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Use case: Ab-initio calculation of a minimal working example OLED stack

Introduction

In this use case we demonstrate how the full Nanomatch software stack is applied to compute essential properties of an OLED stack: J-V characteristics, charge distribution along the device and quantum efficiency. In order to highlight the methodology, we restricted ourselves to the simplest model: a bilayer OLED. While this setup is a rather "bad" OLED in terms of quantum efficiency, it illustrates the application of all modules in the OLED workflow, and extension to arbitrarily complex multilayer stacks is straight forward.

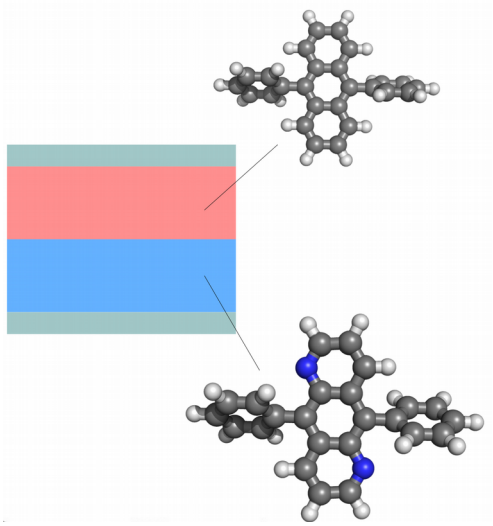

Figure 1: Illustration of the device setup of the minimal bilayer OLED device.

The minimal working example in this use case is a bilayer OLED stack consisting of the molecules ADP and ADP with two carbons substituted by Nitrogen (thereby lowering the orbital energy levels) ADP/N. A schematic setup is illustrated in the Figure to the left.

In order to compute the device characteristings starting from first principles, we follow the standard OLED workflow used in many applications (see http://www.nanomatch.com/index.php/en/products):

1. Single molecule optimization and generation of customized force fields (for bonded and nonbonded interactions)of both molecules using the PARAMETRIZER and the DIHEDRALPARAMETRIZER module.

2. Generation of three morphologies consisting of 1000 molecules each for the bulk materials and 1500 for molecules for the interface with atomistic resolution using DEPOSIT:
   - pristine ADP morphology for the calculation of charge transfer properties within the ADP layer
   - pristine ADP/N morphology for the calculation of charge transfer properties within the ADP/N layer
   - double layer of ADP/N deposited on top of ADP for the calculation of charge transfer properties between ADP and ADP/N at the interface

3. Calculation of density of states and electronic couplings between ADP+ADP, ADP/N+ADP/N and ADP+ADP/N based on the DEPOSIT morphologies using QUANTUMPATCH.
4. Based on the results from QUANTUMPATCH, we performed charge transport simulations of the full bilayer system, including electrodes, using LightForge KMC at different values of the electric field.

Detailed parameters for each step are given below.

Results

EA and IP levels

To approximate EA/IP levels, the orbital energy distributions were computed by QUANTUMPATCH by computing the orbital energy for each molecule in its unique electrostatic environment, and are displayed in the Fig. to the right. The Fermi levels of the electrodes used in the subsequent LightForge calculation are included for comparison.

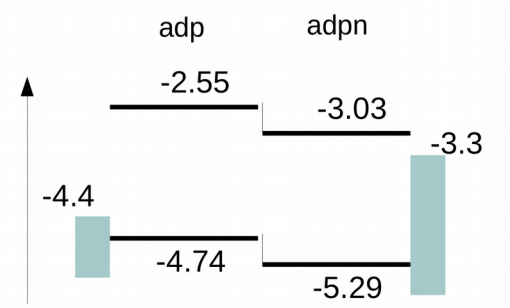

Figure 2: Energy levels of electrodes and layers computed with QuantumPatch.

In this setup, electrons will be injected at the right electrode whereas holes will be injected in the adp layer from the left side. At the interface, electrons and holes can then form excitons and decay under the emission of a photon.

Distribution of charge carriers, J-V characteristics and quantum efficiency (roll-off)

I-V and voltage dependent IQE (roll off) are displayed in Fig. 3. With IQE values of the order of 10%, this OLED setup is very inefficient, but helps to illustrate how thorough analysis of microscopic effects and processes can lead to a strategy for device improvement. The efficiency roll-off towards high voltages can be explained by analyzing the charge distribution and recombination zones shown in Fig.4. At 2.0 V the charge concentration in the device and particular at the interface is lower than at 5.75 V. As most excitons (green) are formed at the interface, quenching is increased in the second case. At higher voltage the interface doesn't block charges effectively leading to exciton formation close to the electrodes. Excitons created there have a high chance to be quenched. At lower voltages the interface acts as an efficient blocking layer thus preventing exciton formation close to the electrodes, where charge concentration is high and exciton quenching thus likely. At high voltages the interface does not block charge as effectively thus increasing exciton formation closer to the electrodes. An increased charge carrier density at the interface also increases exciton quenching for excitons formed there.

From these results we can conclude the following: First, high charge carrier density (of both types) leads to strong exciton quenching and thereby reduces IQE. OLED efficiency can therefore be improved by balancing charge carrier distribution and spatially separate charge accumulation and exciton creation, e. g. by including guest-host emission layers. Second, there is non-vanishing density of holes and electrons at the electrode opposite to

their injection and charge carriers are lost by traveling to the opposite electrode. This can be overcome by including electron- or hole-blocking layers next to the emission layer.

We are well aware that this is not a novel fact, but it illustrates how simulation results can used to identify bottlenecks and improve device efficiency by targeted modification of device setups.

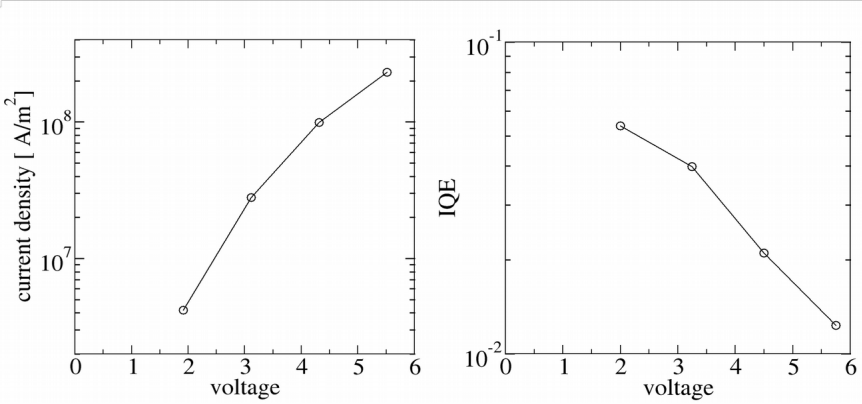

Figure 3: IV-characteristics and efficiency roll of as calculated by our multiscale workflow. At 2V the IQE for singlets is 20%, all triplets are quenched resulting in an overall poor IQE of 5%.

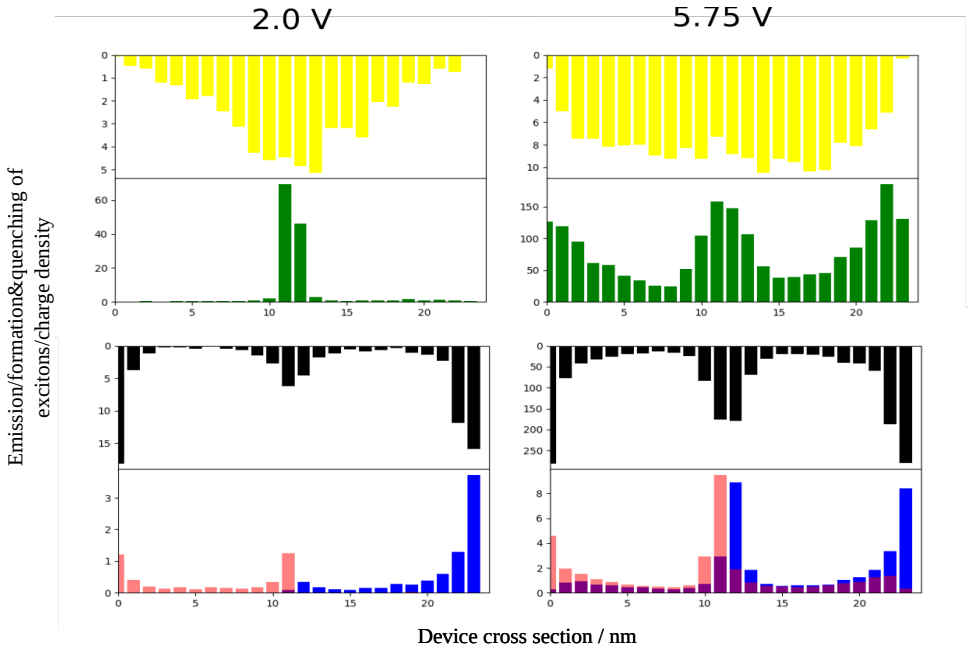

Figure 4: Yellow: Distribution of photon emission over the device cross section. Green: distribution of exciton formation. Red: distribution of holes. Blue: distribution of electrons. Black: exciton quenching. At 2.0 V the charge concentration in the device and particular at the interface is lower than at 5.75 V. As most excitons (green) are formed at the interface, quenching is increased in the second case. At higher voltage the interface doesn't block charges effectively leading to exciton formation close to the electrodes. Excitons created there have a high chance to be quenched.

Parameters used in the calculation:

Parametrizer:

Geometry optimization	DFT (b-p, def2-SV(P))
Partial charges	DFT (b-p, def2-SV(P))
Dihedral parametrization	Hartree-Fock (PM7)

Deposit:

Number of molecules	1000
Number of SA cycles	10
Number of steps per cycle	140.000
Initial temperature	4000K
Final temperature	300K

QuantumPatch:

No. of Mol in inner shell	20
No. of partial charge steps	7
Method	TD-DFT, b-p, def2-SV(P)
No. of excitations	30
Hybrid mode	Off

LightForge:

As LightForge is a multi-purpose KMC tool, the possible range and amount of combinations of settings is large compared to the previous modules. In case of doubt, leave settings on their standard value and only import the necessary files from QuantumPatch. Settings used in this specific use case are displayed in the table below.

general		
Morphology type	Stochastic extension	
physics		
Particles types	Holes, electrons, excitons (leave enabled exciton settings on standard values)	
device		
Layer 1 and 2 (generate second layer with the green „+“)		
Lx,Ly,Lz	12,12,12	
Morphology file	Import com.dat from QuantumPatch module: adp for layer 1, adp_n for layer 2	
Onsite energy source	Gaussian	
Molecule species	Layer 1	Layer 2
Species id	0	1
Ionization potential (values from QP calculation)		
Mean	4.74	5.29
Disorder	0.082	0.12
Electron affinity (values from QP calculation)		
Mean	-2.55	-3.03
Disorder	0.08	0.11
Excitation energies		
Mean	0.8	0.8
Lambda	0.25	0.25
Electrode 1		
Workfunction	-4.4	
Coupling model	QM fit	
Electrode coupling file	Adp/homo_j.dat (import from	

	QuantumPatch)
Electrode 2	
Workfunction	-3.3
Coupling model	QM fit
Electrode coupling file	Adpn/lumo_j.dat (import from QuantumPatch
Topology	
Max neighbours	40
Transfer integral source	QM expansion
Pair parameters 1	
Molecule type id1	0
Molecule type id2	0
Hole transfer integrals	Adp/homo_j.dat
Electron transfer integrals	Adp/lumo_j.dat
excitons	
Foerster radius forwards	1.5
Foerster radius backwards	1.5
Pair parameters 2	
Molecule type id1	1
Molecule type id2	1
Hole transfer integrals	Adpn/homo_j.dat
Electron transfer integrals	Adpn/lumo_j.dat
excitons	
Foerster radius forwards	1.5
Foerster radius backwards	1.5
Pair parameters 3	

Molecule type id1	0
Molecule type id2	1
Hole transfer integrals	interface/homo_j.dat
Electron transfer integrals	interface/lumo_j.dat
excitons	
Foerster radius forwards	1.3
Foerster radius backwards	1.7
Operation	
Simulations	15
Measurement	DC
Temperature	300
Field strength	0.08, 0.13, 0.18, 0.23
Field direction	1,0,0
Initial charges	Holes:0, electrons:0
computational	
IV fluctuation	0.001
Max iterations	4000000
Parallel	true