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Comparative study of hole transport in poly(p-phenylene vinylene) derivatives

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The dc mobility of holes in four poly(p-phenylene vinylene) (PPV) derivatives (three fully conjugated polymers with different side chains and one partially conjugated PPV) is examined as a function of temperature T and applied electrical field E. In all cases the mobility μ follows the empirical $\mu \propto \exp \gamma\sqrt{E}$ law. The specific temperature and electrical field dependence of the mobility is fitted within a (correlated) Gaussian disorder model. From the fits the energetic disorder, localization length, and average transport-site separation are determined. In the case of the fully conjugated polymers the different T and E dependencies of μ are completely determined by the energetic disorder. The relation between the obtained microscopic transport parameters and the specific chemical composition of the polymer material is discussed.

I. INTRODUCTION

The attractive optoelectronic properties of conjugated polymers arise from the extended π-electron system along the polymer backbone. For instance, poly(p-phenylene vinylene), PPV, is considered a suitable candidate for flexible large-area display technologies based on polymer light-emitting diodes (PLED's).[1,2] A lot of effort has been put in the understanding of the device operation; Ref. 3 gives an overview of the current scientific and technological knowledge of PPV based LED's. Recent experiments have demonstrated that charge transport in PLED's is bulk space-charge limited[4] and not injection limited,[5] implying that current flow is directly governed by the charge-carrier mobility. In order to generate light, recombination of electrons and holes in the active layer is required. The capture of oppositely charged carriers in PPV has been shown to be of the Langevin type, meaning that the recombination efficiency is diffusion limited.[6] As both transport and recombination of charge-carriers in PLED's are governed by the mobility, clearly control of this parameter is important for optimizing device performance.

Chemical modification of PPV provides a tool to control the conjugated backbone and (nonconjugated) side chains of the polymer. Different side chains attached to the polymer may influence interchain distance and packing and hence can affect the charge transfer between polymer chains. Furthermore, also on-chain electronic properties may be sensitive to the chemical nature of side groups. On the other hand, tailoring the conjugated backbone naturally has strong impact on the conductive properties as it directly involves modification of the charge transporting fraction of the polymer itself. Therefore, as the molecular structure of the polymer chains is expected to strongly affect the charge-carrier mobility, in order to further optimize PLED performance a handle on the interplay between chemical structure, mobility and device-efficiency (photon/charge-carrier) is needed. This paper focuses on charge-carrier transport properties of PPV derivatives. Among the most widely used PPV derivatives are MEH-PPV (Ref. 7) and OC_1C_{10}-PPV;[8] here we compare the charge transport properties of OC_1C_{10}-PPV and several of its derivatives. By performing current density–voltage measurements the charge-carrier mobility μ as a function of temperature T and electrical field E has been obtained. The obtained T and E dependencies of μ for the different PPV derivatives are analyzed within a Gaussian disorder model, yielding insight in the interplay between chemical structure and microscopic charge transport parameters in these materials. It is obtained that energetic disorder predominantly governs $\mu(T,E)$ of the fully conjugated materials, while both structural changes (increasing hopping distance) and energetic disorder determine the conductive properties when decreasing the conjugated fraction.

II. EXPERIMENT

The chemical structures of the (alkoxy-substituted) PPV derivatives studied here are shown in Fig. 1. In the following the materials are named A, B, C, and D as in Fig. 1, the mobility of the different polymers will be indicated accordingly (μ_A, etc.). Polymer A is ‘‘standard’’ OC_1C_{10}-PPV, a PPV derivative often used in PLED's.[8] Polymer B is a copolymer synthesized from (conjugated) phenylene-vinylene units and (nonconjugated) phenylene-ethylidene units.[9] The intentionally broken conjugation is expected to strongly hamper charge transport due to the decrease of charge transporting sites in the material. In polymer C the short OCH_3 group of OC_1C_{10}-PPV is replaced with a second $OC_{10}H_{21}$ group, for convenience this material will also be indicated as $OC_{10}C_{10}$-PPV. The application of longer side chains could increase the distance between the conjugated regions of neighboring chains and thereby lower the mobility. Polymer

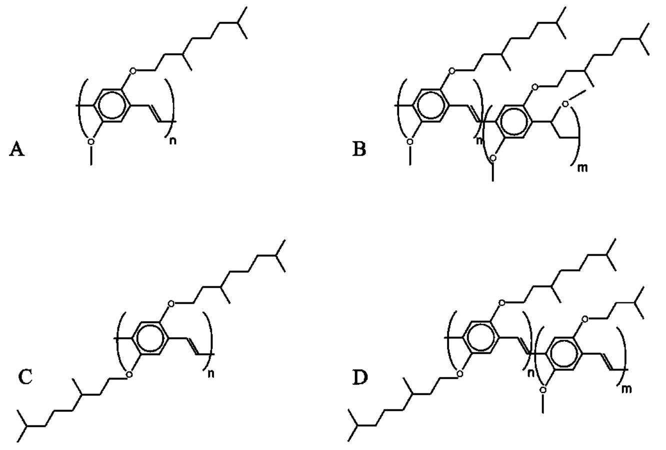

FIG. 1. Chemical structure of the polymers studied: (*A*) Fully conjugated OC_1C_{10}-PPV. (*B*) Partially conjugated OC_1C_{10}-PPV with $n:m=9:1$. (*C*) Dialkoxy-PPV with two $C_{10}H_{21}$ sidegroups ($OC_{10}C_{10}$-PPV). (*D*) Fully conjugated PPV copolymer synthesized of $OC_{10}C_{10}$-PV and OC_1C_5-PV units in the ratio $n:m=3:1$.

D is a derivative of polymer *C* with 25% of the $OC_{10}C_{10}$-PV monomers substituted by a monomer with shorter side chains (OC_5H_{11}/OCH_3) where the above mentioned effect might be partly undone. However, as chemical modifications of the polymer can influence both its electronic and morphologic properties in a complicated way, it is hard to predict beforehand the conductive properties of the material.

The fully conjugated materials were prepared by dehydrohalogenation polymerization of the corresponding monomer(s) with potassium tertiary-butoxide in dioxane at 80 °C, and purified by precipitation in water and in methanol twice. For the partially conjugated material, the nonconjugated units are introduced by a controlled Cl/OCH_3 exchange on the chloro-precursor polymer, prior to elimination. The fraction of conjugation defects (ethylidene concentration) x in the polymers was determined by means of ^{1}H-NMR characterization. For the ''fully conjugated'' polymers *A*, *C*, and *D* $x \approx 2\%$, while in polymer *B* $x \approx 10\%$[9] was obtained due to the intentional decrease of conjugation. The soluble polymers are spin-coated on top of a glass substrate with a patterned ITO contact serving as the anode. Hole-only devices are obtained by applying gold as a top electrode. The typical thickness of the devices studied is $d \sim 100$–300 nm, with an active area $A \sim 10$ mm^2. Current density–voltage (J–V) experiments on the devices were performed with a HP4145A semiconductor parameter analyzer. Both the fabrication procedure and the charge transport experiments were performed in a nitrogen atmosphere.

III. RESULTS AND DISCUSSION

Figure 2 displays a typical example of the temperature-dependent current-density–voltage characteristics of a PPV hole-only device (polymer *A*). The current density depends strongly on both the applied bias voltage and the temperature. Recently, it has been shown that the J–V dependencies of PPV devices can be fitted accurately within a space-charge limited current (SCLC) model[10] taking into account a stretched exponential field dependence of the mobility:

$$\mu(E) = \mu_0 \exp(\gamma \sqrt{E}) \qquad (1)$$

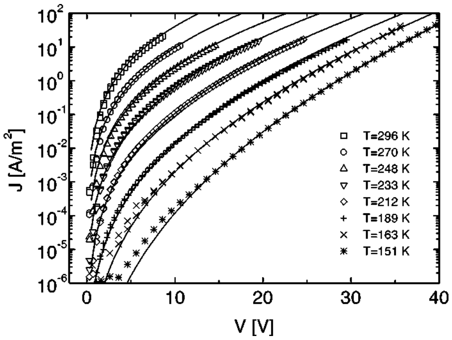

FIG. 2. Temperature dependent $J(V)$ characteristics of a OC_1C_{10}-PPV (polymer *A*) hole-only device, thickness 275 nm and active area 10.9 mm^2. The drawn lines represent the predictions of a space charge limited current model incorporating the stretched exponential field dependence of the mobility, Eq. (1).

with μ_0 the zero-field mobility and γ describing the ''field activation'' of the mobility. This empirical dependence of the mobility on the applied electrical field appears generic for a large class of disordered organic semiconductors such as molecularly doped polymers, pendant group polymers, conjugated polymers and organic glasses, see, e.g., Refs. 11–15. Theoretical models reproducing the stretched exponential E dependence of μ are discussed below. The solid lines in Fig. 2, representing the predictions of the SCLC model, are in excellent agreement with the experimental data. For all PPV derivatives studied here, the current was bulk space-charge limited and the mobility was found to be well described by Eq. (1). The values of μ_0 and γ obtained from the fitting of the J–V curves of all samples studied are plotted as a function of temperature in Fig. 3 and Fig. 4 respectively.

It is directly obtained from Fig. 3 that chemical modification of PPV changes the mobility by orders of magnitude. The (zero-field) mobility of the ''fully conjugated'' OC_1C_{10}-PPV (*A*) is one order of magnitude higher than that of the partially conjugated OC_1C_{10}-PPV (*B*). The effect of different side chains attached to the PV monomer is also considerable. The mobility of polymers *C* and *D*, similar to *A* but with different side chains attached, lies more than one order of magnitude above μ_A. Radiolysis microwave (30 GHz) conductivity experiments on fully and partially conjugated OC_1C_{10}-PPV and fully conjugated $OC_{10}C_{10}$-PPV re-

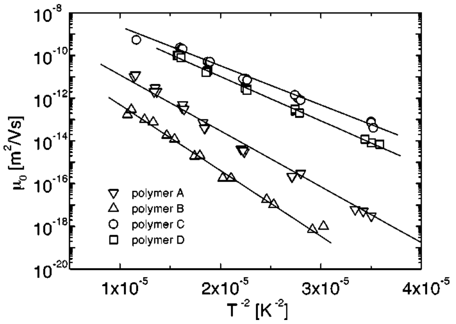

FIG. 3. Temperature dependence of the zero-field mobility of the four PPV derivatives studied. The data are fitted with the $\ln(\mu_0) \sim T^{-2}$ law for hopping transport in a Gaussian DOS. The parameters derived for the different compounds are listed in Table I.

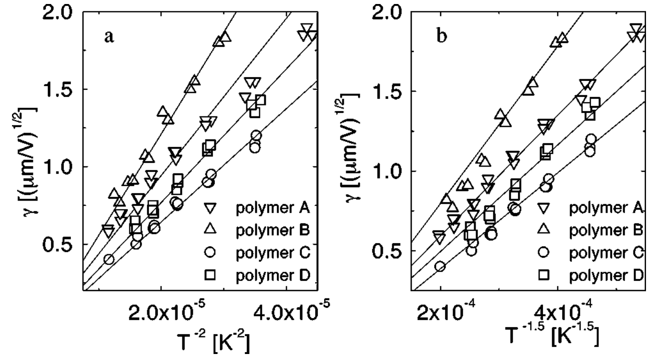

FIG. 4. The coefficient γ (describing the field dependence of μ) against T^{-2} and $T^{-1.5}$ respectively. The solid lines in (a) represent fits to the GDM, Eq. (2), while the solid lines in (b) are fits according to the CDM, Eq. (2).

TABLE I. Parameters μ_∞ (mobility prefactor), σ (energetic disorder bandwidth) and a (site-spacing) describing the temperature dependence of zero-field mobility μ_0 and ''field activation'' factor γ according to the CDM for the different polymers studied. The value of C gives the field dependence of μ in terms of the GDM. The localization length L is derived from the values of a, σ, and μ_∞, see text.

Sample	μ_∞ [m^2/Vs]	σ [meV]	C [(m/V)$^{1/2}$]	a [nm]	L [nm]
A	5.1×10^{-9}	112	4.0×10^{-5}	1.2	0.3
B	4.0×10^{-10}	121	4.3×10^{-5}	1.7	0.3
C	1.6×10^{-7}	93	3.8×10^{-5}	1.1	0.5
D	1.5×10^{-7}	99	4.0×10^{-5}	1.2	0.5

vealed a similar trend (i.e., $\mu_\mathrm{B}<\mu_\mathrm{A}<\mu_\mathrm{C}$) (Ref. 16) although the reported differences were much smaller. This likely stems from the fact that generally in a disordered system the high frequency mobility is less sensitive to disorder present than the dc mobility.[17] Likewise, the field dependence of the mobility, described by γ, shows much variation with the chemical modifications, see Fig. 4. Comparing for the different polymers the absolute value and temperature dependence of μ_0 with the absolute value and temperature dependence of γ (Figs. 3 and 4), a specific trend is observed. A low and strongly activated μ_0 apparently correlates with a high and strongly temperature dependent γ. Qualitatively, this behavior is easily understood. In the case of strong energetic disorder, the barriers for a carrier to overcome in the transport process are high, leading to both a stronger activation and lower absolute value of the mobility. On the other hand, in strongly disordered systems the typical length scale R_t above which the conductive path appears homogeneous increases with growing disorder.[18] Application of an electrical field will be of importance when the potential drop over this length scale, eER_t, becomes comparable to k_BT.[19] Consequently, the field dependence of μ will be stronger in the case of more disorder.

PPV falls in the large class of disordered organic conductors. In these materials the electrical field dependence of the mobility can be described by the empirical relation Eq. (1), which was first observed for poly(N-vinyl carbazole) (Refs. 11 and 12) almost three decades ago. Gill demonstrated that $\gamma=B[1/(k_BT)-1/(k_BT_0)]$ and $\mu_0=\mu_\infty\exp[-\Delta/(k_BT)]$ can give a good description of the experimental data.[12] Numerous experiments (time-of-flight,[11–14] J–V[10] and impedance[15]) have confirmed this functional dependence on E and T with typically $\Delta\approx0.4$ eV, $B\approx3\times10^{-5}$ eV(m/V)$^{1/2}$ and $T_0\approx600$ K. Unfortunately, the empirical Gill formula lacks theoretical justification.

It has been proposed by Bässler that charge transport in disordered organic conductors proceeds by means of hopping in a Gaussian site-energy (ϵ) distribution, $g(\epsilon)=(2\pi\sigma^2)^{-1/2}\exp[-\epsilon^2/(2\sigma^2)]$, with σ the width of this distribution. This density of states (DOS) reflects the energetic spread in the charge transporting levels of chain segments due to the fluctuation in conjugation lengths and structural disorder. Based on Monte-Carlo simulations of transport of carriers injected in an array of sites exhibiting a Gaussian DOS, the following functional dependence of μ was proposed:[20]

$$\mu_{\mathrm{GDM}}=\mu_\infty\exp\left\{-\left(\frac{2\sigma}{3k_BT}\right)^2+C\left[\left(\frac{\sigma}{k_BT}\right)^2-2.25\right]\sqrt{E}\right\} \tag{2}$$

with μ_∞ the mobility in the limit $T\rightarrow\infty$ and $E\rightarrow0$, σ the width of the Gaussian DOS and C a constant (depending a.o. on the lattice constant). A consequence of carriers hopping in a Gaussian DOS is the non-Arrhenius behavior of the mobility. Since the DOS is highest at $\epsilon=0$, in general a carrier must be activated to $\epsilon\approx0$ in order to diffuse through the system. It can be readily calculated that the equilibration energy ϵ_{eq} of a carrier in a Gaussian DOS is $\epsilon_{\mathrm{eq}}=-\sigma^2/k_BT$.[20] Combining these two finding leads to $\ln(\mu_0)\propto-(T_0/T)^2$ dependence. The stretched exponential E dependence of μ depicted by Eq. (2) stems from the reduction of barriers for charge transport in the field direction by the applied electrostatic potential.

Within the temperature range studied, the zero-field mobility of the samples examined is in good agreement with the predictions of the Gaussian disorder model (GDM), see the solid lines in Fig. 3. The values obtained for μ_∞ and σ are listed in Table I for the different polymers (listed are the slightly higher σ values according to μ_{CDM}, see Eq. (3) below). Note that within the temperature range studied it is not possible to discriminate between a $\ln(\mu_0)\propto-(T_0/T)^2$ dependence and Arrhenius behavior, $\ln(\mu_0)\propto-(\Delta/k_BT)$ (fitting to an Arrhenius law yields $\Delta\approx0.48$, 0.52, 0.30, and 0.33 eV for samples A–D, respectively).

According to the GDM the field dependence of μ is controlled by the energetic disorder and a scaling factor C, see Eq. (2). The fits of γ to the GDM formula are shown in Fig. 4(a). The obtained C values are listed in Table I. The fits cannot be performed with one value of C, nor is there a specific trend observable in the obtained values. A drawback of this analysis is that the derived value of C is not clearly linked to a physical parameter of the system. Furthermore, the simulations of the GDM reproduced the $\ln(\mu)\propto\sqrt{E}$ law only in a narrow field range, at relatively high values of $E\sim10^8$ V/m,[20] whereas experiments indicate this universal behavior occurs over a much wider range of fields (10^6–10^8 V/m).[10–15] Recent calculations and simulations demonstrated that the presence of long-range energy correlation gives rise to $\ln(\mu)\propto\sqrt{E}$ behavior over a much broader field range.[21,22] Such long-range energy correlation could for instance arise from charge-dipole interactions in the material. Based on simulations of transport in a correlated Gaussian disorder model (CDM) the following empirical relationship was derived:[22]

$$\mu_{\mathrm{CDM}}=\mu_\infty \exp\left\{-\left(\frac{3\sigma}{5k_BT}\right)^2+0.78\left[\left(\frac{\sigma}{k_BT}\right)^{3/2}-2\right]\sqrt{\frac{eaE}{\sigma}}\right\} \tag{3}$$

with a the intersite spacing. The main difference with Eq. (2) is the predicted temperature dependence of γ. Within the CDM the field dependence of μ is related to the energetic disorder and the site spacing a, which provides a natural measure for the length scale involved in the charge transport process.

Armed with the obtained values for the energetic disorder σ, we fit the temperature dependence of γ to the CDM prediction to extract the average site separation a. In Fig. 4(b) the fits of $\gamma(T)$ to Eq. (3) are shown. For the fully conjugated polymers A and D $a\approx1.2\pm0.1$ nm while for polymer C we find $a\approx1.1\pm0.1$ nm, see Table I. For the partially conjugated polymer (B) a larger inter-site distance is obtained, $a\approx1.7\pm0.1$ nm. Surprisingly, the average transport-site separation in the fully conjugated polymers (A, C, and D) is the same within the error bar, despite the different side chains attached to the PV monomer. It is important to realize the implication of this result: the different electrical field and temperature dependencies of the mobility of the fully conjugated polymers are solely governed by the energetic disorder. On the other hand, the strong electrical field dependence of the mobility of the partially conjugated material (B) stems from both the large energetic disorder and large site separation.

Finally, let us try to analyze the absolute value of the mobility ($\propto\mu_\infty$), which must be linked to the amount of electronic overlap between adjacent chain segments. In the limit $T\rightarrow\infty$ charge transfer rates are only limited by the wave function overlap between neighboring sites. Since for nearest-neighbor hopping the charge-carrier displacement per jump is a and the attempt frequency for such a hop $\nu=\nu_{\mathrm{ph}}\exp(-2a/L)$, with L the localization length and $\nu_{\mathrm{ph}}\sim10^{12}\ \mathrm{s}^{-1}$ a typical (phonon) attempt frequency for hopping, the diffusion constant D of the charge carriers is given by $D=a^2\nu_{\mathrm{ph}}\exp(-2a/L)$. Using the Einstein relation $\mu=(eD/n)\partial n/\partial\mu_{ec}$, with μ_{ec} the electrochemical potential, and realizing that for n carriers in a band of width σ in the limit $k_BT\gg\sigma$ we have $\partial n/\partial\mu_{ec}\approx n/\sigma$, the $T\rightarrow\infty$ mobility becomes

$$\mu_\infty=\frac{ea^2\nu_{\mathrm{ph}}}{\sigma}\exp(-2a/L). \tag{4}$$

Using the derived values of μ_∞, σ and a, the localization length L can now be estimated. We obtain for the fully conjugated and partially conjugated OC_1C_{10}-PPV (samples A and B) $L\approx3$ Å while for samples C and D a localization length $L\approx5$ Å is derived, see Table I. The analysis of the mobility in terms of the microscopic transport parameters σ, a and L allows us to relate the specific chemical composition of a PPV derivative to its charge transport properties, as is discussed in the following.

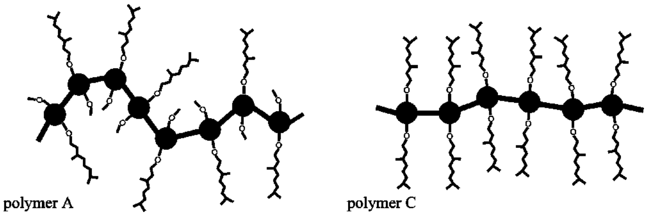

FIG. 5. Schematic representation of OC_1C_{10}-PPV (polymer A) and $OC_{10}C_{10}$-PPV (polymer C) respectively. The conjugated backbone is indicated by the black area. The symmetric $OC_{10}C_{10}$-PPV promotes alignment of the conjugated backbone, which decreases the on-chain energetic disorder. Furthermore, the high degree of regularity of polymer C is expected to lead to less structural disorder in the solid state as compared to polymer A.

Side chains – Contrary to our expectations the presence of the two bulky $OC_{10}H_{21}$ side chains ($OC_{10}C_{10}$-PPV) results in a large increase of the absolute value, and a decrease of the activation of μ as compared to OC_1C_{10}-PPV, see Fig. 3. The lower activation of μ shows that the energetic disorder in $OC_{10}C_{10}$-PPV is significantly less than in OC_1C_{10}-PPV, see Table I. This difference can be understood in view of the possible couplings between monomer units of the two polymers. The asymmetric substitution of the OC_1C_{10}-monomer during synthesis allows three possible dimer configurations in OC_1C_{10}-PPV, whereas in the case of the $OC_{10}C_{10}$ polymer every coupling of two monomers results in the same dimer ($OC_{10}C_{10}$-PPV is a regioregular and stereoregular polymer). In comparison with $OC_{10}C_{10}$-PPV, the increased configurational freedom of the OC_1C_{10} polymer will give rise to a larger energetic spread between the electronic levels of individual chain segments. A second effect of the regularity of $OC_{10}C_{10}$-PPV is the decreased conformational freedom of individual chains, see Fig. 5, which likely also decreases energetic disorder. Furthermore, it can be expected that a high degree of regularity enhances the ordering in the solid state. Since charge transfer between neighboring conjugated chains depends on parameters such as mutual orientation and potential energy barriers, this will favor charge delocalization as is indeed reflected in the larger localization length L of the $OC_{10}C_{10}$-PPV. Finally, let us compare μ_{C} with the charge transport properties of sample D, where 25% of the $OC_{10}C_{10}$ monomers are replaced by ''OC_1C_5''-monomers. The addition of the asymmetrical OC_1C_5-PV monomers decreases the regularity of the polymer chain, resulting in the observed increase of the on-chain energetic disorder. Still, the absolute value of μ_{D} is only slightly below μ_{C} and the other transport parameters derived for this polymer are almost identical to those of $OC_{10}C_{10}$-PPV. Apparently, with the majority of the PV monomers still symmetrical, the electronic and morphologic properties of the polymer material are not strongly altered.

Conjugation – As expected, the intentional decrease of conjugation strongly hampers charge transport. The strongest limitation for charge transport appears to be the increased inter-site distance a, reflecting the effective dilution of the charge transporting fraction of the material upon breaking the conjugation. Furthermore, also the on-chain energetic disorder increases with decreasing conjugation due to the additional vinylene-ethylidene units inserted into the polymer backbone. The localization length L of the carriers is similar to that in ''fully conjugated'' OC_1C_{10}-PPV. As the value of L is determined by mutual orientation and energy barriers between transport sites, apparently these properties are not very sensitive to the decrease of conjugation.

In summary, we have studied the hole mobility in several PPV derivatives by means of $J-V$ experiments. By chemically modifying the PPV material, the dc mobility can be changed by orders of magnitude. In all cases the mobility is well described by the empirical $\ln(\mu)\propto\sqrt{E}$ law. The specific E and T dependence of the data can be fitted within the (correlated) Gaussian disorder model. From the fitting procedure microscopic transport parameters are derived, which give a consistent picture of the influence of the molecular structure of the polymer on the charge transport. Broken conjugation naturally limits charge transport, mainly due to the effective dilution of the charge-transporting fraction of the material. For the fully conjugated materials the amount of energetic disorder appears to govern the charge transport properties. The use of a polymer with a high degree of regularity can significantly increase the mobility, due to the decrease of energetic disorder and likely the better ordering in the solid state, which favors charge delocalization. Ongoing research includes an investigation of the influence of the microscopic transport parameters on the device efficiency (photon/carrier).

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[1] J.H. Burroughes, D.D.C. Bradley, A.R. Brown, R.N. Marks, K. Mackey, R.H. Friend, R.L. Burn, and A.B. Holmes, Nature (London) **347**, 539 (1990).

[2] D. Braun and A.J. Heeger, Appl. Phys. Lett. **58**, 1982 (1991).

[3] R.H. Friend, R.W. Gymer, A.B. Holmes, J.H. Burroughes, R.N. Marks, C. Taliani, D.D.C. Bradley, D.A. Dos Santos, J.L. Brédas, M. Lögdlung, and W.R. Salaneck, Nature (London) **397**, 121 (1999).

[4] P.W.M. Blom, M.J.M. de Jong, and J.J.M. Vleggaar, Appl. Phys. Lett. **68**, 3308 (1996).

[5] I.D. Parker, J. Appl. Phys. **75**, 1656 (1994).

[6] P.W.M. Blom, M.J.M. de Jong, and S. Breedijk, Appl. Phys. Lett. **71**, 1 (1997).

[7] J. Salbeck, Ber. Bunsenges. Phys. Chem. **100**, 1667 (1996).

[8] C. Liedenbaum, Y. Croonen, P. van de Weijer, J. Vleggaar, and H. Schoo, Synth. Met. **91**, 109 (1997).

[9] D. Braun, E.G.J. Staring, R.C.J.E. Demandt, G.L.J. Rikken, Y.A.R.R. Kessener, and A.H.J. Venhuizen, Synth. Met. **66**, 75 (1994).

[10] P.W.M. Blom, M.J.M. de Jong, and M.G. van Munster, Phys. Rev. B **55**, R656 (1997).

[11] D.M. Pai, J. Chem. Phys. **52**, 2285 (1970).

[12] W.D. Gill, J. Appl. Phys. **43**, 5033 (1972).

[13] P.M. Borsenberger and D.S. Weiss, *Organic Photoreceptors for Imaging Systems* (Dekker, New York, 1993), Chap. 8, p. 181, and references therein.

[14] L.B. Schein, A. Peled, and D. Glatz, J. Appl. Phys. **66**, 686 (1989).

[15] H.C.F. Martens, H.B. Brom, and P.W.M. Blom, Phys. Rev. B **60**, R8489 (1999).

[16] G.H. Gelinck and J.M. Warman, J. Phys. Chem. **100**, 20 035 (1996); G.H. Gelinck, J.M. Warmand, and H.F.M. Schoo, J. Radioanal. Nucl. Chem. **232**, 115 (1998).

[17] H. Böttger and V.V. Bryksin, *Hopping Conduction in Solids* (Akademie Verlag, Berlin, 1985), Chap. 2, p. 40.

[18] B.I. Shklovskii and A.L. Efros, *Electronic Properties of Doped Semiconductors* (Springer-Verlag, Berlin, 1985), Chap. 5, p. 130.

[19] B.I. Shklovskii, Fiz. Tekh. Poluprovodn. **10**, 1440 (1976) [Sov. Phys. Solid State **10**, 855 (1976)].

[20] H. Bässler, Phys. Status Solidi B **175**, 15 (1993).

[21] D.H. Dunlap, P.E. Parris, and V.M. Kenkre, Phys. Rev. Lett. **77**, 542 (1996).

[22] S.V. Novikov, D.H. Dunlap, V.M. Kenkre, P.E. Parris, and A.V. Vannikov, Phys. Rev. Lett. **81**, 4472 (1998).