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Unbounded complex modulus of viscoelastic materials and the Kramers–Kronig relations

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Abstract

The Kramers–Kronig (K–K) dispersion relations developed for the complex modulus of elasticity of solid viscoelastic materials connect the frequency dependences of the dynamic modulus and loss modulus. Whether the boundedness of the complex modulus at high ("infinite") frequency is required, or not, for the applicability of the K–K relations is investigated in this paper. The derivation of the K–K relations developed for the complex modulus is presented by examining the physical background of the relations. It is shown that the K–K relations can be applied even if the complex modulus is unbounded at high frequencies. The fractional derivative Kelvin model is used to demonstrate the application of the K–K relations for a class of unbounded complex modulus.

1. Introduction

The Kramers–Kronig (K–K) relations are named after the two authors who developed equations during the early part of the last century in the electromagnetic theory to connect the real and imaginary parts of the complex refraction index [1] and those of the complex susceptibility [2]. It has since been recognized that the K–K relations, also known as dispersion relations, are of general nature, because they rely on the causality, i.e., that no response can occur before the excitation. The causality is the feature of real physical systems and, therefore, the dispersion relations can be applied in different fields, such as optics, electrodynamics, electrical circuit theory, acoustics, etc., to link the real and imaginary parts of the relevant frequency response function under certain conditions [3].

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The complex modulus of elasticity is widely used to characterize the linear dynamic elastic and damping properties of viscoelastic solid materials in the frequency domain. It is known that the complex modulus can formally be considered as a frequency response function of the material, which is evidently causal. It follows that the K–K relations can be applied to the complex modulus or its inverse, the complex compliance–frequency function [4–9]. The K–K relations are a pair of integral equations, which enable the imaginary part of the complex modulus to be calculated from the frequency dependence of the real part if the latter is known for all frequencies, and vice versa. Several forms of the dispersion relations have been developed, and some equations contain the limit value of complex modulus at high ("infinite") frequency as a result of the assumption that the complex modulus is a bounded function of frequency [5,7,8]. This assumption is closely related to the belief that the complex modulus of viscoelastic materials, especially the polymeric ones, approaches a finite limit at high frequencies and which is referred to as the glassy modulus. In contradiction to this, a number of experiments made on polymeric damping materials at very high frequencies, up to around 10^{12} Hz, do not support this belief, but suggest that the complex modulus may be an unbounded function of frequency [10–14]. In addition, the new fractional derivative models enable the prediction of unbounded high-frequency behaviour of the complex modulus [15–19]. Therefore, the question arises: can the K–K relations be applied or not, if the complex modulus–frequency function is unbounded? The aim of the paper is to answer this question.

The paper consists of three parts. In the first part the general definition of the complex modulus is given and experimental data on the high-frequency behaviour of polymeric materials are discussed and shown. In the second part, the derivation of the K–K relations for the complex modulus is reviewed and it is shown that the bounded high-frequency behaviour is not required for the derivation. In the third part, the application of the K–K relations is demonstrated for a class of unbounded complex modulus through the fractional derivative Kelvin model.

2. The complex modulus of viscoelastic materials

The complex modulus concept has been developed to characterize the linear dynamic properties of solid materials in the frequency range. The complex modulus of elasticity is usually defined for harmonic vibration as the ratio of the complex amplitude of stress to that of strain. Nevertheless, a more general definition of the complex modulus for arbitrary time history is needed to link the frequency domain with the time domain, where the causal behaviour is specified. This general definition can be given by means of the Fourier transform [8,9], and is written here for the shear modulus G by

$$\tilde{G}(\mathrm{j}\omega) = \frac{\tilde{\sigma}(\mathrm{j}\omega)}{\tilde{\varepsilon}(\mathrm{j}\omega)}, \tag{1}$$

where $\mathrm{j} = \sqrt{-1}$ is the imaginary unit, $\omega = 2\pi f$, f is the frequency in Hz, and $\tilde{\sigma}(\mathrm{j}\omega)$ and $\tilde{\varepsilon}(\mathrm{j}\omega)$ are the Fourier transforms $\mathbf{F}$ of the stress and strain-time functions, respectively, defined as

$$\tilde{\sigma}(\mathrm{j}\omega) = \mathbf{F}\sigma(t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \sigma(t)\mathrm{e}^{-\mathrm{j}\omega t}\,\mathrm{d}t, \tag{2}$$

where t is the time. Eq. (1) is general from another point of view as it can be related to any modulus of elasticity (Young's, bulk, etc.) regardless of the fact that the material is either isotropic or anisotropic. It is easy to show that for harmonic vibration Eq. (1) reduces to the ratio of the complex amplitudes of the stress and strain. The components of the complex modulus are used to characterize the dynamic elastic and damping properties of the material:

$$\tilde{G}(\mathrm{j}\omega) = G_d(\omega) + \mathrm{j}G_l(\omega) = G_d(\omega)[1 + \mathrm{j}\,\eta(\omega)], \tag{3}$$

where G_d is the dynamic modulus of elasticity, G_l is the loss modulus and η is the loss factor, where

$$\eta(\omega) = \frac{G_l(\omega)}{G_d(\omega)}. \tag{4}$$

The complex modulus describes the dynamic behaviour of the solid material in the frequency domain. The complete characterization of dynamic behaviour requires knowledge of the variation of complex modulus, in principle, for all frequencies; that is, from 0 Hz up to physically meaningful high frequencies. Much effort has been made to determine experimentally the frequency dependences of complex modulus of viscoelastic materials (especially the polymers), over as wide a frequency range as possible. All experimental data show that the dynamic modulus increases with increasing frequency, and the loss modulus and loss factor pass through at least one peak [20–22]. These experimental observations are in good agreement with the theoretical predictions [8,23]. However, the dynamic behaviour of polymeric materials at high frequencies, well away the loss peak, is not so clear. According to a widely accepted belief, the dynamic modulus approaches a finite limit, referred to as the glassy modulus, while the loss modulus and loss factor approach zero at high frequencies. In contrast, the experimental data on a number of polymeric materials concerning very high frequencies, up to around 10^{12} Hz, do not support this belief [10–14]. These data reveal that the dynamic modulus increases monotonically even at very high frequencies, although the increase can be very weak, and the loss modulus may exhibit the same frequency increase above its peak [10,11]. Accordingly, the loss factor appears to approach a finite limit instead of zero in the high-frequency region [10–14]. Fig. 1 illustrates the

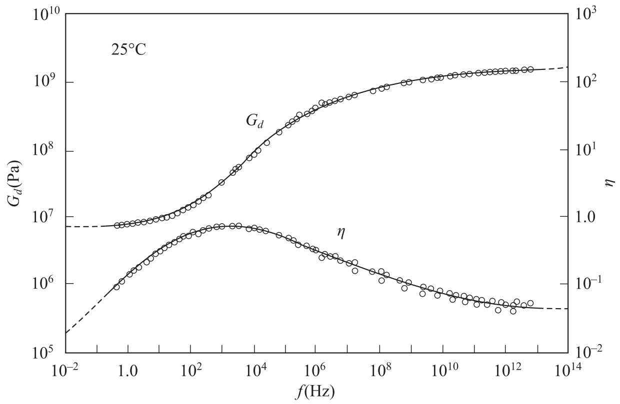

Fig. 1. The frequency variations of dynamic shear properties experienced for a polyurethane damping material [14]. (Some overlapping experimental data have been omitted for the sake of clarity.)

high-frequency behaviour of dynamic shear modulus and loss factor experienced for a polyurethane damping material [14]. It is clear that approximating this behaviour for “infinite” frequency leads to the conclusion that the complex modulus of polymeric materials may, in principle, be an unbounded function of frequency.

3. K–K relations for the complex modulus

Eq. (1) suggests that the complex modulus of elasticity can formally be considered as a frequency response function of the material modelled by a linear, causal system: e.g., if $\tilde{\varepsilon}(\mathrm{j}\omega)$ is the excitation, then $\tilde{\sigma}(\mathrm{j}\omega)$ is the response and vice versa. It is known that the frequency dependences of the real and imaginary parts of a real, and therefore causal system, under certain conditions, are interrelated through the K–K relations [3]. It follows that the K–K relations can also be applied to the complex modulus–frequency function.

The K–K relations for the complex modulus can be derived by different methods [7,8,23]. One of the methods applies the Cauchy’s integral formula to $\tilde{G}(\mathrm{j}\omega)$ [7,8] and uses the theorems of complex functions, and, therefore, the physical background of the derived relations remains somehow hidden. In this paper another method of the derivation is reviewed with the intention to examine the physical background of the K–K relations developed for the complex modulus. The basic idea of this derivation is to show that the relation between the real and imaginary parts of the complex modulus–frequency function is the result of the causality, but the fulfilment of some other conditions defined in the time domain are also required to derive the relations. Consequently, the derivation starts with the specification of material behaviour in the time domain to a proper excitation. Any time function, in principle, could be chosen for the excitation, but for the sake of clear physical interpretation a step function is reasonable to use. In addition, the strain step function of ε_0 magnitude is chosen: namely,

$$\varepsilon(t) = \varepsilon_0 1(t), \tag{5}$$

because then the stress response is the relaxation function, which is well known from both theoretical and experimental studies [20,23]. The relaxation function $\sigma_r(t)$ of a real solid material must have the following properties:

(a) $\sigma_r(t)$ is a real, monotonically decreasing function of time [23],
(b) $\sigma_r(t)$ is causal: i.e., $\sigma_r(t) = 0$ if $t < 0$, and
(c) $\sigma_r(t)$ approaches a finite limit σ_0 for long durations: $\sigma_0 = G_0\varepsilon_0$, where G_0 is the static modulus of elasticity also known as equilibrium modulus.

It is important to note that no restriction has been placed on the initial value $\sigma_r(0_+)$ of the relaxation function. It is usual in the theory of viscoelasticity to assume that $\sigma_r(0_+)$ is finite, although, it is not clear from physical point of view why should it be so. Therefore, at this stage, this question is left open. Fig. 2 illustrates the strain step excitation and the relaxation function for both finite and infinite initial values.

The transformation of the relaxation function of the aforementioned properties into the time domain leads to the K–K relations. As a first step in the derivation, the relation between the complex modulus and the relaxation function is developed. The replacement of the Fourier

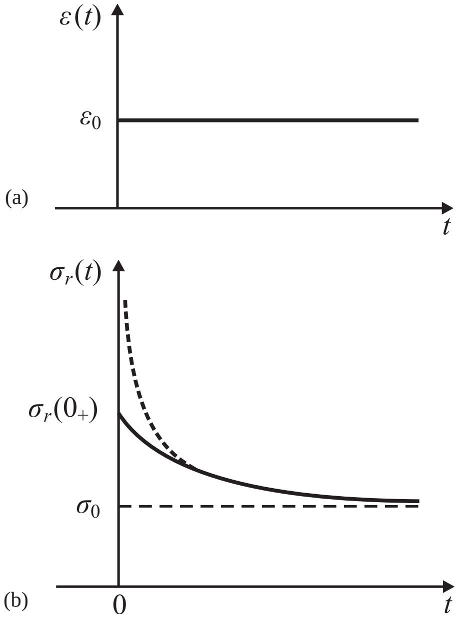

Fig. 2. The strain step excitation (a) and the stress response (b) of a viscoelastic material assuming finite (—) and infinite (- - -) initial values.

transform of the strain step function into Eq. (1) results in

$$\tilde{G}(j\omega) = j\omega \int_{-\infty}^{\infty} g(t) e^{-j\omega t}\, dt, \tag{6}$$

where $g(t)$ is the relaxation modulus defined as

$$g(t) = \frac{\sigma_r(t)}{\varepsilon_0}. \tag{7}$$

A similar procedure (applying the inverse Fourier transform to $\tilde{\sigma}_r(j\omega)$ expressed from Eq. (1)) yields

$$g(t) = \frac{G_0}{2} + \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{\tilde{G}(j\omega)}{j\omega} e^{j\omega t}\, d\omega, \tag{8}$$

where $G_0 = \tilde{G}(0)$. It is clear that $\tilde{G}(0) = G_d(0)$, since $G_l(0) = 0$ (no motion, no energy loss). The separation of the right hand side of Eq. (8) into real and imaginary parts gives

$$\begin{aligned} g(t) = {} & \frac{G_0}{2} + \frac{1}{2\pi} \int_{-\infty}^{\infty} \left(\frac{G_d(\omega)}{\omega} \sin \omega t + \frac{G_l(\omega)}{\omega} \cos \omega t \right) d\omega \\ & - j \frac{1}{2\pi} \int_{-\infty}^{\infty} \left(\frac{G_d(\omega)}{\omega} \cos \omega t - \frac{G_l(\omega)}{\omega} \sin \omega t \right) d\omega. \end{aligned} \tag{9}$$

Using the above equations, the frequency domain consequences of the properties of the relaxation function can be determined. The first property considered is that $g(t)$ is a real, monotonically decreasing function, which results in the convergence of Eq. (6). Then, Eq. (6) can be divided into its real and imaginary parts as

$$G_d(\omega) = \omega \int_{-\infty}^{\infty} g(t) \sin \omega t \, \mathrm{d}t, \tag{10}$$

$$G_l(\omega) = \omega \int_{-\infty}^{\infty} g(t) \cos \omega t \, \mathrm{d}t. \tag{11}$$

From these equations it can be seen that

$$G_d(\omega) = G_d(-\omega) \tag{12a}$$

and

$$G_l(\omega) = -G_l(-\omega). \tag{12b}$$

Using Eqs. (12a) and (12b), the relaxation modulus defined by Eq. (9) can be written as

$$g(t) = \frac{G_0}{2} + \frac{1}{\pi} \int_0^{\infty} \frac{G_d(\omega)}{\omega} \sin \omega t \, \mathrm{d}\omega + \frac{1}{\pi} \int_0^{\infty} \frac{G_l(\omega)}{\omega} \cos \omega t \, \mathrm{d}\omega. \tag{13}$$

The next property considered is the causality: that is $g(t) = 0$ if $t<0$. The application of this property in Eq. (13) leads to two equations namely,

$$g(t) = \frac{2}{\pi} \int_0^{\infty} \frac{G_d(\omega)}{\omega} \sin \omega t \, \mathrm{d}\omega \tag{14}$$

and

$$g(t) = \frac{2}{\pi} \int_0^{\infty} \frac{G_l(\omega)}{\omega} \cos \omega t \, \mathrm{d}\omega + G_0. \tag{15}$$

It follows that the relaxation function can be determined from knowledge of either the dynamic modulus or the loss modulus–frequency function. Consequently, $G_d(\omega)$ and $G_l(\omega)$ are not independent of, but determine, each other. The interrelation between $G_d(\omega)$ and $G_l(\omega)$ can be derived by inserting Eqs. (14) and (15) into Eqs. (11) and (10), respectively. The insertions, and some mathematical manipulations yield

$$G_d(\omega) = \frac{2\omega^2}{\pi} \mathrm{PV} \int_0^{\infty} \frac{G_l(u)/u}{\omega^2 - u^2} \mathrm{d}u + G_0, \tag{16}$$

$$G_l(\omega) = -\frac{2\omega}{\pi} \mathrm{PV} \int_0^{\infty} \frac{G_d(u)}{\omega^2 - u^2} \mathrm{d}u, \tag{17}$$

where the integration variable u has been introduced and PV stands for the principal values of the integrals. Eqs. (16) and (17) are the K–K, or dispersion relations developed for the complex modulus of elasticity of the solid material.

At this stage, it is instructive to summarize the conditions used to derive the K–K relations. The first condition used implicitly is the *linearity*, since all equations quoted above rely on the assumption that the relation between the stress and strain is linear. The second condition is the *convergence of Eqs.* (10) *and* (11), and the third condition is the *causality*. It is emphasized that the convergence of Eqs. (10) and (11), besides the linearity and causality, is absolutely necessary in order to derive the K–K relations. It is clear that these integrals are convergent for monotonically decreasing relaxation functions characterizing the real solid material [23]. Nevertheless, it is important to recognize that Eqs. (10) and (11) can be convergent even if the relaxation function is unbounded, i.e., has singularity at zero time. For example, if the relaxation obeys a simple power function of

$$g(t) \propto t^{-n}, \tag{18}$$

which is singular at $t = 0_+$, then Eqs. (10) and (11) can be convergent, since

$$\int_0^\infty t^{-n} \sin \omega t \, \mathrm{d}t = \frac{\pi \omega^{n-1}}{2\Gamma(n) \sin(n\pi/2)}, \tag{19}$$

$$\int_0^\infty t^{-n} \cos \omega t \, \mathrm{d}t = \frac{\pi \omega^{n-1}}{2\Gamma(n) \cos(n\pi/2)}, \tag{20}$$

where $t > 0$, $0 < n < 1$ and Γ is the gamma function. Therefore, it can be concluded that the K–K relations for the complex modulus can be derived even if the relaxation function is singular at $t = 0_+$. The fact that the relaxation function may be singular has a far-reaching consequence on the possible high-frequency behaviour of the complex modulus, because

$$g(0_+) = \lim_{\omega \to \infty} \tilde{G}(\mathrm{j}\omega). \tag{21}$$

This relation can be proved by the rules of Fourier transform. It follows from the foregoing that the boundedness of the complex modulus is not required to derive the K–K relations. Consequently, the K–K relations may be valid even if the complex modulus is unbounded at high frequencies. It is clear, however that the unboundedness of complex modulus cannot be arbitrary, since the integral of Eqs. (16) and (17) does not exist for any unbounded function. Similarly, the singularity of the relaxation function cannot be arbitrary for the existence of the integral of Eqs. (10) and (11). The development of restrictions on the unboundedness of complex modulus is not subject of this work, but it is important to note that the K–K integrals exist for the unbounded complex modulus characterizing the high-frequency behaviour of real materials. Namely, it is known from experiments [10,11] that both the dynamic modulus and the loss modulus of some polymeric damping materials, at high frequencies, exhibit a weak increase, which obey a power law of form ω^α, where α is small (usually $0 < \alpha < 0.1$ [19]). It is clear that the integrals of the K–K relations exist for this type of high-frequency behaviour.

For the sake of completeness, it is further noted that the unbounded complex modulus raises the problem of the singular relaxation function. Some authors consider this as a disputable question; however, the singular relaxation function appears frequently in the relevant literature, especially in the modern theory of viscoelastic models [15–17]. The singular relaxation function is often refuted by the plausible explanation that the infinite stress in a real material is unacceptable. In contrast to this, a clear physical explanation can be attributed to the singularity of the relaxation function within continuum theory. Namely, the singularity indicates that infinite force would be required to get a perfect strain jump at zero time; consequently such an experiment, in the practice, is impossible to perform with the relevant materials.

4. Demonstration by the fractional Kelvin model

The fractional derivative Kelvin model is the generalization of the well-known Kelvin model consisting of an ideal elastic spring and a viscous dashpot connected in parallel. The fractional derivative Kelvin model, referred to as fractional Kelvin model, has been developed and used successfully to fit experimental data on some solid materials [24], especially damping polymers in the rubbery range [25–28]. It is known that the model is linear and causal [25], and moreover it can be related to the molecular theory of some polymeric materials [26]. Nevertheless, the relaxation function of the fractional Kelvin model is singular at $t = 0_+$ and, thus, the relevant complex modulus is unbounded at "infinite" frequency. It will be shown in this part that the model satisfies the K–K relations.

The constitutive equation for the fractional Kelvin model is [24]:

$$\sigma(t) = G_0\varepsilon(t) + G_0\tau_c^\alpha \frac{\mathrm{d}^\alpha}{\mathrm{d}t^\alpha}\varepsilon(t), \tag{22}$$

where τ_c is the creep time, and $0<\alpha<1$. The αth order derivation of $\varepsilon(t)$ is defined as [26]

$$\frac{\mathrm{d}^\alpha}{\mathrm{d}t^\alpha}\varepsilon(t) = \frac{1}{\Gamma(1-\alpha)}\frac{\mathrm{d}}{\mathrm{d}t}\int_0^t \frac{\varepsilon(\tau)}{(t-\tau)^\alpha}\,\mathrm{d}\tau, \tag{23}$$

in which τ is a dummy variable. The solution of Eq. (22) for a strain step excitation of ε_0 magnitude yields the model relaxation function [24]:

$$\sigma_r(t) = G_0\varepsilon_0\left[1 + \frac{(t/\tau_c)^{-\alpha}}{\Gamma(1-\alpha)}\right]. \tag{24}$$

It is clear that $\sigma_r(t)$ is singular at $t = 0_+$ as illustrated in Fig. 3 for $\alpha = 0.5$.

The complex modulus for the model is easy to derive bearing in mind that [26]

$$\mathbf{F}\frac{\mathrm{d}^\alpha}{\mathrm{d}t^\alpha}\varepsilon(t) = (\mathrm{j}\omega)^\alpha\tilde{\varepsilon}(t). \tag{25}$$

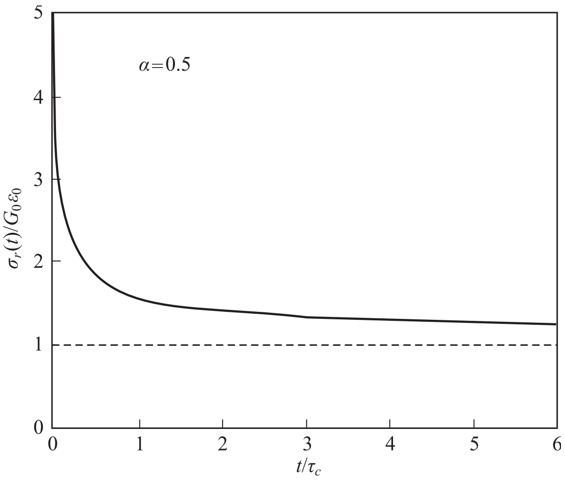

Fig. 3. The relaxation function of the fractional Kelvin model is singular at $t = 0_+$.

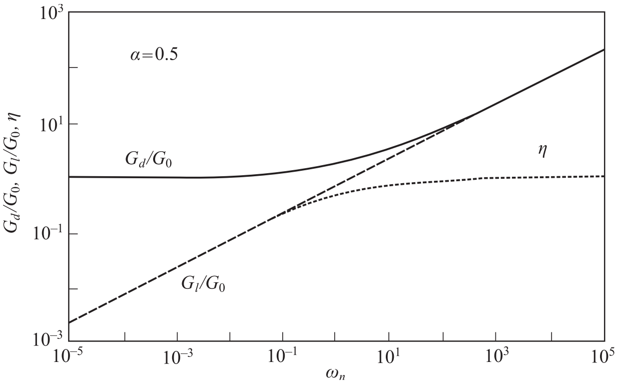

Fig. 4. The frequency variations of the dynamic modulus, loss modulus and loss factor predicted by the fractional Kelvin model.

The model complex modulus is

$$\tilde{G}(\mathrm{j}\omega) = G_0[1 + (\mathrm{j}\omega\tau_c)^{\alpha}], \tag{26}$$

where

$$(\mathrm{j}\omega)^{\alpha} = \cos(\alpha\pi/2)\omega^{\alpha} + \mathrm{j}\sin(\alpha\pi/2)\omega^{\alpha} \tag{27}$$

and so

$$G_d(\omega) = G_0\lfloor 1 + \cos(\alpha\pi/2)\omega_n^{\alpha}\rfloor, \tag{28}$$

$$G_l(\omega) = G_0\sin(\alpha\pi/2)\omega_n^{\alpha}, \tag{29}$$

$$\eta(\omega) = \frac{\sin(\alpha\pi/2)\omega_n^{\alpha}}{1 + \cos(\alpha\pi/2)\omega_n^{\alpha}}, \tag{30}$$

where $\omega_n = \omega\tau_c$ is the normalized frequency. The frequency variations of the components of $\tilde{G}(\mathrm{j}\omega)$ are shown in Fig. 4 for $\alpha = 0.5$ as an example.

It can be seen that $\tilde{G}(\mathrm{j}\omega)$ is unbounded if $\omega \to \infty$ in accordance with the behaviour of the relaxation function at $t = 0_+$. Nevertheless, the fractional Kelvin model obeys the K–K relations, which can be proved by two methods. One method is that the relaxation function is calculated by Eqs. (14) and (15) from $G_d(\omega)$ and $G_l(\omega)$, respectively, developed for the model. It is a simple exercise to show that the calculation of both integrals results in one and the same relaxation function defined by Eq. (24). The other, direct method is to calculate Eqs. (16) and (17) with the dynamic modulus and loss modulus of the model. The analytic calculation is easy to perform, if $\alpha = 0.5$, and then the evaluation of Eqs. (16) and (17) proves that the fractional Kelvin model obeys the K–K relations. These calculations demonstrate that the K–K relations can be applied even if the complex modulus is unbounded at high frequencies.

5. Closing remarks

The role of the high-frequency behaviour of the complex modulus of viscoelastic materials when exploring the validity of the K–K relations is important from the point of view of completeness of the dispersion theory. Nevertheless, it has to be kept in mind that this theory has serious limitations with respect to the physical reality. Apart from the fact that the "infinite" frequency does not exist, the limitation is primarily due to the assumption used throughout the theory that the solid material is considered as a continuum. It is clear that this assumption does not hold true above a certain frequency when the wavelength is comparable with the interatomic distance ($\sim 10^{-10}$ m). This frequency limit is at around 10^{12} Hz in case of a homogeneous solid material provided that the phase velocity is about 10^3 m/s. At higher frequencies, neither the stress field, nor the strain field can be interpreted and, therefore, the complex modulus has no physical meaning. Similarly, the relaxation function has no physical meaning for very short durations, which are smaller than about 10^{-12} s according to the time–frequency equivalency principle. Consequently, the initial value of the relaxation function defined as the response to an ideal strain step function cannot physically be interpreted.

6. Conclusions

Whether the boundedness of the complex modulus at high ("infinite") frequency is required, or not, for the applicability of the K–K relations has been investigated in this paper. The derivation of the K–K relations developed for the complex modulus has been reviewed by examining the physical background of the relations and the conditions of the derivation. It has been shown that, besides the linearity and causality, the convergence of Eqs. (10) and (11) is required to derive the K–K relations for the complex modulus. In addition, it has been shown that these integrals can be convergent even if the relaxation function is unbounded, i.e., has singularity at $t = 0_+$. From this fact it has been concluded that the K–K relations can be applied even if the complex modulus of a viscoelastic material is unbounded at high frequencies. The fractional derivative Kelvin model has been used to demonstrate the application of the K–K relations for a class of unbounded complex modulus.

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