The European Materials Modelling Council

MODA

Modelling data documenting one simulation

STARCELL

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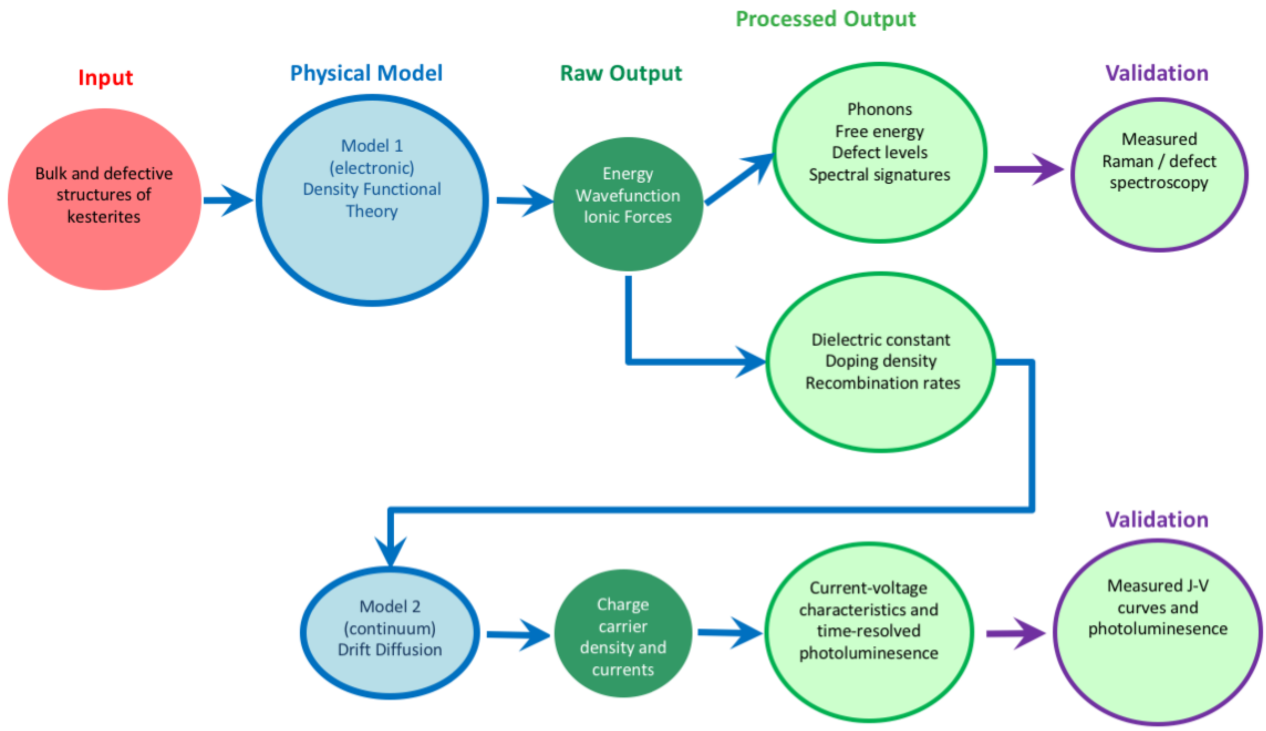
Purpose of this document:
Definition of a data organisation that is applicable to ALL materials modelling simulations. The fiche should contain all elements that are needed to describe a simulation. This information spans from the end-user (manufacturer) information to the computational modelling details.

OVERVIEW of the simulation			
1	**USER CASE**	*1. To understand the effect of point defects and phase competition on the electronic structure of kesterite-structured semiconductors for thin-film photovoltaic applications* *2. To retrieve main efficiency losses in kesterite based solar cells and to support cell engineering for improved performance.*	
2	**CHAIN OF MODELS**	**MODEL 1**	*Electronic model: density functional theory*
		MODEL 2	*Continuum model: Semiconductor electron and hole transport equations in the context of thin-film photovoltaic devices*
3	**PUBLICATION ON THIS ONE SIMULATION**	*Reproducibility in density functional theory calculations of solids, Science 351, 6280 (2016); DOI: 10.1126/science.aad3000*	
4	**ACCESS CONDITIONS**	*A range of density functional theory codes are available under either commercial and open-source licenses. For example, GPAW (https://wiki.fysik.dtu.dk/gpaw/) is available under a GPLv3+ license.* *The basic semiconductor transport equations are implemented in the three-dimensional device simulation tool Synopsys TCAD, which is available at MLU.*	
5	**WORKFLOW AND ITS RATIONALE**		



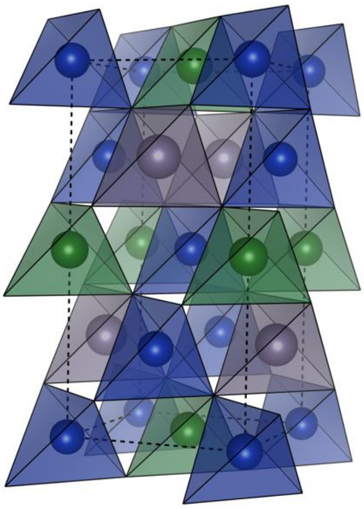
Workflow

Each model used in this simulation can be documented in four chapters:

1. Aspect of the User Case or System simulated with this model
2. Model
3. Computational aspects
4. Post processing

MODEL 1

1	ASPECT OF THE USER CASE/SYSTEM TO BE SIMULATED	
1.1	ASPECT OF THE USER CASE TO BE SIMULATED	*The total energy and electronic structure of pure and defective crystalline semiconductors*
1.2	MATERIAL	*The prototype material is* Cu_2ZnSnS_4*. Crystal structure information from X-ray diffraction data (inorganic crystal structure database).*
1.3	GEOMETRY	*A quaternary metal sulfide with a tetragonal crystal structure derived from the zinc-blende common to many III-V and II-VI semiconductors*

		
1.4	**Time Lapse**	*Static simulations (energy minimisation to a local ground-state)*
1.5	**Manufacturing process or in-service conditions**	*T = 0 and P = 0 for the density functional theory calculations, but external conditions are imposed as post-processing of thermodynamic potentials*
1.6	**Publication on this one simulation**	*Kesterite Thin-Film Solar Cells: Advances in Materials Modelling of Cu_2ZnSnS_4, Advanced Energy Materials 2, 400 (2012)* *http://onlinelibrary.wiley.com/doi/10.1002/aenm.201100630/abstract*

2	**Generic Physics Of The Model Equation**		
2.0	**Model type and name**	*Electronic model: Quantum Density Functional Theory*	
2.1	**Model entity**	*Electrons and ions*	
2.2	**Model Physics/ Chemistry equation PE**	**Equation**	*Kohn-Sham Density Functional Theory Equations*
		Physical quantities	*Boundary conditions including lattice dimensions and symmetry* *Nuclear coordinates and charges* *Number of electrons* *Bloch wave vectors*
2.3	**Materials relations**	**Relation**	*1. Exchange and correlation functional* *2. Core electron pseudopotentials*
		Physical quantities/ descriptors for each MR	*1. Exchange energy, correlation energy, electronic charge density* *2. Kinetic energy, all-electron charge density*
2.4	**Simulated input**		

3	**Solver and Computational Translation of the Specifications**		
3.1	**Numerical Solver**	*Self-consistent field procedure*	
3.2	**Software tool**	*VASP, https://www.vasp.at* *GPAW, https://wiki.fysik.dtu.dk/gpaw*	
3.3	**Time step**	*N/A*	
3.4	**Computational Representation**	**Physics Equation,**	*For a given crystal structure (cell vectors with defined internal atomic positions) the Kohn-Sham equations are solved to yield a set of one-*

	,	**Material Relations, Material**	*electron eigenvalues and a total energy for the system. The electronic wavefunctions are expanded in a plane-wave basis set, and the charge density is discretised on a grid of points. The Hellman-Feynman theorem is used to computed forces on the ions, which are minimized to yield an equilibrium structure.* *In this first-principles approach no materials specific information is required for the Hamiltonian with the exception of the atom identities and core electron pseudopotentials (where used).*
3.5	**Computational boundary conditions**	*Three-dimensional periodic boundary conditions (following Born–von Karman)*	
3.6	**Additional Solver Parameters**	• *Basis set for electronic wavefunctions (e.g. plane wave energy cut-off)* • *Wave vectors (i.e. k-point sampling density)* • *Total energy and force tolerance*	

Post processing

The "raw output" calculated by the model is per definition the physics variable in the PE(s).
This is already specified in the entry 2.2 and will appear in your dark green circle in the workflow picture.

This output is often processed by a post processor in order to calculate values for physics variables for different entities that can be input to the next model. Or the output is homogenised for larger volumes in the form of a MR or Descriptor Rule that are the final output of the total simulation.
This will appear in your light green circle in the workflow picture and also in 2.4 of the next model.

The methodology (often including new physics) used to do this calculation is to be documented.

4	**POST PROCESSING**	
4.1	**The processed output**	*Dielectric constant, Gibbs free energy and dopant density / recombination rates in finite volumes for model 2.*
4.2	**Methodologies**	*The total energy directly obtained from Density Functional Theory calculations represents the static potential. To reconstruct a realistic free energy, lattice vibrations (phonons) must be considered. A thorough overview of the techniques and applications is available in Ab Initio Thermochemistry of Solid-State Materials, Angewandte Chemie 49, 5242 (2010) http://onlinelibrary.wiley.com/doi/10.1002/anie.200906780/full* *Knowledge of the lattice vibrations is used here for (i) assessing phase stability by calculating energies for specific reactions and processes; (ii) simulating bulk spectral signatures (IR and Raman signals); (iii) calculating accurate spectral signatures of defects (transition levels of point defects including vibronic components). We have an on-going collaboration in developing the open-source (BSD license) package Phonopy http://atztogo.github.io/phonopy where these features are being implemented.* *In addition, once the Gibbs free energy of defects in accessible charge states are known the equilibrium concentrations (dopant density); defect levels (charge state transition energies) and electron-hole recombination rates can be readily obtained.*
4.3	**Margin Of Error**	*The leading error is due the approximation of electron-electron interactions in the underlying exchange-correlation functional, which are material and property dependent. We have recently shown that a reliable exchange-correlation functional such as PBEsol can reproduce phonon dispersion and thermal expansion of many semiconductors with good accuracy (within 10% of comparable measurements). See*

		Influence of the exchange-correlation functional on the quasi-harmonic lattice dynamics of II-VI semiconductors, J. Chem. Phys. 143, 064710 (2015) http://dx.doi.org/10.1063/1.4928058

MODEL 2

1	**Aspect of the User Case/System to be Simulated**	
1.1	**Aspect of the User Case to be simulated**	*The photoluminescence decay curves and the efficiency of solar cells based on polycrystalline thin-film semiconductors.*
1.2	**Material**	*The prototype material is Cu_2ZnSnS_4 with dopants and recombination centers*
1.3	**Geometry**	*A quaternary metal sulfide with a tetragonal crystal structure derived from the zinc-blende common to many III-V and II-VI semiconductors as illustrated above.*
1.4	**Time Lapse**	*Static (steady-state) simulations of current-voltage characteristics and time-resolved simulations for time-resolved photoluminescence.*
1.5	**Manufacturing process or in-service conditions**	*The temperature and light source are set according to the conditions of a working solar cell at T = 300 K and the AM1.5 solar spectrum*
1.6	**Publication on this one simulation**	

2	**Generic Physics Of The Model Equation**		
2.0	**Model type and name**	*Continuum model: Drift Diffusion model*	
2.1	**Model entity**	*Electrons, holes*	
2.2	**Model Physics/ Chemistry equation PE**	**Equation**	*Maxwell equations, continuity equation, semi-classical drift-diffusion equations*
		Physical quantities	*Dielectric constant* *Density, energy, and capture cross-sections of defects* *Charge carrier mobilities* *Doping density* *Recombination rates* *Band alignment*
2.3	**Materials relations**	**Relation**	
		Physical quantities/ descriptors for each MR	
2.4	**Simulated input**	*Bulk and surface properties from the application of models 1 (i.e. dielectric constants, doping densities, and recombination rates)*	

3	**Solver and Computational translation of the specifications**		
3.1	**Numerical Solver**	*Bank-Rose-algorithm combined with an iterative linear solver*	
3.2	**Software tool**	*Synopsys TCAD*	
3.3	**Time step**	*N/A*	
3.4	**Computational Representation** ,	**Physics Equation, Material Relations, Material**	*For a given device geometry, a mesh is generated. The Maxwell, continuity, and drift-diffusion equations are discretized using the box method and the TRBDF method. Then, the equations are solved self-consistently on each grid point for each time-step.*

3.5	**COMPUTATIONAL BOUNDARY CONDITIONS**	• *Ohmic boundary conditions at electrical contacts* • *Periodic boundary conditions at non-contact surfaces*
3.6	**ADDITIONAL SOLVER PARAMETERS**	• *Rate of generation by external excitation* • *Applied voltages at the electrical contacts*

Post processing

The "raw output" calculated by the model is per definition the physics variable in the PE(s).
This is already specified in the entry 2.2 and will appear in the third rectangle in the workflow picture.

This output is often processed by a post processor in order to calculate values for physics variables for different entities that can be input to the next model. Or the output is homogenised for larger volumes in the form of a MR or Descriptor Rule that are the final output of the total simulation.
This will appear in the fourth rectangle in the workflow picture and also in 2.4 of the next model.

The methodology (often including new physics) used to do this calculation is to be documented.

4	**POST PROCESSING**	
4.1	**THE PROCESSED OUTPUT**	*Photoluminescence intensity and electrical current.*
4.2	**METHODOLOGIES**	*From the charge carrier densities, the rate of radiative recombination is calculated at each grid point for each time-step. By spatial integration of the recombination rate, the photoluminescence intensity is calculated as a function of time.* *For the current-voltage characteristics, the applied voltage is stepwise increased. For each voltage, the electron and hole current densities at the electric contacts are summed up and integrated. This yields the electrical current as a function of the voltage*
4.3	**MARGIN OF ERROR**	*The leading error will be the optical generation and emission due the approximation of plane layers in the semiconductor stack. Because of the rough interfaces, a combination of coherent and non-coherent effects must be considered. This will affect the photoluminescence emission, generation by light, and photon recycling.*