Relaxation Phenomena and Stability of Probability Densities

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**Abstract:**
A characteristic function whose positive time behavior is proportional to a step response function is constructed in such a way that: all its derivatives at t=0 are finite; it has the usual exponential decay behavior for intermediate times; and it satisfies the Paley-Wienner bound for long times. The constructed characteristic function \( C'(t_0) \) is piecewise continuous with behavior determined by different exponentials of a monomial function of \( t \), namely \( (4/t_k) \), termed monomial exponentials, on appropriate segments of time. Continuity conditions at joining points provide relations among the \( t_k \), so only one \( t_k \) is an independent parameter. The occurrence of \( t_k \) well within a particular segment in positive time determines the monomial exponential that dominates the behavior of \( C'(t_0) \), and the behavior is then called \( k \)-dominant. The \( k \)-dominance property is discussed for the probability density \( P_k(\omega) \) corresponding to \( C_k(t_0) \). A formalism is developed in which the probability density for a summand variable in a sum maintains \( k \)-dominant behavior for its corresponding characteristic function. The property of \( k \)-dominant stability for probability densities is thereby introduced. At this point the identification of the positive \( t \) portion of \( C'(t_0) \) is completely determined.
ABSTRACT (Continued)

\[ C_{\alpha}(t) \] as a step response function is used to make a comparison with a model of relaxation in complex systems which others have called the Ngai model. The latter involves the introduction of interactions that lead to a modification of a constant decay rate for a linear exponential to a time-dependent one appropriate for fractional exponential behavior. Ngai's predicted relation between the two respective relaxation time parameters corresponds here to a continuity condition for \( C_{\alpha}(t) \). \( k \)-dominance and, by implication, \( k \)-dominant stability are compatible with the Ngai model. The changes in the behavior of \( C_{\alpha}(t) \) and its decay rate may be viewed as temporal analogs of phase transformations caused by the changes in the nature of the interactions in the system. Also compatible with the Ngai model and the concept of \( k \)-dominant stability is a cross-over from one-dominant to \( \alpha \)-dominant behavior under change of experimental conditions that is actually observed for selected systems.
RELAXATION PHENOMENA AND STABILITY OF PROBABILITY DENSITIES

I. Introduction

We are interested in the overall time dependence of the relaxation of a perturbed entity to equilibrium. The overall time dependence of the perturbed part of an entity is frequently tracked in terms of the step function response function \( \psi(t) \). The function \( \psi(t) \) has maximal value at \( t=0 \), is continuous, and vanishes as \( t \to \infty \). The function \( \phi(t) \) may be identified as the positive time part of an even function \( \psi(t) \) which may be related to the dissipative part of a scalar impulse response function \( \chi''(t) \) by

\[
\chi''(t) = -\frac{d\psi(t)}{dt}, \quad -\infty < t < \infty
\]  

(1.1)

In the following we only consider situations where

\[
\lim_{t \to \infty} \chi''(t) = 0
\]

(1.2)

so that the Fourier transform of \( \chi''(t) \), namely the dissipative part of the susceptibility \( \tilde{\chi}''(\omega) \), has only a continuous spectrum with no singularity at \( \omega=0 \). Further, since the Fourier transform of \( \chi''(t) \) or the dissipative part of the susceptibility has all moments, all the derivatives of \( \chi''(t) \) and hence also \( \phi(t) \) at \( t=0 \) must be finite.

The dissipative impulse response function \( \chi''(t) \) may be related by perturbation theoretical developments such as those of Kubo,\(^2\) Case,\(^3\) and Martin\(^4\) to the equilibrium average of a two-time scalar commutator of a configurational coordinate operator \( A(t) \),

\[
\chi''(t-t') = \frac{1}{1n} \text{Tr} \rho_0 [A(t), A(t')] \]

(1.3)

Here \( \rho_0 \) is the canonical equilibrium density matrix and we have taken \( A(t) \) to be the perturbed part only. Then it is clear that \( \chi''(t) \) and also \( \phi(t) \) are functions only of a time difference \( t \).

The equation for $\phi(t)$ corresponding to $\chi''(t)$ in Eq. (1.3) is\(^2,3\)

$$
\phi(t) = 2 \sum_{m \neq m'} \frac{W_m - W_{m'}}{|<m|A|m'>|^2} \cos (E_m - E_{m'}) t / \hbar
$$

(1.4)

Here $E_m$ is an eigenvalue of the unperturbed Hamiltonian, $W_m = \exp(-\beta E_m) / Z$, $Z = \sum \exp(-\beta E_m)$ and $\beta = 1/kT$. It is clear that $\phi(t)$ is even in $t$. In the high temperature limit, equivalent to a classical limit, $\phi(t)$ is just $\beta/2$ times the canonical average of the anticommutator $\{A(t), A(0)\}$ which may be identified with a real autocorrelation function. In any event $\phi(t)$ shares with the autocorrelation function of a weakly stationary process the general properties that it is a two-time function dependent only on time differences, i.e. location invariant, and continuous for all time. We have also in addition that $\phi(t)$ has other temporal behavior determined by the physical properties of $\phi(t)$ mentioned above, viz. it is maximal and differentiable to all orders at $t=0$, and monotonically vanishing as $t \to \infty$. Furthermore from the causality of $\phi(t)$, we infer $\sim''(\omega)$ is one of a Hilbert transform pair.\(^2\) Consistency with this latter property of $\sim''(\omega)$ leads us to the requirement that $\phi(t)$ is square integrable for $-\infty < t < \infty$, i.e. $L^2(-\infty, \infty)$. Also from Eq. (1.4) we may verify that since $\phi(t)$ is continuous at $t=0$, it is also continuous at neighboring points, $\epsilon$, and hence for all $t$. Moreover if $X(t)$ is an arbitrary function whose Fourier transform exists, then from Eq. (1.4) it follows that

$$
\int X(t) \phi(t-t') X^*(t') dt' = \sum_{m \neq m'} \frac{(W_m - W_{m'}) |<m|A|m'>|^2}{E_m - E_{m'}} \\
- \frac{i}{\hbar} (E_m - E_{m'}) t \\
\int |X(t)|^2 dt' \geq 0
$$

(1.5)

Bochner's theorem\(^5\) may then be used to prove that $\phi(t)$ is proportional to a characteristic function.
We find it convenient to deal with the normalized function

$$C(t|0) \equiv \frac{\phi(t)}{\phi(0)}$$

where we have made the two time nature of $C(t|0)$ explicit. By its definition $C(0|0) = 1$ so that it is in fact a characteristic function and we shall refer to it as such in the following.

A major purpose of this paper is to construct a class of characteristic functions with behavior that is consistent with the mathematical and phenomenological constraints on the step-response function $\phi(t)$. These requirements are satisfied by allowing a continuous $C(t|0)$ to be only differentiable almost everywhere. This leads to a piecewise character of $C(t|0)$ which will be discussed in detail in the next section. The resulting construction is a $C(t|0)$ which has the form of appropriate exponentials of monomial functions of $t$, namely $-(|t|/\tau_k)^k$, termed monomial exponentials, on appropriate non-overlapping segments of time. For example, for $k=2$ we have a segment of quadratic exponential behavior, etc. Continuity conditions at the joining points of the segments provide relations among the $\tau_k$. It is pointed out that a $C(t|0)$ so defined is in fact the Fourier transform of a probability density.

With a probability density in hand, we turn to another major purpose of the paper in section III and consider the description of a total system built up from subsystems that share the same generic temporal behavior. We find the conditions under which the probability density for the total system also has the same piecewise characteristic function as that of the subsystems. A new type of stability for probability densities is thereby obtained.

In section IV, we compare the general properties of the piecewise $C(t|0)$ for $t>0$ with the step-response function in a model of relaxation in complex systems previously introduced and discussed by us. The latter in fact shares two important features with the $C(t|0)$ constructed in section III. Namely it
has: (1) a long time fractional exponential regime preceded by a linear exponential regime; and (2) a relationship between the relaxation times or \( r \)-parameters in the two regimes. The model is reviewed briefly as a way to gain insight into the meaning of the piecewise \( C(t|0) \) and the new type of stability for probability densities discussed in section IV.

II. Temporal Behavior of the Characteristic Function

We want now to consider the consequences of the various phenomenological and mathematical constraints on the temporal behavior of the characteristic function \( C(t|0) \) introduced in section I. The properties already indicated in that section are the general ones of location invariance and continuity, and the more special ones of evenness in \( t \), differentiability to all orders at \( t=0 \) (from the existence of all moments of the dissipative part of the susceptibility), maximum value at \( t=0 \), and \( L^2 \left( -\infty, \infty \right) \).

It should be noted that the physics of all our considerations is nonrelativistic so the details of behavior at or near \( t=0 \) is not actually included in our formulation. The best we can hope for near \( t=0 \) is a self-consistent phenomenology. However, this self-consistency leads to some interesting constraints. From the differentiability property of \( C(t|0) \) at \( t=0 \), linear (and all other odd) terms in its power series expansion around \( t=0 \) must vanish. In particular, \( C(t|0) \) cannot then be taken to have a linear exponential form at \( t=0 \). An even more stringent condition arises from the fact that all the derivatives of \( C(t|0) \) at \( t=0 \) must be finite. Further as already noted, from the physical meaning of step response, \( C(t|0) \) is also maximal at \( t=0 \). A self-consistent (but not unique) choice for \( C(t|0) \) at or near \( t=0 \) is a quadratic exponential or a Gaussian form centered on \( t=0 \).
For values of \( t > 0 \), we expect from empirical evidence covering most particular cases that \( C(t|0) \) may appear to decay in one of three general forms: (a) quadratic exponential (Gaussian); (b) linear exponential (most usually calculated, and most often observed in simple systems); (c) fractional exponential (for complex systems). Further for very long times, the real \( L^2(-\infty, \infty) \) characteristic function considered here is constrained to be consistent with the Paley-Wiener bound:

\[
\lim_{|t| \to \infty} C(t|0) > \exp(-|t|/\tau_\alpha)^\alpha, \quad 0 < \alpha < 1
\]

(2.1)

where \( \tau_\alpha \) is a positive real scale parameter. It may be noted that the Paley-Wiener bound is a fractional exponential so the latter is the fastest allowable decay as \(|t| \to \infty\).

Thus for any \( C(t|0) \) there are conditions near or at \( t=0 \) and for very long times that must be satisfied. At intermediate times, there may be physically determined behavior that differs from at least one and possibly both of the behaviors respectively around \( t=0 \) and for very long times. We now consider a generic \( C(t|0) \) that incorporates all these features and, dependent upon the choice of parameterization, can be used to describe any of the observed monomial exponential time behaviors.

We consider \( C(t|0) \) to be piecewise continuous with appropriate monomial exponential behavior in given nonoverlapping segments which cover \(-\infty < t < \infty\) completely. The conditions around \( t=0 \) and for a physical decay regime that has a quadratic monomial behavior can be satisfied by taking \( C(t|0) \) to have the form \( \exp(-t/\tau_2)^2 \) in an initial segment pair joined at \( t=0 \), i.e. for \( 0 < |t| < T_0 \). Here \( \tau_2 \) is a positive real scale parameter and \( T_0 \) is also positive real. The possibility of a linear exponential regime that often occurs empirically and is the form obtained in standard decay calculations can be allowed by choosing \( C(t|0) \) to have the form \( \exp(-|t|/\tau_1) \) in a second segment pair \( T_0 < |t| < T_L \). Here
gain $\tau_1$ is a positive real scale parameter and $T_L$ is positive real. To allow for complex physical systems that have a fractional exponential decay and for long time behavior at the Paley-Wiener bound, $C(t|0)$ is taken to have the form $\exp(-|t|/\tau_\alpha)$, $0<\alpha<1$ for $T_L<|t|<\infty$. Again $T_L$ is positive real. The continuity of $C(t|0)$ is assured by continuity conditions at its joining points $\pm T_0$, $\pm T_L$.

The time behavior of our generic $C(t|0)$ may be summarized as follows.

\[
(t|0, T_0, T_L; \tau_k) = \begin{cases} 
\exp(-t/\tau_2)^2, & -T_0 < t < T_0 \\
\exp(-T_0/\tau_2)^2 = \exp(-T_0/\tau_1), & |t| = T_0 \\
\exp(-|t|/\tau_1), & T_0 < |t| < T_L \\
\exp(-T_L/\tau_1) = \exp(-T_L/\tau_\alpha), & 0 < \alpha < 1, |t| = T_L \\
\exp(-|t|/\tau_\alpha), & 0 < \alpha < 1, T_L < |t| < \infty 
\end{cases}
\] 

(2.2)

The symbol $\tau_k$ in $C(t|0, T_0, T_L; \tau_k)$ represents any of the $k=\alpha, 1, 2$ since only one of the $\tau_k$ is an independent parameter. From the continuity conditions, it follows that

\[
\tau_2^2 = T_0 \tau_1 
\] 

(2.3a)

\[
\tau_\alpha = T_L \tau_\alpha^{-1} \tau_1 
\] 

(2.3b)

The choice of $\tau_k$ as a label will be made on physical grounds as described in more detail below. For the moment we write

\[
C(t|0, T_0, T_L; \tau_k) = C_{\tau_k}(t|0) 
\] 

(2.4)

and choose $\tau_k$ to be that $\tau_k$ which falls within the positive $t$ segment with $k$ exponential behavior. This can happen for only one of the $k$ values. For example, if $T_0 < \tau_1 < T_L$, we would write $C_{\tau_1}(t|0)$ and note that $\tau_2^2 = T_0 \tau_1 > T_0^2$, $\alpha = \alpha_\alpha^{-1} \tau_1 \tau_\alpha$ so that $\tau_2$ is not in $[0,T_0]$ and $\tau_\alpha$ is not in $[T_L,\infty)$. Similarly $\alpha_\alpha \tau_\alpha \text{L} \tau_\alpha$ or $C_{\tau_2}(t|0)$, $\tau_2$ is in $[0,T_0]$ but $\tau_1$ is not in $[T_0,T_L]$ and $\tau_\alpha$ is not in $[T_L,\infty)$. 

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Finally for \( C_{\tau_k} (t|0) \), \( \tau_2 \) is in \([T_L, \infty)\) but \( \tau_2 \) is not in \([0,T_0]\) and \( \tau_1 \) is not in \([0,T_0]\). We shall see that \( C_{\tau_k} (t|0) \) is, for centrally placed \( \tau_k \) on a logarithmic time scale, dominated by its behavior in the region in which \( \tau_k \) falls. We shall call \( C_{\tau_k} (t|0) \) a \( k \)-dominant characteristic function in such cases. However, before addressing this particular point, we want to note some general properties of \( C_{\tau_k} (t|0) \).

First it is clear that although \( C_{\tau_k} (t|0) \) is continuous, it is only piecewise differentiable with discontinuities in derivative at \( t=\pm T_0, \pm T_L \). Further, location invariance is maintained by the requirement that a translation of the origin, e.g. by \( \delta \), is accompanied by a like translation of time segment endpoints e.g. \(-T_L+T_L+\delta \) and \( T_L+T_L+\delta \), etc. Also we note that \( C_{\tau_k} (t|0) \) is the Fourier transform of a probability density. This follows because the nature of the probability distribution function associated with the characteristic function depends on the manner in which the latter behaves as \(|t| \to \infty\). If it goes to zero as with our \( L^2 (\infty, \infty) \) class of functions, then a probability distribution function is continuous. For completeness it may be noted that if the characteristic function tends to a constant, \( D \), with \( 0 < D < 1 \), the associated probability distribution function is singular and may be of the form of a Cantor set, for example. If it tends to unity, then the probability distribution function is discrete.

We return now to the discussion of the generic character of \( C_{\tau_k} (t|0) \) which is readily exhibited by considering three mutually exclusive cases of parameter magnitude: (a) \( 0 < \tau_2 < T_1 \); (b) \( T_0 < \tau_1 < T_L \); (c) \( T_L < \tau_\alpha < \infty \). Case (a) corresponds to 2-dominant behavior. The major part of the decay of \( C_{\tau_2} (t|0) \) appears as a quadratic exponential, and the linear and fractional exponential parts are submerged in a long time tail of small magnitude. On the basis of its direct effect, we could here omit the linear exponential part. However,
present construction requires the continuity condition Eq. (2.3a) that proves a relation between \( \tau_2 \) and \( \tau_1 \). Thus the linear exponential region would be revealed by comparing functional relations for a measured \( \tau_2 \) with corresponding functional relations for a calculated \( \tau_1 \). Finally, the fractional exponential or slower behavior must be included at long times to meet the condition of the Paley-Wiener bound.

Case (b) corresponds to \( l \)-dominant behavior. The major part of the decay \( C_\tau(t|0) \) appears as a linear exponential. The latter is of course the most commonly observed behavior in simple systems and is the usual result of theoretical calculations based on a random phase approximation or the introduction of a phenomenological friction term. The quadratic exponential segment of \( C_\tau(t|0) \) is submerged in small deviations at times short compared to \( \tau_1 \) but or some equivalent must be included to meet the conditions on \( C_{\tau_1}(t|0) \) \( t=0 \). Also again a fractional exponential or slower time behavior must be included to meet the Paley-Wiener bound at long times even though that region has a small magnitude.

Finally, case (c) corresponds to \( \alpha \)-dominant behavior. The major part of decay of \( C_\tau(t|0) \) appears as a fractional exponential. The quadratic exponential segment has an immeasurable effect at short times but it or its equivalent is essential to meet conditions at \( t=0 \). The linear exponential regime provides only small deviations that would most probably also not be noted measurements. Thus again as in case (a) the linear exponential segment at first sight appear to be unnecessary. However, if there is a linear exponential regime, the measured parameter \( \tau_\alpha \) can be compared to a calculated by means of the continuity condition Eq. (2.3b). This continuity condition then provides an important relation that is subject to verification. In t, a similar relation between relaxation time parameters respectively in the
linear and fractional exponential regimes is a key feature of the model of relaxation in complex systems\textsuperscript{6,7} to be discussed below in section IV.

We can gain further insight into the time behavior of $C_{T_k}(t|0)$ by consideration of the instantaneous decay rate $W_{T_k}(t|0)$ which is defined to be

$$W_{T_k}(t|0) = \frac{-d C_{T_k}(t|0)/dt}{C_{T_k}(t|0)} \quad (t \neq T_0, T_L)$$

$$= -d \ln C_{T_k}(t|0)/dt$$

We designate $W_{T_k}(t|0)$ to be $W_2(t|0)$, $W_1(t|0)$, and $W_\alpha(t|0)$ respectively in the segments where $C_{T_k}(t|0)$ has quadratic, linear and fractional exponential behavior. Since $C_{T_k}(t|0)$ is even, it is sufficient to consider $t>0$. In the quadratic exponential segment, $W_2(t|0)=2t/\tau_2^2$ which approaches a maximum value $W_2(T_Q|0)=2T_Q/\tau_2^2=2/\tau_1$ as $t$ approaches $T_Q$. On the other hand $W_1(t|0)=1/\tau_1$ for all $T_Q<t<T_L$ so there is a discontinuity in $W_{T_k}(t|0)$ at $t=T_Q$ where it falls to one half its limiting maximum value in the quadratic segment. Similarly there is a discontinuity at $t=T_L$ for $W_\alpha(t|0)=\alpha t^{\alpha-1}/\tau_\alpha$ has a maximum limiting value $W_\alpha(T_L|0)=\alpha/\tau_1 < 1/\tau_1$. It is clear that this sequential behavior for $W_{T_k}(t|0)$ is independent of the choice of magnitude $\tau$'s and so occurs for all possibilities of the $k$ in $k$-dominance.

The sharpness of the discontinuities in $W_{T_k}(t|0)$ is an artifact of our piecewise construction for $C_{T_k}(t|0)$. Nevertheless decay rates depend on system interactions so these or perhaps somewhat more smooth behavior can be given a physical meaning. They can be viewed as due to changes in the nature of the interactions in the system at $|t|=T_Q, T_L$. Such changes can be thought of as temporal transformations of the environment in which the relaxation is occurring. These transformations can be viewed as the analogs of thermodynamic phase transformations.
The $C_T(t|0)$ of Eq. (2.2) could be readily generalized by addition of other monomial exponential intermediate segments or appropriate short time or long time segments. Such a generalization would also involve corresponding additions (and discontinuities) in $W_T(t|0)$. The point we wish to emphasize however is that the generic $C_T(t|0)$ defined in Eq. (2.2) satisfies all the mathematical requirements for, and can provide an adequate description of the most usually phenomenological behaviors of real step-response functions. A simple and useful property of $C_T(t|0)$ so constructed is that it has monomial exponential behavior in all segments. This has provided simple continuity conditions, Eqs. (2.3a,b), and further, provides the basis for consideration in the next section of stability properties for the probability density $P_T(\omega)$ belonging to $C_T(t|0)$.

III. $k$-dominant Stability for Probability Densities

We turn now to a discussion of the stability properties of $P_T(\omega)$. The probability density $P_T(\omega)$ corresponding to $C_T(t|0)$ is a function of $\omega$ as a random variable that incorporates properties that reflect the particular $k$-dominant and piecewise properties of $C_T(t|0)$.

\[
C_T(t|0) = \int_{-\infty}^{\infty} P_T(\omega) \cos \omega t \, d\omega
\]

\[
P_T(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} C_T(t|0) \cos \omega t \, dt
\]

We note that $C_T(t|0)$ remains invariant under scaling of time by a numerical factor. From Eqs. (2.2) and (2.4),

\[
C(At|0, AT_Q, AT_L; AT_k) = C_{AT_k}(At|0) = C_T(t|0)
\]

This property merely reflects the fact that the choice of scale in temporal measurements does not affect the behavior of $C_T(t|0)$. On the other hand from Eq. (3.1a), the probability density must then obey the following scaling relation.
\[ P_{\text{Tr}}(\omega) = A P_{\text{Tr}}(\omega A) \quad (3.3) \]

Consider now \( N \) subsystems each of which is characterized by a member of the set of independent and identically distributed stochastic variables \( \omega_i \). We want then a total system made up of these subsystems and characterized by a normalized summand variable

\[ \omega = B_{N_k} \sum_{i=1}^{N_k} \omega_i \quad (3.4) \]

such that the probability density for \( \omega \) is also the Fourier transform of the same \( k \)-dominant characteristic function as that for the individual systems. The normalization function \( B_{N_k} \) is expected to depend on both \( N \) and \( k \). Such a probability density will be called \( k \)-dominant stable. We write

\[ P_{\text{Tr}}(\omega) = \int_0^\infty \cdots \int_0^\infty \delta(B_{N_k} \sum_{i=1}^{N_k} \omega_i - \omega) \prod_{i=1}^{N_k} P_{\text{Tr}}(\omega_i) d\omega_i, \quad (3.5) \]

which from Eq. (3.3) can also be written in the form

\[ P_{\text{Tr}}(\omega) = B_{N_k} \int_0^\infty \cdots \int_0^\infty \delta(\sum_{i=1}^{N_k} \omega_i - \omega) \prod_{i=1}^{N_k} P_{\text{Tr}}(B_{N_k} \omega_i) d\omega_i \quad (3.6) \]

Here

\[ \omega_i = B_{N_k} \omega_i \quad (3.7) \]

Transforming the \( \delta \)-function to \( t \)-space and carrying out the \( N \omega_i \) integrals, we obtain

\[ P_{\text{Tr}}(\omega) = \frac{1}{2\pi} \int_0^\infty \left[ C_{B_{N_k}^{N_k}}(t|0) \right]^N \cos \omega t \, dt \quad (3.8) \]
The time arguments in the products of \( C_{B_{Nk}}^{T_{k}}(t|0) \) in the integrand are the same so the resulting product of \( N \ C_{B_{Nk}}^{T_{k}}(t|0) \)'s has the piecewise structure of \( C_{B_{Nk}}^{T_{k}}(t|0) \). However, Eq. (3.8) is not a proper equation unless

\[
[C_{B_{Nk}}^{T_{k}}(t|0)]^N = C_{T_{k}}^{T_{k}}(t|0) \tag{3.9}
\]

Using the monomial exponential property of all the segments of \( C_{B_{Nk}}^{T_{k}}(t|0) \) and continuity conditions Eqs. (2.3a,b) as appropriate, we find Eq. (3.9) follows if

\[
B_{Nk} = N^{1/k} \tag{3.10}
\]

We have the result then that \( k \)-dominant stability is achieved for a probability density of a summand variable that is normalized by \( N^{1/k} \). For \( k=2 \), \( N^{1/2} \) provides an enhancement in \( \omega \) with respect to the \( \omega_i \). For \( k=1 \), \( \omega \) and the individual \( \omega_i \) coincide while for \( k=\alpha \), \( 0<\alpha<1 \), \( \omega \) is reduced with respect to an individual \( \omega_i \).

We can view the normalization as a manifestation of an interaction environment that is introduced when the independent subsystems are put together to form the total system. However, once the total system is formed, it can be viewed as a building block subsystem for a larger total system. It follows that the interaction environment for the single system must be self-consistent with the interaction environment for the total system. An important question is how the interaction environment is first set up on a microscopic basis or alternatively, if \( k \)-dominant stability is taken as a primary principle, how a microscopic interaction environment is arranged to form the appropriate building block. In the next section we consider a model of relaxation in complex systems that can be viewed as providing a microscopic basis for \( \alpha \)-dominant behavior. If \( k \)-dominant stability is taken as the primary principle, all models of relaxation must be constructed in such a way to yield \( k \)-dominant behavior.
In general, this complementarity of microscopic basis and k-dominant stability may be viewed in the following physical way. Theoretical descriptions of relaxations in a complex physical systems such as glasses and polymers are usually based on a microscopic relaxation species. The latter can e.g. be a dipolar group, a hopping charge, a spin, or an entire polymer chain or segment of it. A particular relaxation species i in a complex environment and interacting with a nominal heat bath, will have behavior described in terms of a relaxation function \( \phi_i(t) \), or a characteristic function \( C_{rk}^i(t|0) \) or a probability density \( P_{rk}^i(\omega) \). Measurements in relaxation are macroscopic quantities such as transient current, stress, magnetization, volume and shear flow. The stochastic variable \( \omega \) of the macroscopic quantity is a normalized sum of the form of Eq. (3.4). It is generally assumed in all considerations of relaxation that the macroscopic \( \phi(t) \) is identical to the microscopic \( \phi_i(t) \). This assumption is equivalent to the assumption that \( P_{rk}^i(\omega) \) is a stable probability density or in our description, k-dominant stable. From the results of this section, these equivalent assumptions are consistent with the requirement that \( \phi_i(t) \) be the positive time part of a k-dominant \( C_{rk}^i(t|0) \) of the form given in Eq. (2.2).

As a final comment in this section, it is interesting to consider the relationship between Lévy stability\(^{14,11} \) and the k-dominant stability introduced here. While we have restricted ourselves to real characteristic functions that must be piecewise with multiple monomial exponential behaviors, Lévy deals with characteristic functions with magnitude equal to a single monomial exponential with monomial power \( g, 0 < g < 2 \). Except for the possibility of the quadratic exponential behavior, such monolithic behavior cannot meet the physical conditions on the autocorrelation function at \( t=0 \). For a real characteristic function, the monolithic quadratic exponential is ruled out by the
physical unrealistic requirement that its instantaneous decay rate is proportional to \( t \) and therefore blows up as \( t \to \infty \) and such behavior is never observed. For any characteristic function, the limitation to a quadratic exponential magnitude at intermediate times is not sufficiently general to provide a satisfactory basis for the description of decay phenomena. Thus from the viewpoint of physics, the present generalization to \( k \)-dominant stability and its concomitant piecewise characteristic function, or some further generalization yet to be made, is a necessary step beyond Lévy stability.

IV. Comparison with the Ngai Model of Relaxation

We now take advantage of the fact that \( C_{(t|0)} \) for \( t>0 \) can be identified as a step response function to compare our constructed \( C_{(t|0)} \) with the relaxation function obtained in the so-called Ngai model of relaxation. The latter model predicts: (1) a fractional exponential decay in a time regime \( t\gg \omega_c^{-1} \) when \( \omega_c \tau_0 \gg 1 \); (2) a relationship between a relaxation time \( \tau_1 \) in a linear exponential regime \( t<\omega_c^{-1} \) and \( \tau_0 \). Here \( \omega_c \) is a cutoff frequency that is a parameter of the model as described below and we have taken the liberty of using \( \tau_k \) for \( k=a,1 \) to suggest the direct connection with our \( C_{(t|0)} \). It is clear that the model predictions are consistent with \( k \)-dominant behavior for \( C_{(t|0)} \) and, as we shall see in detail below, the continuity condition Eq. (2.3b). Thus the model may indeed provide insight into the physics of the formation of \( C_{(t|0)} \) and the question of the microscopic origin of the building blocks of \( k \)-dominant stability.

To provide context for such a discussion, we briefly review the concepts in the Ngai model. The model considers a relaxing entity that is coupled to a heat bath on a microscopic level. In accordance with the conventional description of relaxation, after some induction time, say \( t>T_Q \), the system initially
relaxes according to a linear exponential. The complexity of the system is modelled in terms of correlations imposed on the simple system. These correlations are represented by modes which are distinct from the heat bath but which also interact with the relaxing entity. These modes were called correlated state excitations in Ref. 6. The correlated state excitations themselves modell in terms of the energy level spacings of the Gaussian Orthogonal Ensemble (GOE) of random matrix Hamiltonian theory.\textsuperscript{12} The level spacing distribution of the GOE gives the energy density function \( N(E) \) of the correlated state excitations which are also called level spacing excitations.\textsuperscript{7} \( N(E) \) varies linearly with \( E \) up to an upper cutoff \( E_C = \omega_C \) that defines the low frequency regime, levels off to a plateau at intermediate energies, and vanishes (linear) exponentially at high energies. As noted above, long and short time regimes for the model are quantified in terms of \( \omega_C \), i.e. \( \omega_Ct \gg 1 \) means \( t \) in the long time regime while \( \omega_Ct \ll 1 \) means \( t \) is in the short time regime (for us, intermediate time regime).

The relaxing entity is then not only coupled to the heat bath but also to the energy level spacing or correlated state excitations. The latter coupling strength is assumed to be independent of the energy of the excitations. It is important to emphasize that the level spacing excitations are not part of the heat bath. The excitations are in equilibrium prior to system excitation, are driven out of equilibrium during the relaxation by their interaction with the measured relaxing entity, and eventually return to equilibrium in a process not addressed specifically in the model.

The interaction between the relaxing entity and the level spacing excitations leads to a long time fractional exponential decay as a manifestation of the modification of the instantaneous decay rate from \( W_1(t) \sim t^{-1} \) for \( \omega_Ct \ll 1 \)

\[
W_a(t) = W_1(t) \exp\left[(\alpha-1)\gamma\right] (\omega_Ct)^{\alpha-1} , \quad 0<\alpha<1 , \quad \omega_Ct>\gamma .
\]  

(4.1)
Here $\gamma$ is the Euler constant. Instead of $\alpha$, the model is usually formulated in terms of the parameter

$$n = 1 - \alpha$$

(4.2)

where $n = 2C|V|^2$.  

(4.3)

Here $C$ is the slope of the linear portion of the density of states $N(E)$ and $|V|^2$ is the averaged squared magnitude of the coupling interaction between the relaxing entity and the level spacing excitations. As mentioned above, $|V|$ is assumed to be independent of the energy of the excitation.

The long time step response function is written as

$$\phi(t) = \exp[-(t/\tau_\alpha)^{1-n}], \omega_c t >> 1$$

(4.4)

It should also be noted that Eq. (4.4) applies for experimental observations where $\omega_c \tau_\alpha >> 1$ so $\phi(t)$ meets the condition (c) of $\alpha$-dominance. Given Eq. (4.4), the instantaneous decay rate for $\omega_c t >> 1$ is

$$W_\alpha(t) = \alpha \tau_\alpha^{-1}/\tau_\alpha$$

(4.5)

Consistency of Eqs. (4.1) and (4.5) requires that

$$\tau_\alpha = \frac{\alpha \exp[(1-\alpha)\gamma \omega_c^{1-\alpha} \tau_1]}{\alpha}$$

(4.6)

Equations (4.4) and (4.6) are the predictions of the Ngai model which are readily susceptible to experimental verification. For example, let us consider the effect of elevated temperature on relaxations in complex systems such as amorphous polymers and glasses which at normal temperatures have the $\alpha$-dominant form of Eq. (4.4) and $\tau_\alpha$ is thermally activated. The Ngai model has an additional prediction given by Eq. (4.6) which relates $\tau_\alpha$ to $\tau_1$. If $\tau_\alpha$ has an Arrhenius temperature dependence with activation energy $E^*_\alpha$, then $\tau_1$ will have a different but predictable activation energy of $E^*_a = \alpha E^*_\alpha$. More generally, a dependence $f(\zeta)$ of $\tau_\alpha$ on a physical parameter $\zeta$, such as molecular weight $M$ of
a polymer chain in polymer melt relaxation or isotope mass $m$ of alkali ion in conductivity relaxation of glasses, will correspond to a different $[f(\zeta)]^{\alpha}$ dependence on $\zeta$ for $\tau_1$. These predictions have been verified in many different complex systems.¹⁴

An interesting possibility that is allowed in the Ngai model is the cross-over from $\alpha$-dominant to $\beta$-dominant behavior by changing the conditions of physical measurement. To discuss this point further, we complete the contact between the Ngai model and the construction of $C_\tau(t|0)$ by identifying $T_L$ in Eq. (2.3b) to be

$$T_L = \alpha^1/(\alpha-1)\exp(-\gamma)\omega_c^{-1}$$  \hspace{1cm} (4.7)

Then we can consider a change in physical conditions by which $T_L$ does not vary significantly but where $\tau_\alpha$ is changed by many orders of magnitude so that a crossover occurs. In such a case, the new $\tau_1$ can obey

$$\tau_1 \ll T_L$$  \hspace{1cm} (4.8)

and the behavior is $\beta$-dominant because it is now $\tau_1$ that is in the interval $(T_Q, T_L)$. Perhaps surprisingly, such a situation occurs in a straightforward way in the temperature dependence of the relaxation of several materials including the glass-forming fused salt¹⁴ $0.4\text{Ca(NO}_3)_2\cdot(0.6)\text{KNO}_3$, the ionic conductor sodium $\beta$-alumina,¹⁵ and in polymeric systems.¹⁶

It may be noted that most presently proposed models¹⁷,¹⁸ of relaxation encompass neither the prediction of Eq. (4.6) nor the possibility of crossover of type of dominant behavior. Although the Ngai model does not require the sudden discontinuity by changing the decay rate from $W_1(\tau_1^{-1})$ to $W_\alpha(t)$ at the single point $t=T_L$, it is compatible with such an interpretation. Then instead of the more abstract discussion of the temporal analog of thermodynamic phase transitions in section II, we may now consider $W_1(t)$ to be suddenly dressed by
means of interaction with correlation or level spacing excitations at $T_L$ to
yield the decay rate $\mathcal{W}_q(t)$.

Correlation excitations may also be viewed as providing the vehicle by
which $\alpha$-dominant stability is introduced microscopically. Thus the Ngai model
may be viewed as providing the first building block. On the other hand, as
indicated in section III, we may view $\alpha$-dominant stability as a primary prin-
ciple. Then the microscopic interactions that enter the first building block
must be compatible with $\alpha$-dominant (or more generally any $k$-dominant) stabil-
ity.
References


9. This statement is correct if the observed behavior is somewhere monomial exponential. A possibility of a monolithic behavior is $C(t|0) = \frac{1}{1+(t/\tau)^2}$ but such behavior does not seem to occur in physical systems.

10. E. Lukacs, ibid., ch. 2.


The effect of level spacing excitations for frequencies above $\omega_c$ are neglected in arriving at Eq. (4.1). The formulation of the Ngai model, given in Ref. 7 includes the effect of high frequencies. The result is to introduce a constant multiplicative factor to the RHS of Eq. (4.1) that has played no role in comparison with experiment.


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