Universality classes for asymptotic behavior of relaxation processes in systems with dynamical disorder: Dynamical generalizations of stretched exponential

Marcel Ovidiu Vlada)

Centre for Nonlinear Dynamics in Physiology and Medicine, McGill University, 3655 Drummond Street, Montreal, Quebec H3G 1Y6, Canada

Ralf Metzler and Theo F. Nonnenmacher

Department of Mathematical Physics, University of Ulm, Albert-Einstein-Allee 11, 89069 Ulm, Germany

Michael C. Mackey

Departments of Physiology, Physics, and Mathematics, McGill University, 3655 Drummond Street, Montreal, Quebec H3G 1Y6, Canada

(Received 26 September 1995; accepted for publication 16 November 1995)

The asymptotic behavior of multichannel parallel relaxation processes for systems with dynamical disorder is investigated in the limit of a very large number of channels. An individual channel is characterized by a state vector \mathbf{x} which, due to dynamical disorder, is a random function of time. A limit of the thermodynamic type in the x-space is introduced for which both the volume available and the average number of channels tend to infinity, but the average volume density of channels remains constant. Scaling arguments combined with a stochastic renormalization group approach lead to the identification of two different types of universal behavior of the relaxation function corresponding to nonintermittent and intermittent fluctuations, respectively. For nonintermittent fluctuations a dynamical generalization of the static Huber's relaxation equation is derived which depends only on the average functional density of channels, $\rho[W(t')]D[W(t')]$, the channels being classified according to their different relaxation rates W = W(t'), which are random functions of time. For intermittent fluctuations a more complicated relaxation equation is derived which, in addition to the average density of channels, $\rho[W(t')]D[W(t')]$, depends also on a positive fractal exponent H which characterizes the fluctuations of the density of channels. The general theory is applied for constructing dynamical analogs of the stretched exponential relaxation function. For nonintermittent fluctuations the type of relaxation is determined by the regression dynamics of the fluctuations of the relaxation rate. If the regression process is fast and described by an exponential attenuation function, then after an initial stretched exponential behavior the relaxation process slows down and it is not fully completed even in the limit of very large times. For self-similar regression obeying a negative power law, the relaxation process is less sensitive to the influence of dynamical disorder. Both for small and large times the relaxation process is described by stretched exponentials with the same fractal exponent as for systems with static disorder. For large times the efficiency of the relaxation process is also slowed down by fluctuations. Similar patterns are found for intermittent fluctuations with the difference that for very large times and a slow regression process a crossover from a stretched exponential to a self-similar algebraic relaxation function occurs. Some implications of the results for the study of relaxation processes in

^{a)}Permanent address: Romanian Academy of Sciences, Centre for Mathematical Statistics, Casa Academiei, Calea 13 Septembrie No. 13, OPB Bucuresti 5, 76100 Bucharest, Romania. Address after January 1, 1996: Department of Chemistry, Stanford University, Stanford, CA 94305-5080.

condensed matter physics and im molecular biology are investigated. © *1996 American Institute of Physics*. © 1996 American Institute of Physics. [S0022-2488(96)01004-3]

I. INTRODUCTION

In the last two decades an enormous amount of experimental evidence has been accumulated showing that the decay of the average survival (relaxation) function $\langle \ell(t) \rangle$ in many diverse systems in condensed matter physics and in molecular biology follows the same stretched exponential law of the Kohlrausch–Williams–Watts (KWW) type

$$\langle l(t)\rangle = \exp[-(\Omega t)^{\beta}], \quad 1 > \beta > 0, \tag{1.1}$$

where β is a positive fractal exponent between zero and unity and Ω is a characteristic frequency. Equation (1.1) was first proposed by Kohlrausch¹ in 1864 to describe the mechanical creep and was later used by Williams and Watts² to describe the dielectric relaxation in polymers and by Weibull³ for describing the failure data in reliability theory. More recently the KWW law has been used to fit the data on remanent magnetization in spin glasses,⁴ the decay of luminiscence in porous glasses,⁵ the relaxation processes in viscoelasticity⁶ on the reaction kinetics of biopolimers,⁷ and on the dynamics of recombination kinetics in radiochemistry.⁸ Further applications include the description of the statistical distributions of open and closed times of ion channels in molecular biophysics⁹ or even the description of the survival functions of cancer patients.¹⁰

The ubiquity of the stretched exponential law (1.1) has led to the idea that there should be a kind of universal mechanism generating it which is independent of the details of an individual process. An argument in favor of this opinion is the close connection between the KWW law (1.1)and the stable probability densities of the Lévy type^{1,11} which emerge as a result of the occurrence of a large number of independent random events described by individual probability densities with infinite moments. Many attempts of searching for such a universal mechanism for the occurrence of the stretched exponential have been presented in the literature. A first attempt is a generalization of a mechanism of parallel relaxation initially suggested by Förster for the extinction of luminescence¹² and improved by other authors.¹³ A second model assumes a complex serial relaxation on a multilevel abstract structure which emphasizes the role of hierarchically constrained dynamics.¹⁴ A third model is a generalization of the defect-diffusion model of Shlesinger and Montroll.¹⁵ All three of these models have been carefully examined by Klafter and Shlesinger;¹⁶ they have shown that in spite of the different details of the three models a universal common feature exists which is the existence of a broad spectrum of relaxation rates described by a scale-invariant distribution. A complementary approach of the universal features of the stretched exponential which is mathematically oriented is based on the powerful technique of fractional calculus and its connections with the theory of Fox functions.¹⁷

An interesting approach has been suggested by Huber;¹⁸ based on a careful examination of the models used for the description of the extinction of luminescence he has derived a general relaxation function

$$\langle l(t)\rangle = \exp\left\{-\int_0^\infty \rho(W)[1-\exp(-Wt)]dW\right\},\tag{1.2}$$

where $\rho(W)dW$ is the number of channels involved in the relaxation process and characterized by an individual relaxation rate between W and W+dW. If the distribution of rates is self-similar and obeys a scaling law of the negative power law type

$$\rho(W)dW \sim \operatorname{const} W^{-(1+\beta)} dW, \qquad (1.3)$$

which is consistent with the general ideas of self-similarity suggested by Klafter and Shlesinger,¹⁶ then Huber's equation (1.2) leads to the stretched exponential law (1.1). The proportionality constant in Eq. (1.3) can be easily determined in terms of the fractal exponent β and of the characteristic frequency Ω entering Eq. (1.1), resulting in

$$\rho(W)dW = [\Gamma(1-\beta)]^{-1}\beta\Omega^{\beta}W^{-(1+\beta)} dW, \qquad (1.3')$$

where $\Gamma(x) = \int_0^\infty t^{x-1} \exp(-t) dt$, x > 0, is the complete gamma function. Although Huber has suggested that his equation might be generally valid for any disordered system with static disorder, the validity range of his derivation, based on the approximation of a product by an exponential, cannot be easily evaluated.

Recently two of the authors of the present paper have shown that Huber's equation (1.2) is exact for a Poissonian distribution of independent channels.¹⁹ Moreover, it has been recently shown that Huber's equation (1.2) also holds beyond the validity range of the Poissonian distribution: it emerges as a universal scaling law for a uniform random distribution of a large number of channels characterized by nonintermittent fluctuations.²⁰ This result is consistent with the general idea that the Huber's equation (1.2) and the stretched exponential relaxation law (1.1) derived from it can be generated by a central limit behavior of the Lévy type which expresses the contribution of a very large number of weakly connected relaxation channels. The analysis presented in Ref. 20 also shows that Huber's equation (1.2) is not the unique universal law which emerges in the limit of a very large number of weakly coupled channels. For intermittent fluctuations of the number of channels at least one supplementary scaling law exists, which is given by

$$\langle \mathscr{O}(t) \rangle = \mathscr{F}_{H} \left[\int_{0}^{\infty} \rho(W) [1 - \exp(-Wt)] dW \right], \tag{1.4}$$

where the function

$$\mathcal{J}_{H}(z) = H[(1+1/H)z]^{-H}\gamma(H,(1+1/H)z)$$
(1.5)

depends on the incomplete gamma function $\gamma(x,u) = \int_0^u t^{x-1} \exp(-t) dt$, x > 0, $u \ge 0$, and *H* is a positive fractal exponent which characterizes the fluctuations of the number of channels. The reciprocal value of the fractal exponent, 1/H, is a measure of the intermittency of fluctuations. In particular in the limit

$$1/H \rightarrow 0 \quad (H \rightarrow \infty), \tag{1.6}$$

the fluctuations are nonintermittent, the function $\mathcal{J}_{H}(z)$ becomes an exponential

$$\lim_{H \to \infty} \mathscr{T}_H(z) = \exp(-z), \tag{1.7}$$

and the scaling law (1.4) reduces to the Huber's scaling equation (1.1). The derivation of the intermittent scaling law (1.4) is based on the searching for a fixed point by means of a stochastic renormalization group approach technique.²¹ Unfortunately the renormalization group technique used in Ref. 20 does not guarantee that the fixed point corresponding to Eq. (1.4) is unique, and thus other intermittent limit scaling laws corresponding to other fixed points may also exist.

By assuming that the distribution of relaxation rates is given by the scale-invariant law (1.3'), the intermittent generalization (1.4) of the Huber's equation leads to the relaxation law

$$\langle \ell(t) \rangle = H(\Omega t)^{-\beta H} (1 + 1/H)^{-H} \gamma(H, (\Omega t)^{\beta} (1 + 1/H)),$$
 (1.8)

which for small times reduces to a stretched exponential

$$\langle \ell(t) \rangle \sim \exp[-(\Omega t)^{\beta}], \quad t \ll \Omega^{-1},$$
(1.9)

and for large times it is given by a negative power law

$$\langle \ell(t) \rangle \sim \Gamma(1+H)(\Omega t)^{-\beta H}(1+1/H)^{-H}, \quad t \ge \Omega^{-1}, \quad H = \text{finite.}$$
 (1.10)

As the fractal exponent *H* increases, the intermittent nature of fluctuations becomes less and less pronounced, the stretched exponential portion of the relaxation function $\langle \ell(t) \rangle$ given by Eq. (1.8) becomes longer and longer and the power law tail becomes shorter and shorter; eventually in the limit $H \rightarrow \infty$, corresponding to nonintermittent fluctuations, the whole relaxation function $\langle \ell(t) \rangle$ can be represented by a stretched exponential.

All these attempts at coming up with a general derivation of the stretched exponential are based on the assumption that the disordered distribution of channels is static, i.e., that an initial fluctuation of the number of channels characterized by different relaxation rates is frozen forever; during the process of relaxation the distribution of channels remains invariant and described by the static density function $\rho(W) dW$. A channel initially characterized by a relaxation rate W is supposed to be characterized by the same rate W at any time in the future. Although reasonable for some problems of condensed matter physics, the validity of this assumption is questionable in molecular biology. In the case of protein–ligand interactions⁷ and of ion channel kinetics⁹ the distribution of relaxation channels with different rates is due to the conformational fluctuations of protein molecules which have a dynamical nature and thus the fluctuations of the numbers of channels characterized by different relaxation rates are continuously generated and destroyed by thermal agitation.

The study of rate or relaxation processes with dynamical disorder is an active field of applied statistical physics.^{22–26} Although at times the possible connection between the stretched exponential relaxation and the dynamical disorder has also been considered,²⁷ little attention has been paid to the derivation of dynamic generalizations of the stretched exponential law which emerge in the limit of a very large number of reaction channels. The purpose of this paper is the searching for such universal scaling laws which are dynamical analogs of the general static limit laws (1.2) and (1.4). The starting point of our approach is the theory developed in Refs. 19 and 20 in which a general approach of rate processes with dynamical disorder has been suggested on the basis of the theory of random point processes.²⁸ In Ref. 19 in the particular case of Poissonian channels a dynamical generalization of the Huber's equation (1.2) has been suggested

$$\langle \mathscr{E}(t) \rangle = \exp\left\{-\overline{\int \int} \rho[W(t')]D[W(t')]\left[1 - \exp\left(-\int_0^t W(t')dt'\right)\right]\right\},\tag{1.11}$$

where, due to dynamical disorder, the relaxation rate corresponding to an individual channel is a random function of time W = W(t'), $t \ge t' \ge 0$, $\rho[W(t')]D[W(t')]$ is an average functional density of channels characterized by different random functions W = W(t'), D[W(t')] is a suitable integration measure over the space of functions W(t'), and $\overline{\int f}$ stands for the operation of path integration. In the following we shall try to derive the dynamic analog (1.11) of Huber's law as a universal limit expression which emerges in the limit of a very large number of weakly interacting channels. We shall also try to derive a universal dynamical intermittent law which is the analog of the static scaling law (1.4):

$$\langle \mathscr{C}(t) \rangle = \mathscr{T}_{H} \left\{ \underbrace{\int \int} \rho[W(t')] D[W(t')] \left[1 - \exp\left(-\int_{0}^{t} W(t') dt' \right) \right] \right\}.$$
(1.12)

Another objective of the article is the application of the universal laws (1.11) and (1.12) to the particular case of a self-similar dynamical distribution of channels which is the analog of the static

J. Math. Phys., Vol. 37, No. 5, May 1996

equation (1.3'). Carrying out this program would lead to dynamical generalizations of the stretched exponential law (1.1) and of its intermittent generalization (1.8).

The structure of the paper is as follows. In Sec. II we give a general formulation of the problem in terms of a functional generalization of the theory of random point processes. In Secs. III and IV the approach developed in Sec. II is used for the derivation of the relaxation functions (1.11) and (1.12) as universal limit laws for nonintermittent and intermittent fluctuations, respectively, valid for a very large number of weakly interacting relaxation channels. In Sec. V explicit dynamical generalizations of the stretched exponential law are derived by computing the path averages in Eqs. (1.11) and (1.12) in the particular case of a stationary self-similar dynamical distribution of relaxation channels. In Sec. VI a comparative numerical analysis of the relaxation equations for static and dynamical disorder is presented. Finally in Secs. VII and VIII some possibilities of application of our approach are analyzed and some open questions are pointed out.

II. FORMULATION OF THE PROBLEM

We consider a relaxation process in which a random (usually very large) number of relaxation modes are involved. By following the usual nomenclature in nuclear physics and molecular dynamics we shall call these modes relaxation channels. The relaxation channels are abstract entities which are characterized by different state vectors $\mathbf{x}_1(t')$, $\mathbf{x}_2(t')$,..., $t \ge t' \ge 0$, which, due to dynamical disorder, are random functions of time. The relaxation channels should not be mistaken for the actual ion channels crossing a cell membrane,⁹ which are concrete objects.

The stochastic properties of the state vectors $\mathbf{x}_1(t'),...,\mathbf{x}_N(t')$ attached to the different individual relaxation channels can be described by a functional generalization of random point processes. A slightly different type of functional random point process has been suggested in Ref. 19. For describing the dynamics of the relaxation channels we introduce a set of grand canonical Janossy probability density functionals

$$Q_0, Q_N[\mathbf{x}_1(t'), \dots, \mathbf{x}_N(t')] D[\mathbf{x}_1(t')] \cdots D[\mathbf{x}_N(t')], \qquad (2.1)$$

with the normalization condition

$$Q_0 + \sum_{N=1}^{\infty} \frac{1}{N!} \underbrace{\int \int}_{\cdots} \underbrace{\int \int}_{\cdots} Q_N[\mathbf{x}_1(t'), \dots, \mathbf{x}_N(t')] D[\mathbf{x}_1(t')] \cdots D[\mathbf{x}_N(t')] = 1.$$
(2.2)

Here $Q_N[\mathbf{x}_1(t'),...,\mathbf{x}_N(t')]D[\mathbf{x}_1(t')]\cdots D[\mathbf{x}_N(t')]$ is the probability that there are N relaxation channels and that these N channels are characterized by state vectors close to $\mathbf{x}_1(t'),...,\mathbf{x}_N(t')$ and $D[\mathbf{x}(t')]$ is a suitable integration measure over the space of functions $\mathbf{x}(t')$. This type of description is based on the implicit assumption that for a given realization of the process the total number N of channels is a random quantity which does not change in time. The initial number N of channels is randomly chosen and then kept constant and only the random vectors $\mathbf{x}_1(t'),...,\mathbf{x}_N(t')$ are variable in time. An alternative description of the stochastic properties of the relaxation channels is given in terms of the generating functional

$$\Lambda[f[\mathbf{x}(t')]] = Q_0 + \sum_{N=1}^{\infty} \frac{1}{N!} \underbrace{\int \int}_{\dots} \cdots \underbrace{\int \int}_{\dots} Q_N[\mathbf{x}_1(t'), \dots, \mathbf{x}_N(t')] \times D[\mathbf{x}_1(t')] \cdots D[\mathbf{x}_N(t')] f[\mathbf{x}_1(t')] \cdots f[\mathbf{x}_N(t')], \qquad (2.3)$$

where $f[\mathbf{x}(t')]$ is a suitable test functional. The main advantage of using the generating functional $\Lambda[f[\mathbf{x}(t')]]$ is that it can be written in a form independent of the integration measure $D[\mathbf{x}(t')]$, which is generally unknown.

Considering a time interval of length t we assume that for each channel u = 1,...,N, there is a fluctuating probability of decay $p_u(t)$. This probability depends on the whole previous history of the channel, that is, $p_u(t)$ is a functional of all previous values $\mathbf{x}_u(t')$, $t \ge t' \ge 0$ of the state vector:

$$p_{u}(t) = p[\mathbf{x}_{u}(t');t].$$
 (2.4)

A realization of the survival (relaxation) function $\ell(t)$, that is, the probability that the relaxation process has not occured in a time interval of length t, is simply given by the product of the complementary probabilities $1-p[\mathbf{x}_u(t');t]$ attached to all channels, which expresses the probability that none of the N channels has led to relaxation:

$$\mathscr{E}(t) = \prod_{u=1}^{N} \{1 - p[\mathbf{x}_{u}(t'); t)]\}.$$
(2.5)

The average relaxation function $\langle \ell(t) \rangle$ can be computed by evaluating the average of the fluctuating function $\ell(t)$ in terms of the grand canonical Janossy probability density functionals (2.1), which describe the random evolution of the channels:

$$\langle \mathscr{C}(t) \rangle = Q_0 + \sum_{N=1}^{\infty} \frac{1}{N!} \overline{\int \int} \cdots \overline{\int \int} Q_N[\mathbf{x}_1(t'), \dots, \mathbf{x}_N(t')] D[\mathbf{x}_1(t')] \cdots D[\mathbf{x}_N(t')]$$

$$\times f[\mathbf{x}_1(t')] \cdots f[\mathbf{x}_N(t')] \prod_{u=1}^{N} \{1 - p[\mathbf{x}_u(t'); t]\}$$

$$= \Lambda[f[\mathbf{x}(t')] = 1 - p[\mathbf{x}(t'); t]],$$

$$(2.6)$$

where we have used the definition (2.3) of the generating functional $\Lambda[f[\mathbf{x}(t')]]$. It follows that the evaluation of the average relaxation function $\langle \ell(t) \rangle$ reduces to the computation of the generating functional $\Lambda[f[\mathbf{x}(t')]]$, which describes the random couplings between the different relaxation channels, and to the computation of the probability $p[\mathbf{x}(t');t]$, which describes the individual behavior of a single channel.

For relating the generating functional, $\Lambda[f[\mathbf{x}(t')]]$, to the fluctuation dynamics of the number of channels we introduce the fluctuating functional density of channels

$$\eta[\mathbf{x}(t')]D[\mathbf{x}(t')] \quad \text{with} \quad N = \underbrace{\int \int} \eta[\mathbf{x}(t')]D[\mathbf{x}(t')], \quad (2.7)$$

characterized by a random vector near $\mathbf{x}(t')$ and the corresponding characteristic functional

$$G[K[\mathbf{x}(t')]] = \left\langle \exp\left(i \int \int K[\mathbf{x}(t')] \eta[\mathbf{x}(t')] D[\mathbf{x}(t')]\right) \right\rangle, \qquad (2.8)$$

where $K[\mathbf{x}(t')]$ is a suitable test functional. The fluctuations of the functional density of channels $\eta[\mathbf{x}(t')]D[\mathbf{x}(t')]$ are described in terms of the corresponding cumulants

$$\langle \langle \eta[\mathbf{x}_1(t')] \cdots \eta[\mathbf{x}_m(t')] \rangle \rangle, \quad m = 1, 2, \dots,$$
(2.9)

which are assumed to exist and be finite. The characteristic functional $G[K[\mathbf{x}(t')]]$ can be expressed in terms of $\langle\langle \eta[\mathbf{x}_1(t')]\cdots\eta[\mathbf{x}_m(t')]\rangle\rangle$, m=1,2,..., by means of the cumulant expansion

Vlad et al.: Universal behavior for dynamical disorder

$$\ln G[K[\mathbf{x}(t')]] = \sum_{m=1}^{\infty} \frac{i^m}{m!} \overline{\int \int} \cdots \overline{\int \int} \langle \langle \eta[\mathbf{x}_1(t')] \cdots \eta[\mathbf{x}_m(t')] \rangle \rangle K[\mathbf{x}_1(t')]$$
$$\times D[\mathbf{x}_1(t')] \cdots K[\mathbf{x}_m(t')] D[\mathbf{x}_m(t')].$$
(2.10)

For establishing a connection between the generating functional $\Lambda[f[\mathbf{x}(t')]]$ of the functional point process and the characteristic functional $G[K[\mathbf{x}(t')]]$ of the functional density of channels $\eta[\mathbf{x}(t')]D[\mathbf{x}(t')]$, we write a realization of the density of channels $\eta[\mathbf{x}(t')]D[\mathbf{x}(t')]$ as a sum of functional Dirac's delta symbols

$$\eta[\mathbf{x}(t')]D[\mathbf{x}(t')] = \sum_{u=1}^{N} \delta[\mathbf{x}_{u}(t') - \mathbf{x}(t')]D[\mathbf{x}(t')].$$
(2.11)

Equation (2.11) is a functional generalization of the well-known relationship from statistical mechanics expressing the particle density fields as sums of delta functions.²⁹ We insert Eq. (2.11) into the definition (2.8) of the characteristic functional $G[K[\mathbf{x}(t')]]$, and compute the average in terms of the grand canonical Janossy probability density functionals (2.1). By using the definition (2.3) of the generating functional $\Lambda[f[\mathbf{x}(t')]]$ after getting rid of the functional integral in the exponent due to the filtration property of the Dirac's functional symbol and computing the resulting sum, we obtain

$$G[K[\mathbf{x}(t')]] = \Lambda[f[\mathbf{x}(t')] = \exp(iK[\mathbf{x}(t')])].$$
(2.12)

It follows that the average relaxation function $\langle \ell(t) \rangle$ can be expressed as

$$\langle \ell(t) \rangle = G[K[\mathbf{x}(t')] = ib[\mathbf{x}(t');t]], \qquad (2.13)$$

where

$$b[\mathbf{x}(t');t] = -\ln(1 - p[\mathbf{x}(t');t]), \qquad (2.14)$$

is the bit number³⁰ of the individual probability of nonrelaxation $1-p[\mathbf{x}(t');t]$ attached to an individual channel with a history characterized by the function $\mathbf{x}(t')$, $t \ge t' \ge 0$. Equation (2.13) is a dynamical generalization of a similar relationship derived in Ref. 20 for systems with static disorder by using a different method that does not make use of the theory of random point processes.

For deriving an expression for the probability of decay $p[\mathbf{x}(t');t]$ attached to an individual channel we generalize an assumption made for systems with static disorder by Huber¹⁸ and by Vlad, Schönfisch, and Mackey.²⁰ We assume that a channel characterized by a state vector \mathbf{x} can be either in an open state with a probability $\lambda(\mathbf{x})$ or in a closed state with a probability $1-\lambda(\mathbf{x})$. Following Ref. 20 we suppose that the state vector \mathbf{x} of a channel belongs to a certain domain Σ of the state space which is simply connected and has the volume

$$V_{\Sigma} = \int_{\Sigma} d\mathbf{x}, \qquad (2.15)$$

and that the probability $\lambda(\mathbf{x})$ that the channel is open is simply given by

$$\lambda(\mathbf{x}) = V^*(\mathbf{x})/V_{\Sigma}, \qquad (2.16)$$

where $V^*(\mathbf{x})$ is a characteristic volume of a neighborhood of the position \mathbf{x} .

We assume that an open channel characterized by a state vector **x** has a rate of relaxation $W(\mathbf{x})$ that depends only on the state vector **x**. Since the state vector **x** is a random function of time, the contribution of an open state to the individual probability of survival (nonrelaxation) $1-p[\mathbf{x}(t');t]$ is given by

$$\mathscr{E}[W(\mathbf{x}(t'))] = \exp\left(-\int_0^t W(\mathbf{x}(t'))dt'\right).$$
(2.17)

The corresponding contribution for a closed state is simply equal to $\mathscr{E}[W(\mathbf{x}(t'))]=1$ and the individual probability of survival $1-p[\mathbf{x}(t');t]$ is given by the average of the $\mathscr{E}[W(\mathbf{x}(t'))]$ -factor corresponding to the two states

$$1 - p[\mathbf{x}(t');t] = \lambda(\mathbf{x}(t))\exp\left(-\int_0^t W(\mathbf{x}(t'))dt'\right) + 1 - \lambda(\mathbf{x}(t)), \qquad (2.18)$$

from which we obtain the following expression for the individual probability of decay

$$p[\mathbf{x}(t');t] = \frac{V^*(\mathbf{x}(t))}{V_{\Sigma}} \left\{ 1 - \exp\left[-\int_0^t W(\mathbf{x}(t'))dt'\right] \right\}.$$
 (2.19)

Now the average survival function $\langle \ell(t) \rangle$ is completely characterized by the collective stochastic properties of the fluctuations of the numbers of channels, expressed by the cumulants $\langle \langle \eta[\mathbf{x}_1(t')] \cdots \eta[\mathbf{x}_m(t')] \rangle \rangle$ given by Eqs. (2.9) or by the cumulant expansion (2.10) of the characteristic functional $G[K[\mathbf{x}(t')]]$ and by the behavior of an individual channel, characterized by the probability of decay given by Eq. (2.19). For investigating the scaling behavior emerging in the limit of a very large average number $\langle N \rangle$ of channels

$$\langle N \rangle = \overline{\int \int} \langle \langle \eta[\mathbf{x}(t')] \rangle \rangle D[\mathbf{x}(t')] \to \infty, \qquad (2.20)$$

we introduce a limit of the thermodynamic type for which both the total volume V_{Σ} available in the **x**-space and the average total number $\langle N \rangle$ of channels tend to infinity, but the average density of channels,

$$\varepsilon = \langle N \rangle / V_{\Sigma},$$
 (2.21)

remains constant

$$V_{\Sigma}, \langle N \rangle \rightarrow \infty \text{ with } \varepsilon = \langle N \rangle / V_{\Sigma} = \text{const.}$$
 (2.22)

For evaluating the different types of asymptotic behavior emerging in the limit (2.22) we assume that the channels are weakly interacting, that is, as the total space volume increases to infinity, $V_{\Sigma} \rightarrow \infty$, the characteristic volumes $V^*(\mathbf{x}_1)$, $V^*(\mathbf{x}_2)$,..., of the neighborhoods of the different channels remain finite and constant; in other words, the increase of the total space volume V_{Σ} does not lead to an increase of the possible overlapping among the neighborhoods attached to the different channels. This assumption of locality generates the two types of asymptotic behavior investigated in Secs. III and IV.

III. LIMIT BEHAVIOR FOR NONINTERMITTENT FLUCTUATIONS

We introduce the relative fluctuations of different orders:

$$c_m[\mathbf{x}_1(t'),\dots,\mathbf{x}_m(t')] = \frac{\langle \langle \eta[\mathbf{x}_1(t')] \cdots \eta[\mathbf{x}_m(t')] \rangle \rangle}{\prod_{u=1}^m \{\langle \langle \eta[\mathbf{x}_u(t')] \rangle \rangle\}}, \quad m \ge 2.$$
(3.1)

If the relative fluctuations $c_m[\mathbf{x}_1(t'),...,\mathbf{x}_m(t')], m \ge 2$, decrease to zero in the thermodynamic limit (2.22)

$$c_m[\mathbf{x}_1(t'), \dots, \mathbf{x}_m(t')] \rightarrow 0, \quad V_{\Sigma}, \langle N \rangle \rightarrow \infty, \text{ with } \varepsilon = \text{const}, m = 2, 3, \dots,$$
 (3.2)

then the fluctuations of the numbers of channels are nonintermittent. For investigating the asymptotic behavior of the survival function $\langle \ell(t) \rangle$ for nonintermittent fluctuations in the thermodynamic limit (2.22) we introduce the average probability density functional of the state vector $\mathbf{x}(t')$ of an individual channel,

$$\xi[\mathbf{x}(t')]D[\mathbf{x}(t')] = \frac{\langle \langle \eta[\mathbf{x}(t')] \rangle \rangle D[\mathbf{x}(t')]}{\underbrace{\int \int \langle \langle \eta[\mathbf{x}(t')] \rangle \rangle D[\mathbf{x}(t')]}, \qquad (3.3)$$

with

$$\overline{\int \int \mathbf{x}(t')]D[\mathbf{x}(t')] = 1, \qquad (3.4)$$

and combine Eqs. (2.10), (2.13), (2.14), (2.19), (2.20), and (3.1). We express the cumulants of the functional density of channels in terms of the relative fluctuations $c_1[\mathbf{x}_1(t'),...,\mathbf{x}_m(t')]$ and of the average probability density functional $\xi[\mathbf{x}(t')]D[\mathbf{x}(t')]$. By inserting the resulting expression for the cumulants into the functional Taylor expansion (2.10) for the logarithm of the characteristic functional $G[K[\mathbf{x}(t')]]$ and expressing the average relaxation function $\langle \ell(t) \rangle$ from Eqs. (2.13), (2.14), and (2.19) we obtain

$$\langle \mathscr{C}(t) \rangle = \exp \left\{ \sum_{m=1}^{\infty} \frac{\varepsilon^m}{m!} \overline{\int \int} \cdots \overline{\int \int} c_m [\mathbf{x}_1(t'), \dots, \mathbf{x}_m(t')] \boldsymbol{\xi} [\mathbf{x}_1(t')] D[\mathbf{x}_1(t')] \dots \boldsymbol{\xi} [\mathbf{x}_m(t')] \right. \\ \times D[\mathbf{x}_m(t')] \prod_{u=1}^m \left\{ V_{\Sigma} \ln \left\{ 1 - \frac{V^*(\mathbf{x}_u(t))}{V_{\Sigma}} \left[1 - \exp \left(- \int_0^t W(\mathbf{x}_u(t')) dt' \right) \right] \right\} \right\} \right\}, \quad (3.5)$$

where

$$c_1 = 1.$$
 (3.6)

From Eqs. (2.22), (3.2), (3.5), and (3.6) it follows that for nonintermittent fluctuations in the thermodynamic limit in Eq. (3.5) only the term corresponding to m = 1 survives and the expression for the average survival function $\langle \ell(t) \rangle$ reduces to the dynamical generalization (1.11) of Huber's equation:

$$\langle \mathscr{C}(t) \rangle \sim \exp\left\{-\overline{\int \int \rho} \left[W(t')\right] D[W(t')] \left[1 - \exp\left(-\int_0^t W(t')dt'\right)\right]\right\} \text{ as } V_{\Sigma}, \langle N \rangle \to \infty, \varepsilon = \text{const},$$
(3.7)

where

$$\rho[W(t')]D[W(t')] = \left\{ \varepsilon \underbrace{\int \int} V^*(\mathbf{x}(t))\xi[\mathbf{x}(t')]D[\mathbf{x}(t')]\delta[W(t') - W(\mathbf{x}(t'))] \right\} D[W(t')]$$
(3.8)

is the average density of channels involved in the relaxation process, the channels being classified according to their relaxation rates W(t'), $t \ge t' \ge 0$.

IV. LIMIT BEHAVIOR FOR INTERMITTENT FLUCTUATIONS

For the study of the asymptotic scaling behavior of the average survival function for intermittent fluctuations a renormalization group technique should be used. In the following we apply a probabilistic version²¹ of the Shlesinger–Hughes stochastic renormalization procedure³¹ which has been recently applied to the study of space-dependent epidemic processes with high migration.³² The method consists of starting out from an initial characteristic functional $G[K[\mathbf{x}(t')]]$ of the functional density of states for which the fluctuations are nonintermittent and constructing, by means of a succession of decimation processes, a renormalized characteristic functional $\tilde{G}[K[\mathbf{x}(t')]]$ for which the fluctuations of the density of states are intermittent. The main steps of such an approach are presented in another context in Ref. 21 and a simplified derivation is also presented in Ref. 32. Here we give only the final expression for the renormalized characteristic functional $\tilde{G}[K[\mathbf{x}(t')]]$:

$$\widetilde{G}[K[\mathbf{x}(t')]] = H \int_0^1 z^{H-1} G[-i \ln[1 - z[1 - \exp(iK[\mathbf{x}(t')])]]] dz; H > 0,$$
(4.1)

where H is a positive fractal exponent similar to the one entering the static equations (1.4)-(1.10).

For evaluating the limit scaling law for the average relaxation function $\langle \ell(t) \rangle$ corresponding to the renormalized expression (4.1) we expand in Eq. (4.1) the nonrenormalized characteristic functional $G[K[\mathbf{x}(t')]]$ in the cumulant expansion (2.10) and express the corresponding cumulants in terms of the nonrenormalized relative fluctuations $c_m[\mathbf{x}_1(t');...;\mathbf{x}_m(t')]$ and in terms of the average renormalized density of channels

$$\varepsilon = \frac{\langle \widetilde{N} \rangle}{V_{\Sigma}} = \frac{H}{H+1} \cdot \frac{\langle N \rangle}{V_{\Sigma}}.$$
(4.2)

Here we have used the relationship between the nonrenormalized average number of channels $\langle N \rangle$ and the corresponding renormalized average $\langle \tilde{N} \rangle$:

$$\langle \widetilde{N} \rangle = \langle N \rangle H/(H+1). \tag{4.3}$$

The relationship (4.3) can be derived from the renormalization group equation (4.1) by means of functional differentiation followed by the application of the relationships

$$\langle N \rangle = \overline{\int \int} \langle \langle \eta[\mathbf{x}(t')] \rangle \rangle D[\mathbf{x}(t')] = \overline{\int \int} \frac{\delta \ln G[K=0]}{\delta K[\mathbf{x}(t')]} D[\mathbf{x}(t')], \qquad (4.4)$$

$$\langle \widetilde{N} \rangle = \overline{\int \int} \langle \langle \widetilde{\eta} [\mathbf{x}(t')] \rangle \rangle D[\mathbf{x}(t')] = \overline{\int \int} \frac{\delta \ln \widetilde{G}[K=0]}{\delta K[\mathbf{x}(t')]} D[\mathbf{x}(t')], \qquad (4.5)$$

which can be derived by expanding the characteristic functionals $G[K[\mathbf{x}(t')]]$ and $\widetilde{G}[K[\mathbf{x}(t')]]$ in cumulant series of the type (2.10).

By using Eqs. (2.10), (2.13), and (2.14) applied for the renormalized characteristic functional $\tilde{G}[K[\mathbf{x}(t')]]$ combined with Eqs. (4.1) and (4.2) and using the same steps as in Sec. III we obtain the following expression for the average relaxation function $\langle \ell(t) \rangle$:

$$\langle \mathscr{C}(t) \rangle = H \int_{0}^{1} z^{H-1} \, \mathrm{d}z \, \exp \left\{ \sum_{m=1}^{\infty} \frac{1}{m!} \left[\varepsilon \left(1 + \frac{1}{H} \right) \right]^{m} \underbrace{\int \int}_{u=1}^{\infty} \cdots \underbrace{\int \int}_{u} c_{m} [\mathbf{x}_{1}(t'), \dots, \mathbf{x}_{m}(t')] \xi [\mathbf{x}_{1}(t')] \\ \times D[\mathbf{x}_{1}(t')] \cdots \xi [\mathbf{x}_{m}(t')] D[\mathbf{x}_{m}(t')] \prod_{u=1}^{m} \left\{ V_{\Sigma} \ln \left[1 - z V^{*}(\mathbf{x}_{u}(t))(V_{\Sigma})^{-1} \\ \times \left(1 - \exp \left(- \int_{0}^{t} W(\mathbf{x}_{u}(t')) dt' \right) \right) \right] \right\} \right\},$$

$$(4.6)$$

from which, by taking into account the nonintermittency conditions (3.2) for the nonrenormalized relative fluctuations of the density of channels we obtain the following scaling law in the thermo-dynamic limit (2.22):

$$\langle \ell(t) \rangle \sim \mathcal{J}_{H} \left\{ \underbrace{\int \int}_{0} \rho[W(t')] D[W(t')] \left[1 - \exp\left(-\int_{0}^{t} W(t') dt' \right) \right] \right\},$$

$$\text{as } V_{\Sigma}, \langle \widetilde{N} \rangle \to \infty \quad \text{with } \varepsilon = \langle \widetilde{N} \rangle / V_{\Sigma} = \text{const},$$

$$(4.7)$$

where the function $\mathcal{J}_{H}(z)$ and the functional density of channels involved in the relaxation process, $\rho[W(t')]D[W(t')]$, are given by Eqs. (1.5) and (3.8), respectively.

Equation (4.7) justifies the conjecture (1.12) made without proof in Sec. I. This equation is the dynamical analog of the intermittent scaling law (1.4) derived for systems with static disorder in Ref. 20. Just like in the static case the reciprocal value of the fractal exponent H, 1/H, is a measure of the degree of intermittency of the fluctuations of the number of channels. In particular in the limit $H\rightarrow\infty$ the fluctuations become nonintermittent and Eq. (4.7) reduces to the dynamical analogue (1.2) of Huber's equation. The renormalization group approach for dynamical disorder used in this paper has the same drawback as the similar static approach developed in Ref. 20: it does not guarantee that the limit for intermittent fluctuations. The renormalization group procedure introduced in Ref. 21 does not provide a hint that the fixed point corresponding to Eq. (4.1) is the unique fixed point of the problem. It is possible that further research may lead to other scaling laws characteristic for intermittent fluctuations.

V. DYNAMICAL GENERALIZATIONS OF STRETCHED EXPONENTIAL

The main difficulty related to the application of the dynamical scaling laws (3.7) and (4.7) is connected to the evaluation of the path integral:

$$I(t) = \underbrace{\int \int}_{0} \rho[W(t')] D[W(t')] \left[1 - \exp\left(-\int_{0}^{t} W(t') dt'\right) \right].$$
(5.1)

The evaluation of such path integrals would be trivial provided that the functional density of states $\rho[W(t')]D[W(t')]$ would have a Gaussian behavior. Unfortunately a Gaussian form for $\rho[W(t')]D[t')]$ must be ruled out because it does not include the static power law distribution (1.3') as a particular case.

A formal solution of the problem can be given by introducing an average probability density functional of the relaxation rates W(t'), $t \ge t' \ge 0$:

Vlad et al.: Universal behavior for dynamical disorder

$$\varphi[W(t')]D[W(t')] = \rho[W(t')]D[W(t')]/\langle N^* \rangle, \qquad (5.2)$$

with the normalization condition

$$\int \int \varphi[W(t')]D[W(t')] = 1,$$
(5.3)

and where

$$\langle N^* \rangle = \overline{\int \int} \rho[W(t')] D[W(t')] = \varepsilon \overline{\int \int} V^*(\mathbf{x}(t)) \xi[\mathbf{x}(t')] D[\mathbf{x}(t')]$$
(5.4)

is the average effective number of channels involved in the relaxation process. Generally the average effective number of channels involved in relaxation, $\langle N^* \rangle$, is at most equal to the total average number of channels, $\langle N \rangle$. By using the expression (5.2) for the average probability density functional $\varphi[W(t')]D[W(t')]$, the factor I(t) can be expressed in terms of a dynamical average of the random function

$$\mathscr{E}[W(t')] = \exp\left(-\int_0^t W(t')dt'\right)$$
(5.5)

[see also Eq. (2.17)]. We have

$$I(t) = \langle N^* \rangle \{ 1 - \langle \mathscr{E}[W(t')] \rangle \}, \tag{5.6}$$

where the dynamical average $\langle \mathscr{E}[W(t')] \rangle$ is given by

$$\left\langle \mathscr{E}[W(t')]\right\rangle = \overline{\int \int \varphi[W(t')]D[W(t')]\mathscr{E}[W(t')]}.$$
(5.7)

In this paper we limit ourselves to the simplest case of dynamical disorder for which the random process corresponding to the average probability density functional $\varphi[W(t')]D[W(t')]$ is Markovian. Moreover we consider that the average effective number of channels involved in the relaxation process, $\langle N^* \rangle$, is time independent:

$$\langle N^* \rangle = \text{const.}$$
 (5.8)

Under these circumstances the probability density functional $\varphi[W(t')]D[W(t')]$ can be represented as

$$\varphi[W(t')]D[W(t')] = \lim_{\substack{m \to \infty \\ (\Delta t \to 0)}} [\varphi(W_m; m\Delta t | W_{m-1}; (m-1)\Delta t)dW_m \cdots \varphi(W_2; 2\Delta t | W_1; \Delta t)dW_2 \\ \times \varphi(W_1; \Delta t | W_0; 0)dW_1\varphi_{st}(W_0)dW_0], \quad t \ge t' \ge 0,$$
(5.9)

where

$$m = t/\Delta t; \tag{5.10}$$

$$\varphi_{st}(W)dW$$
 with $\int \varphi_{st}(W)dW = 1$, (5.11)

J. Math. Phys., Vol. 37, No. 5, May 1996

is an average one-time stationary probability density of an individual relaxation rate attached to a given channel corresponding to static disorder and

$$\varphi(W;t|W';t')dW \quad \text{with} \quad \int \varphi(W;t|W';t')dW = 1$$
(5.12)

is the average conditional probability density of the relaxation rate W at time t provided that at time t' the relaxation rate was W'. For a Markov process both $\varphi_{st}(W)$ and $\varphi(W;t|W';t')$ are the solutions of an evolution equation of the type

$$\partial_t \varphi = \mathbb{L} \varphi, \tag{5.13}$$

where L is a linear Markovian evolution operator of the Liouville, Fokker–Planck, or the master type. In this case the probability $\varphi_{st}(W)$ is the stationary solution of Eq. (5.13), whereas the conditional probability density $\varphi(W;t|W';t')$ is the Green's function of the equation (5.13) corresponding to the initial condition

$$\varphi(W;t=t'|W';t') = \delta(W-W').$$
(5.14)

For the above-mentioned Markovian systems there is a general method for computing dynamical path averages of the type (5.7) without the explicit evaluation of a path integral. The method was suggested by Lax in the sixties³³ in connection with certain problems of quantum optics and rediscovered independently by Van Kampen.^{34,35} For a recent application of this technique to the study of a rate process with dynamical disorder, the passage over a fluctuating activation energy barrier, see Ref. 26. The idea is based on the observation that a realization of the function $\mathscr{E}(t) = \mathscr{E}[W(t')]$ given by Eq. (5.5) obeys a stochastic differential equation with a random coefficient:

$$\frac{d\mathscr{U}(t)}{dt} = -W(t)\mathscr{U}(t) \quad \text{with } \mathscr{U}(0) = 1.$$
(5.15)

Since Eq. (5.15) is local in time and the coefficient W(t) is Markovian it follows that the pair of random variables $(W(t), \mathcal{E}(t))$ is also Markovian and the one-time joint probability density,

$$P(W, \mathscr{E}; t) dW d\mathscr{E} \quad \text{with} \quad \int \int P(W, \mathscr{E}; t) dW d\mathscr{E} = 1, \tag{5.16}$$

obeys a compound stochastic Liouville equation,^{33–35}

$$\partial_t P(W, \ell; t) = \partial_\ell \{ W \ell P(W, \ell; t) \} + \mathbb{L} P(W, \ell; t),$$
(5.17)

with the initial condition

$$P(W, \ell; t=0) = \delta(\ell-1)\varphi_{st}(W).$$
(5.18)

The dynamical average $\langle \mathscr{E}[W(t')] \rangle$ can be expressed as an average value corresponding to the joint probability density $P(W,\mathscr{E};t)$:

$$\langle \mathscr{E}[W(t')] \rangle = \int \int \mathscr{E}P(W,\mathscr{E};t) dW d\mathscr{E} = \int F(W,t) dW, \qquad (5.19)$$

where

$$F(W,t) = \int \mathscr{C}P(W,\mathscr{E};t)d\mathscr{E}$$
(5.20)

is a marginal average. By multiplying Eqs. (5.17) and (5.18) by \mathscr{E} and integrating over \mathscr{E} after a partial integration we obtain a closed equation for the marginal average F(W;t):

$$\partial_t F(W;t) + WF(W;t) = \mathbb{L}F(W;t), \qquad (5.21)$$

with the initial condition

$$F(W;t=0) = \varphi_{st}(W).$$
(5.22)

From the above considerations it turns out that for a stationary Markovian average random process the evaluation of the average survival function $\langle \ell(t) \rangle$ reduces to the evaluation of the marginal average F(W,t) by solving the evolution equation (5.21) with the initial condition (5.22) followed by the application of Eqs. (3.7), (4.7), (5.1), (5.6), and (5.19).

For applying the suggested Markovian approach we should come up with a suitable definition of the Markovian evolution operator L. The simplest possible choice would be a Liouville operator of the type suggested in Ref. 26 determined by starting out from the stationary probability density $\varphi_{st}(W)$ corresponding to the static density of states (1.3') attached to a stretched exponential of the type (1.1) and by assuming that the regression of the fluctuations of the relaxation rate is described by a generally time-dependent regression rate $\omega(t)$. Now we notice a minor difficulty related to the self-similar form (1.3') of the static density of states $\rho(W)dW$: due to the infrared divergence of $\rho(W)$ given by Eq. (1.3') at W=0, the average effective number $\langle N^* \rangle$ of channels involved in the relaxation process is infinite:

$$\langle N^* \rangle = \int_0^\infty \rho(W) dW = \int_0^\infty [\Gamma(1-\beta)]^{-1} \beta \Omega^\beta W^{-(1+\beta)} dW = \infty.$$
 (5.23)

Due to the time independence condition (5.8) for $\langle N^* \rangle$, the divergent behavior carries over for systems with dynamical disorder. This divergence is, however, spurious because the corresponding integral expressions for the average survival function $\langle \ell(t) \rangle$ are well behaved. The problem can be solved by introducing an infrared cutoff $W^* \neq 0$ and by passing to the limit $W^* \rightarrow 0$ after performing the computations.

For a cutoff value $W^* \neq 0$, the total effective average number of channels $\langle N^* \rangle$ is finite:

$$\langle N^* \rangle = \int_{W^*}^{\infty} \rho(W) dW = [\Gamma(1-\beta)]^{-1} \left(\frac{\Omega}{W^*}\right)^{\beta}, \qquad (5.24)$$

and the stationary probability density $\varphi_{st}(W) dW$ is given by

$$\varphi_{st}(W)dW = \rho(W)dW/\langle N^* \rangle = \beta(W^*)^{\beta}W^{-(1+\beta)}dW, \qquad (5.25)$$

which obviously fulfills the normalization condition

$$\int_{W^*}^{\infty} \varphi_{st}(W) dW = 1.$$
(5.26)

By following the approach suggested in Ref. 26 we express $\varphi_{st}(W)$ as the normalized solution of a Bloch-like equation

$$\beta \left(\beta \varphi_{st}(W) + \frac{\partial}{\partial W} \left[W \varphi_{st}(W) \right] \right) = 0, \qquad (5.27)$$

J. Math. Phys., Vol. 37, No. 5, May 1996

and suggest a dynamical generalization of Eq. (5.27) depending on the regression frequency $\omega(t)$:

$$\frac{\partial \varphi}{\partial t} = -\beta \omega(t) \left[\beta \varphi + \frac{\partial}{\partial W} (W\varphi) \right] = \mathbb{L}\varphi, \qquad (5.28)$$

where the Liouville operator \mathbb{L} is given by

$$\mathbb{L}... = -\beta\omega(t) \bigg[\beta... + \frac{\partial}{\partial W}(W...)\bigg].$$
(5.29)

All solutions of the Liouville equation (5.28) should be properly defined, that is, they should be non-negative and conserve the normalization to unity at any time

$$\varphi \ge 0; \int_{W^*}^{\infty} \varphi \ dW = 1.$$
(5.30)

By integrating Eq. (5.28) term by term it is easy to check that it conserves the normalization of φ to unity provided that the following boundary condition is fulfilled:

$$\varphi(W = W^*; t) = \beta/W^*; \quad t \ge 0.$$
 (5.31)

Concerning the non-negativity of φ we express any solution of Eq. (5.28) in terms of the Green's function $\varphi(W;tW';t')$, which is the solution of Eq. (5.28) with the initial condition (5.14), and of the initial condition $\varphi(W;t=0)$:

$$\varphi(W;t) = \int_{W^*}^{\infty} \varphi(W;t|W';0) \varphi(W';t=0) dW'.$$
(5.32)

The Green's function $\varphi(W;t|W';0)$ can be easily evaluated by integrating Eq. (5.28) along the characteristics with the initial condition (5.14) applied for t'=0 and with the boundary condition (5.31), resulting in

$$\varphi(W;t|W';0) = h \bigg[W^* \exp\bigg(\beta \int_0^t \omega(t')dt'\bigg) - W \bigg] \bigg(\frac{\beta}{W^*}\bigg) \bigg(\frac{W^*}{W}\bigg)^{1+\beta} + h \bigg[W - W^* \exp\bigg(\beta \int_0^t \omega(t')dt'\bigg) \bigg] \exp\bigg(-\beta^2 \int_0^t \omega(t')dt'\bigg) \\ \times \delta\bigg[W - W' \exp\bigg(\beta \int_0^t \omega(t')dt'\bigg)\bigg], \qquad (5.33)$$

where h(x) is the Heaviside's step function. From Eqs. (5.32) and (5.33) we obtain

$$\varphi(W;t) = h \bigg[W^* \exp\bigg(\beta \int_0^t \omega(t') dt'\bigg) - W \bigg] \bigg(\frac{\beta}{W^*}\bigg) \bigg(\frac{W^*}{W}\bigg)^{1+\beta} + h \bigg[W - W^*\bigg(\beta \int_0^t \omega(t') dt'\bigg) \bigg] \\ \times \exp\bigg(-\beta^2 \int_0^t \omega(t') dt'\bigg) \varphi\bigg(W \exp\bigg(-\beta \int_0^t \omega(t') dt'\bigg); t=0\bigg).$$
(5.34)

Both Eqs. (5.33) and (5.34) conserve the non-negativity and normalization conditions (5.30) provided that the initial probability density $\varphi(W;t=0)$ is non-negative and normalized to unity and is equal to zero for any rate smaller than the cutoff value $W=W^*$:

Vlad et al.: Universal behavior for dynamical disorder

$$\varphi(W;t=0) \ge 0; \quad \int_{W^*}^{\infty} \varphi(W;t=0) dW = 1; \quad \varphi(W < W^*;t=0) = 0.$$
 (5.35)

By applying the above-mentioned Markovian approach it follows that the marginal average F(W,t) is the solution of the partial differential equation

$$\partial_t F(W,t) + WF(W,t) = -\beta \omega(t) [\beta F(W,t) + \partial_W [WF(W,t)]], \qquad (5.36)$$

with the initial condition

$$F(W,t=0) = \beta(W^*)^{-1}(W^*/W)^{1+\beta}.$$
(5.37)

By integrating Eq. (5.36) by means of the method of characteristics we can express the marginal average F(W,t) in terms of an arbitrary function. By determining this arbitrary function from the initial condition (5.37) we obtain

$$F(W,t) = \beta(W^*)^{-1} \left(\frac{W^*}{W}\right)^{1+\beta} \exp\left\{-Wg^{\beta}(t) \int_0^t g^{-\beta}(t')dt'\right\},$$
(5.38)

where g(t), the attenuation factor of the regression of fluctuations of the relaxation rate attached to a given channel, is given by

$$\frac{dg(t)}{dt} = -\omega(t)g(t), \quad g(0) = 1,$$
(5.39)

that is,

$$g(t) = \exp\left(-\int_0^t \omega(t')dt'\right).$$
(5.40)

From Eqs. (5.1), (5.6), (5.19), and (5.38) if follows that the exponent I(t) is equal to

$$I(t) = \frac{\beta \Omega^{\beta}}{\Gamma(1-\beta)} \int_{W^*}^{\infty} \frac{1 - \exp[-Wg^{\beta}(t)\int_{0}^{t} g^{-\beta}(t')dt']}{W^{1+\beta}} \, dW.$$
(5.41)

As expected in the limit $W^* \rightarrow 0$, the exponent I(t) is well behaved and in this limit the integral over W in Eq. (5.41) can be explicitly computed, resulting in

$$I(t) = \left[\Omega g^{\beta}(t) \int_0^t g^{-\beta}(t') dt'\right]^{\beta}.$$
(5.42)

From the above computations it turns out that for the model considered in this section the universal scaling laws (3.7) and (4.7) for dynamical nonintermittent and intermittent fluctuations become

$$\langle \mathscr{C}(t) \rangle = \exp\left\{-\left[\Omega g^{\beta}(t) \int_{0}^{t} g^{-\beta}(t') dt'\right]^{\beta}\right\}$$
(5.43)

and

$$\langle \mathscr{C}(t) \rangle = \mathscr{T}_{H} \left\{ \left[\Omega g^{\beta}(t) \int_{0}^{t} g^{-\beta}(t') dt' \right] \right\},$$
(5.44)

J. Math. Phys., Vol. 37, No. 5, May 1996

Downloaded-18-Nov-2000--to-18.19.0.171.--Redistribution-subject-to-AIP-copyright,-see-http://ojps.aip.org/jmp/jmpcpyrts.html.

respectively. Equations (5.43) and (5.44) are dynamical analogs of the stretched exponential law (1.1) and of its static intermittent generalization (1.8). The concrete form of these two equations depends on the dynamics of the regression of the fluctuations of the relaxation rate expressed by the attenuation function g(t). A comparison between the relaxation behavior corresponding to some important types of dynamical disorder and the relaxation behavior of the similar systems with static disorder is presented in the following section.

VI. STATIC VERSUS DYNAMICAL DISORDER

The dynamical relaxation equations (5.43) and (5.44) include the stretched exponential (1.1) and its static intermittent analogue (1.8) as particular cases corresponding to a regression rate $\omega(t)$ equal to zero for which there is no attenuation of the fluctuations

$$\omega(t) = 0, \quad g(t) = 1,$$
 (6.1)

and an initial fluctuation of the relaxation rate is frozen forever.

In this paper we limit ourselves to the study of only two types of dynamical disorder. The first case corresponds to a fast regression of the fluctuations for which the frequency $\omega(t)$ is constant and the attenuation function g(t) is exponentially decreasing in time:

$$\omega(t) = \omega_0 = \text{const} \text{ and } g(t) = \exp(-\omega_0 t).$$
 (6.2)

The second case corresponds to a self-similar regression process described by slowly decaying functions $\omega(t)$ and g(t) which obey negative power laws of time as $t \ge t_0$:

$$\omega(t) = \alpha/(t+t_0) \sim \alpha/t \quad \text{as } t \gg t_0, \tag{6.3}$$

$$g(t) = [t_0/(t+t_0)]^{\alpha} \sim (t_0/t)^{\alpha} \quad \text{as } t \ge t_0,$$
(6.4)

where $t_0 > 0$ is a possibly very small but, however, different from zero time constant which has been introduced in order to avoid the divergence of the frequency $\omega(t)$ in the limit $t \rightarrow 0$. The relationships between these two cases can be clarified by requiring that as $t \rightarrow 0$ the regression rates $\omega(t)$ have the same values, resulting in

$$\omega(0) = \alpha/t_0 = \omega_0. \tag{6.5}$$

By using Eq. (6.5) the relationship (6.4) for the attenuation function g(t) becomes

$$g(t) = \left[\alpha / (\omega_0 t + \alpha) \right]^{\alpha}. \tag{6.6}$$

For small values of α the function g(t) given by Eq. (6.6) has a long tail of the negative power law type. As the fractal exponent α increases the tail of the attenuation function g(t) is getting shorter and shorter and in the limit $\alpha \rightarrow \infty$ we recover the exponential decay law (6.2).

In order to outline the analogies and differences between the relaxation processes in systems with static and dynamical disorder we compare the static relaxation equations (1.1) and (1.8) for nonintermittent and intermittent fluctuations, respectively, with the dynamical relaxation equations (5.43) and (5.44) applied in the case of the attenuation functions g(t) given by Eqs. (6.2) and (6.4). It is also of interest to compare the probability densities of the relaxation time

$$\psi(t)dt = -\left[\frac{\partial \langle \ell(t) \rangle}{\partial t}\right]dt, \qquad (6.7)$$

the corresponding moments

Vlad et al.: Universal behavior for dynamical disorder

$$\langle t^m \rangle = \int_0^\infty t^m \psi(t) dt = \lim_{t \to \infty} t^m \langle \mathscr{l}(t) \rangle + m \int_0^\infty t^{m-1} \langle \mathscr{l}(t) \rangle dt,$$
(6.8)

and the effective rates of relaxation

$$W_{\rm eff}(t) = \psi(t) / \langle \ell(t) \rangle = -\partial_t \ln \langle \ell(t) \rangle.$$
(6.9)

For computing these functions we express the average relaxation functions for nonintermittent and intermittent fluctuations in terms of the exponent I(t) given by Eq. (5.42):

$$\langle \ell(t) \rangle = \exp[-I(t)], \qquad (6.10)$$

$$\left\langle \ell(t) \right\rangle = \mathcal{J}_{H}[I(t)]. \tag{6.11}$$

The application of Eqs. (1.5), (5.42), and (6.7)–(6.11) leads to the following relationships for the functions $\psi(t)$ and $W_{\text{eff}}(t)$:

$$\psi(t) = q(t)I(t)\exp[-I(t)],$$
 (6.12)

$$W_{\text{eff}}(t) = q(t)I(t), \qquad (6.13)$$

for nonintermittent fluctuations and

$$\psi(t) = q(t)\mathcal{F}_{H}[I(t)]\mathcal{J}_{H}[I(t)], \qquad (6.14)$$

$$W_{\text{eff}}(t) = q(t)\mathscr{F}_{H}[I(t)], \qquad (6.15)$$

for intermittent fluctuations. Here the functions q(t) and $\mathscr{F}_{H}(z)$ are given by

$$q(t) = \frac{d[\ln I(t)]}{dt} = \beta \left\{ g^{-\beta}(t) \left[\int_0^t g^{-\beta}(t') dt' \right]^{-1} - \beta \omega(t) \right\},$$
(6.16)

$$\mathscr{F}_{H}(z) = (H+1) \left\{ 1 + \left[\left(1 + \frac{1}{H} \right)^{H} z^{H} \right] \frac{\exp[-z(1+1/H)]}{\gamma[H+1;z(1+1/H)]} \right\}^{-1}.$$
(6.17)

For applying these equations for systems with nonintermittent or intermittent static disorder and for systems with nonintermittent or intermittent dynamical disorder with exponential or selfsimilar regression we should evaluate the functions I(t) and q(t) corresponding to all these particular cases. After some computations we come to

$$I(t) = (\Omega t)^{\beta}, \quad q(t) = \beta/t, \tag{6.18}$$

for static disorder;

$$I(t) = \left\{ \frac{\Omega[1 - \exp(-\omega_0 \beta t)]}{[\omega_0 \beta]} \right\}^{\beta}, \tag{6.19}$$

$$q(t) = \beta^2 \omega_0 \exp(-\omega_0 \beta t) [1 - \exp(-\omega_0 \beta t)]^{-1}, \qquad (6.20)$$

for nonintermittent and intermittent dynamical disorder with exponential attenuation; and

$$I(t) = \{ \Omega^* [t + t_0 - t_0 [t_0 / (t + t_0)]^{\alpha \beta}] \}^{\beta},$$
(6.21)

$$q(t) = \beta^2 \alpha(t_0)^{-1} [t_0/(t+t_0)]^{\alpha\beta+2} \{1 - [t_0/(t+t_0)]^{\alpha\beta+1}\}^{-1},$$
(6.22)

J. Math. Phys., Vol. 37, No. 5, May 1996

Case	$\langle l(t) \rangle$ for small times	$\langle l(t) \rangle$ for large times	
(1) Nonintermittent static disorder	$\exp[-(\Omega t)^{\beta}]$	$\exp[-(\Omega t)^{\beta}]$	
(2) Nonintermittent dynamical disorder with exponential regression	$ \exp[-(\Omega t)^{\beta}]; \\ \Omega t \leq \Omega/\omega_0 $	$\exp[-(\Omega/\beta\omega_0)^{\beta}] = \text{const};$ $\Omega t \gg \Omega/\omega_0$	
(3) Nonintermittent dynamical disorder with self-similar regression	$ \begin{split} & \exp[-(\Omega t)^{\beta}]; \\ & \Omega t \leq \Omega t_0 \end{split} $	$\exp[-(\Omega^* t)^\beta];$ $\Omega t \gg \Omega t_0$	
(4) Intermittent static disorder	$\exp[-(\Omega t)^{\beta}];$ $\Omega t \ll 1$	$(\Omega t)^{-\beta H} (1+1/H)^{-H} \Gamma (1+H);$ $\Omega t \ge 1$	
(5) Intermittent dynamical disorder with exponential regression	$\exp[-(\Omega t)^{\beta}];$ $\Omega t \leq \Omega/\omega_0; \Omega t \leq 1$	(a) $(\Omega t)^{-\beta H}(1+1/H)^{-H}\Gamma(1+H);$ $\Omega/\omega_0 \gg \Omega t \gg 1$ (b) $(\Omega/\beta\omega_0)^{-\beta H}(1+1/H)^{-H}\Gamma(1+H);$ $\Omega t \gg \Omega/\omega_0 \gg 1$	
(6) Intermittent dynamical disorder with self-similar regression	$ \begin{split} & \exp[-(\Omega t)^{\beta}]; \\ & \Omega t \leq 1; \Omega t \leq \Omega t_0 \end{split} $	(a) $(\Omega t)^{-\beta H} (1 + 1/H)^{-H} \Gamma (1 + H);$ $\Omega t_0 \ge \Omega t \ge 1$ (b) $(\Omega^* t)^{-\beta H} (1 + 1/H)^{-H} \Gamma (1 + H);$ $\Omega t \ge \Omega t_0 \ge 1$	

TABLE I. Limit behavior of the average relaxation function $\langle l(t) \rangle$ for different types of static and dynamical disorder for small and large times.

for nonintermittent and intermittent dynamical disorder with self-similar attenuation. Here

$$\Omega^* = \Omega/(1 + \alpha\beta). \tag{6.23}$$

By combining Eqs. (6.7)–(6.22) we can derive six sets of functions $\langle \ell(t) \rangle$, $\psi(t)$, and $W_{\text{eff}}(t)$ corresponding to nonintermittent and intermittent static disorder, to the two types of nonintermittent dynamical disorder, and to the two types of intermittent dynamical disorder considered in this article. The resulting equations are rather complicated and to save space they are not given here. We present only two tables with the different types of limit behavior of these functions for short and large times, respectively.

Table 1 shows the asymptotic behavior of the average survival function $\langle l(t) \rangle$ in the six cases considered. For nonintermittent fluctuations the dynamical disorder decreases the efficiency of relaxation both for exponential and self-similar attenuation. The effect is much more pronounced for exponential attenuation for which the relaxation function tends towards a positive value different from zero as $t \rightarrow \infty$ and thus the relaxation process is never complete, not even after an infinitely large period of time. For self-similar attenuation this decrease in efficiency is less pronounced. An interesting effect in this case is that both for small and large times the relaxation process is described by stretched exponentials with the same exponent β and different characteristic frequencies, Ω and $\Omega^* = \Omega/(1 + \alpha\beta) < \Omega$, respectively. For large times the decrease of efficiency due to dynamical disorder is displayed by the decrease of the characteristic frequency from Ω to Ω^* . Similar patterns occur in the intermittent cases for which the dynamical disorder also slows down the relaxation process. For exponential attenuation after an initial stretched exponential behavior for large times a self-similar region exists for which the relaxation function is described by a negative power law of time. The self-similar region is eventually followed by a horizontal asymptote of the relaxation function which has a positive residual value even in the limit $t \rightarrow \infty$, a situation which corresponds to incomplete relaxation. Just as in the nonintermittent

Case	$W_{\rm eff}(t)$ for small times	$W_{\text{eff}}(t)$ for large times	$\langle t^m \rangle, m \ge 1$
(1) Nonintermittent static disorder	$\beta \Omega^{\beta} t^{\beta-1}$	$\beta \Omega^{eta} t^{eta-1}$	$\Omega^{-m}\Gamma(1+m/\beta)=$ finite
(2) Nonintermittent dynamical disorder with exponential regression	$\begin{array}{l} \beta\Omega^{\beta}t^{\beta-1};\\ \Omega t {\ll} \Omega/\omega_0 \end{array}$	$\sim 0;$ $\Omega t \gg \Omega/\omega_0$	∞
(3) Nonintermittent dynamical disorder with self-similar regression	$ \begin{array}{l} \beta \Omega^{\beta} t^{\beta-1};\\ \Omega t \leqslant \Omega t_0 \end{array} $	$ \begin{array}{l} \beta(\Omega^*)^{\beta}t^{\beta-1};\\ \Omega t \! \gg \! \Omega t_0 \end{array} $	finite
(4) Intermittent static disorder	$ \begin{array}{l} \beta \Omega^{\beta} t^{\beta-1};\\ \Omega t \ll 1 \end{array} $	H/t; $\Omega t \gg 1$	∞
(5) Intermittent dynmical disorder with exponential regression	$ \begin{array}{l} \beta \Omega^{\beta} t^{\beta-1};\\ \Omega t \!\ll\! \Omega/\omega_0; \Omega t \!\ll\! 1 \end{array} $	(a) H/t ; $\Omega/\omega_0 \gg \Omega t \gg 1$ (b) ~ 0 ; $\Omega t \gg \Omega/\omega_0 \gg 1$	œ
(6) Intermittent dynamical disorder with self-similar regression	$\beta \Omega^{\beta} t^{\beta-1}; \\ \Omega t \ll 1; \Omega t \ll \Omega t_0$	(a) H/t ; $\Omega t_0 \gg \Omega t \gg 1$ (b) H/t ; $\Omega t \gg \Omega t_0 \gg 1$	∞

TABLE II. Limit behavior for small and large times of the effective relaxation rate $W_{\text{eff}}(t)$ and the values of the positive moments $\langle t^m \rangle$, $m \ge 1$, of the relaxation time for different types of static and dynamical disorder.

case for self-similar attenuation, the decrease in efficiency of the relaxation process is less pronounced in comparison with the case of exponential attenuation. For small times a stretched exponential behavior exists with a characteristic frequency

$$\Omega^{**} = \Omega. \tag{6.24}$$

For large times both the intermittent behavior and the regression of fluctuations lead to the slowing down of the relaxation process. The intermittent behavior is dominant leading to a long time tail of the average survival function.

Table II displays the asymptotic values of the effective relaxation rate $W_{\text{eff}}(t)$ for small and large times as well as the values of the positive moments of the relaxation time. The expressions for the effective relaxation rate are consistent with the data presented in Table I for the average survival function. A survival function of the stretched exponential type corresponds to a negative power law function of time for the effective relaxation rate of the type

$$W_{\rm eff} \sim 1/t^{1-\beta}$$
. (6.25)

Similarly a power law tail of the survival function corresponds to an asymptotic hyperbolic time dependence of the effective relaxation rate

$$W_{\rm eff} \sim 1/t$$
 as $t \to \infty$, (6.26)

whereas a positive residual value of the relaxation function for large times, $\langle l(\infty) \rangle > 0$, corresponds to an asymptotic value of the effective relaxation rate equal to zero:

$$W_{\rm eff} \sim 0$$
 as $t \rightarrow \infty$. (6.27)



FIG. 1. (a) The dependence of the average survival function $\langle l(t) \rangle$ on the dimensionless time Ωt and on the relative frequency of regression ω_0 / Ω for nonintermittent dynamical fluctuations with exponential attenuation, β =0.6. (b) The dependence of the average survival function $\langle l(t) \rangle$ on the dimensionless time Ωt and on the attenuation exponent α for nonintermittent dynamical fluctuations with self-similar regression, β =0.6 and Ωt_0 =0.1.

The analysis of the values of the moments of the relaxation time, $\langle t^m \rangle$, is of interest because their divergence may be related to the possible statistical fractal behavior of the probability density $\psi(t)$ of the relaxation time. To save space the asymptotic values of $\psi(t)$ are not given in Tables I and II; however, the asymptotic expressions for $\psi(t)$ can be easily evaluated from these tables by noticing that from Eq. (6.9) we have

$$\psi(t) = \langle l(t) \rangle W_{\text{eff}}(t). \tag{6.28}$$

Special attention is deserved by the investigation of the asymptotic behavior of the probability density $\psi(t)dt$ of the relaxation time in the case when a residual value different from zero exists for the average survival function, a situation which corresponds to incomplete relaxation. In this case the probability density $\psi(t)dt$ of the relaxation time is apparently not normalized to unity because we have

$$\int_{0}^{\infty} \psi(t)dt = -\int_{0}^{\infty} \partial_{t} \langle l(t) \rangle dt = \langle l(0) \rangle - \langle l(\infty) \rangle = 1 - \langle l(\infty) \rangle < 1.$$
(6.29)

The physical explanation of this result is the following: the factor $\langle l(\infty) \rangle$ expresses the proportion of systems (particles) which never relax. Notice, however, that the violation of normalization of the probability density $\psi(t)dt$ is only apparent because the expression (6.7) for $\psi(t)dt$ does not

J. Math. Phys., Vol. 37, No. 5, May 1996



FIG. 2. (a) The dependence of the average relaxation function $\langle l(t) \rangle$ on the dimensionless time Ωt for nonintermittent fluctuations corresponding to a static process (full line), to a dynamical process with exponential attenuation (dashed line), and to a dynamical process with self-similar attenuation (dash-pointed), $\beta=0.6$, $\omega_0/\Omega=3$, $\alpha=0.3$, $\Omega t_0=0.1$. (b) The dependence of the average relaxation function $\langle l(t) \rangle$ on the dimensionless time Ωt for intermittent fluctuations corresponding to a static process (full line), to a dynamical process with exponential attenuation (dashed line), and to a dynamical process with self-similar attenuation (dash-pointed), $\beta=0.6$, $\omega_0/\Omega=3$, $\Omega t_0=0.1$, and H=0.5.

take into account the contribution of systems (particles) which survive up to infinity. These particles give rise to a contribution to the expression for $\psi(t)dt$ having the form of a delta function displaced to infinity. Equation (6.7) should be rewritten

$$\psi(t) = -\partial \langle l(t) \rangle / \partial t + \langle l(\infty) \rangle \delta(t - t^*) \quad \text{with } t^* \to \infty.$$
(6.30)

This definition of the probability density $\psi(t)dt$ leads to a normalized expression even if $\langle l(\infty)\rangle \neq 0$.

The significance of the values of the moments of the relaxation time displayed in Table II is clear. A stretched exponential relaxation function is relatively fast decreasing and the resulting shape of the tail of $\psi(t)$ ensures the convergence of the moments, a situation which corresponds to static nonintermittent disorder and to dynamical nonintermittent disorder with self-similar regression. In the other four cases presented in Table II the moments are divergent. There are two different causes for this divergence. For nonintermittent and intermittent dynamical disorder with exponential regression it is due to the existence of a finite proportion of particles which never relax. For intermittent static disorder and for intermittent dynamical disorder with self-similar regression the infinite moments are generated by the self-similar features of the tails of the



FIG. 3. The average relaxation function $\langle l(t) \rangle$ for nonintermittent, self-similar fluctuations (full line) in comparison to the static average relaxation function (dashed line). In the logarithmic coordinates used the stretched exponential portions of the relaxation functions appear as straight lines: $\beta = 0.6$, $\Omega^* \equiv \Omega/(1 + \alpha\beta) = 1 s^{-1}$, and $\Omega^* t_0 = 0.1$ for $\alpha = 0.1$, 1, 10, 100, 1000 (from bottom to top).

probability density $\psi(t)dt$. By using Eq. (6.28) and the data displayed in Tables I and II it is easy to see that in both of these cases the large time behavior of the probability density of the relaxation time is described by

$$\psi(t) \sim t^{-(1+\beta H)} \quad \text{as} \ t \to \infty, \tag{6.31}$$

that is, $\psi(t)$ has a power law tail with a fractal exponent $1+\beta H$. It is interesting that this fractal exponent is independent of the exponent α of attenuation; the proportionality coefficient in Eq. (6.31) is, however, generally α -dependent.

For a better understanding of the behavior of the average relaxation function $\langle l(t) \rangle$ in the different cases investigated in this paper we present some graphs of this function. As expected these graphs are consistent with the results of asymptotic analysis presented in Tables I and II. By examining Fig. 1(a) corresponding to nonintermittent dynamical fluctuations with exponential regression we notice that the fraction of particles which never relax increases with the increase of the frequency of regression ω_0 , a result which is consistent with the asymptotic expression of



FIG. 4. The dependence of the difference $\langle l(t) \rangle - \langle l_{\text{stat}}(t) \rangle$ between the relaxation function for dynamical nonintermittent fluctuations with self-similar attenuation and the relaxation function for static systems in terms of the logarithm of dimensionless time $\ln(\Omega^* t)$, $\beta = 0.6$, $\Omega^* \equiv \Omega / (1 + \alpha \beta) = 1 s^{-1}$, and $\Omega^* t_0 = 0.1$ for $\alpha = 0.1$, 1, 10 (from bottom to top).



FIG. 5. Average relaxation functions for nonintermittent dynamical fluctuations with exponential attenuation (dashed) and self-similar attenuation (dash-pointed) in comparison with the static stretched exponential law (full line). In the logarithmic coordinates used, the stretched exponential portions of the relaxation functions appear as straight lines: β =0.6, ω_0/Ω =3, α =0.3, and Ωt_0 =0.1.

 $\langle l(t) \rangle$ for large time corresponding to this case and presented in Table I. A similar effect can be noticed in Fig. 1(b) corresponding to nonintermittent dynamical disorder with self-similar attenuation. Although in this case the decrease of efficiency of relaxation due to dynamical disorder is less pronounced and eventually as $t \rightarrow \infty$ all particles relax, the graph clearly shows that an increase of the attenuation exponent α slows down the relaxation process. Figures 2(a) and 2(b) show some graphs of the average relaxation function for static disorder, dynamical disorder with exponential and self-similar regression for nonintermittent and intermittent fluctuations, respectively. For the consistency of comparison the parameters ω_0 , α , and t_0 fulfill the relationship (6.5) so that for dynamical disorder the initial frequency of regression is the same in all cases. In Fig. 2 the same pattern is observed in both cases, that is, the exponential regression leads to incomplete relaxation and, for relatively low values of the attenuation exponent, $1 > \alpha \ge 0$, the self-similar attenuation leads to relaxation functions which are very close to the functions corresponding to the static case. Significant differences occur only if the attenuation exponent α is bigger than the unity.

The relative insensitivity with respect to the variations of the attenuation exponent α of the average relaxation function for dynamical disorder with self-similar regression is consistent with



FIG. 6. Average relaxation functions for intermittent dynamical fluctuations with exponential attenuation (dashed) and self-similar attenuation (dash-pointed) in comparison with the intermittent static average relaxation function (full line). In the logarithmic coordinates used, the power law portions of the relaxation functions appear as straight lines: $\beta=0.6$, $\omega_0/\Omega=3$, $\alpha=0.3$, $\Omega t_0=0.1$, and H=0.5.

the ubiquity in nature of the stretched exponential. The occurrence of the stretched exponential relaxation for these types of systems is not limited to small values of the exponent α . From Table I it follows that for nonintermittent self-similar dynamical fluctuations a stretched exponential emerges for large times for any values of the exponent α , small or large. In this case the passage from small to large times corresponds to a crossover from a stretched exponential with characteristic frequency Ω to another stretched exponential with a smaller characteristic frequency $\Omega^*=\Omega/(1+\alpha\beta)<\Omega$. Figure 3 displays this crossover phenomenon for different values of the exponent α from very small to very large. For this graph multiple logarithmic coordinates have been used for which the stretched exponential portions of the average relaxation functions exist both for small and large times for any values of the exponent α , small or large and all these stretched exponentials have the same exponent β . We emphasize that, even though all these stretched exponentials have the same exponent β , they may actually look very different because their characteristic frequencies may vary very much. This fact is clearly illustrated in Fig. 4.

Figure 5 displays the departure of the different relaxation functions from a stretched exponential in the nonintermittent case. Even for small values of the attenuation rates the exponential regression leads to a large time saturation behavior which is very different from the one described by a stretched exponential. In contrast the self-similar regression leads to a behavior close to the one corresponding to a stretched exponential. Similarly in Fig. 6 the departure from the power law relaxation is investigated for intermittent fluctuations. In this case, too, even for small regression rates the exponential regression leads to incomplete relaxation whereas the self-similar attenuation generates an average relaxation function with a long tail which is close to the one corresponding to the static intermittent case.

The comparative analysis presented in this section shows that for the approach developed in Sec. V the dynamical disorder decreases the efficiency of relaxation. For small regression rates the self-similar attenuation of fluctuations leads to relaxation patterns similar to the ones corresponding to the static processes. For exponential attenuation, however, even the slowest regression rate leads to a different behavior corresponding to incomplete relaxation. For self-similar nonintermittent dynamical disorder the stretched exponential relaxation behavior emerges for large times even if the attenuation exponent α is very large; the corresponding stretched exponentials, although characterized by the same fractal exponent β as in the static case, may be very different from the static stretched exponential, because their characteristic frequencies may vary very much.

VII. DISCUSSION

In this section we discuss some physical implications of the approach suggested in Sec. V. The physical interpretation of the method of computing path averages based on Eqs. (5.6)-(5.17)is related to an apparently obscure mathematical problem, the choice of the initial and boundary conditions for the evolution equations (5.21) or (5.36) for the marginal average F(W,t). In order to ensure the normalization to unity of the average probability density $\varphi(W)dW$ of an individual relaxation rate, for solving the evolution equation (5.28) we have used the boundary condition (5.31). This boundary condition expresses the generation of new fluctuations which are then destroyed by the regression process. In contrast, for solving the partial differential evolution equations (5.21) or (5.36) for the marginal average F(W,t) no such similar boundary conditions have been used. This omission of a boundary condition is required by the main characteristics of the type of dynamical disorder investigated in Sec. V. The main assumption of our approach is that the fluctuations are generated at the beginning of the relaxation process and then they regress as the relaxation process is going on. We start out by considering an initial fluctuation with statistical properties described by the probability density $\varphi_{\rm er}(W)dW$ given by Eq. (5.25), and then we follow its regression during the relaxation process. As time increases, due to the regression process, the channels with high relaxation rates lose their reactivity and their rates become smaller and smaller. During the relaxation process the size of a set of channels with high relaxation rates is shrinking; of the average number $\langle N^* \rangle$ of channels involved in the process, more and more have low relaxation rates, resulting in the decrease of efficiency described by the model. No mechanism of transition of a channel from a state characterized by a small relaxation rate to a state with a high relaxation rate is supposed to exist for t>0. Such a mechanism acts only at the beginning of the process, for t=0, when the fluctuations are generated and thus we should impose a boundary condition only for this moment:

$$F(W = W^*, t = 0) = \beta/W^*.$$
(7.1)

Such a condition, however, does not need to be taken explicitly into account in the computation because it is contained in the initial condition (5.37).

The above considerations are closely related to the physical interpretation of Eq. (5.28) for the time evolution of the probability density $\varphi(W)dW$. From the physical point of view Eq. (5.28) is a stochastic Liouville equation which describes the regression of fluctuations only and it would lead to a probability loss $\int \varphi(W)dW < 1$ if the generation of new fluctuations is not taken into account. The introduction of the boundary condition (5.31) compensates the "probability loss" due to the regression process by an "influx of probability fluid" into the system. In contrast, the compound stochastic Liouville equation (5.17), which describes the relaxation process and Eq. (5.36) derived from it, cannot accomodate a boundary condition of the type (5.31). This limitation is due to the Markovian approximation introduced in Sec. V. Within its framework a given feature of the regression process can be modeled only by assuming that the regression frequency $\omega(t)$ is generally time dependent, resulting in a time-inhomogeneous evolution equation for the overall relaxation process for which a boundary condition of the type (5.31) cannot be formulated in a simple way.

We emphasize that this type of pure regression mechanism without generation of new fluctuations for t>0 is the only one which includes the case of the static disorder as a particular case, corresponding to the situation when the rate of regression is equal to zero. If the fluctuations are generated for t>0, the system is characterized by dynamical disorder, even if the regression process is missing. Although, at least in principle, this type of dynamical disorder can also be described by the dynamical Huber law (3.7) or by its intermittent analog (4.7), it is different from the type of dynamical disorder considered in Sec. V. Some preliminary research concerning the generation of fluctuations for t>0 is presented in Ref. 19; it has been shown that, as expected, this type of dynamical disorder leads to an increase in the efficiency of relaxation, because it generates an increase in the number of channels with high relaxation rates. In particular, if the regression process is missing, this type of dynamical disorder leads to a compressed exponential relaxation described by the average survival function

$$\langle l(t) \rangle \sim \exp(-\operatorname{const} t^{1+\beta}); \quad 1 > \beta > 0.$$
 (7.2)

Our analysis has shown that the self-similar regression has the remarkable feature that for small regression rates it does not affect the shape of the average relaxation function, generating only small corrections. Moreover, even for very large regression rates, for large times the process is described by a stretched exponential with the same fractal exponent β as in the static case. These results, which might provide an explanation for the universality of the stretched exponential relaxation law, are consistent with the ideas developed by West^{36,37} concerning the insensitivity of the statistical fractal systems to random perturbations. From the mathematical point of view for the model developed in Sec. V, this insensitivity is due to the slow decrease of the relaxation rates in the case of self-similar regression, especially for large times.

At the end of this section we point out an apparent contradiction between the results reported here and the results presented in Ref. 26. In Ref. 26 an analysis of the passage over a fluctuating

activation energy barrier has been suggested based on a path average technique similar to the one used in Sec. V. Although both models assume the existence of a pure regression mechanism for t>0, the analysis from Ref. 26 shows that the dynamical disorder leads to an increase of the transparency factor of the barrier which apparently contradicts the results reported here. The explanation of this apparent paradox is simple. In Ref. 26 the regression of fluctuations leads to a decrease in the height of the activation energy barrier, that is, to an increase of the speed of relaxation, whereas for our model the regression of fluctuations leads to small rates.

VIII. CONCLUSIONS

In this paper an attempt has been made to construct dynamical analogs of the stretched exponential relaxation. The main idea of the suggested approach is to search for the asymptotic relaxation laws which emerge in the limit of a very large number of relaxation modes. The mathematical structure of the theory is based on a formal functional generalization of the theory of random point processes for which to each random point a random function is attached. In the limit of very large numbers of relaxation modes two universal relaxation laws have been identified corresponding to nonintermittent and intermittent dynamical fluctuations, respectively. An attempt to evaluate the path averages entering the asymptotic relaxation laws has been made for Markovian systems with pure regression. It has been shown that the regression of fluctuations leads to a decrease of the efficiency of the relaxation process. For nonintermittent fluctuations, even for high regression rates. This effect might provide an explanation for the wide applicability of stretched exponential law for describing various relaxation processes with dynamical disorder.

Further research should focus on the evaluation of the path averages for the more general case when there is a competition between the generation and the extinction of fluctuations and on the study of suitable applications. Ideal candidates for the application of the theory are the systems in which a large number of degrees of freedom are involved in the relaxation process, for instance, the protein–ligand interactions,⁷ or the ion channel kinetics,⁹ where the relaxation modes correspond to a large number of molecular conformations.

ACKNOWLEDGMENTS

The subject of this article has been suggested by a collaboration of MOV and MCM with Dr. B. Schönfisch concerning the analogies between the theory of competitive risks in population biology and the relaxation dynamics in systems with static disorder.

This research has been supported by the Alexander von Humboldt Foundation, the Sondersforschungsbereich 239, Project No. 310 of the Deutsche Forschungsgemeinschaft, NATO, and the Natural Sciences and Engineering Research Council of Canada.

- ²G. Williams and D. C. Watts, Trans. Faraday Soc. 66, 80 (1970).
- ³D. R. Cox, *Renewal Theory* (Chapman and Hall, London, 1962).
- ⁴R. V. Chamberlain, G. Mozurkewich, and R. Orbach, Phys. Rev. Lett. 52, 867 (1984).

⁷ A. Plonka, Chem. Phys. Lett. **151**, 446 (1988); A. Ansari, J. Berendzden, D. Braunstein, B. R. Cowen, H. Frauenfelder, M. K. Hong, I. E. T. Iben, J. B. Jhonson, P. Ormos, T. B. Sauke, R. Scholl, A. Schulte, P. J. Steinbach, J. Vittitow, and R. D. Young, Biophys. Chem. **26**, 337 (1987).

¹E. W. Montroll and J. T. Bendler, J. Stat. Phys. 34, 129 (1984) and references therein.

⁵U. Even, K. Rademann, J. Jortner, N. Manor, and R. Reisfeld, Phys. Rev. Lett. 52, 2164 (1984).

⁶T. F. Nonnenmacher, in *Rheological Modeling: Thermodynamical and Statistical Approaches, Lecture Notes on Physics*, Vol. 381, edited by J. Casas-Vasquez and D. Jou (Springer, Berlin 1991), pp. 309–320; H. Schiessel and A. Blumen J. Phys. A **26**, 5057 (1993).

⁸A. Plonka, *Time-Dependent Reactivity of Species in Condensed Media, Lecture Notes in Chemistry*, Vol. 40 (Springer, Berlin, 1986).

⁹L. S. Liebovitch, J. Fischbarg, and J. P. Koniarek, Math. Biosci. **84**, 37 (1987); L. S. Liebovitch, Math. Biosci. **93**, 97 (1989); L. S. Liebovitch and T. L. Tóth, J. Theor. Biol. **148**, 243 (1991); Bull. Math. Biol. **53**, 443 (1991); L. S. Liebovitch, J. Stat. Phys. **70**, 329 (1993); ACS Advances in Chemistry No 235, *Membrane Biochemistry*, edited by M. Blank and I. Vodyanovy (American Chemical Society, Washington, DC, 1994), pp. 357–374.

- ¹⁰P. R. J. Burch, *The Biology of Cancer: A. New Approach* (University Park, Baltimore, MD, 1976); M. C. Mackey and J. G. Milton, J. Math. Biol. 28, 33 (1990).
- ¹¹ P. Lévy, *Théorie de l'Addition des Variables Aléatoires* (Gauthier Villars, Paris, 1937); B. V. Gnedenko and A. N. Kolmogorov, *Limit Distributions for Sums of Independent Random Variables* (Addison–Wesley, Reading, MA, 1954); B. J. West, J. Opt. Soc. Am. A **7**, 1074 (1990); M. F. Shlesinger, Ann. Rev. Phys. Chem. **39**, 269 (1988).
- ¹²T. Förster, Z. Naturforsch. A **4**, 321 (1949).
- ¹³ A. Blumen, J. Klafter, and G. Zumofen, in *Optical Spectroscopy of Glasses*, edited by I. Zschokke (Reidel, Amsterdam, 1986), pp. 199–265 and references therein; A. K. Roy and A. Blumen, Phys. D **38**, 291 (1989); A. Blumen, Nuovo Cimen. B **63**, 50 (1981) and references therein.
- ¹⁴R. G. Palmer, D. Stein, E. S. Abrahams, and P. W. Anderson, Phys. Rev. Lett. 53, 958 (1984).
- ¹⁵M. F. Shlesinger and E. W. Montroll, Proc. Natl. Acad. Sci. USA 81, 1280 (1984).
- ¹⁶J. Klafter and M. F. Shlesinger, Proc. Natl. Acad. Sci. USA 83, 848 (1986).
- ¹⁷W. G. Glöckle and T. F. Nonnenmacher, Macromolecules **24**, 6426 (1991); J. Stat. Phys. **71**, 755 (1993); in *Fractals in Biology and Medicine*, edited by T. F. Nonnenmacher, G. A. Losa, and E. R. Weibel (Birkhäuser, Basel, 1994), pp. 22–37; R. Metzler and T. F. Nonnenmacher, J. Chem. Phys. **103**, 7180 (1995).
- ¹⁸D. L. Huber, Phys. Rev. B **31**, 6070 (1985).
- ¹⁹ M. O. Vlad and M. C. Mackey, J. Math. Phys. **36**, 1834 (1995); M. O. Vlad, J. Ross, and M. C. Mackey, J. Math. Phys. **37**, 803 (1996).
- ²⁰M. O. Vlad, B. Schönfisch, and M. C. Mackey, Phys. Rev. E (to be published, 1996).
- ²¹M. O. Vlad, J. Math. Phys. **35**, 796 (1994).
- ²²R. Zwanzig, Acc. Chem. Res. 23, 148 (1990); J. Chem. Phys. 97, 3587 (1992).
- ²³S. Dattagupta, *Relaxation Phenomena in Condensed Matter Physics* (Academic, Orlando, FL, 1987) and references therein.
- ²⁴ R. Kubo, in *Fluctuation, Relaxation and Resonance in Magnetic Systems*, edited by D. Ter. Haar (Olivier and Boyd, Edinburgh, 1962), pp. 23–68; Adv. Chem. Phys. **15**, 101 (1969); R. Lenk, *Brownian Motion and Spin Relaxation* (Elsevier, Amsterdam, 1977).
- ²⁵ R. Czech and K. W. Kehr, Phys. Rev. B **34**, 261 (1986); R. Mazo and C. Van den Broeck, Phys. Rev. A **34**, 2364 (1986); A. G. Kofman, R. Zaibel, A. M. Levine, and Y. Prior, Phys. Rev. A **41**, 6434, 6454 (1990); W. G. Rotschild, M. Perrot, and F. Guillaume, J. Chem. Phys. **87**, 7293 (1987); M. O. Vlad, Int. J. Mod. Phys. B **7**, 2539 (1993).
- ²⁶M. O. Vlad and M. C. Mackey, Phys. Lett. A 203, 292 (1995).
- ²⁷J. Wang and P. Wolynes, Chem. Phys. Lett. **212**, 427 (1993); Chem. Phys. **180**, 141 (1994).
- ²⁸N. G. Van Kampen, *Stochastic Processes in Physics and Chemistry*, 2nd ed. (North-Holland, Amsterdam, 1992), pp. 30–51; R. L. Stratonovich, *Topics in the Theory of Random Noise* (Gordon and Breach, New York, 1963), Vol. 1, Chap. 6; M. O. Vlad, M. C. Mackey, and J. Ross, Phys. Rev. E **50**, 798 (1994); M. O. Vlad and M. C. Mackey, Phys. Rev. E **51**, 3104 (1995).
- ²⁹J. L. Klimontovich, *Kinetic Theory of Nonideal Gases and Nonideal Plasmas* (Pergamon, Oxford 1982).
- ³⁰C. Beck and F. Schlögl, *Thermodynamics of Chaotic Systems* (Cambridge U.P., Cambridge, 1993).
- ³¹M. F. Shlesinger and B. D. Hughes, Phys. A **109**, 597 (1981).
- ³²M. O. Vlad and B. Schönfisch, submitted for publication (1995).
- ³³M. Lax, Rev. Mod. Phys. 38, 359 (1966).
- ³⁴N. G. Van Kampen, Phys. Lett. A 76, 104 (1980).
- ³⁵N. G. Van Kampen, Phys. Rep. C 24, 171 (1976).
- ³⁶B. J. West, Int. J. Mod. Phys. B 4, 1629 (1990); Ann. Biomed. Eng. 18, 135 (1990); M. O. Vlad, Phys. A 184, 290 (1992).
- ³⁷B. J. West and W. Deering Phys. Rep. 246, 1 (1994).