MACROSCOPIC THEORY OF DIELECTRIC RELAXATION AND ITS APPLICATION TO THE ANALYSIS OF KNOWN PHENOMENOLOGICAL MODELS

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It is shown that in the case of linear and uniform dielectric, the Krönig-Kramers relations for dielectric susceptibility are a purely mathematical consequence of the causality principle (§ 2). Treating the above relations as an equation for $\varepsilon(\omega)$, under rather weak assumptions, a general solution of this equation is found (§ 5). The expressions of Cole-Cole (§ 4) and Cole-Davidson (§ 3) were confronted with the Krönig-Kramers relations and it turned out that the both agree with these relations, and that the expression of Cole-Davidson contradicts microscopic interpretation of relaxation function. Putting $\varepsilon(\omega)$ as an analytical function of ω, a simple form of $\varepsilon(\omega)$ is found, having all the correct features of the Cole-Cole expression with exponential but complex relaxation function (§ 6). A more sophisticated model is also discussed, following from the general form of $\varepsilon(\omega)$, which under additional assumptions may probably serve for the microscopic interpretation of "skewed-arc" Cole-Cole diagrams (§ 7).

1. Introduction

In the majority of experimental works devoted to the investigation of dielectric relaxation in liquids, the interpretation of results is based on models of Debye, Cole-Cole or Cole-Davidson (a full list of literature concerning this subject may be found *e.g.* in the monographic work Hill, Vaughan, Davies 1969).

Although in the model of Debye the physical sense of the parameters used is clear, in the remaining expressions the physical sense of the additional parameter β remains problematic. Usually this parameter is connected to continuous distributions of relaxation time calculated by approximate methods from macroscopic theory or from microscopic models (see *e.g.* Glarum 1960a, 1960b), which does not guarantee the validity of such continuous distributions. Moreover, so far the proof is not found in the literature that

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the complex dielectric susceptibilities $\varepsilon(\omega)$ postulated by Cole-Cole or Cole-Davidson, fulfil the Krönig-Kramers relations.

Hence the aim of this work is to draw as many conclusions as possible from the macroscopic theory and to confront it with expressions discussed here.

2. Fundamental macroscopic equation

Let us consider a linear dielectric of no spatial dispersion. It is assumed to be isotropic, in order to simplify the writing of the formulae, and its non-dipole polarization is determined by the dielectric susceptibility ε_{∞}. Then the value of the displacement vector $\mathscr{D}(t)$ in the field $\mathscr{E}(t)$ may be written as follows

$$\mathscr{D}(t) = \varepsilon_{\infty}\,\mathscr{E}(t) + \int\limits_{-\infty}^{t} \Phi(t-t')\,\mathscr{E}(t')dt', \tag{2.1}$$

where $\Phi(t)$ is the derivative of the so-called relaxation function. However, for simplicity, in the following we shall call the function $\Phi(t)$ as well as the function $\Phi_N(t)$ the relaxation functions. With the aid of the Heviside function $\mathscr{H}(t)$, Dirac delta $\delta(t)$ and introducing a new relaxation function

$$\tilde{\Phi}(t) = \varepsilon_{\infty}\delta(t) + \mathscr{H}(t)\Phi(t)$$

equation (2.1) may be written in the form of a convolution

$$\mathscr{D}(t) = \int\limits_{-\infty}^{+\infty} \tilde{\Phi}(t-t')\,\mathscr{E}(t')dt' = (\tilde{\Phi} * \mathscr{E})\,(t). \tag{2.2}$$

From now on we shall treat Eq. (2.2) as an equation in the sense of a distribution which, as we shall soon see, is very convenient from the point of view of calculations. Let us now calculate the Fourier transform $\mathscr{F}$ of (2.2).

$$\mathscr{F}\mathscr{D} = \mathscr{F}(\tilde{\Phi} * \mathscr{E}) = \mathscr{F}\tilde{\Phi}\mathscr{F}\mathscr{E}. \tag{2.3}$$

As is easily seen $\mathscr{F}(\mathscr{E})$ does not exist in the functional sense for periodical fields which are of interest to us, however, it is well determined in the sense of a distribution. We now propose a definition

$$\varepsilon(\omega) \stackrel{\text{df}}{=} \frac{\partial(\mathscr{F}\mathscr{D})}{\partial(\mathscr{F}\mathscr{E})}. \tag{2.4}$$

On its basis

$$\varepsilon(\omega) = \mathscr{F}\tilde{\Phi} = \varepsilon_{\infty} + \int\limits_{0}^{\infty} \Phi(t)e^{-i\omega t}dt \tag{2.5}$$

which is a well known formula for the complex dielectric susceptibility. To simplify notation instead of $\varepsilon(\omega)$ and $\Phi(t)$ we now introduce

$$\eta(\omega) = \frac{\varepsilon(\omega) - \varepsilon_{\infty}}{\varepsilon_s - \varepsilon_{\infty}} \quad \text{and} \quad \Phi_N(t) = \frac{\Phi(t)}{\varepsilon_s - \varepsilon_{\infty}} \tag{2.5'}$$

where ε_s is the static dielectric susceptibility. Thus, instead of (2.5) one may write

$$\eta(\omega) = \int_0^\infty \Phi_N(t)e^{-i\omega t}dt, \tag{2.6}$$

from which

$$\eta'(\omega) = \text{Re}\,\eta(\omega) = \sqrt{\frac{\pi}{2}}\{\mathscr{F}_c[\text{Re}\,\Phi_N] + \mathscr{F}_s[\text{Im}\,\Phi_N]\}, \tag{2.7}$$

$$\eta''(\omega) = -\text{Im}\,\eta(\omega) = \sqrt{\frac{\pi}{2}}\{-\mathscr{F}_c[\text{Im}\,\Phi_N] + \mathscr{F}_s[\text{Re}\,\Phi_N]\}, \tag{2.8}$$

where $\mathscr{F}_c$ and $\mathscr{F}_s$ are Fourier transforms of cosine and sine respectively. On the other hand, the Hilbert transform has the property

$$\mathscr{H}\mathscr{F}_c = -\mathscr{F}_s \quad \text{and} \quad \mathscr{H}\mathscr{F}_s = \mathscr{F}_c. \tag{2.9}$$

With the use of (2.7), (2.8) and (2.9), a simple calculation gives the Krönig-Kramers relations which in the complex form become

$$\mathscr{H}(\eta) = -i\eta. \tag{2.10}$$

From the above consideration one sees that these relations are a purely mathematical consequence of accepting the causality principle in the shape (2.1). Let us further write formulae determining $\Phi_N(t)$ by $\eta(\omega)$. This may be done easily using (2.6) and the properties of Fourtier transforms $\mathscr{F}_c$ and $\mathscr{F}_s$. As a result we obtain

$$\Phi_N(t) = \sqrt{\frac{2}{\pi}}\,\mathscr{F}_s\left[\frac{\eta(-\omega)-\eta(\omega)}{2i}\right] = \sqrt{\frac{2}{\pi}}\,\mathscr{F}_c\left[\frac{\eta(-\omega)+\eta(\omega)}{2}\right]. \tag{2.11}$$

Apart from this, on the basis of (2.6), $\Phi_N(t)$ must be normalized in such a way that

$$\int_0^\infty \Phi_N(t)dt = 1. \tag{2.12}$$

Let us notice further that the existence of $\Phi_N(t)$ fulfilling (2.12) ensures fulfilling by $\eta(\omega)$ the Krönig-Kramers relations[1]. Let us also draw the formula allowing one to calculate the macroscopic relaxation time t_R. This time is, as it is well known, the time after which the polarization amplitude after removing the constant field, falls e^{-1} times. Thus if the field $\mathscr{E}$ is switched off in the moment $t = 0$, then according to (2.1) we have

$$\frac{D(t)}{(\varepsilon_s - \varepsilon_\infty)\mathscr{E}} = \int_{-\infty}^0 \Phi_N(t-t')dt'. \tag{2.13}$$

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[1] Strictly speaking, any function for which $\mathscr{F}_c$ and $\mathscr{F}_s$ exist, leads to Krönig-Kramers relations, and is hence potentially a "relaxation function". Thus tables of $\mathscr{F}_c$ and $\mathscr{F}_s$ provide a large number of formal solutions of (2.10). Choosing solutions appropriate for discussion is however difficult.

Thus the formula seeked is of the form

$$\int_{-\infty}^{0} \Phi_N(t_R - t')dt' = e^{-1}. \tag{2.14}$$

3. Study of the Cole-Davidson expression

For the C-D expression

$$\eta(\omega) = \frac{1}{(1 + i\omega\tau)^\beta}, \quad 0 < \beta \leqslant 1, \tag{3.1}$$

which after insertion into (2.11) and referring to tables of Gradshteyn and Rizhik (1962) gives the relaxation function

$$\Phi_N(t) = \frac{1}{\Gamma(\beta)\tau} \frac{e^{-\frac{t}{\tau}}}{\left(\dfrac{t}{\tau}\right)^{1-\beta}} \tag{3.2}$$

fulfilling also the normalization condition (2.12). It is thus clearly seen that we are dealing here with the case of an non-exponential behaviour of relaxation and not with a conti-
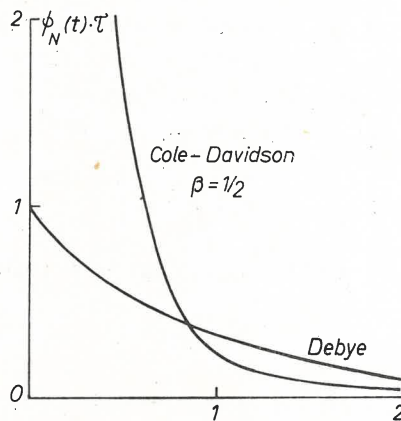

Fig. 1. $\Phi_N(t)$ curves for Cole-Davidson and Debye models

nuous spectrum of relaxation times as used to be accepted up to now. The behaviour of this relaxation function as compared to the exponential relaxation function is shown schematically in Fig. 1. The equation for t_R drawn from (2.14) is of the form

$$\Gamma\left(\beta, \frac{t_R}{\tau}\right) = \Gamma(\beta)e^{-1} \tag{3.3}$$

and, of curse, if we proceed to the Debye model, i.e. put $\beta = 1$, then $t_R = \tau$.

In this case it is also possible to obtain a relaxation function

$$\Psi_N(t) \stackrel{\text{df}}{=} -\int \Phi_N(t)dt \tag{3.4}$$

which from (3.2) is equal

$$\Psi_N(t) = \frac{1}{\Gamma(\beta)\tau}\left(\frac{t}{\tau}\right)^\beta e^{-\frac{t}{\tau}} \sum_{k=0}^{\infty} \frac{\left(\frac{t}{\tau}\right)^k}{\beta(\beta+1)\dots(\beta+k)}. \tag{3.5}$$

The relaxation function (3.4) according to its microscopic interpretation (Glarum 1960a) should hold conditions

$$\Psi_N(0) = 1, \qquad \Psi_N(\infty) = 0. \tag{3.6}$$

Now it is easy to see that (3.5) does not hold the first part of Eq. (3.6).

4. Study of the Cole-Cole expression

For the C-C expression

$$\eta(\omega) = \frac{1}{1+(i\omega\tau)^\beta}, \qquad 0 < \beta \leqslant 1. \tag{4.1}$$

Unfortunately, it was not possible to give the explicit form of $\Phi_N(t)$. On the other hand, one may easily obtain from (2.11)

$$\Phi_N(t) = \frac{2}{\pi} \sin\frac{\pi\beta}{2} \int_0^\infty \frac{(\omega\tau)^\beta \sin\omega t}{1+2(\omega\tau)^\beta \cos\frac{\pi\beta}{2}+(\omega\tau)^{2\beta}}\, d\omega =$$

$$= -\frac{2}{\pi} \sin\frac{\pi\beta}{2} \frac{d}{dt} \int_0^\infty \frac{(\omega\tau)^{\beta-1}\cos\omega t}{1+2(\omega\tau)^\beta \cos\frac{\pi\beta}{2}+(\omega\tau)^{2\beta}}\, d(\omega\tau). \tag{4.2}$$

Let us assume that the first of the integrals written down, exists, otherwise there would be no problem to discuss. The other integral exists, because the integral exists

$$\int_0^\infty \frac{(\omega\tau)^{\beta-1}d(\omega\tau)}{1+2(\omega\tau)^\beta \cos\frac{\pi\beta}{2}+(\omega\tau)^{2\beta}} = \frac{1}{\beta}\sin\frac{\pi\beta}{2}\int_{ctg\frac{\pi\beta}{2}}^\infty \frac{dy}{1+y^2} = \frac{\pi}{2\beta}\sin\frac{\pi\beta}{2}. \tag{4.3}$$

Now we have

$$\int_0^\infty \Phi_N(t)dt = -\frac{2}{\pi}\sin\frac{\pi\beta}{2}\left[\int_0^\infty \frac{(\omega\tau)^{\beta-1}\cos\omega t}{1+2(\omega\tau)^\beta \cos\frac{\pi\beta}{2}+(\omega\tau)^{2\beta}}\, d(\omega\tau)\right]\Bigg|_0^\infty = 1, \tag{4.4}$$

as intuitively, the function in the square brackets vanishes in the upper limit. As the obtained

results is very important, a detailed proof of this intuitive argument is given in the Appendix.

Finally, for $\beta \neq 1$, $\Phi_N(t)$ is normalized and moreover in this case $\Psi_N(t)$, which one may have simple from (4.2), agrees with the conditions (3.6).

5. Macroscopic model of relaxation basing on the causality principle with $\varepsilon(\omega)$ as an analytic function

The basic equation which gives the possibility of determining the shape of the $\eta(\omega)$ function, is equation (2.10). In order to exploit this formula we make three assumptions concerning $\eta(\omega)$:

(1) $\eta(\omega)$ is an analytic function of a complex variable ω,
(2) $\eta(\omega)/\omega$ vanishes at infinity faster than ω^{-1},
(3) $\eta(\omega)$ has no poles on the real axis nor first order poles in the lower part of the Gauss plane.
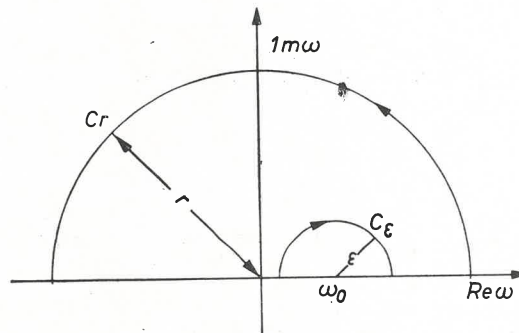
Fig. 2. Integration paths in equations (5.1)

The first part of the last assumption is essential in order for the relaxation function $\Phi_N(t)$ to tend to 0 when $t \to \infty$. Now one may already see that

$$\mathscr{H}[\eta](\omega_0) = \text{P.V.} \int_{-\infty}^{\infty} \frac{\eta(\omega)}{\omega - \omega_0} \, d\omega =$$

$$= -\frac{1}{\pi} \lim_{\varepsilon \to 0} \int_{C_\varepsilon} \frac{\eta(\omega)d\omega}{\omega - \omega_0} - \frac{1}{\pi} \lim_{r \to \infty} \int_{C_r} \frac{\eta(\omega)d\omega}{\omega - \omega_0} + 2i \sum_{\omega_k \in Z_+} \text{res} \frac{\eta(\omega_k)}{\omega_k - \omega_0} =$$

$$= i\eta(\omega_0) + 2i \sum_{\omega_k \in Z_+} \text{res} \frac{\eta(\omega_k)}{\omega_k - \omega_0}, \tag{5.1}$$

where curves C_ε and C_r are marked in Fig. 2, and ω_k are first-order poles in the upper

part of the Gauss plane Z_+. Joining (5.1) and (2.10) we obtain

$$\eta(\omega) = \sum_k{}' \frac{a_k}{\omega_k - \omega},\qquad(5.2)$$

where a_k are residues of the $\eta(\omega)$ function. Thus one sees that $\eta(\omega)$ possesses only first-
-order poles, which is naturally a consequence of the Krönig-Kramers relations. The
relaxation function corresponding to these relations, using (2.11) equals

$$\Phi_N(t) = \sum_k i a_k e^{i\omega_k t}\qquad(5.3)$$

with the normalizing coefficient

$$\sum_k \frac{a_k}{\omega_k} = 1.\qquad(5.3')$$

As in general $\omega_k = \omega_k^R + i\omega_k^I$ and $a_k = a_k^R + i a_k^I$

$$\eta'(\omega) = \sum_k{}' \frac{a_k^R(\omega_k^R - \omega) + a_k^I\omega_k^I}{(\omega_k^R - \omega)^2 + (\omega_k^I)^2},\qquad(5.4)$$

$$\eta''(\omega) = -\sum_k{}' \frac{a_k^I(\omega_k^R - \omega) - a_k^R\omega_k^I}{(\omega_k^R - \omega)^2 + (\omega_k^I)^2}.\qquad(5.5)$$

These formulae are, unfortunately, too complicated for discussion without more specific
assumptions. Thus in this paragraph we shall consider only two cases: (a) if we put $\omega_k^R = a_k^R = 0$ then, as may be seen directly from (5.4) and (5.5), we obtain the Frohlich model
of discrete spectrum of relaxation times. (b) Assuming the existence of only one pole,
which, following (5.3'), leads to $a_1 = \omega_1$, we have

$$\eta'(\omega) = \frac{\omega_1^R(\omega_1^R - \omega) + (\omega_1^I)^2}{(\omega_1^R - \omega)^2 + (\omega_1^I)^2},\qquad(5.6)$$

$$\eta''(\omega) = \frac{\omega_1^I\omega}{(\omega_1^R - \omega)^2 + (\omega_1^I)^2}.\qquad(5.7)$$

These results apply equally well to optical frequencies. Formula (5.7) does not give however
the detailed shape of the Lorentz line, but its close approximation if $\omega_1^R \gg \omega_1^I$, which
is not distinguishable experimentally. It is worth noting at this point that Shimizu (1970)
starting from Louville's equation for the density operator, found for the shape of lines
in NMR a formula differing from the shape of the Lorentz line in the same way as formula
(5.7).

Finally, following Frohlich (1960) we shall formally generalize the formula (5.3)
replacing summing over poles by integrating by Z_+, then

$$\Phi_N(t) = i \int_{Z_+} a(\Omega)e^{i\Omega t} d\Omega.\qquad(5.8)$$

From this, thanks to (2.6)

$$\eta(\omega) = \int\limits_{z_+} \frac{a(\Omega)}{\Omega - \omega}\, d\Omega \qquad (5.9)$$

where on the distribution function $a(\Omega)$ only the condition of the existence of the integral (5.8) is imposed. In this way we have obtained a very general solution of equation (2.10), however of no practical importance.

6. Single-pole model with $\omega_1^R < 0$

Introducing denotations $\beta = -\omega_1^R/\omega_1^I$ and $\omega_1^I = \tau^{-1}$ we obtain from (5.2), (5.6), (5.7) and (5.3)

$$\eta(\omega) = \frac{1+i\beta}{1+i(\omega\tau+\beta)}, \qquad \beta \geqslant 0, \qquad (6.1)$$

$$\eta'(\omega) = \frac{1+\omega\tau\beta+\beta^2}{1+(\omega\tau+\beta)^2}, \qquad \eta''(\omega) = \frac{\omega\tau}{1+(\omega\tau+\beta)^2}, \qquad (6.2)$$

$$\Phi_N(t) = \frac{1+i\beta}{\tau}\, e^{-\frac{1+i\beta}{\tau}t}. \qquad (6.3)$$

It is easy to verify directly that

$$(\eta' - 1/2)^2 + (\eta'' + \beta/2)^2 = \frac{1+\beta^2}{4}. \qquad (6.4)$$

Consequently, according to the proposed model, on the Cole-Cole diagram, η' and η'' given by formulae (6.2) lie on the same fragment of the arc as those for the C-C expression, if the β parameter is connected with the β_{C-C} parameter by the relation $\beta = \mathrm{ctg}\,\dfrac{\pi\beta_{C-C}}{2}$. Hence the single pole model with $\omega_1^R < 0$ takes over the most positive feature of the C-C model, which assures the same degree of accordance with experiments in Cole-Cole diagrams as that offered by the C-C expression, and has exponential but complex relaxation function.

The parameter β, introduced here, may be evaluated from experimental data by the formula

$$\beta = \frac{1}{\eta''}\left[\tfrac{1}{4} - (\eta' - \tfrac{1}{2})^2 - \eta''^2\right]. \qquad (6.5)$$

Hence the characteristic time

$$\tau = \frac{1}{\omega}\,\frac{\eta''(1+\beta)}{\eta' - \eta''\beta}. \qquad (6.6)$$

One thus sees that similarly to the C-C expression, an increase of τ as compated to the Debye time, is observed.

Let us further analyze the properties of absorption and dispersion curves given by formulae (6.2). The absorption curve reaches its maximum equal to $\dfrac{1}{2} \dfrac{1}{1+\beta \sqrt{1+\beta^2}}$ for $\omega_{\max} = \frac{1}{2}(1+\beta^2)^{1/2}$, hence diminishes with the increase of β. Its half-width

$$\Delta\omega(\tau, \beta) = \frac{2}{\tau} \left[4(1+\beta^2) + 4\beta(1+\beta^2)^{\frac{1}{2}} - 1 \right]^{\frac{1}{2}} \tag{6.7}$$

increases with increasing β, for example $\Delta\omega(\tau, \frac{1}{2})/\Delta\omega(\tau, 0) = 1.44$. The dispersion curve for $\omega < \dfrac{1}{\tau} \left[\sqrt{1+(\beta/2)^2} - \beta/2 \right]$ lies under the Debye curve, and later on, after intersection, above it. Thanks to this all the discussed behaviour is at least qualitatively in accordance with experimental results.

To interpret the β parameter let us write down the behaviour of $\mathscr{D}(t)$ for $t > 0$ after immediate removal at the moment $t = 0$ of the constant field $\mathscr{E}_0$. Owing to (2.13) and (6.3) we have

$$\mathscr{D}(t) = (\varepsilon_s - \varepsilon_\infty)\mathscr{E}_0 e^{-\frac{t}{\tau}} \left(\cos \frac{\beta}{\tau} t - i \sin \frac{\beta}{\tau} t \right), \tag{6.8}$$

from which one may see that the motion of vector $\mathscr{D}(t)$ is not a purely damped motion, but a periodic damped motion of period $\tau\beta^{-1}$. The investigated model is thus characterized not by a continuous distribution of relaxation times, as is usually postulated for cases when the Cole-Cole diagram turns out to be a fragment of an arc, but by two times τ and $\tau\beta^{-1}$ showing the way in which the system approaches equilibrium. It is also evident that in practice $\tau\beta^{-1} > \tau$ or even $\tau\beta^{-1} \gg \tau$, hence the damping is very strong.

7. Remarks on the two-pole model

The second model after the single-pole model is naturally, in terms of simplicity, the two-pole model. Inserting into formulae (5.4) and (5.5) denotations $\omega_k^I = \tau_k^{-1}$, $\omega_k^R/\omega_k^I = -\beta_k$, $a_k^R \tau_k = \xi_k$, $a_k^I \tau_k = \zeta_k$ and restricting summation to $k = 1, 2$, we obtain

$$\eta'(\omega) = \sum_{k=1}^{8} \frac{\zeta_k - \xi_k(\tau_k\omega + \beta_k)}{1 + (\omega\tau_k + \beta_k)^2}, \tag{7.1}$$

$$\eta''(\omega) = \sum_{k=1}^{2} \frac{\zeta_k(\omega\tau_k + \beta_k) - \xi_k}{1 + (\omega\tau_k + \beta_k)^2}, \tag{7.2}$$

$$\sum_{k=1}^{2} \frac{\zeta_k - \beta_k\xi_k}{1 + \beta_k^2} = 1; \qquad \sum_{k=1}^{2} \frac{\zeta_k\beta_k + \xi_k}{1 + \beta_k^2} = 0. \tag{7.3}$$

In this model six free parameters[2] are present, thus one may believe the model to be very elastic, and because of this, difficult to apply in practice. To illustrate the adaptibility of the model, two Cole-Cole diagrams are shown in Fig. 3, corresponding to equations (7.1), (7.2) and (7.3). The first (1) is composed for

$$\xi_1 = \xi_2 = \beta_1 = \beta_2 = 0; \ \zeta_1 = \tfrac{4}{5}; \ \zeta_2 = \tfrac{1}{5}; \ \frac{\tau_2}{\tau_1} = \tfrac{1}{5}, \tag{7.4}$$
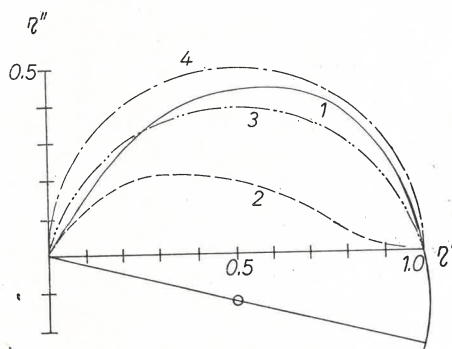

Fig. 3. Cole-Cole diagrams for *1* — two-pole model with conditions (7.4), *2* — two-pole model with conditions (7.5), *3* — single-pole model with $\beta = 0.26$, *4* — Debye model

and, as may be sees, is of "skewed-arc" type. The second one (2) was calculated for

$$\zeta_1 = \zeta_2, \ \xi_1 = \xi_2, \ \beta_1 = \beta_2 = \tfrac{1}{2}, \ \frac{\tau_2}{\tau_1} = \tfrac{1}{5}, \tag{7.5}$$

which leads to $\zeta_k = 2/5$ and $\xi_k = -1/5$. Both these diagrams illustrate the "fitting ability" to experimental data of the two-pole model.

8. Conclusions

The most important conclusion which seems to be apparent is the necessity of confronting models of dielectric relaxation, constructed by a formal generalization of models already known as well as microscopic models, with macroscopic theory (§§ 3 and 4). Such a confrontation, as was shown, may protect against errors and help to interpret results.

The most correct approach would be, naturally to construct an effective statistical theory, however such an enterprise is met with many difficulties, a fact which is well known.

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[2] Dr J. Makosz suggested that, because of physical meaning of $\eta''(\omega)$, the assumption $\zeta_k(\omega\tau_k + \beta_k) + \xi_k \geqslant 0$ can be made. This assumption leads to $\zeta_k\beta_k + \xi_k = 0$ for $\omega = 0$, and in result Eqs (7.1), (7.2) and (7.3) are simplified, what is very important in practice.

APPENDIX

The upper limit in formula (4.4) may be written as

$$\lim_{\xi \to \infty} \int_0^\infty f(x) \cos (x\xi) dx,$$

where

$$f(x) = \left(x^{1-\beta} + x^{1+\beta} + 2 \cos \frac{\pi\beta}{2} \right)^{-1}.$$

It is easily verified that for $x \geqslant 0$ and $0 \leqslant \beta \leqslant 1$, $f(x)$ is a monotonically decreasing function when $x \to \infty$. Apart from this

$$\int_0^\infty f(x) \cos (x\xi) dx = \int_0^{\frac{3\pi}{2\xi}} f(x) \cos (x\xi) dx +$$

$$+ \sum_{n=1}^{\infty} \left\{ \int_{\frac{4n-1}{2\xi}\pi}^{\frac{4n+1}{2\xi}\pi} f(x) \cos (x\xi) dx + \int_{\frac{4n+1}{2\xi}\pi}^{\frac{4n+3}{2\xi}\pi} f(x) \cos (x\xi) dx \right\} \geqslant \int_0^{\frac{3\pi}{2\xi}} f(x) \cos (x\xi) dx,$$

where for lower estimation, in the sum, in positive integrals instead of $f(x)$ the $f(x)$ value on the right edge of the integration range was inserted. In negative integrals the value $f(x)$ at the left edge of the integration range was inserted. By an similar procedure we make an upper estimation

$$\int_0^\infty f(x) \cos (x\xi) dx = \int_0^{\frac{\pi}{2\xi}} f(x) \cos (x\xi) dx +$$

$$+ \sum_{n=1}^{\infty} \left\{ \int_{\frac{4n-3}{2\xi}\pi}^{\frac{4n-1}{2\xi}\pi} f(x) \cos (x\xi) dx + \int_{\frac{4n-1}{2\xi}\pi}^{\frac{4n+1}{2\xi}\pi} f(x) \cos (x\xi) dx \right\} \leqslant \int_0^{\frac{\pi}{2\xi}} f(x) \cos (x\xi) dx.$$

As a result

$$0 = \lim_{\xi \to \infty} \int_0^{\frac{3\pi}{2\xi}} f(x) \cos (x\xi) dx \leqslant \lim_{\xi \to \infty} \int_0^\infty f(x) \cos (x\xi) dx \leqslant \lim_{\xi \to \infty} \int_0^{\frac{\pi}{2\xi}} f(x) \cos (x\xi) dx = 0$$

Q.E.D.

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