

Effect of doping on the density-of-states distribution and carrier hopping in disordered organic semiconductors

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The effect of doping on the density-of-states (DOS) distribution and charge-carrier transport in a disordered hopping system is considered analytically. It is shown that doping such a system produces a random distribution of dopant ions, which Coulombically interact with carriers localized in intrinsic hopping sites. This interaction further increases the energy disorder and broadens the deep tail of the DOS distribution. Therefore, doping of a disordered organic semiconductor, on the one hand, increases the concentration of charge carriers and lifts up the Fermi level but, on the other hand, creates additional deep Coulombic traps of the opposite polarity. While the former effect facilitates conductivity, the latter strongly suppresses the carrier hopping rate. A model of hopping in a doped disordered organic semiconductor is suggested. It is shown that the doping efficiency strongly depends upon the energy disorder and external electric field.

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I. INTRODUCTION

Charge-carrier hopping within a positionally random and energetically disordered system of localized states was shown to be an adequate model for the description of both equilibrium and nonequilibrium conductivity in noncrystalline organic semiconductors.^{1–3} In a positionally random system, the energy disorder is to a large extent caused by random positions and orientations of molecules via Van der Waals and dipole-dipole interactions.^{4,5} Doping such a system by charged moieties will, in addition, create a random distribution of dopant ions that will Coulombically interact with carriers localized in randomly located intrinsic hopping sites^{6,7} and, thus, broaden the effective density-of-states (DOS) distribution. This effect is especially important in molecular semiconductors because the dielectric permittivity is low and, concomitantly, the range of Coulomb potential is large in organic solids.

Doping of a disordered organic semiconductor by charged moieties has two counteracting effects. On the one hand, it increases the concentration of charge carriers and lifts up the Fermi level^{8,9} but, on the other hand, it increases energetic disorder. While the former effect facilitates conductivity, the latter strongly suppresses the carrier hopping rate and, therefore, the mobility. The latter effect can dominate at some dopant concentrations such that doping appears to be even counterproductive as far as the carrier mobility is concerned.^{8,10}

Upon doping by ionized moieties, charge neutrality must be maintained. There are two ways to accomplish this. One is electrochemical doping. If the ionization (or reduction) potential of the electrolyte electrode more or less matches the

highest occupied molecular orbital (HOMO) of the organic semiconductor, mobile majority carriers can be injected provided that the electrolyte supplies appropriate counterions that can diffuse into the semiconductor. An example is the work of Jiang *et al.*,¹⁰ who injected holes from a solution of 0.1 M tetraethylammonium perchlorate into a film of polyhexylthiophene. Charge injection was compensated for by concomitant doping with (permanent) perchlorate anions. The alternative method is doping by a neutral entity whose electron affinity is large enough (or its ionization energy is small enough) to allow for charge transfer from the semiconductor to the dopant.¹¹ Dictated by the redox potential of donor-acceptor systems and, concomitantly, the dissociation enthalpy, complete charge transfer and creation of free carriers in the dark should in practice never be possible, i.e., full charge transfer should be an endothermic and reversible process.

Both modes of doping resemble the situation Onsager had in mind when he developed his 1934 and 1938 treatments of ionic and radiation-induced conductivity.^{12,13} In both cases, excess mobile majority carriers and immobile countercharges (ions) are generated that roughen the energy landscape in which the carriers migrate, but in the “neutral” doping case also charge redistribution can and does occur. The majority of charge carriers will actually form metastable geminate pairs whose dissociation is facilitated by the ambient phonon bath and external electric field. Therefore, the average hopping rate is controlled by the carrier release from the Coulomb traps, i.e., by the Onsager-type dissociation of metastable geminate pairs. This process determines both the field and, together with intrinsic disorder, temperature dependences of the mobility.

A high carrier density can be reached without introducing countercharges due to either the field effect or high level of monopolar charge injection across a contact. Under these circumstances, the Coulomb interaction between carriers can also strongly change the effective potential landscape and even make it strongly fluctuating in time. However, the interaction between charges of the same sign is repulsive and, therefore, cannot create Coulomb traps. Instead, it gives rise to fluctuating in time potential barriers that still affect the mobility, although this effect is significantly weaker than the effect of Coulomb traps. The broadening of the DOS distribution is, therefore, much smaller and the width of its deeper tail is affected much more weakly.

In this article we shall first calculate the dopant-induced DOS distribution and then consider the effect of dopant-induced Coulomb traps on the charge transport in doped disordered organic semiconductors.

II. DOS DISTRIBUTION IN A DOPED DISORDERED ORGANIC MATERIAL

Two important distinctions between crystalline and disordered semiconductors should be borne in mind when considering doping effects: (i) in a disordered organic material, all electronic states are in fact localized hopping sites and (ii) due to typically low values of the dielectric constant the Coulomb interaction between charged species can strongly affect the energy of localized states. Consistent account for these two characteristic features of disordered organics will be the roadmap of the present study.

Let us consider a host material with the HOMO energy of $E_{HOMO}^{(host)}$ doped by a potential dopant with the lowest unoccupied molecular orbital (LUMO) of the energy $E_{LUMO}^{(dop)}$. Obviously, carriers will be transferred from dopants to guest HOMO states and, concomitantly, all dopants will be ionized if $E_{HOMO}^{(host)} > E_{LUMO}^{(dop)}$. However, practically all dopants can be ionized even if this condition is not fulfilled because the energy deficit Δ for charge transfer from HOMO of the host to LUMO of the dopant can be compensated for by the Coulomb interaction between ionized dopants and released charge carriers localized in nearby hopping sites, i.e., if

$$\Delta = E_{LUMO}^{(dop)} - E_{HOMO}^{(host)} \leq \frac{e^2}{4\pi\epsilon_0\epsilon a}, \quad (1)$$

where e is the elementary charge, a is the distance between a dopant and the nearest intrinsic hopping site, ϵ_0 is the dielectric permittivity, and ϵ is the relative dielectric constant. For $a=0.6$ nm and $\epsilon=3$, Eq. (1) yields $\Delta=0.8$ eV, indicating that carriers can be released from dopants even if their LUMOs are well above the HOMO of the host material. It should be noted that the above estimate disregards intrinsic energetic disorder in the host material. Recently, it has been shown that this effect further enhances ionization of “deep” dopants.¹⁴

An ionized dopant, embedded in a random network of localized states, affects the energy of nearby hopping sites due to Coulomb interaction of the dopant with charge carriers localized in those sites. For a given hopping site, the probability density, $w(r)$, of having a nearest dopant ion at a

distance r is determined from the Poisson distribution as

$$w(r) = 4\pi r^2 N_d \exp\left(-\frac{4\pi}{3} N_d r^3\right), \quad (2)$$

where N_d is the concentration of ionized dopant atoms. A carrier, trapped by this localized state, will Coulombically interact with the dopant ion and the potential energy of this interaction, E_c , is

$$E_c = -\frac{e^2}{4\pi\epsilon_0\epsilon r}. \quad (3)$$

This energy, added to the intrinsic disorder energy, E_i , yields the total energy E of the hopping site: $E=E_c+E_i$. In the present work, we consider a relatively weak doping when the concentration of dopant ions remains much smaller than the total density of intrinsic hopping sites, N_i . Under these conditions, the energy of almost every localized state will be essentially affected only by the nearest dopant ion. Combining Eqs. (2) and (3) yields a distribution of the localized states over the Coulomb binding energy, $g_c(E_c)$. The result reads

$$g_c(E_c) = w[r(E_c)] \left| \frac{dr}{dE_c} \right| = \frac{4\pi e^6 N_d}{(4\pi\epsilon_0\epsilon)^3 E_c^4} \exp\left[\frac{4\pi N_d}{3} \frac{e^6}{(4\pi\epsilon_0\epsilon E_c)^3}\right]. \quad (4)$$

The distribution of hopping sites over the total energy must account for both the intrinsic DOS $g_i(E_i)$ and the distribution over the Coulomb energy $g_c(E_c)$ given by Eq. (4). Integrating over E_c and E_i with the condition $E_c+E_i=E$ leads to the following expression for the energy distribution function $g(E)$ in a doped material:

$$g(E) = \frac{4\pi e^6 N_d}{(4\pi\epsilon_0\epsilon)^3} \int_{-\infty}^0 \frac{dE_c}{E_c^4} \exp\left[\frac{4\pi N_d}{3} \frac{e^6}{(4\pi\epsilon_0\epsilon E_c)^3}\right] \times \int_{-\infty}^{\infty} dE_i g_i(E_i) \delta(E - E_c - E_i), \quad (5)$$

where δ is the Dirac delta function. Evaluating the integral over E_i on the right-hand side of Eq. (5) yields

$$g(E) = \frac{4\pi e^6 N_d}{(4\pi\epsilon_0\epsilon)^3} \int_{-\infty}^0 \frac{dE_c}{E_c^4} \exp\left[\frac{4\pi N_d}{3} \frac{e^6}{(4\pi\epsilon_0\epsilon E_c)^3}\right] g_i(E - E_c). \quad (6)$$

It is worth noting that Eq. (6) ignores both the Coulomb interaction between mobile charge carriers and contributions of non-nearest dopant ions to the Coulomb energy of localized states. At low dopant concentrations, the validity of this approximation is obvious. It can be also justified in heavier doped materials because dopant ions and carriers form relatively short pairs. Dipole moments of these pairs do affect the potential energy of more distant carriers but this effect is weaker as compared to the interaction with the nearest dopant ion. This argument is not, of course, valid in very heavily doped amorphous materials in which N_d approaches N_i and

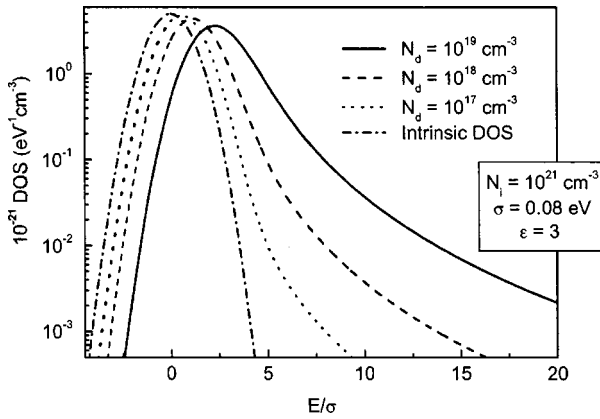


FIG. 1. The effect of doping on the DOS distribution in a disordered organic semiconductor. The Coulomb interaction between ionized dopants and charge carriers creates additional deep traps and broadens the deep tail of the DOS.

different pairs start to overlap. Since the value of N_i in disordered organic semiconductors is typically around 10^{20} – 10^{21} cm^{-3} , the formulated model is applicable at dopant concentrations up to 10^{18} – 10^{19} cm^{-3} .

The effect of doping on an intrinsically Gaussian DOS distribution is shown in Fig. 1 for several doping levels. An increasing concentration of dopant ions converts an increasingly large number of shallow sites into deep states. One should, therefore, expect that the doping-induced change in the DOS distribution would immediately affect the carrier hopping mobility. However, the density of mobile carriers also increases upon doping. Under equilibrium conditions, these carriers fill deep states and lift up the quasi-Fermi energy. For a fixed DOS distribution, this would lead to a higher average hopping rate and, concomitantly, higher carrier mobility. Thus, the mobility is determined by the interplay of two competing doping-induced processes. On the one hand, increasing the density of mobile majority carriers shifts their energy distribution over localized states towards shallower sites, which facilitates the mobility. On the other hand, the Coulomb interaction with localized dopant ions of the opposite polarity effectively raises the density of states in the deep tail of the DOS distribution and produces additional deep traps, which suppresses the jump rate and reduces the mobility. Below, we show that this interplay may lead to a nonmonotonic dependence of the mobility upon the dopant concentration.

III. HOPPING IN A DOPED ORGANIC SEMICONDUCTOR

At first glance, the calculation of the carrier mobility in a doped hopping system is straightforward: One may just use the doping-modified DOS distribution, given by Eq. (6), in the equations of the variable-range hopping model. However, this simple approach would significantly overestimate the role of the dopant-induced deep Coulomb traps. The reason is that the DOS itself does not contain information about correlations between energies and positions of the localized states. Since the typical Onsager radius of a single Coulomb trap in an organic material is 10–20 nm and this trap consists

of several hundreds of hopping sites, the correlations are essential as far as the carrier release from such a trap is concerned.

The energetic requirements for dissociative doping by an originally originally neutral dopant will be discussed first. Experimentally, it is known that an impurity can serve as, for instance, an electron acceptor in an organic semiconductor even if the LUMO of the dopant is ~ 1 eV above the HOMO of the host molecules. Intuitively, it is not clear how the charge transfer can occur from a host molecule to a dopant under such circumstances. In order to clarify the situation, one should bear in mind that both HOMO and LUMO energies are defined for isolated charges disregarding Coulomb interactions and/or intrinsic fields. However, in amorphous organic materials, charge transfer from a host molecule to a dopant should directly produce a strongly Coulombically bound short geminate pair rather than a free carrier. The size of such a pair is equal to the intermolecular distance that is typically 0.6–1.0 nm. The Coulomb binding energy of this pair is then 0.5–0.8 eV if the permittivity retains its typical macroscopic value of 3 and 0.8–1.2 eV if the permittivity goes down to 2 at such short distances. If this energy gain is sufficient to compensate for the charge-transfer energy, the geminate pair of charges rather than a neutral dopant and a neutral host molecule will form the ground state in a doped material.

Even if a carrier has been transferred from a dopant to a host molecule, it cannot immediately contribute to the dc conductivity due to the Coulomb interaction that still bounds it to the parent dopant ion. A carrier can be released from a Coulomb trap in the course of a multijump Onsager-like process facilitated by the external electric field. Exact analytic consideration of this process, including correlations between energies and positions of hopping sites within Coulomb potential wells, is hardly possible and one has to formulate a simplified model that still retains essential details of the carrier kinetics. We suggest a model based on the following simplifications: (i) every collective Coulomb trap surrounding a localized counterion is replaced by a single deep localized state nearest to the ionized dopant and (ii) the energy of this site is a sum of the intrinsic disorder energy and the electrostatic energy Δ counted from the top of the potential barrier, which is formed by the Coulomb and external fields as

$$\Delta = \sqrt{\frac{e^3 F}{\pi \epsilon_0 \epsilon}} - \frac{e^2}{4 \pi \epsilon_0 \epsilon a}. \quad (7)$$

Under these assumptions, the effective DOS distribution in a doped material takes the form

$$g(E) = \frac{N_i - N_d}{N_i} g_i(E) + \frac{N_d}{N_i} g_i \left(E + \frac{e^2}{4 \pi \epsilon_0 \epsilon a} - \sqrt{\frac{e^3 F}{\pi \epsilon_0 \epsilon}} \right). \quad (8)$$

A model of variable-range hopping in a disordered material at a finite carrier density has been formulated in Refs. 8 and 15. This model is based on the concept of the effective

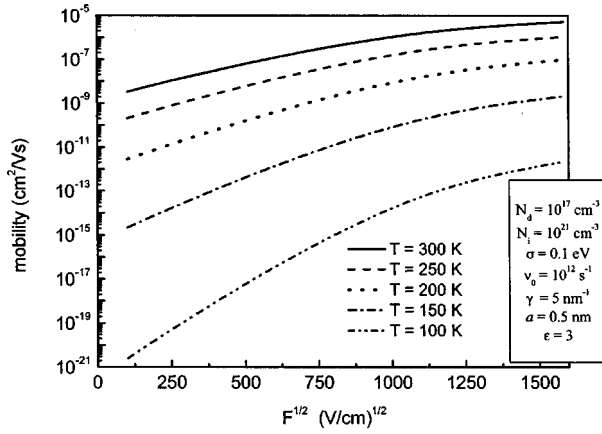


FIG. 2. Field dependences of the charge-carrier mobility in a doped disordered organic semiconductor at different temperatures.

transport energy^{15–18} that virtually reduces the problem to trap-controlled transport with a broad energy distribution of localized states.¹¹ In the following, we always assume that the material is macroscopically neutral, i.e., that the average density of carriers is equal to the concentration of dopants. The field dependence of the mobility, calculated with the DOS distribution given by Eq. (8) at a moderate concentration of dopants $N_d = 10^{17} \text{ cm}^{-3}$, is shown in Fig. 2 parametric in temperature. A Gaussian distribution of the width $\sigma = 100 \text{ meV}$ has been used as an intrinsic DOS distribution. Although the curves follow the Poole-Frenkel-type $\log \mu \propto F^{1/2}$ dependence at weaker fields, they tend toward saturation at stronger fields. Figure 3 illustrates the temperature

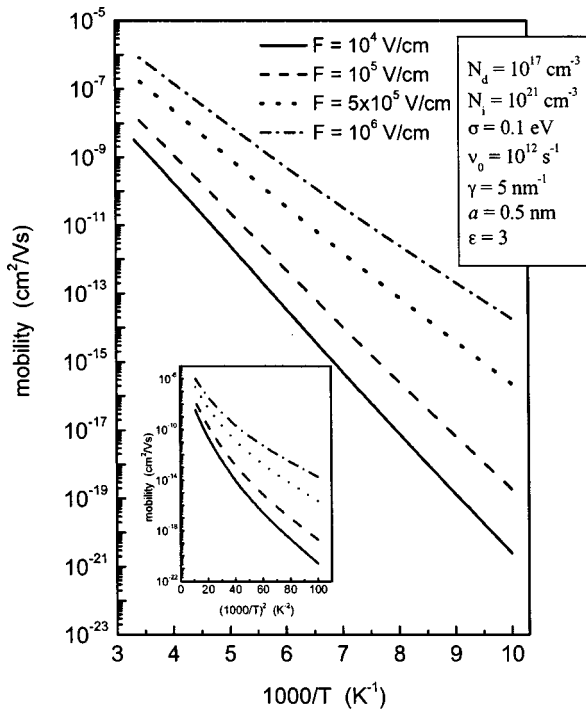


FIG. 3. Temperature dependence of the mobility in a doped disordered organic material. The inset shows the same set of curves replotted in $\log \mu$ vs $1/T^2$ axes.

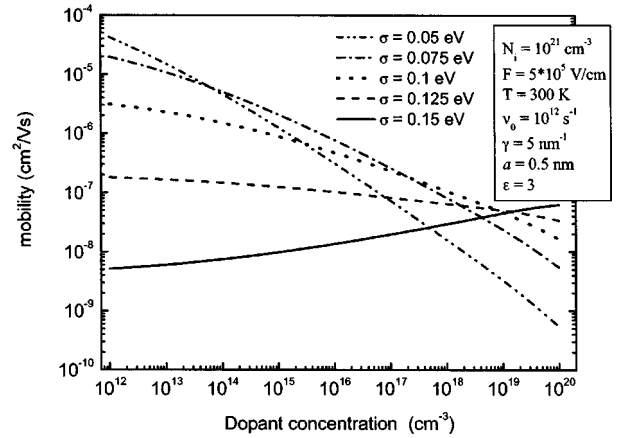


FIG. 4. Dependence of the carrier mobility upon the concentration of dopants in materials with different variations of the intrinsic DOS distribution.

dependence of the mobility at different external fields. Although both the doping-induced Coulomb traps and the intrinsic DOS distribution affect this dependence, most carriers are localized in the former, which gives rise to an almost perfect Arrhenius temperature dependence with the slope affected by the external field. As shown in the inset to Fig. 3, an attempt to visualize these data on a $\log \mu$ versus $1/T^2$ plot fails to yield straight lines, indicating that the mobility is effectively controlled by carrier jumps from states around the Fermi level.^{8,15} One should expect that, at lower temperatures, the effective transport level should approach the Fermi level, and the temperature dependence of the mobility has to almost level off featuring the Mott $T^{-1/4}$ law.

Figure 4 illustrates the dopant concentration dependence of the mobility parametric in the width of the intrinsic Gaussian DOS distribution. These dependencies are strikingly different in materials with weak and strong energy disorder, i.e., with small and large values of the DOS width. While doping a weakly disordered system suppresses the mobility, the latter increases with doping level in strongly disordered materials. It should be noted, however, that the mobility always decreases with doping more weakly than $1/N_d$ and, therefore, the conductivity, which is proportional to the product of μ and N_d , increases upon doping even in materials with small DOS widths. Decreasing carrier mobility upon doping of a weakly disordered conjugated polymer was observed in Ref. 10.

In order to understand why the mobility in weakly and strongly disordered materials is so differently affected by doping, one should bear in mind that dopants provide both charge carriers and deep Coulomb traps. If these traps are deeper than those states that control the mobility in the pristine material, the deep Coulomb traps will still trap the majority of doping-induced carriers and their mobility has to be smaller than the carrier mobility in the undoped material. The electrostatic energy of a Coulomb trap can be estimated from Eq. (7) as 0.5 eV in a field of 1 MV/cm with $a = 0.5 \text{ nm}$ and $\epsilon = 3$. However, the effective depth of a Coulomb trap is smaller due to the fact that carriers can escape from this trap by jumps via localized states with energies below the maximum of the DOS distribution.^{8,15–18} The ac-

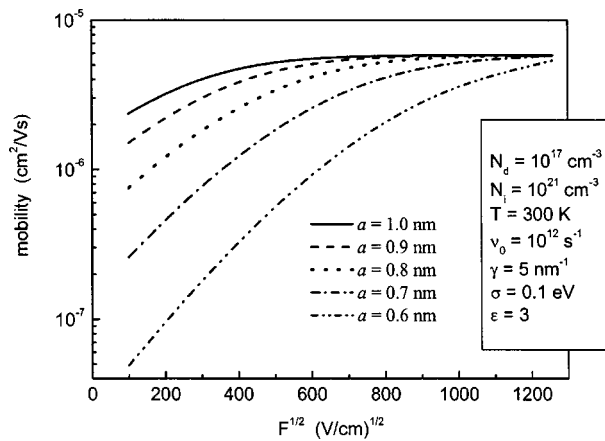


FIG. 6. Field dependences of the mobility calculated for different distances between the dopant ions and nearest hopping sites, i.e., for different effective depths of the Coulomb potential wells.

tivation energy of the mobility can be estimated from the curves plotted in Fig. 3, and for the field of 1 MV/cm this energy is only 0.36 eV.

In a pristine material with a Gaussian DOS, the distribution of localized carriers has a maximum at the energy E_m of σ_i^2/kT below the maximum of the intrinsic DOS function. In a strongly disordered material with $\sigma_i = 120$ meV, the energy E_m is as large as 0.6 eV at room temperature. This energy is larger than the activation energy of the Coulomb traps and carriers can easily leave the latter and fill the deep tail of the intrinsic DOS at energies below and above E_m . Concomitantly, the Fermi level elevates, which leads to increasing mobility upon doping. In other words, disordered organic materials can be efficiently doped by introducing virtually deep Coulomb traps because free equilibrated carriers fill states in the deep tail of the intrinsic DOS distribution that are even deeper than the Coulomb traps. It should also be noted that, at high doping levels, Coulomb potential wells of neighboring dopants strongly overlap, which leads to smoothing of the potential landscape. Under such circumstances, the effect of trap filling takes over and the mobility

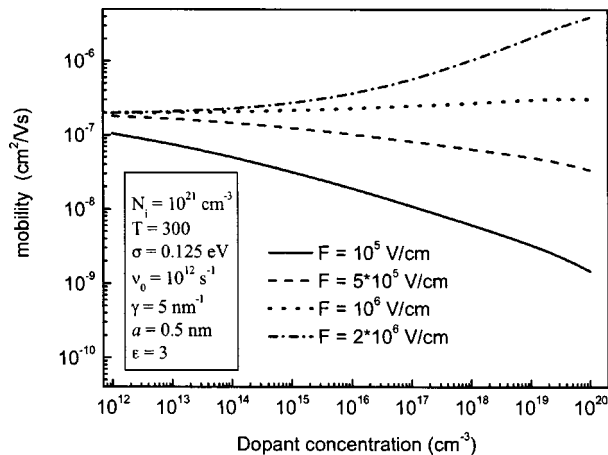


FIG. 5. Dependence of the carrier mobility upon the concentration of dopants at different external fields.

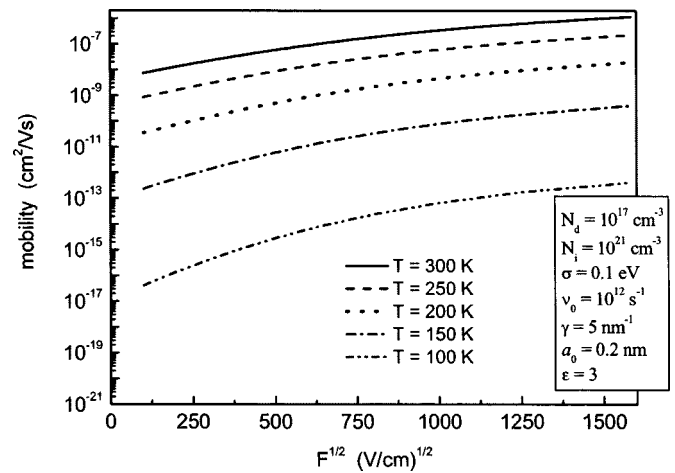


FIG. 7. Field dependences of the mobility calculated for different widths of the Gaussian distribution of distances between the dopant ions and nearest hopping sites.

steeply increases even in weakly intrinsically disordered materials.¹⁰

If the activation energy of the dopant-induced Coulomb traps is larger than E_m , most doping-induced carriers are still localized within Coulomb potential wells of ionized dopants and in the deep tail states below E_m . The dominant effect of doping is then a creation of additional deep states in the DOS and, concomitantly, the mobility decreases with increasing N_d . However, this decrease is weaker than $1/N_d$ and the conductivity, determined by the product of the mobility and carrier density, still increases with increasing dopant concentration.

It is known from both experimental studies and theoretical considerations that the mobility must strongly increase at high doping levels.^{8–10} However, this effect cannot be analyzed within the framework of the present model because the latter is valid only at relatively low doping levels when the Coulomb potential wells of ionized dopants do not overlap. The increase of the mobility at high values of N_d is associated with filling of deep tail states by carriers. This is possible only if adding new dopants does not create new deep Coulomb traps, which is the case at very high dopant concentration when Coulomb potential wells already strongly overlap and additional ionized dopants smooth rather than roughen the potential landscape.⁸

Since the effective depth of Coulomb traps is controlled by the external field, one should expect different dopant-concentration dependencies of the mobility at weak and strong electric fields. This effect is illustrated in Fig. 5. Indeed, at weak external fields, Coulomb potential wells are deep and ionized dopants serve as deep traps for carriers. Strong external fields reduce the barrier for carrier release from Coulomb traps, making them shallower and, thereby, increasing the density of free carriers and the average carrier mobility. It is interesting that the effect of the external field on the effective depth of a Coulomb trap does not depend upon the field direction. Therefore, carriers in the channel of an organic field-effect transistor should not experience the Coulomb trapping by dopant ions due to a strong vertical

field, and their mobility along the channel should increase with doping level even if the lateral field is weak.

The zero-field activation energy of a Coulomb trap strongly depends upon the distance a between an ionized dopant and the nearest intrinsic hopping site, i.e., upon the size of the equilibrium dark “geminate pair” formed by the dopant ion and a charge carrier occupying the nearest site. Concomitantly, this parameter controls the critical value of the external field at which the negative dopant-density dependence of the mobility is changed to positive. The field dependences of the mobility are shown in Fig. 6 for different sizes of the dark geminate pairs. As one could anticipate, the critical value of the field, at which the effect of the Coulomb traps vanishes and the mobility saturates, strongly increases with decreasing a .

It should be noted that all our calculations have been done for a fixed size of the dark geminate pairs. However, one should expect a distribution of the parameter a in a disordered material. The curves shown in Fig. 7 were calculated for the following normalized Gaussian distribution, $f(a)$, of pair sizes:

$$f(a) = 8 \sqrt{\frac{2}{\pi}} \frac{a^2}{a_0^3} \exp\left(-\frac{a^2}{2a_0^2}\right), \quad (9)$$

where a_0 is the width of the distribution. The value of $a_0 = 0.2$ nm was used in the calculations. With this value of a_0 , the function $f(a)$ has a maximum at $a \approx 0.5$ nm, i.e., at the same pair size as has been used in the mobility calculations with a fixed value of a ; see, e.g., Fig. 2. Since larger values of a correspond to shallower Coulomb traps, larger pairs give a major contribution to the mobility at weaker fields. Concomitantly, the weak-field mobility is higher in a material with a distribution of pair sizes. Very deep Coulomb traps, formed by short pairs, can keep carriers even at strong fields and, therefore, the strong-field mobility turns out to be smaller in materials with varying sizes of charge-dopant pairs. As a result, the slope of the $\log \mu$ versus $F^{1/2}$ curves decreases as compared to those calculated with a fixed value of a .

It should be noted that the results, discussed above, were obtained under the assumption that the density of charge carriers is equal to the density of dopants, i.e., that the field-driven carrier ejection from a sample is fully compensated by charge injection and vice versa. This condition can be violated if a blocking contact is used, which is typical for the time-of-flight (TOF) measurements. Upon application of an external electric field, all mobile carriers will sooner or later be extracted from the sample and only Coulomb traps surrounding counterions will remain in the bulk. In a heavily doped material, this will result in the formation of a zone at the blocking contact that is depleted of mobile carriers. However, in an accidentally doped (apparently pristine) material with a low density of dopant ions, the field can still remain almost constant. In order to simulate the TOF mobility, measured in such samples, one has to use the DOS distribution given by Eq. (8) and assume the density of photogenerated carriers much smaller than the dopant concentration. The use

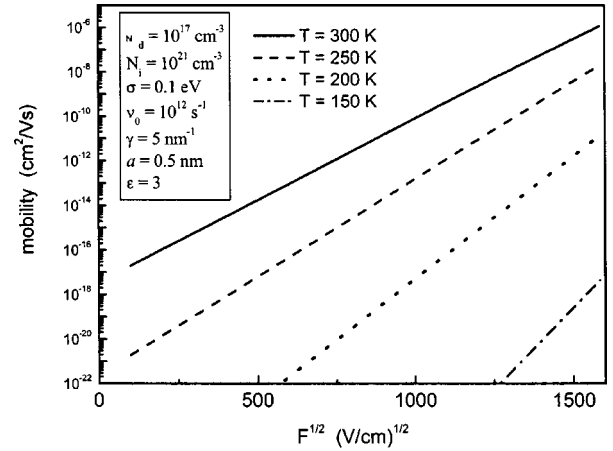


FIG. 8. Field dependences of the single-carrier (TOF) carrier mobility in a doped disordered organic material at different temperatures.

of this model yields the mobility that is orders of magnitude smaller than at higher carrier densities and reveals a perfect Poole-Frenkel field dependence within the entire field range as illustrated in Fig. 8. This result offers a plausible explanation of the notorious difference^{8,9} between both the magnitudes and field dependences of the field-effect and space-charge-limited-current mobility on the one hand, and the mobility measured in TOF experiments on the other.

IV. CONCLUSIONS

The density-of-states distribution in a disordered organic semiconductor is strongly affected by doping. Due to the Coulomb interaction between released charge carriers and ionized dopants, the deep tail of the intrinsically Gaussian DOS distribution broadens and the total density of deep localized states strongly increases with increasing dopant content. Therefore, doping of a disordered organic semiconductor, on the one hand, increases the concentration of charge carriers and lifts up the Fermi level but, on the other hand, creates deep Coulomb traps. While the former effect facilitates conductivity, the latter strongly suppresses the carrier hopping rate. The trade-off between increasing densities of both charge carriers and Coulomb traps is controlled by the intrinsic DOS width and external field strength. In strongly disordered materials and/or at strong electric fields, the carrier mobility increases with increasing dopant concentration. Otherwise, carrier localization in Coulomb potential wells takes over and the mobility decreases upon doping. The TOF measurements in intentionally or accidentally doped samples yield the Poole-Frenkel-like field dependence of the mobility that is orders of magnitude smaller than the mobility measured at high carrier densities.

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