Stochastic aggregation and subsequent recombination of particles generated by pulsed excitation in fractal and homogeneous systems

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The author discusses the accumulation and recombination kinetics of particles generated by pulsed excitation in systems with arbitrary fractal and spectral dimensionality, assuming that the excitation creates particles in pairs and that these particles are annihilated as a result of the bimolecular reaction $A + B \rightarrow 0$. He investigates the cases in which the characteristic size of a pair is the largest (i.e., the creation is spatially independent) and smallest spatial scale in the problem. Intermediate and truly asymptotic expressions are obtained for the decrease of the particle concentration after the excitation is switched off. The spatial distribution of the particles is investigated.

INTRODUCTION

Recently, the effect of density fluctuations of reacting particles on the kinetics of diffusion-controlled reactions has been intensively studied.[1,2] For example, for reactions of the type $A + B \rightarrow 0$ at various initial particle concentrations, fluctuation effects lead to replacement of the asymptotic fall-off $n(t) \propto t^{-1}$ of the concentration, predicted by formal kinetic theory, i.e., by the law $n(t) \propto t^{-d/4}$, where d is the dimensionality of the space.[3–7] This effect is connected with statistical clustering of particles of the same kind.

In this paper we will address the problem of particle clustering when particles are generated under pulsed conditions and recombine after the excitation is switched off. One example of such a process is the production of radiation-induced defects in an irradiated solid and their recombination during a subsequent anneal. Another possible realization is photodissociation or photoionization of molecules in a solution. By carrying out such experiments in solutions of strong electrolytes, we can eliminate electrostatic interactions between the radicals and observe the diffusion-controlled processes. Because of our considerable interest in the study of fractal systems, we were strongly motivated to pay special attention to this particular case. We can list other systems which likewise constitute a realization of these processes; mixed molecular crystals when the concentration of one of the components is close to percolation value solutions located in channels of porous silicate or organic glasses, and reactions at the roughened surfaces of catalyst crystals.

The corresponding problems for homogeneous systems have been investigated in considerable detail. The kinetics of particle annihilation after the switching-off of a very prolonged excitation were studied in Ref. 8, in which the authors investigated the case of independent creation of two types of particle in equal quantities. In Ref. 9 the same problem was investigated within a first-principles formalism both for independent and pairwise creation of particles; in this paper both the one-dimensional and three-dimensional cases were investigated. The asymptotic concentration of the particles, as well as their spatial distribution, were studied in detail.

There is much interest in the question of how particles accumulate within the excitation period. The authors of Ref. 10 arrived at the conclusion that for independent creation of particles in homogeneous one- and two-dimensional systems there occurs within the excitation period a spatial separation and accumulation of particles, while in the three-dimensional case no accumulation occurs and an equilibrium concentration of reagents is established. In Ref. 11 the accumulation was studied using numerical experiments on a number of fractal and Euclidean lattices. It was shown that clustering took place in systems with spectral dimension not exceeding 2. In a three-dimensional cubic lattice this accumulation is absent. Several questions relating to the establishment of equilibrium concentrations were studied in Ref. 12. In Ref. 9 it was proved that accumulation is absent in one- and three-dimensional models with particle creation.

In this paper we will study the intermediate (i.e., within times on the order of the irradiation time) and truly asymptotic behavior of the particle concentration after the excitation pulse is switched off (the pulse duration is t_0), and investigate the problem of accumulation and spatial separation of particles in systems with arbitrary spatial and spectral dimensionality. The types of asymptotic behavior predicted in Ref. 9 are reproduced here as particular cases at fractal and spectral dimensionalities $d_F = d_S = 3$ and $d_F = d_S = 1$.

THE MODEL

Within the framework of the mesoscopic approximation[2] the behavior of the particle concentration in these systems is described by the equations

$$\begin{aligned} \partial n_A(\mathbf{x}, t)/\partial t &= D\Delta n_A(\mathbf{x}, t) - K(n_A, n_B) + i_A(\mathbf{x}, t), \\ \partial n_B(\mathbf{x}, t)/\partial t &= D\Delta n_B(\mathbf{x}, t) - K(n_A, n_B) + i_B(\mathbf{x}, t), \end{aligned} \tag{1}$$

where $n_{A,B}(\mathbf{x},t)$ are the local values of the concentrations, $i_{A,B}(\mathbf{x},t)$ is the number of particles of a given species created per unit time per unit volume, K is an operator which describes the in particle loss resulting from the reaction, and $\langle i_A(\mathbf{x},t)\rangle = \langle i_B(\mathbf{x},t)\rangle = i_0$. The diffusion constants for particles A and B are assumed to be the same, which simplifies the theoretical investigation; however, this has no effect on the asymptotic time dependence of the concentrations if neither concentration reduces to zero. This latter case may belong to a different universality class.[13]

For a system whose spectral dimensionality $d_S > 2$ the operator K equals

$$K = \text{const}\, D R_0^{d_F - 2 - \theta} n_A n_B$$

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(see Ref. 14), where θ is the anomalous diffusion exponent and R_0 is the recombination radius. For $d_S \leqslant 2$ the function has a different character (the operator is nonlocal).

If there were no particle-flux fluctuations in the system, an equilibrium concentration n_{eq} would be established within the excitation time, characterized by equality of the influx of particles and their loss as a result of the reaction. Let us investigate the effect of fluctuations.

The concentration difference $r(\mathbf{x},t) = n_a(\mathbf{x},t) - n_B(\mathbf{x},t)$ obeys the equation

$$\partial r(\mathbf{x}, t)/\partial t = D\Delta r(\mathbf{x}, t) + i_r(\mathbf{x}, t), \qquad (2)$$

where $i_r(\mathbf{x},t) = i_A(\mathbf{x},t) - i_B(\mathbf{x},t)$. The pair correlation function $C(x,t) = \langle r(0,t)r(\mathbf{x},t)\rangle$ equals

$$C(x,t) = \int_0^t d\tau_1 \int_0^t d\tau_2 \int d\mathbf{x}'\, d\mathbf{x}'' G(\mathbf{x}', t-\tau_1)\, c(\mathbf{x}-\mathbf{x}', \tau_1, \tau_2) \times G(\mathbf{x}-\mathbf{x}'', t-\tau_2), \qquad (3)$$

where $G(x,t)$ is the Green's function of the diffusion equation and

$$c(x, t_1, t_2) = \langle i_r(0, t_1)\, i_r(\mathbf{x}, t_2)\rangle.$$

Assuming $r(z,t)$ to be a Gaussian random variable (which is perfectly valid for independent creation of particles or particle pairs), we find that

$$\langle |r(\mathbf{x},t)| \rangle = \left[\frac{2}{\pi}\langle r^2(\mathbf{x},t)\rangle\right]^{1/2} = \left[\frac{2}{\pi} C(0,t)\right]^{1/2}.$$

Because $n_{A,B}(\mathbf{x},t) \geqslant |r(\mathbf{x},t)|$, the average concentration of particles in the system cannot grow slower during the excitation period, or decrease faster after it is switched off, than does the quantity $|\langle r(\mathbf{x},t)\rangle|$.

In a number of cases[3-6,8] particles in the system become segregated in the later stages of the reaction. The resulting spatial distribution is related to the fact that a particle of one kind which has entered into a region where there is a fluctuation excess of particles of another species is annihilated very rapidly, while the fluctuations themselves attenuate slowly. This situation leads to the space being split into regions occupied by particles of only one kind. The reaction is intense only in the boundaries between these regions; these boundaries are found to be narrow compared to the characteristic sizes of the regions themselves. The structure of these boundaries was investigated in detail in Ref. 4. The number of particles in the boundary regions becomes small compared to the total number of particles, so that

$$n_A(\mathbf{x}, t) = r(\mathbf{x}, t)\theta[r(\mathbf{x}, t)], \quad n_B(\mathbf{x}, t) = r(\mathbf{x}, t)\theta[-r(\mathbf{x}, t)]$$

[$\theta(x)$ is the Heaviside step-function].

The distribution functions for like and unlike particles

$$p_{AA,BB}(\mathbf{x}, t) = \langle n_{A,B}(0, t)\, n_{A,B}(\mathbf{x}, t)\rangle,$$

$$p_{AB}(\mathbf{x}, t) = \langle n_A(0, t)\, n_B(\mathbf{x}, t)\rangle$$

are calculated by averaging the corresponding quantities over the Gaussian distribution of $r(\mathbf{x},t)$ using the two-point correlation function $C(x,t)$.[7] The expressions for the distribution functions have the form

$$p_{AA,BB}(x, t) = (1-\gamma^2)^{3/2} + \gamma \operatorname{arctg}[(1-\gamma^2)^{1/2}/\gamma] - \gamma^2(1-\gamma^2)^{1/2} + \pi\gamma, \qquad (4)$$

$$p_{AB}(x, t) = (1-\gamma^2)^{3/2} - \gamma \operatorname{arctg}[(1-\gamma^2)^{1/2}/\gamma] + \gamma^2(1-\gamma^2)^{1/2}, \qquad (5)$$

where $\gamma = C(x,t)/C(0,t)$.

Let us assume that the mass centers of newly-created pairs are randomly distributed in space, that the vector $2\mathbf{a}$ connecting the particles has a random orientation, that the quantity a is distributed according to some function $p(a)$ and has a characteristic value $\langle \mathbf{a}^2\rangle^{1/2} = a_0$, and that the pair-creation events occur in a time-independent way. Let us investigate two limiting cases corresponding to values of the ratio of the quantity a_0 to other characteristic scales in the problem, in particular to the scales n^{-1/d_F}—the average spacing between particles—and $l_D \propto (Dt)^{1/(2+\theta)}$, where t is the observation time. The case of uncorrelated creation of particles corresponds to the case where a_0 is the maximum spatial scale in the problem. In this case

$$c(x, t, t') = 2i_0\delta(t-t')\delta(x). \qquad (6)$$

However, for the case that a_0 is the minimum scale (i.e., for $a_0 < n^{-1/d_F}$ and the system is studied over times $t \gg a_0^{2+\theta}/D$),

$$c(x, t, t') = i_0 a_0^2 \langle \cos^2\vartheta\rangle \delta(t-t')\delta''(x), \qquad (7)$$

where $\delta''(x)$ is the second derivative of the δ-function and $\langle \cos^2\theta\rangle$ is the mean squared cosine of the angle between two vectors oriented arbitrarily in the space under study, in a d-dimensional Euclidean space, $\langle \cos^2\theta\rangle = 1/d$.

We note that by virtue of the finite pulse duration t_0 the values of c are given by Eqs. (6) and (7) for $0 \leqslant t$ and $t' \leqslant t_0$ and are zero otherwise.

Equations (6) and (7) are easy to obtain after investigating the auxiliary problem of calculating the correlation function for the density of pairwise disposed "unlike" points. The mass centers of pairs located at the points x_i are distributed with a density n, and

$$g(x) = \langle \rho(0)\rho(\mathbf{x})\rangle = \left\langle \sum_i [\delta(\mathbf{x}_i-\mathbf{a}) - \delta(\mathbf{x}_i+\mathbf{a})] \times \sum_i [\delta(\mathbf{x}_i-\mathbf{a}-\mathbf{x}) - \delta(\mathbf{x}_i+\mathbf{a}-\mathbf{x})]\right\rangle. \qquad (8)$$

After expanding the sums and passing from summation to integration over the coordinate of the center of a pair, we obtain

$$g(x) = 2n\delta(\mathbf{x}) - n\int [\delta(\mathbf{a}-\mathbf{x}) + \delta(\mathbf{a}+\mathbf{x})]\, p(\mathbf{a})\, d\mathbf{a}. \qquad (9)$$

For $a_0 \gg l_{char}$ the contribution of the second term to these expressions is found to be small to the extent that $a_0^{-d_F}$ is small, so that we can limit ourselves to the first term: $g = 2n\delta(x)$. When $a_0 \ll n^{-1/d_F}$ the converse holds. Noting that the δ-function depends only on the modulus of the argument, and expanding the expression in square brackets up to terms of second order in a, we find in this case that $g(x) = na_0^2\langle \cos^2\vartheta\rangle\delta''(x)$, where ϑ is the angle between the directions of the vectors $\mathbf{x}$ and $\mathbf{a}$. Equations (6) and (7) are obtained as the corresponding limiting cases of (9) by studying the pair creation versus time.

The behavior of the Green's function $G(x,t)$ in a fractal system is given by $G(x,t) \propto (Dt)^{-d_S/2} f(\xi)$, where $\xi = x/$

$(Dt)^{1/(2+\theta)}$ and $f(\xi)\to 0$ as $\xi\to\infty$ faster than a power law. Approximate expressions for the operator Δ and the function $G(x,t)$ are found, e.g., in Ref. 15. Here, d_S is the spectral dimensionality of the system, i.e., $d_S = 2d_F/(2+\theta)$, where the exponent of anomalous diffusion θ determines the mean square drift of a particle in a time t: $\langle x^2\rangle \propto t^{2/(2+\theta)}$. The dimensionality of the diffusion coefficient D for the fractal case is $[D] = [L^{2+\theta}/T]$.

In a homogeneous system $\theta = 0$ and $d_F = d_S = d$. For this case the behavior of $G(x,t)$ is known exactly. The additional results that can be extracted from knowledge of the exact form of $G(x,t)$ in the homogeneous case are presented in the Appendix. For the most part, these results coincide with those obtained in Ref. 9 within the framework of a different approach.

INDEPENDENT CREATION OF PARTICLES

When the particles are created independently, $c(x,t,t') \propto i_0\delta(x)$, and consequently $\langle r^2(t)\rangle = C(0,t)$ behaves like

$$\langle r^2(x,t)\rangle \propto i_0 \int_{t-t_0}^{t} dt \int dx x^{d_F-1} G^2(x,t)$$
$$\propto i_0 D^{-d_S/2} |t^{1-d_S/2} - (t-t_0)^{1-d_S/2}|. \qquad (10)$$

For $d_S < 2$ and $t = t_0$ (i.e., for observations within the generation time of the particles)

$$n(t) \propto \langle r^2(x,\ t)\rangle^{1/2} \propto i_0^{1/2} D^{-d_S/2} t^{(2-d_S)/4}. \qquad (11)$$

In this case an accumulation of particles is observed, which is connected with their statistical aggregation, and an equilibrium concentration of particles is not established. It is clear that $d_S = 2$ is the critical dimensionality for this effect, in full agreement with the results of numerical modeling.[11] On percolation clusters, $d_S \approx 4/3$ (Ref. 16) and $n(t) \propto t^{1/6}$. Within a time $\Delta t = t - t_0 \gg t_0$ after the source is switched on, the function $n(t)$ reaches the fluctuation asymptotic form:

$$n(t) \propto (i_0 t_0)^{1/2} (Dt)^{-d_S/4}. \qquad (12)$$

We recall that in the absence of fluctuations the concentration decreases like $n(t) \propto (Dt)^{-d_S/2}$ for $d_S > 2$ and like $n(t) \propto (DR_0^{d_F-2-\theta} t)^{-1}$ for $d_S > 2$ (Refs. 14 and 17).

The asymptotic behavior of (11) corresponds exactly to the type of fluctuation slowing-down which is observed in homogeneous systems, i.e., $n(t) \propto n_0^{1/2} (Dt)^{-d/4}$ (Refs. 3–7); however, in place of the initial concentration of particles n_0 the irradiation dose $Q = i_0 t_0$ appears in Eq. (12). For irradiation by very short pulses, i.e., when the particles created are unable to recombine within the irradiation time, the result (12) for the homogeneous case leads to well-known results.[3–7] For $d_S > 2$ the integral (9) diverges at its lower limit, which corresponds to the fact that $C(x,t) \to \infty$ as $x \to 0$ and $t \to t_0$. This unphysical divergence is connected with Poisson fluctuations in the number of particles created within small volumes at times immediately preceding the observation time, i.e., with the δ-function-like form of the correlator c. It is clear from (4) and (5) that the presence of a divergence of C means absence of spatial segregation.

Let us investigate the question of particle accumulation in the system. As in Ref. 10, we introduce an observation volume with sides whose length is of order l_w. After calculating the mean square difference of particle concentrations, where the latter are determined by averaging over this volume (technically this is easy to do if we replace the δ-function $\delta(x)$ in Eq. (6) by a bell-shaped function with characteristic scale l_w), we find that

$$\langle r^2(t_0)\rangle_w^{1/2} \propto (i_0/Dl_w^{d_F-2-\theta})^{1/2}. \qquad (13)$$

Comparing this quantity with the quanity n_{eq} obtained by solving (1) under the assumption that fluctuations are absent ($\Delta n_A = \Delta n_B = 0$):

$$n_{eq} \propto (i_0/DR_0^{d_F-2-\theta})^{1/2}, \qquad (14)$$

we see that fluctuation of the number of particles in any volume exceeding the particle volume is small compared to the equilibrium particle concentration and perturbs weakly the equilibrium distribution. Thus, particle accumulation during generation does not take place for systems with $d_S > 2$.

For $\Delta t \gg t_0$ the asymptotic decay of the particle concentration is given by Eq. (12). The critical dimension for this effect is $d_S = 4$. For $d_S > 4$, the concentration follows the usual bimolecular law $n(t) \propto t^{-1}$ for $\Delta t \gg t_0$.

For $\Delta t \ll t_0$, after excitation by a very long pulse (when the first term in (10) can be neglected compared to the second),

$$n(t) \propto \langle r^2\rangle^{1/2} \propto i_0^{1/2} D^{-d_S/4} \Delta t^{(2-d_S)/4}. \qquad (15)$$

In the initial instants after the pulse switches off the relative fluctuations in concentration are small and the concentration decreases like $n(t) \propto t^{-1}$. The intermediate asymptotic form (15) is reached at

$$t \sim t_c \propto (R_0^{d_F-2-\theta} i_0^{1/2} D^{1-d_S/4})^{4/(6-d_S)}. \qquad (16)$$

Since to observe (15) it is necessary that $t_c \ll \Delta t \ll t_0$, the length of the excitation pulse must be such that $t_0 \gg t_c$. It is clear from (15) that the critical dimension for observing this intermediate slowing down is $d_S = 6$. The function $n(t) \propto t^{-1/4}$ which was observed in Ref. 8 corresponds to just this intermediate asymptotic form for the case $d = 3$ and $t_0 \to \infty$. As often happens, in this case the change to a fractal system reduces to a replacement of d by d_S.

PAIRWISE CREATION OF PARTICLES

The case of pairwise creation of particles is much more interesting. In this case,

$$\langle r^2(t)\rangle \propto i_0 a^2 \int_{t-t_0}^{t} dt \int dx x^{d_F-1} (G^2(x,t))''$$
$$= i_0 a_0^2 D^{-(d_F+2)/(2+\theta)} |t^{-f} - \Delta t^{-f}|, \qquad (17)$$

where $f = (d_F - \theta)/(2+\theta)$. Let us turn our attention to the way this atypical combination of exponents arises.

Depending on the sign of f there are two possible cases:

(a) $f < 0$ ($d_F < \theta$). In this case $n(t) \propto \langle r^2\rangle^{1/2} \propto t_0^{-f/2}$ and accumulation of particles takes place within the excitation time. It is not known to us whether this case is observed in real systems. On percolation clusters $d_F \geqslant \theta$ (equality is achieved for $d \geqslant 6$) so that the following case is applicable.

(b) $f > 0$ ($d_F > \theta$). In this case there is no accumulation of particles. For $\Delta t \ll t_0$ the function $\langle r^2(t)\rangle$ has the

form

$$\langle r^2(t)\rangle \propto i_0 a_0^2 D^{-(d_F+2)/(2+\theta)}\Delta t^{-f}.$$

When the inequality $d_F < 4 + 3\theta$ holds, the decrease of $\langle r^2\rangle^{1/2}$ turns out to be slower than the predicted bimolecular kinetic decrease of $n(t)$ (i.e., $n(t) \propto t^{-1}$ for $d_S > 2$ and $n(t) \propto t^{-d_S/2}$ for $d_S < 2$), and the quantity $n(t)$ follows the asymptotic form

$$n(t) \propto i_0^{1/2} a_0 D^{-(d_F+2)/2(2+\theta)}\Delta t^{-f/2}. \tag{18}$$

In particular, for $d = 3$ and $\theta = 0$ we have $n(t) \propto t^{-3/4}$. This expression agrees with that obtained in Ref. 9 for the case of pairwise particle creation in a three-dimensional system. There this expression corresponded to the purely asymptotic form of the decay of the particle concentration after switching off a pulse of infinitely long duration ($t_0 \to 0$); in our case it arises as an intermediate asymptotic form when $\Delta t \ll t_0$.

For $\Delta t \gg t_0$ we obtain from (17)

$$\langle r^2(t)\rangle^{1/2} \propto Q^{1/2} a_0 (Dt)^{-(d_F+2)/2(2+\theta)}.$$

A nontrivial fluctuation asymptotic form corresponding to this behavior

$$n(t) \propto \langle r^2(t)\rangle^{1/2} \propto t^{-(d_F+2)/2(2+\theta)} \tag{19}$$

is observed if the quantity $\langle r^2\rangle^{1/2}$ decreases more slowly than $n(t)$ when the latter is calculated by neglecting fluctuations, i.e., when the inequality

$$2 < d_F < 2 + 2\theta \tag{20}$$

holds. This inequality can be fulfilled if diffusion takes place at a roughened fractal surface (for example, at the surface of a solid catalyst). At such a surface $2 \leqslant d_F \leqslant 3$; it can be shown that $\theta \geqslant d_F - 2$. When this latter relation is a strict inequality, condition (20) will be fulfilled. The asymptotic form (19) cannot be observed in any other homogeneous ($\theta = 0$) system.

CONCLUSION

We have obtained asymptotic forms for the accumulation of particles under irradiation, and expressions for both pure and intermediate asymptotic decays of the particle concentrations after the excitation is switched off in systems with arbitrary fractal and spectral dimensions. We have investigated what conditions must be fulfilled in order to observe the corresponding fluctuation effects, taking note of the fact that for both independent and pairwise creation of particles in fractal and homogeneous systems one can have either particle accumulation during excitation or else a power-law intermediate asymptotic decrease of the particle concentrations.

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APPENDIX

In homogeneous systems the quantity $C(x,t)$ in (3) is easily calculated by passing to a Fourier transform and the functions $C(x,t)$ can be expressed in terms of special functions for $d = 1,2,3$: in the case of uncorrelated creation of particles, for $d = 1$ we have

$$C(x,t) = \frac{i_0}{2D\pi^{1/2}}\left[(8Dt)^{1/2}\exp\left(-\frac{x^2}{8Dt}\right) - \pi^{1/2} x \operatorname{erf}\left(\frac{x}{(8Dt)^{1/2}}\right) - [8D(t-t_0)]^{1/2}\exp\left(-\frac{x^2}{8D(t-t_0)}\right) + \pi^{1/2} x \operatorname{erf}\left(\frac{x}{[8D(t-t_0)]^{1/2}}\right)\right] \tag{21}$$

(this expression coincides with that obtained in Ref. 10); for $d = 2$,

$$C(x,t) = \frac{i_0}{2\pi D}\left[\operatorname{Ei}\left(-\frac{x^2}{8D(t-t_0)}\right) - \operatorname{Ei}\left(-\frac{x^2}{8Dt}\right)\right], \tag{22}$$

and for $d = 3$

$$C(x,t) = \frac{i_0}{8\pi D x}\left[\operatorname{erf}\left(\frac{x}{[8D(t-t_0)]^{1/2}}\right) - \operatorname{erf}\left(\frac{x}{(8Dt)^{1/2}}\right)\right]. \tag{23}$$

By following the system's behavior for $t > t_0$, we see that for $d = 1$ and 2 clusters made up of like particles form in the system after the excitation period; the characteristic spatial scale of these clusters is $\sim (Dt)^{1/2}$. For $d = 3$ this scale enters into the problem only in the sense that the transition from a power-law decrease $C(x,t) \propto x^{-1}$ to the more rapid decrease determined by the expression in the brackets takes place over this spatial interval.

For $d = 1$, $C(0,t_0) \propto t_0^{1/2}$ and the average particle concentration in the system $n \geqslant [2C(0,t_0)/\pi]^{1/2}$ increases with time as $n \propto t_0^{1/4}$.

For $d = 2$ and 3, $C(x,t)$ diverges as $x \to 0$ and $t \to t_0$. For $d = 3$ an equilibrium concentration is established in the system. For $d = 2$

$$\langle r^2(t)\rangle_w^{1/2} = \left(\frac{i_0}{4\pi D}\ln\frac{t_0}{t_w}\right)^{1/2}, \tag{24}$$

where $t_0 = l_w^2/D$; consequently the number of particles in a given specified volume increases no slower than the function $N \propto (\ln t_0)^{1/2}$. After the pulse is switched off, the intermediate asymptotic decrease in the particle concentration for $d = 2$ is

$$n(t) = \left(\frac{i_0}{4\pi D}\right)^{1/2}\ln^{1/2}\left(\frac{t_0}{\Delta t}\right), \tag{25}$$

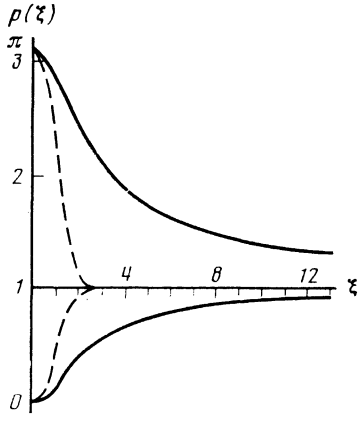

FIG. 1. Pair correlation function for like and unlike particles for the case of independent particle creation in a homogeneous three-dimensional system corresponding to large (the dashed curve) and intermediate (solid curve) times.

while the three-dimensional case is described by Eq. (12) with $d_S = 3$. For pairwise creation of particles

$$C(x,t) = \frac{i_0 a_0^2}{(2\pi)^{d/2} dD^{1+d/2}} \times \left[\Delta t^{-d/2} \exp\left(-\frac{x^2}{8D\Delta t}\right) - t^{-d/2} \exp\left(-\frac{x^2}{8Dt}\right)\right]. \quad (26)$$

An analysis of the divergence as $x \to 0$ and $t \to t_0$, analogous to that carried out for uncorrelated creation of particles, shows that accumulation of particles in a macroscopic volume does not occur and that equilibrium is established for any dimensionality.

Let us discuss pair distribution functions of particles for $d = 3$. The behavior of the pair distribution functions of like and unlike particles calculated using Eqs. (4) and (5) for the cases $t_c \leqslant t \leqslant t_0$ (the intermediate asymptotic form) and $\Delta t \gg t_0$ (the truly asymptotic form) for independent creation of particles are shown in Fig. 1. The corresponding functions are shown plotted against the dimensionless parameter $\xi = x/(8Dt)^{1/2}$.

The particle pair distribution functions corresponding to the intermediate asymptotic form $n(t) \propto t^{-3/4}$ for pairwise creation look the same as the function $P_{AA,BB}$ in its purely asymptotic form for the case of uncorrelated creation of particles.

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