Catastrophe in diffusion-controlled annihilation dynamics: From formation to universal explosion

Boris M. Shipilevsky
Institute of Solid State Physics, Chernogolovka, Moscow district, 142432, Russia

(Received 27 June 2006; revised manuscript received 26 April 2007; published 21 September 2007)

We present a systematic theory of formation of the universal annihilation catastrophe which develops in an open system where species A and B diffuse from the bulk of a restricted medium and die on its surface (desorb) by the reaction $A+B\to 0$. This phenomenon arises in the diffusion-controlled limit as a result of self-organizing explosive growth (drop) of the surface concentrations of, respectively, slow and fast particles (concentration explosion), and manifests itself in the form of an abrupt singular jump of the desorption flux relaxation rate. As striking results we find the dependences of time and amplitude of the catastrophe on the initial particle number, and answer the basic questions of when and how universality is achieved.

DOI: 10.1103/PhysRevE.76.031126 PACS number(s): 82.20.−w, 05.70.Jk, 05.70.Ln

I. INTRODUCTION

In recent decades, it has been shown that in spite of its fundamental simplicity the reaction-diffusion system $A+B\to 0$ exhibits rich cooperative behavior [1]. One of the most impressive examples in the class of systems with in the bulk reaction is the phenomenon of Ovchinnikov-Zeldovich segregation (spontaneous growth of single-species domains which leads to anomalous reaction deceleration). Here we focus on another wide class of systems, where reaction proceeds on the *catalytic surface* of a medium, whereas diffusion proceeds in its bulk. In [2], it was first demonstrated that in this class of systems the interplay between reaction and diffusion acquires qualitatively new features and leads to a new type of self-organization. It has been found that if particles A and B diffuse with different mobilities from the bulk of a restricted medium onto the surface and die on it (desorb) by the reaction $A+B\to 0$, there exists some threshold difference in the initial numbers of A and B particles, Δ_c, above which the loop of positive feedback is switched on, and the process of their death, instead of the usual deceleration, starts to accelerate autocatalytically. Recently, it has been discovered [3] that the deceleration-acceleration transition is a prelude to much more nontrivial dynamical effects: in the diffusion-controlled limit $\Delta\to\infty$, a new critical phenomenon develops—annihilation catastrophe, which arises as a result of self-organizing explosive growth (drop) of the surface concentrations of slow (fast) particles (concentration explosion) and manifests itself in the form of an abrupt singular jump of the desorption flux relaxation rate.

The key features of the annihilation catastrophe have been obtained on the assumption that the initial number of A-B pairs is large, so that after a transient stage the annihilation dynamics crosses over to the universal regime, independent of initial pair number [2,3]. Until now, however, the principal questions remain open: When and how is the universal regime achieved? Moreover, the central question concerned with the time moment of the catastrophe as a function of the initial pair number remains open too. In this paper, we present a systematic theory which gives exhaustive answers to these questions.

II. MODEL

We consider a model in which species A and B are supposed to be initially uniformly distributed in the bulk of an infinitely extended slab of thickness 2ℓ. Both species diffuse to the surface $X=\pm\ell$ ($X\in[-\ell,\ell]$) and irreversibly desorb as a result of the surface reaction $A_{ads}+B_{ads}\to AB$ with a rate proportional to the product of surface concentrations $I=\kappa c_{As}c_{Bs}$ [4] (Fig. 1). Because of the planar spatial homogeneity, the system is effectively one dimensional. The boundary conditions are determined from the equality of diffusion I^D and desorption I flux densities at the surface, $I^D|_s=I$, i.e., it is assumed that the surface layer capacity can be neglected. According to [2,3], after introducing the index H (heavy) for the slower-diffusing species and L (light) for the faster one, the problem of species evolution in the dimensionless units reads (by symmetry, we consider the interval $[0,\ell]$ only)

$$\partial h/\partial\tau=\nabla^2 h,\quad \partial l/\partial\tau=(1/p)\nabla^2 l, \tag{1}$$

$$\nabla h|_s=(1/p)\nabla l|_s=-h_s l_s, \tag{2}$$

with $\nabla(h,l)|_{x=0}=0$ and initial conditions $h(x,0)=h_0$ and $l(x,0)=l_0$. Here, $h(x,\tau)=c_H/c_*$ and $l(x,\tau)=c_L/c_*$ are the reduced concentrations, $\nabla\equiv\partial/\partial x, x=X/\ell\in[0,1]$ is the dimensionless coordinate, $\tau=D_H t/\ell^2$ is the dimensionless time, p

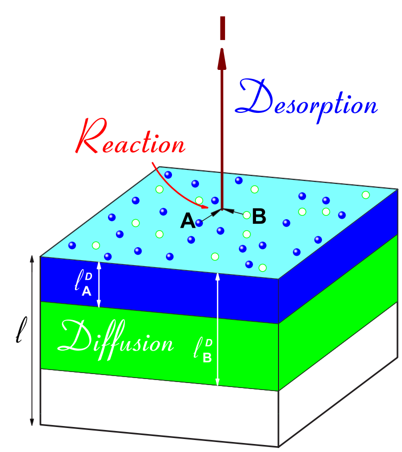

FIG. 1. (Color online) Schematic illustration of the processes of bulk diffusion, surface reaction, and irreversible desorption in the system $A+B\to 0$. Because of the planar spatial homogeneity, the system is effectively one dimensional. $\ell_A^D=\sqrt{D_A t}$ and $\ell_B^D=\sqrt{D_B t}$ are the diffusion lengths of A and B particles, respectively.

$=D_H/D_L \leqslant 1$ is the ratio of diffusivities, and $c_* = D_H/\kappa\ell$ is the characteristic concentration scale. According to (2), particles disappear in pairs only, i.e.,

$$J = h_s l_s = -\langle \dot{h} \rangle = -\langle \dot{l} \rangle,$$

where $J = I/I_*$ is the reduced desorption flux density and $I_* = \kappa c_*^2$ is its characteristic scale. Therefore

$$\langle h \rangle - \langle l \rangle = \Delta = \text{const},$$

i.e., the excess amount stays "inert" in the bulk (here $\langle h \rangle = \int_0^1 h\, dx = \mathcal{N}_H/\mathcal{N}_*$ and $\langle l \rangle = \int_0^1 l\, dx = \mathcal{N}_L/\mathcal{N}_*$ are the total reduced numbers of particles in the bulk per unit surface, and $\mathcal{N}_* = c_*\ell = D_H/\kappa$). This inert part of the majority species $\Delta = \delta\mathcal{N}/\mathcal{N}_*$ acts as a control parameter, whereas its active part $N = \mathcal{N}_{pair}/\mathcal{N}_*$ (equal to the total number of H-L pairs) acts as the only variable decaying from N_0 to 0 as $\tau \to \infty$. We will consider here the annihilation dynamics for $\Delta > 0$ when the pair number N is dictated by the number of L particles $N(\tau) = \langle l \rangle$ ($N_0 = l_0 = h_0 - \Delta$), so that in the final state all L particles disappear, $N(\infty) = l(x,\infty) = 0$, and H particles are distributed uniformly with concentration $h(x,\infty) = \Delta$.

According to [2,3], the key features of nontrivial dynamics developing in the system (1) and (2) in the diffusion-controlled limit $N_0 = c_L(0)\kappa\ell/D_H \to \infty$ may be formulated as follows. When the parameter Δ achieves the threshold value

$$\Delta_c = \sqrt{\omega_0/p}\, \tan(\sqrt{\omega_0 p}) \tag{3}$$

($\omega_0 = \pi^2/4$ is the main eigenfrequency of the diffusion field relaxation), the system undergoes a transition to a state where, after a transient stage, the surface concentration of H particles, h_s, and as a result the rate of death of pairs, starts to grow autocatalytically with time (the growth of h_s accelerates the drop of l_s, the drop of l_s accelerates the growth of h_s, and so on). With growing Δ the autocatalytic stage becomes more and more pronounced, so that far beyond the threshold the self-acceleration acquires an explosive character: at $\Delta \to \infty$ the rates of growth $\Omega_{Hs} = +d \ln h_s/d\tau$ and relaxation $\Omega_{Ls} = -d \ln l_s/d\tau$ are synchronized, growing singularly by the law

$$\Omega_s = 1/|\mathcal{T}|, \quad |\mathcal{T}| = |\tau - \tau_\star| \to 0,$$

where the point of finite-time singularity $\tau_\star$ is achieved at the moment when the reduced number of pairs $n(\tau) = N(\tau)/\Delta$ drops to some critical value $n_\star$. The most spectacular consequence of concentration explosion is singular behavior of the flux relaxation rate $\tau_J^{-1} = -d \ln J/d\tau$, which is sustained constant up to the critical point $\tau_\star$, upon reaching which τ_J^{-1} blows up abruptly to ∞; at $\mathcal{K} = p^{3/2}\Delta/\Delta_c \to \infty$ the width of the jump contracts and its amplitude grows by the laws

$$|\mathcal{T}|_{cat} \propto \Delta^{-2/5} \to 0, \quad \max \tau_J^{-1} \propto \Delta^{1/4} \to \infty.$$

In [3], it has been shown that in the limit $n_0 = N_0/\Delta \to \infty$ this catastrophic jump of τ_J^{-1}, called the *annihilation catastrophe*, proceeds in the *universal*(n_0-independent) regime, and a scaling theory of the universal explosion was given. However, the approach developed in [3] did not allow one to say anything about the dynamics of explosion formation, or about how the universal regime is achieved or how the point of catastrophe depends on the initial conditions. The goal of this paper is to give a closed theory of the annihilation catastrophe formation and, based on it, (a) to reveal the catastrophe universalization regularities and (b) to find the dependence $\tau_\star(n_0)$ for an arbitrary ratio of diffusivities. We show that this strongly nonlinear problem allows for strict and elegant analytical solution, we reveal its surprisingly rich "structure," and we demonstrate remarkable agreement with numerical calculation results.

III. THEORY OF THE UNIVERSAL ANNIHILATION CATASTROPHE FORMATION

We start with the exact formal solution of the problem (1) and (2) in the Laplace space $\hat{f}(s) = \hat{\mathcal{L}} f(\tau)$:

$$\hat{h}(x,s) = h_0/s + \frac{(\hat{h}_s - h_0/s)\cosh(x\sqrt{s})}{\cosh\sqrt{s}},$$

$$\hat{l}(x,s) = l_0/s + \frac{(\hat{l}_s - l_0/s)\cosh(x\sqrt{sp})}{\cosh\sqrt{sp}}. \tag{4}$$

According to (4) the boundary conditions (BCs) (2) acquire the form

$$\hat{J} = (h_0/s - \hat{h}_s)\sqrt{s}\tanh\sqrt{s} = (l_0/s - \hat{l}_s)\sqrt{s/p}\tanh\sqrt{sp} = \hat{\mathcal{L}}(h_s l_s), \tag{5}$$

and in an implicit form completely define the behavior of the surface concentrations h_s and l_s, which in turn via Eqs. (4) define the evolution of the spatial distribution. The strategy for solution of the nonlinear chain (5) is based on the fact that in the H-diffusion-controlled regime the $h_s/\langle h \rangle$ ratio should rapidly drop with time. Therefore, according to (5) we can first (i) calculate $J(\tau)$ and $l_s(\tau)$ neglecting the h_s contribution, then (ii) derive $h_s(\tau)$ from the condition $h_s = J/l_s$, and, finally, (iii) calculate next-to-leading terms, thereby defining the self-consistent picture of the surface concentration evolution.

A. Transient dynamics ($\tau \ll 1$)

At sufficiently small times, the flux density is slightly changed. So, assuming $J \approx J_0 = h_0 l_0$, from (5) one obtains

$$h_s = h_0(1 - v_h + \cdots), \quad l_s = l_0(1 - v_l + \cdots), \tag{6}$$

where $v_i = (2/\sqrt{\pi})\sqrt{\tau/\tau_i}$, $\tau_h = 1/l_0^2$, and $\tau_l = 1/p h_0^2$. According to (6), the relative rate drop for l_s and h_s is governed by the value of the parameter

$$R = v_l/v_h = r\sqrt{p},$$

where $r = h_0/l_0 = (1 + n_0)/n_0$. Therefore, the necessary conditions for the H-diffusion-controlled annihilation regime are the fulfilment of the requirements $l_0 = N_0 \gg 1$ and $R < R_c = 1$. Taking both requirements satisfied, from (6) and (5), we conclude that at $\tau_h \ll \tau \ll 1$ the flux should drop according to the law

$$J = h_0(1-m)/\sqrt{\pi\tau} \approx J_0(\tau_h/\tau)^{1/2}/\sqrt{\pi} \quad (7)$$

with $m|_{\tau/\tau_h\to\infty}\to 0$. On the time interval $\tau_h \ll \tau \ll p$, both species diffuse in the semi-infinite medium regime, so from Eqs. (5) it follows that

$$l_s - h_s\sqrt{p} = l_0\epsilon,$$

where $\epsilon = R_c - R$. Assuming that $\epsilon > 0$ is not too small, from this expression, with account taken of the condition $h_s = J/l_s$ and Eq. (7), we find

$$h_s = \frac{r}{\epsilon\sqrt{\pi\tau}}(1-\phi+\cdots), \quad l_s = l_0\epsilon(1+\phi+\cdots), \quad (8)$$

where $\phi = (R/\sqrt{\pi}\epsilon^2)\sqrt{\tau_h/\tau}$. Using then (6) and (8), from Eq. (5) we self-consistently find $m \sim (R/\epsilon^2)^2(\tau_h/\tau) \ll \phi$, and conclude that the asymptotics (8) is realized at the condition $\epsilon \gg \tau_h^{1/4} = 1/\sqrt{N_0}$. In the opposite limit $0<\epsilon\ll\tau_h^{1/4}$, by the same procedure we come to the critical asymptotics

$$l_s(1-g+\cdots) = \sqrt{p}h_s(1+g+\cdots) = \frac{l_0}{\pi^{1/4}}\left(\frac{\tau_h}{\tau}\right)^{1/4}(1-\phi_c+\cdots),$$

where $g \sim \epsilon(\tau/\tau_h)^{1/4}$ and $\phi_c = m_c/2 = c(\tau_h/\tau)^{1/4}$ with $c = \pi^{1/4}\Gamma(\frac{3}{4})/2\Gamma(\frac{1}{4})$. At sufficiently small p and not too large r (so that $R \ll R_c$) on the time interval $\tau_h, p \ll \tau \ll 1/r^2 < 1$, the L particle distribution becomes uniform. In this regime from (5) and (7) we find

$$h_s = \frac{r}{\sqrt{\pi\tau}}(1+\sigma+\cdots), \quad l_s = l_0(1-\sigma+\cdots), \quad (9)$$

where $\sigma = 2r\sqrt{\tau/\pi} + O(r\sqrt{\tau_h}, R\sqrt{p/\tau})$.

B. Self-accelerating dynamics and the critical transition point ($\tau \gtrsim 1$)

According to Eqs. (8) and (9) at large $N_0\to\infty$, $\epsilon \gg 1/\sqrt{N_0}$, and not too large r (i.e., not too small n_0) by the moment $\tau \sim 1$, when the diffusion length of H particles becomes comparable with the system's size, the ratio $h_s/h_0 \propto r/\epsilon N_0 \to 0$. Neglecting the h_s contribution, it can be shown (see below) that at $\tau>1$ and large n_0 the h_s value has to tend exponentially to a constant $\mathcal{C}$. In view of this, according to (5) we write

$$\hat{J}^{(0)} = (h_0 - \mathcal{C})\tanh\sqrt{s}/\sqrt{s},$$

whence it follows that

$$J^{(0)} = \mathcal{A}e^{-\omega_0\tau}(1+e^{-8\omega_0\tau}+\cdots), \quad (10)$$

where $\mathcal{A} = 2(h_0 - \mathcal{C})$. With the same accuracy, from (5) we have

$$\hat{l}_s^{(0)} = l_0/s - [(h_0-\mathcal{C})/s]\sqrt{p}\tanh\sqrt{s}\coth\sqrt{sp},$$

whence it follows that

$$l_s^{(0)} = (\mathcal{A}/\Delta_c)e^{-\omega_0\tau}(1-\Lambda), \quad (11)$$

where Δ_c is defined by Eq. (3) and the leading contribution to Λ is governed by the sum of exponentially decaying, Λ_-, and exponentially growing, Λ_+, terms [6]

$$\Lambda = \Lambda_- + \Lambda_+,$$

$$\Lambda_- = \varrho_8 e^{-8\omega_0\tau} + \varrho_p e^{-\chi\omega_0\tau}, \quad \Lambda_+ = \lambda e^{\omega_0\tau}, \quad (12)$$

with exponent $\chi = (4/p - 1)$ and amplitudes

$$\varrho_8 = -(\Delta_c/3)\sqrt{p/\omega_0}\cot(3\sqrt{\omega_0 p}),$$

$$\varrho_p = (\Delta_c\sqrt{p}/\pi)\tan(\pi/\sqrt{p}),$$

$$\lambda = \Delta_c(\Delta - \mathcal{C})/\mathcal{A}.$$

From the condition $h_s = J/l_s$ and Eqs. (10) and (11), closing the chain, we find

$$h_s = \frac{\Delta_c(1+e^{-8\omega_0\tau}+\cdots)}{(1-\Lambda)}. \quad (13)$$

According to (12) and (13), in the limit of large $\mathcal{A}/|\Delta - \mathcal{C}|\Delta_c \to \infty$ ($|\lambda|\to 0$), the h_s value at any Δ exponentially achieves the universal asymptotics $h_s^c = \Delta_c$, whence it follows that $\mathcal{C} = \Delta_c$, and, therefore,

$$\lambda = \frac{\Delta_c(\Delta-\Delta_c)}{\mathcal{A}}, \quad \mathcal{A} = 2(h_0 - \Delta_c). \quad (14)$$

According to Eqs. (5), the correction to the diffusion-controlled flux $J^{(0)}$ is defined by the equation

$$\delta\hat{J}(s) = -\delta\hat{h}_s(s)\sqrt{s}\tanh\sqrt{s}, \quad (15)$$

whereas the correction to $l_s^{(0)}$ is defined by the equation

$$\delta\hat{l}_s(s) = \delta\hat{h}_s(s)\sqrt{p}\tanh\sqrt{s}\coth\sqrt{ps}, \quad (16)$$

where $\delta h_s = h_s - \Delta_c$. In Appendix A we give a systematic calculation of these corrections at $|\Lambda| \ll 1$ which leads to the following results.

(1) The joint contribution of the transient stage (8) and (9) and the long-time "tail" (13) is reduced to a relative displacement of the amplitude,

$$\delta\mathcal{A}/\mathcal{A} \sim O(\Delta_c/h_0).$$

One thus concludes that in the diffusion-controlled limit $\Delta_c/h_0 \to 0$ with an accuracy of vanishingly small terms,

$$\lambda = \frac{\Delta_c(\Delta-\Delta_c)}{2h_0}. \quad (17)$$

(2) The main contribution of the long-time tail (13) is reduced to the appearance of the exponentially growing term

$$\varepsilon(\tau) = (\Delta_c^2/2h_0)e^{\omega_0\tau} \quad (18)$$

so that at $|\Lambda| \ll 1$ Eq. (13) acquires the form

$$h_s = \Delta_c\{1 + (1 + \varrho_8)e^{-8\omega_0\tau}[1 + O(\varepsilon)] + \varrho_p e^{-\chi\omega_0\tau}[1 + O(\varepsilon)] + \Lambda_+[1 + O(\varepsilon)] + \cdots\}. \quad (19)$$

Hence one concludes that, in the diffusion-controlled limit $\Delta_c^2/h_0 \to 0$ at any Δ and small $|\lambda| \to 0$, h_s relaxes to the critical asymptotics $h_s^c = \Delta_c$ according to the law

$$\delta h_s/\Delta_c \propto e^{-\alpha\omega_0\tau}. \quad (20)$$

Here, in the $0 < p < p_c = 4/9$ range, $\alpha = 8$ and therefore the relaxation rate does not depend on p, whereas, in the $p_c < p < 1$ range, $\alpha = \chi(p)$ and therefore the relaxation rate decays with growing p $[3 < \chi(p) < 8]$.

From Eqs. (17)–(19) we immediately come to important consequences.

(i) With a growth of Δ, when the *threshold* value $\Delta = \Delta_c$ is reached, the amplitude λ reverses its sign and the behavior of h_s *changes qualitatively*: at $\Delta \leqslant \Delta_c$ h_s always drops monotonically, whereas at $\Delta > \Delta_c$ h_s first reaches a minimum $h_s^{min} \approx \Delta_c$ and then begins to grow exponentially. This rigorously proves the existence of a threshold for self-acceleration for an arbitrary ratio of diffusivities $0 < p < 1$ [note that in the limit $\tau \to \infty (\varepsilon \to \infty)$ a strict derivation of the threshold Δ_c has been given in [5]].

(ii) At $(\Delta - \Delta_c)/\Delta_c \gg (\Delta_c^2/h_0)^\alpha$ and $\lambda \ll 1$ for the time of onset of self-acceleration, τ_s, and departure of the starting point h_s^{min} from the critical asymptotics h_s^c, $\delta_s = (h_s^{min} - \Delta_c)/\Delta_c$, we find, respectively,

$$\tau_s = [1/\omega_0(\alpha + 1)]\ln(\alpha\tilde{\varrho}/\lambda) \quad (21)$$

and

$$\delta_s = (\alpha + 1)\tilde{\varrho}^{1/(\alpha+1)}(\lambda/\alpha)^{\alpha/(\alpha+1)}, \quad (22)$$

where, in the $0 < p < p_c$ range, $\alpha = 8$, $\tilde{\varrho} = 1 + \varrho_8$, whereas, in the $p_c < p < 1$ range, $\alpha = \chi$, $\tilde{\varrho} = \varrho_p$.

C. Annihilation catastrophe

According to (12), (17), and (18) the ratio

$$\varepsilon/\Lambda_+ = \frac{\Delta_c}{\Delta - \Delta_c};$$

therefore at $\Delta/\Delta_c \to \infty$ one has $\varepsilon/\Lambda_+ \to 0$. This suggests that, at finite $\Lambda < 1$ in the diffusion-controlled limit $h_0/\Delta_c \to \infty$, $\Delta/\Delta_c \to \infty$ (which is of prime interest for us), Eqs. (10)–(13) have to give the asymptotically exact description of the annihilation dynamics. In Appendix B we give a rigorous substantiation of this suggestion for small λ in terms of exact expansions in powers of Λ_+,

$$\delta J/J^{(0)} = \sum_{n=2}^{\infty} \mathcal{I}_n \Lambda_+^n,$$

$$\delta l_s/l_s^{(0)} = \sum_{n=2}^{\infty} \mathcal{L}_n \Lambda_+^n,$$

$$\delta h_s/\Delta_c = \Lambda_+ + \sum_{n=2}^{\infty} (1 - \mathcal{H}_n)\Lambda_+^n. \quad (23)$$

It is shown that the coefficients $\mathcal{I}_n$, $\mathcal{L}_n$, and $\mathcal{H}_n$ are proportional to $(\Delta - \Delta_c)^{-1}$ and that they grow slowly with n. From here there follows the remarkable fact that at $\lambda \ll 1$ in the limit $\Delta/\Delta_c \to \infty$ the corrections $\delta J/J^{(0)}$ and $\delta l_s/l_s^{(0)}$ remain vanishingly small up to the point of finite-time singularity $\Lambda \to 1$, where $\dot{h}_s/h_s \to \infty$ (below, for the "boundary" Λ_f of the dominant contribution of the corrections, it will be found that $1 - \Lambda_f \sim p^{1/4}\sqrt{\Delta_c/\Delta} \to 0$). This means that Eqs. (10)–(13) give a complete description of the explosion dynamics. Moreover, as at $\Delta/\Delta_c \to \infty$ the parameter

$$\lambda = \frac{\Delta_c}{2(1 + n_0)} \quad (24)$$

becomes a unique function of the reduced initial number of pairs $n_0 = N_0/\Delta$, Eqs. (10)–(13) allow one to achieve two main goals: (a) to find the time of the catastrophe $\tau_\star(n_0)$ and (b) to obtain a description of the explosion evolution with growing n_0.

Taking $\Lambda(\tau_\star) = 1$ and $\lambda \ll 1$, from Eqs. (12), we find

$$\tau_\star = \tau_\star^\mu(1 + \delta_\tau), \quad (25)$$

where

$$\tau_\star^\mu = (4/\pi^2)\ln[2(1 + n_0)/\Delta_c] \quad (26)$$

and

$$\delta_\tau(n_0) = -(\varrho_8\lambda^8 + \varrho_p\lambda^\chi)/|\ln \lambda|. \quad (27)$$

Introducing then the relative time $\mathcal{T} = \tau - \tau_\star$, from Eq. (13), we find that at any $p < 1$ in the vicinity of $|\mathcal{T}| \ll \omega_0^{-1}$ an explosive growth of h_s sets in according to the law

$$h_s = \frac{(1 + Q)}{\mu|\mathcal{T}|}, \quad |\mathcal{T}| \to 0, \quad (28)$$

where $\mu = \omega_0/\Delta_c \sim 1 - p$ and

$$Q(n_0) = (1 + 9\varrho_8)\lambda^8 + (1 + \chi)\varrho_p\lambda^\chi. \quad (29)$$

According to Eq. (10), in the course of explosion the flux is actually "frozen,"

$$J^{(0)} = J_\star[1 + \omega_0(1 + w)|\mathcal{T}| + \cdots],$$

reaching at the point of singularity the value

$$J_\star = \Delta\Delta_c(1 + G), \quad |\mathcal{T}| \to 0, \quad (30)$$

where $w(n_0) = 8\lambda^8$ and

$$G(n_0) = (1 + \varrho_8)\lambda^8 + \varrho_p\lambda^\chi. \quad (31)$$

From Eqs. (28) and (30) and the condition $h_s l_s = J_\star$, there immediately follows the synchronization of the growth $\Omega_{Hs} = +d\ln h_s/d\tau$ and relaxation $\Omega_{Ls} = -d\ln l_s/d\tau$ rates, which grow singularly by the law

$$\Omega_s = \Omega_{Hs} = \Omega_{Ls} = 1/|\mathcal{T}|, \quad |\mathcal{T}| \to 0.$$

Clearly, the explosive growth of $h_s^{ex} = \delta h_s(|\mathcal{T}| \to 0)$ has to lead to an explosive growth of the flux $J_{ex} = \delta J(|\mathcal{T}| \to 0)$ directed into the medium interior (in what follows we shall call it the "antiflux"). The dominant contribution to the explosion-triggered antiflux J_{ex} occurs in the vicinity $|\mathcal{T}| \ll 1$ where the diffusional response to the explosion forms in a narrow layer proportional to $\sqrt{|\mathcal{T}|}$ [3]. Thus, considering the medium as a semi-infinite one and allowing for (28), we can write [7] (see Appendix C)

$$J_{ex} = -\int_{-\infty}^{\mathcal{T}} \frac{dh_s^{ex}}{d\theta} \frac{d\theta}{\sqrt{\pi(\mathcal{T}-\theta)}} \sim -\frac{(1+Q)}{\mu|\mathcal{T}|^{3/2}}, \quad (32)$$

whence there follows the smallness of $|J_{ex}|/J_\star$ down to $|\mathcal{T}| \propto \Delta^{-2/3} \to 0$. Calculating then a singular contribution to the relaxation rate of the total flux $J = J^{(0)} + J_{ex}$,

$$\tau_J^{-1} = -\frac{d \ln J}{d\tau} = \omega_0(1+w) + [\tau_J^{-1}], \quad (33)$$

we find

$$[\tau_J^{-1}] = -\dot{J}_{ex}/J_\star \sim \frac{(1+Q-G)}{\Delta|\mathcal{T}|^{5/2}}, \quad (34)$$

whence there follows the catastrophic jump of τ_J^{-1} from $\tau_J^{-1} \approx \omega_0$ to $\tau_J^{-1} \to \infty$ with the width

$$|\mathcal{T}|_{cat} \propto \Delta^{-2/5} \to 0.$$

According to [3], the culminating consequence of the explosion in the limit $\mathcal{K} \sim p^{3/2}\Delta/\Delta_c \to \infty$ is the exact scaling description of passage through the point of singularity based on two key conditions: (a) the requirement

$$\sqrt{p}\ddot{h}_s = \ddot{l}_s,$$

providing for equality of diffusional responses $\dot{J}_{ex}^H = \dot{J}_{ex}^L$ [here and in what follows $(\dot{\ }) \equiv d(\ )/d\tau$] and (b) the self-consistent condition of passage through the singularity point at a frozen flux

$$h_s l_s = J_\star.$$

It has to be mentioned, however, that the scaling behavior of Ω_s was postulated in [3] on the basis of indirect arguments, and only later was the postulated scaling function analytically substantiated. Moreover, the length of [3] being concise, an important chain of considerations remained beyond the analysis given there. Below, we shall give a systematic scaling theory of the annihilation catastrophe in the universal limit $n_0 \to \infty$, and then, on its basis, a complete picture of catastrophe universalization with the growth of n_0 will be constructed.

D. Scaling laws of passage through the point of singularity

For simplicity, we shall begin by inferring the scaling laws of the concentration explosion and of the annihilation catastrophe in the universal limit $n_0 \to \infty$. Taking $\lambda, Q, G \to 0$, according to (10) and (11), at the explosion stage $\omega_0|\mathcal{T}| \to 0$ we have

$$J^{(0)} = J_\star(1 + \omega_0|\mathcal{T}| + \cdots) \quad (35)$$

and

$$l_s^{(0)} = \mu J_\star |\mathcal{T}|(1 + \omega_0|\mathcal{T}|/2 + \cdots), \quad (36)$$

whence it follows that

$$h_s = J^{(0)}/l_s^{(0)} = \frac{1}{\mu|\mathcal{T}|} + \omega_0/2\mu + \cdots, \quad (37)$$

and for $h_s^{ex} = h_s - \Delta_c$ we find

$$h_s^{ex} = \frac{1}{\mu|\mathcal{T}|} - \omega_0/2\mu + \cdots, \quad (38)$$

where the index (0) marks the solutions that neglect the contribution of h_s^{ex} [$(h_s^{ex})^{(0)} = 0$]. As has been mentioned above, the explosive growth of h_s^{ex} ought to trigger an explosive growth of the antiflux J_{ex}, in calculating which the medium can be regarded as a semi-infinite one,

$$J_{ex} = -\int_{-\infty}^{\mathcal{T}} \frac{dh_s^{ex}}{d\theta} \frac{d\theta}{\sqrt{\pi(\mathcal{T}-\theta)}} \quad (39)$$

(apparently, to an accuracy of vanishingly small terms the lower limit of the integral can be directed to $-\infty$). Substituting here h_s^{ex}, we find

$$J_{ex} = -\frac{a_J}{\mu|\mathcal{T}|^{3/2}} + \cdots, \quad (40)$$

where $a_J = \sqrt{\pi}/2$. In Appendix C we give a systematic calculation of $\delta J(|\mathcal{T}|)$ following Eq. (15). We find that not too far from the critical point the dominant contribution to $\delta J(|\mathcal{T}|)$ is made by the term

$$\mathcal{G}_+(|\mathcal{T}|) = -\Delta_c \sum_{n=1}^{\infty} \sqrt{n\omega_0} \tanh(\sqrt{n\omega_0}) e^{-n\omega_0|\mathcal{T}|}.$$

In the vicinity of the critical point $\omega_0|\mathcal{T}| \ll 1$, the sum $\mathcal{G}_+(|\mathcal{T}|)$ is reduced to the integral

$$J_{ex} = -(1/\mu) \int_{\omega_0}^{\infty} \sqrt{z} e^{-z|\mathcal{T}|} dz, \quad (41)$$

whence it follows that

$$J_{ex} = -\frac{a_J}{\mu|\mathcal{T}|^{3/2}} + O(1)/\mu + \cdots. \quad (42)$$

Thus, by neglecting the term $O(1)$ as compared with the singular term $\sim 1/|\mathcal{T}|^{3/2}$, in the limit $|\mathcal{T}| \to 0$ one arrives at the result (40) of the semi-infinite medium approximation. So for the total flux we have

$$J = J^{(0)} + J_{ex} = J_\star\left(1 + \omega_0|\mathcal{T}| - \frac{a_J}{\mu J_\star |\mathcal{T}|^{3/2}} + \cdots\right). \quad (43)$$

As the diffusion fluxes of fast and slow particles must be equal, $J_L^D|_s = J_H^D|_s = J$, it is clear that against the background of dropping $l_s^{(0)}$ there must arise an explosive growth of l_s^{ex}, which must initiate exactly the same antiflux $J_{ex}^L = J_{ex}^H = J_{ex}$. Assuming that at a developed explosion stage $\Omega_s p \gg 1$ the dominant contribution to J_{ex} occurs at times $|\mathcal{T}|/p \ll 1$, when for L particles the medium can also be regarded as a semi-infinite one, by full analogy with Eq. (39) we can write

$$J_{ex} = -\int_{-\infty}^{\mathcal{T}} \frac{dl_s^{ex}}{d\theta} \frac{d\theta}{\sqrt{p\pi(\mathcal{T}-\theta)}}. \tag{44}$$

Thus, by requiring the equality of (39) and (44), one is led to the key condition of synchronous growth, $\dot{l}_s^{ex} = \sqrt{p}\dot{h}_s^{ex}$, where, with account taken of the obvious requirement for smallness of l_s^{ex} away from the critical point, in the limit $|\mathcal{T}|/p \to 0$ one obtains

$$l_s^{ex} = \sqrt{p}h_s^{ex} = \frac{\sqrt{p}}{\mu|\mathcal{T}|}. \tag{45}$$

In Appendix C we give a systematic calculation of $\delta l_s(|\mathcal{T}|)$ following Eq. (16). We find that not too far from the critical point the dominant contribution to $\delta l_s(|\mathcal{T}|)$ is made by the term

$$\mathcal{Z}_+(|\mathcal{T}|) = \Delta_c \sum_{n=1}^{\infty} \sqrt{p}\tanh(\sqrt{n\omega_0})\coth(\sqrt{pn\omega_0})e^{-n\omega_0|\mathcal{T}|}.$$

In the vicinity of the critical point $\omega_0|\mathcal{T}| \ll 1$, the sum $\mathcal{Z}_+(|\mathcal{T}|)$ is reduced to the integral

$$l_s^{ex} = (1/\mu)\int_{\omega_0}^{\infty} \sqrt{p}\coth(\sqrt{pz})e^{-z|\mathcal{T}|}dz, \tag{46}$$

whence at $\omega_0|\mathcal{T}| \ll p$ it follows that

$$l_s^{ex} = \frac{\sqrt{p}}{\mu|\mathcal{T}|} + O(1)/\mu\sqrt{p} + \cdots. \tag{47}$$

By neglecting in (47) the term $\sim 1/\mu\sqrt{p}$ as compared with the singular term $\sqrt{p}/\mu|\mathcal{T}|$, we arrive at the result of the semi-infinite medium approximation,

$$l_s^{ex}(|\mathcal{T}|/p \to 0) = \frac{\sqrt{p}}{\mu|\mathcal{T}|}, \tag{48}$$

thus obtaining a rigorous derivation of the condition of synchronous growth (45).

Making use of the result (48), one obtains

$$l_s = l_s^{(0)} + l_s^{ex} = \mu J_\star|\mathcal{T}|\left(1 + \frac{\sqrt{p}}{\mu^2 J_\star \mathcal{T}^2} + \cdots\right), \tag{49}$$

and finally, with account taken of (43), from the condition $h_s = J/l_s$ one finds

$$h_s = \frac{1}{\mu|\mathcal{T}|}\left(1 - \frac{\sqrt{p}}{\mu^2 J_\star \mathcal{T}^2}(1+f_J) + \cdots\right), \tag{50}$$

where $f_J = a_J\mu\sqrt{|\mathcal{T}|/p}$. From Eqs. (49) and (50), it follows that in the vicinity of some characteristic time $\mathcal{T}_f \sim p^{1/4}/\mu\sqrt{J_\star}$, the explosion rate growth begins to drastically decelerate. Due to the requirement $\mathcal{T}_f/p \ll 1$ necessary for the realization of the synchronous explosion regime (45), the explosion deceleration begins long before a noticeable flux departure from the critical one, $|J_{ex}(\mathcal{T}_f)/J_\star| \ll 1$. Introducing the parameter

$$\mathcal{K} = \mu^2 p^{3/2} J_\star,$$

it can easily be seen that at any finite $0 < p < 1$ with a growth of Δ in the limit of large $\mathcal{K} \sim p^{3/2}\Delta/\Delta_c \to \infty$

$$|J_{ex}(\mathcal{T}_f)|/J_\star \sim \sqrt{\mathcal{T}_f/p} \sim 1/\mathcal{K}^{1/4} \to 0, \tag{51}$$

and, therefore, down to $\mathcal{T}_f \to 0$ the flux remains frozen. One of the most important consequences of the drastic deceleration of the explosion rate growth is the drastic deceleration of the flux relaxation rate growth. As will be shown in what follows, in the limit of large $\mathcal{K} \to \infty$, as a result of such deceleration the flux remains frozen and, therefore, the explosion develops synchronously

$$\Omega_s = \Omega_{Hs} = \Omega_{Ls} \tag{52}$$

both *before and after* passage through the point of singularity where Ω_s reaches a maximum. We shall now show that the condition (52) along with the following from (39) and (44)*key condition* of synchronous growth

$$\dot{l}_s^{ex} = \sqrt{p}\dot{h}_s^{ex}, \tag{53}$$

lead to a remarkably complete scaling description of passage through the point of singularity.

Taking $l_s = l_s^{(0)} + l_s^{ex}$, one has

$$\dot{l}_s^{ex} = \dot{l}_s + \mathcal{C}_{(0)}, \tag{54}$$

where $\mathcal{C}_{(0)} = -\dot{l}_s^{(0)} = \mu J_\star$. Substituting (54) into (53) and using then the equalities $\dot{h}_s^{ex} = \dot{h}_s = \Omega_s h_s$ and $\dot{l}_s = -\Omega_s l_s$, following from (52), one obtains

$$\Omega_s(l_s + \sqrt{p}h_s) = \mathcal{C}_{(0)}. \tag{55}$$

Differentiating (55) one finds

$$\dot{\Omega}_s = \Omega_s^2(l_s - \sqrt{p}h_s)/(l_s + \sqrt{p}h_s), \tag{56}$$

whence it follows that the explosion rate goes through the maximum Ω_s^M at the point where

$$l_s^M/h_s^M = \sqrt{p}. \tag{57}$$

Combining of (55) and (57) with the frozen flux condition

$$h_s^M l_s^M = h_s l_s = J_\star \tag{58}$$

enables one to close the chain and to readily derive the scaling law of concentration explosion. Indeed, from (57) and (58) we find

$$h_s^M = p^{-1/4}\sqrt{J_\star}, \quad l_s^M = p^{1/4}\sqrt{J_\star}. \tag{59}$$

Introducing then the scaling function

$$\zeta = h_s/h_s^M = l_s^M/l_s,$$

from (55) and (59) we obtain

$$\Omega_s = \frac{\dot{\zeta}}{\zeta} = \Omega_s^M F(\zeta), \quad F(\zeta) = \frac{2\zeta}{1+\zeta^2}, \tag{60}$$

where

$$\Omega_s^M = \mathcal{C}_{(0)}/2l_s^M = (\mu/2)h_s^M. \quad (61)$$

Integrating (60) under the condition that $\zeta(\mathcal{T}=0)=1$, one easily finds

$$\zeta - 1/\zeta = 2\Omega_s^M \mathcal{T}. \quad (62)$$

From Eq. (62) it follows that the characteristic time scale of explosion is determined by the quantity $\mathcal{T}_f = 1/\Omega_s^M$; therefore, introducing the reduced time $\mathsf{T}=\mathcal{T}/\mathcal{T}_f$, we finally obtain

$$\zeta(\mathsf{T}) = \mathsf{T} + \sqrt{1+\mathsf{T}^2}, \quad (63)$$

whence it follows that

$$\Omega_s = \Omega_s^M S(\mathsf{T}), \quad S(\mathsf{T}) = \frac{1}{\sqrt{1+\mathsf{T}^2}}. \quad (64)$$

Two striking features of this result are symmetrical universalization $|\mathcal{T}|^{-1} \leftrightarrow \mathcal{T}^{-1}$ of Ω_s beyond the scope of interval $[-\mathcal{T}_f, \mathcal{T}_f]$ and remarkable symmetry

$$\mathcal{T} \leftrightarrow -\mathcal{T}, \quad \zeta \leftrightarrow 1/\zeta.$$

Substituting (63) into (39) and using (64), we come to the scaling law of growth of the explosion-triggered antiflux J_{ex},

$$J_{ex} = J_{ex}^M \mathcal{J}(\mathsf{T}), \quad (65)$$

where the amplitude at the point of explosion maximum,

$$J_{ex}^M = -a_M h_s^M \sqrt{\Omega_s^M}, \quad (66)$$

and the scaling function

$$\mathcal{J}(\mathsf{T}) = a_{\mathcal{J}} \int_0^\infty d\theta\, \zeta(\mathsf{T}-\theta) S(\mathsf{T}-\theta)/\sqrt{\theta} \quad (67)$$

has the asymptotics

$$\mathcal{J}(\mathsf{T}) = \begin{cases} (\pi a_{\mathcal{J}}/4)|\mathsf{T}|^{-3/2}, & -\mathsf{T} \gg 1, \\ 4a_{\mathcal{J}}\mathsf{T}^{1/2}, & \mathsf{T} \gg 1. \end{cases}$$

The coefficients a_M and $a_{\mathcal{J}}$ are bound by the relation $a_M a_{\mathcal{J}} = 1/\sqrt{\pi}$; therefore, by satisfying $\mathcal{J}(0)=1$, one finds $a_M = 2\Gamma^2(3/4)/\pi \approx 0.956$ and $a_{\mathcal{J}} = \sqrt{\pi}/2\Gamma^2(3/4) \approx 0.590$.

Differentiating (65), one finds the singular part of the flux relaxation rate in the form

$$[\tau_J^{-1}] = -\dot{J}_{ex}/J_\star = [\tau_J^{-1}]_M W(\mathsf{T}), \quad (68)$$

where the amplitude at the point of explosion maximum,

$$[\tau_J^{-1}]_M = c_M h_s^M (\Omega_s^M)^{3/2}/J_\star, \quad (69)$$

and the scaling function

$$W(\mathsf{T}) = c_W \int_0^\infty d\theta/\sqrt{\theta}[1+(\mathsf{T}-\theta)^2]^{3/2} \quad (70)$$

has the asymptotics

$$W(\mathsf{T}) = \begin{cases} (3\pi c_W/8)|\mathsf{T}|^{-5/2}, & -\mathsf{T} \gg 1, \\ 2c_W \mathsf{T}^{-1/2}, & \mathsf{T} \gg 1. \end{cases}$$

The coefficients c_M and c_W are bound by the relation $c_M c_W = 1/\sqrt{\pi}$; therefore, by satisfying $W(0)=1$, one finds $c_M = \Gamma^2(1/4)/4\pi \approx 1.046$ and $c_W = 4\sqrt{\pi}/\Gamma^2(1/4) \approx 0.539$. The numerical analysis shows that at $\mathsf{T}=0.46205$ the scaling function $W(\mathsf{T})$ reaches the maximum

$$\max W(\mathsf{T}) = 1.156\,27\ldots,$$

whence for the amplitude of catastrophe at the point of maximum one finds

$$\max[\tau_J^{-1}] = (1.156\,27\ldots)[\tau_J^{-1}]_M. \quad (71)$$

Equations (59), (61), and (63)–(70) give a detailed picture of the concentration explosion and of the annihilation catastrophe in the asymptotic limit $\mathcal{K}\to\infty$. Substituting into these expressions $J_\star = \Delta\Delta_c$ and marking the asymptotic values of amplitudes with the index (a), in full agreement with [3] we obtain

$$h_s^M(a) = p^{-1/4}\sqrt{\Delta\Delta_c}, \quad l_s^M(a) = p^{1/4}\sqrt{\Delta\Delta_c}, \quad (72)$$

$$\Omega_s^M(a) = (\mu/2)p^{-1/4}\sqrt{\Delta\Delta_c}, \quad (73)$$

$$[\tau_J^{-1}]_M(a) = (1.433\,40\ldots)p^{-5/8}\Delta_c^{-5/4}\Delta^{1/4}, \quad (74)$$

whence, adopting (71),

$$\max[\tau_J^{-1}](a) = (1.657\,39\ldots)p^{-5/8}\Delta_c^{-5/4}\Delta^{1/4}. \quad (75)$$

From (33), (34), (68), and (74) it follows that the width of the flux relaxation rate jump does not depend on p,

$$|\mathcal{T}|_{cat} \propto \Delta^{-2/5},$$

whereas its amplitude

$$\max \tau_J^{-1} \sim [\tau_J^{-1}]_M \propto p^{-5/8}(1-p)^{5/4}\Delta^{1/4}$$

grows rapidly with diminishing p. So, the smaller p, i.e., the less the L diffusion restrains the explosion development, the more brightly the effect is displayed [3].

According to (44), one of the necessary conditions for the scaling description of passage through the point of singularity is the requirement

$$\Omega_s p \gg 1,$$

which implies that, for the medium to be considered as semi-infinite in the process of explosion, the explosion rate must be much beyond the characteristic rate of the L particle diffusion. Combining this requirement with Eqs. (63) and (64) and, using the equality $\Omega_s^M(a)=\sqrt{\mathcal{K}}/2p$, one can easily see that the applicability limits of the scaling description are described by the inequalities

$$1/\sqrt{\mathcal{K}} \ll \zeta \ll \sqrt{\mathcal{K}}, \ |\mathsf{T}| \ll \sqrt{\mathcal{K}}.$$

A more rigorous requirement on the quantity $\mathcal{K}$ is imposed by the apparent chain of conditions

$$\frac{|J_{ex}^M|}{J_\star} \sim \frac{\Omega_{Ls}^M - \Omega_{Hs}^M}{\Omega_{Hs}^M} \sim \frac{[\tau_J^{-1}]_M}{\Omega_s^M} \sim \mu/\mathcal{K}^{1/4} \ll 1, \quad (76)$$

whence it is to be expected that with a growth of $\mathcal{K}$ the amplitudes (72)–(74) ought to be reached mainly by a law proportional to $\mu/\mathcal{K}^{1/4}$. One of the remarkable analytical advantages of the approach given above is that it enables one not only to determine the exact asymptotic amplitudes (72)–(74) but, also, to answer the question of *when and how* they are reached. A systematic analysis of the crossover to the asymptotics (72)–(74) is given in Appendix D. Here we shall focus on the main results. The central conclusion of the presented analysis is that Ω_{Hs}^M reaches the asymptotic limit $\Omega_s^M(a)$ much faster than Ω_{Ls}^M; therefore the point, of the explosion maximum is defined by precisely the point of the Ω_{Hs} maximum. According to (D7),

$$\Omega_{Hs}^M/\Omega_s^M(a) = 1 - B_\Omega \mu/\mathcal{K}^{1/4} + O_\Omega(\mathcal{K}^{-1/2}), \quad (77)$$

where $B_\Omega \approx 0.0318$. Taking into account the equality $\Omega_{Ls}^M = \Omega_{Hs}^M + \omega_0 + [\tau_J^{-1}]_M$ and Eq. (68), it yields

$$\Omega_{Ls}^M/\Omega_s^M(a) = 1 + (c_M/\sqrt{2} - B_\Omega)\mu/\mathcal{K}^{1/4} + \cdots. \quad (78)$$

According to (D8),

$$[\tau_J^{-1}]_M/[\tau_J^{-1}]_M(a) = 1 + B_J\mu/\mathcal{K}^{1/4} + O_J(\mathcal{K}^{-1/2}), \quad (79)$$

where $B_J \approx 0.776$. From these expressions we conclude that Ω_{Hs}^M always goes to its asymptotics from below whereas $[\tau_J^{-1}]_M$ and Ω_{Ls}^M always go to their asymptotics from above. Remarkably, the coefficient B_Ω appears to be anomalously small, so that the contribution of the $\mathcal{K}^{-1/4}$ term in the case of Ω_{Hs}^M becomes less than 0.01 already at $\mathcal{K} > 10^2$. Below, we shall present extensive numerical calculations in wide ranges of p, Δ, and $\mathcal{K}$, which demonstrate excellent agreement between the numerical data and the analytical predictions.

E. Universalization of the annihilation catastrophe

It is apparent that the scaling theory of the annihilation catastrophe, developed in the previous section for the universal limit $n_0 \to \infty$, holds in the general case of finite n_0 too. Indeed, according to (10), (11), (25), (28), and (30) at finite n_0 and $\lambda \ll 1$ the chain (52)–(62) remains valid, with the sole difference that now

$$J_\star(n_0) = \Delta\Delta_c[1 + G(n_0)] \quad (80)$$

and

$$\mathcal{C}_{(0)}(n_0) = \mu J_\star(n_0)[1 - Q(n_0)] \quad (81)$$

become functions of n_0. We thus have a complete scheme to close the chain (6)–(34) and to answer the question of when and how the universality is reached. It remains for us to find the central characteristic of scaling, namely, the amplitude of the explosion $\Omega_s^M(n_0)$, and then from Eq. (69) to derive the amplitude of the catastrophe $[\tau_J^{-1}]_M(n_0)$.

Taking $\zeta \ll 1$, from Eq. (62), we have

$$h_s = (h_s^M/2\Omega_s^M)/|\mathcal{T}|. \quad (82)$$

Matching this result with Eq. (28), thereby closing the chain (6)–(34), we obtain

$$\Omega_s^M = \frac{\mu}{2}(1-Q)h_s^M = \frac{\mu}{2}(1-Q)p^{-1/4}\sqrt{J_\star}. \quad (83)$$

Using then Eq. (30) we conclude that at $\lambda \ll 1$ the evolution of the explosion with growing n_0 is completely defined by the functions $Q(n_0)$ and $G(n_0)$, and find finally the laws of universalization of the amplitudes of the explosion,

$$\Omega_s^M(n_0) = \Omega_s^M(\infty)(1 + \delta_\Omega), \quad (84)$$

and catastrophe,

$$[\tau_J^{-1}]_M(n_0) = [\tau_J^{-1}]_M(\infty)(1 + \delta_J), \quad (85)$$

in the form

$$\delta_\Omega = G/2 - Q, \quad \delta_J = G/4 - 3Q/2. \quad (86)$$

Leaving aside details here, we distinguish two main consequences of (86).

(1) According to (29) and (31) the drop of δ_Ω and δ_J with growing n_0 is surprisingly rapid:

$$Q,G \propto n_0^{-8}(p < p_c), \quad Q,G \propto n_0^{-\chi}(p > p_c),$$

where $3 < \chi(p) < 8$. Comparing this with the relatively slow decrease of δ_s [Eq. (22)]

$$\delta_s(p < p_c) \propto n_0^{-8/9}, \quad \delta_s(p > p_c) \propto n_0^{-\chi/(\chi+1)},$$

we conclude that universalization of explosion occurs long before h_s^{min} has reached h_s^c.

(2) According to (86), in the range $p < p_c$ with decreasing p some critical values $\varrho_{8,i}^*(p_i^*)$ are reached at which δ_Ω and δ_J reverse their signs $(- \to +)$ so that, contrary to intuitive reasoning, at $p < p_\Omega^*$ and $p < p_J^*$ the amplitudes Ω_s^M and max τ_J^{-1}, respectively, drop with growing n_0. From (29), (31), and (86) we obtain

$$\varrho_{8,\Omega}^* = -1/17, \quad p_\Omega^* = 0.0609,$$

and

$$\varrho_{8,J}^* = -5/53, \quad p_J^* = 0.0217.$$

Note that this correlates with the behavior of the function δ_τ that passes through zero $(- \to +)$ at

$$p_\tau^* = 1/9.$$

IV. NUMERICAL CALCULATIONS

To test analytical predictions we have carried out extensive numerical calculations of Eqs. (1) and (2). The numerical integration of Eqs. (1) and (2) was performed by means of an implicit discretization scheme of increased accuracy with an additional fictitious node at the surface [8,9]. The scheme allowed performing the calculations in a system with strong difference in species diffusivities with an accuracy down to 10^{-3}%. The space and time steps were changed within the ranges 3×10^{-4}–3×10^{-6} and 10^{-4}–10^{-11}, respectively, with the number of time steps being 10^5–10^6.

In Figs. 2 and 3 are shown the results for $\Delta = 10^5$ and $p = 0.25$, giving a detailed picture of the universal explosion formation with growing n_0. It is seen that, in accord with Eqs. (86), already at small departures from $n_0^c(R=R_c)$ the transient dynamics (8) terminates in explosion that *remarkably rapidly* becomes universal: further growth of n_0 leads to a progressing shift of the critical point $\tau_\star(n_0)$ [Figs. 2(a) and

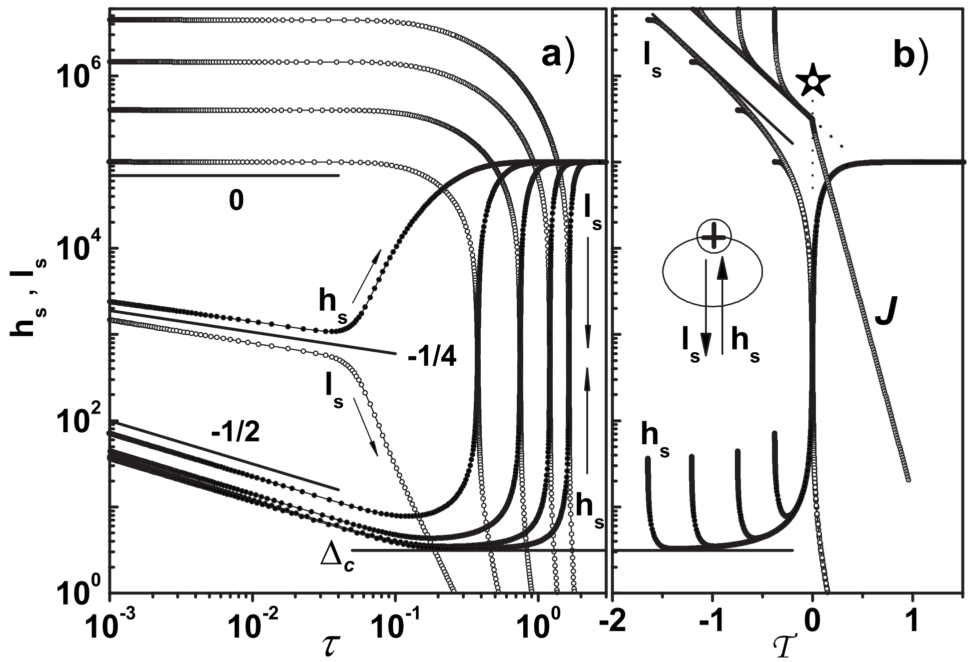

FIG. 2. Formation of universal concentration explosion with growing n_0 at $\Delta=10^5$ and $p=0.25$. (a) Numerical calculation of the behavior of $l_s(\tau)$ (open circles) and $h_s(\tau)$ (filled circles) for $n_0=1$ $(R=R_c),3,9,30,90$ (from left to right). (b) Data for $n_0=3-90$ replotted as $\log_{10}(l_s,h_s)$ vs $\mathcal{T}$ ($\mathcal{T}=0$ corresponds to Ω_{Hs}^M). Data for the flux $J=h_s l_s$ (open hexagons) are also given.

3] without changing the explosive dynamics in its vicinity but gradually universalizing the entire self-acceleration trajectory [Fig. 2(b)]. We distinguish two important points which characterize the universalization process.

(i) In accord with Eqs. (64) and (68), the symmetrical "flash" of the explosion rate Ω_{Hs} $(|\mathcal{T}|^{-1}\leftrightarrow\mathcal{T}^{-1})$ [Fig. 3(a)] and the sharply asymmetrical jump of the flux relaxation rate τ_J^{-1} $(|\mathcal{T}|^{-5/2}\leftrightarrow\mathcal{T}^{-1/2})$ that accompanies it [Fig. 3(b)] form long before the universalization of the corresponding amplitudes, shifting self-similarly with growing n_0.

(ii) In accord with Eqs. (22) and (23), as n_0 grows, the starting point of catastrophe reaches the level ω_0 [Fig. 3(b)] long before the starting point of self-acceleration, h_s^{min}, reaches the level Δ_c (Fig. 2).

In the insets in Fig. 3 are shown the dependences $\Gamma_\Omega(n_0)=\Omega_{Hs}^M(n_0)/\Omega_{Hs}^M(\infty)$ and $\tau_\star(n_0)$, plotted on the basis of the dependences $\Omega_{Hs}(\tau,n_0)$, represented on the main panel and the analogous dependences obtained in a wide p range for $\Delta=10^5$ (according to Appendix D, here and in what follows the critical point $\tau_\star$ is defined to be the point of maximum of Ω_{Hs}). The inset in Fig. 3(a) demonstrates that, in accord with (86), as p decreases, the behavior of $\Omega_{Hs}^M(n_0)$ changes qualitatively: at $p>p_\Omega^*$ the explosion amplitude $\Omega_{Hs}^M(n_0)$ grows monotonically with growing n_0, reaching $\Omega_{Hs}^M(\infty)$ from below, whereas at $p<p_\Omega^*$ the explosion amplitude first goes through a maximum and then drops with growing n_0, reaching $\Omega_{Hs}^M(\infty)$ from above. The inset in Fig. 3(b) demonstrates that at all p with growing n_0 the numerically calculated $\tau_\star$ values come to the function $\tau_\star^u(p,n_0)$ calculated according to Eq. (26). Moreover, in full agreement with Eqs. (25) and (27), at $p>p_\tau^*$ the numerical $\tau_\star$ values come to $\tau_\star^u$ from below, whereas at $p<p_\tau^*$ the numerical $\tau_\star$ values come to $\tau_\star^u$ from above. Below we shall present the results of a detailed numerical study of the catastrophe universalization in a wide range of p and Δ and compare them with the predictions of (22), (25), and (86), to plot on their basis the complete n_0-p diagram of universalization. Before discussing these results, our main goals are as follows.

(1) To demonstrate numerically how with a growth of Δ in the vicinity of the critical point $\tau_\star$ a singularity forms, and to show that with a growth of Δ and n_0 the time dependences $\Omega_{Hs}(\mathcal{T})$ and $[\tau_J^{-1}](\mathcal{T})$ collapse to the predicted scaling functions $S(\mathsf{T})$ (64) and $W(\mathsf{T})$ (70).

(2) Making use of Eqs. (86) for selecting the $n_0^u(p)$ region where the contribution of n_0 can be excluded with the required accuracy, to study numerically the behavior of the Ω_{Hs}^M and $[\tau_J^{-1}]_M$ amplitudes in a wide range of p and Δ, and to demonstrate that with a growth of $\mathcal{K}$ they reach their asymptotic values $\Omega_s^M(a)$ (73) and $[\tau_J^{-1}]_M(a)$ (74) in accord with the predictions of (D7)–(D11).

In Fig. 4 are shown the numerically calculated dependences $\Omega_{Hs}(\tau)$ that demonstrate the formation of singularity with growing Δ at $n_0=4$ and $p=0.25$. An analysis of the given data suggests that, as Δ grows, the critical point of the explosion maximum $\tau_\star(\Delta)$ rapidly (proportional to Δ^{-1}) comes to the point of singularity $\tau_\star(\infty)$, calculated according to Eq. (25), so that already at $\Delta>10^4$ the ratio $\delta\tau_\star(\Delta)/\tau_\star(\infty)$ becomes less than 0.001. In the inset to Fig. 4 are compared the dependences $\Omega_{Hs}(\tau)$ and $\Omega_{Ls}(\tau)$ calculated numerically at $\Delta=10^8$, $n_0=4$, and $p=0.25$. In accord with Eq. (64), at large Ω_s the curves are seen to merge in "synchronous" explosion, asymmetrically coming apart away from the critical point. In Fig. 5(a) are shown the data of Fig. 4 for $\Delta=10^5,10^6,10^7,10^8$, replotted in the coordinates Ω_{Hs}-$\mathcal{T}$ where $\mathcal{T}=\tau-\tau_\star$, $\tau_\star$ being the point of the explosion maximum. Here the data of Fig. 3(a) for $n_0=1.8,2.4,3.1$ $(\Delta=10^5$, $p=0.25)$

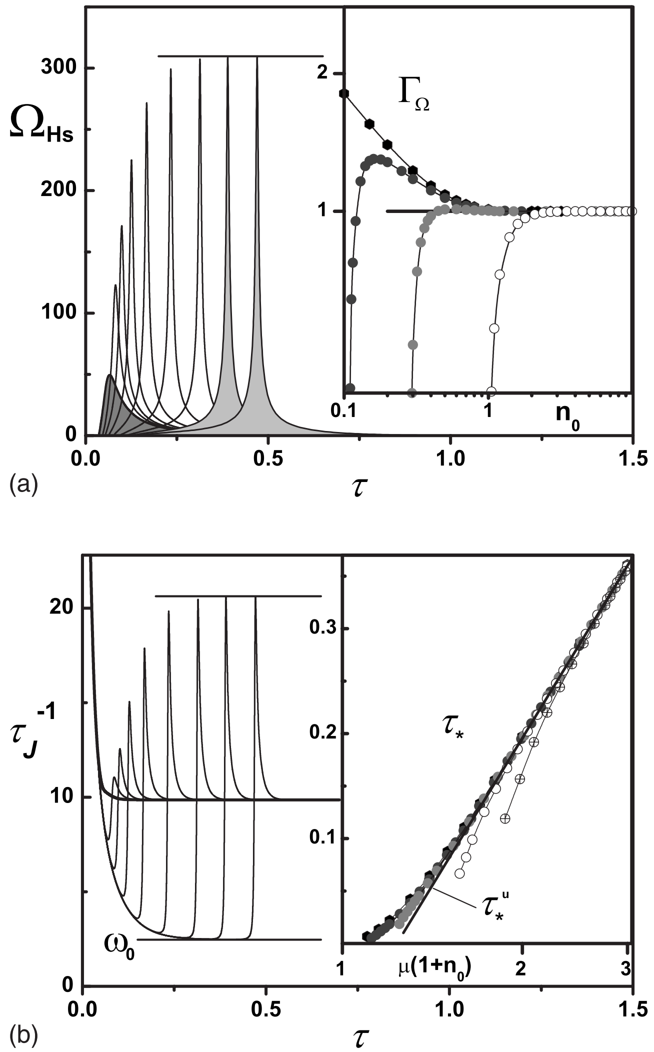

FIG. 3. Evolution of $\Omega_{Hs}(\tau)$ (a) and $\tau_J^{-1}(\tau)$ (b) with growing n_0 at $\Delta=10^5$ and $p=0.25$. The numerical calculation results are shown for $n_0=1$ $(R=R_c)$, 1.05, 1.1, 1.2, 1.4, 1.8, 2.4, 3.1, 4 (from left to right). Inset in (a): Dependences $\Gamma_\Omega(n_0)=\Omega^M_{Hs}(n_0)/\Omega^M_{Hs}(\infty)$ vs n_0 calculated numerically at $\Delta=10^5$ for $p=10^{-3}, 10^{-2}, 0.05, 0.25$ (from top to bottom). Inset in (b): Shift of the critical point $\tau_\star$ with growing n_0. Symbols show the dependences $\tau_\star(n_0)$ calculated numerically at $\Delta=10^5$ for $p=10^{-3}, 10^{-2}, 0.05, 0.25, 0.5$ (from left to right). The lines are the dependence $\tau^\mu_\star(p,n_0)$ calculated from Eq. (26).

are also represented. It is seen that, in full agreement with Eq. (64), (i) the rate of explosion $\Omega_{Hs}(\mathcal{T})$ demonstrates the remarkable symmetry $-\mathcal{T}\leftrightarrow\mathcal{T}$ and (ii) beyond the $[-\mathcal{T}_f,\mathcal{T}_f]$ region, contracting without limit with a growth of Δ, the explosion rate comes to the universal law $1/|\mathcal{T}|$. In the concluding Fig. 5(b) the data of Fig. 5(a) are represented in the scaling coordinates $\Omega_{Hs}/\Omega^M_{Hs}$-$\mathsf{T}$ where $\mathsf{T}=\mathcal{T}/\mathcal{T}_f=\mathcal{T}\Omega^M_{Hs}$. It is seen that the numerically calculated dependences perfectly collapse to the scaling function $S(\mathsf{T})$ (64).

Let us now turn to an analysis of the flux relaxation rate. Figure 6(a) demonstrates the dependences $\tau_J^{-1}(\mathcal{T})$ calculated numerically for the same parameters as in Fig. 5(a) (the data are shown only for $n_0=4$). The data analysis suggests that in accord with Eqs. (33), (34), and (68) with growing Δ in the vicinity of the critical point $\tau_\star$ a singular jump of τ_J^{-1} forms,

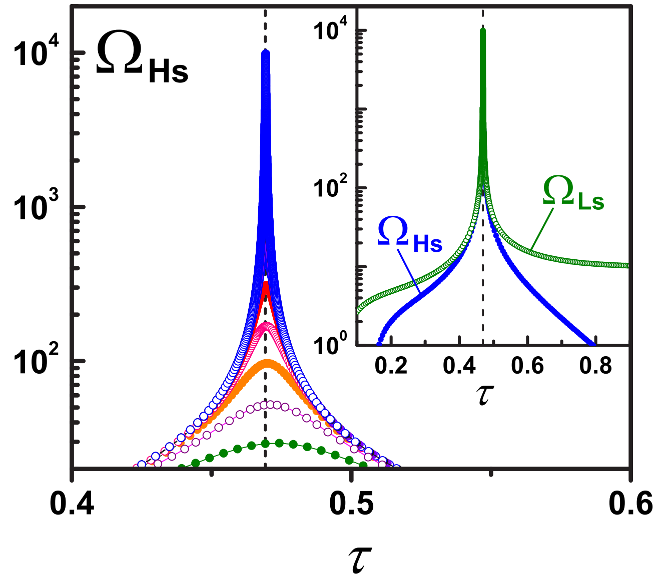

FIG. 4. (Color online) Formation of singular concentration explosion with growing Δ at $n_0=4$ and $p=0.25$. Shown are the dependences $\Omega_{Hs}(\tau)$ calculated numerically at $\Delta=10^3, 3\times10^3, 10^4, 3\times10^4, 10^5, 3\times10^5, 10^6, 10^7$, and 10^8 (from bottom to top). Only a small part of the points with density proportional to the explosion rate are given. The value of the critical point $\tau_\star$ calculated from Eq. (25) is denoted by the vertical dashed line. Inset: Synchronization of the concentration explosion in the vicinity of the critical point $\tau_\star$. Shown are the dependences $\Omega_{Ls}(\tau)$ and $\Omega_{Hs}(\tau)$ calculated numerically at $\Delta=10^8$, $n_0=4$, and $p=0.25$.

the width of which contracts by the law $|\mathcal{T}_{cat}|\propto\Delta^{-2/5}$ and the amplitude of which grows by the law $\max\tau_J^{-1}\propto\Delta^{1/4}$ (see below). Note that, in accord with Eq. (68), after the critical point has been passed ($\mathcal{T}\gg\mathcal{T}_f$), the relaxation rate drops according to the Δ-independent law proportional to $1/\sqrt{\mathcal{T}p}$, reaching at times $\mathcal{T}\sim p/\omega_0^2$ the L-diffusion-controlled limit ω_0/p. As a result, in the limit of small p there arises a most dramatic consequence of the annihilation catastrophe: an abrupt, practically instantaneous (on the scale of ω_0) disappearance of the flux [3]. Based on the data of Fig. 6(a), in accord with Eq. (33) the time dependences of the singular part of the flux relaxation rate $[\tau_J^{-1}](\mathcal{T})=\tau_J^{-1}(\mathcal{T})-\omega_0(1+w)$ were calculated, which were then replotted in the scaling coordinates $[\tau_J^{-1}](\mathsf{T})/[\tau_J^{-1}](0)$-$\mathsf{T}$. The results are demonstrated in Fig. 6(b). It is seen that, in perfect agreement with Eq. (68), with growing Δ the numerical results collapse to the scaling function $W(\mathsf{T})$. For a more detailed illustration, in Figs. 6(c) and 6(d), the data of Fig. 6(b) are represented in double-logarithmic coordinates in a wider range of $|\mathsf{T}|$ separately for $\mathsf{T}<0$ [Fig. 6(c)] and $\mathsf{T}>0$ [Fig. 6(d)]. There are also given the data based on the numerical calculation for $n_0=1.8, 2.4, 3.1$ ($\Delta=10^5$, $p=0.25$), which demonstrate a collapse to the scaling function $W(\mathsf{T})$ with growing n_0. It is seen from Fig. 6(c) that at $\Delta=10^8$ the range of the scaling growth regime of $[\tau_J^{-1}]$ reaches six orders of magnitude. As in this case at the starting point of growth the ratio $[\tau_J^{-1}]/\omega_0$ $\sim10^{-4}$, it implies that the accuracy of the numerical calculation of τ_J^{-1} reaches 10^{-3}%. Extensive numerical calculations in a wide range $10^{-3}<p<1$, a part of which will be given

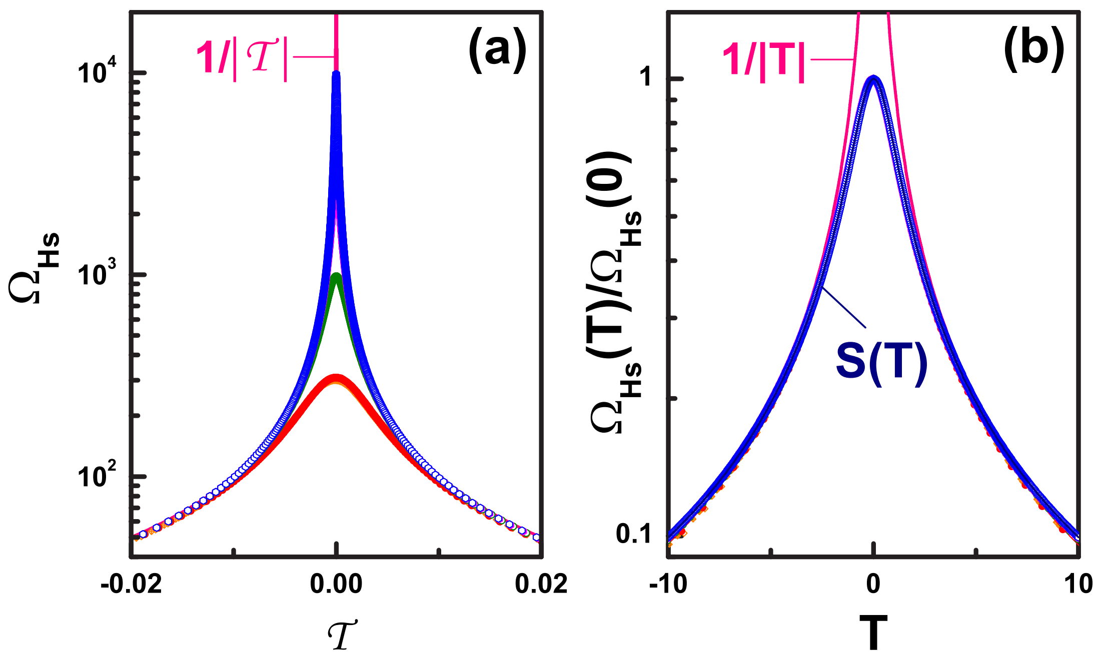

FIG. 5. (Color online) (a) Data of Fig. 4 ($n_0=4$) for $\Delta=10^5$ (red filled circles), 10^6 (green open circles), 10^7 (pink filled hexagons), and 10^8 (blue open hexagons) replotted in the coordinates Ω_{Hs} vs $\mathcal{T}$, where $\mathcal{T}=\tau-\tau_\star$ and $\tau_\star$ is the point of the explosion maximum. Also given are the data of Fig. 3(a) ($\Delta=10^5$) for $n_0=1.8$ (orange open diamonds), 2.4 (orange half-filled diamonds), and 3.1 (orange filled diamonds). (b) Collapse of the data of (a) in scaling coordinates $\Omega_{Hs}(\mathsf{T})/\Omega_{Hs}^M$ vs $\mathsf{T}=\mathcal{T}\Omega_{Hs}^M$ to the scaling function $S(\mathsf{T})$ (solid line) calculated from Eq. (64).

below, have shown that at all the investigated p values with a growth of Δ (and therefore $\mathcal{K}$) the normalized dependences $\Omega_{Hs}(\mathsf{T})$ and $[\tau_J^{-1}](\mathsf{T})$ obtained numerically collapse, respectively, to the scaling functions $S(\mathsf{T})$ and $W(\mathsf{T})$. We thus conclude that the scaling theory of catastrophe perfectly agrees with the numerical results.

Let us now come to a numerical study of the behavior of the amplitudes $\Omega_{Hs}^M(p,\Delta,n_0)$ and $[\tau_J^{-1}]_M(p,\Delta,n_0)$, which are the central characteristics of the scaling regime of catastrophe. Following the above stated program, we shall begin with the results derived in the universal limit $n_0\to\infty$. The numerical calculations were performed within $\Delta=10^4-10^8$ for $p=0.01,0.03,0.1,0.25,0.5,0.75$. In order that the contribution of the initial conditions be excluded, the initial number of particles n_0 was, depending on p, selected from the range $n_0=10-200$ so that, in accord with Eqs. (86), this contribution may not exceed $10^{-3}\%$. In Fig. 7(a) the numerically calculated dependences $\gamma_\Omega=\Omega_{Hs}^M/\Omega_s^M(a)$ and $\gamma_J=[\tau_J^{-1}]_M/[\tau_J^{-1}]_M(a)$ are shown as functions of $\mathcal{K}/\mu^4$. It is seen that with growing $\mathcal{K}$ the numerically calculated amplitudes Ω_{Hs}^M and $[\tau_J^{-1}]_M$ come, respectively, to the asymptotic values $\Omega_s^M(a)$ and $[\tau_J^{-1}]_M(a)$ calculated analytically according to Eqs. (73) and (74). Remarkably, in accord with the predictions of Eqs. (77) and (79), (i) γ_Ω comes to 1 from below whereas γ_J comes to 1 from above; (ii) γ_Ω comes to 1 much faster than γ_J; (iii) the law by which γ_J approaches 1 in a wide range of $\mathcal{K}/\mu^4$ is described with excellent accuracy by the principal term of Eq. (79). For a more detailed illustration of (iii), in Fig. 7(b) the dependences $\epsilon_J=\gamma_J-1$ vs $\mathcal{K}/\mu^4$ are presented in double-logarithmic coordinates. There are also shown the numerically calculated dependences $\epsilon_\star=(J_\star-J_M)/J_\star$ vs $\mathcal{K}/\mu^4$, which demonstrate the law by which J_M approaches $J_\star$ with growing $\mathcal{K}$. It is seen that the numerically calculated ϵ_J and ϵ_* values at p not too close to 1 perfectly fall on the analytic dependences $B_J\mu/\mathcal{K}^{1/4}$ [Eq. (79)] and $B_\star\mu/\mathcal{K}^{1/4}$ [(Eq. (D4))], respectively, shown in thick lines [note that, according to (D10), at $p\to1$, $\mu\sim1-p\to0$ the dominant term in ϵ_J contributing up to $\mathcal{K}\propto\mu^{-4}\to\infty$ becomes proportional to $1/\sqrt{\mathcal{K}}$]. In accord with Eqs. (77), (D9), and (D11), owing to the anomalous smallness of the coefficient B_Ω, the value of $\epsilon_\Omega=1-\gamma_\Omega$ ought to decrease with growing $\mathcal{K}$ according to the law proportional to $p/\sqrt{\mathcal{K}}$ or (at small p) even faster, down to very low values of $\epsilon_\Omega\sim10^{-3}$. To illustrate these predictions in Fig. 7(c) the dependences of ϵ_Ω on $\sqrt{\mathcal{K}}$ are shown in double-logarithmic coordinates. At comparatively high p, the numerically calculated ϵ_Ω values are seen to fairly fall on the analytic dependences shown as dashed lines. As p decreases, the effective "rate" of ϵ_Ω drop grows only insignificantly; the ϵ_Ω value itself becomes very small already at $\mathcal{K}\sim10^2-10^3$; therefore, the analytical description of ϵ_Ω in this region necessitates additional terms. Summarizing, we conclude that in the universal limit the represented theory gives an exhaustive picture of the evolution of the catastrophe and explosion amplitudes.

It only remains for us to complete this section by demonstrating the results of extensive numerical study of the regularities of catastrophe universalization with growing n_0. We have studied the behavior of the dependences $\tau_\star(n_0)$, $\Omega_{Hs}^M(n_0)$, $[\tau_J^{-1}]_M(n_0)$, and $h_s^{min}(n_0)$ on scanning n_0 from n_0^c to 10^4 in wide ranges of $\Delta=10^5-10^8$ and $p=10^{-3}-0.97$. Based on the data obtained for each of the studied p and Δ values, we calculated the dependences $\delta_\tau(n_0)=\tau_\star(n_0)/\tau_\star^u-1$, $\delta_\Omega(n_0)=\Gamma_\Omega(n_0)-1$, $\delta_J(n_0)=[\tau_J^{-1}]_M(n_0)/[\tau_J^{-1}]_M(\infty)-1$, and $\delta_s(n_0)$

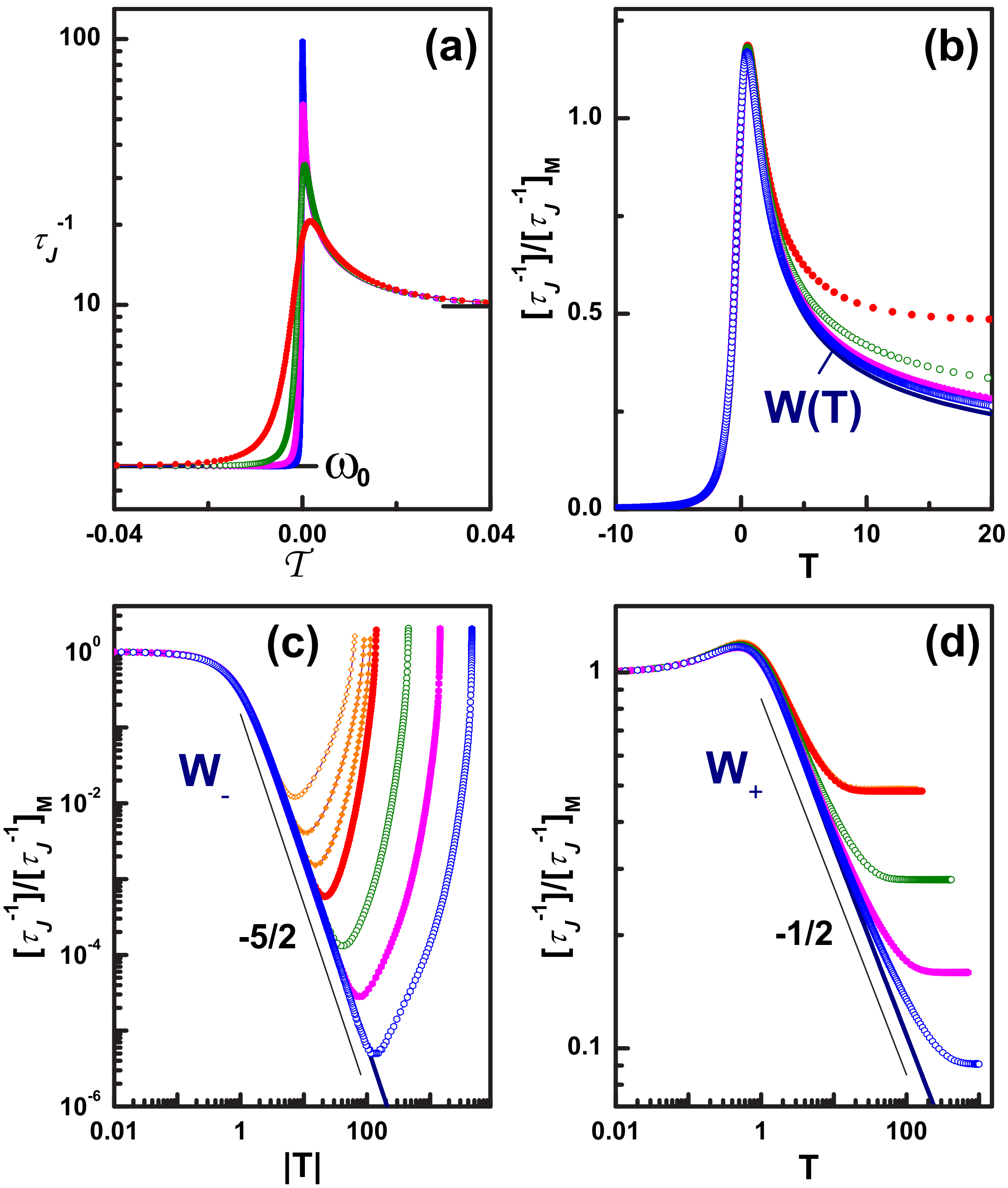

FIG. 6. (Color online) (a) Formation of singular jump of the flux relaxation rate τ_J^{-1} with growing Δ at $n_0=4$ and $p=0.25$. Shown are the dependences $\tau_J^{-1}(\mathcal{T})$ calculated numerically at $\Delta=10^5$ (red filled circles), 10^6 (green open circles), 10^7 (pink filled hexagons), and 10^8 (blue open hexagons) (from left to right). (b) Collapse of the dependences $[\tau_J^{-1}](\mathsf{T})/[\tau_J^{-1}](0)$ vs $\mathsf{T}=\mathcal{T}\Omega_{Hs}^M$ calculated from the data of (a) to the scaling function $W(\mathsf{T})$ (solid line) calculated from Eq. (70). The singular part of the relaxation rate was calculated from Eq. (33). (c),(d) The data of (b) replotted in double-logarithmic coordinates in a wider range of $|\mathsf{T}|$ for the ascending $\mathsf{T}<0$ (c) and descending $\mathsf{T}>0$ (d) catastrophe branches. Also given are the data for $\Delta=10^5$ based on numerical calculation at $n_0=1.8$ (orange open diamonds), 2.4 (orange half-filled diamonds), and 3.1 (orange filled diamonds), which demonstrate collapse to the scaling function $W(\mathsf{T})$ with growing n_0.

$=h_s^{min}(n_0)/\Delta_c-1$, which we then compared with the analytic predictions. We have found that in the region of small $\delta_i(n_0)$ $(i=\tau,\Omega,J,s)$ the behavior of the functions $\delta_i(n_0)$ is described with remarkable exactness by Eqs. (25), (86), and (22). Figure 8 represents the concluding n_0-p diagram of universalization, where are compared the positions of the boundaries $|\delta_i|=0.01$ $(i=\tau,\Omega,J)$ and $\delta_s=0.1$ resulting from a great number of numerical data (some of which are given in the inset) for $\Delta=10^5$ (in the case $i=J$ for $\Delta=10^7$) and calculated from (86), (27), and (13). The excellent agreement of the analytic and numerical results (not shifting with further growth of Δ [10]) needs no comments.

V. DISCUSSION AND CONCLUSION

Finite-time singularities—blowup solutions developing from smooth initial conditions at a particular time—provide probably the most dramatic manifestation of strongly nonlin-

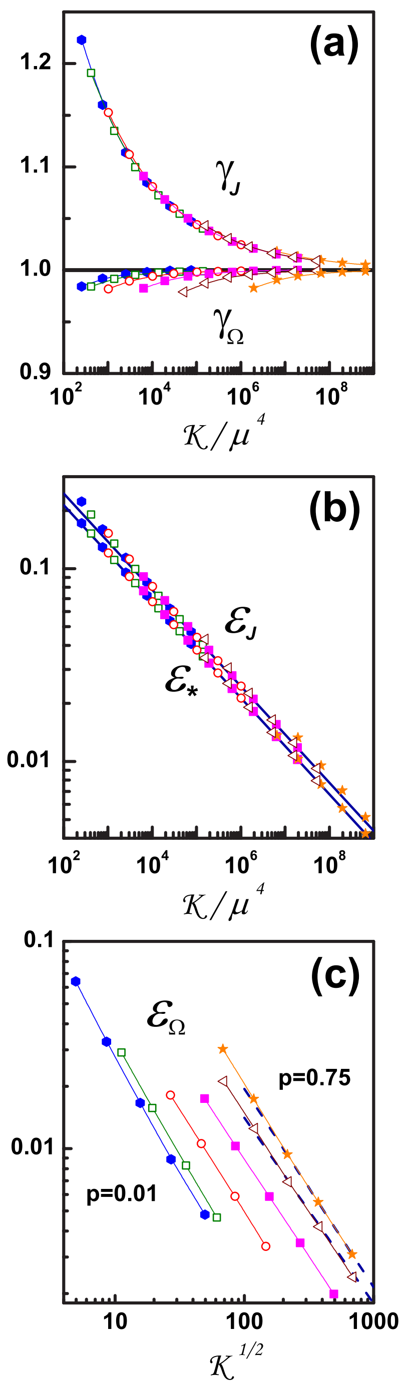

FIG. 7. (Color online) Regularities by which the universal amplitudes Ω_{Hs}^M and $[\tau_J^{-1}]_M$ approach the asymptotic values $\Omega_s^M(a)$ Eq. (73) and $[\tau_J^{-1}]_M(a)$ Eq. (74) with growing $\mathcal{K}$. (a) Dependences $\gamma_\Omega = \Omega_{Hs}^M/\Omega_s^M(a)$ and $\gamma_J = [\tau_J^{-1}]_M/[\tau_J^{-1}]_M(a)$ vs $\mathcal{K}/\mu^4$ calculated numerically within $\Delta = 10^4 - 10^8$ for $p=0.01$ (hexagons), 0.03 (open squares), 0.1 (circles), 0.25 (filled squares), 0.5 (triangles), and 0.75 (stars). Depending on p, the initial number of particles n_0 was selected from the range $n_0 = 10-200$ so that the contribution of the initial conditions could be ignored. (b) Comparison of the numerically calculated dependences $\epsilon_J = \gamma_J - 1$ and $\epsilon_\star = (J_\star - J_M)/J_\star$ vs $\mathcal{K}/\mu^4$ with the analytical laws $B_J\mu/\mathcal{K}^{1/4}$ and $B_\star\mu/\mathcal{K}^{1/4}$ (solid lines). (c) Numerically calculated dependences $\epsilon_\Omega = 1 - \gamma_\Omega$ vs $\sqrt{\mathcal{K}}$. For $p = 0.5, 0.75$ are shown the analytical dependences calculated from Eqs. (D7), (D9), and (D11) (dashed lines).

ear effects that can occur in nature [11]. The formation of finite-time singularities is observable in a wide spectrum of nonlinear systems (Yang-Mills fields [12], black holes [13], self-gravitating Brownian particles [14], turbulent flows [15], jet eruption [16], chemotaxies [17], and earthquakes [18], to name only a few); therefore, the description of scenarios of finite-time singularities development is a fundamental problem which attracts wide interdisciplinary interest.

In this paper, a systematic theory of formation of the universal annihilation catastrophe from a smooth initial distribution has been developed, and extensive numerical calculations of the regularities of the catastrophe formation in a wide range of parameters have been presented. The main results may be formulated as follows.

(1) The exact condition has been found for the H-diffusion-controlled annihilation regime.

(2) A rigorous proof of the threshold for onset of self-acceleration has been given.

(3) An exact expression has been obtained for the time point of the catastrophe.

(4) A closed scaling theory of passage through the point of singularity has been given, and the scaling laws for the concentration explosion and for the annihilation catastrophe have been derived.

(5) The laws of universalization of the concentration explosion and of the annihilation catastrophe have been found; their surprisingly rich structure has been revealed.

(6) A remarkable agreement has been found between the analytical predictions and the results of numerical calculations.

Summarizing, we believe that the analysis presented may pretend to be one of the most striking examples of a detailed description of formation of the finite-time singularity. We shall distinguish here the two most bright features of the annihilation catastrophe.

(i) In the majority of the models which demonstrate the formation of finite-time singularities, an analytical description for the singularity development (based on properties of self-similarity) appears possible only in some narrow vicinity of the critical point, beyond which the solution cannot as a rule be continued or is impossible in principle. One of the main advantages of the theory presented here is the asymptotically exact scaling description of passage through the point of singularity, which yields a complete dynamical picture at *both sides* of the critical point.

(ii) Arising as a result of explosive growth of the antiflux J_{ex} at the background of slow relaxation of the diffusion-controlled flux $J^{(0)}$, the annihilation catastrophe demonstrates a peculiar singular behavior at which two explosive processes (Ω_{Hs} and Ω_{Ls}) are developing simultaneously, effectively compensating one another, so that for an external observer of flux (J) the explosion dynamics goes unnoticed up to the critical point $\tau_\star$, in the vicinity of which decompensation of the explosions is manifested as a sudden singular jump of the flux relaxation rate. In the limit of small p this brings about a most radical consequence—an *abrupt disappearance* of the flux.

Let us discuss in conclusion the conditions and possibilities of experimental observation of the annihilation catastrophe. The irreversible bimolecular reaction $A+B\to 0$ is one of

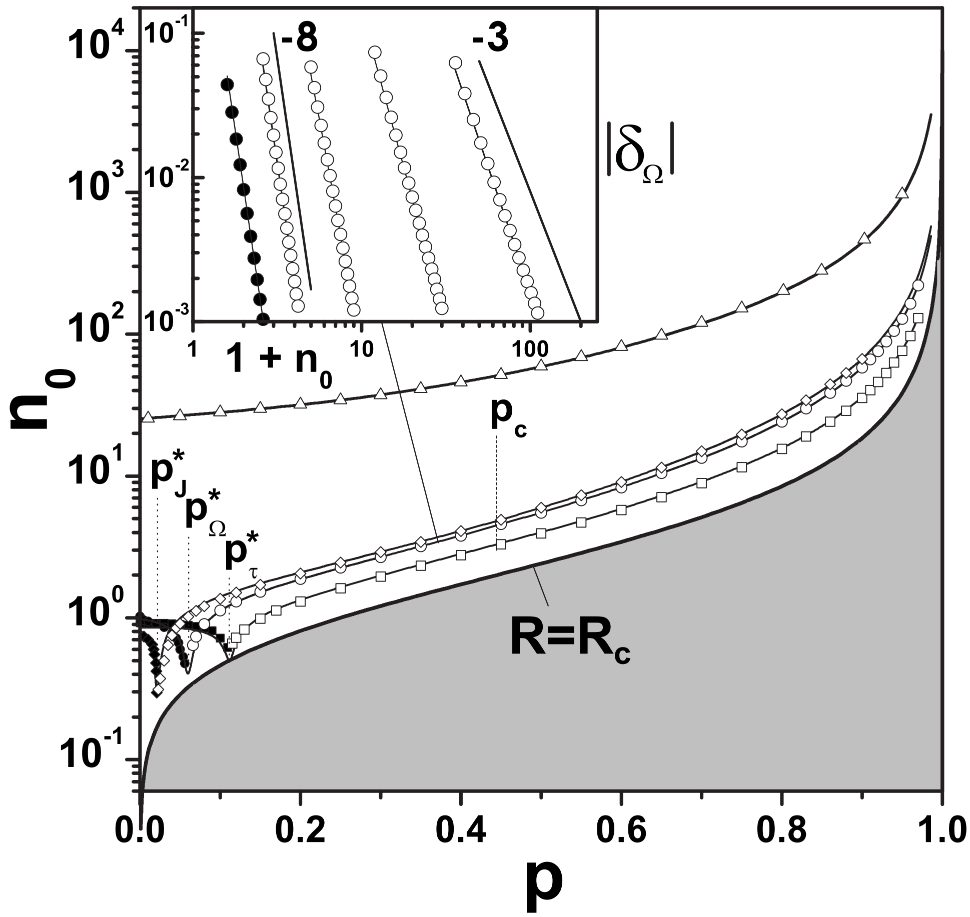

FIG. 8. n_0-p diagram of universalization of the concentration explosion and of the annihilation catastrophe. Shown are numerically (symbols) and analytically (lines) calculated boundaries $|\delta_\tau|=0.01$ (squares), $|\delta_\Omega|=0.01$ (circles), $|\delta_J|=0.01$ (diamonds), and $\delta_s=0.1$ (triangles). Open symbols, $\delta_i<0$; filled symbols, $\delta_i>0$. Inset: Numerically (circles) and analytically (lines) calculated dependences $|\delta_\Omega|$ vs $1+n_0$ at $p=0.01$, 0.25, 0.5, 0.75, and 0.9 (from left to right).

the most abundant reactions. Therefore, it is to be expected that the predicted phenomena can, in principle, be observed in a wide class of physical, chemical, and biological systems with a catalytic interface, which, because of a high energetic barrier, does not let diffusing particles A (B) pass from medium 1 (2) into medium 2 (1), so that the reaction $A+B \to 0$ can occur only at the interface between the media [4,19,20]. Leaving aside here a discussion of systems of this type, we shall focus on the main object of the model in question, namely, adsorption-desorption systems (Fig. 1). Until now, most of the theoretical studies on the $A_{ads}+B_{ads} \to 0$ catalytic reaction (the Langmuir-Hinshelwood process, which is also often referred to as the monomer-monomer catalytic scheme) have been performed under the assumption that diffusion into the bulk can be neglected ([21–29] and references therein). Such an assumption is valid in low-temperature systems with high surface-bulk crossover barriers, i.e., in systems with negligibly small bulk solubility of A and B particles. Here, we address the wide class of catalytic systems where the surface-bulk crossover barriers are not too high and, therefore, adsorption-desorption processes are always followed by a more or less intensive diffusion of A and B particles into or from the bulk, where reaction between A's and B's is energetically forbidden [30]. This class of catalytic system is not only of fundamental interest for surface science but, also, of considerable applied interest for describing the interaction kinetics of gases with metals at high temperatures ([30–33] and references therein). In [34] a theory has been developed for the diffusion-controlled associative desorption of like particles, $A_{ads}+A_{ads} \to A_{2,gas} \to 0$, from the dissolved state into vacuum. Adopting this theory, a complete picture of diffusion-controlled thermodesorption of hydrogen and nitrogen has been constructed, in good agreement with the available experimental data. The theory reported in this work gives a systematic description of the diffusion-controlled kinetics of associative desorption into a vacuum of unlike particles,

$$A_{ads} + B_{ads} \to AB_{gas} \to 0,$$

which are initially uniformly dissolved in the bulk. We shall focus here on discussing the possibility of observation of the predicted effects for one of the most important surface reactions of carbon monoxide CO thermodesorption from metals into vacuum,

$$C_{ads} + O_{ads} \to CO_{gas} \to 0.$$

It is to be mentioned first that the continual description (1) and (2) holds as long as the diffusion length of the explosion at the point of maximum remains much greater than the monolayer thickness a [3], $\delta x_M \sim 1/\sqrt{\Omega_s^M} \gg a/\ell$, whence there follow the limitations

$$\Omega_s^M \ll (\ell/a)^2, \quad \mathcal{K} \ll p^2(\ell/a)^4.$$

Taking, for example, $\ell/a \sim 10^3$ and $p \sim 0.01$, we come to the requirements $\Omega_s^M \ll 10^6$ and $\mathcal{K} \ll 10^8$, to see that at any value of the reaction rate constant κ the specimens must have macroscopic sizes in order for a considerable effect be observed. Based on the data of [31], we shall make estimations for three refractory metals, i.e., niobium, tantalum, and molybdenum, which at elevated temperatures dissolve carbon and oxygen in quite large amounts. According to [31], at temperatures of intensive thermodesorption of CO in the range from $T \sim 1600$ °C to the melting point, for the coefficients of carbon and oxygen diffusion in these metals we find, respectively, $D_{\mathrm{C}} \sim 10^{-7}-10^{-5}$ cm^2/s and $D_{\mathrm{O}} \sim 10^{-5}-10^{-4}$ cm^2/s, whence it follows that $p = D_{\mathrm{C}}/D_{\mathrm{O}} \sim 10^{-2}-10^{-1}$. According to the data of [31–33], the desorption rate constant of CO in the said temperature range alters within $\kappa \sim 10^{-23}-10^{-18}$ cm^4/s. Substituting these values into the expression

$$\Delta = \delta_{\mathrm{C}}(0)\kappa\ell/D_{\mathrm{C}}$$

and taking $\delta_{\mathrm{C}}(0) = c_{\mathrm{C}}(0) - c_{\mathrm{O}}(0) \sim 10^{20}$ cm^{-3} and $\ell \sim 0.1$ cm, we find that in the said temperature range the Δ parameter value changes within $\Delta \sim 10^2-10^6$. For the density of the diffusion-controlled desorption flux of CO at the critical point, we find $I_\star \sim D_{\mathrm{C}}\delta_{\mathrm{C}}(0)/\ell \sim 10^{14}-10^{16}$ particles/cm^2 s. We thus conclude that, in a study of isothermal desorption of CO at elevated temperatures under high vacuum, the predicted sharp jump of the flux relaxation rate can confidently be registered experimentally with a standard measuring technique.

Finally, we would like to point out one of the many lines of generalization of the results represented in this paper. In recent works by O' Shaughnessy and Vavylonis [4,19] it was shown analytically and demonstrated by numerical simulation that, in the problem of diffusion-controlled interfacial annihilation $A+B \to 0$, the critical bulk dimension, below which fluctuation effects become essential, is $d_c = 1$. This implies that the quasi-one-dimensional mean-field theory represented here should give the adequate description of the annihilation catastrophe development in all physical dimensions $d=3$ (catalytic surface), 2 (catalytic line) and 1 (catalytic point) with possible logarithmic corrections in the case $d_c = 1$. It should be noted, however, that the many-particle analysis performed in [4,19] was carried out only for an initial transient stage of annihilation (semi-infinite medium) at equal species diffusivities. Therefore, it would be of great interest to carry out extensive numerical simulations of low-dimensional $(d=1,2)$ discrete systems to reveal the role of fluctuation effects in the annihilation catastrophe development.

ACKNOWLEDGMENTS

This research was financially supported by the RFBR through Grants No. 05-03-33143 and No. 02-03-33122.

APPENDIX A: SYSTEMATIC ANALYSIS OF THE EXPONENTIAL STAGE OF ANNIHILATION AT $|\Lambda| \ll 1$

According to Eq. (13), at $|\Lambda| \ll 1$ and $\tau \gtrsim 1$, dropping the terms $O(\Lambda^2, \Lambda e^{-8\omega_0\tau}, \ldots)$, one has

$$h_s = \Delta_c(1 + e^{-8\omega_0\tau} + \Lambda + \cdots). \tag{A1}$$

From Eqs. (A1) and (12), it follows that at $\Delta = \mathcal{C}$ the value $\Lambda_+ = 0$ and, therefore, $\lim_{\tau\to\infty} h_s = \Delta_c$. But, by the definition, $\lim_{\tau\to\infty} h_s = \Delta$, so we conclude that $\mathcal{C} = \Delta_c$. In what follows we give a systematic analysis of the corrections to Eqs. (10)–(13) arising in the diffusion-controlled limit $h_0/\Delta_c \to \infty$.

According to Eqs. (5) the corrections $\delta J = J - J^{(0)}$ and $\delta l_s = l_s - l_s^{(0)}$ as functions of $\delta h_s = h_s - \Delta_c$ are determined by Eqs. (15) and (16), respectively. We shall begin with the analysis of the contribution of the long-time tail (A1)

$$\delta h_s = \Delta_c(e^{-8\omega_0\tau} + \Lambda + \cdots), \tag{A2}$$

and then we shall separately consider the contribution of δh_s^{tr} occurring at a transient stage.

Substituting (A2) into Eq. (15), for the total flux $J = J^{(0)} + \delta J$ one obtains

$$J = \mathcal{A}e^{-\omega_0\tau}\{1 + a_0\epsilon_{\mathcal{A}} + e^{-8\omega_0\tau}[1 + O(\varepsilon/\Delta_c)] + O(\varepsilon\Lambda/\Delta_c) + \cdots\}. \tag{A3}$$

Substituting then (A2) into Eq. (16), for the total value $l_s = l_s^{(0)} + \delta l_s$ one finds

$$l_s = (\mathcal{A}/\Delta_c)e^{-\omega_0\tau}\{1 + a_0\epsilon_{\mathcal{A}} + e^{-8\omega_0\tau}O(\varepsilon) - \Lambda[1 + O(\varepsilon)] + \cdots\}. \tag{A4}$$

We introduce here the designations

$$\epsilon_{\mathcal{A}} = \Delta_c/\mathcal{A}, \quad \varepsilon(\tau) = \Delta_c\epsilon_{\mathcal{A}}e^{\omega_0\tau},$$

and, for simplicity, in ε terms give only the order of magnitude. The coefficient a_0 is given to emphasize the like shift of the amplitude $\delta\mathcal{A}/\mathcal{A} = a_0\epsilon_{\mathcal{A}}$ in Eqs. (A3) and (A4). Using (A3) and (A4), from the condition $h_s = J/l_s$ instead of (A1) one finds

$$h_s = \Delta_c\{1 + e^{-8\omega_0\tau}[1 + O(\varepsilon)] + \Lambda[1 + O(\varepsilon)] + \cdots\}. \tag{A5}$$

It is easy to check that repeated use of this procedure leads to self-consistent appearance of the terms $O(\epsilon_{\mathcal{A}}^2, \varepsilon^2)$, $O(\epsilon_{\mathcal{A}}^3, \varepsilon^3)$, and so on.

Let us now calculate the contribution of the transient stage. Our goal will be to estimate this contribution in the order of magnitude. It is apparent that extrapolating the $h_s^{tr} \sim r/\epsilon\sqrt{\pi\tau}$ and $h_s \sim \Delta_c$ asymptotes to the point of their intersection, $\tau_{tr} \sim (r/\epsilon\Delta_c)^2/\pi$, one can take with fair approximation (see Fig. 2 for illustration) that

$$\delta h_s^{tr} \sim \begin{cases} r/\epsilon\sqrt{\pi\tau}(1 - \sqrt{\tau/\tau_{tr}}), & \tau \leq \tau_{tr}, \\ 0, & \tau > \tau_{tr}. \end{cases} \tag{A6}$$

Following the definition, $\epsilon = 1 - (1 + 1/n_0)\sqrt{p}$, therefore with allowance for the requirement $n_0 \gg \Delta_c$, necessary for satisfaction of the conditions $\varepsilon \ll 1$ at $\Delta/\Delta_c \sim 1$ and $\lambda \ll 1$ at $\Delta/\Delta_c \gg 1$ [see Eq. (A15)], one has

$$\epsilon \sim \mu/(1+\sqrt{p}), \tag{A7}$$

where $\mu = \omega_0/\Delta_c \sim 1-p$. In the limit $p \to 1$ the value $\Delta_c \sim 2/(1-p) \to \infty$ and $\epsilon \sim (1-p)/2 \to 0$, so that $\lim_{p\to 1}(\epsilon\Delta_c) \sim 1$. We thus conclude that in the range $0<p<1$ the value τ_{tr} changes in the interval

$$1/\pi\omega_0^2 < \tau_{tr} < 1/\pi. \tag{A8}$$

Representing Eqs. (15) and (16) in the form of time convolution and using Eq. (A6), we find

$$\delta J_{tr} \sim -(r/\epsilon)\frac{d}{d\tau}\left(e^{-\omega_0\tau}\int_0^{\tau_{tr}} d\theta\, e^{\omega_0\theta}\varphi(\theta) + e^{-9\omega_0\tau}\int_0^{\tau_{tr}} d\theta\, e^{9\omega_0\theta}\varphi(\theta) + \cdots\right) \tag{A9}$$

and

$$\delta l_s^{tr} \sim (r/\epsilon\Delta_c)\omega_0 e^{-\omega_0\tau}\left(\int_0^{\tau_{tr}} d\theta\, e^{\omega_0\theta}\varphi(\theta) - c_p e^{-\chi\omega_0\tau}\int_0^{\tau_{tr}} d\theta\, e^{(\chi+1)\omega_0\theta}\varphi(\theta) - c_8 e^{-8\omega_0\tau}\int_0^{\tau_{tr}} d\theta\, e^{9\omega_0\theta}\varphi(\theta) + \cdots\right), \tag{A10}$$

where $\varphi(\theta) = 1/\sqrt{\theta} - 1/\sqrt{\tau_{tr}}$, $c_8 = 9\varrho_8$, and $c_p = (\chi+1)\varrho_p$. From Eqs. (A9) and (A10) with account taken of Eq. (A7), it follows that the corrections, arising at the transient stage, are of the order $O(\epsilon_{\mathcal{A}})$ [for example, the relative shift of the amplitude is $\delta\mathcal{A}_{tr}/\mathcal{A} \sim (r/\epsilon\mathcal{A})\sqrt{\tau_{tr}} \propto O(\epsilon_{\mathcal{A}})$]; therefore in the diffusion-controlled limit $\epsilon_{\mathcal{A}} \sim \Delta_c/2h_0 \to 0$ these corrections become vanishingly small.

We conclude that the principal condition of applicability of the diffusion-controlled solution (10)–(13) is the requirement of smallness of the exponentially growing function

$$\varepsilon(\tau) = (\Delta_c^2/2h_0)e^{\omega_0\tau}. \tag{A11}$$

In turn, the necessary condition of smallness of $\varepsilon(\tau)$ is the requirement $\Delta_c^2 \ll h_0$ which, in the vicinity of $p \sim 1$ (due to the condition $\Delta_c\epsilon \sim 1$), is in conformity with the requirement $\epsilon \gg 1/\sqrt{N_0}$. By assuming that ε is small and neglecting the terms $O(\epsilon_{\mathcal{A}}, \varepsilon^2, \ldots)$, as a result of exact calculation we find

$$h_s = \Delta_c[1 + (1+\varrho_8)e^{-8\omega_0\tau}(1 + a_8\varepsilon + \cdots) + \varrho_p e^{-\chi\omega_0\tau}(1 + a_\chi\varepsilon + \cdots) + \Lambda_+(1 + a_\lambda\varepsilon + \cdots) + \cdots], \tag{A12}$$

where the coefficients a_α for $\alpha = 8, \chi$ are given by

$$a_\alpha = \tan\sqrt{\alpha\omega_0}(\sqrt{\alpha\omega_0}/\Delta_c - \sqrt{p}\cot\sqrt{\alpha\omega_0 p}), \tag{A13}$$

the coefficient a_λ is given by

$$a_\lambda = -\tanh\sqrt{\omega_0}(\sqrt{\omega_0}/\Delta_c + \sqrt{p}\coth\sqrt{p\omega_0}), \tag{A14}$$

and the amplitude of the exponentially growing function $\Lambda_+ = \lambda e^{\omega_0\tau}$ is

$$\lambda = \frac{\Delta_c(\Delta - \Delta_c)}{2h_0}. \tag{A15}$$

In the diffusion-controlled limit $\Delta_c^2/h_0 \to 0$, from Eqs. (A11)–(A15), we come to the following conclusions.

(a) At any Δ in the limit $|\lambda| \to 0 (n_0 \to \infty) h_s$ relaxes to the critical asymptotics $h_s^c = \Delta_c$ according to the law

$$\delta h_s/\Delta_c \propto e^{-\alpha\omega_0\tau}, \tag{A16}$$

where, in the $0<p<p_c=4/9$ range, $\alpha=8$, and, in the $p_c<p<1$ range, $\alpha=\chi(p)$. At $p=p_c$ one has a resonant solution

$$\delta h_s/\Delta_c \propto (1+\varrho_c\tau)e^{-8\omega_0\tau} \tag{A17}$$

with $\varrho_c = 2/9\mu$.

(b) With growth of Δ, when the threshold value $\Delta = \Delta_c$ is reached, the amplitude λ reverses its sign and the behavior of h_s changes qualitatively: at $\Delta \leqslant \Delta_c$, h_s always drops monotonically, whereas, at $\Delta > \Delta_c$, h_s first reaches a minimum $h_s^{min} \approx \Delta_c$ and then begins to grow exponentially. Setting $\varepsilon \ll 1$ for the time of start of h_s growth, one finds

$$\tau_s = [1/\omega_0(\alpha+1)]\ln(\alpha\tilde{\varrho}/\lambda), \tag{A18}$$

and for the departure of the starting point h_s^{min} from the critical asymptotics h_s^c, $\delta_s = (h_s^{min} - \Delta_c)/\Delta_c$, one obtains

$$\delta_s = \varsigma\lambda^{\alpha/(\alpha+1)}[1 + O(\delta_s) + O(\varepsilon_s)], \tag{A19}$$

where $\varsigma = (\alpha+1)\tilde{\varrho}^{1/(\alpha+1)}/\alpha^{\alpha/(\alpha+1)}$, $\tilde{\varrho} = 1+\varrho_8$ in the $0<p<p_c$ range and $\tilde{\varrho} = \varrho_p$ in the $p_c<p<1$ range. According to Eqs. (A11) and (A15), the ratio

$$\varepsilon/\Lambda_+ = \frac{\Delta_c}{(\Delta - \Delta_c)}. \tag{A20}$$

Thus, the requirement $\varepsilon_s \ll 1$ reduces to the condition

$$\delta_s \propto \Lambda_+^s \propto \lambda^{\alpha/(\alpha+1)} \ll \frac{(\Delta - \Delta_c)}{\Delta_c}, \tag{A21}$$

or, equivalently, to the condition

$$(\Delta_c^2/h_0)^\alpha \ll \frac{(\Delta - \Delta_c)}{\Delta_c}. \tag{A22}$$

(c) According to [5], at the critical point $\Delta = \Delta_c$ in the long-time limit $\tau \to \infty$ ($\varepsilon \to \infty$), δh_s, regardless of the initial conditions, relaxes by the law

$$\delta h_s/\Delta_c \propto e^{-\pi^2\tau}, \quad \tau \to \infty. \tag{A23}$$

The comparison of (A12) and (A23) leads one to conclude that there is a characteristic value $p_\varepsilon = 4/5$ above which ($p_\varepsilon < p < 1$) and below which ($0<p<p_\varepsilon$) the crossover from (A16)–(A23) is accompanied by a growth and a drop, respectively, of the relaxation rate. Remarkably, at $p = p_\varepsilon(\chi=4)$ the coefficient $a_\chi(p)$ passes through zero; therefore, by neglecting the term $O[e^{-8\omega_0\tau}(1+a_8\varepsilon+\cdots)]$, one obtains the exact law of relaxation $\delta h_s/\Delta_c = \varrho_{4/5}e^{-\pi^2\tau}$. Note that, at $\Delta = \Delta_c$, for any $0<p<1$, at the crossover region $\varepsilon \sim 1$

$$\delta h_s^{\varepsilon}/\Delta_c \propto (\Delta_c^2/h_0)^{\alpha} \tag{A24}$$

so that $\delta h_s^{\varepsilon}/\Delta_c \to 0$ as $\Delta_c^2/h_0 \to 0$. Setting, for example, $h_0 = 10^5$, one finds $\delta h_s^{\varepsilon}/\Delta_c \sim 10^{-6}$ at $p=0.9$ and $\delta h_s^{\varepsilon}/\Delta_c \sim 10^{-40}$ at $p=0.1$.

APPENDIX B: EXACT EXPANSIONS IN POWERS OF $\Lambda_+ < 1$

According to Eq. (A20), in the limit $\Delta/\Delta_c \to \infty$ the ratio $\varepsilon/\Lambda_+ \to 0$. This important fact suggests that, in the diffusion-controlled limit of primary interest here, $h_0/\Delta_c \to \infty$, $\Delta/\Delta_c \to \infty$, $\lambda < 1$, Eqs. (A10)–(A13) ought to give an exact description of the annihilation dynamics to the point of finite-time singularity $\Lambda \to 1$. Below we substantiate rigorously this suggestion in terms of the exact expansions of $\delta J/J^{(0)}$, $\delta l_s/l_s^{(0)}$, and $\delta h_s/\Delta_c$ in powers of Λ_+.

For simplicity, we will consider the limit $\lambda \to 0$ so that the terms $O(e^{-8\omega_0\tau}, \Lambda_-)$ may be neglected. According to (13), in this limit h_s grows via the law

$$h_s = \Delta_c/(1-\Lambda_+). \tag{B1}$$

At $\Lambda_+ < 1$, one has exactly

$$\frac{1}{1-\Lambda_+} = 1 + \sum_{n=1}^{\infty} \Lambda_+^n, \tag{B2}$$

whence it follows that

$$\delta h_s/\Delta_c = \sum_{n=1}^{\infty} \Lambda_+^n. \tag{B3}$$

Our goal will be to calculate the corrections through sequential iterations following Eqs. (15) and (16). We start with the equation

$$\delta h_s/\Delta_c = \Lambda_+ + \cdots. \tag{B4}$$

Substituting (B4) into Eq. (15) and allowing for Eq. (10), we find

$$\delta J/J^{(0)} = \mathcal{I}_2 \Lambda_+^2 + \cdots, \tag{B5}$$

where

$$\mathcal{I}_2 = -\frac{\sqrt{\omega_0}\tanh\sqrt{\omega_0}}{(\Delta-\Delta_c)},$$

and then substituting (B4) into Eq. (16) and allowing for Eq. (11) we obtain

$$\delta l_s/l_s^{(0)} = \mathcal{L}_2 \Lambda_+^2 + \cdots, \tag{B6}$$

where

$$\mathcal{L}_2 = \frac{\Delta_c\sqrt{p}\tanh\sqrt{\omega_0}\coth\sqrt{p\omega_0}}{(\Delta-\Delta_c)}.$$

Using (B5) and (B6), from the condition $h_s = J/l_s$ we derive

$$\delta h_s/\Delta_c = \Lambda_+ + \Lambda_+^2(1-\mathcal{H}_2) + \cdots, \tag{B7}$$

where

$$\mathcal{H}_2 = \frac{\tanh\sqrt{\omega_0}(\sqrt{\omega_0} + \Delta_c\sqrt{p}\coth\sqrt{p\omega_0})}{(\Delta-\Delta_c)}.$$

Starting with Eq. (B7), at the next iteration we find

$$\delta J/J^{(0)} = \mathcal{I}_2\Lambda_+^2 + \mathcal{I}_3\Lambda_+^3 + \cdots, \tag{B8}$$

where

$$\mathcal{I}_3 = -\frac{\sqrt{2\omega_0}\tanh\sqrt{2\omega_0}}{(\Delta-\Delta_c)}(1-\mathcal{H}_2)$$

and

$$\delta l_s/l_s^{(0)} = \mathcal{L}_2\Lambda_+^2 + \mathcal{L}_3\Lambda_+^3 + \cdots, \tag{B9}$$

where

$$\mathcal{L}_3 = \frac{\Delta_c\sqrt{p}\tanh\sqrt{2\omega_0}\coth\sqrt{2p\omega_0}}{(\Delta-\Delta_c)}(1-\mathcal{H}_2) + \mathcal{L}_2.$$

Using (B8) and (B9), from the condition $h_s = J/l_s$ we derive

$$\delta h_s/\Delta_c = \Lambda_+ + \Lambda_+^2(1-\mathcal{H}_2) + \Lambda_+^3(1-\mathcal{H}_3) + \cdots, \tag{B10}$$

where

$$\mathcal{H}_3 = \mathcal{L}_3 - \mathcal{I}_3 + \mathcal{L}_2 - 2\mathcal{I}_2.$$

Clearly, the many-times repeated use of this procedure enables one to obtain exact expansions in powers of Λ_+,

$$\delta J/J^{(0)} = \sum_{n=2}^{\infty} \mathcal{I}_n \Lambda_+^n,$$

$$\delta l_s/l_s^{(0)} = \sum_{n=2}^{\infty} \mathcal{L}_n \Lambda_+^n,$$

$$\delta h_s/\Delta_c = \Lambda_+ + \sum_{n=2}^{\infty}(1-\mathcal{H}_n)\Lambda_+^n, \tag{B11}$$

where the coefficients $\mathcal{I}_n$, $\mathcal{L}_n$, and $\mathcal{H}_n$ are proportional to $(\Delta-\Delta_c)^{-1}$ and slowly grow with n. This suggests that, in the limit $h_0/\Delta_c \to \infty$, $\Delta/\Delta_c \to \infty$, Eqs. (10)–(13) give an exact description of the annihilation dynamics to the point of finite-time singularity $\Lambda_+ \to 1$ [according to (49) and (50), $1-\Lambda_+^f \propto p^{1/4}\sqrt{\Delta_c/\Delta} \to 0$ as $\Delta/\Delta_c \to \infty$]. So, for the critical point $\tau_\star$ one finds

$$\Lambda_+(\tau_\star) = \lambda e^{\omega_0\tau_\star} = 1. \tag{B12}$$

According to (17), in the limit $\Delta/\Delta_c \to \infty$ the λ value with the accuracy of the multiplier $1-\Delta_c/\Delta + O(\Delta_c/h_0)$ becomes a unique function of the reduced initial number of pairs $n_0 = N_0/\Delta$,

$$\lambda = \frac{\Delta_c}{2(1+n_0)}, \tag{B13}$$

whence it follows that $\tau_\star(\lambda)$ is a unique function of n_0. Introducing a relative time $\mathcal{T} = \tau - \tau_\star$, from Eqs. (B1), (B12), and (B13) we find that, in the limit $n_0/\Delta_c \to \infty$, h_s demonstrates a universal (n_0-independent) behavior

$$h_s = \frac{\Delta_c}{1 - e^{-\omega_0|\mathcal{T}|}}, \tag{B14}$$

whence in the vicinity of the critical point $\omega_0|\mathcal{T}| \ll 1$ we obtain the universal law of explosive growth

$$h_s = \frac{1}{\mu|\mathcal{T}|}, \quad |\mathcal{T}| \to 0. \tag{B15}$$

Due to the smallness of $\varepsilon/\Lambda_+ \to 0$, it is apparent that in the limit $\Delta/\Delta_c \to \infty$ at finite $\lambda \ll 1$ the dominant contribution to δJ and δl_s arises at the stage of explosive growth of h_s. It is therefore clear that, in the general case, Eqs. (10)–(13) are asymptotically exact to the point of finite-time singularity $\Lambda \to 1$, so that $\Lambda(\tau_\star) = 1$. Introducing $\mathcal{T} = \tau - \tau_\star$ from Eq. (13) we obtain

$$h_s = \frac{\Delta_c(1 + \xi_8 e^{-8\omega_0 \mathcal{T}})}{1 - \Lambda(\mathcal{T})}, \tag{B16}$$

where

$$\Lambda_-(\mathcal{T}) = \varrho_8 \xi_8 e^{-8\omega_0 \mathcal{T}} + \varrho_p \xi_p e^{-\chi\omega_0 \mathcal{T}},$$

$$\Lambda_+(\mathcal{T}) = \lambda \xi_\lambda e^{\omega_0 \mathcal{T}},$$

and the coefficients $\xi_8 = e^{-8\omega_0 \tau_\star}$, $\xi_p = e^{-\chi\omega_0 \tau_\star}$, $\xi_\lambda = e^{\omega_0 \tau_\star}$ determine the critical point $\tau_\star(\lambda)$ according to the condition

$$\Lambda(0) = \varrho_8 \xi_8 + \varrho_p \xi_p + \lambda \xi_\lambda = 1. \tag{B17}$$

By setting $\lambda \ll 1$ from Eqs. (B16) and (B17) one straightforwardly comes to Eqs. (25) and (28), which explicitly determine the laws of critical point shift and universalization of concentration explosion with growth of the initial number of pairs, n_0.

APPENDIX C: SYSTEMATIC ANALYSIS OF THE SINGULARITY FORMATION

According to Eqs. (38) and (39), in the vicinity of the critical point $\omega_0|\mathcal{T}| \ll 1$, an explosive growth of the surface concentration of slow particles,

$$h_s^{ex} \propto 1/\mu|\mathcal{T}|, \tag{C1}$$

results in an explosive growth of the antiflux J_{ex}, directed into the medium interior. This antiflux is calculated in the semi-infinite medium approximation

$$J_{ex} = -\int_{-\infty}^{\mathcal{T}} \frac{dh_s^{ex}}{d\theta} \frac{d\theta}{\sqrt{\pi(\mathcal{T} - \theta)}}. \tag{C2}$$

The legitimacy of using the semi-infinite medium approximation (C2) stems from the fact that, at the stage of explosion, which makes the dominant contribution to J_{ex}, the explosion rate $\Omega_{Hs} \propto |\mathcal{T}|^{-1}$ dictates a characteristic size of the layer proportional to $\sqrt{|\mathcal{T}|}$, into which particles have had time to diffuse [3]. So at $|\mathcal{T}| \ll 1$ the medium shows up effectively as semi-infinite.

Below we give a systematic calculation of J_{ex} following the exact Eq. (15). We show that, in the limit of small $|\mathcal{T}| \to 0$, the obtained result coincides with the result of the semi-infinite medium approximation with an accuracy to vanishingly small terms. Moreover, in the main body of the paper, the law of explosive growth of l_s^{ex} at $|\mathcal{T}|/p \ll 1$ is found using the corresponding semi-infinite medium approximation

$$J_{ex} = -\int_{-\infty}^{\mathcal{T}} \frac{dl_s^{ex}}{d\theta} \frac{d\theta}{\sqrt{p\pi(\mathcal{T} - \theta)}}. \tag{C3}$$

The requirement of equality (C2) and (C3) leads to the key condition of synchronous growth $\dot{l}_s^{ex} = \sqrt{p}\dot{h}_s^{ex}$, where, with allowance for the apparent requirement of smallness of l_s^{ex} far from the critical point, in the limit $|\mathcal{T}|/p \to 0$ for the universal explosion one obtains

$$l_s^{ex} = \sqrt{p} h_s^{ex} = \sqrt{p}/\mu|\mathcal{T}| + \cdots. \tag{C4}$$

According to (C4), the rate of synchronous explosion $\Omega_{Hs} \propto |\mathcal{T}|^{-1}$; therefore the characteristic size of the layer into which L particles have had time to diffuse is proportional to $\sqrt{|\mathcal{T}|/p}$. So, at $|\mathcal{T}|/p \ll 1$, the medium shows up effectively as semi-infinite for particles of both types, and the whole procedure is self-consistent. In what follows, we give a systematic calculation of l_s^{ex} following the exact Eq. (16). We rigorously show that, in the limit of small $|\mathcal{T}|/p \to 0$, the obtained result leads to (C4) with an accuracy to vanishingly small terms.

According to Eqs. (B11), in the limit $\lambda \to 0$, $\Delta/\Delta_c \to \infty$, the coefficients $\mathcal{H}_n \propto \Delta_c/(\Delta - \Delta_c) \to 0$; therefore to a vanishingly narrow vicinity of the critical point [in conformity with Eq. (50) to the point $|\mathcal{T}| \sim \mathcal{T}_f \propto \sqrt{\Delta_c/\Delta} \to 0$] the growth of h_s is described by Eqs. (B1) and (B3), so that at $\omega_0|\mathcal{T}| \ll 1$

$$h_s = \frac{(1 + \omega_0|\mathcal{T}|/2 + \cdots)}{\mu|\mathcal{T}|}. \tag{C5}$$

From Eq. (B3) one has

$$\delta\hat{h}_s(s) = \Delta_c \sum_{n=1}^{\infty} \frac{\lambda^n}{s - n\omega_0}. \tag{C6}$$

Substituting this result into Eq. (15), one finds

$$\delta\hat{J}(s) = -\Delta_c \sum_{n=1}^{\infty} \frac{\lambda^n}{s - n\omega_0} \sqrt{s} \tanh \sqrt{s}, \tag{C7}$$

whence it follows that

$$\delta J(\tau) = \mathcal{G}_+(\tau) + \mathcal{G}_-(\tau), \tag{C8}$$

where

$$\mathcal{G}_+(\tau) = -\Delta_c \sum_{n=1}^{\infty} \lambda^n \sqrt{n\omega_0} \tanh(\sqrt{n\omega_0}) e^{n\omega_0 \tau} \tag{C9}$$

and

$$\mathcal{G}_{-}(\tau) = -2\Delta_c e^{-\omega_0\tau}\sum_{n=1}^{\infty}\frac{\lambda^n}{(n+1)} - 18\Delta_c e^{-9\omega_0\tau}\sum_{n=1}^{\infty}\frac{\lambda^n}{(n+9)} + \cdots . \tag{C10}$$

From Eqs. (C10) and (25), we find that, at $\lambda \ll 1$ not too far from the critical point,

$$\mathcal{G}_{-}(|\mathcal{T}|) \propto -\Delta_c\lambda^2 e^{\omega_0|\mathcal{T}|}.$$

By adding to (C8) the contribution of the transient stage (A9),

$$\delta J_{tr}(|\mathcal{T}|) \propto \Delta_c\lambda\sqrt{\tau_{tr}}e^{\omega_0|\mathcal{T}|},$$

and the contribution of (A3) of the same order of magnitude, we conclude that at $\lambda \ll 1$ not too far from the critical point the dominant contribution to $\delta J(|\mathcal{T}|)$ is made by the term

$$\mathcal{G}_{+}(|\mathcal{T}|) = -\Delta_c\sum_{n=1}^{\infty}\sqrt{n\omega_0}\tanh(\sqrt{n\omega_0})e^{-n\omega_0|\mathcal{T}|}, \tag{C11}$$

which in the limit $\lambda \to 0$ becomes exact. In the vicinity of the critical point $\omega_0|\mathcal{T}| \ll 1$, the main contribution to $\mathcal{G}_{+}$ is made by the terms with $n \gg 1$; therefore, by setting $\tanh(\sqrt{n\omega_0}) \approx 1$ and coming from summation to integration, one obtains

$$\mathcal{G}_{+}(|\mathcal{T}|) = -(1/\mu)\int_{\omega_0}^{\infty}\sqrt{z}e^{-z|\mathcal{T}|}dz, \tag{C12}$$

whence it follows that

$$\mathcal{G}_{+}(|\mathcal{T}|) = -a_J/\mu|\mathcal{T}|^{3/2} + O(1)/\mu + \cdots , \tag{C13}$$

where $a_J=\sqrt{\pi}/2$. Thus, by neglecting the term $O(1)$ as compared with the singular term $\sim 1/|\mathcal{T}|^{3/2}$, we come to the result of the semi-infinite medium approximation (40),

$$J_{ex} = \mathcal{G}_{+}(|\mathcal{T}| \to 0) = -\frac{a_J}{\mu|\mathcal{T}|^{3/2}}. \tag{C14}$$

Let us now turn to the calculation of δl_s. Substituting (C6) into (16), we obtain

$$\delta\hat{l}_s(s) = \Delta_c\sum_{n=1}^{\infty}\frac{\lambda^n}{s-n\omega_0}\sqrt{p}\tanh\sqrt{s}\coth\sqrt{ps}, \tag{C15}$$

whence it follows that

$$\delta l_s(\tau) = \mathcal{Z}_{+}(\tau) + \mathcal{Z}_{-}(\tau), \tag{C16}$$

where

$$\mathcal{Z}_{+}(\tau) = \Delta_c\sum_{n=1}^{\infty}\lambda^n\sqrt{p}\tanh(\sqrt{n\omega_0})\coth(\sqrt{pn\omega_0})e^{n\omega_0\tau} \tag{C17}$$

and

$$\mathcal{Z}_{-}(\tau) = -2e^{-\omega_0\tau}\sum_{n=1}^{\infty}\frac{\lambda^n}{(n+1)} + 18\varrho_8 e^{-9\omega_0\tau}\sum_{n=1}^{\infty}\frac{\lambda^n}{(n+9)} + 2(\chi+1)\varrho_p e^{-(\chi+1)\omega_0\tau}\sum_{n=1}^{\infty}\frac{\lambda^n}{(n+\chi+1)} + \cdots . \tag{C18}$$

From Eqs. (C18) and (25), we find that at $\lambda \ll 1$ not too far from the critical point

$$\mathcal{Z}_{-}(|\mathcal{T}|) \propto -\lambda^2 e^{\omega_0|\mathcal{T}|}.$$

By adding to (C16) the contribution of the transient stage (A10),

$$\delta l_s^{tr}(|\mathcal{T}|) \propto \lambda\sqrt{\tau_{tr}}e^{\omega_0|\mathcal{T}|},$$

and the contribution of (A4) of the same order of magnitude, we conclude that at $\lambda \ll 1$ not too far from the critical point the dominant contribution to $\delta l_s(|\mathcal{T}|)$ is made by the term

$$\mathcal{Z}_{+}(|\mathcal{T}|) = \Delta_c\sum_{n=1}^{\infty}\sqrt{p}\tanh(\sqrt{n\omega_0})\coth(\sqrt{pn\omega_0})e^{-n\omega_0|\mathcal{T}|}, \tag{C19}$$

which in the limit $\lambda \to 0$ becomes exact. In the vicinity of the critical point $\omega_0|\mathcal{T}| \ll 1$, the main contribution to $\mathcal{Z}_{+}$ is made by the terms with $n \gg 1$; therefore, by setting $\tanh(\sqrt{n\omega_0}) \approx 1$ and coming from summation to integration, one obtains

$$\mathcal{Z}_{+}(|\mathcal{T}|) = (1/\mu)\int_{\omega_0}^{\infty}\sqrt{p}\coth(\sqrt{pz})e^{-z|\mathcal{T}|}dz, \tag{C20}$$

whence at $p \ll \omega_0|\mathcal{T}| \ll 1$ it follows that

$$\mathcal{Z}_{+}(|\mathcal{T}|) = \sqrt{\pi}/\mu\sqrt{|\mathcal{T}|} + O(1)p/\mu|\mathcal{T}|^{3/2} + \cdots \tag{C21}$$

and at $\omega_0|\mathcal{T}| \ll p$ one finds

$$\mathcal{Z}_{+}(|\mathcal{T}|) = \sqrt{p}/\mu|\mathcal{T}| + O(1)/\mu\sqrt{p} + \cdots . \tag{C22}$$

By neglecting in (C22) the term $\sim 1/\mu\sqrt{p}$ as compared with the singular term $\sqrt{p}/\mu|\mathcal{T}|$, we come to the result of the semi-infinite medium approximation

$$l_s^{ex} = \mathcal{Z}_{+}(|\mathcal{T}|/p \to 0) = \frac{\sqrt{p}}{\mu|\mathcal{T}|}. \tag{C23}$$

In this way one obtains the rigorous derivation of the condition of synchronous growth (C4). It is remarkable that at small $p \ll 1$ Eq. (C20) yields an exact analytical description of the crossover $|\mathcal{T}|^{-1/2} \to |\mathcal{T}|^{-1}$ from an intermediate regime (C21) to the synchronous growth regime (C23).

It remains for us to make some important concluding remarks. The central point of the scaling theory of catastrophe is the contention, following from (C2) and (C3), that at $\Omega_s p \gg 1$ the condition of synchronous growth

$$\dot{l}_s^{ex} = \sqrt{p}\dot{h}_s^{ex} \tag{C24}$$

has to be obeyed both before and after passage through the critical point. The analysis presented here, together with the condition (C24), gives a rigorous substantiation of the fact that both before and after passage through the critical point the stronger condition

$$l_s^{ex} = \sqrt{p} h_s^{ex} \tag{C25}$$

must be obeyed. Substituting (C25) into the equation

$$h_s = J/l_s = (J^{(0)} + J_{ex})/(l_s^{(0)} + l_s^{ex}), \tag{C26}$$

and, allowing for the facts that (a) according to (76) in the limit $\mathcal{K}\to\infty$ the flux remains frozen in the vicinity of the critical point, $J = J_\star$, and (b) in the limit $|\mathcal{T}|\to 0$ the ratio $h_s^{ex}/h_s \to 1$, one finds

$$h_s = J_\star/(l_s^{(0)} + \sqrt{p} h_s), \tag{C27}$$

whence the scaling law (63) immediately follows. When deriving Eq. (C4) we, for simplicity, considered the universal limit $\lambda\to 0$. It is, however, clear that Eqs. (C24) and (C25) have to remain valid at finite $\lambda \ll 1$ with the sole difference that according to (28) in the right part of (C4) there appears the multiplier $1+Q(n_0)$ which characterizes the evolution of the explosion with growing n_0.

APPENDIX D: CROSSOVER TO THE SCALING REGIME OF CATASTROPHE

From (73) and (74), it follows that in the limit of large $\mathcal{K}\to\infty$ the ratio

$$\frac{\Omega_{Ls}^M - \Omega_{Hs}^M}{\Omega_{Hs}^M} \sim \frac{[\tau_J^{-1}]_M}{\Omega_s^M} \propto \mu/\mathcal{K}^{1/4} \to 0.$$

We shall show below that Ω_{Hs}^M reaches the asymptotic limit (73) much more rapidly than Ω_{Ls}^M; therefore it is the point of maximum of Ω_{Hs} that defines the point of maximum of the explosion. With allowance for the fact that $\Omega_{Ls}/\Omega_{Hs}\to 1$ only asymptotically, at finite $\mathcal{K}$ instead of (55) one has to write

$$\Omega_{Hs}[\sqrt{p} h_s + (\Omega_{Ls}/\Omega_{Hs}) l_s] = \mathcal{C}_{(0)}, \tag{D1}$$

whence at the point of the explosion maximum $\dot{\Omega}_{Hs}=0$ we find

$$\frac{\sqrt{p} h_s^M}{l_s^M} = \left(\frac{\Omega_{Ls}^M}{\Omega_{Hs}^M}\right)^2 - \frac{[\dot{\tau}_J^{-1}]_M}{(\Omega_{Hs}^M)^2} + \frac{\dot{\mathcal{C}}_{(0)}}{l_s^M(\Omega_{Hs}^M)^2}, \tag{D2}$$

where for completeness the term with the derivative is kept

$$\dot{\mathcal{C}}_{(0)} = -\ddot{l}_s^{(0)} = -\omega_0 \mu J_\star,$$

and it is taken into account that from the condition $\dot{\Omega}_{Hs}=0$ it follows that $\dot{\Omega}_{Ls}=[\dot{\tau}_J^{-1}]$. Differentiating (68) and calculating the arising integral, we find the leading term in $[\dot{\tau}_J^{-1}]_M$ in the form

$$[\dot{\tau}_J^{-1}]_M = \gamma_M h_s^M (\Omega_s^M)^{5/2}/J_\star,$$

where $\gamma_M = 3\Gamma(3/4)^2/2\pi = (3/4)a_M \approx 0.717$. Substituting this result into Eq. (D2), using the equality $\Omega_{Ls}^M = \Omega_{Hs}^M + \omega_0 + [\tau_J^{-1}]_M$, and taking $\Omega_{Hs}^M \approx \Omega_s^M$, after separating out the leading terms we find

$$\sqrt{p} h_s^M / l_s^M = 1 + B_r \mu/\mathcal{K}^{1/4} + O_r(\mathcal{K}^{-1/2}) \tag{D3}$$

where

$$B_r = \frac{2c_M - \gamma_M}{\sqrt{2}} \approx 0.972.$$

At the point of the explosion maximum, allowing for the contribution of J_{ex}^M (66) instead of (58), one has to write

$$h_s^M l_s^M = J_M = J_\star(1 + J_{ex}^M/J_\star).$$

Substituting (66) here we have

$$h_s^M l_s^M = J_\star[1 - B_\star \mu/\mathcal{K}^{1/4} + O_\star(\mathcal{K}^{-1/2})], \tag{D4}$$

where

$$B_\star = a_M/\sqrt{2} \approx 0.676.$$

Equations (D3) and (D4) immediately give

$$h_s^M/h_s^M(a) = 1 + B_h \mu/\mathcal{K}^{1/4} + O_h(\mathcal{K}^{-1/2}), \tag{D5}$$

where

$$h_s^M(a) = p^{-1/4}\sqrt{J_\star}$$

and

$$B_h = \frac{(2c_M - a_M - \gamma_M)}{2\sqrt{2}} \approx 0.148,$$

and

$$l_s^M/l_s^M(a) = 1 - B_l \mu/\mathcal{K}^{1/4} + O_l(\mathcal{K}^{-1/2}), \tag{D6}$$

where

$$l_s^M(a) = p^{1/4}\sqrt{J_\star}$$

and

$$B_l = \frac{2c_M + a_M - \gamma_M}{2\sqrt{2}} \approx 0.824.$$

From (D5) and (D6) it follows that h_s^M always comes to its asymptotics $h_s^M(a)$ from above, whereas l_s^M always comes to its asymptotics $l_s^M(a)$ from below. Essentially, the coefficient B_h is much smaller than B_l and, therefore, with growing $\mathcal{K}$ the asymptotics $h_s^M(a)$ is reached much earlier than the asymptotics $l_s^M(a)$. Substituting then (D3) into (D1) and using (D5) and (D6), we obtain

$$\Omega_{Hs}^M/\Omega_s^M(a) = 1 - B_\Omega \mu/\mathcal{K}^{1/4} + O_\Omega(\mathcal{K}^{-1/2}), \tag{D7}$$

where

$$\Omega_s^M(a) = (\mu/2) p^{-1/4}\sqrt{J_\star}$$

and

$$B_\Omega = \frac{c_M - a_M}{2\sqrt{2}} \approx 0.0318.$$

From (D7) it follows that Ω_{Hs}^M always comes to its asymptotics $\Omega_s^M(a)$ from below. Remarkably, the coefficient B_Ω appears so small that already at $\mathcal{K}>10^2$ the contribution of the $\mathcal{K}^{-1/4}$ term becomes less than 0.01. Substituting now (D4), (D5), and (D7) into the expression

$$[\tau_J^{-1}]_M = c_M h_s^M (\Omega_{Hs}^M)^{3/2}/J_M$$

for the amplitude of the catastrophe at the point of explosion maximum we find

$$[\tau_J^{-1}]_M/[\tau_J^{-1}]_M(a) = 1 + B_J\mu/\mathcal{K}^{1/4} + O_J(\mathcal{K}^{-1/2}), \qquad \text{(D8)}$$

where

$$[\tau_J^{-1}]_M(a) = (0.369\,834\ldots)\mu^{3/2}p^{-5/8}J_\star^{1/4}$$

and

$$B_J = \frac{a_M + (1/8)(2c_M - a_M)}{\sqrt{2}} \approx 0.776.$$

From (D8) it follows that $[\tau_J^{-1}]_M$ always comes to its asymptotics $[\tau_J^{-1}]_M(a)$ from above and, due to the comparatively high B_J value, reaches its asymptotics much more slowly than Ω_{Hs}^M.

The expressions (D5)–(D8) completely define the main picture of the crossover to the scaling catastrophe and explosion regime in the limit of large $\mathcal{K}\to\infty$. To be complete, we shall calculate now the corrections $O_\Omega(\mathcal{K}^{-1/2})$ and $O_J(\mathcal{K}^{-1/2})$ which, as one can easily see, may be of two types $O_i^p(p/\sqrt{\mathcal{K}})$ and $O_i^\mu(\mu^2/\sqrt{\mathcal{K}})$. As $\mu\sim 1-p\to 0$ at $p\to 1$ the O_i^p corrections appear essential at p close to 1. The calculations give

$$O_\Omega^p = -(\pi^2/4)p/\sqrt{\mathcal{K}} \approx -2.467p/\sqrt{\mathcal{K}} \qquad \text{(D9)}$$

and

$$O_J^p = \frac{(4a_M/c_M - 3)\pi^2}{8}p/\sqrt{\mathcal{K}} \approx +0.809p/\sqrt{\mathcal{K}}. \qquad \text{(D10)}$$

Due to the anomalously low value of the coefficient B_Ω, we shall also give the term O_Ω^μ. The calculations yield $O_\Omega^\mu = [B_\Omega(3B_\Omega/2 - B_h - B_\star/8) - B_\star^2/128]\mu^2/\sqrt{\mathcal{K}}$, whence after substituting the coefficients we find

$$O_\Omega^\mu \approx -0.009\,46\mu^2/\sqrt{\mathcal{K}}. \qquad \text{(D11)}$$

[1] E. Kotomin and V. Kuzovkov, *Modern Aspects of Diffusion Controlled Reactions: Cooperative Phenomena in Bimolecular Processes* (Elsevier, Amsterdam, 1996); D. C. Mattis and M. L. Glasser, Rev. Mod. Phys. **70**, 979 (1998); U. C. Tauber, M. Howard, and B. P. Vollmayr-Lee, J. Phys. A **38**, R79 (2005), and references therein.
[2] B. M. Shipilevsky, Phys. Rev. Lett. **73**, 201 (1994).
[3] B. M. Shipilevsky, Phys. Rev. Lett. **82**, 4348 (1999).
[4] B. O'Shaughnessy and D. Vavylonis, Phys. Rev. Lett. **84**, 3193 (2000).
[5] B. M. Shipilevsky, J. Phys. A **30**, L471 (1997).
[6] Taking account of the next-to-leading terms, one has $\Lambda_- = \varrho_8 e^{-8\omega_0\tau}[1+O(e^{-16\omega_0\tau})] + \varrho_p e^{-\chi\omega_0\tau}[1+O(e^{-(3\pi^2/p)\tau})]$, where it follows that at $\tau>\max[1/4\pi^2, p/3\pi^2]$ these terms may be neglected.
[7] L. D. Landau and E. M. Lifshitz, *Fluid Mechanics* (Pergamon Press, Oxford, 1987).
[8] J. Crank, *The Mathematics of Diffusion* (Clarendon Press, Oxford, 1975).
[9] G. D. Smith, *Numerical Solution of Partial Differential Equations with Exercises and Worked Solutions* (Oxford University Press, London, 1965).
[10] Acting in the spirit of Appendix D, it is easy to check that at large $\mathcal{K}$ and small $\delta_{\Omega,J}(n_0)$ the ratio $[\tau_J^{-1}]_M/\Omega_s^M \propto \epsilon_J(1+\delta_J-\delta_\Omega)$, so not too close to the critical points p_Ω^* and p_J^* at large $\Delta/\Delta_c\to\infty$ the relative deviations $|(\delta_i-\delta_i^\infty)/\delta_i^\infty| \propto \epsilon_i \to 0$ $(i=\Omega,J)$ must become vanishingly small.
[11] L. Kadanoff, Phys. Today **50**(2), 11 (1997).
[12] P. Bizon and Z. Tabor, Phys. Rev. D **64**, 121701(R) (2001).
[13] M. W. Choptuik, Phys. Rev. Lett. **70**, 9 (1993).
[14] P. H. Chavanis, C. Rosier, and C. Sire, Phys. Rev. E **66**, 036105 (2002).
[15] R. B. Pelz and Y. Gulak, Phys. Rev. Lett. **79**, 4998 (1997).
[16] B. W. Zeff, B. Kleber, J. Fineberg, and D. P. Lathrop, Nature (London) **403**, 401 (2000).
[17] M. Rascle and C. Ziti, J. Math. Biol. **33**, 388 (1995).
[18] D. Sornette and A. Helmstetter, Phys. Rev. Lett. **89**, 158501 (2002).
[19] B. O'Shaughnessy and D. Vavylonis, Eur. Phys. J. E **1**, 159 (2000).
[20] B. Chopard, M. Droz, J. Magnin, and Z. Racz, Phys. Rev. E **56**, 5343 (1997).
[21] A. A. Ovchinnikov and S. F. Burlatsky, JETP Lett. **43**, 638 (1986).
[22] L. W. Anacker and R. Kopelman, Phys. Rev. Lett. **58**, 289 (1987).
[23] K. Lindenberg, B. J. West, and R. Kopelman, Phys. Rev. Lett. **60**, 1777 (1988).
[24] D. ben-Avraham and C. R. Doering, Phys. Rev. A **37**, 5007 (1988).
[25] E. Clement, L. M. Sander, and R. Kopelman, Phys. Rev. A **39**, 6455 (1989); **39**, 6466 (1989).
[26] P. L. Krapivsky, Phys. Rev. A **45**, 1067 (1992).
[27] J. W. Evans and T. R. Ray, Phys. Rev. E **47**, 1018 (1993).
[28] L. Frachebourg, P. L. Krapivsky, and S. Redner, Phys. Rev. Lett. **75**, 2891 (1995).
[29] P. Argyrakis, S. F. Burlatsky, E. Clement, and G. Oshanin, Phys. Rev. E **63**, 021110 (2001).
[30] R. Gomer, *Chemisorption on Metals*, Solid State Physics Vol. 30 (Academic Press, New York, 1975).
[31] E. Fromm and G. Gebhardt, *Gase and Kohlenstoff in Metallen* (Springer, Berlin, 1976).
[32] H. J. Grabke and G. Horz, Annu. Rev. Mater. Sci. **7**, 155 (1977).
[33] G. Horz, in *Processing and Applications of High-Purity Refractory Metals and Alloys*, edited by P. Kumar, H. A. Jehn, and M. Uz (Minerals, Metals and Material Society, Warrendale, 1998).
[34] B. M. Shipilevsky and V. G. Glebovsky, Surf. Sci. **216**, 509 (1989).