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SOME REMARKS ON AC CONDUCTION IN DISORDERED SOLIDS

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ABSTRACT

A number of remarks are made to both theory and experiment of AC conduction in disordered solids. As regards experiment, it is argued that the observed power-law behavior of the frequency-dependent conductivity, $\sigma(\omega)$, is not fundamental, that the Ngai-relation between DC and AC activation energies follows from independent experimental facts, and that the shape of the modulus peak has no fundamental significance. As regards hopping models for AC conduction, it is shown that three commonly used arguments against the existence of a distribution of activation energies are all wrong. Also, it is shown that $\sigma(\omega) \neq \sigma(0)$ only if there are correlations in the directions of different charge carrier jumps; in particular this result implies $\sigma(\omega) = \sigma(0)$ for all frequencies in the CTRW model. In the final section a number of open problems are listed and suggestions are made for future work.

ABSTRACT

A number of remarks are made to both theory and experiment of AC conduction in disordered solids. As regards experiment, it is argued that the observed power-law behavior of the frequency-dependent conductivity, $\sigma(\omega)$, is not fundamental, that the Ngai-relation between DC and AC activation energies follows from independent experimental facts, and that the shape of the modulus peak has no fundamental significance. As regards hopping models for AC conduction, it is shown that three commonly used arguments against the existence of a distribution of activation energies are all wrong. Also, it is shown that $\sigma(\omega) \neq \sigma(0)$ only if there are correlations in the directions of different charge carrier jumps; in particular this result implies $\sigma(\omega) = \sigma(0)$ for all frequencies in the CTRW model. In the final section a number of open problems are listed and suggestions are made for future work.

1. INTRODUCTION

This paper discusses AC conduction in non-metallic disordered solids. A number of remarks are made, most of which are not new but are still not generally appreciated. The class of disordered solids with interesting AC behavior is very large, including amorphous semiconductors [1,2], ionic conductive glasses [3,4], conducting polymers [5,6], various defective or doped crystals [7-9], and many polycrystals [10,11].

Various representations of AC data are used. One possibility is to use the complex frequency-dependent conductivity, $\sigma(\omega) = \sigma'(\omega) + i\sigma''(\omega)$. A common alternative is the complex electric modulus, $M(\omega) = M'(\omega) + iM''(\omega)$, defined [12] by

$$M(\omega) = i\omega/\sigma(\omega). \quad (1)$$

Data may also be presented in terms of the complex impedance [13,14], or in terms of the complex dielectric constant defined by

$$\epsilon_0 \epsilon(\omega) = \frac{\sigma(\omega) - \sigma(0)}{i\omega}. \quad (2)$$

Here, ϵ_0 is the vacuum permittivity. The negative imaginary part of $\epsilon(\omega)$, $\epsilon''(\omega)$, is referred to as the dielectric loss.

AC conduction in quite different disordered solids shows a number of common features, a surprising fact which is often overlooked. For each of the above listed classes of glassy solids one observes, almost without exception [15-19]: At high frequencies $\sigma'(\omega)$ follows a power-law with an exponent s in the range 0.7-1.0; s goes to one as the temperature goes to zero. At the dielectric loss peak frequency, ω_m , there is a transition to a frequency-independent conductivity below ω_m . The BNN relation [20-22] is satisfied:

$$\sigma(0) = p \Delta\epsilon \epsilon_0 \omega_m \quad (3)$$

where $\Delta\epsilon = \epsilon(0) - \epsilon(\infty)$ and p is a numerical constant of order one. Finally, the time-temperature superposition principle is usually obeyed, i. e. , the shape of the $\sigma'(\omega)$ -curve is temperature-independent when plotted in a log-log plot.

The outline of the paper is the following. In sec. 2 some points relating to experiment are discussed. Section 3 is devoted to hopping models for AC conduction. Section 4 deals with a number of open problems and gives suggestions for future work. Finally, sec.5 is the conclusion.

2. REMARKS RELATING TO EXPERIMENT

1) The observed power-law frequency-dependence of the conductivity is hardly fundamental.

The large frequency power-law

$$\sigma'(\omega) \propto \omega^s \quad (4)$$

is deduced from $\sigma'(\omega)$ following a straight line in the log-log plot. Since both frequency and conductivity usually vary several decades, it is absolutely reasonable to plot data in a log-log plot. However, log-log plots are dangerous; an old saying [23] warns: "Anything is a straight line in a log-log plot". To illustrate this point, Fig. 1 shows a log-log plot of $\sigma'(\omega)$ where $\sigma(\omega)$ is given by

$$\sigma(\omega) = \sigma(0) \frac{i\omega\tau}{\ln(1+i\omega\tau)} \quad (5)$$

This function gives a good fit to many data [19]. The function follows closely a straight line at high frequencies though there is no power-law hidden in it. Most workers would report an exponent around 0.8 for data following Fig. 1. There is no problem with this as long as one speaks only about **approximate** power-laws. But there is no basis for concluding from experiments that power-laws are **fundamental**, as is often done [24-26].

2) The Ngai-relation follows from the BNN-relation and the time-temperature superposition principle.

The Ngai-relation [27] correlates three quantities, the activation energy of the dielectric loss peak frequency, E_{ω_m} , the AC conductivity activation energy, E_{AC} , and the exponent s of Eq. (4), as follows:

$$E_{AC} = (1-s)E_{\omega_m} \quad (6)$$

Equation (6) is confirmed by experiment [27] (E_{AC} is sometimes derived from NMR experiments which, however, give the same activation energy as that of AC conduction [4,28]). As illustrated in Fig. 1, the loss peak frequency ω_m is the characteristic frequency for the onset of AC conduction. Since the dielectric loss strength $\Delta\epsilon$ is only weakly temperature-dependent, the essence of the BNN-relation is an approximate proportionality between $\sigma(0)$ and ω_m . Remember that the time-temperature superposition principle is the fact that, at different temperatures, one observes in the log-log plot parallel displacements of the same $\sigma'(\omega)$ -curve. Because of the proportionality between $\sigma(0)$ and ω_m , the $\sigma'(\omega)$ -curve is displaced in a direction 45° to the x- and y-axis as the temperature changes. Since activation energies are obtained as derivatives of the logarithm, it is now straightforward to show that Eq. (6) is automatically obeyed.

3) The shape of the modulus peak has no fundamental significance.

For all disordered solids the imaginary part of the electric modulus, $M''(\omega)$, has a peak at a frequency of the same order of magnitude as ω_m . The shape of the modulus peak is often attributed to a spectrum of relaxation times [12,29]. This spectrum, however, has no significance reflecting the motion of the charge carriers. This is because there is always, in parallel to the charge transport due to the mobile charge carriers, the current due to the infinitely fast dielectric displacement. The strength of the latter current is given by the infinite frequency dielectric constant, ϵ_∞ . If ϵ_∞ is changed, the shape of

$M''(\omega)$ is affected [30] even if the mobile charge carriers move about in the solid exactly as before.

4) There are close mechanical analogies to the observed AC behavior.

4a) Many ionic conductive glasses have an internal friction loss peak at the dielectric loss peak frequency [4,31,32]. This mechanical loss must be due to ionic motion. In effect, this means that a mechanical stress induces an ionic current.

4b) The frequency-dependent viscosity, $\eta(\omega)$, of a typical highly viscous liquid, e. g., a polymeric liquid, looks very much like $1/\sigma(\omega)$ for a typical disordered solid. Thus, at low frequencies $|\eta(\omega)|$ is constant whereas at higher frequencies $|\eta(\omega)|$ decreases like an approximate power-law [33]. A possible explanation of this analogy is the following. Suppose a foreign microscopic particle is introduced into a viscous liquid. If the particle is described by hydrodynamics, its frequency-dependent mobility (velocity/force) varies as $1/\eta(\omega)$. Because of the fluctuation-dissipation theorem [34], the analogy between $1/\eta(\omega)$ of a viscous liquid and $\sigma(\omega)$ of a disordered solid implies that (in equilibrium) the particle moves about in the liquid just as a charge carrier moves about in a disordered solid (in zero external field). This has been confirmed by conductivity measurements on ions dissolved in viscous liquids, where the observed conductivity is indeed like that of a disordered solid [35,36].

3. REMARKS RELATING TO HOPPING MODELS

In hopping models DC and AC conduction are both due to hopping charge carriers [37]. The disorder is usually reflected by assuming randomly varying transition rates $\Gamma(s' \rightarrow s)$ between two sites, s' and s . If $P(s,t)$ is the probability for a particle to be at site s at time t , a hopping model is described by the master equation [37,38]

$$\frac{\partial P(s,t)}{\partial t} = -\gamma_s P(s,t) + \sum_{s'} \Gamma(s' \rightarrow s) P(s',t) \quad (7)$$

where $\gamma_s = \sum_{s'} \Gamma(s \rightarrow s')$. Equation (7) refers to the zero external field situation; more generally Γ depends on the external field. It can be shown that in hopping models $\sigma'(\omega)$ is always an increasing function of ω [39]. No exact analytical methods are available for evaluating $\sigma(\omega)$ in hopping models, but various approximate methods exist [37,40,41].

1) Three common arguments against hopping models are all wrong.

In most hopping models the variation in hopping rates is assumed to derive from a spread in activation free energies, $P(\Delta F)$. The following three arguments have traditionally been put forward against the existence of any $P(\Delta F)$:

1a) "Any distribution of activation energies implies the DC conductivity is non-Arrhenius."

This is not necessarily true; in some models $\sigma(0)$ is Arrhenius with an activation energy which is simply the maximum barrier encountered on any "percolation" path between the electrodes. In one dimension exact results are available [42,43]. Here, $p(\Delta F) = \text{const.}$, or more generally $P(\Delta F) \propto \exp(-\Delta F/\Delta F_0)$, give

an exactly Arrhenius $\sigma(0)$ if a sharp cut-off at a maximum activation energy is assumed. Similar results are obtained from the approximate analytical methods available in three dimensions [37,40,41].

1b) "The BNN-relation implies that AC conduction is due to processes with activation energy equal to that of $\sigma(0)$."

The BNN-relation implies that the dielectric loss peak frequency has the same activation energy as $\sigma(0)$. But this does not rule out the possibility that a distribution of activation energies is responsible for the frequency dispersion. Thus, in hopping models ω_m corresponds to the lowest effective jump frequency and this quantity is determined by the maximum energy barrier, just as $\sigma(0)$ is itself [44].

1c) "The time-temperature superposition principle contradicts the existence of a distribution of energy barriers."

It is often claimed that the existence of a distribution of activation energies implies a broadening of the distribution of relaxation times as the temperature is lowered, thereby violating the time-temperature superposition principle. Two points are to be noticed. First, for experimental reasons the time-temperature principle is usually checked only over a relatively narrow range of temperatures and frequencies; here any sufficiently broad distribution of activation energies will obey the time-temperature superposition principle rather accurately. Secondly, for the flat distribution of activation energies, $p(\Delta F) \propto \text{const.}$, the time-temperature superposition principle is obeyed exactly. In this case the distribution of jump frequencies varies as Γ^{-1} at all temperatures. In conclusion, as long as one assumes a

sufficiently broad distribution of activation energies, approaching the flat distribution, there is no contradiction with experiment.

Traditionally, the points 1a), 1b) and 1c) have been thought to imply at most a quite **narrow** $p(\Delta F)$, which obviously cannot account for the observed very **broad** loss peaks. This is why an early model like Stevels' and Taylor's random potential energy model from 1957 [45,46] was never considered a serious candidate for explaining experiments.

2) The conductivity is frequency-dependent only if there are correlations between the directions of charge carrier jumps.

Thus, if each jump occurs in a random direction one has $\sigma(\omega) = \sigma(0)$ at all frequencies [38]. To prove this we first recall the fluctuation-dissipation theorem [34] which expresses $\sigma(\omega)$ in terms of the zero-field auto-correlation function of the total current in volume V , $J(t)$, in the following way

$$\sigma(\omega) = \frac{1}{3k_B T V} \int_0^\infty \langle J(0) \cdot J(t) \rangle e^{-i\omega t} dt \quad (8)$$

Here k_B is the Boltzmann constant and T is the temperature. In hopping models the jumps are instantaneous and $J(t)$ is a sum of delta functions. If the i 'th jump occurs at time τ_i and displaces a particle by Δr_i , one has

$$J(t) = q \sum_i \Delta r_i \delta(t - \tau_i) \quad (9)$$

where q is the charge carrier charge. For $\langle J(0) \cdot J(t) \rangle$ to be non-zero at any $t > 0$ one must have $\langle \Delta r_i \cdot \Delta r_j \rangle \neq 0$ for at least one pair of $i < j$. But whenever the direction of the latter jump, Δr_j , is random, one has $\langle \Delta r_i \cdot \Delta r_j \rangle = 0$. Consequently $\langle J(0) \cdot J(t) \rangle \propto \delta(t)$ and the conductivity is frequency-independent according to Eq. (8). This result has two important consequences:

2a) Any random walk in a spatially homogeneous medium has

$$\sigma(\omega) = \sigma(0) .$$

This is true even for non-markovian random walks. An important example is the continuous time random walk model (CTRW) of Montroll and Weiss [47]. This model is characterized by the so-called waiting-time distribution function, $\psi(t)$, which is the probability for a particle to jump at time t , given the particle last jumped at $t=0$. In 1973 Scher and Lax erroneously calculated $\sigma(\omega)$ in terms of $\psi(t)$ [48]; the error was pointed out by Tunaley who proved by direct calculation that there is no frequency dispersion of the conductivity in the CTRW model [49]. While the CTRW model is itself of no use as a model for AC conduction, the formalism developed by Scher and Lax does give rise to a useful approximation, usually referred to as the CTRW approximation or the Hartree approximation [40].

2b) The existence of a distribution of relaxation times is not enough to ensure frequency-dependence of the conductivity. Consider hopping in a potential where all maxima are equal but the minima vary (Fig. 2). Obviously, in this model there is a distribution of waiting times. But the direction of each charge carrier jump is random so $\sigma(\omega) = \sigma(0)$. This has also been shown by explicit calculation [51,52]. In passing we note that the model of Fig. 2 is a useful model for the transient behavior of photo-excited charge carriers in amorphous semiconductors [53]. Here, a brief laser pulse excites the electrons to random states at $t=0$, and the current in an external field subsequently monitors the thermalisation of the charge carriers. This example shows that, in general, transient currents cannot be calculated from

$\sigma(\omega)$, as has been predicted from the study of specific models [54,55].

4. OPEN PROBLEMS AND SUGGESTIONS FOR FUTURE WORK

1) Are reported data always bulk and not due to contact effects?

Electrode effects may cause serious problems for the interpretation of measurements. One might think it could easily be checked, by simply varying the sample size, whether or not the bulk response is measured. For disordered solids, however, it is often difficult to prepare two samples with identical physical properties, so this method does not always work. The contacts are usually modeled as simple RC-elements, implying the bulk response is measured at sufficiently high frequencies. But it has never been proved that this procedure is correct, and it has even been suggested that contacts and interfaces play a dominant role in the whole range of frequencies measured [56]. While this is probably too drastic a statement, it is a fact that even a quite simple model of the electrode/sample interface predicts a non-trivial frequency-dependence of the measured conductivity varying like $\omega^{1/2}$ [31,57]. In conclusion, it is not obvious that all reported data are bulk, and more work is needed to clarify the role of contacts.

2) Are DC and AC conduction always due to the same mechanism?

The BNN-relation shows that DC and AC conduction in disordered solids are strongly correlated. The simplest possibility is that DC and AC conduction are both due to the same mechanism, as is the case in hopping models. The existence of a loss peak supports this; loss peaks are hard to explain otherwise. It should be noted that, when there is no dielectric loss peak, a BNN-like

relation may still exist between $\sigma(0)$ and the characteristic frequency ω'_m defined by

$$\sigma'(\omega'_m) = 2\sigma(0) . \quad (10)$$

[If a loss peak does exist, ω'_m is close to ω_m .] Suppose $\sigma'(\omega) = \sigma(0) + A\omega$. Then clearly ω'_m is proportional to $\sigma(0)$ although this does not reflect any relation between DC and AC conduction.

3) There are theoretical reasons to expect $\epsilon''(\omega) \propto \omega^{1/2}$ on the low-frequency side of the dielectric loss peak.

In hopping models one always has $\sigma(\omega) = \sigma(0) + C(i\omega)^{3/2}$ as $\omega \rightarrow 0$ [38,58], an example of the celebrated "long time tails". As one of the few general predictions in the field, this should ideally be tested on a number of disordered solids. But unfortunately electrode effects cause very serious problems for measuring accurately the low-frequency side of the loss peak, and the prediction may be close to impossible to verify.

4) Does any solid exist which has $\sigma'(\omega) \ll \epsilon_0 \omega$?

A puzzling phenomenon is the fact that, apparently, any solid has a conductivity $\sigma'(\omega)$ which is at least of order $\epsilon_0 \omega$ [16]. Thus, at 1 MHz the conductivity is never much less than $10^{-6} (\Omega \text{ cm})^{-1}$. This rule seems to apply without exception, even to single crystal insulators. It could be a spurious effect due to contact effects [56], or due to experimental problems in distinguishing properly between $\sigma'(\omega)$ and $\sigma''(\omega)$ [$\sigma''(\omega)$ always has a sizable contribution from the infinite frequency dielectric constant]. If the effect is real an explanation is very much needed. Is it possible that even the most "perfect" single crystal contains enough defects to account for this

observation?

5) What kind of measurements could supplement the measurement of $\sigma(\omega)$?

The AC conductivity is the $k=0$ component of the more general quantity $\sigma(k,\omega)$ [which, by the fluctuation-dissipation theorem, is related to equilibrium fluctuations of the k 'th Fourier component of $\mathbf{J}(\mathbf{r},t)$]. It would be interesting to have measurements of $\sigma(k,\omega)$. For electronic systems it is not obvious how to do this, but for ionic conductors neutron scattering can be applied, at least in principle. Other important measurements to supplement $\sigma(\omega)$ are transient current experiments (only available for electronic conductors) [59], excess current noise measurements [60], or large field experiments [31,61].

6) Is the observed AC behavior due to microscopic or macroscopic inhomogeneities?

The mathematical description of, and predictions for, inhomogeneous conductors are quite similar to that of hopping models [11,14]. Therefore, it is not clear from AC measurements alone whether macroscopic or microscopic inhomogeneities are responsible for the observed frequency dispersion. It is not unlikely that, in some amorphous systems, there are inhomogeneities several hundred Angstroms large. One way to distinguish between macroscopic and microscopic inhomogeneities is to measure the large field response; for macroscopic inhomogeneities one expects nonlinearities to set in at much lower fields than for microscopic inhomogeneities.

7) There are two important open problems relating to hopping

models:

7a) How accurate are the presently available approximate analytical solutions of hopping models?

Perhaps the simplest hopping model is the random free energy barrier model which, when solved in the CTRW approximation, yields Eq. (5) [19]. Numerical solutions of this model should be undertaken to assess the validity of Eq. (5). Preliminary work shows that, in one dimension, Eq. (5) works very well [62]. In general the question 7a) remains unanswered.

7b) What is the cause of the quasi-universality among different models?

As noticed by Summerfield in 1985 [63], different models solved in the extended pair approximation (EPA) yield almost identical predictions for $\sigma(\omega)$ (apart from an overall scaling of σ and ω). This "quasi-universality" applies not only to EPA models, but to most models studied so far. The cause of quasi-universality is not clear. The agreement between different theoretical models is generally much better than the agreement between theory and experiment, where quasi-universality does not really apply. This indicates that the present hopping models are too simple. It seems likely that interactions between the charge carriers have to be taken into account to arrive at a realistic model. (Contrary to what is sometimes claimed, Eq. (7) cannot describe interacting particles [64].)

5. CONCLUSION

There are a number of important unsolved problems in the field of AC conduction. Because of this, measurements of $\sigma(\omega)$ do not yet provide unambiguous insight into the conduction process. More work is needed before this goal is reached, for instance along the lines of sec. 4. In this sense, AC conduction is still a field in its infancy.

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FIGURE CAPTIONS

Fig. 1: Real part of the function $\sigma(\omega)$ given by Eq. (5). Though there is no power-law in this function, it follows closely a power-law at high frequencies. The function gives a good fit to many data [19]; thus one cannot conclude from experiment that a power-law frequency-dependence of the AC conductivity of disordered solids is fundamental. - The vertical line marks the dielectric loss peak frequency which is always found where the conductivity starts increasing.

Fig. 2: Potential energy of a hopping model which, because the direction of each charge carrier jump is random, has no frequency-dependence of the conductivity. This example shows that a distribution of waiting times is not enough to ensure frequency-dependence of the conductivity. Also, since the model has non-trivial transient behaviour [53], the example shows that there is no correlation between AC conduction and transient behaviour.

Fig. 1

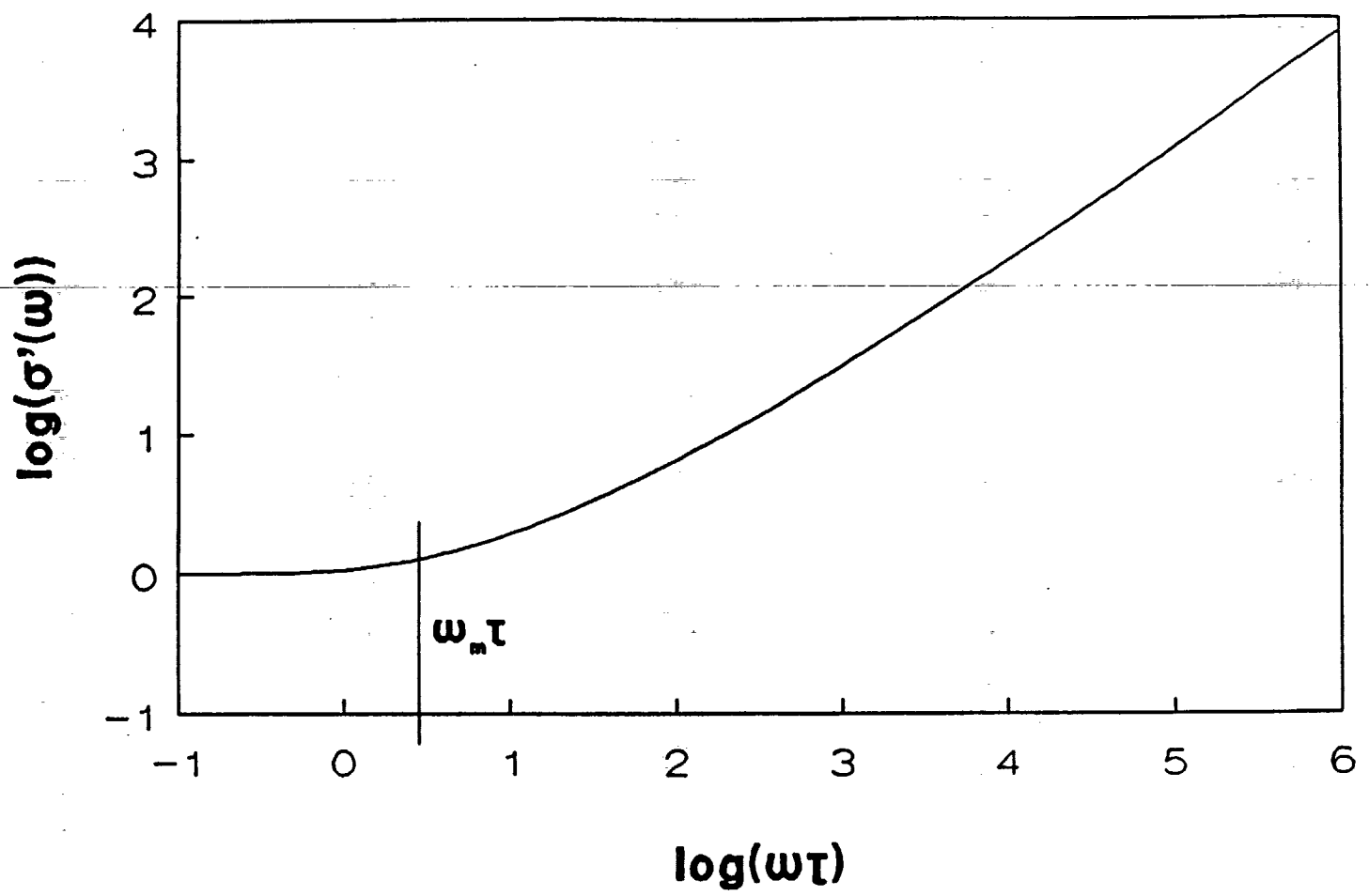
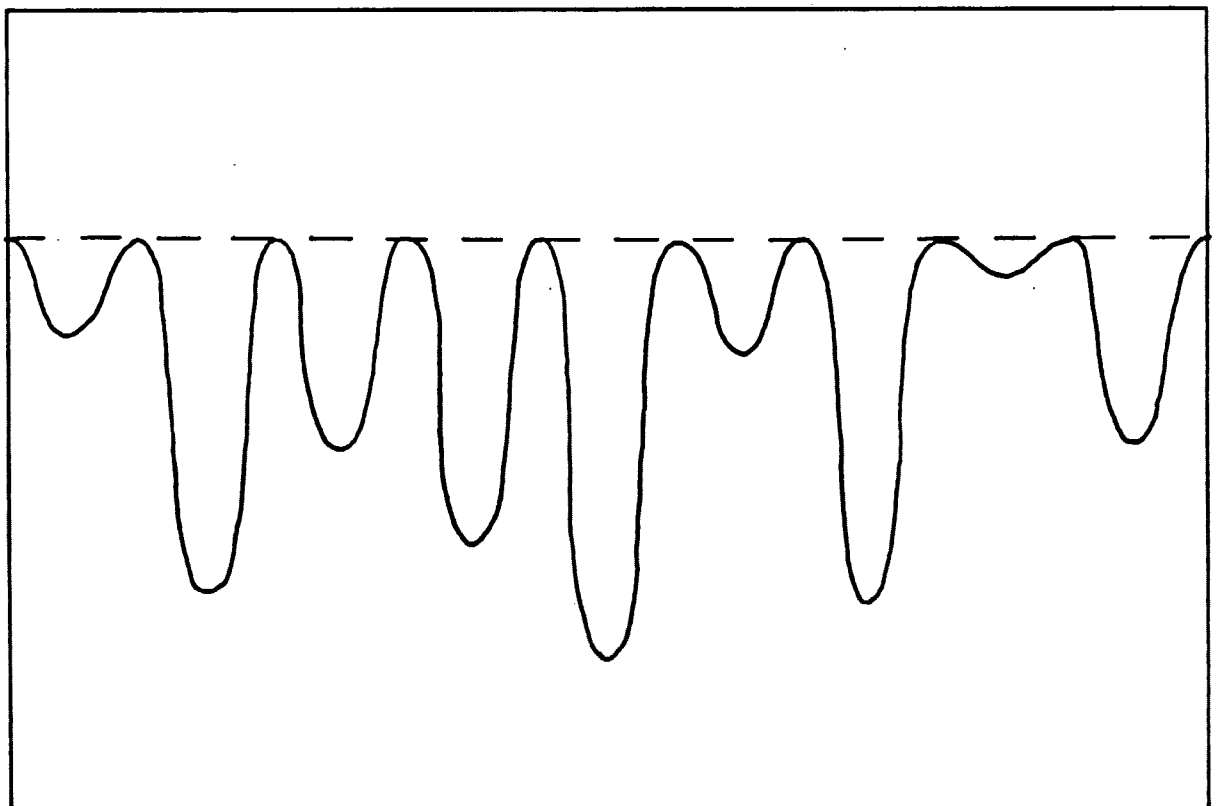


Fig. 2



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