University of Groningen

Carrier-carrier Coulomb interactions reduce power factor in organic thermoelectrics

Koopmans, Marten; Koster, L. Jan Anton

Link to publication in University of Groningen/UMCG research database

Carrier–carrier Coulomb interactions reduce power factor in organic thermoelectrics

Marten Koopmans and L. Jan Anton Koster

COLLECTIONS

Paper published as part of the special topic on Organic and Hybrid Thermoelectrics

View Online   Export Citation   CrossMark

ARTICLES YOU MAY BE INTERESTED IN

Carrier–carrier Coulomb interactions reduce power factor in organic thermoelectrics

Marten Koopmans[a] (iD) and L. Jan Anton Koster[b] (iD)

AFFILIATIONS

Zernike Institute for Advanced Materials, University of Groningen, Groningen 9747 AG, The Netherlands

Note: This paper is part of the APL Special Collection on Organic and Hybrid Thermoelectrics.
[a]marten.koopmans@rug.nl
[b]Author to whom correspondence should be addressed: l.j.a.koster@rug.nl

ABSTRACT

Organic semiconductors are excellent candidates for low temperature thermoelectric generators. However, such thermoelectric applications require materials be doped and highly conductive. Here, we show how doping affects the Seebeck coefficient in organic semiconductors using kinetic Monte Carlo simulations. Employing a hopping transport approach, we demonstrate that at high dopant loading, carrier–carrier interactions can reduce the Seebeck coefficient. This results in systems with intrinsic disorder, still following Heike's formula for thermopower at high dopant density. Reducing these carrier–carrier interactions results in an increased Seebeck coefficient and power factor. Specifically, a realistic reduction in carrier–carrier interactions can increase the power factor by more than a factor 15, increasing ZT above 1 for organic thermoelectrics.

Published under an exclusive license by AIP Publishing. https://doi.org/10.1063/5.0071208

Organic thermoelectrics are starting to attract much attention because of their increasing performance and unique flexibility.[1–3] The performance of thermoelectric generators is characterized by the figure of merit $ZT = \sigma S^2 T / \kappa$, where σ is the electrical conductivity, S is the Seebeck coefficient, T is the temperature, and κ is the thermal conductivity.[4] In inorganic thermoelectrics, it is possible to engineer κ, but in organic thermoelectric materials, the value of κ is generally so low that it is not considered for optimization.[5] Therefore, organic thermoelectric materials are generally characterized via their power factor (PF), which is $S^2\sigma$. The primary method of increasing the PF is through doping, as this increases σ.

While doping increases σ, it also strongly affects the energetic landscape because of strong Coulombic interactions between the introduced charges. Charge carriers are known to get trapped by Coulomb traps created by dopant ions.[6,7] The effect of these Coulomb traps is strongest at low charge carrier densities, because at higher densities, the dopant Coulomb potentials start to overlap. This reduces the potential barrier for removing a charge carrier from a dopant.[6,7] Multiple ways have been shown to reduce the effect of these carrier–dopant (c–d) interactions. It is possible to physically increase the distance between the dopant ion and the host molecules to reduce the Coulombic interaction by adding molecular spacers in-between the host and dopant.[8,9] Another way to influence the Coulomb attraction

between host and dopant is by allowing charge to delocalize on the dopant, where larger molecules allow for more delocalization and, therefore, lower Coulomb attraction between host and dopant.[8]

While c–d interactions become less relevant with increasing carrier density, the opposite is true for carrier–carrier interactions. We have recently shown that under the assumption of hopping transport, the conductivity at typical electron densities for organic thermoelectrics is limited by c–c interactions.[10] Also these c–c interactions rapidly increase in strength upon further increase in the charge carrier density.

Intrinsically, organic materials are generally considered to have a Gaussian distribution of hopping sites, which resemble molecules or molecular segments, where the width of this Gaussian is determined by disorder in the material.[11] Up to medium charge carrier densities, this model can describe charge transport very well.[12–15] Carrier–carrier interactions were also shown to widen the DOS, but the Seebeck coefficient and power factor were not investigated.[10] Widening of the DOS in systems containing dopant ions has both been measured[16–18] and simulated.[10,19,20] Interestingly, the difference between only having c–c interactions or having both c–c and carrier–dopant (c–d) interactions in terms of the width of the DOS is small.[10]

The Seebeck coefficient is strongly, although not exclusively, affected by the DOS. The Seebeck coefficient can be calculated using[21]

$$S = -\frac{k}{q}\int_{-\infty}^{\infty}\frac{(E - E_F)}{kT}\frac{\sigma(E)}{\sigma}dE = -\frac{1}{qT}(E_{tr} - E_F), \quad (1)$$

where k is the Boltzmann constant, q is the unit charge, E is energy, E_F is the Fermi energy, and E_{tr} is the transport energy, which is the energy at which electronic conduction takes place on average. In the supplementary material, details on how we numerically obtain E_F and E_{tr} can be found. In disordered organic materials, E_{tr} is typically independent of charge carrier density, while E_F scales with charge carrier density. The difference increases with increasing with the increasing energetic disorder, leading to an increased S.[22] At a charge carrier density of 2×10^{18} cm^{-3}, lower than that in typical organic thermoelectrics,[23–27] c–c interactions dominate the density of states (DOS)[10] and, therefore, affect both E_F and E_{tr}, but the effect of c–c interactions on the S and power factor has yet to be investigated.

On the other hand, Heike's formula describes the Seebeck coefficient in the absence of interactions:[4] The Seebeck coefficient for a system of identical, non-interacting fermions distributed over equivalent sites is just the change in entropy of mixing upon the addition of a single charge carrier. In terms of the relative occupation of sites, c, one has[4]

$$S = \frac{k}{q}\ln\left(\frac{c}{1 - c}\right). \quad (2)$$

Therefore, Heike's formula contains no information on disorder, chemical structure, or transport mechanism. As a result, it can be argued that if S coincides with Heike's formula, the molecular sites must be equivalent and show no disorder.[28]

In this contribution, we will show how disorder, either intrinsic or as a result from dopant Coulomb potentials, increases the Seebeck coefficient compared to Heike's formula for thermopower [Eq. (2)] through energetic filtering. As dopant density increases, the effect of dopant induced disorder is shown to decrease. We also show that c–c interactions decrease the Seebeck coefficient, such that at 10^{19} cm^{-3} the Seebeck coefficient coincides with Heike's formula. While c–c interactions keep charge carriers apart, mobile charge carriers are shown to shield dopants very strongly. Finally, we show that reducing c–c Coulomb interactions can improve the power factor by more than an order of magnitude, even with dielectric constants close to the currently achievable values.[29]

We study the effect of Coulomb correlation on the Seebeck coefficient and power factor using a Gaussian disorder model with added dopant counter ions. A kinetic Monte Carlo algorithm is used to numerically obtain the results using a T of 300 K, a σ of 3 kT (77.6 meV), and a ε_r of 4 unless specified otherwise.[30] Dopant counter-ions are put in at random positions and share the Coulomb cutoff radius of one tenth of the simulation volume length, beyond which Coulomb interactions are not accounted for, with charge carriers. More details on the specifics of the simulations are in the supplementary material.

Figure 1 shows the simulated Seebeck coefficient vs the density of reacted dopants, which equals the number of free charge carriers. The effects of Coulomb interactions, both c–c and c–d, are modulated using the dielectric constant. First, we validate the applicability of Heike's formula by running KMC simulations without intrinsic disorder ($\sigma = 0$) and Coulomb interactions (neither c–c nor c–d). We observe in Fig. 1 that these simulations indeed show a Seebeck coefficient that exactly follows Heike's formula for thermopower, as all criteria for the validity of Heike's formula are met.
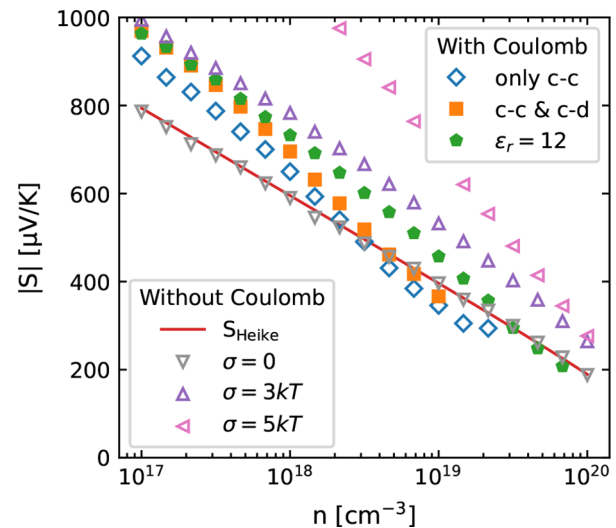
FIG. 1. The Seebeck coefficient vs concentration of charge carriers. A number of dopant ions equal to the number of charge carriers are added (filled symbols). In simulations with Coulomb interactions, only c–c, or both c–c and c–d, the dopants are left out (open symbols) to show the maximum possible effect of reducing c–d interactions and the energetic disorder is 3 kT. In the simulations without Coulomb interactions, c–c and c–d interactions are effectively removed by taking ε_r equal to 1000. The Seebeck coefficient from Heike's formula is plotted as a reference (solid line). The standard error of the mean is smaller than the symbol size.

At a charge carrier density of 10^{17} cm^{-3}, the other simulations show a Seebeck coefficient higher than given by Heike's formula. This is to be expected, as all these KMC simulations have energetic disorder, either intrinsically or induced by dopant ions, which is known to increase the Seebeck coefficient as it effectively applies energetic filtering.[20,31] This makes low energy charge carriers less conductive compared to high energy charge carriers, increasing the Seebeck coefficient as per definition. The KMC simulations with Coulomb interactions and without dopant counter ions (blue diamonds) show a slightly lower S than the simulations with counter ions because of the absent additional disorder induced by dopants.

With increasing density, it can be observed that the simulations with and without dopant counter ions and a ε_r of 4 show a comparable S that is reduced to that of Eq. (2). The simulations without Coulomb interactions (emulated by setting ε_r to 1000) show that c–c interactions cause the reduction in the Seebeck coefficient. The high ε_r effectively eliminates c–c and c–d interactions, leaving only the intrinsic disorder affecting the Seebeck coefficient. While there is some gain to be made by reducing c–c interactions, even completely eliminating them only increases the Seebeck coefficient by about 200 μV/K at 10^{19} cm^{-3} at a disorder of 3 kT. The Seebeck coefficient and especially the slope of the Seebeck coefficient can be seen to increase quite strongly when changing the disorder from 3 to 5 kT. While this might seem favorable, the increased disorder decreases the conductivity by more than an order of magnitude at a charge carrier density of 10^{19} cm^{-3}, resulting in a reduced power factor.

The reduction in the Seebeck coefficient when c–c interactions start to dominate can be explained within the framework of Heike's formula: Some sites are no longer available because the energy cost would be too high. The number of available sites decreases with

increasing carrier density, meaning the derivative of entropy with respect to charge carrier density, the Seebeck coefficient, decreases. This is analogous to increasing c in Heike's formula. The finding that c–c interactions can push the Seebeck coefficient close to that of Heike's formula is interesting, since a Seebeck coefficient being close to Heike's formula is sometimes seen as evidence of a lack of intrinsic disorder,[28] which is still present in the KMC simulations in Fig. 1. The charge carrier density at which the Seebeck coefficient starts to get close to Heike's formula is very close to $10^{18}\,\mathrm{cm^{-3}}$, the density at which c–c interactions were found to become significant in organic materials.[10,32]

Figure 2 shows the Seebeck coefficient vs the carrier density. Both total carrier density (n) and free carrier density (n_{free}) are shown, where n equals the number of ionized dopants and free carriers are defined as not being on a dopant site or a nearest neighbor site to a dopant. This means that a free carrier can still be trapped on a site that has low energy because of energetic disorder.

We show three different datasets, one without dopant ions, two dopant ions with varying c–d interaction strength. The c–d interactions are modulated by changing the depth of the Coulomb potential of dopant ions (E_{dop}), where a lower E_{dop} reduces the Coulomb interaction by capturing charge carriers more strongly.

The open symbols show S vs n. These data show that increasing the c–d interaction strength increases S at a constant n by inducing more energetic disorder. The closed symbols show S vs n_{free}. Interestingly, all data collapse onto the same line when S is plotted vs n_{free}, showing that only free carriers contribute to S. Charge carriers that are on dopants do not contribute as their Coulomb interaction is shielded by the dopant, effectively removing them from the system.

To show the effect of c–c interactions in the KMC simulations, Fig. 3 shows the probability of finding an electron at a certain distance from another electron normalized to the probability if the electrons were randomly placed. Figure 3(a) shows the relative probability at a charge carrier density of $10^{18}\,\mathrm{cm^{-3}}$. It can be observed that unscreened
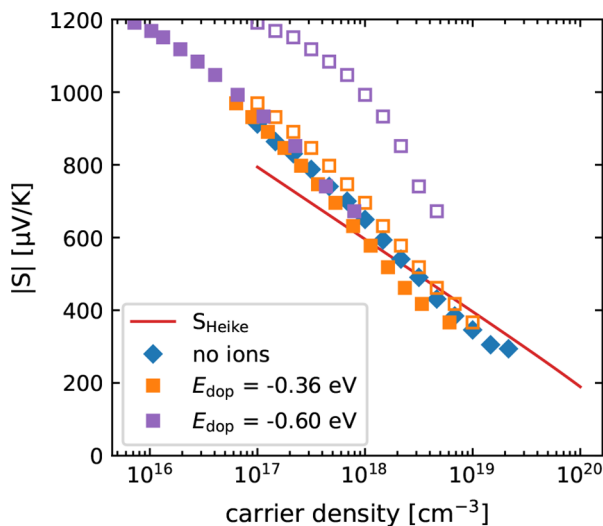
FIG. 2. The Seebeck coefficient vs carrier density. Both free carrier density (solid symbols) and total carrier density (open symbols) are shown, where free carriers are those that are not on a dopant site or on a nearest neighbor site to a dopant. The standard error of the mean is smaller than the symbol size.

electrons ($\varepsilon_r = 4$, no ions) sit relatively far apart. When dopant counter ions are introduced ($\varepsilon_r = 4$), the charge carriers can be observed to sit much closer together. This because the randomly placed dopants effectively screen part of the charge carriers, resulting in a dipole potential that drops off in strength much more strongly with increasing r compared to a monopole from a charge carrier. Interestingly, reducing Coulomb interactions (c–c and c–d, $\varepsilon_r = 12$) increases the probability of finding carriers very close together, but reduces the probability of finding two charge carriers at intermediate distance. This is indicative of less charge carriers being stuck close to a dopant when Coulomb interactions decrease in strength. While carriers sit further apart, still the Seebeck coefficient increases slightly.

Figure 3(b) shows the relative probability at a charge carrier density of $10^{19}\,\mathrm{cm^{-3}}$. When counter ions are present ($\varepsilon_r = 4$), the relative probability does not change strongly when compared to Fig. 3(a). This shows that charge carriers are mostly close to carriers that are screened by a dopant, especially when Coulomb interactions are strong. Even without Coulomb interactions strength ($\varepsilon_r = 1000$), the charge carriers show slight correlation. This can occur even without Coulomb interaction, as only one electron is allowed per grid point.

Since the PF is of critical importance to thermoelectric materials and the conductivity is limited by c–c interactions,[10] the way to improve the PF would be by reducing c–c interactions. In Fig. 1, we observed that the Seebeck coefficient was reduced by c–c interactions, meaning that it is possible to improve both the Seebeck coefficient and conductivity by reducing c–c interactions.

In Fig. 4(a), we show that the conductivity is limited by c–c interactions. As a result, the conductivity can be improved by reducing these interactions. We note that the distribution of charge carriers in a field-effect-transistor configuration is highly non-uniform and the impact of c–c interaction may very well be different.[33] At low density, we observe that the conductivity is a result from a convolution of intrinsic disorder and c–d interactions. At ε_r of 4, the conductivity is slightly lower than for ε_r of 8 and 12. This is because at ε_r of 4, c–d interactions are still strong compared to the energetic disorder, as the depth of the dopant potential is 0.36 eV, making the hop from the bottom of the potential to a site 1 nm away from it have an energy barrier of 180 meV while the intrinsic disorder is 0.078 eV. At ε_r of 8, the energy barrier for the shortest hop away from the dopant becomes 90 meV, which is similar to the energetic disorder, rendering the dopant potentials unimportant.

In Fig. 4(b), we show that the PF instead increases by more than a factor of 15 with decreasing Coulomb interactions (c–c and c–d). Considering the currently achievable values for ZT, this would bring the power factor far beyond the required ZT of 1 for application of organic thermoelectrics.[26] While it could be argued that the c–d interactions are mainly responsible for suppressing the conductivity, we have previously shown that at high dopant loading, c–c interactions are a possible culprit.[10] As this work builds on a Gaussian disorder model and assumes hopping conductivity, the conclusions should not be applied outside their domain of validity. Within this domain of validity, however, we show that reducing c–c interactions might be a promising route toward increasing the PF in disordered organic materials, even for realistically achievable dielectric constants in the low double digits.[29]

We conclude that Heike's formula for thermopower is very closely reproduced by KMC simulations, confirming its validity in hopping systems. Intrinsic disorder increases the Seebeck coefficient when it dominates the DOS, which happens at low charge carrier
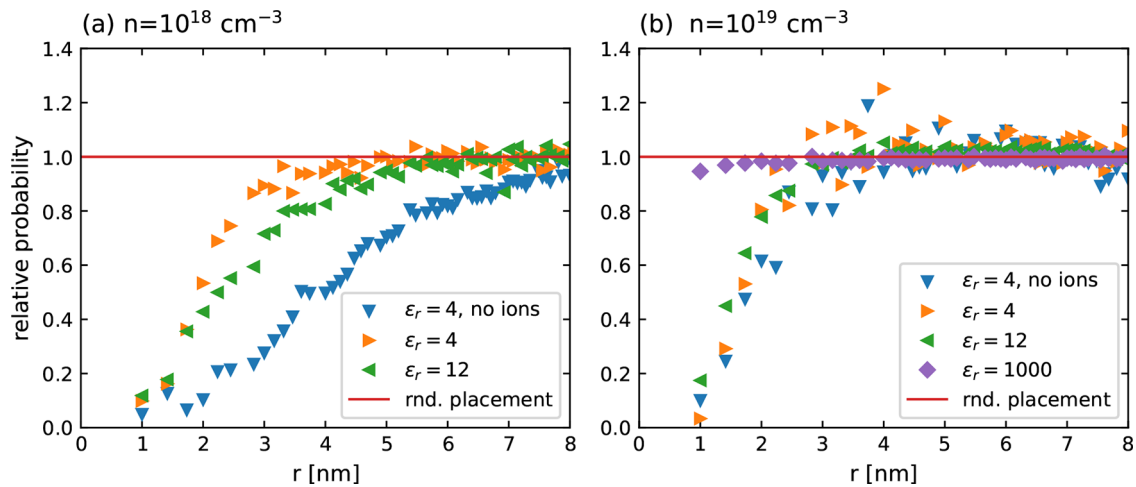
FIG. 3. The probability of finding an electron at a certain distance from another electron normalized to the probability of finding an electron at that distance if the electrons were randomly placed for a carrier density of (a) 10^{18} cm^{-3} and (b) 10^{19} cm^{-3}.
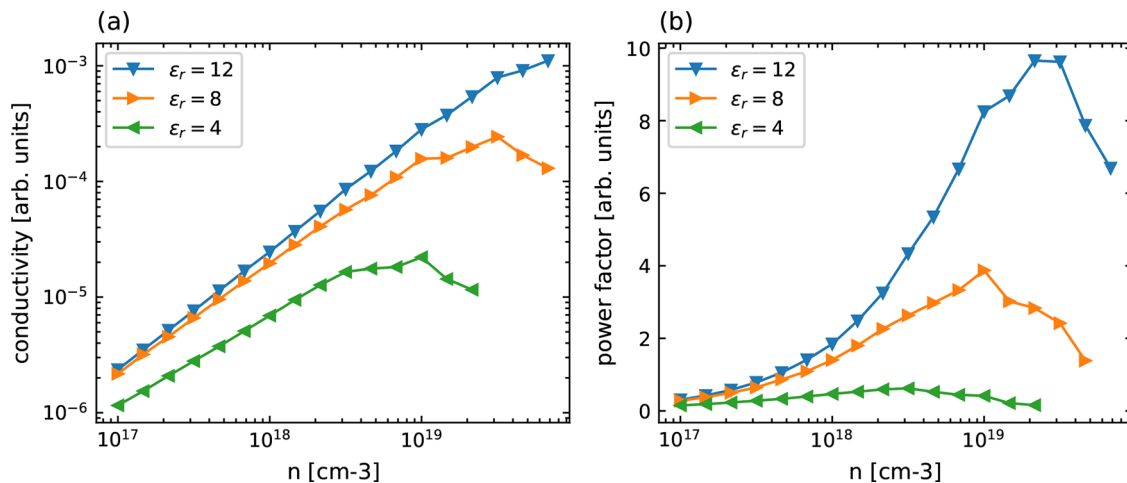
FIG. 4. (a) Conductivity vs charge dopant density for varying degree of Coulomb interaction (modulated by ε_r). (b) The power factor vs carrier density for varying Coulomb interaction (c–c and c–d) strength, which was modulated by ε_r. The standard error of the mean is smaller than the symbol size.

density or with little Coulomb interaction. Dopants are shown to trap charge carriers, but the Seebeck coefficient is uniquely determined by the number of charge carriers not trapped by dopants. Carrier–carrier interactions reduce the Seebeck coefficient, which can be seen at high carrier density. This makes it possible for the Seebeck coefficient for disordered materials to coincide with the value calculated from Heike's formula at higher charge carrier densities. Reducing carrier–carrier interactions increases both the Seebeck coefficient and conductivity at high doping densities, resulting in an increased power factor.

See the supplementary material for details about the KMC simulation parameters, the calculation of the transport energy and Fermi energy, and the relative probability of site occupation.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

REFERENCES

[1]O. Bubnova and X. Crispin, "Towards polymer-based organic thermoelectric generators," Energy Environ. Sci. **5**, 9345–9362 (2012).

[2] R. Kroon, D. A. Mengistie, D. Kiefer, J. Hynynen, J. D. Ryan, L. Yu, and C. Müller, "Thermoelectric plastics: From design to synthesis, processing and structure–property relationships," Chem. Soc. Rev. **45**, 6147–6164 (2016).

[3] B. Russ, A. Glaudell, J. J. Urban, M. L. Chabinyc, and R. A. Segalman, "Organic thermoelectric materials for energy harvesting and temperature control," Nat. Rev. Mater. **1**, 1–14 (2016).

[4] D. M. Rowe, *Thermoelectrics Handbook* (Taylor & Francis, 2006), p. 1008.

[5] Y.-J. Zeng, D. Wu, X.-H. Cao, W.-X. Zhou, L.-M. Tang, and K.-Q. Chen, "Nanoscale organic thermoelectric materials: Measurement, theoretical models, and optimization strategies," Adv. Funct. Mater. **30**, 1903873 (2020).

[6] V. I. Arkhipov, E. V. Emelianova, P. Heremans, and H. Bässler, "Analytic model of carrier mobility in doped disordered organic semiconductors," Phys. Rev. B **72**, 235202 (2005).

[7] V. I. Arkhipov, P. Heremans, E. V. Emelianova, and H. Bässler, "Effect of doping on the density-of-states distribution and carrier hopping in disordered organic semiconductors," Phys. Rev. B **71**, 045214 (2005).

[8] M. Schwarze, C. Gaul, R. Scholz, F. Bussolotti, A. Hofacker, K. S. Schellhammer, B. Nell, B. D. Naab, Z. Bao, D. Spoltore, K. Vandewal, J. Widmer, S. Kera, N. Ueno, F. Ortmann, and K. Leo, "Molecular parameters responsible for thermally activated transport in doped organic semiconductors," Nat. Mater. **18**, 242–248 (2019).

[9] J. Liu, G. Ye, H. G. O. Potgieser, M. Koopmans, S. Sami, M. I. Nugraha, D. R. Villalva, H. Sun, J. Dong, X. Yang, X. Qiu, C. Yao, G. Portale, S. Fabiano, T. D. Anthopoulos, D. Baran, R. W. A. Havenith, R. C. Chiechi, and L. J. A. Koster, "Amphipathic side chain of a conjugated polymer optimizes dopant location toward efficient n-type organic thermoelectrics," Adv. Mater. **33**, 2006694 (2021).

[10] M. Koopmans, M. A. T. Leiviskä, J. Liu, J. Dong, L. Qiu, J. C. Hummelen, G. Portale, M. C. Heiber, and L. J. A. Koster, "Electrical conductivity of doped organic semiconductors limited by carrier–carrier interactions," ACS Appl. Mater. Interfaces **12**, 56222–56230 (2020).

[11] H. Bässler, "Charge transport in disordered organic photoconductors a Monte Carlo simulation study," Phys. Status Solidi (b) **175**, 15–56 (1993).

[12] V. I. Arkhipov, P. Heremans, E. V. Emelianova, G. J. Adriaenssens, and H. Bässler, "Weak-field carrier hopping in disordered organic semiconductors: The effects of deep traps and partly filled density-of-states distribution," J. Phys.: Condens. Matter **14**, 9899–9911 (2002).

[13] M. C. J. M. Vissenberg and M. Matters, "Theory of the field-effect mobility in amorphous organic transistors," Phys. Rev. B **57**, 12964–12967 (1998).

[14] C. Tanase, E. J. Meijer, P. W. M. Blom, and D. M. de Leeuw, "Unification of the hole transport in polymeric field-effect transistors and light-emitting diodes," Phys. Rev. Lett. **91**, 216601 (2003).

[15] W. F. Pasveer, J. Cottaar, C. Tanase, R. Coehoorn, P. A. Bobbert, P. W. M. Blom, D. M. de Leeuw, and M. A. J. Michels, "Unified description of charge-carrier mobilities in disordered semiconducting polymers," Phys. Rev. Lett. **94**, 206601 (2005).

[16] X. Lin, G. E. Purdum, Y. Zhang, S. Barlow, S. R. Marder, Y.-L. Loo, and A. Kahn, "Impact of a low concentration of dopants on the distribution of gap states in a molecular semiconductor," Chem. Mater. **28**, 2677–2684 (2016).

[17] J.-P. Yang, W.-Q. Wang, F. Bussolotti, L.-W. Cheng, Y.-Q. Li, S. Kera, J.-X. Tang, X.-H. Zeng, and N. Ueno, "Quantitative fermi level tuning in amorphous organic semiconductor by molecular doping: Toward full understanding of the doping mechanism," Appl. Phys. Lett. **109**, 093302 (2016).

[18] C. Gaul, S. Hutsch, M. Schwarze, K. S. Schellhammer, F. Bussolotti, S. Kera, G. Cuniberti, K. Leo, and F. Ortmann, "Insight into doping efficiency of organic semiconductors from the analysis of the density of states in n-doped C 60 and ZnPc," Nat. Mater. **17**, 439–444 (2018).

[19] A. Fediai, F. Symalla, P. Friederich, and W. Wenzel, "Disorder compensation controls doping efficiency in organic semiconductors," Nat. Commun. **10**, 4547 (2019).

[20] G. Zuo, H. Abdalla, and M. Kemerink, "Impact of doping on the density of states and the mobility in organic semiconductors," Phys. Rev. B **93**, 235203 (2016).

[21] H. Fritzsche, "A general expression for the thermoelectric power," Solid State Commun. **9**, 1813–1815 (1971).

[22] A. V. Nenashev, J. O. Oelerich, and S. D. Baranovskii, "Theoretical tools for the description of charge transport in disordered organic semiconductors," J. Phys.: Condens. Matter **27**, 093201 (2015).

[23] J. Liu, L. Qiu, G. Portale, M. Koopmans, G. T. Brink, J. C. Hummelen, and L. J. A. Koster, "N-type organic thermoelectrics: Improved power factor by tailoring host–dopant miscibility," Adv. Mater. **29**, 1701641 (2017).

[24] D. Kiefer, A. Giovannitti, H. Sun, T. Biskup, A. Hofmann, M. Koopmans, C. Cendra, S. Weber, L. J. A. Koster, E. Olsson, J. Rivnay, S. Fabiano, I. McCulloch, and C. Müller, "Enhanced n-doping efficiency of a naphthalenediimide-based copolymer through polar side chains for organic thermoelectrics," ACS Energy Lett. **3**, 278–285 (2018).

[25] J. Liu, G. Ye, B. van der Zee, J. Dong, X. Qiu, Y. Liu, G. Portale, R. C. Chiechi, and L. J. A. Koster, "N-type organic thermoelectrics of donor–acceptor copolymers: Improved power factor by molecular tailoring of the density of states," Adv. Mater. **30**, 1804290 (2018).

[26] J. Liu, B. van der Zee, R. Alessandri, S. Sami, J. Dong, M. I. Nugraha, A. J. Barker, S. Rousseva, L. Qiu, X. Qiu, N. Klasen, R. C. Chiechi, D. Baran, M. Caironi, T. D. Anthopoulos, G. Portale, R. W. A. Havenith, S. J. Marrink, J. C. Hummelen, and L. J. A. Koster, "N-type organic thermoelectrics: Demonstration of ZT > 0.3," Nat. Commun. **11**, 5694 (2020).

[27] D. Kiefer, R. Kroon, A. I. Hofmann, H. Sun, X. Liu, A. Giovannitti, D. Stegerer, A. Cano, J. Hynynen, L. Yu, Y. Zhang, D. Nai, T. F. Harrelson, M. Sommer, A. J. Moulé, M. Kemerink, S. R. Marder, I. McCulloch, M. Fahlman, S. Fabiano, and C. Müller, "Double doping of conjugated polymers with monomer molecular dopants," Nat. Mater. **18**, 149–155 (2019).

[28] D. Venkateshvaran, M. Nikolka, A. Sadhanala, V. Lemaur, M. Zelazny, M. Kepa, M. Hurhangee, A. J. Kronemeijer, V. Pecunia, I. Nasrallah, I. Romanov, K. Broch, I. McCulloch, D. Emin, Y. Olivier, J. Cornil, D. Beljonne, and H. Sirringhaus, "Approaching disorder-free transport in high-mobility conjugated polymers," Nature **515**, 384–388 (2014).

[29] S. Rousseva, H. D. Besten, F. S. van Kooij, E. L. Doting, N. Y. Doumon, E. Douvogianni, L. J. Anton Koster, and J. C. Hummelen, "Reaching a double-digit dielectric constant with fullerene derivatives," J. Phys. Chem. C **124**, 8633–8638 (2020).

[30] M. C. Heiber, "Excimontec v1.0: An open-source software tool for kinetic Monte Carlo simulations of organic electronic devices," J. Open Source Softw. **5**, 2307 (2020).

[31] G. Kim and K. P. Pipe, "Thermoelectric model to characterize carrier transport in organic semiconductors," Phys. Rev. B **86**, 085208 (2012).

[32] F. Liu, H. van Eersel, B. Xu, J. G. E. Wilbers, M. P. de Jong, W. G. van der Wiel, P. A. Bobbert, and R. Coehoorn, "Effect of coulomb correlation on charge transport in disordered organic semiconductors," Phys. Rev. B **96**, 205203 (2017).

[33] A. Sharma, F. W. A. van Oost, M. Kemerink, and P. A. Bobbert, "Dimensionality of charge transport in organic field-effect transistors," Phys. Rev. B **85**, 235302 (2012).