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Memory functions for mechanical relaxation in viscoelastic materials

L. A. DISSADO, R. M. HILL
The Dielectrics Group, Department of Physics, King's College London (KQC), The Strand, London WC2R 2LS, UK

The roles of memory functions for compliance retardation and modulus relaxation in viscoelastic materials are examined. It is shown that essential features of the mechanical responses are the components which occur instantaneously on the application of either a stress or a strain. Taking these features into consideration it is shown that at non-zero time the cooperative memory function of compliance retardation is the time differential of the modulus relaxation function and the cooperative memory function of modulus relaxation is the time differential of the compliance relaxation function for step up functions of stress and strain, respectively. Zero time singularities in the memory functions have been eliminated in the derivation of the reduced dynamical equations, whose memory functions are limited to non-singular contributions which are always present.

1. Introduction

It has been shown [1] that when relaxation in a viscoelastic material is restricted to two elementary first-order processes the slower of these experiences a memory of the faster when the processes are allowed to interact in a sequential manner. In this case the time scale of the memory of the system extends into that of the slow response and modifies its relaxation rate without altering its Debye form. It has also been shown [2–4] that non-Debye spectral responses are obtained when relaxation is governed by a continuum or hierarchy of sequential, cooperative processes rather than just two. Here we investigate the nature of memory in mechanical relaxation and show that for any practical viscoelastic system the memory function of the compliance response is related to the modulus relaxation function, and the memory function of the response of the modulus is related to the compliance (creep) retardation function, regardless of whether the response is of Debye or non-Debye form. Hence memory must exist on the time scales of the relaxation/retardation of both compliance and modulus.

The memory function for a single process of relaxation is correctly defined through the general Volterra equation of the second kind

$$\frac{\mathrm{d}\alpha(t)}{\mathrm{d}t} = -\int_{-\infty}^{t} K_J(t-\tau)\,\alpha(\tau)\,\mathrm{d}\tau + \theta_{\mathrm{th}}(t) + \sigma(t)\,C \tag{1}$$

where we have considered relaxation in the strain, $\alpha(t)$, under the application of a time dependent stress, $\sigma(t)$. $K_J(t)$ is the compliance memory function, $\theta_{\mathrm{th}}(t)$ the coupling to the phonon bath and C a strain/stress coupling constant that requires to be determined from the specific experimental situation that is under consideration. The presence of the thermal coupling term in the equation of motion, Equation 1, with the average properties

$$\langle\theta_{\mathrm{th}}(t)\rangle = 0 \tag{2a}$$

and

$$\langle\theta_{\mathrm{th}}(t)\,\theta_{\mathrm{th}}(t_1)\rangle \propto \delta(t - t_1) \tag{2b}$$

is a formal necessity for the correct description of thermal equilibrium [5]. Thus Equation 2b ensures that fluctuations about the average value of α (i.e. $\langle\alpha(t)\rangle$) exist, and use of Equation 2a shows that their average relaxation is governed by an equation of the form of Equation 1 but with $\theta_{\mathrm{th}}(t)$ taken as zero.

The average strain response to the driving force $\sigma(t)$ can now be determined. For example, if we take $\sigma(t)$ to be a delta function of magnitude σ_0 at time zero, a one-sided Fourier transformation of the average strain gives the response $\alpha(\omega)$ to an alternating stress, $\sigma_0\,\mathrm{e}^{i\omega t}$ [6]. This can be obtained from the averaged form of Equation 1 as

$$\frac{\alpha(\omega)}{\sigma_0} = J(\omega) = \frac{C}{K_J(\omega) + i\omega} = J_0 F(\omega) \tag{3}$$

and hence

$$K_J(\omega) = C\,J_0^{-1}[F(\omega)]^{-1} - i\omega \tag{4}$$

In Equation 3, $J(\omega)$, which by definition is the compliance [7], has been expressed as a magnitude, the equilibrium compliance J_0, multiplied by a normalized complex spectral shape function $F(\omega)$ which goes to zero as the frequency becomes infinite and to unity as the frequency approaches zero. This convenient notation, which follows that in [6], will be used throughout. $K_J(\omega)$ is the one-sided Fourier transformation of $K_J(t)$, that is the spectral response of the compliance memory function. In principle, $K_J(t)$ can be obtained from $K_J(\omega)$ by inverse Fourier transformation of Equation 4, where the inverse transformation exists.

On the other hand when $\sigma(t)$ has the form of a step

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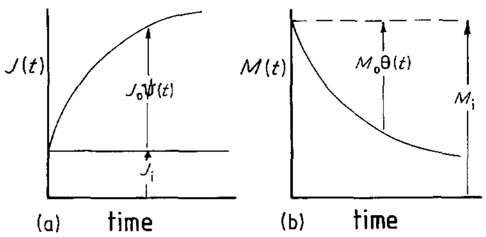

Figure 1 (a) Diagrammatic representation of the compliance retardation as a function of time. J_i is the "instantaneous" response, $\psi(t)$ the unity normalized retardation function with J_0 being the magnitude of the time dependent response. The range of $\psi(t)$ is from zero to unity as the time develops from zero to infinity. (b) Diagrammatic representation of the modulus relaxation as a function of time. M_i is the "instantaneous" response, M_0 the magnitude of the time dependent response and $\theta(t)$ the relaxation function with $\theta(t)$ being zero at time zero and unity at infinite time. Note that the relaxation is a decrease from a high "instantaneous" value to the equilibrium value $M_i - M_0$.

function at zero time, i.e. $\sigma(t) = \sigma_0 \lambda(t)$ with $\mathrm{d}\lambda/\mathrm{d}t = 1 \cdot \delta(t)$, the unit magnitude delta function, Fourier transformation of the macroscopically averaged Equation 1 gives

$$\alpha(\omega) = \frac{\sigma_0 C}{i\omega[K_J(\omega) + i\omega]} \tag{5}$$

Here $\alpha(\omega)$ is the Fourier transformation of the strain response $\alpha(t)$ to the step function stress, and substitution for the compliance $J(\omega)$ from Equation 3 shows that $\alpha(\omega) = (i\omega)^{-1} J(\omega)\, \sigma_0$. Thus, in this case

$$\begin{aligned} J(\omega) &= \sigma_0^{-1}\, \mathrm{FT}\left[\frac{\mathrm{d}\alpha(t)}{\mathrm{d}t}\right] \\ &= J_0\, \mathrm{FT}\, [\dot{\psi}(t)] \end{aligned} \tag{6}$$

where $\alpha(t) \propto \alpha_0 \psi(t)$ is the average strain response to a step function stress, which is defined through the creep retardation function $\psi(t)$ [8] as indicated in Fig. 1a, and $J_0 = \alpha_0/\sigma_0$. The dot notation signifies differentiation with respect to time.

The cases considered above show that the compliance may, correctly, be taken as the one-sided Fourier Transformation (FT) of either the average strain in response to a delta function excitation, or the rate of relaxation of strain under unit step function stress excitation. An alternative approach to the memory function analysis (for example see [8 to 10]), is to consider the right-hand side of Equation 1 to be given solely by an integral of a memory function running from zero to t, i.e.

$$\frac{\mathrm{d}}{\mathrm{d}t}[\alpha(t)/\sigma_0] = -\int_0^t K_J'(t-\tau)[\alpha(\tau)/\sigma_0]\,\mathrm{d}\tau \tag{7}$$

The primed symbol is used here to differentiate between this form of memory function and that defined through Equation 1. From a one-sided Fourier transformation of Equation 7 we have

$$K_J'(\omega) = \frac{i\omega F'(\omega)}{1 - F'(\omega)} \tag{8a}$$

where $F'(\omega)$ is the Fourier transform of the response $\alpha(t)/\sigma_0$. It has been suggested [10] that $F'(\omega)$ should be identified with the dynamic compliance function $F(\omega)$ of Equation 3. However, the dropping of the explicit source terms $\theta_{\mathrm{th}}(t)$ and $\sigma(t)$ on going from Equation 1 to Equation 7 has introduced a serious ambiguity in the boundary conditions and it is not clear whether $\alpha(t)$ should be regarded as the response to a delta function, a step function, or to some other form of excitation. As we have already indicated the interpretation of $\mathrm{FT}[\alpha(t)]$ in terms of the dynamic compliance depends on the form of the excitation, cf. Equations 3 and 5, and thus the identification of $F'(\omega)$ with $F(\omega)$ is not a priori justifiable. In fact by noting that the response must be zero in the absence of a source we can see that Equation 8a implies, erroneously, that the memory function is itself the source, i.e.

$$[i\omega + K_J'(\omega)]\, F'(\omega) = K_J'(\omega) \tag{8b}$$

An equivalent Fourier transformation of the homogeneous portion of Equation 1 gives

$$[J(\omega)]^{-1}\, C\, \alpha(\omega) = [K_J(\omega) + i\omega]\, \alpha(\omega) = 0 \tag{9}$$

and shows that it is essential to include the source term if the compliance is to be obtained by Fourier transformation of the equation of motion for $\alpha(t)$.

It is unfortunate that the simpler but erroneous form of the Volterra equation appears to have been generally accepted. In part this is due to a unique result for an exponentially decaying relaxation process. In this case Equation 1 requires that $K_J(t)$ must have the form

$$K_J(t) = \omega_{\mathrm{p}}\, \delta(t) \tag{10a}$$

and

$$K_J(\omega) = \omega_{\mathrm{p}} \tag{10b}$$

with a complete absence of memory on the time scale of the relaxation, $(\omega_{\mathrm{p}})^{-1}$. Because $F(\omega)$ now has the Debye form, $(1 + i\omega/\omega_{\mathrm{p}})^{-1}$ then Equation 4 gives the constant C as

$$C = \omega_{\mathrm{p}} J_0 = \omega_{\mathrm{p}} J(\omega = 0) \tag{11}$$

It can be seen, from Equation 10, that the primitive form of the Debye relaxation memory function allows the memory function defined by Equation 7 to recover the correct Debye behaviour, because now the apparent source term in Equation 8b has the required delta function form. This fortuitous result is, by definition, limited to an exponential time decay and in any other case $K_J'(t)$ will be significantly different from a delta function and hence $F'(\omega)$ may not be identified with $F(\omega)$.

Equation 10a shows that the description of non-Debye behaviour in terms of a parallel summation of discrete or continuous distributions of Debye responses of differing relaxation rates requires that the memory function takes the form of an equivalent summation or integration of zero time delta functions [1]. On the other hand, Equation 4 clearly shows that a non-Debye spectral function for $F(\omega)$ will give a frequency dependent $K_J(\omega)$ and hence a time dependence in the memory function at times other than zero. The latter implies that some memory of the preceeding steps must influence the course of relaxation. It is our intention here to discuss the formal interpretation of the memory functions that can be determined, with complete generality, from the dynamic compliance and modulus expressions.

In principle, it should be possible to develop a description for the relaxation of the mechanical modulus which would be equivalent to that already contained within Equations 1 to 6 for the creep compliance. However, when one attempts to do so a fundamental difficulty arises because the modulus response to a step function excitation is a relaxation from a high instantaneous value to a low equilibrium value, as indicated in Fig. 1b. Essentially the zero time instantaneous response is an intrinsic part of the relaxation which has been neglected in the creep compliance derivation. Using the notation in [6] the time dependence of the modulus can be expressed as

$$M(t) = M_i\lambda(t) - M_0\theta(t) \qquad (12)$$

where $\theta(t)$ is the modulus relaxation function, M_0 the magnitude of the relaxation process, M_i the instantaneous or zero time response of the ratio of the stress to the strain and $\lambda(t)$ is, as before, the unit step function. There can be no modulus relaxation without the instantaneous component. Fourier transformation of Equation 12 gives the frequency dependence of the modulus response as

$$M(\omega) = M(\infty) - M_0 G(\omega) \qquad (13)$$

with $M(\infty)$ the infinite frequency component, which is equivalent to the instantaneous response in the time domain, and $G(\omega)$ a second, unity scaled complex spectral function which also approaches units at low frequencies and zero at high frequencies [6]. As will be discussed later, the compliance and modulus functions of frequency are reciprocals of one another and hence the infinite frequency component of the modulus requires a finite high frequency response in the compliance which, in turn, necessitates the presence of an instantaneous component in the compliance in the time domain, indicated as J_i in Fig. 1a. It is this feature that has been neglected in the conventional approach to the memory response described through Equations 2 to 6. In the following sections the memory functions of both compliance and modulus will be obtained, taking into consideration not only the principal retardation/relaxation but the instantaneous responses as well. In the remainder of the paper the consequences of memory in practical relaxing systems, that is those with both instantaneous and non-instantaneous components, will be considered and discussed.

2. Memory functions

When the application of a strain/stress to a material results in more than one component of response, the Volterra equation, Equation 1, is unable to describe the system and requires to be replaced (after taking macroscopic averages) by the general expression

$$\mathrm{d}[\alpha_j(t)]/\mathrm{d}t = -\int_{-\infty}^{t} \sum_k K_J^{jk}(t-\tau)\alpha_k(\tau)\,\mathrm{d}\tau + \sigma(t)C \qquad (14)$$

where $K_J^{jk}(t)$ is the jkth element of a memory tensor and summation over the jth component of the response is implied. If it is assumed that the different components evolve independently, i.e. in parallel, then the memory tensor becomes diagonal and Equation 14 reduces to

$$\mathrm{d}[\alpha_j(t)]/\mathrm{d}t = -\int_{-\infty}^{t} K_J^{jj}(t-\tau)\alpha_j(\tau)\,\mathrm{d}\tau + \sigma(t)C \qquad (15a)$$

When $K_J^{jj}(t)$ is a delta function the formalism of Equation 15a describes a distribution of independent components which each relax exponentially, i.e. a distribution of Debye relaxation elements.

As outlined in Section 1, however, an intrinsic feature of any mechanical relaxation is the presence of an "instantaneous" response in the stress/strain. This can be considered to arise from the initial dynamical behaviour (e.g. damped lattice vibrations, rapidly relaxing constraints) which eventually converts to the directly observable relaxation. Under these circumstances the "instantaneous" and "observable" components of the relaxation can be regarded as cooperative sequential elements of one complete process. The development of this process can be described by means of a single element "cooperative" memory function

$$\mathrm{d}\left[\sum_j \alpha_j(t)\right]\Big/\mathrm{d}t = -\int_{-\infty}^{t} K_J(t-\tau) \times \sum_j \alpha_j(\tau)\,\mathrm{d}\tau + \sigma(t)C \qquad (15b)$$

with the equivalent expression for the modulus "cooperative" memory function $K_M(t)$ being obtained by replacing $\alpha_j(t)$ with $\sigma_j(t)$ and vice versa. Here the sum in j runs only over the "instantaneous" response, denoted by subscript i, and an "observable" compliance increment of magnitude J_0, such that the strain response to a step-up stress excitation of magnitude, σ_0, is

$$J(t) = \sum_j \alpha_j(t)/\sigma_0 = J_i\lambda(t) + J_0\psi(t) \qquad (16)$$

Because the "instantaneous" response, J_i, takes place on an extremely fast time scale relative to that of the "observable" increment, we have taken it to occur at time zero and represented it through a step-up function $\lambda(t)$. The time scale of the observable increment itself is defined via its unity-scaled time-dependent retardation function, $\psi(t)$.

Formal relationships between the "cooperative" memory functions, $K_J(\omega)$ and $K_M(\omega)$, and the spectral functions $F(\omega)$ and $G(\omega)$ can be obtained by Fourier transforming Equation 15b for a delta function source ($\sigma(t)$ or $\alpha(t)$), because in this case the strain/stress response is the dynamic compliance/modulus. The constant C is obtained through the equilibrium (static) compliance/modulus as

$$C_J = [J_0 + J(\infty)]K_{J,0} \qquad (17a)$$

and

$$C_M = [M(\infty) - M_0]K_{M,0} \qquad (17b)$$

where $K_{J,0}$ and $K_{M,0}$ are the zero frequency limits of $K_J(\omega)$ and $K_M(\omega)$, respectively. The generalization of

Equation 4, defining the spectral response of the compliance memory function, is thus given by

$$K_J(\omega) = K_{J,0}\frac{J_0 + J(\infty)}{J_0F(\omega) + J(\infty)} - i\omega \qquad (18)$$

with the frequency dependence of the compliance being related to the strain response to a step-up excitation through

$$\begin{aligned} J_0F(\omega) + J(\infty) &= \mathrm{FT}[\mathrm{d}J(t)/\mathrm{d}t] \\ &= \mathrm{FT}[J_0\dot{\psi}(t) + J_i\dot{\lambda}(t)] \quad (19) \end{aligned}$$

In an exactly similar manner the memory function for the modulus relaxation can be derived in the form

$$K_M(\omega) = K_{M,0}\frac{M(\infty) - M_0}{M(\infty) - M_0G(\omega)} - i\omega \qquad (20)$$

and because M_0 is required to be less than $M(\infty)$ no difficulties arise from the sign of the frequency-dependent term. We also have that the response of the modulus after step function excitation in strain is

$$\begin{aligned} M(\infty) - M_0G(\omega) &= \mathrm{FT}[\mathrm{d}M(t)/\mathrm{d}t] \\ &= \mathrm{FT}[M_i\dot{\lambda}(t) - M_0\dot{\theta}(t)] \quad (21) \end{aligned}$$

Inverse transformations of Equations 18 and 20 give the required time domain response of the memory functions of compliance retardation and modulus relaxation. The difficulties that can be experienced in carrying out the inverse transformations lie not with the $(i\omega)$ terms, whose transformation is the differential of the unit delta function $[\mathrm{d}\delta(t)/\mathrm{d}t = \dot{\delta}(t)]$, but with the inversion of the reciprocal of the spectral functions and the high-frequency component.

3. Compliance-modulus relationships

The problems associated with the spectral function reciprocals in Equations 18 and 20 can be removed by utilizing the relationship between dynamic modulus and compliance determined through a consideration of the stress and strain under sinusoidal excitation [8, 9, 11] to be

$$[J_0F(\omega) + J(\infty)][M(\infty) - M_0G(\omega)] = 1 \qquad (22a)$$

which contains, as special cases at zero and infinite frequencies

$$[J_0 + J(\infty)[M(\infty) - M_0] = 1 \qquad (22b)$$

and

$$J(\infty)\,M(\infty) = 1 \qquad (22c)$$

By means of these relationships and Equation 18 we get

$$\begin{aligned} K_J(\omega) &= K_{J,0}\frac{M(\infty) - M_0G(\omega)}{M(\infty) - M_0} - i\omega \qquad (23) \\ &= K_{J,0}\gamma^{-1} - i\omega - K_{J,0}\gamma^{-1}(1 - \gamma)\,G(\omega) \qquad (24) \end{aligned}$$

with $\gamma = J(\infty)/[J_0 + J(\infty)]$. After inverse transformation we then have

$$K_J(t) = K_{J,0}\gamma^{-1}\delta(t) - 1.\dot{\delta}(t) - K_{J,0}\gamma^{-1}(1 - \gamma)\dot{\theta}(t) \qquad (25)$$

as the memory function of compliance (creep) retardation. The equivalent expressions for the frequency dependence of the modulus, from Equation 20, are

$$\begin{aligned} K_M(\omega) &= K_{M,0}\frac{J_0F(\omega) + J(\infty)}{J_0 + J(\infty)} - i\omega \qquad (26) \\ &= K_{M,0}\gamma - i\omega + K_{M,0}(1 - \gamma)F(\omega) \qquad (27) \end{aligned}$$

and hence

$$K_M(t) = K_{M,0}\gamma\delta(t) - 1.\dot{\delta}(t) + K_{M,0}(1 - \gamma)\dot{\psi}(t) \qquad (28)$$

is the memory function of modulus relaxation.

Equations 25 and 28 are general expressions for the memory functions of mechanical response. They show that the creep compliance and the modulus are related not only in the frequency domain, through Equation 22a, but are also directly related in the time domain through their memory functions. This inter-relationship can be made more explicit by substituting for $K_J(t)$ and $K_M(t)$ in Equation 15b and its modulus equivalent to obtain

$$\sum_j \alpha_j(t) = \int_{-\infty}^{t} \bar{K}_J(t - \tau)\sum_j \alpha_j(\tau)\,\mathrm{d}\tau + \bar{C}_J\sigma(t) \qquad (29a)$$

and

$$\sum_j \sigma_j(t) = \int_{-\infty}^{t} \bar{K}_M(t - \tau)\sum_j \sigma_j(\tau)\,\mathrm{d}\tau + \bar{C}_M\alpha(t) \qquad (29b)$$

In these reduced dynamic equations the "instantaneous" response acts as the source for the time development of the system, with

$$\bar{C}_J = J(\infty) \qquad (30a)$$

and

$$\bar{C}_M = M(\infty) \qquad (30b)$$

The reduced memory functions now relate the response to the time development of the "observable" increment of the conjugate variable, i.e.

$$\bar{K}_J(t) = [M_0/M(\infty)]\dot{\theta}(t) \qquad (31a)$$

and

$$\bar{K}_M(t) = [-J_0/J(\infty)]\dot{\psi}(t) \qquad (31b)$$

with Fourier transformation of Equations 29a and 29b leading directly to the reciprocal relationship of Equation 22a when the source term has a $\delta(t)$ behaviour.

It should be noted that the interpretation of the reduced memory functions in terms of the observable quantities depends upon the type of excitation source used for measurement. Thus $\bar{K}_J(t)$ is determined by either the observable stress response to delta function excitation or the observable stress relaxation rate for step-up excitation, with the appropriate conjugate variables applying for $\bar{K}_M(t)$.

4. The Debye response

The expressions derived for the "cooperative" memory functions in the previous section are quite general and do not depend on any specific form for the spectral functions $F(\omega)$ and $G(\omega)$. In particular they imply that even when the "observable" response has a Debye form an allowance for the "instantaneous" component causes the memory function to extend into the relaxation time range, in contrast to its zero time delta function, $\delta(t)$, behaviour for an isolated process.

For a Debye response the relaxation is exponential and thus we may take $\exp(-\omega_\mathrm{p} t)$ for $\psi(t)$ and $(1 + i\omega/\omega_\mathrm{p})^{-1}$ for $F(\omega)$. Because [6]

$$\begin{aligned} &\{J_0 F(\omega) + J(\infty)\}^{-1} \\ &= [1 + i\omega/\omega_\mathrm{p}][J_0 + J(\infty) + iJ_0\omega/\omega_\mathrm{p}]^{-1} \\ &= M(\infty) - M_0[1 + i\omega/\omega_{M,\mathrm{p}}]^{-1} \end{aligned} \tag{32}$$

with

$$\omega_{M,\mathrm{p}} = [J_0 + J(\infty)][J(\infty)]^{-1}\omega_\mathrm{p} = \gamma^{-1}\omega_\mathrm{p} \tag{33}$$

$\theta(t)$ is also exponentially decaying with a relaxation time $\gamma\omega_\mathrm{p}^{-1}$. The relationship between the cooperative memory function and the relaxation of the conjugate variable contained in Equations 25 and 28 can be confirmed directly in this case because inversion of Equations 18 and 20 is possible and yields

$$\begin{aligned} K_J(t) = & -K_{J,0}\omega_\mathrm{p}\gamma^{-2}(1 - \gamma)\exp(-\omega_\mathrm{p}\gamma^{-1}t) \\ & + K_{J,0}\gamma^{-1}\delta(t) - \dot{\delta}(t) \end{aligned} \tag{34a}$$

and

$$\begin{aligned} K_\mathrm{M}(t) = & K_{M,0}\omega_\mathrm{p}(1 - \gamma)\exp(-\omega_\mathrm{p}t) \\ & + K_{M,0}\gamma\delta(t) - \dot{\delta}(t) \end{aligned} \tag{34b}$$

The identification of the modulus relaxation function

$$\omega_\mathrm{p}\gamma^{-1}\exp(-\omega_\mathrm{p}\gamma^{-1}t) = \dot{\theta}(t) \tag{35a}$$

and the creep retardation function

$$\omega_\mathrm{p}\exp(-\omega_\mathrm{p}t) = \dot{\psi}(t) \tag{35b}$$

gives $K_J(t)$ and $K_M(t)$ in the form of Equations 25 and 28. Thus when the "observable" compliance response has the Debye form its memory function extends exponentially into the relaxation region with the time scale $\gamma\omega_\mathrm{p}^{-1}$ appropriate to the similarly exponential modulus relaxation. Equivalently the modulus memory function follows the exponentially relaxing compliance at times greater than zero.

5. Discussion

In the previous sections we have shown that when more than one step is sequentially involved in the complete mechanical relaxation of a viscoelastic system one part of the memory extends over the whole time scale of the relaxation. This result applies even when the slowest mechanical component relaxes exponentially, in contrast to the situation applying when the different components relax independently, for which the memory functions need be non-zero only at time zero. We have also argued that sequential relaxation is the natural behaviour, with fast inertial (and constraint relaxing) motions being essential for a centre to enter its relaxation coordinate. These motions have been denoted as the "instantaneous" and "observable" responses, and it has been shown that the memory function at non-zero time for the response is determined by the "observable" relaxation of its conjugate variable. The relationships that ensue between spectral and memory functions for the compliance and modulus are summarized in Fig. 2, and mean that the determination of any one of four quantities, as a function of frequency or time, is sufficient to determine all the others.

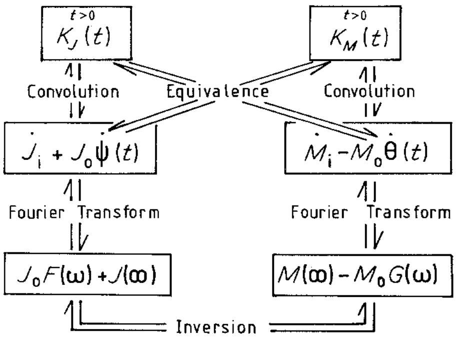

Figure 2 Schematic representation of the compliance/modulus retardation/relaxation functions and their inter-relationships. The symbols are defined in the text.

Elimination of the singular parts of the memory functions, Equations 25 and 28, has resulted in the reduced Equations 29 which have simple interpretation. Here the "instantaneous" response drives the dynamical behaviour of the system. In the first place the magnitude of the source term is rescaled to that of the "instantaneous" component. Secondly the "instantaneous" component of one response variable acts as a step-up source for its conjugate whose slow ("observable") relaxation determines the time dependence of the response through the reduced memory functions, Equations 31a and 31b. This latter feature implies that the "instantaneous" release of stress/strain at a centre drives the further slow relaxation of the centre.

The relationships derived here in Equations 25 and 28 are independent of the form of the relaxation functions $\psi(t)$ and $\theta(t)$, and thus apply equally well to non-exponential as to exponential relaxation. Thus if the relaxation is non-exponential the non-singular part of its memory function is also non-exponential, just as both are exponential in the case of the Debye response considered in Section 4. Furthermore the similarity in form between $K_J(t)/\ K_M(t)$ and the delta function responses $J(t)/M(t)$, Equations 19 and 21, allow us to conjecture that if the memory functions are themselves considered to be dynamic variables in the spirit of reference [12] then all functions in the embedded hierarchy of equations of motion will be either exponential (Debye response) or non-exponential (non-Debye response). If this is the case then it implies that no arbitrary truncation of the Mori series, which will necessarily lead to exponential behaviour for the preceeding response function, can successfully describe non-exponential response functions. However, those theoretical approaches which derive non-exponential relaxation, other than the elementary distribution of relaxation times concept, are all based on a formally infinite hierarchical sequence of processes [13] and therefore avoid arbitrary truncation of the embedded response functions.

6. Conclusions

It has been shown that when describing mechanical relaxation by means of equations of motion with a memory function a proper account of the initial (boundary) conditions is of paramount importance. When due allowance is made for an "instantaneous" component in a sequential relaxation mechanism it has been shown that the memory function always extends into the time scale of the relaxation, and relationships have been derived between the memory function of the response and the relaxation of the conjugate variable. Elimination of singularities in the memory functions can be achieved resulting in a reduced dynamical equation for which the origin of the response is seen to be the "instantaneous" component of the relaxation itself.

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