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Combined Impact of Field and Carrier Concentration on Charge Carrier Mobilities in Amorphous Organic Thin Films

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1. Introduction

A critical process governing amorphous organic electronic devices is charge transport, and for this reason the theory of charge transport in disordered molecular solids has been the subject of intense research over the last thirty years. Extensive Monte Carlo simulations have been performed to obtain the equilibrium charge carrier mobility as a function of field in the limit of negligible charge carrier concentration (see e.g. [1] and [2]). Sophisticated analytic theories for equilibrium charge carrier mobilities as a function of carrier concentration in the low field limit have also been reported (see e.g. [3]).

However, many amorphous organic electronic devices (e.g. organic light emitting devices and photodetectors) operate in a regime where both high fields and high carrier concentrations are present. As a result, to properly model current flow in such devices one must take into account the influence of both factors when calculating carrier mobilities. We report what are to our knowledge the first Monte Carlo simulations of equilibrium charge carrier mobilities in amorphous organic solids as a fucntion of *both* field and carrier concentration. We also compare these Monte Carlo calculations with existing analytic methods and find that the analytic calculations have significant errors, attributable to: (1) inadequate correction of hopping cycles; and (2) improper use of Fermi statistics.

2. Calculation Method

We analyze charge transport in amorphous organic materials assuming as model of incoherent hopping between molecular sites subject to energetic disorder. We employ the Miller-Abrahams[4], field–assisted hopping rate to calculate the rate of transfer of a charge carrier to an unoccupied molecule through a displacement $\vec{R}$, subject to an applied field $\vec{F}$, and having charge q:

$$\Gamma_{hop} = \frac{1}{\tau_{hop}} \exp[-\gamma R]\chi(\Delta E) \qquad (1)$$

where ΔE is the difference in the site energy levels,

$$\chi(\Delta E) \equiv \begin{cases} 1 & if \quad \Delta E^* < 0 \\ \exp[-\Delta E^*/k_B T] & if \quad \Delta E^* \geq 0 \end{cases} \qquad (2)$$

$k_B T$ is the thermal energy, and,

$$\Delta E^* \equiv \Delta E - q\vec{R} \cdot \vec{F}. \qquad (3)$$

We model the impact of non-zero carrier concentration by prohibiting transfer to sites that are already occupied.

We assign the site energies using either the Gaussian Disorder Model (GDM)[1] or the Correlated Disorder Model (CDM)[5]. In both cases, the charge carrier density of states, $g(E)$, is a Gaussian function:

$$g(E) = N_{mol} \frac{1}{\sqrt{2\pi\sigma}} \exp[-E^2/2\sigma] \qquad (4)$$

where N_{mol} is the density of molecular sites. For the GDM, the site energies are assigned randomly. For the CDM, the site energies are subject to spatial correlations arising from the interaction between the charge carrier and randomly oriented surrounding dipoles, as previously described by Novikov *et al.*[5].

We apply this model to the calculation of equilibrium charge carrier mobilities using Monte Carlo simulations. Cubic lattices of 40x40x40 sites are used in this study, with an intersite spacing of 1 nm (yielding $N_{mol} = 1x10^{21} cm^{-3}$.) A random fraction of the lattice sites are initially populated with charge carriers. The simulation then proceeds in time steps until the average mobility of the carrier population (calculated by dividing the average

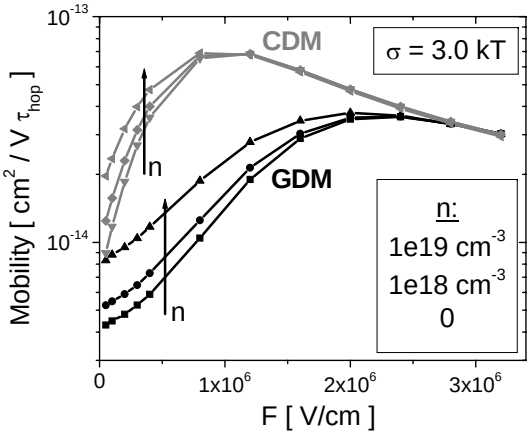

Figure 1: Equilibrium mobility as a function of field and carrier concentration, n, under the GDM and CDM for $\sigma = 3k_BT$, calculated by Monte Carlo simulation.

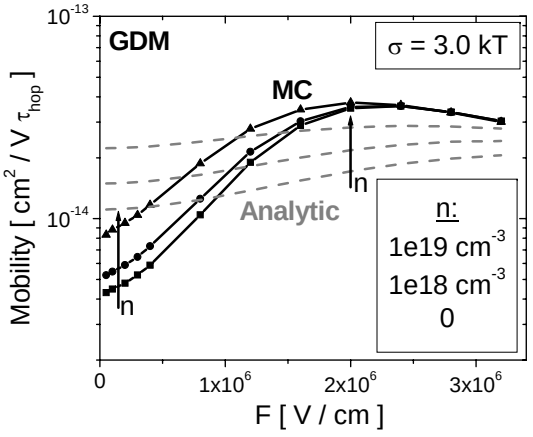

Figure 2: Comparison of equilibrium mobility as a function of field and carrier concentration, n, under the GDM for $\sigma = 3k_BT$, calculated by Monte Carlo simulation (black lines and symbols) and using the analytic treatment reported in [6].

carrier velocity by the applied field) is found to equilibrate.

3. Results and Analysis

Shown in Fig. 1 are our calculations of the equilibrium mobility as a function of field for three carrier concentrations, $n = 0$, $n = 1x10^{18}cm^{-3}$, and $n = 1x10^{19}cm^{-3}$, for both the GDM and CDM with $\sigma = 3k_BT$. We have performed calculations for a range of other σ and n values and obtained similar results. We find that increasing n universally increases the equilibrium mobility, and that this effect is more dramatic for larger σ and smaller fields. From these simulation results, we have also developed approximate empirical relationships for the mobility as a function of field and carrier concentration over the Poole-Frenkel regime for both the GDM and the CDM.

To our knowledge, the only analytic treatment of the charge carrier mobility in amorphous organic materials as a function of carrier concentration at high fields is due to Roichman *et al.*[6]. In this report, the authors employ a simple variable range hopping model to calculate equilibrium mobilities under the GDM. The impact of applied field is treated through the same field-assisted Miller-Abrahams hopping rate that we have used. The impact of the carrier concentration is similarly introduced by prohibiting transfer to occupied states, and the equilibrium condition is expressed by using Fermi statistics to described the state occupancy.

We compare the results of this analytic treatment with our Monte Carlo calculations, and find that the the analytic results are significantly different from the Monte Carlo results (see Fig. 2.) We find that this disagreement arises from two shortcomings of the analytic treatment reported by Roichman *et al.*[6]. First, the method does not account for the occurance of hopping cycles (i.e. sequences of hops which return a carrier to a previously visited site). Second, the method assumes Fermi statistics, which is found to be inconsistent with charge carrier hopping in the presense of an applied field.

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