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Frequency-dependent dielectric behavior of amorphous silicon thin films

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The admittance $Y = G + j\omega C$ of amorphous silicon films, produced by electron-beam evaporation, has been measured at frequencies between dc and 10^7 Hz, and at temperatures between 150 and 400 K. At low frequencies the conductance G was nearly independent of frequency, an exponential function of temperature, and influenced by contacts. At medium frequencies G increased almost proportionally to ω^s, with $s \simeq 0.9$. In this region G was found to be contact independent and superlinearly dependent on temperature. In the high-frequency region a saturation behavior of G was observed. The capacitance C decreased monotonically from low to high frequencies. These measured properties have been attributed to a hopping between sites, randomly distributed in space and energy, and over the barriers separating the sites. The barrier height has been correlated to the intersite separation by assuming Coulomb-like potential wells. This model accounts both for the ω^s behavior and for the saturation of G. However, to obtain good agreement between theory and experiment it was necessary to envisage a minimum relaxation time τ_o not much smaller than 10^{-6} sec. In contrast, τ_o has in previous work been interpreted to be of the order of an atomic vibrational period ($\sim 10^{-12}$ sec).

I. INTRODUCTION

A dependence of the real part σ' of the complex conductivity σ on the angular frequency ω in the form $\sigma' \propto \omega^s$ (where $s \lesssim 1$) can be observed in various amorphous semiconductors[1] and, indeed, in a multitude of other materials.[2,3] In particular, as far as tetrahedrally bonded amorphous semiconductors are concerned, there is experimental evidence that thin amorphous films, even though prepared in different ways, exhibit this behavior. The examples include Ge,[4-9] Si prepared by glow discharge decomposition of SiH_4,[10,11] Si prepared by sputtering,[12] and the Si layers discussed below, prepared by electron-beam evaporation.

Since such a frequency response is common to a very wide range of materials, with entirely different molecular structures, and since it may in principle be due to either electronic or ionic movement, the microscopic mechanism of the effect cannot be uniquely identified from the observations themselves. Experimental data have been analyzed on the basis of different models; a summary is given by Jonscher[3] and by Pollak.[13] Amongst other things, it has been shown that the superposition of randomly distributed activation energies gives rise to $\sigma' \propto \omega$ in a certain frequency range.[13-16] In *crystalline n*-type Si a $\sigma' \propto \omega^{0.8}$ behavior was observed by Pollak and Geballe[17] at temperatures near absolute zero, and was explained as a phonon-assisted tunneling of charge carriers between spatially localized states. This model has been shown to be in satisfactory agreement with experiment.[17] Austin and Mott,[18] and also other authors,[19,20] adapted this theory to amorphous semiconductors. However, in several cases these models (tunneling models) have been proved incompatible with experimental facts.[9,21-24] An alternative model, addressed in particular to chalcogenide glasses, has been proposed by Elliott.[22,23] His theory is based on the classical hopping model of Pike[24] for scandium oxide, and has been shown to be in better agreement with experiments than the models mentioned above. It will be one of the purposes of the present paper to examine the applicability of these theories to the ac behavior of the amorphous Si (a-Si) films investigated.

The results presented in Sec. III below describe in the main a $\sigma' \propto \omega^{0.9}$ behavior, but at frequencies around 100 kHz some unexpected saturation effects are observed. A model is developed to account for the $\omega^{0.9}$ term as well as for the saturation (Sec. IV). However, these matters are in need of confirmation.

II. EXPERIMENTAL PROCEDURE

To obtain capacitances and conductances of reasonable values, our experiments were performed on samples with a sandwich structure, the a-Si layer being sandwiched between two electrodes. Several identical sandwich devices, and one planar device (gap sample) for comparative measurements, were deposited on the same substrate (Corning glass 7059).

The a-Si films were produced by evaporating polycrystalline high-purity Si with a 1 kW electron-beam gun, and condensing the Si vapor on the substrate heated to 100 °C. After deposition, the films were annealed *in situ* for two hours at 300 °C. Standard photolithographic techniques were used to obtain the device geometry, and the amorphous character of the Si films was verified by x-ray diffraction. An estimate of oxygen content was obtained by Auger spectroscopy, and yielded approximately 1% at the surface. This value decreased to about 0.5% at 100 Å inside the film, and to values undetectably low (<0.3%) some hundred Å away from the surface.

The evaporation parameters were systematically varied between the following limits: Evaporation rate 0.5–5 Å/ sec, vapor pressure during evaporation 10^{-6}–10^{-3} Torr. Further details of preparation may be found elsewhere.[25]

The effect of substrate conductance on the measurements was shown to be negligible, and the influence of contacts was tested in three ways: (i) by using various combinations of the metals Au, Al, Mo, and Cr as contact materials; (ii) by comparative measurements on planar devices of much greater interelectrode spacing (1 mm); and (iii) by testing the field dependence of the admittance. In the gap system the bulk contribution to the total resistance is at least two orders of magnitude larger than in sandwich systems, which justifies neglecting the electrode effects. It should be noted that a-Si is stable only in the form of thin films,[26] and that the electrical properties are very sensitive to preparation.[27,28] It is therefore impracticable to vary the film thickness for the purpose of monitoring contact effects.

The voltage-current relationship of the specimens as a whole was nonlinear, but care was taken to perform all measurements within the linear range $V<kT/e$ near the origin. The applied voltages were thus in the mV range, yielding currents of the order of nA at room temperature, smaller at low temperatures.

At frequencies up to 100 kHz data were obtained by current-voltage-phase angle measurements: From dc to 1 kHz with a high-impedance (10^{11} Ω) and low-bias current (0.5 pA) operational amplifier; overlapping this range, from 10–100 kHz by correlative measurements with a lock-in-amplifier (PAR 124). Above 500 kHz a vector-impedance-meter (HP 4815) was employed. Measurements at frequencies higher than 10 MHz proved to be meaningless, due to the inductance of the sample connection leads. During the measurements the substrate with the samples was mounted in a temperature chamber, in which a pressure lower than 10^{-3} Torr was maintained. The parallel (stray) capacitance was about 0.5 pF. The sample temperature was measured with a small thermocouple, glued onto the substrate surface with silver paste. Various admittances, formed by known capacitors and known high-value resistors, were connected in place of the sample in order to check the capability of the measurement system.

The Si layer of all the sandwich and gap samples investigated here had the same thickness (6500 Å). The conducting area of the a-Si film, determined by the overlap of bottom and top electrodes of the sandwich samples, had the dimensions 0.6 × 0.4 mm.

III. EXPERIMENTAL RESULTS

Because of contact effects, it is not in general permissible to translate the presented admittance data $Y=G+j\omega C$, directly into conductivities, but such a translation is justified for the gap specimens, and even for the sandwich specimens at high frequencies. Table I embodies the appropriate geometrical factors.

Typical data for admittance components G and C are shown in Fig. 1. The conductance can be expressed as $G_{dc}+G_{ac}$. At low frequencies G is evidently determined by the frequency independent term G_{dc}, which is influenced by the contact material. Thus G_{dc} values differ by one order of magnitude for samples with Mo/Cr and Cr/Cr electrodes (the first material indicates the bottom electrode). The difference was usually smaller between samples with other contact materials. In contrast, the subsequent ac term G_{ac} was always observed to be nearly contact independent. Furthermore, the temperature dependence of G_{ac} was much weaker than that of G_{dc}. Due to the relatively high conductance of the samples at low frequencies the capacitance could not be determined with any degree of accuracy (or not at all), and the form of the C curves of Fig. 1 thus permit no conclusion as to whether or not the contacts influence the capacitance.

Different preparation conditions and different thicknesses of the a-Si film influenced G and C quantitatively, but did not change their characteristics. The measured admittance properties are therefore regarded as characteristic of the atomic disorder in the a-Si, and not as the result of contamination or film geometry.

Comparative measurements on a planar device are presented in Fig. 2. At frequencies above 1 kHz the C curve tends to approach a value of about 0.5 pF, which is determined by stray capacitances, as previously mentioned. The character of the conductance is similar to that of the sandwich samples of Fig. 1. There is a difference for the

TABLE I. Correspondence between admittance and conductivity values.

	Sandwich samples	Gap sample
Admittance	1 Ω^{-1}	1 Ω^{-1}
Conductivity	27.4×10^{-3} $(\Omega\,\mathrm{cm})^{-1}$	1.9×10^{3} $(\Omega\,\mathrm{cm})^{-1}$

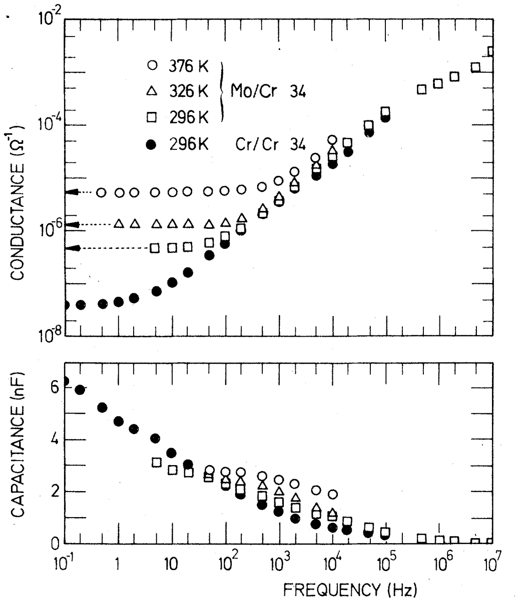

FIG. 1. Conductance G and capacitance C of two sandwich samples, prepared in the same way but with different base electrodes. The arrows indicate the *dc* conductances.

gap sample due to the high value of G_{dc}, which suppresses the ac conductance G_{ac} up to 10 kHz. This confirms our conclusion that the G_{dc} values measured on sandwich samples are strongly influenced by contact effects.

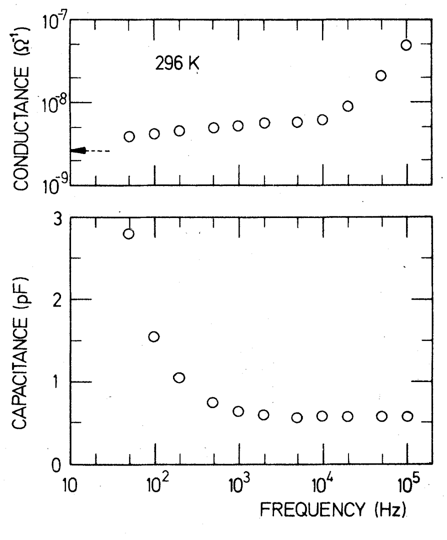

FIG. 2. Conductance G and capacitance C of a planar (gap) sample. The arrow indicates the dc conductance.

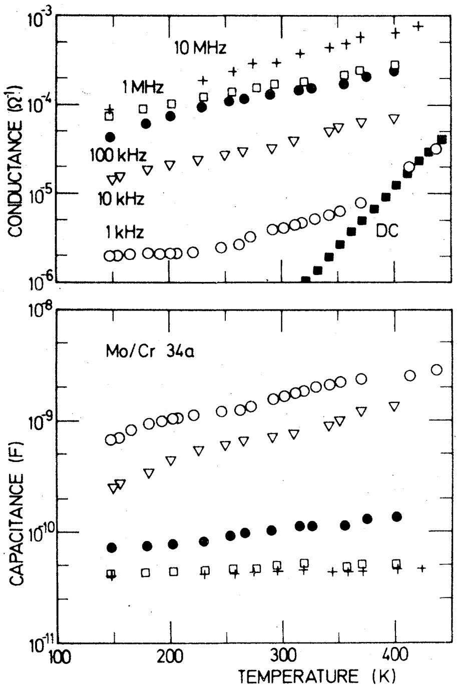

FIG. 3. Temperature dependence of conductance G and capacitance C of a sandwich sample.

ac measurements on dc biased samples[29] show that at sufficient high frequencies, where $G \simeq G_{ac}$ holds, the admittance is nearly independent of biasing voltage. This behavior suggests that G and C are indeed characteristic of bulk properties at high frequencies.

The temperature dependence of G and C is shown in Fig. 3, at frequencies for which $G \simeq G_{ac}$. At low temperature (150 K) and high frequency (10 MHz) the measured C value (39.8 pF) agrees well with the C value of 38.5 pF, computed from the geometrical dimensions of the sandwich device and the permittivity of crystalline Si (11.8). A sharp increase of capacitance was observed at temperatures above 400 K (not shown in Fig. 3), but further investigation will be necessary to establish this result.

More measurements were performed on the Mo/Cr 34*a* samples to obtain the exponent s at 1 kHz for two different temperatures. They yielded: $T_1 = 150$ K, $s_1 = 0.94 \pm 0.02$; $T_2 = 350$ K, $s_2 = 0.88 \pm 0.02$. The quoted tolerances are governed by the measurement technique.

IV. ANALYSIS OF THE RESULTS

A. Application of current models

The difficulty of interpreting ac data observed on various amorphous semiconductors and other materials with the tunneling models[18–20] has been recognized by several authors.[9,21–24] The following considerations indicate that these models are likewise incompatible with the above experimental results.

At moderately high frequencies, for which the ac properties of our samples are believed to be essentially bulk properties, the observed frequency and temperature dependence of σ' can be written

$$\sigma' = A(T)\omega^{s(T)}, \tag{1}$$

where $A(T)$ shows a superlinear dependence on temperature T. Thus, at $\omega = 2\pi \times 10^3\ \mathrm{sec}^{-1}$ from Fig. 3 we see that $G_1 = 2 \times 10^{-6}\ \Omega^{-1}$ for $T_1 = 150$ K, and $G_2 = 6.33 \times 10^{-6}\ \Omega^{-1}$ for $T_2 = 350$ K. This gives $T_2/T_1 = 2.33$ and $G_2/G_1 = 3.17$. The exponent s of Eq. (1) is close to 0.9, and tends to decrease with increasing temperature. However, Eq. (1) represents the experimental observations only up to frequencies of some 10 kHz, at higher frequencies saturation effects occur, which will be discussed below. For the moment we will focus on Eq. (1) and its explanation by current theories.

For an uncorrelated hopping mechanism the tunneling models[18–20] predict a conductivity of the form

$$\sigma' \propto \omega T[N(E_f)]^2 \ln^4(\nu_{\mathrm{ph}}/\omega), \tag{2}$$

where $N(E_f)$ is the density of defect states at the Fermi level E_f and ν_{ph} a frequency, of the order of a molecular vibration frequency.

Two main disagreements arise. The measured temperature dependence is superlinear, not linear as suggested. However, this is of minor importance compared with the fact that one obtains very unrealistic values for ν_{ph}, if one tries to interpret the experimental value of s ($s \simeq 0.9$) by means of Eq. (2). From Eq. (2) we obtain

$$s \equiv \frac{d \ln\sigma'}{d \ln\omega} = 1 - \frac{4}{\ln(\nu_{\mathrm{ph}}/\omega)}, \tag{3}$$

and upon rearranging,

$$\nu_{\mathrm{ph}} = \omega \exp(4/\beta), \tag{4}$$

where $\beta = 1 - s$. Therefore, $s = 0.9$ and $\omega = 2\pi \times 10^3\ \mathrm{sec}^{-1}$ yields $\nu_{\mathrm{ph}} = 10^{21}$ Hz, which seems unrealistic for a phonon frequency. Moreover, due to the temperature dependence of β, ν_{ph} becomes an exponential function of temperature.

The assumption of a correlated hopping mechanism[19] does not eliminate or solve these controversies. In this case the right side of Eq. (3) becomes $1 - 3/[\ln(\nu_{\mathrm{ph}}/\omega)]$, which does give rise to lower ν_{ph} values, but this model gives a temperature independent σ', which means that theory and experiment are even more divergent.

An alternative classical model for the hopping mechanism was proposed by Pike.[24] This theory has been shown to be in better agreement with data measured on scandium oxide. In Pike's treatment the transport is attributed to the hopping of electrons *over* barriers separating different sites, rather than to be due to a tunneling through the barrier. The potential of the sites has been assumed to be Coulombic. For $\omega\tau_0 \ll 1$, where τ_0 is assumed to be an inverse phonon frequency, Pike finds

$$\sigma' \propto \frac{\omega^s}{\tau_0^\beta \cos(\beta\pi/2)}, \tag{5}$$

with

$$\beta \propto T. \tag{6}$$

This result seems to be in good agreement with our data, as the following example demonstrates: Our experimental values for 1 kHz are $s_1 \simeq 0.94$ at $T_1 = 150$ K, and $s_2 \simeq 0.88$ at $T_2 = 350$ K. Putting the first value into Eq. (6) we obtain $s_2 = 0.86$, and with this value, the measured s_1, and $\tau_0 = 10^{-12}$ sec, Eq. (5) yields $\sigma_2'/\sigma_1' = 4.53$, compared with the measured quantity $\sigma_1'/\sigma_2' \equiv G_2/G_1 = 3.17$ as quoted above. Thus, this model accounts for the general trend of σ', namely $\sigma' \propto \omega^s$, with $s \lesssim 1$, and for the decrease of s with increasing temperature. In contrast, the tunneling models lead to reasonable ν_{ph} values only for $s \simeq 0.8$, and does not explain the temperature dependence of s. Moreover, Elliott[22,23] has shown that, with proper modifications, Pike's theory is applicable to chalcogenide glasses, for which Elliott assumes that a bipolaron mechanism is responsible for the ac conduction. However, as will be discussed below, our experiments yield $\tau_0 \sim 10^{-6}$sec, which is several orders of magnitude larger than an atomic vibrational period. Using this value in Eq. (5) we calculate $\sigma_2'/\sigma_1' = 1.5$, which underestimates the observed temperature dependence of σ'. Our further analysis will show that this divergence between theory and experiment may be attributed to the fact that both Pike[24] and Elliott[22] consider hopping only between sites of *equal* energy.

B. Modified model with varying site energies

In the following we will derive a hopping model, based on a hopping over barriers, which includes both, a variation of site energies, barrier heights, and of intersite distances. We will show that, to a certain extent, such calculations are possible without explicit knowledge of the distribution of these parameters. The assumption of Coulomb-like potential wells will then be demonstrated to give results which are very close to the experimental data.

First we idealize the situation, and assume that hopping occurs only between pairs of sites, and that the jumping events are independent of each other (uncorrelated hopping). In this case the total conductivity will be an additive function of the contribution of each pair. The first steps of the following calculations are similar to the derivations of Pollak and Geballe,[17] and will therefore be only briefly outlined. Adopting the notation of these authors, the occupancy probability f_i of site i will change per unit time as

$$\dot{f}_i = \sum_j w_{ji} f_j - \sum_j w_{ij} f_i , \tag{7}$$

where w_{ji} is the transition rate from site j to site i. The normalizing condition is

$$V^{-1} \sum_j f_j = N_c , \tag{8}$$

where V is the volume over which the summation extends, and N_c the total number of hopping carriers. In contrast to the Pollak and Geballe[17] model, where the transition rates are determined by the tunneling probability between two states, the transition rate for hopping over the barrier in our classical model is

$$w_{ij} = [2\tau_0 \exp(\psi_i/kT)]^{-1} . \tag{9}$$

The energy ψ_i is the difference between the top of the barrier which separates the states i and j and the effective (perturbated in the presence of an electric field) energy level of site i. For a two level system the situation is illustrated in Fig. 4. Since it is assumed that a transition over the barriers involves many phonons the constant τ_0 is usually considered to be of the order of an atomic vibrational period.[30] However, some of the presented experimental data seem to be incompatible with this interpretation, as will be discussed below.

We consider the two level system of Fig. 4, together with its symmetrical counterpart, which must always be present in the material for the sake of isotropy. We denote the transition rates from (1) to (2) and *vice versa*, respectively, as w'_{12} and w'_{21}, and those of the symmetrical problem as w''_{12} and w''_{21}. The following assumptions are also made:

$$ERe \ll kT \tag{10}$$

(a linearization condition), and

$$w'_{12} + w'_{21} \simeq w''_{12} + w''_{21} . \tag{11}$$

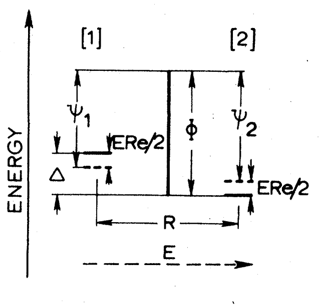

FIG. 4. Two level system for electrons. An electric field E shifts the energies of the unperturbed levels (solid lines) by $ERe/2$ (dotted lines). The site distance is denoted by R, e is the charge of an electron. The energies ψ_1 and ψ_2 are the difference between the top of the barrier Φ and the energies of the perturbed levels (1) and (2).

For a harmonically varying field $E \propto \exp(j\omega t)$ (we assume the applied field to be approximately equal to the local field) we obtain a complex conductivity due to this hopping process, namely

$$\sigma = \sigma' + j\sigma'' = \frac{ne^2R^2}{4kT\cosh^2(\Delta/2kT)} \frac{j\omega}{1+j\omega\tau} , \tag{12}$$

where the relaxation time τ is given by

$$\tau = 2\tau_0 \exp(\Phi/kT)/[1+\exp(\Delta/kT)] . \tag{13}$$

The number n in Eq. (12) denotes the total number of pairs. Since R is the projection of the actual separation r in the direction of the applied field, we still have to summarize over all directions. Thus,

$$R^2 = r^2/3 . \tag{14}$$

Since the real part σ' and the imaginary part σ'' of σ are related to each other by the Kramers-Kronig integral transform,[31] the discussion that follows may be limited to σ'. Assuming that N_c is smaller than the number of sites N within an effective hopping distance, the density of pairs becomes equal to N_c. The differential conductivity $d\sigma'$ may then be written

$$d\sigma' = \frac{N_c r^2 e^2}{12kT\cosh^2(\Delta/2kT)} \frac{\omega^2\tau}{1+\omega^2\tau^2} p(r,\Phi,\Delta)\, dr\, d\Phi\, d\Delta , \tag{15}$$

with τ given by Eq. (13). The number of pairs having an intersite distance between r and $r+dr$, barriers between Φ and $\Phi+d\Phi$, and differences in site energies between Δ and $d\Delta$, is given by $p(r,\Phi,\Delta)\,dr\,d\Phi\,d\Delta$, where p is the probability density.

In Eq. (15) we have assumed r, Φ, and Δ to be independent variables but, in practice, this may not be the case. For instance, by assuming Coulomb-like barriers (see below), r and Φ become dependent on each other. The triple integration necessary to obtain σ' from Eq. (15) then reduces to a double integration.

To calculate σ' we have to integrate Eq. (15) over the whole r, Φ, and Δ spectrum. But these spectra are not in fact known to us in an amorphous semiconductor. We have, therefore, attempted to find some approximations which seem physically reasonable.

However, as long as $p(r,\Phi,\Delta)$ is not a very strong function of Φ and Δ, an approximate integration in the Φ,Δ region can be obtained by making use of the property that $\operatorname{sech}^2(\Delta/2kT)\omega\tau/(1+\omega^2\tau^2)$ is sharply peaked at $\omega\tau=1$ and $\Delta=0$. With the notation

$$\Phi_\omega=-kT\ln(\omega\tau_0) \tag{16}$$

and the condition

$$\Phi_{\min}\ll\Phi_\omega\ll\Phi_{\max}, \tag{17}$$

where $\Phi_{\min}$ and $\Phi_{\max}$ are the limits of the actual barrier distribution, the main contribution to the integral will come from the close neighborhood of Φ_ω and $\Delta=0$. In this region we approximate

$$p(r,\Phi,\Delta)\simeq p(r,\Phi_\omega,0)\,. \tag{18}$$

Moreover, if Φ_ω satisfies Eq. (17), and Δ varies between 0 and an upper limit Δ_0, we may take $\Phi_{\min}=0$ and $\Phi_{\max}=\infty$. The integration over Φ and Δ can then be done exactly, yielding

$$\sigma'=\frac{\omega N_c e^2 kT\pi}{12}\tanh\left(\frac{\Delta_0}{2kT}\right)\int_r p(r,\Phi_\omega,0)r^2\,dr\,, \tag{19}$$

or, if we have only two independent variables Φ and Δ,

$$\sigma'=\frac{\omega N_c e^2 kT\pi r_\omega^2}{12}\tanh\left(\frac{\Delta_0}{2kT}\right)p(\Phi_\omega,0)\,, \tag{20}$$

where $r_\omega=r(\Phi=\Phi_\omega)$.

Before proceeding to the treatment of Eq. (20) it is desirable to consider the behavior of σ' for Φ_ω values outside the limits given by Eq. (17). Since in this region a variation of Δ does not have any important effect on the characteristics of σ', we put $\Delta=0$. Then, for $\Phi_\omega\ll\Phi_{\min}$, Eq. (13) yields

$$\omega\tau_{\min}\gg1\,, \tag{21}$$

where $\tau_{\min}$ is the relaxation time corresponding to $\Phi_{\min}$. Thus, for high frequencies where Eq. (21) holds, $\omega^2\tau/(\omega^2\tau^2+1)$ may be approximated by $1/\tau_{\min}$ and σ' becomes

$$\sigma'\propto1/(T\tau_{\min})\propto1/[T\exp(\Phi_{\min}/kT)]\,. \tag{22}$$

In a similar way, Eq. (15) leads to an estimate of σ' in the low frequency region, with

$$\omega\tau_{\max}\ll1\,. \tag{23}$$

Here

$$\sigma'\propto\omega^2\tau_{\max}/T\propto\omega^2\exp(\Phi_{\max}/kT)/T\,. \tag{24}$$

Thus, at low and high frequencies, the response of the hopping system to an applied field is similar to that of a Debye dipole.[32]

Our measurement yields no evidence for a $\sigma'\propto\omega^2$ behavior at low frequencies. However, our model considers hopping between pairs of sites, which is a good approximation only for appropriately high frequencies.[13] At lower frequencies multiple hopping processes are assumed to predominate. These mechanisms increase σ' and finally lead to the dc term σ'_{dc} which always suppresses possible $\sigma'\propto\omega^2$ terms.

There are some experimental indications of saturation effects at higher frequencies (see Fig. 3). Saturation would imply that Eq. (21) is fulfilled. From Eqs. (13) and (22) we can see that, both, the transition frequency $\omega_t=1/\tau_{\min}$ and approximately also the saturation value of σ' are activated with an energy $\Phi_{\min}$. Thus, the experimental data allow us to estimate $\Phi_{\min}$ and τ_0. Replotting Fig. (3) in the σ', ω region (not shown here) we find that $\Phi_{\min}\simeq0.05$ eV and $\tau_0\simeq10^{-6}$ sec fit the experimental data. In contrast, if we assume τ_0 to be in the order of a typical inverse phonon frequency ($\sim10^{-12}$sec), we need at room temperature $\Phi_{\min}\simeq0.4$ eV to obtain saturation above 100 kHz. Thus, the measured τ_0 is incompatible with an atomic vibrational period, an experimental fact which is not yet understood. It also remains unclear why our experiments imply a barrier $\Phi_{\min}\simeq0.05$ eV. Further it should be noted that, after a more or less pronounced saturation behavior, σ' tends to increase again (see, e.g., Fig. 1). Qualitatively, this characteristic may be explained by superimposing hopping processes with different time constants τ_0. The results in this range are not yet sufficient for an attempt at quantitative analysis.

Experimental data for *a*-Ge and *a*-Si presented by other workers are inconclusive, as far as saturation and, consequently, information on τ_0 is concerned. This is because bridge measurement techniques, usually applied to determine σ', are limited to some 100 kHz, which may be too low to observe saturation. Further, at high frequencies contact effects can give rise to a $\sigma'\propto\omega^2$ dependence, which often leads to misinterpretation. For instance, the high frequency data of Arizumi *et al.*[6] may be explained in this way. However, some experiments do indicate saturation of σ' at high frequencies, and saturation would also be a possible explanation for the "anormalous" frequency dependence of σ' of *a*-Ge films reported by Narasimhan *et al.*[9] These authors observed a decrease of s with decreasing temperature (s varied from 0.7 at 154 K to 0.4 at 138 K). Unfortunately the presented data cover almost only one decade of frequency (10^4–10^5 Hz). At 77 K the *a*-Ge samples of Chopra and Bahl[4] exhibited a σ' saturation around 100 kHz. However, these data

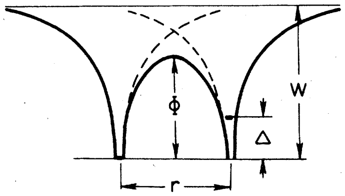

FIG. 5. Two level system with Coulomb-like potential wells. The energy of the ground state and the intersite separation are denoted by W and r, respectively.

are too incomplete to permit conclusions. Incidentally, also the ac conductivity of grain boundaries in crystalline Ge[33] shows saturation at some MHz, but here the measurements have been made only at room temperature.

The further procedure is addressed to the behavior of σ' at medium frequencies due to Eq. (1). In this range Eq. (17) is satisfied and, consequently, σ' is expressed by Eq. (19) or Eq. (20).

Above we have shown that a model with Coulomb-like potential wells and *equal* site energies is in fair agreement with our experimental data, if $\tau_0 \simeq 10^{-12}$ sec. It underestimates the observed temperature dependence of σ' if we take $\tau_0 \simeq 10^{-6}$ sec, as implied by our experiments. However, our theory includes also a variation of site energies, and it will be shown below that in this case a Coulombic potential gives results which are close to the experiment, also as regards the temperature dependence.

If we assume sites with Coulomb-like potential wells (see Fig. 5) the barrier height Φ is related to r as

$$W - \Phi = e^2/(\pi\epsilon_0\kappa r)\,, \tag{25}$$

where W is the energy of the ground state, ϵ_0 the permittivity of free space and κ an effective dielectric constant. Assuming randomness in r and in Δ, and a variation of Δ between 0 and Δ_0, we get a probability density

$$g(r,\Delta) = 4\pi r^2 N/\Delta_0\,. \tag{26}$$

Thus, $p(\Phi_\omega, 0)$ in Eq. (20) becomes

$$p(\Phi_\omega, 0) = 4\pi^2\epsilon_0\kappa r_\omega^4 N/(e^2\Delta_0)\,. \tag{27}$$

As a result, the conductivity is given by

$$\sigma' = \frac{NN_c kT\omega}{3\Delta_0\epsilon_0^5\kappa^5\pi^3}\left(\frac{e^2}{W + kT\ln(\omega\tau_0)}\right)^6 \tanh\left(\frac{\Delta_0}{2kT}\right). \tag{28}$$

With the approximation[24]

$$[W + kT\ln(\omega\tau_0)]^{-6} \simeq W^{-6}(\omega\tau_0)^{-\beta}\,, \tag{29}$$

where

$$\beta = 6kT/W\,, \tag{30}$$

we may write

$$\sigma' \propto T\tanh(\Delta_0/2kT)\,\omega^s/\tau_0^\beta\,. \tag{31}$$

This is almost the same result as that obtained by Pike[24] and Elliott,[22] but multiplied by $T\tanh(\Delta_0/2kT)$. Thus, a variation of site energies, as included in our model, increases the temperature dependence of σ'.

Due to Eq. (28), σ' should vary with NN_c, provided the other parameters remain unchanged. We have no information about N and N_c in our samples. However, Abkowitz *et al.*[10] reported that their a-Si films, produced from SiH_4 by glow discharge, exhibited a dependence of approximately $\sigma' \propto [N(E_f)]^2$. If we assumed that $\sqrt{NN_c} \sim N \propto N(E_f)$, this could amount to a confirmation of our model. It should be noted that in Elliott's model[12] for the ac conductivity in a-Si (this author assumes a simultaneous hopping of two electrons between, so called, intimate pairs of sites) σ' varies linearly with N, which seems to be in contrast to the above data. Moreover, the application of Elliott's theory to our experimental results leads to unrealistically high N values.

From our measurements we are able to estimate some of the parameters of Eq. (28). Values for W between 1.5 and 1.7 eV give s values which are very close to those measured. For further calculations, we take $W = 1.6$ eV. In chalcogenide glasses W has been interpreted as the energy of the bound state, which may be approximated by the gap energy.[22] A value of 1.6 eV seems to be somewhat too large to be explained by the pseudogap of a-Si. If, on the other hand, one considers that in small clusters the energy difference between bonding and nonbonding states, and thus the gap, may be considerably larger than in the infinite crystal,[34] this could be a possible interpretation. However, we have to keep in mind that the application of band models to amorphous semiconductors is accompanied by great complications, and has not, so far, been satisfactorily solved.

With the experimental values for sample Mo/Cr 34a: $\tau_0 = 10^{-6}$ sec, $\omega = 2\pi \times 10^3\ \mathrm{sec}^{-1}$, $T = 150$ K, $\sigma' = 5.5 \times 10^{-8}\ (\Omega\mathrm{cm})^{-1}$, and with the assumption $\Delta_0 = 0.1$ eV,[35] we obtain from Eq. (28) $NN_c/\kappa^5 = 2.6 \times 10^{40}\ \mathrm{cm}^{-6}$. This value leads to reasonable defect densities if we assume that the defects responsible for the hopping are immersed in a medium with a dielectric constant between 1 and 11.8 (relative permittivity of crystalline Si). For instance, with $\kappa = 3$ we obtain $\sqrt{NN_c} = 2.5 \times 10^{21}\ \mathrm{cm}^{-3}$ and $r_\omega = 13$ Å. This value for the hopping distance remains nearly constant over the whole frequency and temperature range considered.

Table II shows the comparison between experi-

TABLE II. Comparison between calculated and measured data for conductivity σ' and exponent s ($\sigma' \propto \omega^s$) at 1 kHz. The σ' value measured at 150 K has been used to scale the other calculated σ' values.

Temperature/K	s_{meas}	s_{calc}	$\sigma'_{\text{meas}} \times \Omega$ cm	$\sigma'_{\text{calc}} \times \Omega$ cm
150	0.94	0.95	5.5×10^{-8}	•••
250	•••	0.915	6.9×10^{-8}	10.8×10^{-8}
350	0.88	0.88	1.73×10^{-7}	1.8×10^{-7}

mental σ' and s values, and the values calculated from Eq. (28). For the calculation we simply have used σ' measured at 150 K as a scaling factor. Further we have assumed $\Delta_0 \gg kT$, and, consequently, $\tanh(\Delta_0/2kT) \simeq 1$. The agreement between theory and experiment may be considered as satisfactory. It should be noted that τ_0 has to be in the μ sec range to bring the measured and the computed data close together. In contrast, if τ_0 is assumed to be an inverse phonon frequency ($\tau_0 = 10^{-12}$sec) Eq. (28) overestimates the temperature dependence of s and σ' considerably. For instance, with $W = 1.6$ eV and $\tau_0 = 10^{-12}$sec we calculate for $T_2/T_1 = 350/150$ K: $\sigma_2'/\sigma_1' = 12$ and $s_2/s_1 = 0.83/0.94$; whereas the experimental values are $\sigma_2'/\sigma_1' = 3.16$ and $s_2/s_1 = 0.88/0.94$.

An attempt to apply the present model to ac measurements on a-Si and a-Ge by other workers has been made, especially with a view to comparing the measured and the calculated temperature dependence of σ' and s. From the cited papers,[4–12] only the results of Abkowitz *et al.*[10] proved to be complete enough to yield some reliable values for $\sigma'(T)$ and $s(T)$. Several other works present data exclusively for one temperature,[5,7] in other instances the frequency region in which the $\sigma'(T) \propto \omega^{s(T)}$ behavior has been investigated was too small (one order of magnitude or less) to be conclusive.[4,9,11,12]

Table III shows that our model is in good agreement with experimental data obtainable from Fig. 1 of Ref. 10. The calculations have been made for different Δ_0. For $\Delta_0 \ll kT$ we have approximated $\tanh(\Delta_0/2kT) \simeq \Delta_0/2kT$. Thus, Eq. (28) becomes

$$\sigma' = (NN_c\omega/6\kappa^5\epsilon_0^5\pi^3)\{e^2/[W + kT\ln(\omega\tau_0)]\}^6. \quad (32)$$

Obviously the same and correct temperature dependence of s can be approached by τ_0 values which differ from one another by many orders of magnitude. The calculated σ_2'/σ_1' values are close to the experimental results with either large τ_0 and $\Delta_0 \gg kT$, or small τ_0 and $\Delta_0 \ll kT$. Consequently, it seems not possible to estimate τ_0 exclusively from the $\sigma' \propto \omega^s$ region. Additional information about the saturation behavior of σ' is necessary, e.g., of the kind provided by our experimental results.

V. CONCLUSION

The ac conductivity of the a-Si films investigated has been attributed to a classical hopping between localized states, randomly distributed in space and energy. The model presented accounts for both the $\sigma' \propto A(T)\omega^{s(T)}$ behavior observed at medium frequencies, and for the saturation effects measured in the high-frequency region. At medium frequencies close numerical agreement between theory and experiment could be obtained only with relaxation time constants τ_0 in the order of 10^{-6} sec, which value could also be deduced from the saturation behavior measured at high frequencies. So far, τ_0 has always been assumed to be in the range of an atomic vibrational period. However,

TABLE III. Application of the hopping model to the data of Abkowitz, Le Comber, and Spear (Ref. 10), measured on glow discharge amorphous silicon at 1 kHz. For $T_1 = 84$ K and $T_2 = 255$ K these experiments yielded $\sigma_1' = 6.4 \times 10^{-11}$ $(\Omega\,\text{cm})^{-1}$, $s_1 = 0.98$ and $\sigma_2' = 1.55 \times 10^{-10}$ $(\Omega\,\text{cm})^{-1}$, $s_2 = 0.93$, respectively.

				Calculation parameters		
	σ_2'/σ_1'	s_1	s_2	τ_0/sec	W/eV	Δ_0
Measured	2.42	0.98	0.93			
Calculated	3.82	0.978	0.931	10^{-6}	2	$\gg kT$
	1.26	0.978	0.931	10^{-6}	2	$\ll kT$
	6.93	0.980	0.931	10^{-12}	2.3	$\gg kT$
	2.28	0.980	0.931	10^{-12}	2.3	$\ll kT$

more experimental information is necessary, in order to get a better physical understanding of the meaning of τ_0.

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