Non-Markovian stochastic Liouville equation and its Markovian representation: Extensions of the continuous-time random-walk approach

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Some specific features and extensions of the continuous-time random-walk (CTRW) approach are analyzed in detail within the Markovian representation (MR) and CTRW-based non-Markovian stochastic Liouville equation (SLE). In the MR, CTRW processes are represented by multidimensional Markovian ones. In this representation the probability density function (PDF) $W(t)$ of fluctuation renewals is associated with that of reoccurrences in a certain jump state of some Markovian controlling process. Within the MR the non-Markovian SLE, which describes the effect of CTRW-like noise on the relaxation of dynamic and stochastic systems, is generalized to take into account the influence of relaxing systems on the statistical properties of noise. Some applications of the generalized non-Markovian SLE are discussed. In particular, it is applied to study two modifications of the CTRW approach. One of them considers cascaded CTRWs in which the controlling process is actually a CTRW-like one controlled by another CTRW process, controlled in turn by a third one, etc. Within the MR a simple expression for the PDF $W(t)$ of the total controlling process is obtained in terms of Markovian variants of controlling PDFs in the cascade. The expression is shown to be especially simple and instructive in the case of anomalous processes determined by the long-time tailed $W(t)$. The cascaded CTRWs can model the effect of the complexity of a system on the relaxation kinetics (in glasses, fractals, branching media, ultrametric structures, etc.). Another CTRW modification describes the kinetics of processes governed by fluctuating $W(t)$. Within the MR the problem is analyzed in a general form without restrictive assumptions on the correlations of PDFs of consecutive renewals. The analysis shows that fluctuations of $W(t)$ can strongly affect the kinetics of the process. Possible manifestations of this effect are discussed.

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I. INTRODUCTION

Relaxation and transport phenomena in condensed media are of great importance for the understanding of various processes in physics, chemistry, biology, etc. [1–3]. For a description of these phenomena a large number of approaches have been proposed in the literature [1,2,4,6]. The most popular are Markovian approaches based on Langevin and Fokker-Planck [1,2] equations assuming a negligibly short memory in the processes under study. A great advantage of these approaches consists in the possibility of a relatively simple treatment of relaxation in dynamical and (Markovian) stochastic systems induced by interactions whose fluctuations are Markovian processes (MPs). This type of relaxation, which in what follows for brevity is called MP-affected MPs, is known to be described by the stochastic Liouville equation (SLE) [5].

Recent investigations show, however, that conventional Markovian theories do not properly describe the kinetics of relaxations and fluctuations of various observables in many nonequilibrium processes [6,7]. The non-Markovian relaxation kinetics in these processes results from strong memory effects. Moreover, in a number of systems the kinetics is anomalously long-time tailed which implies long-time memory in them. The anomaly manifests itself in some interesting phenomena such as nonergodicity [7–9], aging [10–12], slow relaxation [7,13–15], etc.

The anomalous specific features of relaxation in highly nonequilibrium systems have attracted the close attention of an increasing number of scientists [7,13,14]. An analysis of these features can be made within different models and approximations. One of the most popular and, probably, adequate is the continuous-time random-walk (CTRW) approach in which the process is represented by jumplike fluctuations (or relaxation) treated as a sequence of renewals [4,7,16] characterized by the probability density function (PDF) $W(t)$ of the waiting times of renewals [this PDF is often denoted as $\psi(t)$ [7,16,17]].

During the last years different variants have been analyzed and applied to treating a large number of processes [7,13,18,19]. In some recent studies special attention has been paid to the above-mentioned anomalously long-time tailed relaxation [7,14]: within the CTRW approach such behavior is assumed to result from that of the PDF $W(t) \sim 1/t^{1+\alpha}$ with $\alpha < 1$ [7,20].

It is worth pointing out that, despite the great success of the CTRW models, in the majority of most interesting applications their validity is not quite clear. Typically, it is justified by the use of intuitive arguments concerning the statistical properties of the systems under study. Furthermore, some recent investigations show that, the conventional CTRW approach proves to be not quite accurate in the description of the kinetics of a number of non-Markovian processes [21,12]. In such a case any further extensions of this approach are of course desirable.

Additional problems also arise in CTRW treatment of multistate processes for which one has to introduce the PDF matrix $\hat{W}(t)$ in the corresponding space. The commutation rules for $\hat{W}(t)$ and other matrices describing evolution of systems are usually not evident and cannot easily be understood with conventional probabilistic arguments [22], so that a more rigorous analysis is needed.

Certain modifications of the CTRW approach have already been discussed in the literature. In some CTRW variants the nonhomogeneity of the process, consisting in the dependence of the jumplike fluctuations on the fluctuation number, have been taken into account [23,24]. In other variants modification of simple CTRW time sequences of renewals is proposed [25,26]. These modifications are very interesting and essentially clarify specific features of CTRW-like processes although they still imply the conventional probabilistic description of the problems under study.

Recently another method of analysis and extension of the CTRW models has been proposed, which is based on the multistate Markovian approach [23,27,28]. The idea of the method lies in the treatment of non-Markovian CTRW processes as MPs with fluctuating parameters, whose fluctuations are described by other (controlling) MPs—i.e., as MP-affected MPs mentioned above. In so doing one finds an unambiguous representation of CTRW processes in terms of some multstate MPs [23], referred to as Markovian representation [23,27,28], in which the statistics of renewals is associated with that of reoccurrences in certain transition states during the controlling MPs. These multistate MPs are described by the corresponding equations (termed above SLEs).

The proposed Markovian approach and representation are shown to be very useful for the analysis of non-Markovian CTRW-like models [23,27,28]. In particular, they permit a rigorous derivation of the non-Markovian SLE describing relaxation in dynamical and Markovian stochastic systems induced by CTRW-type fluctuating interactions with a thermal bath [23,27,28] (this relaxation is an important example of the above-mentioned multistate processes). In what follows, for brevity, MPs in which relaxation is induced by CTRW-fluctuating interactions will be called CTRW-affected MPs.

In this work we discuss a compact formulation of the Markovian representation which simplifies and generalizes the derivation of the CTRW-based non-Markovian SLE, thus allowing for an extension of the SLE to treat properly the effects of different noncommuting stochastic interactions and, in particular, to take into account the influence of relaxing systems on the effective statistical properties of fluctuations. This formulation clearly demonstrates that CTRW processes can be considered as MP-affected MPs (in the above-proposed brief terminology) and basic equations of the CTRW approach are, in fact, SLEs.

An important goal of this paper is to exhibit potentialities of the proposed multistate Markovian approach and generalized non-Markovian SLE, as applied to analyzing CTRW-type approaches and to describing a large number of realistic processes. In so doing we, first, have studied the validity of CTRW approaches by means of the generalized SLE. It is shown that under fairly general assumptions on the model of fluctuations, the long-time behavior of the kinetics of the above-mentioned anomalous processes is correctly treated by the CTRW approach. Second, some examples of realistic processes have discussed, which cannot be described by the previously proposed non-Markovian SLE [23], and one needs to apply the generalized variant of the SLE. Third, two important extended modifications of the CTRW approach have been proposed and discussed in detail.

(i) The first modification describes the effect of cascaded controlling processes in which the process determining the statistics of renewals is assumed to be a CTRW-like one controlled by the second CTRW process, which in turn is controlled by the third CTRW processes, etc. In the cascaded control model the compact representation for the PDF $W(t)$ [actually for the Laplace transform $\widetilde{W}(\epsilon)$] is obtained in terms of PDFs of controlling processes $W_j(t)$ at all cascade steps j, found assuming these processes to be Markovian (i.e., neglecting control). This model is very suitable for the analysis of non-Markovian relaxation kinetics in structured and disordered systems [13], random walk in ultrametric spaces [29,30], etc.

(ii) The second modification treats CTRW-like processes governed by fluctuating PDFs $W(t)$. In this type of process fluctuations of $W(t)$ are assumed to result from the additional effect of the nonequilibrium medium and are modeled by the dependence of the system on the Markovian stochastic variable of a special type. Note that, in general, the proposed model implies some correlations of PDFs $W(t)$ of successive renewals of the process whose description is outside the possibilities of the conventional CTRW approach. In this model and within the Markovian representation the processes are reduced to CTRW-type ones, but in the space of larger dimensionality, thus reducing the problem to solving the non-Markovian SLE. The PDF fluctuations are shown to strongly change the kinetics of CTRW relaxation. An especially significant effect is predicted in the case of anomalous long-time tailed processes. The results of this analysis appear to be very helpful for studying reactions in the presence of fluctuating reactivity (called gating [31,32]).

II. GENERAL FORMULATION

We consider relaxation processes in a dynamical or stochastic Markovian system induced by a fluctuating interaction with a classical thermal bath. The Markovian evolution of the system is assumed to be governed by the fluctuating Liouville operator $L(t)$. Our general formulation is applicable to both quantum and classical systems. In particular, in the dynamic systems the only difference of these two cases is in the form of the operators L: for a quantum system $L=i[H,\ldots]$ (here $[H,\ldots]$ is the commutator with the Hamiltonian H of the system in frequency units in which $\hbar=1$), while for a classical system $L=\{H,\ldots\}$ (with $\{H,\ldots\}$ being classical Poisson brackets). In what follows, for definiteness and brevity, we will concentrate on general results as applied to classical systems.

Fluctuations of $L(t)$ are assumed to result from the dependence on the fluctuating bath coordinate $\mathbf{x}$, $L(t)\equiv L_{\mathbf{x}(t)}$, whose changes are modeled by stochastic jumps between states $|\nu\rangle\equiv|\mathbf{x}_\nu\rangle$ with different $L=L_\nu$ in multidimensional space $\{x\}$ which is assumed to be discrete [the continuum case can considered as a corresponding limit of the discrete variant (see Sec. III B 1)]. The adjoint vectors $\langle\mathbf{x}_\nu|$, which determine the projection on the states, are defined by the relations $\langle\mathbf{x}_{\nu'}|\mathbf{x}_\nu\rangle=\delta_{\nu'\nu}$. In accordance with the general rules of linear algebra the scalar product of two vectors $|\mathbf{x}_a\rangle=\Sigma_\nu p_\nu^a|\mathbf{x}_\nu\rangle$ and $\langle\mathbf{x}_b|=\Sigma_\nu \bar{p}_\nu^b\langle\mathbf{x}_\nu|$ is defined as $\langle\mathbf{x}_b|\mathbf{x}_a\rangle=\Sigma_\nu \bar{p}_\nu^b p_\nu^a$.

Hereafter we will use "bra-ket" notation for the states in $\{x\}$ space suitable for treating relaxation phenomena determined by not self-adjoint evolution operators.

In the model under study the system evolution is described by the probability distribution $\rho(t)$ (or density matrix for quantum systems) which satisfies the linear equation

$$\dot{\rho} = -\hat{L}\rho, \tag{2.1}$$

with

$$\hat{L} \equiv \hat{L}_{\mathbf{x}} := \sum\nolimits_{\nu} |\mathbf{x}_{\nu}\rangle L_{\nu} \langle \mathbf{x}_{\nu}|.$$

For simplicity of presentation, $\hat{L}$ is considered to be diagonal in $\{x\}$ space although the formulas obtained are valid in the case nondiagonal $\hat{L}$ as well.

The operator solution of Eq. (2.1) is represented as

$$\rho(t) = \hat{U}(t)\rho_0, \tag{2.2}$$

with

$$\hat{U}(t) := \hat{T}\left[\exp\left(-\int_0^t d\tau\, \hat{L}(\tau)\right)\right],$$

where $\hat{T}$ is the time-ordering operator and $\rho_0 = \rho(t=0)$ is the initial condition.

Experimentally measured observables are usually described by the evolution operator $\hat{U}(t)$ averaged over stochastic fluctuations of $\mathbf{x}(t)$ which is expressed in terms of the conditional evolution operator $\hat{\mathbb{G}}(x, x'|t)$ as

$$\langle \hat{U}(t) \rangle_{\mathbf{x}} = \sum_{\mathbf{x},\mathbf{x}_0} \hat{\mathbb{G}}(\mathbf{x}, \mathbf{x}_0|t)\sigma_i(\mathbf{x}_0) \equiv \langle \mathbf{x}_e|\hat{\mathbb{G}}|\mathbf{x}_i\rangle, \tag{2.3}$$

where $\sigma_i(\mathbf{x}) \equiv |\mathbf{x}_i\rangle = \Sigma_{\nu}\sigma_{i_{\nu}}|\mathbf{x}_{\nu}\rangle$ is the initial probability distribution of the system in $\{x\}$ space normalized by the condition $\Sigma_{\mathbf{x}}\sigma_i(\mathbf{x}) = 1$. In Eq. (2.3) we have also applied the adjoint (equilibrium state) vector which in bra-ket notation is represented as [see below Eq. (3.9)] $\langle \mathbf{x}_e| = \Sigma_{\nu}\langle \mathbf{x}_{\nu}|$ and can be introduced even in the absence of an equilibrium state.

In accordance with Eq. (2.3) the problem reduces to evaluating the operator $\hat{\mathbb{G}}(\mathbf{x}, \mathbf{x}_i|t)$. Unfortunately this can be done relatively easily only for very few models of stochastic $\mathbf{x}(t)$ fluctuations. In this work we will analyze some of those models based on the CTRW approach.

III. MODELS OF FLUCTUATIONS

A. Markovian models

The Markovian approach is based on the assumption that $\hat{L}(t)$ fluctuations are described by the stochastic MPs in $\{x\}$ space and $\hat{L}(t)$ evolution is represented by the probability distribution $\sigma(\mathbf{x}, \mathbf{x}_0|t)$ satisfying the equation

$$\dot{\sigma} = -\hat{\mathcal{L}}\sigma, \tag{3.1}$$

with

$$\sigma(\mathbf{x}, \mathbf{x}_0|0) = \delta_{\mathbf{x},\mathbf{x}_0},$$

where $\hat{\mathcal{L}} \equiv \hat{\mathcal{L}}_{\mathbf{x}}$ is some linear operator in $\{x\}$ space. The principal simplification of the problem results from the fact that in the Markovian approach (3.1)—i.e., in the case of MP-affected MPs—$\hat{\mathbb{G}}(\mathbf{x}, \mathbf{x}_0|t)$ obeys the SLE [5]

$$\dot{\hat{\mathbb{G}}} = -(\hat{L} + \hat{\mathcal{L}})\hat{\mathbb{G}}, \tag{3.2}$$

so that

$$\hat{\mathbb{G}}(t) = e^{-(\hat{L}+\hat{\mathcal{L}})t},$$

which yields for $\langle \hat{\tilde{U}} \rangle = \int_0^\infty dt \langle \hat{U}(t) \rangle \exp(-\epsilon t)$

$$\langle \hat{\tilde{U}} \rangle = \langle \hat{\tilde{\mathbb{G}}} \rangle_{\mathbf{x}} = \langle \mathbf{x}_e|(\epsilon + \hat{L} + \hat{\mathcal{L}})^{-1}|\mathbf{x}_i\rangle. \tag{3.3}$$

In the form (3.2) the SLE is valid for any dependence $\hat{L}_{\mathbf{x}}$ on the coordinate $\mathbf{x}$ although, in general, it is still very complicated for either a numerical or analytical analysis.

Significant simplification can be gained within some special models, for example, in the CTRW approach.

B. CTRW-based models

Non-Markovian $\hat{L}(t)$ fluctuations can conveniently be described by the CTRW approach [4,7,17]. It treats fluctuations as a sequence of sudden, jumplike changes of $\hat{L}_{\mathbf{x}}$ resulting from the changes of $\mathbf{x}$. In the simplest variant of the CTRW (more complicated variants are discussed below) the onset of any particular sudden change of number β is described by the probability $P_{\beta-1}(t)$ (in $\{x\}$ space) not to have any change during time t and its derivative $W_{\beta-1}(t) = -\dot{P}_{\beta-1}(t)$—i.e., the PDF for times of waiting for the change. These functions are independent of the change number β for $\beta > 1$ and for $\beta = 1$ depend on the problem considered [4,7]:

$$W_{\beta\geq 1}(t) = W_n(t), \quad W_0(t) = W_i(t), \tag{3.4}$$

and $P_j(t) = \int_0^t d\tau\, W_j(\tau)$ $(j = n, i)$. In what follows we will mainly discuss the nonstationary CTRW variant in which $W_{\beta\geq 0}(t) = W_n(t)$. Note that in the stationary variant

$$W_i(t) = \tau_e^{-1}\int_t^\infty d\tau\, W_n(\tau), \tag{3.5}$$

where $\tau_e = \int_0^\infty dt\, tW(t)$ is the average time of waiting for the change [4,7]

The Laplace transforms of $W_j(t)$ and $P_j(t)$ $(j = n, i)$ are related by the simple equation $\tilde{P}_j(\epsilon) = [1 - \tilde{W}_j(\epsilon)]/\epsilon$ with

$$\tilde{W}_j(\epsilon) = [1 + \Phi_j(\epsilon)]^{-1} \quad (j = n, i). \tag{3.6}$$

In Eq. (3.6), $\Phi_j(\epsilon)$ is the important auxiliary function [23].

The stochastic changes of $\mathbf{x}$ are also controlled by the matrix $\hat{P}_{\mathbf{x}}$ of the probabilities of jumps in $\mathbf{x}$ space, which is discussed in detail below (in Sec. III B 1).

1. Markovian representation

The results of our earlier studies [23,27] show that important non-Markovian generalizations of the SLE (3.2) can be obtained by assuming the operator $\hat{\mathcal{L}}(t)$ to be a stochastic function of time.

In this work we will analyze a quite natural generalization of the Markovian SLE (3.2) based on the assumption that fluctuations of $\hat{\mathcal{L}}(t)$ are Markovian. In other words $\hat{\mathcal{L}}(t) \equiv \hat{\mathcal{L}}_{\mathbf{z}(t)}$ is assumed to be a function of the Markovian stochastic (controlling) variable $\mathbf{z}(t)$ which, in general, is a vector, so that generalizing the solution (3.2) of the Markovian SLE one can write

$$\hat{\mathbb{G}}(t) = \hat{T}\left\{ \exp\left[-\int_0^t d\tau (\hat{L}_{\mathbf{x}(\tau)} + \hat{\mathcal{L}}_{\mathbf{z}(\tau)}) \right] \right\}. \tag{3.7}$$

In the major part of the analysis we will assume that both $\{x\}$ and $\{z\}$ spaces are discrete and use bra-ket notation with Greek and Latin characters for states in these two spaces—for example, $|\mathbf{x}_\nu\rangle$ and $|\mathbf{z}_j\rangle$, respectively (although, in cases when needed, continuous variants of $\{x\}$ and $\{z\}$ spaces will also be considered).

We start our analysis with the simple Markovian stochastic migration process in $\{x\}$ space, which is described by the Kolmogov-Feller jump matrix

$$\hat{\mathcal{L}} := \hat{k}^d_{\mathbf{z}(t)} - \hat{P}_x \hat{k}^n_{\mathbf{z}(t)}, \tag{3.8}$$

with

$$\hat{P}_x = \sum_{\nu \neq \mu} p_{\nu\mu} |\mathbf{x}_\nu\rangle\langle \mathbf{x}_\mu|.$$

In Eq. (3.8), $\hat{k}^d_{\mathbf{z}(t)} \equiv \hat{k}^d[\mathbf{z}(t)]$ and $\hat{k}^n_{\mathbf{z}(t)} \equiv \hat{k}^n[\mathbf{z}(t)]$ are the matrices of z-dependent (i.e., fluctuating) jump rates diagonal and nondiagonal in $\{\mathbf{z}\}$ space, respectively, with $\hat{k}^n_{\mathbf{z}_j\mathbf{z}_{j'}} = 0$ for $j \neq j'$ and $\hat{k}^d_{\mathbf{z}_j\mathbf{z}_j} = \Sigma_{j \neq j'} \hat{k}^n_{\mathbf{z}_{j'}\mathbf{z}_j}$. Both matrices are assumed to be diagonal in $\{x\}$ space. The matrix $\hat{P}_x$ determines the distribution of jumps in $\{x\}$ space, whose elements $p_{\nu\mu}$ are the probabilities of jumps normalized by the relation $\Sigma_\nu p_{\nu\mu} = 1$. For simplicity, we assume that $\hat{P}_x$ is independent of z, although most general results obtained in this section are valid in the case of z-dependent $\hat{P}_x$ as well (see below).

The matrix $\hat{\mathcal{L}}(t)$ describes relaxation in $\{x\}$ space. This relaxation reduces to equilibration in the presence of the equilibrium state

$$|\mathbf{x}_e\rangle := \sum_\nu p^e_\nu |\mathbf{x}_\nu\rangle, \tag{3.9}$$

with

$$\langle \mathbf{x}_e| := \sum_\nu \langle \mathbf{x}_\nu|,$$

for which $(1-\hat{P}_x)\hat{k}|\mathbf{x}_e\rangle = 0$ and $\langle \mathbf{x}_e|(1-\hat{P}_x)\hat{k} = 0$. This state is assumed to be independent of $\mathbf{z}$.

It is important to note that the theory developed is quite applicable in the absence of the equilibrium state $|\mathbf{x}_e\rangle$—for example, when $\hat{\mathcal{L}}$ describes diffusive migration in infinite space. As for the adjoint vector $\langle \mathbf{x}_e|$ defined in Eq. (3.9) it can still be introduced in this case. It satisfies the relation $\langle \mathbf{x}_e|(1-\hat{P}_x)\hat{k} = 0$ which means nothing else but the conservation of population in the process of migration in $\{x\}$ space.

Within the considered Markovian approximation for $\mathcal{L}_{\mathbf{z}(t)}$ fluctuations the evolution of the system in controlling $\{z\}$ space is governed by the probability distribution $\varphi(\mathbf{z}, \mathbf{z}_0|t)$ satisfying the equation

$$\dot{\varphi} = -\hat{\Lambda}\varphi, \tag{3.10}$$

with

$$\varphi(\mathbf{z}|0) = |\varphi_i(\mathbf{z})\rangle \equiv |\mathbf{z}_i\rangle,$$

in which $\hat{\Lambda} \equiv \hat{\Lambda}_\mathbf{z}$ is some linear operator describing relaxation in $\{z\}$ space and $\varphi_i(\mathbf{z}) \equiv |\mathbf{z}_i\rangle$ is the initial state of the controlling process (in this $\{z\}$ space) in the bra-ket representation. Similar to the process in $\{x\}$ space, considered above, in the presence of the equilibrium state of the operator $\hat{\Lambda}$,

$$|\mathbf{z}_e\rangle := \sum_j p^{\{\mathbf{z}\}}_j |\mathbf{z}_j\rangle, \quad \langle \mathbf{z}_e| := \sum_j \langle \mathbf{z}_j|, \tag{3.11}$$

in which $p^{\{\mathbf{z}\}}_j$ are the corresponding equilibrium population probabilities for states in $\{\mathbf{z}\}$ space, relaxation reduces to equilibration. As in the case of $\{x\}$ space, the vector $\langle \mathbf{z}_e|$ is still meaningful even in the absence of the equilibrium state of the operator $\hat{\Lambda}$.

The operator $\hat{\Lambda}$ can, in general, depend on $\mathbf{x}$. Moreover, $\hat{\Lambda}$ can be a nondiagonal matrix in $\{x\}$ space although, for simplicity of further discussion, we will assume $\hat{\Lambda}$ to be diagonal in this space.

In our analysis, in the case of need we will use the simple and flexible continuous Smoluchowski model for the controlling process in $\{z\}$ space in which

$$\hat{\Lambda} = -D_z z^{1-n_z} e^{-u(z)} \nabla_z [z^{n_z-1} e^{u(z)} \nabla_z], \tag{3.12}$$

where $z = |\mathbf{z}|, u(z)$ is the effective potential and $\nabla_z = \partial/\partial z$ is the radial part of the operator describing diffusion in n_z-dimensional $\{z\}$ space with the diffusion coefficient D_z. This model is quite sufficient for our qualitative and semiquantitative analysis. The correspondence between the continuous model implying smooth functions D_z and $u(z)$ and its discrete variant considered above is formulated as follows: $\mathbf{z} \leftrightarrow b|\mathbf{z}_j\rangle$, where b is the spacing in the discrete model. Naturally, in this limit the scalar product of two vectors $|\mathbf{z}_a\rangle = \Sigma_j p^{\{\mathbf{z}\}}_{a_j} |\mathbf{z}_j\rangle$ and $\langle \mathbf{z}_b| = \Sigma_j \bar{p}^{\{\mathbf{z}\}}_{b_j} \langle \mathbf{z}_j|$ reduces to the properly normalized integral of the product $p^{\{\mathbf{z}\}}_{a_j} \bar{p}^{\{\mathbf{z}\}}_{b_j}$ over $\{z\}$ space (over the continuum variable j).

Similarly to the simplest model discussed in Sec. III A one can easily see that in this approximation the evolution operator $\hat{\mathbb{G}}_\mathbf{r}(\mathbf{r}, \mathbf{r}_i|t)$, which determines $\langle \hat{U}(t)\rangle$, obeys the Markovian SLE in the extended space $\{\mathbf{r}\}$ including the dynamical subspace $\{x\}$ and additional stochastic subspace $\{z\}$, $\{\mathbf{r}\} = \{x \otimes z\}$:

$$\dot{\hat{\mathbb{G}}}_\mathbf{r} = -(\hat{L} + \hat{\mathcal{L}} + \hat{\Lambda})\hat{\mathbb{G}}_\mathbf{r}, \tag{3.13}$$

with

$$\hat{\mathbb{G}}_{\mathbf{r}}(\mathbf{r},\mathbf{r}_i|0) = \delta_{\mathbf{r}\mathbf{r}_i},$$

so that we get for $\hat{\mathbb{G}}$ and the Laplace transform $\tilde{\hat{U}}=\langle\tilde{\hat{\mathbb{G}}}\rangle_x$

$$\hat{\mathbb{G}} = \langle \mathbf{z}_e|\hat{\mathbb{G}}_{\mathbf{r}}|\mathbf{z}_i\rangle, \quad \langle\tilde{\hat{U}}\rangle = \langle(\hat{\Omega} + \hat{\mathcal{L}} + \hat{\Lambda})^{-1}\rangle_{\mathbf{r}}, \tag{3.14}$$

where $\hat{\Omega}=\epsilon+\hat{L}$. In the form (3.14) the SLE is valid for any dependence of coupling $\hat{\mathcal{L}}_z$ on the coordinate z although, in general, it is still very complicated for either a numerical or analytical analysis.

Significant simplification can be gained within some special models for the jump-rate matrix $\hat{k}_z$ [see Eq. (2.3)]. One of the most well known is the CTRW model discussed below in Sec. IV B.

2. Localized jump rates: Non-Markovian SLE

Very important results can be obtained in the model of interaction $\hat{\mathcal{L}}_z$ highly localized in $\{z\}$ space—i.e., highly localized jump rates $\hat{k}_z^d$ and $\hat{k}_z^n$. The most interesting for our further discussion is the simplest variant of this model, which in the case of discrete $\{z\}$ space is written as

$$\hat{k}_{\mathbf{z}}^d = \hat{k}_0\hat{P}_{\mathbf{z}}^{tt}, \quad \hat{k}_{\mathbf{z}}^n = \hat{k}_0\hat{P}_{\mathbf{z}}^{nt}, \tag{3.15}$$

with

$$\hat{P}_{\mathbf{z}}^{jt} := |\mathbf{z}_j\rangle\langle\mathbf{z}_t|$$

and $j=t,n$. Here $\hat{k}_0$ is the matrix diagonal in $\{x\}$ space, while $|\mathbf{z}_n\rangle$ and $|\mathbf{z}_t\rangle$ are the states $\{z\}$ space. The model (3.15) approximates $\mathcal{L}(t)$ fluctuations by the set of jumps (blips) with the relaxation effect of each jump described by the $\hat{k}_0$ matrix. As to the properties of jumps statistics, they are determined by the controlling operator $\hat{\Lambda}$ and the localized matrix $\hat{P}_{\mathbf{z}}^{nt}$ which implies that jumps occur as long as the system appears at the state $|\mathbf{z}_t\rangle$ and each jump results in the transition $|\mathbf{z}_t\rangle \to |\mathbf{z}_n\rangle$ in $\{z\}$ space.

Evidently, for the case of continuum $\{z\}$ space in Eq. (3.15) [with $\hat{\Lambda}$, for example, of the type of that given by Eq. (3.12)] the term $\hat{P}_{\mathbf{z}}^{nt}$ should be replaced by the corresponding δ-function-type one.

Within the model (3.15) the general formula (3.14) can be represented in a more suitable CTRW-like form

$$\begin{aligned}\tilde{\hat{\mathbb{G}}}(\epsilon) &= \langle\mathbf{z}_e|\hat{G}(\epsilon)[1 - \hat{P}_x\hat{k}_0\tilde{\hat{G}}(\epsilon)]^{-1}|\mathbf{z}_i\rangle \\ &= \tilde{\hat{\mathbb{P}}}_i(\epsilon) + \tilde{\hat{\mathbb{P}}}_n(\epsilon)[1 - \tilde{\hat{\mathbb{W}}}_n(\epsilon)]^{-1}\tilde{\hat{\mathbb{W}}}_i(\epsilon), \end{aligned} \tag{3.16}$$

where

$$\tilde{\hat{G}}(\epsilon) := (\epsilon + \hat{L} + \hat{k}_{\mathbf{z}}^d + \hat{\Lambda})^{-1} \tag{3.17}$$

is the Green's function, which essentially determines two matrices: the effective PDF matrix $\tilde{\hat{\mathbb{W}}}_j(\epsilon)$ of sudden fluctuation jumps (analogs of renewals) and the matrix $\tilde{\hat{\mathbb{P}}}_j(\epsilon)$ of probabilities not to observe any fluctuation until time t, playing an important role in CTRW-based theories [4,7,17]] [see Eq. (3.4)]. These two matrices are expressed in terms of the Green's function of the controlling process in $\{z\}$ space:

$$\tilde{\hat{\mathbb{W}}}_j(\epsilon) = \hat{P}_x\tilde{\hat{w}}_j(\epsilon) \quad (j=i,n), \tag{3.18}$$

in which

$$\tilde{\hat{w}}_j(\epsilon) := \hat{k}_0\langle\mathbf{z}_t|\hat{G}(\epsilon)|\mathbf{z}_j\rangle = [1 + \hat{\Phi}_j(\epsilon)]^{-1}, \tag{3.19}$$

where

$$\hat{\Phi}_j(\epsilon) := [\hat{g}_{tt}(\epsilon)\hat{g}_{tj}^{-1}(\epsilon) - 1] + [\hat{k}_0\hat{g}_{tj}(\epsilon)]^{-1} \tag{3.20}$$

with

$$\hat{g}_{tj} := \langle\mathbf{z}_t|\hat{g}(\epsilon)|\mathbf{z}_j\rangle \quad \text{and} \quad \hat{g}(\epsilon) := (\epsilon + \hat{L} + \hat{\Lambda})^{-1} \tag{3.21}$$

and

$$\tilde{\hat{\mathbb{P}}}_j(\epsilon) := \langle\mathbf{z}_e|\hat{G}(\epsilon)|\mathbf{z}_j\rangle = (\epsilon + \hat{L})^{-1}[1 - \tilde{\hat{w}}_j(\epsilon)]. \tag{3.22}$$

In Eq. (3.22), $\langle\mathbf{z}_e|$ is an equilibrium ket vector in $\{z\}$ space (with $\langle\mathbf{z}_e|\Lambda=0\rangle$ defined by a formula similar to Eq. (3.9) but with the $\mathbf{x}$ vectors replaced by those in $\{z\}$ space.

Expression (3.16), which in what follows will be called the generalized non-Markovian SLE (in resolvent form), looks similar to the simpler one derived earlier within the Markovian representation [23]. Moreover, for $\hat{L}=0$ it reduces to that obtained in Refs. [23,33] in which the PDF matrices

$$\tilde{\hat{\mathbb{W}}}_{0j}(\epsilon) \equiv \tilde{\hat{\mathbb{W}}}_j(\hat{L}=0,\epsilon) = \hat{P}_x\tilde{\hat{W}}_j(\epsilon) \quad (j=i,n), \tag{3.23}$$

$$\tilde{\hat{\mathbb{P}}}_{0j}(\epsilon) \equiv \tilde{\hat{\mathbb{P}}}_j(\hat{L}=0,\epsilon) = \epsilon^{-1}[1 - \tilde{\hat{W}}_j(\epsilon)], \tag{3.24}$$

where

$$\tilde{\hat{W}}_j(\epsilon) := \int_0^\infty dt\langle\mathbf{z}_e|\hat{G}_0(t)|\mathbf{z}_j\rangle e^{-\epsilon t} \equiv \langle\mathbf{z}_e|\tilde{\hat{G}}_0(\epsilon)|\mathbf{z}_j\rangle \tag{3.25}$$

is the PDF matrix in which $\hat{G}_0(t)=e^{-(\hat{k}_{\mathbf{z}}^d+\hat{\Lambda})t}$ and $\tilde{\hat{G}}_0(\epsilon) =(\epsilon+\hat{k}_{\mathbf{z}}^d+\hat{\Lambda})^{-1}$ are the Green's functions describing the stochastic process in $\{z\}$ space. Equations (3.23) and (3.24) can be considered as a generalized variant of relation (3.6).

Despite the similarity of the formulas obtained with those known in the CTRW theory [4,7], there are, however, some important specific features of the general expression (3.16) as opposed to the conventional CTRW formulas (3.4)–(3.6), (3.23), and (3.24).

(i) The matrix

$$\tilde{\hat{\mathbb{P}}}_j(\epsilon) = \hat{\Omega}^{-1}(\epsilon)\{1 - \hat{k}_0\langle\mathbf{z}_t|[\hat{\Omega}(\epsilon) + \hat{k}_0 + \hat{\Lambda}]^{-1}|\mathbf{z}_j\rangle\}, \tag{3.26}$$

in which $\hat{\Omega}(\epsilon)=\epsilon+\hat{L}$ does not necessarily commute with $\hat{\Lambda}$, unlike a similar matrix in the conventional CTRW approach (see below). Moreover, in general, elements $\hat{\mathbb{P}}_{j_{ik}}(t)$ of the matrix $\hat{\mathbb{P}}_j(t)$ do not satisfy the evident relation of the type of $\hat{\mathbb{P}}_{j_{ik}}(0)=1$ which would allow one to interpret these elements as probabilities. In addition, $\hat{\mathbb{P}}_j(t)$ can be complex valued, for example, in quantum process for which the elements of $\hat{L}$ are complex.

(ii) The relation between $\hat{\tilde{\mathbb{W}}}_j(\epsilon)$ and $\hat{\tilde{\mathbb{P}}}_j(\epsilon)$ implied by Eqs. (3.18) and (3.22) does not coincide with that between the PDF-matrix of waiting times and the matrix of probabilities known in the CTRW theory. Strictly speaking, the conventional relation [see Eqs. (3.23) and (3.24)] is reproduced only in the evident case $\hat{L}=0$.

(iii) Equation (3.16) is of the form of the CTRW expression, however with the matrices $\hat{\tilde{\mathbb{W}}}_j(\epsilon)$ and $\hat{\tilde{\mathbb{P}}}_j(\epsilon)$ strongly modified by the dynamic operator $\hat{L}$ (in contrast to the non-Markovian SLE appealing to the conventional PDF matrices $\hat{\mathbb{W}}_{0j}(t)$ and $\hat{\mathbb{P}}_{0j}(t)$ [23] [see Eq. (3.27)]).

The above-mentioned effects of dynamic evolution can result in a significant change of the time dependences $\hat{\mathbb{W}}_{0j}(t)$ and $\hat{\mathbb{P}}_{0j}(t)$, thus leading to a strong change of the kinetics of the relaxation processes under study.

3. CTRW-based non-Markovian SLE

The simplified variant of formula (3.16) can be obtained in the special case of the matrix $\hat{\tilde{W}}_j(\epsilon)$ diagonal in $\{x\}$ space, which is realized when the matrices $\hat{\Lambda}$, $\hat{L}$, and $\hat{k}_0$ commute with each other: $[\hat{\Lambda},\hat{L}]=[\hat{L},\hat{k}_0]=0$. In this case one can obtain the representation for $\hat{\tilde{\mathbb{W}}}_j(\epsilon)$ in terms of the Laplace transform of the conventional PDF-matrix of fluctuation blips $\hat{\tilde{\mathbb{W}}}_{0j}$ [see Eqs. (3.23) and (3.25)] [23]:

$$\hat{\tilde{\mathbb{W}}}_j(\epsilon) = \hat{\tilde{\mathbb{W}}}_{0j}(\hat{\Omega}(\epsilon)) := \hat{P}_x \int_0^\infty dt \hat{W}_j(t) e^{-\hat{\Omega}(\epsilon)t}, \qquad (3.27)$$

where $\hat{\Omega}(\epsilon)=\epsilon+\hat{L}$.

The simplified non-Markovian SLE (3.27) is nevertheless more general than the variant of this equation in which $\hat{\tilde{W}}_{0j}(\epsilon)$ is independent of the x coordinate [i.e., $\hat{\tilde{W}}_{0j}(\epsilon)$ proportional to the unity matrix in $\{x\}$ space] [34].

Noteworthy is that, in general, if $\hat{\Lambda}$, $\hat{L}$, and $\hat{k}_0$ do not commute with each other, the representation (3.27) is not valid and one should use the original expression (3.16).

IV. EXTENSIONS OF CTRW MODELS

The proposed Markovian representation, based on a description of CTRWs as MP-affected MPs, enables one to significantly extend the CTRW approach and the non-Markovian SLE. It reduces the treatment of system evolution to an averaging exponential functional over Markovian fluctuations and offers the expression of CTRW equations in terms of a multidimensional Markovian SLE.

There are some straightforward extensions which do not need detailed analysis. For example, an evident variant of extension is the model of several highly localized jump states in $\{z\}$ space, $\langle \mathbf{z}_{t_i}|$ and $|\mathbf{z}_{n_i}\rangle$ $(1\le i\le i_z, i_z>1)$, in which $k_z=\Sigma_j k_0^{(j)}\hat{P}_{nt}^{(j)}$ with $\hat{P}_{nt}^{(j)}=|\mathbf{z}_n\rangle\langle\mathbf{z}_t|$. In this model the general formula (3.14) is also simplified by reducing the problem to solving the linear equation for the matrix $\hat{\mathbf{G}}$ with elements $\hat{\mathbf{G}}_{ij}(\epsilon)=\langle\mathbf{z}_{t_i}|\hat{G}(\epsilon)|\mathbf{z}_{n_j}\rangle$. Unfortunately, in this model the expression for $\hat{\tilde{\mathbb{G}}}$ is fairly cumbersome and not quite suitable for applications.

In this section we will mainly discuss less evident and more general extensions which allow for a description of effects of Markovian and non-Markovian fluctuating interactions on the kinetics of relaxation in CTRW-like systems—i.e. (in our brief terminology), MP- and CTRW-affected CTRWs. In the Markovian representation the problem is still reduced to an analysis of MP-affected MPs though in multidimensional space.

Because of the large number of parameters in this type of multidimensional MPs, it is practically senseless to discuss the problem in general. For this reason, here we will restrict ourselves to consideration of two modifications and extensions of the CTRW approach interesting for theoretical analysis and for applications. For simplicity of notation and presentation, we will put $\hat{L}=0$ although one should keep in mind that all proposed extensions can actually be considered to result from fluctuations of some additional operators—i.e., whose effect is evaluated as if they enter the equations instead of $\hat{L}$. Below, in each particular case this fact will pointed out separately.

A. CTRW control: Coupled Markovian processes

1. General results

So far in our analysis we have discussed the Markovian model for $\hat{\mathcal{L}}(t)$ fluctuations which suggests that the controlling stochastic process $z(t)$, responsible for fluctuations of the jump rates $\hat{k}_{\mathbf{z}(t)}^{d,n}$ [see Eq. (3.8)], is Markovian.

Here we will consider an extension of the Markovian model based on the assumption that the controlling process $z(t)$ is represented as a sequence (cascade) of controlling Markovian processes in multidimensional space $\{\mathbf{Z}_1\}=\{z_1,z_2,\ldots,z_N\}$ with evolution in each space $\{z_{q-1}\}$ being controlled by the process in $\{z_q\}$ space whose mechanism is similar to that responsible for CTRW-type motion in $\{x\}$ space as described above in Sec. III B. This mechanism implies that stochastic jumps in $\{z_q\}$ space happen as long as the system appears in the transition state $|\mathbf{z}_{q_t}\rangle$ (in $\{z_q\}$ space).

As compared to the mechanism presented in Sec. III B, however, here, for simplicity, we will assume that jumps are not accompanied by a change of state in $\{z_{q+1}\}$ space—i.e., the final state $|\mathbf{z}_{q_n}\rangle=|\mathbf{z}_{q_t}\rangle$. For the same reason we will also assume that the initial state $|\mathbf{z}_q^i\rangle$ of evolution in $\{z_q\}$ subspace coincides with the transition state: $|\mathbf{z}_{q_i}\rangle=|\mathbf{z}_{q_t}\rangle$ $(1\le q\le N)$; i.e., we will analyze nonstationary processes (the stationary case will be discussed at the end of this section).

The above-formulated model of cascaded controlling processes is described by the following jump operators in spaces $\{\mathbf{Z}_q\}=\{z_q,z_2,\ldots,z_N\}$ $(q\ge 1)$:

$$\hat{\mathcal{L}}_q = \textstyle\sum_{i=q+1}^N \hat{\Lambda}_{i-1}\hat{P}_{i_{tt}}, \qquad (4.1)$$

where

$$\hat{\Lambda}_i = (1 - \hat{\mathcal{P}}_{z_i})\hat{k}_{0_i},$$

with

$$\hat{P}_{i_{tt}} = |\mathbf{z}_{i_t}\rangle\langle\mathbf{z}_{i_t}|. \tag{4.2}$$

Here $\hat{\mathcal{P}}_{z_i}$ is the matrix of distribution functions of jump lengths (in $\{z_i\}$ space) and $\hat{k}_{0_i}$ is the matrix of jump rates diagonal in $\{z_i\}$ space.

Similar to the model discussed in Sec. III B, the control of motion in $\{x\}$ space will be described by the z dependence of the operator $\mathcal{L}$:

$$\hat{\mathcal{L}}_{\mathbf{z}}: = (1 - \hat{\mathcal{P}}_x)\hat{k}_0\hat{P}_{1_{tt}}. \tag{4.3}$$

Noteworthy is that in accordance with the results obtained the model (4.1)–(4.3) can be considered as a generalized variant of CTRW-type models for the controlling process.

Recall that the problem under study reduces to evaluating the evolution operator $\hat{\mathbb{G}}(t)$ defined in Eq. (3.7). In the proposed model of cascaded controlling processes this operator can be found in analytical form.

A formula for $\hat{\mathbb{G}}(t)$ can be obtained with the use of the general expression (3.16):

$$\tilde{\hat{\mathbb{G}}}(\epsilon) = \epsilon^{-1}[1 - \hat{w}(\epsilon)][1 - \hat{P}_x\hat{w}(\epsilon)]^{-1} \tag{4.4}$$

$$= \epsilon^{-1}\hat{\Phi}(\epsilon)[\hat{\Phi}(\epsilon) + \hat{\mathcal{L}}_x\hat{k}_0^{-1}]^{-1}, \tag{4.5}$$

in which $\hat{\mathcal{L}}_x: = (1-\hat{P}_x)\hat{k}_0$,

$$\hat{w}(\epsilon) = [1 + \hat{\Phi}(\epsilon)]^{-1}, \tag{4.6}$$

with $\hat{\Phi}(\epsilon) = \hat{k}_0^{-1}\hat{\phi}_1(\epsilon + \hat{\mathcal{L}}_2)$, and

$$\hat{\phi}_1(\epsilon) = \langle\mathbf{z}_{1_t}|\hat{g}_1(\epsilon)|\mathbf{z}_{1_t}\rangle^{-1}, \tag{4.7}$$

where $\hat{g}_1(\epsilon) = (\epsilon + \hat{\Lambda}_1)^{-1}$.

The expressions (4.4)–(4.7) relate the evolution operator $\tilde{\hat{\mathbb{G}}}(\epsilon)$ of the total system in the combined space $\{x \otimes \mathbf{z}\}$ to that $[\hat{g}_1(\epsilon)]$ in the subspace $\{\mathbf{Z}_1\}$.

The procedure presented above allows one to express $\hat{\phi}_1(\epsilon + \mathcal{L}_2)$ in terms of the evolution operator $\hat{g}_2(\epsilon) = (\epsilon + \hat{\Lambda}_2)^{-1}$ in the reduced subspace $\{\mathbf{Z}_2\}$. Expressions similar to Eqs. (4.4)–(4.7), as applied to the operator $\hat{g}_1^{tt}(\epsilon)$, yield

$$\hat{\phi}_1(\epsilon) = \langle\mathbf{z}_{2_t}|[\hat{\phi}_2(\epsilon + \hat{\mathcal{L}}_3) + \hat{\Lambda}_2]^{-1}|\mathbf{z}_{2_t}\rangle^{-1}, \tag{4.8}$$

with

$$\hat{\phi}_2(\epsilon) = \hat{g}_{2_{tt}}(\epsilon)^{-1}, \quad \hat{g}_{2_{tt}}(\epsilon) = \langle\mathbf{z}_{2_t}|\hat{g}_2(\epsilon)|\mathbf{z}_{2_t}\rangle. \tag{4.9}$$

By continuing the proposed procedure one gets a formula for the function $\hat{\Phi}(\epsilon)$ in terms of $\hat{g}_q(\epsilon) = (\epsilon + \hat{\Lambda}_q)^{-1}$ (with $q \geq 1$) and therefore in terms of the functions

$$\hat{\phi}_q(\epsilon) = \langle\mathbf{z}_{q_t}|(\epsilon + \hat{\Lambda}_q)^{-1}|\mathbf{z}_{q_t}\rangle^{-1}, \tag{4.10}$$

which describe memory effects in the system as a result of Markovian motion in $\{z_q\}$ subspace—i.e., without controlling the interaction with other subspaces $\{z_i\}$ with $i > q$:

$$\hat{\Phi}(\epsilon) = \hat{k}_0^{-1}\hat{\phi}_1(\hat{\phi}_2(\hat{\phi}_3(\cdots)))(\epsilon). \tag{4.11}$$

Now we would like to briefly discuss the case of stationary processes. The formulas obtained in this section can easily be modified to describe the stationary case by taking into account that (as has been mentioned in Sec. III B) the only difference between nonstationary and stationary variants is in the initial W_i. In particular, for stationary CTRW, W_i is given by Eq. (3.5). It is worth recalling that, according to our earlier results [23], the relation (3.5) is easily interpreted within the Markovian representation in which the stationary variant is realized by assuming the equilibrium initial condition for the controlling process in $\{z\}$ space—i.e., by taking $|\mathbf{z}_i\rangle = |\mathbf{z}_e\rangle$, where $|\mathbf{z}_e\rangle$ is the equilibrium state of the controlling operator $\hat{\Lambda}$ [23]. Note that in the model (4.1) the equilibrium state can straightforwardly be written as a product: $|\mathbf{z}_e\rangle = \Pi_{q=1}^N|\mathbf{z}_{q_e}\rangle$, in which $|\mathbf{z}_{q_e}\rangle$ is the equilibrium state of the operator $\hat{\Lambda}_q$.

2. Examples

To illustrate the results obtained we will consider two limiting examples of two-state exponential and anomalously slow inverse-power-type controlling processes.

a. Two-state controlling processes. The exponential two-state model for controlling processes corresponds to a simple expression for the evolution operator:

$$\hat{\phi}_q(\epsilon) \sim \langle\mathbf{z}_{q_t}|(\epsilon + \hat{\Lambda}_q)^{-1}|\mathbf{z}_{q_t}\rangle^{-1} = [\epsilon_q/\epsilon - \kappa_q/(\epsilon + \xi_q)]^{-1}, \tag{4.12}$$

where ϵ_q, κ_q, and ξ_q are some constant parameters. Obviously, formula (4.11) with $\hat{\phi}_q(\epsilon)$ presented in Eq. (4.12) gives an expression for $\hat{\Phi}(\epsilon)$ of the algebraic type which predicts multiexponential behavior of the PDF matrix $\hat{w}(t)$.

b. Anomalously slow controlling processes. Another very important model of controlling processes describes anomalously slow inverse-power-type behavior of the PDF $\hat{w}_1(t)$. This model can easily be constructed within the Markovian representation [23]. It corresponds to a weak fractional-power-type dependence of $\hat{\phi}_q(\epsilon)$ [7,23]:

$$\hat{\phi}_q(\epsilon) = w_q(\epsilon/w_q)^{\alpha_q}, \quad \alpha_q < 1, \tag{4.13}$$

where $w_q = \zeta_q k_{0_q}$ and $\zeta_q \sim 1$. Substitution of Eq. (4.13) into formula (4.11) yields

$$\hat{\Phi}(\epsilon) = R_0(\epsilon/w_N)^{\alpha_0}, \tag{4.14}$$

with

$$\alpha_0 = \prod_{q=1}^{N} \alpha_q < 1$$

and $R_0 = \Pi_{q=2}^N(w_q/w_{q-1})^{\gamma_q}$, where $\gamma_q = \Pi_{i=1}^{q-1}\alpha_i$.

Note that in the simplest case of identical rates w_q, $w_q = \bar{w}$ $(q \geq 1)$, when $R_0 = 1$ and $w_N = \bar{w}$, formula (4.14) is represented in a simple form $\hat{\Phi}(\epsilon) = (\epsilon/\bar{w})^{\alpha_0}$.

B. Fluctuating CTRW jumps

1. General formulas

Another interesting problem in the CTRW theory, which can thoroughly be analyzed with the use of the proposed Markovian representation, concerns the kinetics of CTRW processes governed by stochastically fluctuating PDF matrices $\hat{\mathbb{W}}_\beta(t)$ of jumps (renewals) or corresponding matrices $\hat{w}_\beta(t)$ (here β is the number of jumps).

The fact is that in conventional CTRW theories the PDF $\hat{W}_\beta(t)$ and the probability $\hat{P}_\beta(t)$ are assumed to be the same for all renewals $\beta>1$ except the first one (β=0) and fixed functions of time [Eq. (3.4)]. In this section we will discuss the extension of CTRW processes in which the fluctuation jump kinetics is controlled by the PDF and probability matrices whose functional form fluctuates, leading to a stochastic change of $\hat{\mathbb{W}}_\beta(t)$ for different β for fixed β, to the difference of $\hat{\mathbb{W}}_\beta(t)$ for different β, and, in the end, to nontrivial statistical properties renewals.

The main difficulty in modeling PDF matrices, fluctuating in time, consists in the necessity to take into account the normalization condition $\int_0^\infty dt\, W_\beta(t)=\tilde{\hat{W}}_\beta(\epsilon=0)=1$, which ensures population conservation in the process.

The Markovian representation, discussed in Sec. III, allows us to make the above problem tractable. Within this representation the fluctuations of PDF functions $\hat{w}_\beta(t)$ are assumed to be determined by the Markovian controlling process which is governed by the fluctuating operator $\hat{\Lambda}$. The representation offers a quite natural and fairly simple way of describing fluctuating $\hat{\Lambda}(t)$ by suggesting this operator to depend on the parameter $\mathbf{y}(t)$ which undergoes stochastic Markovian fluctuations. In this model the validity of the normalization relation for $\hat{w}_\beta(t)$ can be ensured by taking the proper form of the operator $\hat{\Lambda}(t)$. It is clear from Eq. (3.10) that this relation will be fulfilled if during evolution in $\{z\}$ space, described by fluctuating $\hat{\Lambda}(t)$, the population is conserved; i.e., Eq. (3.10) can be represented in the form $\dot{\varphi}=-[\boldsymbol{\nabla}_z\cdot\mathbf{J}_z(\varphi,t)]$, where $\mathbf{J}_z(\varphi,t)$ is the fluctuating flux in $\{z\}$ space. For instance, the population is certainly conserved in processes governed the Smoluchowski operator (3.12). More clearly this fact will be illustrated below with some examples.

In general, in the Markovian model proposed above, the probability distribution $\sigma(y|t)$ representing the $y(t)$-fluctuation process satisfies an equation of the type of (3.10):

$$\dot{\sigma}=-\hat{\Lambda}_y\sigma, \tag{4.15}$$

with

$$\sigma(\mathbf{y},0)=\sigma_i(\mathbf{y}),$$

where $\hat{\Lambda}_y$ is the operator responsible for the evolution in $\{y\}$ space.

Similar to the case of $\hat{\mathcal{L}}(t)$ fluctuations caused by the Markovian fluctuating $\mathbf{z}(t)$ parameter, consideration of the effect of $\mathbf{y}(t)$ fluctuations reduces to analyzing the SLE for the evolution operator $\hat{\mathbb{G}}_\mathbf{r}(t)$ in the combined space $\{\mathbf{r}\}=\{x\otimes z\otimes y\}$:

$$\dot{\hat{\mathbb{G}}}_\mathbf{r}=-(\hat{\mathcal{L}}+\hat{\Lambda}_z+\hat{\Lambda}_y)\hat{\mathbb{G}}_\mathbf{r}, \tag{4.16}$$

with

$$\hat{\mathbb{G}}_\mathbf{r}(\mathbf{r},\mathbf{r}_i|0)=\delta_{\mathbf{r}\mathbf{r}_i},$$

so that

$$\tilde{\hat{\mathbb{G}}}=\langle\mathbf{y}_e|\langle\mathbf{z}_e|(\epsilon+\hat{\mathcal{L}}+\hat{\Lambda}_y+\hat{\Lambda}_z)^{-1}|\mathbf{z}_i\rangle|\mathbf{y}_i\rangle, \tag{4.17}$$

where $|\mathbf{y}_i\rangle\equiv\sigma_i(\mathbf{y})$ is the initial $\mathbf{y}(t)$ state in bra-ket notation and

$$\langle\tilde{\hat{U}}\rangle=\langle(\epsilon+\hat{\mathcal{L}}+\hat{\Lambda}_y+\hat{\Lambda}_z)^{-1}\rangle_\mathbf{r}. \tag{4.18}$$

In Eq. (4.16) both the jump operator $\hat{\mathcal{L}}$ and the controlling operator Λ_z can, in principle, depend on the variable y as a parameter: $\hat{\mathcal{L}}\equiv\hat{\mathcal{L}}_z^y$ and $\Lambda_z\equiv\Lambda_z^y$.

Just these dependences of $\hat{\mathcal{L}}$ and Λ_z on y model the effect of $\mathbf{y}(t)$ evolution [i.e., fluctuations of PDF functions $\hat{w}_j(t)$] on the kinetics of the processes under study. In general, both dependences can strongly contribute to the effect; however, in this work we will concentrate on an analysis of the effect of $\hat{\mathcal{L}}_z^y$ dependence only, assuming that Λ_z is independent of $\mathbf{y}$.

In the Markovian model of $\hat{w}_j(t)$ fluctuations the problem reduces to an analysis of the SLE (4.16) which is similar to Eq. (3.13) considered above in detail. In general, Eq. (4.16) can be solved only numerically. In this work we will restrict ourselves to discussing some most important specific features of the manifestation of $\hat{w}_j(t)$ fluctuations in the kinetics of the process within the simple analytically solvable variant of the proposed model.

2. Localized rate of fluctuation jumps

The variant of the Markovian model which allows for an analytical analysis of the problem can be considered as an extension of the approximation of highly localized jump rates discussed above. Here we assume high localization of fluctuation jump rates in both spaces $\{z\}$ and $\{y\}$—i.e., in the combined space $\{\mathbf{r}\}=\{z\otimes y\}$. In this limit the expression for the $\hat{\mathcal{L}}_z^y$ dependence is given by

$$\hat{\mathcal{L}}_\mathbf{r}=(1-\hat{P}_x)\hat{k}_0\hat{P}_{\mathbf{r}_{tt}}, \tag{4.19}$$

with

$$\hat{P}_{\mathbf{r}_{tt}}=|\mathbf{y}_t\mathbf{z}_t\rangle\langle\mathbf{y}_t\mathbf{z}_t|,$$

where $|\mathbf{y}_t\mathbf{z}_t\rangle=|\mathbf{y}_t\rangle|\mathbf{z}_t\rangle$ is the jump state in $\{z\otimes y\}$ space which, for simplicity, is suggested to remain unchanged after jumps.

In the model (4.19), $y(t)$ fluctuations are represented as a set of jumps (blips). The statistics of jumps is characterized by the PDF $w_y(t)$ of waiting times of jumps whose Laplace transform can be written as

$$\tilde{\hat{w}}_y(\epsilon)=[1+\hat{\Phi}_y(\epsilon)]^{-1}, \tag{4.20}$$

where

$$\hat{\Phi}_y(\epsilon) = [\hat{k}_0\langle \mathbf{y}_t|(\epsilon + \hat{\Lambda}_y)^{-1}|\mathbf{y}_t\rangle]^{-1}. \quad (4.21)$$

It is easily seen that from a mathematical point of view the proposed model is close that formulated in Sec. III. The difference consists only in a larger dimensionality of the space of the controlling process ($\{z\otimes y\}$ space instead of the $\{z\}$ one in the model considered in Sec. III) and in the corresponding difference in the form of the operator Λ which describes the controlling process ($\Lambda_z+\Lambda_y$ instead of Λ_z).

This similarity enables us to use the results obtained above. It follows from these results that the model (4.19) predicts the CTRW expression for the evolution operator $\hat{\tilde{\mathbb{G}}}$ of type of Eq. (3.16):

$$\hat{\tilde{\mathbb{G}}}(\epsilon) = \hat{\tilde{\mathbb{P}}}_i(\epsilon) + \hat{\tilde{\mathbb{P}}}_t(\epsilon)[1 - \hat{\tilde{\mathbb{W}}}_t(\epsilon)]^{-1}\hat{\tilde{\mathbb{W}}}_i(\epsilon), \quad (4.22)$$

where

$$\hat{\tilde{\mathbb{W}}}_j(\epsilon) = \hat{P}_x\hat{\tilde{w}}_{f_j}(\epsilon),$$

$$\hat{\tilde{\mathbb{P}}}_j(\epsilon) = \epsilon^{-1}[1 - \hat{\tilde{w}}_{f_j}(\epsilon)] \quad (j = i,t), \quad (4.23)$$

and

$$\hat{\tilde{w}}_{f_j}(\epsilon) = [1 + \hat{\Phi}_{f_j}(\epsilon)]^{-1}, \quad (4.24)$$

however with matrices

$$\hat{\Phi}_{f_j}(\epsilon) = [\hat{k}_0\langle \mathbf{y}_t\mathbf{z}_t|(\epsilon + \hat{\Lambda}_y + \hat{\Lambda}_z)^{-1}|\mathbf{y}_t\mathbf{z}_j\rangle]^{-1}, \quad (4.25)$$

whose behavior essentially depends on the specific features of the stochastic evolution of $\mathbf{y}(t)$.

This expression makes it possible to easily analyze some specific features of the effect of $\mathbf{y}(t)$ fluctuations. First of all, it is of special interest to discuss two natural limits: slow and fast $\mathbf{y}(t)$ fluctuations.

a. The limit of slow $y(t)$ and $\hat{w}_j(t)$ fluctuations. In the limit of slow fluctuations $y(t)$, when the characteristic time of evolution in $\{y\}$ space, τ_y, is larger than that of the PDFs $\hat{w}_j(t)$, τ_w, $\tau_y \gg \tau_w$, one can neglect the term $\hat{\Lambda}_y$ in Eqs. (4.16)–(4.18), thus arriving at the expression for $\hat{\mathbb{G}}$ and $\langle\hat{\tilde{U}}\rangle$ of the type of those obtained in the CTRW approach (see Sec. III), but averaged over the y distribution $|\sigma_i(\mathbf{y})\rangle \equiv |\mathbf{y}_i\rangle$. In the short-range model considered in this limit one gets expressions (4.22)–(4.25) for $\hat{\tilde{\mathbb{G}}}(\epsilon)$ with

$$\hat{\Phi}_{f_j}(\epsilon) \approx [\hat{k}_0\langle \mathbf{y}_t|\mathbf{y}_j\rangle\langle \mathbf{z}_t|(\epsilon + \hat{\Lambda}_z)^{-1}|\mathbf{z}_j\rangle]^{-1}. \quad (4.26)$$

b. The limit of fast $y(t)$ and $\hat{w}_j(t)$ fluctuations. In the opposite limit of fast fluctuations $\mathbf{y}(t)$, corresponding to $\tau_y \ll \tau_w$, the SLE (4.16) still yields the CTRW-like formula for the evolution operator $\hat{\mathbb{G}}$. In this formula, however, the matrices $\hat{\tilde{\mathbb{W}}}_j(\epsilon)$ and $\hat{\tilde{\mathbb{P}}}_j(\epsilon)$ are expressed in terms of the PDF matrix $\hat{w}_j(t)$ obtained by means of relations (3.18)–(3.22) which are evaluated with transition matrices $\hat{k}_z^d(\mathbf{y})$ ana $\hat{k}_z^n(\mathbf{y})$ [see Eq. (3.15)] averaged over the equilibrium y distribution $\sigma_e(\mathbf{y}) \equiv |\mathbf{y}_e\rangle$. In the fast-fluctuation limit the short-range model results in expressions (4.22)–(4.25) for $\hat{\tilde{\mathbb{G}}}(\epsilon)$ with

$$\hat{\Phi}_{f_j}(\epsilon) \approx [\hat{k}_0\langle \mathbf{y}_t|\mathbf{y}_e\rangle\langle \mathbf{z}_t|(\epsilon + \hat{\Lambda}_z)^{-1}|\mathbf{z}_j\rangle]^{-1}, \quad (4.27)$$

where $|\mathbf{y}_e\rangle$ is the equilibrium state in $\{y\}$ space.

It is seen from formulas (4.26) and (4.27) that in the simple model of localized jump rates considered the kinetics of the process is similar in both limits. The difference is only in the characteristic parameters. However, in general, in the case of delocalized jump rates—i.e., delocalized dependence of $\mathcal{L}_z^y$ on $\mathbf{y}$—the kinetics in these two limits is, of course, different although the asymptotic behavior at long times is expected to be similar. Below we will discuss the effect of the delocalization of this dependence (see next section).

In addition, strictly speaking, in the above analysis of two limits we have assumed that $\mathbf{y}(t)$ fluctuations are stationary which implies the existence of the equilibrium state $|\mathbf{y}_e\rangle$ of the operator Λ_y. It is of special interest to study the manifestation of nonstationary $\mathbf{z}(t)$ fluctuations which can be fairly strong especially in the case anomalously slow fluctuations [7].

3. Anomalous y(t) fluctuations

Here we will analyze in detail the effect of an anomalously slowly fluctuating coordinate $\mathbf{y}(t)$. In the Markovian representation this type of fluctuations can be described with the use of the free-diffusion model for $y(t)$ evolution discussed in Sec. III B 1. In this model the operator Λ_y is given by

$$\hat{\Lambda}_y = -y^{1-n_y}\nabla_y(D_y y^{n_y-1}\nabla_y), \quad (4.28)$$

where $\nabla_y = \partial/\partial y$ and n_y is the $\{y\}$-space dimensionality (it is assumed that $n_y \le 2$).

The model (4.28) predicts the anomalous long-time tailed dependence of the characteristic PDF $w_y(t)$ [see formulas (4.20) and (4.21)]:

$$w_y(t) \sim 1/t^{1+n_y/2}. \quad (4.29)$$

These anomalous fluctuations of PDFs $w_\beta(t)$ of CTRW-jumps very strongly affect the kinetics of the CTRW process. The effect can clearly be revealed by analyzing the behavior of the matrices $\Phi_{f_j}(\epsilon)$ [see Eq. (4.25)]. Of special interest and importance is the asymptotic behavior of $\Phi_{f_j}(\epsilon)$ at $\epsilon \to 0$ which determines the long-time asymptotic behavior of the process.

To demonstrate the specific features of the small-ϵ behavior of $\Phi_{f_j}(\epsilon)$ we will consider two examples of controlling processes in $\{z\}$ space: exponential (Poissonian) and anomalous long-time-tailed. Both these examples can properly be treated within the diffusion model.

a. Poissonian $\mathbf{z}(t)$ controlling processes. Poissonian-like controlling processes can be described by the model of diffusive motion of a Brownian particle confined within a well in $\{z\}$ space. In this model at small ϵ, which corresponds to times longer than the time τ_z of the diffusive relaxation within the well $U(z)$ in $\{z\}$ space, the behavior of $\Phi_{f_i}(\epsilon)$ is determined by free diffusion in $\{y\}$ space and therefore

$$\Phi_{f_j}(\epsilon) \sim \epsilon^{n_y/2}, \quad w_{f_j}(t) \sim 1/t^{1+n_y/2} \quad (j = i,t). \quad (4.30)$$

This result shows that originally normal CTRW processes become anomalous as a result of anomalous $w_j(t)$ fluctuations and the resultant behavior of $w_j(t) \equiv w_{f_j}(t)$ coincides with that of $y(t)$ fluctuations; i.e., anomalous $y(t)$ fluctuations strongly modify the kinetics of normal CTRW processes.

b. Long-time-tailed $\mathbf{z}(t)$ *controlling processes.* The long-time tailed case is represented by the free diffusion model in $\{z\}$ space $[U(z)=0]$. In this model the small-ϵ behavior of $\Phi_{f_i}(\epsilon)$ is determined by free diffusion in the total $\{y \otimes z\}$ space of dimensionality n_y+n_z. This means that in the model of localized jump rates

$$\Phi_{f_j}(\epsilon) \sim \epsilon^{n_{yz}/2}, \quad w_{f_j}(t) \sim 1/t^{1+n_{yz}/2}, \quad (4.31)$$

where $n_{yz}=n_y+n_z$. Formula (4.31) demonstrates the strong effect of $w_j(t)$ fluctuations on the kinetics of the originally anomalous CTRW processes. Noteworthy is that $w_j(t)$ fluctuations result in an increase of the anomaly parameter α which determines the long-time behavior of $w_j(t)$: $w_j(t) \sim 1/t^{1+\alpha}$.

It is interesting to note that the effect of fluctuations can lead to a crucial change of fluctuation jump statistics. The fact is that usually CTRW theories assume that $\int_0^\infty dt\, W_\beta(t) = 1$. This relation ensures conservation of normalization (or population) in CTRW processes. In the free-diffusion approximation for the controlling process this conservation relation is fulfilled if the dimensionality of the space is small, when the statistics of reoccurrences in the jump state is recurrent. That is why we have assumed $n_y<2$ and $n_z<2$. These two inequalities, however, do not guarantee the same inequality for $n_{yz}=n_y+n_z$. In principle, one can get $n_{yz}>2$, and in this case the statistics of reoccurrences becomes transient which means that $\int_0^\infty dt\, w_j(t) < 1$. Such CTRW processes with decay are known in probability theory although are not applied widely [22].

V. DISCUSSION

The results obtained in this work have shown that the Markovian representation is very useful for a general investigation of non-Markovian processes and, in particular, for studying and extending the CTRW approach.

Special attention in this paper has been paid to application of this representation to the analysis of approaches well known in the theory of CTRW processes. For example, subordination, conventionally used as a basis for formulation of the CTRW approach [17,22,35], can conveniently be described and generalized in the Markovian representation with controlling processes (in $\{\mathbf{z}\}$ space) of certain type. In particular, multiple subordination is closely related to cascaded processes [realized with the cascaded scheme of coupling (see below)]. This representation also helps to extend the CTRW modeling by combining the cascaded scheme of coupling with some other schemes, as proposed in Sec. V C 1.

The important advantage of the proposed method consists in simplification of the description of non-Markovian kinetic problems by reducing the treatment to manipulations with linear operators. Moreover, in many cases the representation allows for deep understanding and modeling real stochastic processes in clear physical terms.

In this section we will discuss some specific features of the proposed method and obtained results.

A. Validity of the CTRW approach

The Markovian equation (3.13), applied to deriving the Markovian representation, is of course more general than the CTRW approach and therefore is quite suitable for the analysis of applicability conditions of this approach. Here we will present some comments on this point.

The problem reduces to analyzing the statistics of fluctuation jumps controlled by the stochastic process in $\{z\}$ space which is determined by the evolution operator $\hat{G}(\epsilon)=(\epsilon+\hat{k}_z^d+\hat{\Lambda})^{-1}$, where $\hat{k}_z^d$ is the jump-rate matrix diagonal in $\{x\}$ and $\{z\}$ spaces [see Eq. (3.17)].

In the limit of a high localization of the jump-rate matrix $\hat{k}_z^d$ (in $\{z\}$ space) defined by Eq. (3.15) the Markovian equation (3.13) leads to the evolution operator $\hat{\bar{\mathbb{G}}}(\epsilon)$ corresponding to the CTRW approach. In the case of extended $\hat{k}_z^d$, however, the CTRW approach is not valid, strictly speaking.

To find the conditions which can ensure the applicability of the CTRW approach we will consider the above-proposed simple and fairly flexible diffusion model for the controlling operator $\hat{\Lambda}$ [Eq. (3.12)].

(i) *Poissonian-like statistics.* Within the diffusion model the Poissonian-like jump statistics is described by the evolution operator $\hat{G}(\epsilon)$ with the Smoluchowski operator $\hat{\Lambda}$, in which the potential $u(z)$ is of type of infinitely deep potential wells, say, of width a_u—i.e., $u(a_u) \sim 1$. At long times this model predicts an exponentially decreasing function $w_n(t)$ with characteristic time $\tau_n \sim a_u^2/D_z$.

It is easily seen that in this model the limit of high localization is realized for $a_k \ll a_u$, where a_k is the characteristic width of the function $\hat{k}_z^d$.

(ii) *Anomalous long-time tailed statistics.* The case of long-time tailed jump statistics is represented by the free-diffusion variant of the diffusion model $[u(z)=0]$, which in the limit of highly localized $\hat{k}_z^d$ predicts the PDF $w_j(t) \sim 1/t^{1+n_z/2}$, where n_z is the $\{z\}$-space dimensionality.

In the case of long-time tailed statistics the condition of applicability of the CTRW approach (validity of the approximation of highly localized jump rates) is less trivial than that formulated above for Poissonian-like statistics. The fact is that in the absence of the potential one needs to compare the characteristic width of $\hat{k}_z^d$ in $\{z\}$ space with that of the probability distribution $\varphi(z,t)$ of the controlling process [see Eq. (3.10)] which yields the condition $a_k^2 \ll D_z t$.

Noteworthy is that to satisfy this condition the function $\hat{k}_z^d$ does not need to be short range: for example, in the case $\hat{k}_{\mathbf{z}}^d \sim 1/z^m$ the proper parameter a_k can be introduced for $m>3$ [36]. In addition in the case $m>3$ the long-time behavior of the PDF is shown to be identical to that for really short range $\hat{k}_z^d$ with the corresponding size a_k. This means that for $m>3$ at long times $a_k \ll D_z t$ the CTRW approach is quite applicable.

This brief analysis shows that *in the large class of anomalous non-Markovian models the long-time behavior of the evolution operator* $\hat{\mathbb{G}}(t)$ is correctly described by the CTRW approach.

B. Applications of the generalized SLE

Here we will discuss some processes which cannot be described by the conventional CTRW-based SLE and the application of the generalized SLE (3.16) is required. In principle, the generalized SLE is necessary when one needs to describe the relaxation in addition to that presented by the considered CTRW process. The effect of this additional relaxation, assumed to be governed by the fluctuating operator $\hat{L}(t)$, is described by the functional (2.2). If the PDF matrix $\hat{w}_n(t)$ describing the original CTRW induced relaxation, or the operator $\hat{\Lambda}$ which determines this matrix [see Eqs. (3.16)–(3.21)], does not commute with $\hat{L}(t)$, then the combined effect of both types of relaxation can only be described by the generalized SLE (3.16). Below we will briefly discuss some examples of such processes, both quantum and classical.

1. Spin evolution in radical pairs

The non-Markovian SLE is known to be able to describe relaxation in quantum systems induced by fluctuating interactions with a long-time tailed correlation function. So far only very simple examples of quantum systems have been considered [37,38]. Here we will discuss a more complicated process, whose kinetics can be described only by the generalized SLE. This is the geminate recombination of a pair of radicals (particles with electron spin 1/2) undergoing relative diffusive motion. The spin evolution in the pair is governed by the Hamiltonian [39] $\hat{H}(r)=\hat{H}_0-\hat{J}(r)$, where $\hat{H}_0$ is the spin Hamiltonian of noninteracting radicals and $\hat{J}(r)$ is the electron exchange interaction depending on the interradical distance r. This Hamiltonian describes transitions between singlet ($|S\rangle$) and triplet ($|T_{0,\pm}\rangle$) states of the pair.

The recombination is assumed to result in a transition (reversible or irreversible) from the $|S\rangle$ state to the ground state $|S_0\rangle$ of the product molecule localized at a contact distance $r=d$ and is usually modeled by the recombination operator $\hat{K}(r)=\hat{K}_s\delta(r-d)$.

The evolution of this system can be treated as a CTRW process governed by the diffusive reencounters (renewals) which are controlled by the relative diffusion operator $\hat{\Lambda}$ of type (3.12) with $z\equiv r$ and the "jump" operator $\hat{K}_s$ (the analog of the operator $\hat{P}_x$). In this formulation the spin and space evolution is determined by averaging the functional $U(t)=T\{\exp[-\int^t d\tau\,\hat{L}_q(r(\tau))]\}$, where $\hat{L}_q(r)=i[H(r),\ldots]$, over this CTRW process. The problem, however, arises due to the fact that $\hat{\Lambda}$ does not commute with $\hat{H}(r)$. In such a case the conventional CTRW-based SLE [23] is not applicable and one needs to use the generalized SLE (3.16) which predicts essential modification of the diffusive probability distribution by the quantum subsystem.

2. Non-Markovian Fokker-Planck equation

Here we will analyze the one-dimensional stochastic motion of a particle moving with fluctuating velocity $v(t)$ whose fluctuations are assumed to be of CTRW type with the PDF $W(t)$ depending on the coordinate $\{x\}$: $W(t)\equiv W_x(t)$. In other words, we would like to derive the non-Markovian variant of the Fokker-Planck equation.

The non-Markovian phase evolution operator for the particle can be obtained by averaging the functional $U(t)=T\{\exp[-\int^t d\tau\,\hat{L}_d(\tau)]\}$, with $\hat{L}_d(t)=v(t)\nabla_x$, describing dynamical motion.

In the case of $W(t)$ being independent of x the expression for the evolution operator is given by the simple variant of the SLE (3.16) with $\hat{\tilde{\mathbb{W}}}_j(\epsilon)$ determined by the simplified formula (3.27)—i.e., $\hat{\tilde{\mathbb{W}}}_j(\epsilon)=\hat{\tilde{\mathbb{W}}}_{0j}(\hat{\Omega}(\epsilon))=\widetilde{W}(\Omega(\epsilon))$, where $\hat{\Omega}(\epsilon)=\epsilon+\hat{L}_d=\epsilon+v\nabla_x$ [23,27,28]. In the simplest variant, when $W(t)$ is just a function rather than a matrix (or operator), this expression was recently reproduced in Ref. [40].

Of special interest is a more complicated variant of x-dependent $W_x(t)$, which in the Markovian representation is modeled by taking the x-dependent operator $\hat{\Lambda}\equiv\hat{\Lambda}(x)$. This nonhomogeneous case is not described by the simple SLE, and one should use the generalized one (3.16)–(3.21) (since $[\hat{L}_d,W_x]\neq 0$). The SLE (3.16) is still of CTRW form, but with the PDF matrix $\tilde{\mathbb{W}}_j(\epsilon)$ strongly modified by dynamical motion (the PDF matrix essentially depends on the operator $\hat{L}_d=v\nabla_x$): $\hat{\tilde{\mathbb{W}}}_j(\epsilon)=\hat{P}_x\hat{k}_0\langle\mathbf{z}_t|[\epsilon+\hat{L}_d+\hat{k}^d_{\mathbf{z}}+\hat{\Lambda}(x)]^{-1}|\mathbf{z}_j\rangle$. This formula makes the problem tractable, reducing it to calculating the PDF matrix $\hat{\tilde{\mathbb{W}}}_j(\epsilon)$ by means of solving some differential equation if $\hat{\Lambda}$ is assumed to be differential operator [for example, of the form (3.12) with x-dependent D_z and $u(z)$].

C. Extended CTRW approaches

In Sec. IV we have studied the most general features of two extensions: CTRW with cascaded controlling processes and CTRW processes with fluctuating PDFs. Below we will discuss in detail some particular predictions of the extended CTRW approaches.

1. Cascaded controlling processes

The model of cascaded control is a good tool for the analysis of kinetic processes in fractal structures. The cascade of coupled processes can properly model the kinetic coupling of structures of different sizes. The model is very useful for the analysis of processes in self-similar and complex structures [13]. It can also be fairly helpful in the analysis of so called "basin-to-basin kinetics" or diffusion in ultrametric spaces which describes relaxation in systems with a hierarchal energy landscape [29,30].

As an example, we discuss anomalous relaxation in disordered structures in which the controlling processes are often quite adequately described by the anomalous model considered in Sec. IV B with the controlling evolution functions $\hat{\phi}_q(\epsilon)\sim\epsilon^{\alpha_q}$, where $\alpha_q<1$ $(q=1,\ldots,N)$ [see Eq. (4.13)] [7].

In this model formula (4.14) predicts interesting behavior of the total evolution function $\hat{\Phi}(\epsilon)\sim\epsilon^{\alpha_0}$, where $\alpha_0=\Pi_{i=1}^{N}\alpha_i$. This product is convergent for properly behaving α_i as a function of i [41], but it is evident, in advance, that the necessary condition is $\alpha_{i\to\infty}\to 1$. This condition has clear physical meaning: if we ascribe the controlling functions corresponding to larger numbers i to structures of smaller size, then the behavior $\alpha_{i\to\infty}\to 1$ results from the evident fact that in structures of smaller size disorder-induced anomalous effects are expected to be weaker. The weakness of the effects manifests itself in a reduction of processes to Markovian with an increase of i—i.e., just in the relation $\alpha_{i\to\infty}\to 1$ or $\alpha_i=1$ at i larger than some characteristic number N.

Noteworthy is also another interesting prediction of the expression (4.14): the anomaly of the processes is accumulated in the cascade. This effect manifests itself in a decrease of α_0 with an increase of the number N of coupled processes. In particular, for a large number N of weakly anomalous cascaded processes with $\delta_i=1-\alpha_i\ll 1$ the value

$$\alpha_0\approx\exp\left(-\sum_{j=1}^{N}\delta_j\right)\approx\exp\left(-\int_0^N dj(1-\alpha_j)\right) \quad (5.1)$$

can be fairly small, corresponding to strongly anomalous processes. Formula (5.1) sheds light on the mechanism of formation of the relaxation anomaly in disordered systems.

Concluding the discussion notice that the proposed model of cascaded control has something in common with the approach based on coupled Langevin equations [42,43], at least as applied to anomalous processes. Noteworthy, however, is that unlike this approach, analyzing the stochastic trajectories of the process, the proposed method and, in particular, the Markovian representation deal with equations for distribution functions from the very beginning. In general, it is more suitable for both fundamental analysis and applications. Typically the Langevin approach is applied to phenomenological investigations while the proposed method allows one to reveal, in addition, microscopic mechanisms of the processes under study. The results obtained in this work (fairly general and flexible) confirm this conclusion. They can be applied both to stationary and nonstationary processes and can describe not only the asymptotic behavior of the kinetics, but also transient jumplike processes. Besides, the proposed method is not restricted to the cascaded (sequential) coupling scheme. It also enables one to consider more complicated schemes of coupling, for example, by combining cascaded schemes with those assuming multidimensional coupling (corresponding to parallel scheme of coupling) as in the model of fluctuating PDFs.

2. Fluctuations of waiting-time PDF matrices

The results obtained in Sec. IV B demonstrate that fluctuations of waiting-time PDF matrices can strongly manifest themselves in the kinetics of CTRW-like processes. It is, however, still worth pointing out some specific features of this effect.

(i) First we should emphasize that the assumption of correlated fluctuations of $W_\beta(t)$ implies some correlations between renewals of fluctuation blips (sudden changes). After such a modification the model under study is, strictly speaking, not of CTRW type any more. One of the important ideas of our analysis presented above is just to demonstrate that a large class of at first sight non-CTRW models with fluctuating $W_\beta(t)$ are actually the CTRW ones, but in some extended space.

(ii) In our consideration in Sec. IV B fluctuation PDF matrices are assumed to result from those of the jump rate $\hat{k}_0$—i.e., from the dependence $\hat{k}_0(\mathbf{y})$. For simplicity, we have used the model of highly localized $\hat{k}_0(\mathbf{y})=\hat{\kappa}_0|\mathbf{y}_t\rangle\langle\mathbf{y}_t|$. However, in accordance with the conditions of applicability of CTRW approaches (Sec. V A), the main conclusions about the effect of fluctuations on the kinetics of the process remain valid for delocalized dependences $\hat{k}_0(\mathbf{y})$ as well, if these dependences are sharp enough (for details see Sec. IV B)—i.e., for the wide class of models based on the Markovian representation.

(iii) To simplify the analysis, the operators $\hat{\Lambda}_z$ and $\hat{\Lambda}_y$ have been suggested to be independent of y and z variables, respectively. Nevertheless, the major conclusions of Sec. IV B are valid in the case of coupled processes in $\{y\}$ and $\{z\}$ spaces (i.e., for $[\hat{\Lambda}_z,\hat{\Lambda}_y]\neq 0$). The most general formulation in this case consists in the replacement of the sum $\hat{\Lambda}_z+\hat{\Lambda}_y$ by some operator $\hat{\Lambda}_{yz}$ in the combined space $\{y\otimes z\}$.

(iv) To illustrate possible modifications of the fluctuation mechanism within the above general formulation we will briefly discuss the simple model which describes fluctuations occurring at moments of jump transitions. In principle, this model corresponds to the highly localized z dependence of the operator $\hat{\Lambda}_y(\mathbf{z})$, $\hat{\Lambda}_y(\mathbf{z})\sim\hat{P}_z^{tt}$, but in the end the dependence reduces to the following modification of the operator $\hat{\mathcal{L}}$ [see Eqs. (3.8) and (3.15)]:

$$\hat{\mathcal{L}}=\hat{k}_0(\mathbf{y})\hat{P}_z^{tt}-\hat{P}_x\hat{P}_y\hat{k}_0(\mathbf{y})\hat{P}_z^{nt}. \quad (5.2)$$

Here $\hat{P}_y$ describes a sudden change of the $\mathbf{y}$ coordinate simultaneously with the jump in $\{x\}$ space. Of course, $\hat{P}_y$ satisfies the normalization condition $\langle\mathbf{y}_e|(1-\hat{P}_y)=0$, where $\langle\mathbf{y}_e|=\Sigma_i\langle\mathbf{y}_j|$ is the adjoint equilibrium vector in $\{y\}$ space. For example, in the simplest variant of the sudden relaxation in $\{y\}$ space, $\hat{P}_y=|\mathbf{y}_e\rangle\langle\mathbf{y}_e|$.

Substitution of the expression (5.2) into the SLE (3.13) and subsequent manipulations similar to those presented in Sec. III yield

$$\hat{\tilde{\mathbb{G}}}(\epsilon)=\langle\mathbf{y}_e|\hat{\tilde{\mathbb{G}}}_y(\epsilon)|\mathbf{y}_i\rangle, \quad (5.3)$$

where

$$\hat{\tilde{\mathbb{G}}}_y(\epsilon)=\hat{\tilde{\mathbb{P}}}_{y_i}(\epsilon)+\hat{\tilde{\mathbb{P}}}_{y_n}(\epsilon)[1-\tilde{\mathbb{W}}_{y_n}(\epsilon)]^{-1}\hat{\tilde{\mathbb{W}}}_{y_i}(\epsilon). \quad (5.4)$$

The parameters in this formula are similar to those defined in Eqs. (3.16)–(3.22). The only difference consists in an additional term $\hat{P}_y$ in the expression for $\hat{\tilde{\mathbb{W}}}_{y_j}$ $(j=i,n)$ and in the y dependence of the parameters resulting from the dependence $\hat{k}_0(\mathbf{y})$. According to Eq. (3.18), $\hat{\tilde{\mathbb{W}}}_{y_j}=\hat{P}_x\hat{P}_y\hat{\tilde{w}}_{y_j}(\epsilon)$, where

$\widetilde{w}_{y_j}(\epsilon)=\hat{k}_0(y)\langle \mathbf{z}_t|\hat{G}_y(\epsilon)|\mathbf{z}_j\rangle$ and $\hat{G}_y(\epsilon)$ is obtained with $\hat{k}_0(\mathbf{y})$. This means that formula (5.4) coincides with Eq. (3.18) in which $\hat{P}_x$ is replaced by $\hat{P}_{xy}=\hat{P}_x\hat{P}_y$. In other words, this formula describes jumplike migration in $\{x\otimes y\}$ space although with jump rate $\hat{k}_0(y)$ which has addition dependence on y. Moreover, the expression (5.4) can also be represented in the form (4.5) suitable for an analysis of the limit of diffusion-like motion governed by the operator $\hat{\mathcal{L}}_{xy}=(1-\hat{P}_{xy})\hat{k}_0(\mathbf{y})$ describing diffusion with the coefficient $\hat{D}(\mathbf{y})\sim\hat{k}_0(\mathbf{y})$ in some effective potential [44].

Notice that in this model (unlike that discussed in Sec. IV B) $\hat{\mathbb{W}}(t)$ fluctuations, caused by stochastic motion in $\{y\}$ space, do not lead to a significant and universal change of the long-time behavior of $\hat{w}_{f_j}(t)$ defined in Eq. (4.23) [see Eqs. (4.30) and (4.31)]. Some change of the $\hat{w}_{f_j}(t)$ behavior is, in principle, possible due to the effect of the $\hat{D}(\mathbf{y})\sim\hat{k}_0(\mathbf{y})$ dependence, but this change is not universal, depending on the specific features of the $\hat{k}_0(\mathbf{y})$ behavior.

It is also worth noting that the model, which has something in common with the particular variant considered of our general model, was recently discussed in Ref. [25]. Restricting ourselves to brief comments we would only like to emphasize that, as the analysis of the above simple variant shows, the general method developed in our work enables one to represent the results obtained in this paper in a very compact and general form.

(v) From the very beginning of our considerations we have discussed extensions within the Markovian representation reducing the problem to an analysis of multidimensional SLEs. In so doing we have not interpreted the results in terms of the conventional approach appealing to the stochastic properties of fluctuating PDFs of consecutive jumps $\hat{\mathbb{W}}_\beta(t)$ (see Sec. III). It is clear that the case of the most pronounced effect of fluctuations corresponds to strong long-time tailed correlations of the PDFs and of course in this case the expression can be found without difficulty. For example, one can consider the model assuming a stochastic change of $\hat{\mathbb{W}}_\beta(t)$ localized (in time) near the time of fluctuation jumps. In this model the change of PDFs can be described by introducing an additional fluctuation matrix $\hat{\mathbb{W}}_c(t)$ with which conventional convolution terms of type $\int_0^{t_2}dt_1\hat{\mathbb{W}}(t-t_1)\hat{\mathbb{W}}(t_1-t_0)$ are transformed into $\int_0^{t_2}dt_1\hat{\mathbb{W}}(t-t_1)\int_0^{t_1}d\tau\,\hat{\mathbb{W}}_c(t_1-\tau)\hat{\mathbb{W}}(\tau-t_0)$. It is clear that in the limit of short-range dependences $\hat{\mathbb{W}}_c(t)$ this model reduces to that considered above. However, even in the most general formulation it can be treated as a particular variant of the proposed approach based on the Markovian representation.

(vi) Concluding the discussion we present some examples of realistic processes which can be interpreted within the CTRW models with fluctuating PDF $W(t)$.

One of them is related to that discussed in Sec. V B 1: this is a diffusion-controlled liquid-phase reaction resulting in transitions in the space of molecular states (the analog of $\{x\}$ space). Assume, however, that the reactivity is anisotropic [45–47]. In this case the diffusive rotational motion of reacting molecules leads to stochastic modulation of the reaction rate, thus resulting in fluctuations of the PDF $W(t)$ describing the statistics of diffusion-controlled blips (jumps) of the rate. The kinetics of these reactions can be treated within the proposed model of fluctuating CTRW jumps (Sec. IV B) by reducing the problem to solving the diffusionlike equation in the extended space of the relative coordinates and orientations of molecules.

The processes described are, actually, a particular case of a more general class of processes whose kinetics is strongly affected by the fluctuating reaction rate (this phenomenon is usually called gating [31,32]). The CTRW approach is very useful for an analysis of these processes. In particular, the effect of gating can properly be described just with the use of the proposed model of fluctuating CTRW jumps (fluctuating waiting-time PDFs).

VI. CONCLUSIONS

This work concerns a detailed discussion and applications of the Markovian representation of non-Markovian CTRW-like processes and, in particular, non-Markovian CTRW-based SLEs. In reality, however the Markovian representation, reducing the problem to a study of the multidimensional Markovian SLE, is more general than the CTRW approach and allows one not only to analyze the applicability of this approach, but also to develop some extensions. In our work we have considered two of them describing the effect of cascaded controlling processes and fluctuations of jump PDFs. The number of extensions is, however, very large. For example, interesting effects can be predicted in variants of CTRW approaches combining the two above-mentioned extensions. Further generalizations and applications will be the subject of subsequent publications.

Furthermore, in the analysis of a number of problems it is, may be, more insightful and convenient to solve (at least numerically) the general multidimensional Markovian equations of the Markovian representation themselves rather than the CTRW-like SLEs which can only be considered as a particular case of these equations (corresponding to the highly localized transition region). In this respect the situation is similar to that concerning the generalized Langevin equation [48]. The fact is that the memory term in this equation is adequately modeled by coupling of the system under study with the oscillatory bath [48] (in the quantum case this approach is often called the spin-boson model). Many of studies with the use of this model [48] have shown that the majority of problems are more easily analyzed within the oscillatory model itself rather than the corresponding Langevin equation. The same situation can also be realized in applications of multidimensional Markovian equations and the Markovian representation discussed in this work.

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