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The effects of *p*-diethylaminobenzaldehyde diphenylhydrazone, a 0.22 eV trap, on the charge transport properties of disordered organic materials consisted of a mixture of N,N′-diphenyl N,N′-bis(3-methylphenyl)-(1,1′-biphenyl)-4,4′-diamine and polycarbonate are described. Trap-perturbed, trap-limited, and trap-controlled (trap-to-trap) transports were observed, where the trap is less effective at very low trap concentrations but appears to have increasing trap depth with respect to the concentration increase in the trap-limited regime. Most photocurrent transients showed non-dispersive transport behaviors; however, some were dispersive, especially at low electric fields or near the cross-over region between the trap-limited and trap-controlled transports. Field dependency of the mobilites at the trap-limited regime is higher than that of the trap-controlled regime, suggesting some type of superexchange phenomenon or field induced detrapping for the latter transport. For most known systems, charge transport reaches at a minimum mobility that is higher than anticipated from the trap depth. Analysis of the experimental results by theoretical models based on molecular crystals and disordered materials strongly suggests the influence of disorder but only gives qualitative agreements. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4757412]

INTRODUCTION

Conjugated organic materials are being used or under development for a number of optoelectronic applications such as electrophotographic photoreceptors,[1] emissive displays,[2] photovoltaic devices,[3] solid state lighting,[4] photorefractive materials for 3-D displays,[5] and flexible thin film transistors.[6] Many challenges remain, including the need to improve charge mobility, reduce impurities and charge traps, and durability. Particularly, charge traps can cause loss of the image development field in electrophotography, efficiency loss and defects in displays and lighting, and low performance in photovoltaic and transistor devices. Therefore, investigation of charge transport and trapping properties of these materials is of broad technological and fundamental interest.[7–13]

Charge transport in conjugated organic materials is governed by their disordered energy landscape, where electron hops among active molecules or molecular moieties and usually exhibits Poole-Frenkel type behavior with logarithm of mobility linear to the square root of electric field, $\log \mu \sim E^{1/2}$. Hopping is influenced by the energetic disorder of the electronic states of active molecules, and earlier theories, attempted to explain the seemingly universal square root dependency, most notably the Gaussian disorder model by Bässler and coworkers[10] can only predict the behavior over a narrow range of electric field. In the model, the energetic disorder is described by a Gaussian distribution, σ. Later, Dunlap and coworkers proposed that the wide range field dependency was due to dipolar disorder, removing the assumption of the absence of spatial correlation and setting on a premise that the interaction of charge carriers with randomly located but oriented dipoles induces a highly correlated energy landscape.[14,15]

With the presence of traps, intrinsic or extrinsic, the charge transport becomes more complicated and the mobility is strongly correlated with the trap depth and concentration. Historically, there have been only few investigations of charge trapping behaviors on doubly doped conjugated materials.[16–22] Generally, there are three stages of trap influenced transport related to the trap concentration. First, at very low concentrations, ca. 10^{-5} mole fraction depending on the sample thickness (the distance a charge needs to go through) and trap depth, mobility only decreases slightly from that of the zero trap concentration, and the phenomenon is noted as trap-perturbed transport. As the concentration increases, mobility is now significantly affected by the trap and decreased substantially, where the transport behavior is noted as trap-limited transport. Mobility reaches a minimum at a certain trap concentration usually around 10^{-1} mole fraction of traps, a phenomenon that the underlying mechanism is still not well understood. The minimum mobility is also significantly higher than that implied by the trap depth, which appears to be universal but also not very well understood. As the trap concentration continues to increase, the mobility will increase as the charge transport now crosses over to the trap-controlled regime and dictates mainly by the transport behaviors of the trap with diminishing contributions from the host.

For trap-influenced charge transport, the early work by Hoesterey and Letson on molecular crystals provides a good foundation for mechanistic understanding; however, the model does not take into consideration of disorder.[23] Based

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on the Gaussian disorder model, numerical simulation and experimental results related to the trap depth and concentration were reported and showed qualitative agreements with the experimental data.[24] Similar to the original trap-free model, the model assumes that despite weak electron-phonon couplings, thermal equilibrium can be achieved for charge and heat bath but with a broadening factor associated with the energetic disorder and trap depth. A theory based on effective medium approach (EMA) has also been developed to describe the nondispersive charge transport in disordered organic materials containing extrinsic traps.[25] This approach can account qualitatively for the dependence of charge carrier mobility on the trap concentration, temperature, and electric field. Later, the EMA was extended to include the dependence of the overlap integral due to trap dilution, which provides a more accurate estimate of the effective mobility.[26] More recently, a bimodal Gaussian density of states was used to investigate host-guest systems.[27] The study found excellent agreements with the EMA results for high and low trap concentrations. However, at the cross-over region, between the trap-limited and trap-controlled hopping the model predicts a much larger mobility. Surprisingly, there have been very few investigations of the above models with experimental results. Furthermore, very little is currently known about the underlying mechanism and critical parameters of the trap-influenced transport.

In this paper, we report charge trapping properties of N,N′-diphenyl N,N′-bis(3-methylphenyl)-(1,1′-biphenyl)-4,4′-diamine (TPD), a widely investigated hole transport material, in a polycarbonate binder, doped with a trap dopant of *p*-diethylaminobenzaldehyde diphenylhydrazone (DEH), where the trap depth is 0.22 eV, determined from solution-phase electrochemical potential measurements. The charge trapping behaviors of the doubly doped system were analyzed with the Hoestery-Letson formalism and EMA model and compared with previous results. We found that both treatments can qualitatively describe the observed mobility variations with respective to the trap concentration, but quantitatively both show some discrepancies or deficiencies in describing critical concentration and effective mobility.

EXPERIMENTS

Molecular structures of TPD and DEH are shown in Figure 1(a). DEH is a better donor with a 0.22 eV higher electronic ground state than that of TPD. For sample preparation, the weight ratio between the polycarbonate binder and the transport-active small molecules (TPD or TPD + DEH) was always fixed at 1 to 1. The molar ratio between DEH and TPD is described as the total mole of DEH with respect to that of the total mole of TPD and DEH, which varies from 0 to 1 mole fraction (mole/mole). Devices were prepared by draw bar coating of solution mixtures of the molecular doped polymer on a pre-coated charge generating layer consisted of a phthalocyanine on a titanium and zirconium alloy coated polyethylene terephthalate substrate and after cured at 120 °C for about 10 min and cooled down to room temperature, a gold electrode was deposited on the films. Most of the devices are about 10 μm in thickness as
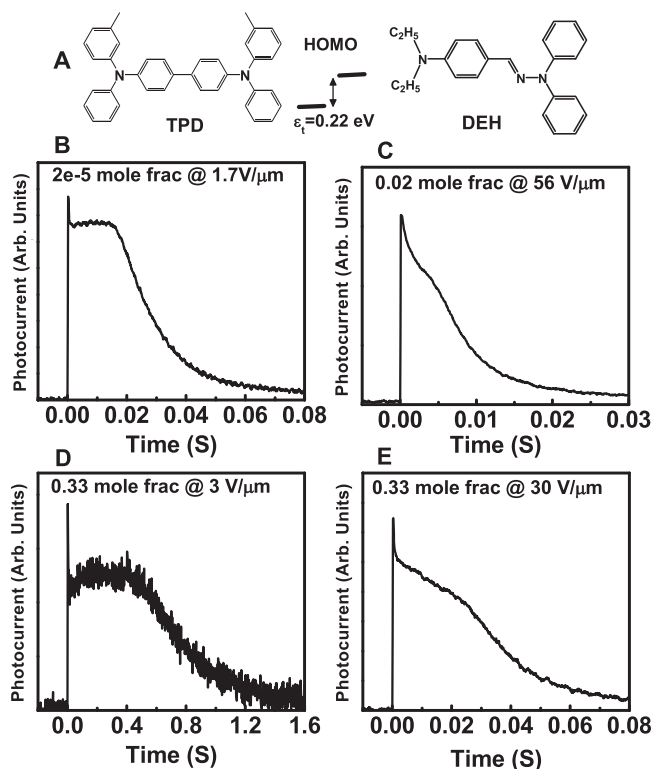

FIG. 1. (a) Molecular structures of DEH and TPD and the trap depth determined from electrochemical oxidation potential measurements. (b)–(e) Representative photocurrent transients of DEH doped TPD in polycarbonate.

measured by a permascope thickness gauge. Charge mobility was measured in ambient by the conventional time-of-flight technique with negative bias applied at the gold electrode side. Care was taken to limit the light exposure to a minimal value so that the number of free electron pairs generated was less than 2% CV, rendering the charge transport studies in the emission limited region. Electric fields applied were typically from 1 to 60 V/μm. The nature of electronic states dictates charge transport behaviors, where the hopping rates are not unique for all holes participated in the transport and resulted in various transit times. Photocurrent transit time is usually determined by the asymptotic intersection of the photocurrent plateau and tail, which is referred as t_0 and represents transit time of those high mobility holes first traversed the film. Alternatively, the time where the photocurrent intensity decreases by half to its plateau value, which is referred as $t_{1/2}$ and represents the median transit time of the holes, can also be used. A careful study of transit times derived by the two methods found that the difference between the two types of transit times stayed constant for most data, and therefore, only $t_{1/2}$ transit time was used to determine the mobility. In the few cases of slightly dispersive transport, semi-logarithmic plots were used to discern the transit times.

RESULTS AND DISCUSSION

Majority of the photocurrent transients showed non-dispersive charge transport behaviors, suggesting holes in the trap and host have reached steady state. Figures 1(b)–1(e) show several representative photocurrent transients at various

electric field strengths versus DEH mole fractions. Typically, for low mole fraction samples, the photocurrent transients showed non-dispersive transport feature, especially at low and high electric fields, ca. 1–2 V/μm (Fig. 1(b)) and >20 V/μm, respectively, then appeared to show some slightly dispersive transport behaviors at medium electric fields, ca. 10 V/μm. The non-dispersive transport behavior suggests that the trap and host have reached a steady state even at the low electric fields. As the trap concentration further increased, reaching the trap-limited regime, some slightly dispersive transport was also found, such as the photocurrent transient shown in Fig. 1(c). When the concentration increased high enough so that the trap-to-trap transport or trap dominated transport became the major transport mechanism, and depending on the charge transport behaviors of the trap usually, the transport was again non-dispersive (Fig. 1(d)). In general, with the presence of a significant concentration of an extrinsic trap, the photocurrent transients at all electric fields tended to become more dispersive than without (Fig. 1(e)). We speculate that the dispersive behaviors could be attributed to sample preparation, purity, and insufficient thickness, but we cannot rule out potential contributions from intrinsic factors such as phase separation resulted from insufficient or inhomogeneous molecular packing.

Mobilities of the DEH doped TPD at various trap mole fractions versus square root of electric field strength are shown in Figs. 2 and 3, where obviously the E$^{1/2}$ dependency is preserved even with the presence of the 0.22 eV trap at all the concentrations studied. Only at very low electric fields, deviation from the field dependency is observed, where the phenomenon is perhaps due to longer trap (from both intrinsic and extrinsic) release times at the low electric fields. Note that at low electric fields, such a deviation usually exists even without the presence of extrinsic traps, as indicated by the 0 mole fraction (pure TPD in polycarbonate) results. It is unclear why at low electric fields, the mobility becomes lower than the trend predicted by the mobilities at higher fields, but speculated that it is due to the presence of deep traps (intrinsic and extrinsic) or delay injection of holes by an energetic bar-

rier between the phthalocyanine charge generation layer and charge transport layer. Note that at very low trap concentrations, such as the 0.002 mole fraction results indicated, the electric field dependency of mobilities is similar to that of the pure host, suggesting a low probability of hopping to traps that does not change the general charge transport characteristics—a feature usually describes as trap-perturbed transport. The field dependency increases as the trap concentration increases to 0.01–0.1 mole fraction, a regime described as trap-limited transport. The similar field dependency for the three trap concentrations at 0.01, 0.033, and 0.1 mole fractions suggests that there is one dominating underlying mechanism, i.e., all hops see the same type of perturbation in this range. As the trap concentration further increases, at around 0.33 mole fraction, the field dependency actually decreases with respect to those in the trap-limited regime, indicating that in this trap-controlled transport regime, the host is providing some sort of "boost" to the trap transport, and therefore the field dependency actually decreases. The effect can also be attributed to electric field induced detrapping.[28] For pure trap, the field dependency steepens, reflecting the nature of charge transport properties of DEH.

To further analyze the results, mobilities at the electric fields of 0, 9, and 64 V/μm are plotted against various trap concentrations, where the zero field mobility was obtained by extrapolating the trend predicted by the high field mobilities to zero electric field, and the results are shown in Fig. 4. Other than the magnitude of mobility, no significant differences among the data for the three electric fields are seen, and only for pure DEH (mole fraction = 1), since its mobility is strongly dependent on electric field, the differences among the electric fields are significantly differ for different fields. The mobility only decreases slightly at trap concentration smaller than ca. 10^{-3} mole fraction. One possible reason is that, as discussed earlier, the trap and host may not be fully equilibrated at the thicknesses studied, which is why that the decrease in mobility is not obvious at the low concentrations. Strong trap influenced transport is observed at concentrations above 10^{-3} mole fraction, where the decrease in mobility is
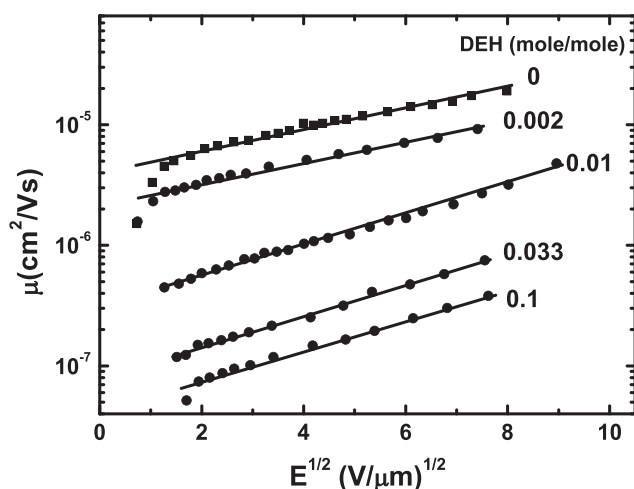

FIG. 2. Hole mobilities of DEH doped TPD in polycarbonate as a function of the square root of the electric field strength (E) at 0 to 0.1 mole fraction of trap concentration.
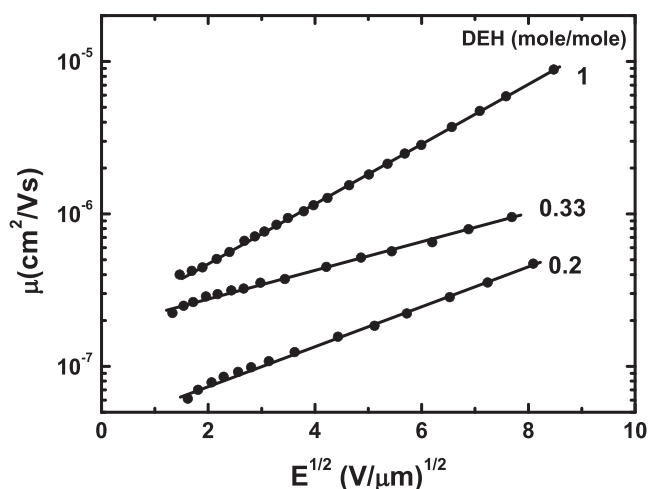

FIG. 3. Hole mobilities of DEH doped TPD in polycarbonate as a function of the square root of the electric field strength (E) at 0.2 to 1 mole fraction of trap concentration.
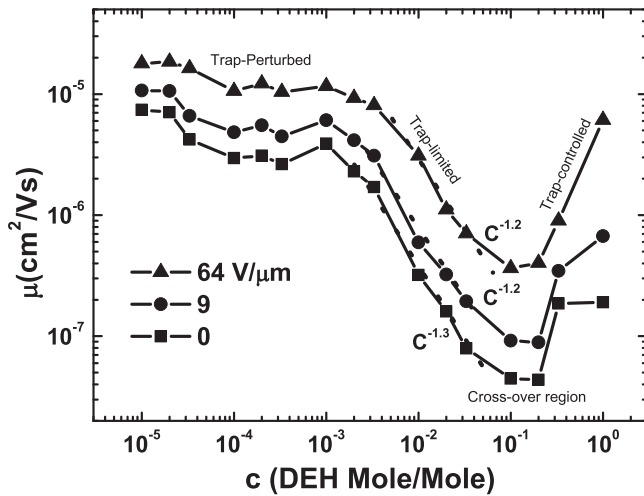
FIG. 4. Effective hole mobilities of DEH doped TPD in polycarbonate as a function of 0 to 1 mole fraction trap concentration at the electric field strengths of 0, 9, and 64 V/μm.

about 1 order of magnitude at about 1 order of magnitude increase in concentration (10^{-3} to 10^{-1} mole fraction). The trap concentration where the mobility reduces by a half of its trap-free value is referred as $c_{1/2}$. The $c_{1/2}$'s are equal to about 6×10^{-4} to 1×10^{-3}, 2×10^{-3}, and 4×10^{-3} at the electric fields of 0, 9, and 64 V/μm, respectively. The trap-influenced mobility predicted by the Hoesterey-Letson formalism is given as[23]

$$\mu_c(T) = \mu_0[1 + c \exp(\varepsilon_t/kT)]^{-1}, \tag{1}$$

where c is the relative trap concentration defined as the number density of trap, N_t, over the total number density of traps and hosts, N, thus $c = N_t/N$, μ_c the mobility at a certain trap concentration, c, μ_0 the trap-free mobility, ε_t the trap depth, and k the Boltzmann constant. For mobility to be reduced to half of its trap-free value, $c_{1/2}$ will be equal to the Arrhenius factor, $\exp(-\varepsilon_t/kT)$, which is about 2×10^{-4} for a trap depth of 0.22 eV at 298 K for the DEH/TPD system. The value is smaller but not substantially different from the experimental values. Alternatively, the EMA model provides an expression[26]

$$c_{1/2} = \exp\left[-\frac{\varepsilon_t}{kT} - \frac{1}{2}\left(\frac{\sigma_0}{kT}\right)^2(\eta^2 - 1)\right], \tag{2}$$

where $\sigma_0(\sigma_1)$ is the width of the density of states of the host (trap), $\eta = \sigma_1/\sigma_0$, and ε_t is the trap depth. Taking the values, $\sigma_0 = 0.089$ eV and $\sigma_1 = 0.11$ eV from the literature,[29] $c_{1/2}$ is $\sim 6.8 \times 10^{-8}$, which is way too small from the experimental results, suggesting the effect of disorder could be overestimated by the EMA model at the low trap concentrations.

We next discuss the trap-limited regime. According to the Hoesterey-Letson expression in the Eq. (1), under the criteria of a large trap depth and high trap concentration, μ_c is approximately proportional to $1/c$. The trap depth is 0.22 eV for DEH relative to TPD, and therefore μ_c should be proportional to $1/c$ at concentrations greater than 10^{-3} mole fraction. The resulting fits give slopes greater than 1 for the

mobility data in the 10^{-3} to 10^{-1} mole fraction range (Fig. 4). The slopes also appear to have a very little field dependency for all the electric fields studied. The results suggest that DEH is a more efficient trap than the model predicted in the trap-limited regime. Alternatively, it acts as if the trap depth is increasing with respect to the increase in concentration. However, the uncertainties in the trap depth determination—such as errors in the oxidation potential determination or the different environments between the electrochemical measurements (in solution) and the charge transport studies (in solids) are being made—could also lead to the discrepancy. Nevertheless, it is worth noting that traps can be more efficient than the trap depth implied or the appearance of an increasing trap depth with respect to the trap concentration in the trap-limited transport regime. This type of over effectiveness has also been observed earlier in a trap study of DEH in di-p-tolylphenylamine (DTP), where the trap depth is 0.32 eV.[30] Assuming all carriers are thermalized, for a 0.1 eV difference in the trap depths between the DEH/TPD and DEH/DTP systems, at room temperature the ratio of the minimum mobilities between the two traps should be about 1/50 based on the energy gap ($\exp^{0.1eV/kt} \sim 47$). However, the difference in the measured mobilites between the two studies is calculated to be about 3 orders of magnitude for the minimum mobilities ($3 \times 10^{-5}/2 \times 10^{-8} \sim 1000$).

The minimum mobility in the cross-over region is at about 0.09 mole fraction trap concentration from the experimental results, which has only been qualitatively predicated by the effective medium theory[25] and the percolation model.[27] From most of the known studies, the location of the minimum mobility for this type of extrinsic trap doped systems varies from ~ 0.02 to 0.1 mole fraction and usually higher the trap depth lower the critical concentration.[17,19–21] Based on the EMA model, the critical concentration, c_{cr}, where the mobility reaches a minimum is expressed by the following transcendental equation:

$$c_{cr} = \exp\left[-\frac{\varepsilon_t}{kT} + \frac{a}{b}\left(\frac{1}{c_{cr}^{1/3}} - 1\right)\right], \tag{3}$$

where a is the average spatial distance between transport sites ($a = N^{-1/3}$), and b is the localization radius defined as the localized wave function decay length of a charge carrier and is assumed to be the same for both host and trap sites. The critical concentration for the DEH/TPD is calculated as 0.25 from Eq. (3), which is significantly higher than the 0.09 value observed from the experimental results. We have also compared two earlier studies that contain the cross over region, and similarly the calculated values are much higher than the experimental data.[17,30] Summary of the $c_{1/2}$ and c_{cr} values obtained from this work and previous publications is shown in Table I. We infer from the results that the EMA model underestimates the effect of traps in the trap-limited regime but overestimates the role of traps in the trap-perturbed regime.

As the trap concentration continues to increase the mobility increases, a clear indication of trap-to-trap or trap controlled transport, where the host becomes inert or "antitrap" and presumably the "host" now plays a lesser role in transport.

TABLE I. Trapping parameters for several hole transporting materials obtained from experimental data or calculated from the Hoesterey-Letson formalism and EMA model (the experimental data for DEH/DTP, TPD/PVK, and (TAA/TTA, DTA/TTA, and DAT/TTA) were taken from Refs. 17, 21, and 30, respectively. PVK: polyvinylcarbazole; TTA: titolylamine; TAA: tri-*p*-anisylamine; DTA: di-*p*-anisyl-tolylamine; DAT: di-*p*-tolylphenylamine.).

Trap/Host	ε_{t}(eV)	$c_{1/2}$ (H-L)	$c_{1/2}$ EMA	$c_{1/2}$ exp	b (Å)	c_{cr} EMA	c_{cr} exp
DEH/TPD	0.22	1.9×10^{-4}	4.3×10^{-5}	10^{-4}–10^{-3}	2.4	0.25	0.09
DEH/DTP	0.32	4.2×10^{-6}	3.5×10^{-9}	9.0×10^{-6}	2.1	0.15	0.068
TPD/PVK	0.5	2.9×10^{-9}	6.6×10^{-6}	No data	2.0	0.069	0.02
TAA/TTA	0.07	4.5×10^{-9}	6.9×10^{-3}	3.0×10^{-2}	1.9	0.62	No data
DTA/TTA	0.15	0.003	6.1×10^{-5}	2.7×10^{-3}	1.9	0.39	No data
DAT/TTA	0.22	0.0002	1.7×10^{-7}	2.8×10^{-4}	1.9	0.27	No data

Previously, mobility versus trap concentration results were analyzed with EMA and a master-equation approach and percolation model and showed qualitative agreements between the experimental data and theories.[25–27] The EMA is a two-site cluster model, where hops along and against the electric field are both considered. The effective drift mobility is defined as[25,26]

$$\mu_e = \mu_2 \frac{Y_e^+ - Y_e^-}{f} \;, \tag{4}$$

where $Y_e^+(Y_e^-)$ is the jump rate along (against) the electric field described by

$$
\begin{aligned}
Y_e^{\pm} =\; & A(1-c)^2 i_1^{\pm} \exp\left[-2\frac{a}{b}\left(\frac{1}{(1-c)^{1/3}} - 1\right)\right] \\
& + A(1-c)c[i_2^{\pm} i_3^{\pm} \exp(xy)] \\
& \times \exp\left[-\frac{a}{b}\left(\frac{1}{(1-c)^{1/3}} + \frac{1}{c^{1/3}} - 2\right)\right] \\
& + Ac^2 i_4^{\pm} \exp(xy) \exp\left[-2\frac{a}{b}\left(\frac{1}{c^{1/3}} - 1\right)\right],
\end{aligned} \tag{5}
$$

$$
A = \frac{1}{(1-c)\exp\left[\frac{1}{2}\left(\frac{\sigma_0}{k}\right)^2\right] + c\exp\left[\frac{\epsilon_t}{k} + \frac{1}{2}\left(\frac{\sigma_1}{k}\right)^2\right]},
$$

$$
x = \frac{\sigma_0}{k_B T}, \quad y = \frac{\varepsilon_t}{\sigma_0}, \quad \mu_2 = \frac{ea^2 v_0}{\sigma_0}\exp\left(-2\frac{a}{b}\right), \quad \text{and} \quad f = \frac{eaE}{\sigma_0}. \tag{6}
$$

In Eq. (5), the $i_n^{\pm}$'s are equations related to the host-to-host, trap-to-trap, and host-to-trap (or trap-to-host) charge transport,[31] and in Eq. (6), v_0 is the average energy spacing between hopping sites, and all other parameters have been defined in previous sections. The first and third terms in the right side of the Eq. (5) describe the jump rates for the host-to-host and trap-to-trap hops, respectively, whereas the second term is related to the host-to-trap hops and vice versa.

We have attempted to fit the mobility versus trap concentration data by Eq. (4) for many different electric fields obtained in this work without success. For comparison purposes, we have calculated the effective mobility by Eq. (4) using the values of $\sigma_0 = 0.089$ eV, $\sigma_1 = 0.11$ eV, $a = 11.8$ Å, and $b = 2.4$ Å, which is 1/5 of a, a reasonable estimate. The resulting curve in Fig. 5 shows a huge discrepancy to the experimental data (64 V/μm). The model appears to overly

estimate the contribution of trap in the trap-perturbed regime and underestimate the effect of trap in the trap-limited regime, which results in a wide and deep bell curve. We have also done reference fitting using data appeared in previous works, and only the DEH/DTP study could be reproduced, where the results are similar to those obtained in the Ref. 26 (see the inset of Fig. 5). Equation (4) is also weakly dependent on electric field as the f function and error functions (in the $Y_e^{\pm}$ functions) both contain the electric field term and will cancel each other out (see Eq. (5) and Ref. 31). We infer from this study that the EMA model works well for high trap depth systems but not for those with a low trap depth or a large difference in the width of density of states between the host and trap.

In summary, charge trapping of an arylamine doped polycarbonate by a hydrazone of a trap depth of 0.22 eV was studied. Under the presence of such trap, as the concentration increases, the mobility first decreases slightly by a factor or few, a charge transport regime described as trap-perturbed. Both the Hoesterey-Letson formalism and EMA model, especially the latter, overestimate the effect of trap concentration in this regime. At above 10^{-3} mole fraction, the trap transport enters into the trap-limited regime and the mobility reduces at a very steep rate, where the slope of the decline is greater than 1/c, suggesting the trap is more effective than the trap depth prescribed based on the Hoesterey-Letson
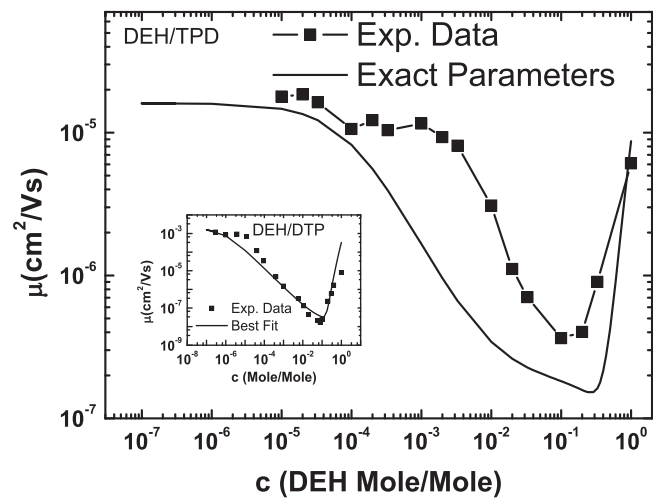

FIG. 5. Comparison of the experimental results for the mobility versus trap concentration at 64 V/μm with the simulation results obtained using exact parameter values from literature. Inset: Comparison of experimental and simulation results obtained using DEH/DTP data from Ref. 30.

formalism. Alternative, it can be viewed as an increasing trap depth with respect to the increase of trap concentration. The trap-limited mobility reaches a minimum at ~0.1 mole fraction with a value about 1/50 to that of the host, implying a mere 0.1 eV effective trap depth. The critical concentration, c_{cr}, is about 0.09, which is much smaller than that calculated from the EMA model, implying that the effect of trap is being underestimated. From most known studies, the minimum mobilities for this type extrinsic trap doped systems are about 0.02-0.1 mole fraction, depending on the trap depths. As the concentration further increases, charge transport becomes trap to trap and the mobility increases at a steep rate, and its field dependency lowered with respect to those at the trap-limited region. The phenomenon is perhaps due to a superexchange interaction between the host and trap or because of a field-induced detrapping effect. There remain a number of unresolved issues that will require further investigation.

ACKNOWLEDGMENTS

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[31]The $i_n^{\pm}$ functions are expressed as $i_1^{\pm} = \frac{1}{2}\exp(\frac{x^2}{2}) \left\{ \left[1 - \mathrm{erf}(\frac{\pm f}{\sqrt{2}})\right] \exp\left(-\frac{x^2}{2} \pm xf\right) + \left[1 - \mathrm{erf}(\frac{x\mp f}{\sqrt{2}})\right] \right\}$, $i_2^{\pm} = \frac{1}{2}\exp(\frac{x^2}{2}) \left\{ \left[1 - \mathrm{erf}(\frac{\pm f + y}{\sqrt{2}})\right] \exp\left(-\frac{x^2}{2} \pm x(\pm f + y)\right) + \left[1 - \mathrm{erf}(\frac{x\mp f - y}{\sqrt{2}})\right] \right\}$, $i_3^{\pm} = \frac{1}{2}\exp(\frac{x^2\eta^2}{2}) \left\{ \left[1 - \mathrm{erf}(\frac{\pm f + y}{\eta\sqrt{2}})\right] \exp\left(-\frac{x^2\eta^2}{2} \pm x(\pm f + y)\right) + \left[1 - \mathrm{erf}(\frac{\eta^2 x\mp f - y}{\eta\sqrt{2}})\right] \right\}$, and $i_4^{\pm} = \frac{1}{2}\exp(\frac{x^2\eta^2}{2}) \left\{ \left[1 - \mathrm{erf}(\frac{\pm f}{\eta\sqrt{2}})\right] \exp\left(-\frac{x^2\eta^2}{2} \pm xf\right) + \left[1 - \mathrm{erf}(\frac{\eta^2 x\mp f}{\eta\sqrt{2}})\right] \right\}$, where $\mathrm{erf}(z) = \frac{2}{\sqrt{\pi}} \int_0^z dt \exp(-t^2)$ is the error function.