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Letter

On Analytical Modeling of Hopping Transport of Charge Carriers and Excitations in Materials with Correlated Disorder

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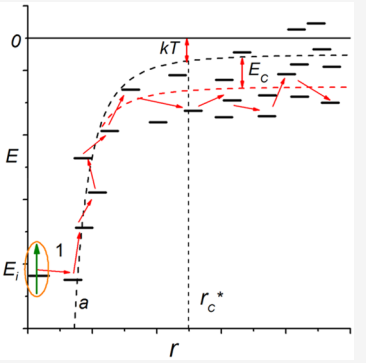
ABSTRACT: Spatial-energy correlations strongly influence charge and exciton transport in weakly ordered media such as organic semiconductors and nanoparticle assemblies. Focusing on cases with shorter-range interparticle interactions, we develop a unified analytic approach that allows us to calculate the temperature and field dependence of charge carrier mobility in organic quadrupole glasses and the temperature dependence of the diffusion coefficient of excitons in quantum dot solids. We obtain analytic expressions for the energy distribution of hopping centers, the characteristic escape time of charge/exciton from the energy well stemming from energy correlations around deep states, and the size of the well. The derived formulas are tested with Monte Carlo simulation results, showing good agreement and providing simple analytic expressions for analysis of charge and exciton mobility in a broad range of partially ordered media.



Organic semiconductors, quantum dot (QD) solids, and other partly ordered condensed matter systems are actively studied as promising materials for solar energy, light-emitting, and other optoelectronic devices.[1−5] Transport of both charge and excitation has a large effect on the performance of such devices. Despite the differences in chemical composition, it has been shown that, from the point of view of transport, disordered organic semiconductors and QD solids have much in common: energetic disorder can lead to localization of electron and hole states and, consequently, to the hopping transport mechanism.[6,7] It is known that spatial-energy correlations significantly affect transport in organic semiconductors, especially materials composed of molecules with large dipole moments, when the correlations are the most long-range (dipole glasses).[8,9] Typically, hopping transport in materials with correlated disorder is modeled by numerical Monte Carlo (MC) methods,[8,9] but recently an analytic approach has been developed.[10] Correlations arise due to electrostatic interactions (in the case of dipole glass, interactions of charges with randomly oriented dipoles). The effect of correlations arising from weaker interactions, the range of which is shorter, is less clear. In this work, an analysis has been carried out for two such cases: charge transfer in quadrupole glasses[11,12] and diffusion of triplet excitons in QD solids, using a unified formalism. The energy density of states (DOS), which controls the hopping transport of excitons in QD solids, is also calculated.

It is known that excitons in QD solids can diffuse over distances much greater than the size of a QD.[13−15] Note that the transfer of triplet excitons between QDs occurs according to the Dexter mechanism, which is a charge exchange mechanism; therefore, the hopping transport model can also be considered for triplet excitons.[16] Large intrinsic dipole moments of QDs[17−19] create energy correlations. In previous work,[10] a model was proposed for calculating the mobility of charge carriers in organic dipole glasses, which considered a multistep process of charge carrier exit from a potential well caused by energy correlations. Description of the hopping transport of triplet excitons in QD solids can be obtained by using a similar approach; however, it is necessary to estimate the corresponding spatial size of the potential well and determine its shape. One can consider an exciton as a pointlike dipole with the moment, $d = er_{ex}$ and its interaction with randomly oriented dipole moments of QDs, p, situated at the nodes of a simple cubic lattice with constant a. The lattice constant corresponds to the distance between the centers of QDs, such that $a = d_{QD} + l$, where d_{QD} is the QD diameter and l is the edge-to-edge distance (ligand length). The density of energy states in this system can be calculated as[11]

$$g(E) = \left\langle \delta\left(E - \sum_n E_n\right)\right\rangle \quad (1)$$

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where E_n is the interaction energy between the exciton (pointlike dipole) and the dipole at the node n:

$$E_n = \frac{1}{4\pi\varepsilon\varepsilon_0 r_n^3}\left((\vec{d}\vec{p}_n) - \frac{3(\vec{d}\vec{r}_n)(\vec{p}_n\vec{r}_n)}{r_n^2}\right) \tag{2}$$

The summation is carried out over all nodes except the one at which the exciton is located. Averaging over all random vector directions $\boldsymbol{r}_n$ and $\boldsymbol{p}_n$ gives

$$g(E) = \frac{1}{\sqrt{2\pi\sigma^2}}\exp\left[-\frac{E^2}{2\sigma^2}\right], \quad \sigma = 2.4\frac{er_{ex}p}{4\pi\varepsilon\varepsilon_0 a^3} \tag{3}$$

Thus, the presence of large permanent dipole moments of QDs leads to the formation of Gaussian disorder for exciton states, and the scale of the disorder depends on the size of the exciton, the dipole moments of the QDs, and the dielectric constant of the medium.

Determination of the shape of the potential well, formed around a deep energy state, requires calculation of the correlation function in the system:[11]

$$C(r) = \frac{1}{(4\pi\varepsilon\varepsilon_0)^2}\left\langle \sum_{m,n}\frac{1}{r_m^3|\vec{r}_n - \vec{r}|^3}\left((\vec{d}\vec{p}_m) - \frac{3(\vec{d}\vec{r}_m)(\vec{p}_m\vec{r}_m)}{|\vec{r}_m|^2}\right)\cdot \left((\vec{d}\vec{p}_n) - \frac{3(\vec{d}(\vec{r}_n - \vec{r}))(\vec{p}_m(\vec{r}_n - \vec{r}))}{|\vec{r}_n - \vec{r}|^2}\right)\right\rangle \tag{4}$$

Carrying out statistical averaging and replacing summation with integration gives

$$C(r) = \beta\sigma^2\left(\frac{a}{r}\right)^3, \quad \beta = 0.48 \tag{5}$$

This type of correlation function is also typical for quadrupole glasses,[11] with a slight difference in the coefficient β (for quadrupole glasses, $\beta = 0.5$). The form of the correlation function and the energy distribution suggests that the conditional probability of a state situated at a distance r from the initial state with energy E_i to have the energy E has the following form:[9]

$$P(E|E_i) = \frac{1}{\sqrt{2\pi\sigma_t^2(r)}}\exp\left[-\frac{(E - U_{av}(r, E_i))^2}{2\sigma_t^2(r)}\right] \tag{6}$$

$$U_{av}(r, E_i) = -\beta|E_i|a^3/r^3, \quad r > a \tag{7}$$

$$\sigma_t = \sigma\sqrt{1 - (\beta^{1/3}a/r)^2} \tag{8}$$

This means that deep states with energies $E_i \approx -\sigma^2/kT$ controlling the conductance in a weak external electric field are surrounded by a potential well, defined by eq 7. One can define the spatial size of this well by the condition $U_{av}(r_C^*, E_i) = kT$, in analogy to the Coulomb radius (the Onsager radius):

$$r_C^* = a\beta^{1/3}\left(\frac{\sigma}{kT}\right)^{2/3} \tag{9}$$

The diffusion coefficient is calculated according to the ubiquitous (for the low-field limit) expression:

$$D = \frac{1}{6}\frac{r_C^{*2}}{\langle t\rangle} \tag{10}$$

where $\langle t\rangle$ is the average time required for a carrier to escape from the potential well. Thus, as in ref 10, the release of a carrier from a deep state is considered as a multistep process. One can distinguish two stages: (1) the first jump from the initial state to the nearest neighbor (most likely having the same energy due to the strong correlations), with a frequency ν; (2) drift-diffusion over a region of size r_C^*, which depends on the depth of the well (i.e., on the energy of the initial state E_i). Note that the escape probability is low. The scheme of the process is shown in Figure 1.

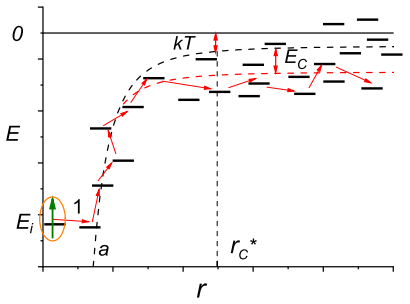

Figure 1. Scheme of exciton escape from a potential well formed around the deep state E_i. The dashed line shows the energy dependence given by eq 7, and the number 1 corresponds to a typical first jump from the initial state.

The typical number of attempts to escape is inversely proportional to the probability of a release after the first jump. The average escape time can be calculated as[10]

$$\langle t\rangle = \nu^{-1}\int_{-\infty}^{\infty} dE_i\, g(E_i)\eta^{-1}(E_i) \tag{11}$$

where $\nu = \nu_0\exp(-2\gamma r_0)$, and the typical tunnelling hopping length r_0 is defined by the condition $U(r_0) \approx U_{av}(r_0) = E_i$. For the triplet transport in QD solids, $r_0 = \beta^{1/3}l$. The probability of leaving the well (η) is calculated as the separation probability of a pair of particles interacting according to eq 7. To calculate the separation probability, it is necessary to solve the Smoluchowski equation with potential (eq 7) in the space of initial separations r_0:[20,21]

$$\frac{d^2\eta}{dr_0^2} + \left(\frac{2}{r_0} - \frac{1}{kT}\frac{dU_{av}}{dr_0}\right)\frac{d\eta}{dr_0} = 0 \tag{12}$$

with the boundary conditions:

$$\varphi = 0 \quad \text{at } r_0 = 0$$

$$\varphi \to 1 \quad \text{as } r_0 \to \infty$$

Using the method from previous work,[21] we obtain

$$\eta(r_0) = \frac{1}{3}\frac{\Gamma(1/3, (r_C^*/r_0)^3)}{\Gamma(4/3)} \tag{13}$$

Since the main contribution to the integral described by eq 11 is made by deep states, we will use the asymptote for the Γ function in expression described by eq 13. We also take into account, similarly to work,[10] that the transport level $E_C < 0$ (calculated according to previous work[22]), which lies below the maximum of the Gaussian distribution, reduces the height of the energy barrier that the carrier must overcome, see Figure 1, as noted in ref 23. Thus, energetic disorder increases the separation probability:

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$$\eta^{-1}(E_i) \approx 3\Gamma(4/3)\left(\frac{|E_i|}{kT}\right)^{2/3} \exp\left(\frac{|E_i| + E_C}{kT}\right) \quad (14)$$

Equations 11 and 14 give

$$\langle t \rangle^{-1} = \frac{\nu}{3\Gamma(4/3)} \exp\left[-C\left(\frac{\sigma}{kT}\right)^2 - \frac{E_C}{kT}\right] \quad (15)$$

$$C = \frac{1}{2} + \frac{4}{3}\left(\frac{kT}{\sigma}\right)^2 \ln\left(\frac{\sigma}{kT}\right) \quad (16)$$

Finally, from eqs 9, 10, 15, and 21, we obtain

$$D(T) = \frac{1}{6}\nu_0 \exp(-2\gamma r_0)\left[\frac{a^2\beta^{2/3}}{3\Gamma(4/3)}\right] \exp\left[-\frac{1}{2}\left(\frac{\sigma}{kT}\right)^2 - \frac{E_C}{kT}\right] \quad (17)$$

In eqs 15 and 17, E_C is the effective transport level,[22] $E_C = E_{tr} - 2\gamma l$, where the tunneling length (l) replaces the intersite distance (a), because hopping sites are not pointlike, in contrast to previous work;[22] E_{tr} is defined from the following equation:

$$B = \frac{4\pi}{3} N \int_{-\infty}^{E_{tr}} dE\, g(E)[(E_{tr} - E)/2\gamma kT]^3 \quad (18)$$

where $B = 2.77$, $N = a^{-3}$.

For CdSe QDs with diameter $d_{QD} = 3$ nm, the reported parameters are the following: dielectric constant, $\varepsilon \approx 6.2$;[24] and QD dipole moment, $p \approx 25$ D.[18] These parameters along with the short ligands ($l = 0.5$ nm) give the disorder scale $\sigma \approx 20$ meV (see eq 3). The proposed approach assumes that the condition $r_C^* > a$ is met. Estimation of the size of the potential well from eq 9 gives $r_C^* > 2a$ at $T < 120$ K, indicating that the effect of correlations is strongly pronounced at low temperatures. The obtained result is qualitatively consistent with the conclusions of ref 16, which showed that, at low temperatures, the transport of triplet excitons in organic materials is more influenced by disorder and, at high temperatures, by polaronic effects.

As for the hopping parameters, the values of the inverse localization radius (γ) of ~0.5 Å^{-1} were reported,[6] depending on the chemical structure of the ligand molecule. The value of the phonon frequency $\nu_0 = 7.5 \times 10^{11}$ s^{-1} for the 3 nm CdSe QD was reported in the calculations.[25] Using these parameters, we derived the absolute values for the diffusion coefficient $D = 1.5 \times 10^{-4}$ sm^2/s for temperature $T = 100$ K and $D = 6.7 \times 10^{-4}$ sm^2/s for $T = 298$ K, which are qualitatively consistent with the data of some experimental works. For example, previous work[13] has reported a value of $D \approx 3 \times 10^{-4}$ sm^2/s at room temperature for core/shell CdSe/ZnCdS QDs. Note that the proposed model works best at low temperatures, at which experimental data are limited, making the comparison difficult.

Since the resulting form of the correlation function and the density of states are in good agreement with the formulas for charge transport in quadrupole organic glasses (see, for example, refs 11 and 12), in order to verify the approach, the results of the calculations of the diffusion coefficient according to eq 11 were compared to the results of Monte Carlo simulation in the quadrupole glass model[12] with the parameters typical for the disordered organic media: $\varepsilon = 3$, $p = 3$ D, $\sigma = 0.078$ eV, $a = a_0 = 1$ nm (a_0 is the intermolecular distance), $\gamma a = 5$, $r_0 = \beta^{1/3} a_0$. This estimated value of σ is calculated for the quadrupole moment $Q = pr_{ex}$, where quadrupoles are formed by two oppositely directed dipoles with moment p, located at a distance r_{ex} from each other, $r_{ex} = 1$ nm. The estimation of the size of the potential well from eq 9 gives $r_C^* > 3a_0$ at $T < 125$ K. Figure 2 shows the good

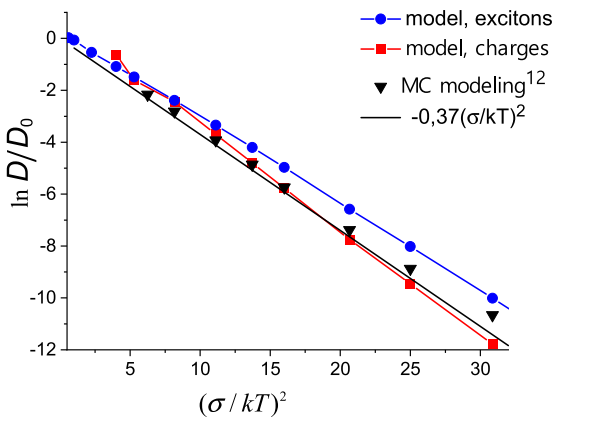

Figure 2. Temperature dependence of the diffusion coefficient for exciton transport in QD solids and charge transport in organic quadrupole glasses, in comparison with the MC simulation results;[12] $D_0 = \left(\frac{\nu_0 r_0^2}{6}\right) \exp(-2\gamma r_0)$.

agreement with the Monte Carlo results, as well as with the empirical formula for the case of zero external electric field[12] $\ln(D/D_0) = -0.37(\sigma/kT)^2$ at temperatures below room temperature ($T < 278$ K) (even for $r_C^* \approx a_0$). The difference with the Monte Carlo results for large values of σ/kT is explained by the fact that the data were taken for a weak but nonzero field, the influence of which on mobility increases along with σ/kT.

In the first approximation, the temperature dependences of the diffusion coefficient both in the case of quadrupole glasses and QD solids can be expressed by the well-known for organic materials dependence,[8,12,26] $\ln(D/D_0) = -\text{const}(\sigma/kT)^2$. For the case of QD solids, one obtains const ≈ 0.34, whereas for the case of quadrupole glasses, const ≈ 0.42 is the same as in the Gaussian disorder model[26] and is slightly different from $\ln(D/D_0) = -0.37(\sigma/kT)^2$ (see Figure 2). The difference in the constants follows from the difference in the temperature dependences of E_C, which, in turn, follows from the difference in the localization parameters used: $\gamma a_0 = 5$ for the quadrupole glass and $\gamma l = 2.25$ for the QD solid.

We can also obtain the field dependence of the mobility by using the field-dependent effective temperature concept (ETC) modified in this work. The essence of the ETC is that the electric field F reduces the activation energy of jumps by eFh (in a disordered medium and, in the case of a uniform field, the length scale h is the localization radius γ^{-1}). Consequently, the jump frequency and mobility increase. Qualitatively, an increase in the temperature has the same effect. In the limit $T = 0$, one has $T_{eff}(0, F) \approx eFh/k$.[27] For the general case, one obtains interpolation expressions such as

$$T_{eff}(T, F) = T\left[1 + \lambda\left(\frac{eFh}{k}\right)^n\right]^{1/n} \quad (19)$$

where λ is a numerical constant, and usually, $n = 2$.[28,29] When a particle leaves a potential well, the electric field is nonuniform and the process is multistep. By analogy with the Onsager model, in which the quantum yield depends on the parameter

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$eFr_0/2kT$,[20] one assumes that the exponent n in eq 19 depends on this parameter. According to the fitting data (see Figure 3), the exponent n increases from 1 to 4 with increasing field F. The distance r_0 is the spatial scale. Thus, in eq 19,

$$n = \frac{1 + 4c_1\left(\frac{eFr_0}{2kT}\right)}{1 + c_1\left(\frac{eFr_0}{2kT}\right)}, \quad c_1 = 0.29, \ \lambda = 0.32, \ h = r_0 \tag{20}$$

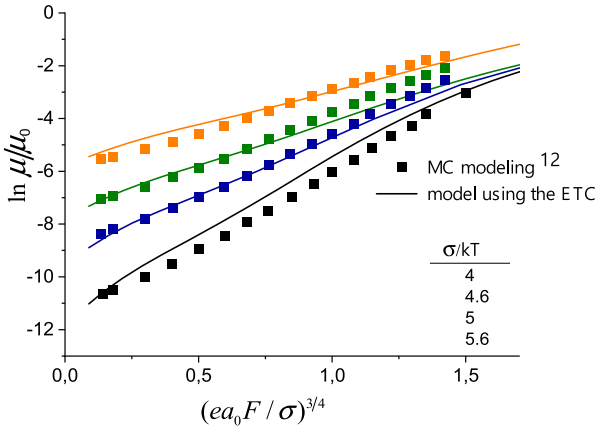

Figure 3. Field dependence of charge carrier mobility in quadrupole glasses, in comparison with the MC simulation data.[12]

The mobility for a weak electric field is calculated from eq 17, using the Einstein relation:

$$\mu(T) = \frac{\mu_0 \beta^{2/3}}{3\Gamma(4/3)} \exp\left[-\frac{1}{2}\left(\frac{\sigma}{kT}\right)^2 - \frac{E_C}{kT}\right] \tag{21}$$

with $\mu_0 = e\left(\frac{\nu_0 a_0^2}{6}\right) \exp(-2\gamma r_0)/kT$. Here, $E_C = E_{tr} - 2\gamma a$, the same as in previous work.[22] We obtain the field dependence of mobility in a quadrupole glass by replacing the temperature T in eq 21 with the effective temperature, $T_{eff}(T, F)$. The field dependence of mobility with the above-mentioned parameters for quadrupole glasses, obtained using the modified ETC, are compared in Figure 3 with the Monte Carlo simulation data for quadrupole glasses from ref 12. The agreement is good, especially at moderate field strengths. Equation 21 gives the correct value and field dependence of hole mobility in nonpolar organic material 4,4′-bis(*N*-carbazolyl)-1,1′-biphenyl (CBP), as measured in previous work,[30] providing the independently determined[31] values $\sigma = 0.125$ eV and $\mu_0 = 0.8$ cm^2 V^{-1} s^{-1}. We obtain the inverse localization radius $\gamma = 2.5/a_0$ from fitting; hence, $\nu_0 = 10^{15}$ s^{-1} from eq 21. This is an example of the application of our model for the estimation of physical parameters that are difficult to measure for a given material.

In summary, the energy distribution of hopping centers that control the triplet exciton diffusion has been calculated in this work. This distribution is in the form of a Gaussian function. The energy disorder is spatially correlated. It arises due to the electrostatic interaction of the exciton dipole moment with the randomly oriented molecular dipole moments. When the width of the DOS was calculated, other possible contributions to disorder have not been taken into account, primarily the spread in QD sizes and spatial disorder in QD positions. This approach is justified by the fact that one aims to achieve the most ordered QD-solid structures for applications. On the other hand, random orientation of QD dipoles is assumed, while their partial ordering can reduce disorder. Nevertheless, the resulting expression for the width of the Gaussian DOS appears to be a lower estimate.

The characteristic escape time of an exciton from the energy well, which arises as a result of energy correlations around deep states, has been calculated together with the characteristic size of the well. These quantities determine the magnitude and temperature dependence of the exciton diffusion coefficient. A simple analytical expression for the diffusion coefficient of triplet excitons in QD solids has been obtained. This expression (see eq 17) is reminiscent of the expression for the coefficient of hopping diffusion of charge carriers in disordered organic semiconductors. The proposed approach has been tested by comparison with the known[12] results of MC modeling of charge carrier mobility in organic quadrupole glasses (where a similar type of correlation exists). Using the same approach, an analytic model of the diffusion coefficient and mobility for quadrupole glasses has been developed and justified by comparison with the experimental data.[30,31]

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Notes

The authors declare no competing financial interest.

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REFERENCES

(1) Ge, F. Y.; Han, Y. Y.; Feng, C. S.; Zhang, H.; Chen, F. F.; Xu, D.; Tao, C. L.; Cheng, F.; Wu, X. J. Halide Ions Regulating the Morphologies of Pbs and Au@Pbs Core-Shell Nanocrystals: Synthesis, Self-Assembly, and Electrical Transport Properties. *J. Phys. Chem. Lett.* **2023**, *14*, 9521−9530.

(2) Chen, Y. A.; Filip, M. R. Tunable Interlayer Delocalization of Excitons in Layered Organic-Inorganic Halide Perovskites. *J. Phys. Chem. Lett.* **2023**, *14*, 10634−10641.
(3) Nicolaidou, E.; Parker, A. W.; Sazanovich, I. V.; Towrie, M.; Hayes, S. C. Unraveling Excited State Dynamics of a Single-Stranded DNA-Assembled Conjugated Polyelectrolyte. *J. Phys. Chem. Lett.* **2023**, *14*, 9794−9803.
(4) Lee, Y. H.; Park, J. Y.; Niu, P. R.; Yang, H. J.; Sun, D. W.; Huang, L. B.; Mei, J. G.; Dou, L. T. One-Step Solution Patterning for Two-Dimensional Perovskite Nanoplate Arrays. *ACS Nano* **2023**, *17*, 13840−13850.
(5) Mondal, S.; Chowdhury, U.; Dey, S.; Habib, M.; Perez, C. M.; Frauenheim, T.; Sarkar, R.; Pal, S.; Prezhdo, O. V. Controlling Charge Carrier Dynamics in Porphyrin Nanorings by Optically Active Templates. *J. Phys. Chem. Lett.* **2023**, *14*, 11384−11392.
(6) Guyot-Sionnest, P. Electrical Transport in Colloidal Quantum Dot Films. *J. Phys. Chem. Lett.* **2012**, *3*, 1169−1175.
(7) Chistyakov, A. A.; Zvaigzne, M. A.; Nikitenko, V. R.; Tameev, A. R.; Martynov, I. L.; Prezhdo, O. V. Optoelectronic Properties of Semiconductor Quantum Dot Solids for Photovoltaic Applications. *J. Phys. Chem. Lett.* **2017**, *8*, 4129−4139.
(8) Novikov, S. V.; Dunlap, D. H.; Kenkre, V. M.; Parris, P. E.; Vannikov, A. V. Essential Role of Correlations in Governing Charge Transport in Disordered Organic Materials. *Phys. Rev. Lett.* **1998**, *81*, 4472−4475.
(9) Novikov, S. V.; Vannikov, A. V. Cluster Structure in the Distribution of the Electrostatic Potential in a Lattice of Randomly Oriented Dipoles. *J. Phys. Chem.* **1995**, *99*, 14573−14576.
(10) Nikitenko, V. R.; Saunina, A. Y.; Tyutnev, A. P.; Prezhdo, O. V. Analytic Modeling of Field Dependence of Charge Mobility and Applicability of the Concept of the Effective Transport Level to an Organic Dipole Glass. *J. Phys. Chem. C* **2017**, *121*, 7776−7781.
(11) Novikov, S. V.; Dunlap, D. H.; Kenkre, V. M. Charge-Carrier Transport in Disordered Organic Materials: Dipoles, Quadrupoles, Traps, and All That. *SPIE Proc.* **1998**, *3471*, 181−191.
(12) Novikov, S. V.; Vannikov, A. V. Hopping Charge Transport in Disordered Organic Materials: Where Is the Disorder? *J. Phys. Chem. C* **2009**, *113*, 2532−2540.
(13) Akselrod, G. M.; Prins, F.; Poulikakos, L. V.; Lee, E. M. Y.; Weidman, M. C.; Mork, A. J.; Willard, A. P.; Bulovic, V.; Tisdale, W. A. Subdiffusive Exciton Transport in Quantum Dot Solids. *Nano Lett.* **2014**, *14*, 3556−3562.
(14) Kholmicheva, N.; Moroz, P.; Bastola, E.; Razgoniaeva, N.; Bocanegra, J.; Shaughnessy, M.; Porach, Z.; Khon, D.; Zamkov, M. Mapping the Exciton Diffusion in Semiconductor Nanocrystal Solids. *ACS Nano* **2015**, *9*, 2926−2937.
(15) Kholmicheva, N.; Moroz, P.; Eckard, H.; Jensen, G.; Zamkov, M. Energy Transfer in Quantum Dot Solids. *ACS Energy Lett.* **2017**, *2*, 154−160.
(16) Fishchuk, I. I.; Kadashchuk, A.; Sudha Devi, L.; Heremans, P.; Bässler, H.; Köhler, A. Triplet Energy Transfer in Conjugated Polymers. II. A Polaron Theory Description Addressing the Influence of Disorder. *Phys. Rev. B* **2008**, *78* (045211), 1−8.
(17) Colvin, V. L.; Cunningham, K. L.; Alivisatos, A. P. Electric Field Modulation Studies of Optical Absorption in CdSe Nanocrystals: Dipolar Character of the Excited State. *J. Chem. Phys.* **1994**, *101*, 7122−7138.
(18) Blanton, S. A.; Leheny, R. L.; Hines, M. A.; Guyot-Sionnest, P. Dielectric Dispersion Measurements of Cdse Nanocrystal Colloids: Observation of a Permanent Dipole Moment. *Phys. Rev. Lett.* **1997**, *79*, 865−868.
(19) Kortschot, R. J.; Van Rijssel, J.; Van Dijk-Moes, R. J. A.; Erné, B. H. Equilibrium Structures of Pbse and Cdse Colloidal Quantum Dots Detected by Dielectric Spectroscopy. *J. Phys. Chem. C* **2014**, *118*, 7185−7194.
(20) Onsager, L. Initial Recombination of Ions. *Phys. Rev.* **1938**, *54*, 554−557.
(21) Tachiya, M. General Method for Calculating the Escape Probability in Diffusion-Controlled Reactions. *J. Chem. Phys.* **1978**, *69*, 2375−2376.
(22) Nikitenko, V. R.; Strikhanov, M. N. Transport Level in Disordered Organics: An Analytic Model and Monte-Carlo Simulations. *J. Appl. Phys.* **2014**, *115* (7), 073704.
(23) Albrecht, U.; Bässler, H. Yield of Geminate Pair Dissociation in an Energetically Random Hopping System. *Chem. Phys. Lett.* **1995**, *235*, 389−393.
(24) Casas Espínola, J. L.; Hernández Contreras, X. A. Effect of Dielectric Constant on Emission of CdSe Quantum Dots. *J. Mater. Sci.: Mater. Electron.* **2017**, *28*, 7132−7138.
(25) Yang, J.; Wise, F. W. Effects of Disorder on Electronic Properties of Nanocrystal Assemblies. *J. Phys. Chem. C* **2015**, *119*, 3338−3347.
(26) Bässler, H. Charge Transport in Disordered Organic Photoconductors a Monte Carlo Simulation Study. *Phys. Status Solidi B* **1993**, *175*, 15−56.
(27) Marianer, S.; Shklovskii, B. I. Effective Temperature of Hopping Electrons in a Strong Electric Field. *Phys. Rev. B* **1992**, *46*, 13100−13103.
(28) Jansson, F.; Baranovskii, S. D.; Gebhard, F.; Österbacka, R. Effective Temperature for Hopping Transport in a Gaussian Density of States. *Phys. Rev. B* **2008**, *77* (19), 195211.
(29) Khan, M. D.; Nikitenko, V. R.; Tyutnev, A. P.; Ikhsanov, R. S. Joint Application of Transport Level and Effective Temperature Concepts for an Analytic Description of the Quasi- and Non-equilibrium Charge Transport in Disordered Organics. *J. Phys. Chem. C* **2019**, *123*, 1653−1659.
(30) Hoping, M.; Schildknecht, C.; Gargouri, H.; Riedl, T.; Tilgner, M.; Johannes, H.-H.; Kowalsky, W. Transition Metal Oxides as Charge Injecting Layer for Admittance Spectroscopy. *Appl. Phys. Lett.* **2008**, *92* (21), 213306.
(31) Stankevych, A.; Vakhnin, A.; Andrienko, D.; Paterson, L.; Genoe, J.; Fishchuk, I.; Bässler, H.; Köhler, A.; Kadashchuk, A. Density of States of OLED Host Materials from Thermally Stimulated Luminescence. *Phys. Rev. Appl.* **2021**, *15* (4), 044050.

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