluster is given by

$$\Delta N = n (0) \exp \int_{0}^{\Delta t} \lambda (\tau) d\tau. \qquad (2.4.3)$$

If we describe an individual cluster by introducing the characteristic parameter

$$s = \int_{-\infty}^{+\infty} \lambda(t) \, \mathrm{d}t, \qquad (2.4.4)$$

we find that (2.4.3) can be written as  $\Delta N = n(0)e^s$ , where n(0) is the mean density in the medium just before the appearance of the given cluster.

Suppose that p = p(s) is the probability per unit volume per unit time that a strong cluster with parameter s will appear in the medium. The average increase in density on strong clusters per unit time is then

$$\overline{\Delta n} = \int \Delta N (s) p(s) ds. \qquad (2.4.5)$$

The average rate of increase Q is therefore

$$Q \sim \int e^s p(s) \, \mathrm{d}s. \tag{2.4.6}$$

The explosive instability threshold determined from the condition that the decay rate  $\alpha$  must be equal to the total rate of increase in the amount of material, which includes contributions of  $\bar{f}$  and Q, is given by (2.3.14), provided  $Q < \bar{f}$ .

The expression for the average rate of increase of the material on strong clusters, given by (2.4.6), contains the exponentially large factor  $e^s$ . The quantity  $\vec{f}$  cannot be exponentially large under the same conditions. Hence the contribution of strong clusters must be negligible provided only they are exponentially rare, i.e., provided  $p(s) = \exp(-\Phi(s))$ , where  $\Phi(s) > 1$  and s > 1.

However, even this requirement will not completely quarantee that the contribution of Q can be totally neglected. In fact, in this situation,

$$Q \sim \int_{1}^{\infty} \exp(s - \Phi(s)) \, \mathrm{d}s_{\bullet}$$
 (2.4.7)

and the order of magnitude of Q is determined by competition between two factors, namely, the fact that strong clusters are exponentially rare and that the increase in the material on each of them is exponential. We shall see that this competition is of fundamental significance in three-dimensional media.

The estimates of Q given below refer to the limit of high concentration of breeding centers for which individual centers exhibit considerable overlap (c > 1). The field  $f(\mathbf{r}, t)$  in

<sup>&</sup>lt;sup>8</sup>We note that it is clear from (2.3.14) that the correction associated with the first diagram is always small if  $J/J^{\bullet} < 1$ .

the medium can then be looked upon as Gaussian. Similar considerations can be repeated in the more general case where the field  $f(\mathbf{r}, t)$  is Poissonian.

In the Gaussian limit, it is convenient to measure the rate of decay from the mean breeding rate  $\tilde{f} = \zeta_1 cJ$ , i.e., it is convenient to define the new rate  $\alpha_{eff} = \alpha - \overline{f}$ . The function  $f(\mathbf{r}, t)$  can then be interpreted as the fluctuating component of the breeding rate field  $\delta f = f(\mathbf{r}, t) - \overline{f}$ , which is a Gaussian random process with pair correlation function

$$\langle \delta f(\mathbf{r}, t) \, \delta f(0, 0) \rangle = c J^2 \mathscr{P}(\mathbf{r}, t), \qquad (2.4.8)$$

where

$$\mathscr{P}(\mathbf{r}, t) = r_0^{-d} \tau_0^{-1} \int \chi \left( \mathbf{r} - \mathbf{r}' \right) \chi \left( \mathbf{r}' \right) d\mathbf{r}' \int \sigma \left( t - t' \right) \sigma \left( t' \right) dt'$$
(2.4.9)

is a dimensionless overlap interval. The intensity S of this random process is

$$S = cJ^{2}\mathscr{P}(0, 0) = \zeta_{2}cJ^{2},$$
  

$$\zeta_{2} = r_{0}^{-d} \int \chi^{2}(\mathbf{r}) \, \mathrm{d}\mathbf{r}.$$
(2.4.10)

Its correlation radius is  $r_0$  and its correlation time is  $\tau_0$ .

Since, in the Gaussian limit, the breeding centers show considerable overlap, the phrase "cluster of breeding centers" must be interpreted as describing rare strong positive bursts of the random field  $\delta f(\mathbf{r}, t)$ , due to an anomalously high concentration of breeding centers.

Let  $\overline{\lambda}$  be the characteristic depth of the lowest-lying level in the "potential well" due to a *typical* field fluctuation  $\delta f(\mathbf{r}, t)$ . To ensure that the fluctuational reduction in the threshold is small, typical fluctuations must definitely be weak, i.e., the condition  $\overline{\lambda}\tau_0 < 1$ , must be satisfied for them because the correlation time  $\tau_0$  can be looked upon as the characteristic lifetime of a typical fluctuation.

For strong fluctuations,  $s \ge 1$  and, since  $s = \lambda \tau$ , this condition can be satisfied in two ways. Firstly, some of the fluctuations with typical value  $\lambda \sim \overline{\lambda}$  may turn out to be unusually long-lived so that their lifetime  $\tau$  is much greater than  $\tau_0$ . Secondly, very "deep" fluctuations for which  $\lambda \ge \overline{\lambda}$  may appear and, since they are very rare events, their lifetime must be of the order of  $\tau_0$ .

The next step is to recognize that we have separately estimated the contributions to the mean rate of growth of the material that are due to strong fluctuations (clusters) of two limiting types. It turns out that the most "hazardous" are clusters of the second type which may be referred to as spatial (in contrast to "temporal" clusters belonging to the first type). It is precisely these spatial clusters that determine the transition to the state with considerably reduced explosive instability threshold.

The probability of appearance of temporal clusters with lifetime  $\tau \gg \tau_0$  was estimated for the Gaussian random process in Ref. 20, where it was found that

$$p(\tau) \sim \exp(-\varphi(\tau)), \quad \varphi(\tau) \sim \frac{\tau}{\tau_0} \quad \text{for} \quad \tau \gg \tau_0.$$
  
(2.4.11)

The contribution of such fluctuations to Q is given by the order-of-magnitude formula

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$$Q \sim \int_{\overline{\lambda}^{-1}}^{\infty} \exp\left(\overline{\lambda}\tau - \varphi\left(\tau\right)\right) d\tau. \qquad (2.4.12)$$

It is readily noted that this cluster provides an exponentially small contribution, provided  $\bar{\lambda}\tau_0 \leq 1$ .

When we consider the contribution due to spatial clusters, we can use the results obtained in the theory of singleparticle spectra of the Schrödinger equation with a stationary random potential. In fact, for times of the order of correlation time  $\tau_0$ , the picture of the breeding field  $\delta f(\mathbf{r}, t)$  is almost stationary. The "frozen" spatial distribution in this case is Gaussian.

It is well known<sup>14,16</sup> that exponentially rare low-lying levels belong to the fluctuational part of the spectrum of the Schrödinger equation with random potential. The probability of finding such levels is  $p(\lambda) = \exp(-\Phi(\lambda))$ , where, in the Gaussian limit, we have

$$\Phi(\lambda) = a \frac{\lambda^{2 - (d/2)} D^{d/2}}{Sr_0^d} \quad \text{for} \quad \frac{\lambda r_0^2}{D} \ll 1, \qquad (2.4.13a)$$

$$\Phi(\lambda) = \frac{\lambda^2}{2S} \qquad \text{for} \quad \frac{\lambda r_b^2}{D} \gg 1. \qquad (2.4.13b)$$

The dimensionless coefficient *a* is of the order of unity. For  $\lambda r_0^2/D \sim 1$ , the two expressions for  $\Phi(\lambda)$  agree to within an order of magnitude. The formulas given by (2.4.13) have been deduced from those given in Ref. 14 with allowance for the fact that (2.4.1) reduces to the Schrödinger equation if  $\hbar^2/2m \rightarrow D, \Psi \rightarrow n, U(\mathbf{r}, t) \rightarrow -f(\mathbf{r}, t), E \rightarrow -\lambda$ . They are valid only for exponentially rare levels, i.e., provided  $\Phi(\lambda) \ge 1$ . We recall that, according to (2.4.10), the intensity S is proportional to the concentration c of the breeding centers.

The contribution of spatial clusters to the mean rate of increase of the material is estimated by

$$Q \sim \int_{\tau_0^{-1}}^{\infty} \exp\left(F\left(\lambda\right)\right) d\lambda, \qquad (2.4.14)$$

where

$$F(\lambda) = \lambda \tau_0 - \Phi(\lambda). \qquad (2.4.15)$$

Let us start by analyzing the situation where the breeding centers are long-lived, i.e.,  $\tau_0 \gg r_0^2/D$ . In this case, there are two regions for  $\lambda \gtrsim \tau_0^{-1}$  for which (2.4.13a) and (2.4.13b) apply. The form of the functions  $F(\lambda)$  for media of different dimensionality is shown in Fig. 1.

In the one-dimensional case, the contribution of Q is exponentially small if  $\lambda = \tau_0^{-1}$  for the function  $F(\lambda)$  is negative and  $|F(\lambda)| \ge 1$ . This requirement is actually equivalent to the requirement that levels of depth  $\lambda = \tau_0^{-1}$  should be exponentially rare. Using (2.4.13) for d = 1, we find that this condition is satisfied if

$$S \ll \frac{D^{1/2}}{\tau_0^{3/2} r_0}$$
, (2.4.16)

or, using (2.4.10) and (2.4.7), if

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$$(cJ) \tau_0 \frac{J}{J^*} \ll 1.$$
 (2.4.17)

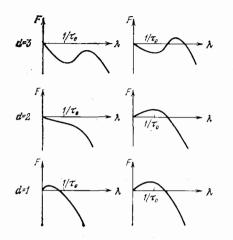


FIG. 1. Change in the form of  $F(\lambda)$  as a result of transition to a situation where the explosive instability threshold is determined by clusters of breeding centers.

d = 1, 2, 3—dimensionality of the medium;  $\tau_0 > r_0^2/D$ .

When the contributions to the rate of increase due to rare clusters are small, the explosive instability threshold is given by  $\alpha = \overline{f} = cJ\zeta_1$ . Substituting  $cJ \sim \alpha$  in (4.17), we arrive at (2.3.16) which was obtained earlier from estimates of the perturbation-theory diagrams. It defines the range of values of the decay rate  $\alpha$  in which the threshold exhibits only a slight fluctuational depression.

The critical concentration increases with incressing decay rate  $\alpha$  and, eventually, the condition given by (2.4.17) is violatd. Strong spatial clusters cease to be exponentially rare, and it is precisely on such clusters that the main increase in the amount of material in the medium occurs near the explosive instability threshold.

Analysis of the two-dimensional case leads to analogous conclusions. The only difference is that, when this analysis is performed, it does not reveal the logarithmic factor in the expression for  $J^*$  given by (2.1.8) for d = 2. However, this is not unexpected because the expressions given by (2.4.14) are valid only with logarithmic precision [they do not take into account the preexponential factor in the expression for the probability  $p(\lambda)$ ].

The three-dimensional model presents a special case. The violation of (2.3.16) is again connected with the exponential increase in the contribution of rare strong clusters but, in contrast to the one- and two-dimensional cases, strong clusters with  $\lambda \sim \tau_0^{-1}$  remain ineffective in this case (cf. Fig. 1). As the decay rate increases, the most "hazardous" clusters are those for which  $\lambda \sim D/r_0^2 \gg \tau_0^{-1}$ . It is precisely for these clusters that the function  $F(\lambda)$  first reaches zero as the intensity S of the breeding-rate field increases. Estimates based on (2.4.13) show that this occurs for  $S \sim D / r_0^2 \tau_0$ . The intensity S is related to the concentration c by the formula  $S = \zeta_2 c J^2$ , and the threshold concentration  $c_{\rm crit}$  depends on the decay rate  $\alpha$ . Up to the time at which the contribution due to rare strong clusters is no longer exponentially small, the critical concentration is given by  $c_{\rm crit} = \alpha/J\zeta_1$ . Comparison of th above expressions shows that the contribution of rare strong clusters becomes dominant for  $\alpha \gtrsim D / \tau_0 r_0^2 J$ . It is also readily seen that, by virtue of (2.1.9), this condition can be given the form  $\alpha \gtrsim \alpha^{**}$ , where  $\alpha^{**} = J^*/(J\tau_0)$ . We thus again arrive at the criterion given by (2.3.16).

It is clear from the figure that, in a three-dimensional medium, there is a range of values of the decay rate  $\alpha$  in which the threshold is determined exclusively by exponentially rare strong clusters. The probability of appearance of such clusters is described by (2.4.13) which gives the level density in the fluctuation region. Within this interval, in which  $(D\tau_0/r_0^2)^{3/2} \ge \ln(\alpha/\alpha^{**}) \ge D\tau_0/r_0^2 \ge 1$ , the threshold concentration of breeding centers can be determined from (2.4.14) by the method of steepest descents:

$$c_{\rm crit} = \frac{2\ln(\alpha/\alpha^{**})}{\zeta_2 J^2 \tau_0^2} \,. \tag{2.4.18}$$

As can be seen, the linear relation  $c_{\rm crit} \sim \alpha/J$  is replaced in this interval by the much slower logarithmic relation. When  $\ln (\alpha/\alpha^{**}) \gtrsim (D\tau_0/r_0^2)^{3/2}$ , clusters for which  $\lambda \sim \tau_0^{-1}$  cease to be exponentially rare.

It may be shown that, in the case of short-lived breeding centers  $(\tau_0 \ll r_0^2/D)$ , the relative reduction in the explosive instability threshold when (2.3.16) ceases to be satisfied in a medium of any dimensionality is always connected with the fact that spatial centers with levels  $\lambda \sim \tau_0^{-1}$  cease to be exponentially rare.

In conclusion, we emphasize once again that rare but strong clusters of breeding centers can ensure a substantial depression of the explosive instability threshold; this depression occurs when (2.3.16) ceases to be valid.

# 2.5. Kinetic transition of the "medium populating" type

Let us suppose that the medium supports a nonlinear mechanism restricting explosive instability, so that (2.9) must be replaced with

$$\mathbf{n} = -\alpha n - \beta n^2 + f(\mathbf{r}, t) n + D\Delta n. \qquad (2.5.1)$$

The origin of the quadratic term in n may be different in different cases. We note, to begin with, that this term appears in the kinetic equation when the coalescence reaction  $(X + X \rightarrow X)$ , which is the reverse of the breeding reaction, takes place. One would naturally expect that, in contrast to the breeding reaction, the coalescence reaction does not require activation, and will therefore occur throughout the medium. Moreover, there are possible situations where the decay products enhance the decay rate (for example, for some biochemical fermentation reactions<sup>29,30</sup>). If we suppose that the restriction on the growth of density sets in for sufficiently small values of n, we can expand the decay rate into a series in powers of the density n and retain only the linear and quadratic terms.

When the limiting mechanism is present above the explosive instability threshold, the unlimited growth in the mean density within the volume is suppressed. Depending on the amount  $\Delta c = c - c_{\rm crit}$  by which the threshold is exceeded, a mean density  $\bar{n}$  is eventually established in the medium. It decreases with decreasing  $\Delta c$ , and vanishes for  $\Delta c \leq 0$ .

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The appearance of nonzero mean density  $\overline{n}$  for  $c \ge c_{crit}$  as  $t \to \infty$  will be interpreted below as a kinetic transition of the "medium populating" type.

In one particular respect, the asymptotically established mean density  $\overline{n}$  may be looked upon as an order parameter of the kinetic transition. In fact when it passes through  $c = c_{crit}$ , the medium that we are considering acquires an *additional* quantitative characteristic as  $t \rightarrow \infty$ , namely, the density of the material populating it.<sup>9)</sup>

From the formal point of view, it is clear that the kinetic transition of the "medium populating" type is accompanied by a reduction in the symmetry of the asymptotic solution established in the limit as  $t \rightarrow \infty$ . Below the transition point, when  $\overline{n} = 0$ , the solution is clearly invariant under the group of scaling transformations involving multiplication by an arbitrary nonzero number. This invariance is lost above the transition point.

We emphasize, however, that, in contrast to secondorder phase transitions in equilibrium systems, the above symmetry under scaling transformations is valid only for (asymptotic) solutions of the original equation given by (2.5.1). The equation itself never exhibits this invariance. This distinction must be borne in mind when we refer to  $\overline{n}$  as the order parameter of the kinetic transition under consideration. We must also remember that (2.5.1) itself ceases to be valid for limiting small concentrations  $\bar{n} \leq r_0^{-d}$ , for which the concentration of the breeding material becomes so low that only a few particles are present in an individual breeding center. In the immediate neighborhood of a kinetic transition of the "medium populating" type, we must take into account the discrete nature of the reactions. However, we shall confine our attention to (2.5.1) which is valid in the hydrodynamic limit.

Since the breeding rate is random, local density fluctuations  $\delta n$  persist in the steady asymptotic state with the stationary mean density  $\bar{n}$  evaluated over the entire volume. In this section, we shall examine mean square density fluctuations ( $\delta n^2$ ) in the steady state as we approach the kinetic transition point.<sup>10</sup>

It is well known that, in second-order equilibrium phase transitions, the order parameter  $\bar{\eta}$  vanishes as we pass through the transition point, whereas the pair correlation function for fluctuations  $\delta n$  in the order parameter is nonzero at the transition point. In this respect, the kinetic transition of the "medium populating" type exhibits an important specific feature. Since, in this case, the analog of the order parameter is a *nonnegative* quantity (the density *n* of the breeding material), the fact that the mean density  $\bar{n}$  within the volume is zero implies that all these density fluctuations must be zero:

$$\overline{n} = 0 \to \langle \delta n^2 \rangle = 0.$$
(2.5.2)

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Thus, the fluctuation behavior near the kinetic transition of the "medium populating" type is very distinctive.

We shall now confine our attention to the situation where the breeding centers are weak and (2.3.16) is satisfied. The latter condition guarantees that the increase in the amount of material on rare strong clusters of breeding centers is exponentially small.

According to (2.5.1), the steady value of the mean density  $\overline{n}$  is the stationary solution of the equation

$$\overline{n} = -\alpha \overline{n} + \overline{fn} - \beta \overline{n^2} + \langle \delta f \delta n \rangle - \beta \langle \delta n^2 \rangle.$$
(2.5.3)

If we neglect the density fluctuations  $\delta n$  in (2.5.3), which is equivalent to the self-consistent field approximation in the theory of second-order equilibrium phase transitions,<sup>31</sup> the steady value of the mean ensity is given by

$$\overline{n} = \begin{cases} 0, & c < c_{\text{crit}}^{(0)} \\ \frac{\alpha}{\beta} \left( \frac{c}{c_{\text{rit}}^{(0)}} - 1 \right), & c \ge c_{\text{crit}}^{(0)} \end{cases}$$
(2.5.4)

where  $c_{\rm crit}^{(0)}$  is found from the condition  $\alpha = \overline{f}$ .

Inclusion of fluctuations leads to two effects. Firstly, there is the fluctuation reduction in the explosive instability threshold, i.e., the kinetic transition points, calculated in Section 2.3. When (2.3.16) is satisfied, the fluctuation shift of the transition point is small [see (2.3.24)]. Secondly, the mean density  $\bar{n}$  given by (2.5.4) may change because of the presence in (2.5.3) of both the term  $\beta \bar{n}^2$  and the term  $\beta \langle \delta n^2 \rangle$ . It is only when the condition

$$\sqrt{\langle \delta n^2 \rangle} \ll \overline{n} \tag{2.5.5}$$

is satisfied that the self-consistent field approximation, used to obtain (2.5.4), will be valid.

It is well known that the self-consistent field approximation is always invalid in the theory of second-order phase transitions in a sufficient proximity of the transition point when the dimensionality of the medium is less than four. We shall see below that the situation is different in the case of a "medium populating" kinetic transition.

The density fluctuations  $\delta n$  satisfy the stochastic differential equation

$$\delta n = - (\alpha + 2\beta \overline{n} - \overline{f}) \,\delta n + D\Delta\delta n + \delta f (\mathbf{r}, t) \,\overline{n} \\ + (\delta f \,\delta n - \langle \delta f \,\delta n \rangle) - \beta (\delta n^2 - \langle \delta n^2 \rangle). \quad (2.5.6)$$

We can again use the perturbation theory graphs to find  $\langle \delta n^2 \rangle$  with the aid of this equation. Consider the function  $U_q$  defined by

$$\langle \delta n_q \delta n_{q'} \rangle = U_q \delta \left( q + q' \right) \bar{n^2}, \qquad (2.5.7)$$

so that

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$$\langle \delta n^2 \rangle = \overline{n^2} \int U_q \, \mathrm{d}q. \tag{2.5.8}$$

Successive iterations in the equation for  $\delta n_q$ , obtained by taking the Fourier transform of (2.5.6) with quadratic terms in  $\delta n$  discarded by virtue of (2.5.5), enable us to construct a formal solution for  $\delta n_q$  in the form of an infinite series in powers of the random fields  $\delta f_{q^r}$ . Multiplying two

<sup>&</sup>lt;sup>99</sup>We know one further example of a transition with a nonnegative order parameter. The order parameter can be looked upon as the relative fraction of elements belonging to an infinite cluster as the percolation threshold is traversed. Its magnitude is zero below the threshold whereas, above the threshold, it is finite and increases with increasing probability of bonding.

<sup>&</sup>lt;sup>10</sup>We note that, since the density fluctuations  $\delta n$  in our problem are due to an agency that is external to the system, the correlator  $\langle \delta n (\mathbf{r}) \delta n (0) \rangle$ need not diverge for coincident arguments.

such infinite series for  $\delta n_q$  term by term, and evaluating the statistical average, we obtain an infinite series of graphs for the function  $U_q$ . Summation of this series of weakly coupled graphs leads us to the integral equation

$$U_{q} = |G_{q}|^{2} \left[ \widetilde{S}(q) + \int S(q-q') U_{q'} \, \mathrm{d}q' \right].$$
 (2.5.9)

The function  $\widetilde{S}(q)$  in this expression is given by the following series of graphs:

where S(q) is the Fourier transform of the pair correlator  $\langle \delta f(\mathbf{r}, t) \, \delta f(0, 0) \rangle$ .

Let us discard all the graph terms in (2.5.10), i.e., let us approximately put  $\tilde{S}(q) = S(q)$ . If we then introduce the new function  $Z_q$  defined by  $U_q = |G_q|^2 Z_q$ , we find that it must satisfy the integral equation

$$Z_{q} = S(q) + \int S(q-q') |G_{q'}|^{2} Z_{q'} dq'. \qquad (2.5.11)$$

Since the Green's function  $G_q$  has a pole at the threshold when q = 0, the principal contribution to the integral near the threshold is provided by the region of small values of q. Taking S(q) out from under the integral sign, and performing some simple algebraic manipulations, we find  $Z_q$  and then  $\langle \delta n^2 \rangle$ :

$$\langle \delta n^2 \rangle = \bar{n^2} \, \frac{\mu}{1-\mu} \,, \tag{2.5.12}$$

where

$$\mu = \int S(q) |G_q|^2 dq. \qquad (2.5.13)$$

Thus, the criterion given by (2.5.5), which ensures the validity of the self-consistent field approximation, is satisfied if  $\mu < 1$ .

Calculations yield the following values of  $\mu$  in media of different dimensionality:

a) Long-lived breeding centers  $(\tau_0 \ge r_0^2/D)$ , or  $l \ge r_0$ , where  $l = \sqrt{D\tau_0}$ :

$$\mu \sim \begin{cases} \gamma\left(\frac{r_0}{l}\right), & d = 3, \\ \frac{\gamma \ln (r_c/l)}{\ln (l/r_0)}, & d = 2, \\ \gamma\left(\frac{r_c}{l}\right), & d = 1. \end{cases}$$
(2.5.14)

b) Short-lived breeding centers  $(l \lt r_0)$ :

$$\mu \sim \begin{cases} \gamma \left(\frac{r_0}{l}\right)^2, & d = 3, \\ \gamma \left(\frac{r_0}{l}\right)^2 \ln \frac{r_c}{r_0}, & d = 2, \\ \gamma \left(\frac{r_0}{l}\right)^2 \frac{r_c}{r_0}, & d = 1. \end{cases}$$
(2.5.15)

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The proportionality symbol indicates that we have discarded numerical factors of the order of unity. The quantity  $l = \sqrt{D\tau_0}$  has a simple interpretation: it is the mean distance traversed by a diffusing particle during the lifetime  $\tau_0$  of an individual breeding center.

The expressions given by (2.5.14) and (2.5.15) involve (with the exception of the three-dimensional medium) the correlation length of density fluctuations:

$$r_{\rm c} = \sqrt{\frac{D}{\alpha \left[ (c/c_{\rm crit}) - 1 \right]}}, \qquad (2.5.16)$$

which becomes infinite at the kinetic transition point at which  $c = c_{crit}$ . This means that, for d = 1 and d = 2, the quantity  $\mu$  increases as the critical point is approached, whereas for the three-dimensional medium (d = 3), it becomes constant in the limit as  $c \rightarrow c_{crit}$ .

The parameter  $\gamma$  in (2.5.14) and (2.5.15) has the form

$$\gamma = c \left(J\tau_0\right) \frac{J}{J^*}.$$
 (2.5.17)

According to (2.3.13), it is small near the threshold  $(\gamma < 1)$ , provided we satisfy (2.3.16) which ensures that the fluctuational shift of the explosive instability threshold is small.

By estimating the contribution of the different diagrams in the series given by (2.5.10), we can show that the contributions due to diagrams with irreducible high-order correlators [3, 4, and 6 in (2.5.10)] are small with respect to the parameter  $J/J^*$ . When the contributions due to diagrams with crossing broken lines are estimated [1, 2, and 5 in (2.5.10)], we must distinguish between two situations, namely, those involving long-lived and short-lived breeding centers. When the lifetime of an individual center is long  $(l > r_0)$ , these graphs are small with respect to the parameter  $\gamma$ . On the other hand, when the centers are short-lived  $(l < r_0)$ , the parameter  $\gamma$  is replaced with the combination  $\gamma(r_0/l)^2$ , in which case the contribution of such graphs near the explosive instability threshold is negligible, provided

$$\frac{\alpha r_{\delta}^2}{D} \frac{J}{J^*} \ll 1, \qquad (2.5.18)$$

which is a more stringent condition than (2.3.16).

In view of the foregoing, the steady value of the mean density  $\bar{n}$  of the breeding material is given by (2.5.4) with  $c_{\rm crit}^{(0)}$ replaced by  $c_{\rm crit}$  (i.e., the fluctuational depression of the threshold has been taken into account). The fact that this stationary solution is stable can be verified by linearizing (2.53) with respect to a small deviation from  $\bar{n}_{\rm st}$ . Simple considerations can then be used to verify not only that the solution (2.5.4) is stable, but also to exhibit the critical slowing down effect, i.e., the fact that the characteristic time for a change in  $\bar{n}(t)$  becomes infinite at the transition point. The corresponding formula for this time is given below.

Thus, the principal features of the "medium populating" kinetic transition that we have been considering are as follows:

1) Critical slowing down is observed near the transition point, i.e., the relaxation time increases in accordance with

$$\mathbf{r}_{c} = \left[\alpha \left(\frac{c}{c_{crit}} - 1\right)\right]^{-1}.$$
(2.5.19)

The onset of critical slowing down is connected with the presence of the q = 0 pole in the Green's function at the transition point.

2) It is clear from (2.5.16) that, like  $\tau_c$ , the correlation length  $r_c$  becomes infinite at the transition point.

3) The fluctuation shift of the transition point (explosive instability threshold) is independent of the specific mechanism responsible for nonlinear limitation; the fluctuation shift is small when (2.3.16) is satisfied.

4) According to the terminology adopted in the theory of second-order equilibrium phase transitions, the region near the transition point, in which the self-consistent field approximation is violated, is referred to as the fluctuation region. It follows from (2.5.14) that the fluctuation region is completely absent from the three-dimensional medium for long-lived breeding centers  $(l \ge r_0)$ , provided (2.3.16) is satisfied. When d = 2, it is found to be exponentially narrow with respect to the small parameter  $\gamma$ , its width being given by

$$\frac{c}{c_{\text{crit}}} - 1 \sim (\alpha \tau_0)^{-1} \exp\left[-\frac{2\ln\left(l/r_0\right)}{\gamma}\right].$$
(2.5.20)

In the one-dimensional case, the width of the fluctuation region is given by

$$\frac{c}{c_{\rm crit}} - 1 \sim (\alpha \tau_0)^{-1} \gamma^2 = \gamma \frac{J}{J^*}. \qquad (2.5.21)$$

Consequently, these fluctuations are much weaker than in the case of the phase transition. They become more important when the dimensionality of the medium is smaller. In actual fact, the fluctuation region appears only in the onedimensional case, and not beginning with d = 3 as in the case of second-order phase transitions.

5) For short-lived breeding centers  $(l \ll r_0)$ , the conditions ensuring that the fluctuation shift of the transition point is small are insufficient to ensure that the self-consistent field approximation will be valid for phenomena occurring near this point. The analog of the Ginzburg number in this case is the combination  $\gamma(r_0/l)^2$ . When  $(l/r_0)^2 \leq \gamma \ll 1$ , the shift of the threshold is small, and the threshold is given by (2.3.14), but the self-consistent field approximation is invalid whatever the dimensionality of the medium. If, on the other hand,  $\gamma \ll (l/r_0)^2 \ll 1$ , i.e., (2.5.18) is satisfied, the self-consistent field approximation is valid at all points as close to the transition point as desired, so that the fluctuation region is absent for d = 3. The width of this region is exponentially small for d = 2:

$$\frac{c}{c_{\text{crit}}} - 1 \sim \left(\frac{\alpha r_0^2}{D}\right)^{-1} \exp\left[-\frac{2}{\gamma} \left(\frac{l}{r_0}\right)^2\right], \qquad (2.5.22)$$

whereas for d = 1 we have

$$\frac{c}{c_{\text{crit}}} - 1 \sim \left(\frac{\alpha r_0^2}{D}\right)^{-1} \gamma^2 \left(\frac{r_0}{l}\right)^4 = \gamma \left(\frac{r_0}{l}\right)^2 \frac{J}{J^*}.$$
 (2.5.23)

Renormalization-goup methods can probably be developed for the description of phenomena within the fluctuation region in one-dimensional media and when  $(l/r_0)^2 \leq \gamma < 1$ .

We emphasize that we have left outside the scope of our

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discussion all fluctuation phenomena during "medium populating" kinetic transitions in the case of strong breeding centers and in the case where, even though the breeding centers are weak, the condition given by (2.3.16) is violated, so that the explosive instability threshold is determined by rare strong clusters. Analysis of fluctuation phenomena in these situations gives rise to serious difficulties.

#### 2.6 Gaussian fluctuations in decay and breeding rates

To conclude this section, let us briefly consider calculations of the explosive instability threshold and analyses of "medium populating" kinetic transitions in systems in which the decay and breeding rates are subject to Gaussian fluctuations.<sup>20</sup>

Let  $K_1(\mathbf{r}, t)$  be the rate of decay of the diffusing material at time t at the point **r** in the medium and  $K_2(\mathbf{r}, t)$  the rate of breeding at the same point. The diffusion equation then takes the form

$$n = -(K_1 - K_2) n + D \Delta n.$$
 (2.6.1)

Generally speaking, the quantities  $K_1$  and  $K_2$  depend on the local concentration  $n(\mathbf{r}, t)$  of the diffusing material. If we confine our attention to low concentrations, we have the linear relation

$$K_1 - K_2 = k_1 - k_2 + \beta n. \tag{2.6.2}$$

We shall examine the situation where the coefficient  $\beta$  is positive, i.e., an increase in the concentration suppresses breeding or increases the rate of decay.

In a closed system, breeding cannot continue indefinitely and must come to an end when the substrate necessary for it to occur becomes exhausted. We shall assume, however, that the system is open and the rate of breeding is maintained by the influx of substrates from outside.

In contrast to Sections 2.1–2.5, where breeding was looked upon as localized at individual centers and the rate of decay was constant throughout the medium, we shall now examine the situation where breeding and decay occur over all space, but the corresponding rates exhibit fluctuations.

Let us suppose that the reaction constants  $k_1$  and  $k_2$ fluctuate in a given fashion in space and time (fluctuations in  $\beta$  may be ignored because the concentration *n* is low). We can then separate the regular and the fluctuating components:

$$k_{1,2} = \overline{k_{1,2}} + \delta k_{1,2} (\mathbf{r}, t), \quad \overline{k_{1,2}} = \langle k_{1,2} \rangle, \quad (2.6.3)$$

and rewrite (2.6.1) in the form

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$$\dot{n} = -\Gamma n - \beta n^2 + D \Delta n + g (\mathbf{r}, t) n,$$
 (2.6.4)

where

$$\Gamma = \overline{k}_1 - \overline{k}_2, \quad g(\mathbf{r}, t) = \delta k_2(\mathbf{r}, t) - \delta k_1(\mathbf{r}, t). \quad (2.6.5)$$

Henceforth, we shall suppose that the coefficient  $\Gamma$  is positive, so that the mean decay rate exceeds the mean breeding rate. The random field  $g(\mathbf{r}, t)$  is external in relation to (2.6.4) because it is independent of the concentration distribution  $n(\mathbf{r}), t$ ). The mean value of this field is zero by definition, and its pair correlation function will be assumed to be given by

the following exponential function of position and time:

$$\langle g(\mathbf{r}, t) g(\mathbf{r}', t') \rangle = S \exp(-r_0^{-1}|\mathbf{r} - \mathbf{r}'| - \tau_0^{-1}|t - t'|).$$

(2.6.6)

We shall assume that the random field  $g(\mathbf{r}, t)$  is Gaussian.

In the absence of fluctuations in the decay and breeding rates (i.e., when S = 0), the explosive instability threshold is observed in the system when these two rates are equal (when  $\Gamma = 0$ ). Fluctuations reduce the explosive instability threshold, and the following problem is therefore interesting. What is the critical intensity  $S_{\rm crit}$  of the fluctuations for which the system reaches the explosive instability threshold despite the fact that the mean decay rate exceeds the mean breeding rate, i.e.,  $\Gamma > 0$ ? What is the behavior of the system for  $S > S_{\rm crit}$  when the nonlinear suppression of explosive instability is taken into account?

These questions were examined in Re. 20. We shall not go into the details of this analysis which, in many ways, is analogous to that given in Sections (2.3)–(2.5). We shall merely reproduce the final results.<sup>11)</sup>

Starting with the parameters  $\Gamma$ , D,  $r_0$  and  $\tau_0$  of the problem, we can construct three characteristic goupings with the dimensions of length:

1) The diffusion length  $r_{\text{diff}} = \sqrt{D/\Gamma}$  gives the mean depth of penetration from the boundary of the medium in the absence of fluctuations.

2) The stationary length  $l = \sqrt{D\tau_0}$  is the mean distance traversed by a diffusing particle in the characteristic lifetime  $\tau_0$  of the individual fluctuation.

3) The correlation length  $r_0$  determines the characteristic spatial size of an individual fluctuation in the decay and breeding rates.

The critical fluctuation intensity  $S_{crit}$  for which the medium attains the explosive instability threshold depends on the relationship between the lengths  $r_{diff}$ , l, and  $r_0$ . When  $r_{diff} \gg r_0 \gg l$ , the critical intensity  $S_{crit}$  is given by the following expression for a medium of any dimensionality (d = 1, 2, 3):

$$S_{\rm crit}^{1/2} = \Gamma\left(\frac{r_{\rm diff}}{l}\right) \,. \tag{2.6.7}$$

On the other hand, when  $r_{\text{diff}} \ge l \ge r_0$ , the expression for  $S_{\text{crit}}$  turns out to be different for different dimensionalities:

$$S_{\rm crit}^{1/2} = \Gamma\left(\frac{r_{\rm diff}}{r_0^{1/2}l^{1/2}}\right), \qquad d = 1, \qquad (2.6.8)$$

$$S_{\text{crit}}^{1/2} = \Gamma\left(\frac{r_{\text{diff}}}{l}\right) \left(\ln\frac{2l}{r_0}\right)^{-1/2}, \quad d = 2, \quad (2.6.9)$$

$$S_{\text{crit}}^{1/2} = \Gamma\left(\frac{r_{\text{diff}}}{l}\right), \qquad d = 3. \qquad (2.6.10)$$

When  $l \gg r_{\text{diff}} \gg r_0$ , the explosive instability threshold is largely determined by rare, strong, positive bursts of the random field  $g(\mathbf{r}, t)$  (compare this with the discussion given in Section 2.4); for this relationship between the characteristic lengths, a simple estimate cannot be obtained for  $S_{\text{crit}}$ . Finally, diffusion is unimportant for  $r_0 \gg r_{\text{diff}}$ , and the problem reduces to

those examined in the introduction to Section 2.

When the nonlinear limiting mechanism described by the term  $\beta n^2$  in (2.6.4) is present, the medium is populated above the explosive instability threshold. In the mean-field approximation, the steady mean concentration  $\bar{n}$  is then given by

$$\overline{n} = \begin{cases} 0, & S < S_{krit} \\ (\Gamma/\beta) \left( \frac{S}{S_{crit}} - 1 \right), & S \geqslant S_{crit} \end{cases}$$
(2.6.11)

In the same approximation, the correlation length  $r_c$  determined by the pair correlation function  $\langle n(\mathbf{r}, t) n(\mathbf{r}', t') \rangle$  is given by

$$r_{\rm c} = r_{\rm diff} \left| 1 - \frac{S}{S_{\rm crit}} \right|^{-1/2}$$
. (2.6.12)

It becomes infinite at the kinetic transition point.

Analysis of the mean-field approximation shows that it is valid for one-deminsional media outside the fluctuation region defined by the inequalities

$$\frac{S}{S_{\text{crit}}} - 1 \leqslant \left(\frac{l}{r_{\text{diff}}}\right)^2, \quad r_{\text{diff}} \gg l \gg r_0, \quad (2.6.13)$$

$$\frac{S}{S_{\text{crit}}} - 1 \leqslant \left(\frac{r_0}{r_{\text{diff}}}\right)^2, \quad r_{\text{diff}} \gg r_0 \gg l.$$
(2.6.14)

The fluctuation region is exponentially narrow [see (2.5.20)] for two-dimensional systems, and can then be neglected. In three-dimensional media, the fluctuation region is not present when  $r_{\text{diff}} \ge l$ ,  $r_0$ , and the mean-field approximation remains valid up to the kinetic transition point itself.

#### 3. Competition processes in fluctuating media

In this section, we examine a further example<sup>17,32,33</sup> of the "medium populating" kinetic transition that involves the phenomenon of competition.

Competition and selection processes are frequently encountered in the behavior of highly nonequilibrium open systems. It will be sufficient to recall the competition between modes in a laser,<sup>5</sup> chemical models of evolution,<sup>34</sup> and models of competitive selection in ecology.<sup>35</sup> In fact, the very process of self-organization, i.e., spontaneous formation of ordered structures,<sup>5</sup> may be looked upon as the result of competition between a large number of unstable growing modes leading to the establishment of the macroscopic amplitude of one of these modes and the suppression of all others.

When the system is subject to the effects of external noise, this may result in a qualitative modification of the competition process, which alters the course of its evolution. We shall not give a general analysis, and will confine our attention to a simple example which is of major biological significance.

As a specific model, we shall use the Lotky-Volterra type equations that describe competition in ecological communities and the effects of prebiological chemical evolution. To be specific, we shall use ecological terminology. A reformulation of the problem to the case of a set of chemical reactions is given in Ref. 33. It will be clear from the ensuing account [see Eqs. (3.6)] that the "ecological model" will

<sup>&</sup>lt;sup>11)</sup>We draw attention to the fact that, as noted in Section 2.4, Eq. (2.5.1) reduces to (2.6.4) in the limit of high concentration (c > 1) of weak breeding centers. The correspondence rules are given by (2.4.8)-(2.4.10).

serve as a prototype of a whole class of models of open systems in which the "medium populating" kinetic transition can take place.

Consider a system consisting of two biological species that compete for the same type of food. We shall suppose that individuals of the weaker species are mobile (capable of diffusion) and the rate of increase in the amount of food available fluctuates in space and time. This system is described by

$$\begin{array}{l}
\dot{N} = (BM - A) N, \\
\dot{n} = (bM - a) n + D\Delta n, \\
\dot{M} = Q - GM - CN - cn + f(\mathbf{r}, t);
\end{array}$$
(3.1)

where N and n the population densities of strong and weak species, and M is the food density. All the coefficients in (3.1)are positive and it is assumed that A/B < a/b.

The first two equations in (3.1) are traditional: the rate of increase of the populations is a linear function of the amount of available food, and negative when there is no food. The second equation in (3.1) contains the additional diffusion term  $D \Delta n$  which represents the mobility of the individuals of the weaker species. The third equation in (3.1)describes the dynamics of the variations in the density of food. Individuals belonging to the two species consume amounts C and c of food per unit time, respectively. The food itself "grows" at a constant rate Q and, even in the absence of the two species, its maximum density is limited by some decay mechanism represented by the term-GM in the above equation. The random Gaussian force  $f(\mathbf{r}, t)$  represents fluctuations in the rate at which the food grows in space and time. We shall assume that the correlation function for this noise is

$$\langle f(\mathbf{r}, t) f(\mathbf{r}', t') \rangle = 2G\Theta \exp\left(-k_{\mathbf{f}} | \mathbf{r} - \mathbf{r}' |\right) \delta(t - t').$$
(3.2)

The parameter  $r_f = k_f^{-1}$  determines the typical spatial size of an individual fluctuation, whereas  $\Theta$  characterizes the fluctuation intensity. It can be shown using (3.1) that, when individuals belonging to both species are absent (N = n = 0), the mean square fluctuation in food density is

$$\langle \delta M^2 \rangle_0 = \Theta. \tag{3.3}$$

We note that the random process  $f(\mathbf{r}, t)$  is asumed to be deltacorrelated in time. This means that its correlation time is much shorter than all the characteristic times of the problem.

When there are no fluctuation ( $\Theta = 0$ ), the only stable stationary solution of (3.1) is<sup>12</sup>

$$n = 0, \quad M = M_1 = \frac{A}{B}, \quad N = N_1 = (Q - GM_1) A^{-1}.$$
 (3.4)

This is the classical result:<sup>35</sup> competition results in the extinction of the weaker species. This is frequently formulated as the Gause theorem, according to which two biological species that totally rely on the same resource cannot coexist in a stationary state.

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The situation is radically different when the rate of growth of the food supply fluctuates both in space and time. The basic result then is that, beginning with a certain critical noise intensity  $\Theta_c$ , it is possible for two competing species to coexist statistically in a stationary state, i.e., a new state is established in which  $\langle n \rangle \neq 0$  and  $\langle N \rangle \neq 0$ . The transition to this state, which is accompanied by the appearance of a non-zero average density  $\langle n \rangle$  that is asymptotic in time and is evaluated over the entire volume, is an example of a "medium populating" kinetic transition.

By making a number of mathematical estimates, it is possible to show<sup>33</sup> that, when there is no diffusion (D = 0), fluctuations in the rate of growth of food do not prevent the asymptotic extinction of the weaker species so that, in contrast to Section 2.5, this particular kinetic transition is fundamentally related to the presence of diffusion.

Our conclusion is important from the standpoint of mathematical ecology. Under the conditions of a fluctuating environment, simple mobility turns out to be an essential factor ensuring evolutionary advantage and the possibility of coexistence with a stronger species. In reality, mobile individuals belonging to the weaker species survive because they are capable of "eating up" the fluctuations!

It is now convenient to introduce the new variables

$$p = M - M_1, \quad q = \ln \frac{N}{N_1},$$
 (3.5)

which describe deviations of the densities M and N from their stationary values given by (3.4) in the absence of fluctuations. We shall suppose that these deviations are small,<sup>13)</sup> so that  $\langle p^2 \rangle \ll 1$  and  $\langle q^2 \rangle \ll 1$ . After linearization in p and q, the model given by (3.1) reduces to the following three equations in terms of the new variables:

$$\left.\begin{array}{l}
q = Bp, \\
\dot{p} = -G'p - vq - cn + f(\mathbf{r}, t), \\
\dot{n} = b(p - p_1)n + D\Delta n,
\end{array}\right\}$$
(3.6)

where

$$p_1 = \frac{a}{b} - \frac{A}{B}, \quad v = CN_1M_1. \tag{3.7}$$

The parameter  $p_1$  characterizes the deficiency of food in the stationary state (3.4) for the propagation of individuals of the weaker species. We shall assume that this parameter is small, i.e., the mean square fluctuations in food density M(and, consequently, in the quantity  $p = M - M_1$ ) may exceed  $p_1$ . Hence, in the third equation in (3.6) we have both the term  $-bp_1n$  and the nonlinear term bpn.

The equations given by (3.6) describe a set of identical damped oscillators of frequency  $\omega_0^2 = vB$ , located at each point of space and interacting through the field  $n(\mathbf{r}, t)$ . When all the oscillators are in the unexcited state, this field decays exponentially to the value n = 0. Excitation of the oscillators by the random force  $f(\mathbf{r}, t)$  provides us, however, with the possibility of the "propagation" of this field. The spatial structure of the propagation regions (in which  $p > p_1$ ) varies randomly in space and time. A kinetic transition may take

ΠE

<sup>&</sup>lt;sup>12)</sup>We are assuming that Q > GA / B.

<sup>&</sup>lt;sup>13)</sup>This assumption imposes definite restrictions on the intensity of noise  $f(\mathbf{r}, t)$ , which will be discussed below.

place if the increase in n in the propagation regions begins to compensate the reduction in the field n outside such regions.

It is important to note that (3.6) has a much wider range of validity than the original model (3.1) from which (3.6) was obtained. Actually, these equations constitute an example of a distinctive excitable medium that can undergo a transition to a qualitatively new "organized" state under the influence of external noise, where the field *n* that couples the oscillators is nonzero and decreases in time. This situation may arise in different problems.

The analysis that follows is performed in close analogy with the theory of second-order equilibrium phase transitions. The field  $f(\mathbf{r}, t)$  is looked upon as the order parameter, and we perform an adiabatic elimination of the rapidly oscillating variables that are subject to it. This eventually yields a Ginzburg-Landau type equation for the slow component of the order parameter.

It is readily seen that the spatial Fourier modes of the field *n* with large values of the wave vector **k** are damped out more rapidly than the corresponding modes with small **k**. This occurs as a result of diffusion. There is also fast motion of the oscillators (q, p), and this naturally allows us to perform a separation of fast and slow variables. The characteristic "microscopic" scales in (3.6) with dimensions of the reciprocal of time are  $bp_1$ , G,  $\omega_0$  and  $Dk_f^2$ . Slow variables can be determined as the time averages of n, p, and q within a time interval T chosen so that  $(1/T) \triangleleft bp_1$ , G,  $\omega_0$ ,  $Dk_f^2$ . In particular, we take

$$\eta(\mathbf{r}, t) = \frac{1}{T} \int_{0}^{T} n(\mathbf{r}, t+\tau) d\tau \equiv \langle n(r, t) \rangle_{T}$$
(3.8)

and, similarly,  $\tilde{q} = \langle q \rangle_T$ ,  $\tilde{p} = \langle p \rangle_T$ . Fast variables are sometimes defined by  $\delta n = n - \eta$ ,  $\delta p = p - \tilde{p}$ ,  $\delta q = q - \tilde{q}$ .

Our aim is to obtain a closed equation for the slow component  $\eta$  of the order parameter *n*. We begin the derivation by taking the time average (3.8) of the third equation in (3.6), which yields

$$\dot{\eta} = -bp_1\eta + b\widetilde{p}\eta + D \ \Delta \eta + b \ (\delta p \ \delta n)_T. \tag{3.9}$$

The slow component p can be expressed in terms of  $\eta$  by taking the time average of the first two equations in (3.6), noting that the averaging time T is much longer than all the "microscopic" time scales. In view of this, we find that

$$\widetilde{p} = -\frac{c}{\omega_0^2} \dot{\eta} + \frac{1}{\omega_0^2} \dot{\widetilde{f}}, \qquad (3.10)$$

where the slow component  $\overline{f}$  of the random force is defined by  $\overline{f} = \langle f \rangle_T$ .

The equations for the fast components have the form

$$\begin{split} \dot{\delta q} &= B \delta p, \\ \dot{\delta p} &= -G \delta p - v \delta q - c \delta n + \delta f(\mathbf{r}, t), \\ \dot{\delta n} &= -b \left( p_1 - \widetilde{p} \right) \delta n + b \eta \delta p + D \Delta \delta n \\ &+ b \left( \delta n \delta p - \langle \delta n \delta p \rangle_T \right). \end{split}$$

$$(3.11)$$

The slow variables  $\eta$  and  $\tilde{p}$  must be looked upon here as given external parameters, so that the quasistationary distribution of fast variables adjusts itself adiabatically to the slow evolu-

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tion of the variables  $\eta$  and  $\tilde{p}$  in time. The time average can therefore be replaced with the statistical average  $\langle \delta n \ \delta p \rangle_{\eta}$ evaluated with the stationary fast-variable probability distribution that is established for given  $\eta$  and p.

The averages  $\langle \delta n^2 \rangle_{\eta}$  and  $\langle \delta n \, \delta p \rangle_{\eta}$  can be determined by using an approximation analogous to the self-consistent field theory for second-order phase transitions. If

$$\sqrt{\langle \delta n^2 \rangle_{\eta}} \ll \eta, \tag{3.12}$$

the validity of which is verified below, the only nonlinear term  $b (\delta n \, \delta p - \langle \delta n \, \delta p \rangle)$  in (3.11) is, on average, small in comparison with the term  $b\eta \, \delta p$  in the same equation, and can be neglected.<sup>14)</sup> As a result, (3.11) reduces to a set of stochastic linear differential equations for which one can calculate both the stationary probability distribution and all the correlation functions corresponding to it.

If we evaluate the correlator  $\langle \delta n \, \delta p \rangle_{\eta}$  in this way, we obtain it in the form of a certain function of  $\eta$ . Since  $\eta$  is small near the transition point, we can expand  $\langle \delta n \, \delta p \rangle_{\eta}$  in powers of  $\eta$ , retaining only terms up to the second order. Substituting the expansion in (3.9), we obtain the Ginzburg-Landau equation for the "medium populating" transition:

$$\eta = -b (p_1 - \mu \Theta) \eta - R\Theta \eta^2 + D \Delta \eta + \Phi (\mathbf{r}, t) \eta.$$
(3.13)

If we suppose that

$$Dk_1^2 \gg \omega_0 \gg G \gg bp_1 \tag{3.14}$$

and that the medium is three-dimensional, we find that

$$\mu = \frac{b}{Dk_f^2},\tag{3.15}$$

$$R = \frac{3\sqrt{2}}{\cdot 4} \frac{b^{3c}}{G\omega_{0}^{1/2} (Dk_{1}^{2})^{3/2}}$$
(3.16)

which are independent of  $p_1$ .

The random Gaussian force  $\Phi$  in (3.13) is defined by

$$\Phi(\mathbf{r}, t) = \frac{1}{\omega_0^2} \frac{\partial}{\partial t} \widetilde{f}(\mathbf{r}, t)$$
(3.17)

and its spectrum is

$$\langle \Phi(\omega, \mathbf{k}) \Phi(\omega', \mathbf{k}') \rangle = S(\omega, \mathbf{k}) \delta(\mathbf{k} + \mathbf{k}') \delta(\omega + \omega'), \quad (3.18)$$

$$S(\omega, \mathbf{k}) = \begin{cases} 0 & |\omega| > \frac{2\pi}{T}, \\ 2G\Theta\left(\frac{\omega}{\omega_{e}}\right)^{2} h_{3}(\mathbf{k}), & |\omega| < 2\pi/T, \end{cases}$$
(3.19)

where

$$h_{3}(\mathbf{k}) = \frac{8\pi k_{f}}{(k^{2} + k_{f}^{2})^{2}}$$

is the Fourier transform of the function  $\exp(-k_f r)$ .

It is clear from (3.14) that, when the noise intensity reaches the critical value given by

<sup>&</sup>lt;sup>14</sup>Further calculations show that, near the transition point, i.e., when the fluctuations  $\delta p$  are large in comparison with  $p_1$ , we can actually neglect the term  $b (p_1 - \bar{p}) \delta n$  in this equation, as well.

$$\Theta_{\rm c} = \frac{p_1}{b} Dk_{\rm f}^2 \tag{3.20}$$

kinetic transition occurs in the medium. Near the transition point, the volume average of the density of the weak species is given by

$$\langle n \rangle = \begin{cases} 0, & \Theta < \Theta_{\rm c}, \\ \frac{bp_1}{R} \left( \frac{1}{\Theta_{\rm c}} - \frac{1}{\Theta} \right), & \Theta \geqslant \Theta_{\rm c}. \end{cases}$$
(3.21)

We draw attention to the fact that the critical noise intensity  $\Theta_c$  is proportional to the diffusion coefficient D and increases with increasing mobility.<sup>15)</sup> This has a simple explanation. If the mobility is too large, weak individuals traverse the breeding region in which  $p > p_1$  very rapidly, and cannot efficiently use the food excess available in the propagation region. If we take (3.3) into account in (3.20), we may also write

$$(\langle \delta M^2 \rangle_{0.\,\mathrm{crit}})^{1/2} = p_1 \frac{l}{r_f},$$
 (3.22)

where  $l = (D/bp_1)^{1/2}$  is the average displacement by diffusion during the extinction time  $(bp_1)^{-1}$  of the weak species in the stationary state (3.4). Thus, at the transition threshold, the root mean square fluctuation in food density  $(\langle \delta M^2 \rangle)^{1/2}$  is not simply equal to the food shortage  $p_1$  for the weak species, but exceeds it by a factor equal to the ratio of the diffusion length l to the mean radius  $r_f$  of an individual reproduction region.

At the transition point, the correlation length  $r_c$  determined from the reaction of the system to the introduction of the external source of density n is found to diverge:

$$r_{\rm c} = l \left( \mathbf{1} - \frac{\Theta}{\Theta_{\rm c}} \right)^{-1/2}. \tag{3.23}$$

There is also the critical slowing down effect. We note that (3.13) itself is valid only near the transition point, when the characteristic relaxation time

$$\mathbf{r}_{c} = (bp_{i})^{-1} \left(\mathbf{1} - \frac{\Theta}{\Theta_{c}}\right)^{-1}$$
(3.24)

is much greater than the "microscopic" time scales of the problem.

The above analysis was based on the assumption that fluctuations in the density *n* were relatively small. Let us now check the validity of this assumption. Starting with the linearized equation (3.11), we can readily determine the mean square  $\langle \delta n^2 \rangle_{\eta}$ . Near the transition point, when  $\eta$  is sufficiently small, this mean square is given by<sup>16</sup>

$$\langle \delta n^2 \rangle_{\eta} = \Theta b^2 \sqrt{\frac{2}{\omega_v (Dk_f^2)^3}} \, \eta^2. \tag{3.25}$$

Substituting for  $\Theta$  in this expression the critical noise intensity  $\Theta_c$ , we find that

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$$\frac{\left(\left(\delta n^{2}\right)_{\eta}\right)^{1/2}}{\eta} = 2^{1/4} \left(\frac{bp_{1}}{\omega_{0}}\right)^{1/2} \left(\frac{\omega_{0}}{Dk_{1}^{2}}\right)^{1/4}.$$
(3.26)

Consequently, when (3.14) is satisfied, the condition for weak fluctuations (Ginzburg-Levanyuk criterion) is valid up to the transition point itself. We arrived at a similar conclusion in Section 2.5 when we discussed another example of a "medium populating" kinetic transition in the three-dimensional case.

The partially linearized equations (3.6) were obtained on the assumption that  $\langle q^2 \rangle \ll 1$  and  $\langle p^2 \rangle \ll 1$ . It is readily verified that this is satisfied if  $\Theta_c \ll \min(v/B, A/B)$ . The last condition imposes a definite limitation on coefficients in the original model.

# 4. DIFFUSION IN A MEDIUM WITH RANDOMLY DISTRIBUTED TRAPS

In the preceding sections, we examined critical phenomena in media in which either the breeding rates had randomly fluctuating increments or the breeding process itself occurred in randomly appearing breeding centers. We have shown that explosive instability can occur in the medium even if, on average, decay predominates over breeding. However, there are frequent situations where traps are randomly distributed within a medium and can capture particles of the diffusing material. Such problems arise when one examines the trapping of excitons by impurities in crystals,<sup>38</sup> in the theory of diffusion-controlled reactions,<sup>37</sup> and in a number of other applications. We shall examine such problems below, concentrating our attention on manifestations of randomness in the distribution of traps in a medium.

Let us suppose that particles diffusing through a medium can be captured by traps distributed randomly throughout the medium, and are thus removed from the subsequent diffusive random walk. In the continuum limit, the diffusing material is described by the local particle density field  $n(\mathbf{r}, t)$ , whose variation in time is described by

$$n = -\alpha (\mathbf{r}) n + D \Delta n.$$
 (4.1)

The coefficient  $\alpha$  is proportional to the local trap density and characterizes the rate of loss of particles. Traps are located at randomly distributed independent points  $\{\mathbf{r}_j\}$  in the medium, so that

$$\alpha (\mathbf{r}) = \sum g (\mathbf{r} - \mathbf{r}_j). \tag{4.2}$$

The mean number of traps per unit volume is m.

We note that the model defined by (4.1)–(4.2) will also describe a medium with randomly-distributed decay centers.

Since there is no breeding, the mean density  $\langle n \rangle$  of the material, evaluated over the volume, tends to zero as time increases. The problem is to find  $\langle n \rangle$  as a function of time, given that the initial particle distribution is homogeneous, i.e., $n(\mathbf{r}, 0) = n_0$ .

It might be thought that the decay process will be exponential and characterized by a mean decay constant  $\langle \alpha \rangle$ :

$$\langle n(t) \rangle = n_0 \exp(-\langle \alpha \rangle t).$$
 (4.3)

<sup>&</sup>lt;sup>15)</sup>The quantity D has a lower bound set by (3.14), and the expression given by (3.20) was derived on the assumption that this condition was valid.

<sup>&</sup>lt;sup>16)</sup>We recall once again that the fluctuations that we are examining are of external origin, so that the correlator of the density fluctuations does not diverge when the arguments are equal.

However, this is not correct, as can be seen from the following considerations.

When traps are distributed randomly, it is always possible to find regions that are free of traps. Particles found in the interior of large empty regions will be lost only because the diffusive random walk of any such particle will take it outside the limits of such a region. Hence, the number of particles originally found in the interior of sufficiently large empty regions will decrease very slowly. It is precisely these particles that eventually determine the temporal evolution of the mean density  $\langle n(t) \rangle$ .

The importance of these rare "empty" regions that are free of traps was first noted by Balagurov and Vaks,<sup>38</sup> who also proposed a method of determining the mean density  $\langle n \rangle$ as a function of time. This method was based on the analogy with the quantum-mechanical motion of a particle in a random potential. This problem has recently been examined in Ref. 39, which is virtually a repetition of the derivation given in Ref. 38.

If we write (4.1) in the form

$$\dot{-n} = \mathcal{H}n \tag{4.4}$$

with the linear operator

$$\mathscr{H} = -D\Delta + \alpha (\mathbf{r}), \qquad (4.5)$$

we reduce the problem, as in Section 2, to the study of the properties of the Schrödinger equation with imaginary time and random potential. However, in contrast to the situation we have encountered in connection with the explosive instability threshold in a medium with breeding centers, the potential  $\alpha(\mathbf{r})$  in (4.5) is nonnegative [see (4.2)]. From this, it follows that all the eigenvalues  $E_i$  of the equation

$$\mathscr{H} \psi_l = E_l \psi_l, \qquad (4.6)$$

are also nonnegative, i.e., the spectrum of the operator  $\mathcal{H}$  has the lower limit E = 0. This property of the spectrum persists for any random realization of the field  $\alpha(\mathbf{r})$ . According to the terminology adopted in the theory of the Schrödinger equation with a random potential,<sup>14</sup> the value E = 0 gives the finite fluctuational limit of the spectrum.

The spectrum of the operator  $\mathcal{H}$  is discrete near the E = 0 limit. The eigenvalues  $E_i$  lying near this limit are determined by fluctuations of the potential for which  $\alpha$  is close to zero in large regions of space, i.e., by "empty" regions free of traps. It has been shown<sup>14</sup> that, for a Poisson random field of the form given by (4.2), the probability of finding a fluctuation with the lowest level E close to zero is exponentially small and, apart from preexponential factors, can be estimated for a medium of dimensionality d from the formula

$$p(E) = \exp(-\mu_d m E^{-d/2}),$$
 (4.7)

where

$$\mu_d = \frac{\pi^{d/2}}{\Gamma\left(\frac{d}{2}+1\right)} \varepsilon_d^{d/2},\tag{4.8}$$

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and  $\varepsilon_d$  is the ground-state energy in the *d*-dimensional potential well of unit radius and infinitely high walls.

The general solution of (4.1) can be written in the form of an expansion<sup>17)</sup> in terms of th eigenfunctions of the above operator:

$$n(\mathbf{r}, t) = \sum_{l} C_{l} \psi_{l}(\mathbf{r}) \exp(-E_{l}t) + \int C_{l} \psi_{l}(\mathbf{r}) e^{-E_{l}t} dl$$
(4.9)

where the coefficients  $C_1$  are determined by the initial condition  $n(\mathbf{r}, 0) = n_0$ :

$$C_l = n_0 \int \psi_l^* (\mathbf{r}) \, \mathrm{d}\mathbf{r}. \tag{4.10}$$

Terms in 4.9) that correspond to the discrete spectrum with  $E_i$  close to zero decrease to zero more slowly than any other terms. The eigenfunctions corresponding to such values of  $E_i$  are localized on individual fluctuations of the field  $\alpha(\mathbf{r})$  which constitute large regions free of traps. Because they are rare, such fluctuations must be considered separately. Apart from the preexponential factor, the mean density  $\langle n \rangle$  evaluated over the entire volume can be estimated from the following expression as  $t \to \infty$ :

$$\langle n(t)\rangle = n_0 \int_0^\infty p(E) e^{-Et} dE,$$
 (4.11)

where the probability p(E) is given by (4.7).

Evaluation of the integral by the method of steepest descents yields

$$\langle n(t) \rangle = n_0 \exp\left[-v_d \left(Dtm^{2/d}\right)^{d/(d+2)}\right],$$
 (4.12)

where  $v_d$  is a numerical factor that depends on the dimensionality of the medium.

Thus, inclusion of the contribution due to empty regions leads to the replacement of the simple exponential law (4.3) with (4.12) which is characterized by a slower falling off with time. Detailed analysis shows that (4.12) is valid for  $t \gg m^{-2/d} D^{-1}$ .

The mean square displacement of a particle in a time t was estimated in Ref. 39 in the limit of large t, and it was shown that

$$\langle r^{\mathbf{r}}(t) \rangle \sim \left(\frac{Dt}{m}\right)^{2/(d+2)}.$$
 (4.13)

It was assumed in the calculation that, if a particle is captured by a trap, its coordinates remain constant. The mean square displacement increases with time because some particles do not reach a trap in time t. Long displacements correspond to particles found in the interior of large regions free of traps.

A numerical simulation of this system was also performed in Ref. 39. The authors investigated the random walk on a square lattice with traps occupying one-eighth of the total number of lattice sites. Particles were allowed to jump to one of the four nearest-neighbor sites in successive instants of time. Having hit a site occupied by a trap, the particle was absorbed and computer simulation of the process showed that, for large t, the mean square displacement  $\langle r^2(t) \rangle$  and the logarithm of the mean number of free particles were proportional to the square root of time. This is in agreement with predictions given by (4.12) and (4.13). At the same time, a considerable discrepancy was found between the numerical coefficients and the theoretical results. The dimensionality d of the medium plays an important role in the case that we have considered, as it does in any diffusion problem. It is clear from (4.12) that  $|\ln (n(t))\rangle | \sim t^{d/(d+2)}$ . This is why the result  $|\ln \langle n(t) \rangle| \sim t$  obtained in the mean-field approximation reappears in the formal limit <sup>18)</sup> as  $d \rightarrow \infty$ . The reason for this is that, as the dimensionality of the medium is increased, diffusion becomes increasingly more effective, and the probability that a particle will remain in the interior of a region free of traps for a sufficiently long interval of time will decrease. In the same limit, the mean square displacement (4.13) offer a long time will eventually cease to depend on time.

It has recently been noted<sup>40</sup> that statistical configuration effects similar to those discussed in this section play an important role in the long-term asymptotic behavior of the kinetics of two-body reactions of the form  $A + B \rightarrow C$ .

### **5. CONCLUSION**

From the mathematical standpoint, the problems that arise in connection with studies of critical phenomena in media with random breeding are no less interesting and profound than classical problems in the theory of stationary disordered media. We have attempted in this review, with the aid of a number of simple models, to demonstrate the specificity of these problems, and have carried out a classification of qualitatively different types of critical behavior in such systems. There are many unsolved problems, and in fact only the simplest questions have been answered. Many interesting results are expected when the renormalization group and the  $\varepsilon$ -expansion are applied to these problems. The construction and analysis of discrete percolation-type schemes providing a closer approximation to the phenomena observed in continuum models with random breeding would be exceedingly interesting. Finally, the specificity of many particular applications will require partial modification of the models that we have discussed.

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<sup>&</sup>lt;sup>18)</sup>Problems involving diffusion in multidimensional media arise, for example, in mathematical models of biological evolution.<sup>41,42</sup>

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