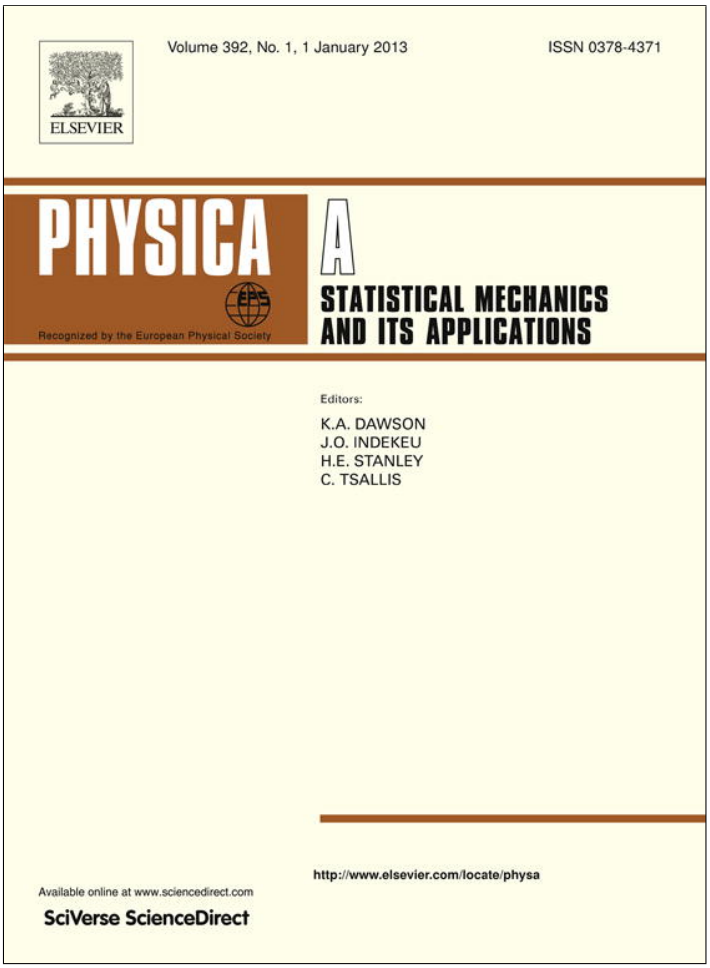

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Log-periodic corrections to the Cole–Cole expression in dielectric relaxation

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ABSTRACT

A model of the self-similar process of relaxation is given, and a method of derivation of the kinetic equations for the total polarization based on the ideas of fractional kinetics is suggested. The derived kinetic equations contain integro-differential operators having non-integer order. They lead to the Cole–Cole expression for the complex dielectric permittivity. It is shown rigorously that the power-law exponent α in the Cole–Cole expression coincides with the dimension of the mixed space-temporal fractal ensemble. If the discrete scale invariance for the temporal-space structure of the dielectric medium considered becomes important, then the expression for the complex dielectric permittivity contains log-periodic corrections (oscillations) and, hence, it generalizes the conventional Cole–Cole expression. The corrections obtained in this model suggest another way of interpretation and analysis of dielectric spectra for different complex materials.

1. Introduction

At the present time, detailed research in soft condensed physics matter [1] as a means of understanding the structure dynamics and relaxation phenomena in complex systems is observed. These complex systems represent a wide class of different materials with disordered structure. In particular, the investigation of (bio)polymers, colloids (emulsions and microemulsions), biologic cells, porous materials and liquid crystals can be also related to a complex system. In most methods used to investigate disordered materials, dielectric spectroscopy plays a dominant role on the mesoscopic scale [1]. However, the simple exponential relaxation law and classical model of Brownian diffusion cannot describe a wide class of relaxation phenomena and kinetics in these soft condensed materials. So, it is necessary to develop other approaches for the description of nonexponential relaxation behavior and anomalous diffusion processes (which nowadays are referred to as "strange kinetics" [2]).

Usually, for detailed descriptions of the kinetic processes taking place in different complex systems, it is necessary to use many different experimental methods to cover the corresponding frequency range. From this point of view, dielectric spectroscopy has an undoubted advantage over all other methods, because modern dielectric spectrometers allow one to cover a very wide frequency range (from 10^{-6} to 10^{11} Hz) [3–5].

The total polarization $P(t)$ of a dielectric placed in an external electric field $E(t)$ contains two parts [6]:

$$P(t) = P_0(t) + P_1(t), \tag{1}$$

where $P_0(t)$ defines the instantaneous component while $P_1(t)$ determines its retarded component. In the classic theory of relaxation [6], it is supposed that the rate of change of the retarded component is proportional to the difference between its

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limiting value ($P^*(t) = \chi_\infty E$) and the current value $P_1(t)$, i.e.

$$\frac{dP_1(t)}{dt} = \frac{1}{\tau}\left(\Delta\chi E - P_1(t)\right), \tag{2}$$

where $\Delta\chi = \chi_\infty - \chi_0$, and τ is the relaxation time. Integration of Eq. (2) leads (for constant E) to the following expression for the total polarization relaxation:

$$P(t) = P_0(t) + P_1(t) = \left[\chi_0 + \Delta\chi\left(1 - \exp(-t/\tau)\right)\right]E. \tag{3}$$

If the applied alternating field has the form $E = E_0 e^{i\omega t}$, then we obtain the following expression for the total polarization:

$$P(t) = P_0(t) + P_1(t) = \left[\chi_0 + \frac{\Delta\chi}{1 + i\omega\tau}\right]E(t). \tag{4}$$

Hence, the complex dielectric permittivity is determined by the expression [6]

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{1 + i\omega\tau}, \tag{5}$$

where $\varepsilon_\infty = \lim_{\omega\to\infty}\varepsilon$ and $\varepsilon_0 = \varepsilon|_{\omega=0}$. General expressions like (2) and (5) correspond to Debye relaxation.

However, for most complex systems, relationship (5) is not applicable, because the data associated with measurements of complex permittivity are not in general described by (5). This fact forced researchers to use different empirical expressions, which formally correspond to some distribution of relaxation times; see [7]. For a relatively wide class of complex systems, the dielectric spectra are described by the Cole–Cole expression [8],

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{1 + (i\omega\tau)^\alpha}, \tag{6}$$

where α is an empirical power-law exponent, which usually serves as a measure of the broadening loss peak.

For a physically meaningful description of nonexponential relaxation processes, one can use more sophisticated approaches. For example, one can generalize the kinetic equation, like in Refs. [9–12], or use hypotheses relating to the self-similar character of relaxation processes occurring in many complex systems [13–18]. In the context of the last approach, the kinetic equation for the retarded component of the total polarization (2) is rewritten in terms of a fractional derivative as

$$D_t^\alpha P_1(t) = \frac{1}{\tau^\alpha}\left(\Delta\chi E - P_1(t)\right), \tag{7}$$

where

$$D_t^\alpha P_1(t) = \frac{1}{\Gamma(1-\alpha)}\frac{d}{dt}\int_{t_0}^{t}\frac{P_1(t')}{(t-t')^\alpha}dt', \quad 0 < \alpha < 1, \tag{8}$$

determines the fractional derivative taken in the Riemann–Liouville form [19]. Steady-state solution of Eq. (7) for an alternating field leads to the following expression for the total polarization,

$$P(t) = P_0(t) + P_1(t) = \left[\chi_0 + \frac{\Delta\chi}{1 + (i\omega\tau)^\alpha}\right]E(t), \tag{9}$$

and, correspondingly, to an expression for the complex dielectric permittivity in the Cole–Cole form (6). Moreover, it is supposed that the power-law exponent α coincides with the dimension of a fractal set which determines the self-similar character of the relaxation process. In Ref. [18], it was associated with the set of relaxation times.

It is necessary to note that the presentation of the relaxation/kinetic equation in the form (7) and the relationship of the power-law exponent α to the fractal dimension is an intuitive supposition that requires rigorous (mathematical) justification. In Refs. [20,21], hypotheses connecting the fractal nature of the relaxation process and fractional kinetics for the total polarization have been suggested. The hypotheses allow one to derive closed Mori–Zwanzig kinetic equations and to obtain equations with non-integer operators for the total polarization. However, only initial steps have been made in this direction.

An inherent property of self-similar systems with discrete-scale invariance is the existence of log-periodic oscillations that accompany the power-law (fractal) behavior of these scale-invariant systems [22]. Moreover, if $\Phi(x)$ is a function that characterizes the behavior of a fractal system, then the scale invariance is determined by the functional equation

$$\Phi(\lambda x) = \gamma\Phi(x), \tag{10}$$

where the constant γ shows how the properties of a fractal system alter as a function of the scaling parameter λ. The general solution of the functional equation (10) has the form [23,24]

$$\Phi(x) = x^\nu W\left(\frac{\ln x}{\ln\lambda}\right), \tag{11}$$

where the first multiplier coincides with the usual power-law dependence characterizing the continuous scale invariance with the power-law exponent $\nu = \ln\gamma / \ln\lambda$, while $W(y)$ is a log-periodic function having unit period. The regular fractals exhibit discrete scale invariance, and so relationship (10) is not satisfied for an arbitrary scale transformation (expressed in terms of the scaling parameters λ, γ). As a result we have power-law behavior x^{ν} affected by a perturbation which is periodic in the log-scale representation. These log-periodic oscillations are evoked by coincidence of continuous scales with discrete ones, where the latter ones form a countable subset. The general solution (11) characterizes the discrete scale invariance as a whole.

The aim of this paper (in accordance with our supposition about the self-similar character of the relaxation process that takes place in a fractal system) is to attach a certain mathematical rigor to the correspondence of the power-law exponent α figuring in the Cole–Cole relationship (6) to a space-temporal self-similar structure having the property of discrete scale invariance. The log-periodic oscillations can modify the real power-law exponent in the Cole–Cole empirical relationship and serve as a definite indication of the presence of the discrete-scale invariance related to the space-temporal structure of the complex systems analyzed.

2. Memory function

The concept of the memory function was introduced by Zwanzig [25] and Mori [26]. They suggested an effective and very general theoretical formalism for the calculation of correlation functions based on projection operator techniques. The memory function is used to analyze correlation functions at short times. For example, the exponential decay of a correlation function is determined by a delta-like function, and this behavior is not correct at short times because the correlation function being a smooth function should have zero slope at $t = 0$. The replacement of the delta-function by a less singular function (e.g. a step function, the Gaussian function, etc.) adds more realistic features to its behavior at short times. The memory function $K(t)$ for the normalized correlation function $\phi(t)$ is determined by the equation [27]

$$\frac{d\phi(t)}{\mathrm{d}t} = -\int_{t_0}^{t} K(t-t')\phi(t')dt'. \tag{12}$$

For investigation of the influence of memory effects on dielectric relaxation, we put $\phi(t) = P_1(t)/P_1(t_0)$. Having in mind also the influence of the external electric field, one can rewrite Eq. (12) for the total polarization as

$$\frac{\mathrm{d}P_1(t)}{\mathrm{d}t} = -\int_{t_0}^{t} K(t-t')\left[P_1(t') - \Delta\chi E(t')\right]dt'. \tag{13}$$

If the memory function is absent, then $K(t) = \tau^{-1}\delta(t)$, and from (13) we retrieve the classical relaxation laws (3) and (4). In real situations we, as a rule, are dealing with processes where memory effects play an essential role.

The determination of the analytical form of the memory function is a nontrivial problem [28]. Many researches restore the behavior of the desired memory function by means of an experimentally known correlation function, using for this purpose Eq. (12). Besides this option, the memory function can also be restored with via computer simulations [29].

The Mori–Zwanzig formalism suggests the calculation of the desired memory function from the solution of the infinite chain of integro-differential equations of the type [25,26]

$$\frac{\mathrm{d}K_n(t)}{\mathrm{d}t} = -\int_{t_0}^{t} K_{n+1}(t-t')K_n(t')dt', \quad n = 0, 1, 2, \ldots, \tag{14}$$

where $K_n(t)$ determines the memory function of the nth order and $K_0(t) = K(t)$. This approach to the memory function is the most successful and rigorous; however, it is accompanied by serious computational difficulties. For solution of the infinite chain of equations (14), it is necessary to use some decoupling procedure in order to close the infinite system of integro-differential equations. The decoupling procedure is based on the reduction of a memory function of higher order to a combination of the memory functions of the lower order, and in general it does not have a clear physical meaning. As an example, one can demonstrate the decoupling procedure in system (14) by choosing the correlation function $K_1(t) = \kappa^2 K_0(t)$, where κ is some dimensionless correlation parameter. Finally, we have the closed equation for the function $K_0(t) = K(t)$,

$$\frac{\mathrm{d}K(t)}{\mathrm{d}t} = -\kappa^2\int_{0}^{t} K(t-t')K(t')dt', \tag{15}$$

where, for convenience, we put $t_0 = 0$. Then the integro-differential equation (15) is solved easily using the Laplace transform:

$$p\aleph(p) - K(0) = -\kappa^2\left(\aleph(p)\right)^2, \qquad \aleph(p) = \int_{0}^{\infty} K(t)e^{-pt}\mathrm{d}t. \tag{16}$$

The solution of the operator equation (16) has the form

$$\Re(p) = \frac{2K(0)}{\sqrt{p^2 + 4K(0)\kappa^2} + p}. \tag{17}$$

Returning to the time domain, we obtain

$$K(t) = \frac{\sqrt{K(0)}}{\kappa t} J_1\left(2\kappa\sqrt{K(0)}t\right), \tag{18}$$

where $J_1(x)$ is the Bessel function of the first kind of order 1.

Another way of selection of the memory function is based on replacement by some model function. For example, taking into account the basic approximations of the delta-function [30]

$$\delta(t) = \begin{cases} \dfrac{1}{\sqrt{\pi}} \lim\limits_{\tau\to 0} \dfrac{1}{\tau} e^{-t^2/\tau^2}, \\ \dfrac{1}{\pi} \lim\limits_{\tau\to 0} \dfrac{\tau^{-1}}{t^2/\tau^2 + 1}, \\ \dfrac{1}{\pi} \lim\limits_{\tau\to 0} \dfrac{\sin(t/\tau)}{t}, \end{cases} \tag{19}$$

and its normalization

$$\tau \int_0^\infty K(t, \tau)\mathrm{d}t = 1, \tag{20}$$

one can define the memory function via the following expressions:

$$K(t, \tau) = \begin{cases} \dfrac{2}{\sqrt{\pi}\tau^2} e^{-t^2/\tau^2}, & \text{(a)} \\ \dfrac{2}{\pi\tau^2} \dfrac{1}{t^2/\tau^2 + 1}, & \text{(b)} \\ \dfrac{2}{\pi\tau^2} \dfrac{\sin(t/\tau)}{(t/\tau)}. & \text{(c)} \end{cases} \tag{21}$$

From Eq. (21), it is clear that this set of memory functions has the following structure:

$$K(t, \tau) = \frac{1}{\tau^2} F\left(\frac{t}{\tau}\right), \tag{22}$$

where τ is a characteristic relaxation time and $F(x)$ is a dimensionless function having the following properties:

$$\begin{aligned} &\lim_{x\to\infty} F(x) = 0, \\ &F(x) \le \lim_{x\to 0} F(x). \end{aligned} \tag{23}$$

One can also suggest the following model representation for the desired model function. Let us suppose that memory exists during some period T (T is a dimensionless parameter) and after this period is absent. In this case, $F(x)$ can be approximate by a step function,

$$F(x) = \begin{cases} 1/T, & 0 \le x \le T, \\ 0, & x > T, \end{cases} \tag{24}$$

and by varying the parameter T in wide limits one can change the temporal region width affected by the memory effect.

At present, a universal criterion and the corresponding approach for selection of the proper memory function are absent. The detailed investigation of probable functional dependences for the memory function is a self-contained problem that is outside the scope of the present work. In the further consideration, mainly for demonstration purposes, we consider only the model forms of the memory function.

Based on modern theoretical conceptions [31,29] and experimental observations [6], one can put forward a reasonable supposition that in many cases the memory function is expressed by a power-law function depending on time. This fact is confirmed also by agreement with a large quantity of actual data obtained for different dielectric spectra. Usually this dependence is introduced empirically without any serious arguments justifying this manipulation. Below, we want to show that the power-law dependence has a clear physical interpretation and is a consequence of the self-similar structure of the medium considered and the relaxation processes that take place in it.

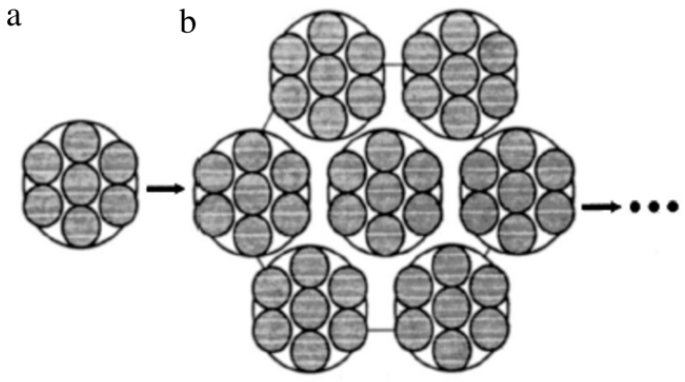

Fig. 1. Schematic image illustrating the cluster generation of the self-similar structure in a dielectric medium: (a) a cluster of the nth level, (b) a cluster of the $(n+1)$th level.

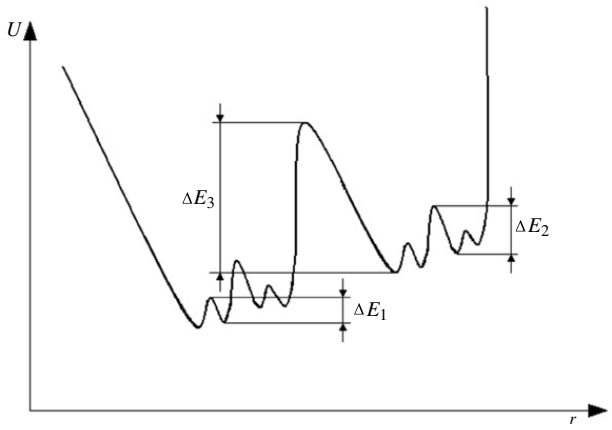

Fig. 2. The schematic cross-section of the potential profile in the applied electric field. Here, r is a configuration coordinate.

3. The model of self-similar relaxation process

Following Ref. [18], we represent the dielectric medium as a hierarchically organized set of dipole clusters. In many disordered media we have a local order in the location of particles (elementary dipoles), and the nearest-neighbor dipoles form clusters of the first order. It is natural to suppose that between clusters of the first level spatial correlation exists. This correlation leads to generation of the second-level clusters from the clusters belonging to the first level. Using a self-similar construction of such type, one can construct the desired nth-level cluster (see Fig. 1).

For further analysis of the microscopic kinetics of dielectric relaxation for such a hierarchical structure, we consider Fröhlich's relaxator model [32]. Here, the fixed relaxator is in two stable equilibrium points separated by a potential barrier. When the external electric field is applied, the energy difference ΔE arises between the minima of the potential landscape, and the system starts the relaxation process with the relaxation time

$$\tau = \tau_0 \exp\left(\frac{\Delta E}{T}\right). \tag{25}$$

Now we *suppose* that the minima of the potential landscape are also organized according to a self-similar hierarchy, as shown in Fig. 2. This self-similar organization of the potential landscape corresponds to increasing the scales of the geometrical structure considered above. Hence we have the hierarchy of relaxation times:

$$\tau_1 < \tau_2 < \tau_3 < \cdots < \tau_n < \tau_{n+1} < \cdots. \tag{26}$$

This sequence of relaxation times is monotonically increasing because the values of the potential profiles and energy difference are also increasing. Hence, in the self-similar system considered many interacting relaxation processes exist.

Accepting this physically reasonable model, let us consider the relaxation kinetics of this hierarchically organized structure. We suppose that the initial polarization was created in the recent past and that separate particles and their clusters interact with each other via both dipole and multi-pole interactions, correspondingly. The relaxation processes starting at the time t_0 correspond to the state of absence (or "switching off") of the applied external field. Initially, we have the process of relaxation of the first level (defined by the minimum value of relaxation time τ_1 in (26)). On the first level, elementary dipoles can easy overcome the potential barriers formed by neighboring dipoles.

However, the relaxation process on the second level does not take place, because for the cluster belonging to this level the probability to overcome the corresponding potential barrier formed by a neighboring cluster is low (because the vast majority of dipoles are oriented in one direction). The second level of the hierarchically organized dipoles starts to relax (with characteristic relaxation time τ_2) only when a relatively large number of dipoles belonging to the first level have relaxed. So, if the multi-pole correlations between clusters have decayed then transition of the whole almost isolated cluster in the depolarized state is highly probable. Then the self-similar process of relaxation is repeated. Thus, the set of relaxation times can form a hierarchically organized fractal set distributed in time in accordance with Eq. (26) above.

Now we want to formulate the physical model outlined above. We suppose that the mean number of dipoles in a cluster of the lth level is N_l and that its characteristic relaxation time is defined as $\tau = \tau_l$. In accordance with the hierarchically organized structure of clusters, we require that the values N_l and τ_l satisfy the following self-similarity conditions,

$$N_l = N_0 b^l, \quad \tau_l = \tau_0 \xi^l, \quad -M \le l < L, \quad L, M \gg 1, \tag{27}$$

where the parameters $b, \xi > 1$, and b is an integer. The conditions (27) imply that the system is considered on the mesoscopic scale with characteristic parameters $\tau_0, N_0 (N_0 \ge b^M)$ and the low and that upper limits of the space-temporal structure considered are *infinite*. We want to note here that the hierarchy structure of dynamic degrees of freedom expressed by relationships (27) leads to log-periodic oscillations in the relaxation laws [33].

Hence, as a result of this supposition we obtain a set of relaxation processes, and each process has its own memory function. The effective relaxation process is determined as the averaged process with a memory function having the following form:

$$\overline{K}(t) = \sum_l \frac{N_l}{N} K(t, \tau_l) = c \sum_{l=-M}^{L} b^l K(t, \tau_0 \xi^l) = \frac{c}{\tau_0^2} \sum_{l=-M}^{L} \left(\frac{b}{\xi^2}\right)^l F(\theta \xi^{-l}), \tag{28}$$

where N is the total number of dipoles on all levels, $c = N_0/N$ is the averaged concentration of dipoles on the mesoscopic scale ($l = 0$), and $\theta = t/\tau_0$ is the dimensionless time. The effective (averaged) expression for the memory function (28) satisfies (in the limit $L, M \to \infty$) the functional equation (10):

$$\overline{K}(\theta\xi) = b\xi^{-2}\overline{K}(\theta). \tag{29}$$

We determine the "space-temporal" fractal dimension of the self-similar relaxation process by the following relationship:

$$d_f = \frac{\ln b}{\ln \xi} > 0. \tag{30}$$

Here, the parameter b can be associated with the geometrical location of the relaxing clusters, while the parameter ξ is associated with the self-similar dynamical process taking place inside them. This mixed dimension, introduced for the first time in Refs. [20,21], can be used for characterization of the relaxation process in self-similar structures. Further, it is convenient to regard the parameter d_f as the independent variable, and so the geometrical parameter b is expressed as $b = \xi^{d_f}$. In accordance with the requirement (23), the series (28) must converge at $0 < d_f < 2$.

The detailed calculations of the averaged memory function (28) are given in the Appendix. Using Mellinn's transform, the series (28) can be calculated analytically without specifying the concrete form of the function $F(x)$. Finally, the expression for $\overline{K}(\theta)$ takes the form

$$\overline{K}(\theta) = \frac{c\Omega\tau_0^{-2}}{2\pi} \sum_{k=-\infty}^{\infty} \mathfrak{F}\left(2 - d_f + i\Omega k\right) \theta^{-(2-d_f+i\Omega k)}, \quad \Omega = \frac{2\pi}{\ln \xi}, \tag{31}$$

where $\mathfrak{F}(s) = \int_0^\infty F(\theta)\theta^{s-1}d\theta$ is the Mellin transform of function $F(x)$.

4. Kinetic equation

Eq. (31) for the memory function allows one to rewrite the kinetic equation (13) in the more compact form

$$\frac{dP_1(t)}{dt} + \frac{c\Omega}{2\pi} \sum_{k=-\infty}^{\infty} \mathfrak{F}\left(2 - d_f + i\Omega k\right) \tau_0^{-d_f+i\Omega k} \int_{-\infty}^{t} \left(t - t'\right)^{-(2-d_f+i\Omega k)} \left(P_1(t') - \Delta\chi E(t')\right) dt' = 0, \tag{32}$$

where we put the lower time limit $t_0 \to -\infty$. Using the definition of the fractional integral in the Riemann–Liouville form [19],

$${}_{t_0}I_t^{\nu} f(t) = \frac{1}{\Gamma(\nu)} \int_{t_0}^{t} \frac{f(t')}{(t - t')^{1-\nu}} dt', \quad \nu > 0, \tag{33}$$

one can rewrite this equation as

$$\frac{dP_1(t)}{dt} + \sum_{k=-\infty}^{\infty} C_k(d_f)\tau_0^{-d_f+i\Omega k}\,{}_{-\infty}I_t^{d_f-1-i\Omega k}\left(P_1(t) - \Delta\chi E(t)\right) = 0. \tag{34}$$

Because of the definition of the fractional integral as (33), the power-law exponent $\nu > 0$, and Eq. (34) is valid when $1 < d_f < 2$. For generalization of the kinetic equation (34) for the case $0 < d_f < 1$ (when the Riemann–Liouville integral diverges at the upper limit), it is necessary to introduce a regularization procedure. In accordance with the recommendations considered in Ref. [19], the regularization procedure reduces to replacement of the divergent integral by its finite part in accordance with Hadamard's recommendations [19] (accurate within a sign).

$$p.f.\int_{-\infty}^{t} \frac{f(t')}{(t-t')^{\nu+1}}dt' = \int_{-\infty}^{t} \frac{f(t)-f(t')}{(t-t')^{\nu+1}}dt', \quad \nu > 0. \tag{35}$$

We remark that in the definition (35) alternation of the integral sign is taken into account. Using the regularization procedure in the Riemann–Liouville fractional integral for the case $0 < d_f < 1$, we obtain the fractional derivative in the form suggested by Marchaud [19]:

$${}_{-\infty}\mathfrak{D}_t^{\nu}f(t) = p.f.\,{}_{-\infty}I_t^{-\nu}f(t) = \frac{1}{\Gamma(-\nu)}\int_{-\infty}^{t}\frac{f(t')-f(t)}{(t-t')^{\nu+1}}dt', \quad 0 < \nu < 1. \tag{36}$$

Hence, one can rewrite the kinetic equation for the case $0 < d_f < 1$ as

$$\frac{dP_1(t)}{dt} + \sum_{k=-\infty}^{\infty} C_k(d_f)\tau_0^{-d_f+i\Omega k}\,{}_{-\infty}\mathfrak{D}_t^{1-d_f+i\Omega k}\left(P_1(t) - \Delta\chi E(t)\right) = 0. \tag{37}$$

The coefficients $C_k(d_f)$ in Eqs. (34) and (37) are determined by the single expression

$$C_k(d_f) = \mathrm{sgn}(d_f - 1)\frac{c\Omega}{2\pi}\mathfrak{F}\left(2 - d_f + i\Omega k\right)\Gamma(d_f - 1 - i\Omega k). \tag{38}$$

Let us now suppose that the external electric field is switched on at $t_0 = -\infty$ and varies harmonically, i.e. $E(t) = E_0\exp(i(\omega - i\delta)t)$, $(\delta \to +0)$. Using the simple relationships

$${}_{-\infty}I_t^{\nu}e^{i(\omega-i\delta)t} = (i\omega)^{-\nu}e^{i(\omega-i\delta)t}, \qquad {}_{-\infty}\mathfrak{D}_t^{\nu}e^{i(\omega-i\delta)t} = (i\omega)^{\nu}e^{i(\omega-i\delta)t}, \tag{39}$$

the kinetic equations (34) and (37) describing the evolution of the total polarization become

$$P_1(t) + \sum_{k=-\infty}^{\infty} C_k(d_f)(i\omega\tau_0)^{-d_f+i\Omega k}\left(P_1(t) - \Delta\chi E(t)\right) = 0. \tag{40}$$

From kinetic equation (40), one can now obtain the compact expression for the complex dielectric permittivity,

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{1 + R(i\omega)},$$

with

$$R(i\omega) = \left[\sum_{k=-\infty}^{\infty} C_k(d_f)(i\omega\tau_0)^{-d_f+i\Omega k}\right]^{-1} = \frac{C_0^{-1}(d_f)(i\omega\tau_0)^{d_f}}{1 + 2\sum\limits_{k=1}^{\infty}\left|\frac{C_k(d_f)}{C_0(d_f)}\right|\cos\left(\phi_k + k\Omega\ln(i\omega\tau_0)\right)}. \tag{41}$$

Here, $\varphi_k = \arg\left[C_k(d_f)\right]$. As stated above, the Mellin image defining the behavior of the coefficients $C_k(d_f)$ (38) is expressed in the general case in terms of a product of combinations of hypergeometric functions of the type (A.7). This combination decreases rapidly (with the growth of k) in the complex plane. So, in expression (41), it is sufficient to keep only the first few terms. We want to justify this statement, taking as an example the memory function expressed in the form of the step function (24). The Mellin image of this function has the following form:

$$\mathfrak{F}(s) = \frac{T^{s-1}}{s}, \quad \mathrm{Re}\ s > 1. \tag{42}$$

Then the ratio $\left|C_k(d_f)/C_0(d_f)\right|$ for the chosen memory function in accordance with expressions (38) and (42) takes the form

$$\left|\frac{C_k(d_f)}{C_0(d_f)}\right| = \left|\frac{\mathfrak{F}\left(2 - d_f + i\Omega k\right)\Gamma(d_f - 1 - i\Omega k)}{\mathfrak{F}\left(2 - d_f\right)\Gamma(d_f - 1)}\right| = \left|\frac{\Gamma(d_f - 2 - i\Omega k)}{\Gamma(d_f - 2)}\right|. \tag{43}$$

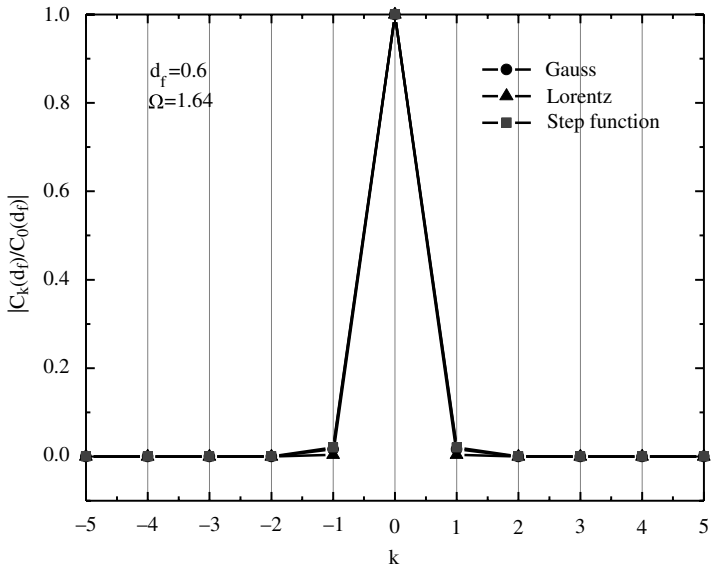

Fig. 3. The modulus dependence of the ratio $\left|C_k(d_f)/C_0(d_f)\right|$ containing the log-periodic corrections in the Cole–Cole expression against the number of modes k for the various model memory functions. Clearly, the specific form of the memory function chosen has no practical effect on the value of this ratio.

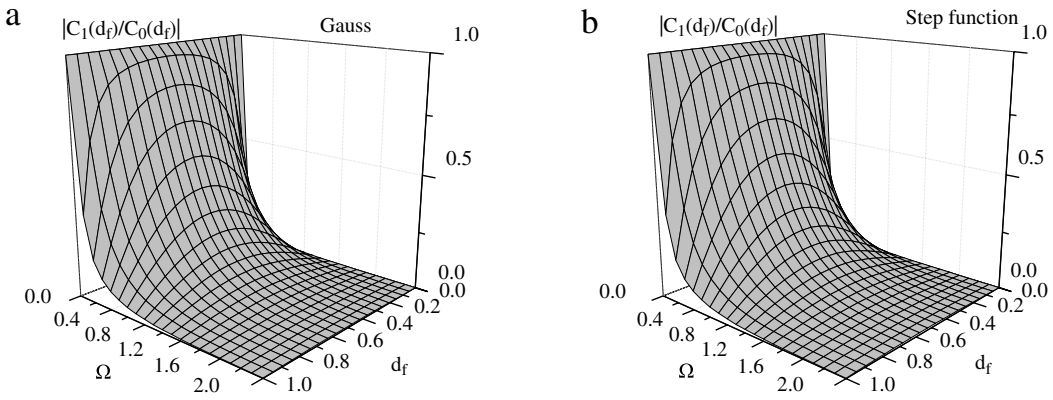

Fig. 4. The dependence of the ratio $\left|C_1(d_f)/C_0(d_f)\right|$ for the first mode containing a log-periodic term with respect to variations of the values d_f and Ω: (a) for the Gaussian memory function and (b) for the step function. Again we notice how the behaviors resemble each other.

From this expression, we obtain the following estimation:

$$\left|\frac{C_k(d_f)}{C_0(d_f)}\right| \sim \exp\left(-A(\Omega, d_f)k\right), \tag{44}$$

where $A(\Omega, d_f)$ is some constant close to unity. From (44), it follows that the contribution of the higher modes becomes negligible as k increases. So, one can approximate in the sum, in view of Eq. (41), to take into account only zero and the first modes, correspondingly:

$$R(i\omega) \approx C_0^{-1}(d_f)(i\omega\tau_0)^{d_f}\left[1 + 2\left|\frac{C_1(d_f)}{C_0(d_f)}\right|\cos\left(\varphi_1 + \Omega\ln(i\omega\tau_0)\right)\right]^{-1}. \tag{45}$$

Fig. 3 demonstrates the sharp slope of the amplitudes of the higher modes containing the log-periodic terms with respect to the number of modes k. This normalized ratio (see (44)) is calculated for the different memory functions for realistic values of parameters d_f and Ω ($d_f = 0, 6$ and $b = 10$, $\Omega \approx 1, 64$). From the figure it is apparent that different memory functions do not essentially change this ratio. In Fig. 4, we show the behavior of ratio (44) at $k = 1$ for different values of parameters d_f and Ω for two model memory functions: the Gaussian function (21)(a) and the step function (24)(b). Clearly, the form of the memory function chosen does not in practice alter the behavior of the ratio $\left|C_1(d_f, \Omega)/C_0(d_f, \Omega)\right|$.

If one keeps only the zero term in (41), then we have usual the Cole–Cole function:

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{1 + C_0^{-1}(d_f)(i\omega\tau_0)^{d_f}}. \tag{46}$$

Now consider expression (41) in the classical case corresponding to Debye relaxation. As remarked earlier, in Ref. [18], the Debye relaxation time can represent the mean relaxation time averaged over all the ensemble of relaxation times defined by Eq. (26). Taking into account the spatial self-similarity, it is necessary to average the ensemble of relaxation times with the factor N_l/N. This multiplier takes into account the number of dipoles forming a cluster. Finally, for the averaged time we have the following expression:

$$\frac{1}{\langle\tau\rangle} = \sum_{l=-M}^{L} \frac{N_l}{N}\frac{1}{\tau_l} = \frac{c}{\tau_0}\sum_{l=-M}^{L} \xi^{(d_f-1)l} = \frac{c}{\tau_0}\frac{\left(\xi^{-(d_f-1)M} - \xi^{(d_f-1)(1+L)}\right)}{1-\xi^{(d_f-1)}}. \tag{47}$$

Taking into account the normalization factor, we have

$$1 = \sum_{l=-M}^{L} \frac{N_l}{N} = c\sum_{l=-M}^{L} \xi^{d_f l} = c\frac{\left(\xi^{-d_f M} - \xi^{d_f(1+L)}\right)}{1-\xi^{d_f}}. \tag{48}$$

Excluding from these expressions the value L, we find

$$\tau_0 = \frac{c\,\langle\tau\rangle}{1-\xi^{(d_f-1)}}G(d_f, M), \quad G(d_f, M) = \xi^{-(d_f-1)(M+1)} - \left(\xi^{-d_f M} - \frac{1-\xi^{d_f}}{c}\right)^{\frac{d_f-1}{d_f}}. \tag{49}$$

Putting the last expression into Eq. (41), we obtain

$$R(i\omega) = \left[\sum_{k=-\infty}^{\infty} \frac{c\Omega}{2\pi}\mathfrak{F}\left(2-d_f+i\Omega k\right)\Gamma(d_f-1-i\Omega k)\left(\frac{i\omega\,\langle\tau\rangle\, cG(d_f, M)}{1-\xi^{(d_f-1)}}\right)^{-d_f+i\Omega k}\right]^{-1}. \tag{50}$$

For consideration of the Debye case in accordance with the classic kinetic equation (2), it is necessary to have $d_f \to 1$, and in accordance with the definition of the fractional derivatives [19] we should consider the right-hand limit ($d_f \to 1+0$). Besides, in accordance with the chosen model (27), it is necessary that $M \to \infty$ (see the definition of the parameter M in expressions (47) and (48)). Taking into account the value of the limit

$$\lim_{\substack{d_f\to 1+0\\ M\to\infty}} G(d_f, M) = -1, \tag{51}$$

we obtain

$$\lim_{\substack{d_f\to 1+0\\ M\to\infty}} R(i\omega) = \left[\sum_{k=-\infty}^{\infty} \lim_{d_f\to 1+0}\frac{c\Omega}{2\pi}\mathfrak{F}\left(2-d_f+i\Omega k\right)\Gamma(d_f-1-i\Omega k)\left(\frac{i\omega\,\langle\tau\rangle\, c\Omega}{2\pi\,(d_f-1)}\right)^{-d_f+i\Omega k}\right]^{-1}. \tag{52}$$

In this sum, all terms except the zero term ($k=0$) vanish, and so we have, finally,

$$\lim_{\substack{d_f\to 1+0\\ M\to\infty}} R(i\omega) = \left[\frac{1}{i\omega\,\langle\tau\rangle}\mathfrak{F}(1)\lim_{d_f\to 1+0}\left(\frac{\Gamma(d_f-1)}{(d_f-1)}\right)^{-1}\right]^{-1} = i\omega\,\langle\tau\rangle, \tag{53}$$

where the normalization of the memory function (20), i.e. $\mathfrak{F}(1)=1$, is taken into account. Clearly, $d_f = 1$ corresponds to the situation when the number of dipoles in the cluster increases uniformly with the same velocity as their relaxation time rate ($b=\xi$). In other words, this rather general physical requirement corresponds to the Debye law for dielectric permittivity. The same law is observed when the memory effect is absent, and from a mathematical point of view, in accordance with Eq. (19), it corresponds to vanishing $\tau_0 \to 0$ in Eq. (41).

The calculated function (41) for the dielectric permittivity contains only six fitting parameters, namely ε_0, ε_∞, τ_0, c, d_f, Ω. All these parameters have a clear physical meaning, and they characterize the collective relaxation process occurring in the mesoscale region. For the complete evaluation of expression (41) it is necessary to know the Mellin image $\mathfrak{F}(s)$ of the memory function, which in most cases presents itself as the ratio of different gamma-functions (A.7). As follows from Eqs. (38) and (44), the log-periodic oscillations are effective when $\Omega \ll 1$. In this case the corresponding coefficients $C_k(d_f)$ decay very slowly as k increases. However, in real situations, from our point of view, the case $\Omega \geq 1$ is more probable. Really, it is logical to suppose that the values of relaxation times (26) of two neighboring levels are close to each other. This supposition, in turn, implies that the scaling parameter ξ from (27) is close to unity, and this fact leads to large values of Ω (as follows from definition (31)). The large values of Ω lead to the fast oscillations and, consequently, to the fast decay of the coefficients $C_k(d_f)$ with increasing k (as shown in Fig. 4). In this case, the log-periodic corrections in the Cole–Cole expressions appear in the form of specific small oscillations which accompany the basic peak. As a demonstration of Fig. 5, we show the dielectric permittivity function (41) for two types of memory function (21)(a) and (24) having different values of parameter Ω. Naturally, the character of these oscillations depends on the type of memory function chosen. The step-type memory function leads to more pronounced log-periodic oscillations in comparison with the Gaussian memory function.

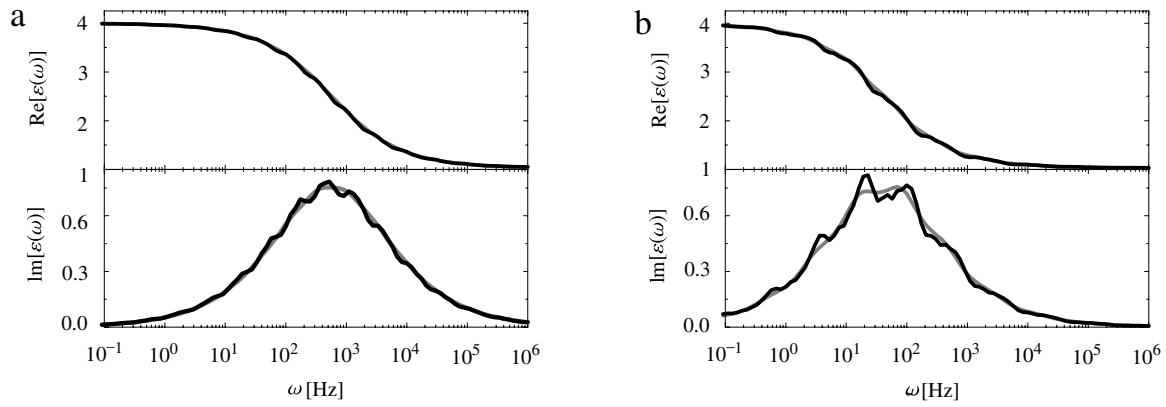

Fig. 5. The real and imaginary parts of the complex dielectric permittivity function defined by expression (41). For both figures two memory functions are chosen: the Gaussian in the form of (21) (a) (bold gray line) and the step function in the form of (24) for $T = 1$ (solid black line). For direct calculation of these expressions the following values of the fitting parameters were used: $\varepsilon_0 = 4$, $\varepsilon_\infty = 1.03$, $\tau_0 = 10^{-3}$, $c = 0.3$, $d_f = 0.6$; for (a), $\Omega = 5.4$, and for (b), $\Omega = 3.4$.

The analysis of the experimentally measured different dielectric spectra does not reveal the specific behavior of $\varepsilon\ (i\omega)$ shown, for example, in Fig. 5(a) by the bold black line. So, at the present stage, one can conclude that log-periodic oscillations are manifested as distortions that accompany the basic contour line. This specific behavior is illustrated schematically in Fig. 5(a) and (b). We think that the amplitudes of these oscillations are comparable with the noise level of the measurement equipment used, and in most cases these oscillations are smoothed or ignored as an understandable (artifact) phenomenon.

5. Basic remarks and conclusions

One of the basic results obtained in this paper is associated with the general structure of expression (41), which is practically independent of the memory function selected. This general result is directly associated with the "universal response" phenomenon discovered and properly investigated by Jonscher [6,34–37]. Another important result is related to log-periodic oscillations in the Cole–Cole expression (41), which were confirmed experimentally in Ref. [38]. If the log-periodic oscillations decay very rapidly and the complex permittivity function is described by Cole–Cole expression (46), then the memory function affects only the value of the relaxation time and can lead to asymmetry of a loss peak. This asymmetry is not contained in the conventional Cole–Cole expression. In the opposite case, when the log-periodic oscillations are noticeable, then the influence of the memory function becomes important, changing the form of these oscillations. The last conclusions allow one to extract the desired memory function from basic expression (41) and hence to obtain information about the microscopic processes that take place in the substance studied. We want to note also that the parameters determining a specific scale of these oscillations depend on the temperature. Based on this fact, we suppose that one can expect a strongly pronounced effect of log-periodic oscillations in the frequency dependence of dielectric response in some temperature interval in the vicinity of the phase temperature region. This supposition is based, in turn, on the experimentally confirmed observation that log-periodic oscillations are becoming noticeable near the critical temperature [22].

Acknowledgments

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Appendix. Calculation of the averaged memory function

For evaluation of the sum in (28), it is convenient to use the Mellin transform:

$$F(\theta) \overset{MT}{=} \mathfrak{F}(s) = \int_0^\infty F(\theta)\theta^{s-1}d\theta, \quad \sigma_1 < \gamma = \mathrm{Re}\, s < \sigma_2, \tag{A.1}$$

where σ_1, σ_2 determine the limits of existence of the Mellin image $\mathfrak{F}(s)$. Hence (28) yields, for the Mellin image,

$$\overline{K}(\theta) \overset{MT}{=} \mathfrak{K}(s) = \frac{c}{\tau_0^2} \sum_{l=-M}^{L} \xi^{(d_f+s-2)l} \mathfrak{F}(s). \tag{A.2}$$

We took into account the following property of the Mellin transform: $F(\theta\xi^{-l}) \overset{MT}{=} \xi^{l\cdot s}\mathfrak{F}(s)$. By summing the series (A.2), and taking into account that $M \gg 1$, we obtain

$$\mathfrak{K}(s) \simeq \frac{c}{\tau_0^2}\frac{\xi^{(d_f+s-2)L}\mathfrak{F}(s)}{\left(1-\xi^{-(d_f+s-2)}\right)}, \qquad \max(\sigma_1, 2-d_f) < \gamma = \operatorname{Re} s < \sigma_2, \tag{A.3}$$

where the convergence conditions for the geometrical progression were taken into account. Applying the inverse Mellin transform to Eq. (A.3), we obtain

$$\mathfrak{K}(s) \overset{IMT}{=} \overline{K}(\theta) = \frac{1}{2\pi i}\int_{\gamma-i\infty}^{\gamma+i\infty}\mathfrak{K}(s)\theta^{-s}ds = \frac{c\tau_0^{-2}}{2\pi i}\int_{\gamma-i\infty}^{\gamma+i\infty}\frac{\xi^{(d_f+s-2)L}\mathfrak{F}(s)}{1-\xi^{-(d_f+s-2)}}\theta^{-s}ds. \tag{A.4}$$

For the calculation of the Mellin–Barnes integrals in the right-hand side of (A.4), we redefine the integration path as the semicircular integration contour located correspondingly on the left-hand side (contour $L_{-i\infty}$) or on the right-hand side (contour $L_{+i\infty}$) and then use Cauchy's theorem, namely

$$\overline{K}(\theta) = \pm\sum_k \operatorname*{Re}_{s_k} s\left[\mathfrak{K}(s)\theta^{-s}\right]. \tag{A.5}$$

Here, the set of s_k determines the poles of the Mellin image $\mathfrak{K}(s)$, which should be located inside the chosen contour. The poles s_k are divided on two groups: $s_k^{(1)}$ are the poles of the function $\mathfrak{F}(s)$, and $s_k^{(2)}$ are associated with the poles of expression $\left(1-\xi^{-(d_f+s-2)}\right)^{-1}$ which, in turn, are easily found:

$$s_k^{(2)} = 2-d_f+i\Omega k, \quad \Omega = \frac{2\pi}{\ln\xi}, \ k\in\mathbb{Z}. \tag{A.6}$$

From the conditions (A.3) imposed on the range of convergence of the Mellin image, one can conclude that the integration line in (A.4) is located always on the right-hand side, in contrast to poles $s_k^{(2)}$, which are located on the left-hand side.

Let us consider the structure of the poles $\mathfrak{F}(s)$. In the most common cases (some of them have been discussed in Section 2), the Mellin image $\mathfrak{F}(s)$ belongs to the class of the hypergeometric functions having the form

$$\mathfrak{F}(s) = \frac{\prod_{j=1}^{m}\Gamma(a_j+\alpha_j s)\prod_{j=1}^{n}\Gamma(b_j-\beta_j s)}{\prod_{j=1}^{p}\Gamma(c_j+\gamma_j s)\prod_{j=1}^{q}\Gamma(d_j-\delta_j s)}, \tag{A.7}$$

where the parameters $a_j, b_j, c_j, d_j \in R$, $\alpha_j, \beta_j, \gamma_j, \delta_j \in R_+$. For example, for the memory functions defined in Eq. (21), the corresponding Mellin images have the form [39]

$$\mathfrak{F}(s) = \begin{cases} \dfrac{1}{\sqrt{\pi}}\Gamma\left(\dfrac{s}{2}\right), & \operatorname{Re} s > 0, \quad \text{(a)} \\ \dfrac{1}{\pi}\Gamma\left(\dfrac{s}{2}\right)\Gamma\left(1-\dfrac{s}{2}\right), & 0 < \operatorname{Re} s < 2, \quad \text{(b)} \\ -\dfrac{2\Gamma(s-1)}{\Gamma\left(\frac{1+s}{2}\right)\Gamma\left(\frac{1-s}{2}\right)}, & 0 < \operatorname{Re} s < 2 \quad \text{(c)}, \end{cases} \tag{A.8}$$

while for a memory function written in the form (18) the corresponding Mellin image has the form [39]

$$\mathfrak{F}(s) = \kappa^{1-s}\frac{\Gamma(s/2)}{2\Gamma(2-s/2)}. \tag{A.9}$$

In addition to these observations, it has been shown in Refs. [40,41] that the Mellin images of frequently used functions for the description of dielectric susceptibility (Cole–Cole, Cole–Davidson, Havriliak–Negami and the stretched exponential function) can also be expressed in the form of function (A.7). The function (A.7) has the set of poles

$$s_k^{(1)} = -(a_j+k)/\alpha_j, \quad j=\overline{1,m}, \ k=0,1,2,\ldots, \tag{A.10}$$

$$s_k^{(1)} = (b_j+k)/\beta_j, \quad j=\overline{1,n}, \ k=0,1,2,\ldots, \tag{A.11}$$

which, as a rule, do not intersect and, in consequence, are simple ones. Besides, the poles (A.10) are separated from the poles (A.11) by a curve. Hence, one may conclude that the contour $L_{i\infty}$ leaves all poles defined by Eq. (A.10) on the left-hand side, while the poles like (A.11) are located to the right-hand side of the contour.

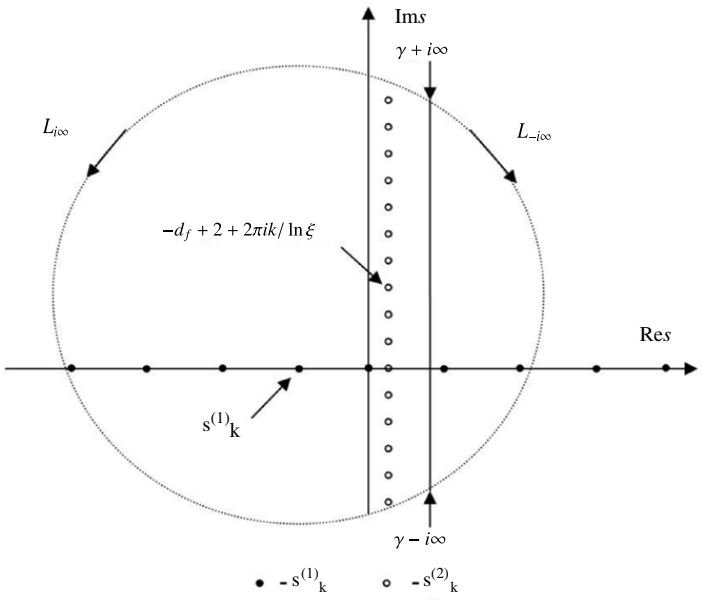

Fig. A.1. The schematic location of poles $s_k^{(1)}$ and $s_k^{(2)}$.

Fig. A.1 illustrates the location of all poles and contours in the complex plane. If one supposes that the poles $s_k^{(2)}$ from (A.6) located to the left-hand side of contour $L_{i\infty}$ do not intersect with poles (A.10), then from (A.5) (choosing the contour $L_{-i\infty}$ appropriately) we have

$$\overline{K}(\theta) = \overline{K}_1(\theta) + \overline{K}_2(\theta),$$

$$\overline{K}_1(\theta) = \sum_k \operatorname*{Re\,s}_{s_k^{(1)}} \left[\Re(s)\theta^{-s}\right] = c\tau_0^{-2} \sum_k \frac{\xi^{-((2-d_f)-s_k^{(1)})L} \operatorname*{Re\,s}_{s=s_k^{(1)}} [\mathfrak{F}(s)]}{1 - \xi^{-(s_k^{(1)}-(2-d_f))}} \theta^{-s_k^{(1)}},$$

$$\overline{K}_2(\theta) = \sum_k \operatorname*{Re\,s}_{s_k^{(2)}} \left[\Re(s)\theta^{-s}\right] = \frac{c\tau_0^{-2}}{\ln\xi} \sum_{k=-\infty}^{\infty} \mathfrak{F}\left(2 - d_f + i\Omega k\right) \theta^{-(2-d_f+i\Omega k)}. \tag{A.12}$$

Clearly, the components of the memory function $\overline{K}_2(\theta)$ do not depend on the limiting value of L (from (27)) and contain log-periodic oscillations, which are combined with the power-law dependence in the form of the product

$$\overline{K}_2(\theta) = \frac{c\tau_0^{-2}}{\theta^{2-d_f}\ln\xi} w_{d_f}\left(\frac{\ln\theta}{\ln\xi}\right), \qquad w_{d_f}(x) = \sum_{k=-\infty}^{\infty} \mathfrak{F}\left(2 - d_f + i\Omega k\right) e^{2\pi ikx}. \tag{A.13}$$

If one supposes that the function $\mathfrak{F}(s)$ has a form similar to Eq. (A.7), then choosing the corresponding contour $L_{-i\infty}$, and taking into account that

$$\operatorname*{Re\,s}_{s=-(a_j+k)/\alpha_j} \left[\Gamma(a_j + \alpha_j s)\right] = \frac{(-1)^k}{\alpha_j k!}, \tag{A.14}$$

we have

$$\overline{K}_1(\theta) = c\tau_0^{-2}\xi^{-(2-d_f)(L+1)} \sum_{j_1=1}^{m}\sum_{k=0}^{\infty} \frac{\prod_{j=1, j\neq j_1}^{m} \Gamma\left(a_j - \frac{\alpha_j a_{j_1}}{\alpha_{j_1}} - \frac{\alpha_j}{\alpha_{j_1}}k\right) \prod_{j=1}^{n} \Gamma\left(b_j + \frac{\beta_j a_{j_1}}{\alpha_{j_1}} + \frac{\beta_j}{\alpha_{j_1}}k\right)}{\prod_{j=1}^{p} \Gamma\left(c_j - \frac{\gamma_j a_{j_1}}{\alpha_{j_1}} - \frac{\gamma_j}{\alpha_{j_1}}k\right) \prod_{j=1}^{q} \Gamma\left(d_j + \frac{\delta_j a_{j_1}}{\alpha_{j_1}} + \frac{\delta_j}{\alpha_{j_1}}k\right)}$$

$$\times \frac{(-1)^{k+1}\left(\theta\xi^{-(L+1)}\right)^{(a_{j_1}+k)/\alpha_{j_1}}}{\alpha_{j_1} k! \left(1 - \xi^{((a_{j_1}+k)/\alpha_{j_1}+2-d_f)}\right)}. \tag{A.15}$$

This series converges for all $0 \leq \theta < \infty$, if the following constraints are fulfilled:

$$\Delta_1 = \sum_{j=1}^{m} \alpha_j + \sum_{j=1}^{q} \delta_j - \sum_{j=1}^{n} \beta_j - \sum_{j=1}^{p} \gamma_j > 0, \tag{A.16}$$

$$\Delta_2 = \sum_{j=1}^{n} \beta_j - \sum_{j=1}^{p} \gamma_j + \sum_{j=1}^{m} \alpha_j - \sum_{j=1}^{q} \delta_j \geq 0. \tag{A.17}$$

For $\Delta_1 = 0$, series (A.15) converges only in the interval $0 \leq \theta < \zeta$, where

$$\zeta = \sqrt{\xi^{-1}} \prod_{j=1}^{m} \beta_j^{-\beta_j} \prod_{j=1}^{p} \gamma_j^{-\gamma_j} \prod_{j=1}^{n} \alpha_j^{\alpha_j} \prod_{j=1}^{p} \delta_j^{\delta_j}. \tag{A.18}$$

In order to find an analytical continuation of the function $\overline{K}_1(\theta)$ in the region $\theta > \zeta$, it is necessary to select the contour $L_{+i\infty}$ containing only the poles (A.11):

$$\overline{K}(\theta) = \overline{K}_1(\theta) = c\tau_0^{-2}\xi^{-(2-d_f)(L+1)} \sum_{j_1=1}^{n} \sum_{k=0}^{\infty} \frac{\prod_{j=1,}^{m} \Gamma(a_j + (b_{j_1}+k)\alpha_j/\beta_{j_1}) \prod_{j=1, j\neq j_1}^{n} \Gamma(b_j - (b_{j_1}+k)\beta_j/\beta_{j_1})}{\prod_{j=1}^{p} \Gamma(c_j + (b_{j_1}+k)\gamma_j/\beta_{j_1}) \prod_{j=1}^{q} \Gamma(d_j - (b_{j_1}+k)\delta_j/\beta_{j_1})}$$

$$\times \frac{(-1)^k \left(\theta\xi^{-L}\right)^{-(b_{j_1}+k)/\beta_{j_1}}}{\beta_{j_1} k! \left(1 - \xi^{(-(b_{j_1}+k)/\beta_{j_1}+2-d_f)}\right)}. \tag{A.19}$$

Clearly, conditions (A.16) and (A.17) are satisfied only for the first and third functions from (A.8) ((a) and (c)) and (A.9). As for the second function from (A.8) (b), $\Delta_1 = 0$, and so the effective memory function is represented in the form (A.12) and contains log-periodic oscillations for the dimensionless time in the interval $0 < \theta < 1/\sqrt{\xi}$. For times $\theta > 1/\sqrt{\xi}$, the log-periodic oscillations vanish, and finally the memory function can be written in the form of Eq. (A.19).

Now we assume that conditions (A.16) and (A.17) are fulfilled and in addition the limitations ($L \to \infty$) are removed. Then, in this limit, one can see from (A.15) that $\overline{K}_1(\theta) \to 0$, and finally the effective memory function is determined by expression

$$\overline{K}(\theta) = \frac{c\Omega\tau_0^{-2}}{2\pi} \sum_{k=-\infty}^{\infty} \mathfrak{F}\left(2 - d_f + i\Omega k\right) \theta^{-(2-d_f+i\Omega k)}. \tag{A.20}$$

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