Exact correlations in the one-dimensional coagulation-diffusion process by the empty-interval method

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Article with J-Y. Fortin, D. Del Biondo, M. Henkel, J. Richert : in preparation

CompPhys09, Leipzig, 2009

Motivation, previous studies

- One-dimensional diffusion-limited coagulation process
  1. Particle concentration : $c(t) \sim t^{-1/2}$
     $\longrightarrow$ mean field description : $c(t) \sim t^{-1}$
     $\longrightarrow$ fluctuations

     D. Toussaint and F. Wilczek, 1983

     D. ben Avraham, M. Burschka and C.R. Doering, 1990
  2. Correlation function : $C(r,t) \sim t^{-1} f(r^2/t)$

     D. ben Avraham, 1998

- Theoretical prediction confirmed by experiments
  1. kinetics of excitons on long polymer chains

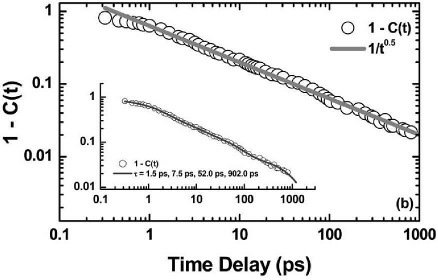
     R. Kroon, H. Fleurent and R. Sprik, 1993

     J. Prasad and R. Kopelman, 1989
  2. photoluminescence saturation in carbon nanotubes

     A. Srivastava and J. Kuno, 2009
  3. relaxation of photoexcitations in suspensions of carbon nanotubes

     R.M.Russo *et al.*, 2006

Motivation, previous studies



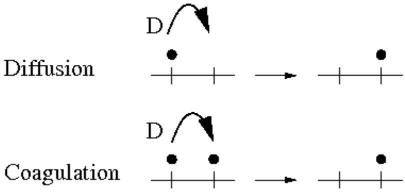
I.C. : high density of excited states
$1 - C(t)$: remaining excited population
behaviour : $t^{-1/2}$

R.M. Russo *et al.*, 2006

- Influence of initial conditions
- Perspective : Study of the ageing phenomena
  $\longrightarrow$ requires the knowledge of the one-time quantities

Model

One dimensional lattice of spacing a



One-interval probability

$E_n(t)$: time-dependent probability of having an interval of n consecutive empty sites at time t
$\longrightarrow$ give access to particle concentration

D. ben Avraham *et al.* 1990

Two-interval probability

$E_{n_1,n_2,d}(t)$: time-dependent probability of having two intervals of n_1 and n_2 consecutive empty sites at distance d at time t
$\longrightarrow$ give access to correlation function

I. Peschel *et al.* 1994

I.1 Differential equation : closed system

For $n > 1$

$$\partial_t E_n(t) = (2D/a^2)\left(E_{n-1} - 2E_n + E_{n+1}\right).$$

For $n = 1$, the equation is

$$\partial_t E_1(t) = (2D/a^2)\Big[1 - 2E_1(t) + E_2(t)\Big]$$

This gives the constraint : $E_0(t) = 1$

Equation of motion in the continuum limit

$$\partial_t E(x,t) = 2D\partial_{xx} E(x,t), \quad \text{and} \quad E(0,t) = 1.$$

$$E(x,t) = \int_{-\infty}^{\infty} \frac{dx'}{\sqrt{\pi}\ell_0} \exp\left[-\frac{1}{{\ell_0}^2}(x-x')^2\right] E(x',0).$$

where ℓ_0 is the scaling length

$$\ell_0 := \sqrt{8Dt}$$

I.2 General solution

We have to take into account the constraint : $E_0(t) = 1$.

Idea

assume that the differential equation is valid for $n \leq 0$

For $n = 0$

$$\partial_t E_0(t) = (2D/a^2)\,(E_{-1} - 2E_0 + E_1) = 0$$

which implies

$$E_{-1}(t) = 2E_0(t) - E_1(t) = 2 - E_1(t)$$

Redefine the meaning of $E(x, 0)$ for negative x such that

$$\boxed{E_{-n}(t) = 2 - E_n(t)} \qquad \text{and} \qquad E(-x,t) = 2 - E(x,t)$$

$$E(x,t) = \mathrm{erfc}(x/\ell_0) + \int_0^{+\infty} \frac{dx'}{\sqrt{\pi}\ell_0} E(x',0)\left[e^{-\frac{1}{\ell_0{}^2}(x-x')^2} - e^{-\frac{1}{\ell_0{}^2}(x+x')^2}\right].$$

I.3 General expression for the particle concentration

$$c(t) = -\,\partial_x E(x,t)|_{x=0}$$

$$c(t) = \frac{2}{\sqrt{\pi}\ell_0}\left(1 - \int_0^\infty dx E(x\ell_0, 0) 2xe^{-x^2}\right)$$

$E(x,0)$ is related to $P(x',t) = \mathrm{Pr}(\bullet \boxed{\ x'\ })$

$$E(x\ell_0, 0) = \int_{x\ell_0}^\infty P(x',t)dx'$$

such that when ℓ_0 is large, $E(x\ell_0) \ll 1$ and we obtain

$$c(t) = \frac{2}{\sqrt{\pi}\ell_0} + o(1/\ell_0) \sim t^{-1/2}$$

independent of initial condition

I.4 Particle concentration for a special initial condition

p probability of having a particle on a site

discrete case

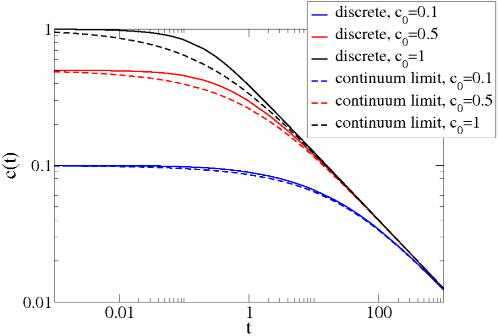
$$E_n(0) = (1-p)^n$$

$$c(t) = e^{-4Dt}\Big(I_0(4Dt) + I_1(4Dt) - \sum_{m=1}^{\infty}(1-p)^m \frac{2m}{4Dt} I_m(4Dt)\Big)$$

continuum limit

$$E(x,0) = \exp(-c_0 x)$$

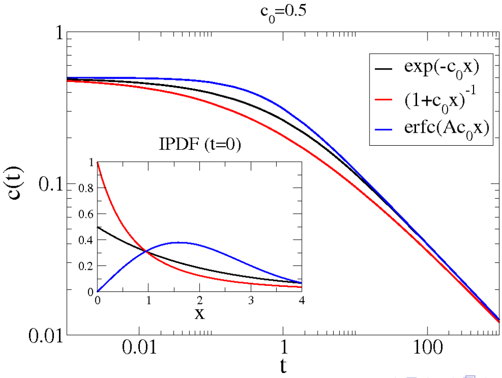
$$c(t) = c_0 e^{\frac{c_0^2 \ell_0^2}{4}} \mathrm{erfc}(\frac{c_0 \ell_0}{2})$$



$p(x,t)$: interparticle distribution functions (IPDF) (probability that the nearest particle to a given particle is at distance x)

$$p(x,t)c(t) = \partial^2_{xx} E(x,t)$$

$$E_0(x) = \left(\frac{1}{1+c_0 x}\right) \qquad E_0(x) = \mathrm{erfc}(\sqrt{\pi} c_0 x/2)$$



II. Correlation funtion

Connected two-point correlation function defined in the discrete case : probability to have two particles separated by d

$$C_2(d) = \mathrm{Pr}(\bullet\ d\ \bullet) - \mathrm{Pr}(\bullet)\mathrm{Pr}(\bullet)$$

Using $E_{n_1,n_2,d}(t) = \mathrm{Pr}(\boxed{\ n_1\ }\ d\ \boxed{\ n_2\ })$
we obtain

$$C_2(d) = 1 - E_{0,1}(d) - E_{1,0}(d) + E_{1,1}(d) - (1 - E_{1,0}(d))(1 - E_{0,1}(d))$$

Correlator in the continuum limit

$$C_2(z) = \partial^2_{xy} E(x,y,z)\big|_{x=0,y=0} - \partial_x E(x)\big|_{x=0}\, \partial_y E(y)\big|_{y=0}$$

II.1 Two-intervals probability

Consider $E_{n_1,n_2,d}(t) = \mathrm{Pr}(\boxed{\ n_1\ }\ d\ \boxed{\ n_2\ })$

Symmetries and auto-consistency relation

$$E_{n_1,n_2}(d,t) = E_{n_2,n_1}(d,t)$$
$$E_{n_1,0}(d,t) = E_{n_1}(t) \quad \text{and} \quad E_{0,n_2}(d,t) = E_{n_2}(t)$$
$$E_{n_1,n_2}(0,t) = E_{n_1+n_2}(t).$$

In the continuum limit, setting $x = n_1\mathrm{a}$, $y = n_2\mathrm{a}$ and $z = d\mathrm{a}$

$$\partial_t E(x,y,z,t) = 2D\left[\partial_x^2 + \partial_y^2 + \partial_z^2 - \left(\partial_x\partial_z + \partial_y\partial_z\right)\right]E(x,y,z).$$

$\longrightarrow$ subject to boundary and consistency conditions

II.4 General solution and compatibility conditions

General solution without any constraint

$$E(x,y,z,t) = \int_{-\infty}^{\infty} \frac{dx'dy'dz'}{\pi\sqrt{\pi/2}{\ell_0}^3} \mathcal{W}(x-x', y-y', z-z') E_0(x', y', z')$$

where the Gaussian kernel $\mathcal{W}(u,v,w)$ is given by

$$\mathcal{W}(u,v,w) = \exp \frac{1}{{\ell_0}^2}\Big[-(u+v+w)^2 - w^2 - \frac{1}{2}(u-v)^2\Big]$$

Correspondence between negative and positive variables in the continuum limit

$$\begin{aligned}
E(-x) &= 2 - E(x), \\
E(-x,y,z) &= 2E(y) - \mathbf{E}(\mathbf{x},\mathbf{y},\mathbf{z}-\mathbf{x}), \\
E(x,-y,z) &= 2E(x) - \mathbf{E}(\mathbf{x},\mathbf{y},\mathbf{z}-\mathbf{y}), \\
E(-x,-y,z) &= 4 - 2E(x) - 2E(y) + \mathbf{E}(\mathbf{x},\mathbf{y},\mathbf{z}-\mathbf{x}-\mathbf{y}), \\
E(x,y,-z) &= 2E(x+y-z) - \mathbf{E}(\mathbf{x}-\mathbf{z},\mathbf{y}-\mathbf{z},\mathbf{z}).
\end{aligned}$$

Using the compatibility conditions, we can write $E(x, y, z, t)$ as

$$E(x, y, z, t) = E^{(0)}(x, y, z, t) + E^{(1)}(x, y, z, t) + E^{(2)}(x, y, z, t)$$

where

- $E^{(0)}(x, y, z, t)$ is independent of the initial conditions
- $E^{(1)}(x, y, z, t)$ depends on the initial one-interval probability $E_0(x)$
- $E^{(2)}(x, y, z, t)$ depends on the initial **two-intervals** probability $E_0(x, y, z)$

II.6 Contribution independent of the initial conditions

$E^{(0)}(x,y,z,t)$

$$E^{(0)}(x,y,z,t) = \mathrm{erfc}(\frac{x}{\ell_0})\mathrm{erfc}(\frac{y}{\ell_0}) + \mathrm{erfc}(\frac{z}{\ell_0})\mathrm{erfc}(\frac{x+y+z}{\ell_0}) - \mathrm{erfc}(\frac{x+z}{\ell_0})\mathrm{erfc}(\frac{y+z}{\ell_0}).$$

- Full solution for initially completely filled system
- Shows the required symmetries
- For large z, it decouples : $E^{(0)}(x,y,z,t) \simeq E(x,t)E(y,t)$

$$E^{(1)}(x,y,z,t) = \operatorname{erfc}(\frac{x}{\ell_0})I(y) + I(x)\operatorname{erfc}(\frac{y}{\ell_0})$$
$$-\operatorname{erfc}(\frac{x+z}{\ell_0})I(y+z) - I(x+z)\operatorname{erfc}(\frac{y+z}{\ell_0})$$
$$+\operatorname{erfc}(\frac{z}{\ell_0})I(x+y+z) + I(z)\operatorname{erfc}(\frac{x+y+z}{\ell_0})$$

where

$$I(x) = \int_0^\infty \frac{dx'}{\ell_0\sqrt{\pi}} E_0(x')\left[e^{-(x'-x)^2/{\ell_0}^2} - e^{-(x'+x)^2/{\ell_0}^2}\right]$$

$$E^{(2)}(x,y,z,t) = \int_0^\infty \frac{\sqrt{2}dx'dy'dz'}{{\ell_0}^3\sqrt{\pi}^3} E_0(x',y',z')$$
$$\times \mathcal{W}(x-x',y-y',z-z')K_2(x',y',z';x,y,z)$$

Correction to the leading behaviour in the asymptotic regime

II.8 One-time correlation function

Correlation function

$$C_2(z,t) = \partial^2_{xy} E(x,y,z,t)\big|_{x=0,y=0} - \partial_x E(x,t)\big|_{x=0}\, \partial_y E(y,t)\big|_{y=0}$$

In the case of an initially completely filled system, $E(x,0)=0$ and $E(x,y,z,0)=0$, we recover the expression

$$C_2(z,t) = \frac{4}{\pi {\ell_0}^2}\left[-e^{-2z^2/{\ell_0}^2} + \sqrt{\pi}\frac{z}{\ell_0}\mathrm{erfc}\left(\frac{z}{\ell_0}\right) e^{-z^2/{\ell_0}^2}\right]$$

D. ben Avraham, 1998

exact in asymptotic regime for all initial conditions.

Connected correlator

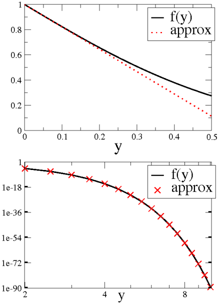
$$\begin{aligned} C_2(z,t) &= \left(\frac{2}{\sqrt{\pi}\ell_0}\right)^2 f(z/\ell_0) \\ \text{with} \quad f(y) &= -e^{-2y^2} + \sqrt{\pi} y e^{-y^2}\mathrm{erfc}(y) \end{aligned}$$

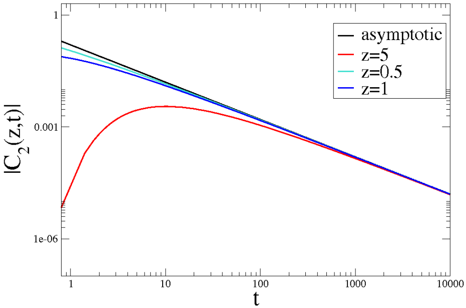
II.9 One-time correlation function

$$C_2(z,t) \;=\; \left(\frac{2}{\sqrt{\pi}\ell_0}\right)^2 f(z/\ell_0)$$

with

$$f(y) \simeq \begin{cases} -1+\sqrt{\pi}y & \text{for } y \ll 1 \\ -\frac{1}{2y^2}e^{-2y^2} & \text{for } y \to \infty \end{cases}$$





Algebraic behaviour when t large $|C_2(t)| \sim t^{-1}$.

Conclusion

- Solvable model through closed equations of motion for empty-interval probabilities
- We have extended the technique to find correlations directly, for arbitrary initial conditions
- Exact calculation of
    1. $E(x,t) \longrightarrow$ exact expression of the particle concentration
       $\longrightarrow$ new treatment of the boundary condition
    2. $E(x,y,z;t) \longrightarrow$ exact correlation function
- confirm scaling description from explicit expressions

Perspectives

- include other reactions : $A \longrightarrow A+A$, $\emptyset \longrightarrow A$, $A\emptyset A \longrightarrow AAA$
- Link between physical quantities and initial two-interval probability
- Two-time quantities and ageing