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THE D.C. AND THE A.C. ELECTRICAL TRANSPORT IN AsSeTe SYSTEM

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Abstract

The electrical charge transport in AsSeTe glasses have been investigated by correlating the d.c. and the a.c. conductivity measurements performed at different temperatures.
It was possible to seperate the bulk properties from those at the insulator-metal interface. The deduced hopping parameters favour a single channel hopping model of charges (strongly localised through finite charge-lattice interaction) hopping in a narrow band of states. The importance of the knowledge of the space distribution of the charge in the contact region for the correct interpretation of the experimental results is stressed.

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The electrical charge transport in AsSeTe glasses have been investigated by correlating the d.c. and the a.c. conductivity measurements performed at different temperatures. It was possible to separate the bulk properties from those at the insulator-metal interface. The deduced hopping parameters favour a single channel hopping model of charges (strongly localised through finite charge-lattice interaction) hopping in a narrow band of states. The importance of the knowledge of the space distribution of the charge in the contact region for the correct interpretation of the experimental results is stressed.

1. INTRODUCTION

The electrical charge transport in the chalcogenide based

glasses is of considerable interest both from technological point of view as well as from the point of view of underlying physical principles governing the dynamics of the transport. There exist at present two different theoretical models which are usually used to interpret various experiments. Although both of these approaches assume charges to hop among localised states, they differ somewhat in the underlying physics (interactions) governing the transport. The first of the two models[1,2] assumes a multichannel transport of the charges in the rigid lattice (hopping in the band of localised states and extended transport at the band edges) while the other[3] is based on the finite effects of the charge-lattice interaction and involves only a single band of localised states. In order to help to distinguish between the two models when applied to $AsSe_xTe_{1-x}$ system, the d.c. electrical measurements were combined with a.c. admittance spectroscopy performed at different temperatures for two different compositions (x=0.6 and x=0.5). It will be shown that the combinaton of the d.c. and the a.c. measurements offers a new way how to determine the charge density and the charge mobility activation energies in the studied samples independently and selfconsistently. At the same time the bulk properties can be separated from those characterising the depleted contact regions when the samples are equipped with metal electrodes.

2. PREPARATION AND STRUCTURE

The glassy samples of $AsSe_xTe_{1-x}$ were prepared by first sealing the appropriate mixtures in the silica tubes, then annealing the assembly at 950° C for 8 hours and finally air

quenching the sealed melts to obtain corresponding amorphous ingots. The amorphous structure was verified through elastic neutron and X-ray scattering. The specimens for the experimental investigaton of transport properties were cut out from these ingots in form of thin discs and the sandwich geometry of a simple condenser was defined by evaporating gold electrodes on the two flat surfaces. In another set of experiments a conducting silver paste was used as electrode material in order to investigate the effect of diferent electrodes on the measured transport properties. All the measurements were performed in the liquid nitrogen cryostat with samples being placed in between two gold plated spring contacts.

3. D.C. ELECTRICAL CONDUCTIVITY

A typical temperature T dependence of the d.c. electrical conductivity $\sigma_{d.c.}$ for the two chosen compositons is shown in Fig.1. In all the studied samples the d.c. electrical transport was approximately simply activated with the activation energy somewhat dependent on the electrode material used (ΔE=0.554 eV - Au electrodes, ΔE=0.524 eV - silver paste electrodes in case of x=0.5 composition and ΔE=0.611 eV - Au el., ΔE=0.547 eV - silver paste el. in case of x=0.6 composition). The d.c. conductivity was found to be strongly dependent on the applied d.c. electrical bias. In order to investigate this dependence further and also to establish whether there was a finite contribution to the total conductivity from transport in the extended states (d.c. conductivity field independent up to the field strengths of 10^6 V/cm)[4], the

measurements of the d.c. electrical field dependence of the d.c. conductivity were also performed and the results are shown in Fig. 2. There are two distinguishable regimes in both compositions. The first highly non-linear region at low biases indicate the finite effect of the charge depleted contact layer on the d.c. el. transport (the effect being stronger for x=0.5 composition). In the second region the conductivity $\sigma_{d.c.}$ is exponentially dependent on the first power in the applied field indicating that the dominant transport process for both compositions is simple hopping of the charges among strongly localised levels with a well defined, temperature independent average hopping distance (nearest neighbour hopping). The slope of the curve log $\sigma_{d.c.}$ versus d.c. el. field E is in the case of constant distance hopping equal to $e.\langle r\rangle/k.T$ (e- elemental charge, k. Boltzmann constant, T- absolute temperature, $\langle r\rangle$- average hopping distance) and gives a value of $\langle r\rangle$ of approximately 7.5 Å with the predicted temperature dependence observed experimentally (calculation has been performed for the x=0.6 composition in view of the fact that the charge depleted contact region non-linearity extended to higher fields in the x=0.5 composition pushing the region where $\langle r\rangle$ could be determined easily to the higher fields where the samples began to switch). This is a very important result since it shows clearly that in neither of the two compositions is there experimental evidence for any sizable contribution to the transport from the conduction in the extended states. The d.c. transport seems to be dominated by the charge depleted contact regions at low d.c.

biases and by the simple nearest neighbour hopping throughout the entire sample at higher fields. Another important experimental result of the d.c. electrical measurements is concerned with the switching phenomenen. Switching was observed in some samples at the threshold fields of appr. 10^4 V/cm. The process was investigated at the values of the field near the threshold. In this case the on-state (some 4 orders of magnitude higher conductivity than the dark off-state conductivity) was of an intermittent nature in that it gave rise only to series of current spikes with the off-state current as a base-line. The spikes in the current vs. time trace at near (slightly above) threshold value of the d.c. field were random in nature, piling up as the d.c. field was further increased. This result shows that contrary to the previous models the switching in its initial phases is not caused by thermal effects, but rather that it is a highly non-linear phenomenon involving non-linear coupling of the charge to the lattice. It is possible that certain structural pre-cursor in the amorphous matrix is needed in order to initiate the fast transport process.

4. A.C. CONDUCTIVITY (Dielectric admittance spectroscopy)

To investigate further the nature of the transport process itself and the role played by the charge depleted contact regions in the d.c. experiments, the a.c. conductivity measurements were performed between 5 and 10^7 Hz at various temperature. Although the temperatures range covered was limited from below by the largest impedance (10^7 Ohm) still

measurable by the Hewlett-Packard model 4192A LF impedance analyser, it was wide enough to obtain sufficient information about the dynamical behaviour in the studied samples. The results of these measurements (using Au electrodes) are summarised in Figs. 3 to 6, where the log of the capacitance C (~ real part of the complex dielectric constant) and hte log of the conductance G (~ imaginary part of the complex dielectric constant) are displayed as functions of the measuring frequency for both compositions. By inspecting Fig. 3 a double relaxation can be identified for the x=0.6 composition (one at low frequencies at about 100-300 Hz and the second coming up at around 10^5 Hz). It should be pointed out that the double relaxation spectra have been observed for both compositions and seem to be a feature observed in general in high resistivity relaxation type semiconductors (also in ultra pure crystalline silicon) indicating that the phenomenon is independent of the nature of the transport process and the material but dependes only on such parameters as the resistivity, the geometry and contact metal material. Since the high frequency relaxation was occurring at the limit of the present measurements, it was not possible to obtain any quantitative information. Its possible origin will be discussed elsewhere[5], while here the attention will be focussed on the low frequency results. The following important experimental findings are worth pointing out before discussing the physical model:

1) The two conductance levels for both compositions moved up (down) with the increasing (decreasing) temperature at almost the same rate indicating that the same

transport process was operative.

2) When different electrode material was used, the lowest lying conductance level could change by more than an order of magnitude while the upper lying level scaled more or less with the geometrical factor.

3) As both conductance levels moved up with increasing temperature the transition between them moved towards higher frequencies (as can be best seen in the capacitance results). The movement of the transition region was simply activated with approximately the same activation energy as that of the d.c. conductivity, dependent somewhat on the electrode material used and on the composition (ΔE=0.664 eV - Au electrodes, ΔE=0.605 eV - silver paste electrodes in case of the x=0.5 composition and ΔE=0.619 eV - Au electrodes, ΔE=0.685 eV - silver paste electrodes in case of the x=0.6 composition). A typical temperature dependence of the characteristic time (frequency^{-1}) is shown in Fig.7. One more point should be mentioned in this context namely that larger was the difference between low and high frequency capacitance, smaller was the difference between the corresponding conductance levels.

4) When the levelling-off of the low frequency capacitance was seen experimentally (most clearly seen in Fig.6), a finite decrease in the limiting capacitance was observed with increasing temperature.

5. DISCUSSION

In the discussions of the dielectric admittance spectroscopy it is often assumed that the experimental results

such as depicted in Figs.3 to 6 reflect the dynamical behaviour of the various relaxation processes in the bulk of the material under study. Such an assumption is however difficult to reconcile with the present results where low frequency lying conductance level is clearly strongly dependent on the contact material used while the higher frequency level stays more or less the same. It is the main purpose of this paper to propose a qualitatively different interpretation based on the simple linear response of the passive linear a.c. network consisting of coupled constant (frequency independent!) resistors and capacitors (describing the sample) to applied external a.c. field. It is suggested that the present results and similar results in other materials reflect a completely static situation in homogeneous media (in thermodynamic equilibrium at each measured frequency) with rising (charge depleted region) or decreasing (charge accumulation region) resistance near the contact regions (but also near free surfaces!). The change in the resistance is caused by the charge transfer because of the difference in the work functions of the contacting metal and the studied insulator/semiconductor (somewhat different arguments apply for free surfaces) and the precise resistance ($\simeq$ 1/charge density) profile is determined through Maxwell's field equations[5] (Poisson's equation sufficient in this case). *It is therefore the static* (time independent) *charge density changing in space and not the microscopic mobility changing in time* that gives rise to the observed experimental results. Because the classical

electrodynamics field equations which govern most of the presently discussed phenomena work with conductivities (charge density and the charge mobility lumped together in a product) this important qualitative difference has been overlooked in the past. The characteristic thickness of the charge depleted contact region can be roughly approximated by the Debye screening length[5]

$$d = \sqrt{\frac{\varepsilon_o \cdot \varepsilon_r \cdot kT}{N \cdot e^2}} \qquad (1)$$

($\varepsilon_o \cdot \varepsilon_r$ - static dielectric constant, N- charge density). The thickness of the charge depleted region and the total sample thickness are the two characteristic lengths of the problem. In the framework of the presented model it now becomes clear why the relaxation phenomenon discussed here should depend only on such parameters as the geometry of the sample, its resistance, the nature of the contacting metal and the charge depleted region thickness and should be independent of the microscopic transport process (microscopic mobility).

6. THE MODEL

In view of the above presented discussion the sample can be modelled as a lumped circuit consisting of a large number of elements each element representing a finite slice of the sample characterised by its own resistance (decreasing with distance down to the bulk resistance) and the corresponding geometrical capacitance both connected in parallel.

Alternatively one can, for the purpose of the qualitative discussion and in cases where the ratio of the sample thickness to the charge depleted region thickness becomes large, lump the whole of the charge depleted region into one element coupled to the second element representing the bulk. With this reticulation of the sample it is relatively an easy task to interpret the physical meaning of the measured conductances and the capacitances in Figs. 3 to 6. The large value of the low frequency capacitance corresponds to the charge depleted region capacitance which is larger than the bulk capacitance by the inverse ratio of the Debye screening length d to the total sample thickness minus d. Correspondingly, the low value of the conductance at these low frequencies is identified with the high resistance of the charge depleted contact region. The characteristic bulk properties with temperature dependent conductivity and constant bulk capacitance are seen first at higher frequencies. To interpret the d.c. measurements as representative of the bulk would be therefore quite wrong. Approximating the sample by two elements (contact region and the bulk), the expressions for the real and the imaginary part of the admittance are relatively simple

$$G = \frac{1}{R_S+R_B} \cdot \frac{1+\omega^2[\tau(\tau_B+\tau_S)-\tau_B.\tau_S]}{1+\omega^2\tau^2}$$

$$C = \frac{1}{R_B+R_S} \cdot \frac{\tau_B+\tau_S-\tau(1-\omega^2\tau_B\tau_S)}{1+\omega^2\tau^2} \qquad (2)$$

where $\tau = (C_B+C_S)\frac{R_S R_B}{R_S+R_B}$

(here $\tau_B = R_B \cdot C_B$ - bulk element, $\tau_S = R_S \cdot C_S$ - charge depleted region element and τ is the characteristic time of the transition between the two conductance levels), with the limits $G(\omega << \tau) \rightarrow 1/R_B + R_S$ and $C(\omega >> \tau) \rightarrow C_S \cdot C_B/(C_S + C_B)$. For the composition x=0.6 this very crude reticulation was already sufficient to model successfully (by an analogue circuit) the experimental results as is shown by dashed lines in Figs.3 and 4. For the composition x=0.5 in order to reproduce the slopes of the transition region more closely, the contact region had to be simulated by three elements with exponentially decreasing resistance (the results of the analogue circuit simulation is shown by dashed lines in Figs. 5 and 6). Further simulations were performed numerically using ten and hundred elements and a very satisfactory agreement with experiments was obtained ("free" parameters being d (eq.(1)) and R_B-the bulk resistance). From the numerical simulations it was found that the slopes in the transition regions were quite sensitive to the exact form of the resistance distribution and that a simple exponential decrease of the resistance with thickness in the charge depleted contact region gave the best agreement with the experiment. The difference in the low frequency capacitance between x=0.6 and x=0.5 compositions indicates that the charge depleted region in the x=0.6 composition is appreciably smaller ($\simeq$ 1-2μ) than in the x=0.5 composition ($\simeq$ 20μ). This difference is believed to be due to the difference in the position of the hopping bands relative to the Fermi level of the contacting metal rather than due to a large diffe-

rence in the charge densities in the two compositions. It has to be stressed that throughout the discussion the capacitance of the medium was assumed to be un-affected by the resistance changes (through possible changes of the dielectric constant) and that the medium conductivity was time (frequency) independent and was of a usual form

$$\sigma = N.e.\mu \ , \quad \text{where } N=N_o \cdot e^{-\Delta E_1/k.T} \text{ and } \mu=\mu_o e^{-\Delta E_2/k.T}. \quad (3)$$

(N - charge density, μ - charge mobility).

The proposed model has one interesting consequence. It is the possibility of independent determination of the charge density and the charge mobility in the relaxation type semiconductors. This possibility rests on the fact that while the thickness of the charge depleted region (as measured by low frequency capacitance) d is proportional to $1/\sqrt{N}$ (eq.(1)), the bulk conductivity involves the product of both the charge density and the mobility. In the materials where the charge density is constant and independent of temperature (such as crystalline doped silicon in the extrinsic temperature region), no change in the low frequency capacitance is predicted. In those materials on the other hand where the mobility is temperature independent and where all of the major temperature dependence of the conductivity is due to the temperature dependence of the charge density (ultra pure crystalline silicon in the intrinsic temperature region) a simple square root scaling should exist between the change in the bulk conductivity and the thickness of the charge depleted contact region. The present case lies between these two limits

since the conductivity is given by (3). With the physical parameters appropriate to the x=0.5 composition (the decrease of the low frequency capacitance with increasing temperature most apparent) it was possible to determine the two activation energies in eq. (3) independently. The deduced values were 0.253 eV for the activation energy of mobility and 0.267 eV for the activation energy of the charge density. These values are quite consistent with the values found in other chalcogenide based glasses and together with the d.c. measurements give yet another supporting evidence for a single channel hopping transport model involving charges strongly localised through appreciable charge-lattice interaction. The important point here is that the evidence comes in a sense from a qualitatively new experiment (or new experimental technique).

7. CONCLUSION

It has been demonstrated that in the high resistivity materials such as $AsSe_xTe_{1-x}$ system of the present investigation, the d.c. measurements do not reflect necessarily the bulk properties but can be completely dominated by the charge depleted (accumulation) contact regions effects. Using the d.c. measurements in combination with a.c. admittance spectroscopy it was possible to separate the effects of these two regions and to describe them quantitatively. It has been stressed that the observed a.c. behaviour stems from the spatial distribution of the charge density within the contact

regions and not, as it is usually assumed, from the various relaxation time distributions entering the microscopic conductivity. For the system studied in the present investigation a single channel hopping of charges strongly localised through finite charge-lattice interaction was deduced with the following hopping parameters:

1) The conductivity σ is of the form as described by the Eq.(3) giving the specific resistivities at 70°C of $\rho = 5.35 \cdot 10^5$ Ωcm and $\rho = 5.6 \cdot 10^6$ Ωcm for the composition x=0.5 and x=0.6 respectively (compare with Fig.1).

2) Average hopping distance is approximately 7.5 Å.

3) Activation energy of the mobility (x=0.5 composition) is $\Delta E_1 = 0.253$ eV, the hopping mobility being of the form $\mu[cm^2V^{-1}SEC^{-1}] = 0.3 \exp(-0.253 \text{ eV}/kT)$.

4) Activation energy of the charge density (x=0.5 composition) is $\Delta E_2 = 0.267$ eV, the charge density itself folloving $N[cm^{-3}] = 1.34 \cdot 10^{21} \cdot \exp(-0.267 \text{ eV}/kT)$ relationship.

8. ACKNOWLEDGEMENTS

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FIGURE CAPTIONS

Figure 1 : A typical temperature T dependence of the d.c. electrical conductivity $\sigma_{d.c.}$ in the two $AsSe_xTe_{1-x}$ compositions.

Figure 2 : The d.c. electrical field E dependence of the d.c. electrical conductivity $\sigma_{d.c.}$ in the two $AsSe_xTe_{1-x}$ compositions.

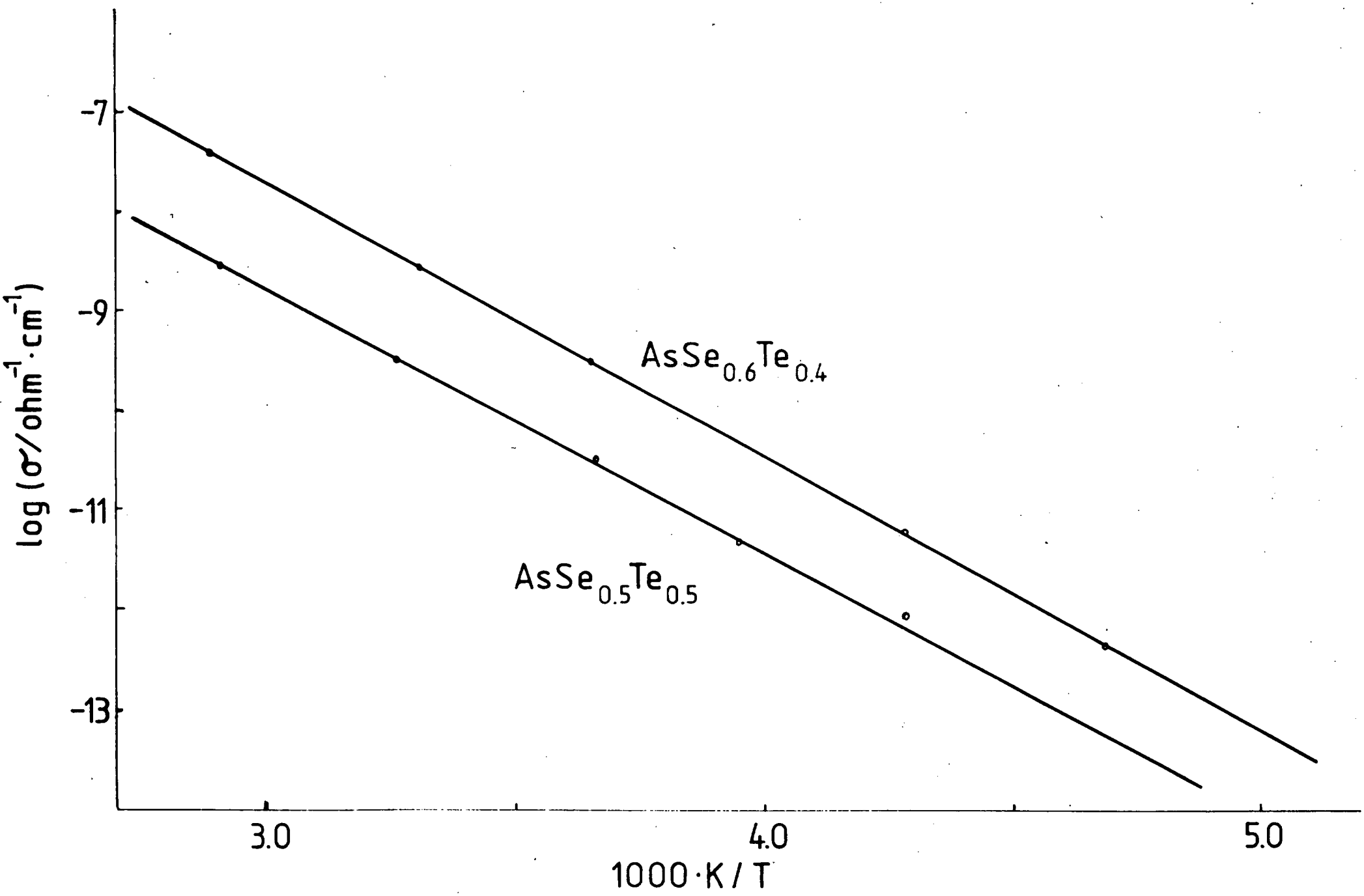
Figure 3 : The frequency dependence of the conductance G in the $AsSe_{o.6}Te_{o.4}$ composition at various temperatures.

Figure 4 : The frequency dependence of the capacitance C in the $AsSe_{o.6}Te_{o.4}$ composition at various temperatures.

Figure 5 : The frequency dependence of the conductance G in the $AsSe_{o.5}Te_{o.5}$ composition at various temperatures.

Figure 6 : The frequency dependence of the capacitance C in the $AsSe_{o.5}Te_{o.5}$ composition at various temperatures.

Figure 7 : A typical temperature T dependence of the characteristic time τ in the two $AsSe_xTe_{1-x}$ compositions.


-7
-9
-11
-13
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AsSe0.5Te0.5
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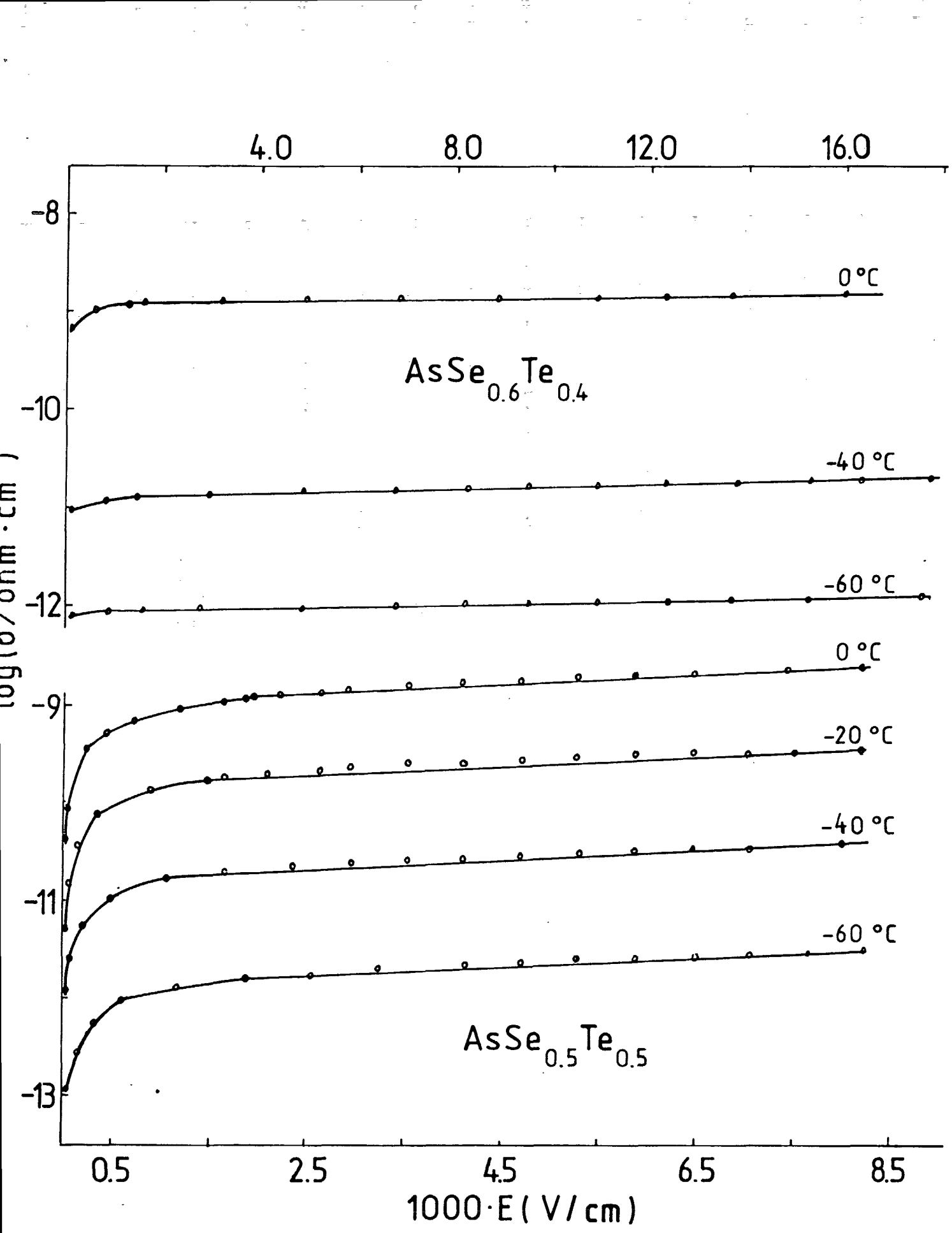
4.0
5.0
1000·K / T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8.0
12.0
16.0
-8
-10
-12
-9
-11
-13
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0 °C
-20 °C
-40 °C
-60 °C
AsSe0.6Te0.4
AsSe0.5Te0.5
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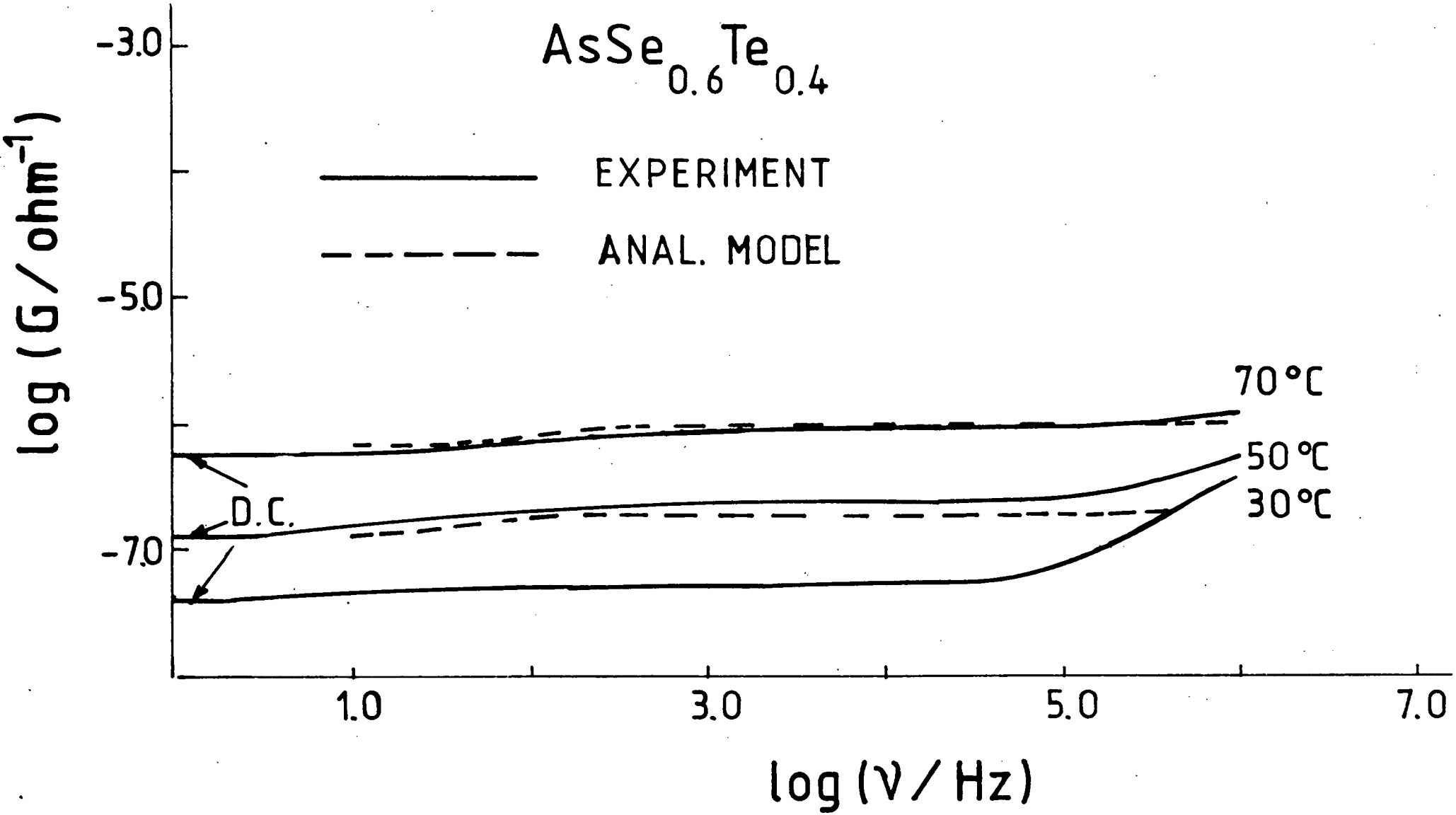
2.5
4.5
6.5
8.5
1000·E (V/cm)
log(σ/ohm·cm)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EXPERIMENT
ANAL. MODEL
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50 °C
30 °C
D.C.
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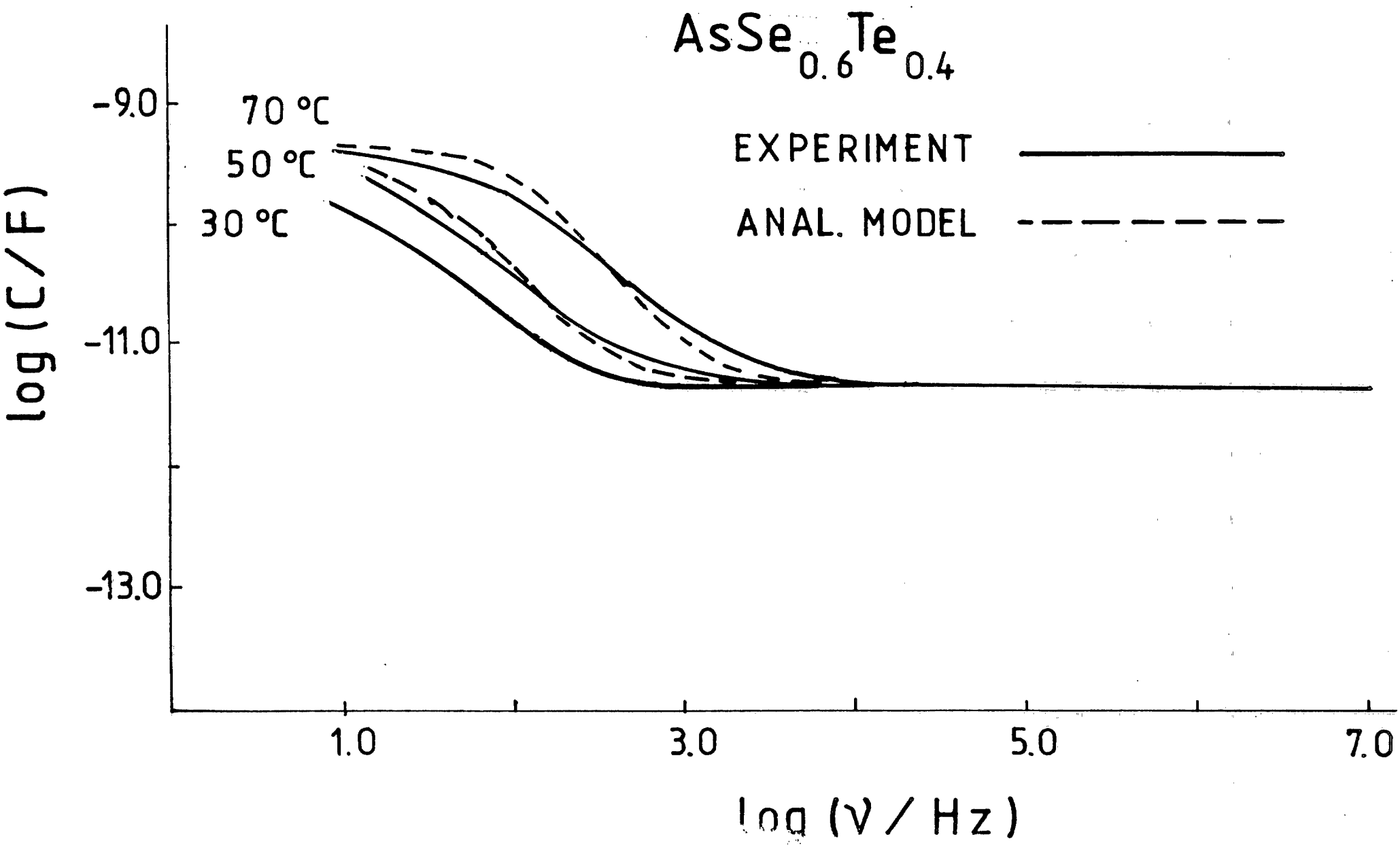
3.0
5.0
7.0
log (ν / Hz)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50 °C
30 °C
EXPERIMENT
ANAL. MODEL
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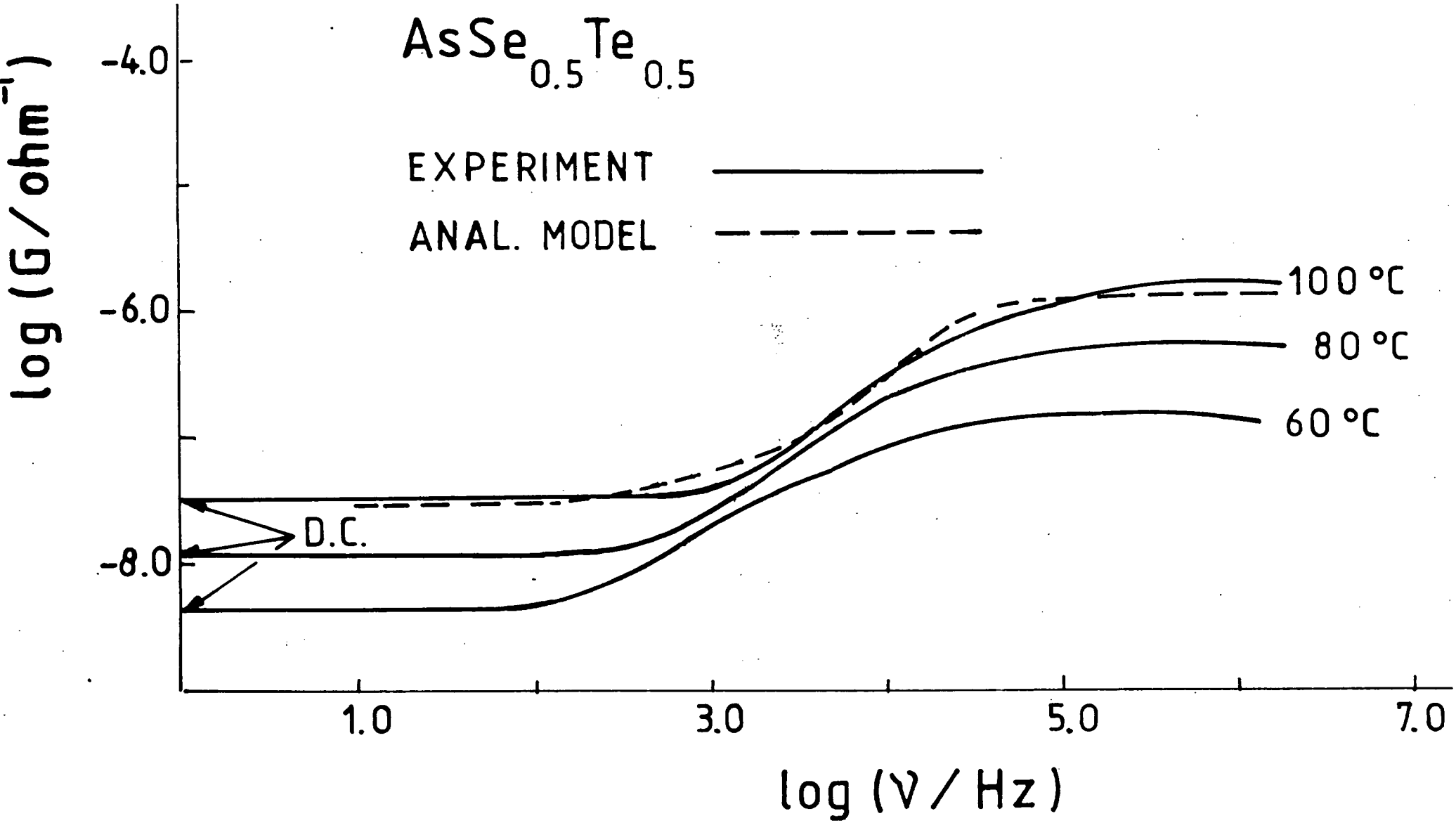
1.0
3.0
5.0
7.0
log (ν / Hz)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ANAL. MODEL
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D.C.
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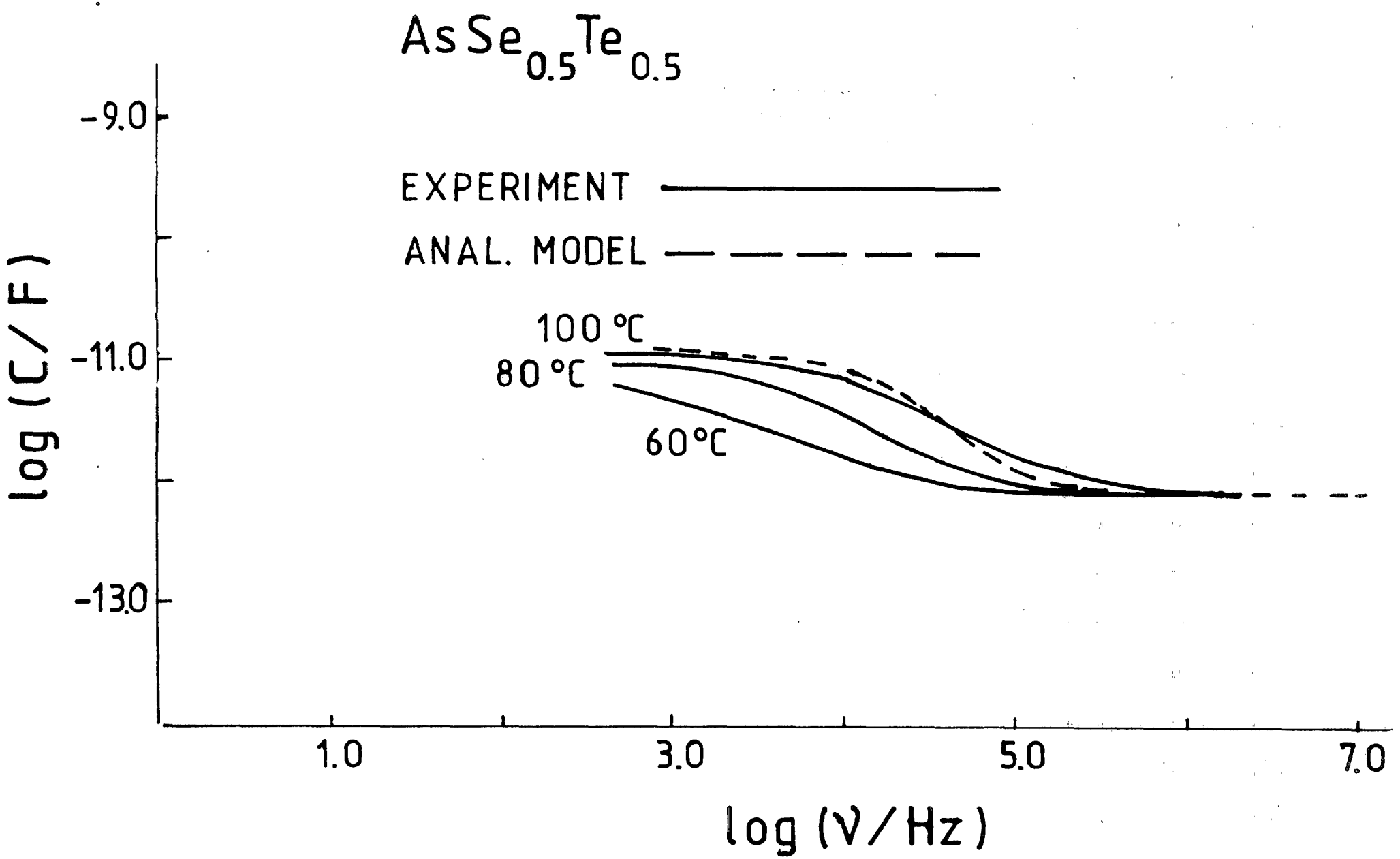
5.0
7.0
log (ν / Hz)


AsSe0.5Te0.5
EXPERIMENT
ANAL. MODEL
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80 °C
60°C
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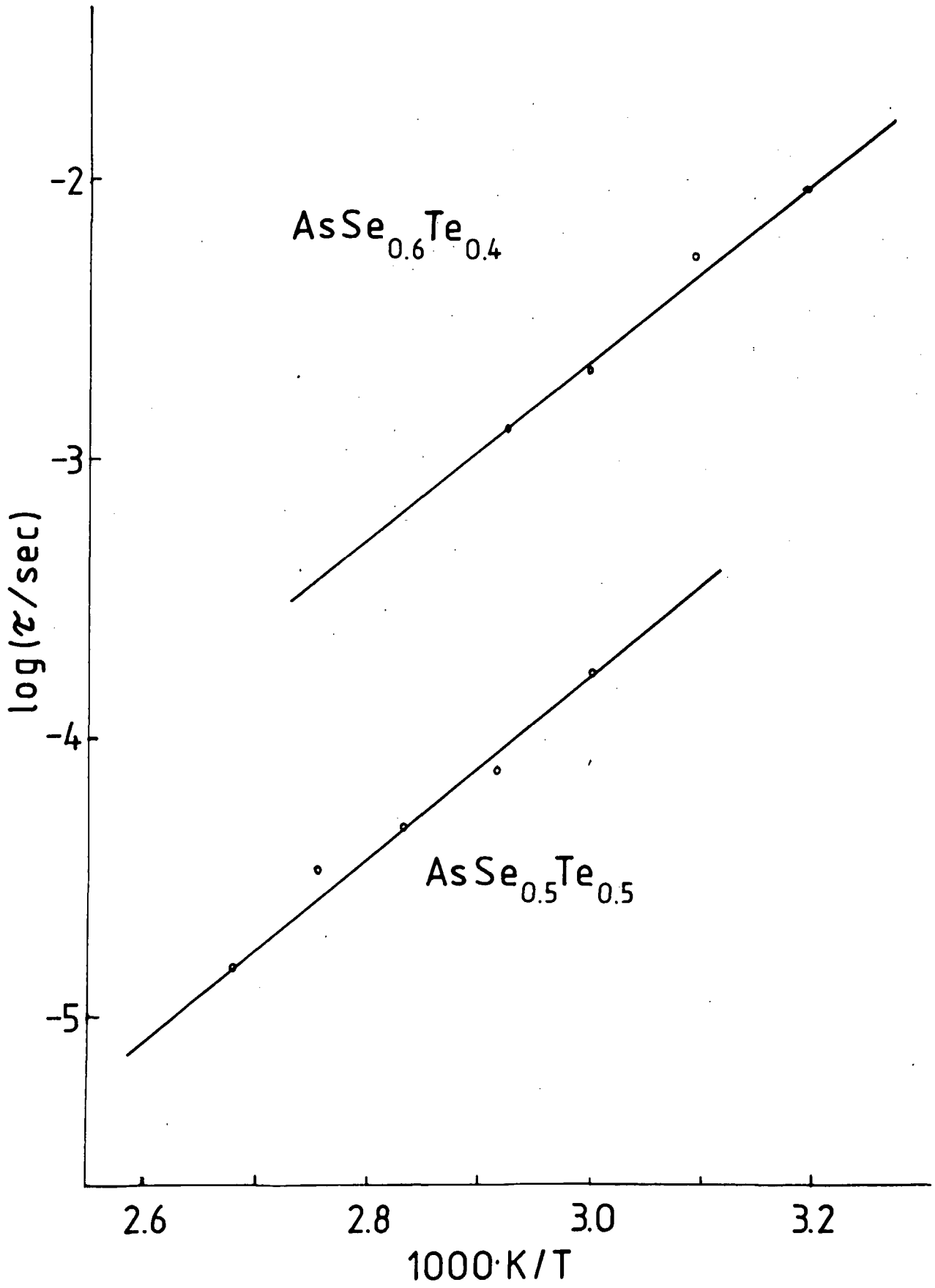
5.0
7.0
log (V/Hz)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-3
-4
-5
log(τ/sec)
AsSe0.6Te0.4
AsSe0.5Te0.5
2.6
2.8
3.0
3.2
1000·K/T

THE D.C. AND THE A.C. ELECTRICAL TRANSPORT IN AsSeTe SYSTEM

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1. D.C. ELECTRICAL CONDUCTIVITY

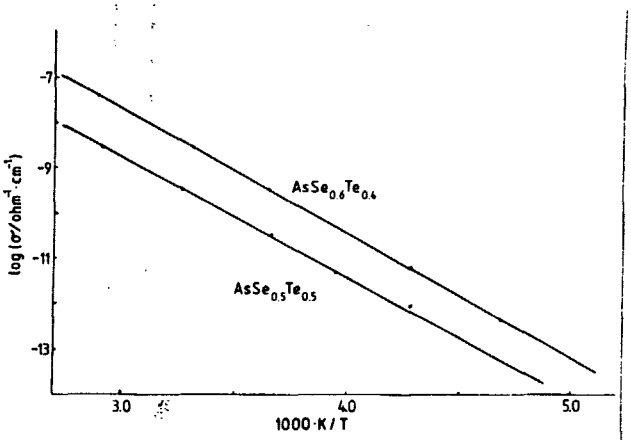

FIGURE 1

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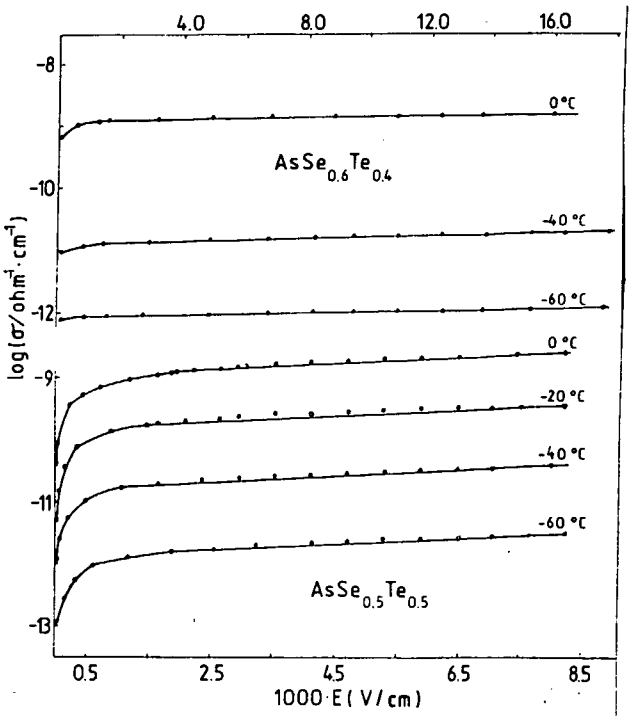

FIGURE 2

stant distance hopping (e-elemental charge, k- Boltzmann constant, T- absolute temperature, <r>- average hopping distance) and gives a value of <r> of approximately 7.5 Å with the predicted temperature dependence observed experimentally. This is a very important result since it shows clearly that in neither of the two compositions is there experimental evidence for any sizable contribution to the transport from the conduction in the extended states.

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To investigate further the nature of the transport process itself and the role played by the charge depleted contact regions in the d.c. experiments, the a.c. conductivity measurements were performed between 5 and 10^7 Hz at various temperatures. The results of these measurements (using Au electrodes) are summarised in Figs. 3 to 6. By inspecting Fig. 3 a double relaxation can be identified for the x=0.6 composition.

It should be pointed out that the double relaxation spectra have been observed for both compositions and seem to be a feature observed in general in high resistivity relaxation time semiconductors (also in ultra pure crystalline silicon). The high frequency relaxation was occurring at the limit of the present measurements and therefore it was not possible to obtain any quantita-

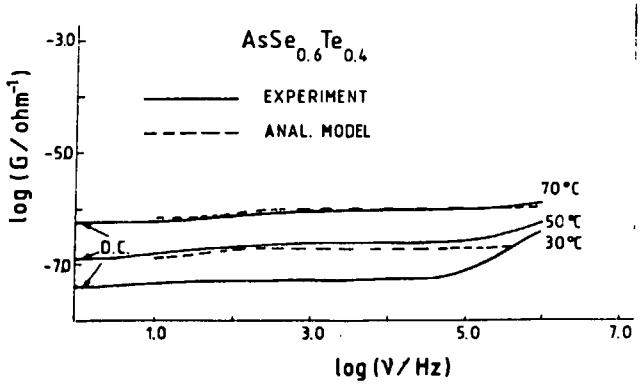

FIGURE 3

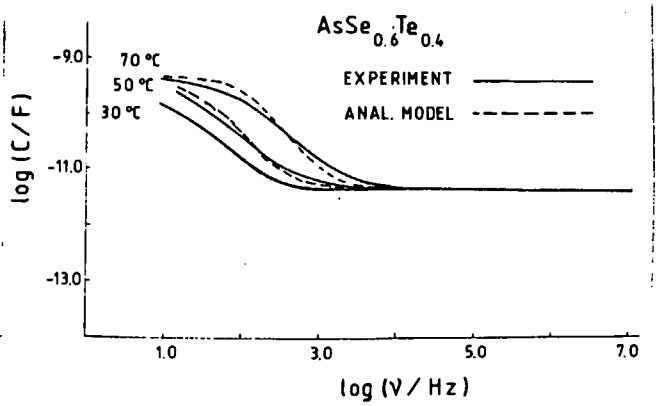

FIGURE 4

tive information. Its possible origin will be discussed elsewhere[2], while here the attention will be focussed on the low frequency results.

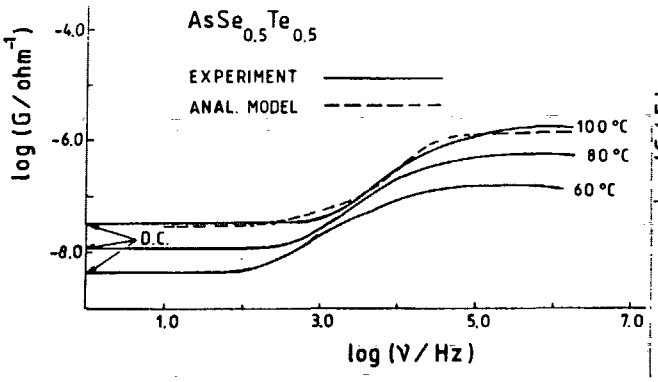

FIGURE 5

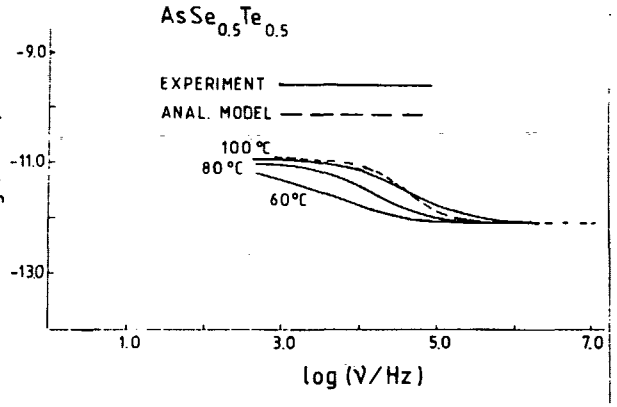

FIGURE 6

3. MODEL

In the discussions of the dielectric admittance spectroscopy it is often assumed that the experimental results such as depicted in Figs. 3 to 6 reflect the dynamical behaviour of the various relaxation processes in the bulk of the material under study. Such an assumption is however difficult to reconcile with the present results where the low frequency lying donductance level was clearly strongly dependent on the contact material used while the higher frequency lying level stayed more or less the same. It is the main purpose of this paper to propose a qualitatively different interpretation.

It is suggested that the present results and similar results in other materials reflect a completely static situation in homogeneous media with rising (charge depleted region) or decreasing (charge accumulation region) resistance near the contacts. *It is therefore the static* (time independent) *charge density changing in space and not the microscopic mobility changing in time* that gives rise to the observed experimental results. The sample can be modelled as a lumped circuit consisting of a large number of elements each element representing a finite slice of the specimen characterised by its own resistance (decreasing with distance down to the bulk resistance) and the corresponding geometrical capacitance, both connected in parallel. Alternatively one can, for the purpose of qualitative discussion and in cases where the ratio of the sample thickness to the charge depleted region thickness d becomes large, lump the whole of the charge depleted region into one element coupled to the second element representing the bulk. For the composition x=0.6 this very crude retardation was already sufficient to model successfully (by an analogue circuit) the experimental results as is shown by the dashed lines in Figs. 3 and 4. For the composition x=0.5 in order to reproduce the slopes of the transition region more closely, the contact region had to be simulated by three elements

with exponentially decreasing resistance (the results of the analogue circuit simulation are shown by dashed lines in Figs. 5 and 6). Further simulations were performed numerically using ten and hundred elements and a very satisfactory agreement with experiments was obtained ("free" parameters being d and R_B-the bulk resistance). The difference in the low frequency capacitance between x=0.6 and x=0.5 compositions indicated that the charge depleted region in the x=0.6 composition is appreciably smaller ($\simeq 1-2\mu$) than in the x=0.5 composition ($\simeq 20\mu$). This difference is believed to be due to the difference in the position of the hopping bands relative to the Fermi level of the contacting metal rather than due to a large difference in the charge densities in the two compositions. The proposed model has one interesting consequence. It is the possibility of independent determination of the charge density and the charge mobility in the dielectric relaxation time-type semiconductors. This possibility rests on the fact that while the capacitance of the charge depleted region is approximately proportional to $1/\sqrt{N}$, the bulk conductivity involves the product of both the charge density N and the mobility. With the physical parameters appropriate to the x=0.5 composition (the decrease of the low frequency capacitance with increasing temperature most apparent) it was possible to determine a value of 0.253 eV for the activation energy of mobility and 0.267 eV for the activation energy of the charge density.

4. CONCLUSION

It has been demonstrated that in the high resistivity materials such as $AsSe_xTe_{1-x}$ system of the present investigation, the d.c. measurements do not reflect necessarily the bulk properties but can be dominated completely by the charge depleted (accumulation) contact region effects. Using the d.c. measurements in combination with a.c.admittance spectroscopy it was possible to separate the effects of these two regions and to describe them quantitatively. For the system studied in the present investigation a single channel hopping of charges strongly localised through finite charge-lattice interaction was deduced.

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