Limitations on distinguishing between representations of relaxation data over narrow frequency ranges

Chad R. Snyder[a)] and Frederick I. Mopsik
National Institute of Standards and Technology, Polymers Division, Gaithersburg, Maryland 20899

In this article, we examine the ability to distinguish between relaxation functions with data over a limited range of frequency. It is demonstrated that over these limited frequency ranges under a variety of conditions, the Cole–Cole equation can be used to fit data generated by the Havriliak–Negami equation. These results show that discerning between several very different broad relaxation functions fit to data obtained over narrow time or frequency ranges is nearly impossible within experimental accuracy. Therefore, the uniqueness of the fit parameters, and hence the ability to verify model predictions, is brought into question. Furthermore, as this conclusion is drawn from comparison of exact functions that experience no dispersion overlaps or instrumental systematic errors that can mask exact fits, the true situation with experimental data is even worse. The same conclusion can be applied to time domain data. [S0021-8979(98)00420-4]

I. INTRODUCTION

In dielectric relaxation studies there are four commonly used interpolating (extrapolating) functions: the Cole–Cole (CC),[1] the Havriliak–Negami (HN),[2] the Cole–Davidson (CD),[3] and the Kohlrausch–Williams–Watts (KWW)[4] equations. (The first three are expressed in the frequency domain and the last two in the time domain.) There have been many articles in the literature that have singled out one of these functions and attempted to give it a physical basis.[5–11] These various theories have been used by others to extract molecular information from the experimental data. However oftentimes, dielectric relaxation data are obtained over a range of 3–4 decades in frequency, because of either measurement limitations or the presence of overlapping relaxations. In this article, we will demonstrate several things. First, over a limited range of frequencies the data cannot usually be used to distinguish meaningfully between these very different functions within experimental accuracy. Consequently, the conclusions regarding the various theoretical models that have been drawn from experimental data that were measured over a narrow frequency range and fitted to these functions become uncertain. Second, because of the experimental indistinguishability of these functions, extrapolation far beyond the frequency window is questionable. Finally, we address the issue of the proper metric for comparing relaxation functions. We will not specifically look at time–domain functions, but as we indicate later, similar conclusions should be obtainable in the time domain.

We also would like to point out that these results do show that as the data range is extended, different relaxation functions can become experimentally distinguishable. We will examine these questions, in a subsequent publication, as they are now arising quite frequently.[12–15]

These questions arose in practice when one of the authors was fitting relaxation data to the HN equation using nonlinear least squares techniques.[16] In that work, it was discovered that when a restricted frequency range was available due to experimental conditions, the least squares routine would not reliably converge and give both exponents of the HN relaxation function. However, when one of the exponents was kept fixed, convergence was obtained. This strongly suggested that the data were not adequate to determine all the parameters in the fit.

Since the exponent parameters in these distributions represent the limiting behavior at high and low frequencies as a power law, the data surrounding the maximum are quite insensitive to the actual limiting slope. Therefore as a measure of how broad a frequency range is required for adequate fitting of a distribution, consider the width at half height of the CC distribution (ϕ_{CC}) which is given by:[1]

$$\phi_{CC}(\omega) = \frac{1}{1+(i\omega\tau)^\chi},\tag{1}$$

where ω is the angular frequency, τ is the relaxation time, and χ is a measure of the distribution width and is also the limiting exponent. For $\chi = 1$, 0.5, and 0.3, the width at half height is 1.14, 2.88, and 5.98 decades, respectively. Therefore, for broad distributions, data centered on the maximum make it extremely difficult to determine these exponents unless the data are much broader than these widths. We will illustrate this point later when we show how the effective power law varies as a function of $\omega\tau$.

In this article, we will fit exact forms of the relaxation functions to each other, to within stated tolerance, without adding any noise or dispersion overlap. We will limit our calculations to the frequency domain since analytical forms of the HN and CC functions in general are only available in the frequency domain. Also, these forms can be quite different and, as will be seen, the results of the fit over the full

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a)Electronic mail: chad.snyder@nist.gov

TABLE I. Results of Cole–Cole fit to Havriliak–Negami equation over limited frequency ranges for HN dispersion strength of 1 and ϵ_∞ of 0.

Frequency range	α	β	χ	τ (s)	ϵ_∞	ϵ_0-ϵ_∞	Maximum $\|\epsilon''_{HN}\text{-}\epsilon''_{CC}\|$ $\times 10^4$
0.01–10 Hz	0.4	0.8	0.387	0.0920	0.043 09	0.9613	8.53
	0.6	0.8	0.579	0.1088	0.040 20	0.9640	33.5
	0.8	0.8	0.767	0.1184	0.035 23	0.9704	91.0
	0.8	0.6	0.736	0.083 44	0.102 05	0.9069	132
	0.8	0.4	0.709	0.055 39	0.231 46	0.7777	138
0.1–100 Hz	0.4	0.8	0.372	0.088 56	0.021 47	0.9979	11.1
	0.6	0.8	0.554	0.105 89	0.017 09	1.0036	42.4
	0.8	0.8	0.739	0.116 23	0.016 36	1.0028	82.8
	0.8	0.6	0.671	0.075 83	0.074 91	0.9882	147
	0.8	0.4	0.601	0.041 34	0.122 42	0.9218	192
1–1000 Hz	0.4	0.8	0.354	0.105 53	0.008 28	1.0553	10.6
	0.6	0.8	0.520	0.130 90	0.004 76	1.0889	30.0
	0.8	0.8	0.688	0.144 53	0.004 05	1.1145	59.8
	0.8	0.6	0.571	0.118 15	0.013 24	1.2097	126
	0.8	0.4	0.453	0.073 61	0.041 76	1.2403	175

range will not be even that close. While other forms could be used, the conclusions will not be affected unless they differ even more strongly than the ones we used, in which case our conclusions are strengthened. We will use the HN function to generate representative data, since it is so versatile and is explicitly in the frequency domain. In fact, it has been claimed that the KWW behavior can be represented by it.[12,13] It should be realized that for real data sets, the requirements for discriminating the functions would be more difficult than what we estimate, due to both experimental error and possible dispersion overlap. It should also be noted that the ability to fit one relaxation function to another over a limited frequency range does not mean to imply that they are equivalent. Moreover, for these functions, if the frequency range is broad enough or covers a suitable frequency range and the data have readily achievable precision, we can easily discriminate among them.

We are not specifically addressing the issue of time–temperature superposition, since relaxation functions are properly defined at a given temperature. In order to have time–temperature superposition, a relaxation function must be invariant with respect to temperature. Therefore, establishing time–temperature superposition over the range of data available will be subject to similar considerations that we discuss in this article. It should be noted that in a previous article by one of us,[17] it was demonstrated that fitting data over a restricted range of time or frequency gave the appearance of time–temperature superposition while a broader range indicated otherwise.

II. PROCEDURE

A. Data generation and fitting

Data sets were generated using the HN equation:

$$\phi_{HN}(\omega) = \frac{1}{[1+(i\omega\tau)^\alpha]^\beta} \tag{2}$$

with the values listed in Table I and with $\tau=1/2\pi$ s. The values for α and β were chosen to represent experimentally realizable values as was the generated frequency range ($f = \omega/2\pi = 0.01$ Hz–10 kHz). The imaginary portion (ϵ'') of the data was then fit [over the following frequency (f) windows: 0.01–10 Hz, 0.1–100 Hz, and 1–1000 Hz] using an unweighted Levenberg–Marquardt algorithm[18] minimizing the squared deviations to the following equation:

$$\frac{\epsilon^*(\omega)-\epsilon_\infty}{\epsilon_0-\epsilon_\infty} = \phi(\omega), \tag{3}$$

where ϵ_0-ϵ_∞ is the dispersion strength, ϵ_∞ is the high frequency dielectric constant, and $\epsilon^*(\omega)$ is the complex frequency dependent dielectric constant:

$$\epsilon^*(\omega) = \epsilon'(\omega) + i\epsilon''(\omega), \tag{4}$$

where ϵ' and ϵ'' are the real and imaginary components of the complex dielectric constant, respectively. The high frequency dielectric constant was then obtained through a linear least squares fit of the real data using the CC (or CD) parameters obtained from the fit of the imaginary portion. The residuals on the real component are comparable to the imaginary fit and will not be presented, as they contribute nothing extra to the discussion.

We have deliberately set our frequency ranges to be off center but include the loss maximum. This is perhaps the most commonly seen situation experimentally. In addition, from the functional forms involved, if only one wing of the dispersion is observed, the CC function always would be expected to fit the data no matter what the underlying relaxation function is like. Note that for the data generated by the HN equation we have set the dispersion strength equal to unity and ϵ_∞ equal to zero. However, in the fits these parameters were allowed to be adjustable, since for real data sets, the dispersion width and ϵ_∞ are not known *a priori* and are

thus determined from the curve fits. The forms of $\phi(\omega)$ used in the above equation were the CC equation [Eq. (1)] and the CD equation:

$$\phi_{CD}(\omega) = \frac{1}{(1+i\omega\tau)^{\delta}}.$$ (5)

B. Least squares

A major point of confusion is the use of least squares to fit one exact function to another exact function.[12–14] Typically this is done with a numeric tool kit using regression analysis which reports out both a standard deviation of fit, and fitted coefficients with an associated estimate of their standard deviation. There even seems to be a claim of one least squares package[14] being better than another package.[12]

We would like to point out that, for a constant set of sampling points, any procedure should produce the same results for the fitted parameters within round-off limits and numerical stability since these are exact functions. Furthermore, within these constraints, the coefficients have no standard deviation associated with them since they are entirely defined by the functions themselves and the sampling set that is chosen.

In addition, the actual value of the reported standard deviation loses any statistical meaning other than a minimization parameter. The fit after all is not being made to a set of data that is capable of being represented by an exact model plus a random deviate with a sampled population. This is the assumption, along with a constant expected variance at each sampled point, associated with the statistical application of least squares.

For example, if two functions have the same asymptotic limits, with a fitting range chosen so that over most of the fitting range the two functions have nearly the same values, the calculated standard deviation approaches a small number simply because the interval over which one function differs significantly from the other is bounded, while the normalization is determined by the size of the sampling window. This is the case for a normalized relaxation function where each end goes to either 0 or 1. This point is emphasized by a well-known result for complete orthonormal Fourier expansions where the series expansion is allowed to differ from the reference by an arbitrary number of fixed points, even though the expansion converges in the mean squared sense.[19]

C. Criterion of fit

While we have employed a least squares technique to find a representation of one function to another, we have not used the computed standard deviation as a measure of goodness of fit. The calculated standard deviation does not have its usual meaning as when data, subject to a random uncertainty, are fit to an assumed, correct functional form. In that case, the computed standard deviation is a measure of the width of the distribution of the data points about their expectation values.[20] For our case, neither the fitted data nor the assumed forms have random deviates. Aside from the question of discrete versus continuous fitting, the calculated fit is subject only to computational errors that are not believed to

be important. Furthermore, our comparisons are not dependent on how optimally the least squares was performed.

For real data sets that have a statistical uncertainty (σ), if the absolute deviation between two fitting functions is less than σ, then the standard deviation for large sets takes a limiting value of σ regardless of how well either function fits the data. This should be contrasted to fitting one exact function to another where the computed standard deviation can go to zero as the regions of significant difference, which may have large values, become small relative to the total interval. Indeed for data where σ is greater than the maximum deviation between the two curves, no fitting procedure will be able to discriminate between the two because the change in computed σ would be insignificant and σ will approach a constant. Therefore, we are using what we feel is a more suitable metric related to the question of uniform convergence, the absolute maximum deviation between the two functions over the interval, a metric commonly used in numerical approximation.[21] While others can be chosen, this is in fact a quite restrictive metric since any widely deviating point will be viewed as a measure of poor fit. Consequently, for a measured data set characterized by a population standard deviation greater than our metric, one will be unable to distinguish between them.

Just as is the case for the least squares, we are making no claims as to whether our fits are the best fits for our metric. Rather, we are simply stating that the comparisons are at least this good. Again, as before, better fits would only strengthen our results.

D. Effective power law exponent

To better examine the functionality of the relaxation function over its range, the HN data were also locally fitted to the power law form:

$$\phi_{PL} = A + Bx^{\gamma}$$ (6)

as a function of normalized frequency $x(x=\omega\tau)$. This was done by choosing three points spaced as a geometric progression with multiplier k. For the assumed functional form, Eq. 6, with values of x_1, x_2, and x_3 equal to x_1, kx_1, k^2x_1, the exponent γ can be evaluated as:

$$\gamma = \ln\left(\frac{\phi_{PL,2}-\phi_{PL,1}}{\phi_{PL,2}-\phi_{PL,3}}\right) \cdot \frac{1}{\ln k}$$ (7)

where $\phi_{PL,1}$, $\phi_{PL,2}$, and $\phi_{PL,3}$, are the relaxation functions at x_1, x_2, and x_3, respectively. The curves we will display utilize a k of $(3/2)^{1/2}$. Numerical computations show that the value of γ is not changed appreciably by changing k.

III. RESULTS AND DISCUSSION

Over the three frequency ranges and the varying values of α, the CC equation always fit the generated data better than the CD equation, with the exception of $\alpha=0.8$, $\beta=0.6$. (The results of CC fits are shown in Table I. The CD results have been omitted.) Because the generated data were defined to have unity dispersion strength, it is a normalized distribution and hence the difference between the generated data and the fit data is the relative error in terms of the
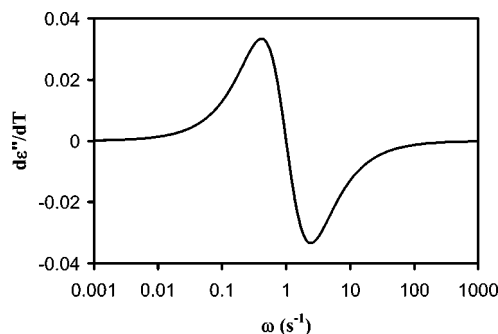
FIG. 1. $d\epsilon''/dT$ as a function of angular frequency (ω) for a Debye relaxation at a temperature T of 300 K, an activation energy of 100 kJ/mol, and unit relaxation time at T.

FIG. 3. Residual in ϵ'' for a Cole–Cole fit to the Havriliak–Negami equation ($\epsilon''_{HN} - \epsilon''_{CC}$) over the frequency range 0.1–100 Hz.

dispersion width. While we do not wish to make an arbitrary criterion for the maximum allowable value for the absolute deviation, we consider a good fit anything less than 1%. For data that are either better or worse than these values, the frequency range would have to be adjusted as needed. In the work by Havriliak and Havriliak[14] a maximum error of 3.5% of ϵ'' is claimed to normally be unmeasurable. This is claimed to be a consequence of limited instrumental precision as well as limitations of temperature control. However, the uncertainty in the time domain spectrometer (TDS),[22] which is commercially available, is less than 0.1% of the measured relaxation. Additionally, Havriliak and Havriliak claimed that temperature control limits the precision of the measurements. A simple calculation shows that this is not correct.

Consider an exponential decay (Debye), where the time constant τ is given by an Arrhenius equation:

$$\tau = \tau_0 \exp\left(\frac{-E_a}{RT}\right). \tag{8}$$

For a temperature T of 300 K, representative activation energy E_a of 100 kJ/mol, and unit relaxation time $d\epsilon''/dT$ is given by:

$$\frac{d\epsilon''}{dT} = \frac{(\omega - \omega^3\tau^2)\dfrac{d\tau}{dT}}{(1 + \omega^2\tau^2)^2}, \tag{9}$$
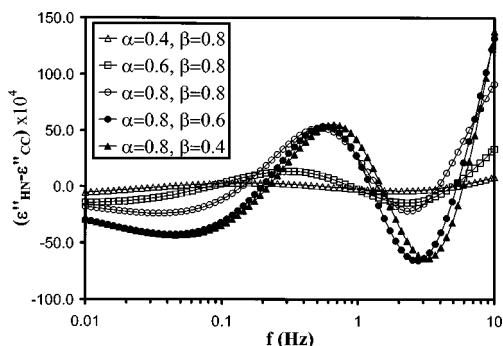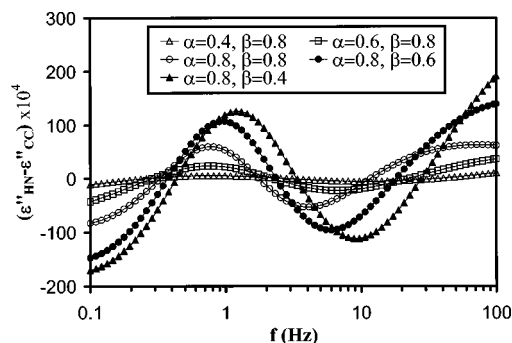
where $d\tau/dT$ is given by:

$$\frac{d\tau}{dT} = \frac{E_a\tau_0}{RT^2} \exp\left(\frac{-E_a}{RT}\right), \tag{10}$$

and where R is the gas constant and $\tau_0 = 2.6 \times 10^{17}$ s to yield a unit relaxation time at 300 K. As can be seen in Fig. 1, the error is sharply peaked at 3.3% for a 1 °C uncertainty. Temperature controllers with control to within 0.1 °C are readily available, showing that the contribution from temperature uncertainty can be easily less than the quoted value of 3%. It should be realized that polymeric materials have relaxations much broader than a Debye and therefore this is an overestimate of the error due to temperature as long as one is not too near a glass transition.

Thus, we have plotted ($\epsilon''_{HN} - \epsilon''_{CC}$) as a function of frequency (f) in Figs. 2–4. In Table I we list the absolute value of the maximum value determined for ($\epsilon''_{HN} - \epsilon''_{CC}$), where ϵ''_{HN} and ϵ''_{CC} are the dielectric loss components from the Havriliak–Negami data and Cole–Cole fit, respectively. As stated previously, the Cole–Davidson fit results have been omitted, because in all but one case the CC function fit the data better.) It should be noted that the highest error observed for the Cole–Cole fits was 1.92×10^{-2} (with $\alpha = 0.8$ and $\beta = 0.4$) and that this value was at the edge of the fitting window. Much lower errors were observed towards the center of the fit window. To better present the nature of the fits, we have plotted in Figs. 5, 6, and 7, the best, average, and worst fits obtained for the Cole–Cole equation, respectively. It is apparent from Fig. 7 that the data can be distinguished from either a Cole–Cole or Cole–Davidson and is therefore fit only by a Havriliak–Negami.
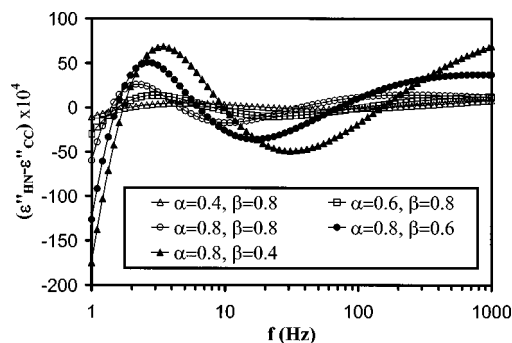

FIG. 2. Residual in ϵ'' for a Cole–Cole fit to the Havriliak–Negami equation ($\epsilon''_{HN} - \epsilon''_{CC}$) over the frequency range 0.01–10 Hz.

FIG. 4. Residual in ϵ'' for a Cole–Cole fit to the Havriliak–Negami equation ($\epsilon''_{HN} - \epsilon''_{CC}$) over the frequency range 1–1000 Hz.
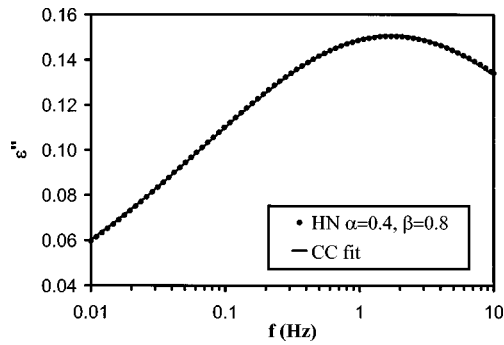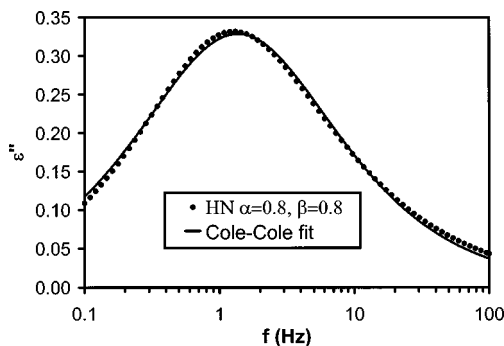
FIG. 5. ϵ'' vs frequency for a Cole–Cole fit to data generated by the Havriliak–Negami equation with $\alpha = 0.4$ and $\beta = 0.8$ over a frequency range of 0.01–10 Hz.

FIG. 7. ϵ'' vs frequency for Cole–Cole and Cole–Davidson fits to data generated by the Havriliak–Negami equation with $\alpha = 0.4$ and $\beta = 0.8$ over a frequency range of 0.1–100 Hz.

Furthermore, Table I clearly illustrates that the dispersion strength and ϵ_∞ obtained by the Cole–Cole fits are reasonable values. Trends are readily observable as well. For a given frequency window (with β fixed), as α is increased, χ increases (as does the dispersion strength) whereas ϵ_∞ decreases. As the frequency window is shifted to higher frequencies, for a fixed value of α and β, χ decreases (as does ϵ_∞), and the dispersion strength increases. If there were good grounds to rule out such trends, these trends could be used as an indicator that the assumed functional form is not adequate to represent the entire relaxation process.

These results indicate several things. First, over limited frequency ranges and for a wide range of conditions, the Cole–Cole equation reproduces the Havriliak–Negami generated data with reasonable values for χ, the dispersion strength, and ϵ_∞. Second, because of this ability to reproduce the data with reasonable fit parameters, it is impossible to distinguish between the HN and CC equations over limited frequency ranges within experimental uncertainty. Furthermore, if we cast the HN equation in the form of the CC or CD equations we obtain the following:

$$\phi_{\omega \to \text{infinity}} = \frac{1}{1 + (i\omega\tau)^{\alpha\beta}}$$

$$= \frac{1}{(1 + i\omega\tau)^{\alpha\beta}}$$

$$= \frac{1}{[1 + (i\omega\tau)^{\alpha}]^{\beta}} \qquad (11)$$

for the high frequency limit. As can be seen in Table I, χ is always greater than $\alpha\beta$, but as the frequency window is shifted to higher frequencies, χ decreases towards $\alpha\beta$, thus properly approaching the asymptotic limit.

We have added an additional plot with rather unrealistic parameters ($\alpha = 0.099$ and $\beta = 0.309$). These values were chosen because Havriliak and Havriliak[14] quote these values as the fit values to the KWW equation with an exponent of 0.04. To display the entire relaxation, Havriliak and Havriliak showed that a frequency range of 10^{-40}–10^{30} Hz was necessary. This range is experimentally inaccessible. The most that could be observable is a small portion of such a relaxation, if the relaxation exists. Using their HN parameters, data were generated for the frequency range from 0.001 Hz to 10 kHz and fit to the Cole–Cole function. As can be seen in Fig. 8, the Cole–Cole fit represents the data extremely well, with a maximum deviation of 5×10^{-5}—a value that under ordinary circumstances cannot be resolved by experimental techniques. Even a linear fit to the data yields only a 7×10^{-4} maximum deviation. This supports the statement we made in Sec. I. If the relaxation is broad enough, it is nearly impossible to determine the exponents since, within experimental error, a curve with $\alpha = 0.099$ and $\beta = 0.306$ is indistinguishable from a curve with $\alpha = 0.086$ and $\beta = 1$ even over a much wider range of frequencies than used above.

As a further example, we have computed the apparent exponent as a function of frequency for our test cases using

FIG. 6. ϵ'' vs frequency for a Cole–Cole fit to data generated by the Havriliak–Negami equation with $\alpha = 0.8$ and $\beta = 0.8$ over a frequency range of 0.1–100 Hz.
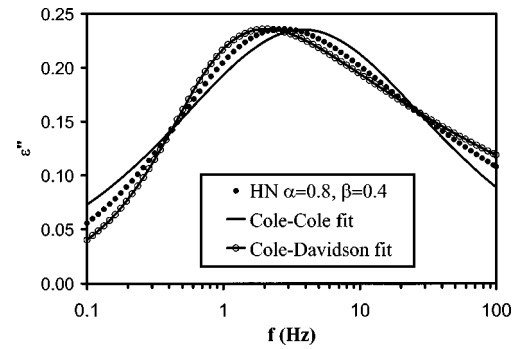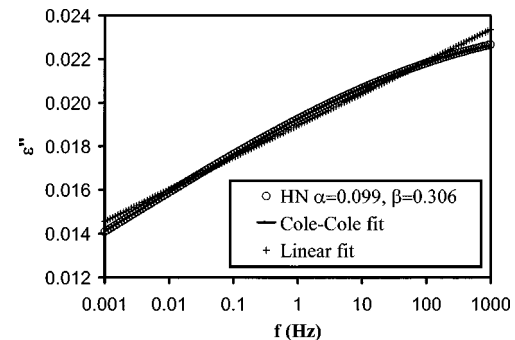

FIG. 8. ϵ'' vs frequency for Cole–Cole and linear fits to data generated by the Havriliak–Negami equation with $\alpha = 0.099$ and $\beta = 0.306$ over a frequency range of 0.001–1000 Hz.
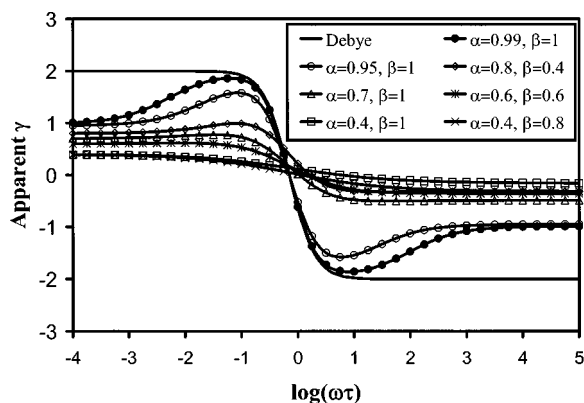
FIG. 9. Apparent power law exponent (γ) as a function of $\log(\omega\tau)$ for various values of α and β.

ϵ' with Eqs. (6) and (7). If the variation of the exponent along a pair of functional curves is similar, the two should be able to be made to resemble each other over that frequency range. From Fig. 9 and our results from direct fitting, it can be seen that this is indeed the case. For this comparison, we display only ϵ' as ϵ'' has a change in sign for the exponent near the maximum. Otherwise, away from the maximum, the results are similar.

Thus the low frequency limit of γ is α, therefore any relaxation curves with the same value of α can be made to resemble each other at low frequencies. Similarly, the high frequency limit of γ is the $\alpha\beta$ product and hence any relaxation curves with the same $\alpha\beta$ product can be made to resemble each other at high frequencies (see Fig. 9 for the two curves: $\alpha=0.6$, $\beta=0.6$ and $\alpha=0.4$, $\beta=0.8$).

A final additional point worth noting is the power law dependence approaching these limits. Power laws are often imputed by measuring slopes on data that do not get too close to the end due to experimental limitations. However, these slopes are assumed to be significant.[23]

In Fig. 9, maxima are observed for limiting values of the exponent greater than 0.5 and most clearly for values greater than 0.7. This surprising result can be understood by observing that for a Debye function, the limiting slope is 2 and not 1 as would be predicted from the above argument. In fact, as the exponent in the HN function or the CC function approaches unity, the maximum approaches a value of 2 and broadens out so that in the limit of an exponent of unity, the correct Debye function result is recovered.

IV. CONCLUSIONS

When data of limited time or frequency range are fit to broad relaxation functions, the significance of the fitted form may be less certain than is appropriate to properly characterize relaxation functions that could otherwise be distinguished. Many of the comparisons we have displayed do have large differences. It is just that they are outside our frequency range of observation.

We have neglected completely the effects of dispersion overlaps and instrumental systematic errors. These will further complicate the process in ways that will be hard to predict. For overlap, the difficulties in separation make the de-

termination of any relaxation function more difficult and can result in parameters that are quite different from those obtained with no overlap present. For systematic errors, such as those due to deviations from an assumed calibration curve near the ends of a frequency range, the deviations need not even follow any expected relaxation function. While a relaxation function always obeys the Kramers–Krönig relations,[24] these latter deviations need not.

We are leaving open the question of how well any function can represent the other. It is clear just from elementary considerations that the KWW, HN, CC, and CD functions are quite different, especially in their limiting behavior. Since it is now possible to directly measure accurate time and frequency data, we have to address this issue in another article.

For all of these reasons, the ultimate appeal must be made to the measured data and how well they can be fitted. If the data are not fit by a given form then the issue disappears. However, it is entirely likely that the data can be fit to more than one in which case the issue should be properly left open.

It should be remembered, however, that whatever form is chosen it must be used in both the time and frequency domains at the same time. The time and frequency responses are connected by transforms that require the transform pair to be uniquely related and using one functional form in one domain does not alter the use of another in the other. Significant deviations in one domain will lead to comparable ones in the other and will be of similar magnitude. A consideration of the relative success of the Hamon approximation[25] will show this to be the case. Furthermore, in this approximation the instantaneous current at time t, is considered to be proportional to $\epsilon''(\omega)/t$. It has been more exactly demonstrated in the development of time–domain techniques to determine the frequency response of dielectrics.[22,26,27] In that work, small deviations in one domain imply corresponding deviations in the other as the integral transform is dominated by behavior at the center of the transformation matrix.

ACKNOWLEDGMENT

[1] K. S. Cole and R. H. Cole, J. Chem. Phys. **9**, 341 (1941).
[2] S. Havriliak, Jr. and S. Negami, J. Polym. Sci., Part C: Polym. Symp. **14**, 99 (1966).
[3] D. W. Davidson and R. H. Cole, J. Chem. Phys. **18**, 1417 (1950).
[4] G. Williams and D. C. Watts, Trans. Faraday Soc. **66**, 80 (1970).
[5] K. L. Ngai and C. T. White, Phys. Rev. B **20**, 2475 (1979).
[6] K. L. Ngai, A. K. Rajagopal, and S. Teiler, J. Chem. Phys. **88**, 5086 (1988).
[7] K. L. Ngai and C. M. Roland, Macromolecules **26**, 6824 (1993).
[8] A. Schönhals and E. Schlosser, Colloid Polym. Sci. **267**, 125 (1989).
[9] R. G. Palmer, D. L. Stein, E. Abrahams, and P. W. Anderson, Phys. Rev. Lett. **53**, 958 (1984).
[10] M. F. Shlesinger and J. Klafter, in *Fractals in Physics*, edited by L. P. Pietronero and E. Tosatti (North-Holland, Amsterdam, 1986), p. 393.
[11] J. F. Douglas and J. B. Hubbard, Macromolecules **24**, 3163 (1991).
[12] F. Alvarez, A. Alegría, and J. Colmenero, Phys. Rev. B **44**, 7306 (1991).
[13] F. Alvarez, A. Alegría, and J. Colmenero, Phys. Rev. B **47**, 125 (1993).
[14] S. Havriliak, Jr. and S. J. Havriliak, Polymer **37**, 4107 (1996).

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[15] C. P. Lindsey and G. D. Patterson, J. Chem. Phys. **73**, 3348 (1980).

[16] B. J. Factor, F. I. Mopsik, and C. C. Han, Macromolecules **29**, 2318 (1996).

[17] R. Zorn, F. I. Mopsik, G. B. McKenna, L. Willner, and D. Richter, J. Chem. Phys. **107**, 3645 (1997).

[18] W. H. Press, S. A. Teukolsky, W. T. Vetterling, and B. P. Flannery, *Numerical Recipes in FORTRAN* (Cambridge University Press, New York, 1992).

[19] R. Courant and D. Hilbert, *Methods of Mathematical Physics* (Interscience, New York, 1953), Vol. I.

[20] A. M. Mood and F. A. Graybill, *Introduction to the Theory of Statistics* (McGraw-Hill, New York, 1963), p. 341.

[21] E. K. Blum, *Numerical Analysis and Computation: Theory and Practice* (Addison-Wesley, London, 1972).

[22] F. I. Mopsik, Rev. Sci. Instrum. **55**, 79 (1984).

[23] A. K. Jonscher, Nature (London) **267**, 673 (1977).

[24] R. De L. Kronig, J. Opt. Soc. Am. **12**, 547 (1926).

[25] B. V. Hamon, Proc. Inst. Electr. Eng. **99**, 27 (1952).

[26] F. I. Mopsik, IEEE Trans. Dielectr. Electr. Insul. **1**, 3 (1994).

[27] P. J. Hyde, Proc. IEEE **117**, 1891 (1970).