Frequency response of hopping systems: Application to polyacetylene

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Hopping mechanisms for conductivity are characterized by a broad distribution of hopping rates. We have calculated the ratio of the conductivity in the high-frequency regime (frequency greater than the maximum hopping frequency) and its dc value. The results for isoenergetic hopping and variable-range hopping are shown to differ, although both are found to be independent of the details of the hopping attempt frequency. These results are applied to the experimental data for doped *trans*-polyacetylene. Charge conduction in undoped and lightly doped *trans*-polyacetylene is in agreement with isoenergetic hopping consistent with intersoliton electron hopping. In contrast, the data for *trans*-polyacetylene doped to 0.01—0.05 I_3 per carbon are in better agreement with variable-range hopping, consistent with the proposal of variable-range hopping among pinned solitons. These results support a change in the charge transport mechanism as polyacetylene is doped.

I. INTRODUCTION

Hopping mechanisms are all characterized by a broad (log-normal) distribution of hopping rates and hence exhibit many features that are common to all hopping models.[1,2] We have considered the expected behavior of the conductivity (σ) in the dc ($\omega=0$) and high-frequency limits. The high-frequency limit is reached when ω is greater than the maximum hopping rate or attempt frequency Γ_0. The ratio $\sigma(\omega=0)/\sigma(\omega\gg\Gamma_0)$ is a useful quantity because its value is a sensitive measure of the distribution of site energies, but it is independent of the complicated details of the hopping mechanism. In particular, we consider the two extreme cases, isoenergetic hopping, where the distribution of site energies is narrow on the scale of k_BT, and variable-range hopping, in which the distribution is much broader than k_BT. Since the ratio obtained is very different in the two limits, it is a useful diagnostic of the charge conduction mechanism.

Trans-polyacetylene is one of the few materials we know in which hopping conduction has been measured[3–5] for a broad range of frequency (dc to 10^{13} Hz) and temperature (80—300 K). This has been facilitated by the large absolute values of the hopping conductivity, in contrast to the much lower values in more traditional hopping systems.[2] Polyacetylene has been of particular interest since the report[6] that upon doping its dc conductivity can be varied from 10^{-5} through 10^{+3} $\Omega^{-1}\,\mathrm{cm}^{-1}$. For light to moderate doping levels (up to approximately one dopant per 20 carbon atoms) the addition of charge leads to the formation of charged soliton levels or domain walls.[7–12] Several detailed mechanisms, some involving the presence of solitons, have been proposed to account for the charge transport in different regimes of doping concentration. These include isoenergetic phonon-assisted hopping among solitons[13–15] and variable-range hopping among fixed defect sites[5,6] (these defect sites could also be pinned solitons[16]). Comparison of the predicted ratio with the experimental results for $\sigma(\omega)$ of undoped *trans*-$(CH)_x$ shows agreement with isoenergetic hopping and disagreement with variable-range hopping. In the range of 1—5 % I_3^- per carbon atom, the results are in accord with variable-range hopping and in disagreement with isoenergetic hopping. The same diagnostics could be applied to other hopping systems such as amorphous semiconductors,[2] provided accurate high-frequency (infrared) and dc-conductivity data are available on identically prepared samples.

The only feature of our results that depends on anything but the distribution of site energies is the dependence of the maximum hopping rate Γ_0 on the various parameters such as temperature. Thus, in comparing the results of theory and experiment in polyacetylene, no discussion of the physical nature of the hopping process is included. The interested reader is referred to Refs. 13—15 for a discussion of intersoliton hopping and to Ref. 2 for a discussion of variable-range hopping.

In Sec. II we summarize our principal results for the conductivity ratio. The details of the calculation have been left to the Appendices. In Sec. III the experimentally measured conductivity ratio is compared with the calculated ratios for both undoped *trans*-polyacetylene and iodine-doped *trans*-polyacetylene. The results are summarized in Sec. IV.

II. CONDUCTIVITY RATIO: THEORY

In the class of hopping models we will consider, the electrons are assumed to hop between localized states or sites which we label by their spatial location $\vec{R}$. The set of site-occupation probabilities satisfy a master equation specified by the hopping rates $\gamma_{\vec{R}\,\vec{R}'}$, between each pair of sites, $\vec{R}$ and $\vec{R}'$. The thermally averaged transition rate is thus

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$$\Gamma_{\vec{R}\,\vec{R}'}=\bar{f}_{\vec{R}}\gamma_{\vec{R}\,\vec{R}'}(1-\bar{f}_{\vec{R}'}),$$

where $\bar{f}_{\vec{R}}$ is the thermally averaged site-occupation probability of site $\vec{R}$. These transition rates are random variables which can be expressed in the form

$$\Gamma_{\vec{R}\,\vec{R}'}=\Gamma_0\exp(-X_{\vec{R}\,\vec{R}'}),$$

where X is a well-behaved random variable which thus implies an extremely broad distribution of transition rates. In the case of isoenergetic hopping, we will assume that

$$X_{\vec{R}\,\vec{R}'}=2\,|\,\vec{\alpha}\cdot(\vec{R}-\vec{R}')\,|\,, \tag{2.1}$$

where

$$\vec{\alpha}=\sum_{j=1}^{3}\hat{e}_j\xi_j^{-1},$$

$\hat{e}_j$ is a unit vector in the j direction, and ξ_j is the decay length of the wave function in that direction. [Note, there may be logarithmic corrections to Eq. (2.1) in some cases, but these are small. Also, note that the results are insensitive to the form of the anisotropy of the falloff. For example, if

$$X_{\vec{R}\,\vec{R}'}=\left[\sum_{j=1}^{3}\alpha_j^2(R_j-R_j')^2\right]^{1/2},$$

our conclusions are unchanged.]

The distribution of X's is thus determined by the probability of finding a site at $\vec{R}'$ given that there is a site at $\vec{R}$. At large distances, this probability is simply the concentration $C_{\rm im}$ of (impurity) sites. In the case of variable-range hopping, we will take

$$X_{\vec{R}\,\vec{R}'}=2\,|\,\vec{\alpha}\cdot(\vec{R}-\vec{R}')\,|+\frac{|E_{\vec{R}}|+|E_{\vec{R}'}|+|E_{\vec{R}}-E_{\vec{R}'}|}{2k_BT}, \tag{2.2}$$

where $E_{\vec{R}}$ is the energy of the state at site $\vec{R}$. [In cases of strong electron-phonon coupling, Eq. (2.2) may not be an adequate approximation, but we will not treat that complication here.] The distribution of X's is determined by the density of states $\rho(E)$, which we assume to be independent of energy in the vicinity of E_F.

At high frequency, the conductivity can be evaluated simply using the Einstein relation between the conductivity and the diffusion constant

$$\sigma(\omega)=\frac{e^2}{6k_BT}\frac{1}{\Omega}\left\langle\sum_{\vec{R},\vec{R}'}(\vec{R}-\vec{R}')^2\Gamma_{\vec{R}\,\vec{R}'}\right\rangle\times\left[1-\frac{G\Gamma_0}{\omega}+\cdots\right], \tag{2.3}$$

where G is a model-dependent number of order 1, $\langle\ \rangle$ represents a configuration average, Ω is the volume, and σ is averaged over polarizations. This expression is evaluated for the case of isoenergetic hopping in Appendix A and variable-range hopping in Appendix B.

The dc conductivity is considerably more difficult to calculate. It can be evaluated asymptotically in the limit of extreme disorder using a percolation analysis.[17] A similar analysis can be applied in the general case, and the resulting expression is known[17] to interpolate smoothly between the large and small disorder limits, and to agree well with numerical experiments in the intervening regimes. In this spirit, approximate expressions for the dc conductivity are also obtained in the Appendixes.

The results can be summarized as follows. For isoenergetic hopping,

$$\frac{\sigma(\infty)}{\sigma(0)}=\frac{9D}{32\pi A}\left[\frac{\xi}{R_0}\right]^4F\left[\frac{2BR_0}{\xi}\right]e^{2BR_0/\xi}, \tag{2.4}$$

where $A=0.45$, $B=1.39$, $D\approx1$, and $F(x)$ is given by Eq. (A11) and is equal to 1 for large x. For variable-range hopping (Appendix B),

$$\frac{\sigma(\infty)}{\sigma(0)}=\frac{3\pi(2.3)}{0.2}\left[\frac{T}{T_0}\right]^{3/2}\times G\exp[+(T_0/T)^{1/4}]\left[1+O\left[\left[\frac{T}{T_0}\right]^{1/4}\right]\right], \tag{2.5}$$

where $G\approx1$.

In both cases, $\sigma(\omega)$ is an increasing function of ω, and only approaches its infinite-frequency value when $\omega>\Gamma_0$. Note again that the conductivity ratio is independent of the complicated physics that determines the absolute magnitude of the transition rates. Only the frequency at which $\sigma(\omega)$ saturates depends on Γ_0.

III. APPLICATION TO POLYACETYLENE

A. Undoped and lightly doped *trans*-polyacetylene

The frequency-dependent conductivity (0–10^6 Hz) has earlier been reported for room temperature[18] and as a function of temperature.[3] At 295 K, σ is independent of ω to 10^6 Hz, with $\sigma(0)\approx8\times10^{-6}\ \Omega^{-1}\,\mathrm{cm}^{-1}$, while the fair-infrared data of Hoffman *et al.*[4] gives $\sigma(10^{13}\ \mathrm{Hz})\sim10^{-1}\ \Omega^{-1}\,\mathrm{cm}^{-1}$). The $\sigma(\omega)$ is only weakly dependent on ω for greater than 10^{13} Hz, suggesting $\Gamma_0\lesssim10^{13}$ Hz. These results give

$$\sigma(\infty)/\sigma(0)=1.3\times10^4\,.$$

This experimental result can be compared with the prediction of isoenergetic hopping [Eq. (2.4)] and variable-range hopping [Eq. (2.5)].

A detailed analysis of this material within isoenergetic hopping was done in Ref. 3, which leads to the value of the parameters $C_{\rm im}=1.3\times10^9\ \mathrm{cm}^{-3}$ (2.35×10^{-4} per carbon), and the dimensionally averaged decay length $\xi=(\xi_1\xi_2\xi_3)^{1/3}\approx3.58$ Å, so $2BR_0/\xi=20.6$, $F(2BR_0/\xi)=1.065$, and, from Eq. (2.3),

$$\sigma(\infty)/\sigma(0)\sim6\times10^4\,,$$

in good agreement with experiment. On the other hand, if the dc conductivity is assumed to be due to variable-range hopping, T_0 is found[19] to be $T_0=1.9\times10^9$ K, and hence

$$\sigma(\infty)/\sigma(0)\sim4\times10^{13}\,,$$

in clear disagreement with experiment. Since it seems unlikely that the value of Γ_0 can be much greater than an optical-phonon frequency [typically of order 5×10^{13} in $(CH)_x$], the fact that $\sigma(\omega)$ has reached its infinite-frequency value for $\omega\sim10^{13}$ Hz is consistent with these expectations.

The dc conductivity in polyacetylene is strongly temperature dependent. If the conductivity is due to isoenergetic hopping, this necessarily implies that Γ_0 must be strongly temperature dependent. For instance, at 84 K, $\sigma(0)\approx2\times10^{-14}\ \Omega^{-1}\,\mathrm{cm}^{-1}$, almost 9 orders of magnitude smaller than its value at 295 K. The highest frequency at which $\sigma(\omega)$ has been measured[3] at 84 K is $\omega=10^5$ Hz. There is indication that at this value of ω, the conductivity is beginning to saturate (see Fig. 1). Hence it seems reasonable to assume that $\sigma(\infty)\approx\sigma(10^5\ \mathrm{Hz})$. The conductivity ratio at this frequency is

$$\sigma(10^5\ \mathrm{Hz})/\sigma(0)=4\times10^3\ ,$$

which is nearly the same as that at 295 K, and in good agreement with the (temperature-independent) ratio predicted for isoenergetic hopping. By contrast, if the conductivity were due to variable-range hopping with $T_0=1.9\times10^9$ K, the ratio would be

$$\sigma(\infty)/\sigma(0)\sim5\times10^{22}\ .$$

In the case of isoenergetic hopping, the majority of the temperature dependence of the dc conductivity is determined by the temperature dependence of Γ_0 [see Eq. (A8)]. In Ref. 3 it was found that this temperature dependence could be fit by a high power-law dependence $\Gamma_0(T)=(\mathrm{const})T^{14.7}$ (a form suggested in Ref. 15 as the result of a crude approximate evaluation of the integral over the electron-phonon coupling constants that determine the rate of intersoliton hopping). Thus, a second independent consequence of the assumption that the conducting mechanism is isoenergetic hopping is that $\Gamma_0(84\ \mathrm{K})/\Gamma_0(300\ \mathrm{K})\approx7\times10^{-9}$. Thus a value of $\Gamma_0(84\ \mathrm{K})\approx7\times10^4$ Hz is the largest possible value consistent with $\Gamma_0(295\ \mathrm{K})\sim10^{13}$ Hz. We see, therefore, that the constraints imposed by these two observations can just be satisfied by a model of isoenergetic hopping. We note, in fact, that from an analysis of the ac conductivity over the full range of temperature and frequencies measured in Ref. 3, that it was concluded that the proportionality constant in Eq. (2) of Ref. 3 is $1\times10^{24}\ \mathrm{sec\,K}^{14.7}$, or in other words, that $\Gamma_0(84\ \mathrm{K})\approx2\times10^4$ Hz and $\Gamma_0(295\ \mathrm{K})\approx3\times10^{12}$ Hz, in reasonable agreement with the results of the present paper.

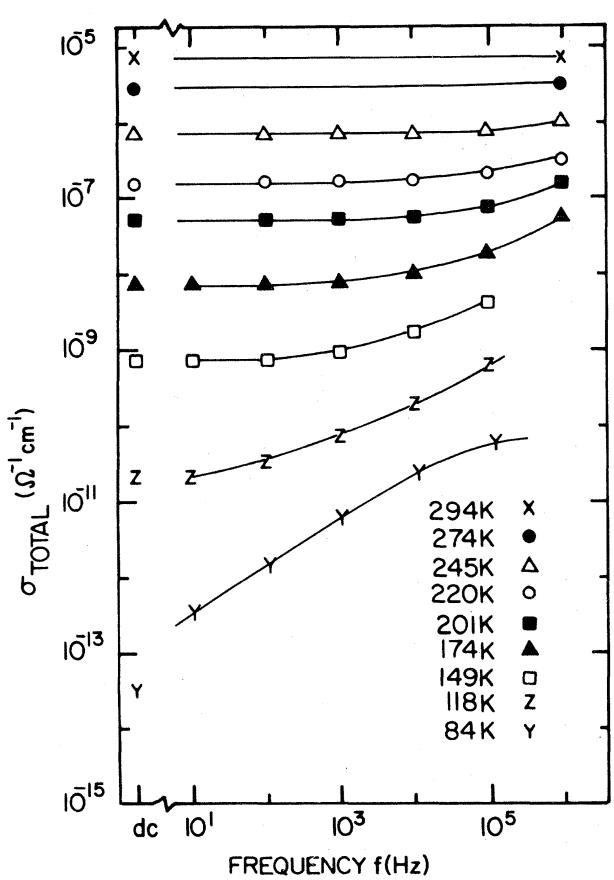

FIG. 1. $\ln\sigma_{ac}$ vs $\ln f$ for *trans*-$(CH)_x$ (Ref. 3). The solid lines are drawn as a guide for the eye.

B. Moderately doped polyacetylene

Polyacetylene doped to the range of 0.005–0.05 dopant per carbon has been shown to have high dc conductivity together with a magnetic susceptibility much less than that of the metallic state.[5,11,16,20–22] The conductivity of iodine-doped samples had earlier been shown[5] to be frequency independent up to 10^9 Hz. Infrared conductivity data show[16] σ to be independent of frequency up to 10^{13} Hz for samples doped in the range of 0.03–0.05 per carbon. Hence for this regime,

$$\sigma(\infty)/\sigma(0)\approx1\ . \tag{2.5}$$

In this regime, the predictions of Eq. (2.4) and (2.5) are less sensitive to choice of parameters. For isoenergetic hopping, Eq. (8) gives

$$\sigma(\infty)/\sigma(0)\approx20\ ,$$

for $\xi\sim3.6\times10^{-8}$ cm and $R_0\approx1.2\times10^{-7}$ cm. Here ξ was chosen as the three-dimensionally averaged soliton decay length,[15] and R_0 was chosen as appropriate for 0.05 dopants per carbon.[16] Larger values of R_0 (lower doping levels) lead to larger values of $\sigma(\infty)/\sigma(0)$. Doubling the assumed three-dimensional decay length to 7 Å and assuming that there is a minimum hop distance, $R_{\min}=1.2\times10^{-7}$ cm, still leads to a value of $\sigma(\infty)/\sigma(0)\approx3.5$ somewhat larger than the experimental ratio.

The predictions for variable-range hopping may also be compared with the available iodine-doped data[16] for 0.03–0.05 I_3^- per carbon. Here, T_0 varies from 3.8×10^5 to 2.9×10^6 K, with the result that

$$\sigma(\infty)/\sigma(0)\approx1$$

for $T_0\sim10^6$ K and $\sigma(\infty)/\sigma(0)\approx2.3$ for $T_0=2.9\times10^6$ K. Contrasting these results with those for isoenergetic hopping shows better agreement between theory and experiment for the variable-range–hopping approach. We note, however, that due to the high concentration of sites in these samples, we are in both cases using the expressions

for the dc conductivity beyond the regime where they are formally justified. Indeed, no good treatment of hopping conduction at high concentration of sites exists; in this limit the theoretical results should be used with caution.

IV. SUMMARY

Hopping mechanisms for conductivity are all characterized by a broad distribution of hopping rates. We have calculated the ratio of the conductivity at high frequency (frequency greater than the maximum hopping rate Γ_0) to its dc value. The results for isoenergetic hopping and variable-range hopping differ, although both are found to be independent of the details of Γ_0.

Trans-$(CH)_x$ and doped *trans*-$(CH)_x$ span a wide range of conductivities. Several different regimes have been identified, and it has been suggested that in each the conductivity is dominated by a different charge transport mechanism. The results for isoenergetic intersoliton electron hopping are in good agreement with the detailed experimental frequency and temperature-dependent conductivity of undoped *trans*-$(CH)_x$. The experiments are inconsistent with variable-range hopping. In *trans*-$(CH)_x$ doped to 0.01—0.05 I_3 per carbon, the data are consistent with variable-range hopping, while the ratio is smaller than that which would be expected for isoenergetic hopping. These results then support a change in the dominate charge transport mechanism as *trans*-$(CH)_x$ is doped to the nearly metallic regime.

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APPENDIX A: ISOENERGETIC HOPPING

An essential aspect of the intersoliton electron hopping developed by one of the authors is the isoenergetic nature of the movement of the charge among soliton sites.[13–15] A similar situation can occur in the case of polaron hopping. For isoenergetic hopping, the hopping rate between sites separated by displacement $\vec{R}$ is approximately

$$\Gamma_{nm}(0)=\Gamma_0 e^{-2|\vec{\alpha}\cdot\vec{R}|}\ , \tag{A1}$$

where

$$\vec{\alpha}=\frac{\hat{e}_1}{\xi_1}+\frac{\hat{e}_2}{\xi_2}+\frac{\hat{e}_3}{\xi_3}\ , \tag{A2}$$

$\hat{e}_i$ is the unit vector in the ith direction, and

$$\xi\equiv(\xi_1\xi_2\xi_3)^{1/3} \tag{A3}$$

is the effective dimensionally averaged localization length, $\vec{\xi}=\xi_1\hat{e}_1+\xi_2\hat{e}_2+\xi_3\hat{e}_3$.

By assuming that the probability of finding a site at the origin $(\vec{0})$ is c (concentration of sites per unit volume) and that the probability of finding a site at $\vec{R}$, given that there is a site at $\vec{0}$, is $P(\vec{R})d\vec{R}$, Eq. (2.3) becomes

$$\sigma_j(\infty)=\frac{e^2\Gamma_0 c}{2k_BT}\int d\vec{R}(R_j)^2P(\vec{R})e^{-2|\vec{\alpha}\cdot\vec{R}|}\ . \tag{A4}$$

Here σ_j is the conductivity in the jth direction, and

$$\sigma(\infty)=\tfrac{1}{3}(\sigma_1+\sigma_2+\sigma_3)\ . \tag{A5}$$

By replacing $\vec{R}$ by $\vec{\xi}r$ where r is dimensionless,

$$\sigma_j(\infty)=\frac{e^2\Gamma_0c^2}{k_BT}\left[\frac{\pi}{2}\right]\xi^3\xi_j^2D_j$$
$$=\frac{9}{32\pi}\frac{e^2\Gamma_0}{k_BTR_0}\left[\frac{\xi}{R_0}\right]^3\left[\frac{\xi_j}{R_0}\right]^2D_j\ . \tag{A6}$$

Here R_0 is the average separation between impurities and

$$D_j=\int d\vec{R}(r_j)^2e^{-2r}\left[\frac{P(\vec{\xi}r)}{\pi c}\right]\ . \tag{A7}$$

Equations (A6) and (A7) were defined so that D_j is very close to 1. That is, D_j depends only on the short-range behavior of $P(\vec{R})$, (where $|\vec{\alpha}\cdot\vec{R}|\sim 1$). It is chosen so that if there are no short-range correlations in the placement of sites [that is, if $P(\vec{R})=c$], then $D_j=1$.

At zero frequency (the dc limit), $\sigma_j(0)$ is given asymptotically by the expression

$$\sigma_j(0)=\frac{A\Gamma_0e^2}{k_BTR_0}\left[\frac{\xi}{R_0}\right]\left[\frac{\xi_i}{\xi}\right]^2e^{-2BR_0/\xi}\ , \tag{A8}$$

where $A=0.45$ and $B=1.39$. This is in agreement with Eqs. (B8) and (B9) of Ref. 23 for the low site density limit, where $S_c\gg 1$, where

$$S_c=\left[\frac{6P_c}{\pi c\xi^3}\right]^{1/3} \tag{A9}$$

and P_c is the critical percolation number. For higher concentrations (larger c, smaller S_c), $\sigma_j(0)$ is approximately

$$\sigma_j(0)=\frac{A\Gamma_0e^2}{k_BTR_0}\left[\frac{\xi}{R_0}\right]\left[\frac{\xi_j}{\xi}\right]^2e^{-2BR_0/\xi}[F(2BR_0/\xi)]^{-1}\ , \tag{A10}$$

where

$$F(x)=[1+x^{-1}+7x^{-2}+12x^{-3}+12x^{-4}-x^2e^{-x}]^{-1}\ . \tag{A11}$$

Dividing Eq. (A6) by Eq. (A10), we obtain

$$\frac{\sigma_j(\infty)}{\sigma_j(0)}=\frac{9D_j}{32\pi A}\left[\frac{\xi}{R_0}\right]^4F\left[\frac{2BR_0}{\xi}\right]e^{2BR_0/\xi}\ . \tag{A12}$$

APPENDIX B: VARIABLE-RANGE HOPPING

When there is a broad distribution of site energies and weak electron-phonon interactions, the hopping rate is approximately given by an expression of the form

$$\Gamma_{nm}^{(0)}=\Gamma_0\exp[-2|\vec{\alpha}\cdot\vec{R}|$$
$$-\tfrac{1}{2}(|E_n|+|E_m|+|E_n-E_m|)/k_BT]\ . \tag{B1}$$

If the probability of finding a site with energy E at the origin is ρ_0 [independent of E within several k_BT of E_F and $\int \rho_0(E)dE = C$] and the probability of finding a site at $\vec{R}$ with energy E' given a site $\vec{0}$ with energy E is $P(E \mid E',\vec{R})$, then

$$\sigma_j(\infty) = \frac{e^2\Gamma_0\rho_0}{2k_BT} \times \int dE_1 dE_2 d\vec{R}(R_j)^2 P(E_1 \mid E_2,\vec{R}) \times e^{[-2|\vec{\alpha}\cdot\vec{R}| + (1/2)(|E_1|+|E_2|+|E_1-E_2|)/k_BT]} \tag{B2}$$

or

$$\sigma_j(\infty) = (3\pi)\frac{e^2\Gamma_0}{k_BT}(\rho_0 k_BT)^2\xi^3(\xi_j)^2 G_j \ , \tag{B3}$$

where

$$G_j = \int dE_1 dE_2 \frac{d\vec{R}}{18\pi}\frac{P(E_1k_BT \mid E_2k_BT, r\vec{\xi})}{\rho_0}\vec{R}^2 \times e^{-2r} e^{-(1/2)(|E_1|+|E_2|+|E_1-E_2|)} \tag{B4}$$

and $E_ik_BT = E_i$. If we define

$$k_BT_0 = 2.3/\rho\xi^3 \ , \tag{B5}$$

then

$$\sigma_j(\infty) = (3\pi)(2.3)^2\left[\frac{e^2\Gamma_0}{k_BT\xi}\right]\left[\frac{T}{T_0}\right]\left[\frac{\xi_j}{\xi}\right]^2 G_j \ . \tag{B6}$$

For the simple case where $P(E \mid E',\vec{R}) = \rho_0$, $G_j = 1$.

The dc conductivity for the case of variable-range hopping is given approximately by Eqs. (15a) and (15b) or Ref. 23,

$$\sigma_j(\omega=0) = 0.2e^2\Gamma_0\rho_0\xi_j^2\left[\frac{T_0}{T}\right]^{1/2}\exp\left[-\left[\frac{T_0}{T}\right]^{1/4}\right] . \tag{B7}$$

The ratio of Eqs. (B6) to (B7) then gives

$$\frac{\sigma_j(\infty)}{\sigma_j(0)} = \frac{3\pi(2.3)}{0.2}\left[\frac{T}{T_0}\right]^{3/2} G_j \exp\left[+\left[\frac{T_0}{T}\right]^{1/4}\right] . \tag{B8}$$

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