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Computational Design of Quantum Defects in Low-dimensional Semiconductors

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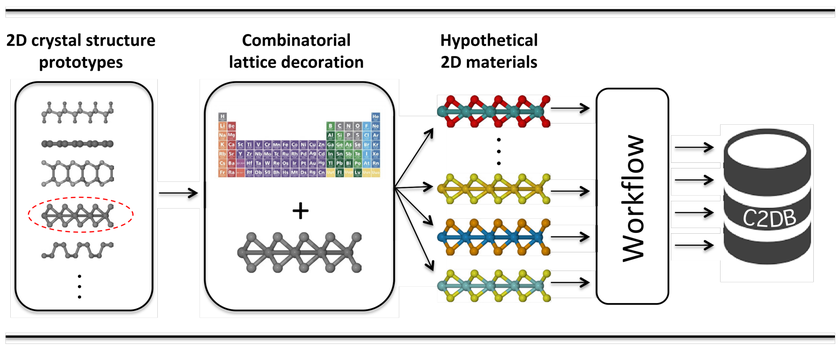
ABSTRACT

2D materials are known to host intriguing electronic properties and thus offer a fascinating platform for quantum photonics. In particular, 2D materials have been shown to host single-photon emitters (SPE). It is therefore vital to investigate the influence of defects within different host materials which are much easier to create and control in monolayers compared to bulk systems. Based on the computational 2D materials database (C2DB) [1] we first perform a computational screening for intrinsic point defects of stable theoretically predicted and experimentally known low-dimensional semiconductors. We will present a tool within the atomic simulation environment (ASE) [2] to automatically identify intrinsic point defects for given structures and calculate properties like formation energies, charge transition levels, and more to pave the way towards creating a database of intrinsic defects in 2D semiconductors.

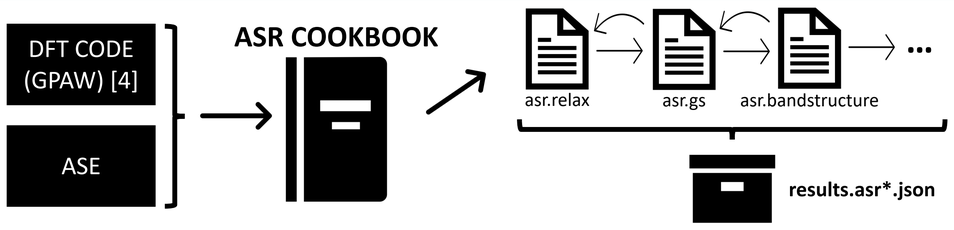
INTRODUCTION – THE COMPUTATIONAL 2D MATERIALS DATABASE (C2DB) [1]



C2DB is an open database and contains more than 4000 two-dimensional (2D) monolayer materials. Those materials include lattice decorated combinations of different 2D structure prototypes. The calculated properties for materials within C2DB range from relaxed structures, theremodynamic and dynamic stabilities, electronic and magnetic structures, optical properties and many more. C2DB is the starting point for our defect considerations.

ATOMIC SIMULATION RECIPES (ASR) [3]

- collection of standardized scripts to perform standard tasks in electronic structure calculations
- dependencies figure out which order lists of recipes need to run in
- easy collection of results in results files

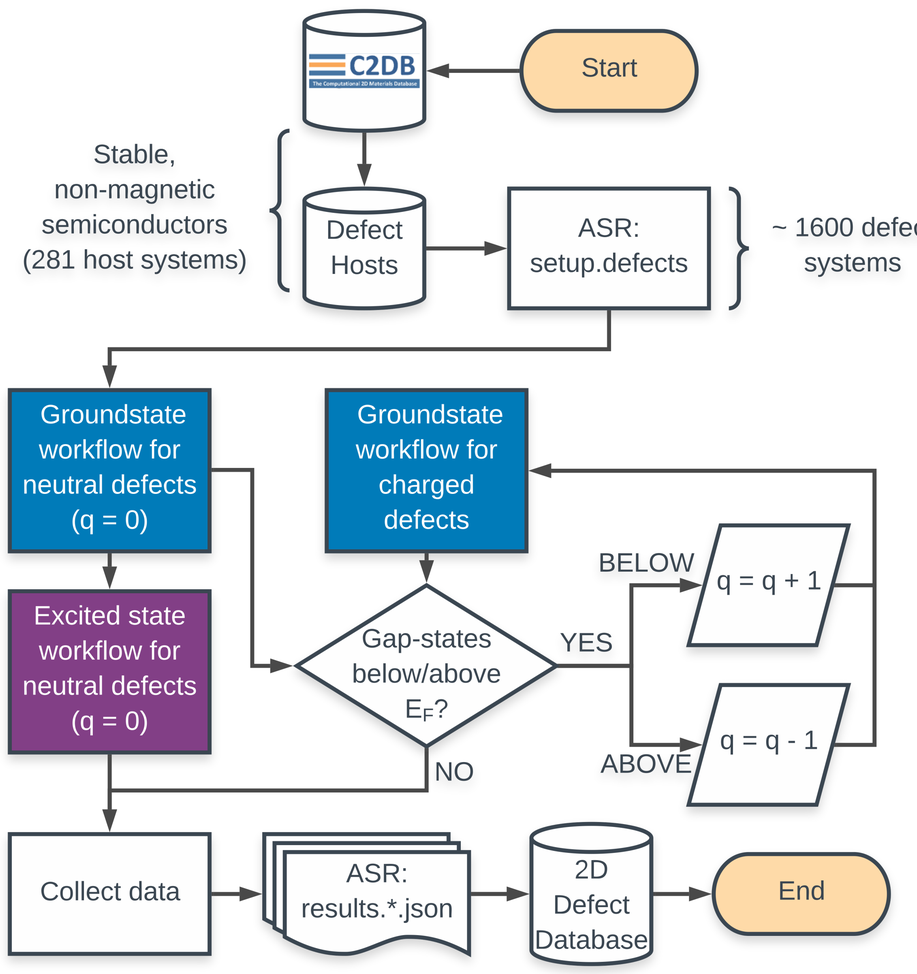


DEFECTS IN 2D SEMICONDUCTORS

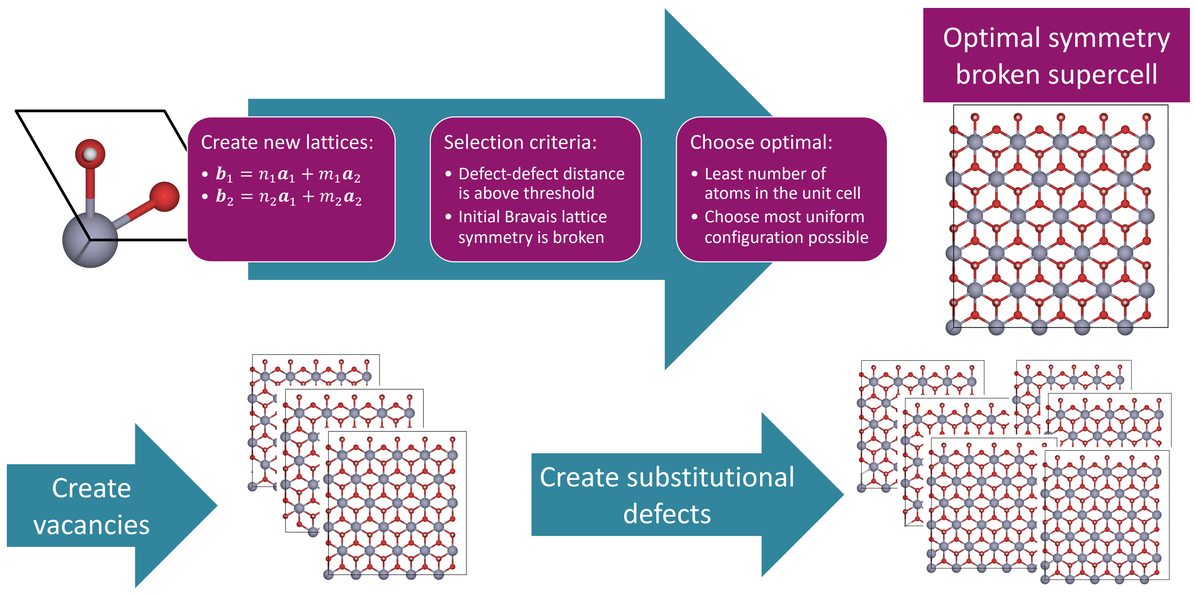
- Single-photon emitters (SPE) play central role for many quantum technologies (quantum communication, quantum information processing, …)
- The ideal single-photon emitter (i) delivers individual photons at high rate and narrow frequency distribution at RT and (ii) is stable over long time periods
- 2D materials with defects present were shown to host SPE
- Defects are easier to control in 2D materials compared to bulk materials

Conduct computational screening of intrinsic defects in 2D materials

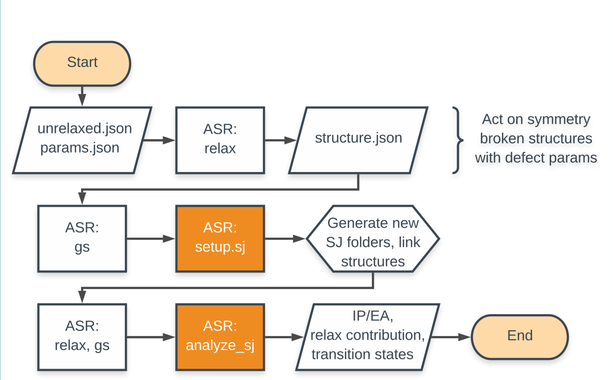
THE WORKFLOW

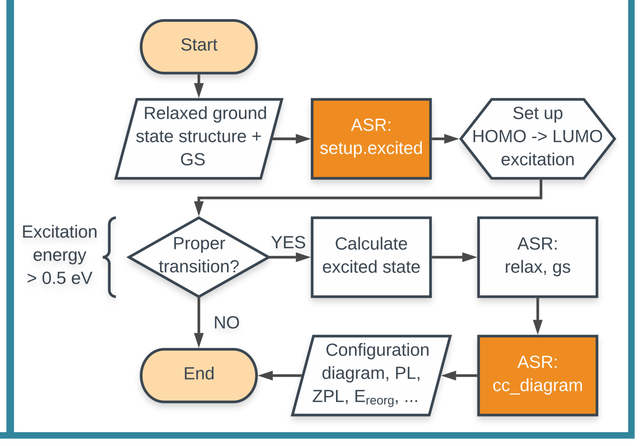


AUTOMATIC GENERATION OF DEFECTS AND SUPERCELLS



GROUNDSTATE AND EXCITED STATE WORKFLOW





OVERVIEW OF PROPERTIES

Slater-Janak (SJ) Transition State Theory

- Kohn-Sham eigenvalues are related to the derivative of the total energy E with respect to occupation number η_i of the respective orbital

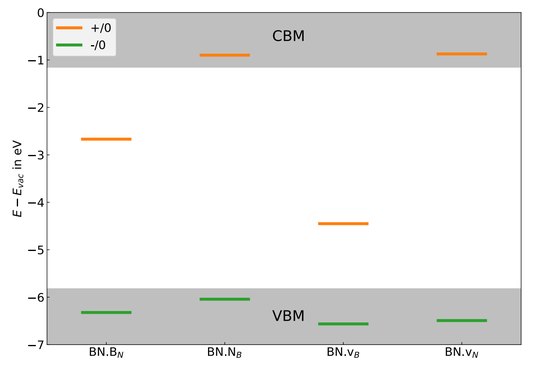
$$\frac{\partial E[N]}{\partial \eta_i} = \varepsilon_i$$

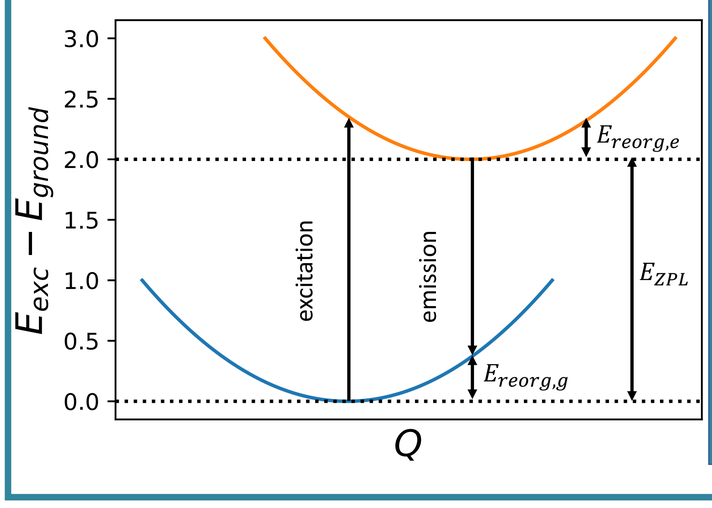
- Assume linear relation between highest occupied state ε_H and occupation number

$$E^{N+1} - E^N = \int_0^1 \varepsilon_H(\eta) d\eta = \varepsilon_H\left(\frac{1}{2}\right)$$

$$E^{N-1} - E^N = \varepsilon_L\left(-\frac{1}{2}\right)$$

- Obtain charge transition levels of defects (see right for hBN defects) [5]





Excited State Properties

- Excite HOMO -> LUMO single electron excitation
- Calculate photoluminescence spectrum for a given defect system:

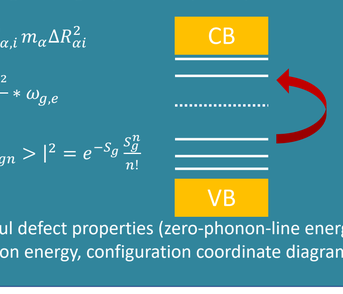
$$L(\hbar\omega) = \sum_n |< \chi_{e0} | \chi_{gn} >|^2 \delta\left(E_{ZPL} + \hbar(\omega_e - n\omega_g - \omega)\right)$$

- Use 1D approximation (displacement along main phonon mode) for large Huang-Rhys factors ($S \gg 1$)

$$\Delta Q^2 = \sum_{\alpha,i} m_\alpha \Delta R_{\alpha i}^2$$

$$S_{g,e} = \frac{\Delta Q^2}{2} * \omega_{g,e}$$

$$|< \chi_{e0} | \chi_{gn} >|^2 = e^{-S_g} \frac{S_g^n}{n!}$$



- Extract useful defect properties (zero-phonon-line energy, reorganization energy, configuration coordinate diagram, …)

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