Evolution of the A-particle island – B-particle island system at propagation of the sharp annihilation front $A+B\rightarrow 0$

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We consider diffusion-controlled evolution of the A-particle island – B-particle island system in a semi-infinite medium at propagation of the sharp annihilation front $A+B\rightarrow 0$. We show that depending on the initial particle number ratio, the system demonstrates three asymptotic regimes: self-accelerating collapse of one of the islands, synchronous power-law relaxation of both islands, and exponential death of one of the islands at a constant velocity of front propagation. We find asymptotic scaling laws of evolution in these regimes and reveal the limits of their applicability for the cases of mean field and fluctuation fronts.

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The reaction-diffusion system $A+B\rightarrow 0$, where species A and B diffuse and irreversibly annihilate in a d-dimensional medium, has attracted great interest owing to the remarkable property of the effective *dynamical repulsion* of unlike species [1,2]. In systems with initially statistically homogeneous particle distribution, this property brings about spontaneous growth of A and B particles domains (Ovchinnikov-Zeldovich segregation). In systems with initially spatially separated reactants, this property results in the formation and propagation of a *localized reaction front* which is a key feature of a broad spectrum of problems. The simplest model of a reaction front, introduced by Galfi and Racz (GR) [3], is the quasi-one-dimensional model

$$\partial a/\partial t = D_A\nabla^2 a - R, \quad \partial b/\partial t = D_B\nabla^2 b - R, \tag{1}$$

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

$$R = R_f Q\left(\frac{x-x_f}{w}\right), \tag{2}$$

where $x_f \propto t^{1/2}$, $R_f \propto t^{-2/3}$, and $w \propto t^{1/6}$ are, respectively, position, height, and width of the reaction front. Subsequently, it has been shown [4–8] that the mean-field approximation can be adopted at $d>d_c=2$, whereas in one-dimensional (1D) systems fluctuations play the dominant role. Nevertheless, scaling law (2) takes place at all dimensions so that at any d on the diffusion length scale $L_D \propto t^{1/2}$ the width of the reaction front asymptotically contracts: $w/L_D\rightarrow 0$ as $t\rightarrow\infty$. Based on this fact a general concept of the front dynamics, the quasistatic approximation (QSA), has been developed [4,5,8,9]. The key property of the QSA is that w and R_f depend on t only through the time-dependent boundary current of particles arriving at the reaction front, $J_A=|J_B|=J(t)$, the calculation of which is reduced to solving the *external* diffusion problem with the moving boundary. In the mean-field case with $D_{A,B}=D$ from the QSA it follows that [4,5,9]

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

whereas in the 1D case w acquires the k-independent form $w\sim(D/J)^{1/2}$ [4,5]. Following the GR model the majority of the authors employed the QSA mainly for 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 [10].

Recently, it has been shown [11] that based on the QSA the scope of the $A+B\rightarrow 0$ problems which allow for analytic description can be appreciably broadened including the systems with *finite* number of particles and *nonmonotonous* front propagation where asymptotically the QSA is violated. In the work [11] the problem of the death of an A-particle island in the B-particle sea was considered, and it was established that at large initial number of A particles and high-reaction constant the death of the majority of island particles proceeds in the universal scaling regime which extends almost up to the critical point of the island collapse. The more general A-particle island – B-particle island problem was considered in the recent work [12] where was given an exhaustive picture of the islands death on the finite interval $x\in[0,L]$ with the impenetrable-to-particles boundaries. The special case of this problem was also analyzed in the work [13] where an excellent agreement with the results of numerical simulation was found. As principally distinct from the island-island model [12] where the centers of both islands hold *immobile*, in this work we investigate the evolution of the island-island system where the center of one of the islands is *moving*. More precisely, we study the evolution of the quasi-one-dimensional island-island system in the semi-infinite space $x\in[0,\infty)$ and reveal a surprisingly rich scaling behavior of the system at propagation of the sharp annihilation front $A+B\rightarrow 0$.

Let in the interval $x\in[0,\infty)$ 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. We will assume that concentrations change only in one direction (flat front) and the boundary $x=0$ is impenetrable. We will also assume that $D_A=D_B=D$. Then, by measuring the length, time, and concentration in units of L, L^2/D, and b_0, respectively, and defining the ratio of initial concentrations $a_0/b_0=r$ and the ratio $\ell/L=q$, we come from Eq. (1) to the simple diffusion equation for the difference concentration $s=a-b$

$$\partial s/\partial t = \nabla^2 s, \tag{4}$$

in the interval $x\in[0,\infty)$ at the initial conditions

$$s_0(x\in[0,q))=r, \quad s_0(x\in(q,1])=-1,$$
$$s_0[x\in(1,\infty)]=0, \tag{5}$$

with the boundary conditions

$$\nabla s|_{x=0}=0, \quad s(\infty,t)=0. \tag{6}$$

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 $b=|s|(x>x_f)$. The general solution to Eqs. (4)–(6) for arbitrary r, q, and t has the form

$$s(x,t)=(r+1)\mathcal{F}_q(x,t)-\mathcal{F}_1(x,t), \tag{7}$$

where

$$\mathcal{F}_q(x,t)=\frac{1}{2}\left[\mathrm{erf}\left(\frac{q+x}{2\sqrt{t}}\right)+\mathrm{erf}\left(\frac{q-x}{2\sqrt{t}}\right)\right]. \tag{8}$$

It can easily be convinced that at $t\gg q^2$ and $x<t/q$ the function $\mathcal{F}_q(,x,t)$ has the asymptotics

$$\mathcal{F}_q(x,t)=\frac{q(1-\chi_q)}{\sqrt{\pi t}}e^{-x^2/4t}, \tag{9}$$

where $\chi_q=q^2\frac{(1-x^2/2t)}{12t}-q^4\frac{[1-x^2(1-x^2/12t)/t]}{160t^2}+\cdots$. Following the work [12] we shall focus here on the initial island-sea configuration $q\ll 1$. The remarkable property of this configuration is that at $q\ll 1$ and $t\gg q^2$ the system's evolution is determined by the sole parameter $\Delta=N_{A0}-N_{B0}=rq-(1-q)$ which at $r\gg 1$ defines the ratio of the initial particle numbers $\rho=1+\Delta\approx N_{A0}/N_{B0}$. In the limit $q\ll 1$ at $t\gg q^2$ we obtain from Eqs. (7) and (9)

$$s(x,t)=\rho e^{-x^2/4t}/\sqrt{\pi t}-\mathcal{F}_1(x,t). \tag{10}$$

By substituting into Eq. (10) the condition $s(x_f,t)=0$ we find the law of front motion in the form

$$x_f=\sqrt{2t\ln[\rho^2/\pi\mathcal{F}_1^2(x_f,t)t]}. \tag{11}$$

From Eq. (11) it follows that at $\rho<\rho_\star=1$ the trajectory of the front center $x_f(t)$ after the initial expansion of the island A comes back to the origin of the coordinates $x_f(t_c)=0$ so that at $k\to\infty$ the island A dies within a finite time $t_c(\rho)$ which is defined by the expression

$$\sqrt{\pi t_c}\,\mathrm{erf}(1/2\sqrt{t_c})=\rho. \tag{12}$$

In the time interval $q^2\ll t\ll 1$ the finiteness of the island B is not yet revealed, $\mathcal{F}_1(x_f)\approx 1$, therefore the island A death has to proceed in the scaling island-sea regime [11] with $t_c=\rho^2/\pi$, amplitude of the maximal island width $x_f^M=\rho\sqrt{2/\pi e}$, and the universal ratio $t_M/t_c=1/e$. Taking $t_c\ll 1$ we derive from Eq. (12) $t_c(\rho)=\rho^2(1+4\rho e^{-\pi/4\rho^2}/\pi+\cdots)/\pi$ whence for the region of the island-sea regime we find $\rho<\rho_u\approx 0.4$. In the opposite limit $t_c\gg 1$ we derive from Eq. (12)

$$t_c(\Delta)=\frac{1-9|\Delta|/10+\cdots}{12|\Delta|}. \tag{13}$$

Here we are mainly interested in the evolution of islands at $t\gg 1$ and $|\Delta|\ll 1$ when the diffusion length exceeds the initial sizes of both islands and the both islands are "long lived." At $t\gg 1$ and $x<t$ from Eqs. (9) and (10) we find

$$s(x,t)=\frac{[\Delta+\chi_1(x,t)]e^{-x^2/4t}}{\sqrt{\pi t}}, \tag{14}$$

where $\chi_1(x,t)$ can conveniently be presented in the form

$$\chi_1(x,t)=\frac{(1-3/40t+\cdots)}{12t}-\frac{x^2(1-3/20t+\cdots)}{24t^2}-\frac{x^4}{1920t^4}+\cdots.$$

From Eq. (14) at small $|\Delta|$ we come to the compact expression for the front trajectory

$$x_f=\sqrt{2t(\gamma+\Lambda t+1/20t+\cdots)}, \tag{15}$$

where $\gamma=1+6\Delta/5+O(\Delta^2)$ and $\Lambda=12\Delta[1-3\Delta/10+O(\Delta^2)]$. Equation (15) demonstrates three characteristic regimes of front propagation: (1) At $\Delta<0$ the island A first expands to a maximal amplitude $x_f^M\propto|\Delta|^{-1/2}$ and then dies within a finite time t_c according to Eq. (13); (2) At $\Delta_\star=0$ the front moves asymptotically by the law $x_f^\star=\sqrt{2t}$; (3) At $\Delta>0$ the front moves asymptotically by the law $x_f=\sqrt{2\Lambda}t$ at a constant velocity $v_f^\infty=\sqrt{2\Lambda}\approx\sqrt{24\Delta}$ [note that from the exact Eq. (10) at large $\rho\approx\Delta\gg 1$ we find $e^{v_f^{\infty 2}/2}/v_f^\infty=\rho$ and therefore $v_f^\infty\propto\ln\Delta$]. The calculated from Eqs. (11) and (15) front trajectories $x_f(t)$ are shown in Fig. 1 which gives the exhaustive picture of their evolution with growing ρ and demonstrates that in the interval $-0.1<\Delta<0.1$ Eq. (15) gives a quite precise description of the trajectories beginning with $t\sim 1$. By neglecting in parentheses of Eq. (15) the terms $O(|\Delta|,1/t)$ we arrive at a remarkable conclusion that at small $0<|\Delta|\ll 1$ and large $t>1$ the front trajectories $x_f(t)$ behave *self-similarly* and are described by the universal scaling laws

$$x_f=\sqrt{2t(1\mp t/t_c)}=|\Delta|^{-1/2}\Phi_\mp(\tau), \tag{16}$$

where $\tau=t/t_c$ and $t_c=1/|\Lambda|\approx 1/12|\Delta|$ defines the collapse time of the island at $\Delta<0$ and the crossover time to the constant front velocity regime at $\Delta>0$ (here and in what follows the upper sign "–" corresponds to the case $\Delta<0$ and the lower sign "+" corresponds to the case $\Delta>0$). From Eq. (16) it follows that at $\Delta<0$ the coordinates of the front turning point are $t_M/t_c=1/2$, $x_f^M=\sqrt{t_M}$ whence for the universal front trajectory we find

$$x_f/x_f^M=\zeta_f^u(\tau)=2\sqrt{\tau(1-\tau)} \tag{17}$$

and conclude that as ρ grows there occurs a crossover from the scaling regime island-sea $t_M/t_c=1/e$ to the scaling re-

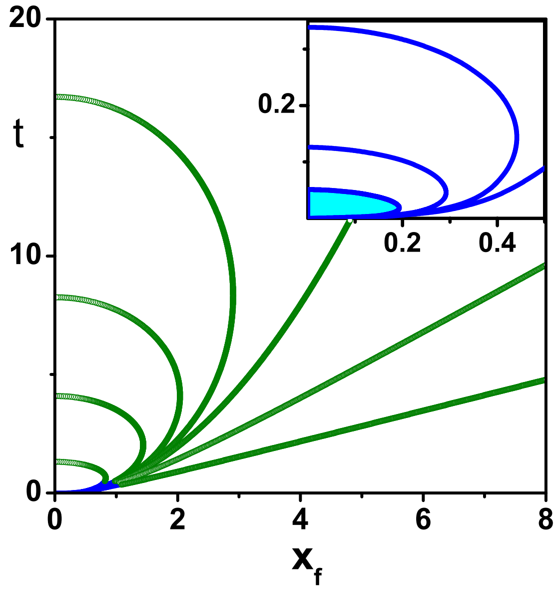

FIG. 1. (Color online) Evolution of the front trajectories $x_f(t)$ with growing $\rho=1+\Delta$, calculated from Eq. (11) (blue lines) and Eq. (15) (green circles) at $\rho=0.4,0.6,0.8,0.94$ (inset) and $\rho=0.94,0.98,0.99,0.995,1,1.02,1.1$ (main panel) (from left to right). The region of the scaling IS regime is colored.

gime island-island $t_M/t_c=1/2$ with the remarkable symmetry $\tau\leftrightarrow 1-\tau$ (Fig. 1). In Fig. 2(a) are shown the calculated from Eq. (11) trajectories of the front $x_f(t)$ in the rescaled coordinates x_f/x_f^M vs t/t_c. It is seen that with the decreasing $|\Delta|$ the trajectories rapidly collapse to the scaling function $\zeta_f^u(\tau)$.

Let us now turn to regularities of the decay of the particle number in islands $N_{A,B}$ and the boundary diffusion current J, which, the width of the reaction front being neglected, are defined by the expressions $N_A=\int_0^{x_f} s dx=N_B+\Delta$ and $J=-\partial s/\partial x|_{x=x_f}$. Taking $0<|\Delta|\ll 1$, $t>1$ and retaining in Eq. (14) only the leading terms, we come from Eqs. (14) and (16) to the scaling laws [Fig. 2(b)]

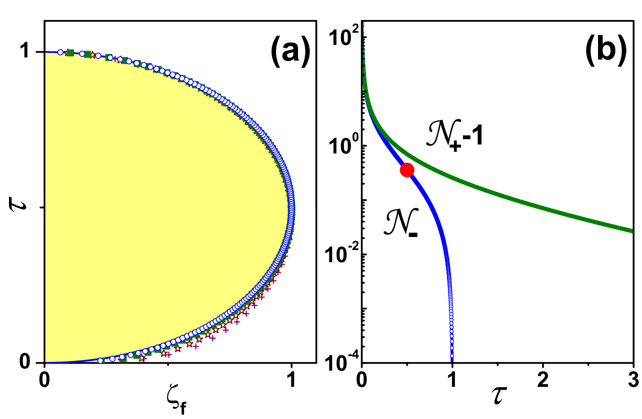

FIG. 2. (Color online) (a) Collapse of the calculated from Eq. (11) trajectories $x_f(t)$ to the scaling function $\zeta_f^u(\tau)$ (line) in the rescaled coordinates $\zeta_f=x_f/x_f^M$ vs $\tau=t/t_c$: $\rho=0.9$ (crosses), 0.94 (stars), 0.98 (squares), and 0.99 (circles). The region restricted by $\zeta_f^u(\tau)$ is colored. (b) Time dependences of the scaling functions $\mathcal{N}_-(\tau)$ and $\mathcal{N}_+(\tau)-1$. The circle marks the turning point of the front.

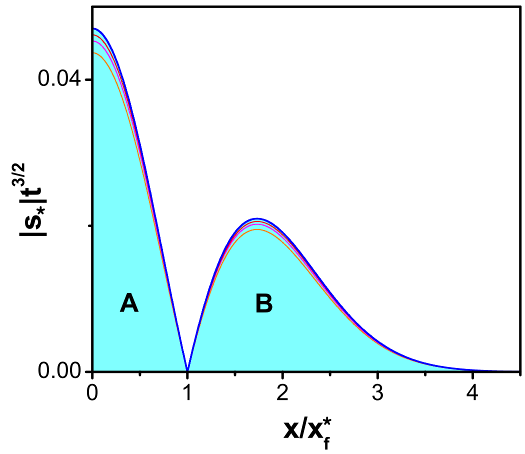

FIG. 3. (Color online) Collapse of the calculated from Eq. (11) dependences $|s_\star|t^{3/2}$ vs $x/x_f^\star$ to the scaling function $|\mathcal{S}|$ (thick line): from bottom to top $t=1,2$, and 4 (thin lines). The region under the function $|\mathcal{S}|$ is colored.

$$N_A=|\Delta|\mathcal{N}_\mp(\tau),\quad J=\Delta^2\mathcal{J}_\mp(\tau),\tag{18}$$

where the scaling functions

$$\mathcal{N}_\mp(\tau)=\frac{2\xi_f e^{-\xi_f^2}}{\sqrt{\pi\tau}}\mp \mathrm{erf}\,\xi_f,\quad \mathcal{J}_\mp(\tau)=\frac{24\xi_f e^{-\xi_f^2}}{\sqrt{\pi}\tau^2}$$

with $\xi_f=x_f/2\sqrt{t}=\sqrt{(1\mp\tau)/2}$. Equations (18) immediately lead to the following important consequences: (i) At $\Delta<0$ in the turning point of the front we find $N_A^M/|\Delta|\approx 0.358$ and therefore, independently of $|\Delta|$ in the turning point of the front $(N_A/N_B)_M\approx 0.263$. The asymptotics of the island A death in the vicinity of the collapse point $\mathcal{T}=(t_c-t)/t_c\ll 1$ have the form

$$N_A=\alpha_-|\Delta|\mathcal{T}^{3/2},\quad J=\beta_-\Delta^2\mathcal{T}^{1/2},\tag{19}$$

where $\alpha_-=\beta_-/18=4/3\sqrt{2\pi}$. (ii) At $\Delta>0$ in the point of crossover to the regime with the constant velocity of the front we find $N_B^c/\Delta\approx 0.257$ and therefore, independently of Δ at the crossover point $(N_B/N_A)_c\approx 0.205$. The asymptotics of the island B death at $t\gg t_c$ have the form of exponential relaxation

$$N_B=\frac{\alpha_+\Delta e^{-\tau/2}}{\tau^{3/2}},\quad J=\frac{\beta_+\Delta^2 e^{-\tau/2}}{\tau^{3/2}},\tag{20}$$

where $\alpha_+=\beta_+/6=2\sqrt{2/\pi e}$. Exactly at the particular point $\Delta=\Delta_\star=0$ from Eq. (14) we find that asymptotically the island-island system relaxes *self-similarly* by the law

$$s_\star(x,t)=t^{-3/2}\mathcal{S}(x/x_f^\star)\tag{21}$$

with the scaling function $\mathcal{S}(z)=(1-z^2)e^{-z^2/2}/12\sqrt{\pi}$ (Fig. 3) whence there follows the synchronous power-law death of both islands

$$N_\star/N_0=\alpha_\star/t,\quad J_\star=\beta_\star/t^2,\tag{22}$$

where $\alpha_\star=\beta_\star=\sqrt{2/\pi e}/12\approx 0.04$. As it must be, in the limit $|\Delta|\to 0$ $(t_c\to\infty)$ Eqs. (18) are reduced to the power-law asymptotics (22) which at any $0<|\Delta|\ll 1$ are intermediate asymptotics in the interval $1\ll t\ll t_c$.

We shall define the center x_m of the island B by the natural condition $|s|_{x=x_m}=\max|s(x>x_f)|$ (Fig. 3). In the point x_m the current of particles B reverses its sign, so it is clear that along with the characteristic size x_f of the island A the distance x_m-x_f defines the second characteristic size of the problem with respect to which the front width ought to be quite small. Taking $|\Delta|\ll 1$ and $t>1$ we find from Eq. (14) the law of motion of the island B center in the form

$$x_m=\sqrt{2t(\gamma_m+\Lambda t+3/20t+\cdots)}, \qquad (23)$$

where $\gamma_m=3+3\Delta/10+O(\Delta^2)$. With the same exactness as in Eq. (16) we come from Eq. (23) to the scaling laws

$$x_m=\sqrt{2t(3\mp t/t_c)}=|\Delta|^{-1/2}\Phi_m^{\mp}(\tau). \qquad (24)$$

The comparison of Eqs. (15) and (23) suggests the following conclusions: (1) at $\Delta<0$ the centers of the front and of the island B having passed through the turning point of the front are moving in the opposite directions. In the vicinity $\mathcal{T}\to 0$ of the island A collapse point $x_m/x_f\to\infty$; (2) at $\Delta>0$ the centers of the front and of the island B are asymptotically moving at the same velocity so that $x_m/x_f\to 1$ and $x_m-x_f\approx 1/\sqrt{6\Delta}=\text{const}$; (3) at the particular point $\Delta=\Delta_\star=0$ the ratio of velocities of the front and the island B centers holds constant so that asymptotically $x_m^\star/x_f^\star\to\sqrt{3}$.

To complete the outlined picture we have to reveal the applicability limits for the key condition of the sharp annihilation front

$$\eta=w/\min(x_f,x_m-x_f)\ll 1. \qquad (25)$$

Mean-field front. We shall estimate the applicability of the sharp front approximation for a prefect 3D diffusion-controlled reaction with dimensionless (in units of D/L^2b_0) constant $k\sim r_aL^2b_0$ where r_a is the annihilation radius [7,12,14]. Substituting here $r_a\sim 10^{-8}$ cm, $L\sim 0.1$ cm and $b_0\sim 10^{20}$ cm^{-3} we obtain $k\sim 10^{10}$. At $\Delta<0$ from Eqs. (3), (15), and (19) we find

$$w_-^{\rm MF}\propto\mathcal{T}^{-1/6}, \quad \eta_-^{\rm MF}\sim(\mathcal{T}_Q/\mathcal{T})^{2/3}, \qquad (26)$$

where $\mathcal{T}_Q=(\beta_-^2k^2|\Delta|)^{-1/4}$. Taking $k=10^{10}$ we obtain $\mathcal{T}^{\rm MF}_{\eta=0.1}\sim\mathcal{T}_Q/\eta^{3/2}\sim 10^{-4}/|\Delta|^{1/4}$ and $N_A^{\rm MF}|_{\eta=0.1}/|\Delta|\sim\alpha_-(\mathcal{T}^{\rm MF}_{\eta=0.1})^{3/2}\sim 10^{-6}/|\Delta|^{3/8}$ and conclude that at not too small $|\Delta|$ the front holds sharp almost up to the island A collapse point. At $\Delta>0$ from Eqs. (3), (15), (20), and (23) we find

$$w_+^{\rm MF}\propto\sqrt{\tau}e^{\tau/6}, \quad \eta_+^{\rm MF}\sim\sqrt{\lambda}\tau e^{\tau/6}, \qquad (27)$$

where $\lambda=(\beta_+^2k^2\Delta)^{-1/3}$. Taking $k=10^{10}$ we obtain $\tau^{\rm MF}_{\eta=0.1}\sim 3\ln(\eta^2/\lambda\tau_\eta)\sim 3\ln(10^5\Delta^{1/3}/\tau_\eta)$ and $N_B^{\rm MF}|_{\eta=0.1}/\Delta\sim 1/k\eta^3\sqrt{\Delta}\sim 10^{-7}/\sqrt{\Delta}$ and conclude that at not too small Δ the exponential relaxation stage is reached in the sharp-front regime. At the particular point $\Delta=\Delta_\star=0$ from Eqs. (3), (15), and (22) we find

$$w_\star^{\rm MF}\propto t^{2/3}, \quad \eta_\star^{\rm MF}\sim(t/t_Q)^{1/6}, \qquad (28)$$

where $t_Q=(\beta_\star k)^2$. Taking $k=10^{10}$ we obtain $t^{\rm MF}_{\eta=0.1}\sim t_Q\eta^6\sim 10^{11}$ and $N^{\rm MF}_{\eta=0.1}/N_0\sim\alpha_\star/t^{\rm MF}_{\eta=0.1}\sim 10^{-13}$ and conclude that on the power relaxation stage the vast majority of the particles die in the sharp-front regime.

Fluctuation front. By performing the analogous calculations for a 1D fluctuation front with $n_0=b_0L$ we find $\eta_-^{\rm F}\sim(\mathcal{T}_Q/\mathcal{T})^{3/4}$ where $\mathcal{T}_Q=(\beta_-\Delta n_0)^{-2/3}$, $\eta_+^{\rm F}\sim(\omega\tau)^{3/4}e^{\tau/4}$ where $\omega=(\beta_+\Delta n_0)^{-2/3}$, and $\eta_\star^{\rm F}\sim\sqrt{t/t_Q}$ where $t_Q=\beta_\star n_0$. Taking $n_0=10^6$ we finally obtain $\mathcal{T}^{\rm F}_{\eta=0.1}\sim 10^{-3}/|\Delta|^{2/3}$, $\tau^{\rm F}_{\eta=0.1}\sim 3\ln(10^3\Delta^{2/3}/\tau_\eta)$, and $t^{\rm F}_{\eta=0.1}\sim 10^2$. We conclude that although the regime of fluctuation front imposes more severe restrictions, the presented theory has a wide applicability scope from electron-hole systems with characteristic times of the order of milliseconds to chemical systems with characteristic times of the order of hours.

In summary, the problem of diffusion-controlled evolution of the A-particle island – B-particle island system in semi-infinite medium has been considered and three characteristic regimes of propagation of the sharp annihilation front $A+B\to 0$ have been revealed. As a central result it has been found out that at $|\Delta|\ll 1$ the evolution of the island-island system in each of these regimes proceeds *self-similarly* and is described by the *universal* scaling laws. Within the framework of the quasistatic approximation the laws of growth of the relative front width have been derived self-consistently and it has been shown that all the three regimes may be realized in a wide range of parameters. One of the most interesting consequences of the analysis presented is the discovery of the regime with a constant velocity of front propagation (regime of exponential relaxation), which offers a possibility to observe nontrivial Liesegang patterns (rhythmic precipitation patterns in the wake of the moving front) for the irreversible reaction $A+B\to C$ [15]. It should be emphasized, however, that to experimentally realize this regime the initial concentrations of particles in the islands should be high enough. As in the case of the island-island system on a finite interval [12], here the evolution of the island-island system has been considered at equal species diffusivities. Although we believe that the analysis presented reflects the key features of the island-island system evolution the study of the much more complicated problem for unequal species diffusivities remains an intriguing and a challenging problem for the future.

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