

Multiscale Simulation and Optimization of OLED Devices with Quantum Chemistry and Kinetic Monte Carlo Simulations

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Abstract

The development of efficient and durable Organic Light-Emitting Diodes (OLEDs) necessitates a comprehensive understanding of processes spanning multiple scales, from molecular interactions to device-level performance. This presentation introduces an integrated multiscale simulation approach utilizing the Amsterdam Modeling Suite and Bumblebee to model, simulate, and optimize OLED devices. By combining quantum mechanical calculations with 3D kinetic Monte Carlo simulations, this methodology provides insights into charge transport, exciton dynamics, device degradation, and optical outcoupling, facilitating the design of high-performance OLEDs.

Author Keywords

OLED; multiscale simulation; virtual screening; 3D kinetic Monte Carlo; quantum chemistry; phosphorescence; spin-orbit coupling; emission width, charge transport; exciton dynamics; device degradation; external quantum efficiency

1. Introduction

At present, the development of new organic electronic materials and devices largely occurs by following empirical recipes and by trial and error. This is a costly and cumbersome process that often does not lead to the expected results and unpredictably delays the release of a new material or device. Rational design, based on predictive atomistic and device simulations, is currently gaining more and more interest due to the large acceleration it can bring to research and development cycles, thus reducing time-to-market and wasteful experiments. Among the top priorities in the OLED industry is to understand, predict and optimize the efficiency and lifetime of devices. This is a hard challenge due to the multiple processes involved in the operation and various degradation pathways, all originating at the material level, which ultimately affect operation at the device level. Degradation or poor transport or emission at a few key molecular sites can result in a large and unexpected performance drop. Although experimental characterization of the optoelectronic and degradation pathways is possible, it is usually insufficient to allow for developing a strategy towards devices with improved lifetime. Integrating multiscale simulations in R&D becomes critical and allows to virtually investigate the effects of many material and stack parameters on the efficiency and degradation lifetime of OLEDs.

2. Multiscale Simulation Methodology

Atomistic Simulations: To further improve the efficiency and lifetime of OLEDs, ultimately the properties of the underlying materials need to be optimized at the atomistic level. In materials science, as in other fields, modeling has become a widespread tool that is integrated with experiments for a holistic R&D approach.

In particular, high-throughput computational screening may considerably reduce experimental costs for synthesizing and testing new materials. For OLED materials, the Amsterdam Modeling Suite (AMS) provides a variety of methods, including quantum chemistry, semi-empirical, force-field based and machine learning methods. Relevant material properties such as charge mobilities [1], phosphorescent lifetimes [2], and emission spectra can be accurately predicted with density-functional theory (DFT) based methods (Figure 1), and recent so-called GW approaches [3], including spin-orbit coupling (SOC) and environment effects.

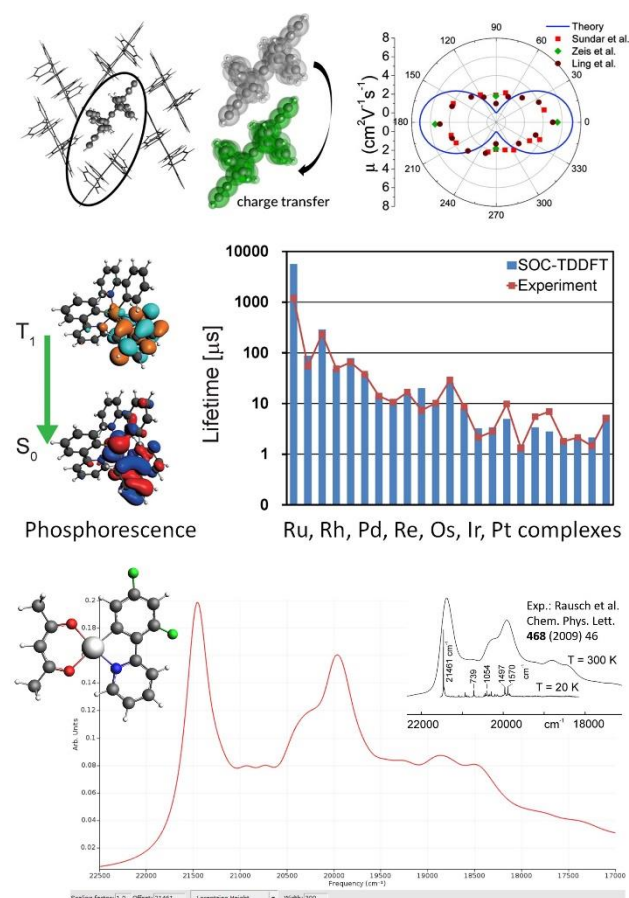


Figure 1. Comparing experimental and calculated charge mobilities (top), phosphorescence lifetimes (middle), and emission spectra (bottom)

For the deposited organic electronic materials, the local disorder and anisotropy need to be taken into account, as orientations and local dipole effects will affect the optoelectronic properties. We developed an OLED workflow tool [4] that simulates the physical vapor deposition processes. The workflow creates (doped) organic

electronic thin films and automatically calculates optoelectronic properties. For a set of known materials, calculated properties such as densities, ionization potentials (IPs) and electron affinities (EAs) are in good agreement with experiment.

Device Simulations: To study physical processes at the device level, various approaches are available, most notably 1D drift diffusion (1D DD), Master Equation (ME), and 3D kinetic Monte-Carlo (3D KMC) approaches. 1D DD is extensively used for simulations at the continuum level, but this method has limitations in situations where three-dimensional percolation effects become important [5]. Charge-carrier relaxation effects and the treatment of molecular-scale mixed materials such as those used in the emissive host-guest layers in OLEDs are also notable cases where 1D DD fails.

To address these shortcomings, methods like 3D ME and 3D KMC are employed [6-8]. While 3D ME treats the charge transport by simulating the probability of charge occupation at each molecular site, 3D KMC explicitly tracks the hopping events of discrete charges in between molecular sites. The mechanistic approach of 3D KMC allows for great versatility in simulating complex and highly correlated processes that can affect each other. Moreover, 3D KMC simulations provide the highest level of detail to the modeler and fundamental insights for understanding the device behavior at the microscopic level. This opens up many opportunities for addressing the device shortcomings and can lead to a very quick and targeted device optimization. In contrast with 1D models, 3D KMC provides time and 3D spatially resolved views of the device operation. 3D KMC does not have any limitation in terms of use cases. However, due to the inclusion of all microscopic-scale events, it can be computationally expensive. Recent acceleration methods in the 3D KMC code Bumblebee [9] enable fast virtual screening of different, complex OLED stacks under a broad range of operating conditions, including transient processes.

Bumblebee can simulate different complex stack designs and access many performance characteristics:

- Time-resolved electro- and photoluminescence
- J(V) curves and color point
- Efficiency roll-off
- Degradation
- Optical outcoupling

These properties depend on various physical properties and processes:

- Energy levels (IP, EA) and gaps
- Disorder (charge carriers, excitons)
- Exciton diffusion
- Exciton quenching and annihilation
- Degradation mechanisms

which can be obtained from the atomistic simulations, experiments, or both.

Using physical properties from the OLED workflow in AMS in conjunction with Bumblebee 3D KMC, researchers can design new host materials, blocking layers, charge transport materials, sensitizers, and emitters, **simultaneously** with the stack design for a holistic, multiscale approach (Figure 2). By zooming in on the

most promising materials and combinations to make and test in the lab, valuable time and resources can be saved.

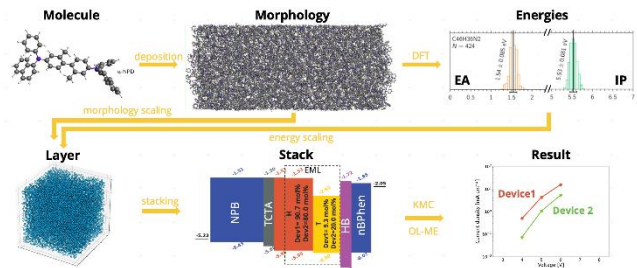


Figure 2. Multiscale simulation of OLED devices

3. OLED device case studies

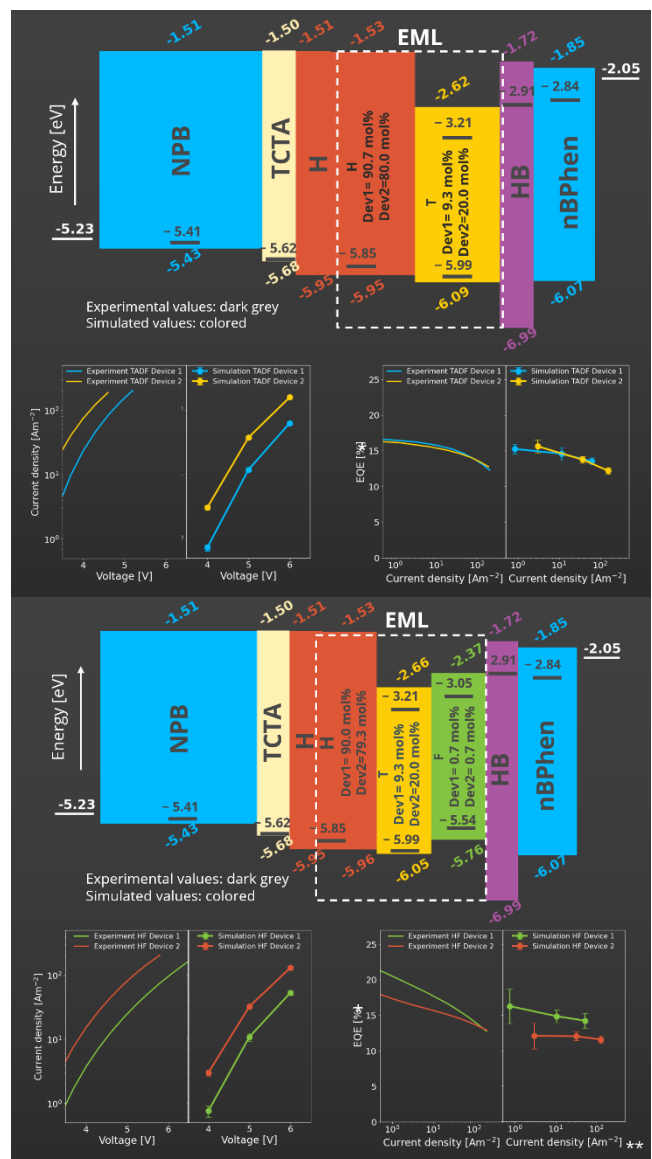


Figure 3. Case studies of TADF (top) and HF (bottom) devices from Cynora

Hyperfluorescent (HF) and Thermally-activated delayed fluorescence (TADF) devices: The effect of doping: To validate the initial multiscale simulation tool, we conducted a pilot project with Cynora to study the effect of host-guest concentrations on J(V) and external quantum efficiency (EQE) for thermally activated delayed fluorescence (TADF) and hyperfluorescence (HF) devices.

As shown in Figure 3, excellent qualitative and good quantitative agreement was obtained with the experiment. The 3D KMC results furthermore give insight into the percolative transport of electrons and holes, selective emission processes, and the main degradation pathways and loss mechanisms. These insights could help improve performance through rational design at the material and stack levels, which can in turn be tested virtually to save previous lab resources.

4. Conclusion and outlook

We have developed a parameter-free, multiscale modeling tool that enables researchers to virtually test new materials and stacks from atomistic to device level. The simulations can be used to predict the most promising material combinations as well as stack designs and give insight into the underlying fundamental mechanisms, aiding rational design. To further accelerate and improve predictions across length and time scales, we are working on Machine Learning (ML) models for forward predictions, with the ultimate aim to develop inverse prediction models that suggest the best material combinations and stack designs to achieve desired properties at the device level.

5. References

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