



# Multiscale Concepts in Simulations of Organic Semiconductors

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## Abstract

We critically review simulation approaches developed to study organic solar cells, light emitting diodes, and field effect transistors. Special attention is paid to multiscale techniques. In particular, we discuss how to parametrize coarse-grained models for morphology and charge transport simulations, to account for finite-size effects, and to treat long-range interactions in small systems.

## 1 Introduction

Organic semiconductors are conjugated molecular compounds constituted of by carbon and hydrogen atoms and heteroatoms such as nitrogen, sulfur, and oxygen. Examples of organic semiconducting molecules are shown in Fig. 1. Conjugation is the key feature of these materials: Connected  $\pi$ -orbitals with delocalized electrons can donate or accept electrons easily, thus facilitating charge transfer reactions. Efficient charge transfer enables charge transport on a mesoscale. In their pristine

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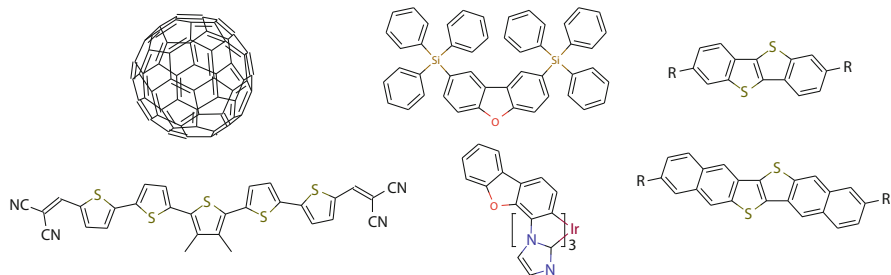
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W. Andreoni, S. Yip (eds.), *Handbook of Materials Modeling*,

[https://doi.org/10.1007/978-3-319-42913-7\\_39-1](https://doi.org/10.1007/978-3-319-42913-7_39-1)



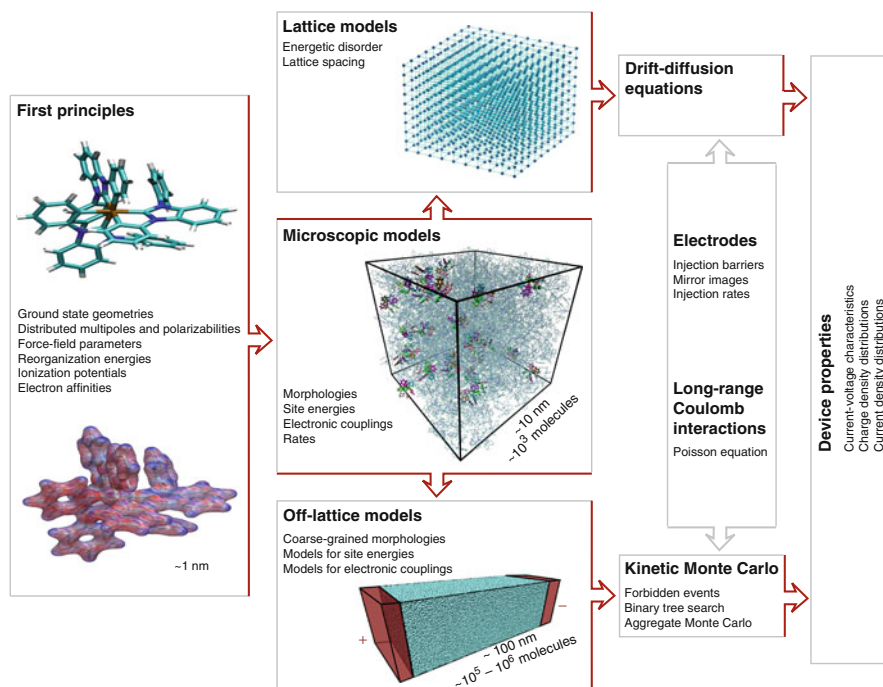
**Fig. 1** Example of a donor-acceptor combination used in an efficient bulk heterojunction solar cell (Fitzner et al. 2011). Host and guest materials forming an active layer of a blue phosphorescent light emitting diode (May et al. 2012a). Self-assembling organic molecules with high charge mobilities measured in field effect transistors (Ebata et al. 2007; Abe et al. 2015)

state, however, organic semiconductors are insulators. They become semiconducting only upon charge injection – from electrodes, by doping, or by photoexcitation.

Besides charge carrier mobility, other relevant physical properties of organic semiconductors are their absorption and emission spectra, ionization potential and electron affinity, solubility, self-assembling abilities, and glass transition temperature. These quantities depend on the molecular arrangement in the solid state, which can vary from crystalline to liquid crystalline to completely amorphous. The prime task of material design is to relate these quantities to the chemical composition and processing conditions, hence helping to prescreen compounds prior to their synthesis. Computer simulations can assist in finding such relationships by providing information which is not accessible to experimental techniques.

In silico prediction of properties of organic semiconducting devices requires multiscale approaches. Indeed, solving the time-dependent Schrödinger or the relativistic Dirac equation is computationally infeasible for an entire device. Likewise, timescales of dynamic processes such as charge or energy transfer span several orders of magnitude. Hence, charge/exciton kinetics cannot be treated by numerical methods with a fixed time step. It is normally coarse-grained into a master equation, as illustrated in Fig. 2, with rates evaluated with the help of the Fermi's golden rule (Athanasopoulos et al. 2007; Kirkpatrick et al. 2007; Nelson et al. 2009; Rühle et al. 2011; Schrader et al. 2012a,b; May et al. 2012a,b; Poelking and Andrienko 2013; Poelking et al. 2013; Kordt et al. 2015a). Multiscaling is essential not only to electronic processes, but also when addressing material processing, since self-assembling properties of conjugated materials, especially polymers can only be studied using coarse-grained models.

In spite of the implicit treatment of the electronic degrees of freedom, the approach based on the master equation is still computationally demanding. Indeed, electronic coupling elements must be evaluated for all pairs of neighboring molecules using first principles calculations. In addition, the evaluation of the self-consistent electrostatic and induction contributions to the free energy of localized charges require large interaction cutoffs. To reduce the computational overhead,



**Fig. 2** Multiscale simulations of organic semiconductors. Polarizable force-fields and electronic properties of isolated molecules obtained from first principles are used to generate morphologies and evaluate charge transfer rates in small systems (microscopic models). Coarse-grained models are parametrized either by matching macroscopic observables, e.g., charge mobility, of the microscopic and coarse-grained (lattice) models. After incorporating long-range electrostatic effects and electrodes, the resulting analytical expressions for mobility are then used to solve drift-diffusion equations for the entire device. Alternatively, off-lattice models can be developed by matching distributions and correlations of site energies, electronic couplings, and positions of molecules. The master equations for this model can be solved using the kinetic Monte Carlo algorithm, yielding macroscopic characteristics of a device. (Reproduced with permission from Adv. Funct. Mater. 25, 1955–1971, 2015)

we can parametrize a lattice model using the solutions to the master equation and then apply this model to study charge/exciton dynamics in larger systems. We will discuss this approach and its pitfalls in Sect. 2.

We can also design a stochastic model, an algorithm which generates the morphologies and the rates of the master equation without computationally demanding evaluations of electronic couplings and site energies. Together with an efficient kinetic Monte Carlo solver, the stochastic model can be used to simulate an entire device. Alternatively, it can be employed as an intermediate model to parameterize the closures of drift-diffusion equations. We will review this approach in Sect. 3.

To further reduce computational costs, we can mimic a macroscopically large system by replicating a relatively small simulation box in two or three spatial

directions. The use of periodic boundary conditions might, however, truncate the tails of the density of states, smoothen the free energy landscape, and artificially increase the charge carrier mobility. These artifacts can be corrected by various extrapolations, which will be discussed in Sect. 4.

In addition, periodic boundary conditions complicate correct treatments of long-range Coulomb forces. To accurately evaluate the density of states, the algorithm should be able to treat charged systems. Unlike the neutral surrounding of the charge, the charge itself should not be periodically repeated. Approaches applicable to partially periodic systems will be discussed in Sect. 5.

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## 2 Regular Lattices

From the computational point of view, the most efficient device modeling is based on the drift-diffusion equations (Knapp et al. 2010). Material properties enter these equations through closures, such as dependencies of diffusion constants and mobilities on charge density, temperature, and pressure. Closures provide a link to the chemical composition and material morphology and can be measured experimentally or computed using lower-resolution models. Lattice models are typical minimalist models required to compute such dependencies. In these models, a regular lattice represents material morphology, charge transfer rates decay exponentially with the intermolecular separation, and energy differences obey a Gaussian distribution. This approach was initiated by Bäessler and has been used by several groups to understand the role of traps, finite charge carrier density, energetic disorder, and other mesoscopic parameters on charge mobility (Pasveer et al. 2005; Cottaar et al. 2011; van der Holst et al. 2011; Bäessler 1993; Yimer et al. 2009; Novikov et al. 1998; Freire and Tonezer 2009).

Fits of phenomenological expressions for charge carrier mobility to the kinetic Monte Carlo simulations of lattice models are known as extended (correlated) Gaussian disorder models, EGDM, and ECDM. These expressions depend parametrically on the lattice constant  $a$ , the energetic disorder  $\sigma$ , and a prefactor  $\mu_0$ . In principle,  $a$  and  $\sigma$  can be evaluated in a relatively small system:  $a$  as the mean distance between neighboring molecules and  $\sigma$  as the width of the DOS that results from perturbative energy calculations.  $\mu_0$  can be extracted from charge transport simulations performed at different temperatures. This approach, however, does not lead to reliable parameterizations (Kordt et al. 2014, 2015a). Indeed, a multidimensional fit of simulated mobilities to the EGDM or ECDM expressions yields a very different set of parameters: the EGDM underestimates the energetic disorder, while the ECDM overestimates it. In both cases spatial site energy correlations are responsible for this discrepancy. EGDM does not include correlations and compensates for higher mobility values by reducing the energetic disorder  $\sigma$ . ECDM overestimates spatial correlations and compensates this by reducing the lattice constant (Kordt and Andrienko 2016). The discrepancy between microscopic values and fits to EGDM and ECDM tells us that parameters of these models do not have a clear physical interpretation. Nevertheless, they still provide

reasonable parameterizations and can eventually be used in conjunction with drift-diffusion equations.

A more conceptual drawback of EGDM and ECDM resides in the approximations incorporated in the rates of the master equation of the underlying lattice model, which constrain the functional form of  $\mu(\rho, \mathbf{F}, T)$ . To avoid such constraints, we can simply tabulate the mobility in a wide range of charge densities, temperatures, and electric fields with the help of the microscopic model. The tabulated function can then be used in the drift-diffusion equations solver (Kordt et al. 2015b). Computationally, the tabulation is only feasible with the help of a stochastic model, which we will discuss in the next section.

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### 3 Stochastic Models

In heterogeneous organic semiconducting devices, charges are nonuniformly distributed and charge density variations span several orders of magnitude. To cover the required density range in simulations, we need to simulate relatively large systems. This quickly becomes computationally demanding if all rates are evaluated from first principles (Kordt et al. 2015b). To remedy the situation, we can devise a phenomenological algorithm to parametrize the master equation (Baumeier et al. 2012; Stenzel et al. 2014; Brereton et al. 2014; Stenzel et al. 2014).

In case of morphologies, this algorithm reproduces given correlation functions, densities, and coordination numbers. For (approximately) spherically shaped molecules, the pair correlation function, or radial distribution function,  $g(r)$ , contains the most relevant structural information. To reproduce this function approximately, we can use a Poisson process (Baumeier et al. 2012; Kordt et al. 2014). More accurate coarse-graining techniques, such as iterative Boltzmann inversion (Tschoep et al. 1998; Reith et al. 2003) or inverse Monte Carlo (Lyubartsev and Laaksonen 1995; Soper 1996; Murtola et al. 2009), allow an exact reproduction of the radial distribution function (Kordt et al. 2015a). These methods optimize a pair interaction potential,  $U(r)$ , in a way that the corresponding  $g(r)$  is reproduced. The approach can also be applied to non-spherical molecules, by using several interacting sites per molecule (Rühle et al. 2009).

The second ingredient of the stochastic model is the connectivity. In the atomistic model only molecules within a certain cutoff distance are used for calculating charge transfer rates, while the rest of the rates is set to zero. This is justified by the fact that electronic coupling elements decrease roughly exponentially with molecular separation (Kordt et al. 2015a). The distance that determines whether or not two molecules are connected is given by their two closest atoms. Since this information is not present in the coarse-grained model, the resulting probability of two sites being connected is given by the corresponding probability extracted from the atomistic system.

For charge transfer rates, a simple procedure can be developed by analyzing the distributions of parameters entering the charge transfer rate. For example, in an amorphous mesophase of the organic semiconductor Alq<sub>3</sub>, the distribution

of electronic couplings at every particular intermolecular separation is Gaussian. The dependence of the width and the mean of this Gaussian on intermolecular separation can be determined from microscopic simulations. The spatial correlations of site energies can be introduced by using a moving-average procedure. In this approach, the site energies of the neighbors within a certain cutoff are mixed into initially independent Gaussian site-energy distribution (Kordt and Andrienko 2016). Electronic coupling elements can also be generated using appropriate distributions. These distributions are, however, separation-dependent: the logarithm of squared transfer integrals,  $\log J^2$  (which is often Gaussian distributed) depends on molecular separation. In the stochastic model, transfer integrals are then drawn from the distant-dependent distributions.

Stochastic models developed for amorphous mesophases of Alq<sub>3</sub> and DCV4T could reproduce the mobility-field and mobility-density dependencies (Baumeier et al. 2012; Stenzel et al. 2014; Kordt et al. 2014). This indicates that they indeed can serve as an intermediate step between the completely microscopic descriptions and macroscopic, drift-diffusion-equation-based models. The current challenge is to extend such models to anisotropic and heterogeneous systems.

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## 4 Finite-Size Effects

The dependence of macroscopic observables on the system size has been experimentally observed in organic semiconductors in systems with large energetic disorder or broad density of states. For example, time-of-flight measurements of mobility are impossible in thin organic films, where charge transport is dispersive and transients do not have a characteristic plateau used to determine the transient time (Scher and Montroll 1975; Borsenberger et al. 1991, 1992, 1993). Using thicker samples normally remedies the situation.

A similar effect is also observed in simulations of small systems. It turns out that replicating the simulation box still gives incorrect (higher) values of charge carrier mobility (Lukyanov and Andrienko 2010). Indeed, all duplicated boxes have exactly the same (and small) number of independent site energies. Statistical averages are performed over this small set of site energies, and charge carriers traverse the sample at a different (higher) temperature than in an infinitely large system. Surprisingly, an additional averaging over different, statistically uncorrelated, but still repeated snapshots does not help, since the origin of the problem lies in the limited number of available for every distinct Monte Carlo simulation sites.

This type of finite-size effects becomes much smaller at higher temperatures, since the relevant dimensionless parameter (at least in the Gaussian disorder model) is the width of the site energy distribution  $\sigma$  divided by  $k_B T$ . In fact, an empirical expression,  $(\sigma/k_B T_{ND})^2 = -5.7 + 1.05 \ln N$ , can be used to estimate at what temperature the “transition” between the dispersive and non-dispersive transport occurs, where  $N$  is the number of the hopping sites (molecules) in the system. This observation has been used to perform an empirical correction of finite-size effects (Lukyanov and Andrienko 2010): Nondispersive mobilities were calculated

for a set of temperatures above  $T_{\text{ND}}$ . Then, an explicit temperature dependence was used to extrapolate the value of mobility to room temperature.

This method relies on an explicit knowledge of the temperature dependence of mobility. While the exact analytical expression of this dependence is available only for one-dimensional systems (Derrida 1983; Seki and Tachiya 2001),

$$\mu(T) = \frac{\mu_0}{T^{3/2}} \exp \left[ - \left( \frac{a}{T} \right)^2 - \left( \frac{b}{T} \right) \right],$$

it can still be used in a three-dimensional case in a rather broad temperature range, as was verified by performing simulations for systems of different sizes and at different temperatures (Lukyanov and Andrienko 2010).

An alternative method is to determine the mobility as a function of the box size. For one-dimensional systems and Gaussian DOS, the mobility is inversely proportional to the number of visited sites  $N$  (Kordt et al. 2016),

$$\mu(N) = \mu_\infty \left( 1 + \frac{c}{N} \right).$$

Using this relation, it is possible to extrapolate the finite-size mobilities to its macroscopic value  $\mu_\infty$ .

To illustrate the relevance of such extrapolation, charge transport in the amorphous mesophase of Alq<sub>3</sub> was simulated in systems of different sizes (Lukyanov and Andrienko 2010). In a system of 512 molecules, the simulated mobility was of the order of  $10^{-6}$  cm<sup>2</sup>/Vs, while in a box of 4096 molecules, an order of magnitude lower value was measured. Extrapolation procedure resulted in a mobility of  $10^{-9}$  cm<sup>2</sup>/Vs, which is three orders of magnitude lower than the one simulated in a small system. Note that the magnitude of the correction is very sensitive to the value of the energetic disorder  $\sigma$ . While in Alq<sub>3</sub>  $\sigma = 0.14$  eV, in an amorphous DCV4T  $\sigma = 0.25$  eV, the value of mobility can be overestimated by seven(!) orders of magnitude (Kordt et al. 2014).

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## 5 Long-Range Interactions

An important ingredient of charge and energy transfer reactions is the free energy difference between the final and initial states or the driving force. In organic semiconductors this energy difference is often evaluated in a perturbative way, starting from an isolated molecule and then calculating the electrostatic and induction contributions of the environment. Coulomb interactions of partial charges or higher distributed multipoles are inherently long-range and require special summation techniques, e.g., Ewald summation, which is widely used in atomistic molecular dynamics simulations (Ewald 1921). In this approach, the Coulomb potential is split into two terms, one of which is converging fast in real and the other in reciprocal space. Induction contribution can also be incorporated in such a

scheme (Ren and Ponder 2003). While this method is well developed, it is designed for neutral systems, whereas for charge transport, our interest lies in energies of a localized charge interacting with the neutral environment. The presence of a charged excitation and its polarization cloud violates the periodicity of the system, which is essential for calculations in the reciprocal space.

A computationally more demanding solution is to evaluate electrostatic sums for spheres centered around the charged molecule (bulk) or cylinders (slab) of radius  $R$ . In both cases charge-quadrupole interactions scale as  $1/R$ , making it possible to extrapolate to  $R \rightarrow \infty$  (D’Avino et al. 2016). A more elegant solution to this problem has been proposed by Poelking and Andrienko (2016). The nonperiodic (foreground) part of the system which incorporates the charged excitation and its induction cloud is superimposed onto a periodic, neutral background, which is computed using the Ewald summation method. The real-space interaction between these two regions is mediated by fields created by the background charge distribution, including induced moments. A modified shape term (Smith 1981) is added to account for the net charge and quadrupole of the simulation box. This term takes into account surface effects and depends on the summation geometry.

By applying this technique to organic/organic interfaces, it has been shown that a cutoff of 4–8 nm is sufficient to converge the energy of a periodic three-dimensional system (Schwarze et al. 2016). For ordered interfaces, the convergence turns out to be significantly slower: for a 12 nm thick slab, the energy is far from converged even for the cutoff of 22 nm (Poelking and Andrienko 2016, 2015; Poelking et al. 2014). Hence, this method should be used for two-dimensional periodic systems or, in general, heterogeneous systems with a long-range molecular ordering. The differences between using simulations with a cutoff and without it are remarkable: the additional electrostatic/induction contribution can change from 0.5 eV (10 nm cutoff) to  $-0.7$  eV (infinite system). The correct treatment of electrostatic can therefore *reverse* the role of donor and acceptor as conditioned by gas-phase energy levels and energy levels calculated with a seemingly ample cutoff. It also predicts that the energy profiles for electrons and holes are flat at the organic-organic interfaces, while cutoff-based calculations lead to a significant level bending. Finally, it allows to establish a relationship between structural coherence and state energetics: the structural coherence is probed up to a  $\mu\text{m}$  scale, hence emphasizing the role of extended crystallites (and their alignment) at interfaces.

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## 6 Outlook

There are still a few directions where substantial method development is required to achieve a parameter-free modeling of realistic devices. An important step is to explicitly include the many-body induction interactions to the solution of the master equation. This, however, requires the reevaluation of rates at every Monte Carlo step and is computationally demanding. Another issue is the quantitative treatment of excited states embedded in a heterogeneous polarizable molecular environment, including descriptions of charge-exciton and exciton-exciton interactions. These



advancements are absolutely vital for devising accurate structure-property relationships for organic semiconductors.

**Acknowledgements** This work was supported in part by the BMBF grants MEDOS (FKZ 03EK3503B), MESOMERIE (FKZ 13N10723), and InterPhase (FKZ 13N13661). The project has received funding from the NMP-20-2014 – “Widening materials models” program under Grant Agreement No. 646259 (MOSTOPHOS). DFG is acknowledged for financial support through the collaborative research center TRR 146.

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