

Electric field dependence of charge carrier hopping transport within the random energy landscape in an organic field effect transistor

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We extended our analytical effective medium theory [Phys. Rev. B **81**, 045202 (2010)] to describe the temperature-dependent hopping charge carrier mobility at *arbitrary electric fields* in the large carrier density regime. Special emphasis was made to analyze the influence of the lateral electric field on the Meyer–Neldel (MN) phenomenon observed when studying the charge mobilities in thin-film organic field-effect transistors (OFET). Our calculations are based on the average hopping transition time approach, generalized for large carrier concentration limit finite fields, and taking into account also spatial energy correlations. The calculated electric field dependences of the hopping mobility at large carrier concentrations are in good agreement with previous computer simulations data. The shift of the MN temperature in an OFET upon applied electric field is shown to be a consequence of the spatial energy correlation in the organic semiconductor film. Our calculations show that the phenomenological Gill equation is clearly inappropriate for describing conventional charge carrier transport at low carrier concentrations. On the other hand a Gill-type behavior has been observed in a temperature range relevant for measurements of the charge carrier mobility in OFET structures. Since the present model is not limited to zero-field mobility, it allows a more accurate evaluation of important material parameters from experimental data measured at a given electric field. In particular, we showed that both the MN and Gill temperature can be used for estimating the width of the density of states distribution.

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I. INTRODUCTION

The charge carrier mobility (μ) is the critical parameter that limits the efficiencies of organic electronic devices as organic field-effect transistors (OFETs)¹ or organic photovoltaic cells.² It is known that the disorder has a strong impact on the charge transport in organic semiconducting films. It is generally accepted that charge carrier transport in such materials occurs by hopping between localized states that are disordered in space and energy. Therefore charge carrier mobilities in these materials are, as a rule, very low and strongly temperature and field dependent. This fact has been consistently described by a formalism based on hopping in a Gaussian density of states (DOS) distribution.^{3–5} The Gaussian disorder model (GDM) originally suggested by Bässler and coauthors^{3,4} has been developed specifically for the low, carrier density hopping transport regime in a random organic system. Such limiting conditions are realized, for example in time-of-flight (ToF) photocurrent transient experiments. However, this might not always be the case in real electronic devices in which space charge effects are important for large carrier concentrations obtained in some organic electronic devices, e.g., in the conductive channel of an OFET, a sizeable fraction of available hopping states is occupied and a Fermi level is established. Therefore, the charge carrier mobility depends also on carrier concentration. This has been described within an extended GDM (EGDM).^{6–10}

Despite the success of the disorder formalism in rationalizing many aspects of the charge transport in organic

semiconductors, the electrical transport mechanisms in realistic organic electronic devices are still not completely understood. For instance, the dependence of the charge carrier mobility on the electric field (F) is of particular interest as it reveals the type of charge hopping transport in organic materials. Numerous ToF measurements reported for a wide range of small molecules and polymer-based organic solids demonstrated^{4,11} that mobilities of charge carriers typically increase with electric field, according to $\ln \mu \propto F^{1/2}$ law, called the “Poole–Frenkel” (PF)-type dependence, though the PF description itself is strictly not applicable to organic semiconductors.⁴ The field dependence of the hopping charge mobility arises due to the electric field lowering of the average barrier height for energetic uphill jumps in the field direction. Extensive Monte Carlo simulations of the charge transport within the GDM approximation using the Miller–Abrahams jump rate model¹² have been corroborated in the PF behavior, yet for a rather limited field interval,⁴ which turns out to be considerably narrower than that relevant to experimental observations. This limitation of the classical GDM has been overcome by introducing some correlations between the energies of nearby sites, as suggested first by Conwell and coauthors.^{13,14} Later, Parris *et al.*¹⁵ and Novikov *et al.*¹⁶ have suggested a so-called correlated disorder model (CDM). This suggestion was based on computer simulations of the charge transport in the low carrier concentration limit with accounting for long-range spatial energy correlations. The CDM model (being an amended version of the GDM) has extended the range of validity of the PF law down to moderate electric fields

for the charge carrier mobility. Long-range energy correlations in organic disordered solids imply slowly varying static spatial fluctuation in the potential energy landscape and can arise due to charge dipole¹⁶ or charge quadrupole interactions.¹⁷ Another origin can be inhomogeneities in the electronic polarization energy,¹⁴ resulting from molecular density fluctuations in a material, due to microscopic regions that are under compression or dilation. The consideration of the energy correlation effects was proven to be important, especially for an adequate description of electric field dependence of the charge carrier mobility, at least in the *low carrier concentration* limit.

Although a field dependence of the charge carrier mobility is expected also in the *high carrier concentration* transport regime, e.g., for the OFET mobility, the reports of such a behavior in literature are rare^{18–20} and systematic studies of the OFET mobility upon the lateral source-drain electric field are still missing. Numerical simulations of the charge carrier mobility in an energetically noncorrelated hopping transport system by Pasveer *et al.*⁶ and also an analytical theory,²¹ based on the EGDM approach and the Miller–Abrahams jump rate, have demonstrated a similarly strong increase of the charge carrier mobility with increasing both the carrier concentration and electric field. Nonetheless, the field dependence of the charge carrier mobility at large carrier densities was not in the spotlight of research as it has been a widespread opinion⁶ that at room temperature it is mainly the dependence on carrier concentration that plays an important role, whereas the electric field dependence could become important only at (i) low temperatures and (ii) at high electric fields. The lateral electric field is conventionally believed to be very low in OFETs due to relatively large channel length in OFETs. This is probably one reason why researchers paid little attention to this issue and why the effect of including the energy correlations for the description of charge transport at high carrier concentrations was addressed just recently. Using numerical simulations Novikov²² and Bouhassoune *et al.*²³ suggested a so-called extended CDM (ECDM).^{22,23} This model has been applied to describe the charge carrier mobility measured in an organic sandwich-type diode device.^{23–25} Note that the electric field in such diode devices is normally much larger than the lateral electric field in an OFET due to very different interelectrode distances.

In this paper we suggest an analytical effective medium theory formulated for arbitrary electric fields using the method of configurational average into energy and hopping transition time. To consider rigorously the electric field dependence of the temperature-dependent hopping mobility at high carrier concentrations, spatial energy correlations are taken into account. Another focus of our present study is the influence of the electric field on the so-called Meyer–Neldel (MN) compensation rule.²⁶ This is typically observed for the temperature dependence of the charge carrier mobility in a high carrier concentration regime.^{27–29} In context of the charge transport in OFETs, the MN rule suggests a specific empirical relation between the Arrhenius activation energy E_a and the mobility prefactor. More specifically, it implies that Arrhenius-type $\mu(1/T)$ dependences, measured at different gate voltage and, concomitantly, at different charge carrier densities, intersect at a given finite temperature T_0 . Our recent theoretical treatment²⁸ based on the EGDM approximation has

demonstrated that the MN effect is recovered regarding the temperature dependences of the charge carrier mobility upon *varying the carrier concentration* but not regarding varying the width of the DOS. The latter actually implies that this MN effect is apparent rather than genuine. Note that the real MN effect predicts a correlation between the mobility prefactor and the activation energy regardless of how the change in E_a is accomplished.²⁶ Nonetheless, we have demonstrated^{28,29} that the operationally applied MN energy is a measure of the energetic disorder in organic films and could be used as a useful sensitive testing parameter characterizing the quality of active organic semiconductor layers in OFETs.

Since our previous treatment²⁸ was limited to zero-electric field, we now extended it for arbitrary electric fields to study simultaneously the effect of the carrier concentration and the electric field on the MN phenomenon for the OFET mobility. The latter is seemingly not an unexpected phenomenon, because the carrier concentration and electric field should have a rather similar impact on the magnitude of the charge carrier mobility in the high carrier concentration transport regime. It is worth noting that the effect of electric field on temperature dependences of charge carrier mobility has already been puzzling since the early 1970s and historically was first described by the phenomenological Gill³⁰ equation

$$\mu(F, T) = \mu_0 \exp\left(-\frac{E_a - \beta\sqrt{F}}{k_B T_{\text{eff}}}\right), \quad (1)$$

$$\frac{1}{T_{\text{eff}}} = \frac{1}{T} - \frac{1}{T^*},$$

where E_a is the zero-field activation energy, and β is a constant. The isokinetic temperature T^* is the so-called ‘‘Gill-temperature’’ at which extrapolations of the $\ln \mu$ vs $1/T$ lines intersect.

Note, that the Gill equation [Eq. (1)] is actually a particular case of the MN compensation rule²⁶ when the activation energy of the charge carrier mobility is changed by an applied electric field. Therefore, for the sake of convenience, we hereafter discriminate between the isokinetic temperature, i.e., the MN temperature that results from varying the charge carrier concentration on the one hand and the Gill temperature that results from varying the applied electric field on the other hand. Apart from the fact that Eq. (1) has no theoretical foundation, its application to experimental data analysis caused serious problems (see discussion in Refs. 11 and 31) For instance, it tacitly implies that transport is limited by traps that are charged when empty. Although the Gill equation is clearly inappropriate for the description of charge carrier mobilities at low carrier concentrations, we show in the present paper that an apparent Gill-type effect has been revealed by a hopping transport-based theory for the large carrier concentration transport regime.

The paper is organized as follows. In Sec. II we describe the effective medium approximation (EMA). We first consider the drift charge carrier hopping mobility for arbitrary electric fields and carrier concentrations in an energy noncorrelated organic random system. Then we extend our approach to account for the space-energy correlations in the distribution of localized states. In Sec. III the results of EMA calculations of the electric field dependence of the charge carrier mobility at

various carrier concentration and temperatures are presented, and the PF-type behavior is discussed for both an energy-correlated and noncorrelated disordered organic system. The results of EMA calculations are compared with available computer simulations and analytic theory data from literature in order to verify the adequateness of the suggested theoretical formalism. Then the major focus of our study is thorough consideration of the MN phenomena upon varying both the carrier concentration and the applied electric field for $\mu(T)$ dependences in the high carrier concentration transport regime. We show that the MN and Gill temperature can depend on electric field and carrier concentration, respectively; and that this is a consequence of spatial energy correlations in the material. Finally, we show that fitting of experimental data with the present theory allows estimating important material parameters as the width of the DOS distribution, characterizing the degree of energetic disorder. The implications of our findings are discussed in Sec. IV.

II. THEORETICAL FORMULATION

The present theoretical treatment is focused on the description of the electric field dependence of the charge carrier drift mobility in a disordered organic solid at different temperatures and carrier concentrations. It is based on the EMA using the concept of the effective transport energy. A key point of the present model compared to the previous analytic treatment^{7,28} is that it is extended for arbitrary electric fields and is able to describe consistently both the carrier concentration and field dependences of charge carrier mobility. Another important modification of our EMA formalism is that the present treatment based on method of the configurational averaging of hopping *transition times*. This method was suggested before for zero-field and low carrier concentration limits, according to Ref. 32, and now is generalized to calculate the drift mobility at finite electric fields for the high carrier concentration transport regime. For zero electric field this method gives the results basically identical to that obtained also by the average hopping *transition rate* approach. We found, however, that the former provides a better agreement with the relevant computer simulation data¹⁶ with respect to the charge mobility electric field dependences in the low carrier concentration regime. This might justify the superiority of this method for the calculation of the electric field dependence of charge carrier mobility especially in the case of high carrier concentrations.

A. General considerations

Within the EMA approach a disordered organic system is replaced by an effective three-dimensional (3D) manifold of localized sites with an average intersite distance $a = N^{-1/3}$, where N is the density of the localized states. We consider an energetically disordered system of localized states characterized by the DOS distribution $g(\varepsilon)$ in the framework of the extended GDM (EGDM),^{6,7} which accounts for the dependence of the mobility on the relative carrier concentration n/N , where n is the density of charge carriers. Positional disorder is neglected in this model description.

Let us consider a random 3D hopping transport system with an applied electric field. In general the effective drift hopping

mobility μ_e can be obtained as

$$\mu_e = ak_0 \frac{W_e^+ - W_e^-}{F}, \quad (2)$$

where W_e^+ and W_e^- describe the effective jump rates along and opposite to the electric field direction, respectively, for an arbitrary electric field $\mathbf{F} = \{F, 0, 0\}$. An additional coefficient k_0 emerges in Eq. (2) to include the generalized Einstein equation, as recently suggested by Roichman and Tessler,³³ relating the mobility and diffusion coefficient at arbitrary carrier concentration. This coefficient is essential at large carrier concentrations, whereas in the case of vanishing carrier concentration $k_0 \rightarrow 1$. Within the average hopping times method,³² the effective jump rates W_e^+ and W_e^- can be calculated using the effective transport energy ε_t level as given by

$$W_e^\pm = \langle \tau_{12}^\pm \rangle^{-1}, \quad (3)$$

$$\langle \tau_{12}^\pm \rangle = \frac{\int_{-\infty}^{\varepsilon_t} P(\varepsilon) \{W_{12}^\pm(\varepsilon_t, \varepsilon)\}^{-1} d\varepsilon}{\int_{-\infty}^{\varepsilon_t} P(\varepsilon) d\varepsilon},$$

where W_{12}^+ and W_{21}^- are effective jump rates between two neighboring localized sites along and opposite to the electric field direction, respectively. We use a Miller–Abrahams-type jump rate

$$W_{12}^\pm(\varepsilon_t, \varepsilon) = W_0 \exp \left[-\frac{|\varepsilon_t - \varepsilon \mp eaF| + (\varepsilon_t - \varepsilon \mp eaF)}{2k_B T} \right] \quad (4)$$

to describe an elementary charge transfer with an applied electric field between sites with energy ε and ε_t being defined in the limit of zero field. Here $W_0 = \nu_0 \exp(-2r_t/b)$, where ν_0 is the attempt-to-escape frequency, r_t is the jump distance below the effective transport energy ε_t , and b is the localization radius of the charged site.

Zero-field charge carrier drift under thermal equilibrium conditions occurs by carrier hopping transitions between ε and ε_t levels via successive hops downward in energy towards an empty state ε and then by upward hops to ε_t .³² As the downwards hops from the level ε_t occur much faster, the major contribution to the drift time in such paired jumps is determined by the upward hops to the state ε_t . Therefore, configurational averaging of the hopping transitions' times in Eq. (3) has to be done over the energy distribution of empty localized states, namely by using the function

$$P(\varepsilon) = g(\varepsilon) [1 - f(\varepsilon, \varepsilon_F)], \quad (5)$$

where $f(\varepsilon, \varepsilon_F)$ is given by the Fermi–Dirac statistics

$$f(\varepsilon, \varepsilon_F) = \frac{1}{1 + \exp\left(\frac{\varepsilon - \varepsilon_F}{k_B T}\right)}. \quad (6)$$

The Fermi level ε_F position can be determined from the following transcendental equation for the carrier

concentration n

$$n = \int_{-\infty}^{\infty} d\varepsilon g(\varepsilon) f(\varepsilon, \varepsilon_F). \quad (7)$$

The coefficient k_0 [cf. Eq. (2)] in this case can be determined as

$$k_0 = 1 - \frac{\int_{-\infty}^{\infty} d\varepsilon g(\varepsilon) f^2(\varepsilon, \varepsilon_F)}{\int_{-\infty}^{\infty} d\varepsilon g(\varepsilon) f(\varepsilon, \varepsilon_F)}. \quad (8)$$

In the present study we assume a Gaussian DOS distribution $g(\varepsilon)$ with the width σ as generally accepted to be appropriate for disordered organic media,

$$g(\varepsilon) = \frac{N}{\sigma\sqrt{2\pi}} \exp\left[-\frac{1}{2}\left(\frac{\varepsilon}{\sigma}\right)^2\right], \quad (9)$$

$$-\infty < \varepsilon < \infty.$$

Combining Eq. (2) with Eqs. (3)–(8) and (9), one can express the effective charge mobility μ_e as

$$\mu_e = \mu_0 k_0 r_t^2 \exp\left(-2\frac{r_t}{b}\right) \frac{(Y_e^+)^{-1} - (Y_e^-)^{-1}}{f}, \quad (10)$$

where

$$Y_e^\pm = \frac{\int_{-\infty}^{x_t} dt \frac{\exp\{-\frac{t^2}{2} + \frac{1}{2}[(x_t - t \mp f] + (x_t - t \mp f)x]\}}{1 + \exp[-(t - x_F)x]}}{\int_{-\infty}^{x_t} dt \frac{\exp\{-\frac{t^2}{2}\}}{1 + \exp[-(t - x_F)x]}}. \quad (11)$$

Here

$$Y_e^\pm = \frac{W_e^\pm}{W_0}, \quad f = \frac{eaF}{\sigma}, \quad x = \frac{\sigma}{k_B T}, \quad (12)$$

$$x_F = \frac{\varepsilon_F}{\sigma}, \quad x_t = \frac{\varepsilon_t}{\sigma}, \quad \mu_0 = \frac{ea^2 v_0}{\sigma}.$$

The effective transport energy ε_t in our method does not depend on the applied electric field F and hence for a Gaussian DOS distribution can be determined from the following transcendental equation derived for zero-field mobility:⁷

$$\frac{1}{\sqrt{2\pi}} \frac{\exp(-\frac{1}{2}x_t^2)}{1 + \exp[-(x_t - x_F)x]} \left\{ \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{x_t} dt \frac{\exp(-\frac{1}{2}t^2)}{1 + \exp[-(t - x_F)x]} \right\}^{-\frac{4}{3}} = \frac{3}{2} \left(\frac{4\pi}{3B}\right)^{\frac{1}{3}} x \frac{b}{a}. \quad (13)$$

Here, parameter $B = 2.7$ is being determined according to percolation criteria.³⁴ Factor r_t is calculated by

$$r_t = a \left\{ \frac{4\pi}{3B} \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{x_t} dt \frac{\exp(-\frac{1}{2}t^2)}{1 + \exp[-(t - x_F)x]} \right\}^{-\frac{1}{3}}. \quad (14)$$

It should be noted that the percolative effects have been considered in Ref. 34, not rigorously in that the consideration was limited to the renormalization solely of the factor r_t . Since r_t and ε_t are quantities interrelated by definition, the accounting for the influence of the percolative effects must be done for an effective transport energy ε_t as well. A combination of Eqs. (13) and (14) duly takes this into account.

B. Spatial energy correlations

The results of our analytical calculation by Eq. (10) are found to be in a very good agreement with the computer simulations data by Pasveer *et al.*⁶ obtained within the EGDM approximation for temperature, field, and carrier concentration dependences of the charge carrier mobility in a random (noncorrelated) organic system. The EGDM-based calculations describe well also the experimentally observed temperature and carrier concentration dependences of the mobility. However, this model predicts a specific $\ln \mu \propto F$ field dependence,⁶ whereas numerous experimental data typically feature a $\ln \mu \propto \sqrt{F}$ PF-type behavior in an experimentally accessible range of electric fields. On the other hand, it is well established that a PF-type field dependence of the charge carrier mobility can be reproduced over an

extended interval of electric fields only when some kind of correlated disorder is taken into account in the framework of the CDM¹⁶ or the ECDM.^{22,23,25} Conventional noncorrelated disorder models ignoring the energy correlation effects, i.e., assuming that site energies are distributed independently, predict the PF law only over a very narrow field interval at high fields ($> 3 \times 10^5$ V/cm). Therefore, accounting for the energy correlation effects is necessary for an adequate description of the PF mobility field dependence being experimentally observed down to moderate fields. We should emphasize that this aspect should be especially relevant in the case of the OFET mobility measurements, because the latter is typically measured at rather low lateral electric fields owing to typically large transistor channel length used in OFETs devices if compared to other organic electronic devices as, for example, sandwich-type diodes.

For this reason we will use the concept of energy correlations. Generally, accounting for the energy-correlated disorder effects is a rather complicated task and could hardly be treated strictly analytically, although it might be solved by numerical computer simulations.^{16,23} We therefore will use here in our analytical treatment some key ingredients from the numerical simulations studies performed before within the CDM^{16,23} for a low carrier density limit. They show^{16,23} that the size of an “energetic valley” (cluster) comprising many localized sites, scales algebraically with the depth of fluctuations. As we suggested before,³⁵ by using the simulations results of Parris *et al.*,¹⁵ one could account for the energy correlation effects by substituting the parameters $x = \sigma/k_B T$ and $f = eaF/\sigma$ (for noncorrelated disordered system) by $x_c = \sigma_c/k_B T$ and $f_c = h_c \sqrt{x_c}/2$, respectively, where $h_c = \sqrt{eaF/\sigma_c}$. It is

worth mentioning that such a substitution yields a coefficient $1/\sqrt{2} \cong 0.71$ in the slope of the electric field dependence of the charge carrier mobility. It agrees reasonably well with the relevant coefficient 0.78 in the approximated expression obtained by numerical simulation studies within the CDM¹⁶ (see, for details, Ref. 35).

It is well known that the presence of energy correlations results in some flattening of the local potential energy landscape within larger scale energetic structures, therefore the width of the so-called correlated DOS, σ_c , has to be somewhat smaller than the initial σ value featured in the noncorrelated GDM approximation. Coehoorn and coworkers²³ have shown that one can use $\sigma_c \cong 0.83\sigma$ due to the charge mobility under the presence of the energy correlations in the limit of zero field, and zero carrier density follows a $\mu_e \propto \exp(-0.29x^2)$ temperature dependence instead of that $\mu_e \propto \exp(-0.44x^2)$ inherent for the GDM.

Another important consequence of the energy correlation effects on the field dependence of the charge carrier mobility $\mu(F)$ at *high* carrier concentrations has been recently demonstrated by Bouhassoune *et al.*²³ and Novikov.²² Based on their computer simulations, they found that the typical size of the relevant energy valley, and hence the jump length, governing the charge carrier mobility depends not only on electric field *but also on carrier concentration*. The typical jump length within ECDM was found to decrease with increasing carrier concentration. This leads to a decrease of the slope S of the field mobility dependence $\ln \mu$ vs $S \times \sqrt{F}$ compared to the slope S_0 of the corresponding curve in the case of vanishing carrier concentration $n/N \rightarrow 0$, provided that the energy correlations have been taken into account. For a noncorrelated disordered system within the EGDM approach the slope remains the same upon varying carrier concentration.²² This effect can hardly be treated analytically. Therefore in the present study we use the relevant result from Ref. 23 as follows: One can express $S = yS_0$, where $y = (1 - n/N)^m$ with exponent m being temperature dependent. In limiting cases of small and large carrier concentrations one obtains $y \rightarrow 1$ and $y \rightarrow 0$, respectively. This agrees qualitatively with the simulation results. Further, we can use S values obtained in Ref. 23 for $n/N = 10^{-5}$ and $n/N = 0.05$ at a certain $\sigma/k_B T$ to formulate a system of two transcendental equations to obtain S_0 and m quantities for given $\sigma/k_B T$. Making similar calculations for different $\sigma/k_B T$ values, one can obtain the following expressions for the temperature dependence of S_0 and m : $S_0 = -5.42 + 2.995(\sigma/k_B T) - 0.115(\sigma/k_B T)^2$ and $m = -9.69 + 10.81(\sigma/k_B T) - 0.905(\sigma/k_B T)^2$. In the range $2.0 < \sigma/k_B T < 5.0$ the calculated parameter S_0 perfectly agrees with the relevant slope $S_1 = 0.78[(\sigma/k_B T)^{3/2} - 2]$ obtained by computer simulations¹⁶ for the small carrier concentration limit.

Coulombic interactions between charge carriers might start to play an essential role at large carrier concentrations,^{22,36} which is not accounted for in the present theoretical consideration. The recent Monte Carlo simulations by Zhou *et al.*³⁶ has, however, demonstrated that the effect of Coulomb interactions is not significant if the carrier density is below $n/N < 10^{-2}$. Since the computer simulations by Novikov²² that include Coulomb interactions show that for carrier

concentrations as large as $n/N \leq 10^{-1}$ the charge mobility increases with increasing n/N exactly in the same manner as in the case of noninteracting carriers,⁶ we henceforth disregard the Coulombic interactions.

In the present treatment we also ignore the influence of local shifts of the Fermi level (electrochemical potential) caused by the electric field and the energy disorder.^{37,38} Such an effect could be accounted for only by the numerical computer simulations. Nonetheless, as it was recently demonstrated,²¹ the neglecting of this aspect does not lead to any notable difference regarding both field dependences and charge carrier mobility magnitude from those obtained by computer simulations studies⁶ for low and high carrier densities. The observed good agreement between analytical calculations and numerical simulations confirms the validity of the approximation made in the context of the EMA treatment.²¹

Thus, for calculation of the field, temperature, and carrier concentration dependences of the charge carrier mobility in disordered organic semiconductors, we will hereafter use Eqs. (10)–(14) obtained within EGDM approximation with accounting for energy correlation effects by substituting the parameters σ , f by σ_c , $f_c y$, respectively, as described previously.

III. RESULTS OF CALCULATIONS

A. Field dependence of OFET mobility.

Let us start with a consideration of the electric field dependence of the charge carrier mobility $\mu(F)$ within the model suggested previously. Figure 1(a) presents the electric field dependence in $\ln \mu \propto \sqrt{E}$ representation for an energy correlated organic disordered system. The curves (solid lines) were calculated by Eqs. (10)–(14) for different carrier concentrations relevant to OFET operation, taking into account the dependence of the jump length on carrier concentration according to Ref. 23. The same dependences were also calculated by ignoring the latter effect and are given for comparison [Fig. 1(a), dashed curves]. As one can see, the PF field dependences are well reproducible within the range $0.4 < (eaF/\sigma)^{1/2} < 1.0$ for the considered charge carrier concentrations and feature a clear decrease in the slope of the curves with increasing carrier concentration [Fig. 1(a), solid curves], whereas the slope remains virtually the same if the carrier concentration dependence of the jump length is ignored [dashed curves in Fig. 1(a)]. These analytically calculated dependences agree well with the relevant computer simulation results of Ref. 22. Assuming representative material parameters for a disordered solid, viz. $\sigma = 0.07$ eV and $a = 1.4$ nm, one obtains a corresponding electric field range $8 \times 10^4 \text{V/cm} < F < 5 \times 10^6 \text{V/cm}$ for which the PF-type field dependence is valid.

Figure 1(b) shows the electric field dependences of the charge carrier mobility, calculated at the same parameters as in Fig. 1(a), but for an energy noncorrelated organic disordered system. As one can see from Fig. 1(b), a $\ln \mu \propto \sqrt{E}$ PF dependence is revealed for such a system but within a narrower range of strong electric fields only, consistent with previous computer simulations.⁴ Note that in the present study we used a representative ratio $a/b = 5$ for organic disordered

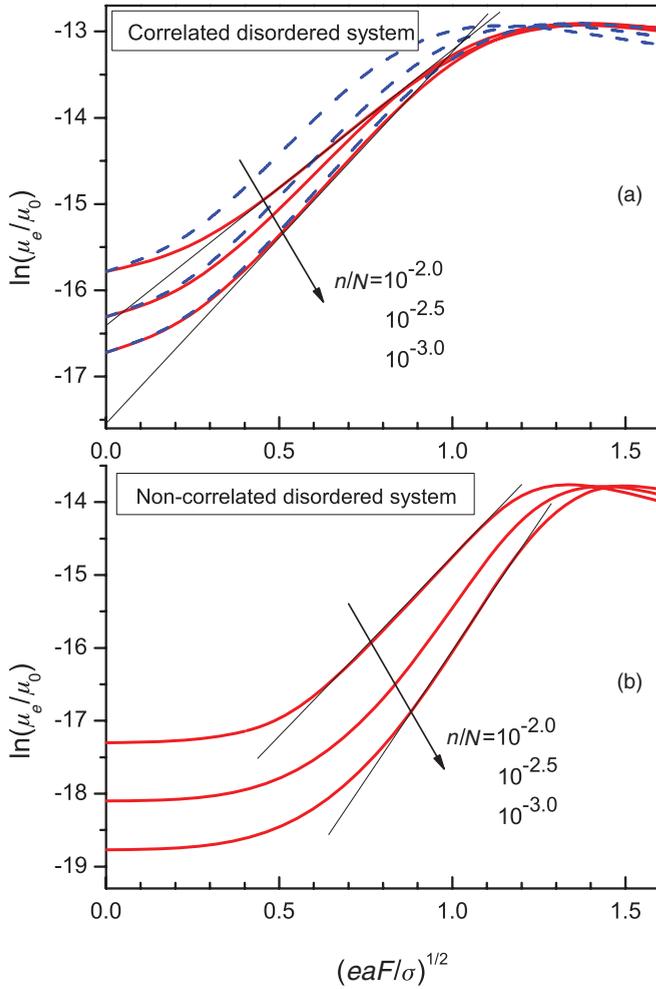


FIG. 1. (Color online) (a) Poole-Frenkel plots of electric field dependences of the effective charge carrier mobility $\ln(\mu_e/\mu_0)$ calculated by Eqs. (10)–(14) for an energy-correlated organic disordered system for several carrier concentrations ($n/N = 10^{-3}$, $10^{-2.5}$, and 10^{-2}) at $a/b = 5$ and $\sigma/k_B T = 5$ with accounting (see text) for the carrier concentration dependence of the jump length (solid curves) and ignoring the latter effect (dashed curves); (b) field dependences calculated for a noncorrelated disordered system are given for comparison.

semiconductors, which has also extensively been employed before in Monte Carlo simulations’ studies of the charge transport.⁴

The electric field dependences of the charge carrier mobility calculated by Eqs. (10)–(14) for different temperatures at a moderate carrier density, $n/N = 10^{-5}$, and a very high carrier density, $n/N = 10^{-1.5}$, are shown in Fig. 2. As one can see, a $\ln \mu \propto \sqrt{E}$ dependence of the charge carrier mobility has been revealed at large and moderate carrier concentrations. It is basically similar to the PF-type field dependences conventionally observed in the limit of low carrier concentrations, i.e., in ToF mobility measurements.

The results of the calculations, described previously, within the ECDM using Eqs. (10)–(14) with accounting for energy correlations, indicate that (i) the PF type of field dependence can be reproduced by the present model within a rather broad range of electric fields, and (ii) the PF mobility

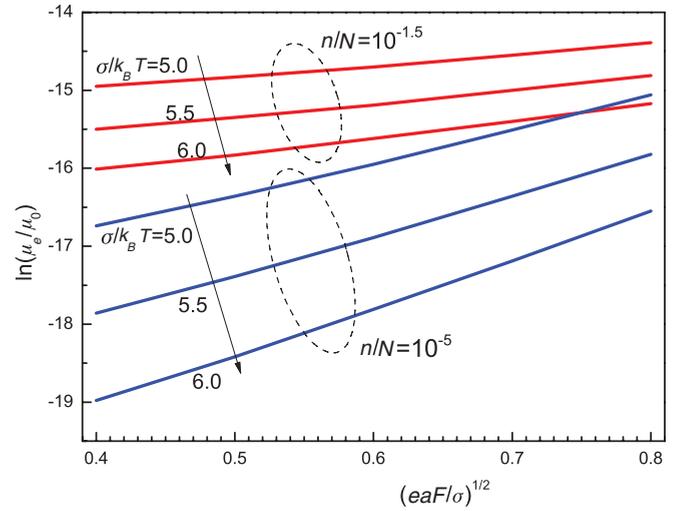


FIG. 2. (Color online) Poole-Frenkel plots of electric field dependences of the effective charge carrier mobility $\ln(\mu_e/\mu_0)$ in an organic disordered system calculated by Eqs. (10)–(14) at different temperatures with accounting for energy correlations for two carrier concentrations: $n/N = 10^{-1.5}$ (upper branch) and 10^{-5} (lower branch) $a/b = 5$.

field dependence becomes notably weaker at higher carrier concentrations.

B. MN effect upon varying the charge carrier concentration

1. Small carrier concentrations

Charge carrier mobility at low carrier concentration is conventionally measured by the ToF technique, which has been applied extensively to study charge transport in disordered organic solids, for instance, in vapor-deposited molecular glasses.^{4,11} Small charge carrier concentration is required in the ToF method to avoid any space charge inside the sample that may distort the field and concomitantly, the ToF signal. To limit space charge effects it has become practice to limit the number of migrating carriers to 5% of the capacitor charge, i.e., ca. 10^{10} charge carriers/cm² in an electric field of 10^5 V/cm. This is equivalent to a concentration of $n = 10^{13}$ cm⁻³ in a $10\text{-}\mu\text{m}$ -thick sample. Taking $N \approx 10^{22}$ cm⁻³ as a representative value for molecular glasses, one gets a relative carrier concentration of $n/N \approx 10^{-9}$. At such a carrier concentration the charge carrier mobility is independent on carrier densities because the Boltzmann statistic dominates the hopping transport; that is the reason why it is called the “small carrier concentration limit.”

Figure 3(a) presents the temperature dependences of the charge carrier mobility calculated by Eqs. (10)–(14) at $n/N = 10^{-9}$ for different electric fields. As one could expect, the temperature dependences are perfect straight lines in $\ln(\mu) \propto T^{-2}$ representation, in agreement with a number of previous theoretical and computer simulation data as well as with ToF experiments.^{4,5,11} The temperature dependences intersect at infinite temperature, as suggested by BäSSLER’s model,⁴ and no MN rule (or Gill-type) behavior is observed. If one replots these data in simple Arrhenius coordinates $\ln(\mu) \propto T^{-1}$ then, as one can see from Fig. 3(b), (i) the calculated

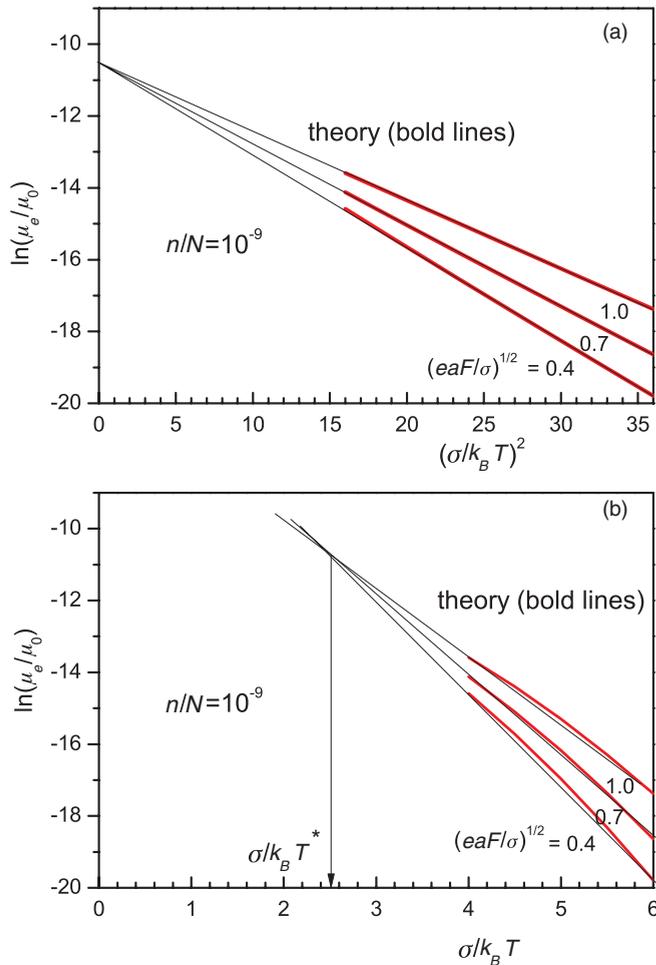


FIG. 3. (Color online) (a) Temperature dependences of the charge carrier mobility $\ln(\mu_e) \propto 1/T^2$ calculated by Eqs. (10)–(14) for an energy correlated system at low carrier concentration $n/N = 10^{-9}$ parametric in electric fields and at $a/b = 5$; (b) the same data replotted in Arrhenius ($\ln(\mu_e) \propto 1/T$) representation.

T dependences are no longer perfect straight lines, and (ii) their formal extrapolation to higher temperatures shows an apparent intersect at some intermediate temperature T^* . It is clear that the latter is a deceptive effect due to improper $\ln(\mu) \propto T^{-1}$ representation of the data, which in fact feature functionally different temperature dependence. Thus, as it was already stated long ago,⁴ the Gill relation is not appropriate for the description of the ToF mobility data.

2. Large carrier concentrations

The temperature dependence of the charge carrier mobility at large carrier densities differs from that in the low concentration limit: (i) It does obey an Arrhenius-type dependence, as discussed already in literature,^{8,28} and (ii) while in the low carrier density limit, the slope of $\mu(T)$ depends on the electric field only, that is, it depends on both the electric field and the carrier concentration at high carrier concentrations.

Figure 4 (bold curves) depicts the temperature dependences of the charge carrier mobility calculated by Eqs. (10)–(14) at a *finite* constant electric field taking into account the energy correlations for several large carrier concentrations relevant for

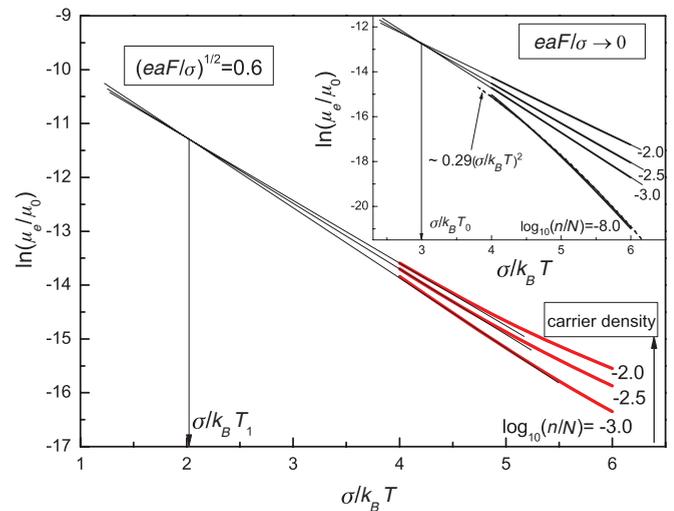


FIG. 4. (Color online) Effective charge carrier mobility $\ln(\mu_e/\mu_0)$ vs $\sigma/k_B T$ calculated by Eqs. (10)–(14) at finite electric field ($(eaF/\sigma)^{1/2} = 0.6$) for different effective carrier concentrations (bold curves) with accounting for energy correlations and $a/b = 5$. Inset: $\ln(\mu_e/\mu_0)$ vs $\sigma/k_B T$ calculated at vanishing electric field ($F \rightarrow 0$) for different effective carrier concentrations (bold curves) with accounting for energy correlations. Fitting the lower curve by $\ln(\mu_e) \propto -0.29(\sigma/k_B T)^2$ is given by the dashed curve (see text for details).

OFET operation The calculated curves are virtually straight lines in Arrhenius representation and their asymptotes, due to an extension to higher temperatures (given by thin lines in Fig. 4), intersect at some finite temperature T_1 featuring, thus, a MN-type behavior.

Temperature dependences of the charge carrier mobility for vanishing electric field ($F \rightarrow 0$) calculated by Eqs. (10)–(14) taking into account the energy correlations at different carrier concentrations are shown in the inset in Fig. 4 for comparison. As one can see, the asymptotes to the calculated dependences at high carrier concentrations ($n/N = 10^{-3} \dots 10^{-2}$) can be linearly extrapolated to higher temperatures and intersect at finite temperature T_0 , demonstrating thus a MN effect. These results agree well with our previous calculations of the temperature-dependent charge carrier mobility²⁸ parametric in carrier concentration using an EMA theory developed *solely* for zero-electric field and for a noncorrelated energy disordered system. This verifies that the present EMA model at vanishing electric field provides virtually the same results regarding the MN rule for $\mu(T)$ upon varying the charge carrier concentration as the model described before.²⁸ It is of interest that the theoretical curve calculated at very low carrier concentration $n/N = 10^{-8}$ can perfectly be fitted by the relation $\ln(\mu_e) \propto -C(\sigma/k_B T)^2$ (inset in Fig. 4, dashed curve) with the parameter $C = 0.29$. This parameter has been obtained beforehand from computer simulations of the charge mobility in an energy-correlated hopping system.¹⁵ This parameter differs from $C = 0.44$, inherent for the uncorrelated GDM.⁴ This supports the adequateness of the present analytical theory accounting for the energy correlation effects. Thus, accounting for energy correlations does result in a notably weaker temperature dependence of the charge

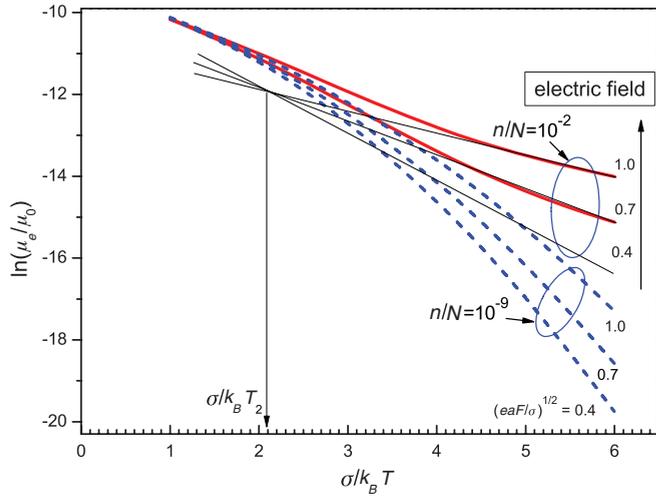


FIG. 5. (Color online) Effective charge carrier mobility $\ln(\mu_e/\mu_0)$ vs $\sigma/k_B T$ calculated by Eqs. (10)–(14) parametric in electric fields for two different carrier concentrations $n/N = 10^{-9}$ and 10^{-2} (dashed and bold curves, respectively) at $a/b = 5$ in an energy correlated hopping system. The thin straight lines show extrapolation of the calculated dependences at $n/N = 10^{-2}$ to higher temperatures.

carrier mobility as compared to that for noncorrelated systems, notably for the charge carrier mobility at vanishing electric field.

C. MN effect upon varying the electric field

Temperature dependences of the charge carrier mobility parametric in electric fields calculated by Eqs. (10)–(14) are plotted in Fig. 5 for two different carrier concentrations: $n/N = 10^{-9}$ (dashed curves, lower branch) and 10^{-2} (bold curves, upper branch). These dependences were calculated over a broad temperature range up to very high temperatures not accessible for experiments. As one can see from Fig. 5, at large carrier concentration asymptotes to the calculated $\ln(\mu) \propto T^{-1}$ dependences extrapolated to higher temperatures (thin straight lines) intersect at finite temperature T_2 implying that the MN- (or Gill-)type behavior is reproduced upon varying the electric field over a range of moderate temperatures. No Gill-type behavior occurs for low carrier concentration ($n/N = 10^{-9}$; Fig. 5), because the charge carrier mobility in this regime does not follow $\ln(\mu) \propto T^{-1}$ dependence, as discussed in Sec. III B 1.

At very high temperatures the calculated charge carrier mobilities merge into the same value independent of electric field and carrier concentration (Fig. 5). This means that, according to the present treatment, there is no finite critical temperature above which the mobility would feature a negative field dependence as it follows from the empirical Gill equation [Eq. (1)]. The reason for such a temperature dependence is that at higher temperatures the average energy ε_m of the equilibrium occupational DOS (ODOS), which derives from $\varepsilon_m = \int_{-\infty}^{\infty} d\varepsilon \varepsilon g(\varepsilon) n(\varepsilon, \varepsilon_F) / \int_{-\infty}^{\infty} d\varepsilon g(\varepsilon) n(\varepsilon, \varepsilon_F)$, is no longer approached to the T -independent Fermi level ε_F but is determined by the T -dependent shift of $\varepsilon_m \rightarrow \varepsilon_0 = -\sigma^2/k_B T$ towards the transport energy²⁸ and, concomitantly, towards the center of the DOS.

The electric field dependence of the charge carrier mobility is consistent with the reasoning described previously. Under the applied field the average equilibrium energy ε_m increases and, as a consequence, the charge carrier mobility determined by jumps from ε_m to ε_t should also increase with increasing electric field. At high carrier concentrations the EGDM formalism predicts a lowering of the barrier height for carrier jumps because ε_m increases not only as a result of an increase of the carrier density but also due to an increase of the applied electric field (lateral field in the case of an OFET).

D. Effect of electric field and carrier concentration on MN energy and Gill energy

As mentioned in the Introduction, for the sake of convenience we discriminate between the isokinetic temperatures resulting from intersection of $\ln(\mu) \propto T^{-1}$ upon varying carrier concentration on the one hand and that upon varying applied electric field on the other hand. The former is referred to as the MN temperature (T_1); the latter is termed Gill temperature (T_2). According to the results presented in Figs. 4 and 5, the MN temperature (denoted as T_1) and Gill temperature (denoted as T_2) are not constant but depend on the electric field F and carrier concentration n/N , respectively. This effect indeed can be well reproduced within the present EMA model provided that the carrier concentration dependence of the jump length is taken into account according to Ref. 23, as described in Sec. II, and which is responsible for changing the slope of the field dependences of the OFET mobility with increasing carrier concentration shown in Fig. 1. Figure 6(a) (solid curve 1) shows the MN temperature T_1 vs applied electric field calculated within the present model. Apparently, the MN temperature shifts to lower values with increasing F . The field dependence of T_1 can be parameterized as follows,

$$\frac{k_B T_1}{\sigma} = 0.5 + 0.029 \frac{eaF}{\sigma} - 0.039 \left(\frac{eaF}{\sigma} \right)^2. \quad (15)$$

If the carrier concentration dependence of the jump length is ignored (cf. dashed lines in Fig. 1 calculated at $y = 1$) the $k_B T_1/\sigma$ quantity demonstrates just a very weak field dependence, as shown by dashed curve 2 in Fig. 6(a). Virtually no field dependence was found for $k_B T_1/\sigma$ calculated for an energy noncorrelated disordered system and $k_B T_1/\sigma \cong 0.42$ in such case. Thus, we conclude that the change of the MN temperature T_1 upon applied electric field results from the presence of energy correlation effects, namely due to the decrease of the typical jump length with increasing carrier concentration.

In line with that, the Gill temperature T_2 tends to decrease with increasing carrier concentration n/N [Fig. 6(b), solid curve 1]. This carrier density dependence of T_2 can be well approximated by

$$\frac{k_B T_2}{\sigma} = 0.416 - 0.037 \log_{10} \left(\frac{n}{N} \right) - 0.0031 \left[\log_{10} \left(\frac{n}{N} \right) \right]^2. \quad (16)$$

We found that $k_B T_2/\sigma$ shows very weak concentration dependence [dashed curve 2 in Fig. 6(b)] if the carrier concentration dependence of the typical jump length is

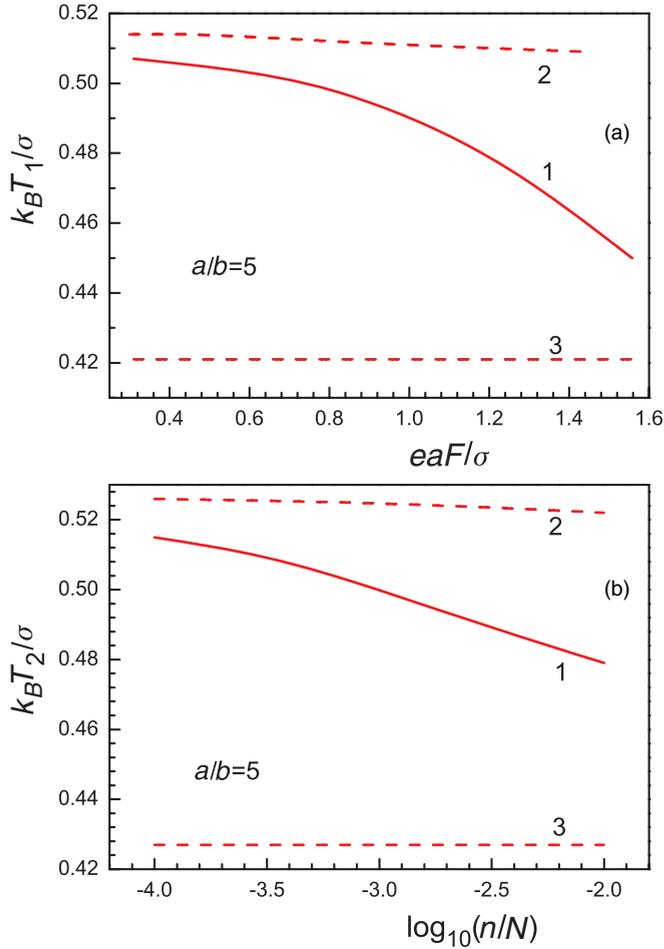


FIG. 6. (Color online) Calculated dependence of MN temperature T_1 (a) on electric field (solid curve 1) and the dependence of Gill temperature T_2 on carrier concentration (b) for an energy correlated disordered hopping system (solid curve 1). The same dependences calculated upon ignoring the carrier concentration dependence of the jump length are shown by dashed curves 2, and upon ignoring any correlation effects are shown by dashed curves 3.

ignored. If energy correlations are absent, the Gill temperature is virtually independent on carrier concentration and reaches the constant value $k_B T_2 / \sigma \cong 0.43$ [Fig. 6(b), dashed curve 3]. Both $k_B T_1 / \sigma \cong 0.42$ and $k_B T_2 / \sigma \cong 0.43$ values are slightly smaller than that obtained for an energy correlated system because the former ones ignore the decrease of σ due to presence of the energy correlations. It should be pointed out that the above calculated dependences for T_1 and T_2 are only relevant to the range of electric fields where the PF-type field dependence holds.

The results of the above calculated charge carrier mobility in the high carrier concentration limit can be used to estimate the energetic disorder parameter σ from experimental data basically by two different methods.

(1) The experimentally measured MN temperature T_1 ($E_{MN} = k_B T_1$) at a given electric field F (within the field interval where a PF-type dependence is obeyed) is inserted into Eq. (15). It results in a quadratic equation to calculate the

parameter σ . The solution reads

$$\sigma = E_{MN} A \left(1 + \sqrt{1 + \left(\frac{eaF}{E_{MN}} \right)^2 \frac{0.078}{A^2}} \right), \quad (17)$$

$$A = 1 - 0.029 \frac{eaF}{E_{MN}}.$$

It should be noted that the present extended theoretical model yields $E_{MN}/\sigma \cong 0.33$ for zero-field mobility. Previous theoretical treatment limited to the zero-field case,²⁸ which disregarded the energy correlations and percolation effects, yielded a somewhat different ratio, $E_{MN}/\sigma \cong 0.40$.

(2) The experimentally measured Gill temperature T_2 ($E_G = k_B T_2$) at a given high carrier concentration n/N [being determined by a gate voltage (V_G) in an OFET] is substituted into Eq. (16) and yields the following relation for calculating σ :

$$\sigma = \frac{E_G}{0.416 - 0.037 \log_{10} \left(\frac{n}{N} \right) - 0.0031 \left[\log_{10} \left(\frac{n}{N} \right) \right]^2}. \quad (18)$$

To use Eq. (18), one has, however, to know the effective carrier concentration in a thin conductive channel of an OFET, which demonstrates a highly nonuniform distribution—strongly decreasing from the semiconductor/insulator interface into the bulk.³⁹

Thus, the first of the aforementioned methods seems to be more appropriate for estimating the disorder parameter σ from experimental data. A big advantage of the present theoretical model is that it does not require an extrapolation of experimental data to zero electric field. It allows fitting of experimental data, obtained at a given (not too low) electric field, in order to evaluate material parameters of organic semiconductors.

Finally we should note that the above consideration suggests that the MN and Gill energies are inherently interrelated in disordered organic semiconductors. Indeed, combining Eqs. (15) and (16) one obtains

$$\frac{E_G}{E_{MN}} \equiv \frac{T_1}{T_2} = \frac{0.416 - 0.037z - 0.0031z^2}{0.5 + 0.029h - 0.039h^2}, \quad (19)$$

where $z = \log_{10}(n/N)$ and $h = eaF/\sigma$ for $-4 \leq z \leq -2$ and $0.3 \leq h \leq 1.6$. Figure 7 presents a 3D plot of the ratio E_G/E_{MN} upon carrier concentrations and applied electric fields calculated by Eq. (19).

IV. CONCLUSION AND FINAL REMARKS

An analytical EMA theory was formulated to describe the effect of electric field on the charge carrier mobility at high carrier concentrations in disordered organic semiconductors at large carrier concentrations taking into account energy correlation effects. The results of theoretical calculations are found to be in good agreement with relevant computer simulations on the PF-type field dependence of the mobility observed in a certain range of electric fields at different temperatures and carrier concentrations. The principal results of this study are the following.

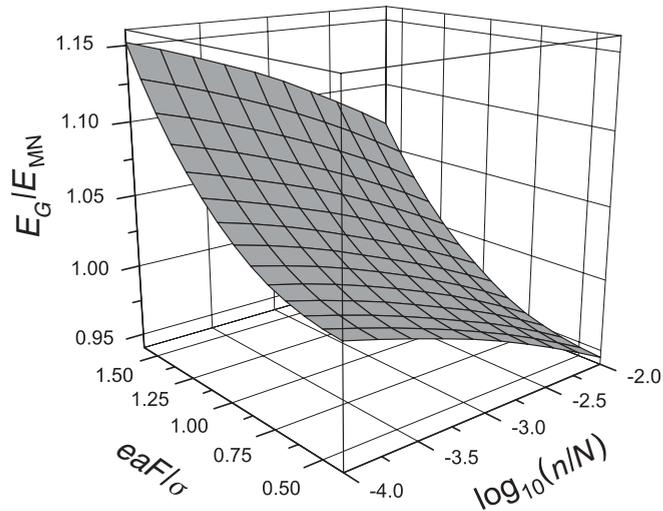


FIG. 7. Ratio of E_G/E_{MN} calculated by Eq. (19) for different electric fields and carrier concentrations.

(i) The present theoretical formalism reproduces the MN compensation rule for the temperature-dependent charge mobility upon *changing the electric field* provided that the carrier concentration is large enough. This is reminiscent of the prediction of the empirical Gill relation [Eq. (1)], and we show that the Gill-type dependence can be used at large carrier densities, as realized in conductive channels of OFETs devices, while it is not applicable for ToF mobilities when carrier concentrations are very low.

(ii) The EMA theory predicts that the MN temperature (T_1) and Gill temperature (T_2) are not constant but depend on the electric field F and carrier concentration n/N , respectively. This turns out to be a consequence of spatial energy correlations in organic disordered materials. We show that both above temperatures can be used for estimating the energy disorder parameter—i.e., the width of the DOS distribution. The present model allows more accurate evaluation of this important material parameter from experimental data measured at any electric field and does not require an extrapolation of experimental data to the zero-electric field.

Finally we should note that a quantitative description of OFET mobility in organic semiconductor film with *strongly*

inhomogeneous morphology could additionally require for accounting for an inhomogeneous lateral electric field along the conductive channel. As we showed recently,⁴⁰ the OFET mobility in multiple-grain channels can be controlled not by the lateral field averaged over the transistor channel (as conventionally assumed) but rather by the much stronger effective local electric fields generated in such inhomogeneous media. The concept of strong local electric fields has been recently proven experimentally for a polycrystalline silylethynyl-substituted pentacene by performing combined charge transport and scanning Kelvin probe microscopy (SKPM) studies on OFET devices based on films from the same organic semiconductor but with and without grain boundaries.⁴⁰ The SKPM measurements were done under device operation and have clearly revealed strong voltage drops exactly at grain boundaries, meaning that the local electric field in these boundaries is high. At the same time the electric field was found to be homogenous along the transistor channel containing a single crystallite/grain without a grain boundary. As we showed in Ref. 40, the lateral field dependence of the OFET mobility in such materials could be quantitatively described well provided that just the local fields are used in Eqs. (10)–(14) instead of the average one. Since the actual ratio between local field at the grain boundaries and the averaged field is not amenable to analytical treatment, one can use a phenomenological field magnification parameter $q \gg 1$ as a fitting parameter. Evidently that employment of q parameter just results in renormalization of the electric field F used in our calculations. In our farther work we plan to investigate an application of our model for description of the field-dependent OFET mobility recently observed also in C60-based OFETs.^{41,42}

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¹C. D. Dimitrakopoulos, S. Purushotaman, J. Kymissis, A. Callegari, and J. M. Shaw, *Science* **283**, 822 (1999).

²C. J. Brabec, N. S. Sariciftci, and J. C. Hummelen, *Adv. Funct. Mater.* **11**, 15 (2001).

³R. Richert, L. Pautmeier, and H. Bässler, *Phys. Rev. Lett.* **63**, 547 (1989).

⁴H. Bässler, *Phys. Status Solidi B* **175**, 15 (1993).

⁵V. I. Arkhipov, I. I. Fishchuk, A. Kadashchuk, and H. Bässler, in *Semiconducting Polymers: Chemistry, Physics and Engineering*, 2nd ed., edited by G. Hadziioannou and G. Malliaras (Wiley-VCH Verlag, Weinheim, 2007).

⁶W. F. Pasveer, J. Cottaar, C. Tanase, R. Coehoorn, P. A. Bobbert, P. W. M. Blom, D. M. de Leeuw, and M. A. J. Michels, *Phys. Rev. Lett.* **94**, 206601 (2005).

⁷I. I. Fishchuk, V. I. Arkhipov, A. Kadashchuk, P. Heremans, and H. Bässler, *Phys. Rev. B* **76**, 045210 (2007).

⁸N. I. Craciun, J. Wildeman, and P. W. M. Blom, *Phys. Rev. Lett.* **100**, 056601 (2008).

⁹R. Coehoorn, W. F. Pasveer, P. A. Bobbert, and M. A. J. Michels, *Phys. Rev. B* **72**, 155206 (2005).

¹⁰V. I. Arkhipov, P. Heremans, E. V. Emelianova, G. J. Adriaenssens, and H. Bässler, *Appl. Phys. Lett.* **82**, 3245 (2003).

- ¹¹P. M. Borsenberger and D. S. Weiss, *Organic Photoreceptors for Xerography* (Dekker, New York, 1998).
- ¹²A. Miller and E. Abrahams, *Phys. Rev.* **120**, 745 (1960).
- ¹³Y. N. Gartstein and E. M. Conwell, *Phys. Rev. B* **51**, 6947 (1995).
- ¹⁴S. V. Rakhmanova and E. M. Conwell, *Appl. Phys. Lett.* **76**, 3822 (2000).
- ¹⁵P. E. Parris, D. H. Dunlap, and V. M. Kenkre, *Phys. Status Solidi B* **218**, 47 (2000).
- ¹⁶S. V. Novikov, D. H. Dunlap, V. M. Kenkre, P. E. Parris, and A. V. Vannikov, *Phys. Rev. Lett.* **81**, 4472 (1998).
- ¹⁷S. V. Novikov and A. V. Vannikov, *J. Phys. Chem.* **113**, 2532 (2009).
- ¹⁸B. H. Hamadani, C. A. Richter, D. J. Gundlach, R. J. Kline, I. McCulloch, and M. Heeney, *J. Appl. Phys.* **102**, 044503 (2007).
- ¹⁹L. Wang, D. Fine, D. Basu, and A. Dodabalapur, *J. Appl. Phys.* **101**, 054515 (2007).
- ²⁰T. Minari, T. Nemoto, and S. Isoda, *J. Appl. Phys.* **99**, 034506 (2006).
- ²¹I. I. Fishchuk, A. Kadashchuk, V. N. Poroshin, and H. Bässler, *Philos. Mag.* **90**, 1229 (2010).
- ²²S. V. Novikov, *Phys. Status Solidi C* **5**, 740 (2008).
- ²³M. Bouhassoune, S. L. M. van Mensfoort, P. A. Bobbert, and R. Coehoorn, *Organic Electronics* **10**, 437 (2009).
- ²⁴S. L. M. van Mensfoort, V. Shabro, R. J. de Vries, R. A. J. Janssen, and R. Coehoorn, *J. Appl. Phys.* **107**, 113710 (2010).
- ²⁵J. C. Blakesley, H. S. Clubb, and N. C. Greenham, *Phys. Rev. B* **81**, 045210 (2010).
- ²⁶W. Meyer and H. Neldel, *Z. Tech. Phys.* **18**, 588 (1937).
- ²⁷E. J. Meijer, M. Matters, P. T. Herwig, D. M. de Leeuw, and T. M. Klapwijk, *Appl. Phys. Lett.* **76**, 3433 (2000).
- ²⁸I. I. Fishchuk, A. K. Kadashchuk, J. Genoe, Mujeeb Ullah, H. Sitter, Th. B. Singh, N. S. Sariciftci, and H. Bässler, *Phys. Rev. B* **81**, 045202 (2010).
- ²⁹Mujeeb Ullah, I. I. Fishchuk, A. K. Kadashchuk, P. Stadler, A. Pivrikas, C. Simbrunner, V. N. Poroshin, N. S. Sariciftci, and H. Sitter, *Appl. Phys. Lett.* **96**, 213306 (2010).
- ³⁰W. G. Gill, *J. Appl. Phys.* **43**, 5033 (1972).
- ³¹L. B. Schein, A. Peled, and D. Glatz, *J. Appl. Phys.* **66**, 686 (1989).
- ³²S. D. Baranovskii, H. Cordes, F. Hensel, and G. Leising, *Phys. Rev. B* **62**, 7934 (2000).
- ³³Y. Roichman and N. Tessler, *Appl. Phys. Lett.* **80**, 1948 (2002).
- ³⁴O. Rubel, S. D. Baranovskii, P. Thomas, and S. Yamasaki, *Phys. Rev. B* **69**, 014206 (2004).
- ³⁵I. I. Fishchuk, D. Hertel, H. Bässler, and A. K. Kadashchuk, *Phys. Rev. B* **65**, 125201 (2002).
- ³⁶J. Zhou, Y. C. Zhou, J. M. Zhao, C. Q. Wu, X. M. Ding, and X. Y. Hou, *Phys. Rev. B* **75**, 153201 (2007).
- ³⁷V. Ambegaokar, B. I. Halperin, and J. S. Langer, *Phys. Rev. B* **4**, 2612 (1971).
- ³⁸B. I. Shklovskii and A. L. Efros, *Electronic Properties of Doped Semiconductors* (Springer, Heidelberg, 1984).
- ³⁹C. Tanase, E. J. Meijer, P. W. M. Blom, and D. M. de Leeuw, *Org. Electr.* **4**, 33 (2003).
- ⁴⁰X. Li, A. Kadashchuk, I. I. Fishchuk, W. T. T. Smaal, G. Gelinck, D. J. Broer, J. Genoe, P. Heremans, and H. Bässler, *Phys. Rev. Lett.* **108**, 066601 (2012).
- ⁴¹A. Pivrikas, Mujeeb Ullah, H. Sitter, and N. S. Sariciftci, *Appl. Phys. Lett.* **98**, 092114 (2011).
- ⁴²M. Ullah, A. Pivrikas, I. I. Fishchuk, A. Kadashchuk, P. Stadler, C. Simbrunner, N. S. Sariciftci, and H. Sitter, *Appl. Phys. Lett.* **98**, 223301 (2011).