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From stretched exponential to inverse power-law: fractional dynamics, Cole–Cole relaxation processes, and beyond

Ralf Metzler [a,*], Joseph Klafter [b]

[a] *Department of Physics, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Room 12-109, Cambridge, MA 02139, USA*

[b] *School of Chemistry, Tel Aviv University, Ramat Aviv, 69978 Tel Aviv, Israel*

Abstract

We present a generalisation of the classical exponential relaxation based on the fractional Fokker–Planck equation framework. We show how fractional dynamics modifies the Brownian dynamics underlying standard relaxation processes, and gives rise to the Mittag–Leffler relaxation of modes and moments. The latter is characterised through a turnover from an initial stretched exponential to a final inverse power-law pattern and the associated complex susceptibility corresponds to the Cole–Cole pattern. © 2002 Published by Elsevier Science B.V.

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Classically, relaxation processes are described in terms of the exponential function

$$\phi(t) = \mathrm{e}^{-t/\tau}, \quad t \geqslant 0, \tag{1}$$

which is often referred to as Maxwell–Debye relaxation [1]. In Eq. (1), the relaxation function $\phi(t)$ possesses the characteristic time scale τ, and the normalised initial condition $\phi(0) = 1$. The exponential function (1) fulfills the relaxation equation

$$\frac{\mathrm{d}}{\mathrm{d}t}\phi(t) = -\tau^{-1}\phi(t); \quad \phi(0) = 1. \tag{2}$$

The Maxwell–Debye relaxation is fully characterised through the relaxation time scale τ, and in this sense it is universal. In many systems, however, the dynamical behaviour shows pronounced deviations from the ideal exponential pattern. Experimental results in the time domain are often described in terms of the Kohlrausch–Williams–Watts (KWW) or stretched exponential function [2]

$$\phi(t) = \mathrm{e}^{-(t/\tau)^{\beta}}, \quad 0 < \beta < 1, \tag{3}$$

or through asymptotic power-laws

$$\phi(t) = \frac{1}{1 + (t/\tau)^{\delta}}, \quad \delta > 0, \tag{4}$$

which show the long-time inverse power-law behaviour $\phi(t) \sim (t/\tau)^{-\delta}$, the Nutting law [3].

* Corresponding author. Present address: Nordita, Blegdamsvej 17, DK-2100 Copenhagen, Denmark.

E-mail addresses: metz@nordita.dk (R. Metzler), klafter@post.tau.ac.il (J. Klafter).

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Experimentally, systems are often probed in frequency space under an harmonic external driving, yielding the complex susceptibility $\chi(\omega) = \chi'(\omega) - \mathrm{i}\chi''(\omega)$ as a function of the driving frequency ω. By definition, $\chi(\omega)$ is connected to the temporal relaxation function through the relation

$$\chi(\omega) \equiv \int_0^\infty \mathrm{e}^{-\mathrm{i}\omega t}\,\mathrm{d}(-\phi(t)). \tag{5}$$

Due to this definition, the complex susceptibility (5) is dimensionless. Mathematically, we note that the transformation (5) is equivalent to the Laplace transformation $\tilde{\phi}(p) \equiv \mathfrak{L}\{\phi(t)\} = \int_0^\infty (-\mathrm{d}\phi(t)/\mathrm{d}t)\mathrm{e}^{-pt}\,\mathrm{d}t$ of the negative time derivative of the relaxation function $\phi(t)$ followed by the rotation $p \to \mathrm{i}\omega$. Due to the differentiation theorem $\mathfrak{L}\{\mathrm{d}\phi(t)/\mathrm{d}t\} = p\tilde{\phi}(p) - 1$, we can rephrase the definition for the complex susceptibility in the more convenient form

$$\chi(\omega) = \left[1 - p\tilde{\phi}(p)\right]_{p=\mathrm{i}\omega}. \tag{6}$$

Following Eq. (6), the spectral distribution corresponding to the exponential function (1) is given through the behaviour

$$\chi(\omega) = \frac{1}{1 + \mathrm{i}\omega\tau}. \tag{7}$$

Note that the initial amplitude $\phi(0) = 1$ (normalisation) can be recovered in the low frequency limit $\omega \to 0$. Moreover, the associated power spectrum is given in terms of the Lorentzian (Cauchy distribution)

$$|\chi(\omega)|^2 = \frac{1}{(1 + \omega^2\tau^2)}. \tag{8}$$

While Eqs. (3) and (4) introduce generalisations to the Maxwell–Debye pattern (1) in the time domain, in frequency space the Cole–Cole pattern [4]

$$\chi(\omega) = \frac{1}{1 + (\mathrm{i}\omega\tau)^\alpha} \tag{9}$$

with $0 < \alpha \leqslant 1$ has been widely used to modify Eq. (7) in order to phenomenologically fit experimental data for the complex susceptibility.

We note that in the limits $\alpha = \beta = 1$, both the KWW form (3) and the Cole–Cole pattern (9) reduce to the corresponding classical results (1) and (7). In contrast, the power-law form (4) stands out in that it does not contain the exponential relaxation function as a limiting case.

In what follows, we report a dynamic framework which leads to relaxation functions of the Mittag–Leffler type. These interpolate between an initial stretched exponential behaviour with KWW index α $(0 < \alpha \leqslant 1)$ and a terminal inverse power-law pattern $\sim(\tau/t)^\alpha$, and the corresponding complex susceptibility is exactly given by the Cole–Cole form (9) with Cole–Cole index α [5]. The generalised dynamical equation fulfilled by the Mittag–Leffler function is the fractional relaxation equation which reduces to Eq. (2) in the limit $\alpha = 1$. It will also be argued that a combination of Mittag–Leffler processes with different time scales reproduces numerically more complicated behaviours such as the Havriliak–Negami (HN) behaviour of the complex susceptibility [6].

We start off by generalising the classical Markoffian diffusion-relaxation models to cases with scale-invariant memory with infinite moments and replace the dynamical equations by fractional ones [7]. After discussing some basic properties of the fractional framework, we proceed by deriving the Mittag–Leffler relaxation pattern and argue that it fulfills the Kramers–Kronig relation.

In a rather general perspective, relaxation theory can be based on the diffusion of defects in the system under consideration. Originally introduced by Zener [8] to explain the relaxation of the strain field in a linear solid by considering the interplay between strain and thermodynamical heat field, the model was generalised by Glarum [9] assuming that vacancies such as microscopic cavities, or the random orientation of crystallites diffuse within the system, and when they meet an initially prepared excited state (microscopic dipole, stress, etc.), the latter is allowed to relax. This formalism was generalised by Shlesinger [10] and Blumen et al. [11], who introduced the target model to processes with anomalous statistics. On the basis of the latter, Glöckle and Nonnenmacher [12] derived fractional relaxation processes.

Similarly, we want to consider the relaxation of an excitation undergoing diffusion in the presence of an external potential. In this procedure, the temporal mode relaxation or its frequency space analogue can be measured. Consider first the standard Markoffian model, in which it is assumed that the spatial spreading of the walkers (defects, excitations etc.) follows the diffusion equation

$$\frac{\partial P}{\partial t} = K\frac{\partial}{\partial x^2}P(x,t), \tag{10}$$

where K is the diffusion constant. In Eq. (10), the probability density function (pdf) $P(x,t)$ to find a given excitation at position x at time t is given in terms of the Gaussian $P(x,t) = (4\pi Kt)^{-1/2} \times \exp(-x^2/(4Kt))$. Consequently, a relaxation function $\phi(t)$ can be defined which is connected to the temporal decay of a given mode k, through

$$\phi(t) \equiv P^*(k,t) = \mathrm{e}^{-Kk^2t}, \tag{11}$$

where P^* is the Fourier transform $\int_{-\infty}^{\infty} \mathrm{e}^{\mathrm{i}kx}P(x,t)\,\mathrm{d}x$ of P. As experimental techniques probe the behaviour of the system for a given mode, Eq. (11) gives the temporal relaxation of a macroscopic excitation.

Let us now discuss generalisations of Eqs. (10) and (11) for systems which exhibit some form of disorder which gives rise to slowly decaying, scale-free memory effects in the dynamical behaviour which are manifest in diffusion and relaxation processes in these materials. In particular, we consider the case that the system is not homogeneous but assume that the diffusing excitations are subject to some external force field [13], like an external electric field. Self-similar dynamical evolutions of the power-law type are widely known in a broad range of systems [14]. A dynamical framework which accounts for such patterns has recently been introduced: fractional dynamics [7,15]. The central equation of fractional dynamics is the fractional Fokker–Planck equation [16]: [1]

$$\frac{\partial P}{\partial t} = {}_0D_t^{1-\alpha}\left(\frac{\partial}{\partial x}\frac{V'(x)}{m\gamma_\alpha} + K_\alpha\frac{\partial^2}{\partial x^2}\right)P(x,t), \tag{12}$$

which governs the temporal evolution of the pdf $P(x,t)$ in the presence of the external potential $V(x)$. The basic ingredient of Eq. (12) is the fractional Riemann–Liouville operator ${}_0D_t^{1-\alpha} \equiv (\mathrm{d}/\mathrm{d}t({}_0D_t^{-\alpha}))$ defined through the convolution [17]

$${}_0D_t^{-\alpha}P(x,t) \equiv \frac{1}{\Gamma(\alpha)}\int_0^t \mathrm{d}t'\frac{P(x,t')}{(t-t')^{1-\alpha}}, \tag{13}$$

which possesses the important property

$$\int_0^\infty {}_0D_t^{-\alpha}P(x,t)\mathrm{e}^{-pt}\,\mathrm{d}t = p^{-\alpha}\tilde{P}(x,p) \tag{14}$$

under Laplace transformation. Mathematically, the definition (13) represents a convolution of the pdf P with a power-law memory kernel. The latter decays slowly in the course of time and therefore gives rise to the deviations from the exponential mode relaxation which is characteristic for the Markoffian case. In Eq. (12), K_α is the generalised diffusion constant of dimension $[K_\alpha] = \mathrm{cm}^2/\mathrm{s}^\alpha$. As the fractional Fokker–Planck equation describes systems close to thermal equilibrium, it obeys a generalised Einstein–Stokes relation, $K_\alpha = k_\mathrm{B}T/(m\gamma_\alpha)$, with the generalised friction constant γ_α of dimension $[\gamma_\alpha] = \mathrm{s}^{\alpha-2}$ [7,16,18], and therefore the fractional Fokker–Planck equation is a natural generalisation of the standard case.

In the force-free case, the fractional Fokker–Planck Eq. (12) describes subdiffusion,

$$\langle x^2(t)\rangle = 2K_\alpha\frac{t^\alpha}{\Gamma(1+\alpha)}, \tag{15}$$

i.e., the random walker spreads less efficiently than a Brownian random walker whose mean squared displacement grows linearly in time. The fractional Fokker–Planck equation can be derived from a continuous time random walk scheme [19], and it can be shown to be connected to a multiple trapping process whose waiting times are broadly distributed [18]. The fractional Fokker–Planck equation is equivalent to the generalised master equation with power-law memory and therefore corresponds to a well-defined stochastic process [20].

[1] Here and in the following, we restrict the discussion to the one-dimensional case.

Eq. (12) can be solved by the method of separation of variables. By help of the separation ansatz $P(x,t) = T(t)\varphi(x)$ [16], the fractional Fokker–Planck Eq. (12) can be decoupled. The spatial eigenequation is the same as for the classical Fokker–Planck equation; however, for the temporal eigenequation for the eigenvalue λ_n, the fractional relaxation equation [12,21],

$$\frac{\mathrm{d}T_n(t)}{\mathrm{d}t} = -\lambda_n {}_0D_t^{1-\alpha} T_n(t), \tag{16}$$

obtains. In the limit $\alpha = 1$, the fractional operator reduces to the identity operator, and Eq. (16) is but the standard relaxation equation (2). Eq. (16) can be solved via Laplace transformation, making use of the property (14), or through a power series ansatz. In Laplace space, Eq. (16) is algebraic, and its solution is

$$\tilde{T}_n(p) = p^{-1}/(1 + (p\tau)^{-\alpha}) \tag{17}$$

with $T_n(0) = 1$ and $\tau^{-\alpha} \equiv \lambda_n$. Via inverse Laplace transformation, the solution

$$T_n(t) = E_\alpha(-(t/\tau)^\alpha) \tag{18}$$

is recovered. Here, E_α denotes the Mittag–Leffler function [22] whose series expansion reads

$$E_\alpha(-z) = \sum_{n=0}^{\infty} \frac{(-z)^n}{\Gamma(1+\alpha n)}. \tag{19}$$

For $0 < \alpha \leqslant 1$, the Mittag–Leffler function is positive and strictly monotonically decreasing [22]. The series expansion (19) shows the proximity between the Mittag–Leffler function and the exponential function, and it is obvious that in the limit $\alpha = 1$, it reduces to the the exponential function. For $\alpha = 1/2$, the Mittag–Leffler function can be expressed in terms of the complementary error function, $E_{1/2}(-(t/\tau)^{1/2}) = \mathrm{e}^{t/\tau}\mathrm{erfc}((t/\tau)^{1/2})$. In Fig. 1, we compare the Mittag–Leffler relaxation for $\alpha = 1/2$ with the KWW relaxation of index 1/2 and the asymptotic inverse power-law $(t/\tau)^{-1/2}$. In general, $E_\alpha(-(t/\tau)^\alpha)$ exhibits the limiting behaviours

$$E_\alpha(-(t/\tau)^\alpha) \sim \begin{cases} 1 - t^\alpha/\Gamma(1+\alpha), & t \ll \tau, \\ (\Gamma(1-\alpha)(t/\tau)^\alpha)^{-1}, & t \gg \tau. \end{cases} \tag{20}$$

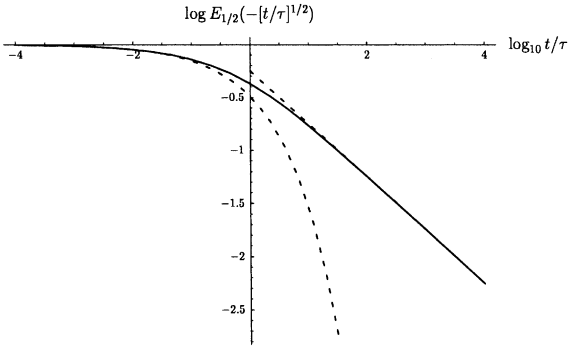

Fig. 1. Mittag–Leffler relaxation in $\log_{10}$–$\log_{10}$ representation. The full line represents the Mittag–Leffler function for index 1/2. The dashed lines depict the initial stretched exponential behaviour and the final inverse power-law pattern, demonstrating the interpolating character between both limiting forms.

Thus, the Mittag–Leffler function interpolates between an initial stretched exponential (KWW) pattern and a terminal inverse power-law decay, both of index α. This interpolating behaviour is well known from rheological modelling, see, for instance, Ref. [23]. We note that by Tauberian theorems, the power-law asymptote $(t/\tau)^{-\alpha}$ for $t \gg \tau$ corresponds to the $p \ll \tau^{-1}$ behaviour of the Laplace transform (17).

The Mittag–Leffler pattern dominates the mode relaxation, and the relaxation of moments in systems with a restoring force of the fractional Fokker–Planck equation (12). The multiple trapping model underlying Eq. (12) states that a diffusing particle can get occasionally trapped at a given space point, and only be released after a given waiting time t. After release, the particle diffuses until it gets trapped again, and so forth. Thereby, the duration of individual waiting periods, t, is a random variable distributed according to the waiting time pdf $\psi(\mathrm{t}) \sim A_\alpha \mathrm{t}^{-1-\alpha}$ [18]. The slow relaxation manifested in the Mittag–Leffler decay (18) is thus directly related to the waiting times whose characteristic scale $\mathfrak{T} = \int_0^\infty \psi(\mathrm{t})\mathrm{t}\,\mathrm{dt}$ diverges, and therefore also the Mittag–Leffler pattern is scale-free, i.e., it possesses a diverging $\mathfrak{T}$.

Due to the dynamic origin of the scale-free memory which gives rise to the fractional equations, the Mittag–Leffler relaxation might also be connected to the crossing of an activation barrier in the generalisation of the Kramers model if the

low diffusivity limit applies, as was demonstrated in Ref. [24].

The complex susceptibility corresponding to the Mittag–Leffler pattern (18) is obtained from Eq. (17) by virtue of relation (6), the result being exactly the Cole–Cole function (9), as obtained earlier by Weron and Kotulski [5] in a similar context.

As mentioned above, the Mittag–Leffler function is a strictly monotonically decreasing function [22], so that all poles of the complex susceptibility $\chi(\omega)$ must necessarily lie in the upper half of the complex plane. Therefore, the real part

$$\chi'(\omega) = \frac{1 + (\omega\tau)^{\alpha}\cos(\pi\alpha/2)}{1 + 2(\omega\tau)^{\alpha}\cos(\pi\alpha/2) + (\omega\tau)^{2\alpha}} \tag{21}$$

and the imaginary part

$$\chi''(\omega) = \frac{(\omega\tau)^{\alpha}\sin(\pi\alpha/2)}{1 + 2(\omega\tau)^{\alpha}\cos(\pi\alpha/2) + (\omega\tau)^{2\alpha}} \tag{22}$$

are connected through the Kramers–Kronig relation [25]

$$\chi''(\omega) = -\frac{1}{\pi}\mathfrak{P}\int_{-\infty}^{\infty}\frac{\chi'(v)}{v-\omega}\,\mathrm{d}v, \tag{23}$$

where $\mathfrak{P}\int$ denotes the Cauchy principal value.

In Fig. 2, we use the Cole–Cole plot which relates the behaviour of the real and imaginary parts of the complex susceptibility (9) for different values of the fractional order α. Accordingly, the semicircle which corresponds to the exponential relaxation pattern is increasingly compressed vertically for decreasing values of α. However, the property that the parametric plot increases strictly monotonically up to 1/2 and then decays strictly monotonically is preserved.

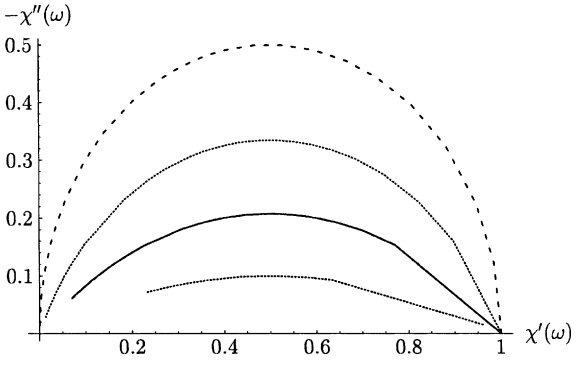

Fig. 2. Cole–Cole plot of the real and imaginary parts (21) and (22) of the susceptibility $\chi(\omega)$ associated with the Mittag–Leffler relaxation, in comparison to the semicircular shape corresponding to the exponential relaxation pattern, $\alpha = 1$ (- - -). The values for α are, from the bottom curve, 1/4, 1/2, 3/4.

For not too low frequencies, both the real part (21) and the imaginary part (22) scale like $\chi'(\omega) \sim \chi''(\omega) \sim \omega^{-\alpha}$, and their ratio is given through

$$\frac{\chi''(\omega)}{\chi'(\omega)} \sim \tan\left(\frac{\pi\alpha}{2}\right), \tag{24}$$

i.e., it is independent of ω, in contrast to the Debye relaxation where the same ratio is $\sim\omega\tau$. Relation (24) corresponds to the universality advocated by Jonscher [26], and it is a direct consequence of the Kramers–Kronig relation (23).

The Cole–Cole behaviour (9) and the corresponding Mittag–Leffler relaxation function are the characteristic response patterns which stem from anomalous diffusion related to the waiting time pdf $\psi(t) \sim A_{\alpha}t^{-1-\alpha}$ within the continuous time random walk scheme. In the system under consideration, there might be combinations of diffusion processes which have a different value for this internal time scale, and they might also be affected differently by the disorder in the system, i.e., exhibit a different α. The combined relaxation process might therefore be composed of two or more individual Mittag–Leffler patterns, giving rise to more elaborate relaxation forms.

A more general a priori form used to fit experimental data is the HN pattern [6]

$$\chi(\omega) = \frac{1}{[1 + (\mathrm{i}\omega\tau)^{\alpha}]^{\gamma}}, \tag{25}$$

which has become a widely used formula to describe experimental data. Usually, one restricts the parameters to $\alpha > 0$ and $\alpha\gamma \leqslant 1$. A special case of the HN pattern (25) for $\gamma = 1$ is therefore the Cole–Cole function (9). The HN parameters α and γ determine the slopes of $\chi''(\omega)$ in the double-logarithmic plot, these being α on the low frequency side and $-\alpha\gamma$ on the high frequency side of the peak. It can be shown that a combination of two fractional processes with different internal time scales can reproduce this behaviour but has the advantage that all related functions are known

analytically [27]. The HN law (25) can be expressed in terms of Fox H-functions in terms of which an exact, closed form analytical representation of (25) in the time domain can be obtained, as is shown in the paper of Hilfer in the same volume [28].

We note that systems which exhibit long-tailed memory kernels feature an inverse power-law behaviour for modes and moments, for long times. Consequently, no characteristic time scale for the relaxation process $\phi(t)$ exists. In such cases, typical materials parameters have to be defined in a more general way. Such a generalisation has recently been introduced for the Deborah number used in rheology [13]. A similar procedure is possible for any materials numbers involving the characteristic relaxation time in the corresponding classical, exponential case.

At this point, we comment on some common bottlenecks encountered in the phenomenological description of dielectric relaxation processes and their spectral analogues. As remarked, e.g., in Ref. [29], the two commonly used fit functions, KWW and HN in time and frequency domain, respectively, are not a Fourier transform pair. This is obvious just from the fact that the former contains only one free parameter whereas the latter includes two; however, there exist exact, analytical Fourier transforms of each function [12,28]. Thus, occasionally there arise problems in the (numerical) Fourier transformation of either a fitted KWW to an HN, or vice versa. By using the Mittag–Leffler function which possesses series expansions for both large and small arguments, relatively simple special representations for rational orders, and an algebraic spectral representations, such difficulties can be avoided; a similar reasoning holds for the combination of two fractional processes which nearly approximate the HN form [27]. The numerical advantage of having exact analytical expressions for all transform pairs including relaxation/retardation time spectra was demonstrated in Ref. [21].

Concluding, we have introduced a connection between relaxation processes and fractional dynamics, giving rise to the ubiquitous Mittag–Leffler behaviour which interpolates between an initial KWW and a final inverse power-law decay. Fractional dynamics occurs in systems which are characterised through multiple trapping events which issue a long-tailed waiting time distribution. Mittag–Leffler patterns are therefore expected to prevail in a wide range of systems. To account for more complex dynamical patterns due to different trap types or different classes of diffusing defects, combinations of fractional processes with different α might arise, and account for the often complex patterns observed in experiments.

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