H-function representations for stretched exponential relaxation and non-Debye susceptibilities in glassy systems

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(Received 28 June 2001; published 28 June 2002)

Analytical expressions in the time and frequency domains are derived for non-Debye relaxation processes. The complex frequency-dependent susceptibility function for the stretched exponential relaxation function is given for general values of the stretching exponent in terms of *H*-functions. The relaxation functions corresponding to the complex frequency-dependent Cole-Cole, Cole-Davidson, and Havriliak-Negami susceptibilities are given in the time domain in terms of *H*-functions. It is found that a commonly used correspondence between the stretching exponent of Kohlrausch functions and the stretching parameters of Havriliak-Negami susceptibilities are not generally valid.

DOI: 10.1103/PhysRevE.65.061510 PACS number(s): 64.70.Pf, 77.22.Gm, 71.55.Jv, 78.30.Ly

Amorphous polymers and supercooled liquids near the glass transition temperature are well known to exhibit nonexponential relaxation behavior in many experiments [1]. Dielectric spectroscopy, viscoelastic modulus measurements, quasielastic light scattering, shear modulus and shear compliance, as well as specific heat measurements all show strong deviations from the exponential Debye relaxation function $f(t)=\exp(-t/\tau)$ where τ is the relaxation time [2].

Most experimental works on glassy dynamics utilize only a small number of empirical nonexponential expressions when fitting to the observed experimental relaxation data. All of these phenomenological fitting formulas are obtained by the method of introducing a fractional ''stretching'' exponent into the Debye expression in the time or frequency domain. In the time domain this method leads to the ''stretched exponential,'' or Kohlrausch, relaxation function, given as

$$f(t)=\exp[-(t/\tau_\beta)^\beta], \quad 0<\beta\leqslant 1, \tag{1}$$

with exponent β and time constant τ_β [3]. Of course all formulas obtained by the method of stretching exponents are constructed such that they reduce to the exponential Debye expression when the stretching exponent becomes unity. Relaxation in the frequency domain is described in terms of a normalized complex susceptibility

$$\hat{\chi}(u)=\frac{\chi(\omega)-\chi_\infty}{\chi_0-\chi_\infty}=1-u\mathcal{L}\{f(t)\}(u), \tag{2}$$

where $u=-\mathrm{i}\omega$, ω is the frequency, $\chi(\omega)$ is a dynamic susceptibility normalized by the corresponding isothermal susceptibility, $\chi_0=\lim_{\omega\to 0}\mathrm{Re}\,\chi(\omega)$ is the static susceptibility, $\chi_\infty=\lim_{\omega\to\infty}\mathrm{Re}\,\chi(\omega)$ gives the ''instantaneous'' response, and $\mathcal{L}\{f(t)\}(u)$ is the Laplace transform of the relaxation function $f(t)$. Extending the method of stretching exponents to the frequency domain, one obtains the Cole-Cole susceptibility [4]

$$\hat{\chi}(u)=\frac{1}{1+(u\tau_\alpha)^\alpha}, \quad 0<\alpha\leqslant 1, \tag{3}$$

the Davidson-Cole expression [5]

$$\hat{\chi}(u)=\frac{1}{(1+u\tau_\gamma)^\gamma}, \quad 0<\gamma\leqslant 1, \tag{4}$$

or the combined Havriliak-Negami form [6] given in Eq. (22) below. Most surprisingly, the analytical transformations between the time and frequency domains for general values of the parameters in these simple analytical expressions seem to be unknown [7], and authors working in the time domain usually employ the stretched exponential function while authors working in the frequency domain use the stretched susceptibilities.

Despite the fact that inserting the Kohlrausch function into Eq. (2) does not yield (3) or (4) [or the related Havriliak-Negami susceptibility in Eq. (22) below], practitioners have tried to establish a relationship between these functions in order to facilitate the transition between the time and the frequency domains [7]. Equally important for practical purposes is the transformation from expressions (3), (4), or (22) in the frequency domain to the corresponding relaxation functions in time [8]. It seems, therefore, that analytical expressions for the Kohlrausch susceptibility in the frequency domain and for the Havriliak-Negami relaxation functions in the time domain are of general importance and broad interest.

Great research activities have ensued from the observation of Williams and Watts [3] that the Kohlrausch susceptibility, obtained by inserting Eq. (1) into Eq. (2), has an analytical expression when $\beta=1/2$. Let me briefly recall their result. One defines the normalized relaxation function as

$$f(t)=\begin{cases}\phi(t)/\phi(0) & \text{for } t\geqslant 0,\\ 0 & \text{for } t<0,\end{cases} \tag{5}$$

where $\phi(t)$ denotes an experimental relaxation function (such as the electrical polarization in dielectric experiments) normalized by the isothermal susceptibility $\phi(0)=\chi_0-\chi_\infty$. Recall now the well known Laplace transform [9]

$$\mathcal{L}\{\exp(-\sqrt{t})\}(u)=\frac{1}{u}-\frac{\sqrt{\pi}}{2}u^{-3/2}\exp\left(\frac{1}{4u}\right)\text{erfc}\left(\frac{1}{2\sqrt{u}}\right), \tag{6}$$

where

$$\text{erfc}\ (x)=\frac{2}{\sqrt{\pi}}\int_x^\infty \exp(-y^2)dy \tag{7}$$

denotes the complementary error function. Inserting this into Eq. (2) and restoring τ_β yields the known result [3]

$$\hat{\chi}(u)=\frac{1}{2}\sqrt{\frac{\pi}{u\tau_\beta}}\exp\left(\frac{1}{4u\tau_\beta}\right)\text{erfc}\left(\frac{1}{2\sqrt{u\tau_\beta}}\right) \tag{8}$$

for the complex susceptibility. According to [7] there are no other cases of $\beta\neq 1$ for which an analytical expression is known for the Kohlrausch susceptibility. My objectives in this paper are (i) to provide analytical expressions for the Kohlrausch susceptibility in the frequency domain in terms of H-functions for all β, (ii) to derive analytical expressions for the Davidson-Cole, Cole-Cole, and Havriliak-Negami relaxation functions in the time domain, and (iii) to show that the approximate correspondence between Kohlrausch and Havriliak-Negami expressions in [7] is limited to a narrow frequency range.

The objectives of this paper are achieved by employing a method based on so-called H-functions [10]. The H-function of order $(m,n,p,q)\in\mathbb{N}^4$ and with parameters $A_i\in\mathbb{R}_+$ $(i=1,\dots,p)$, $B_i\in\mathbb{R}_+$ $(i=1,\dots,q)$, $a_i\in\mathbb{C}$ $(i=1,\dots,p)$, and $b_i\in\mathbb{C}$ $(i=1,\dots,q)$ is defined for $z\in\mathbb{C}$, $z\neq 0$ by the contour integral [10,11]

$$H^{m,n}_{p,q}\left(z\,\middle|\,\begin{matrix}(a_1,A_1),\dots,(a_p,A_p)\\(b_1,B_1),\dots,(b_q,B_q)\end{matrix}\right)=\frac{1}{2\pi i}\int_{\mathcal{L}}\eta(s)z^{-s}ds, \tag{9}$$

where the integrand is

$$\eta(s)=\frac{\prod_{i=1}^{m}\Gamma(b_i+B_is)\prod_{i=1}^{n}\Gamma(1-a_i-A_is)}{\prod_{i=n+1}^{p}\Gamma(a_i+A_is)\prod_{i=m+1}^{q}\Gamma(1-b_i-B_is)}. \tag{10}$$

In Eq. (9) $z^{-s}=\exp\{-s\log|z|-i\arg z\}$ and $\arg z$ is not necessarily the principal value. The integers m,n,p,q must satisfy

$$0\leqslant m\leqslant q,\quad 0\leqslant n\leqslant p, \tag{11}$$

and empty products are interpreted as being unity. For the conditions on the other parameters and the path of integration the reader is referred to the literature [10] (see [12], p. 120ff for a brief summary). The importance of these functions for the present purpose arises from the facts that (i) they contain most special functions of mathematical physics as special cases and (ii) their Laplace transform is again an H-function. Moreover, they possess series expansions that are generalizations of hypergeometric series.

Based on the convenient properties of H-functions, the first objective can now be tackled. An analytical expression for the Laplace transform of the Kohlrausch function is obtained as

$$\mathcal{L}\{\exp[-(t/\tau_\beta)^\beta]\}(u)=H^{11}_{11}\left(u^{-\beta}\,\middle|\,\begin{matrix}(1,\beta)\\(1/\beta,1)\end{matrix}\right). \tag{12}$$

The result is readily obtained from calculating formally

$$\begin{aligned}\mathcal{L}\{H^{m,n}_{p,q}(z)\}(u)&=\frac{1}{2\pi i}\int_0^\infty\int_{\mathcal{L}}\eta(s)e^{-uz}z^{-s}ds\,dz\\&=\frac{1}{2\pi i}\int_{\mathcal{L}}\eta(s)u^{s-1}\Gamma(1-s)ds\\&=\frac{1}{u}H^{m,n+1}_{p+1,q}\left(\frac{1}{u}\,\middle|\,\begin{matrix}(0,1)(a_1,A_1),\dots,(a_p,A_p)\\(b_1,B_1),\dots,(b_q,B_q)\end{matrix}\right)\end{aligned} \tag{13}$$

using the identification

$$\exp[-(t/\tau_\beta)^\beta]=H^{10}_{01}\left[\left(\frac{t}{\tau}\right)^\beta\,\middle|\,\begin{matrix}-\\(0,1)\end{matrix}\right], \tag{14}$$

and then employing identities among H-functions [11,12]. Equation (12) answers the question raised in Ref. [7] concerning the existence of an analytical expression. It will be seen that H-functions are not more difficult to compute than other transcendental functions. Inserting Eq. (12) into (2) leads, after some transformations involving H-function identities, to the Kohlrausch susceptibility in the simple form

$$\hat{\chi}(u)=1-H^{11}_{11}\left((u\tau_\beta)^\beta\,\middle|\,\begin{matrix}(1,1)\\(1,\beta)\end{matrix}\right). \tag{15}$$

This analytical result reduces the calculation of the Kohlrausch susceptibility to a Mellin-Barnes integral of the form (9).

For practical purposes it is also of interest to have series expansions for the analytical results. A Taylor series expansion can be obtained from Eq. (9) using the calculus of residues. It reads for the H^{11}_{11} function

$$H^{11}_{11}\left(z\,\middle|\,\begin{matrix}(a,A)\\(b,B)\end{matrix}\right)=\sum_{k=0}^{\infty}\frac{(-1)^k\Gamma(b+(1-a+k)B/A)}{A\Gamma(k+1)}\times z^{-(1-a+k)/A} \tag{16}$$

TABLE I. Normalized relaxation functions $[f(0)=1]$ with relaxation time τ. Series expansions in the rightmost column are asymptotic series whenever the range of validity of a series expansion is given as a limit.

	$f(t)$	H-function	Series
Debye	$\exp(-t/\tau)$	$H^{10}_{01}\left(\frac{t}{\tau}\middle\vert\begin{matrix}-\\(0,1)\end{matrix}\right)$	$\sum_{k=0}^{\infty}\frac{(-1)^k}{\Gamma(k+1)}\left(\frac{t}{\tau}\right)^k, \quad \frac{t}{\tau}<\infty$ $\exp(-t/\tau), \quad \frac{t}{\tau}\to\infty$
Kohlrausch	$\exp(-(t/\tau_\beta)^\beta)$	$H^{10}_{01}\left(\left[\frac{t}{\tau}\right]^\beta\middle\vert\begin{matrix}-\\(0,1)\end{matrix}\right)$	$\sum_{k=0}^{\infty}\frac{(-1)^k}{\Gamma(k+1)}\left(\frac{t}{\tau_\beta}\right)^{\beta k}, \quad \frac{t}{\tau_\beta}<\infty$ $\exp[-(t/\tau_\beta)^\beta], \quad \frac{t}{\tau_\beta}\to\infty$
Cole-Cole	$E_\alpha[-(t/\tau_\alpha)^\alpha]$	$H^{11}_{12}\left(\left[\frac{t}{\tau_\alpha}\right]^\alpha\middle\vert\begin{matrix}(0,1)\\(0,1)(0,\alpha)\end{matrix}\right)$	$\sum_{k=0}^{\infty}\frac{(-1)^k}{\Gamma(\alpha k+1)}\left(\frac{t}{\tau}\right)^{\alpha k}, \quad \frac{t}{\tau_\alpha}<\infty$ $\sum_{k=1}^{\infty}\frac{(-1)^{k+1}}{\Gamma(1-\alpha k)}\left(\frac{t}{\tau_\alpha}\right)^{-\alpha k}, \quad \frac{t}{\tau_\alpha}\to\infty$
Cole-Davidson	$\frac{\Gamma(\gamma,t/\tau_\gamma)}{\Gamma(\gamma)}$	$1-\frac{1}{\Gamma(\gamma)}H^{11}_{12}\left(\frac{t}{\tau_\gamma}\middle\vert\begin{matrix}(1,1)\\(\gamma,1)(0,1)\end{matrix}\right)$	$1-\frac{1}{\Gamma(\gamma)}\sum_{k=0}^{\infty}\frac{(-1)^k}{(k+\gamma)\Gamma(k+1)}\left(\frac{t}{\tau_\gamma}\right)^{k+\gamma}, \quad \frac{t}{\tau_\gamma}<\infty$ $\frac{\exp(-t/\tau_\gamma)}{\Gamma(\gamma)}\left(\frac{t}{\tau_\gamma}\right)^{\gamma-1}\left[1+\sum_{k=0}^{\infty}\prod_{j=1}^{k}(\gamma-j)\left[\frac{t}{\tau_\gamma}\right]^{-k}\right], \quad \frac{t}{\tau_\gamma}\to\infty$
Havriliak-Negami		$1-\frac{1}{\Gamma(\gamma)}H^{11}_{12}\left(\left[\frac{t}{\tau_H}\right]^\alpha\middle\vert\begin{matrix}(1,1)\\(\gamma,1)(0,\alpha)\end{matrix}\right)$ $\alpha\neq 1$	$-\frac{1}{\Gamma(\gamma)}\sum_{k=0}^{\infty}\frac{(-1)^k\Gamma(k+\gamma)}{\Gamma(\alpha k+\alpha\gamma+1)\Gamma(k+1)}\left[\frac{t}{\tau_H}\right]^{\alpha(k+\gamma)}, \quad \frac{t}{\tau_H}<\infty$ $\frac{1}{\Gamma(\gamma)}\sum_{k=1}^{\infty}\frac{(-1)^{k+1}\Gamma(k+\gamma)}{\Gamma(1-\alpha k)\Gamma(k+1)}\left(\frac{t}{\tau_H}\right)^{-\alpha k}, \quad \frac{t}{\tau_H}\to\infty$

for $B-A\leqslant 0$. Using this result, the Kohlrausch susceptibility is found to have the series expansion (for $|u\tau_\beta|>0$)

$$\hat{\chi}(u)=1-\sum_{k=0}^{\infty}\frac{(-1)^k\Gamma(\beta k+1)}{\Gamma(k+1)}(u\tau_\beta)^{-\beta k}, \tag{17}$$

which reduces its computation to elementary additions and multiplications. The result agrees with a direct evaluation of the Laplace transform of the series expansion for the stretched exponential function. Finally, the asymptotic expansion

$$\hat{\chi}(u)=1-\frac{1}{\beta}\sum_{k=0}^{\infty}\frac{(-1)^k\Gamma[(k+1)/\beta]}{\Gamma(k+1)}(u\tau_\beta)^{k+1} \tag{18}$$

holds for $|u\tau_\beta|\to 0$. It shows that the imaginary part increases linearly at low frequencies similarly to the Cole-Davidson susceptibility.

Using the method of H-functions sketched above also allows one to find analytical expressions for the relaxation functions corresponding to stretched susceptibilities. The results are summarized in the two tables below. Table I gives all relaxation functions, their H-function representations, and their power series expansions, while Table II summarizes the susceptibilities in the frequency domain, their H-function representations, and their power series expansions. In these tables the notation

$$\Gamma(a,x)=\int_a^\infty y^{a-1}e^{-y}dy \tag{19}$$

denotes the complementary incomplete gamma function, and the abbreviation

$$E_a(x)=\sum_{k=0}^{\infty}\frac{x^k}{\Gamma(ak+1)} \tag{20}$$

is the Mittag-Leffler function. In addition, the shorthand notation

$$H_a(x)=H^{11}_{11}\left(-x\middle\vert\begin{matrix}(1,1)\\(1,a)\end{matrix}\right) \tag{21}$$

was introduced for writing the Kohlrausch susceptibility.

TABLE II. Normalized frequency-dependent complex susceptibilities ($u=-\mathrm{i}\omega$). Series expansions in the rightmost column are asymptotic series whenever the range of validity of a series expansion is given as a limit.

	$\hat{\chi}(u)$	H-function	Series
Debye	$\frac{1}{1+u\tau}$	$H^{11}_{11}\left(u\tau \,\middle\vert\, \begin{matrix}(0,1)\\(0,1)\end{matrix}\right)$	$\sum_{k=0}^{\infty}(-1)^k(u\tau)^k, \quad \lvert u\tau\rvert<1$
			$-\sum_{k=0}^{\infty}(-1)^k(u\tau)^{-k-1}, \quad \lvert u\tau\rvert>1$
Kohlrausch	$1-H_\beta[-(u\tau_\beta)^\beta]$	$1-H^{11}_{11}\left((u\tau_\beta)^\beta \,\middle\vert\, \begin{matrix}(1,1)\\(1,\beta)\end{matrix}\right)$	$1-\sum_{k=0}^{\infty}\frac{(-1)^k\Gamma((k+1)/\beta)}{\beta\Gamma(k+1)}(u\tau_\beta)^{k+1}, \quad \lvert u\tau_\beta\rvert\to 0$
			$1-\sum_{k=0}^{\infty}\frac{(-1)^k\Gamma(\beta k+1)}{\Gamma(k+1)}(u\tau_\beta)^{-\beta k}, \quad \lvert u\tau_\beta\rvert>0$
Cole-Cole	$\frac{1}{1+(u\tau_\alpha)^\alpha}$	$H^{11}_{11}\left((u\tau_\alpha)^\alpha \,\middle\vert\, \begin{matrix}(0,1)\\(0,1)\end{matrix}\right)$	$\sum_{k=0}^{\infty}(-1)^k(u\tau_\alpha)^{\alpha k}, \quad \lvert u\tau_\alpha\rvert<1$
			$-\sum_{k=0}^{\infty}(-1)^k(u\tau_\alpha)^{-\alpha(k+1)}, \quad \lvert u\tau_\alpha\rvert>1$
Cole-Davidson	$\frac{1}{(1+u\tau_\gamma)^\gamma}$	$\frac{1}{\Gamma(\gamma)}H^{11}_{11}\left(u\tau_\gamma \,\middle\vert\, \begin{matrix}(1-\gamma,1)\\(0,1)\end{matrix}\right)$	$\sum_{k=0}^{\infty}\frac{(-1)^k\Gamma(k+\gamma)}{\Gamma(\gamma)\Gamma(k+1)}(u\tau_\gamma)^k, \quad \lvert u\tau_\gamma\rvert<1$
			$-\sum_{k=0}^{\infty}\frac{(-1)^k\Gamma(k+\gamma)}{\Gamma(\gamma)\Gamma(k+1)}(u\tau_\gamma)^{-(k+\gamma)}, \quad \lvert u\tau_\gamma\rvert>1$
Havriliak-Negami	$\frac{1}{[1+(u\tau_H)^\alpha]^\gamma}$	$\frac{1}{\Gamma(\gamma)}H^{11}_{11}\left((u\tau_H)^\alpha \,\middle\vert\, \begin{matrix}(1-\gamma,1)\\(0,1)\end{matrix}\right)$	$\sum_{k=0}^{\infty}\frac{(-1)^k\Gamma(k+\gamma)}{\Gamma(\gamma)\Gamma(k+1)}(u\tau_H)^{\alpha k}, \quad \lvert u\tau_H\rvert<1$
			$-\sum_{k=0}^{\infty}\frac{(-1)^k\Gamma(k+\gamma)}{\Gamma(\gamma)\Gamma(k+1)}(u\tau_H)^{-\alpha(k+\gamma)}, \quad \lvert u\tau_H\rvert>1$

Having computable analytical expressions at hand for the Kohlrausch susceptibility, it becomes possible to investigate the mappings between the Kohlrausch susceptibility and the Havriliak-Negami susceptibility [6]

$$\hat{\chi}(u)=\frac{1}{(1+(u\tau_H)^\alpha)^\gamma} \tag{22}$$

that were postulated in Ref. [7]. Table I and Fig. 5 of Ref. [7] present fits for the Kohlrausch susceptibility using the Havriliak-Negami expression as a fit function. Figure 1 shows the real and imaginary parts of the Kohlrausch susceptibility with $\beta=0.25$ plotted as crosses ($\times$) in a doubly logarithmic plot. The corresponding Havriliak-Negami fit from [7] with $\alpha=0.5164$, $\gamma=0.3706$, and $\tau_H/\tau_\beta=10$ is shown as the solid line. In all calculations $\chi_\infty=1$ and $\chi_0=10$ unless stated otherwise.

Because it is known that the phenomenological susceptibility functions are often inadequate for fitting experimental relaxation spectra, some researchers prefer to discuss not stretching exponents but the width of the imaginary part [13]. Figure 2 plots three characteristic frequencies for all three stretched susceptibility functions against their respective stretching exponent. The first is f_0, the location of the maximum of the imaginary part. The second is f_-, the location of the lower half-width point of the imaginary part. The third is f_+, the location of the upper half-width point of the imaginary part. The half-width points are defined as the frequencies at which the imaginary part has decayed to half of its maximum value.

Figure 2 shows that while the Cole-Cole susceptibility (dashed line for maximum, solid line with triangles for the half widths) is symmetric, the other two susceptibilities are asymmetric. For small values of β (respectively, γ) the Cole-Davidson is more strongly asymmetric than the Kohlrausch susceptibility. Note also that the lower half-width point moves to higher frequencies for diminishing γ in the Cole-Davidson case. The total width of the relaxation peak in decades is the difference between the upper and the lower half width. For $\alpha=\beta=\gamma=0.2$ the total width of the Cole-Cole function is roughly 7 decades, the width of the Kohlrausch susceptibility is roughly 5 decades, and that of the Cole-Davidson is roughly 2.5 decades. Figures 2 and 1 demonstrate that the mapping between the Kohlrausch parameter β and the Cole-Davidson parameter γ that is often employed by practitioners [2] becomes increasingly inaccurate for small values of the stretching exponents.

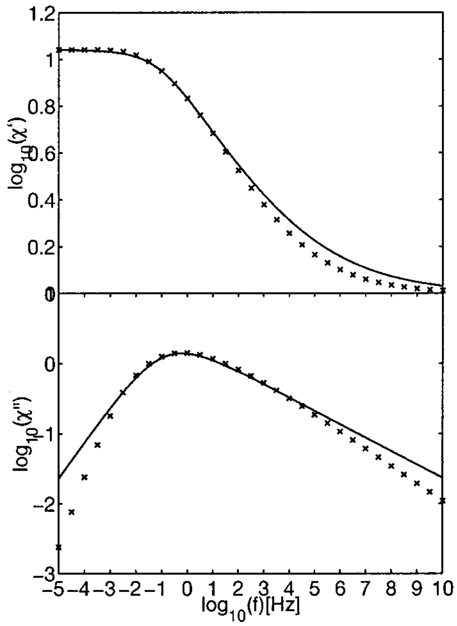

FIG. 1. Comparison of the Havriliak-Negami fit (solid line) with $\alpha = 0.5164$, $\gamma = 0.3706$, and $\tau_H = 10$ for a Kohlrausch susceptibility ($\times$) with $\beta = 0.25$ and $\tau_\beta = 1$. The values were taken from Ref. [7], p. 7310, Table I. In all cases $\chi_\infty = 1$ and $\chi_0 = 10$.

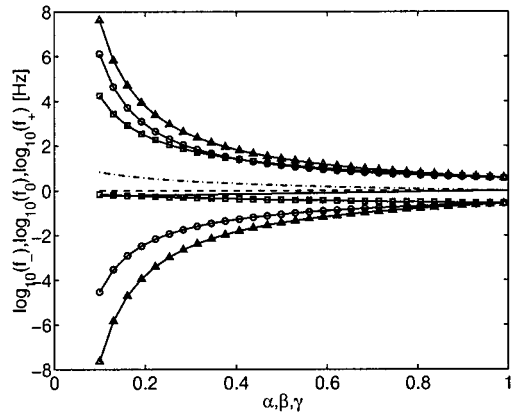

FIG. 2. Location of the frequency of the maximum f_0, frequency f_- of the lower half width, and frequency f_+ of the upper half width of the imaginary part of Cole-Cole, Kohlrausch, and Cole-Davidson susceptibility functions as a function of the respective stretching exponents α, β, γ. The upper and lower half-width frequencies $f_\pm$ are indicated by solid lines connected with symbols [Cole-Cole susceptibility function ($\triangle$), Kohlrausch susceptibility ($\bigcirc$), Cole-Davidson ($\square$)]. The location of the maximum of the imaginary part is indicated by a dashed line for the Cole-Cole function, by a solid line without symbols for the Kohlrausch function, and by a dash-dotted line for the Cole-Davidson function. The relaxation time is always $\tau = 1$.

In summary, the present paper has given unified representations of nonexponential relaxation and non-Debye susceptibilities in terms of *H*-functions. These representations lead to computable expressions that were used to investigate the relations between the Kohlrausch susceptibility and other fit functions. The *H*-function representations given here can help to facilitate the computational transformation between the frequency and time domains in theoretical considerations and experiment.

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