Elements in materials modelling of doped organic semiconductors

Purpose of this document:

Definition of a data organisation that is applicable to ALL materials modelling simulations. The fiche should contain all elements that are needed to describe a simulation. This information spans from the end-user (manufacturer) information to the modelling details.

| THE SIMULATION			
1	**USER CASE**		Device characteristics for organic light emitting diodes made with small molecule films
2	**CHAIN OF MODELS**	**MODEL 1** QC-ABINITIO	Higher level ab-initio models (model 1.1.2 in ROMM)
		MODEL 2 MC/MD	Molecular Dynamics (MD) methods to grow atomistic morphologies (model 2.2 in ROMM)
		MODEL 3 CGMD	Coarse Grained Molecular Dynamics, CGMD, (mesoscopic particle model 3.2 in ROMM) finds packing arrangements of entities which are ellipsoids representing groups of atoms
		MODEL 4 QC-POLARON	Time-independent Schrödinger Equation TISE, (linear scaling, constrained DFT, electrostatic embedding) finds charge and energy transfer rates and on site energies (thousand atoms) (electronic model 1.1.3 in ROMM)
		MODEL 5 QC-EXCITON	Electronic model (effective Hamiltonian) (model 1.2 in ROMM)
		MODEL 6 BTE	Semi-classical Boltzmann transport equation, BTE (statistical mechanics electronic model 1.4.2 in ROMM[1]) predicts charge and energy transport with as post processor an optical model, OPT, model 8
		MODEL 7 DD	Drift diffusion model, DD, (continuum model 1.4.1 in ROMM) predicts charge and energy transport with as post process or an optical model, OPT, model 8.
		MODEL 8 OPT	Optical model (continuum model, 4.6 in ROMM) predicts light emission and absorption used as post processor to the BTE and DD model.
3	**PUBLICATIONS**		
4	**ACCESS CONDITIONS**		Models 1,3,5. Fiesta, BigDFT, Mescal resp.are open source. DEPOSIT model x and QuantumPatch model Y will be made available as open source code. The complete EXTMOS package will be covered by a restrictive licence enabling commercialisation by the firm Nanomatch to pay for staff to support and update the model after Extmos has finished.

Each model used in a simulation is documented in three chapters below:

1. Aspect of the User Case/ System simulated with this model
2. Model
3. Computation

Between two simulations the post/pre-processing shown as 4 takes place. This processes the output of one simulation into input for the next simulation.

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[1] ROMM with revisions proposed 14.6.16: Proposal for text in RoMM: Ch 1.4.2 Fermi Golden Rule (FGR) for quasi particle transport (conduction) 1.4 Statistical charge transport model, 1.4.1 Semi-classical drift-diffusion model
1.4.2 Hopping models based on Fermi's Golden Rule for quasi particle transport 1.4.3 Percolation models

Workflow for models needed for the EXTMOS package
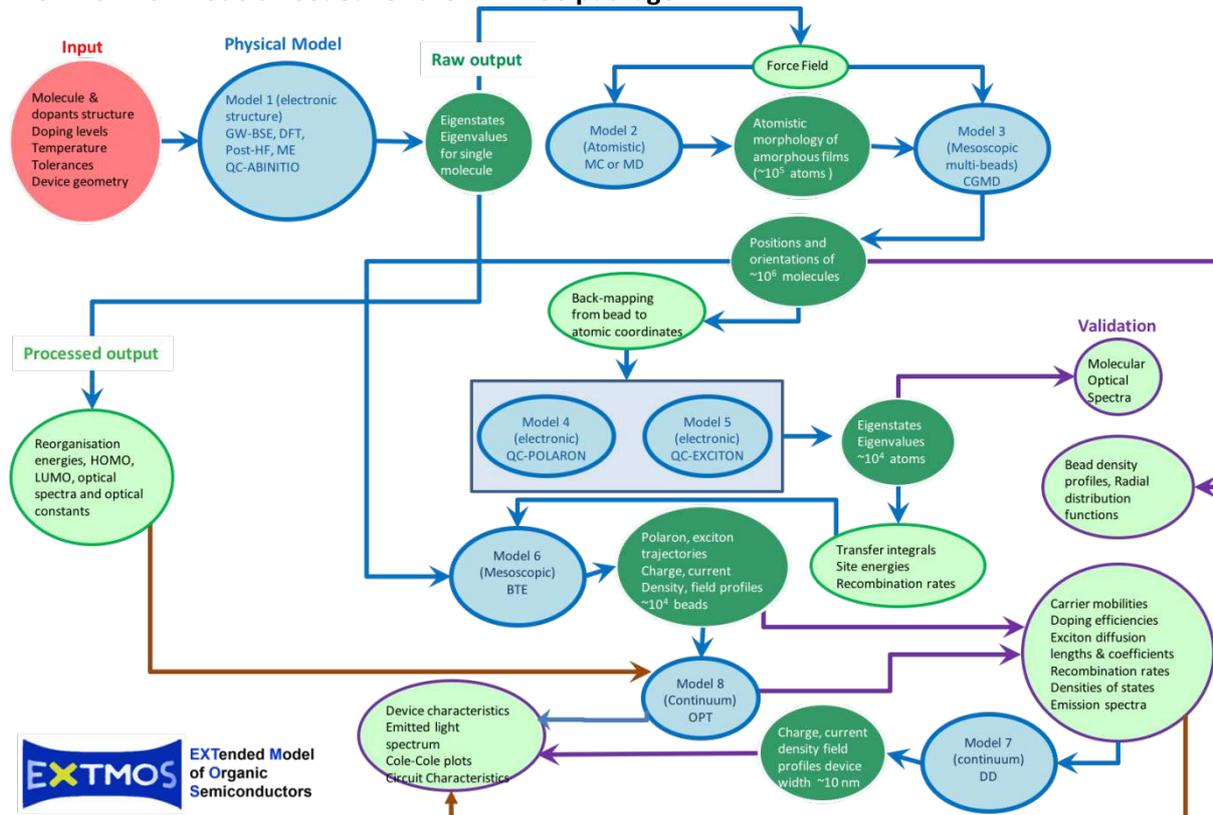
The Extmos package consists of 8 linked models that predict the behaviour of doped and pristine organic semiconductors and devices. These models examine materials phenomena at different length scales, with an emphasis on the mesoscale. This workflow shows the links between these models

Elements in the simulation with one single materials model in the chain

1	ASPECT OF THE USER CASE/SYSTEM TO BE SIMULATED: QC-ABINITIO	
1.1	ASPECT OF THE USER CASE TO BE SIMULATED	Quantitative understanding of the doping mechanism and of the excited states responsible for light emission in doped organic semiconductors/Validation of lower-level techniques (e.g. DFT)
1.2	MATERIAL	Validation system: F4TCNQ doped pentacene atomic geometries partitioned into a subsystem (a few hundred atoms) treated at the GW/BSE level and a surrounding subsystem (a few thousand atoms with their atomic polarizability) treated at the microelectrostatic level
1.3	GEOMETRY	Embedded cluster (a dopant surrounded by nearest-neighbour shell + microelectrostatic embedding)
1.4	TIME LAPSE	
1.5	MANUFACTURING PROCESS	
1.6	PUBLICATION	I. Duchemin, D. Jacquemin, X. Blase, J. Chem. Phys. (2016), **144**, 164106.

2	GENERIC PHYSICS OF THE MODEL EQUATION: QC-ABINITIO		
2.0	MODEL TYPE AND NAME	Higher level ab-initio models (model 1.1.2 in ROMM)	
2.1	MODEL ENTITY	Polarons and excitons: quasiparticles in the form of electrons or holes or electron-hole pairs coupled to lattice modes	
2.2	MODEL PHYSICS/ CHEMISTRY EQUATION PE1	Equation	Time-independent Schrödinger Equation (TISE): $H\psi = E\psi$ where ψ is either an electronic (GW) or electron-hole (excitonic, BSE) state.
		Physical quantities	Wavefunctions, energies and Hamiltonian
2.3	MATERIALS RELATIONS MR1	MR Equations	GW correction to DFT Kohn-Sham electronic energy levels*:* $$\varepsilon_n^{QP} = \varepsilon_n^{DFT} + <\phi_n^{DFT}|\Sigma^{GW}(\mathbf{r},\mathbf{r}';\varepsilon_n^{QP}) - V_{XC}^{DFT}|\phi_n^{DFT}$$ Bethe-Salpeter Hamiltonian (with electron-hole interaction) $$H_{ij,kl}^{diag} = \delta_{i,k}\delta_{j,l}\left(\varepsilon_i^{QP} - \varepsilon_j^{QP}\right),$$ $$H_{ij,kl}^{direct} = -\left\langle\varphi_i^e\varphi_j^h\left|W(r,r')\right|\varphi_k^e\varphi_l^h\right\rangle,$$ $$H_{ij,kl}^{exchange} = 2\left\langle\varphi_i^e\varphi_j^h\left|V^C(r,r')\right|\varphi_k^h\varphi_l^e\right\rangle,$$
		Physical quantities or descriptors for each MR	GW: the DFT electron-electron interaction potential (V_{XC}) is replaced by the higher-level self-energy Σ=iGW to produce accurate electronic energy levels; BSE: the screened (W; 2nd term) and exchange (V^C;3rd term) electron-hole interactions are added to the GW electron and hole energies (1st term) to generate excitonic energies and wavefunctions.
2.4	SIMULATED INPUT		

3	COMPUTATIONAL MODELLING METADATA QC-ABINITIO	
3.1	NUMERICAL SOLVER	Gaussian-basis implementation of the GW and Bethe-Salpeter formalisms with microelectrostatic (ME) semi-empirical embedding.
3.2	SOFTWARE TOOL	FIESTA combined with MESCAL for microelectrostatic embedding
3.3	GRID SIZE	Atomic-like Gaussian basis (typically 30 orbitals/atom).

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3.4	TIME STEP		
3.5	**COMPUTATIONAL REPRESENTATION** *refers to how your computational solver represents the material, properties, equation variables,…..*	**PHYSICS EQUATION**	Electronic wavefunction expressed in an atomic-like basis => GW equation recast into a linear-algebra eigenvalue problem; the BSE hamiltonian is expressed in the electron-hole wavefunction-product basis => excitonic eigenstates expressed in terms of weighted transitions between occupied and unoccupied electronic energy levels.
		MATERIAL RELATIONS	The eigenvalues of the GW problem provides the energy levels and wavefunction for the electron and holes in the system; the Bethe-Salpeter excitonic Hamiltonian provides the energies, oscillator strengths and electron-hole wavefunctions associated with optical transitions, providing information on the electron and hole localization and the nature of the excitation (localized/charge-transfer, associated dipole, bright/dark, etc.)
		MATERIAL	Atom-scale model comprising a number of atomic nuclei with defined relative positions both for the QM subsystem treated at the GW/BSE level and the environment treated at the ME level.

4	**POST PROCESSING** QC-ABINITIO	
4.1	**THE PROCESSED OUTPUT IS CALCULATED FOR**	Parameters (q, σ) and optimized starting atom coordinates r for the force fields for model 2 and 3
4.2	**METHODOLOGIES**	Force field parameters associated with lower-level methods (exchange-correlation functional in DFT, onsite/hopping parameters in tight-binding models, etc.) will be adjusted so as to reproduce the GW/BSE interatomic potentials (force fields) on representative systems (e.g. F4TCNQ-doped pentacene, etc.)
4.3	**MARGIN OF ERROR**	0.1-0.2 eV for force fields

1	**ASPECT OF THE USER CASE/SYSTEM TO BE SIMULATED: MOLECULAR DYNAMICS SOLVED BY MC**	
1.1	**ASPECT OF THE USER CASE TO BE SIMULATED**	Simulation of thin-film morphologies of small organic molecules. A user wants to have an atomic representation of an organic thin-film, which was either solution processed or processed using physical vapour deposition.
1.2	**MATERIAL**	Organic small molecules (<200 atoms), specifically HTL, Hole transport layer host Alpha-NPD N, N'-di- (alpha-naphthyl) -N, N'-diphenyl-1,1'-biphenyl- 4,4'-diamine, p dopant F6TCNNQ 2,2'-(perfluoronaphthalene-2,6-diylidene) dimalononitrile. HBL ETL, Electron transport layer host TPPhen 2,4,7,9-tetraphenyl-1,10-phenanthroline, n-dopant Tetrakis (1,3,4,6,7,8-Hexahydro-2H-pyrimido[1,2-a]pyrimidinato)–di-tungsten(II). EL, Emissive layerB Alq_3 doped with NPB:$Ir(MDQ)_2$(acac) where NPB is N,N'-di(naphthalen-2-yl)-N,N-diphenyl-benzidine and :$Ir(MDQ)_2$(acac) is iridium(III)bis(2-methyl-dibenzo-[f,h]quinoxaline)(acetylacetonate in an all-atom representation.
1.3	**GEOMETRY**	Thin films of size < $20\times20\times20nm^3$, xyz periodicity for MD, xy periodicity for MC
1.4	**TIME LAPSE**	none
1.5	**MANUFACTURING PROCESS OR IN-SERVICE CONDITIONS**	MD: simulation in multiple thermodynamic ensembles, such as NPT or NVT
1.6	**PUBLICATION**	

2	*Generic Physics Of The Model Equation* MD SOLVED BY MC		
2.0	**Model type and name**	MOLECULAR DYNAMICS (molecular mechanics model 2.2 in ROMM)	
2.1	**Model entity**	Atoms	
2.2	**Model Physics/ Chemistry equation PE**	*Equation*	Newton's equations of motion. Detailed Balance
		Physical quantities	atom coordinates, atom velocities.

2.3	Materials relation MR to PE	*Equation*	Parameterized force fields and energies, such as the OPLS and GAFF force fields, general vacuum electrostatics (Coulomb equation), VdW interactions and Pauli repulsion (Lennard Jones equations) $F(r,q,\sigma)$, $E(r,q,\sigma)$
		physical quantities/ descriptors	F is the force acting on the atoms. E is the energy of the system r are the coordinates of all atoms q are all charges of all atoms σ are the Lennard-Jones parameters (radii, interactions strength) of all atoms
2.4	Simulated Input		Input parameters (q, σ) and optimized starting atom coordinates r for the force fields can be obtained via ab-initio models (models 1.1)
2.5	Publication		
3	**Computational Modelling Metadata MD**		
3.1	Numerical Solver		Metropolis Monte Carlo Verlet Integrator
3.2	Software tool		GROMACS, LAMMPS DEPOSIT, available at Nanomatch (www.nanomatch.com)
3.3	grid size		Optional, 0.1nm
3.4	Time step		MD: 1 fs, total simulation time 40-60 ns MC: N/A
3.5	Computational Representation	Physics Equation	Newton's equations are discretized, e.g. in a Verlet scheme. VdW and Electrostatic interaction can be discretized in a uniform grid format.
		Material Relations	MD: Integration of the timestep using an integrator provides the coordinates of the next timestep based on previous coordinates. MC: A random displacement of the coordinates provides the coordinates at the next step, if accepted according to the detailed balance criterion.
		Material	Atomic coordinates and parameters, energies, forces are stored as floating point values. Both single and double precision are supported.
		COMPUTATIONAL BOUNDARY CONDITIONS	optionally periodic boundary conditions. The atom positions are concatenated and optionally periodically extended.
3.6	Publication		

4	**POST PROCESSING MC/MD**		
4.1	THE PROCESSED OUTPUT IS CALCULATED FOR		The raw output is positions of atoms in a finite volume (atomistic morphology), and is mapped to beads in model 3, CGMD. Back-mapping from bead to atomic coordinates for model 4 is used in which a correspondence between models 3 and 4; i.e. identifying which atoms make up which bead
4.2	METHODOLOGIES		The radial distribution function yields the distribution of centre of mass pair distances between molecules. The back-mapping procedure is addressed in the CGMD description
4.3	MARGIN OF ERROR		Atomic densities typically have <20% margin of error

1	**ASPECT OF THE USER CASE/SYSTEM TO BE SIMULATED COARSE GRAINED MOLECULAR DYNAMICS, CGMD**	
1.1	**ASPECT OF THE USER CASE TO BE SIMULATED**	Coarse Grained Molecular Dynamics, CGMD, finds packing arrangements of entities which are ellipsoids representing groups of atoms
1.2	**MATERIAL**	HTL, Hole transport layer host Alpha-NPD N, N'-di- (alpha-naphthyl) -N, N'-diphenyl-1,1'-biphenyl- 4,4'-diamine, p dopant F6TCNNQ 2,2'-(perfluoronaphthalene-2,6-diylidene) dimalononitrile. HBL ETL, Electron transport layer host TPPhen 2,4,7,9-tetraphenyl-1,10-phenanthroline, n-dopant Tetrakis (1,3,4,6,7,8-Hexahydro-2H-pyrimido[1,2-a]pyrimidinato)–di-tungsten(II). EL, Emissive layerB Alq_3 doped with NPB:Ir(MDQ)$_2$(acac) where NPB is N,N'-di(naphthalen-2-yl)-N,N-diphenyl-benzidine and :Ir(MDQ)$_2$(acac) is iridium(III)bis(2-methyl-dibenzo-[f,h]quinoxaline)(acetylacetonate) The organic molecules are represented by one or more entities referred to as beads. A bead represents a group of atoms, typically a functional organic group. The size of the group depends on the extent of coarse graining of the simulation, that in turn depends on the system size. A bead is often chosen to represent a whole molecule in the case of small and/or rigid molecules.
1.3	**GEOMETRY**	Vertical device consisting of cathode: Ag, 100 nm, ETL: 40-265 nm,
1.4	**TIME LAPSE**	0.9 ns/day for 5.25 million beads (without charges) on 256 cores
1.5	**MANUFACTURING PROCESS OR IN-SERVICE CONDITIONS**	
1.6	**PUBLICATION**	

2	**Generic Physics Of The Model Equation** CGMD		
2.0	**Model type and name**	COARSE GRAINED MOLECULAR DYNAMICS, CGMD (mesoscopic particle model 3.2 in ROMM)	
2.1	**Model entity**	Coarse grained (CG) representation of grouped atoms called beads (each bead corresponds to a group of atoms, typically a functional organic group)	
2.2	**Model Physics/ Chemistry equation PE**	*Equation*	Newton's equations of motion.
		Physical quantities	Positions, orientations, and velocities of CG-entity
2.3	**Materials relation MR to PE**	*Equation*	Force fields appropriate to beads $$U(\boldsymbol{u}_i, \boldsymbol{u}_j, \boldsymbol{r}) = 4\varepsilon_0 \varepsilon_{\mu\nu}(\boldsymbol{u}_i, \boldsymbol{u}_j, \boldsymbol{r}) \left[\left(\frac{\sigma_c}{r - \sigma(\boldsymbol{u}_i, \boldsymbol{u}_j, \boldsymbol{r}) + \sigma_c} \right)^{12} - \left(\frac{\sigma_c}{r - \sigma(\boldsymbol{u}_i, \boldsymbol{u}_j, \boldsymbol{r}) + \sigma_c} \right)^{6} \right]$$ Here U is the biaxial Gay-Berne (GB) potential, parameters $\sigma(\boldsymbol{u}_i \boldsymbol{u}_j \boldsymbol{r})$ and $\varepsilon_{\mu\nu}(\boldsymbol{u}_i, \boldsymbol{u}_j, \boldsymbol{r})$ are the contact distances and well depths, and ε_0, σ_c scaling energies and lengths.
		physical quantities/ descriptors	U is the Gay-Berne potential of a given pair of CG-entities. σ is the effective radius, S is contact distance, ε is interaction energy, r is the distance between the CG-entity (bead) centres of mass. The parameters of the GB potential are fitted to reproduce energy profiles of interaction for the corresponding full-atom model. Additional terms are added to U to allow for electrostatic contributions.
2.4	**Simulated Input**		
2.5	**Publication**		

3	Computational Modelling Metadata CGMD			
3.1	**Numerical Solver**	Velocity Verlet, predictor-corrector Verlet algorithm or predictor-corrector equations combined with appropriate thermostat and barostat to control temperature and pressure respectively.		
3.2	**Software tool**	SIMONA, GBMAP, LAMMPS, DL-POLY SIMONA and GBMAP convert between atomistic and coarse grained, CG, representations LAMMPS and DL-POLY solve the equations of motion in the CG representation.		
3.3	**grid size**	0.1 nm		
3.4	**Time step**	20 fs, total simulation time 40-60 ns		
3.5	**Computational Representation**	**Physics Equation**	To be determined	
		Material Relations		
		Material		
3.6	**Publication**			

	POST PROCESSING CGMD	
4.1	**THE PROCESSED OUTPUT IS CALCULATED FOR**	Molecule coordinates based on the back mapped output for beads. The internal energy, number density profiles and radial distribution function for the modelled material. Molecular positions are required by the QC-POLARON and QC-EXCITON

		models, whereas bead coordinates are required for the BTE model. These quantities will be used to validate the model. CG-entity positions (raw output) are processed into atom positions for the BTE model 6 and into electron positions required by the DFT-SE QC-POLARON and QC EXCITON model 4 and 5 . For the latter, the morphology determines the sites on which electrons, holes, excitons are localised between hops.
4.2	METHODOLOGIES	Averaged ellipsoid trajectories over around 1000 timesteps once equilibrium achieved identified from total energy vs time are required for the BTE model.
4.3	MARGIN OF ERROR	Typically 1%, depends on simulated cell size, accuracy of force fields, simulation time.

1	**Aspect of the User Case/System to be Simulated: QC-POLARON**	
1.1	**Aspect of the User Case to be simulated**	Electronic and atomistic structure of films of ~10^4 molecules of doped and pristine organic semiconductors.
1.2	**Material**	HTL, Hole transport layer host α-NPD N, N'-di- (alpha-naphthyl) -N, N'-diphenyl-1,1'-biphenyl- 4,4'-diamine, p dopant F6TCNNQ 2,2'-(perfluoronaphthalene-2,6-diylidene) dimalononitrile. HBL ETL, Electron transport layer host TPPhen 2,4,7,9-tetraphenyl-1,10-phenanthroline, n-dopant Tetrakis (1,3,4,6,7,8-Hexahydro-2H-pyrimido[1,2-a]pyrimidinato)–di-tungsten(II). EL, Emissive layerB Alq_3 doped with NPB:Ir(MDQ)$_2$(acac) where NPB is N,N'-di(naphthalen-2-yl)-N,N-diphenyl-benzidine and :Ir(MDQ)$_2$(acac) is iridium(III)bis(2-methyl-dibenzo-[f,h]quinoxaline)(acetylacetonate) described by atomic positions, grid spacing, radius around each atom, exchange-correlation functional, net charge
1.3	Geometry	Vertical device consisting of cathode: Ag, 100 nm, ETL: 40-265 nm,
1.4	Time Lapse	N/A
1.5	**Manufacturing process or in-service conditions**	
1.6	**Publication**	

2	*Generic Physics Of The Model Equation* QC-POLARON		
2.0	**Model type and name**		Electronic model (effective Hamiltonian) (model 1.2 in ROMM) combined with polarisable continuum model (model 1.2.6 in ROMM)
2.1	**Model entity**		Polarons: quasiparticles in the form of electrons or holes coupled to lattice modes
2.2	**Model Physics/ Chemistry equation PE**	*Equation*	Schrödinger Equation (SE) for electron states, Kohn-Sham equations. $$H\Psi_i = \left[-\frac{\hbar^2}{2m_e}\nabla^2 + V_{KS}(r) \right]\Psi_i = E_i\Psi_i ,$$ $$V_{KS}(r) = \sum_{i=1}^{N_{atoms}} V_{ps}(r - R_i) + \int \frac{e^2 n_s(r')}{|r-r'|}\,dr' + V_{XC}[n_s(r)]$$
		Physical quantities	Ψ_i are the Kohn-Sham electronic orbitals, H is the mean-field Kohn-Sham Hamiltonian, E_I the Kohn-Sham eigenvalues, V_{KS} the effective Kohn-Sham one-electron potential, $V_{ps}(r - R_i)$ the pseudopotential energy from the external field due to the nuclei located at R_i and corresponding inner electrons .
2.3	**Materials relation MR to PE**	*Equation*	Kohn-Sham formulation of the Hamiltonian using all-electron (Turbomole, Gaussian) or pseudopotential (BigDFT)
		physical quantities/ descriptors	Exchange-correlation functional, pseudopotentials
2.4	**Input**		Positions of atomic and electronic sites from the back mapped CGMD model

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		Orbitals and their energies from GW-BSE model 1 that are linked to charge transfer states close to dopants and to excited states.
2.5	Publication	

3	Computational Modelling Metadata QC-POLARON		
3.1	Numerical Solver	BigDFT calculates electronic structure in systems of more than 10,000 atoms going beyond DFT. It uses Constrained DFT methods. localized support functions expressed in an underlying Daubechies wavelet basis which offers ideal properties for accurate linear scaling calculations. Atom positions are determined by conjugate gradients or molecular dynamics implemented self consistently within the code. QuantumPatch computes environmental effects on polarons and excitons in systems of more than 1000 atoms. It partitions the total charge density into charge densities of single molecules. For each molecule, the charge densities of the other molecules are replaced by a set of point charges. Intermolecular electrostatic interactions are allowed for by partial charges. Initial partial charges use vacuum partial charges for each molecule, and then solve the SE for each molecule. Partial charges are updated from the new electron densities for a molecule with Kohn–Sham orbitals. MESCal coupled with BigDFT finds the self-consistent polarization state of a system of atoms. It implements a polarizable points description of electronic polarization to compute the polarization energy of a localized charge carrier and dielectric tensor. Constrained DFT, where a Lagrange multiplier term is added to the Kohn Sham energy functional, is used to evaluate on-site energies and charge-transfer integrals. The electron density is self consistently calculated for clusters of nearest neighbours and then on-site energies and charge-transfer integrals obtained.	
3.2	Software Tools	BigDFT, QuantumPatch, MESCal and Deposit are being used to create MESO-EL	
3.3	Grid Size, Time Step	0.05 nm, 4 fs per step for molecular dynamics simulations	
3.4	Computational Representation Refers To Your Computational Solver	*Physics Equation*	As for QC-ABINITIO
		Material Relations	As for QC-ABINITIO
		Material	As for QC-ABINITIO

4	POST PROCESSING QC-POLARON	
4.1	THE PROCESSED OUTPUT IS CALCULATED FOR	In principle, any experimentally observable quantity, e.g. optical spectra and optical constants, may be computed as a function of the energies. Charge transfer rates and recombination rates are provided to the BTE model. Force fields are provided for the back mapped CGMD model.
4.2	METHODOLOGIES	Charge transfer rates are determined by Marcus theory and require charge transfer and overlap integrals obtained from the orbitals using time dependent perturbation theory. HOMO and LUMO offsets at interfaces are deduced in the same way as for the GW-BSE model.
4.3	MARGIN OF ERROR	Bond lengths 10^{-4} nm, on-site energies 10 meV, transfer integrals 10 meV

1	ASPECT OF THE USER CASE/SYSTEM TO BE SIMULATED: QC-EXCITON	
1.1	ASPECT OF THE USER CASE TO BE SIMULATED	Quantitative understanding of the doping mechanism and of the excited states responsible for light emission in doped organic semiconductors.
1.2	MATERIAL	HTL, Hole transport layer host Alpha-NPD N, N'-di- (alpha-naphthyl) -N, N'-diphenyl-1,1'-biphenyl- 4,4'-diamine, p dopant F6TCNNQ 2,2'-(perfluoronaphthalene-2,6-diylidene) dimalononitrile. HBL ETL, Electron transport layer host TPPhen 2,4,7,9-tetraphenyl-1,10-phenanthroline, n-dopant Tetrakis (1,3,4,6,7,8-Hexahydro-2H-pyrimido[1,2-

		a]pyrimidinato)–di-tungsten(II). EL, Emissive layerB Alq$_3$ doped with NPB:Ir(MDQ)$_2$(acac) where NPB is N,N'-di(naphthalen-2-yl)-N,N-diphenyl-benzidine and :Ir(MDQ)$_2$(acac) is iridium(III)bis(2-methyl-dibenzo-[f,h]quinoxaline)(acetylacetonate)
1.3	**GEOMETRY**	Vertical device consisting of cathode: Ag, 100 nm, ETL: 40-265 nm,
1.4	**TIME LAPSE**	N/A
1.5	**MANUFACTURING PROCESS**	
1.6	**PUBLICATION**	

2	*Generic Physics Of The Model Equation* **QC-EXCITON**	
2.0	**Model type and name**	Electronic model (effective Hamiltonian) (model 1.2 in ROMM)
2.1	**Model entity**	Excitons: quasiparticles in the form of electron-hole pairs coupled to lattice modes
2.2	**Model Physics/ Chemistry equation PE**	*Equation*	Schrödinger Equation (SE) for exciton states. $$H|n> = \left[\hbar\omega_{0-0} + \hbar D + \sum_{n=1}^{N} J_0(|n><n+1| + |n+1><n|) + \sum_{n=1}^{N} \Lambda_n(|n><n|) \right] |n>$$
		Physical quantities	|n> represents a pure (S$_1$) electronic excitation at site n with all other molecules electronically unexcited in state S$_0$, *H* is the Hamiltonian, *J$_0$* the nearest neighbour coupling, $\hbar\omega_{0-0}$ is the energy corresponding to the S$_0 \to$S$_1$ transition, typically dipole allowed in the UV-Vis spectral region, *D* is a non-resonant gas-to-crystal (red) shift which is due to the greater stabilization of S$_1$ compared to S$_0$ upon aggregation and *Λ_n* is the transition energy offset for the *n*th chromophore.
2.3	**Materials relation MR to PE**	*Equation*	$$H = \sum_{m=1}^{N} H_m + \sum_{m=1}^{N}\sum_{n<m}^{N} \hat{V}_{mn} + \hbar\omega_{vib}\sum_{n<m}^{N} b_n^{\dagger}b_n + \hbar\lambda\omega_{vib}\sum_{n} |n><n|(b_n^{\dagger}+b_n) + \hbar\lambda^2\omega_{vib}$$
		physical quantities/ descriptors	H_m describes the intramolecular Hamiltonian for site *m* and $\hat{V}_{mn}$ is an operator representing the intermolecular interactions between sites *m* and *n*. The third term represents the vibrational energy, with $b_n^{\dagger}$ (b_n) creating (annihilating) a vibrational quantum on the *n*th chromophore. The local, linear exciton-vibrational coupling term is represented by the third term.
2.4	**Input**		Orbitals and their energies from GW-BSE that are linked to excitons. The positions of the atoms and quasi-particle sites are deduced from/based on the centres of mass of the ellipsoids obtained from the CGMD model.
2.5	**Publication**		

3. COMPUTATIONAL MODELLING METADATA QC-EXCITON		
NUMERICAL SOLVER	Gaussian-basis implementation with microelectrostatic (ME) semi-empirical embedding	
SOFTWARE TOOL	FIESTA combined with MESCAL for microelectrostatic embedding	
GRID SIZE	N/A	
TIME STEP	N/A	
COMPUTATIONAL REPRESENTATION *refers to how your computational solver represents the material, properties, equation variables,.....*	**PHYSICS EQUATION**	$$\sum_{\{b\}} <\phi_{(a\}}|H|\,\phi_{(b\}}> = E <\phi_{(a\}}|\,\phi_{(b\}}>$$
		The exciton Hamiltonian is written in terms of the adiabatic electronic states, m,a, of the single chromophores m.
	MATERIAL RELATIONS	$$|\phi_{(n\}} \geq \frac{1}{\sqrt{N_p!}} \sum_{mn} (-1)^p P[\prod_{m=1}^{N} |\varphi_{m,n}>$$ $$\rho_{m,ge}(r_m) = \varphi_{m,e}^*(r_m)\,\varphi_{m,g}(r_m),\ \mu_m^x = \int dx\,\hat{\mu}_m\,\rho_{m,ge}(r_m)$$
		P generates a permutation over electronic coordinates of the molecules in the aggregate and N_p is the number of such permutations. The aggregate electronic states $|\psi> = \sum_{\{a\}} C(\{a\})|\phi_{\{a\}}>$ with variational coefficients C({a}) are solutions of the Schrödinger equation. The transition density ρ for ground, g, to excited, e, states is obtained from frontier molecular orbitals φ and is used to find the x,y,z components of the transition dipole moment **μ**$_m$ and the Coulomb term in the Hamiltonian.
	MATERIAL	Atom-scale model comprising a number of atomic nuclei with defined relative positions and the same number of electrons to ensure overall charge neutrality.

4	POST PROCESSING QC-EXCITON	
4.1	**THE PROCESSED OUTPUT IS CALCULATED FOR**	$$A(\omega)/N = \sum_{\alpha} f_{k=0,\alpha}\,\Gamma(\omega - \omega_{k=0,\alpha}),\ \ \overline{S}(\omega)/N = \sum_{v_l} I_{PL}^{0-v_l}\,\Gamma(\omega - \omega_{em} + v_l \omega_{vib})$$ $A(\omega)$ is the absorption spectrum required for validation, $f_{k=0,\alpha}$ the oscillator strength, α represents the multitude of eigenstates which all have a centre of mass (quasi) momentum **k** equal to zero but with varying amounts of vibrational dressing. $\Gamma(\omega)$ is a line shape function usually taken to be Gaussian for the case of inhomogeneous broadening and Lorentzian for homogeneous broadening. $\overline{S}(\omega)$ is the reduced photoluminescence, PL spectrum. Exciton transfer rates are provided for the BTE model.Transfer integrals Site energies Recombination rates for BTE model 6
4.2	**METHODOLOGIES**	Optical spectra and optical constants are obtained from excited state energies by calculating the time dependent dipole moment output by the solver.
4.3	**MARGIN OF ERROR**	On-site energies 10 meV, transfer integrals 10 meV

1	**ASPECT OF THE USER CASE/SYSTEM TO BE SIMULATED Boltzmann Transport Equation, BTE**	
1.1	ASPECT OF THE USER CASE TO BE SIMULATED	Transfer of electrons and excitons between point representations of molecules, leading to current density and electrical field profiles and colour and intensity of optical emission
1.2	MATERIAL	HTL, Hole transport layer host Alpha-NPD N, N'-di- (alpha-naphthyl) -N, N'-diphenyl-1,1'-biphenyl- 4,4'-diamine, p dopant F6TCNNQ 2,2'-(perfluoronaphthalene-2,6-diylidene) dimalononitrile. HBL ETL, Electron transport layer host TPPhen 2,4,7,9-tetraphenyl-1,10-phenanthroline, n-dopant Tetrakis (1,3,4,6,7,8-Hexahydro-2H-pyrimido[1,2-a]pyrimidinato)–di-tungsten(II). EL, Emissive layerB Alq_3 doped with NPB:Ir(MDQ)$_2$(acac) where NPB is N,N'-di(naphthalen-2-yl)-N,N-diphenyl-benzidine and :Ir(MDQ)$_2$(acac) is iridium(III)bis(2-methyl-dibenzo-[f,h]quinoxaline)(acetylacetonate) represented by electrons and excitons on ellipsoids (molecules)
1.3	GEOMETRY	Vertical device consisting of cathode: Ag, 100 nm, ETL: 40-265 nm,
1.4	TIME LAPSE	50 ps to ms
1.5	MANUFACTURING PROCESS OR IN-SERVICE CONDITIONS	
1.6	PUBLICATION	

2	*Generic Physics Of The Model Equation BTE*		
2.0	*Model type and name*	Semi-classical Boltzmann transport equation, BTE (statistical mechanics electronic model 1.4.1 in ROMM. Loosely coupled to an optical model, OPT, model 6 .	
2.1	*Model entity*	Electron polarons, hole polarons, singlet and triplet excitons	
2.2	*Model Physics/ Chemistry equation PE*	*Equation*	Boltzmann transport equation Waiting time before a reaction with reaction rate k for a uniformly distributed random number R between 0 and 1 $$\tau = -\frac{\ln(R)}{k}$$
		Physical quantities	Charge, exciton, current density and field profiles
2.3	*MR*	Hopping rates and generation and recombination rates that are taken from DFT-SE Energy barrier between neighbouring sites	
2.4	*Simulated Input*	Exciton and charge transfer rates and recombination rates from DFT-SE. The positions of the sites on which electrons, holes, excitons are localised between hops from CGMD.	
2.4	*Publication*		

3	Computational Modelling Metadata BTE		
3.1	**Numerical Solver**	Kinetic Monte Carlo, KMC	
3.2	**Software tool**	Merged Bath KMC and KIT KMC codes	
3.3	**grid size**	N/A	
3.4	**Time step**	ps to ms	
3.5	**Computational Representation** refers to how your computational solver	**Physics Equation**	
		Material Rela-	

	represents the material, prop-erties, equation variables,.....	**tions**	
		Material	Each site has a label determining which material it is and which particle species occupies it that varies with elapsed time
3.6	**Publication**		

4	**POST PROCESSING BTE**	
4.1	**THE PROCESSED OUT-PUT IS CALCULATED FOR**	Material/layer characteristic scalars/parameters for continuum models: Carrier mobilities, doping efficiencies, recombination rates , exciton diffusion lengths and coefficients. Finite volume resolved current densities, emitted light intensity and spectrum.
4.2	**METHODOLOGIES**	Current densities found from particle fluxes, charge mobilities from net drift ve-locity d of ensemble of electron trajectories deduced from net distance travelled over a fixed time period at a given external field. The optical model is needed to find the emitted light intensity and spectrum.
4.3	**MARGIN OF ERROR**	Typically 1% for all quantities, depending on number of trajectories calculated

1	**ASPECT OF THE USER CASE/SYSTEM TO BE SIMULATED DD**	
1.1	**ASPECT OF THE USER CASE TO BE SIMU-LATED**	Device characteristics for doped organic semiconductor organic light emitting devices
1.2	**MATERIAL**	HTL, Hole transport layer host Alpha-NPD N, N'-di- (alpha-naphthyl) -N, N'-diphenyl-1,1'-biphenyl- 4,4'-diamine, p dopant F6TCNNQ 2,2'-(perfluoronaphthalene-2,6-diylidene) dimalononitrile. HBL ETL, Electron transport layer host TPPhen 2,4,7,9-tetraphenyl-1,10-phenanthroline, n-dopant Tetrakis (1,3,4,6,7,8-Hexahydro-2H-pyrimido[1,2-a]pyrimidinato)–di-tungsten(II). EL, Emissive layerB Alq_3 doped with NPB:$Ir(MDQ)_2$(acac) where NPB is N,N'-di(naphthalen-2-yl)-N,N-diphenyl-benzidine and :$Ir(MDQ)_2$(acac) is iridium(III)bis(2-methyl-dibenzo-[f,h]quinoxaline)(acetylacetonate) described as continuum with specific values fo part s ofteh device (describe which pls)
1.3	**GEOMETRY**	Vertical device consisting of cathode: Ag, 100 nm, ETL: 40-265 nm, described as continuum with specific values for the parts of the device nl......
1.4	**TIME LAPSE**	Not applicable
1.5	**MANUFACTURING process or in-service conditions**	Voltage bias
1.6	**PUBLICATION**	

2	**GENERIC PHYSICS OF THE MODEL EQUATION DD**		
2.0	**MODEL TYPE AND NAME**	Semi-classical Drift-diffusion model (1.4.1 in RoMM)	
2.1	**MODEL ENTITY**	This is a materials model for concentration of electrons or excitons and the equation is written up for finite volumes (the model entity) corresponding to the active layers in the device	
2.2.a	**MODEL PHYSICS/ CHEMIS-**	**Equation**	**Current equations** for electron current J_n and hole current J_p $$J_n = qn(\boldsymbol{r})\mu_n\boldsymbol{E} + qD_n\boldsymbol{\nabla}n$$ $$J_p = qn(\boldsymbol{r})\mu_p\boldsymbol{E} + qD_p\boldsymbol{\nabla}p$$

	TRY EQUATION PE1	**Physical quantities**	q Electron charge $J_{n(p)}$ Electron (hole) current density $\mu_{n(p)}$ Electron (hole) mobility $\mathbf{E}$ Electric field, given by $\mathbf{E} = -\boldsymbol{\nabla}V$ where V is potential $D_{n(p)}$ Electron (hole) diffusion coefficient
2.3.a	**MATERIALS RELATION 1 MR1 TO PE1**	**Equation**	**Einstein relation** $$\mu_{n(p)} = \frac{qD_{n(p)}}{k_B T}$$
		Physical quantities/ descriptors	Einstein relation used to relate values of mobility $\mu_{n(p)}$ and diffusion coefficient $D_{n(p)}$ to each other; T = temperature
	MATERIALS RELATION 3 MR3 TO PE1	**Equation**	Relation between carrier densities n, p and potential V
		Physical quantities/ descriptors	*Range of different approaches used; all include a dependence on intrinsic carrier density, n_i, potential V and electron (hole) quasi-Fermi energies, $\phi_{n(p}$*
2.2.b	**MODEL PHYSICS EQUATION PE 2**	**Equation**	**Poisson equation**, describing how the potential varies with position $$\nabla.\varepsilon\nabla V = -q(p - n + N_D - N_A)$$
		Physical quantities	ε Dielectric constant N_D Donor doping density N_A Acceptor doping density
2.3.b	**MATERIALS RELATION 1 MR1 TO PE2**	**Equation**	Relation between carrier densities n, p and potential V
		Physical quantities/ descriptors	Dependence on intrinsic carrier density, n_i, potential V and electron (hole) quasi-Fermi energies, $\phi_{n(p)}$, from Boltzmann approximation $$n = n_i \exp\left(\frac{q(V - \phi_n)}{k_B T}\right)$$
2.2 c	**MODEL PHYSICS EQUATION PE 3**	**Equation**	**Continuity equations**, to ensure conservation of charge: $$\frac{\partial n}{\partial t} = \frac{1}{q}\boldsymbol{\nabla}.J_n - rnp, \frac{\partial p}{\partial t} = -\frac{1}{q}\boldsymbol{\nabla}.J_p - rnp$$
		Physical quantities	r is the electron-hole recombination rate
	EQUATION PE 4		$$\frac{\partial S}{\partial t} = D_S\nabla^2 S + \eta_{st}rnp - \frac{Q}{h\nu} - \frac{S}{\tau_S}$$
		Physical quantities	S is exciton concentration η_{st} singlet-triplet ratio Q is the generated photon flux E_{opt} is field arising from photons α is absorption coefficient
2.4	**INPUT**		From Model 1 GW-BSE Interface energy offsets, Optical constants From Model 4 BTE: Carrier mobilities, doping efficiencies, Exciton diffusion lengths and coefficients, Recombination rates, Densities of states
2.5	**PUBLICATION**		

3	**COMPUTATIONAL MODELLING METADATA DD**	
3.1	**NUMERICAL SOLVER**	Finite element model
3.2	**SOFTWARE TOOL**	SIlvaco Atlas code
3.3	**GRID SIZE**	nm
3.4	**TIME STEP**	N/A

3.5	COMPUTATIONAL REPRESENTATION	PHYSICS EQUATION	Finite element discretisation of equations given in part 2 where variable e.g. charge density defined for each finite element.
		MATERIAL RELATIONS	Finite element discretisation of relations given in part 2 where equation defined for each finite element and its neighbours.
		MATERIAL	Material determines which input parameter set is selected
3.6	PUBLICATION		http://www.silvaco.com/examples/tcad/section38/example1/index.html

4	POST PROCESSING DD	
4.1	THE PROCESSED OUTPUT IS CALCULATED FOR	a finite volume, representing the active layers and contacts and is the current-voltage curves. Outputs are current-voltage-Luminance characteristics, the emitted light spectrum, Cole-Cole plots giving impedance variation with frequency of a time varying external bias, circuit characteristics.
4.2	METHODOLOGIES	Run the DD model for a series of voltages to obtain current-voltage curves, or for time following a stimulus, e.g. change in applied bias in the dark for dark injection transients, equivalent circuit models provide impedance of device in a circuit. The optical model produces the luminance and emitted light spectrum.
4.3	MARGIN OF ERROR	The error is sensitive to the accuracy of the input parameters, and to the extent to which these parameters represent real devices. An error of 10% is attainable.

1	Aspect of the OPTICAL MODEL used as post processor	
1.1	Case to be simulated	Calculation of intensity variations for visible light produced by OLEDs and for absorption of incident light and of emitted photons
	Raw output to be processed of BTE	Exciton density profiles
	Raw output to be processed of DD	Exciton and electron? density profiles
1.6	Publication	

2	GENERIC PHYSICS OF THE MODEL EQUATION OPT		
2.0	MODEL TYPE AND NAME	Maxwell's equations in a microcavity (Continuum model, 4.6 in ROMM)	
2.1	MODEL ENTITY	This is a materials model for electric and magnetic field and intensity variations and the equation is for finite volumes (the model entity)	
2.2	MODEL PHYSICS/ CHEMISTRY EQUATION PE1	Equation	$\nabla . \boldsymbol{D} = \rho$, $\nabla . \boldsymbol{B} = 0$, $\nabla \times \boldsymbol{E} = -\partial \boldsymbol{B}/\partial t$, $\nabla \times \boldsymbol{H} = \partial \boldsymbol{D}/\partial t + \boldsymbol{J}$
		Physical quantities	$\boldsymbol{D}$ Displacement, $\boldsymbol{B}$ magnetic flux density, $\boldsymbol{J}$ Current density, ρ charge density $\boldsymbol{E}$ Electric field, given by $\boldsymbol{E} = -\nabla V$ where V is potential t time
2.2.1	MATERIALS RELATION 1 MR1 TO PE1	Equation	$\boldsymbol{D} = \epsilon \boldsymbol{E}$
		Physical quantities/ descriptors	ϵ dielectric constant
	MATERIALS RELATION 2 MR2 TO PE1	Equation	$\Gamma_{cav} = \Gamma_{nr} + F\Gamma_r$
		Physical quantities/ descriptors	Γ excited state decay rate, subscripts cav for cavity formed by device contacts, nr nonradiative, r radiative
	MATERIALS RELATION 3	Equation	$F = \int_0^\infty K(\kappa) d\kappa^2$

MR3 TO PE1	**Physical quantities/ descriptors**	total power F supplied by the dipole antenna (including dissipation in absorbing media and radiation into the far field)
2.3	**Input**	Optical constants from GW-BSE. Positions of excitons undergoing radiative decay from BTE or exciton recombination profile from DD. Here radiating excitons are modelled as radiating dipoles, device geometry including the contacts and their compositions, luminosity function for photopic vision

3	**COMPUTATIONAL MODELLING METADATA OPT**		
3.1	**NUMERICAL SOLVER**	Transfer matrix	
3.2	**SOFTWARE TOOL**	Code in MESO-TRANS or ATLAS	
3.3	**GRID SIZE**	nm width	
3.4	**TIME STEP**	N/A	
3.5	**COMPUTATIONAL REPRESENTATION**	**PHYSICS EQUATION**	Reflection and transmission matrices for each layer
		MATERIAL RELATIONS	As given in part 2
		MATERIAL	Material determines which complex dielectric constant and radiative and nonradiative decay rate obtained from TISE is selected
3.6	**PUBLICATION**		

4	**POST PROCESSING OPT**	
4.1	**THE PROCESSED OUTPUT IS CALCULATED FOR**	Luminance, angular dependence of emission, colour coordinates, reflection transmittance and absorbance spectra of device
4.2	**METHODOLOGIES**	Luminance obtained from emission spectrum obtained from energy of radiating exciton, out-coupling efficiency, i.e. probability of a photon generated in the device being emitted into free space calculated by averaging over finite elements
4.3	**MARGIN OF ERROR**	Typically 5%, main sources of error are from the BTE or DD models where the morphologies and defect concentrations, energies and types which are hard to establish