

Werten bei 5,4 Hz etwa 5%, bei 16,2 Hz etwa 13%. Damit ist anhand der Meßergebnisse an einem realen Werkstoff gezeigt, daß es in Übereinstimmung mit theoretischen Vorstellungen notwendig ist, die jeweilige Schwingungsart anzugeben, bei der der komplexe E -Modul eines Stoffes ermittelt wurde. Dies ist insbesondere dann nötig, wenn aus den Meßergebnissen ein Modell zur Beschreibung des mechanischen Verhaltens ermittelt werden soll (7).

In Gl. [13] sind die exakten Umrechnungsbeziehungen für übliche Dämpfungsmaße angegeben. Hierbei handelt es sich jedoch ausschließlich um Dämpfungsmaße bei freien Schwingungen. Die Möglichkeit, die Dämpfungsmaße bei freien Schwingungen in entsprechende Größen bei erzwungenen Schwingungen, z. B. den mechanischen Verlustfaktor $\operatorname{tg} \delta$ umzurechnen, ist, wie sich leicht aus den obigen Ausführungen zeigen läßt, nicht allgemein gegeben, da diese Umrechnungsbeziehungen modellabhängig sind.

So gilt z. B. für das einfache *Voigt-Kelvin*-Modell (Parallelschaltung von Feder und Dämpfer)

$$\operatorname{tg} \delta = \frac{\frac{A}{\pi}}{1 + \left(\frac{A}{2\pi}\right)^2}; \quad [14]$$

und für das einfache *Maxwell*-Modell (Reihenschaltung von Feder und Dämpfer)

$$\operatorname{tg} \delta = \frac{A}{\pi}. \quad [15]$$

Die den Gln. [14] und [15] entsprechenden Umrechnungsbeziehungen zwischen dem mechanischen Verlustfaktor bei erzwungenen Schwingungen $\operatorname{tg} \hat{\delta}$ und dem bei freien Schwingungen ermittelten logarithmischen Dekrement A können daher nicht allgemein angewandt werden, sondern erst, wenn er-

wiesen ist, daß sich der untersuchte Werkstoff wie ein *Voigt-Kelvin*- oder *Maxwell*-Körper verhält (8). Die in Gl. [13] angegebene Beziehung für freie Schwingungen gilt dagegen allgemein für alle linear-viskoelastischen Stoffe.

Zusammenfassung

Das mechanische Verhalten linear-viskoelastischer Stoffe kann mit Modellen, die aus Federn und Dämpfern aufgebaut sind, beschrieben werden. Berechnet man die komplexe Federkonstante dieser Modelle, so ergeben sich bei freien und erzwungenen Schwingungen verschiedene Werte. Es ist daher notwendig, zur komplexen Federkonstante zusätzlich die jeweils betrachtete Schwingungsart anzugeben. Dies wird durch Versuche, die an Hart-PVC bei +110 °C durchgeführt wurden, bestätigt.

Da die Federkonstante und damit auch der Elastizitätsmodul vom zeitlichen Verlauf der Beanspruchung abhängt (komplexer E -Modul bei freien und erzwungenen Schwingungen, *Retardationsmodul*, *Relaxationsmodul*), sollte man nicht den E -Modul, sondern die Konstanten des Stoffmodells (*Relaxationsspektrum*) als Stoffkenngrößen bezeichnen.

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Free Damped Vibrations of Linear Viscoelastic Materials*

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With 6 figures in 7 details and 2 tables

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1. Introduction

Free damped vibrations are often used in rheology for the determination of dynamic

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mechanical properties of linear viscoelastic materials. This method has a serious difficulty: experiments using damped vibrations cannot give direct information about dynamic mechanical properties which are

only defined for undamped sinusoidal oscillations. This problem has already been discussed by various authors previously; see for example *Staverman* and *Schwarzl* (1) and *Markovitz* (2). To our knowledge no satisfactory solution has so far been given in the literature; it offers no general theory about free vibrations of which the results can be readily used in rheological measurements.

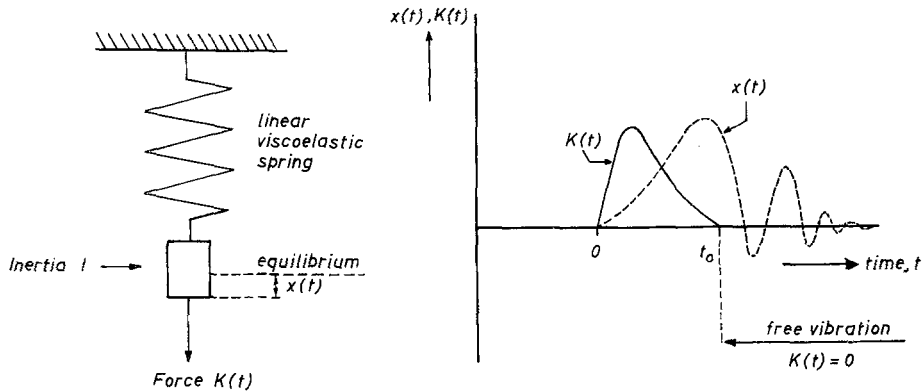


Fig. 1. Schematical illustration of the free vibrating system and the start conditions

We will attempt to present such a theory in this paper.

The mechanical problem under consideration is illustrated in fig. 1. An inert body I is attached to a linear viscoelastic spring fixed at one side. All inertia of the system is concentrated in I . Free vibrations are generated as follows: The system is at rest for time $t \leq 0$. For time $t > 0$ a time-dependent force $K(t)$ acts on I . This force is finite and only non-zero over the limited time interval $0 < t \leq t_0$. These conditions are called the *Start Conditions*. The response after release of the force $K(t)$, i. e. for $t > t_0$, is called the *Free Vibration*.

Most of the free vibrating systems used in rheological measurements, can be represented in this way. A typical example is the torsional pendulum (3, 10). In this case, I is a rotational inertia and the spring stands for the viscoelastic specimen deformed by torsion.

In Chapter 2, we will discuss the equation of motion of this system, and further its formal solution for free vibrations. We will show that the free damped vibration is governed by an *eigen frequency*, ω_0 , and a *logarithmic decrement*, Λ . Part of these considerations can already be found in the literature. In Chapter 3 we will derive formulae to calculate the dynamic mechanical properties, viz. the dynamic modulus, from ω_0 and Λ .

All derivations in this paper are based on only two assumptions for the mechanical relaxation of the viscoelastic material. The first is that *Boltzmann's superposition principle* can be applied, the second that the relaxation has a *positive discrete relaxation spectrum*. The latter assumption can be justified from irreversible thermodynamics, see *Staverman* and *Schwarzl* (1).

2. Equation of Motion and its Formal Solution for Free Damped Vibrations

2.1. The equation of motion¹⁾

Combining *Newton's second law*, *Boltzmann's superposition principle* and the *start conditions* discussed above, we find for the system of fig. 1 the following equation of motion:

$$K(t) = I\ddot{x}(t) + g[M(0)x(t) + \int_0^t \dot{M}(t-\xi)x(\xi)d\xi]. \quad [1]$$

In this formula $x(t)$ is the deviation from equilibrium (see fig. 1), $M(t)$ is the stress relaxation modulus of the viscoelastic material and g is a form factor, depending on the deformation geometry of the specimen²⁾. Eq. [1] may easily be *Laplace-transformed*. It follows³⁾:

$$x(p) = \frac{K(p)}{Ip^2 + gM^*(p)} \quad [2]$$

¹⁾ This equation of motion has already been derived by *Brinkman* (4) in 1955; see also *Markovitz* (2).

²⁾ A summary of form factors for various deformation geometries is given by *Ferry* (3).

³⁾ The term $I\{px(0+) + \dot{x}(0+)\}$, arising by *Laplace-transformation* of the term $I\ddot{x}(t)$ in [1], is zero, because the force $K(t)$ is finite and the inertia-containing system was at rest for $t \leq 0$ (start conditions).

with

$$\left. \begin{aligned} K(p) &= L\{K(t)\} \stackrel{\text{def}}{=} \int_0^\infty e^{-pt} K(t) dt \\ x(p) &= L\{x(t)\} \\ M^*(p) &= L\{M(t)\} + M(0) \end{aligned} \right\} [2A]$$

2.2. The Analytical Expression for the Free Damped Vibration (Formal Solution of the Equation of Motion)

To find an analytical expression for the free damped vibration we use the assumption that the stress relaxation modulus, $M(t)$, has a positive discrete relaxation spectrum. In formula⁴):

$$M(t) = M(\infty) + \sum_{k=1}^N \alpha_k e^{-t/\tau_k} \quad [3]$$

with

$$\left. \begin{aligned} \alpha_k, \tau_k &> 0 \quad k = 1, 2, \dots, N \\ \tau_{k+1} &> \tau_k \quad k = 1, 2, \dots, (N-1) \\ M(\infty) &\geq 0 \end{aligned} \right\} [4]$$

For $M^*(p)$ according to [2A] and [3] we find:

$$M^*(p) = M(0) - \sum_{k=1}^N \frac{\alpha_k}{1 + p\tau_k} \quad [5]$$

Substitution of [5] into the equation of motion [2] gives:

$$x(p) = K(p) \rho \frac{\prod_{k=1}^N (1 + p\tau_k)}{\prod_{j=1}^{N+2} (p + \beta_j)} \quad [6]$$

In this formula the constant ρ is real and positive. The numbers $-\beta_1, -\beta_2, \dots, -\beta_i, \dots, -\beta_{N+2}$ in [6] are the $(N+2)$ roots of the equation:

$$\frac{Ip^2}{g} + M(0) - \sum_{k=1}^N \frac{\alpha_k}{1 + p\tau_k} = 0 \quad [7]$$

It can be shown⁵) that at least N of these roots, i. e. $-\beta_1, -\beta_2, \dots, -\beta_N$, are non-positive real with:

$$\left. \begin{aligned} -1/\tau_1 &< -\beta_1 < -1/\tau_2 \\ -1/\tau_k &< -\beta_k < -1/\tau_{k+1} \\ -1/\tau_N &< -\beta_N \leq 0; -\beta_N = 0 \text{ if } M(\infty) = 0 \end{aligned} \right\} [8]$$

Further⁵) the two other roots $-\beta_{N+1}$, and $-\beta_{N+2}$ may be conjugate complex with non-positive real part as well as non-positive real.

⁴) The number of time constants is taken finite; as can be verified easily, the subsequent derivations also hold if it is infinite.

⁵) Part of the proof is given by Elder (5); by considering eq. [7] for real p -values it may be verified easily.

Two conjugate complex roots

We first consider the case of conjugate complex roots⁶):

$$\left. \begin{aligned} -\beta_{N+1} &= p_0 = -\lambda_0 + i\omega_0 \\ -\beta_{N+2} &= \bar{p}_0 = -\lambda_0 - i\omega_0 \\ i &= \sqrt{-1} \\ \lambda_0 &\geq 0; \omega_0 > 0 \end{aligned} \right\} [9]$$

Because all $(N+2)$ roots are different from each other, and as a matter of course also from the τ_k 's, we can separate the rational part of [6] into partial fractions as follows:

$$x(p) = K(p) \left[\frac{\gamma_1}{p - p_0} + \frac{\gamma_2}{p - \bar{p}_0} + \sum_{k=1}^N \frac{\sigma_k}{p + \beta_k} \right] \quad [10]$$

The constants $\gamma_1, \gamma_2, \sigma_1, \sigma_2, \dots, \sigma_N$ in [10] are the residues of the rational function in [6] at the points $p_0, \bar{p}_0, -\beta_1, -\beta_2, \dots, -\beta_N$. From [8] and [9] it follows that:

$$\left. \begin{aligned} \gamma_1 &= \bar{\gamma}_2 \\ \sigma_k &\text{ real and positive for } k = 1, 2, \dots, N \end{aligned} \right\} [11]$$

Laplace-inversion of [10] leads to:

$$\begin{aligned} x(t) &= A \int_0^t K(\xi) e^{-\lambda_0(t-\xi)} \cos\{\omega_0(t-\xi) + \varphi\} d\xi \\ &+ \sum_{k=1}^N \sigma_k \int_0^t K(\xi) e^{-(t-\xi)\beta_k} d\xi \end{aligned} \quad [12]$$

In [12] the real constants A and φ depend on γ_1 and γ_2 by elementary goniometric formulae.

From [12] we derive the expression for the free vibration. For $t > t_0$, $K(t) = 0$, and [12] reduces to:

$$\begin{aligned} x(t) &= e^{-\lambda_0 t} \{\zeta_1 \cos(\omega_0 t + \varphi) + \zeta_2 \sin(\omega_0 t + \varphi)\} \\ &+ \sum_{k=1}^N d_k e^{-\beta_k t} \end{aligned} \quad [13]$$

with:

$$\zeta_1 = A \int_0^{t_0} K(\xi) e^{+\lambda_0 \xi} \cos(\omega_0 \xi) d\xi \quad [14A]$$

$$\zeta_2 = A \int_0^{t_0} K(\xi) e^{+\lambda_0 \xi} \sin(\omega_0 \xi) d\xi \quad [14B]$$

$$d_k = \sigma_k \int_0^{t_0} K(\xi) e^{+\beta_k \xi} d\xi \quad [14C]$$

Formula [13] can be simplified to:

$$\boxed{x(t) = B e^{-\lambda_0 t} \cos(\omega_0 t + \psi) + \sum_{k=1}^N d_k e^{-\beta_k t} \quad [15]} \\ \lambda_0 \geq 0; \omega_0 > 0; \beta_k > 0; k = 1, 2, \dots, N-1 \\ \beta_N \geq 0$$

⁶) \bar{p}_0 means conjugate complex to p_0 ; labelling with a dash, as common for conjugate complex numbers, is used in another sense in this paper.

In [15] the real constants B and ψ depend on ζ_1 , ζ_2 and φ by simple goniometric formulae.

In [15] the constants ω_0 and λ_0 do not depend on $K(t)$ as they are the imaginary and the real part of a complex root of eq. [7] in which neither $K(t)$ nor $K(p)$ appears. We may therefore formulate our result as follows:

The free damped vibration consists of two parts: first, an exponentially damped harmonic vibration of which the frequency ω_0 and the damping term λ_0 do not depend on $K(t)$ (the manner in which the free vibration was initiated); second, a sum of exponentials.

From eq. [14C] it does not follow that all constants d_k must have the same sign. This means that the sum of exponentials (each absolutely not increasing because the β_k 's are non-negative) is not necessarily monotonic⁷). A special case is that of a $K(t)$ with no change of sign over the interval $(0, t_0)$; the integral in [14C] then has the same sign for each k . Consequently because of [11], all constants d_k have the same sign now. In this special case the sum of exponentials is monotonic.

All roots non-positive real

As can be verified easily, no damped harmonic vibration as given in [15] will result in this case of „over-critically damping“.

3. Theory Concerning the Determination of the Complex Dynamic Modulus by Free Damped Vibrations

3.1. The dynamic modulus; its determination with free vibration experiments

The complex dynamic modulus $\bar{M}(\omega)$ of a linear viscoelastic material is defined for a stationary harmonic vibration with angular frequency ω , see *Staverman and Schwarzl* (1). The stress-strain amplitude ratio being A , the positive phase angle δ , $\bar{M}(\omega)$ is given by:

$$\bar{M}(\omega) = A e^{+i\delta} \quad [16]$$

$$i = \sqrt{-1}$$

The complex dynamic modulus $\bar{M}(\omega)$ can be measured directly by experiments, using forced sinusoidal oscillations [see *Staverman and Schwarzl* (1)]. However, especially for materials with low damping, i. e. with small values of phase angle δ , and further at very

⁷) If the sum of exponentials is not monotonic, it has extrema. The separation of the exponentially damped harmonic vibration from the non-monotonic sum of exponentials may then become a difficult problem in the practice of free vibration measurements.

low frequencies, the measurement of phase angle δ is a difficult problem. Therefore, the dynamic modulus $\bar{M}(\omega)$ is often measured by free damped vibration techniques. A specimen is made part of the mechanical system, illustrated schematically in fig. 1; it plays the role of viscoelastic spring. A free damped vibration is initiated by applying a force $K(t)$, obeying the start conditions specified in the Introduction. Finally, the free damped vibration is recorded.

We have already discussed that (parameters of) this free damped vibration do not give direct information on the dynamic modulus, as it is not defined for this case. However, it remains to be seen if there is any other uniquely defined material property that can be obtained directly.

3.2. The Material Property $M^(p_0)$ as Obtained Directly from Free Vibration Measurements*

From the theory given in Section 2.2. we know that the free vibration contains at most one simple damped harmonic vibration, characterized by a frequency ω_0 and a damping term λ_0 ; see formula [15]. It is only present if the system is not over-critically damped. If we measure⁸) λ_0 and ω_0 we have experimentally found the root

$$p_0 = -\lambda_0 + i\omega_0$$

of eq. [7]. (Follow the reasoning in 2.2. in the reverse direction.) When we remember [5], it follows that:

$$\frac{I p_0^2}{g} + M^*(p_0) = 0. \quad [17A]$$

In other words, from p_0 , calculated from the measured values of λ_0 and ω_0 , we immediately find the value of the function $M^*(p)$ for $p = p_0$. The very simple formula reads⁹):

$$M^*(p_0) = -\frac{I p_0^2}{g}. \quad [17B]$$

Splitting [17B] in real and imaginary parts and introducing the *logarithmic decrement*, Λ , according to¹⁰)

$$\Lambda = \frac{2\pi\lambda_0}{\omega_0} \quad [17C]$$

⁸) The measurement of λ_0 and ω_0 implies that the damped harmonic vibration in [15] can be separated from the sum of exponentials. We do not discuss under what circumstances this is actually possible.

⁹) Λ is the natural logarithm of the decay per period of the damped harmonic vibration in [15].

¹⁰) Notice that [17B] is a generalization of the classical equation for free vibrating systems with a Hookean spring.

we find:

$$M_1(p_0) = \text{Re} \{M^*(p_0)\} = \frac{I \omega_0^2}{g} \left(1 - \frac{A^2}{4 \pi^2}\right) \quad [17D]$$

$$M_2(p_0) = \text{Im} \{M^*(p_0)\} = \frac{I \omega_0^2}{g} \frac{A}{\pi} \quad [17E]$$

From the definition formula [2A] for $M^*(p)$ we observe that the function $M^*(p)$ and therefore also its value $M^*(p_0)$ in the point p_0 is a material property uniquely defined.

3.3. The Significance of the Function $M^*(p)$ and its Connection with the Dynamic Modulus $\bar{M}(\omega)$

To begin with, we give two formulae; the first, [18A], is identical with the definition eq. [2A] of $M^*(p)$ for $p = \lambda + i\omega$; the second, [18B], follows from Boltzmann's superposition principle and the definition formula [16] for the dynamic modulus $\bar{M}(\omega)$.

$$M^*(\lambda + i\omega) = M(0) + \int_0^\infty e^{-(\lambda + i\omega)t} \dot{M}(t) dt \quad [18A]$$

$$\bar{M}(\omega) = M(0) + \int_0^\infty e^{-i\omega t} \dot{M}(t) dt. \quad [18B]$$

Taking $\lambda = 0$ in [18A], it follows that

$$\bar{M}(\omega) \equiv M^*(i\omega). \quad [18C]$$

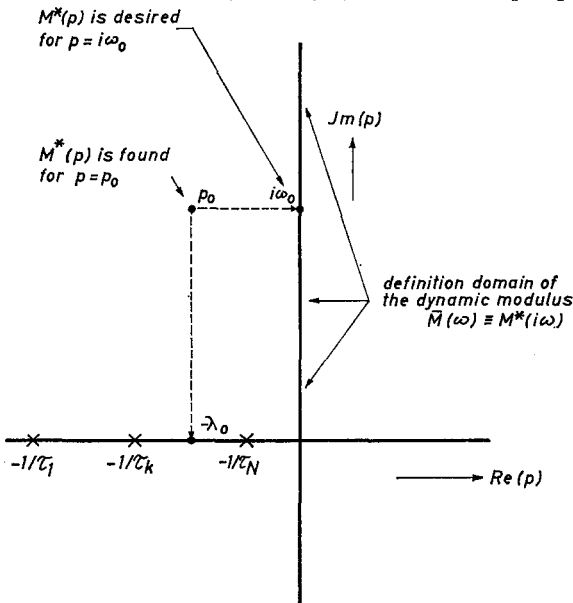


Fig. 2. The complex p -plane

Consider now fig. 2; plotted is the complex p -plane. The function $M^*(p)$ is defined by [18A] for complex values of p . For materials with a positive discrete spectrum the integral [18A] only converges for $Re(p) > -1/\tau_N$; see [3]. For other p -values, $M^*(p)$ can be defined by analytic continuation. From [18C]

it follows that the dynamic modulus $\bar{M}(\omega)$ is a special case of $M^*(p)$ for $p = i\omega$, i. e. for p -values on the positive imaginary axis. Therefore $M^*(p)$ is called the (generalized) complex modulus for the complex frequency p ¹¹⁾.

The conclusion of Section 3.2. may now be reformulated: *With free vibration techniques one measures the complex modulus $M^*(p_0)$ for the complex eigen frequency p_0 of the mechanical system.* While we intended to determine the dynamic modulus, for example at a frequency ω_0 , we have found the value of $M^*(p)$ in the wrong point: in the point $p_0 = -\lambda_0 + i\omega_0$ instead of in the desired point $i\omega_0$ (see fig. 2). To obtain the dynamic modulus we must know how to calculate $M^*(p_0)$ and $M^*(i\omega_0)$, one from the other.

3.4. Derivation of Formulae to Calculate $M^*(p_0)$ and $\bar{M}(\omega_0)$, one from the other Analytical Expressions

To begin with, we specify some notations.

$$M_1(p) = \text{Re} \{M^*(p)\}; \quad \bar{M}_1(\omega) = \text{Re} \{\bar{M}(\omega)\} \quad [19A]$$

$$M_2(p) = \text{Im} \{M^*(p)\}; \quad \bar{M}_2(\omega) = \text{Im} \{\bar{M}(\omega)\} \quad [19B]$$

$$\tan \delta(p) = M_2(p)/M_1(p); \quad \tan \delta(\omega) = \bar{M}_2(\omega)/\bar{M}_1(\omega) \quad [20]$$

As indicated, all quantities related to the dynamic modulus are labelled with a horizontal dash; this notation should not be confused with the usual notation of complex conjugated quantities in the same manner.

From the assumption that $M(t)$ has a positive discrete relaxation spectrum, it follows that the function $M^*(p)$, given in eq. [5], is analytic in the entire p -plane, except in the simple poles $-1/\tau_k$ ($k = 1, 2, \dots, N$) on the negative real axis; see fig. 2. As a consequence, we can expand $M^*(p)$ around the point $p_0 = -\lambda_0 + i\omega_0$, as well as around the point $i\omega_0$ in an infinite convergent Taylor series. The radius of convergence R is at least ω_0 in both cases, because the distance of the nearest pole to p_0 as well as to $i\omega_0$ is at least ω_0 . Writing according to [17C]:

$$i\omega_0 - p_0 = +\lambda_0 = \left(\frac{A}{2\pi}\right) \omega_0 \quad [21]$$

we find the following series to calculate the dynamic modulus $\bar{M}(\omega_0) \equiv M^*(i\omega_0)$ from $M^*(p)$ and its derivatives in the point $p = p_0$:

$$\bar{M}(\omega_0) = M^*(p_0) + \sum_{k=1}^{\infty} \frac{(-i)^k}{k!} \times \left(\frac{A}{2\pi}\right)^k \omega_0^k \left[\frac{\partial^k M^*(p)}{\partial \omega^k} \right]_{p_0}. \quad [22]$$

¹¹⁾ For details see Hiedemann and Spence (6) or König and Meixner (7).

The derivatives to p are expressed as partial derivatives to ω , with λ constant; this is allowed because $M^*(p)$ is analytic.

For the calculation of $M^*(p_0)$ from $\bar{M}(\omega)$ we obtain in the same manner:

$$M^*(p_0) = \bar{M}(\omega_0) + \sum_{k=1}^{\infty} \frac{(i)^k}{k!} \times \left(\frac{\Lambda}{2\pi}\right)^k \omega_0^k \left[\frac{d^k \bar{M}(\omega)}{d\omega^k} \right]_{\omega_0} \quad [23]$$

Because $\bar{M}(\omega)$ is a function of ω only, the partial derivatives as in [22] are replaced by normal derivatives in [23].

Both series, [22] and [23], converge for $\Lambda < 2\pi$ because $R \geq \omega_0$. Therefore, the use of these series in the next theory implies the restriction $\Lambda < 2\pi$. As will be discussed in Section 3.5., experimental values for Λ are generally smaller than about 1 to 1.5. Accordingly, the restriction $\Lambda < 2\pi$ has no significance regarding to the applicability of the theory.

Successive Approximations

In practice, the series [22] and [23] cannot be summed rigorously, because we never know more than a limited number of higher derivatives. Therefore, we now consider the successive approximations for $\bar{M}(\omega_0)$ and $M^*(p_0)$, which arise if we sum in respectively [22] and [23] only the first n terms. For the 0-order and n^{th} -order approximation it follows that:

For the calculation of $\bar{M}(\omega_0)$ from $M^*(p_0)$:

$\{\bar{M}(\omega_0)\}_0 = M^*(p_0)$	[24A]
$\{\bar{M}(\omega_0)\}_n = M^*(p_0) + \sum_{k=1}^n \frac{(-i)^k}{k!} \left(\frac{\Lambda}{2\pi}\right)^k \omega_0^k \left[\frac{\partial^k M^*(p)}{\partial \omega^k} \right]_{p_0}$	[24B]
$n = 1, 2, \dots$	

For the calculation of $M^*(p_0)$ from $\bar{M}(\omega_0)$:

$\{M^*(p_0)\}_0 = \bar{M}(\omega_0)$	[25A]
$\{M^*(p_0)\}_n = \bar{M}(\omega_0) + \sum_{k=1}^n \frac{(i)^k}{k!} \left(\frac{\Lambda}{2\pi}\right)^k \omega_0^k \left[\frac{d^k \bar{M}(\omega)}{d\omega^k} \right]_{\omega_0}$	[25B]
$n = 1, 2, \dots$	

Truncation errors in the approximation formulae

Truncation errors in the approximation formulae can be found by using a general relation derived in the Appendix:

$$\left| \frac{\omega^k}{k!} \left[\frac{\partial^k M^*(p)}{\partial \omega^k} \right]_p \right| \leq M_2(p) \quad [26]$$

$p = -\lambda + i\omega$

For the error e_n in [24B], defined by:

$$e_n^{def} \bar{M}(\omega_0) - \{\bar{M}(\omega_0)\}_n \quad [27]$$

we find from [22], [24B] and [26] that:

$$|e_n| \leq M_2(p_0) \sum_{k=n+1}^{\infty} \left(\frac{\Lambda}{2\pi}\right)^k = M_2(p_0) \frac{(\Lambda/2\pi)^{n+1}}{(1-\Lambda/2\pi)} \quad [28]$$

In the same manner we find for the error e_n' in [25B] and defined by:

$$e_n^{def} M^*(p_0) - \{M^*(p_0)\}_n \quad [29]$$

the following:

$$|e_n'| \leq \bar{M}_2(\omega_0) \frac{(\Lambda/2\pi)^{n+1}}{(1-\Lambda/2\pi)} \quad [30]$$

To derive [30], [26] is used for the special case $p = i\omega_0$.

Approximation errors in storage and loss components

First we specify a notation. The absolute error in the storage modulus $\bar{M}_1(\omega_0)$ calculated by the n^{th} -order approximation formula [25B] is written as:

$$\bar{M}_1(\omega_0) - \{\bar{M}_1(\omega_0)\}_n = \delta_n \bar{M}_1(\omega_0)$$

In the same manner we use the abbreviations $\delta_n \bar{M}_2(\omega_0)$, $\delta_n M_1(p_0)$ and $\delta_n M_2(p_0)$.

From these definitions, together with formulae [27] to [30], we obtain upper limits for the absolute errors:

$$|\delta_n \bar{M}_1(\omega_0)|, |\delta_n \bar{M}_2(\omega_0)| \leq |e_n| \leq M_2(p_0) \frac{(\Lambda/2\pi)^{n+1}}{(1-\Lambda/2\pi)} \quad [31A]$$

$$|\delta_n M_1(p_0)|, |\delta_n M_2(p_0)| \leq |e_n'| \leq \bar{M}_2(\omega_0) \frac{(\Lambda/2\pi)^{n+1}}{(1-\Lambda/2\pi)} \quad [31B]$$

Further, we could derive upper limits for the relative errors in storage and loss components. Table 1 gives a summary¹²⁾. The upper half of this table concerns the calculation of $\bar{M}(\omega_0)$ from $M^*(p_0)$ by the n^{th} -order approximation formula [24], the lower half the reverse problem. Two types of relative errors are distinguished. The first deals with the error in the quantity obtained, relative to the original quantity. An example is:

$$\frac{\delta_n \bar{M}_1(\omega_0)}{\bar{M}_1(\omega_0)}$$

Limits for these errors are given by formulae [32A], [32B], [32D], [33A] and [33C]. The second type concerns the error in the obtained quantity relative to itself,

¹²⁾ The limits given in table 1 are not the best upper limits. Better formulae could be derived but are very complicated. For the case $\Lambda \leq 1$, in which we are mainly interested, (see Section 3.5.), the differences with the formulae of table 1 are small.

Table 1. Upper limits for various relative errors concerning the n^{th} -order approximation; $n = 1, 2, \dots$

Approximation formula	Type of rel. error	Upper limit for the absolute value of the rel. error		Form. no.
		general formulae, $\Lambda < 2\pi$	simplification, $\Lambda \leq 1$	
[24 B] $M^*(p_0) \rightarrow \bar{M}(\omega_0)$ $\Lambda < 2\pi$ $n = 1, 2, \dots$	$\frac{\delta_n \bar{M}_1(\omega_0)}{M_1(p_0)}$	$\tan \delta(p_0) \frac{(\Lambda/2\pi)^{n+1}}{(1 - \Lambda/2\pi)}$	—	[32 A]
	$\frac{\delta_n \bar{M}_1(\omega_0)}{M_1(p_0)}$	$\frac{2 (\Lambda/2\pi)^{n+2}}{(1 - \Lambda/2\pi)^2 (1 + \Lambda/2\pi)}$	2.44 $(\Lambda/2\pi)^{n+2}$	[32 B] ²⁾
	$\frac{\delta_n \bar{M}_1(\omega_0)}{\bar{M}_1(\omega_0)}$	$\frac{2 (\Lambda/2\pi)^{n+2}}{1 - \Lambda/2\pi - 3 (\Lambda/2\pi)^2 + (\Lambda/2\pi)^3}$ ¹⁾	2.60 $(\Lambda/2\pi)^{n+2}$	[32 C] ²⁾
	$\frac{\delta_n \bar{M}_2(\omega_0)}{M_2(p_0)}$	$\frac{(\Lambda/2\pi)^{n+1}}{(1 - \Lambda/2\pi)}$	1.19 $(\Lambda/2\pi)^{n+1}$	[32 D]
	$\frac{\delta_n \bar{M}_2(\omega_0)}{\bar{M}_2(\omega_0)}$	$\frac{(\Lambda/2\pi)^{n+1}}{(1 - \Lambda/2\pi)^2}$	1.41 $(\Lambda/2\pi)^{n+1}$	[32 E]
	$\frac{\delta_n \bar{M}_1(p_0)}{\bar{M}_1(\omega_0)}$	$\tan \delta(\omega_0) \frac{(\Lambda/2\pi)^{n+1}}{(1 - \Lambda/2\pi)}$	—	[33 A]
[25 B] $\bar{M}(\omega_0) \rightarrow M^*(p_0)$ $\Lambda < 2\pi$ $n = 1, 2, \dots$	$\frac{\delta_n M_1(p_0)}{M_1(p_0)}$	$\frac{2 (\Lambda/2\pi)^{n+2}}{(1 - \Lambda/2\pi)^3 (1 + \Lambda/2\pi)}$	2.8 $(\Lambda/2\pi)^{n+2}$	[33 B] ²⁾
	$\frac{\delta_n M_2(p_0)}{\bar{M}_2(\omega_0)}$	$\frac{(\Lambda/2\pi)^{n+1}}{(1 - \Lambda/2\pi)}$	1.19 $(\Lambda/2\pi)^{n+1}$	[33 C]
	$\frac{\delta_n M_2(p_0)}{M_2(p_0)}$	$\frac{(\Lambda/2\pi)^{n+1}}{(1 - \Lambda/2\pi)^2}$	1.41 $(\Lambda/2\pi)^{n+1}$	[33 D]

¹⁾ Only valid if $\Lambda < 0.9\pi$.

²⁾ Only valid if $M^*(p_0)$ is measured by free vibration methods, i. e. if [34] holds.

an example is:

$$\frac{\delta_n \bar{M}_1(\omega_0)}{\bar{M}_1(\omega_0)}$$

Limits of this type are given by formulae [32 C], [32 E], [33 B] and [33 D].

Two sets of formulae for the error limits are given in table 1. The left ones, except [32 C], are valid for $\Lambda < 2\pi$, the right ones are simplified formulae only valid for $\Lambda \leq 1$. Three of the formulae in table 1, viz. [32 B], [32 C] and [33 B], are only valid if $M^*(p_0)$ is measured by free vibration techniques. To understand that this represents a restriction, let us summarize what was exactly done in the preceding theory. We considered the function $M^*(p)$ and tried to find its value in some point from its value in another point. The only assumption for $M^*(p)$ was that it was analytic in the complete complex p -plane, except for a series of simple poles at the negative real p -axis. But the very fact that $M^*(p_0)$ is measured by free vibrations gives *additional information*, viz.:

$$\tan \delta(p_0) = M_2(p_0)/M_1(p_0) = \frac{(\Lambda/\pi)}{1 - (\Lambda/2\pi)^2} \quad [34]$$

Eq. [34] follows immediately from [17 D] and [17 E].

In table 1, the general formulae [32 A], [32 D], [33 A] and [33 C] are obtained immediately from [31 A] and [31 B]. Formula [32 B] is a special case of [32 A] if $M^*(p_0)$ is measured by free vibrations; it follows by substituting [34] in [32 A]. Formulae [32 E] and [33 D] are derived from respectively [32 D] and [33 C] by using the following relation¹³⁾:

$$1 - \left(\frac{\Lambda}{2\pi}\right) \leq \frac{M_2(p_0)}{\bar{M}_2(\omega_0)} \leq \frac{1}{1 - \Lambda/2\pi} \quad [35]$$

The left hand side of [35] is derived from [24 A] and [31 B] for $n = 0$; the right hand side from [25 A] and [31 B] for $n = 0$. Formula [33 B] is found from [31 B], [34] and [35] by writing:

$$\left| \frac{\delta_n M_1(p_0)}{M_1(p_0)} \right| = \left| \frac{\delta_n M_1(p_0)}{\bar{M}_2(\omega_0)} \frac{\bar{M}_2(\omega_0)}{M_2(p_0)} \frac{M_2(p_0)}{M_1(p_0)} \right|$$

Again this formula is only valid if $M^*(p_0)$ is measured by free vibrations. Finally, we derive formula [32 C]. From [29 A] and [32 B]

¹³⁾ By using [25 B] for the case $n = 1$ (first order approximation) it follows from $d\bar{M}_1/d\omega \geq 0$ that:

$$1 - \frac{(\Lambda/2\pi)^2}{1 - \Lambda/2\pi} \leq \frac{M_2(p_0)}{\bar{M}_2(\omega_0)} \leq \frac{1}{1 - \Lambda/2\pi} \quad [35 A]$$

This formula is used in section 3.7.

for the case $n = 0$, it follows that:

$$\left. \begin{aligned} 0 &\leq \frac{1}{1 + 2 \frac{(\Lambda/2\pi)^2}{(1 - \Lambda/2\pi)^2 (1 + \Lambda/2\pi)}} \leq \frac{M_1(p_0)}{\bar{M}_1(\omega_0)} \\ &\leq \frac{1}{1 - 2 \frac{(\Lambda/2\pi)^2}{(1 - \Lambda/2\pi)^2 (1 + \Lambda/2\pi)}} \end{aligned} \right\} [36]$$

if $\Lambda < 0.9\pi$.

Formula [32C] follows from the right hand side of this inequality and from [32B]. The simplified formulae, only valid for $\Lambda \leq 1$, were found by substituting $\Lambda = 1$ in the denominator of the general formulae.

3.5. Usefulness of the Approximation Formulae

From table 1 it can be seen that for the approximation formulae [24] and [25] the error limits continuously decrease with the order of the approximation. However, the usefulness of the formulae in practice strongly depends on the velocity of convergence. Table 1 shows that the logarithmic decrement, Λ , is the dominating factor in this connection. We therefore first consider the range of values for Λ covered in the measurement practice.

There are two reasons to assume that Λ is not greater than about 1. *First*, if we wish to measure λ_0 and ω_0 of a damped sinusoidal vibration according to [15] with some accuracy, the decay per period, viz. exp. (Λ), may not be too large. If Λ becomes large, the number of periods for which the oscillation is above the noise level of the apparatus becomes too small for accurate measurements. Of course, the maximum admissible value of Λ depends on the accuracy of the recording system; a practical limit turned out to be in the order of 1.

Second, it will be shown in 3.7. that the quantity Λ/π is approximately equal to the loss factor $\tan \delta$ of the dynamic modulus. If Λ exceeds 1, consequently $\tan \delta$ must be greater than approximately 0.3. But, in this case, the measurement of the dynamic modulus $\bar{M}(\omega)$ directly by forced sinusoidal vibrations becomes much more practical than the free damped vibration technique¹⁴⁾.

From table 1 we see that all error limits decrease by a factor of $2\pi/\Lambda$, if the order of the approximation is increased by 1.

¹⁴⁾ Many investigators, including the present author, actually use free vibration techniques to measure the dynamic modulus of materials with a loss factor of 1 or more, for example polymers in the glass-rubber transition. We shall discuss this matter in Section 3.7.

Therefore, if $\Lambda \leq 1$, in all cases each approximation is at least $2\pi \simeq 6$ times better than the foregoing; this means that the convergence is rapid and that the formulae are suitable for practical calculations.

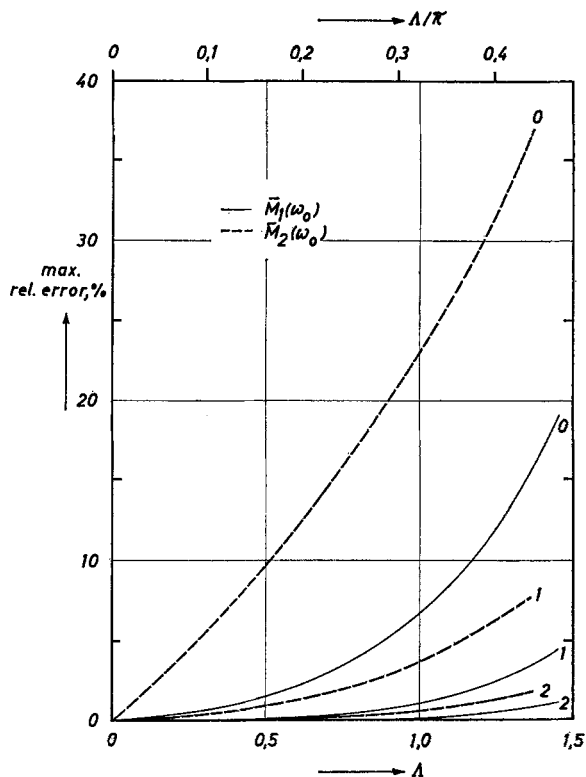


Fig. 3. Limits for $\delta_n \bar{M}_1 / \bar{M}_1$ and $\delta_n \bar{M}_2 / \bar{M}_2$, respectively the relative errors in the dynamic storage and loss modulus, if calculated from $M^*(p_0)$ by the 0th-, 1st- and 2nd-order approximation formula (24B)

Some additional information is summarized in fig. 3 and table 2. It concerns the calculation of $\bar{M}(\omega_0)$ from $M^*(p_0)$, to be performed if the dynamic modulus should be determined by free vibration techniques. Plotted in fig. 3 are error limits versus logarithmic decrement Λ for 0th-, 1st- and 2nd-order approximations of \bar{M}_1 and \bar{M}_2 . We clearly observe that the error limit for the n^{th} -order approximation of the loss modulus \bar{M}_2 is approximately two times smaller than the error limit for the $(n - 1)^{\text{th}}$ -order approximation of the storage modulus \bar{M}_1 (compare also [32C] and [32E]). This means that approximations of the same order for \bar{M}_1 and \bar{M}_2 considerably differ in accuracy, the one for \bar{M}_1 being the best.

Table 2 lists the error limits for the 0th-, 1st-, 2nd- and 3rd- order approximations at a fixed value of Λ , viz. $\Lambda = 1$. This value of Λ being the maximum experimental value,

table 2 also summarizes the maximum errors that are possible in practical calculations.

Table 2. Limits of relative errors in % for $\Lambda = 1$

n	$M^*(p_0) \rightarrow \bar{M}(\omega_0)$	
	$\bar{M}_1(\omega_0)$	$\bar{M}_2(\omega_0)$
0	6.6	22.4
1	1.04	3.6
2	0.17	0.57
3	0.025	0.09

We observe that the 2nd-order approximations have an accuracy better than about 0.5%. This order of approximation is therefore sufficient for calculations with experimental data. A scheme for this will be given in a subsequent paper.

3.6. General Conclusions About the Differences between $M^*(p_0)$ and $\bar{M}(\omega_0)$, $\Lambda \leq 1$

For the differences between the moduli $M^*(p_0)$ and $\bar{M}(\omega_0)$, as measured respectively with free damped and forced undamped vibrations we now shall draw some definite conclusions. Λ is restricted not to exceed a value of 1.

Quantitative Conclusions

The relative differences in storage and loss moduli do not exceed values of respectively 6 and 19%. These values were taken relative to the dynamic storage and loss moduli $\bar{M}_1(\omega_0)$ and $\bar{M}_2(\omega_0)$. This conclusion is based on formulae [35] and [36].

Qualitative Conclusions

To find qualitative conclusions, we use the first order approximation formula [25B]. Split into real and imaginary components it reads:

$$\{M_1(p_0)\}_1 = \bar{M}_1(\omega_0) - (\Lambda/2\pi) \left[\frac{d\bar{M}_2(\omega)}{d \ln \omega} \right]_{\omega_0} \quad [37A]$$

$$\{M_2(p_0)\}_1 = \bar{M}_2(\omega_0) + (\Lambda/2\pi) \left[\frac{d\bar{M}_1(\omega)}{d \ln \omega} \right]_{\omega_0} \quad [37B]$$

Whereas these formulae are accurate within 1 and 3% respectively, (see table 1), the same is true for conclusions drawn from them. The results are illustrated in fig. 4. Plotted are the dynamic storage and loss moduli \bar{M}_1 and \bar{M}_2 both vs angular frequency ω for a hypothetical material; the diagram used is double logarithmic. As indicated in fig. 4. a material with a positive discrete

relaxation spectrum shows various maxima in \bar{M}_2 , accompanied by steplike increments of \bar{M}_1 .

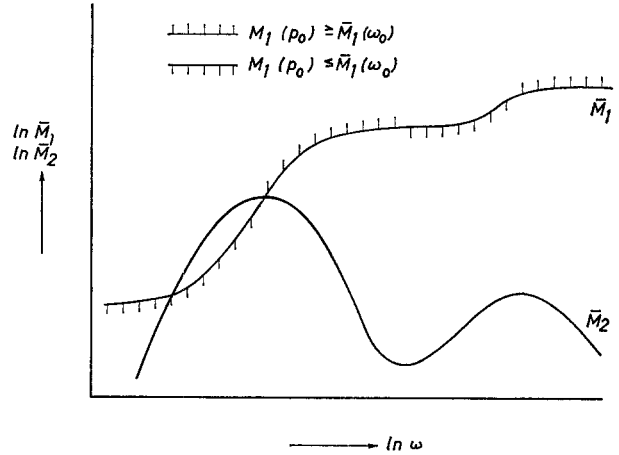


Fig. 4. The difference between $\bar{M}_1(p_0)$ and $\bar{M}_1(\omega_0)$, as it is determined by the slope $dM_2/d \ln \omega$ in the dynamic loss modulus

From [37A] it follows that: the storage modulus $M_1(p_0)$ may be smaller, greater or equal to $\bar{M}_1(\omega_0)$, depending on $d\bar{M}_2/d \ln \omega$ being respectively positive, negative or zero. At the low frequency side of a peak in \bar{M}_2 , $M_1(p_0) \leq \bar{M}_1(\omega_0)$, at the top of the peak $M_1(p_0) = \bar{M}_1(\omega_0)$; at the high frequency side $M_1(p_0) \geq \bar{M}_1(\omega_0)$. (See fig. 4.)

From [37B] it follows that: the loss modulus $M_2(p_0)$ is always greater than $\bar{M}_2(\omega_0)$ because $d\bar{M}_1/d \ln \omega \geq 0$.

3.7. The Relation $\Lambda/\pi \simeq \overline{\tan \delta}(\omega_0)$

Combining [34], [35A] and [36], we obtain:

$$1 - \frac{(\Lambda/2\pi)^2}{(1 - \Lambda/2\pi)^2} \{4 - 5(\Lambda/2\pi) - 2(\Lambda/2\pi)^2 + (\Lambda/2\pi)^3\} \leq \frac{(\Lambda/\pi)}{\overline{\tan \delta}(\omega_0)} \leq 1 + \frac{(\Lambda/2\pi) \{1 + (\Lambda/2\pi)^2\}}{(1 - \Lambda/2\pi)^2} \quad [38]$$

Fig. 5 gives an illustration; plotted are the upper and lower bounds for $\frac{\Lambda/\pi}{\overline{\tan \delta}}$ as functions of Λ . As we are mainly interested in the case $\Lambda \leq 1$, we may simplify [38] to:

$$\Lambda/\pi \simeq \overline{\tan \delta}(\omega_0) \quad [39]$$

The maximum error in this formula is less than 23% if $\Lambda \leq 1$, see fig. 5.

Formula [39] tells us that the restriction $\Lambda \leq 1$, introduced in Section 3.5., is equivalent with a restriction for the loss tangent of the viscoelastic material:

$$\overline{\tan \delta}(\omega_0) \lesssim 0.3 \quad [40]$$

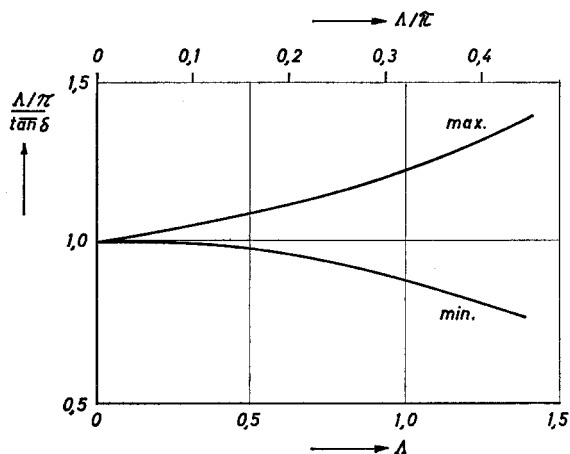


Fig. 5. Upper and lower bounds for $\frac{A/\pi}{\tan \delta}$ as functions of A

In many cases, materials with a loss factor in the order of 1 or more are investigated by free vibration techniques. To avoid a A greater than 1 a *Hookean*, extra stiffness, is then placed parallel to the specimen; the loss tangent of the combination being smaller than about 0.3 in this way. The complex modulus of the combination is measured, that of the specimen is calculated afterwards. It should, however, be realized that this method introduces sources for serious errors; it would be better to use forced sinusoidal vibrations for a direct measurement of the dynamic modulus.

3.8. The Reasonable Accuracy of the Usual Theory of Free Vibrations

The usual theory of free damped vibrations is incorrect, as the dynamic modulus is introduced illegitimately into the classical equation of motion for free vibrating systems. For a description of this theory and its limitations, see *Staverman and Schwarzl* (1), and also *Markovitz* (2). Whereas the introduction of the dynamic modulus is illegitimate it can be performed in several, conflicting ways. It is therefore not surprising that two different formulae to calculate the dynamic modulus from ω_0 and A are current in the literature, the formula with the + being more widely used than that with a - sign.¹⁵⁾

$$\{\bar{M}_1(\omega_0)\}_u = \frac{I\omega_0^2}{g} \left(1 \pm \frac{A^2}{4\pi^2}\right) \quad [41A]$$

$$\{\bar{M}_2(\omega_0)\}_u = \frac{I\omega_0^2}{g} \frac{A}{\pi} \quad [41B]$$

¹⁵⁾ $\{\bar{M}_1(\omega_0)\}_u$ means $\bar{M}_1(\omega_0)$, calculated with the usual theory, so also $\{\bar{M}_2(\omega_0)\}_u$.

The strong analogy of these formulae with [17D] and [17E] is obvious. In the usual theory, the formula with a - sign interprets $M^*(p)$ for $p = -\lambda_0 + i\omega_0$ as $\bar{M}(\omega_0)$. The formula with the + sign does the same thing but it moreover calculates the real part of the modulus incorrectly. Formulae [41A] with the - sign and [41B] therefore are identical with our zero-order approximation [24A]. Consequently, if we use them, it follows from 3.6, that we find \bar{M}_2 too large; \bar{M}_1 may be found smaller, equal or greater than its actual value, this depending on the slope $d\bar{M}_2/d \ln \omega$ of the dynamic loss modulus; see fig. 4.

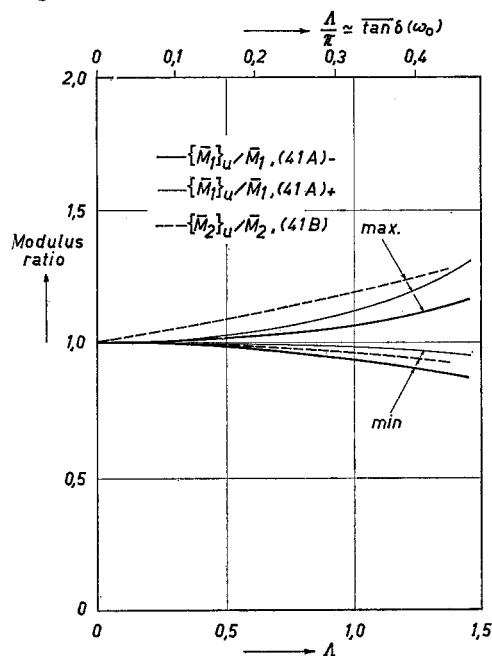


Fig. 6. Bounds for the errors in the usual theory of free vibrations as functions of A

Fig. 6 gives further illustration. Plotted versus A are upper and lower bounds for $\{\bar{M}_1(\omega_0)\}_u/\bar{M}_1(\omega_0)$ if calculated with formula [41A] respectively with the - and + sign, and further for $\{\bar{M}_2(\omega_0)\}_u/\bar{M}_2(\omega_0)$ calculated with [41B].

For the maximum errors at $A = 1$ we find from [35] and [36]:

$$|\delta_{\text{rel}}(41A)_-| \leq 6.5\% \quad |\delta_{\text{rel}}(41A)_+| \leq 11.5\%$$

$$|\delta_{\text{rel}}(41B)| \leq 19\%.$$

Obviously, formula [41A] with the - sign is better than that with the + sign.

Finally, it is surprising that the *usual theory, although incorrect, gives results which in most cases are accurate enough*. For example for organic and inorganic glasses, metals

and some rubbers well above the glass transition, the errors are negligible because $\tan \delta$ and therefore also Λ is much smaller than 1 (see 3.7.). Of course, this is the reason why the usual theory has been used so long without revision.

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Appendix

A Formula for the Higher Derivatives of the Complex Modulus for Materials with a Positive Discrete Relaxation Spectrum

For a material with a positive discrete relaxation spectrum it follows (see formula [5]) for:

$$p = -\lambda + i\omega \quad \lambda \geq 0; \omega > 0 \quad i = \sqrt{-1} \quad [A, 1]$$

that:

$$M^*(p) = M(0) - \sum_{k=1}^N \frac{\alpha_k (1 - \lambda\tau_k)}{(1 - \lambda\tau_k)^2 + (\omega\tau_k)^2} + i \sum_{k=1}^N \frac{\alpha_k \omega\tau_k}{(1 - \lambda\tau_k)^2 + (\omega\tau_k)^2} \quad [A, 2]$$

Consequently:

$$M_2(p) > 0 \quad \text{if } \omega > 0 \quad [A, 3]$$

$$\left| \frac{\omega^n}{n!} \frac{d^n M^*(p)}{dp^n} \right| = \left| \sum_{k=1}^N \frac{\alpha_k (\omega\tau_k)^n}{(1 + p\tau_k)^{n+1}} \right| \leq \sum_{k=1}^N \frac{\alpha_k (\omega\tau_k)^n}{\{(1 - \lambda\tau_k)^2 + \omega^2\tau_k^2\}^{(n+1)/2}} = \sum_{k=1}^N \frac{\alpha_k \omega\tau_k}{(1 - \lambda\tau_k)^2 + \omega^2\tau_k^2} \times \left[\frac{\omega^2\tau_k^2}{(1 - \lambda\tau_k)^2 + \omega^2\tau_k^2} \right]^{\frac{n-1}{2}} \leq M_2(p) \quad [A, 4]$$

Formula [A, 4] contains a number of formulae for the higher derivatives of the dynamic modulus $M^*(i\omega)$. This subject will be discussed extensively in a subsequent article (9).

Summary

A mechanical system consisting of an inert component, attached to a linear viscoelastic spring, is studied theoretically. Basic assumptions about the viscoelastic material are Boltzmann's superposition principle and a positive discrete relaxation spectrum. The equation of motion and its formal solution for free damped vibrations are discussed.

The theory focusses on the determination of the complex dynamic modulus, defined for undamped sinusoidal vibrations, by free damped vibrations. Simple approximation formulae to calculate the dynamic modulus from free vibration data, i. e. eigen frequency and logarithmic decrement, are given; upper limits for the approximation errors could be derived.

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