Temporal fractal model for the anomalous dielectric relaxation of inhomogeneous media with chaotic structure

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The potential of the fractional derivative technique is demonstrated on the example of derivation of all three known patterns of anomalous, nonexponential dielectric relaxation of an inhomogeneous medium in the time domain. It is explicitly assumed that the fractional derivative is related to the dimensionality of a temporal fractal ensemble (in a sense that the relaxation times are distributed over a self-similar fractal system). The proposed fractal model of a microstructure of inhomogeneous media exhibiting nonexponential dielectric relaxation is built by singling out groups of hierarchically subordinated ensembles (subclusters, clusters, superclusters, etc.) from the entire statistical set available. Different relaxation functions are derived assuming that the real (physical) ensemble of relaxation times is confined between the upper and lower limits of self-similarity. It is predicted that at times, shorter than the relaxation time at the lowest (primitive) self-similarity level, the relaxation should be of a classical, Debye-like type, whatever the pattern of nonclassical relaxation at longer times.

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I. INTRODUCTION

Anomalous (nonexponential) relaxations have long been and still are a hot topic in the physics of inhomogeneous media [1–26]. Broadly speaking, one may refer to three general relaxation laws that are encountered in the experimental studies of complex systems:

(i) stretched exponential [1,2,12]:

$$f(t)\approx\exp\left[-\left(\frac{t}{\tau}\right)^{\beta}\right],\quad 0<\beta<1,\ t>\tau, \tag{1.1}$$

(ii) exponential-logarithmic [3–5]:

$$f(t)\approx\exp\left[-B\ln^{\alpha}\left(\frac{t}{\tau}\right)\right], \tag{1.2}$$

(iii) algebraic decay [8]:

$$f(t)\approx\left(\frac{t}{\tau}\right)^{-\alpha}, \tag{1.3}$$

where α's, β's, τ's, and B are the appropriate fitting parameters.

Currently, there seems to be no quantitative microscopic theory for the cited laws [6,7,15]; moreover, sometimes even the possibility of such a theory is denied [13–15]. The main argument is that a spatial inhomogeneity (such as, e.g., a random distribution of impurities within a matrix, or of interatomic spacings in amorphous semiconductors) will necessarily result in an extremely broad range of microscopic transition rates. Hence, a spatial disorder is expected to induce a temporal energetic disorder.

Another approach to the problem of anomalous relaxations makes use of fractal concepts [9–11,22–25]. In this case, the problem is analyzed using the mathematical language of fractional derivatives [16,22–25] based on the Riemann-Liouville fractional differentiation operator [27–29]

$$D^{\alpha}[f(t)]=\frac{1}{\Gamma(1-\alpha)}\frac{d}{dt}\int_{c}^{t}(t-\tau)^{-\alpha}f(\tau)d\tau, \tag{1.4}$$

where $\Gamma(x)$ is the gamma function

In spite of a reasonable success of the latter approach, use of the fractional derivative as represented by Eq. (1.4) makes the interpretation of differentiation procedures difficult (for example, the nonzero value of a fractional derivative of a constant), as well as their relevance to the assumed fractal ensemble. One may also note that so far the fractional derivatives were analyzed in essentially phenomenological terms; moreover, the equations based on fractional derivatives were constructed more by intuition (guessed), rather than obtained by derivation.

The fractional derivative technique is used for description of different physical phenomena (e.g., [30–38]). Apparently, Blumen *et al.* [11] were the first to use fractal concepts for the analysis of anomalous relaxations. The same problem was treated in [12,16,22–25] using the fractional derivative approach. Excellent review of the use of fractional derivative operators for the analysis of various physical phenomena can be also found in [30]. However, up until now there seems to be little understanding of the relationship between the fractional derivative operator and/or differential equations derived therefrom (which are used for the description of various transport phenomena such as transport of a quantum

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particle through the potential barrier in fractal structures, or transport of electromagnetic waves through a medium with a fractal-like profile of dielectric permittivity, etc.), and the fractal dimensionality of a medium.

In this context, attempts to construct fractional derivatives and to clarify their relevance to the assumed fractal ensemble are believed to remain feasible for the treatment of the problem of anomalous relaxations. In our previous papers [37], the analysis of a classical problem of polarization of an inhomogeneous medium permitted us to establish the relationship between anomalous relaxation and dimensionality of a temporal fractal ensemble, which characterizes a nonequilibrium state of a medium. Thus, the main aim of the present paper is the further extension and generalization of these results within the frame of a fractal model.

II. GENERAL OVERVIEW: DERIVATIVE OF FRACTAL FUNCTIONS

In general, functions for which the total increment

$$\Delta_h f(x) = f(x+\Delta x) - f(x) \tag{2.1}$$

can be represented as

$$\Delta_h f = A[\Delta x]^h + \alpha(x)[\Delta x]^h, \quad (\lim_{\Delta x \to 0} \alpha(x) = 0) \tag{2.2}$$

may be subdivided into two classes:

(i) $h=1;0: f(x)$ belongs to the classical ensemble of differentiated functions; and

(ii) $h \neq 1$ (Hoelder index): $f(x)$ belongs to the ensemble of functions for which it is not the classical derivative but only the fractional derivative that exists,

$$\frac{d^h f(x)}{dx^h} = \lim_{\Delta x \to 0} \frac{\Delta_h f}{[\Delta x]^h}. \tag{2.3}$$

It is pertinent to recall here that fractals are defined, sometimes, as continuous functions characterized by the absence of derivatives (tangents) at any point, with a curvilinear cone serving as a tangent to the fractal curve trajectory [38]. Apparently, Czech scientist Bolzano was the first to study such continuous, nondifferentiated functions around 1830 (the corresponding manuscript was discovered only in 1920 [39]). Wiener's process (i.e., Brownian motion) and Kolmogorov's turbulence (i.e., nonsmooth vector field) may be cited as examples of phenomena that can be described by continuous, nondifferentiated functions (fractal functions).

The displacement $y(t)$ of a Brownian particle in the former (Wiener's) process is defined as [40]

$$|y(t+\Delta t) - y(t)| \approx [\Delta t]^{\alpha}, \tag{2.4}$$

whereas the singular velocity of the latter phenomenon (Kolmogorov's turbulent flow) is characterized by [41]

$$\langle |\Delta \vec{\nu}|^p \rangle \approx [\Delta x]^{p/3}, \tag{2.5}$$

where $\Delta \vec{\nu} = \vec{\nu}(x+\Delta x) - \vec{\nu}(x)$ is the difference of velocities between two points separated by distance Δx.

Assume that a function $f(x)$ is defined on a fractal ensemble Ω_f, of dimensionality $0 < d_f < 1$. Let the function $f(x)$ (hereafter referred to as a fractal function) be continuous through Ω_f, be self-similar at different scales, and have no tangent at any point of its trajectory. It is assumed that $f(x)=0$ if $x<0$, and $|f(x)|<\infty$.

Let us divide a segment $[x,x_0]$ so that the length of each qth fragment at the nth scale level is

$$\Delta x_q^{(n)} = \xi^n (x_0 - x), \tag{2.6}$$

where $\xi<1$ is the scaling factor (i.e., the index of similarity of the ensemble Ω_f,).

The number of dividing points of the segment $[x,x_0]$ at the nth step is therefore,

$$m_n = 1,2,\ldots,j^{n+1}, \tag{2.7}$$

where j is the number of blocks (i.e., the branching index) involved in the construction of the fractal unit cell ($j=2$ for the Cantor's ensemble).

Let the unit scale at the nth step be Δx^{α},

$$[\Delta x_q^{(n)}]^{\alpha} = \frac{1}{N_n}(x_0 - x)^{\alpha}, \tag{2.8}$$

where $N_1 = j^1, \ldots; N_n = j^n$ (that is, $N_n = j^n$ determines the number of fragments at the nth scale level). This definition of the unit scale for the segment $[x,x_0]$ allows us to associate each point (element) of the fractal ensemble with a point of an ultrametric space which can be represented by the Cayley Tree [11,42].

It follows from Eq. (2.8) that $\lim_{n\to\infty} \Delta x_q^{(n)} = 0$; hence, $\Delta x_q^{(n)}$ is an infinitesimal quantity. From now on, the increment of the function argument $\Delta x_q^{(n)}$ at the nth step will be denoted as Δx (that is, $\Delta x = \Delta x_q^{(n)}$), while the corresponding coordinates of dividing points will be defined as

$$x_q = x_0 - q\Delta x_q^{(n)} = x_0 - q\Delta x, \tag{2.9}$$

where $q = 0,1,2,\ldots,j^{n+1}$. Recognition of fractal dimensionality as $d_f = \alpha$ implies, further,

$$\Delta x = (x_0 - x) \Big/ \left(\frac{1}{\xi}\right)^n, \quad [\Delta x]^{\alpha} = [(x_0 - x)^{\alpha}/N_n],$$

$$\left(\frac{1}{\xi}\right)^{n\alpha} = j^n = N_n. \tag{2.10}$$

Consider an increment $\Delta_{\alpha} f(x) = f(x_0) - f(x_0 - \Delta x)$; then the qth increment $\Delta_{\alpha}^q f(x)$ will be determined through binomial coefficients with alternating signs [37]

$$\Delta_{\alpha}^m f(x_0) = \sum_{q=0}^{m} [-1]^q C_m^q (f(x_0 - q\Delta x)),$$

$$C_m^q = \frac{m!}{q!(m-q)!}, \quad m = j^{n+1}, \tag{2.11}$$

and the function $f(x)$ in the vicinity of point x_0 will be

$$f(x)=(1-\Delta_\alpha)^m f(x_0). \tag{2.12}$$

Using Eqs. (2.6)–(2.12), one can derive an analog of the Taylor series for function $f(x)$

$$f(x)=\sum_{q=0}^{\infty} a_q (x_0-x)^{\alpha q}, \tag{2.13}$$

where $a_q=(j^q/q!)f^{(\alpha q)}(x_0)$, and $f^{(\alpha q)}(x_0)$ defines the fractional derivative of qth order of the fractal function $f(x)$ at the point $x=x_0$ as

$$f^{(\alpha q)}(x_0)=\lim_{\Delta x\to 0}\frac{\Delta_\alpha^q f(x_0)}{([\Delta x]^\alpha)^q}. \tag{2.14}$$

The coefficients of the series Eq. (2.13) depend both on the fractional derivative of the qth order of the fractal function $f(x)$ at the point $x=x_0$ and on the branching index j of the fractal ensemble for which the function $f(x)$ is specified.

It follows from Eq. (2.13) that the first derivative ($q=1$) is

$$\frac{d^\alpha f(x_0)}{dx^\alpha}=f^{(\alpha)}(x_0)=\lim_{\Delta x\to 0}\frac{\Delta^\alpha f(x_0)}{[\Delta x]^\alpha}=\lim_{\Delta x\to 0}\frac{f(x_0)-f(x_0-\Delta x)}{[\Delta x]^\alpha}. \tag{2.15}$$

thus, Eq. (2.3) is recovered.

In a similar way, one can also specify the integral of function $f(x)$ on fractal ensemble Ω_f as a limit of integral summation [37]

$$\int_y^x f(t)[dt]^\alpha=\lim_{\Delta t\to 0}\sum_{q=1}^{n} f(y-(q-1)\Delta t)[\Delta t]^\alpha, \qquad \lim_{\Delta t\to 0}(n[\Delta t]^\alpha)=(x-y)^\alpha. \tag{2.16}$$

For convenience in the further use of a fractional derivative, let us introduce the differentiation operator D^α_{x-y}:

$$D^\alpha_{x-y} f(x)=\lim_{x\to y}\frac{f(x)-f(y)}{[x-y]^\alpha}, \quad 0<\alpha\leqslant 1.$$

The integration operator I^α_{x-y} will be defined as

$$I^\alpha_{x-y} f(x)=\int_y^x f(t)[dt]^\alpha, \tag{2.17}$$

$$I^\alpha_{x-y} f(x)=D^{-\alpha}_{x-y} f(x). \tag{2.18}$$

Consider the function

$$\Phi_y^{(\alpha)}(x)=\frac{1}{\Gamma(\alpha)}\int_y^x (x-t)^{\alpha-1}[f(x)-f(t)]dt. \tag{2.19}$$

Taking into account that $t=(x-y)z+y$, and

$$\Phi_y^{(\alpha)}(x)=\frac{1}{\Gamma(\alpha)}\int_y^x (x-t)^{\alpha-1}[f(x)-f(t)]dt=\frac{1}{\Gamma(\alpha)}(x-y)^\alpha\int_0^1 (1-z)^{\alpha-1}[f(x)-f^*(z)]dz,$$

$$f^*(z)=f[(x-y)z+y],$$

it can be shown that

$$D^{1-\alpha}_{x-y} f(x)=D^1_{x-y}\Phi_y^{(\alpha)}(x) \tag{2.20}$$

or

$$D^\beta_{x-y} f(x)=D^1_{x-y}\Phi_y^{(1-\beta)}(x),$$

where $\alpha+\beta=1$, $0<\alpha\leqslant 1$, and $f(x)=0$, if $x<0$.

If $f(y)=0$, the following should hold:

$$D^{-\alpha}_{x-y} f(x)=\Phi_y^{(\alpha)}(x), \quad f(x)=D^\alpha_{x-y}\Phi_y^{(\alpha)}(x), \quad f(x)=D^{-\alpha}_{x-y}D^\alpha_{x-y} f(x), \quad D^\alpha_{x-y} f(x)=D^{2\alpha}_{x-y}\Phi_y^{(\alpha)}. \tag{2.21}$$

Summarizing, the developed fractional differential-integral concepts establish the link with the procedure of construction of the fractal ensemble that determines the function $f(x)$.

III. DIELECTRIC RELAXATION

The potential of fractional derivatives will become evident, and the relationship between the exponents β and α in Eqs. (1.1)–(1.3) and the fractal dimensionality d_f will be established, in the subsequent treatment of the classical problem of polarization $P(t)$ of a dielectric medium (which is, in fact, equivalent to the general problem of relaxation of internal parameters of a nonequilibrium phase).

Assume that $P(t)$ contains two contributions [7]

$$P(t)=P_0+P_1(t), \tag{3.1}$$

where the former one ($P_0=\chi_0 E$) varies exactly (at least, with negligibly small retardation) as the applied field E, while the latter time-dependent one $P_1(t)$ is retarded. Let $P^*=\chi_\infty E$ be the upper limit (at fixed E); then the instantaneous rate of approach of the contribution $P_1(t)$ to this limit will be higher, the larger the amplitude $(\chi_\infty E-P(t))$. Hence, the corresponding relaxation equation may be written as

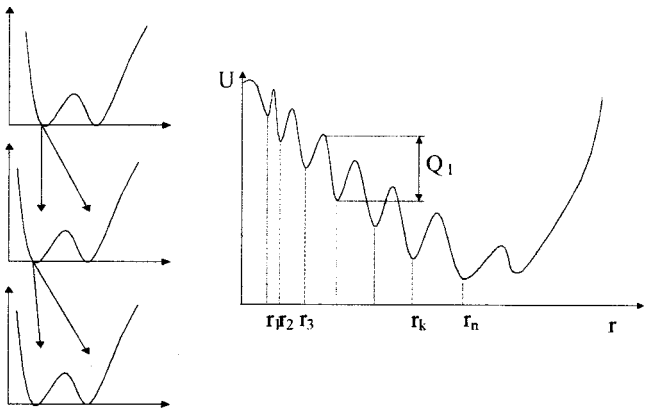

FIG. 1. Schematic of the potential barrier landscape.

$$\frac{dP_1(t)}{dt}=\frac{1}{\tau}(\Delta\chi E-P_1(t)).$$
$$\Delta\chi=\chi_\infty-\chi_0, \tag{3.2}$$

where τ is the relaxation time. After integration, one derives from Eq. (3.2)

$$P(t)=P_0+P_1(t)=\left[\chi_0+\Delta\chi\left(1-\exp\left(-\frac{t}{\tau}\right)\right)\right]E \tag{3.3}$$

(for the field fixed at E), and

$$P(\omega)=P_0+P_1(\omega)=[\chi_0+\Delta\chi/(1+i\omega\tau)]E \tag{3.4}$$

(for the field alternating as $E=E_0e^{i\omega t}$).

Therefore, the dielectric permittivity of a medium may be defined, finally, as [7]

$$\varepsilon=\varepsilon_\infty+\frac{\varepsilon_0-\varepsilon_\infty}{1+i\omega\tau}, \tag{3.5}$$

where

$$\varepsilon_\infty=\lim_{\omega\to\infty}\varepsilon;\quad \varepsilon_0=\varepsilon|_{\omega=0}.$$

The next issue concerning us will be the case of an anomalous relaxation in which the ''smearing out'' of relaxation spectrum (i.e., the deviation of complex susceptibility from its Debye form) is associated with the concept of relaxation times distribution (RTD). As is well known, this concept implies a continuous distribution of dipoles by their relaxation times.

Consider the Froelich's relaxation model [43] which is based on the RTD concept. It is usually assumed that the relaxators are homogeneously distributed along the height of the potential barrier U; however, this assumption is not strictly correct. As can be seen from Fig. 1, many shallower minima may exist between two main minima; therefore, a system is involved in a continuous chain of transitions $r_1 \to r_2 \to r_3 \to \ldots \to r_k \to r_n$ between adjacent minima (Fig. 1). Let $S_l(t)$ be the probability of transition from the lth minimum into the $(l+1)$th one, so that

$$S(t)=e^{-(t/\tau)l}, \tag{3.6}$$

where the relaxation time is

$$\tau_l=\tau_0\exp\left(\frac{Q_l}{kT}\right), \tag{3.7}$$

Q_l is the barrier height, k is the Boltmann's constant, and T is the absolute temperature.

Implicit in the RTD concept is the assumption of comparable magnitudes of barrier heights Q_l (Fig. 1); hence, the characteristic times $\Delta t_l=t_{l+1}-t_l$ of transitions over the barriers will also be of comparable magnitudes. Therefore, during the time $t\gg n\tau_l$ a system will pass through n barriers with a probability $1-S(t)$ so that

$$S(t)=\prod_{l=1}^{n} S_l,$$
$$S_l=\exp(-\Delta t_l/\tau_l). \tag{3.8}$$

Assuming $\Delta t_l=t/n$, it becomes clear that the dispersion of intervals Δt_l may be neglected in the limit of $n\gg 1$; as a result, Eq. (3.8) will regain its Debye form with the mean relaxation time $\langle\tau\rangle$ defined as

$$\langle\tau\rangle^{-1}=\frac{1}{n}\sum_{l=1}^{n}\tau_l^{-1}. \tag{3.9}$$

Thus, the chain of transitions considered above is effectively reduced to the exponential, Debye-like relaxation with the mean relaxation time $\langle\tau\rangle$. In other words, the concept of RTD implies the Debye-like relaxation of a system. However, it is evident that the relaxation will become nonexponential, should a system be characterized by a complex susceptibility of, say, Cole-Cole type.

Thus, it can be conclude that the RTD concept applies in the case of a Debye-like relaxation (even though its frequency dependence may be smeared out), whereas it becomes inapplicable in the case of still slower relaxation patterns. In this latter case, the distribution of relaxation times over a self-similar, fractal ensemble seems a physically more reasonable assumption. As is well known, the fractality of geometrical objects implies their noninteger dimensionality; however, a more exact definition of the fractal concept with respect to the ensemble of relaxation times is in order.

As proved by Nigmatullin [22–25], fractional derivatives by time in Newton's equations imply that the interactions between a system and an external field are not continuous but occur in discrete time intervals. In this context, the fractality of an ensemble of relaxation times simply means that the relaxation is not a single process with a unique relaxation time, but a series of successive relaxation events with different relaxation times.

Let us consider now the nonequilibrium state of a fractal-like medium assuming that this nonequilibrium state is char-

acterized by many events such that each next event is separated by a certain time interval τ_i from a previous event. In this case, some intervals will be eliminated from a continuous process of system evolution by a definite law. Assume that such a process is caused by a temporal fractal state of dimensionality d_f; the corresponding relaxation equation can be written as

$$D^{\alpha}_{x-t}P_1(x)=\frac{1}{\tau^{\alpha}}(\Delta\chi E-P_1(t)), \tag{3.10}$$

and rearranged as

$$[1+(\tau D_{x-t})^{\alpha}]P_1(t)=\Delta\chi E. \tag{3.11}$$

The latter Eq. (3.11) can be solved using the Laplace transform [27–30]

$$[1+(\tau p)^{\alpha}]\overline{P_1}(p)=\frac{\Delta\chi E}{p},\quad \overline{P_1}(p)=\int_0^{\infty}e^{-pt}P_1(t)dt, \tag{3.12}$$

which yields

$$\overline{P_1}(p)=\frac{\Delta\chi E}{p}\frac{1}{1+(\tau p)^{\alpha}}. \tag{3.13}$$

Insofar as

$$\frac{1}{1+(p\tau)^{\alpha}}=\frac{(p\tau)^{-\alpha}}{1+(p\tau)^{-\alpha}}=\sum_{n=0}^{\infty}(-1)^n(p\tau)^{-\alpha(n+1)}, \tag{3.14}$$

the solution of Eq. (3.13) in the domain of originals will have the following form:

$$P_1(t)=\Delta\chi E\sum_{n=0}^{\infty}\frac{(-1)^n\left(\frac{t}{\tau}\right)^{\alpha(n+1)}}{\Gamma[\alpha(n+1)+1]}, \tag{3.15}$$

where $\Gamma(x)$ is the gamma function. Therefore,

$$P(t)=P_0+P_1(t)=\left[\chi_0+\Delta\chi\sum_{n=0}^{\infty}\frac{(-1)^n\left(\frac{t}{\tau}\right)^{\alpha(n+1)}}{\Gamma[\alpha(n+1)+1]}\right]E. \tag{3.16}$$

After substitution of $\alpha=1$ into Eq. (3.16), Eq. (3.3) may be recovered; in fact,

$$P(t)=\left[\chi_0+\Delta\chi\sum_{n=0}^{\infty}\frac{(-1)^n\left(\frac{t}{\tau}\right)^{(n+1)}}{\Gamma(n+2)}\right]E$$
$$=\left[\chi_0+\Delta\chi\left(1-\exp\left(-\frac{t}{\tau}\right)\right)\right]E \tag{3.17}$$

[in the derivation, the standard Eq. (3.18) was used]

$$\sum_{n=0}^{\infty}\frac{(-1)^nz^n}{\Gamma[(n+1)]}=\exp(-z),\quad z=\frac{t}{\tau}. \tag{3.18}$$

Thus, the crossover from a strictly exponential to an anomalous relaxation pattern can be associated with the change of a continuous distribution of relaxation times ($\alpha=1$) into a fractal-like one ($0<\alpha=d_f<1$).

It follows from Eq. (3.16) that

$$P(t)\sim\left[\chi_0+\frac{\Delta\chi}{\Gamma(\alpha+1)}\left(\frac{t}{\tau}\right)^{-\alpha}\right]E, \tag{3.19}$$

which can be compared with Eqs. (1.1)–(1.3)

In the case of alternating field, the Fourier transform of Eq. (3.10) yields

$$P(\omega)=[\chi_0+\Delta\chi/(1+i\omega\tau)^{\alpha}]E, \tag{3.20}$$

and the dielectric permittivity will be

$$\varepsilon=\varepsilon_{\infty}+\frac{\varepsilon_0-\varepsilon_{\infty}}{1+(i\omega\tau)^{\alpha}}. \tag{3.21}$$

The latter Eq. (3.21) describes the frequency dependence of the Cole-Cole type. The real $\mathrm{Re}\,\varepsilon(\omega)$ and imaginary $\mathrm{Im}\,\varepsilon(\omega)$ parts of the total dielectric permittivity in Eq. (3.21) are, respectively,

$$\mathrm{Re}\,\varepsilon(\omega)=\varepsilon_0\left[\eta+\frac{(1-\eta)\left[1+(\omega\tau)^{\alpha}\cos\frac{\pi\alpha}{2}\right]}{1+2(\omega\tau)^{\alpha}\cos\frac{\pi\alpha}{2}+(\omega\tau)^{2\alpha}}\right], \tag{3.22}$$

$$\mathrm{Im}\,\varepsilon(\omega)=\varepsilon_0\left[\frac{(\eta-1)\left[1+(\omega\tau)^{\alpha}\sin\frac{\pi\alpha}{2}\right]}{1+2(\omega\tau)^{\alpha}\cos\frac{\pi\alpha}{2}+(\omega\tau)^{2\alpha}}\right]; \tag{3.23}$$

therefore, the dielectric loss tangent will be

$$\tan\delta=(\eta-1)\left[\frac{(\omega\tau)^{\alpha}}{1+2(\omega\tau)^{\alpha}\cos\frac{\pi\alpha}{2}+(\omega\tau)^{2\alpha}}\right], \tag{3.24}$$

where $\eta=\varepsilon_{\infty}/\varepsilon_0$.

Equations (3.22)–(3.24), respectively, were used to construct the plots of the real, $\mathrm{Re}\,\varepsilon(\omega)/\varepsilon_0$ (Fig. 2) and of the imaginary $\mathrm{Im}\,\varepsilon(\omega)/\varepsilon_0$ (Fig. 3), parts of complex dielectric permittivity, as well as of the dielectric loss tangent $\tan\delta$ (Fig. 4) in the function of $\log\omega\tau$ for a medium with $\eta=\varepsilon_{\infty}/\varepsilon_0=10$. As can be easily verified, the relaxation spectrum pattern strongly depends on the dimensionality of a temporal fractal ensemble $\alpha=d_f$.

It is pertinent to mention at this point that by using other operators of fractional differentiation, it is possible to derive

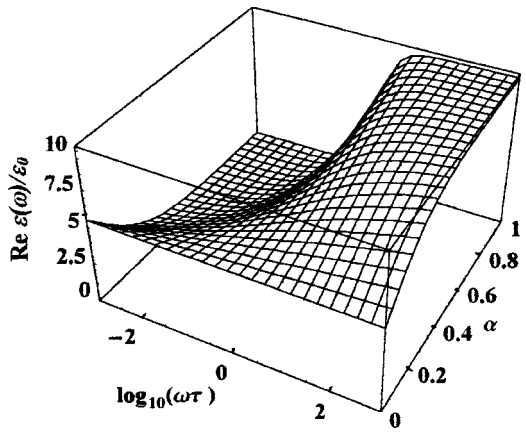

FIG. 2. Dispersion dependence of Re $\varepsilon/\varepsilon_0$ at different values of parameter α.

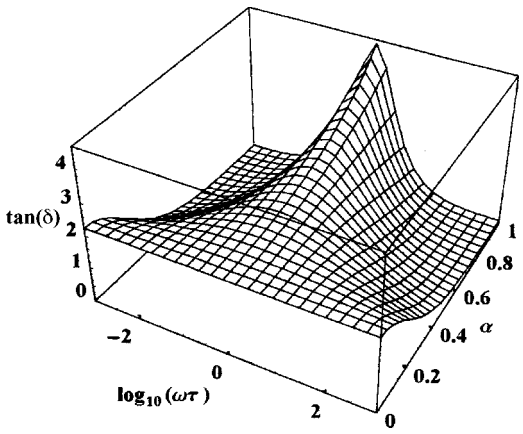

FIG. 4. Dispersion dependence of tan δ at different values of parameter α.

various laws of anomalous relaxation, such as the popular Havriliak-Negami law [25] (see the Appendix).

IV. FRACTAL MODEL

Let us consider in more detail the issue of relaxation times relevant to our fractal model of anomalous relaxation. As can be seen from the potential energy landscape for a system under the action of an external electrical field (Fig. 5), the energy differences between minima separated by energy maxima of different levels of self-similarity diminish, the larger the number of a self-similarity level. In view of the standard definition of a relaxation time, $\tau \approx e^{U/kT}$, one may write the following chain of inequalities:

$$\tau_0 > \tau_1 > \tau_2 \ldots \ldots \tau_i > \tau_{i+I}. \tag{4.1}$$

It is easy to see that these inequalities meet the model requirement on the difference between relaxation times insofar as both the width and the height of energy maxima are assumed to decrease, the larger the number of self-similarity level (Fig. 5). Note, however, the existence of the upper limit for an ensemble of relaxation times; that is, a self-similar process of a growing complexity of the potential energy landscape is arrested at a certain level $N<\infty$.

Thus, the proposed fractal relaxation model satisfies the criterium of self-similarity; moreover, its validity is restricted by asymptotic lower and upper limits. Let us analyze now the physical meaning of the self-similar potential energy landscape [44].

Assume that a system evolves by passing over a successive series of potential barriers, each next one of higher height. In this context, the initial relaxation processes with short relaxation times are assumed to be followed by those with ever increasing relaxation times.

Consider a relaxing ensemble of $N<\infty$ particles. Let this system consist of smaller subsystems (clusters), each of which, in its turn, consists of still smaller subsystems (subclusters), and so on. In principle, this kind of tesselation might have been repeated down to the infinitesimal scale; however, as mentioned above, the accepted model requires that such a self-similar increase of system complexity should be arrested at a certain level. Stated otherwise, the relaxation at the $n+1$ st level would not set on until a certain fraction of particles at the previous nth level would have relaxed (here it is implicitly assumed that the numeration of relaxation levels starts at the lower self-similarity limit for a subcluster comprising a minimum number of relaxing particles, $n=1$, and goes up to the upper limit of self-similarity for a cluster comprising all smaller subclusters, $n=N$, where N is the total number of hierarchical levels).

Let ω_q be the probablity of existence of each relaxation level corresponding to the kth statistical ensemble; then, the probability for a system to pass from qth to pth relaxation level during time t may be defined as

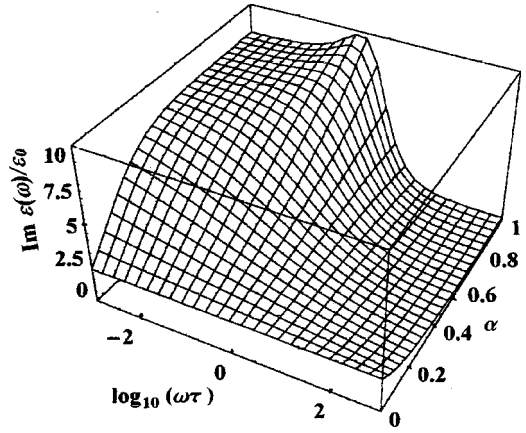

FIG. 3. Dispersion dependence of Im $\varepsilon/\varepsilon_0$ at different values of parameter α.

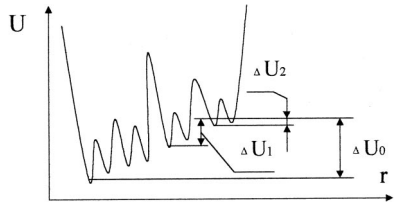

FIG. 5. Schematic of the potential barrier landscape under the action of an external electrical field.

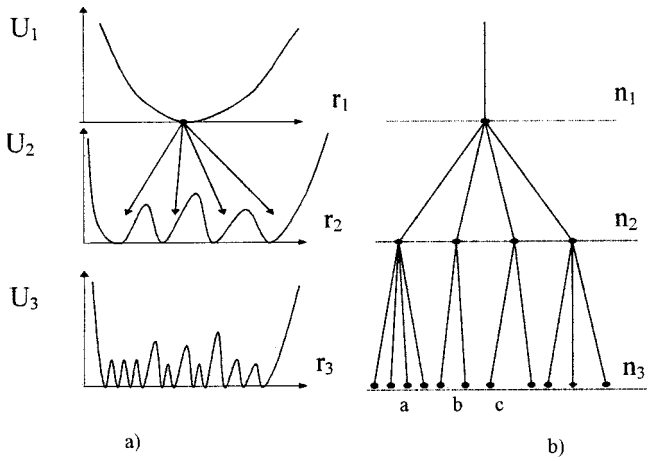

FIG. 6. Schematic of the self-similar potential energy landscape and of the Cayley Tree.

$$S_{qp}(t)=\exp(-t/\tau_{qp}), \tag{4.2}$$

where τ_{qp} is the relaxation time defined as

$$\tau_{qp}=\tau_0\exp(Q_{qp}/kT), \tag{4.3}$$

and Q_{qp} is the barrier height between levels q and p.

The probability for a system to pass to the level $n\leqslant N$ after time t will be

$$S(t)=\sum_{q=1,p=1}^{n}\omega_q\omega_p S_{qp}(t); \tag{4.4}$$

that is, the function $S(t)$ is assumed to account for contributions from all available relaxation channels. Therefore, the relaxation process may be specified, provided functions ω_q and Q_{qp} are known.

It can be noted that the self-similar potential energy landscape (Fig. 6) resembles the so called Cayley Tree [Fig. 6(b)], provided each minimum at a certain self-similarity level of the former is associated with a branch of the same number on the latter [45]. The coordinates of branches on the Cayley Tree form the ultrametric space; the metrics of this space is specified by interbranch distances which are defined as the numbers of steps between the branches and a common origin (for example, the distances between branches *a* and *b*, and between *a* and *c* in Fig. 6(b) are unity and two, respectively).

As can be inferred from Fig. 5, statistical ensembles $\{q, p\}$ may merge into clusters, each of which is characterized by the maximum barrier height Q_{qp} separating this particular cluster from its neighbor. In view of correspondence between the ensembles $\{q,p\}$ and the branches on Cayley Tree as referred to above, the former may be also characterized by points $\{q, p\}$ in the ultrametric space separated by distances l_{pq}. In this context, the barrier heights Q_{qp}, as well as corresponding relaxation times τ_{qp}, turn out to be the functions of distances l_{qp} in the ultrametric space. Insofar as elimination of clusters from this space may be achieved by the increase of corresponding barrier heights, one may conclude that the $Q(l)$ should be a smoothly increasing function.

It follows from the above analysis that parallel contributions of various relaxation channels may be possible only on the condition of hierarchical subordination of the corresponding series of statistical ensembles. In this case, the smallest statistical ensembles (subclusters) merge, and the system passes through to a higher hierarchical level of the Cayley Tree (Fig. 6). After passing over barriers of higher height, Q_{qp}, the newly born clusters merge again into larger entities (superclusters) corresponding to the next hierarchical level, and so on. It is this kind of a hierarchical subordination which is believed to be the main cause of critical slowing down of the relaxation process, which manifests itself as the transformation of exponential, Debye-like behavior into a slower, nonexponential decay.

Within the frame of the phenomenological approach, consider possible patterns of temporal dependence $S(t)$ at variable distribution majorants $\omega(l)$ and $Q(l)$. Assume that the descending tails of the probability distribution may be approximated as

$$\omega_w(l)\approx\exp(-l/l_0); \quad \omega_s(l)\approx l^{-D}, \tag{4.5}$$

where the first and the second functions apply to weakly hierarchical and to strongly hierarchical systems, respectively (here l_0 and D are positive parameters). The reason is that the former exponential function $\omega_w(l)$ decays at distances $l\approx l_0$ and, therefore, links only a limited number of hierarchical levels, while the latter, slowly changing power dependence $\omega_s(l)$ accounts, in effect, for the entire set of levels available.

The barrier heights $Q(l)$ will be approximated by three major types of ascending functions

$$Q_l(l)=Q\ln l; \quad Q_p(l)=Ql^a; \quad Qe(l)=Qe^l, \tag{4.6}$$

where Q is the characteristic barrier height, and $a=\text{const}>0$.

Asymptotics at $t\to\infty$ derived by the saddle-point method after substitution of Eq. (4.6) into Eqs. (4.3) and (4.2), and of the obtained result together with Eq. (4.5) into Eq. (4.4), are shown in Table I. It can be seen that all relaxation laws derived in this fashion are non-Debye-like, the weakest slowing down corresponding to a logarithmic growth of landscape heights in weakly hierarchical systems [i.e., the Kohlrausch-Williams-Watts (KWW) stretched exponential law [1,2]]. The descending function $S(t)$ transforms into a power law, should the hierarchical links become stronger and/or should the increase of peaks on the landscape follow a power law. The alternative cases of exponential and power-law increases of the landscape peaks in weakly and strongly hierarchical systems, respectively, would correspond to a logarithmic relaxation law, as described elsewhere [7]. Finally, a double-logarithmic slowing down (i.e., the virtual arrest) of the relaxation process is expected for strongly hierarchical systems characterized by exponential growth of barrier heights.

It would be instructive now to discuss the relevance of these model predictions to structural features of inhomogeneous media (IM). Let us define a statistical ensemble as a set of particles in the state of constant motion. The model of

TABLE I. Asymptotics of the correlator $S(t)$ at $t\to\infty$.

$S(t)$	$Q_l(l)$	$Q_P(l)$	$Qe(l)$
W_w	$\exp\left[-\left(\frac{t}{\tau_0}\right)^{\beta}\right]$ $\beta=\left[1+\left(\frac{Q}{kT}\right)\right]^{-1}$	$\exp\left[-\left(\frac{kT}{Q}\ln\frac{t}{\tau_0}\right)^{1/a}\right]$	$\left(\frac{kT}{Q}\ln\frac{t}{\tau_0}\right)^{-1/l_0}$
$W_s(l)$	$t^{-\gamma};\gamma=\frac{DT}{Q}$	$\left(\frac{Q}{kT}\ln\frac{t}{\tau_0}\right)^{-D/a}$	$\left[\ln\left(\frac{kT}{Q}\ln\frac{t}{\tau_0}\right)\right]^{-D}$

hierarchical subordination was constructed by singling out groups of ensembles from the entire statistical set available; hence, a similar tesselation procedure should be applied to the ensemble of particles making up the IM. For this purpose, a principle of singling out clusters and subclusters in the latter should be developed.

In simple terms, solid bodies may be characterized by two main features: by the pattern of mutual arrangement (packing) of particles and by the pattern of interparticle interactions. These features are complementary, rather than independent; nevertheless, it is the former one that will be used as a criterion to partition the entire IM system into smaller subsystems. According to current concepts, an IM may be considered as a structureless body at large length scales (i.e., above the characteristic correlation length ξ), whereas regions of a short-range order are assumed to exist at smaller scales (below ξ). In this context, it is the regions of a short-range order which will be identified as the primitive (first level) clusters; a set of primitive clusters will be defined as the second level clusters, and so on. Thus, the nth level cluster corresponding to the statistical ensemble of the nth hierarchical level may be built using such a process of a self-similar increase of complexity. The ''blob'' model based on these concepts was introduced elsewhere [46].

It is possible now to find a correspondence between functions $\omega(l), Q(l)$ and the accepted model of a short-range order hierarchy. As can be inferred from Fig. 7, the primitive cluster comprises seven particles, the next one at the second level $7^2=49$ particles, and the lth level cluster 7^l particles (here l is the level number). Obviously, for a level l comprising $N=7^l$ particles, the level number may be defined as

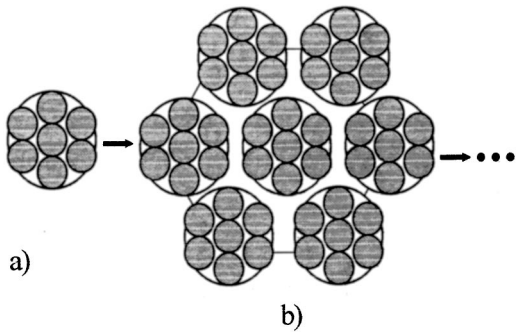

FIG. 7. Schematic of the self-similar structure of a dielectric substance.

$$l=a\ln(N),$$

$$a^{-1}=\ln 7. \tag{4.7}$$

In a general case, $l\sim\ln N$; therefore Eq. (4.5) may be rewritten as

$$\omega_w(n)\sim N^{-G};\quad \omega_S(N)\sim(\ln N)^{-D}. \tag{4.8}$$

The results obtained imply a rather low probability of large-size clusters comprising many particles for a weakly hierarchical system; the reverse is true for a strongly hierarchical system. Thus, structural implications of the concept of strongly and weakly hierarchical systems become more transparent.

The physical meaning of the function accounting for the increase of potential barrier height will be clarified by consideration of microscopic kinetics of dielectric relaxation of a hierarchical structure. Assume that the initial polarization is induced in the latter, and that a single particle and clusters of particles interact through dipole and multipole interactions, respectively. The relaxation processes start after the field is switched off at $t=0$. Initially, the relaxation sets on at the primitive, first level, insofar as the elementary dipoles can easily pass over a potential barrier created by their neighbors. In contrast, for the second level clusters the barrier heights created by neighboring clusters with preferential orientation of the major fraction of dipoles is so high that no relaxation can occur. Therefore, relaxation at the second level may set on after completion of relaxation in the major fraction of dipoles at the primitive level. Stated otherwise, it is only after sufficient weakening of multipole correlations of a given cluster with its neighbors that its transition into a depolarized state becomes possible. Such self-similar processes occur in succession at each next higher level, and so on. Thus, the form of function $Q(l)$ depends not only on the number of dipoles in a cluster but on the form of a multipole potential and temperature.

Finally, in view of Eq. (4.7), Eq. (4.6) can be rewritten as

$$Q_l(N)=Q\ln[\ln N];\quad Q_p(N)=Q(\ln N)^a;\quad Q_e(N)=QN. \tag{4.9}$$

The model considerations outlined above permit us to clarify the results presented in Table I. For example, from the explicit definition of the KWW stretched exponent

$$\beta=\left[1+\frac{Q}{kT}\right]^{-1}, \tag{4.10}$$

it can be inferred that $\beta\to 1$ at $T\gg 0$; that is, at sufficiently high temperatures the anomalous relaxation becomes Debye-like. Physically, this effect may be associated with the increase of fluctuation density of dipole reorientations; as a result, the relaxation of all available dipoles occurs already at the first level, so that the entire chain of remaining parallel relaxation channels becomes ineffective.

In a similar fashion, it becomes easy to predict the pattern of anomalous, nonexponential relaxation at times shorter than the relaxation time τ_1 at the lowest (i.e., first) self-similarity level. This level may be considered as primitive (in a sense that it cannot be tesselated further into subclusters); hence, the relaxation should be of a classical, Debye-like type,

$$P(t)\sim e^{-(t/\tau)}, \tag{4.11}$$

whatever the pattern of nonclassical relaxation at longer times.

V. CONCLUSIONS

The potential of fractional derivative technique is demonstrated on the example of derivation of three known patterns of anomalous, nonexponential dielectric relaxation of an inhomogeneous medium in the frequency domain. It is explicitly assumed that the fractional derivative is related to the dimensionality of a temporal fractal ensemble (in a sense that the relaxation times are distributed over a self-similar fractal system). The proposed fractal model of a microstructure of inhomogeneous media exhibiting nonexponential dielectric relaxation is built by singling out groups of hierarchically subordinated ensembles (subclusters, clusters, superclusters, etc.) from the entire statistical set available. In this context, it is the regions of a short-range order which are identified as the primitive (first level) clusters; a set of primitive clusters are defined as the second level clusters, and so on.

Different relaxation functions are derived assuming that the real (physical) ensemble of relaxation times is confined between the upper and lower limits of self-similarity. In this respect, the temporal fractal differs from the geometrical fractal (''Cantor dust'') for which only an upper limit (i.e., the initial segment before its subdivision) is assumed to exist. It is predicted that at times shorter than the relaxation time at the lowest (primitive) self-similarity level the relaxation should be of a classical, Debye-like type, whatever the pattern of nonclassical relaxation at longer times.

ACKNOWLEDGMENT

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APPENDIX A

Hereafter, substitution of $D^{\alpha}_{x-y}=D^{\alpha}$ will be implied. Consider the operator

$$(\tau^{-\alpha}+D^{\alpha})^{\nu}=\sum_{n=0}^{\infty}(\tau^{-\alpha})^{n}\binom{\nu}{n}D^{\alpha(\nu-n)}, \tag{A1}$$

where $\binom{\nu}{m}$ is the binomial coefficient.

Making use of Eq. (A1), the relaxation law for complex susceptibility may be written as

$$\tau^{-\alpha}+D^{\alpha\ \nu}[\chi\exp(i\omega t)]=\frac{\chi_0 E_0}{\tau^{\alpha\nu}}\exp(i\omega t). \tag{A2}$$

The solution of Eq. (A2) yields the standard definition of complex susceptibility

$$\chi(i\omega)=\chi_{\infty}+\frac{\chi_0-\chi_{\infty}}{(1+(i\omega\tau)^{\alpha})^{\nu}}, \tag{A3}$$

which is identical to the empirical Havriliak-Negami law [7].

In this case, the dielectric permittivity will be

$$\varepsilon\ \ i\omega=\varepsilon_{\infty}+\frac{\varepsilon_0-\varepsilon_{\infty}}{(1+(i\omega)^{\alpha})^{\nu}}, \tag{A4}$$

with the real and the imaginary parts, respectively,

$$\mathrm{Re}[\varepsilon(i\omega)]=\varepsilon_{\infty}+(\varepsilon_0-\varepsilon_{\infty})\times\frac{\cos\left[\nu\,\mathrm{arctg}\left(\dfrac{\sin\frac{\alpha\pi}{2}}{\cos\frac{\alpha\pi}{2}+(\omega\tau)^{-\alpha}}\right)\right]}{1+(\omega\tau)^{2\alpha}+2(\omega\tau)^{\alpha}\cos\frac{\alpha\pi}{2}} \tag{A5}$$

$$\mathrm{Im}[\varepsilon(i\omega)]=(\varepsilon_0-\varepsilon_{\infty})\times\frac{\sin\left[\nu\,\arctan\left(\dfrac{\sin\frac{\alpha\pi}{2}}{\cos\frac{\alpha\pi}{2}+(\omega\tau)^{-\alpha}}\right)\right]}{1+(\omega\tau)^{2\alpha}+2(\omega\tau)^{\alpha}\cos\frac{\alpha\pi}{2}} \tag{A6}$$

and the dielectric loss tan is

$$\tan\delta=\frac{\sin\left[\nu\arctan\left(\dfrac{\sin\dfrac{\alpha\pi}{2}}{\cos\dfrac{\alpha\pi}{2}+(\omega\tau)^{-\alpha}}\right)\right]}{\dfrac{\varepsilon_\infty}{\varepsilon_0-\varepsilon_\infty}\left[1+(\omega\tau)^{2\alpha}+2(\omega\tau)^{\alpha}\cos\dfrac{\alpha\pi}{2}\right]+\cos\left[\nu\arctan\left(\dfrac{\sin\dfrac{\alpha\pi}{2}}{\cos\dfrac{\alpha\pi}{2}+(\omega\tau)^{-\alpha}}\right)\right]}. \tag{A7}$$

The relaxation equation for an initially polarized dielectric is

$$(\tau^{-\alpha}+D^{\alpha})^{\nu}P(t)=0, \tag{A8}$$

and its solution is

$$P(t)=P_0 t^{\alpha\nu-1}\sum_{n=0}^{\infty}(-1)^n\frac{\nu(\nu+1)\ldots(\nu+n-1)}{n!}\times\frac{\left(\dfrac{t}{\tau}\right)^{\alpha n}}{\Gamma(\alpha(n+\nu)+1)}, \tag{A9}$$

where P_0 is the initial polarization (this solution also diverges at $t\to 0$).

The case of a dielectric without initial polarization (when the field is switched on at $t=0$), is described by Eq. (A10)

$$(\tau^{-\alpha}+D^{\alpha})^{\nu}P(t)=\frac{\chi_0 E}{\tau^{\alpha\nu}}, \tag{A10}$$

and its solution is the function

$$P(t)=\chi_0 E\sum_{n=0}^{\infty}(-1)^n\frac{\nu(\nu+1)\ldots(\nu+n-1)}{n!}\times\frac{\left(\dfrac{t}{\tau}\right)^{\alpha(n+\nu)}}{\Gamma(\alpha(n+\nu)+1)}. \tag{A11}$$

APPENDIX B

Consider functions $f(x)$, $g(x)$ with the Laplace transformation $L[f(x)]L[g(x)]$, and the convolution,

$$f(x)*g(x)=\int_0^x g(x-\tau)f(\tau)d\tau=\int_0^x g(\tau)f(x-\tau)d\tau. \tag{B1}$$

The Laplace transformation for the convolution is the product $L[f(x)]\cdot L[g(x)]$, i.e.,

$$L[f(x)*g(x)]=L[f(x)]L[g(x)] \tag{B2}$$

Thus, the function $\Phi(x)$ from Eq. (2.19) can be defined as

$$\Phi_y^{(\alpha)}(x)=\frac{1}{\Gamma(\alpha)}\int_y^x g(x-\tau)f^*(\tau)d\tau. \tag{B3}$$

where

$$g(x-\tau)=(x-\tau)^{\alpha-1},\quad f^*(\tau)=f(\tau)-f(y). \tag{B4}$$

It follows from Eq. (B2) that

$$L[\Phi_y^{(a)}(x)]=L[g(x)]L[f^*(x)]=p^{-\alpha}L[f^*(x)], \tag{B5}$$

where

$$L[x^{\alpha-1}]=p^{-\alpha}\Gamma(\alpha). \tag{B6}$$

In view of Eq. (2.20) and of Eq. (B7)

$$L\left[\frac{d\Phi(x)}{dx}\right]=pL[\Phi(x)], \tag{B7}$$

one obtains if $f(y)=0$

$$L[D_{x-y}^{1-\alpha}f(x)]=p^{1-\alpha}L[f(x)], \tag{B8}$$

or

$$L[D_{x-y}^{\beta}f(x)]=p^{\beta}L[f(x)],\quad \beta=1-\alpha.$$

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