

## ON THE EXISTENCE AND UNIQUENESS OF THE RELAXATION SPECTRUM OF VISCOELASTIC MATERIALS PART II: OTHER EXISTENCE CONDITIONS

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**Summary.** In the first part of this paper the necessary and sufficient condition of the existence and uniqueness of the relaxation spectrum of linear viscoelastic materials is given based on the fundamental for such materials concept of fading memory and the notion of completely monotonic functions. In this paper, using the known conditions for a scalar-valued infinitely differentiable function to be the Laplace transform of an integrable function, other necessary and sufficient conditions which guarantee the existence of the nonnegative relaxation spectrum of viscoelastic material are given. Moreover, we give simple (sufficient) conditions under which the square integrable relaxation spectrum there exists. The above is of an important role for the synthesis of the relaxation spectrum identification algorithms. All the conditions refer to the Boltzmann relaxation modulus, which is accessible in experiment. Three illustrative examples are given.

**Keywords:** viscoelasticity, relaxation modulus, relaxation spectrum, existence and uniqueness

### INTRODUCTION

In rheological literature, it is generally assumed that the linear relaxation modulus  $G(t)$  has the following representation [Ter Haar 1950, Christensen 1971, Ferry 1980]:

$$G(t) = \int_0^{\infty} H(\nu) e^{-\nu t} d\nu, \quad (1)$$

where:  $H(\nu)$ ,  $\nu \geq 0$ , is the spectrum of relaxation frequencies. The modulus  $G(t)$  can be also represented in equivalent form as a function of the spectrum of relaxation times  $N(\tau)$ ,  $\tau \geq 0$  as follows:

$$G(t) = \int_0^{\infty} N(\tau) e^{-t/\tau} d\tau. \quad (2)$$

In the first part of this paper necessary and sufficient condition of the existence and uniqueness of nonnegative relaxation spectrum of linear viscoelastic materials is given based on the notion of completely monotonic Boltzmann relaxation modulus [Anderssen and Loy 2002]. In this paper two next necessary and sufficient conditions for the existence of nonnegative spectrum as well as sufficient conditions for the existence of square integrable spectrum are given. The last property is

of fundamental role for the known and still constructed algorithms for relaxation spectrum identification [Stankiewicz 2003, 2005, 2007, 2009]. All the conditions refer to the relaxation modulus, which is accessible in experiment.

## OTHER EXISTENCE CONDITIONS

Another known in the literature necessary and sufficient conditions for a scalar-valued infinitely differentiable function on  $(0, \infty)$  to be the Laplace transform of a function, can be used to obtain the next relaxation spectrum existence conditions. The proofs are not necessary since the conditions follow immediately from the known results.

On the basis of the Post-Widder conditions [Widder 1946, 1971] the following theorem can be stated.

**Theorem 1.** *Nonnegative integrable relaxation frequencies spectrum  $H(\nu)$  defined by the eq. (1) there exists iff the linear relaxation modulus  $G(t)$  is a function of class  $C^\infty(0, \infty)$  and the conditions*

$$\int_0^\infty |H_P^n(\nu)| d\nu < \infty, \quad n = 1, 2, \dots, \quad (3)$$

$$\lim_{n, k \rightarrow \infty} \int_0^\infty |H_P^n(\nu) - H_P^k(\nu)| d\nu = 0, \quad (4)$$

are satisfied, where:

$$H_P^n(\nu) = \frac{(-1)^n}{n!} \left( \frac{n}{\nu} \right)^{n+1} G^{(n)}\left(\frac{n}{\nu}\right), \quad (5)$$

is so called  $n$ -th Post-Widder approximation of the inverse Laplace transform of  $G(t)$ .

As it was in the case of theorem 1 in the first part of the paper applying theorem 1 to relaxation modulus from example 3 in the first part of the paper we can easily conclude that the integrable relaxation spectrum of this viscoelastic material these exists.

**Example 1.** Let us consider again the relaxation modulus  $G(t) = t^{-1}$ . On the basis of formula  $d^n G(t)/dt^n = (-1)^n n! t^{n-1}$  the  $n$ -th Post-Widder approximation is as follows:

$$H_P^n(\nu) = \frac{(-1)^n}{n!} \left( \frac{n}{\nu} \right)^{n+1} (-1)^n n! \left( \frac{n}{\nu} \right)^{(n+1)} - 1.$$

Thus, for every integer  $n \geq 1$  the condition (3) is not satisfied. Therefore, on the basis on theorem 2 nonnegative integrable relaxation spectrum of this material does not exist. For the modulus  $G(t) = t^{-1}$  the kernel of the eq. (1) is singular Hilbert's kernel  $(t - \tau)^{-1}$ , which was studied by Boltzmann even in 1876 year [Boltzmann 1876].

The existence of nonnegative integrable relaxation spectrum has been considered above. We now wish to show that such a spectrum may be unbounded.

**Example 2.** Consider again the viscoelastic relaxation modulus  $G(t) = 1/(t+a)^p$  from example 2 in the first part of the paper, where the parameters  $a > 0$ . Suppose  $p = 1/2$ . Since  $\Gamma(1/2) = \sqrt{\pi}$  on the basis of eq. (9) in the first part of the paper, the corresponding relaxation spectrum  $H(\nu) = e^{-a\nu} / \sqrt{\pi\nu}$ ,  $H(\nu)$  is plotted in Figure 1. The spectrum is integrable, however it is unbounded and singular for  $\nu = 0$ .

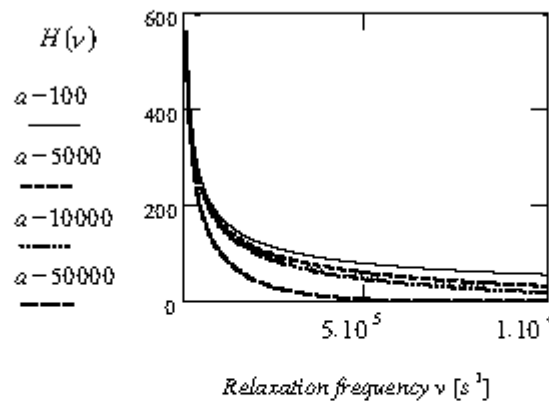


Fig. 1. The relaxation spectrum  $H(\nu) = e^{-\alpha\nu} / \sqrt{\pi\nu}$  for a few parameters  $\alpha$

In the next theorem the necessary and sufficient conditions for the existence of bounded relaxation spectrum are formulated on the basis of Arendt's conditions [1987].

**Theorem 2.** *Nonnegative bounded relaxation frequencies spectrum  $H(\nu)$  defined by the eq. (1) there exists iff linear relaxation modulus  $G(t)$  is a function of class  $C^\infty(0, \infty)$  and for some constant  $M > 0$  the Widder's condition [Widder 1946]*

$$\left| \frac{d^n G(t)}{dt^n} \right| \leq \frac{Mn!}{t^{n+1}} \text{ for } n \geq 0 \text{ and } t > 0, \quad (6)$$

is satisfied.

Using the condition (6) it is easy to check if the relaxation spectrum of the considered material there exists and is bounded in the case when the corresponding modulus  $G(t)$  is infinitely differentiable function. The effectiveness of this criterion is demonstrated by the next example.

**Example 3.** Let us consider again the exponential relaxation modulus described by infinite Dirichlet-Prony series [Gerlach and Matzenmiller 2005]:

$$G(t) = \sum_{j=1}^{\infty} E_j e^{-\nu_j t} + E_{\infty}. \quad (7)$$

According with the remark 4 in the first part of the paper, under the assumption  $E_j \geq 0$ ,  $\nu_j \geq 0$  and  $E_{\infty} \geq 0$ , the modulus (7) is completely monotonic function. By (7)  $G(0+) = \sum_{j=1}^{\infty} E_j + E_{\infty}$  thus on the basis of theorem 1 in the first part of the paper there exists integrable relaxation spectrum  $H(\nu)$  defined by the eq. (1) and in view of the assertion 1 from the same paper the spectrum is unique. We now check if for the relaxation modulus  $G(t)$  (7) the Widder's condition (6), that is:

$$\left| \frac{d^n G(t)}{dt^n} \right| t^{n+1} \leq M n! \text{ for } n \geq 0 \text{ and } t > 0, \quad (8)$$

is satisfied. It is proved in Appendix A that for any  $k \geq 1$ , such that  $\nu_k \neq 0$  and for any finite number  $M$  there exists  $\hat{n}$  such that for any  $n \geq \hat{n}$  and  $t = \hat{t}_k(n+1)/\nu_k$  the following estimation:

$$\left| \frac{d^n G(\hat{t}_k)}{dt^n} \right| \hat{t}_k^{n+1} > \frac{E_k}{\nu_k e} \sqrt{n+1} n! \leq M n!, \quad (9)$$

holds, what contrary the inequality (8). This leads to the final conclusion, that on the basis of the theorem 2 the relaxation spectrum corresponding to the relaxation modulus (7) is not bounded. It is

not difficult to check, using the selectivity property of Dirac delta function  $\delta(\nu)$  and the definition formula (1), that the relaxation spectrum takes the form:

$$H(\nu) = \sum_{j=1}^{\infty} E_j \delta(\nu - \nu_j) + E_{\infty} \delta(\nu), \quad (10)$$

where  $\delta(\nu - \nu_j)$  is the Dirac delta function displaced to the point  $\nu = \nu_j$ . Discrete relaxation spectrum (10) is depicted symbolically on Figure 2; here Dirac delta functions are replaced by Kronecker delta functions of one unit value in the singular points  $\nu = \nu_j$  of  $\delta(\nu - \nu_j)$ .

A number of different algorithms have been proposed during the last few years for identification of the relaxation frequencies and relaxation times spectra [Stankiewicz 2003, 2005, 2009], for other references and classification see [Stankiewicz 2007]. All the methods are based on the expansion of the unknown spectrum in finite series of properly selected basic functions of the space  $L^2(0, \infty)$ . Applying theorem 1 in the first part of the paper and theorem 1 given above we can assert the conditions under which the relaxation spectrum  $H(\nu)$  is not only integrable but also square integrable on  $(0, \infty)$ . They are stated in our next theorem.

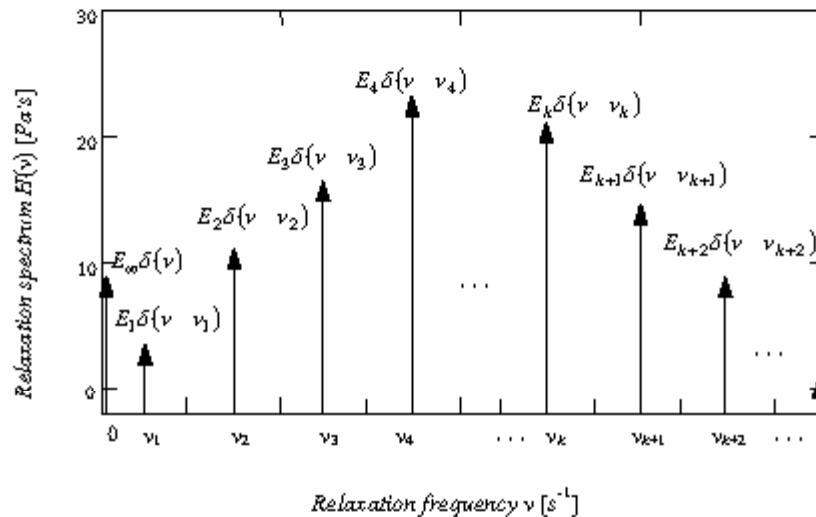


Fig. 2. Schematic representation of the relaxation spectrum  $H(\nu)$  (10)

**Theorem 3.** *If the linear relaxation modulus  $G(t)$  is completely monotonic function with fading memory,  $G(0+) < \infty$  and the Widder condition (6) holds, then there exists a unique nonnegative relaxation spectrum  $H(\nu) \in L^2(0, \infty)$  defined by the eq. (1).*

**Proof.** Existence of the nonnegative relaxation spectrum  $H(\nu)$  follows immediately from theorem 1 in the first part of the paper, applying assertion 1 in the first part of the paper implies the uniqueness. Since on the basis of theorem 2 spectrum  $H(\nu)$  is bounded and by virtue of theorem 1 in the first part of the paper it is also integrable, using the inequality:

$$\int_0^{\infty} H^2(\nu) d\nu \leq \tilde{M} \int_0^{\infty} H(\nu) d\nu,$$

where:  $\tilde{M} = \sup_{\nu > 0} H(\nu)$ , we obtain immediately the square integrability of  $H(\nu)$ . The proof is now completed.

Relaxation spectrum  $H(\nu) = e^{-\alpha} / \sqrt{\pi \nu}$  (see Example 2) is an example of integrable spectrum, which is not square integrable on  $(0, \infty)$ . Clearly, the Euler's integral of second kind  $\Gamma(z) = \int_0^\infty \nu^{z-1} e^{-\nu} d\nu$  is not convergent for any  $z \leq 0$ .

#### EXISTENCE OF THE RELAXATION TIMES SPECTRUM

On the basis of definition equations (1) and (2) the spectra of relaxation times  $N(\tau)$  and relaxation frequencies  $H(\nu)$  are related by:

$$N(\tau) = H(1/\tau)/\tau^2 \quad \text{and} \quad H(\nu) = N(1/\nu)/\nu^2. \quad (11)$$

It is easy to establish, using the definitions (1) and (2), that nonnegative integrable relaxation frequencies spectrum there exists iff there exists nonnegative spectrum of relaxation times. The existence conditions of theorem 1 and theorem 1 in the first part of the paper refer to the relaxation modulus and not to the relaxation spectrum, therefore they are also valid for the spectrum of relaxation times. The next theorem provides the necessary and sufficient condition for  $N(\tau) \in L^2(0, \infty)$ .

**Theorem 4.** Suppose that the linear relaxation modulus  $G(t)$  is completely monotonic function and  $G(0+) < \infty$ . The relaxation times spectrum  $N(\tau) \in L^2(0, \infty)$  iff the function  $\nu H(\nu) \in L^2(0, \infty)$ . The spectrum of relaxation frequencies  $H(\nu) \in L^2(0, \infty)$  iff the function  $\tau N(\tau) \in L^2(0, \infty)$ .

**Proof.** Under the taken assumptions it follows from theorem 1 in the first part of the paper that the spectra of relaxation times  $N(\tau)$  and frequencies  $H(\nu)$  there exist. The spectra are related by eqs. (11). By simple change of variables  $\tau = 1/\nu$  in the left-hand side of (11), we have the sequence of equalities:

$$\int_0^\infty N(\tau)^2 d\tau = \int_0^\infty H(1/\tau)^2 / \tau^4 d\tau = \int_0^\infty H(\nu)^2 \nu^4 / \nu^2 d\nu = \int_0^\infty H(\nu)^2 \nu^2 d\nu.$$

Therefore,  $N(\tau) \in L^2(0, \infty)$  iff  $\nu H(\nu) \in L^2(0, \infty)$ . To complete the proof it is enough to note that, using the second equation of (11), that  $H(\nu) \in L^2(0, \infty)$  iff  $\tau N(\tau) \in L^2(0, \infty)$ .

#### FINAL REMARK

However, the relaxation spectrum is not measurable directly. Therefore it must be determined from the appropriate response functions, measured either in time or frequency-domain. The literature concerned with different algorithms for the relaxation spectrum computation using the data both from a small-amplitude oscillatory shear experiment, [Honerkamp and Weese 1989, Elster et al. 1991, Brabec et al. 1997] for example, as well as from relaxation modulus and creep compliance data [Yamamoto and Masuda 1971, Fujihara et al. 1995, Zi and Bažant 2002, Stankiewicz 2003, 2005, 2009] and papers cited therein, is quite extensive now. In almost all known methods the relaxation spectrum model  $H_M(\nu)$  is selected in such a way that the respective model of relaxation modulus:

$$G_M(t) = \int_0^\infty H_M(\nu) e^{-\nu t} d\nu, \quad (12)$$

ensures the best fit to the measurement results. A relation between the true relaxation spectrum  $H(\nu)$  defined by (1), and its model  $H_m(\nu)$  provides the next assertion, in which the necessary condition is obvious in view of (1) and (12), while the sufficient condition follows from the Laplace transform invertibility.

**Assertion 1.** *If the true relaxation spectrum there exists, then  $H(\nu) = H_m(\nu)$  iff  $G(t) = G_m(t)$ , where  $G_m(t)$  is the relaxation modulus model.*

Of course, if the true relaxation modulus  $G(t)$  of viscoelastic material under study is not completely monotonic function, according to the remark 5 in the first part of the paper the true relaxation spectrum of this material does not exist. However, we may still describe the mechanical properties of the material using the relaxation spectrum model  $H_m(\nu)$ , taking into account that it is only the relaxation spectrum of the model (12), which is only an approximation of the reality.

## APPENDIX A

**Derivation of inequality (9).** Let  $k \geq 1$ ,  $\nu_k \neq 0$  and  $n \geq 1$ . On the basis of equation (7):

$$\left| \frac{d^n G(t)}{dt^n} \right| t^{n+1} = \sum_{j=1}^{\infty} (\nu_j)^n E_j e^{-\nu_j t^{n+1}}.$$

Let  $t = \hat{t}_k = (n+1)/\nu_k$ . We have the following estimation:

$$\left| \frac{d^n G(\hat{t}_k)}{dt^n} \right| \hat{t}_k^{n+1} = \sum_{j=1}^{\infty} (\nu_j)^n E_j e^{-\nu_j \hat{t}_k^{n+1}} \geq (\nu_k)^n E_k e^{-\nu_k \hat{t}_k^{n+1}} = \alpha_k \quad (\text{A.1})$$

where:

$$\alpha_k = \frac{E_k}{\nu_k} \sqrt{n+1} e^{-(n+1)} (n+1)^{n+1/2}.$$

Monotonically decreasing sequence  $b_n = [(n+1)/n]^{n+1/2}$  tends to the number  $e$ , whence for any  $n \geq 1$  we have  $(n+1)^{n+1/2} > e n^{n+1/2}$ , what implies the estimation:

$$\alpha_k = \frac{E_k}{\nu_k} \sqrt{n+1} e^{-(n+1)} (n+1)^{n+1/2} > \frac{E_k}{\nu_k} \sqrt{n+1} e^{-n} (n)^{n+1/2},$$

and hence, using again the same convergence property of the sequence  $b_n$  we obtain

$$\alpha_k = \frac{E_k}{\nu_k} \sqrt{n+1} e^{-n} (n)^{n+1/2} = \frac{E_k}{\nu_k} \sqrt{n+1} e^{-n} n (n)^{n-1/2} > \frac{E_k}{\nu_k} \sqrt{n+1} e^{-n+1} n (n)^{n-1/2}.$$

This implies for any  $1 \leq j \leq n-1$  the inequality:

$$\alpha_k = \frac{E_k}{\nu_k} \sqrt{n+1} e^{-n+1} \prod_{i=0}^j (n-i)(n-j)^{n-j-1/2},$$

whence, for  $j = n-1$  we see that:

$$\alpha_k = \frac{E_k}{\nu_k} \sqrt{n+1} e^{-1} n! = \frac{E_k}{\nu_k e} \sqrt{n+1} n!. \quad (\text{A.2})$$

Combining (A.2) and (A.1) we obtain:

$$\left| \frac{d^n G(\hat{t}_k)}{dt^n} \right| \hat{t}_k^{n+1} > \frac{E_k}{\nu_k e} \sqrt{n+1} n!,$$

that is, for any finite number  $M$  there exists  $\hat{n}$ , such that for any  $n \geq \hat{n}$  and  $t = \hat{t}_1$  the inequality (9) holds. And the proof is completed.

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O ISTNIENIU I JEDNOZNACZNOŚCI SPEKTRUM RELAKSACJI  
MATERIAŁÓW LEPKOSPĘŻYSTYCH  
CZĘŚĆ II: INNE WARUNKI ISTNIENIA SPEKTRUM RELAKSACJI

**Streszczenie.** W pierwszej części pracy wychodząc z fundamentalnego dla materiałów lepkospężystych pojęcia zanikającej pamięci i wykorzystując własności funkcji w pełni monotonicznych sformułowano podstawowy warunek konieczny i dostateczny istnienia i jednoznaczności spektrum relaksacji materiałów liniowo lepkospężystych. W tej pracy, wykorzystując znane warunki istnienia odwrotnej transformaty Laplace'a z funkcji rzeczywistej, podano inne warunki konieczne i dostateczne istnienia nieujemnego spektrum relaksacji. Podano także warunki (wysarczające) istnienia spektrum relaksacji całkowitego z kwadratem; warunki te mają istotne znaczenie dla konstrukcji algorytmów identyfikacji spektrum relaksacji. Wszystkie warunki odnoszą się do modułu relaksacji Boltzmanna, czyli wielkości dostępnej pomiarowo. Rozważania zilustrowano trzema przykładami.

**Słowa kluczowe:** lepkospężystość, moduł relaksacji, spektrum relaksacji, istnienie i jednoznaczność