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The Physical Significance of a for Electron Transport

USU Candidacy Exam Report

Zachary Gibson

Abstract:

Charge transport in crystalline materials can be well understood through the use of Bloch functions, band theory, and extended state transport. Disordered materials do not allow the same luxury, and different methods have to be considered. It turns out that band structures still exist. Although extended state conduction can still occur, localized states within the band gap allow for alternative mechanisms of charge transport in disordered materials. Conduction can be understood through such models as hopping, multiple trapping, and percolation, but all of them can lead to a broad distribution of event times. For example, the hopping-time distribution proposed by Scher and Montroll based on continuous time random walk formalism to describe transient photocurrent time-of-flight experiments, $\psi(t) \propto t^{-(1+\alpha)}$, where ' α ' is referred to as the dispersion parameter. This form of distribution exhibits a long tail. If the distribution of event times is broad enough to contain times on the order of the experiment then dispersive transport is observed. At sufficiently high temperatures or electric fields a transition from dispersive to normal transport can occur due to a shift in the occupancy of the states extending into the mobility gap to shallow traps. This results in shorter event times, leaving out times on the order of the experiment and dispersive transport is no longer observed. In the case of an exponential energetic density of states extending into the mobility gap, the dispersion parameter α is linearly proportional to either temperature or electric field. In overlapping regimes, descriptions are more complex. The dispersion parameter is scaled by the reciprocal of a characteristic energy that corresponds to the width of the density of states in the mobility gap and defines both a temperature and electric field corresponding to a transition from dispersive to normal transport when the dispersion parameter is one. These transition temperatures and fields appear to have significant meaning for a given material, such as the electric field strength associated with the onset of breakdown or a glassy transition temperature. While the dispersion parameter shows up in models involving other density of states, the simplicity of the insight is lost in more complex descriptions.

1. Introduction

A concrete understanding of the nature of electrical properties is elusive when it comes to disordered media. Insight has been gained through the help of dispersive transport models which can rely on a single parameter, the dispersion parameter. This gives rise to universal plots and hints at the energetic density of states in the mobility gap [1, 2, 9]. Transport models attempt to link the microscopic nature of the underlying physical processes to macroscopic properties such as measured conductivity and permittivity. The significance of the dispersion parameter in describing anomalous measurements and electrical properties of disordered media is the focus of this paper.

Charge transport in crystalline material is well understood through the use of extended state Bloch functions. This leads to the concept of band structure and band gaps, which helps to navigate the electrical properties of crystalline materials [10]. These methods are all but thrown out the window when there is a departure from crystalline structure, such as that introduced by defects or outright disordered materials with no long-range order. The conduction mechanisms in disordered materials have come to be understood through various mechanisms such as thermally assisted hopping, variable range hopping, multiple trapping, and percolation theories. The common theme of these conduction mechanisms revolves around the idea of localization. These localized states appear within the band gap, that is fittingly referred to as the mobility gap for disordered media (refer to Fig. 1) [1, 11, 12].

It turns out that small variations from crystalline/ordered materials can result in a broad distribution of event times. Event times can be hopping times, relaxation times, capture/release transition rates, etc. corresponding to characteristic times of an event related to a conduction mechanism. These broad event times result in dispersive transport [13], which can be seen in I(t) current versus time plots for experiments such as constant voltage conductivity (CVC) [4, 5, 14, 15], time-of-flight transient photoconduction [2, 9, 16, 17], and charge decay experiments [18-20]. Broad distribution functions of the form $\psi(t) \propto t^{(1+\alpha)}$ have been shown by Scher and Montroll [2] to reduce the dispersive transport problem to a single free parameter ' α '. This was also accomplished by Cole and Cole [4, 15] with their semi-empirical description of the anomalous properties of the real and imaginary permittivity. This parameter is often referred to as the dispersion parameter. The dispersion parameter can then be used to define a characteristic width of the energetic density of states extending into the mobility gap [20, 21]. For example, in the case of an exponential density of states the temperature dependence of the dispersion parameter is scaled by the reciprocal of the width of the distribution (width defined as energy where density of states drops to 1/e). In other words, given that $\alpha = T/T_c$, where $k_B T_c$ = characteristic energy [6, 9, 20-28], the width of an exponential energetic density of states in the mobility gap is $k_B T_c$, where k_B is the Boltzmann constant, T is temperature, and T_c is the characteristic temperature. It

may be that T_c is the glassy transition temperature in some materials. It appears that T_c corresponds to a characteristic energy ($k_B T_c$) related to the energetic density of states in the mobility gap and α relates to the occupancy of these states.

The following sections will give a brief overview of crystalline conduction theory and conduction mechanisms that arise in disordered media, which lead to dispersive transport models. These models will be discussed in detail, particularly that of Scher and Montroll [2] based on continuous time random walks. Comparisons of various models and experiments will be made. Since a number of these models depend on the parameter α , this naturally leads to the discussion of the physical significance of this parameter.

2. Electron Transport Theory

Electron transport in crystalline materials can be understood through the introduction of a periodic potential in a lattice. It was discovered by F. Bloch that the Schrödinger equation can be solved analytically for any infinite periodic potential. The solutions are wavefunctions of the form,

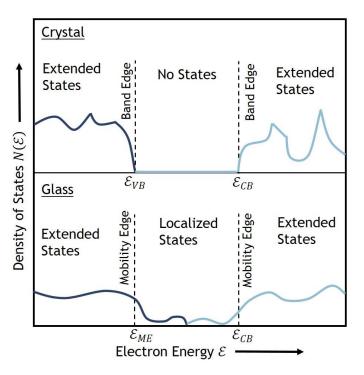


Figure 1. Idealized band diagrams for periodic (top) and amorphous non-conducting materials (bottom). From (Allen).

$$\psi_k(t) = u_k(r) \mathrm{e}^{ikr},\tag{1}$$

where, $u_k(\mathbf{r})$ is the periodic potential with the same period of the crystal lattice, \mathbf{k} is the wave vector, and \mathbf{r} is the position vector. This leads to the existence of band structures [10]. This allows for the differentiation of electronic properties according to the electron occupancies of the outermost band. In other words, the outermost band (in energy) is either partially filled (conductors) or exactly filled (semiconductors and insulators). The difference between semiconductors and insulators is the difference in energy between the filled band and the next available band. Semiconductors are somewhat arbitrarily defined as having a bandgap of ~100 times $k_B T_{room}$ or less, and insulators above that. In metals, electrons can move around essentially freely in extended states in the partially filled conduction band.

In disordered materials, there is no longer any long-range order (refer to Fig. 2). This means that Bloch functions (Eqn. (1)) no longer apply due to a lack of a periodicity. However, it turns out that something like band structures still exist in amorphous materials as validated by experiments measuring a dispersion like relation, relating the real electron momentum, rather than the usual crystal momentum, to the energy [29-32]. For disordered materials, the band gap is referred to as the mobility gap and states within the mobility gap are localized (refer to Fig. 1).

2.1 Amorphous Materials – Localization

The understanding of materials through use of Bloch functions falls apart when disorder is introduced. The underlying assumption of periodicity can no longer be used. Some insight can still be gained through simple energetic and spatial perturbations from periodicity. These two perturbations correspond to two transitions from metal to insulators described by Mott [33, 34] and Anderson [35] by increasing spatial separation and adding energetic disorder respectively. Figures 3 and 4 show Mott and Anderson transitions respectively, as explained by Table 1. If the width 'W' or energy 'U' is greater than the bandwidth 'B', then a transition to insulator with localized states occurs. This is qualitatively simple to understand, since it seems intuitive that at some point the states will no longer interact with each other to form extended states if they are separated enough in either energy or space. The underlying mathematics are much more complex [1, 11, 33-36].

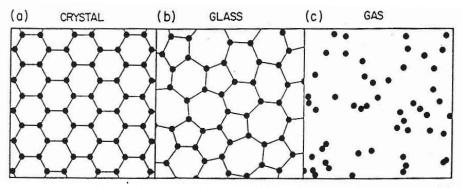
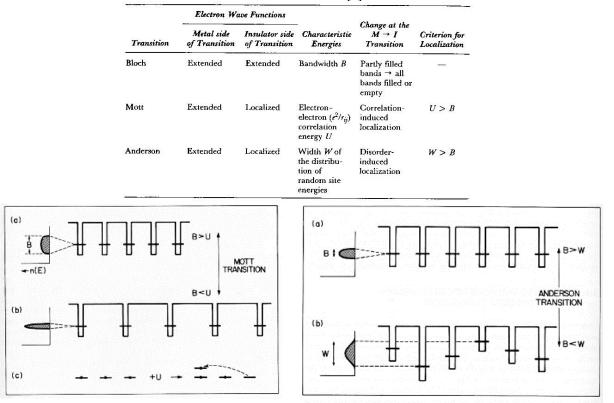


Figure 2. 2D representation of atoms in (a) crystalline material (b) glassy/disordered material (c) and gas. Crystals exhibit long range order. Glass can be seen to have short range order. Gas shows no order and the atoms do not have a steady state equilibrium point. From [1].

Table 1. Metal insulator transitions. From [1]



When the spatial separation is enough to decrease the the electron-electron energy U, correlation-induced induced localization takes place. From [1]. localization takes place. From [1].

Figure 3. Schematic diagram of the Mott transition. Figure 4. Schematic diagram of the Anderson transition. When the energetic disorder causes the width W to be electron bandwidth B to be sufficiently smaller than sufficiently larger than the bandwidth B, disorder-

Often in crystalline materials, conduction via extended states dominate. In disordered materials where localized states are present, conduction can be dominated by hopping or multiple trapping. This is the topic of discussion in the following section.

2.2 Amorphous Materials – Conduction Mechanisms

Extended state conduction is still possible in disordered materials. However, sufficient energy must be given to an electron to excite it into the conduction band. This is more easily accessible for electrons in shallow traps near the mobility edge. Often, thermally assisted hopping (TAH) refers to the process of electrons excited due to thermal

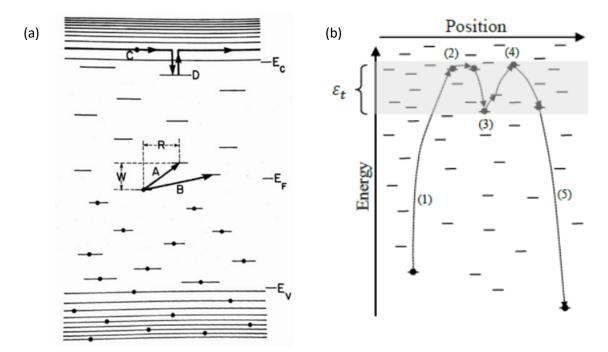


Figure 5. (a) Schematic diagram of electronic conduction in an amorphous material. Energy is the vertical direction and position in the horizontal. From [1]. (b) Depiction of VRH. (1) Charge carrier excited from deep trap; (2)-(4) TAH like conduction occurs; (5) Charge carrier falls back into deep trap [6].

energy from shallow trap states up into the conduction band where they can conduct for a time before they fall back into a trap state, as seen in paths C and D in Fig. 5 (a) [1]. This is more accurately referred to as multiple trapping (MT). MT is a more fitting name as hopping seems to imply a quantum tunneling aspect which is not applicable in this description of TAH. TAH also refers to hopping conduction such as depicted in paths A and B in Fig. 5 (a) with energetic and spatial separations of W and R respectively [7]. TAH will be referred to in this way (hopping) exclusively throughout the rest of the paper. Variable range hopping (VRH) dominates when the temperature is sufficiently low that MT is negligible and quantum effects take over. VRH is phonon assisted quantum tunneling (hopping). Referring to Fig. 5 (b), VRH includes a phonon assisted jump to a transport energy (highlighted in grey), where TAH like conduction occurs for a time before falling back into a deep trap state [7]. If excess energy is added through irradiation then radiation induced conductivity (RIC) can occur [14, 37-39]. Similarly to VRH, an electron in a deep trap state is excited into the conduction band in an extended state via irradiation (as opposed to a phonon assisted jump to a localized state within the mobility gap for VRH), where it will conduct for a time but will eventually fall back into a trap state. Schematically, this would correspond to a jump from a deeper trap, such as that shown in Fig 5 (b) with the grey area replaced with extended states in the conduction band. From there, the electron may continue to undergo MT or fall back into a deep trap state. Analogously, these same mechanisms can apply to hole transport if the electron vacancies are now considered hole occupancies. In other words, the valence band is essentially the conduction band for holes [7]. The focus will be on electron transport throughout this paper.

Another way to look at conduction in disordered materials is through percolation theories [1, 9, 28]. Zallen [1] explains it simply as if a saboteur was trying to cut communications between two locations, how many links would need to be cut before connection was completely cut-off? Refer to Fig. 6. This can be described as the fraction of links/bonds that need to be severed for a metal-insulator transition to occur. In other words, this critical percolation threshold defines an Anderson transition. This idea of blocked and unblocked sections of networks (or networks of resistors) can be extended to understand many different things. This will not be explored here, but a brief list of applications of percolation theory are shown in Table 2. An in depth review of percolation and fractal theory in the context of diffusion in disordered media is given by Havlin [40].

It has been shown by Pfister and Scher [9] that any of these transport models (hopping, MT, and percolation) can lead to broad distributions of event times, and therefore dispersive transport.

Table 2. Applications of percolation theory. From [1].

Phenomenon or System	Transition
Flow of liquid in a porous medium	Local/extended wetting
Spread of disease in a population	Containment/epidemic
Communication or resistor networks	Disconnected/connected
Conductor-insulator composite materials	Insulator/metal
Composite superconductor-metal materials	Normal/superconducting
Discontinuous metal films	Insulator/metal
Stochastic star formation in spiral galaxies	Nonpropagation/propagation
Quarks in nuclear matter	Confinement/nonconfinement
Thin helium films on surfaces	Normal/superfluid
Metal-atom dispersions in insulators	Insulator/metal
Dilute magnets	Para/ferromagnetic
Polymer gelation, vulcanization	Liquid/gel
The glass transition	Liquid/glass
Mobility edge in amorphous semiconductors	Localized/extended states
Variable-range hopping in amorphous semiconductors	Resistor-network analog

3. Anomalous Transport

Anomalous transport in dielectrics has been studied for well over a century. The effects appear in experiments such as measurements of AC and DC conductivity [4, 5, 8, 15], photoconductivity [2, 9, 16, 17], radiation induced conductivity [14, 37-39, 41-43], charge decay [3, 18], and electrostatic breakdown [44, 45]. The models of such anomalous behavior rely on a broad distribution of event times [2, 9, 13]. It turns out that the underlying mechanisms are of secondary importance for modeling. As Pfister [9] puts it "...all disordered solids will exhibit a broad dispersion of

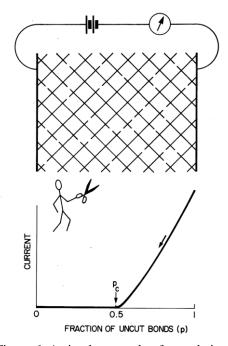


Figure 6. A simple example of percolation theory. At a critical fraction p_c of uncut bonds there is a threshold to transition from current flowing or not. From [1].

carrier transit times. These features are independent of the transport mechanism ... trivial fluctuation in x [defects, trap states, etc.] induce large fluctuations in the event times. The resulting event time distribution can be sufficiently broad that the experimental transit time becomes part of the distribution." In other words, disorder in materials causes broad event time distributions regardless of the underlying transport mechanisms. Note, an important point here is the distribution includes a time on the order of the experiment, such as the transit time. This is key as this is the determining factor of whether dispersive transport is observed [13].

The first mention of a broad distribution of event times as a reason for the observed anomalous/dispersive effects was in 1907 by v. Schweidler [46]. Later on in 1941, Cole and Cole [4, 15] published a few papers on the anomalous dispersion and absorption of dielectrics in AC and DC conditions. They use a modified version of Debye's equation for dielectric dispersion and absorption to derive an equation for the current I(t) with dependencies on t and α identical to those in Scher and Montroll's continuous time random walk (CTRW) theory describing transient photocurrent time-of-flight experiments [2]. Note that the relation between the Cole and Cole ' α_{cole} ' and the Scher and Montroll ' α_{SM} ' is $\alpha_{SM} = 1 - \alpha_{cole}$. Throughout this paper $\alpha_{cole} = \alpha'$ and $\alpha_{SM} = \alpha$.

An overview of models for the range of experiments, and their dependencies on the dispersion parameter, is presented here.

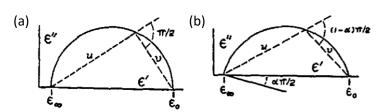
3.1 AC and DC Experiments

Interestingly enough, the description of AC and DC conductivity was explained in terms of the dispersion parameter over three decades [4, 15] before the landmark paper written by Scher and Montroll [2]. The semi-empirical formula starts from the classic theory of Debye for polar liquids [47]. This leads to equations for the real and imaginary parts of the permittivity.

$$\varepsilon^*(\omega) - \varepsilon_{\infty} = \frac{(\varepsilon_0 - \varepsilon_{\infty})}{(1 + i\omega\tau_0)}.$$
(2)

Separating the real and imaginary components the equations are,

$$\varepsilon' - \varepsilon_{\infty} = \frac{(\varepsilon_0 - \varepsilon_{\infty})}{[1 + (\omega\tau_0)^2]}$$
$$\varepsilon'' = \frac{(\varepsilon_0 - \varepsilon_{\infty})\omega\tau_0}{[1 + (\omega\tau_0)^2]}$$
(3)



where ε^* is the complex permittivity defined as, $\varepsilon^* = \varepsilon' - i\varepsilon''$, with ε' and ε'' being the real and imaginary parts of the permittivity, respectively. The parameters ε_0 and ε_{∞} are the limiting values of the

Figure 7. Above are plotted the (a) semi-circle Debye theory, and (b) circular arc as required by experimental data. From [4].

permittivity at very low and very high frequencies, respectively, $\omega = 2\pi$ frequency, and $\tau_0 \equiv 2\pi/\omega_0$ is a characteristic constant which may be called the relaxation time. Note that the relaxation time here differs from that defined by Debye by a constant factor which depends on the theory assumed for the static permittivity [48]. These equations have widespread applicability to explain dispersion, differing only in the significance of the parameters ε_0 , ε_{∞} , and τ_0 , ranging from paramagnetic dispersion [49, 50], to that of elastic dispersion and damping in metals subjected to alternating stress [51].

It is convenient to view these results in the form of an Argand diagram. This is where the real and imaginary parts of the complex permittivity are plotted against each other, each point corresponding to a different frequency. Cole and Cole [4] explain that Eqn. (2) can be written as $u + v = \varepsilon_0 - \varepsilon_\infty$, where $u \equiv \varepsilon^* - \varepsilon_\infty$, $v \equiv i\omega\tau_0(\varepsilon^* - \varepsilon_\infty)$. The vectors *u*, *v*, are perpendicular in the complex plane with their sum being the constant real quantity $\varepsilon_0 - \varepsilon_\infty$. Therefore these vectors inscribe a semi-circle of diameter $\varepsilon_0 - \varepsilon_\infty$, as shown in Fig. 7 (a). For Eqn. (2), the semi-circle is centered on the ε ' axis (real axis) at $1/2(\varepsilon_0 + \varepsilon_\infty)$ where ε_0 and ε_∞ are the ε' intercepts. However, for many materials this is not an accurate representation of the data. The center of the circle may shift down below the ε' axis. Materials exhibiting this anomalous behavior can be modeled with a circular arc rather than a full semi-circle [refer to Fig. 7(b)].

With consideration of Fig. 7, it can be easily deduced through a geometrical argument that,

$$\varepsilon^* - \varepsilon_{\infty} = \frac{(\varepsilon_0 - \varepsilon_{\infty})}{[1 + i^{(1 - \alpha_{cole})} f(\omega)]},\tag{4}$$

where $f(\omega)$ is a (real) undetermined function of the frequency and other parameters. Note that this leads to the conclusion that there is a distribution of relaxation times rather than a single relaxation function as assumed in Debye theories [52]. Cole and Cole [4] then go on to argue that the frequency dependence resulting from an assumed applied field of $E = E_0 e^{i\omega t}$ will be of the form $\omega^{1-\alpha_{cole}}$ (which is in agreement with Pfister, Scher, and Lax if $1 - \alpha_{cole} = s$ [9, 16, 17]). Cole and Cole state that any theory in which ω appears as the result of linear operations on the complex exponential must lead to the same functional dependence on both *i* and ω . This leads to a new equation for the complex permittivity.

$$\varepsilon^* - \varepsilon_{\infty} = \frac{(\varepsilon_0 - \varepsilon_{\infty})}{[1 + (i\omega\tau_0)^{1 - \alpha_{cole}}]}.$$
(5)

This is verified with experiment as seen in [4]. It can also be seen that for values of $\alpha_{cole} = 0$ ($\alpha_{SM} = 1$), the equations reduce to that of the Debye expressions for a single relaxation time.

With Eqn. (5) established, a theoretical description of the current for the DC case can be made. Starting with Eqn. (5), one can obtain an expression for the current as a function of time [15].

$$I(t) = \frac{\varepsilon_0 - \varepsilon_\infty}{\tau_0} (1 - \alpha_{cole}) \left(\frac{t}{\tau_0}\right)^{-(2 - \alpha_{cole})} \sum_{n=1}^{\infty} \frac{n(-1)^{n-1}}{\Gamma[1 - n(1 - \alpha_{cole})]} \left(\frac{t}{\tau_0}\right)^{-(n-1)(1 - \alpha_{cole})}.$$
 (6)

If Eqn. (6) is then taken to its asymptotic limits in t/τ_0 then,

$$I(t) = \frac{\varepsilon_0 - \varepsilon_\infty}{\tau_0} \frac{1}{\Gamma(1 - \alpha_{cole})} \left(\frac{t}{\tau_0}\right)^{-\alpha_{cole}}, \quad t \ll \tau_0$$

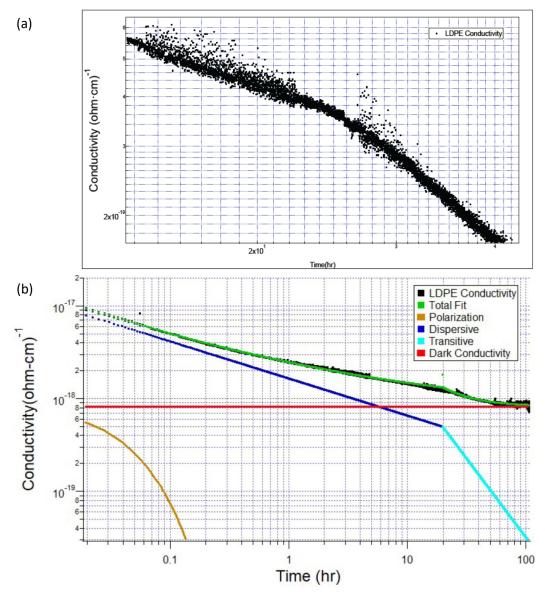


Figure 8. Double logarithmic plots of conductivity as a function of time with a constant voltage applied for lowdensity polyethylene (LDPE). Plot (a) is a zoomed in portion of the transition/kink denoting the transit time [5]. Plot (b) has fits using Eqn. (8) with each contribution to the conductivity shown separately and all together [8].

$$I(t) = \frac{\varepsilon_0 - \varepsilon_\infty}{\tau_0} \frac{(1 - \alpha_{cole})}{\Gamma(\alpha_{cole})} \left(\frac{t}{\tau_0}\right)^{-(2 - \alpha_{cole})}, \quad t \gg \tau_0, \tag{7}$$

where Γ is the gamma function. Note that this is similar to the mathematical approach taken by Scher and Montroll and one can easily show equivalent t and α dependencies by replacing $\alpha = 1 - \alpha'$ in Eqns. (7) [2]. This is shown in Appendix A. There is an easy check to this theory by observing that the slopes before and after the transition/kink in a double logarithmic I(t) plot add up to -2 because $(-\alpha') + (-2 + \alpha') = -2$. This is independent of the α' (or α) value.

The 'kink' in the double logarithmic I(t) curves can be seen in CVC data taken by the Materials Physics Group at Utah State University (MPG) (refer to Fig. 8) [5, 8]. The data can be understood using the equation [5, 7, 8, 53],

$$\sigma(t) = \sigma_P^{\frac{-t}{\tau_P}} + \left\{ \sigma_{disp} t^{-(1-\alpha)} \theta(\tau_{transit} - t) + \sigma_{trans} t^{-(1+\alpha)} \theta(t - \tau_{transit}) \right\} + \sigma_{DC}, \tag{8}$$

where $\tau_{transit}$ is the transit time, τ_p is the polarization time constant, σ_p is the polarization conductivity, σ_{disp} is the initial dispersive transport conductivity before the transit time, σ_{trans} is the initial dispersive transport conductivity after the

transit time, σ_{DC} is the dark current DC conductivity, and θ is the Heaviside function. The first term is due to polarization, the terms enclosed by the brackets are the conductivity before and after the transit time respectively (making use of the Heaviside function), and the last term is the dark conductivity. Each of these terms can be seen plotted along with the data in Fig. 8 (b). It is clear that the dispersive transport terms have the same t and α dependencies as that of Eqn. (7). Although this is conductivity and not current, it is clear through Ohm's law, $J = \sigma E$, that they should share the same dependencies.

Another form of the complex permittivity ϵ '' (for polar polymers) is modeled by Kirkwood and Fuoss as [54, 55],

$$\varepsilon'' \approx A \operatorname{sech}(\alpha x) \to H(x) = H(0) \operatorname{sech}(\alpha x)$$
 (9).

Where H(x) is an approximation to the imaginary permittivity and x is the natural logarithm of the ratio of frequency that maximizes H(x) over the measured frequency, $x = \ln \omega_m/\omega$. This can be seen to be equivalent to the Debye case when $\alpha = 1$, which corresponds to a single relaxation time. In the limit of $\alpha=0$, there is an infinitely broad distribution of relaxation times. It is shown in Appendix B that this relation can be derived starting from Eqn. (5), but is only a good approximation when $\alpha\approx 1$. Kirkwood and Fuoss [55] show that $H(0) = \alpha/2$, where H(0) is the maximum value of the function H(x). They found only qualitative agreement with their data; the exact form derived in Appendix B should be tried as a potentially better fit for experimental data. There is an interesting discussion in [56] about the relation between Cole and Cole equations, Fuoss and Kirkwood approximation and how these models can also apply to mechanical properties of polymers.

Note that the α values determined by CVC and other data are not always self-consistent. In other words, α will occasionally be different before and after the transit time [9, 28]. This is indicative that the density of states is not exponential and may be Gaussian or some other distribution [57]. A good overview of various density of states is given in [14] and [7].

3.2 Irradiation Experiments

Various conduction mechanisms in disordered media were described in Section 2. These hopping mechanisms are considered stochastic processes. Macroscopic properties of conduction in disordered media have been described semi-empirically in the previous section with use of a perturbed Debye equation for permittivity via the dispersion parameter, but to start to understand what is happening on a microscopic level a new approach is needed. This is why the Scher and Montroll paper [2] was deemed by Zallen [1] as the "anomalous to obvious" transition paper to describe dispersive transport via time of flight experiments in disordered media. This section will present a stochastic charge transport model via a random walk through a disordered material.

The stochastic theory of continuous time random walks (CTRW) is paramount for the modeling of time-of-flight transient photocurrent data, here on referred to as TOF experiments. Montroll has published a series of papers spanning two decades on random walks on lattices to develop CTRW formalism and show its equivalency to random walks [58-61]. A random walk describes a walker (in this case a charge carrier, either a hole or electron) on a spatially random lattice, while a continuous time random walk shifts to adding disorder via a distribution of event times. In other words, random walks are continuous (disordered) in space and discrete in time while CTRW are discrete in space (ordered lattice) and continuous in time (broad event-time distribution function). The distribution of event times contains the disorder. A small variation in properties related to the conduction mechanisms leads to broad distributions of event times [9, 16, 17, 61]. Before discussing the model and novel distributions presented by Scher and Montroll [2], a discussion of a series of papers by Scher and Lax [16, 17] is necessary. Scher and Lax start from first principles to build their theory, and their model can be shown to be equivalent to Scher and Montroll's results when t~t_{transit}. This comparison is used as a physical justification of the Scher and Montroll model.

Scher and Lax use CTRW on a lattice to approach the problem of transport in disordered solids. The approach is to model charge transport through stochastic theory in a way that is tractable analytically. This is the reason a CTRW on a discrete lattice is used rather than a random walk on a spatially disordered lattice with discrete event times. This allows a distribution of event times to be used to represent the stochastic processes of conduction rather than an ensemble average of spatial disorder. This is justified by equivocating random walks and CTRW in their appendix [17]. The mathematical justification will not reproduced here. This allows all of the disorder to be included through the use of a distribution of event times.

The goal is to obtain a function for the conductivity as a function of frequency $\sigma(\omega)$ for a disordered solid. It turns out that the conductivity can be described by the equations,

$$\sigma(\omega) = \frac{ne^2}{kT} D(\omega) \tag{10}$$

$$D(\omega) = -\frac{1}{6}\omega^2 \sum_{s,s_0} (s - s_0)^2 \tilde{P}(s,\omega;s_0) f(s_0)$$
(11).

Here, D is the diffusion constant, n is the density of effective carriers, e is the elementary charge, k is the Boltzmann constant, T is the temperature, s is the position vector of the walker, s₀ is the position vector of the carrier at time zero, \tilde{P} is the Fourier transform of P, P is the probability of a walker that was at position s₀ at t = 0 to be at position s at time t, and f(s₀) is the distribution of walkers at time t = 0. Eqns. (10) and (11) are the first principles physical basis of the model.

There are then two ways to find a solution to Eqns. (10) and (11). One is to approximate a self-consistent solution through the use of transport equations. The second, and the one used here, is to find an exact solution through the calculation of $P(s, t; s_0, 0)$. No transport equations are used. The approach is then to calculate P using a CTRW stochastic model. This turns out to be a mathematically involved problem and one can review the papers if they wish [16, 17]. It turns out that,

$$\tilde{P}(\boldsymbol{s},\boldsymbol{u}) = \tilde{R}(\boldsymbol{s},\boldsymbol{u})\frac{1-\tilde{\psi}(\boldsymbol{u})}{\boldsymbol{u}}$$
(12).

Where $\tilde{R}(\mathbf{s},\mathbf{u})$ is the Fourier transform of the random walk recursion relation and

$$\psi(\mathbf{s},t) = p(\mathbf{s})\psi(t) \tag{13},$$

where $\tilde{\psi}(u)$ is the Fourier transform of the hopping time distribution function $\psi(t)$, and $\psi(s,t)$ is the probability per unit time that the displacement and time between hops is s and t. Eqn. (13) just states that the event time distribution is independent of position. With this in place, $D(\omega)$ and therefore $\sigma(\omega)$ can be determined by

$$D(\omega) = \frac{\frac{1}{6}\sigma_{RMS}^2(\omega)i\omega\tilde{\psi}(i\omega)}{1-\tilde{\psi}(i\omega)}$$
(14)

$$\sigma_{RMS}^2 \equiv \sum_{s} s^2 \frac{\tilde{\psi}(s, i\omega)}{\tilde{\psi}(i\omega)}$$
(15).

Now the conductivity is determined by the 0th and 2nd spatial moments of the Fourier transform of the distribution of event times. This can then be applied to impurity conduction in a semiconductor to calculate $\psi(t)$ [16]. This will not reproduced here. It turns out that the form of $\psi(t)$ for impurity conduction in a semiconductor with the time approximately that of the transit time can be shown to be equivalent to the $\psi(t)$ used by Scher and Montroll.

if
$$\alpha \sim \frac{1}{3}\gamma(\ln t)^2$$
 and $t \sim t_{transit}$ then $\psi(t) \sim \frac{\gamma(\ln t)^2}{\tau^{1+(\frac{\gamma}{3})(\ln t)^2}} \sim constant \cdot t^{-(1+\alpha)}$

This form is equivalent to that proposed by Scher and Montroll. Therefore, the Scher and Montroll model is physically justified.

The approach of Scher and Montroll [2] is two pronged. First, it attacks the problem through CTRW formalism that leads to the broad distribution of event times, and second, an absorbing boundary layer is introduced to describe the anomalous transport seen in TOF experiments. The basic quantity of their theory is the propagator $\tilde{G}(l, t)$, the probability that a walker is found at l at time t if at time t = 0 it was at the origin. The propagation of the carrier is completely specified by this function (in the absence of the absorbing boundary). It is defined by,

$$\tilde{G}(l,t) = \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} \frac{du}{u} e^{ut} [1 - \psi^*(u)] G(l,\psi^*(u))$$
(16).

9

This function is determined by the Laplace transform of the hopping-time distribution function $\psi(t)$, and G(l, z), a random-walk generating function. The random-walk generating function is determined by p(l), which is the probability of a hop from a given cell to one displaced by a vector l away from it, and is normalized to 1. Therefore the hopping process is characterized by only two functions, p(l) and $\psi(t)$. All cells are treated as equivalent so that both are universal functions for all cells. The spatial hopping probability is simply handled with

$$p(0, \pm 1, 0) = p(0, 0, \pm 1) = q \tag{17}$$

$$p(1,0,0) = 2\tilde{\eta}p, p(-1,0,0) = 2(1-\tilde{\eta})p$$
(18),

with all other probabilities vanishing. This corresponds to an electric field in the l_1 direction (x-direction). This is for the situation of an electric field in the x-direction and $\tilde{\eta}$ is then a measure of field strength $\tilde{\eta} = \tilde{\eta}(E)$ and must be greater than $\frac{1}{2}$.

Two starkly different hopping-time distribution functions are studied, that of a single event time τ and that of a broad distribution of event times given by

$$\psi(t) \sim e^{-\tau} \tag{19}$$

$$\psi(t) \sim t^{-(1+\alpha)} \tag{20}$$

respectively.

To understand the anomaly at hand, let us first consider the case of "normal" (Gaussian) transport with a single intersite transition rate τ in a TOF experiment as in Eqn. (19). This can be seen in Fig. 9 (a), (b), and (d). In this case, the transit time is clearly defined as the Gaussian charge packet is absorbed at the boundary. This is not the case with dispersive transport, refer to Fig. 9 (c) and (e). For the single intersite transition rate (Gaussian charge packet), $\sigma/\langle l \rangle = \tau^{-1/2}$, but for a broad distribution of event times as in Eqn. (20) then $\sigma/\langle l \rangle = constant$.

To get from the description of a propagator in an infinite medium undergoing a CTRW, to TOF experiment, an absorbing boundary must be introduced. This is done by taking the original propagator, and subtracting from it an integral of the propagator multiplied by a first time of arrival function, so that any carriers that have reached the absorbing boundary are no longer taken into account. In then turns out that for the distribution of Eqn. (20) this leads to the equation,

$$I(t) = \begin{cases} \frac{\bar{l}}{c\Gamma(\alpha)\tau^{1-\alpha}} & \tau \ll \tau_{transit} \\ \frac{1}{2}\bar{l}\left(\frac{N-l_0}{\bar{l}}\right)^2 \frac{c}{-\Gamma(-\alpha)\tau^{1+\alpha}} & \tau \gg \tau_{transit} \end{cases}$$
(21).

Where \bar{l} is the mean displacement for a single hop, τ is a unitless time scaled by the reciprocal of a transition rate, l_0 is the initial position of the walker, N is the number of cells in the model, and c is an undetermined constant. Notice that the α and t dependence are exactly the same as derived by Cole and Cole for DC conductivity [4, 15], as shown in Appendix A.

The importance of these dependencies comes from the universality of their applicability. There are universal double logarithmic I(t) plots if the time and current are scaled by the reciprocal of the time and current at the transit time, where here the transit time is defined as the transition point between $t^{-(1-\alpha)}$ to $t^{-(1+\alpha)}$. This is found in the double logarithmic I(t) plot by finding the intersection of the two straight lines with slopes $-(1-\alpha)$ and $-(1+\alpha)$ (refer to Fig. 10 (a)). This universality applies to both transient TOF experiments and DC conductivity phenomena as outlined in the prior subsection. The time scales can be nanoseconds to decades and the universality still applies. Refer to Fig. 10. Note that there are other interesting dependencies such as $t_{transit} \sim (E/L)^{1/\alpha}$, refer to Fig 10 (b) [2, 9].

Other transient phenomena that can be studied using the idea of broad distributions of event times and the dispersion parameter include surface voltage potential measurements [7] and photocurrent decay measurements. The latter of which can be modeled using stretched exponential functions [18-20, 62, 63],

$$I_{ph}(t) = I_{ph}(0)e^{-\left(\frac{t}{\tau}\right)^{\beta} + constant}$$
(22)

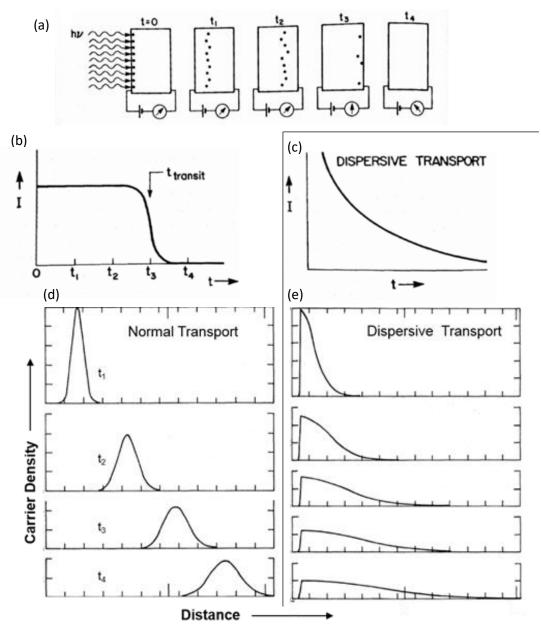
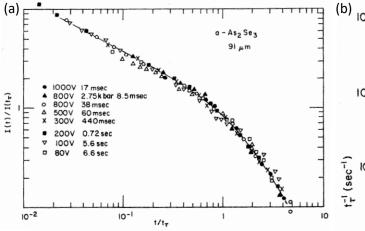


Figure 9. (a) Depicts a TOF experiment with normal transport. Current versus time are plotted for TOF experiments with (b) normal and (c) dispersive transport. (d) Charge packet propagation for TOF experiments are shown for normal transport. (e) Charge packet propagation for TOF experiments shown for dispersive transport.

where β is the decay exponent ($0 < \beta < 1$) and related to α by $\alpha = 1 - \beta$. I_{ph} is the photocurrent, t is the measurement time, and τ is the characteristic time constant of the process (which is likely some sort of average). Using DataThief software [64], photocurrent data of LDPE was taken from Wintle 1977 [3] and fit with a stretched exponential to compare the α value to that obtained by MPG [5, 8, 14]. The results are presented in Appendix C. More information on surface voltage potential measurements modeling can be found here [7].

Radiation induced conductivity (RIC) can also be explained with the use of the dispersion parameter [37]. It turns out that RIC can be explained by the simple relation,

$$\sigma_{RIC} = k_{RIC}(T)D^{\Delta} \tag{23}$$



 $10^{2} = a - As_{2} Se_{3}$ $10^{2} = 34 \mu m$ $10^{2} = 72 \mu m$ $91 \mu m$ $10^{0} = 5LOPE = 2.2$ $10^{0} = 6$ $10^{0} = 6$ $10^{0} = 6$ $10^{0} = 6$ $10^{0} = 6$ $10^{0} = 6$ $10^{0} = 6$ $10^{0} = 6$ $10^{0} = 6$ $10^{0} = 6$ $10^{0} = 6$ $10^{0} = 6$

Figure 10. Data is shown for As₂Se₃ from [2]. (a) double logarithmic I(t) data scaled by the transit time and current at transit time is plotted. The curves are universal for a given material. Transit times range from 17 ms to 6.6 s. (b) (E/L) versus transit time in double logarithmic plot. The slope is the reciprocal of the dispersion parameter, giving a value of $\alpha = 0.45$.

where k_{RIC} is a temperature dependent coefficient and \dot{D} is the dose rate [14, 39]. It can also be seen from [39, 41] and the definition $\alpha(T) = T_c/T$ that,

$$\Delta = \frac{T_c}{T_c + T} = \frac{1}{1 + \frac{T}{T_c}} = \frac{1}{1 + \alpha}$$

where T_c is the characteristic energy at which localized states are thought to be "frozen in" [14, 38, 39]. Tyutnev states this relation to the dispersion parameter explicitly [37, 41, 42]. This shows that RIC is dependent on the dispersion parameter as well. To further prove the point of equivalence, Fowler and Rose state that the relation $0.5 < \Delta < 1$ must hold [38, 39]. It is already known that $0 < \alpha < 1$ must be true, and in fact leads to the stated restrictions on Δ . The qualitative understanding also tracks for both parameters. In other words, as $\Delta \rightarrow 1$ then $\alpha \rightarrow 0$, and as as $\Delta \rightarrow 0.5$ then $\alpha \rightarrow 1$. Rose and Fowler describe this as infinitely broad (constant) or sharply exponential energetic density of states for $\Delta = 1$ and 0.5 respectively. The relation should not be surprising as RIC and TOF experiments are quite similar.

3.3 Other Approaches

There are many different ways to approach the problem of dispersive transport in disordered solids and no lack of attempted solutions. Other methods that have been shown to be equivalent to that of Scher and Montroll [2] include MT, hopping, and percolation theories [28], which have a nice overview in Pfister's paper [9]. An equivocation of MT, hopping and CTRW is given by Schmidlin [13]. Other approaches include fractional dynamic equations [65, 66] and effective medium approach (EMA) [57, 67-69]. Fractional dynamic equations are interesting as they apply to everything from geology and astronomy to dispersive transport in disordered materials. In this framework, dispersive transport in disordered materials is categorized as a subdiffusive process with $\alpha < 1$. What is quite interesting is this same parameter α is sprinkled into numerous theories about subdiffusive/superdiffuse processes.

4. Physical Significance of α

4.1 Introduction - Word of Warning

For dispersive transport to occur there must be transition rates/event times on the order of the experiment time. For example, there must be a transition rate that is on the order of the transit time in a TOF experiment. This means that even for relatively few trap states, dispersive transport can occur if the transition rate of one of the traps is on the order of the transit time and is shown for as few as 7 discrete trap energies in [13]. Clearly, a broad distribution of states can accomplish this as well. Any small deviation can cause this to occur, as Pfister [9] puts it:

"However complicated the form of the transition rates and the details of the molecular charge transfer, it is assumed that these rates depend sensitively on a number of parameters that are statistically distributed. Thus, even rather mild variations of some system parameters 'map' onto a broad distribution of transition rates. This mapping is not unique. A number of different parameter dispersions can produce very similar transition rate distributions."

This means that even relatively small deviations from crystalline structure can cause large effects resulting in dispersive transport. Since the underlying mechanisms for

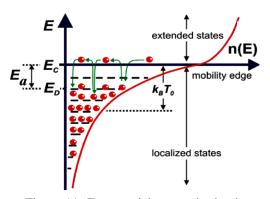


Figure 11. Exponential energetic density of states in the mobility gap with indications for MT and characteristic width kT_0 . From [6].

transport are more or less unimportant for the modeling of dispersive transport, it is difficult to extract significant physical meaning from the dispersion parameter α that is used simply to model the data. Many papers guess at the underlying transport mechanisms and some back it up with theory, but while they may not necessarily be wrong, it is also quite hard to determine if they are right without a deep understanding and control of microscopic properties of the material. This is why many tests of the models are done on impurity conduction in semiconductors, as there is a high degree of control and understanding of the sample [9, 16]. The key takeaway is that macroscopic effects measured can come from a multitude of microscopic mechanisms.

4.2 Equations of α – Exponential Density of States

Not all hope is lost, some insight into the energetic density of states can be gained through the understanding of the dependencies of the dispersion parameter on the temperature and electric field. Therefore, some light can be shed on material properties and transport. Many papers have shown that for an exponential energetic density of states (refer to Fig. 11) the temperature [6, 9, 22-25, 27, 70] and electric field dependence [23-25] are of the form,

$$\alpha(T) = \frac{kT}{E_c} = \frac{T}{T_c}$$
(24)

$$\alpha(E) = \frac{qaE}{2kT_c} \tag{25}$$

where E_c is a characteristic energy and T_c is the temperature corresponding to that energy (Note that there are theoretical descriptions of the characteristic energy such as the one presented by Arkhipov [22], but many simply state it to be the width of the exponential energetic density of states [25]), q is the elementary charge, a is the localization length, E is the electric field strength, and T is the temperature. When the temperature is low and the field is high, Eqn. (25) is applicable and when the temperature is high and the field is low Eqn. (24) is applicable. It gets more complicated in the transition region between the two regimes. Attempts have been made to introduce an effective temperature as the E-field has a temperature like effect [23-25]. An effective temperature equation can be made for materials but does not seem to have the universal dependence of the pure T or E dependent dispersion parameter equations.

It is useful to introduce the demarcation energy (DE) and quasi-fermi level (QFL). This is explained in great detail in Ch. 3 of Sim's dissertation [7], as well as in these references [38, 43, 71]. Essentially, the DE is the centroid of the trapped charge within the mobility gap. The QFL is an effective Fermi level, corresponding to the DE. In wide band gap materials such as highly disordered insulating materials, the Fermi level does not deviate significantly from the Fermi energy. Deviations are instead caused by external charge injection, RIC, or similar processes that modify the carrier distribution of the material. This is where the use of the QFL comes in handy.

In Fig. 12, the thermalization of a charge distribution can be seen. For example, imagine that a charge layer has been injected into shallow traps. The electrons will thermalize and fall into deeper trap states, moving from the red curve progressively to the blue curve. During thermalization transient dispersive transport occurs. Once thermalization

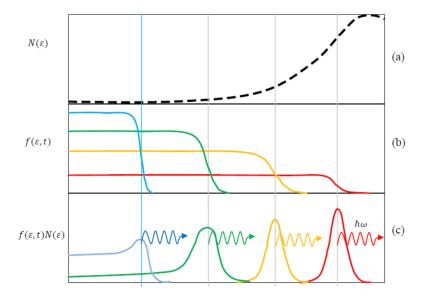


Figure 12. Depiction of the thermalization of trapped charge carriers. (a) Exponential density of states as a function of energy. (b) Mean occupation of states as a function of time and energy. (c) Product of the density of states and the mean occupation, which results in a description of the distribution of occupied states near the demarcation energy during thermalization. Time flows from red (earliest time) to blue (latest time). From [7].

is complete, the QFL is at the equilibrium Fermi level and the transient dispersive phenomena is no longer observed. This baseline dark current conductivity is then described by equations based on VRH or MT [7, 72-74].

Consider Fig. 5 (b). The VRH mechanisms looks much like MT but entirely within localized states rather than excitation into the conduction band. The shaded area where TAH like conduction occurs is sometimes called the transport energy [7]. It should be noted the transport energy has a finite width and can be called the transport band. With added energy via increased temperature or electric field this transport band moves towards the conduction band. Once the two overlap, this defines the transition from VRH to MT [73, 74]. This also describes the transition from dispersive to normal transport [7]. This transition can be predicted as a function of T and E by setting $\alpha = 1$ in Eqns. (24) and (25).

This T and E dependence naturally leads one to the conclusion that the dispersion parameter is related to the occupancy of the density of states. If the density of states is an exponential function, then the dispersion parameter equations define the characteristic energy as the width (1/e drop) of the energetic density of states in the mobility gap, refer to Fig. 11. This also means that at the characteristic energy, where transition rates are increased enough due to the increased occupation of shallow trap states (VRH is negligible), a transition from dispersive to normal transport occurs [21, 23-25]. This makes sense as the temperature, or electric field equivalent, is equal to the characteristic energy, then $\alpha = 1$ and dispersive transport no longer occurs.

During the transient dispersive transport before reaching steady state conductivity, as shown in Fig. 8 (b), thermalization is not complete. The 'kink' in these plots indicates the transition from MT (extended state transport in conduction band, punctuated by shallow trapping) to VRH as depicted by Fig. 5 (b). This transition occurs as the DE moves below the transport energy. Note that another explanation is that this is when the carriers first reach the other side of the material, for example the transit time in TOF experiments [2]. In general the transport energy is a complex function of many parameters such as temperature, electric field, and density of states [7].

If after thermalization occurs the DE is above the transport energy, then normal (packet-like) transport occurs and VRH is negligible. The steady-state conductivity is then described by a T^{-1} dependence indicative of MT. If the DE is less than the transport energy after thermalization then the conductivity has a $T^{-1/4}$ dependence indicative of VRH [7, 72-74]. The time needed to reach steady-state conductivity is denoted the segregation time and this time uniquely defines the conductivity. That is, if the segregation time is known then the conductivity is known and vice versa [7].

It turns out that the transitions from dispersive to normal transport at $\alpha = 1$, corresponding to T_{trans} and E_{trans} have physical significance for the material. The temperature T_{trans} is potentially a glassy transition temperature in some materials. The electric field E_{trans} defines the onset of packet-like charge behavior, or in other words normal transport.

This transition has been validated experimentally in low-density polyethylene in conductivity measurements [72, 73], and direct measurements of charge distributions via pulsed electroacoustic method under DC stress with electric field strength of ~10⁸ V/m [75]. This electric field strength also indicates the onset of breakdown in materials, as validated by theory and experiment [45, 75]. The onset of breakdown presented by Andersen [45] bears a striking resemblance to Eqn. (25) with $\alpha = 1$. The temperature found experimentally for the VRH to MT transition in low-density polyethylene [72, 73] corresponds well with a β -phase transition [76, 77] that can be considered a glassy transition temperature of sorts since such a temperature cannot actually exist in this material due to sections of amorphous and crystalline lamellae (no true amorphous form) [77].

4.3 Gaussian Density of States

It turns out that the modeling of some materials is more complex. There are often materials where the dispersion parameter is different before and after the transit time, and the log dependence of the ratio of the thickness to the electric field is superlinear rather than linearly proportional to α (refer to Fig. 10 (b) for linear case) [9, 28]. This is indicative that the energetic density of states in the mobility gap is not that of an exponential but of a different form. Specifically in materials with an exponential trap distribution, the dispersion parameter is useful and gives us insight as outlined above. If this energetic density of states is instead a Gaussian, as is assumed in the case of hopping transport in organic glasses and molecularly doped polymers, then the dispersion parameter offers at best a zero-order approximation and does not offer much insight [57]. A different approach is needed in this case.

In the case of a Gaussian energetic density of states, through a combination of analytic functions and Monte Carlo simulations the effective medium approach (EMA) can be employed [57, 67, 69]. EMA has been shown to provide an excellent description of hopping processes in dense systems [68]. This theory will not be covered in detail here as it does not help us gain insight into the physical significance of α . It should be noted that the dispersion parameter does appear in other formalisms with Gaussian or other arbitrary density of states but in a less simple way [7].

5. Other Concerns

Concerns that should be noted but that are not directly addressed in this paper include effects from electrode injection and ohmic/non-ohmic contacts directly related to the electrode material and interface effects/surface defects [9, 78]. The Poole-Frenkel effect and space charge limited current should be noted as well [7, 12]. Another concept not discussed is activation energy [2, 9, 22, 78], likely closely related to the demarcation energy discussed previously. There is occasionally some confusion in the literature related to values of the dispersion parameter. Typically it is expected that $0 < \alpha < 1$, but in some cases there have been reports of values outside this range which are likely due to a naming convention confusion [63, 79, 80]. It also appears that the Cole and Cole theory, which seems to imply dispersive transport with a circular arc rather than a semi-circle with Argand diagram, applies to crystalline materials [4, 15]. In other words, crystalline materials can produce circular arcs in Argand diagrams as well. It is likely that these crystalline materials have defects that would cause this sort of behavior.

I would also like to note that there seems to be connections of the dispersion parameter to other electronic properties of materials including secondary electron yield and photoluminescence. This is not surprising as the dispersion parameter plays in important role in describing the occupied energetic density of states and the density of states is paramount to the mentioned processes.

6. Conclusions

There has clearly been an expansive amount of work done to attempt to gain an understanding of anomalous properties of disordered materials. In this context, α has proven to be a useful parameter on many occasions. In the case of exponential density of states α tells us a characteristic energy of which a dispersive to normal transport transition occurs. It tells us qualitatively how dispersive a material is, with a more dispersive material having an α value closer to zero.

The key importance of the dispersion parameter is the insight it gives to the energetic density of states in the mobility gap that help provide a description of the charge transport in disordered materials. From a textbook published just two years ago, "The absence of reliable information on the energy spectrum and on the structures of the wavefunctions in the vicinity and below the mobility edges can be considered to be the main problem for researchers attempting to quantitatively describe the charge transport properties of disordered material" [81]. In other words, the energetic density of states in the mobility gap is considered to be of utmost importance for electrical properties of disordered materials.

The dispersion parameter aids in the qualitative and quantitative description of electrical properties in noncrystalline materials. Universal plots of dispersive transport in TOF and CVC data are explained via the dispersion parameter, as well as the temperatures and electric fields necessary to transition to normal transport. These correspond to other physically important characteristics such as a glassy transition temperature or the onset of electrostatic breakdown. The dispersion parameter can shed light on AC and DC conductivity, RIC, photoconductivity, charge decay, electrostatic breakdown, and pulsed electroacoustic experiments. It appears the dispersion parameter is a dream tool for an engineer when applied correctly as it explains the response of a disordered material macroscopically (practically), but leaves a physicist wanting more since it fails to shine direct light on the underlying microscopic processes. This is due to the fact there are a multitude of underlying factors that can lead to dispersive transport.

7. Appendix

A. DC and Transient Equivocation

In TOF experiments it has been shown by CTRW theory and validated by experiment that the t and α dependencies are that of Eqn. (21). Shown again below for convenience with coefficients omitted, aside from those with α dependence.

$$I(t) \propto \begin{cases} \frac{1}{\Gamma(\alpha)t^{1-\alpha}} & t \ll t_{transit} \\ \frac{1}{-\Gamma(-\alpha)t^{1+\alpha}} & \tau \gg t_{transit} \end{cases}$$
(A1)

It has also been presented that the dependencies of DC measurements are shown to have the dependencies as in Eqn (7),

$$I(t) = \begin{cases} \frac{\varepsilon_0 - \varepsilon_{\infty}}{\tau_0} \frac{1}{\Gamma(1 - \alpha')} \left(\frac{t}{\tau_0}\right)^{-\alpha'} & t \ll t_{transit} \\ \frac{\varepsilon_0 - \varepsilon_{\infty}}{\tau_0} \frac{(1 - \alpha')}{\Gamma(\alpha')} \left(\frac{t}{\tau_0}\right)^{-(2 - \alpha')} & t \gg t_{transit} \end{cases}$$
(A2)

Eqn. (A1) will be shown to be of an equivalent form to Eqn. (A2), but will be done so with all constants carried through. This is because there are no free parameters without physical meaning (aside from α) in Eqn. (A2), and therefore is a more complete description. For Eqn. (A1) there are undetermined constants (without physical meaning) and it is not useful to carry them through.

Two things must be addressed here to show the equivalency. First, it must be assumed that $\alpha = 1 - \alpha'$, which is not hard to do as Scher and Montroll [2] and Cole and Cole [4, 15] describe the same physical dependencies of their respective dispersion parameters. That is to say, for $\alpha = 1$ and $\alpha' = 0$ the materials are no longer dispersive and are progressively more dispersive as the values approach $\alpha = 0$ and $\alpha' = 1$. The second thing is to equate the gamma function coefficients. This is trivial in the pre-transit time dependencies as it is already met after the first substitution for $\alpha = 1 - \alpha'$. For the post-transit dependency,

$$\frac{(1-\alpha')}{\Gamma(\alpha')} = \frac{(\alpha'-1)(-1)}{(\alpha'-1)(\alpha'-2)(\alpha'-3)\cdots} = \frac{(-1)}{(\alpha'-2)(\alpha'-3)\cdots} = \frac{-1}{\Gamma(\alpha'-1)} = \frac{-1}{\Gamma(\alpha)}$$

showing that the coefficients are equivalent.

The exponential time dependence is trivial and for the DC case,

$$I(t) = \begin{cases} \frac{\varepsilon_0 - \varepsilon_\infty}{\tau_0} \frac{1}{\Gamma(\alpha)} \left(\frac{t}{\tau_0}\right)^{-(1-\alpha)} & t \ll \tau_0 \\ \frac{\varepsilon_0 - \varepsilon_\infty}{\tau_0} \frac{(-1)}{\Gamma(-\alpha)} \left(\frac{t}{\tau_0}\right)^{-(1+\alpha)} & t \gg \tau_0 \end{cases}$$
(A3)

This can clearly be recognized as the same t and α dependence of the TOF experiment case, Eqn. (A1). This is one explanation for the same transit time/kink as seen in the double logarithmic I(t) plots for TOF and CVC experiments.

B. Cole-Cole Equation and the Fuoss-Kirkwood Approximation

To show the validity of the form of the ratio of the imaginary permittivity over the max value $\varepsilon''/\varepsilon_m''$ put forward by Fuoss and Kirkwood [55], refer to Eqn. (9), starting from the equation for the complex permittivity ε^* given by Cole and Cole [4, 15], refer to Eqn. (5). There are three steps, starting with Eqn. (5); first, find the equation for ε'' ; second, find ε_m'' ; third, show the form of $\varepsilon''/\varepsilon_m''$ derived with limits of $\alpha \to 0$ and $\alpha \to 1$.

For Debye theory it is easy to show,

$$\frac{\varepsilon''}{\varepsilon_m''} = \operatorname{sech}(x) \tag{B1}$$

where $x = \ln(\omega_m/\omega)$. The frequency ω_m is when ε_m '' occurs. This is the motivation for the form of Eqn. (9).

Starting with Eqn. (5), the imaginary number *i* is replaced with its exponential form $\exp(i\pi/2)$. Then with use of the Euler identity,

$$\varepsilon^{*} - \varepsilon_{\infty} = \frac{\varepsilon_{0} - \varepsilon_{\infty}}{1 + e^{\frac{i\alpha\pi}{2}}(\omega\tau_{0})^{\alpha}} = \frac{(\varepsilon_{0} - \varepsilon_{\infty})\left[1 + \cos\left(\frac{\alpha\pi}{2}\right)(\omega\tau_{0})^{\alpha}\right]}{\left[1 + \cos\left(\frac{\alpha\pi}{2}\right)(\omega\tau_{0})^{\alpha}\right]^{2} + \sin^{2}\left(\frac{\alpha\pi}{2}\right)(\omega\tau_{0})^{2\alpha}}$$

Now using $\varepsilon^* = \varepsilon' - i \varepsilon''$ and solving for ε'' (the imaginary part of the equation),

$$\varepsilon'' = \frac{(\varepsilon_0 - \varepsilon_\infty) \sin\left(\frac{\alpha \pi}{2}\right) (\omega \tau_0)^{\alpha}}{\left[1 + \cos\left(\frac{\alpha \pi}{2}\right) (\omega \tau_0)^{\alpha}\right]^2 + \sin^2\left(\frac{\alpha \pi}{2}\right) (\omega \tau_0)^{2\alpha}}$$
(B2)

To find the max value ϵ_m ", one can simply take the derivative of ϵ " with respect to frequency and set equal to zero.

$$\frac{d(\varepsilon'')}{d(\omega\tau_0)} = \frac{\alpha(\varepsilon_0 - \varepsilon_\infty)\sin\left(\frac{\alpha\pi}{2}\right)(\omega\tau_0)^{\alpha-1}[1 - (\omega\tau_0)^{2\alpha}]}{\left[(\omega\tau_0)^{2\alpha} + 2\cos\left(\frac{\alpha\pi}{2}\right)(\omega\tau_0)^{\alpha}\right]^2} = 0$$

Evaluating at $\omega = \omega_m$ and solving for ω_m ,

$$(\omega_m \tau_0)^{\alpha - 1} [1 - (\omega_m \tau_0)^{2\alpha}] = 0$$

The zeroes of $(\omega \tau_0)$ are 0 and 1 The solution $\omega_m \tau = 0$ is trivial. Therefore, the solution below is used.

$$\omega_m \tau_0 = 1 \qquad \rightarrow \qquad \omega_m = \frac{1}{\tau_0}$$

This makes sense as this essentially says the characteristic frequency is the reciprocal of the characteristic relaxation time. To get the value ε_m ", simply plug in ω_m into the equation for ε ". So that,

$$\frac{\varepsilon''}{\varepsilon_m''} = \left(\frac{\omega}{\omega_m}\right)^{\alpha} \frac{\left[1 + \cos\left(\frac{\alpha\pi}{2}\right)(\omega_m\tau_0)^{\alpha}\right]^2 + \sin^2\left(\frac{\alpha\pi}{2}\right)(\omega_m\tau_0)^{2\alpha}}{\left[1 + \cos\left(\frac{\alpha\pi}{2}\right)(\omega\tau_0)^{\alpha}\right]^2 + \sin^2\left(\frac{\alpha\pi}{2}\right)(\omega\tau_0)^{2\alpha}}$$

Recall that $\omega_m \tau_0 = 1$. This means,

$$\frac{\varepsilon''}{\varepsilon_m''} = \left(\frac{\omega}{\omega_m}\right)^{\alpha} \frac{2\left[1 + \cos\left(\frac{\alpha\pi}{2}\right)\right]}{\left[1 + \cos\left(\frac{\alpha\pi}{2}\right)(\omega\tau_0)^{\alpha}\right]^2 + \sin^2\left(\frac{\alpha\pi}{2}\right)(\omega\tau_0)^{2\alpha}}$$

If the substitution $e^x = \omega_m / \omega$ is made (or since $\omega_m = 1/\tau_0$, $\omega \tau_0 = exp(-x)$),

$$\frac{\varepsilon''}{\varepsilon_m''} = e^{-\alpha x} \frac{2\left[1 + \cos\left(\frac{\alpha \pi}{2}\right)\right]}{\left[1 + \cos\left(\frac{\alpha \pi}{2}\right)e^{-\alpha}\right]^2 + \sin^2\left(\frac{\alpha \pi}{2}\right)e^{-2\alpha}} = \frac{2\left[1 + \cos\left(\frac{\alpha \pi}{2}\right)\right]}{e^{\alpha x} + 2\cos\left(\frac{\alpha \pi}{2}\right) + e^{-2\alpha x}}$$

To get to a more appealing form of this equation, it is useful to manipulate the reciprocal,

$$\frac{\varepsilon_m''}{\varepsilon''} = \frac{e^{\alpha x} + 2\cos\left(\frac{\alpha \pi}{2}\right) + e^{-2\alpha x}}{2\left[1 + \cos\left(\frac{\alpha \pi}{2}\right)\right]} = \frac{\cos\left(\frac{\alpha \pi}{2}\right)}{1 + \cos\left(\frac{\alpha \pi}{2}\right)} + \frac{\cosh(\alpha x)}{1 + \cos\left(\frac{\alpha \pi}{2}\right)} = \frac{\cos\left(\frac{\alpha \pi}{2}\right) + \cosh(\alpha x)}{1 + \cos\left(\frac{\alpha \pi}{2}\right)}$$

Then taking the reciprocal again,

$$\frac{\varepsilon''}{\varepsilon_m''} = \frac{1 + \cos\left(\frac{\alpha\pi}{2}\right)}{\cos\left(\frac{\alpha\pi}{2}\right) + \cosh(\alpha x)}$$
(B3)

The limits of $\alpha \approx 1$ and $\alpha \approx 0$ are respectively,

$$\frac{\varepsilon''}{\varepsilon_m''} \approx \operatorname{sech}(\alpha x) \quad \text{and} \quad \frac{\varepsilon''}{\varepsilon_m''} \approx \frac{2}{1 + \cosh(\alpha x)}$$
(B4)

This shows that the approximation put forward by Fuoss and Kirkwood, Eqn. (9), is valid around $\alpha \approx 1$. In other words, this model is only good for materials only slightly dispersive/disordered. Note that for the extreme when $\alpha = 1$, the familiar Debye form is recovered. Another check, when $\omega = \omega_m$ (x = 0) each equation should reduce to $\varepsilon'' = \varepsilon_m''$. Which is easily seen to be the case since sech(0) = cosh(0) = 1.

Interestingly, if you stare at Eqn. (B4) long enough, the two limits resemble the Boltzmann and Fermi-Dirac distribution functions respectively. In a bad argument, when $\alpha \approx 0$, $\cosh(\alpha x) \approx 1 \approx e^{\alpha x}$. Giving the form of the Fermi-Dirac distribution function. Then by a similar argument, near x = 0, for $\alpha \approx 1$, then $\operatorname{sech}(\alpha x) = 1/\cosh(\alpha x) = e^{-\alpha x}$. This is the form of the Boltzmann distribution. It is likely just an interesting coincidence, but these limits approximate regimes where quantum effects are unimportant ($\alpha \approx 1$, normal transport) and when they are very important ($\alpha \approx 0$, highly dispersive transport such as VRH). These correspond to the Fermi-Dirac and Boltzmann distributions respectively. Sim notes that there is a Boltzmann-like and Fermi-Dirac like distribution of mean occupation for electrons in a highly disordered material for MT and VRH regimes respectively [7].

C. Photocurrent Decay Modeling with Stretched Exponential

Data of an infrared charge/decay experiment on low density polyethylene (LDPE) [3] was extracted using DataThief [64] and fit with a stretched exponential function, refer to Eqn. (22). The results are shown in Fig. C1. The best fit gives a dispersion parameter of about 0.15. This does not correspond well to the data found by Wood and King for a value of 0.5 [5, 8]. However, it is clear that this parameter depends on field and temperature and it is unclear what temperature and field the sample was exposed to in Wintle's data [3]. It seems the data from Wintle was likely taken at a temperature lower than room temperature. This may agree better with data from Gillespie [14], but there are large experimental uncertainties and in some cases unphysical values of α . Note that Tyutnev [41] is in agreement with King and Wood data [5, 8].

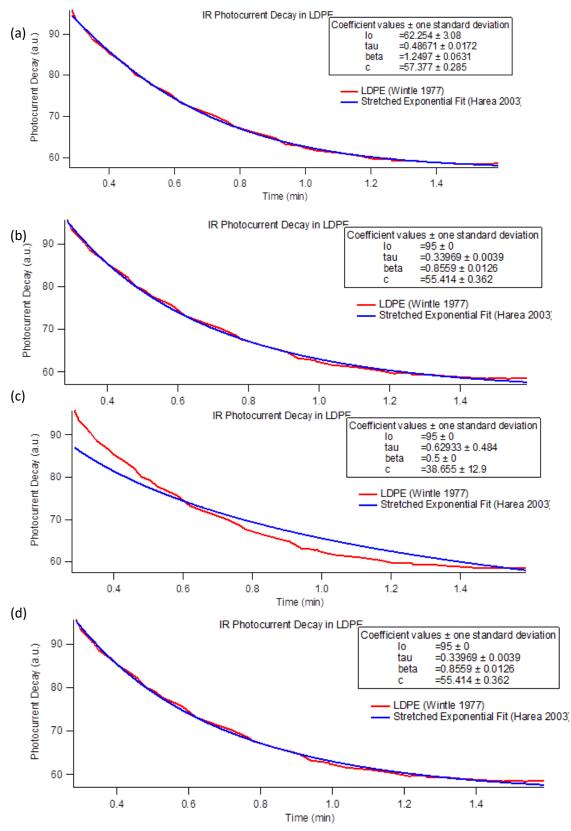


Figure C1. Stretched exponential fit to LDPE data from [3]. Two fits are shown, (a) is with no constraints on the parameters, (b) is with $0 < \beta < 1$ and $I_0 = 95$, (c) is with β held at 0.5 and I_0 held at 95, and (d) is with I_0 held at 95 respectively. Note that the same results for (d) are obtained if β is constrained between 0 and 1.

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