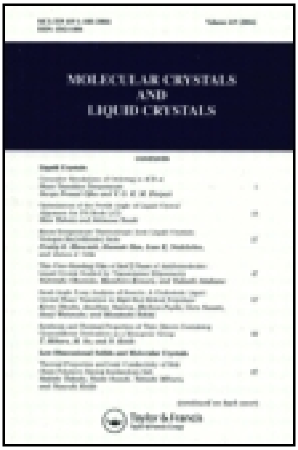

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Frequency Dependent Conductivity of Polyacetylene

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FREQUENCY DEPENDENT CONDUCTIVITY OF POLYACETYLENE

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We report on an extensive study of the frequency and temperature dependent conductivity of largely cis-$(CH)_x$, trans-$(CH)_x$, and NH_3 compensated and iodine doped materials. The results reveal a strongly temperature (T) dependent dc conductivity and weakly T-dependent ac conductivity for largely cis-$(CH)_x$ and for NH_3 compensated samples, similar to the behavior that is observed in many crystalline and amorphous semiconductors. The trans-$(CH)_x$ and lightly iodine doped trans-$(CH)_x$ have larger dc and ac conductivity, with a "weakly" T-dependent dc conductivity and a strongly T-dependent ac conductivity. These latter results contrast with the usual expectations of variable range hopping and polaronic hopping, but are in good agreement with the predictions of Kivelson's Theory of charge transport via intersoliton electron hopping.

INTRODUCTION

The charge transport properties of polyacetylene films have been under intense study for several years.[1,2] This activity was in large part motivated by the report[1] that the dc conductivity increases by up to eight orders of magnitude upon doping, from that of a semiconductor to metallic behavior. The mechanisms for doping and transport are of fundamental interest.

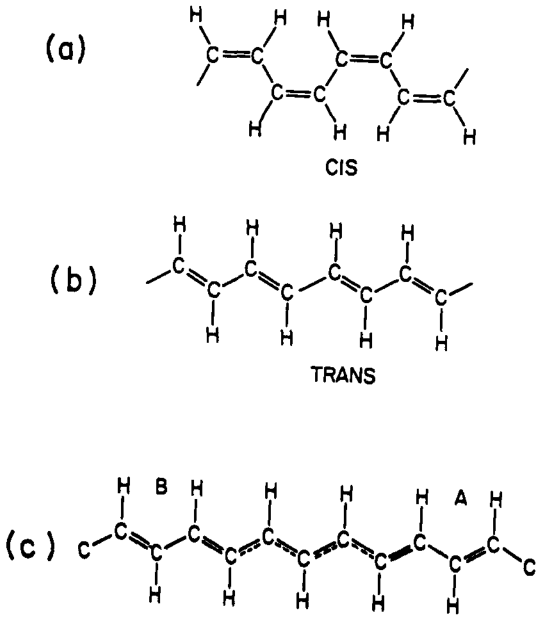

FIGURE 1. Idealized polyacetylene structures: (a) cis-$(CH)_x$; (b) trans-$(CH)_x$; (c) a soliton separates two regions (A and B) of bond alternation with opposite phase (schematic).

There are two isomers of polyacetylene, cis-$(CH)_x$ and trans-$(CH)_x$, Figure 1. The trans-$(CH)_x$ is obtained from cis-$(CH)_x$ through thermal isomerization in vacuo. According to the soliton model[3,4] undoped $(CH)_x$ is a semiconductor due to a commensurate Peierls distortion which opens up a band gap at the Fermi energy, E_F. For the trans isomer, the two possible phases of the dimerization are degenerate in energy and a soliton is the boundary between regions of the two phases, Figure 1c. The energy level associated with the soliton is at mid-gap. When singly occupied the soliton is neutral with spin 1/2. If the state is doubly occupied or empty, the soliton is charged and spinless. For the cis-$(CH)_x$, the two possible phases of dimerization (cis-transoid and trans-cisoid) are not degenerate in energy. Hence the formation of solitons is not favored energetically in cis-$(CH)_x$. Magnetic,[5,6,7] infrared[8,9] luminescence[10] and photoconductivity[11] studies have been interpreted as evidence for the presence of solitons in the trans isomer and their absence in the cis isomer.

We report here the results of an extensive series of measurements of the frequency (f) and temperature (T) dependence of the conductivity ($\sigma = \sigma_{DC} + \sigma_{AC}$) of trans-$(CH)_x$, predominantly cis-$(CH)_x$, NH_3 compensated $(CH)_x$ and iodine doped $(CH)_x$. The results reveal that both the ac and dc components of σ of the trans are larger than those of the cis and the NH_3 compensated trans. The σ (f,T) of cis and NH_3 compensated trans material are very similar to the well known behavior of amorphous and insulating materials. The trans-$(CH)_x$ has a very strongly T-dependent σ_{AC}. Detailed analysis shows that this behavior is not consistent with usual models for σ_{AC}. On the other hand, the measured σ_{AC} (f,T) is consistent with behavior predicted by the model of phonon assisted hopping of electrons between solitons proposed by Kivelson.

MODELS FOR CONDUCTIVITY

DC and ac electrical conductivity in crystalline and amorphous semiconductors occurs via several possible mechanisms.[12,13,14] Table I summarizes the most commonly applied models[12-14] and their functional dependence upon temperature, T, and frequency, f, for frequencies less than 10^6 hz. In Table I, k_B is the Boltzmann constant, E_a and E'_a are activation energies, E_b is the polaron binding energy, T_o is a constant ($\propto$ ($\alpha^{-3} N(E_F)^{-1}$)where α is the localization length and $N(E_F)$ is the density of states at the Fermi level , and s is a constant between zero and one.

Table 1 Functional Dependence of the DC and AC Conductivities on Temperature and Frequency

$\sigma = \sigma_{DC}(T) + \sigma_{AC}(T,f)$

Model	DC	AC
Thermal activation to extended states	$\exp[-E_a/k_BT]$	0 ($\sigma = \sigma_{DC}$)
Thermally activated hopping in band tails	$\exp[-E'_a/k_BT]$	$f^s\, T \exp[-E'_a/k_BT]$
Polaron hopping (ordered lattice)	$\exp[-E_b/2k_BT]$	$f^s \exp[-E_b(1-s)k_BT]$
Hopping in a manifold of states at E_F	$\exp[-(T_o/T)^{0.25}]$	Tf^s

EXPERIMENTAL PROCEEDURES

Polyacetylene was prepared by the Shirakawa technique.[15] The conductivity was measured in a sandwich cell configuration as well as along the length of the film. Scanning electron micrographic, magnetic, and transport studies demonstrated that these films had the previously characterized morphology,[16,17] magnetic[7] and transport[18] properties. Samples were maintained in an inert atmosphere at all times. Measurements on several samples produced nearly identical results. The $\sigma(f)$ was obtained at fixed temperature using a General Radio Capacitance-Conductance Bridge in its three terminal configuration. The σ_{DC} was obtained using a Keithley high impedance electrometer. Care was taken to assure that contact sheet resistance effects were not leading to spurious behavior.[19]

EXPERIMENTAL RESULTS - CIS AND NH_3 COMPENSATED POLYACETYLENE

The experimental results for predominately cis-$(CH)_x$ and for NH_3 compensated cis-$(CH)_x$ or trans-$(CH)_x$ are similar. For cis-$(CH)_x$, σ_{DC} (295K) is 10^{-10} $ohm^{-1} cm^{-1}$. Trans-$(CH)_x$ has σ_{DC} (295K) $\approx 10^{-5}$ $ohm^{-1} cm^{-1}$. From thermoelectric power measurements, it is a p-type conductor.[20] Although the detailed chemistry is not yet well understood, it is known[21] that exposure of trans-$(CH)_x$ to NH_3 leads to compensation of a large fraction of the charge carriers resulting in σ_{DC} (295 K) $\approx 10^{-10}$ $ohm^{-1} cm^{-1}$, similar to cis-$(CH)_x \cdot NH_3$ compensation of partially isomerized polyacetylene leads to even lower conductivity.

Figure 2 displays the total conductivity versus frequency at constant temperature for a sample of 70% trans-$(CH)_x$ (by NMR) compensated with NH_3. This result is typical for cis and NH_3 compensated samples.[22] The $\sigma(f)$ has a marked frequency dependence even at 100 Hz with $\sigma_{AC} \propto f^{0.8}$. As T is varied, σ_{DC} changes rapidly, while σ_{AC} changes very slowly with temperature. This behavior for $\sigma(f,T)$ is very similar to that common to a broad class of disordered semiconductors and insulators (for example, selenium,[23] anthracene,[19] impurity conduction in crystalline silicon,[24] and amorphous silicon[25]). Diverse physical mechanisms[12,14] can underlie this behavior, including variable range hopping, the presence of surface bariers and the presence of ionic dipoles.

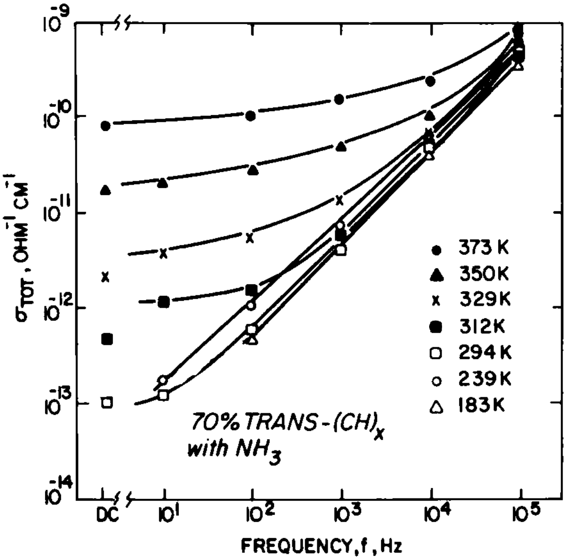

FIGURE 2. Log σ vs. log f for NH_3 compensated (0.70 trans-0.30 cis)-polyacetylene at constant temperature. Note the break in the abscissa for dc. The solid lines are drawn as a guide to the eye.

EXPERIMENTAL RESULTS - TRANS - POLYACETYLENE

The conductivity of trans samples was measured both along the films and through the film thickness. Generally, the conductivity through the films was found to be a factor of two to three times lower than along the film. This is probably due to the fibrils lying in the plane of the films,[16,17] so that the effective path length through the film is several times the measured thickness.

Figure 3 displays the log $\sigma_{DC}(T)$ vs T^{-1} for a typical sample of trans-$(CH)_x$. The room temperature value is five orders of magnitude larger than that of cis-$(CH)_x$ and five to eight orders of magnitude larger than that of ammonia compensated polyacetylene samples. The solid line is a fit of a simply activated expression,

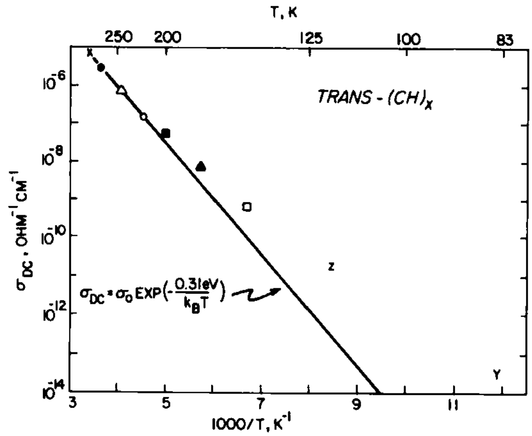

FIGURE 3. Log σ_{DC} vs. T^{-1} for trans-$(CH)_x$.

$\sigma_{DC} = \sigma_o \exp(-E_a/k_BT)$. The best fit is obtained with E_a=0.31 eV in agreement with earlier reports.[26,27] However, data in the extended temperature range displayed in Figure 3 makes it clear that the actual T-dependence is much weaker than this. This data is replotted as log $\sigma_{DC}(T)$ versus $T^{-0.25}$ in Figure 4. The functional form,

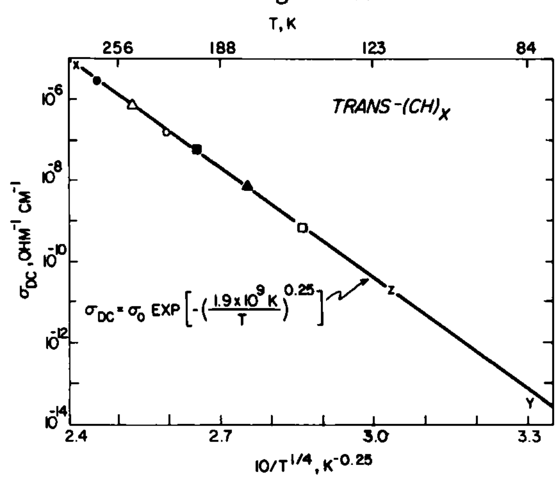

FIGURE 4. Log σ_{DC} vs. $T^{-0.25}$ for trans-$(CH)_x$ data displayed in Fig. 3.

exp $[-(T_o/T)^{0.25}]$ provides a fit with $T_o = 1.9\times10^9$ K. Referring to Table I, this weak temperature dependence of $\sigma_{DC}(T)$ suggests that hopping in a manifold of states at the Fermi-level[12,14] may be the conduction mechanism. If this is appropriate, then $\sigma_{AC}(f,T)$ should be weakly T-dependent (approximately proportional to T). The experimental results for $\sigma_{AC}(f,T)$ of trans-$(CH)_x$ are shown in

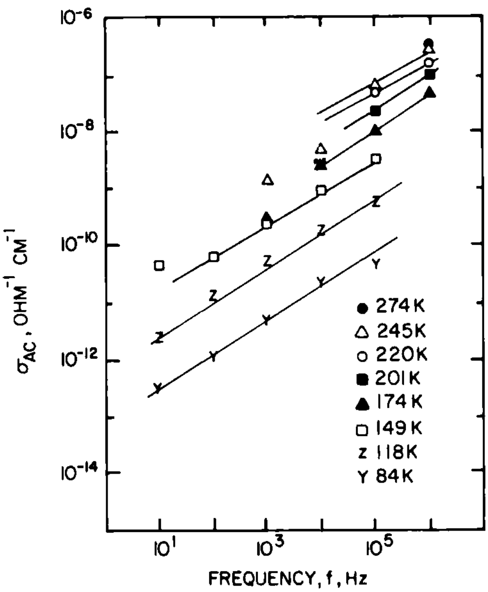

FIGURE 5. Log σ_{AC} vs. log f for the same sample utilized for Figs. 3 and 4. The solid lines are drawn as guides to the eye.

Figure 5. The frequency dependence was measured at constant temperature at various temperatures. At low temperatures the ac term is larger than the dc term for all frequencies measured, with $\sigma_{AC} \propto f^{0.5}$ at constant temperature, as for example seen at 84K and 118K. As the temperature is increased, increasing σ_{DC} makes it difficult to directly measure σ_{AC} at low frequency, although higher frequency data is still obtainable. The slope of σ_{AC} vs f increases with increasing T to, for example, $\sigma_{AC} \propto f^{0.7}$ for T > 200K. Examining the data as

a function of temperature at constant frequency reveals a very strong T-dependence. For example, at 10^5 Hz, σ_{AC} varies by a factor of more than 10^3 upon lowering the temperature from 220 K to 84 K, i.e., less than a factor of three in temperature.

This $\sigma_{AC}(f,T)$ for trans-$(CH)_x$ is a marked contrast to the $\sigma_{AC}(f,T)$ of cis and NH_3 compensated polyacetylene, shown in Figure 2. It also differs from the weak T-dependence of σ_{AC} usually associated with hopping at a manifold of states at the Fermi level (Table I). The observed strong T-dependence can, in principal, arise from thermal activation of charge carriers from localized states in the energy gap to the mobility edge.[14] The σ_{AC} and σ_{DC} would then both be proportional to $\exp(-E'_a/k_BT)$ with E'_a the activation energy. However, this functional form is inconsistant with our observed $\sigma_{DC}(T)$, Figure 3. Another alternative model is the hopping of polarons.[13,14] Although polaron hopping in an ordered lattice can lead to strongly T-dependent σ_{AC}, σ_{DC} would

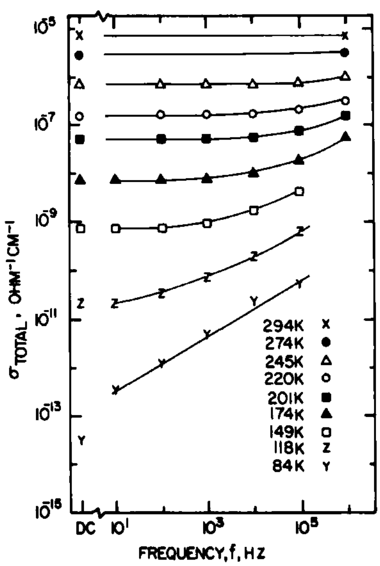

FIGURE 6. Log σ vs. log f for the data of trans-$(CH)_x$ displayed in Figs. 3-5. Note the break in the abscissa for dc. The solid lines are drawn as guides to the eye.

then be simply activated, i.e., $\sigma_{DC} \propto \exp(-E_b/2k_BT)$ with E_b the small polaron binding energy (Table I). This is inconsistent with the experimental results for $\sigma_{DC}(T)$, Figure 3. It remains to be seen if addition of disorder to the polaron model, or a more complete calculation of $\sigma_{AC}(f,T)$ for variable range hopping model could account for the observed data.

The experimental results for trans-$(CH)_x$ are summarized as log σ_{TOTAL} versus log f in Figure 6. They are readily contrasted with the results for NH_3 compensated material shown in Figure 2.

INTERSOLITON ELECTRON HOPPING

Recently, Kivelson has proposed[28-30] a phenomenological model for three-dimensional hopping conduction based upon the separation of the energy (temperature) and spatial dependence of the rate of hopping among sites. The wavefunction overlap is assumed to vary exponentially with distance while the rate at which the hopping transition occurs is averaged over the thermal distribution of initial and final site energies and assumed to be proportional to a power law in temperature, T^{n+1}. Kivelson[28-30] associated this formalism with the phonon assisted hopping of electrons between soliton sites. In this model, charged solitons are coulombically bound to charged impurity sites. The excess charge on the soliton site makes a phonon assisted transition to a neutral soliton. If this neutral soliton is near another charged impurity, the energy of the charged carrier before and after the hop is unchanged. The presence of disorder, as represented by a spatially random distribution of dopant molecules, causes the hopping-conduction pathways to be essentially three-dimensional. The T-dependence of the conductivity is then determined[28-30] by the probability, $\gamma(T)$, that the neutral soliton is near the charged impurity and the initial and final energies are within k_BT of each other:

$$\sigma_{DC} = Ae^2(\gamma(T)/(k_BT\,N)\,(\xi/R_0^2)\,y_n y_{ch}(y_n+y_{ch})^{-2}\,e^{-2BR_0/\xi} \qquad (1)$$

Here A=0.45, B=1.39, y_n and y_{ch} are the concentrations of neutral and charged solitons per carbon atom respectively, $R_o \equiv (4\pi/3\ c_{im})^{-1/3}$ is the typical separation between impurities where c_{im} is the concentration of impurities, ξ is the dimensionally averaged decay length for a soliton, and N is the number of carbon atoms per $(CH)_x$ chain. γ (T)/N is then proportional to the fraction of time a pair of solitons are so situated that the initial and final soliton states are within k_BT of each other. Kivelson shows[28-30] that band center optical phonons are most important leading to $\gamma(T) \propto T^{n+1}$, so that, from Eq. 1, $\sigma(T) \propto T^n$, with $n \sim 10$.

The σ_{DC} of trans-$(CH)_x$ shown in Figs. 3 and 4, is replotted in Fig. 7 as a log-log plot. It is seen that $\sigma_{DC}(T) \propto T^{13.7}$ is a very good fit to the data (reproducible in many samples). The lower measured σ_{DC} for T=84K may reflect the onset of a decrease in the contribution of acoustic phonons to the hopping in addition to the T-dependence of the optical phonons. The dc conductivity may be quantitatively analyzed using Equation 1. Each of the parameters in Equation 1 are known from prior work except c_{im} which is related to y_{ch} through $c_{im}=y_{ch}\ \rho$

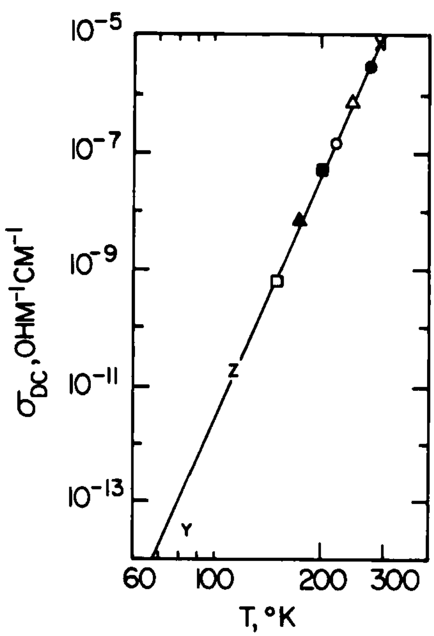

FIGURE 7. Log σ_{DC} vs. log T for the data in Figs. 3 and 4. The solid line is the best fit with $\sigma_{DC} \propto T^{13.7}$

where ρ is the volume density of carbon atoms. This assumes that each charged impurity gives rise to a charged soliton. Using $\xi = 3.56\times10^{-8}$ cm (Ref. 4, 7, 29), $y_n = 5\times10^{-4}$ (Ref. 7, 31) and $\hbar\gamma_0(T) = 500\,\mathrm{eV}(T/300\mathrm{K})^{14.7}$ (Ref. 28, 29), a value of $c_{im} = 1.3\times10^{19}$ cm^{-3} ($y_{ch} = 2.35\times10^{-4}$) is obtained. This is in good agreement[32] with depletion measurements on heterojunctions.

Assuming pairwise hopping, Kivelson also obtained an expression for σ_{AC} within this model:

$$\sigma_{AC} = (e^2/\hbar)\,(c_{im}{}^2/(384k_BT))\; n^{(o)}(1-n^{(o)})\,\xi_{\parallel}{}^3\xi_{\perp}{}^2\,\hbar f\,[\ln(4\pi fN/(n^{o}(1-n^{o})\gamma(T))]^4 \quad (2)$$

$$= K'\ (f/T)\ [\ln(Df/T^{n+1})]^4 \equiv Kx \quad (3)$$

where $n^{(o)}$ is the fraction of solitons that are charged and K' and D are constants. Here $\xi_{||}$ and $\xi_{\perp}$ are the decay lengths of solitons parallel and perpendicular to the polymer chain direction respectively. The dimensionally averaged decay length for a soliton is given by $\xi = (\xi_{||}\xi_{\perp}{}^2)^{1/3}$. This result may be compared with pairwise phonon-assisted hopping between localized states near the Fermi level (see also Table I):

$$\sigma_{AC} = 2(\pi^2/3)\, e^2 k_B\, T\, N^2(E_F)\,\alpha^{-5}\, f\,[\ln(\nu_{ph}/2\pi f)]^4, \quad (4)$$

with k_B the Boltzman constant, $N(E_F)$ the density of localized states at the Fermi energy, ν_{ph} is the phonon assisted attempt frequency, and α^{-1} is the effective localization length. The forms of Equations 2 and 4 are similar with $k_BT\ N(E_F)$ replaced by C_{im} and the ν_{ph} replaced by $n^{(o)}\ (1-n^{(o)})\,\gamma(T)/N$. This transforms a weakly T-dependent ac conductivity to a strongly T-dependent behavior.

The experimental σ_{AC} (f,T) for trans-$(CH)_x$, Figure 5, is readily compared with the Kivelson result, Eqs. 2 and 3. Utilizing the variable x defined in Equation 3 as $x = (f/T)\,[\ln(Df/T^{n+1})]^4$ a value for x can be calculated for each data point in Figure 5 using n=13.7 obtained by fitting $\sigma_{DC}(T)$ to Eq. 1. The parameter D was varied to obtain a linear relation between σ_{AC} and x. It was found that the results are sensitive to the choice of D; even a

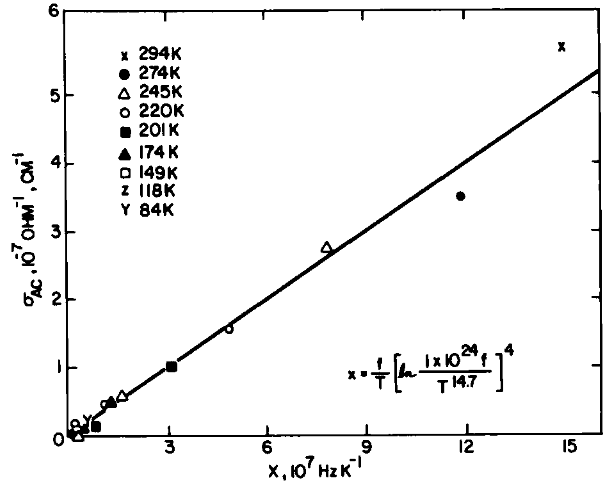

FIGURE 8. σ_{AC} vs. x, with $x \equiv (f/T)\,[\ln(10^{24} f/T^{14.7})]^4$ for the data in Fig. 5. The best fit is given by the solid line.

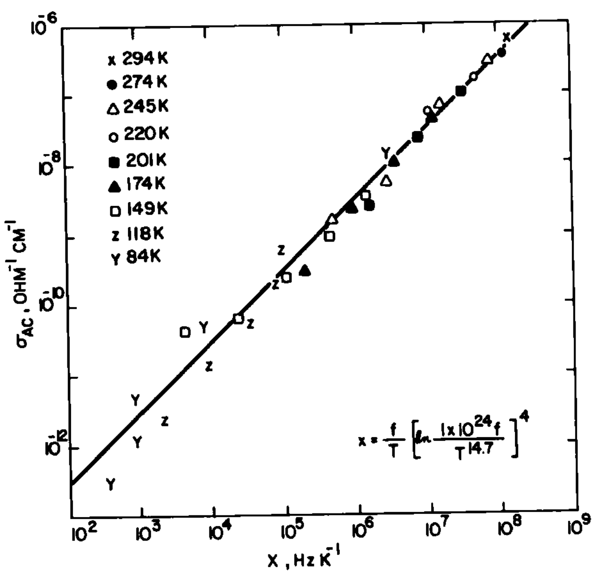

FIGURE 9. Log σ_{AC} vs. log x, $x \equiv (f/T)[\ln(10^{24} f/T^{14.7})]^4$, for the data in Fig. 5. The solid line is the same linear fit displayed by the solid line in Figure 8.

change of a factor of two in D leads to significant differences in the fit to the data. In Figure 8 we replot the σ_{AC} data of Figure 5 versus x for D=1x10^{24}sec. K$^{14.7}$. The results show a good fit to a linear relationship, given by the solid line. In order to test the uniqueness of the fit at low values of σ_{AC} and x, the data is replotted on a log-log plot in Figure 9. The results demonstrate a linear relationship between σ_{AC} and x for over six orders of magnitude in variation of σ_{AC}, f and x. The solid line in Figure 9 has the same slope as that in Figure 8 (linear relation between σ_{AC} and x) with D=1x10^{24} sec K and K$'$=3x10^{-15} ohm^{-1} cm^{-1} sec K. This self-consistent agreement in the functional form of σ_{AC}(f,T) and σ_{DC}(T) demonstrates the utility of the Kivelson phenomenological formalism.

The Equation (2) can be evaluated without any adjustable parameters. Using ξ, y_n, N and $\hbar$ γ(T) given above, together with c_{im}, y_{ch} and n from σ_{DC}(T) [Figure 7], and $\xi_{||} \simeq$ 10x10^{-8} cm (Ref. 28,29) and $\xi_{\perp} \simeq$ 2.5x10^{-8}cm (Ref. 28,29) we calculate the constants K$'$ and D as K$'$=6.3x10^{-16}ohm^{-1}cm^{-1}sec K and D=4.0x10^{23}sec K$^{14.7}$. This compares very well with the experimental values given above. The small difference between the experimental and calculated K$'$ and D are within the uncertainty of the values of the parameters used for input to the calculated K$'$ and D. Thus very good agreement with the theory of phonon assisted hopping of electrons among soliton sites is observed.

EFFECTS OF LIGHT DOPING WITH IODINE

After completing the measurement of $\sigma = \sigma_{DC}$(T)+ σ_{AC}(f,T) for <u>trans</u>-polyacetylene, several samples were doped <u>in situ</u> with iodine. The conductivity was then remeasured as a function of temperature and frequency. A typical result is shown in Figure 10. As seen here the temperature dependence of σ_{DC} is unchanged upon doping despite the fourteen-fold increase in absolute value of conductivity. The σ_{AC}(f,T) of the undoped <u>trans</u> was very similar to that shown in Figure 5. Upon doping, the σ_{AC}(f,T) increased by approximately a factor of 3.5.

Although quantitative analysis is complicated by uncertainty in the exact quantity of iodine absorbed several points can readily be made. The maintenance of the same temperature dependence for σ_{DC} after doping makes it difficult to apply usual models for disordered semiconductors such as variable range hopping states near the Fermi level. In this case the T-dependence is

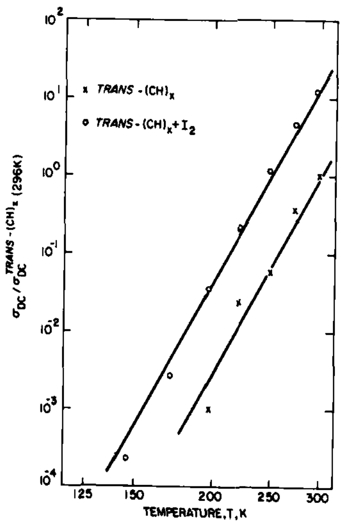

FIGURE 10. Log conductivity vs. log T for undoped trans-$(CH)_x$ (x) and the same sample after doping with a small amount of iodine (o). The conductivity data are normalized to the value of the conductivity of the undoped trans-$(CH)_x$ at 296K. The solid lines have the same slope with $\sigma_{DC} \sim T^{14}$, see text.

determined by $\sigma_{DC} \propto \exp[-(To/T)^{0.25}]$. The constant T_o is inversely proportioned to α^3 and $N(E_F)$. With increase doping, both α^{-1} and $N(E_F)$ should change leading to a change in the T-dependence of σ_{DC}.

For the intersoliton electron hopping model, Eq. 1, the T-dependence is independent of dopant concentration, consistent with the data in Figure 10. Using Eq. 1, we calculate the concentration of charged solitons as $3.0x10^{-4}$ per carbon. Comparing with $2.35x10^{-4}$ per carbon for the undoped $(CH)_x$, this indicates an increase in the number of charged solitons of 65 ppm. This value of y_{ch} (from σ_{DC}) may be used in Equation 2 to predict the change of $\sigma_{AC}(f,T)$ upon doping. Evaluating Equation 2 for the observed increase for y_{ch}, σ_{AC} (f,T) is expected to increase by a factor of 3.5. The measured increase of a factor of 2.5 in σ_{AC} (f,T) is in reasonable agreement with the intersoliton electron hopping model.[28-30]

SUMMARY

A large strongly temperature dependent ac conductivity was observed in trans-polyacetylene and lightly doped trans polyacetylene. The data are inconsistent with the usual ac transport models for hopping near the Fermi level. Very good agreement with the Kivelson theory of intersoliton electron hopping is found. In contrast, cis-polyacetylene and NH_3 compensated polyacetylene do not exhibit this behavior.

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