Specific features of the effect of time dependent field on subdiffusing particles. The stochastic Liouville equation approach.

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We analyze the effect of time dependent external field on non-Markovian migration described by the continuous time random walk (CTRW) approach. The rigorous method of treating the problem is proposed which is based on the Markovian representations of the CTRW approach and field modulation. The method is applied to the case of subdiffusive migration in which the exact formulas for the first and second moments of spatial distribution are derived. For oscillating external field they predict unusual dependence of the first moment on oscillation phase and anomalous field dependent contribution to the dispersion. Similar formulas are also derived fluctuating field.

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I. INTRODUCTION.

Brownian motion in external time-dependent field is the important stage of many physical and chemical processes which often strongly affect their kinetics [1, 2]. Last years close attention is given to the anomalous (subdiffusive) jump-like motion typical for disordered systems [3, 4] and, in particular, to the effect of time-dependent field on this type of migration [3, 4]. Usually the motion anomaly is assumed to be a manifestation of the long memory in the kinetics of jumps. In such a case the serious difficulty in theoretical treatment of time-dependent field effects occurs because of subtle interplay of field and anomalous memory effects which should be properly described.

Subdiffusive processes in time-independent potential \( V(x) \) are traditionally described by the fractional Smoluchowski equation (FSE) for the probability distribution function (PDF) \( \rho(x,t) \) [4]

\[
\frac{\partial \rho}{\partial t} = -D_1^{1-\alpha} \hat{L}_\alpha \rho,
\]

where \( D_1^{1-\alpha} \) is the Riemann-Liouville fractional derivative defined by

\[
D_1^{1-\alpha} \psi = \frac{1}{\Gamma(\alpha)} \frac{\partial}{\partial t} \int_0^t dt_1 \frac{\psi(t_1)}{(t-t_1)^{1-\alpha}}
\]

and

\[
\hat{L}_\alpha = -D_\alpha \nabla_x \left[ \nabla_x - F(x) \right]
\]

is the Smoluchowski operator, in which \( D_\alpha \) is a subdiffusion constant, \( \nabla_x \equiv \partial/\partial x \), and \( F(x) = -\nabla_x V(x)/(k_BT) \) is a force. The FSE (1) can be derived within the continuous time random walk (CTRW) approach [4] assuming the long time tailed behavior of the waiting time distribution \( W(t) \) for CTRW-jumps: \( W(t) \sim 1/t^{1+\alpha} (\alpha < 1) \).

In the case of time-dependent field \( F(x,t) \) [i.e. time-dependent \( \hat{L}_\alpha(t) \)], however, no analogs of the FSE are rigorously derived as yet. The main difficulty is in correct treatment of the effect of \( \hat{L}_\alpha(t) \)-evolution during the time of waiting for jumps. Only approximate variants of these FSEs have been proposed so far [3, 4].

In this work within the CTRW approach we derive the exact FSE (describing the influence of time-dependent field) with the use of recently proposed Markovian representation of the CTRW and the non-Markovian stochastic Liouville equation (SLE) [4]. The solutions of this FSE for different time dependences of the force \( F(t) \), for simplicity, assumed to be independent of \( x \), are proposed and discussed in detail.

II. MARKOVIAN SLE.

Here we will present the method of treating the effect of time-dependent field \( F(x,t) \) on CTRW-like processes by reduction of the problem to solving the SLE with time-independent operators.

To clarify the method we first consider the Markovian (normal diffusion) case: \( \alpha = 1 \), in which the evolution of the system is described by the Smoluchowski equation

\[
\frac{\partial \rho}{\partial t} = -\hat{L}_1 \rho = D_1 \nabla_x \left[ \nabla_x - F(x) \right] \rho
\]

(4)

The method is based on the representation of the time dependence of \( F(x,t) \) in terms of the dependence on some Markovian (in general, stochastic) variable \( z(t) \):

\[
F(x,t) \equiv F(x,z(t)), \quad \text{satisfying the Markovian equation}
\]

\[
\dot{\sigma} = -\hat{L} \sigma, \quad \text{with} \quad \sigma(0) = \sigma(z),
\]

(5)

in which \( \hat{L} \) is the linear operator in \( \{z\} \)-space and \( \int dz \sigma(z) = 1 \). For brevity, formulas are written assuming that \( \{z\} \)-space is one dimensional, though they are, evidently, valid for any dimensionality of \( \{z\} \)-space. The corresponding examples will be discussed below. In addition, in what follows we will restrict ourselves to the simple model of \( x \)-independent force:

\[
F(x,t) \equiv F(x,z(t)) = F_0 z(t)
\]

(6)
In this representation the kinetics of the process described by eq. \( \Phi \) is determined by the average evolution operator which in the space \( \{ x \otimes z \} \) is given by formula

\[
U(x, z; x_i, z_i|t) = \langle x, z \mid (Te^{-\int_0^t d\tau \hat{\Lambda}(\tau)} \rangle |x_i, z_i \rangle,
\]

(7)

where the average (denoted as \( \langle \ldots \rangle \)) is taken over trajectories of the stochastic Markovian process in \( \{ z \} \)-space with fixed initial \( (z_i) \) and final \( (z) \) coordinates. In particular, the PDF of interest, \( \Phi_F(x, z_i|t) \) averaged over \( F(t) \)-fluctuations, can be calculated as

\[
\Phi_F(x, z_i|t) = \int dz \int dz_i U(x, z; x_i, z_i|t) \sigma_i(z_i).
\]

(8)

The important point of the proposed representation consists in the fact that for Markovian processes in \( \{ x \otimes z \} \)-space the operator \( \hat{U} \) satisfies the Markovian SLE with time-independent operators

\[
\dot{\hat{U}} = -\{ \hat{\Lambda}_1 + \hat{L} \} \hat{U}
\]

(9)

with \( U(x, z; x_i, z_i|0) = \delta(x-x_i)\delta(z-z_i) \).

Thus we have reduced the problem to solving the SLE \( \Phi \) with time-independent operators, though at the cost of the extension of the space of the process, which describes the evolution of the system.

Noteworthy is that the representation \( \Phi \) is valid not only for stochastic functions \( z(t) \) but also for dynamical ones, which are known to be Markovian as well. For example, in the model of harmonically oscillating force:

\[
z(t) = z_c(t) = z_0 \sin(\omega t + \varphi),
\]

(10)

the dependence \( z_c(t) \) can be considered as a coordinate part of the trajectory of dynamical motion (in the harmonic potential), described by the operator

\[
\hat{L} = -(v \nabla z + \omega^2 z \nabla \psi_z),
\]

(11)

in the phase space \( \{ z \} \) \( = (z, v_z) \) \( (v_z = \dot{z} \) is the velocity) with \( \psi_z(z) = \delta(z-z_0 \sin \varphi)\delta(v_z-z_0 \cos \varphi) \). Evidently, the case of \( z(t) \) represented as a linear combination of, say, \( N \) oscillating functions \( z_j(t) = z_{j0} \sin(\omega_j t + \varphi_j) \): \( z(t) = \sum_{j=1}^N z_j(t) \), can be modeled by coupling to \( N \) harmonic coordinates \( z_N = (z_1, z_2, \ldots, z_N) \).

### III. NON-MARKOVIAN CTRW.

#### A. Markovian representation.

The main goal of this work is the analysis of the effect of time-dependent field on CTRW-type (subdiffusive) migration.

In the CTRW approach the stochastic motion in \( \{ x \} \)-space is treated as a set of jumps with jump statistics described by the waiting time distribution \( W(t) \). For time-independent driving force the non-Markovian equation for the PDF \( \rho(x, t) \) is in conventionally derived by summing up the contributions of all sets of jumps. In terms of the Laplace transform \( R(e) = \int_0^\infty dt \rho(t)e^{-et} \) this equation is written as

\[
\epsilon R(e) = \rho_i - M(e) \hat{\Lambda}_e R(e).
\]

(12)

In this equation \( \rho_i(x) \) is the initial PDF and

\[
M(e) = [1 - \hat{\Lambda}W]/[\epsilon \hat{\Lambda}W],
\]

(13)

where \( \hat{\Lambda}W = \int_0^\infty dt W(t)e^{-et} \). Note that in the case of subdiffusion, when \( M(e) = e^{1-\alpha} \), eq. \( \Phi \) reduces to the Laplace transform variant of the FSE \( \Phi \).

CTRW-type processes can conveniently be analyzed within the Markovian representation \( \Phi \) in which these processes are assumed to result from jump-like \( \hat{\Lambda}_1(t) \)-fluctuations determined by the dependence of \( \hat{\Lambda}_1(y(t)) \) on some Markovian stochastic variable \( y(t) \) whose PDF \( \eta(y, t) \) satisfies equation

\[
\dot{\eta} = -\hat{\Lambda}\eta, \text{ with } \eta(y(0) = \eta_i(y).
\]

(14)

In this equation \( \hat{\Lambda} \) is a linear operator in \( \{ y \} \)-space and \( \eta_i(y) \) is the initial condition \( \int dy \eta_i(y) = 1 \).

The dependence \( \hat{\Lambda}_1(y) \) is taken in the form \( \hat{\Lambda}_1(y(t)) \equiv \delta(y_0 - y(t))\hat{\Lambda}_1 \), where \( y_0 \) is the coordinate at which the system undergoes the jump described by \( \hat{\Lambda}_1 \). Similar to the above-considered model of \( z(t) \)-modulation of \( \hat{\Lambda}_1 \), in the case of \( y(t) \)-modulation the evolution of the system is described by the PDF \( p(x, y(t)) \) in the combined space \( \{ x \otimes y \} \). This PDF satisfies the SLE of type of eq. \( \Phi \), which as applied to the Laplace transform

\[
P(e) = \int_0^\infty dt p(t)e^{-et}
\]

(15)

Of special interest is the PDF averaged over \( y(t) \)-process

\[
\Phi_y(x, y_i|t) = \int dy \int dy_i P(x, y; y_i, y_i|t)\eta_i(y_i).
\]

(16)

The \( y(t) \)-controlled (or modulated) process in \( \{ x \} \)-space proves to be of CTRW type. Thus obtained CTRW depends on the initial condition \( p_i(y) \) and the form of the operator \( \hat{\Lambda} \). In what follows we will consider the non-stationary variant realized for \( p_i(y) = \delta(y-y_0) \). In this variant the average PDF \( \Phi_y(x, y_i|t) \) is known to satisfy the CTRW-like equation usually written as applied to \( R(e) = \hat{R}_y(e) = \int_0^\infty dt \Phi_y(t)e^{-et} \)

\[
\epsilon R(e) = \rho_i - M(e) \hat{\Lambda}_e R(e),
\]

(17)

in which

\[
M(e) = \hat{M}(e) = (D_1/D_\alpha)e(y_0)(e + \hat{\Lambda})^{-1}|y_0|.
\]

(18)

The behavior of \( \hat{M}(e) \) is completely determined by the specific features of the controlling process \( \Phi \). Various models of this process are discussed in ref. \( \Phi \).
IV. CTRW-BASED SLE.

The Markovian representation is very suitable for treating the effect of time-dependent field on CTRW-like processes.

The corresponding equation is straightforwardly derived by taking into account that, in accordance with the Markovian representation, this equation describes Markovian process in \((x \otimes y)\)-space affected by the driving force which can be modeled by interaction with the Markovian \(z(t)\) variable. This means that the equation sought is actually the Markovian SLE \([8]\) for the PDF \(q(r; r_i|t)\) in the extended space \(\{r\} \equiv \{x \otimes y \otimes z\}\)-space. For the Laplace transform \(Q(\epsilon) = \int_0^\infty dt q(t)e^{-\epsilon t}\) this equation is written as:

\[
\epsilon Q = \delta_{rr} - (\hat{\Lambda} + \delta_{yy}\hat{L}_1 + \hat{L})Q
\]

(19)

This equation is seen to differ from eq. \([15]\) for \(P(x, y|\epsilon)\) only in the replacement \(\epsilon = \hat{\Omega} = \epsilon + \hat{L}\) and the evident change of \(\delta\)-function describing the initial condition. Naturally, for the PDF averaged over \(y(t)\)-process:

\[
R(\epsilon) = \overline{P_y}(\epsilon) = \int_0^\infty dt \overline{P_y}(t)e^{-\epsilon t},
\]

(20)

one gets CTRW-like equation (sometimes called the non-Markovian SLE \([7]\)) similar to eq. \([17]\)

\[
[\hat{\Omega} + \hat{L}_a M(\hat{\Omega})]R = \rho_\omega \sigma_\omega \text{ with } \hat{\Omega} = \epsilon + \hat{L}.
\]

(21)

Notice that the order of operators \(\hat{L}_a\) and \(M(\hat{\Omega})\) is important since they do not commute with each other.

To qualitatively interpret eq. \([21]\) within the CTRW approach it is worth noting that, according to the SLE representation \([9]\), the time-dependent-field affected CTRW can be considered as sequence of jumps governed by the \(z\)-dependent operator \(\hat{L}_1(z)\). The CTRW process is accompanied by the simultaneous evolution of the parameter \(z(t)\), determined by the operator \(e^{-L_1t}\). This operator will enter in formulas describing CTRW evolution in the form of the product \(W(t)e^{-L_1t}\) which for the Laplace transform \(\hat{W}(\epsilon)\) just corresponds to the replacement \(\epsilon\) by \(\hat{\Omega} = \epsilon + \hat{L}\) in \(\hat{W}(\epsilon)\) thus resulting in eq. \([21]\).

Exact eq. \([21]\) essentially differs from that proposed earlier \([5, 6]\) to treat the effect of time-dependent field, i.e. the results of these works are, in general, incorrect, except, may be, some special cases (see below).

A. General results.

In this brief communication we will restrict ourselves to analyzing the time evolution of the moments of the PDF \(\overline{P}_{\gamma z}(x|t)\) (averaged over \(y(t)\)- and \(z(t)\)-processes) \(m_n(t) = \int dx x^n \overline{P}_{y z}(t)\). For this analysis we need to specify the initial PDF \(\rho_0(x)\). For simplicity we will assume \(\rho_0(x) = \delta(x)\).

Instead of moments \(m_n(t)\), it is more convenient to analyze their Laplace transforms \(\tilde{m}_n(\epsilon)\) which can be expressed in terms of moment operators in the \(\{z\}\)-space

\[
\dot{\tilde{m}}_n(t) = \int dx x^n \overline{R}_y(x, \hat{\Omega}):
\]

(22)

\[
\tilde{m}_n(\epsilon) = \langle \tilde{M}_n \rangle_z = \int dz \int dz' \langle |\tilde{M}_n (z)| \sigma(z) \sigma(z')\rangle.
\]

(23)

As is seen from eq. \([21]\) the operators \(\tilde{M}_n(\epsilon)\) satisfy simple equations

\[
\dot{\tilde{M}}_n = n f z M(\hat{\Omega}) M_{n-1} + n(n - 1) D_\alpha \tilde{M}_{n-2}
\]

(24)

for \(n \geq 2\) with \(\tilde{M}_{-1} = 0\) and \(\tilde{M}_0 = \hat{\Omega}^{-1}\), in which \(f = D_n F_0 > 0\). Solution of these equations and substitution into eqs. \([23]\) yields for Laplace transforms of time derivatives of the moments

\[
\tilde{m}_1(\epsilon) = f \langle z \Phi(\hat{\Omega}) \rangle_z, \quad \tilde{m}_2(\epsilon) = \tilde{m}_0(\epsilon) + \tilde{m}_2(\epsilon),
\]

(25)

with

\[
\tilde{m}_0(\epsilon) = 2 D_\alpha \langle \Phi(\hat{\Omega}) \rangle_z, \quad \tilde{m}_2(\epsilon) = f^2 \langle (z \Phi(\hat{\Omega}))^2 \rangle_z.
\]

(26)

Here \(\Phi(\hat{\Omega}) = \hat{\Omega}^{-1}M(\hat{\Omega})\).

In what follows we will concentrate on the analysis of force dependent terms. The force independent contribution \(\tilde{m}_0(\epsilon)\) was discussed in detail earlier \([4]\).

After the inverse Laplace transformation of expressions \([23]\) and \([26]\) one gets \(\tilde{m}_0(t) = 2 D_\alpha f(t)\),

\[
\tilde{m}_1(t) = \int_0^t dt' \int dz dz' z g(z, z'|t) \sigma(z),
\]

(27)

\[
\tilde{m}_2(t) = f^2 \int_0^t dt' \int dz dz' z z' \sigma(z),
\]

(28)

In these formulas

\[
g(z, z'|t) = \phi(t) \langle |z| e^{-L_1t} \rangle,
\]

(29)

where \(\langle |z| e^{-L_1t} \rangle\) is controlled by the model of \(z(t)\)-modulation, while \(\phi(t) = (2\pi i)^{-1} \int_{-i}^{i} \overline{d\epsilon} e^{-\epsilon \hat{M}(\epsilon)}\) is determined by the CTRW model considered and for the subdiffusion model \([M(\epsilon) = e^{\epsilon - \alpha}, \alpha < 1]\)

\[
\phi(t) = \Gamma^{-1}(\alpha) t^{\alpha - 1}.
\]

(30)

B. Applications

a. Harmonically oscillating force. In the model of harmonically oscillating force \([10]\) in which \(L \ [eq. \([11]\)]\) describes dynamical motion in the harmonic potential, one gets \(\langle |z| e^{-L_1t} \rangle = \delta(z - z_c(z)|t)\), where \(z_c(z|t) = (z_c(z|t), v_{z_c}(z|t))\) is the trajectory of dynamical motion with \(z_c(t = 0) = z\) in the phase space \(\{z\}\).

Substituting this formula into eqs. \([27]\)-\([29]\) one obtains \(\mu_0(t) = 2 D_\alpha \Gamma^{-1}(1 + \alpha) t^\alpha\),

\[
m_1(t) = f \int_0^t dt' z_c(t') \phi(t'),
\]

(31)

\[
\mu_2(t) = f^2 \int_0^t dt' z_c^2(t') f \int_0^t dt'' z_c(t'') \phi(t - t'') \phi(t''),
\]

(32)
where \( z_c(t) \) is given by eq. \((10)\).

For brevity, we will restrict ourselves to the discussion of the long time behavior of the moments only:

\[
\begin{align*}
m_1(t) &\xrightarrow{t \to \infty} (f/\omega^\alpha) \sin(\varphi + \pi \alpha/2) \\
m_2(t) &\xrightarrow{t \to \infty} \gamma_2(\alpha) f^2(t/\omega)^\alpha
\end{align*}
\]

where \( f = D_\alpha F_0 z_0 \) and \( \gamma_2(\alpha) = \cos(\pi \alpha/2)/[2\Gamma(1 + \alpha)] \).

These formulas predict some peculiarities of the subdiffusion response to oscillating force. First, \( m_1(t) \) appears to be nonzero with asymptotic value \((at t \to \infty)\) independent of time and harmonically oscillating as a function of the initial phase \( \varphi \) of force oscillations. Second, the force dependent part of \( m_2(t) \) is anomalously large increasing in time so that \( m_2(t)/m_0(t) = \cos(\pi \alpha/2)[f^2/(4D_\alpha \omega^\alpha)] \) independent of time. Third, in the case of conventional diffusion, i.e. at \( \alpha \to 1 \), \( m_2(t)/m^* \to 0 \), as should be.

The exact formulas \((33)\) and \((34)\) significantly differ in their analytical form from those derived earlier with not quite correct kinetic equation \([5] \), though, surprisingly, the results obtained with this equation appeared to be qualitatively correct.

b. Stepwise oscillating force. Here we will briefly discuss the model of stepwise oscillating force to check the results obtained in ref. \([6] \) with the equation which is not quite correct, in general. The model is defined as \( z(t) = z_0(1 - |2t/\tau_0|) \), where \( \tau_0 \) is the oscillation period and \([x]\) denotes the integer part of \( x \). It can also be represented as

\[
z(t) = z_c(t) = \sum_{n=-\infty}^{\infty} z_n e^{in\omega t}, \tag{35}
\]

where \( \omega = 2\pi/\tau_0 \) and \( z_n \) are given by: \( z_{2n} = 0 \), \( z_{2n+1} = -2i/(\pi(2n + 1)) \) with \( z_{-n} = z^*_n \).

As mentioned above, the case of \( z_c(t) \) represented as a superposition of harmonically oscillating functions can be described by assuming \( z_c(t) \)-modulation to result from dynamical motion in a harmonic potential in the multidimensional space \( z = (z_1, v_{z_1}; \ldots; z_n, v_{z_n}; \ldots). \) In this case formulas \((27)-(28)\) predict the same expressions \((31)-(32)\) for the moments. Evaluation with these expressions yields: \( m_1(t) \xrightarrow{t \to \infty} \bar{\gamma}_1(\alpha)(2f/\omega^\alpha) \), and

\[
m_2(t) \xrightarrow{t \to \infty} \bar{\gamma}_2(\alpha) f^2(t/\omega)^\alpha. \tag{36}
\]

Here the functions \( \bar{\gamma}_1(\alpha) \) and \( \bar{\gamma}_2(\alpha) \) are defined as:

\[
\begin{align*}
\bar{\gamma}_1(\alpha) &= \zeta(1 + \alpha)(2 - 2^{-\alpha}) \sin(\pi \alpha/2), \\
\bar{\gamma}_2(\alpha) &= \zeta(2 + \alpha)(4 - 2^{-\alpha})/[4\Gamma(1 + \alpha)],
\end{align*}
\]

where \( \zeta(x) \) is the Riemann’s zeta-function.

Obtained results agree with those of ref. \([6] \) thus confirming the correctness of the method proposed in this work as applied to the model of stepwise oscillating force.

c. Fluctuating force. Of great interest is also the case of fluctuating force \( F(t) \), i.e. fluctuating \( z(t) \). This case is conveniently analyzed using eqs. \((25)\) and \((26)\) for the Laplace transforms.

For brevity, we will analyze only the long time limit. In addition, for simplicity, we will assume that the initial condition \( \sigma_i(z) \) in \((z)\)-space is equilibrium i.e. \( \bar{\sigma}_i = 0 \), and \( m_1(0) = \langle z_0 \rangle = 0 \). In such a case the first moment \( m_1(t) = m_1(0) = 0 \) so that the only value to be analyzed is \( \mu_2(\epsilon) \). At small \( \epsilon \to 0 \) this term can be estimated as

\[
\bar{\mu}_2(\epsilon) \approx f^2 e^{-1} \Phi(\epsilon) \langle |z| z \Phi(\Omega)z \rangle |e_z\rangle, \tag{39}
\]

where \( |e_z\rangle \) and \( \langle e_z| \) are the equilibrium states of the operator \( \hat{L} \) (in this bra-ket notation \( \langle e_z| \equiv \int dz\hat{z}[\hat{z}]^{\dagger} \) and \( |\sigma_i\rangle = |e_z\rangle \) therefore

\[
\mu_2(t) \xrightarrow{t \to \infty} \Gamma^{-1}(1 + \alpha) f^2(\tilde{\tau} t)^\alpha \tag{40}
\]

with \( \tilde{f} = D_\alpha F_0 \sqrt{\langle z^2 \rangle} \) and the parameter \( \tilde{\tau} \) is expressed in terms of the correlation function \( \bar{K}(t) = \langle zz(t)\rangle = \langle e_z|ze^{-Lt}z|e_z\rangle \):

\[
\tilde{\tau}^\alpha = \int_0^\infty dt \bar{K}(t)\phi(t)/(z^2). \tag{41}
\]

It is seen that the characteristic features of \( \mu_2(t) \) are similar to those obtained in the case of oscillating force with \( f \) and \( \omega \) replaced by \( \tilde{f} \) and \( \tilde{\tau}^{-1} \), respectively. Note-worthy is, however, that unlike this case, for fluctuating force \( \mu_2(t)/t^\alpha \) is finite as \( \alpha \to 1 \).

V. CONCLUDING REMARKS.

We have analyzed the response of CTRWs on time-dependent field using the rigorous method based on the Markovian representations of CTRW and the modulated field. This method is applied to describing the field effect on subdiffusive motion.

Obtained formulas \((33)-(40)\) clearly demonstrate some specific features of the response of anomalous subdiffusive systems: 1) in the case of oscillatory force modulation the first moment (average displacement) is, in general, non-zero (even in the long time limit) and depends on the oscillation phase, 2) the modulated force results in the anomalously strong contribution to the second moment (dispersion) growing in time.

In conclusion, it is worth noting that the proposed Markovian SLE approach \([9] \) for describing the influence of modulated external fields is applied to only one particular problem of the theory of force induced effects in stochastic systems. This approach is, however, fairly general and can be very suitable in studying many time-dependent-field affected stochastic processes \([1]-[3] \) since it reduces the study to the analysis of characteristic features of time-independent operators (their spectra, eigenfunctions, etc.). Some of applications of the SLE approach \([9] \) are currently under consideration.
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