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Diffusion-controlled death of *A*-particle and *B*-particle islands at propagation of the sharp annihilation front $A+B\rightarrow 0$

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We consider the problem of diffusion-controlled evolution of the *A*-particle-island–*B*-particle-island system at propagation of the sharp annihilation front $A+B\rightarrow 0$. We show that this general problem, which includes as particular cases the sea-sea and island-sea problems, demonstrates rich dynamical behavior from self-accelerating collapse of one of the islands to synchronous exponential relaxation of both islands. We find a universal asymptotic regime of the sharp-front propagation and reveal the limits of its applicability for the cases of mean-field and fluctuation fronts.

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For the last decades the reaction-diffusion system $A+B\rightarrow 0$, where unlike species *A* and *B* diffuse and annihilate in a *d*-dimensional medium, has acquired the status of one of the most popular objects of research. This attractively simple system, depending on the initial conditions and on the interpretation of *A* and *B* (chemical reagents, quasiparticles, topological defects, etc.), provides a model for a broad spectrum of problems [1,2]. A crucial feature of many such problems is the dynamical *reaction front*—a localized reaction zone which propagates between domains of unlike species.

The simplest model of a reaction front, introduced almost two decades ago by Galfi and Racz (GR) [3], is a quasi-one-dimensional (quasi-1D) model for two initially separated reactants which are uniformly distributed on the left side ($x<0$) and on the right side ($x>0$) of the initial boundary. Taking the reaction rate in the mean-field form $R(x,t)=ka(x,t)b(x,t)$, GR discovered that in the long-time limit $kt\rightarrow\infty$ the reaction profile $R(x,t)$ acquires the universal scaling form

$$R=R_f\mathcal{Q}\left(\frac{x-x_f}{w}\right), \tag{1}$$

where $x_f\propto t^{1/2}$ denotes the position of the reaction front center, $R_f\propto t^{-\beta}$ is the height, and $w\propto t^{\alpha}$ is the width of the reaction zone. Subsequently, it has been shown [4–8] that the mean-field approximation can be adopted at $d>d_c=2$, whereas in 1D systems fluctuations play the dominant role. Nevertheless, the scaling law (1) takes place at all dimensions with $\alpha=1/6$ at $d>d_c=2$ and $\alpha=1/4$ at $d=1$, so that at any *d* the system demonstrates a remarkable property of the effective *dynamical repulsion* of *A* and *B*: on the diffusion length scale $L_D\propto t^{1/2}$ the width of the reaction front asymptotically contracts unlimitedly: $w/L_D\rightarrow 0$ as $t\rightarrow\infty$. Based on this property a general concept of the front dynamics, the quasistatic approximation (QSA), has been developed [4,5,8,9] which consists in the assumption that for sufficiently long times the kinetics of the front is governed by two characteristic time scales. One time scale $t_J=-(d\ln J/dt)^{-1}$ controls the rate of change in the diffusive current, $J=J_A=|J_B|$, of particles arriving at the reaction zone. The second time scale $t_f\propto w^2/D$ is the equilibration time of the reaction front. Assuming that $t_f/t_J\ll 1$ from the QSA in the mean-field case with $D_{A,B}=D$ it follows that [4,5,9]

$$R_f\sim J/w, \quad w\sim(D^2/Jk)^{1/3}, \tag{2}$$

whereas in the 1D case *w* acquires the *k*-independent form $w\sim(D/J)^{1/2}$ [4,5]. On the basis of the QSA a general description of spatiotemporal behavior of the system $A+B\rightarrow 0$ has been obtained for arbitrary nonzero diffusivities [10] which was then generalized to anomalous diffusion [11], diffusion in disordered systems [12], diffusion in systems with inhomogeneous initial conditions [13] and to several more complex reactions. Following the simplest GR model [3] the main attention has been traditionally focused on the systems with *A* and *B* domains having an unlimited extension—i.e., with an *unlimited number* of *A* and *B* particles, where asymptotically the stage of monotonous quasistatic front propagation is always reached: $t_f/t_J\rightarrow 0$ as $t\rightarrow\infty$.

Recently, in [14] a new line in the study of the $A+B\rightarrow 0$ dynamics has been developed under the assumption that the particle number of one of the species is *finite*; i.e., an *A*-particle island is surrounded by a uniform sea of *B* particles. It has been established that at sufficiently large initial number of *A* particles, N_0, and a sufficiently high reaction rate constant *k* the death of the majority of island particles $N(t)$ proceeds in the *universal scaling regime* $N=N_0\mathcal{G}(t/t_c)$, where $t_c\propto N_0^2$ is the lifetime of the island in the limit $k,N_0\rightarrow\infty$. It has been shown that while dying, the island first expands to a certain maximal amplitude $x_f^M\propto N_0$ and then begins to contract by the law $x_f=x_f^M\zeta_f(t/t_c)$ so that on reaching x_f^M (the turning point of the front)

$$t_M/t_c=1/e, \quad N_M/N_0=0.19886\ldots, \tag{3}$$

and, therefore, irrespective of the initial particle number and dimensionality of the system $\approx 4/5$ of the particles die at the stage of the island expansion and the remaining $\approx 1/5$ at the stage of its subsequent contraction.

In this Rapid Communication we consider a much more general problem of the $A+B\rightarrow 0$ annihilation dynamics with the initially separated reactants under the assumption that the particle number of *both species* is finite. More precisely, we consider the problem of the diffusion-controlled death of *A*-particle and *B*-particle islands at propagation of the sharp annihilation front $A+B\to 0$. We show that this island-island (II) problem, of which particular cases are the GR sea-sea (SS) problem and the island-sea (IS) problem [14], exhibits rich dynamical behavior and we reveal its most essential features.

Let in the interval $x\in[0,L]$ particles A with concentration a_0 and particles B with concentration b_0 be initially uniformly distributed in the islands $x\in[0,\ell)$ and $x\in(\ell,L]$, respectively. Particles A and B diffuse with diffusion constants D_A and D_B, and when meeting they annihilate $A+B\to 0$ with a reaction constant k. We will assume, as usual, that concentrations $a(x,t)$ and $b(x,t)$ change only in one direction (flat front), and we will consider that the boundaries $x=0,L$ are impenetrable. Thus, our effectively one-dimensional problem is reduced to the solution of the problem

$$\partial a/\partial t = D_A\nabla^2 a - R, \quad \partial b/\partial t = D_B\nabla^2 b - R, \tag{4}$$

in the interval $x\in[0,L]$ at the initial conditions $a(x,0)=a_0\theta(\ell-x)$ and $b(x,0)=b_0\theta(x-\ell)$ and the boundary conditions $\nabla(a,b)|_{x=0,L}=0$ where $\theta(x)$ is the Heaviside step function. To simplify the problem essentially we will assume $D_A=D_B=D$. Then, by measuring the length, time, and concentration in units of L, L^2/D, and b_0, respectively—i.e., assuming $L=D=b_0=1$—and defining the ratio of initial concentrations $a_0/b_0=r$ and the ratio $\ell/L=q$, we come from Eqs. (4) to the simple diffusion equation for the difference concentration $s=a-b$,

$$\partial s/\partial t = \nabla^2 s, \tag{5}$$

in the interval $x\in[0,1]$ at the initial conditions

$$s_0(x\in[0,q)) = r, \quad s_0(x\in(q,1]) = -1, \tag{6}$$

with the boundary conditions

$$\nabla s|_{x=0,1} = 0. \tag{7}$$

According to the QSA for large $k\to\infty$ at times $t\propto k^{-1}\to 0$ there forms a sharp reaction front $w/x_f\to 0$ so that the solution $s(x,t)$ defines the law of its propagation, $s(x_f,t)=0$, and the evolution of particle distributions, $a=s(x<x_f)$ and $b=|s|(x>x_f)$. In the limits sea-sea [3] ($\ell\to\infty, L\to\infty$) or island-sea [14] problem (ℓ finite, $L\to\infty$) the corresponding solutions $s_{SS}(x,t)$ and $s_{IS}(x,t)$ describe the initial stages of the system's evolution at times $\sqrt{t}\ll q$, $1-q$, and $q\ll\sqrt{t}\ll 1$, respectively. The general solution to Eqs. (5)–(7) for arbitrary r, q, and t has the form

$$s(x,t) = \Delta + \sum_{n=1}^{\infty} A_n(r,q)\cos(n\pi x)e^{-n^2\pi^2 t}, \tag{8}$$

where coefficients $A_n(r,q)=2(r+1)\sin(n\pi q)/n\pi$ and $\Delta(r,q)=N_A-N_B=rq-(1-q)$ is the difference of the reduced number of A and B particles which remains constant. At $t>1/\pi^2$ the main mode in Eq. (8) becomes dominant, so neglecting the contribution of small-scale modes we find

$$s = \Delta + A_1(r,q)\cos(\pi x)e^{-\pi^2 t} + \cdots . \tag{9}$$

Taking $s(x_f,t)=0$ we obtain from Eq. (9) the law of the front motion,

$$\cos(\pi x_f) = \mathcal{C}e^{\pi^2 t} + \cdots , \tag{10}$$

where coefficient $\mathcal{C}$ can be represented in the form

$$\mathcal{C} = -\Delta/A_1 = q(r_\star - r)/A_1 = (q_\star - q)/q_\star A_1, \tag{11}$$

where $q_\star=1/(r+1)$ and $r_\star=(1-q)/q$ are the critical values of q and r at which $\mathcal{C}$ reverses its sign. From Eq. (10) it follows that at $|\mathcal{C}|<1/e$ and $r\neq r_\star, q\neq q_\star$, when the ratio of the initial particle numbers,

$$\rho = \frac{N_{A0}}{N_{B0}} = \frac{r}{r_\star} = \frac{(1-q_\star)q}{(1-q)q_\star} \neq 1, \tag{12}$$

the front $x_f(t)$ moves either towards the boundary $x=0$ ($\rho<1$) or towards the boundary $x=1$ ($\rho>1$) so that in the limit $k\to\infty$ the island of a smaller particle number (A or B, respectively) dies within a finite time

$$t_c = (1/\pi^2)|\ln|\mathcal{C}||. \tag{13}$$

From Eqs. (10) and (13) in the time interval $1/\pi^2 < t \leq t_c$ we obtain

$$x_f = (1/\pi)\arccos(\pm e^{\pi^2(t-t_c)}) \tag{14}$$

(here and in what follows the upper sign corresponds to $\rho<1$ and the lower sign corresponds to $\rho>1$), from which for the front velocity $v_f=\dot{x}_f$ we find

$$v_f = -\pi\cot(\pi x_f) = \mp\pi/(\sqrt{e^{2\pi^2(t_c-t)}-1}). \tag{15}$$

Making use then of Eq. (13), for the distribution of particles $[a=s(x<x_f), b=|s|(x>x_f)$ [14]] at $\rho\neq 1$ we obtain

$$s = \Delta(1\mp\cos(\pi x)e^{\pi^2(t_c-t)}) + \cdots . \tag{16}$$

Thus from the condition $N_A=\int_0^{x_f} s dx = N_B+\Delta$ we find the laws of decay of the A and B particle numbers,

$$N_A = (|\Delta|/\pi)(\sqrt{e^{2\pi^2(t_c-t)}-1}\mp\pi x_f), \tag{17}$$

and then we derive finally the diffusive boundary current in the vicinity of the front,

$$J = -\partial s/\partial x|_{x=x_f} = \pi|\Delta|\sqrt{e^{2\pi^2(t_c-t)}-1}, \tag{18}$$

which according to (2) defines the evolution of the amplitude $R_f(t)$ and of the width of the front $w(t)$.

From Eqs. (13)–(18) we immediately come to the following important conclusions: for arbitrary r and q which satisfy the condition $|\mathcal{C}(r,q)|<1/e$, at $\rho<1$ or $\rho>1$, (i) the motion of the front is the *universal* function of the "distance" to the collapse time t_c-t with the remarkable property $x_f^<(t_c-t)=1-x_f^>(t_c-t)$; moreover, the front velocity v_f is the *unique* function of x_f with the remarkable symmetry $x_f\leftrightarrow 1-x_f$, $v_f\leftrightarrow -v_f$; (ii) the reduced particle number $N_A/|\Delta|$ and the reduced boundary current $J/|\Delta|$ are *universal* functions of t_c-t with the remarkable properties $N_A^<(t_c-t)=N_A^>(t_c-t)-|\Delta|$ and $J^<(t_c-t)=J^>(t_c-t)$. Introducing the relative time $\mathcal{T}=t_c-t$, from Eqs. (13)–(18) in the vicinity $\mathcal{T}\ll 1/\pi^2$ of the critical point t_c we come to the universal power laws of self-accelerating collapse ($|v_f|\propto\mathcal{T}^{-1/2}$):

$$x_f^<, 1-x_f^> = \sqrt{2\mathcal{T}} + \cdots , \tag{19}$$

$$N_A^{<}, N_B^{>} = (\sqrt{8}/3)\pi^2|\Delta|\mathcal{T}^{3/2} + \cdots , \qquad (20)$$

$$J = \sqrt{2}\pi^2|\Delta|\sqrt{\mathcal{T}} + \cdots . \qquad (21)$$

At large $t_c \gg 1/\pi^2$ far from the critical point $\mathcal{T} > 1/\pi^2$ according to Eqs. (13)–(18) there is realized the intermediate exponential relaxation regime ($|v_f| \propto e^{-\pi^2\mathcal{T}}$)

$$x_f^{<,>} = 1/2 \mp e^{-\pi^2\mathcal{T}}/\pi + \cdots , \qquad (22)$$

$$N_A^{<,>} = (|\Delta|/\pi)e^{\pi^2\mathcal{T}}(1 \mp \pi e^{-\pi^2\mathcal{T}}/2 + \cdots), \qquad (23)$$

$$J = \pi|\Delta|e^{\pi^2\mathcal{T}}(1 - e^{-2\pi^2\mathcal{T}}/2 + \cdots), \qquad (24)$$

which in the limit $t_c \to \infty (|\mathcal{C}|, |\varrho - 1| \to 0)$ becomes dominant. Thus, at large $t_c \gg 1/\pi^2$ the point $x_f \approx 1/2$ (*stationary front*) is an "attractor" of trajectories. Exactly at the critical point $\rho_\star = 1$ from Eqs. (9) and (10) we find $x_f^\star = 1/2$ and obtain

$$N_\star/N_0 = \left(\frac{2}{\pi^2}\right)\frac{\sin(\pi q)}{q(1-q)}e^{-\pi^2 t} + \cdots , \qquad (25)$$

$$J_\star = 2[\sin(\pi q)/q]e^{-\pi^2 t} + \cdots . \qquad (26)$$

In order to answer the question of when and how the attractor $x_f^\star = 1/2$ is reached it is necessary to retain the next term ($n=2$) in the sum (8). With allowance for the first two terms one can easily obtain

$$x_f^\star = 1/2 - \mathcal{D}(q)e^{-3\pi^2 t} + \cdots , \qquad (27)$$

where $\mathcal{D}(q) = (A_2/\pi A_1) = \sin(2\pi q)/2\pi \sin(\pi q)$. According to Eq. (27) at $q = 1/2$ the coefficient $\mathcal{D}$ reverses its sign; therefore, as is to be expected, at $q < 1/2$ and $q > 1/2$ the front reaches the attractor $x_f^\star = 1/2$ from the left and right, respectively. By combining Eqs. (22) and (27), at small but finite $|\mathcal{C}|$ we have $x_f^{<,>} = 1/2 - \mathcal{C}e^{\pi^2 t}/\pi - \mathcal{D}e^{-3\pi^2 t} + \cdots$. We thus conclude that under the condition $\mathcal{DC} > 0$ there arises the turning point of the front ($v_f^M = 0$) with the coordinates

$$t_M = (1/4\pi^2)\ln(\lambda_M|\mathcal{D}/\mathcal{C}|) + \cdots , \qquad (28)$$

$$x_f^M = 1/2 - m_M\mathcal{D}|\mathcal{C}/\mathcal{D}|^{3/4} + \cdots , \qquad (29)$$

where $\lambda_M = 3\pi, m_M = 4/(3\pi)^{3/4}$, whereas at $\mathcal{DC} < 0$ there arises the inflection point of the front trajectory ($|v_f^s| = \min|v_f|$) with the coordinates t_s and x_f^s which are determined by Eqs. (28) and (29) with the coefficients $\lambda_s = 3\lambda_M$ and $m_s = 2m_M/(3)^{3/4}$. The analysis presented demonstrates the key points of the evolution of the island-island system at arbitrary r and q which satisfy the condition $|\mathcal{C}(r,q)| < 1/e$ [according to Eqs. (11) and (12) this condition restricts the interval $\rho_l < \rho < \rho_u$ to the values of $\rho_{l,u}$ which are not too different from unity: at $q \ll 1$ we find $\rho_l \approx 0.6$ and $\rho_u \approx 4$]. Below we will focus on a detailed illustration of this evolution from the initial island-sea configuration ($q \ll 1$).

A remarkable property of the island-sea configuration $q \ll 1$ is that at $r \gg 1$ the $\Delta(\rho) = \rho - 1$ value and all the coefficients $A_n(\rho) = 2\rho$ up to $n \propto 1/q \gg 1$ become unique functions

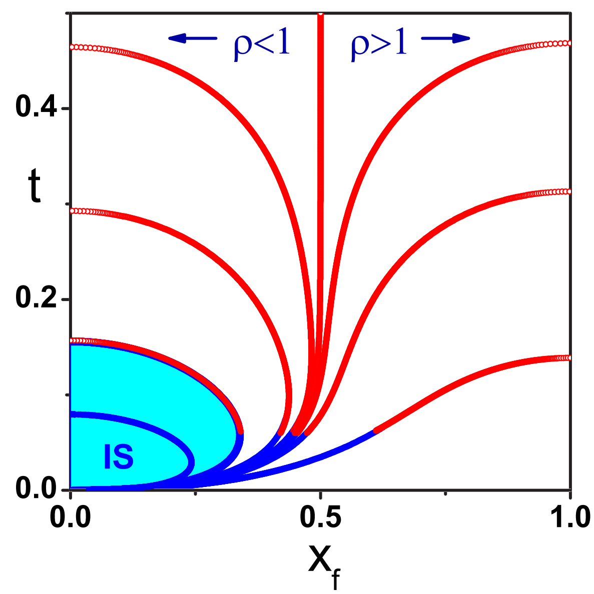

FIG. 1. (Color online) Evolution of the front trajectories $x_f(t)$ with growing ρ, calculated from Eqs. (30) (blue lines) and (31) (red circles) at ρ=0.5, 0.7, 0.9, 0.98, 1, 1.02, 1.1, and 2 (from left to right). The region of the scaling IS regime is shaded.

of ρ. Therefore, the system's evolution at $t \gg q^2$ is determined by the sole parameter ρ. At $q^2 \ll t \ll 1$ we have the scaling IS regime [14]

$$x_f = \sqrt{2t}\ln^{1/2}(\rho^2/\pi t), \quad t_c(\rho) = \rho^2/\pi, \qquad (30)$$

with $x_f^M = \rho\sqrt{2/\pi e}$ and $t_M = \rho^2/\pi e$. For $t > 1/2\pi^2$ with allowance for two principal modes ($n = 1,2$) we obtain from Eq. (8)

$$x_f = (1/\pi)\arccos[G(\rho,t)e^{3\pi^2 t}/4], \qquad (31)$$

where $G(\rho,t) = \sqrt{1 + 8\mathcal{C}(\rho)e^{-2\pi^2 t} + 8e^{-6\pi^2 t}} - 1$ and $\mathcal{C}(\rho) = (1-\rho)/2\rho$. For the time of collapse $t_c(\rho)$ we derive from Eq. (8) the general equation for arbitrary $\rho \neq 1$,

$$\sum_{n=1}^{\infty} (\pm 1)^n e^{-n^2\pi^2 t_c(\rho)} = \pm |\mathcal{C}(\rho)|, \qquad (32)$$

from which in accordance with Eq. (31) for the leading (at small $|\mathcal{C}|$) correction to Eq. (13) we find

$$t_c(\rho) = (|\ln|\mathcal{C}|| \pm |\mathcal{C}|^3 + \cdots)/\pi^2. \qquad (33)$$

Using small-t representations of the series (32), one can easily show that, with growing ρ, t_c initially grows by the law $t_c(\rho) = \rho^2(1 + 4e^{-\pi/\rho^2} + \cdots)/\pi$; then, it passes through the critical point $t_c(\rho_\star) \to \infty$ according to Eq. (33) and finally at large ρ decays by the law $t_c(\rho) \propto 1/\ln \rho$. From Eqs. (31) and (17) for the starting points $t_{M,s}$ of front self-acceleration at small $|\mathcal{C}|$ we find

$$t_{M,s}/t_c = 1/4 + \beta_{M,s}/|\ln|\mathcal{C}|| + \cdots , \qquad (34)$$

with the number of A particles, $N_A^{M,s}/N_{A0} \propto |\mathcal{C}|^{1/4}$, where $\beta_M = \beta_s/2 = \ln 3/4$. Remarkably, the same as for the scaling IS regime (3) and (30) in the vicinity $|\rho - \rho_\star| \ll 1$ the ratio t_M/t_c

reaches the *universal limit* $t_M/t_c=1/4$. In Fig. 1 are shown the calculated from Eqs. (30) and (31) trajectories of the front $x_f(t)$, which illustrate the evolution of the front motion with the growing ρ. It is seen that to $\rho\approx 0.7$ the death of the island A proceeds in the scaling IS regime (30) ($t_M/t_c=1/e$); then, the $x_f(t)$ trajectory begins to deform, and at small $|\rho-\rho_\star|\ll 1$ the regime of the dominant exponential relaxation (22)–(24) and (34) ($t_M/t_c\approx 1/4$) is reached. After the critical point $\rho_\star=1$ has been crossed, the death of the island A is superseded by the death of the island B, so the front trajectory becomes monotonous and the stopping point of the front x_f^M, t_M ($v_f^M=0$) "transforms" to the point of maximal deceleration of the front x_f^s, t_s ($v_f^s=\min v_f \propto |\mathcal{C}|^{3/4}$) which at large ρ shifts by the law $1-x_f^s\propto 1/\sqrt{\ln\rho}$ with $t_s\propto 1/\ln\rho$.

One of the key features of the island-island problem is a rapid growth of the front width w while the islands are dying. Therefore, to complete the analysis we have to reveal the applicability limits for the sharp front approximation $\eta=w/\min(x_f,1-x_f)\ll 1$. By substituting Eq. (21) into (2) we obtain for the self-accelerating collapse $\eta\sim(\mathcal{T}_Q/\mathcal{T})^{\mu}$ where for the mean-field front $\mu_{\text{MF}}=2/3$ and $\mathcal{T}_Q^{\text{MF}}=1/\sqrt{|\Delta|k}$. For a perfect diffusion-controlled 3D reaction $k\sim Dr_a$ where r_a is the annihilation radius. Thus, as our k is measured in units of D/L^2b_0 [14] for the dimensionless k we have $k=r_aL^2b_0$. Substituting here $r_a\sim 10^{-8}$ cm, $L\sim 10$ cm, and $b_0\sim 10^{22}$ cm^{-3} we find $k\sim 10^{16}$ and derive $\mathcal{T}_Q^{\text{MF}}\sim 10^{-8}/\sqrt{|\Delta|}$ so that for not too small $|\Delta|$ ($|\rho-\rho_\star|\gg 10^{-8}$) the sharp front is not destroyed almost down to the point of collapse. Clearly at small $|\Delta|\to 0$ the "destruction" of the front has to occur already at the stage of exponential relaxation (22)–(26). Substituting Eq. (26) into (2) for the exponential relaxation we find $\eta\sim e^{\nu\pi^2(t-t_Q)}$ where $\nu_{\text{MF}}=1/3$ and $t_Q^{\text{MF}}=(\ln k)/\pi^2$. Substituting here $k\sim 10^{16}$ we obtain $t_Q^{\text{MF}}\sim 3.7$ and then from Eq. (25) we find $N_\star^{\text{MF}}(\eta=0.1)/N_0\sim 10^{-13}$. An analogous calculation for the fluctuation 1D front gives $\mu_{\text{F}}=3/4$, $\mathcal{T}_Q^{\text{F}}\sim 1/(|\Delta|n_0)^{2/3}$ and $\nu_{\text{F}}=1/2$, $t_Q^{\text{F}}=(\ln n_0)/\pi^2$ where $n_0=Lb_0$. Substituting here $n_0\sim 10^6$ we find $\mathcal{T}_Q^{\text{F}}\sim 10^{-4}/|\Delta|^{2/3}$, $t_Q^{\text{F}}\sim 1.4$, and $n_\star^{\text{F}}(\eta=0.1)/n_0\sim 10^{-4}$. We conclude that both for the mean-field and the fluctuation fronts the vast majority of the particles die in the sharp-front regime; therefore, the presented theory has a wide applicability scope.

In summary, the evolution of the island-A–island-B system at the sharp annihilation front $A+B\to 0$ propagation has been considered and a rich dynamical picture of its behavior has been revealed. The theory presented may have a broad spectrum of applications—e.g., in the description of electron-hole luminescence in quantum wells [15], the formation of nontrivial Liesegang patterns [16], and so on. Of special interest is the analogy of the island-island problem with the problem of annihilation on the catalytic surface of a restricted medium where for unequal species diffusivities in a recent series of papers [17] the phenomenon of annihilation catastrophe has been discovered. Study of the much more complicated case of unequal diffusivities and comparison with the annihilation dynamics on the catalytic surface is a generic and challenging problem for the future.

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