Vol. 36 (2005) ACTA PHYSICA POLONICA B No 5

ON MODELING OF GROWTH PROCESSES DRIVEN BY VELOCITY FLUCTUATIONS*

M. Niemiec, W. Olchawa

Institute of Physics, Opole University
Oleska 48, 45-052 Opole, Poland

and J. Łuczka

Institute of Physics, University of Silesia
Uniwersytecka 4, 40-007 Katowice, Poland

(Received January 5, 2005)

This work is dedicated to Prof. Andrzej Fuliński on the occasion of his 70th birthday

In the classical theory of diffusion limited growth, it is assumed that the concentration field of solution is described by the standard diffusion equation. It means that particles of the solution undergo a random walk described by the Wiener process. In turn, it means that the velocity of particles is a stochastic process being Gaussian white noise. In consequence, the velocity–velocity correlation function is the Dirac δ-function and velocity correlation time is *zero*. In many cases such modeling is insufficient and one should consider models in which velocity is correlated in space and/or time. The question is whether correlations of velocity can change the kinetics of growth, in particular, whether the long-time asymptotics of the growth kinetics displays the power-law time dependence with the classical exponent 1/2. How to model such processes is a subject of this paper.

PACS numbers: 81.10.Aj, 05.40.–a

1. Introduction

Growth processes such as aggregation processes, growth of clusters and crystals, grain growth and the like have been studied both experimentally and theoretically [1–3]. In these irreversible processes objects are built up from elementary ingredients like particles, molecules or micro-aggregates. Growth from the vapor phase, from saturated solution or from supercooled

---

[*] Presented at the XVII Marian Smoluchowski Symposium on Statistical Physics, Zakopane, Poland, September 4–9, 2004.

melt is the most widespread method of crystal growing. Commonly used solvents are water, multicomponent aqueous or non-aqueous solutions, melts of some chemical components, colloidal suspensions and complex electrolytes. Among many aspects, the growth kinetics have been intensively studied including evolution of the interface dynamics, asymptotics of the growth rate of the interface, the time-dependence of the crystal radius, existence of regimes with power-law growth behavior and determination of various growth exponents.

In any modeling, the complexity of the process is usually reduced by supposing elementary processes to be dominant mechanisms responsible for the growth kinetics. The classical theory of crystal growth assumes the crystal formation by addition of individual particles or molecules from the saturated solution (the concentration field around the growing crystal, which is usually metastable fluid). Mass transfer from surroundings to the crystal is achieved by a diffusive flow, *i.e.* by the particle self-diffusivity or by the particle gradient diffusivity [4] and is described by a standard diffusion equation in the form

$$\frac{\partial c(\boldsymbol{r})}{\partial t} = D\Delta c(\boldsymbol{r})\,, \tag{1}$$

where $c(\boldsymbol{r})$ is the concentration of solution around the crystal at the point determined by the vector $\boldsymbol{r}$ and D is the diffusion coefficient of the solution particles.

The growth kinetics is determined from the conservation law of the crystallizing substance at the moving growth front. It has the form [1]

$$\frac{D\partial c(\boldsymbol{r})}{\partial n_0} = [C - c(\boldsymbol{r})]\; V(\boldsymbol{n}_0, \Delta\mu)\,, \qquad \text{for} \qquad \boldsymbol{r} = \boldsymbol{R}\,, \tag{2}$$

where the derivative is taken in the direction of the outer normal $\boldsymbol{n}_0$ to the surface at the given point $\boldsymbol{r} = \boldsymbol{R}$, C is the density of the growing crystal on the interface $\boldsymbol{R}$. The interface velocity or growth rate $V(\boldsymbol{n}, \Delta\mu)$ depends on its crystallographic orientation $\boldsymbol{n}$ and the deviation $\Delta\mu$ from equilibrium at the point $\boldsymbol{r} = \boldsymbol{R}$. Hence, if we know a solution of the diffusion equation (1) then from (2) we can determine the growth rate $V(\boldsymbol{n}, \Delta\mu)$.

One has to impose boundary conditions on Eq. (1): *(i)* at the interface between the crystal and the solution and *(ii)* at infinity. The concentration $c(\boldsymbol{R})$ of the saturated solution over the interface $\boldsymbol{R}$ is determined from thermodynamical conditions and geometry of the interface. The universally accepted expression for $c(\boldsymbol{R})$ has been derived under an assumption of local thermodynamical equilibrium near the interface. It depends on the surface curvature due to the Gibbs–Thomson effect and is given by the formula [1,5]

$$c(\boldsymbol{R}) = c_0[1 + \mathit{\Gamma}(\boldsymbol{R})K(\boldsymbol{R})]\,, \tag{3}$$

where $\mathit{\Gamma}(\boldsymbol{R})$ is the capillary coefficient, $K(\boldsymbol{R})$ is twice the mean curvature of the surface $\boldsymbol{R}$ at a given point and c_0 is the concentration of the saturated solution over the flat surface (the equilibrium concentration for the planar interface). The second boundary condition is imposed on the concentration far from the crystal, *i.e.*,

$$c(\boldsymbol{r}) \to c_\infty = \text{const.} \quad \text{when} \qquad |\boldsymbol{r}| \to \infty \,. \tag{4}$$

The problem is non-trivial because in its most general formulation it requires to solve Eq. (1) with the moving boundary condition (because the crystal grows and the interface changes with increase of time). It can be simplified for the spherical symmetry and in the so-called quasi steady state approximation [1]. The most important conclusion is the time-dependence of the growth rate,

$$V = V(t) \sim t^{-1/2} \tag{5}$$

and in consequence the radius R of the crystal grows according to the power law as

$$R = R(t) \sim t^{1/2} \tag{6}$$

with the growth exponent 1/2. It is universal behavior, valid for a wide class of growth processes. However, the deviation from this diffusion-like kinetics has been observed.

In models based on the above approach, the crystal growth is a process of attachment of particles from isotropic medium which usually is liquid or gas (growth from saturated solution or any metastable fluid). After the nucleation stage, a small crystal is formed. Particles of the surroundings perform a random walk and when they arrive in contact with the crystal, they stick permanently. Then another particles arrive and stick, and so on. Because the concentration field is described by the standard diffusion equation (1), it means that particles of the solution undergo a random walk which is the Wiener process in the position space. In turn, in the velocity space it is Gaussian white noise. It means that the velocity–velocity correlation function of the fluid particles is the Dirac δ-function and both the correlation time and the correlation length is *zero*. However, this idealization is never exactly realized and in many situations such modeling is insufficient. Therefore, a modified theory is desirable which includes models with time and/or space correlations of the particle velocity. The fundamental problem appears: do correlations change the kinetics of the growth process? In other words, does the long-time asymptotics of the growth kinetics display the power-law time dependence? If yes, then the next question appears: what is the value of the growth exponent? Is it the classical exponent 1/2 resembling the standard diffusion limited growth or not?

A mathematical model could be formulated by a suitable modification of the classical theory. Firstly, the diffusion equation (1) could be replaced by a master equation which describes a random walk of the solution particles with a specific form of the velocity correlation function. Secondly, in Eq. (2), the left-hand side should be replaced by a more general expression, namely

$$-\boldsymbol{J} \cdot \boldsymbol{n}_0 = [(C - c(\boldsymbol{r})]V(\boldsymbol{n}_0, \Delta\mu)\,, \qquad \text{for} \qquad \boldsymbol{r} = \boldsymbol{R}\,, \tag{7}$$

where $\boldsymbol{J} = J[c(\boldsymbol{r})]$ is the flux of particles whose dependence on the fluid concentration $c(\boldsymbol{r})$ follows from the master equation. For some models of the velocity correlations (*e.g.* the temporal exponential correlations), we can write a closed set of evolution equations. Then the typical problem is how to solve this set of equations but it is a secondary problem. For some models of velocity correlations, the problem is indeed complicated because we do not know master equations for processes of prescribed velocity correlations. So, we even cannot present a closed set of equations. Therefore, a radically new formulation is desired because in the standard description, which couples the diffusion equations in the bulk phases with the interfacial boundary conditions, it is rather difficult to include more general diffusion processes, which are described by much more complicated evolution equations (*e.g.* integro-differential equations) than the standard parabolic diffusion equation (1) with a constant (state-independent) diffusion coefficient. One of such a novel formulation of the problem has been proposed in [6], where the case of only temporal and not spatial correlations has been worked out. Here we want to present the modeling which can include the spatial velocity correlations.

2. Model

For growing aggregates of an ideal or perturbed spherical symmetry, it is convenient to use the parametrization of the spherical coordinate system. The surface of the aggregate is described by the vector equation

$$\tilde{\boldsymbol{r}} = \tilde{r}\cos\phi\sin\vartheta\;\boldsymbol{e}_x + \tilde{r}\sin\phi\sin\vartheta\;\boldsymbol{e}_y + \tilde{r}\cos\vartheta\;\boldsymbol{e}_z\,, \tag{8}$$

where the function

$$\tilde{r} \equiv \tilde{r}(\vartheta, \phi, t) \tag{9}$$

specifies the aggregate surface. In the spherical coordinate system (r, ϑ, ϕ), the surface is determined if we know the function

$$r = \tilde{r}(\vartheta, \phi, t)\,. \tag{10}$$

An equation describing evolution of a growing object can be derived exploiting the mass conservation law. Its form reads [6]

$$\tilde{r}^2 \sin\vartheta\,[C(\tilde{r},\vartheta,\phi) - c(\tilde{r},\vartheta,\phi)]\frac{d\tilde{r}}{dt} = \boldsymbol{J}[c(\tilde{r},\vartheta,\phi)]\cdot\boldsymbol{n}\,, \tag{11}$$

where $C(r,\vartheta,\phi)$ is the density of the growing crystal at the point $\boldsymbol{r} = (r,\vartheta,\phi)$. We can assume that the aggregate is homogeneous and the density is uniform, *i.e.* $C(r,\vartheta,\phi) = \mathcal{C} = \text{const}$. The concentration $c(\boldsymbol{r}) \equiv c(r,\vartheta,\phi)$ of the saturated solution over the interface $(\tilde{r},\vartheta,\phi)$ is determined from thermodynamical conditions and geometry of the surface. It depends on the surface curvature due to the Gibbs–Thomson effect and is given by the formula [1,5]

$$c(\tilde{r},\vartheta,\phi) = c_0[1 + \mathit{\Gamma}(\vartheta,\phi)\mathcal{K}(\tilde{r},\vartheta,\phi)]\,, \tag{12}$$

where $\mathit{\Gamma}(\vartheta,\phi)$ is the capillary coefficient, $\mathcal{K}(\tilde{r},\vartheta,\phi)$ is twice the mean curvature of the surface at the point $(\tilde{r},\vartheta,\phi)$ and c_0 is the concentration of the saturated solution over the flat surface (the equilibrium concentration for the planar interface), *i.e.* when the curvature of the interface is zero.

The particle flux $\boldsymbol{J}$ depends on the concentration $c(\boldsymbol{r}) = c(r,\vartheta,\phi)$ of the solution at the position (r,ϑ,ϕ). Our suggestion is to consider not a diffusional flux of particle but rather convective flow for which [7]

$$\boldsymbol{J}[c(\boldsymbol{r})] = c(\boldsymbol{r})\boldsymbol{v}(\boldsymbol{r},t)\,, \tag{13}$$

where $\boldsymbol{v}(\boldsymbol{r},t)$ is a velocity of particles of the solution at the position $\boldsymbol{r}$ at the moment t. Finally, the vector

$$\boldsymbol{n} = \frac{\partial\boldsymbol{r}}{\partial\phi}\times\frac{\partial\boldsymbol{r}}{\partial\vartheta} \tag{14}$$

is the inward normal to the interface (the symbol "×" denotes the vector product). Generally, it is not a unit vector.

3. Spherical growth

The evolution equation (11) is a complicated equation which in a general case is difficult to handle. Therefore, we simplify our model assuming that the aggregate grows as a spherical object. Consequently, the interface is a sphere for which $\tilde{r}(\vartheta,\phi,t) = R(t)$, where $R(t)$ is a radius of the sphere at the instant t. The Gibbs–Thomson relation (12) takes the form

$$c(\tilde{r},\vartheta,\phi) = c_0\left[1 + \mathit{\Gamma}\mathcal{K}(R)\right]\,, \tag{15}$$

where $\varGamma$ is the capillary constant and $\mathcal{K}(R) = 2/R$ is twice the mean curvature of the aggregate. The reduced form of the evolution equation (11) now reads

$$\frac{dR}{dt} = A(R)\, v(R,t)\,, \tag{16}$$

where

$$\begin{aligned} A(R) &= \alpha \frac{R + 2\varGamma}{R - R^*}\,, \qquad R > R^*\,, \\ \alpha &= \frac{c_0}{\mathcal{C} - c_0}\,, \\ R^* &= 2\varGamma\alpha\,, \end{aligned} \tag{17}$$

and we assume that the velocity field of the solution particles

$$v(r,t) = \boldsymbol{v}(\boldsymbol{r},t) \cdot \boldsymbol{n}\,, \qquad \boldsymbol{n} = -\boldsymbol{e}_r\,, \qquad r \geq R\,, \tag{18}$$

and $\boldsymbol{e}_r$ stands for a radial unit vector. Let us remember that $v(R,t) = \lim_{r\to R} v(r,t)$.

If the velocity field $v(r,t) =$const. then the growth process displays the so called kinetic-regime growth, *i.e.* for long times $R(t) \propto t$. Other examples of the deterministic convection fields are studied in [8]. The case when $v(r,t) = v(t)$ is a stochastic Gaussian white noise is analyzed in [9]. The temporally correlated stochastic processes $v(r,t) = v(t)$ have been included in [6]. Here, we propose the subsequent extension assuming that the velocity field is a purely random (zero-mean) Gaussian field,

$$\langle v(r,t) \rangle = 0\,. \tag{19}$$

Its correlation function

$$\big\langle v(r,t) v(r',t') \big\rangle = \mathcal{F}(r,t;r',t')\,. \tag{20}$$

It implies that the deterministic part of the velocity field is zero and $v(r,t)$ describes no-bias velocity fluctuations of the zero-mean. Effectively, the mass transfer from the fluid to the growing object is not convective but implicitly it is diffusive because diffusion is induced by velocity fluctuations of the fluid particles in the vicinity of the interface. Notice that the fluid particles of the interface region will stick to the object if their velocity (and in consequence energy and momentum) is sufficiently large in order to overcome the potential barrier of the interface. In this picture, it can be achieved by fluctuations of the particle velocity.

4. Kinetic equations

Eq. (16) defines a non-stationary stochastic process $R(t)$. To obtain its one-dimensional probability density $P(R,t)$ one can use the relation [10]

$$P(R,t) = \langle \delta(R - R(t)) \rangle \,, \tag{21}$$

where $R(t)$ is a solution of the stochastic equation (16) with a given initial condition and a given realization of the random field $v(r,t)$. The relation (21) can easily be proved using the relation between $P(R,t)$ and the characteristic function of the process $R(t)$, and the representation of $\delta(R)$ as a Fourier transform of the unit function $f(R) = 1$.

The process $R(t)$ as a solution of (16) is a functional of the random field $v(r,t)$. The variable R has no relation with the function $R(t)$. The brackets $\langle \ldots \rangle$ indicate averages over all realizations of the random field $v(r,t)$ and all possible initial conditions. Differentiating with respect to time the above equation and using the differential equation (16) yields the relation

$$\frac{\partial}{\partial t} P(R,t) = -\frac{\partial}{\partial R} A(R) \langle \delta(R - R(t)) v(R,t) \rangle \,, \tag{22}$$

where we used the properties of the δ-function and replaced the functional derivative $\delta / \delta R(t)$ by derivative $-\partial / \partial R$ which could be taken out of the average. Applying the Furutsu–Novikov–Donser formula [11] (see Appendix) we get

$$\begin{aligned} \frac{\partial}{\partial t} P(R,t) &= \frac{\partial}{\partial R} A(R) \int dR' \int\limits_0^t d\tau \; \langle v(R,t) v(R',\tau) \rangle \\ &\quad \times \frac{\partial}{\partial R} \left\langle \delta(R - R(t)) \frac{\delta R(t)}{\delta v(R',\tau)} \right\rangle . \end{aligned} \tag{23}$$

The functional derivative can be calculated in the same way as in [12] leading to the relation

$$\begin{aligned} &\frac{\partial}{\partial t} P(R,t) = \frac{\partial}{\partial R} A(R) \int dR' \int\limits_0^t d\tau \; \langle v(R,t) v(R',\tau) \rangle \\ &\times \frac{\partial}{\partial R} \left\langle \delta(R - R(t)) A(R(\tau)) \delta(R' - R(\tau)) \exp\left[\int\limits_\tau^t ds \frac{dA(R(s))}{dR(s)} v(R(s),s) \right] \right\rangle . \end{aligned} \tag{24}$$

Let us observe that, using Eq. (16), one can write

$$A(R(\tau)) = A(R(t)) \exp\left[\int\limits_t^\tau ds \frac{dA(R(s))}{dR(s)} v(R(s),s) \right] . \tag{25}$$

Inserting it into Eq. (24) gives

$$\begin{aligned}\frac{\partial}{\partial t}P(R,t) &= \frac{\partial}{\partial R}A(R)\int dR' \int\limits_0^t d\tau\ \langle v(R,t)v(R',\tau)\rangle \\ &\quad\times\frac{\partial}{\partial R}A(R)\left\langle\delta(R-R(t))\delta(R'-R(\tau))\right\rangle\,, \end{aligned}\tag{26}$$

where

$$\left\langle\delta(R-R(t))\delta(R'-R(\tau))\right\rangle \equiv P(R,t;R',\tau)\tag{27}$$

is a two-event probability distribution. Hence, a single-event probability distribution $P(R,t)$ for the process $R(t)$ obeys in a general case the following evolution equation

$$\frac{\partial}{\partial t}P(R,t)=\frac{\partial}{\partial R}A(R)\int dR'\int\limits_0^t dt'\mathcal{F}(R,t;R',t')\frac{\partial}{\partial R}A(R)P(R',t';R,t)\,.\tag{28}$$

It is an integro-differential equation, both with respect to time and space coordinates. Using the same procedure for the two-event probability distribution, we get the evolution equation in the form

$$\begin{aligned}\frac{\partial}{\partial t}P(R',t';R,t) &= \frac{\partial}{\partial R}A(R)\int dR'' \int\limits_0^t dt''\ \mathcal{F}(R,t;R'',t'') \\ &\quad\times\left[\frac{\partial}{\partial R}A(R)+\frac{\partial}{\partial R'}A(R')\right]P(R'',t'';R',t';R,t)\,,\end{aligned}\tag{29}$$

where $t'' < t' < t$. Generally, an m-event probability distribution obeys the equation

$$\begin{aligned}\frac{\partial}{\partial t_m}\quad P(R_1,t_1;\ldots;R_m,t_m) &= \frac{\partial}{\partial R_m}A(R_m)\int dr_0\int\limits_0^t dt_0\ \mathcal{F}(R_m,t_m;R_0,t_0) \\ \times&\left[\frac{\partial}{\partial R_m}A(R_m)+\frac{\partial}{\partial R_{m-1}}A(R_{m-1})+\cdots+\frac{\partial}{\partial R_1}A(R_1)\right] \\ \times&\ P(R_0,t_0;\ldots;R_m,t_m)\,,\end{aligned}\tag{30}$$

where $t_0 < t_1 < \ldots < t_{m-1} < t_m$. In consequence we get an infinity chain of equations.

5. Special cases

In a general case, the problem (16) is equivalent to the infinite hierarchy of the evolution equations (28)–(30). However, for particular forms of the correlations functions, one can get a closed and finite set of equations. We have constructed two models of the velocity fluctuations which allow to close the system (28)–(30).

5.1. Temporally correlated fluctuations

The first model refers to the case of only time-correlated fluctuations, *i.e.* when the velocity fluctuations do not depend on the space coordinate. This problem was investigated in our earlier paper [6] for which

$$v(R,t) = V(t)\,, \tag{31}$$

$$\langle V(t)\rangle = 0, \qquad \langle V(t)V(s)\rangle = \mathcal{F}(\mid t-s\mid)\,, \tag{32}$$

Then $P(R,t)$ obeys the Fokker–Planck equation in the form [13]

$$\frac{\partial}{\partial t}P(R,t) = D(t)\frac{\partial}{\partial R}A(R)\frac{\partial}{\partial R}A(R)P(R,t)\,, \tag{33}$$

where the diffusion function $D(t)$ is

$$D(t) = \int\limits_0^t \mathcal{F}(s)ds\,. \tag{34}$$

Imposing proper initial and boundary conditions, the solution of (33) reads

$$P(R,t) = \frac{1}{A(R)}\frac{1}{\sqrt{\pi\tau(t)}}\exp\left[-\frac{(x(R)-x(R_0))^2}{4\tau(t)}\right]\,, \tag{35}$$

where

$$x(R) = \frac{1}{\alpha}[R-(R^*+2\mathit{\Gamma})\ln(R+2\mathit{\Gamma})], \tag{36}$$

and

$$\tau(t) = \int\limits_0^t D(s)ds\,. \tag{37}$$

The function $\tau(t)$ is a rescaled time for the growth process. Kinetics of growth process controlled by fluctuations (31) with the statistical moments (32) was investigated in details in [6].

5.2. Spatially correlated fluctuations

Let the correlation function (20) of the velocity fluctuations be of the form

$$\mathcal{F}(R,t;R',t') = 2\,\delta(t-t')\,G(R,R')\,, \tag{38}$$

which consists of two parts: the time-dependent and the space-dependent ones. The time-dependent part is realized by the Dirac-delta function. It means that the velocity fluctuations of the fluid particles are non-correlated in time. The second space-dependent part can be any function of the space coordinates. In this case, the evolution equation for the probability distribution of the process (16) reduces to a diffusion equation in the form

$$\frac{\partial}{\partial t}P(R,t) = \frac{\partial}{\partial R}A(R)G(R,R)\frac{\partial}{\partial R}A(R)P(R,t)\,. \tag{39}$$

This equation can be analyzed, at least numerically and compared with those for the time-correlated fluctuations. The results will be presented elsewhere.

5.3. Concluding remarks

Although Eq. (16) seems to look simple, its analysis in a general case is extremely difficult. The two above presented examples of fluctuations can be treated analytically. Other cases could be studied under some simplified approximations (as *e.g.* decoupling, decorrelation, *etc.*). The open problem remains under what limiting cases, such approximate schemes can rigorously be exploited.

In summary, we have presented models and problems related to growth processes driven by velocity fluctuations of particles of solution in which the object grows. It would allow to investigate a mechanism of normal and anomalous kinetics of growing crystals and investigate a large class of growth processes controlled by different concentration fields. A rich behavior in the growth kinetics might appear due to various statistics of the particles of the saturated solution. In general case the evolution equations for growth process controlled by velocity fluctuations do not form a closed system. However, for some specific models of velocity fluctuations or by approximation procedures one can close a system of evolution equations.

This work supported in part by the Project CLUSTERICS, The National Linux Cluster, and the ESF program: Stochastic Dynamics.

Appendix

The Furutsu–Novikov–Donsker formula

Let $F[v]$ be an arbitrary functional of any Gaussian field $v(r,t)$. Then [11]

$$\langle v(r,t)F[v]\rangle = \int dr' \int\limits_0^t d\tau \, \langle v(r,t)v(r',\tau)\rangle \left\langle \frac{\delta F[v]}{\delta v(r',\tau)} \right\rangle . \tag{40}$$

In this relation, $\langle v(r,t)v(r',\tau)\rangle$ is a two-point correlation function of the process $v(r,t)$ and $\delta F[v]/\delta v(r',\tau)$ denotes a functional derivative.

REFERENCES

[1] A.A. Chernov, *Modern Crystallography III. Crystal Growth*, Springer, Berlin 1984.

[2] T. Vicsek, *Fractal Growth Phenomena*, World Scientific, Singapore 1992.

[3] J. Schmeltzer, G. Röpke, R. Mahnke, *Aggregation Phenomena in Complex Systems*, Wiley-VCH, Weinheim 1999.

[4] B.J. Ackerson, K. Schätzel, *Phys. Rev.* **E52**, 6448 (1995); D.J.W. Aastuen, N.A. Clark, L.K. Cotter, B.J. Ackerson, *Phys. Rev. Lett.* **57**, 1733 (1986); N.M. Dixit, Ch.F. Zukoski, *Phys. Rev.* **E64**, 041604 (2001).

[5] W.W. Mullins, R.F. Sekerka, *J. Appl. Phys.* **34**, 323 (1963).

[6] J. Łuczka, M. Niemiec, R. Rudnicki, *Phys. Rev.* **E65**, 051401 (2002).

[7] F. Rosenberger, *J. Cryst. Growth* **76**, 618 (1986).

[8] A. Gadomski, Z.J. Grzywna, J. Łuczka, *Chem. Eng. Sci.* **48**, 3713 (1993).

[9] A. Gadomski, J. Łuczka, *Acta Phys. Pol. B* **24**, 725 (1993).

[10] P. Hänggi, P. Jung, *Adv. Chem. Phys.* **89**, 239 (1994).

[11] K. Furutsu, *J. Res. Natl. Bur. Standards* **67D**, 303 (1963); E.A. Novikov, *Zh. Eksp. Teor. Fiz.* **47**, 1919 (1964); M. Donsker, in *Proceedings of the Conference on The Theory and Applications of Analysis in Function Space*, MIT Press, Cambridge MA 1964, p. 17.

[12] P. Hänggi, *Noise in Nonlinear Dynamical System*, Eds. F. Moss and P.V.E. McClintock, Cambridge University Press, Cambridge 1989, Vol. 1, p. 307.

[13] P. Hänggi, H. Thomas, *Phys. Rep.* **168**, 207 (1982).