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ON THE MECHANISM OF GLASS IONIC CONDUCTIVITY

JEPPE C. DYRE
INSTITUTE OF MATHEMATICS AND PHYSICS
IMFUFA
UNIVERSITY OF ROSKILDE
POSTBOX 260
DK-4000 ROSKILDE
DENMARK

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IMFUFA, Roskilde Universitetscenter, Postbox 260, 4000 Roskilde

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af: Jeppe C. Dyre

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Abstract

It is shown that most models can not explain the broad dielectric loss peaks and the correlation between dieletric loss and d.c. conductivity, which are both universal features of glass ionic conductivity. However, these features are reproduced by Stevels' and Taylor's old "random potential energy model" as is shown from a recent approximate solution of the model. It is argued that experimental data should preferably be presented in terms of the frequency-dependent conductivity instead of as dielectric loss peaks.

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Introduction

Despite several years of research the mechanism of glass ionic conductivity is still not understood (some reviews of the subject are found in refs. 1-7). In ionic conductive glasses one always observes both d.c. conductivity and dielectric relaxation. It is very interesting that these two phenomena are correlated, an observation of seemingly universal validity. If $\sigma(0)$ denotes the d.c. conductivity, Barton, Nakajima and Namikawa (BNN) discovered that most glasses obey [8,9,10]

$$\sigma(0) = p\, \varepsilon_0\, \Delta\varepsilon\, \omega_m \qquad (1)$$

where ε_0 is the vacuum permittivity, $\Delta\varepsilon$ is the relaxation strength, ω_m is the loss peak frequency, and p is a temperature-independent numerical constant of order one. We remind that in media with a non-zero d.c. conductivity, the complex dielectric constant $\varepsilon^* = \varepsilon' - i\,\varepsilon''$ is defined by [4]

$$\varepsilon_0\, \varepsilon^*(\omega) = \frac{\sigma(\omega) - \sigma(0)}{i\omega} \qquad (2)$$

where ω is the angular frequency and $\sigma = \sigma' + i\,\sigma''$ is the frequency-dependent conductivity. Equation (1) will be referred to as the **BNN-relation**. In particular it implies that $\sigma(0)$ and ω_m have the same activation energy. This was actually discovered before the BNN-relation, demonstrating that the dielectric relaxation is due to ionic motion [11]. In fig. 1 the

correlation between $\sigma(0)$ and $\varepsilon_0 \Delta\varepsilon\, \omega_m$ is examined for several glasses. It is clear that most glasses have a p-value of order one. The BNN-relation is thus experimentally very well-founded.

There is little doubt that the BNN-relation provides a key, perhaps the key, to a deeper understanding of ionic conductivity in glasses. Another observation of general validity is the very broad dielectric loss peaks. In this paper it will be shown that most current models are not able to explain both the BNN-relation and the broad loss peaks. In contrast, one of the oldest and simplest models works. This is Stevels' and Taylor's "random potential energy model" [2,12]. In this model, the loss peak frequency is simply the minimum ion jump frequency and the proportionality between $\sigma(0)$ and ω_m becomes physically obvious.

Critique of current models

Tomozawa has reviewed the most important models for the BNN-relation [1]. Three models predict simple Debye relaxation, namely Isard's, Charles' and Nakajima's model. In Isard's model the dielectric relaxation is assumed to be due to glass inhomogeneities which may derive e.g. from phase separation. If the different phases have different d.c. conductivity, dielectric relaxation of the bulk glass will result [13]. In Charles' model of alkali ionic conductivity in oxide glasses, the dielectric relaxation is due to defect jumps where a defect

is defined as a non-bridging oxygen atom surrounded by two alkali ions [14]. Finally, Nakajima has proposed a model for the BNN-relation which assumes that the ions are forced to follow certain random zig-zag paths frozen in the glass [9].

Because each of the above mentioned models predicts Debye relaxation, it is necessary to introduce a distribution of relaxation times into the models if the observed broad dielectric loss peaks are to be reproduced. Unfortunately, this is not possible without violating the BNN-relation. The reason is the following. Dielectric relaxation implies that the real part of the conductivity, $\sigma'(\omega)$, is an increasing function of frequency. For Debye relaxation with relaxation time τ, the increase in conductivity from low to high frequencies, $\Delta\sigma$, is easily found from eq. (2):

$$\Delta\sigma = \varepsilon_0 \Delta\varepsilon \tau^{-1} . \qquad (3)$$

The dielectric loss peak frequency is equal to τ^{-1} so from eqs. (1) and (3) one finds

$$\Delta\sigma = \frac{1}{p}\sigma(0) . \qquad (4)$$

Since $\sigma'(\omega)$ is monotonically increasing we therefore have

$$\sigma'(\omega) < \left(1 + p^{-1}\right)\sigma(0) \qquad (5)$$

which is valid for all frequencies. Now, if the dielectric relaxation is assumed to be a sum of elementary Debye processes

each of which satisfies the BNN-relation, the resulting conductivity will still obey eq. (5) with a p of order one. But this is inconsistent with experiment because the broad loss peaks imply that $\sigma'(\omega) >> \sigma(0)$ whenever $\omega >> \omega_m$.
The only way to save the models is to assume that the d.c. conductivities do **not** simply add algebraically. But then a whole new theory is needed for calculating $\sigma(0)$ and one may question the relevance of starting out with Debye processes each of which satisfies the BNN-relation. In conclusion, this procedure does not seem to work and one must look for a model which from the very beginning incorporates a broad distribution of relaxation times.

Tomozawa has proposed a model [1] based on Debye's and Falkenhagen's old theory of ionic solutions. This model is interesting because it has a broader loss peak than simple Debye relaxation. Still, the model predicts an excess conductivity $\Delta\sigma$ of the same order of magnitude as $\sigma(0)$. This means that the loss peak is far from broad enough; if it is attempted to add several "Tomozawa" relaxation processes with different relaxation times one runs into the same problems as above, namely that the BNN-relation is violated.

A model which predicts a very broad dielectric loss peak is Doremus' model which is based on the space charge mechanism [5]. This model has $\varepsilon''(\omega) \propto \omega^{-1/2}$ as $\omega \to \infty$ and a loss peak frequency which is related to the d.c. conductivity. However, because of the very nature of the model, the magnitude of the dielectric loss depends on sample dimensions while the BNN-relation is a true bulk property. Therefore, Doremus' model can

not explain the BNN-relation [1].

Stevels' and Taylor's "random potential energy model" [2,12] is perhaps the oldest model. In the next section it will be shown that this model is able to explain both the BNN-relation and the broad loss peaks.

The random potential energy model

In this model it is assumed that the ions feel a more or less random potential energy deriving from the random network structure of the glass. An ion spends most time at potential energy minima, but occasionally it gains energy by thermal fluctuations to pass the energy barrier which separates adjacent potential energy minima. The ion jump frequency, γ , depends on the energy barrier, ΔE , as

$$\gamma = \gamma_0 \, e^{-\Delta E/kT} \qquad (6)$$

where γ_0 is the attempt frequency (usually $\cong 10^{12}$ Hz), k is the Boltzmann constant, and T is the temperature. Because of the randomness of the potential energy surface all energy barriers are equally likely, so from eq. (6) it is concluded that the jump frequency probability distribution, $p(\gamma)$, varies as

$$p(\gamma) \propto \gamma^{-1} . \qquad (7)$$

If the ions can jump to more that one nearest neighbour energy

minimum, the resulting $p(\gamma)$ changes only insignificantly from eq. (7) which forms the basis of the present treatment of Stevels' and Taylor's model.

The model of a random medium with the jump frequency distribution of eq. (7) has recently been considered by the author [15,16]. The treatment makes use of the CTRW approximation [17]. In order to normalize $p(\gamma)$ it is necessary to introduce two cut-off's, γ_{min} and γ_{max}. The existence of γ_{min} in real glasses is obvious: if $\gamma_{min} = 0$ a zero d.c. conductivity results. On the other hand, experimentally there is no sign of any maximum jump frequency so γ_{max} should be put equal to γ_0. Since laboratory frequencies are always much smaller than γ_0, it is convenient to let γ_0 go to infinity. In this limit one finds [15]

$$\sigma(\omega) = \sigma(0)\,\frac{i\omega\tau}{\ln(1+i\omega\tau)} \qquad (8)$$

where

$$\tau = \gamma_{min}^{-1} . \qquad (9)$$

Equation (8) does not, of course, include the contribution to the conductivity from the high frequency dielectric constant. Substituting now eq. (8) into eq. (2) we get

$$\varepsilon_0 \Delta\varepsilon = \frac{1}{2}\sigma(0)\tau \qquad (10)$$

and for the dielectric loss

$$\varepsilon''(\omega) = 2\Delta\varepsilon \left[\frac{\arctan(\omega\tau)}{\left(\ln\sqrt{1+(\omega\tau)^2}\right)^2 + \left(\arctan(\omega\tau)\right)^2} - \frac{1}{\omega\tau} \right]. \quad (11)$$

From eq. (11) the loss peak frequency can easily be determined numerically. It is given by $\omega_m \tau = 4{,}71$ which corresponds to a p-value of the BNN-relation of

$$p = 0{,}42. \quad (12)$$

Since 0,42 is a numerical constant of order one, the BNN-relation is satisfied by the random potential energy model.

The model predicts a very broad dielectric loss peak as is clear from fig. 2. In this figure the model prediction is compared to experimental data from a typical sodium silicate glass. There is a qualitative agreement between theory and experiment.

Discussion

The derivation of eq. (8) makes use of a number of simplifying assumptions: 1) All ion-ion interactions are ignored, 2) all energy barriers are assumed to be equally likely in the interval from zero to a sharply defined maximum energy barrier, and 3) the model is solved within the CTRW

approximation which is the simplest possible non-trivial mean-field approximation for calculating $\sigma(\omega)$ in random media. The validity of these approximations will not be critically examined here. One point is perhaps worth noticing. The present use of the CTRW approximation (letting $\gamma_0 \to \infty$) is formally equivalent to the electric modulus approach of Macedo, Moynihan and Bose [18]. The physical interpretation of this analogy is not quite clear at present [16,19]. One possibility is that the electrical equivalent circuit of ref. 18 (fig.3) reflects a one-dimensional character for conduction processes in glasses [19]. Another possibility is to regard the electric modulus approach as the relevant description when inhomogeneities are present, i.e. as a generalization of Isard's model [16,18]. Because of the analogy to the CTRW, if the inhomogeneities have d.c. conductivities with a probability distribution $p(\sigma(0)) \propto \sigma(0)^{-1}$, the bulk frequency-dependent conductivity is again given by eq. (8). As regards electrical properties, the random potential energy model is therefore indistinguishable from a generalized Isard model (if this model really is faithfully represented by the equivalent circuit of fig. 3).

For a number of years it has been thought that the random potential energy model, while appealing, can not possibly be correct [1,6]. The BNN-relation implies that the activation energies of $\sigma(0)$ and ω_m are equal, and this has been interpreted as implying that the a.c. conductivity must be due to ion jumps with the same activation energy as the d.c. conductivity. The argument is incorrect, however. As is clear from the preceding section, while the activation energy of

$\sigma(0)$ is certainly equal to that of γ_{min}, there is absolutely no problem with having larger jump frequencies involved in the conduction process, corresponding to smaller energy barriers. Another common objection to Stevels' and Taylor's model is that no model based on a distribution of energy barriers can predict a loss peak with a temperature-independent shape. This is wrong; in the present model the jump frequency distribution is given by eq. (7) for all temperatures and therefore the shape of the loss peak indeed is temperature-independent.

Glass ionic conductivity data are usually presented in terms of the dielectric loss when the frequency dependence is discussed. However, because d.c. conductivity and dielectric loss are both due to ionic motion, the subtraction of $\sigma(0)$ in eq. (2) really has no physical justification. In the author's opinion, data should preferably be presented in terms of the real part of the conductivity. As an example, in fig. 4 the data of fig. 2 are shown together with model predictions (eq. 8). A number of important features of glass ionic conductivity become immediately clear when data are presented this way. The characteristic frequency which marks the onset of a.c. conductivity is just the dielectric loss peak frequency ω_m. On a time-scale larger than ω_m^{-1}, $\sigma(\omega)$ is frequency-independent so the glass "looks" homogeneous to the ions. On a time-scale smaller than ω_m^{-1}, $\sigma(\omega)$ is strongly frequency-dependent. The obvious physical interpretation is that ω_m is the **minimum** ion jump frequency; then many jumps must be involved for times >> ω_m^{-1}. In order to account for the strong

increase of $\sigma'(\omega)$ for $\omega > \omega_m$, jump frequencies much larger than ω_m must however also be present in the glass. The random potential energy model has exactly these features. In this model a wide range of jump frequencies is present, deriving from a range of activation energies.

The spread in activation energies in Stevels' and Taylors' model is not an arbitrary postulate. On the contrary, it is a simple fact which follows immediately when experiments are presented in terms of the frequency-dependent conductivity in a log-log plot as in fig. 4 but without the normalization: The shape of the conductivity curve is always temperature-independent. Because of the BNN-relation, when the temperature changes the conductivity curve is simply displaced in the direction 45° to the positive x- and y-axis. It is now clear by inspection that the a.c. conductivity indeed has a smaller activation energy than the d.c. conductivity. And from the curvature of the conductivity curve it can be concluded that a whole range of activation energies must be involved. Thus, the experimental facts themselves apparently forces one to adopt Stevels' and Taylor's approach and base the theory of glass ionic conductivity on a distribution of energy barriers.

Another reason for preferring to discuss glass ionic conductivity in terms of $\sigma(\omega)$, is the conceptual dilemma involved in the use of $\varepsilon(\omega)$ as defined by eq. (2). One would like to interpret $\varepsilon(\omega)$ in terms of a dielectric polarisation but it is difficult, if not impossible, to tell which ion jumps contribute to $\sigma(0)$ and which contribute to $\varepsilon(\omega)$. This dilemma is old and well-known. It derives from the fact that it

is not possible to distinguish between bound and free charges in a.c. electric fields [20]. The problem disappears when one operates only with $\sigma(\omega)$.

Conclusions

Most current models of glass ionic conductivity can not explain both the BNN-relation and the observed broad dielectric loss peaks. On the other hand, the random potential energy model is rather successful. This model involves no ad hoc assumptions and it is probably the simplest possible model which is not totally unrealistic. The model is based on a fact which can be deduced directly from experiment when presented in terms of the frequency-dependent conductivity, namely the existence of a distribution of energy barriers. The BNN-relation is satisfied by the model. Here, the relation is nothing but the statement that the d.c. conductivity is proportional to the minimum ion jump frequency. The jump frequency distribution eq. (7) strongly emphasizes the smallest jump frequencies which are also the most important for $\sigma(0)$ because they partially act as traps, so this proportionality is by no means surprising. In addition to the BNN-relation, the model also reproduces the observed broad asymmetric loss peaks, the temperature independence of the loss peak shape, and the almost frequency-independent dielectric loss as $\omega \to \infty$. These are all well-known characteristics of glass ionic conductivity [1,4]. The quantitative model predictions are not entirely successful, though (fig. 2 and fig.

4). This is undoubtedly because it is too simple to regard all energy barriers as equally likely. Refinements of the model must make use of a more realistic energy barrier distribution. Admittedly, this introduces a temperature dependence into the loss peak shape, but in many cases the effect is only weak and may still be consistent with experiment. The real challenge for the future is to make a coherent theory in which the energy barrier distribution is correlated to the glass structure and conditions of glass formation.

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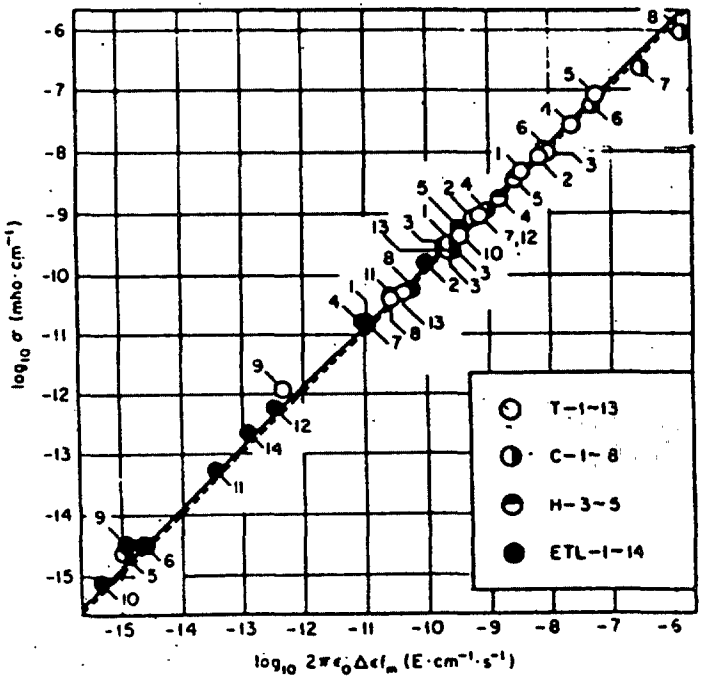
Figure Captions

Fig. 1 : Test of the BNN-relation for several glasses (note that $2\pi f_m = \omega_m$) (reproduced from ref. 9).

Fig. 2 : Model prediction for the dielectric loss (full curve) and experimental data from a typical sodium silicate glass (data taken from ref. 4). The dashed curve is the Debye dielectric loss peak.

Fig. 3 : Equivalent circuit of the CTRW approximation in the $\chi_0 \to \infty$ limit [16]. The circuit was suggested by Macedo, Moynihan and Bose in their electric modulus approach towards glass ionic conductivity [18].

Fig. 4 : Model prediction for the real part of the conductivity (full curve) and the experimental data of fig. 2. For the data points the conductivity has been calculated assuming $p=1$ in the BNN-relation and utilizing the Kramers-Kronig relation for calculating $\Delta\varepsilon$ ($= 3{,}7 \cdot \varepsilon''_{max}$). The characteristic time τ is a fitting parameter for the data points in this figure.


-6
-7
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-14
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-15 -14 -13 -12 -11 -10 -9 -8 -7 -6
$\log_{10}\sigma$ (mho·cm^{-1})
$\log_{10} 2\pi\epsilon_0\Delta\epsilon f_m$ (E·cm^{-1}·s^{-1})
T-1-13
C-1-8
H-3-5
ETL-1-14

Fig. 1

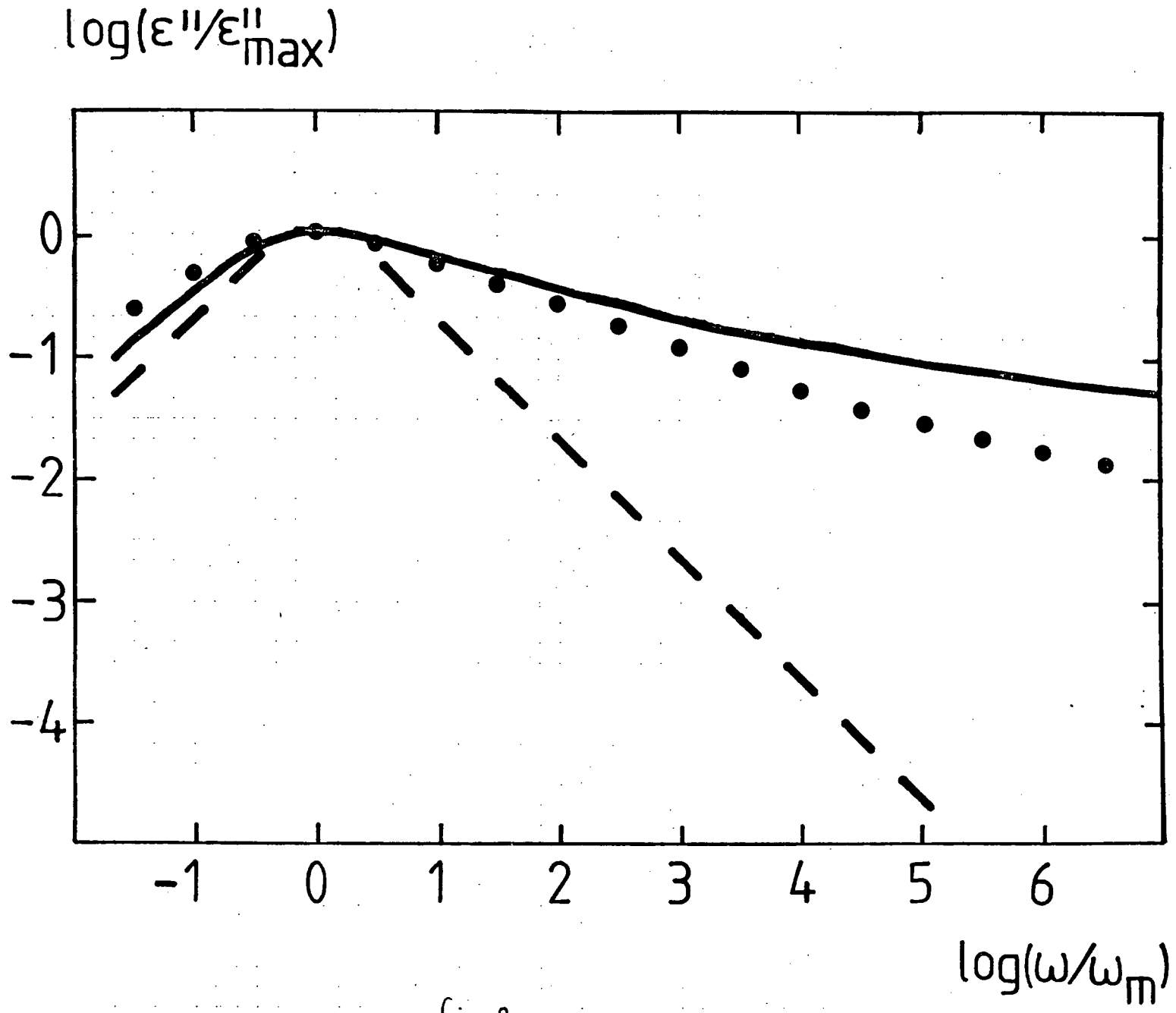

fig. 2

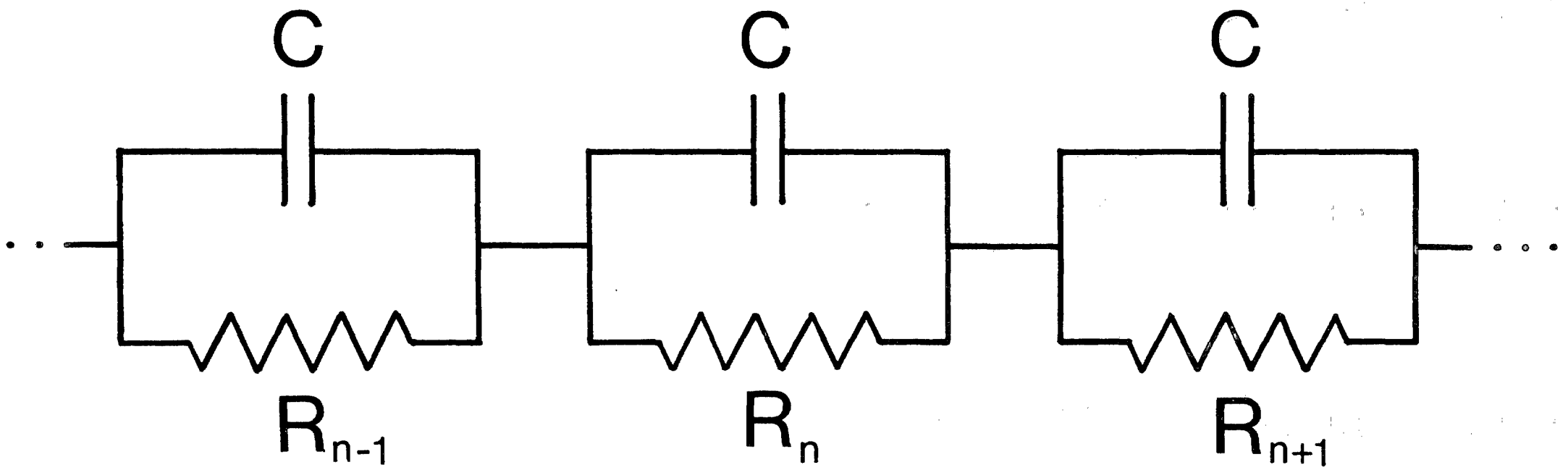

fig. 3

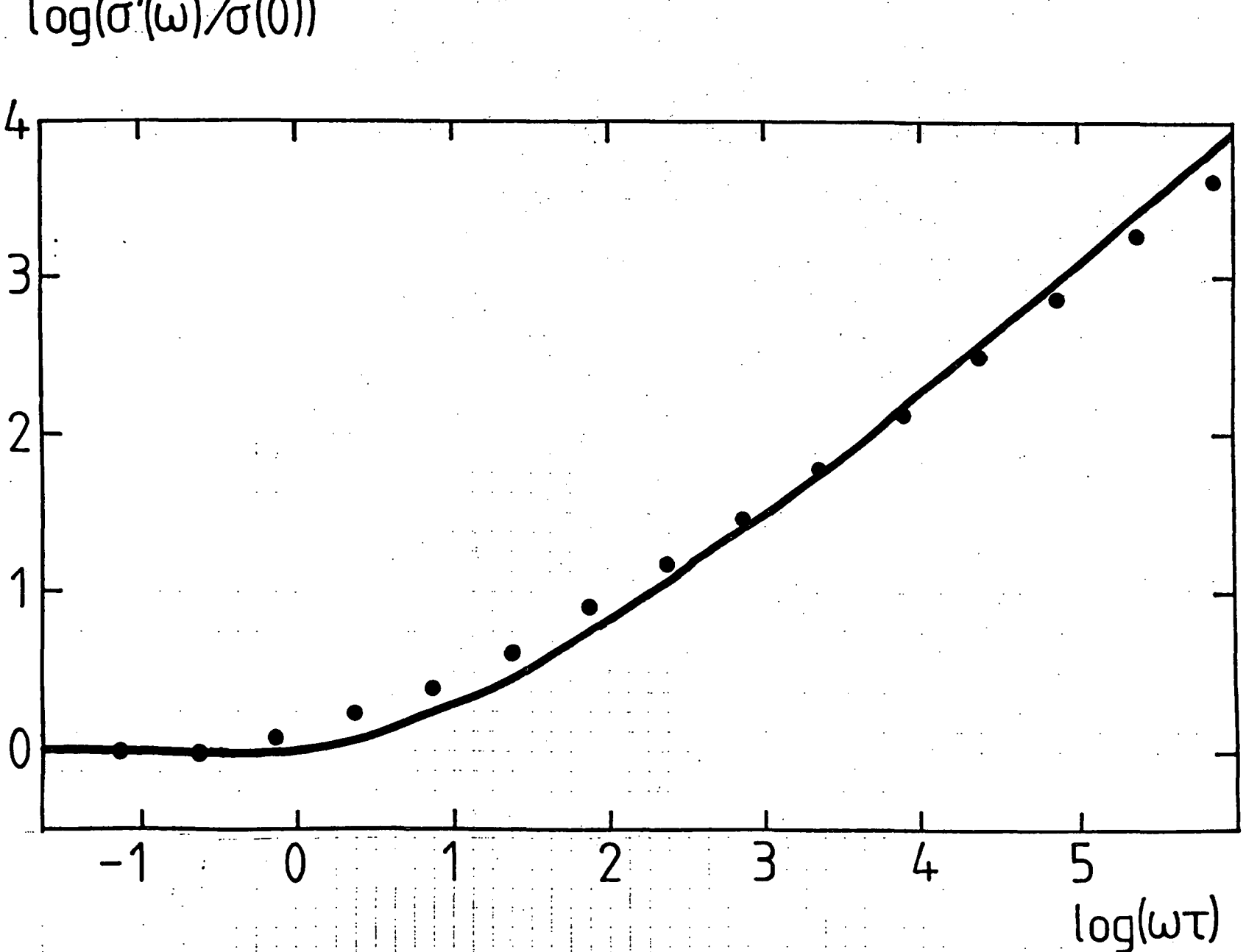

fig. 4

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