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Renormalization-group approach to simple reaction-diffusion phenomena

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A field-theoretical model describing simple one-species reaction-diffusion systems [$A + A \rightarrow O$ (inert) or $A + A \rightarrow A$] with an external source is analyzed from a renormalization-group point of view. It is shown that when the dimension of the system is larger than the upper critical dimension $d_u = 2$, the behavior of the system is governed by a trivial fixed point dominated by diffusion. Below the upper critical dimension, a line of fixed points governs the behavior. Reaction and diffusion processes play an equally important role resulting in a so-called anomalous kinetic behavior. This approach confirms previous scaling arguments. Possible generalizations to more complicated models are discussed.

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I. INTRODUCTION

Simple reaction-diffusion models in which chemical species diffuse in a solvent and react have been the subject of a vast body of work. Although very simple, these nonequilibrium systems have nontrivial dynamics. Among many possible reaction-diffusion processes [1], we shall mainly consider here two particular cases, namely, the annihilation reaction [2]: $A + A \rightarrow O$ (inert) and the coagulation reaction [3]: $A + A \rightarrow A$. We shall also make several comments on the closely related two-species annihilation reaction [4]: $A + B \rightarrow O$ (inert).

Different situations occur depending on the initial conditions. The simplest case is the one-species problem in which the A particles are initially randomly distributed. The number of particles decreases algebraically in time. Moreover, if the dimensionality of the system is smaller than an upper critical dimension d_u, the fluctuations play an important role and the predictions given by the usual rate equations (law of mass action) are not correct [2]. The situation is similar for the two-species problem, providing that the initial numbers of particles of both species are the same [4]. This non-mean-field behavior is called anomalous kinetics [5].

For the two-species problem, a more complicated situation arises in the case where the two constituents are initially spatially separated. A reaction front appears [6], the properties of which are strongly affected by the fluctuations below an upper critical dimension d_{uf} [7, 8].

Let us now return to the case $A+A \rightarrow O$. The simplest question concerns the time dependence of the number of particles. The conventional approach consists in writing a rate equation for the density $n(t)$ of particles at time t:

$$\partial_t n(t) = -kn^2(t), \tag{1}$$

where k is the reaction rate. In the long time limit one finds $n(t) \sim 1/kt$. However, the conventional approach does not take the fluctuations into account, and simple heuristic arguments [2] show that

$$n(t) \sim t^{-\alpha}, \quad \alpha = \min(1, d/2). \tag{2}$$

A systematic study of the role played by fluctuations in this model has been undertaken along several lines. Rigorous studies of the asymptotic decay have been reviewed by Bramson and Lebowitz [9]. For one-dimensional systems, the problem has been exactly solved by several means [1, 10–12]. In all these approaches one assumes that the particles are moving on a discrete lattice. The master equation associated with this reaction-diffusion process is similar to the one describing the kinetics of an Ising chain with Glauber dynamics at zero temperature [1, 10, 12]. Both problems can also be mapped onto a quantum chain Hamiltonian which can be transformed into a free fermion problem [1]. In more than one dimension, the mapping onto a quantum system is always possible; however, this quantum problem can no longer be exactly solved.

An interesting generalization of the model is obtained by adding source terms [10]. Single particles are created at a rate h per lattice site. In the large-time limit a steady state is reached in which particle production is balanced by diffusive annihilation. The $h \rightarrow 0$ limit can be considered as a critical limit in the sense that the steady-state particle density n and the relaxation time τ governing the homogeneous density fluctuations behave as

$$n(h) \sim h^{1/\delta}, \qquad \tau(h) \sim h^{-\Delta}. \tag{3}$$

Scaling arguments show that $\alpha = 1/\delta\Delta$ and $\Delta + 1/\delta = 1$, offering an alternative way of computing α exactly [10, 13, 14].

However, one would like to have a unifying approach, allowing one to predict the scaling behavior and the role of the fluctuations for all the models in arbitrary dimension.

A good strategy is to develop a field-theoretical description of these reactions by means of a path-integral representation of birth-death processes introduced by Doi [15, 16]. This approach has been used by Peliti [17, 18] to study the annihilation reaction $A + A \rightarrow O$ as well as the coagulation reaction $A + A \rightarrow A$ without sources. The

corresponding Lagrangians, in the continuous limit, can be renormalized to all orders in perturbation theory by resummation of the "parquet" diagrams. The two reactions belong to the same universality class: the exponent α is the same for both models and agrees with previous predictions.

Recently, Friedman, Levine, and O'Shaughnessy [19] made an attempt to calculate the explicit time dependence of the particle density in the case of the annihilation reaction $A + A \to O$, using renormalized perturbation theory. They have found that besides the reaction rate, which can be exactly renormalized, a second parameter, proportional to the product of the initial particle density and time, should be treated to all orders in perturbation theory. This complicated problem has not been solved in a totally satisfactory way. Indeed, at $d = 2$, the expression proposed by Friedman *et al.* on the basis of a low-order perturbative calculation and renormalization-group arguments reproduces known results at long times, but the description of short-time behavior remains inconsistent.

In this paper we propose a different approach. Instead of investigating time-dependent quantities for the model without sources, we study the stationary state reached in the presence of external sources. A simple and consistent renormalization-group approach is obtained. Scaling arguments relate these two problems.

Some of the results with sources, in particular the exponent δ, have been previously also derived by Mikhailov [14], using the quantum field-theory method.

The paper is organized as follows. In Sec. II we define the model and discuss the associated field theory. Section III is devoted to the renormalization-group analysis. Section IV is reserved for conclusions.

II. FIELD THEORY FORMALISM

The way to describe a reaction-diffusion process in terms of a field theory is by now well established [15–18]. Thus we shall only summarize the key steps without going into too many details.

One starts with a master equation describing the reaction-diffusion process on a lattice. As the number of particles is not conserved, it is convenient to introduce a Fock-space representation. To each site of the lattice one associates annihilation and creation operators which obey the usual commutation relations. A basis of the Fock space is formed by the vectors $|n_1, n_2, \ldots\rangle = |\underline{n}\rangle$, where n_j is the occupation number of the lattice site j. The macroscopic state $|\Phi\rangle$ corresponding to the probabilities $\phi(\underline{n})$ of finding the system in the state $|\underline{n}\rangle$ is given by $|\Phi\rangle = \sum_{\underline{n}} \phi(\underline{n})|\underline{n}\rangle$. The equation of evolution for $|\Phi\rangle$ can be formally written as $|\Phi(t)\rangle = \mathcal{U}_{t,t_0}|\Phi(t_0)\rangle$. It turns out to be useful to consider a representation of this Fock space in terms of a Hilbert space of the generating functions. The time evolution operator can then be cast in a path integral form [17], which, in the (coarse-grained) continuous limit, takes the following form:

$$\mathcal{U}_{t,t_0} = \int \mathcal{D}\eta\mathcal{D}\hat{\eta} \exp - \int_{t_0}^{t} dt\mathcal{L}(\eta, i\hat{\eta}), \tag{4}$$

where $\mathcal{L}$ is the Lagrangian. The field $\eta(\mathbf{r}, t)$ corresponds (only approximately) to the local particle density, while the auxiliary field $\hat{\eta}(\mathbf{r}, t)$ has no particular physical meaning.

We shall consider a model in which the following two processes can occur:

(a) The annihilation process: $A + A \to 0$, with reaction rate k.

(b) The coagulation process: $A + A \to A$, with reaction rate g.

Particles diffuse and react locally. Moreover, a homogeneous steady state with a finite density of A particles will be reached when a source is introduced, producing particles with a homogeneous time-independent rate h. The corresponding Lagrangian is

$$\mathcal{L} = \int d^d r \left[i\hat{\eta}\left(\frac{\partial \eta}{\partial t} - D\Delta\eta + Dv\eta^2 - h\right) + Du(i\hat{\eta})^2\eta^2 \right], \tag{5}$$

where D is the diffusion constant, and h is the homogeneous constant source. u and v are linear combinations of the reaction rates, namely,

$$Dv \sim g + 2k, \qquad Du \sim g + k. \tag{6}$$

The explicit forms of the proportionality factors arising from the passage, from the discrete lattice level to the coarse-grained continuous limit, are unimportant. The presence of a term proportional to $\hat{\eta}^2$ in the Lagrangian is very important. Indeed, if this term were absent, only the paths $\eta(\mathbf{r}, t)$ being solutions of the diffusive rate equation would contribute to the path integral, and important correlations would be lost.

Explicit calculations can be carried out using standard perturbation expansion [17] for $\mathcal{U}_{t,t_0}$ in powers of the reaction rates u and v. The starting point is a Gaussian approximation, which retains only the diffusive motion of the particles. Thus the unperturbed Lagrangian reads

$$\mathcal{L}_0 = \int d^d r \left[i\hat{\eta}\left(\frac{\partial \eta}{\partial t} - D\Delta\eta)\right)\right]. \tag{7}$$

Diagrams are built up with the vertices Du and Dv and the bare propagator G_0, the Fourier-Laplace transform of which is given by $G_0^{-1}(k, s) = s + Dk^2$. One can use simple dimensional analysis to determine the upper critical dimension d_u below which the reaction rates become relevant. Fixing the scales of length and time, respectively, by $1/\Lambda$ and $1/(D\Lambda^2)$, where Λ is the cutoff imposed by the lattice, one finds $[\eta] = \Lambda^d$, $[\hat{\eta}] = 1$, $[u] = [v] = \Lambda^{2-d}$, hence $d_u = 2$.

III. RENORMALIZATION-GROUP ANALYSIS

The renormalization-group analysis follows the usual Wilson-like procedure [20]. First, we eliminate the Fourier components of the fields belonging to the shell $\Lambda/b < k < \Lambda$. Then, we rescale the lengths and time according to $k' = bk$, $t' = b^{-z}t$. Fields and vertices are rescaled as required by their dimensions. Since no two-leg diagram is generated, fields do not have an anomalous

dimension and thus $\eta' = b^d\eta$, $\hat{\eta}' = \hat{\eta}$. Moreover, the new couplings of the form $\hat{\eta}^m\eta^n$ with $n \geq m$, which are absent in the initial Lagrangian and generated during the renormalization procedure, are irrelevant.

The diagrams contributing to the renormalization of u and v are given by the bubble series shown in Figs. 1(a) and 1(b). In the limit $b = e^l \to 1$, diagrams with n loops are proportional to l^n. Thus only one-loop diagrams contribute to the differential recursion relations, which read

$$\frac{dD}{dl} = (z-2)D, \tag{8}$$

$$\frac{du}{dl} = u\left((2-d) - \frac{u}{2}K_d\Lambda^{d-2}\right), \tag{9}$$

$$\frac{dv}{dl} = v\left((2-d) - \frac{u}{2}K_d\Lambda^{d-2}\right), \tag{10}$$

where $K_d = [(2^{d-1}\pi^{d/2}\Gamma(d/2))]^{-1}$.

The diffusion coefficient D, which was used to fix the time scale, can be kept finite by choosing $z = 2$. The above recursion relations have a unique stable fixed point for u:

$$u^* = \begin{cases} 0\,, & \text{if } d \geq 2\,; \\ \dfrac{2(2-d)}{K_d\Lambda^{(d-2)}}\,, & \text{if } d < 2\,. \end{cases} \tag{11}$$

For $l \to \infty$, v approaches a nonuniversal asymptotic value, depending on the initial value of the parameters. Indeed, combining the recursion relations of u and v we find that the ratio u/v is invariant under renormalization, i.e.,

$$v(l) = \frac{v(0)}{u(0)}u(l). \tag{12}$$

In $d > 2$, the trivial fixed point $u^* = v^* = 0$ is stable. This corresponds to the limiting case in which the reaction is very slow in comparison with the diffusion. Thus the diffusion mixes the particles efficiently enough so that a homogeneous reaction takes place for which the law of mass action is valid.

In $d < 2$, the behavior of the system is governed by the nontrivial fixed points. We have, in fact, a line of fixed points parametrized by the initial values $v(0)$, $u(0)$. On this fixed line, diffusion and reaction proceed at a comparable rate, resulting in an interplay of the two processes, hence the breakdown of the law of mass action.

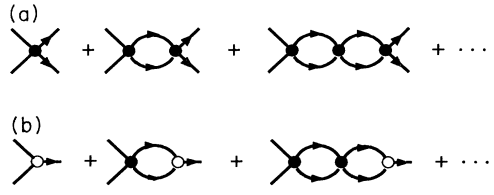

FIG. 1. (a) and (b) Bubble diagrams contributing to the renormalization of u and v. The black (white) dots correspond respectively to the four-point vertex (Du) and the three-point vertex (Dv), the internal line to the bare propagator G_0.

Let us now consider the problem with sources. Our main goal is to characterize the steady state, which develops in the presence of a homogeneous source. The source h has the same dimension as $\hat{\eta}$, and its fixed-point value is $h^* = 0$. It is a relevant term in the vicinity of both fixed points. Thus the source term plays a role similar to the magnetic field for an Ising model. Apart from a simple rescaling, no renormalization occurs to linear order in h. Thus in the vicinity of the fixed points, the recursion relation for h is

$$\frac{dh}{dl} = (d+2)h + O(h^2). \tag{13}$$

(i). Scaling theory for d<2.

Knowing the recursion relations and their linearized forms near the fixed points, we can write the scaling forms of the physical quantities of interest. The density of particles n rescales as follows:

$$n(t,h,u,v) = e^{-dl}n(e^{-zl}t, h(l), u(l), v(l)), \tag{14}$$

where $z = 2$ and $h(l)$,$u(l)$,$v(l)$ are solutions of the recursion relations with the initial conditions $h(0) = h$, $u(0) = u$, $v(0) = v$.

We choose $l = l^*$ such that $h(l^*) = e^{(d+2)l^*}h = 1$. For sufficiently small values of h $u(l^*) \simeq u^*$, $v(l^*) \simeq v^*$, and one finds that

$$\begin{aligned} n(t,h,u,v) &\simeq h^{d/(d+2)}n(h^{2/(d+2)}t, 1, u^*, v^*) \\ &\equiv h^{d/(d+2)}\mathcal{F}\left(h^{2/(d+2)}t\right). \end{aligned} \tag{15}$$

With the assumption that the scaling function $\mathcal{F}(y)$ approaches a finite value when $y \to \infty$, we obtain in the limit $t \to \infty$,

$$n \sim h^{1/\delta}, \qquad \delta = 1 + 2/d. \tag{16}$$

The characteristic time to approach the steady state, τ, is determined by the condition $\tau h^{2/(d+2)} \sim 1$, i.e.,

$$\tau \sim h^{-\Delta}, \qquad \Delta = 2/(d+2). \tag{17}$$

Finally, in the limit $h \to 0$, n should be independent of h. This requires that $\mathcal{F}(y) \simeq y^{d/2}$ for $y \ll 1$; hence $n(t) \sim t^{-\alpha}$, $\alpha = d/2$.

One recovers the scaling laws postulated phenomenologically by Rácz [10],

$$\alpha = \frac{1}{\Delta\delta}, \qquad \Delta + \frac{1}{\delta} = 1. \tag{18}$$

In order to check the assumptions made on the scaling function we calculated the exponents δ and Δ to lowest order in $\epsilon = 2 - d$, using standard expansion techniques, and found agreement with the above results.

(ii). Scaling theory for d ≥2.

In these cases the asymptotic behavior is controlled by the trivial fixed point $u^* = v^* = 0$. Nevertheless, it is clear that the reactions play a vital role in developing a steady state with finite density in the presence of sources; therefore u and v must be kept finite. In other words, in contrast to the the case $d < 2$, it is not allowed to approximate them by their fixed-point values for $l \gg 1$. Using the terminology of critical phenomena, u and v are

dangerous irrelevant variables [21].

Let us consider in detail the marginal case $d = 2$. The steady-state value of the density obeys the scaling relation

$$n(h,u,v) = e^{-2l} n(h(l),u(l),v(l)), \tag{19}$$

where $h(l) = e^{4l}h$ and, for large l, $u(l) \simeq 4\pi/l$, $v(l) \simeq 4\pi v/ul$. For small h, we choose $l = l^*$ such that $h(l^*) = e^{4l^*}h = 1$. Moreover, for a source of amplitude of order unity, the steady-state density takes its mean-field value, determined by the law of mass action:

$$h(l^*) - Dv(l^*)n^2(h(l^*),u(l^*),v(l^*)) = 0. \tag{20}$$

This yields

$$n(h,u,v) = e^{-2l^*}\sqrt{\frac{h(l^*)}{Dv(l^*)}} \sim e^{-2l^*} l^{*1/2} \sim h^{1/2}|\ln h|^{1/2}, \tag{21}$$

i.e., the mean-field result with logarithmic corrections. Note that the same logarithmic correction is implicitly given in Ref. [14].

For $d > 2$ similar arguments lead to the simple mean-field result, $n(h,u,v) \sim h^{1/2}$.

IV. CONCLUSIONS

The above calculation has shown that it is possible to construct a consistent renormalization-group approach for simple reaction-diffusion problems. This approach has the advantage of giving a unified description of the problem, allowing one not only to predict scaling and determine exponents, but also to compute the corresponding scaling functions.

Note that this type of renormalization-group approach is tailored to completely treat the fluctuations, which are a key ingredient below the upper critical dimension, and is not related to the renormalization-group method proposed by Chen, Goldenfeld, and Oono [22] to extract the asymptotic scaling behavior of the solutions of deterministic partial differential equations. Nevertheless, the latter method may prove to be fruitful when, within the conventional (mean-field) approach ignoring fluctuations, scaling properties of solutions of the diffusive rate equations are investigated under various initial and boundary conditions [23].

In the present work the renormalization-group approach was applied to the time-evolution operator $\mathcal{U}_{t,t_0}$. This gives reliable results only in situations where the behavior is independent of the initial state and fluctuations are completely controlled by internal dynamics, as is the case for steady state reached in the presence of particle sources. Scaling theory, supported by the existence of the stable fixed point, was then used to determine the asymptotic decay of particle density in the absence of sources.

It is then natural to try to extend this approach to more complicated problems as, for example, the two-species annihilation model defined in the Introduction. However, the generalization is not obvious. Indeed, in this case the initial conditions play a crucial role [4, 9] and any approach which is not keeping track of the fluctuations in the initial state will not describe correctly the long time dynamics.

Another challenging problem is the applicability of the renormalization-group approach to inhomogeneous systems; for example, to the case where the two constituents are initially spatially separated and a reaction front is formed. Both problems are presently under investigation.

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