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# Relation between charge-dipole interactions and the $\sqrt{E}$-dependent mobility in molecularly-doped polymers

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## ABSTRACT

Time-of-flight measurements on a wide variety of molecularly-doped polymers reveal carrier mobilities that exhibit an exponential dependence on the square root of the applied electric field. Recent attempts to explain the observed field dependence have focused on the role played by spatial and energetic disorder. It has also been conjectured that the charge-dipole interactions often identified as the source of energetic disorder could be of sufficient range to lead to correlations in the energies of neighboring hopping sites. We have analytically explored the effect of such correlations on high field carrier transport in random potentials, and discuss how particular features of the correlations associated with charge-dipole interactions might lead to behavior similar to that seen in experiment.

## 1    Introduction

High-field time-of-flight experiments have been used for over two decades to characterize hopping mobilities of photoexcited charge carriers in molecularly-doped polymers and amorphous molecular glasses.[1-3] Numerous measurements over a large range of fields and temperatures have established several nearly universal features that appear to characterize these materials. In particular, the field-dependent mobility $\mu$ of these systems is typically found to exhibit a thermally-activated behavior with a strong exponential dependence[1-4]

$$\mu = \mu_0 \exp\left[-\frac{\Theta}{kT}\right] \exp\left[\gamma\sqrt{E}\right],  \tag{1}$$

on the square root of the electric field $E$. In (1), $T$ is the absolute temperature, $k$ is Boltzmann's constant, and $\mu_0$ is a prefactor that tends to reveal an exponential dependence

$$\mu_0 = a_0 \rho^2 e^{-2\rho/\rho_0}  \tag{2}$$

upon the mean interdopant distance $\rho = n^{-1/3}$. In a particular form of this phenomenological expression proposed by Gill,[5] the activation energy $\Theta$ is temperature independent, and the "Poole-Frenkel" factor is written

$$\gamma = B(\beta - \beta_0),  \tag{3}$$

where $\beta = 1/kT$, and $B$ and $\beta_0 = 1/kT_0$ are constants.

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Many recent theoretical attempts to explain this observed field dependence have explored the role played by spatial and energetic disorder.[2,6–8] Numerical simulations by Bässler and coworkers have focused on a Gaussian disorder model (GDM), which seeks to describe the transport as a biased random walk among dopant molecules having independent Gaussian-distributed random site energies.[2] One result of extensive analysis and characterization of numerical work on the GDM, has been the introduction and widespread use of an empirical relation for the mobility, formally identical to (1) except for a quadratic temperature dependence, i.e., $\Theta/kT = (2\sigma/3kT)^2$, and a slightly different parameterization of the Poole-Frenkel factor $\gamma = C(\beta^2\sigma^2 - \Sigma^2)$, where $\sigma$ is the width of the energetic disorder, $\Sigma$ is a constant characterizing the off-diagonal (spatial and orientational) disorder, and $C$ is a universal constant. Simulations based upon the GDM satisfactorily explain many features of experiment, such as the time-of-flight transients. Unfortunately, mobilities deduced from simulation on the GDM[2,6] and other numerical work which treats the spatial disorder in a somewhat different fashion[8] predict a field dependence similar to (1) only in a relatively narrow range at rather large fields ($E > 10^5$V/cm). By contrast, the experimentally observed linear dependence of $\log\mu$ on $\sqrt{E}$ has been clearly established[9] down to some of the lowest fields probed experimentally ($E \sim 8 \times 10^3$ V/cm).

Simple theoretical arguments would suggest that, since the electric field enters the hopping rates only through the combination $eEr/kT$, where $e$ is the electronic charge and $r$ the hopping distance, a strong field dependence should be expected only when the potential drop $F = eE\ell$ across a relevant length scale in the system is comparable to $kT$. Identifying $\ell$ with a typical *nearest-neighbor* distance leads, through this simple argument, to a field dependent mobility only at the relatively higher fields where it is, in fact, observed in simulation. The *observation* of a field-dependent mobility at lower fields, on the other hand, seems to provide evidence for a larger relevant length scale in the problem. Such a length scale, it turns out, arises naturally from one of the mechanisms often proposed as the source of the energetic disorder in these materials.

In particular, it has been recently shown that a Gaussian-like density of states of the type assumed in the GDM can arise from the interaction of charge carriers with a random distribution of permanent electric dipoles, having magnitudes typical of those associated with the host and dopant molecules from which these materials are typically formed.[10–12] In the most general form of what has been referred to as the "dipolar disorder model", these permanent dipole moments are associated either with dopant molecules, with repeat units of the host polymer, or with both. Considerable data and analysis establishing a relationship between carrier mobilities and group dipole moments of molecular constituents has lent support to this view of charge-dipole interactions as the source of energetic disorder in these systems.[10–17]

Although many recent investigations of the dipolar disorder model have focused on establishing its consistency with specific assumptions and parameters of the standard Gaussian disorder model, it was pointed out recently by Novikov and Vannikov[17] from simulations of the fluctuating potential experienced by a charge in the presence of randomly distributed dipoles, that the long-range nature of the charge-dipole interaction leads to positive algebraic correlations among the energies of neighboring hopping sites. This is in contrast to one of the basic assumptions of the GDM, in which site energies are treated as uncorrelated random variables. These ideas were carried an important step further by Gartstein and Conwell[18] who showed that finite-range correlations imposed upon the standard GDM push the regime over which the field dependence is described by (1) to lower fields, in better agreement with experiment.

In a recent paper,[19] Dunlap, Parris, and Kenkre presented a simple analytical theory describing the effects that algebraic correlations of the type that arise in the dipolar disorder model have on carrier transport. In particular, it was shown that the specific correlation that arises in the dipolar disorder model can lead, even at relatively low fields, to the specific $\sqrt{E}$ field dependence of the mobility observed in experiment. In this paper we review the arguments leading to that result.

## 2 Field-Dependent Mobility

We consider a simplified model describing a photoexcited carrier migrating among dopant molecules as it crosses a sample in the presence of an electric field $E$. We assume that the actual percolating path taken by the carrier is determined primarily by the fluctuation in interdopant distances, upon which the hopping rates are exponentially dependent. The resulting critical transport path will then be of reduced dimensionality relative to a typical three dimensional random walk, with the next site to which the particle hops being, typically, one of the two neighboring sites to which it is closest in real space. At this stage energetic considerations are less important, since the energies of neighboring sites are strongly correlated. In the model, this correlation arises because the energy of a carrier occupying any one of the molecular sites along this path will be a random variable arising from the sum of the many long-range interactions that the particle experiences due to a fixed concentration $n_0$ of randomly distributed point dipoles, each with dipole moment $\vec{p}$. Formally, we treat the concentration of dipoles as being distinct from the concentration of dopant molecules among which the carrier moves. This is done for two reasons: first, as discussed above, we envision situations in which there is a contribution due to the molecular constituents of the host polymer, and secondly we recognize that the typical density of dopant molecules forming the critical pathway may well differ from the actual dopant density itself. Thus, a carrier at the $n$th dopant molecule experiences a random energy $u_n$ of zero mean associated with an overall distribution of site energies of significant width $\sigma > kT$.

Focusing only on those dopant molecules associated with the low dimensional path along which the carrier drifts, we write a one-dimensional master equation

$$\frac{dP_n}{dt} = -(W_{n-1,n} + W_{n+1,n})P_n \; + \; W_{n,n-1}P_{n-1} + W_{n,n+1}P_{n+1}, \tag{4}$$

for the probability $P_n(t)$ of finding the carrier at the $n$th site along this path. The hopping rates which appear in this relation obey a detailed balance relation $W_{n\pm 1,n}/W_{n,n\pm 1} = \exp[-\frac{1}{2}\beta(u_{n\pm 1} - u_n \mp F)]$, where $F = eE\rho$ is the energy change induced between two sites by the field. (Because we are focusing on the effects associated with energetic correlations we ignore any fluctuations in the distances associated with neighboring sites.) Independent of how this detailed balance condition is implemented, the one-dimensional aspect of the transport equation that we have assumed allows the steady state drift velocity to be obtained exactly. In what follows we give a brief derivation of the drift velocity similar to that originally given by Derrida.[20] Seeking the bulk thermodynamic properties, we periodically repeat the sample (assumed to be of length $L = N\rho$) and consider the steady-state probability current flowing through the resulting infinite system in response to the driving field. In steady state, the net probability current $J$ flowing through each bond in the chain must be the same, i.e., the quantity

$$J = W_{n+1,n}P_n - W_{n,n+1}P_{n+1}. \tag{5}$$

must be independent of $n$. Solving this difference equation for

$$P_n = \frac{J}{W_{n+1,n}} + \frac{W_{n,n+1}}{W_{n+1,n}}P_{n+1} \tag{6}$$

and iterating $N$ times gives

$$P_n = J\left[\frac{1}{W_{n+1,n}} + \cdots + \frac{W_{n,n+1}\cdots W_{n+N-2,n+N-1}}{W_{n+1,n}\cdots W_{n+N-1,n+N-2}}\frac{1}{W_{n+N,n+N-1}}\right] + \prod_{j=1}^{N}\frac{W_{j,j+1}}{W_{j+1,j}}P_n, \tag{7}$$

where we have used a boundary condition $P_{n+N} = P_N$ valid for the periodically repeated system. Collecting terms in $P_n$ we obtain the relation

$$P_n\left[1 - \prod_{j=1}^{N}\frac{W_{j,j+1}}{W_{j+1,j}}\right] = \frac{J}{W_{n+1,n}}\left[1 + \sum_{i=1}^{N-1}\prod_{j=1}^{i}\left(\frac{W_{n+j-1,n+j}}{W_{n+j+1,n+j}}\right)\right]. \tag{8}$$

Summing over $n$ and invoking an arbitrary normalization $\sum_n P_n = 1$ appropriate to a carrier density of one independent particle per $N$ sites yields the current density $J = v/N$, where

$$v = \frac{\left[1 - \prod_{n=1}^{N} \frac{W_{n,n+1}}{W_{n+1,n}}\right]}{\frac{1}{N}\sum_{n=1}^{N}\frac{1}{W_{n+1,n}}\left[1 + \sum_{i=1}^{N-1}\prod_{j=1}^{i}\left(\frac{W_{n+j-1,n+j}}{W_{n+j+1,n+j}}\right)\right]} \tag{9}$$

is the associated drift velocity. This expression can be simplified if we assume *symmetric hopping rates* $W_{n\pm1,n} = \nu_0 \exp[-\frac{1}{2}\beta(u_{n\pm1} - u_n \mp F)]$, for which the last expression reduces, after some work, to[19]

$$v = \frac{\nu_0\rho e^{\frac{\beta F}{2}}\left[1 - e^{-\beta N F}\right]}{\frac{1}{N}\sum_{m=0}^{N-1}e^{-m\beta F}\sum_{n=1}^{N}e^{-\beta u_n}e^{\beta u_{n+m}}e^{\beta\delta_{n+m}}} \tag{10}$$

in which $2\delta_n = u_{n+1} - u_n$. The exponential $e^{-\beta N F}$ in the numerator is entirely negligible, with $\beta N F$ representing the ratio of the potential drop across the sample to the mean thermal energy. In the limit that the energy difference between neighboring sites is small compared to $kT$ (as will tend to occur when the energies of those sites are positively correlated) we ignore factors involving $\delta_{n+m}$, so that

$$v = \frac{\nu_0\rho e^{\frac{\beta F}{2}}}{\sum_{m=0}^{N-1}e^{-m\beta F}\langle e^{-\beta u_n}e^{\beta u_{n+m}}\rangle}, \tag{11}$$

where we have identified the sum

$$\langle e^{-\beta u_n}e^{\beta u_{n+m}}\rangle = \frac{1}{N}\sum_{n=1}^{N}e^{-\beta u_n}e^{\beta u_{n+m}} \tag{12}$$

over the macroscopic crystal ($N \to \infty$) with the ensemble average of the associated exponentials. Focusing on fields small enough that the field-induced potential drop across a nearest-neighbor distance is small, i.e., $\beta F < 1$, we approximate the exponential in the numerator of (11) by unity, and approximate the remaining sum in the denominator by an integral, to obtain an expression for the field dependent mobility[19]

$$\mu = v/E = \frac{\mu_0}{\epsilon\int_0^{\infty}dy\, e^{-\epsilon y}\langle e^{-\beta U(0)}e^{\beta U(y)}\rangle} \tag{13}$$

which involves the Laplace transform of the correlation function (12), with $U(y)$ representing the zero-field energy of a site at $y$, the Laplace variable $\epsilon = \beta e E$ representing the applied field, and the prefactor $\mu_0 = \beta\nu_0 e\rho^2$. (Obviously, for significantly larger fields, Eq. (11) can still be used to numerically evaluate the drift velocity, and thus the mobility, once the associated correlation function has been evaluated). We note, in passing, the interesting structure of Eq. (13), which relates the *nonlinear* (i.e., high field) response of the system in the presence of a driving field to the *Laplace* transform of an equilibrium correlation function of the *undriven system*. This is reminiscent of the familiar Kubo relation which relates the *linear* response of a system to the *Fourier* transform of a different equilibrium correlation function.

# 3    Dipolar Disorder

Equation (13) is applicable to any disordered one-dimensional chain with sufficient correlations that the underlying variation in the potential between neighboring sites is small compared to the thermal energy. We now consider, within the context of the dipolar disorder model, evaluation of the correlation function $\langle e^{-\beta U(0)}e^{\beta U(y)}\rangle = \langle e^{\beta\Delta_y}\rangle$ appearing in (13), where $\Delta_y = U(y) - U(0)$ represents the spatially-fluctuating energy difference between two sites separated by a displacement $y$. As we have noted, numerical investigations suggest that the overall distribution of site energies is well approximated by a Gaussian. The mean energy difference $\langle\Delta_y\rangle$ between

all sites separated by a distance $y$ is easily shown to vanish. We assume, as in the lattice calculations of Novikov and Vannikov,[17] that the distribution of energy differences is, for each value of $y$, also well-approximated by a Gaussian, so that the correlation function of interest can be described by a relation

$$\langle e^{\beta \Delta_y} \rangle = e^{\frac{1}{2}\beta^2 \langle \Delta_y^2 \rangle} \tag{14}$$

involving only the second moment $\langle \Delta_y^2 \rangle$. To calculate this latter quantity we compute the potential energy

$$U(\vec{r}_0) = -\sum_m \vec{p}_m \cdot \vec{E}_{\vec{r}_0}(\vec{r}_m) \tag{15}$$

of a carrier of charge $e$ located at a point $\vec{r}_0$ in a medium containing independently distributed and randomly oriented point dipoles $\vec{p}_m$. In (15),

$$\vec{E}_{\vec{r}_0}(\vec{r}) = \frac{e}{4\pi\epsilon} \frac{\vec{r} - \vec{r}_0}{|\vec{r} - \vec{r}_0|^3} \tag{16}$$

is the field at $\vec{r}$ due to the charge at $\vec{r}_0$. Equation (15) can be rewritten exactly in the form

$$U(\vec{r}_0) = -\int d^3r \ \vec{p}(\vec{r}) \cdot \vec{E}_{\vec{r}_0}(\vec{r}), \tag{17}$$

where $\vec{p}(\vec{r}) = \sum_m \vec{p}_m \delta(\vec{r} - \vec{r}_m)$ represents a fluctuating polarization density. We exclude the interaction energy of the charge with the molecule on which it is sitting, which will be the same for all similar dopant molecules, by excluding from the region of integration a volume of some radius $a$ comparable to the size of a dopant molecule. This is mathematically equivalently to treating the field in (16) as arising from a uniformly charged sphere of radius $a$, inside of which the field vanishes. The energy difference $\Delta_y = U(\vec{y}) - U(0)$ between two points separated by an arbitrary displacement $\vec{y}$ then becomes,

$$\Delta_y = \int d^3r \ \vec{p}(\vec{r}) \cdot \vec{E}(\vec{r}) \tag{18}$$

where we have introduced a hypothetical field $\vec{E}(\vec{r}) = \vec{E}_0(\vec{r}) - E_y(\vec{r})$, which can be mathematically interpreted as the field due to a positive sphere of radius $a$ located at the origin and a negative sphere located at $\vec{y}$. This interpretation is useful for evaluating $\langle \Delta_y^2 \rangle$, which may now be written as the double integral,

$$\langle \Delta_y^2 \rangle = \int d^3r \int d^3r' \ \vec{E}(\vec{r}) \cdot \langle \vec{p}(\vec{r}) \, \vec{p}(\vec{r}') \rangle \cdot \vec{E}(\vec{r}'). \tag{19}$$

Evaluation of the average

$$\langle \vec{p}(\vec{r}) \, \vec{p}(\vec{r}') \rangle = \sum_{n,m} \langle \vec{p}_m \vec{p}_m \rangle \langle \delta(\vec{r} - \vec{r}_n) \delta(\vec{r}' - \vec{r}_m) \rangle \tag{20}$$

over the random orientations and positions of the independently distributed dipoles is straightforward, insofar as orientational and spatial averages can be performed separately. Since the orientationally-averaged dipole moment vanishes, i.e., $\langle \vec{p}_m \rangle = 0$, the average reduces to

$$\langle \vec{p}(\vec{r}) \, \vec{p}(\vec{r}') \rangle = \langle \vec{p}_m \vec{p}_m \rangle \sum_{m,n} \delta_{nm} \langle \delta(\vec{r} - \vec{r}_n) \delta(\vec{r}' - \vec{r}_m) \rangle = \langle \vec{p}_m \vec{p}_m \rangle \delta(\vec{r} - \vec{r}') \langle \sum_n \delta(\vec{r} - \vec{r}_n) \rangle. \tag{21}$$

The average of the last sum just gives the mean dipole density $n_0$, while the orientational average of the dyadic product $\langle \vec{p}_m \vec{p}_m \rangle$ yields $\frac{1}{3}p^2 \mathbf{1}$, where $\mathbf{1}$ represents the unit tensor of second rank. With this result, (19) can be expressed in the form

$$\langle \Delta_y^2 \rangle = \frac{2p^2 n_0}{3\epsilon} \int d^3r \ \frac{1}{2}\epsilon |\vec{E}(\vec{r})|^2, \tag{22}$$

which is now easily identified at the energy needed to set up the field $\vec{E}(\vec{r}) = \vec{E}_{\vec{r}_0}(\vec{r}) - E_{\vec{y}}(\vec{r})$ of two oppositely charged spheres of radius $a$ separated by a distance $y$. Provided the two spheres do not overlap this is a textbook electrostatics problem, with the result[17,19]

$$\langle \Delta_y^2 \rangle = \frac{2p^2 n_0}{3\epsilon} \left( \frac{e^2}{4\pi\epsilon a} - \frac{e^2}{4\pi\epsilon y} \right) = 2\sigma^2 \left( 1 - \frac{a}{y} \right), \tag{23}$$

in which $\sigma = \left(e^2 p^2 n_0 / 12\pi\epsilon^2 a\right)^{1/2}$. Thus, for $y > 2a$, the correlation function (14) takes the form

$$\langle e^{\beta \Delta_y} \rangle = \exp\left[\beta^2\sigma^2 \left(1 - \frac{a}{y}\right)\right].$$

(24)

We now substitute (24) into (13) and evaluate the resulting Laplace transform to obtain[21,19]

$$\mu = \frac{\mu_0 e^{-\beta^2\sigma^2}}{2\beta\sigma \left(\beta e E a\right)^{1/2} K_1 \left[2\beta\sigma \left(\beta e E a\right)^{1/2}\right]},$$

(25)

where $K_1(z)$ is the first-order modified Bessel function of the third kind. Finally, for $2\beta\sigma \left(\beta e E a\right)^{1/2} > 1$, the Bessel function in (25) is well represented by its asymptotic expansion [see, e.g., Ref. 21, p. 963.] $K_1(z) \sim \sqrt{\pi/2z}$ $\exp(-z)$, leaving a mobility[19]

$$\mu = \mu_0\left(E\right) \exp\left(-\beta^2\sigma^2\right) \exp\left(2\beta\sigma\sqrt{\beta e E a}\right)$$

(26)

which displays the characteristic field dependence observed in molecularly-doped polymers, in which $\mu_0(E) = \mu_0 \left(\pi\beta\sigma\sqrt{\beta e E a}\right)^{1/2}$ is a prefactor that is slowly varying relative to the exponential factors. Note that the field independent prefactor $\exp\left(-\beta^2\sigma^2\right)$ supports the quadratic temperature dependence associated with the GDM, but omits the factor $2/3$ that appears in that model.

The simple analysis presented above and in Ref. 19 allows a number of conclusions to be drawn regarding charge transport in disordered systems. First, it provides an analytical confirmation of the idea that site energy correlations, when present, can play an important role in determining the precise field dependence of the mobility. The magnitude and concentration of the dipole moments associated with dopant molecules in typical molecularly-doped polymers insures that, at the very least, they will provide a background of correlated energetic disorder sufficiently strong that it cannot be legitimately neglected in describing these systems. The specific correlations that arise in the charge-dipole model lead, in this simple one-dimensional model, to the precise field dependence observed in the molecularly-doped polymers. We note in passing that the analysis presented here can be easily extended to treat other types of correlations arising, e.g., from induced dipole interactions, and charged defects. It remains to be seen whether the agreement in form suggested by Eq. (25) will be quantitatively validated in terms of the dopant dependence of the mobility observed in a wide variety of physical systems. While the general relationship observed between dipole moments of dopant molecules and the inferred activation energies is captured in the calculation presented above [Note the relation between the strength of the energetic disorder and the properties of the dipole distribution implicit in the expression following Eq. (23)], it is not yet clear whether the concentration dependence of the activation energy predicted from the present calculation can be quantitatively reconciled with that deduced in experiment. In addition, while we have made plausible arguments regarding the potential applicability of our analysis to describe the bulk behavior of a system, the rigorous extension of these ideas to three-dimensional transport remains an important area for future theoretical studies.

# 4    ACKNOWLEDGMENT

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