Parametrization of the charge-carrier mobility in organic disordered semiconductors

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An appropriately parametrized analytical equation (APAE) is suggested to account for charge-carrier mobility in organic disordered semiconductors. This equation correctly reproduces the effects of temperature T, carrier concentration n, and electric field F on the carrier mobility $\mu(T, F, n)$, as evidenced by comparison with analytical theories and Monte Carlo simulations. The set of material parameters responsible for charge transport is proven to be at variance with those used in the so-called extended-Gaussian-disorder-model (EGDM) approach, which is widely exploited in commercially distributed device-simulation algorithms. While the EGDM is valid only for cubic lattices with a specific choice of parameters, the APAE describes charge transport in systems with spatial disorder in a wide range of parameters. The APAE is user-friendly and, thus, suitable for incorporation into device-simulation algorithms.

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I. INTRODUCTION

A correct parametrization of carrier mobility in organic disordered semiconductors (ODSs) with hopping charge transport is of vital importance for the development of algorithms to simulate devices based on ODSs, such as organic light-emitting diodes, organic solar cells, and organic field-effect transistors. Theoretical equations for the charge-carrier mobility $\mu(T, n, F)$, dependent on the concentration of carriers n, on temperature T, and on the applied electric field F, are at the heart of device-simulation algorithms. The choice of the appropriate theoretical description for $\mu(T, n, F)$ suitable for applications in device-simulation software was recently addressed in several studies [1–6]. Despite the progress achieved in these studies, the parametrization of the dependences $\mu(T, n, F)$ still needs improvement.

Most of the device-simulation algorithms, including commercially distributed software packages, are based on the EGDM equation [7]. This equation has been suggested [7] as a "unified description of charge-carrier mobilities in disordered semiconducting polymers." Several review papers [8,9] promote the EGDM equation as the gold standard for the description of carrier mobilities in ODSs, and the EGDM is the basis of commercially distributed device-simulation packages [2,8].

However, the EGDM equation [7,8] opposes the basic theoretical concepts developed so far for hopping transport in disordered materials [10–12]. First, the EGDM equation is based on an irrelevant parametrization [10–12]. Some parameters responsible for $\mu(T, n, F)$ are missing in the EGDM, while some parameters present in the EGDM are not responsible for the effects. Second, the EGDM equation was formulated to fit simulation data on regular cubic lattices without spatial disorder [7]. However, hopping mobility on regular lattices deviates significantly from that in materials with spatial disorder [6,12]. Therefore, it is necessary to determine whether the EGDM based on regular lattices with deficient parametrization could be of use for systems with spatial disorder.

The regular cubic lattice used in the EGDM is a specific case of the Gaussian-disorder model (GDM), in which charge transport is due to incoherent hopping of carriers via randomly distributed localized states with a Gaussian energy spectrum. This model is widely used due to its ability to account for experimentally observed dependences $\mu(T, n, F)$ [13–15]. A parametrization of the carrier mobility $\mu(T, n, F)$ in the framework of the GDM is given in Sec. II.

While much effort has been focused on computer simulations, the transport problem in the GDM can be easily solved analytically in the form of a simple closed-form system of equations [1,2,4,12,16–22]. This solution is formulated in Sec. III. Although this simple closed-form system of equations can easily be solved numerically, it has not yet become a state of the art for the device-simulation

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community. Single user-friendly equations, such as the EGDM equation, can be more easily implemented in device-simulation software than systems of interconnected analytical equations, but even so the user-friendliness is achieved at the cost of accuracy.

The challenging task is to replace the system of analytical equations for $\mu(T, n, F)$ by a single user-friendly appropriately parametrized analytical equation (APAE) that can be easily embedded into device-simulation software. The APAE is formulated in Sec. IV, providing the main result of this paper.

In Sec. V, the validity of the APAE is proven by comparison with results of computer simulations. The agreement of this heuristic equation with analytical theories and with computer simulations suggests the APAE for using in device-simulation algorithms.

In Sec. VI, the widely used alternative description of $\mu(T, n, F)$ based on the EGDM is discussed. Despite the insufficient parametrization and despite the reduction of simulated systems to regular lattices, the EGDM could, by chance, be, in rare cases, applicable to systems with spatial disorder.

Section VII is dedicated to comparison with recent studies in the literature [1–3] conducted in the framework of the same model as the one used in our study to develop the APAE.

II. CRUCIAL PARAMETERS FOR THE CARRIER MOBILITY

A. Gaussian-disorder model

Charge transport in single-component and multicomponent ODSs is due to incoherent hopping of carriers via randomly distributed localized states with a Gaussian energy spectrum [1–4,7–9,12,13,20–25],

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

where σ is the energy scale of the density of states and N is the concentration of randomly distributed localized states, henceforth called "hopping sites." The estimates for σ range between approximately 0.05 eV and approximately 0.15 eV [1,7,13], and the estimates for N range between approximately 1.7×10^{20} cm^{-3} and approximately 4.6×10^{21} cm^{-3}, depending on the material [2,7,13]. For simplicity, we follow most previous studies considering the GDM without correlations between the spatial positions of hopping sites and their energies [1–4,7–9,12,13,20–22,24,25].

Phonon-assisted hopping suggested by Miller and Abrahams [26] is usually considered to be the dominant charge-transport mechanism in ODSs [1–4,7–9,12,13,20–22,24,25]. The expression for the rate of carrier transfer from an occupied site with energy ε_i to an empty site with energy ε_j over the distance r_{ij} has the form

$$\nu_{ij} = \nu_0 \exp\left(-\frac{2|\mathbf{r}_{ij}|}{a}\right) \chi\left(\frac{\varepsilon_j - \varepsilon_i - e\mathbf{F}\cdot\mathbf{r}_{ij}}{kT}\right), \tag{2}$$

with

$$\chi(X) = \begin{cases} \exp(-X) & \text{if } X > 0, \\ 1 & \text{if } X \le 0. \end{cases}$$

Here $\mathbf{F}$ is the applied electric field, e is the elementary charge, a is the localization length of charge carriers in the localized states, k is the Boltzmann constant, and T is the temperature. Estimates of a in the range 0.1 nm $\le a \le$ 0.75 nm have been suggested in the literature [2,7,18,27]. The prefactor in Eq. (2) is usually described by a single parameter, the so-called attempt-to-escape frequency ν_0. Precise quantum-mechanical calculation of ν_0 can be found elsewhere [26,28]. The energy ε_i of the starting site and the energy ε_j of the target site in Eq. (2) are counted without contributions of the applied electric field. The effect of the electric field $\mathbf{F}$ on the hopping rates is expressed explicitly by the term $e\mathbf{F}\cdot\mathbf{r}_{ij}$ in the exponent on the right-hand side in Eq. (2).

The validity of the GDM determined by Eqs. (1) and (2) is justified by its ability to account for a broad variety of experimental observations. Among those is the transition from the dependence $\ln[\mu(T)] \propto 1/T^2$ to the dependence $\ln[\mu(T)] \propto 1/T$ with rising carrier concentration [17,20,21]. Another pronounced phenomenon predicted by Eqs. (1) and (2) is the transition from the mobility μ being independent of carrier concentration n at small values of n to the mobility μ being strongly dependent on n at large values of n [4,12,17,20–22]. Experimental data related to the latter effect [29] are indicative [4,12,21,22] of the Gaussian shape of the density of states given by Eq. (1). Therefore, our consideration is based on Eqs. (1) and (2), in agreement with several recent studies [1–3,12,30].

B. Parametrization of the mobility $\mu(T, n, F)$

For the discussion of the proper parametrization of $\mu(T, n, F)$, we focus on the strong exponential dependences of the carrier mobility $\mu(T, n, F)$—namely, on temperature T, electric field F, and carrier concentration n.

1. Parametrization at small electric fields

As is evident from the exponents in Eqs. (1) and (2), the transport problem at small electric fields, $F \to 0$, is determined by only two dimensionless parameters: $\alpha = kT/\sigma$ and $\beta = N^{-1/3}/a$. This is true at low carrier concentrations [16–18], $n \ll N$, when the mobility μ does not depend on n. At large values of n, a third dimensionless parameter, $\gamma = n/N$, enters the carrier mobility $\mu(T, n)$.

Remarkably, the dependence of the carrier mobility μ on temperature T is affected by the parameter $N^{-1/3}/a$. [10] This is true in the so-called variable-range-hopping (VRH) regime, when the characteristic hopping length depends on T. Charge transport in ODSs is dominated by the VRH process, as has been proven by Monte Carlo simulations and analytical calculations [5,6,16–18]. Therefore, the temperature dependence $\mu(T,n)$ is sensitive to β [5,6,16–18]. This effect is often overlooked [7,13].

2. Parametrization of the field dependence

The appropriate parametrization of the dependence $\mu(F)$ in hopping transport was revealed in 1973 by Shklovskii [11], who concluded that the effect of the electric field F on the carrier mobility μ is determined by the product eaF, where a is the localization length. Shklovskii considered for simplicity the case $T=0$, recognizing that a charge carrier gains the amount of energy $\Delta = eFx$ tunneling in the field direction over some distance x. The tunneling rate $\nu(x) \propto \exp(-2x/a)$ can be rewritten as $\nu(x) \propto \exp(-\Delta/kT_{\text{eff}})$, with $T_{\text{eff}} \simeq eFa/2$. Apparently, the field-dependent effective temperature $T_{\text{eff}} \simeq eFa/2$ accounts for the effect of the electric field F on hopping transport at $T=0$.

For the case $T \neq 0$, Marianer and Shklovskii [31] suggested that the combined effects of the electric field F and temperature T can be expressed in the form of the effective temperature:

$$T_{\text{eff}} = T\left[1+\left(c_1\frac{eFa}{kT}\right)^2\right]^{1/2}, \tag{3}$$

with $c_1 \approx 0.67$. Several studies performed by numerical simulations [5,32–34] confirmed the validity of this approach in the range $0.5 \leq c_1 \leq 0.9$.

Apparently, the localization length a and not the intersite distance $N^{-1/3}$ governs the effect of the electric field on the hopping conductivity. This fact is nontrivial because the electric field enters the theory only via the combination $e\mathbf{F}\cdot\mathbf{r}_{ij}$, in which the length of a hop $|\mathbf{r}_{ij}|$ is of the order of the intersite distance $N^{-1/3}$. Therefore, one might expect the combination of the parameters e, $N^{-1/3}$, and F to be essential for the field-dependent mobility. However, it has been rigorously proven by straightforward computer simulations [5] that the localization length a, i.e., the feature of a single localized state, and not the intersite distance $N^{-1/3}$, is responsible for $\mu(F)$. This counterintuitive result has not yet been adopted by the broad scientific community despite its rigorous proof [5,12]. Equation (3) along with Eqs. (1) and (2) implies that the combined effects of the electric field $\mathbf{F}$ and temperature T are described by a single parameter $\alpha = kT_{\text{eff}}/\sigma$. This means that the effect of the electric field on the hopping conductivity is governed in accord with Eq. (3) by the parameter

$$\delta = eFa/kT. \tag{4}$$

Herewith, only three parameters are responsible for $\mu(T,n,F)$—namely,

$$\alpha = kT_{\text{eff}}/\sigma, \quad \beta = N^{-1/3}/a, \quad \gamma = n/N. \tag{5}$$

III. ANALYTICAL DESCRIPTION OF $\mu(T,n,F)$

The analytical theory for the description of hopping transport in amorphous materials with strongly-energy-dependent density of states $g(\varepsilon)$ has been known for decades [19,20,35,36]. In particular, it has been proven [4,12,21,22] that the charge-carrier mobility can be described in the framework of the GDM as

$$\mu = \mu_0\gamma^{-1}\exp\left(-\frac{2B_c^{1/3}}{a}r(\varepsilon_t) - \frac{\varepsilon_t-\varepsilon_F}{kT}\right), \tag{6}$$

where

$$r(\varepsilon_t) = \left[\frac{4\pi}{3}\int_{-\infty}^{\varepsilon_t} g(\varepsilon')[1-f(\varepsilon',\varepsilon_F)]d\varepsilon'\right]^{-1/3}, \tag{7}$$

and the transport energy ε_t is calculated from [4,12,21]

$$\frac{2}{3}\left(\frac{4\pi}{3B_c}\right)^{-\frac{1}{3}}\frac{kT}{a}\left[\int_{-\infty}^{\varepsilon_t}[1-f(\varepsilon,\varepsilon_F)]g(\varepsilon)d\varepsilon\right]^{-\frac{4}{3}} \times [1-f(\varepsilon_t,\varepsilon_F)]g(\varepsilon_t) = 1, \tag{8}$$

where $f(\varepsilon,\varepsilon_F)$ is the Fermi function

$$f(\varepsilon,\varepsilon_F) = \left[1+\exp\frac{(\varepsilon-\varepsilon_F)}{kT}\right]^{-1}, \tag{9}$$

with the Fermi energy ε_F determined by the relation

$$\int_{-\infty}^{\infty} g(\varepsilon)f(\varepsilon,\varepsilon_F)d\varepsilon = n, \tag{10}$$

which accounts for the finite charge-carrier concentration n. The coefficient $B_c \simeq 2.7$ is due to the percolation nature of the hopping transport [18].

The pre-exponential factor in Eq. (6) is given by

$$\mu_0 = B\frac{e\nu_0}{kTN^{2/3}}, \tag{11}$$

where B is a numerical factor, which can be determined by comparison with computer simulations.

The structure of the prefactor μ_0 in Eq. (11) relies on the conventional form of the Einstein relation between the carrier mobility and the carrier diffusion coefficient that is

valid for nondegenerate systems, i.e., at low carrier concentrations $n \ll N$. In general, the ratio e/kT should be replaced by the generalized Einstein relation [15,21,37]. We nevertheless leave Eq. (11) in the given form because the final result for $\mu(T,n,F)$ in Sec. IV is obtained from a comparison of the parameters in the analytical theory with numerical data from computer simulations. In this respect, our approach is essentially similar to that used in recent studies by Upreti *et al.* [1] and Lee *et al.* [2], who calibrated parameters of a similar analytical theory by comparison with numerical simulations.

Following the well-known recipe [1,12,21,38], we replace T by T_{eff} in the above equations, in order to describe appropriately the dependence $\mu(F)$.

IV. APAE FOR $\mu(T,n,F)$

The set of equations (6)–(11) determines $\mu(T,n,F)$ in terms of the relevant parameters given by Eq. (5). Remarkably, the function $\mu(T,n,F)$ obtained from this set of equations can be approximated in realistic parameter ranges by a single APAE:

$$\mu(T,n,F) = \frac{e\nu_0}{\sigma N^{2/3}}\Phi(\gamma)\exp\left[-A(\beta) - \alpha^{-2}C(\beta)\right], \tag{12}$$

where

$$A(\beta) = c_2\beta^2 + c_3\beta + c_4, \tag{13}$$

$$C(\beta) = c_5\beta^{-2} + c_6\beta^{-1} + c_7. \tag{14}$$

The constant $\exp(c_4)$ replaces the factor B in Eq. (11).

The function $\Phi(\gamma)$ in Eq. (12), responsible for the dependence $\mu(n)$, is determined by the equation

$$\Phi(\gamma) = \exp\left[(c_8 \ln\alpha^{-1} - c_9)\xi^2\Theta(\xi)\right], \tag{15}$$

where

$$\xi = \frac{\alpha^{-1}}{2} + 2 - \left|\ln\left(\gamma - \frac{c_{10}\gamma^2}{\max(\beta, c_{11})}\right)\right|^{1/2}, \tag{16}$$

$$\Theta(x) = \begin{cases} 1 & \text{if } x \geq 0, \\ 0 & \text{if } x < 0. \end{cases}$$

Numerical parameters c_1–c_{11} were further optimized by comparison with computer simulations, attaining the following values:

$$\begin{aligned} c_1 &= 0.6, \\ c_2 &= -0.066, \\ c_3 &= 2.65, \\ c_4 &= -1.35, \\ c_5 &= 0.89, \\ c_6 &= -0.86, \\ c_7 &= 0.54, \\ c_8 &= 0.6, \\ c_9 &= 0.15, \\ c_{10} &= 13, \\ c_{11} &= 3. \end{aligned} \tag{17}$$

Recall that $\alpha = kT_{\text{eff}}/\sigma$, $\beta = N^{-1/3}/a$, and $\gamma = n/N$ [see Eq. (5)] and T_{eff} is determined by Eq. (3) with c_1 given by Eq. (17). Equation (12) is the central result of our work.

To understand the motivation for the choice of the APAE, we first consider the limiting cases of low carrier concentration n and low electric field F. Then we introduce a correction due to finite carrier concentration n. Finally, we consider a modification due to a finite electric field.

In the low-carrier-concentration and low-electric-field limits, dimensionality considerations impose the condition that the mobility must be equal to the factor $e\nu_0/(\sigma N^{2/3})$ multiplied by a dimensionless function of α and β. It is well known that the mobility, as a function of temperature, in this limit is proportional to $\exp[-C(\sigma/kT)^2] \equiv \exp(-C\alpha^{-2})$. The parameter C here depends on β, and the coefficient of proportionality, which we denote as $\exp(-A)$, also can depend on β. These considerations are motivation for the representation of the mobility μ as

$$\mu = \frac{e\nu_0}{\sigma N^{2/3}} \exp[-A(\beta) - \alpha^{-2}C(\beta)] \tag{18}$$

in the low-carrier-concentration and low-electric-field limits. This is Eq. (12) up to the concentration-dependent factor $\Phi(\gamma)$.

The functions $A(\beta)$ and $C(\beta)$ appear sufficiently smooth, allowing a Taylor expansion. We performed Taylor expansion in parameter β as well as in β^{-1} up to quadratic terms for both functions, and found that expansion in β is more accurate for function A, whereas expansion in β^{-1} is more accurate for function C. These findings justify Eqs. (13) and (14).

Let us now turn to the dependence of the mobility on the carrier concentration n described by the factor $\Phi(\gamma)$ in Eq. (12). Looking at Eq. (6), one can see three reasons for

the concentration dependence of the mobility: (1) the factor γ^{-1}, where $\gamma = n/N$, (2) the factor $\exp(\varepsilon_F/kT)$, where the Fermi energy ε_F depends on the carrier concentration n in accord with Eq. (10), and (3) the transport energy ε_t determined by Eq. (8). It has been proven [39] that ε_t becomes concentration dependent only at extremely high concentrations n. When the concentration n is lower than some critical value n_c, the contributions of the factors γ^{-1} and $\exp(\varepsilon_F/kT)$ to the dependence $\mu(n)$ compensate each other [4], yielding $\Phi(\gamma) = 1$. At higher concentrations, the Fermi energy can be roughly estimated [4] from the relation $\int_{-\infty}^{\varepsilon_F} g(\varepsilon)d\varepsilon \approx n$. Ignoring the pre-exponential factors, one can approximate the left-hand side of this relation as $N\exp(-\varepsilon_F^2/2\sigma^2)$, which provides the following estimate for the Fermi energy:

$$\varepsilon_F \approx -\sigma\sqrt{2|\ln(n/N)|} \equiv -\sigma\sqrt{2|\ln\gamma|}. \quad (19)$$

Hence, for $n < n_c$, the concentration-dependent factor $\Phi(\gamma)$ is equal to unity, and for $n > n_c$, it can be estimated as an exponent of a quadratic function of $\sqrt{|\ln\gamma|}$, in which the linear term is due to the factor $\exp(\varepsilon_F/kT)$ in Eq. (6) and the quadratic term is due to the factor $\gamma^{-1} \equiv \exp(-\ln\gamma) \equiv \exp[(\sqrt{|\ln\gamma|})^2]$ in Eq. (6). These two cases must be smoothly connected to each other at $n \approx n_c$. The latter circumstance gives rise to the following guess for the shape of the function $\Phi(\gamma)$:

$$\Phi(\gamma) = \exp[f\ \xi^2\ \Theta(\xi)], \quad (20)$$

where ξ is a linear function of $\sqrt{|\ln\gamma|}$, and the coefficient f can depend on $\alpha = kT/\sigma$. We found that f is well approximated as a linear function of $\ln\alpha$, which gives rise to Eq. (15). In the expression for ξ as a linear function of $\sqrt{|\ln\gamma|}$, the free term (independent of γ) depends on α, and this dependence is well approximated by a linear function of α^{-1}. As a result, we arrive at Eq. (16). Also in Eq. (16), there is a β-dependent correction that accounts for the concentration dependence of the transport energy ε_t in Eq. (6), and this becomes important only for very high concentrations.

Finally, the effect of the finite electric field is fully incorporated in the APAE by our replacing the actual temperature T by the effective temperature T_{eff} in parameter α, as stated in Eq. (5).

To optimize the numerical parameters c_1–c_{11}, we, in the first step, used the numerical data for the mobility obtained in the limit $n \to 0$, $F \to 0$. From the linear approximation of the dependence of $\ln\mu$ on $(\sigma/kT)^2$, we found the coefficients A and C in the dependence $\mu = (e\nu_0/\sigma N^{2/3})\exp(-A - \alpha^{-2}C)$ at different values of β. Approximating the dependences $A(\beta)$ and $C(\beta^{-1})$ by quadratic polynomials, we found coefficients c_2–c_7. In the next step, we considered the concentration dependences of the mobility in the range $10^{-7}N \leq n \leq 10^{-2}N$. For the different values of α, we optimized the coefficient f in the approximation $\mu(n)/\mu(0) = \exp[f\,\xi^2\Theta(\xi)]$, where ξ was calculated by Eq. (16) with the omission of the last (β-dependent) term in parentheses. This term is not important at concentrations $n < 10^{-2}N$. From the linear approximation of the dependence of f on $\ln\alpha^{-1}$, we obtained c_8 and c_9. Then c_{10} and c_{11} were found by optimization of the APAE for the concentration dependence of the mobility in the concentration range $10^{-2}N \leq n \leq 10^{-1}N$. Finally, the value of c_1 was chosen to fit the data on the electric field dependence of the mobility shown in Fig. 1.

V. TESTING THE APAE BY COMPUTER SIMULATIONS

A. APAE and $\mu(F)$

In Fig. 1, the dependences of the normalized mobility $\mu(F)/\mu(0)$ obtained by Monte Carlo simulations at different values of the parameter $\beta = N^{-1/3}/a$ are depicted by symbols. Simulations were performed in the framework of the model with spatial and energy disorder formulated in Sec. II A. The simulation algorithm is the same as the one used in previous studies [5]. In particular, we used the HOPHOP algorithm [40]. Simulation data obtained at $\beta^{-1} \leq 0.3$ agree with those from previous studies [5], while the data obtained at $\beta = 2$ are novel. At all values of β in Fig. 1, $\mu(F)/\mu(0)$ steeply increases with rising field F at $F \leq 2\sigma/eN^{-1/3}$. At larger fields, the field dependences in Fig. 1 saturate. Solid lines in Fig. 1 depict the results obtained with the APAE. Apparently,

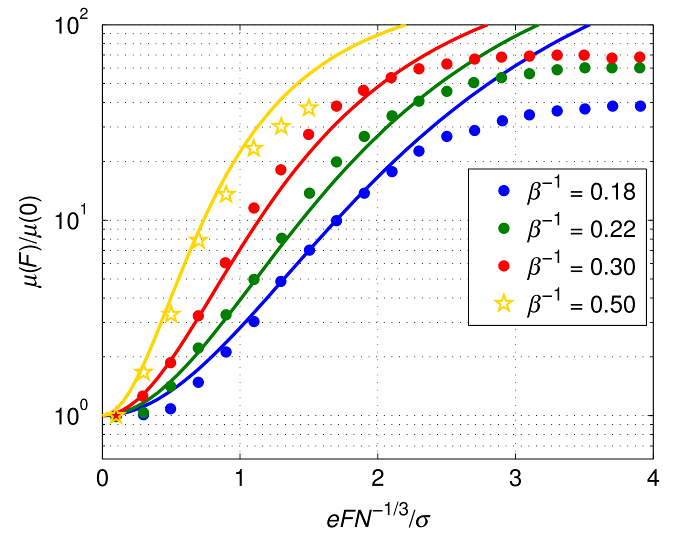

FIG. 1. Normalized mobility $\mu(F)/\mu(0)$ at $\sigma/kT = 4$ and $n \to 0$ as a function of $eFN^{-1/3}/\sigma$ at different values of $\beta = N^{-1/3}/a$. Symbols represent the results of Monte Carlo simulations and solid lines represent results obtained with the APAE (12).

the APAE appropriately describes the simulation data at $F \leq 2\sigma/eN^{-1/3}$, while at larger fields, the APAE predicts a stronger dependence $\mu(F)/\mu(0)$ than the one obtained by simulations.

The weakening of the field dependency at high fields in Fig. 1 is a well-known effect [13,41,42]. As is evident from Eq. (2), the hopping rates do not depend on site energies at very high fields, $F \gg \sigma/eN^{-1/3}$, leading to saturation of the drift velocity at large F. Concomitantly, mobility $\mu(F)$ at very high fields should decrease with increasing F [13,41–45]. The apparent saturation of $\mu(F)$ in Fig. 1 is the result of a transition from the increasing dependence $\mu(F)$ at small and moderate fields to the decreasing $\mu(F)$ at very high fields. This effect is not unique for the Miller-Abrahams (MA) hopping rates given by Eq. (2). Marcus expressions for hopping rates were shown [42] to cause an even-more-pronounced decrease of $\mu(F)$ at large F than the MA rates. Hopping transitions caused by nuclear tunneling also lead [45] to decreasing $\mu(F)$ at large F.

The width of the range of electric fields with increasing mobility $\mu(F)$ depends on the disorder parameter σ in Eq. (1). The larger is σ, the broader is this range [13,41]. In the absence of energy disorder, i.e., at $\sigma = 0$, mobility always decreases [13] with increasing F.

We do not address such high-field effects in our study, because the range of electric fields, $F < 2 \times 10^6$ V cm^{-1}, that is relevant to experimental studies [46,47] and to device simulations [1] corresponds in Fig. 1 to the field range with increasing mobility. The data depicted in Fig. 1 suggest for the width of this range the estimate $F \leq F^* \simeq 2\sigma/eN^{-1/3}$. Taking the relevant for ODS parameter values [2,7], $N \approx 10^{21}$ cm^{-3} and $\sigma \approx 0.1$ eV, one obtains $F^* \approx 2 \times 10^6$ V cm^{-1}, which perfectly agrees with the upper limit of the field interval relevant for experimental studies and for device simulations [1,46,47]. In this range of electric fields, the APAE provides a good description of $\mu(F)$ with the parameter values $\beta > 3.3$ relevant for practical applications. Therefore, the APAE can be successfully used to interpret experimental data and it can be applied in device-simulation algorithms.

B. APAE and $\mu(T)$

The scale of energy disorder σ is usually estimated in ODSs as between approximately 50 meV and approximately 150 meV [1,2,13]. For σ/kT at room temperature $T \approx 300$ K, this gives values between approximately 2 and approximately 6.

In Fig. 2, the mobility $\mu(F \to 0, n \to 0)$, as obtained by computer simulations, is plotted as a function of σ/kT at different values of $\beta = N^{-1/3}/a$. The results obtained with the APAE are in perfect agreement with the simulation data. In contrast to the Monte Carlo simulations used for the data in Fig. 1, numerical calculations at $F \to 0$ were performed by solution of Kirchhoff equations in a resistor network proposed by Miller and Abrahams [28].

The data in Fig. 2 correspond to the well-known dependence $\mu(T)$, which can be approximated by the expression

$$\mu(T) \propto \exp\left[-C\left(\frac{\sigma}{kT}\right)^2\right]. \tag{21}$$

By fitting the Monte Carlo simulation data for $\mu(T)$ obtained on a cubic lattice at $\beta = N^{-1/3}/a = 5$, Bässler [13] suggested the value $C = (2/3)^2 \simeq 0.44$. With use of analytical theory based on the use of the transport energy given by Eq. (8), it was shown [16] that in the GDM with spatial and energy disorder described in Sec. II A, the coefficient C depends on the parameter $\beta = N^{-1/3}/a$, with $C \simeq 0.46$ at $\beta = 10$, $C \simeq 0.41$ at $\beta = 5$, and $C \simeq 0.38$ at $\beta \simeq 3.7$. These values have been confirmed by analytical calculations based on the percolation theory [17]. The data depicted in Fig. 2 are in good agreement with these previous results yielding the values of C specified in the figure.

C. APAE and $\mu(n)$

In Fig. 3, the normalized mobility $\mu(n)/\mu(n \to 0)$ at $F \to 0$ is plotted as a function of $\gamma = n/N$ for a typical ODS parameter value [13] $\beta = 5$ and different values of $\alpha = kT/\sigma$. The choice of $\beta \simeq 5$ is further supported by the estimates [18] 0.1 nm $\leq a \leq$ 0.3 nm and 0.5 nm $\leq N^{-1/3} \leq$ 1.8 nm in ODSs [7,18].

The computer simulations confirm the capability of the APAE to describe the dependences $\mu(T, n, F)$ in the

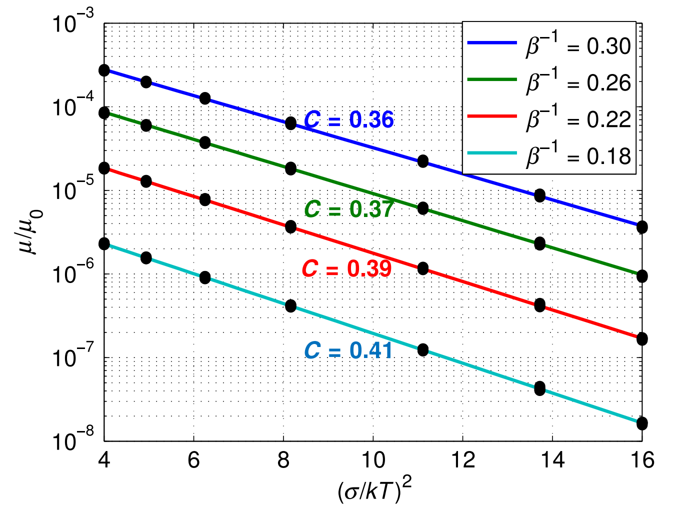

FIG. 2. Mobility $\mu(T)$ at $n \to 0$, $F \to 0$ as a function of $\alpha^{-2} = \sigma^2/(kT)^2$ for different values of $\beta = N^{-1/3}/a$. Symbols represent results of simulations and solid lines represent results obtained with the APAE.

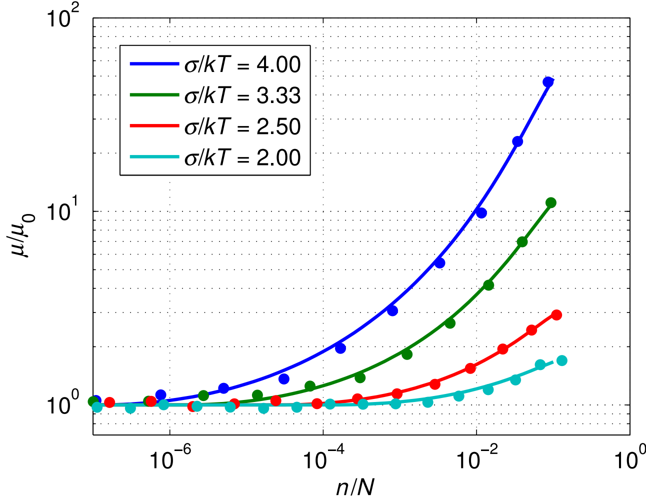

FIG. 3. Normalized mobility $\mu(n)/\mu(n \to 0)$ at $F \to 0$ as a function of $\gamma = n/N$ at $\beta = 5$ and different values of $\alpha = kT/\sigma$. Symbols represent results of simulations and solid lines represent results obtained with the APAE.

framework of the GDM, i.e., for hopping transport via randomly distributed localized states with a Gaussian energy spectrum.

Most algorithms developed so far for the simulation of devices based on ODSs, such as organic light-emitting diodes, organic solar cells, and organic field-effect transistors, are based on the EGDM equation [7,8]. In the following sections, we analyze the EGDM equation and compare it with the APAE.

VI. COMPARISON WITH THE EGDM

A reduced version of the GDM on a cubic lattice without spatial disorder at the fixed value $N^{-1/3} = 10a$ is what one calls the EGDM [8]. Charge transport on a regular cubic lattice was simulated for the case $b = 10a$, where b is the lattice spacing and a is the localization length in Eq. (2). To make a bridge to the case of the randomly placed localized states, we note that $b = N^{-1/3}$. Simulation results for the carrier mobility $\mu(T, n, F)$ were fitted in the form [7]

$$\mu(T, n, F) \approx \widetilde{\mu}(T, n)\phi(T, F), \tag{22}$$

where $\phi(T, F)$ is given by

$$\phi(T, F) = \exp\left\{0.44\left[\widehat{\sigma}^{3/2} - 2.2\right]\left[\sqrt{1 + 0.8\left(\frac{Feb}{\sigma}\right)^2} - 1\right]\right\}, \tag{23}$$

with $\widehat{\sigma} = \sigma/(kT)$. The function $\widetilde{\mu}(T, n)$ is given by [7]

$$\widetilde{\mu}(T, n) = \mu_0(T)\exp[(\widehat{\sigma}^2 - \widehat{\sigma})(2nb^3)^{\widetilde{\delta}}/2], \tag{24a}$$

$$\mu_0(T) = \mu_0 \mathfrak{c}_1 \exp[-\mathfrak{c}_2\widehat{\sigma}^2], \tag{24b}$$

$$\widetilde{\delta} \equiv 2\frac{\ln(\widehat{\sigma}^2 - \widehat{\sigma}) - \ln(\ln 4)}{\widehat{\sigma}^2}, \quad \mu_0 \equiv \frac{b^2\nu_0 e}{\sigma}, \tag{24c}$$

with $\mathfrak{c}_1 = 1.8 \times 10^{-9}$ and $\mathfrak{c}_2 = 0.42$. Equations (22)–(24c) were named [8] the "extended Gaussian-disorder model." These equations fit numerical data for $\mu(T, n, F)$ obtained on a cubic lattice at $b/a = 10$.

Remarkably, the localization length of charge carriers, a, does not enter the EGDM equations at all, although a is known [10,11] to determine the temperature and the field dependences of the hopping mobility via the parameters $\alpha = kT_{\text{eff}}/\sigma$ and $\beta = N^{-1/3}/a$ present in Eqs. (3) and (5).

The absence of a in the EGDM was supported by two arguments. First, it was stated that presumably in all published 3D-modeling work on the (E)GDM, a fixed value $N^{-1/3}/a = 10$ had been used, suggesting $N^{-1/3}/a = 10$ as a standard one for ODSs [8]. In this respect, one has to remark, however, that published 3D-modeling work on the GDM mostly used a fixed value $N^{-1/3}/a = 5$ instead [13].

Second, it was stated, referring to unpublished work, that variation of the ratio $N^{-1/3}/a$ has no significant effect on the temperature, field, or carrier-density dependence of the mobility [8]. Figure 1 evidences, however, that variation of the ratio $N^{-1/3}/a$ does have a significant effect on the field dependence of the mobility in the GDM with spatial disorder. A similar effect has also been proven on cubic lattices [5,12].

This analysis raises the question of whether the EGDM can be used for the description of $\mu(T, n, F)$ in ODSs. The answer to this question is of great importance not only for academic researchers but also particularly for the community dealing with device simulations, where the EGDM is considered as the state of the art. It is a lucky coincidence that the EGDM equation could, by chance, be applicable to ODSs, in some cases.

In Fig. 4, we depict by symbols the results of Monte Carlo simulations [5,6] for the system of random sites at $0.1 \le \beta^{-1} \le 0.3$ along with the result obtained with the EGDM (dashed line), which fits the Monte Carlo simulations [7] on a cubic lattice at $\beta^{-1} = 0.1$. The case $\sigma/kT = 4$ is considered as typical for ODSs at room temperature, since σ is usually estimated as approximately 0.1 eV [2,7].

Apparently, $\mu(F)$ on the lattice is not equal to $\mu(F)$ in a system of random sites. Equation (23) fits the simulation data on a cubic lattice at $\beta^{-1} = 0.1$, but it does not fit the simulation data in a spatially disordered system at $\beta^{-1} = 0.1$ (black circles). The physical mechanism responsible for the drastic difference in $\mu(F)$ between lattices and random sites has been discussed in detail elsewhere [6].

From Fig. 4 we, however, learn that the EGDM, although desired to fit $\mu(F)$ on a cubic lattice at $\beta^{-1} = 0.1$,

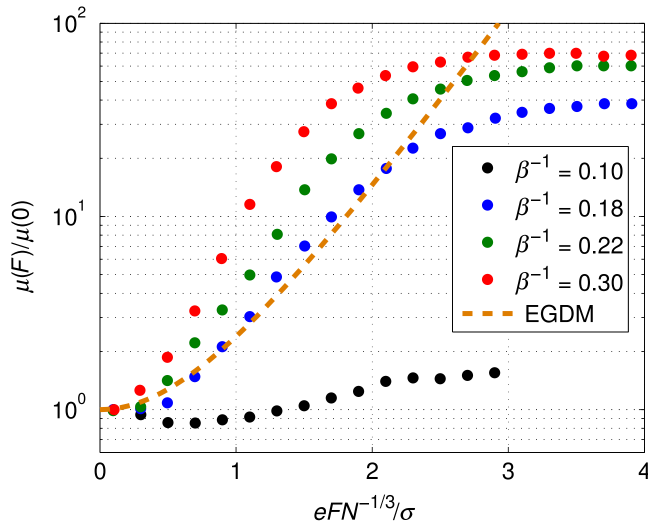

FIG. 4. Normalized mobility $\mu(F)/\mu(0)$ at $n \to 0$ for $\sigma/kT = 4$ as a function of electric field at different values of β. Symbols represent data from Monte Carlo simulations obtained for random sites at $\beta^{-1} = 0.1$ (black), $\beta^{-1} = 0.18$ (blue), $\beta^{-1} = 0.22$ (green), and $\beta^{-1} = 0.3$ (red). The dashed orange line is the result obtained with the EGDM, i.e., the data for a cubic lattice at $\beta^{-1} = 0.1$.

fits, by chance, $\mu(F)$ on random sites at $\beta^{-1} = 0.18$. While $\beta = N^{-1/3}/a = 10$ was standardized in the EGDM [8], the previously used $\beta = N^{-1/3}/a = 5$ looks more relevant to ODSs [13]. This conclusion is supported by the estimates for a [2,7,18,27], 0.1 nm $\leq a \leq$ 0.75 nm, and estimates for N between approximately 1.7×10^{20} cm^{-3} and approximately 4.6×10^{21} cm^{-3} depending on the material [2,7,13]. The data in Fig. 4 suggest, therefore, that the EGDM could, by chance, be applied to ODSs with a realistic value $\beta^{-1} = a/N^{-1/3} \simeq 0.18$. For materials with $\beta^{-1} > 0.18$, one should, instead, use the APAE for $\mu(F)$, as illustrated in Fig. 4.

Furthermore, the EGDM occasionally appears capable of accounting, in some cases, for the dependence $\mu(T)$ in a system with spatial disorder. The localization length a affects not only the field dependence $\mu(F)$ but also the temperature dependence $\mu(T)$ [2,3,16,17]. This effect is ignored in the EGDM. The effect of a on the dependence $\mu(T)$ can be taken into account, for instance [2,3,16,17], by replacing the constant c_2 in Eq. (24b) by the appropriate function of the parameter $\beta = N^{-1/3}/a$. An illustration is provided by Fig. 2, where the slopes of the straight lines depend on $\beta = N^{-1/3}/a$.

At low carrier concentrations and low electric fields, the EGDM describes the dependence $\mu(T)$ by Eq. (21) with $C = 0.42$. As is evident in Fig. 2, the dependence $\mu(T)$ for carrier mobility on random sites with $\beta \simeq 5$, which is typical for ODSs [13], is described by Eq. (21) with $C \simeq 0.40$, which is very close to $C \simeq 0.42$ given by the EGDM. At β values different from approximately 5, the APAE should be preferred.

VII. DISCUSSION

It is necessary to emphasize that the APAE is neither a new model nor a new theory. The APAE reproduces the well-known theory [1,2,4,12,16–22,36] represented by Eqs. (3)–(11) applied to the well-known model (GDM) [1–4,7–9,12,13,20–25] described by Eqs. (1) and (2). The remarkable feature of the APAE is its ability to replace Eqs. (1)–(11) in the form of a single user-friendly equation checked by computer simulations in the framework of the GDM. The GDM is chosen because of its success in accounting for the broad manifold of experimental data, as has been highlighted in numerous monographs, edited books, and review articles. There is no need to review once again the successful applications of the GDM in the form of the APAE.

The simple structure of the APAE, as a single equation that includes all the relevant parameters of the GDM, might be favorable for implementation in device-simulation software packages. Currently, such packages use the EGDM, praising the latter as the state of the art in the theoretical description of $\mu(T, n, F)$ in ODSs.

It is further necessary to emphasize that the APAE and the EGDM are not two different models, or two different theories. The APAE and EGDM are solutions for $\mu(T, n, F)$ in the framework of the very same model, the GDM, described by Eqs. (1) and (2). While the APAE is the result for $\mu(T, n, F)$ in the framework of the GDM that contains spatial randomness and allows for variation of all the parameters in Eq. (5), the EGDM is the result for $\mu(T, n, F)$ in the framework of a reduced GDM. In this reduced version, sites are placed on a cubic lattice and the parameter $\beta = N^{-1/3}/a$ has a fixed value of 10. The comparison in Fig. 4 between the dashed orange line (lattice) and the black dots (random sites) illustrates how strongly the lattice assumption affects the dependence $\mu(F)$ at $\beta = 10$. However, $\beta = 10$ is probably not the optimal choice for ODSs. Estimates for N and a in the literature [2,7,13,18,27] favor rather values of $\beta = N^{-1/3}/a$ close to 5. Remarkably, the data in Fig. 4 for the lattice model at $\beta = 10$ (EGDM) do not strongly deviate from the data for spatially random systems at $\beta \approx 5$. This means that the EGDM can be used, by chance, for spatially disordered ODSs with $\beta \approx 5$. The same cannot be claimed for systems with $\beta \approx 10$.

With respect to the validity of the GDM described by Eqs. (1) and (2) and used for both the EGDM and the APAE, the following should be noted. While Eq. (1) has been thoroughly validated [4] by comparison with experimental data on $\mu(T, n, F)$ in polymers [29,48], we are not aware of solid proofs for the validity of Eq. (2), i.e., of the MA expression for hopping rates in ODSs. One could

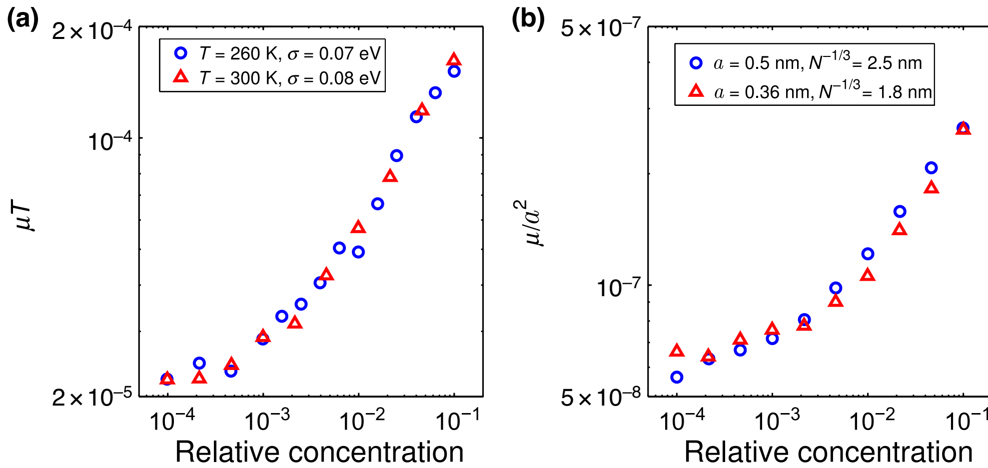

FIG. 5. Proof that $\alpha = kT/\sigma$ and $\beta = N^{-1/3}/a$ are the critical parameters of the GDM and not separately kT, σ, $N^{-1/3}$, and a. (a) $\alpha = kT/\sigma$ has the same value of approximately 0.32 for both sets of data. (b) $\beta = N^{-1/3}/a$ has the same value of 5 for both sets of data.

suppose that, for instance, Marcus transfer rates, which include polaron effects, would be more suitable for soft organic materials [49]. A discussion of the appropriateness of the use of MA hopping rates described by Eq. (2) is beyond the scope of the present work. One can find some analysis in review articles [15,21,50]. It is the success of the GDM with MA rates to explain experimental data that makes this model so popular. The MA rates were used in the most-cited reviews in the context of charge transport in ODSs [9,13]. We use Eq. (2) because this expression for hopping rates was used in the widely used EGDM [7–9] implemented in device-simulation software. We suggest use of the APAE instead of the EGDM, not questioning the GDM with MA rates, but rather questioning the reduced version of this model in the form of a regular cubic lattice with fixed $\beta = N^{-1/3}/a = 10$ (EGDM). The GDM with MA rates was recently used in numerous theoretical studies of $\mu(T, n, F)$ in ODSs [30,50–53], particularly, in three recent studies published in this journal [1–3].

These very recent studies [1–3] were based on the same model with the same set of parameters as our study. Two of them [1,2] used the same set of analytical equations, although not replacing this set by a single equation, like the APAE. Remarkably, these studies [1–3] analyzed recent experimental observations in ODSs desired for applications in transistors and photovoltaic devices. Considering analytical equations (1)–(11) for a system of spatially random sites, encoded in our paper in the form of the APAE, Upreti *et al.* [1] and Lee *et al.* [2] concluded that these analytical equations account for experimental observations in all cases, while the EGDM does not. Sun *et al.* [3], although not going beyond the model based on a cubic lattice, concluded that the value of $\beta = N^{-1/3}/a$ necessary to account for experimental data is not 10 as fixed in the EGDM, but rather approximately 3.6. These conclusions might encourage researches to pay attention to the APAE, which is not restricted to a cubic lattice and to a fixed value $\beta = 10$. Therefore, we discuss the recent studies [1–3] based on the same set of parameters in more detail.

Upreti *et al.* [1] considered an analytical theory for $\mu(T, n, F)$ similar to that described in Sec. III and performed Monte Carlo simulations used to calibrate parameters in the analytical theory. Their theory was further implemented in the drift-diffusion solver. Along with the theoretical development, Upreti *et al.* [1] fabricated hole-only and electron-only organic devices and performed measurements of temperature-dependent space charge–limited currents. It was found that the suggested theory can adequately describe electron and hole transport in a wide variety of organic semiconductor blends, which are used as the active layer in typical bulk heterojunction organic solar cells [54]. It was also recognized that the EGDM fails to produce an acceptable fit for experimental data obtained on electron-only devices [1]. Upreti *et al.* supposed that the reason for the failure of the EGDM in electron-only devices is a small value of the parameter $\beta = N^{-1/3}/a = 2$, which favors the VRH mechanism, while the EGDM with the large fixed parameter $\beta = 10$ presumably favors the nearest-neighbor hopping mechanism. However, Monte Carlo simulations have proven [6] that the EGDM is based on the VRH transport mechanism, although on a cubic lattice without spatial disorder. Therefore, more study is needed to clarify why the EGDM fails to account for experimental data in electron-only devices, despite being capable of describing the data in hole-only devices reported by Upreti *et al.* [1].

Lee *et al.* [2] also addressed an analytical theory for $\mu(T, n, F)$ described in Sec. III. They implemented the results in a technology-computer-aided-design simulation tool, ATLAS [version 5.30.0.R (2020)], from Silvaco and deduced parameters of the GDM by comparison with experimental data for ODSs with high carrier mobilities. Only the case of low electric fields was considered by Lee *et al.* [2], who highlighted that the GDM with spatial and energy disorder is superior to the EGDM, which

lacks spatial disorder and uses only one fixed value $\beta = N^{-1/3}/a = 10$ on a simple cubic lattice.

Neither Upreti *et al.* [1] nor Lee *et al.* [2] attempted to replace the set of analytical equations by a single closed-form expression for $\mu(T, n, F)$ as, for example, the APAE derived in Sec. IV.

Sun *et al.* [3] improved the EGDM, although not going beyond the model based on a cubic lattice. One improvement is the replacement of the constants $\mathfrak{c}_1$ and $\mathfrak{c}_2$ in Eq. (24b) by functions of the parameter $\beta = N^{-1/3}/a$. The other improvement is the replacement of the lattice constant b in Eq. (23) by $b = 10a$ for use of the localization length a as an adjustable parameter [3]. Sun *et al.* [3] solved analytically the degenerate drift-diffusion equation and extracted model parameters for several organic diodes by comparison with experimental data.

These studies have not revealed the combinations of parameters given in Eq. (5) as parametrization of the theoretical model. Instead, the parameters $N^{-1/3}$ and a and the parameters σ and kT were treated separately from each other. To emphasize once again the validity of the parametrization given in Sec. II B, we use in Fig. 5 the data of Upreti *et al.* [1] obtained by Monte Carlo simulations for the dependence $\mu(n)$ at different values of the parameters $N^{-1/3}$, a, σ, and kT. While Upreti *et al.* [1] plotted their numerical data for $\mu(n)$ as a function of $N^{-1/3}$ at fixed a and as a function of a at fixed $N^{-1/3}$, as well as a function of σ at fixed T and a function of T at fixed σ, we have chosen their combinations of $N^{-1/3}$ and a, as well as the combinations of T and σ, which correspond to the same values of the ratios $\alpha = kT/\sigma$ and $\beta = N^{-1/3}/a$. The plots illustrate that $\alpha = kT/\sigma$ and $\beta = N^{-1/3}/a$ control $\mu(n)$ and not separately $N^{-1/3}$, a, σ, and kT. This is the reason why the APAE introduced in Sec. IV is formulated in terms of only three parameters—α, β, and γ—determined in Eq. (5).

VIII. CONCLUSIONS

The APAE formulated for $\mu(T, n, F)$ in Sec. IV is well justified by classical theories of hopping transport. It is calibrated by Monte Carlo simulations in the framework of the GDM. It is user-friendly and can be easily implemented in device-simulation software.

The EGDM [7], desired to fit simulation data for $\mu(T, n, F)$ in the framework of the GDM on a cubic lattice at fixed $\beta = N^{-1/3}/a = 10$, occasionally agrees with simulation data for a system of random sites, although at $\beta \simeq 5$. For values of β other than approximately 5, the APAE should be preferred.

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