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Tage Christensen:
The Frequency Dependence of the Specific Heat at the Glass Transition.

Jeppe C. Dyre:
A simple model of ac hopping conductivity.

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THE FREQUENCY DEPENDENCE OF THE SPECIFIC HEAT AT THE GLASS TRANSITION.

Af: Tage Christensen

A SIMPLE MODEL AF AC HOPPING CONDUCTIVITY.

Af: Jeppe C. Dyre

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Abstract:

This IMFUFA-text contains two papers presented at the Third International Conference on the Structure of Non-Crystalline Materials held july 1985 in Grenoble, France. The first paper deals with the frequency dependent specific heat at the glass transition (p. 1-3), and the second paper deals with ac hopping conductivity (p. 4-8). Each paper starts with a short abstract.

The Frequency Dependence of the Specific Heat at the Glass Transition

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ABSTRACT - The frequency dependence of the specific heat of glycerol at the glass transition has been measured. A time-temperature equivalence is found - the mean relaxation time having an activation temperature of $2.1 \cdot 10^4$ K. In the high frequency limit $c - c_\infty$ decays as a power law with exponent 0.28

The glass transition shows up in various thermodynamic responses such as the specific heat c_p /1/. When c_p is measured at a certain cooling rate the glass transition temperature T_g is found at the "soft discontinuity" in $c_p(T)$. T_g is dependent on the cooling rate, being higher at fast cooling than at slow cooling. Futhermore, hysteresis effects are seen, i.e. cooling and heating gives different curves $c_p(T)$, see fig. 1 taken from /2/.

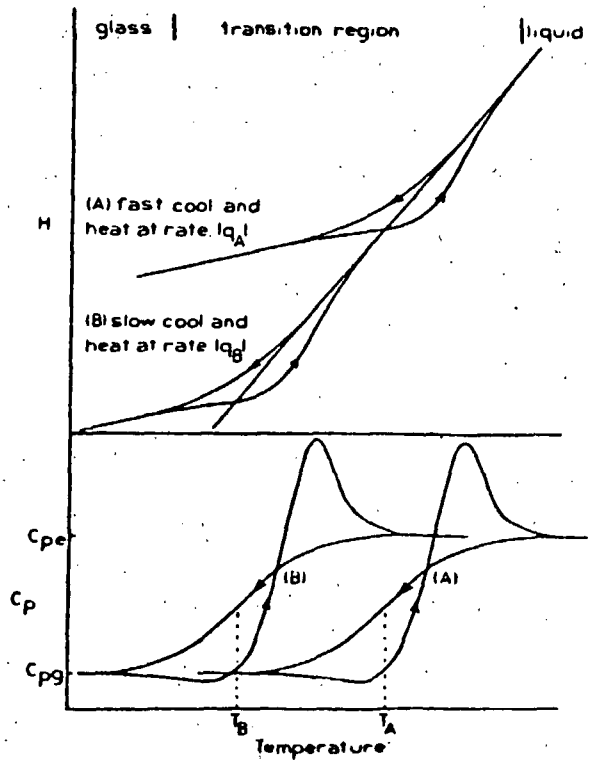

Fig. 1 - Enthalpy and heat capacity vs. temperature plots for a glass cooled and then reheated through the transition region at different rates.

The physical picture of the glass transition is that near T_g some degrees of freedom are so slowly approaching equilibrium (times exceeding the duration of the experiment), that they cease to contribute to c_p.
Usually the enthalpy relaxation near T_g is measured in the time domain. In this work we have considered the frequency domain.
Doing linear nonequilibrium thermodynamics, we apply an AC-heating current $I_o e^{i\omega t}$, small enough for the temperature response $T_o e^{i(\omega t+\phi)}$, to be linear. The heat impedance is defined as $Z = T_o e^{i\phi}/Q_o$ and the complex heat capacity becomes $c = 1/i\omega Z$.
In a substance with structural relaxation c will have a nonvanishing imaginary part. As for more commonly considered susceptibilities, Im(c) is proportional to the entropy production during a cycle due to irreversible processes.
AC-temperature calorimetry is a technique known to have the advantage of a high signal-to-noise ratio /3/, and has been used to determine critical exponents of equilibrium phase transitions /4/, where c_p is independent of ω. It turns out to be a powerfull method for a relaxing heat capacity allso.

EXPERIMENTAL

$c(\omega)$ has been measured for glycerol. 130 mg glycerol was put between two concentric aluminia cylinders 0.3 mm apart. The heat current were send into the liquid at the inner cylinder, and the temperature measured at the outer cylinder. The frequency range was 2,4 mHz to 35 mHz, - the lower time limit set by the heat diffusion time in the sample and the upper time limit by the patience of the experimentalist. The temperature was measured every half second by a computer, and through a Fourier analysis the amplitude and phase were determined.

REDUCED DATA

After subtracting the background admittance consisting of container-capacity and heat conductivity to temperature bath, the data were subjected to the following procedure. The liquid heat capacity $c_o(T)$ and the glass heat capacity $c_\infty(T)$ were extrapolated into the transition region and c_p normalized to

$$\tilde{c}(\omega,T) = \frac{c_p(\omega,T) - c_\infty(T)}{c_o(T) - c_\infty(T)}$$

The real and imaginary part of $\tilde{c}$ is presented in fig. 2 in a Cole-Cole plot. The segments belonging to different temperatures rall on the same mastercurve indicating that the principle of corresponding states (time - temperature equivalence) is valid; that is $\tilde{c}$ is of the form $\tilde{c}(\tilde{\omega} \cdot \bar{\tau}(T))$, $\bar{\tau}(T)$ being a kind of mean relaxation time /5/.

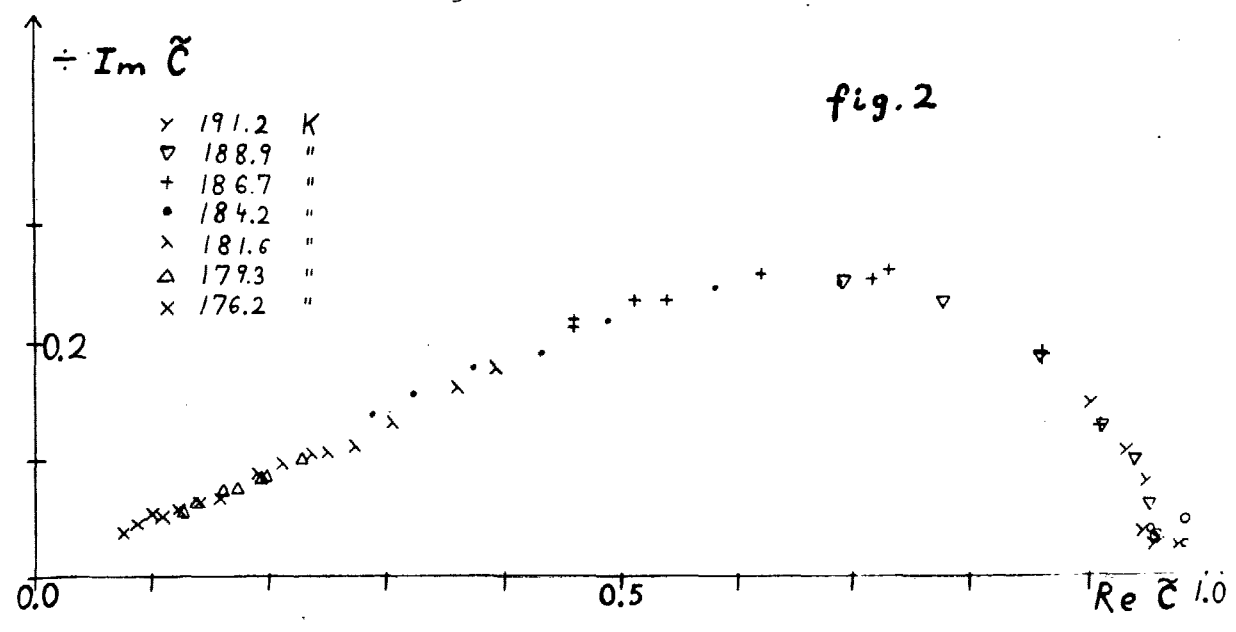

fig. 2

In the high frequency end the Cole-Cole plot is a straight line which shows $\tilde{c}(\tilde{\omega}) \propto (i\tilde{\omega})^{-\beta}$ $(= \tilde{\omega}^{-\beta} e^{-i\beta\pi/2})$ for $\omega \to \infty$. The angle of this line with the Re $\tilde{c}(\tilde{\omega})$ axis is $-\beta\pi/2$. β is found to be 0,28.
The time - temperature equivalence have now been used to make the mastercurve of fig. 3.
The various segments of $\tilde{c}(\tilde{\omega})$ measured at different temperatures were displaced along the $\log(\tilde{\omega})$ axis to form a single curve.
The displacements followed an Arrhenius law $\tau \propto \exp(\frac{A}{T})$ with $A = 2.1 \times 10^4$ K. At $T = 184.2$ the $\tilde{\omega}$-axis can be read in Hz.

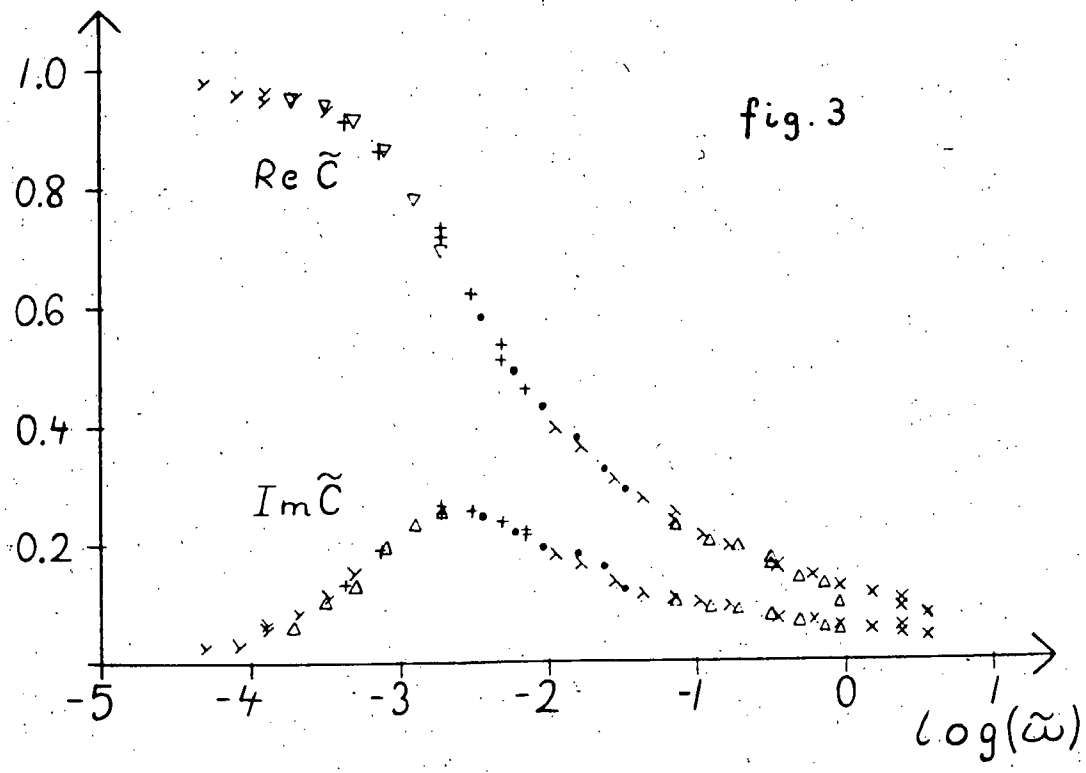

fig. 3

The intention of future work is to relate $\tilde{c}(\tilde{\omega})$ to other known responsefunctions at the glass transition.

Acknowledgement : The idea of measuring the frequency dependence of c grew out of discussions with Niels Boye Olsen and Jeppe Dyre, whom I would like to thank .

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A simple model of ac hopping conductivity

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Abstract - The physics of ac hopping conductivity in disordered solids is discussed in the light of a recently proposed model. Several new comments are made.

One of the most interesting facts about conductivity in disordered solids is the surprising similarity between ionic and electronic conductivity. The same kinds of non-linearities in strong electric fields are observed, and ionic and electronic conductivity have similar temperature and frequency dependence /1,2/. In a recent paper by the author it was suggested that the latter similarities are observed because ionic and electronic hopping conductivity have the same jump frequency distribution $p(\gamma)$ /3/. In the ionic case, the jump frequency γ is given by $\gamma \propto \exp(-\beta\Delta E)$ where β is the inverse temperature and ΔE is the energy barrier. In the case of tunneling electrons, γ is essentially given by $\gamma \propto \exp(-\alpha r)$ where α is the decay parameter for the wavefunctions and r is the jump distance. If ΔE resp. r are randomly varying, one finds in both cases that $p(\gamma)$ varies as $1/\gamma$. From a phenomenological point of view, ionic and electronic conductivity in disordered solids are thus claimed both to be characterized by randomly varying free energy barriers.

In order to check this hypothesis, the frequency-dependent conductivity $\sigma(\omega)$ must be calculated. In the CTRW approximation one finds /3-6/

$$\sigma(\omega) = \frac{nq^2a^2}{2dkT}\left\{-i\omega + i\omega\ln\lambda\left[\ln\left(\frac{1 + i\omega/\gamma_1}{1 + i\omega/\gamma_0}\right)\right]^{-1}\right\}. \qquad (1)$$

Here q and n are charge resp. density of the charge carriers, a^2 is the mean square jump distance in the x-direction, d is the dimensionality, k is the Boltzmann constant, T is the temperature, γ_1 and γ_0 are the smallest and largest jump frequencies, and $\lambda = \gamma_0/\gamma_1$. By letting γ_0 go to infinity the following expression is obtained where $\tau = 1/\gamma_1$ /3/

$$\sigma(\omega) = \sigma(0)\frac{i\omega\tau}{\ln(1+i\omega\tau)} \quad . \qquad (2)$$

This formula reproduces the qualitative features of ac hopping conductivity. In particular one finds an approximate power law of the conductivity at high frequencies, $\mathrm{Re}\,\sigma(\omega) \propto \omega^s$, where $s \cong 1 - 2/\ln(\omega\tau)$ /3/.

In the research field of ionic conductivity in glasses, the idea of randomly varying ion jump activation energies has been around since the fifties. At that time Stevels and Taylor suggested this as a reasonable hypothesis /7,8/ but they never actually calculated $\sigma(\omega)$. The idea was not generally accepted however, because it was thought to be inconsistent with the frequent observation of temperature-independent dielectric loss peaks /9,10/. But this argument is not correct. Equation (2) does indeed imply temperature-independent dielectric loss peaks. At the same time the activation energy for ac conductivity is smaller than that for dc conductivity (compare the discussion of the temperature-dependence of $\sigma(\omega)$ in ref. /3/). Thus there is no paradox, the point is that the relevant variable to consider is the conductivity and not the dielectric loss, at least when activation energies are discussed.

It has been known experimentally for several years that there is a connection between the dc conductivity and the dielectric loss peak frequency ω_0 /11,12/. Namikawa finds that most amorphous solids satisfy the following relation /12/

$$\sigma(0) = p\,\omega_0\,\varepsilon_0\,\Delta\varepsilon, \qquad (3)$$

where p is a numerical constant of order one, ε_0 is the vacuum permittivity, and $\Delta\varepsilon$ is the dielectric loss. The above model fits nicely into eq. (3). From eq. (2) one finds that $\Delta\varepsilon$ is given by

$$\varepsilon_0\,\Delta\varepsilon = \tfrac{1}{2}\sigma(0)\tau. \qquad (4)$$

When ω_0 is determined numerically, eq. (3) is found to be satisfied with $p = 0{,}42$.

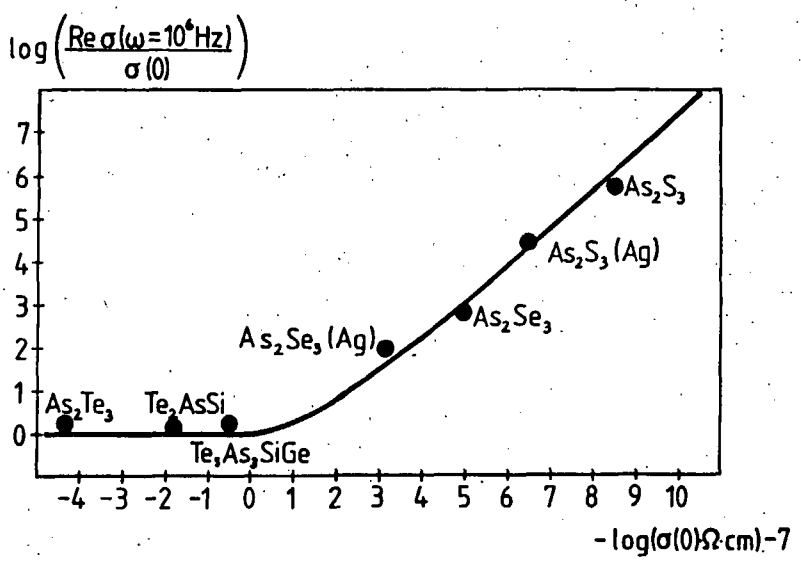

Fig. 1: This figure is a reinterpretation of an old figure (fig. 3 in ref. /13/) which shows a correlation for a number of chalcogenide glasses (at T = 300 K) between the dc conductivity and the ac conductivity at 10^6 Hz. The solid line shows the prediction of eq. (2) if it is assumed that the glasses have the same dielectric loss $\Delta\varepsilon$ (= 0,6).

In a famous paper, Davis and Mott plotted the dc conductivity as a function of $\mathrm{Re}\,\sigma(\omega = 10^6\ \mathrm{Hz}) - \sigma(0)$ at 300 K in a log-log plot for a number of chalcogenide glasses (fig. 3 in ref. /13/). Their figure reveils a correlation between the dc and the ac conductivity of these glasses. It will now be shown that this correlation can be interpreted within the above model. Suppose that the glasses have the same $\Delta\varepsilon$. Then it follows from eq. (4) that

$$\omega\tau|_{\omega=10^6\ \mathrm{Hz}} = K/\sigma(0) \qquad (5)$$

where K is independent of the glass *). If the ratio between the ac and the dc conductivity is plotted against $1/\sigma(0)$ in a log-log plot, the points are thus expected to lie on the curve which gives the real part of the conductivity (relative to $\sigma(0)$) as a function of $\omega\tau$. This is approximately the case as is clear from fig. 1. The constant K in eq. (5) is a fitting parameter; the value used in fig. 1 corresponds to $\Delta\varepsilon = 0{,}6$ - not an unreasonable value.

In the derivation of eqs. (1) and (2) the CTRW approximation was used. This approximation is known to be poor in some cases /15/. It may therefore be a good idea to try to apply a more reliable approximation, for instance the EMA /5/. If the jump frequency distribution is given by $p(\gamma) \propto 1/\gamma$, one finds that $\sigma(\omega)$ in this approximation in the limit $\gamma_0 \to \infty$ is determined by /16/ (for $d \geq 3$)

$$\frac{\sigma(\omega)}{\sigma(0)} \ln\left[\frac{\sigma(\omega)}{\sigma(0)}\right] = i\omega\tau' . \qquad (6)$$

Here τ' is a characteristic time of the same order of magnitude as τ. Equation (6) is easily solved numerically by means of the Newton-Raphson method. When the solution is plotted in a log-log plot it turns out to be almost indistinguishable from eq. (2), except for a rescaling of ω. Phenomenologically, the CTRW approximation is thus satisfactory in our use where $p(\gamma) \propto 1/\gamma$ and $\gamma_0 \to \infty$.

As the temperature goes to zero, the frequency-dependence of the conductivity becomes particularly simple in the model. In order to show this it is important to return to eq. (1) and keep γ_0 fixed throughout the calculation ($\gamma_0 \approx 10^{12}$ Hz). Assuming zero activation entropy we have

$$\gamma_1 = \gamma_0 e^{-\frac{\Delta E}{kT}} . \qquad (7)$$

When eq. (7) is substituted into eq. (1) one finds that the temperature-dependence cancels as $T \to 0$. The following results are obtained (for $\omega \ll \gamma_0$)

$$\lim_{T\to 0} \mathrm{Re}\,\sigma(\omega) = \frac{\pi}{4}\frac{nq^2a^2}{d\,\Delta E}\,\omega \sim \varepsilon_0\,\omega \qquad (8)$$

$$\lim_{T\to 0} \mathrm{Im}\,\sigma(\omega) = \frac{nq^2a^2}{2d\,\Delta E}\,\omega \ln\left[\frac{\gamma_0}{\omega}\right] . \qquad (9)$$

In particular we find that

$$\lim_{T\to 0} \frac{\mathrm{Im}\,\sigma(\omega)}{\mathrm{Re}\,\sigma(\omega)} = \frac{2}{\pi}\ln\left[\frac{\gamma_0}{\omega}\right] . \qquad (10)$$

Experimentally it is often found that the conductivity is almost proportional to the frequency at low temperatures with an only weakly temperature-dependent constant of proportionality. For instance, Long and coworkers find that $\mathrm{Re}\,\sigma(\omega) \propto \omega^{0,9}\,T^{0,4}$ for amorphous germanium at

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*) A similar scaling law has recently been discussed by Steve Summerfield /14/.

low temperatures /17/. After all, eqs. (8) and (9) can only be valid at very low temperatures. This is easy to see if it is remembered that the exponent s of the ac conductivity is given by $s \cong 1 - 2/\ln(\omega\tau)$ /3/. Substituting eq. (7) into this we find that (at ω fixed)

$$s \cong 1 - T/T_0, \quad T_0 = \tfrac{1}{2}\Delta E \tag{11}$$

at low temperatures.

Finally it will be shown that eq. (2) fits nicely into the framework proposed by Macedo et al. for the description of ionic conductivity in glasses /18/. Consider an infinite network as shown in fig. 2.

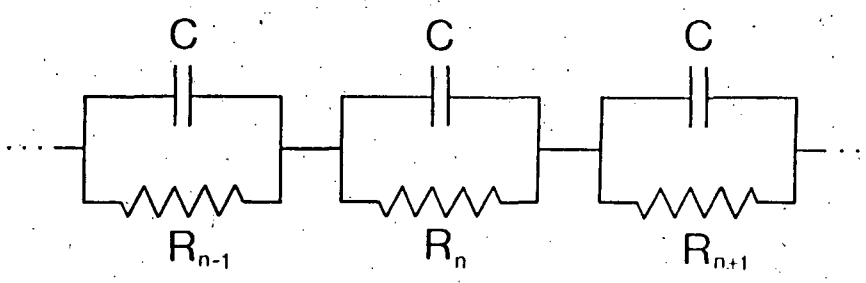

Fig. 2: Electrical equivalent-circuit used in the alternative derivation of eq. (2). The circuit was proposed by Macedo et al. to describe dielectric loss in ionically conducting oxide glasses /18/.

Each capacitance is equal to C, while the resistances R_n vary according to a probability distribution p(R). The electrical properties of the network are determined by the average impedance per RC-unit, $Z(\omega)$, which is given by

$$Z(\omega) = \left\langle \frac{1}{1/R + i\omega C} \right\rangle = \frac{1}{C}\int_0^\infty \frac{p(R)\,dR}{1/RC + i\omega} \,. \tag{12}$$

Suppose each RC-unit somehow corresponds to an activated process in the solid. If the free energies of activation vary randomly, the characteristic time $t = RC$ is distributed according to $p(t) \propto 1/t$. Equation (12) then becomes

$$Z(\omega) = \frac{K}{C}\int_0^\tau \frac{1}{1/t + i\omega}\,\frac{dt}{t} = \frac{K}{C}\int_0^\tau \frac{dt}{1 + i\omega t}, \tag{13}$$

where K is a constant and τ is the maximum value of t, the existence of which follows if a finite average dc conductivity is required. Because $p(t) \propto 1/t$ is not normalizable, the constant K is unknown and must be determined selfconsistently. When this is done after the integration has been carried out, eq. (13) reduces to eq. (2).

It must be emphasized that the physical interpretation of the electrical circuit is different from that of the above discussed "random walk" model. The circuit corresponds to macroscopic inhomogeneities in the solid, and in this case it is the overall potential difference which can be controlled experimentally. In the case of the random walk model, the fluctuation-dissipation theorem is used to derive $\sigma(\omega)$ from microscopic fluctuations. The implicit assumption is here that the local electric field is the experimentally controllable parameter. Thus, the electrical circuit of fig. 2 is not conceptually equivalent to the random walk model, although both gives rise to the same frequency-dependence of the conductivity.

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