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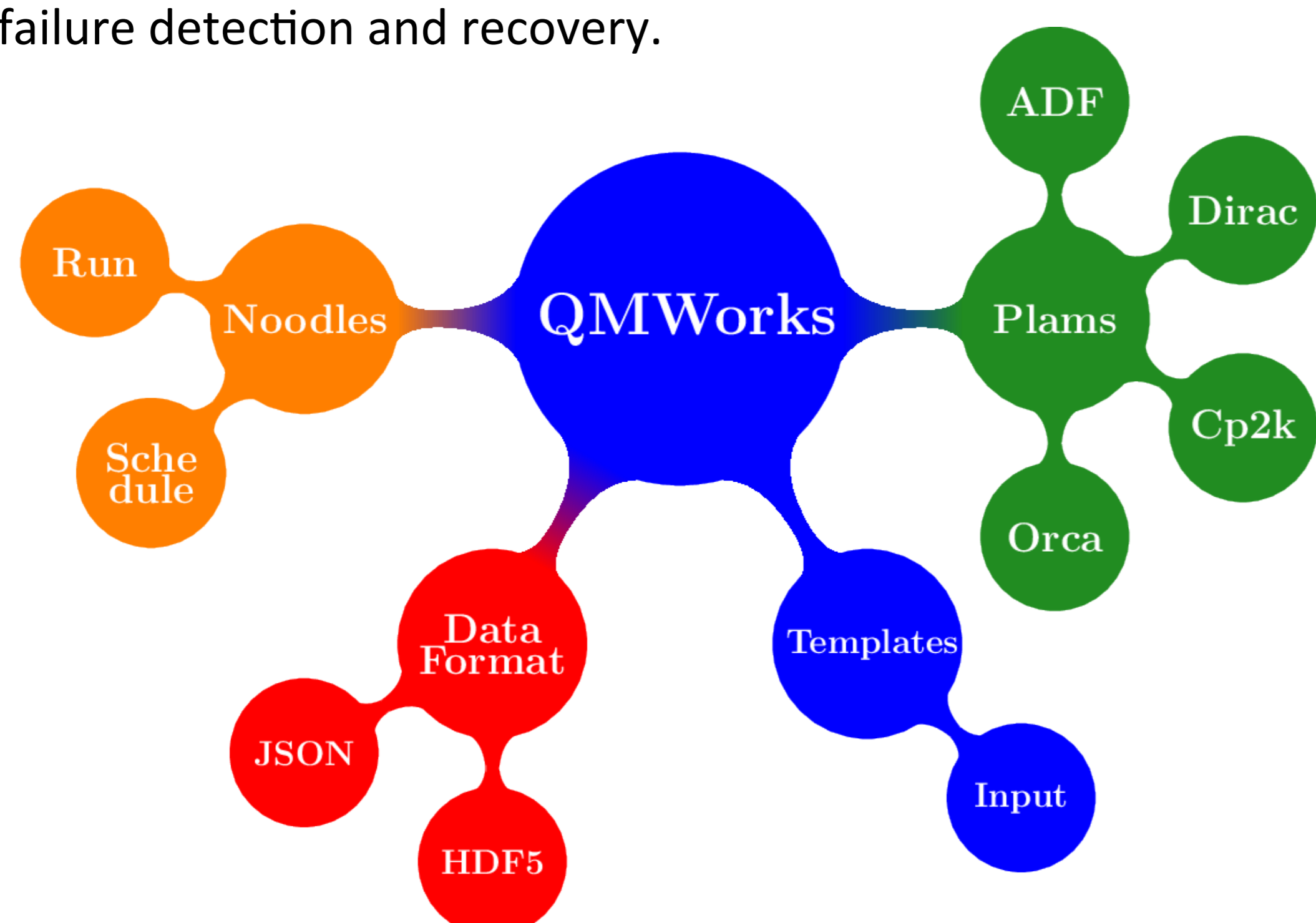
Introduction

Research on modern computational quantum chemistry relies on a diverse set of computational packages to carry out calculations. The complexity of the workflows usually requires intercommunication between the aforementioned tools. The communication is usually carried out through shell scripts that try to automate input/output actions like: launching computations in a cluster, reading the resulting output, and feeding a subset of numerical results to another program. Such *ad hoc* scripts are difficult to maintain and extend, requiring a significant programming expertise to work with them. It is thus desirable to have a set of general and extensible tools that allows to perform complex simulations on heterogeneous hardware platforms.

Aims

The goal of our project is to create a modular and extensible software called **qmworks** to automate the following tasks using Python:

- Input generation (*Templates* and *Plams*).
- Dependencies management (*Noodles*).
- Efficient data storage (*HDF5*).
- Job failure detection and recovery.



Case Study

Excited states molecular dynamics are carried out using the fewest switches surface hopping algorithm (FSSH)¹, together with the classical path trajectory approximation, using the *PYXAID* package². In this approach the Schrödinger time-dependent equation is expanded in a set of N adiabatic states:

$$\Psi(\mathbf{R}, t) = \sum_{i=1}^N c_i(t) \phi_i(\mathbf{R}(t)) \quad (1)$$

Where the time-dependent coefficient are expressed in the following way:

$$\frac{dc_j(t)}{dt} = -i\hbar^2 c_j(t) E_j(t) - \sum_{i=1}^N c_i(t) \sigma_{ji}(t) \quad (2)$$

Where the nonadiabatic coupling matrix σ_{ij} is given by

$$\sigma_{ji}(t) \approx \frac{1}{4\Delta t} (3\mathbf{S}_{ji}(t) - 3\mathbf{S}_{ij}(t) - \mathbf{S}_{ji}(t - \Delta t) + \mathbf{S}_{ij}(t - \Delta t)) \quad (3)$$

Being $\mathbf{S}_{ij}(t)$ the overlap matrix between two consecutive time steps.

Solutions of this equation provides the time-evolution of both electron and holes and serves as a powerful tool to analyze photophysical processes that occur in the pico- and subpico- timescales like electron injection between a donor and an acceptor, charge trapping induced by defects, etc.

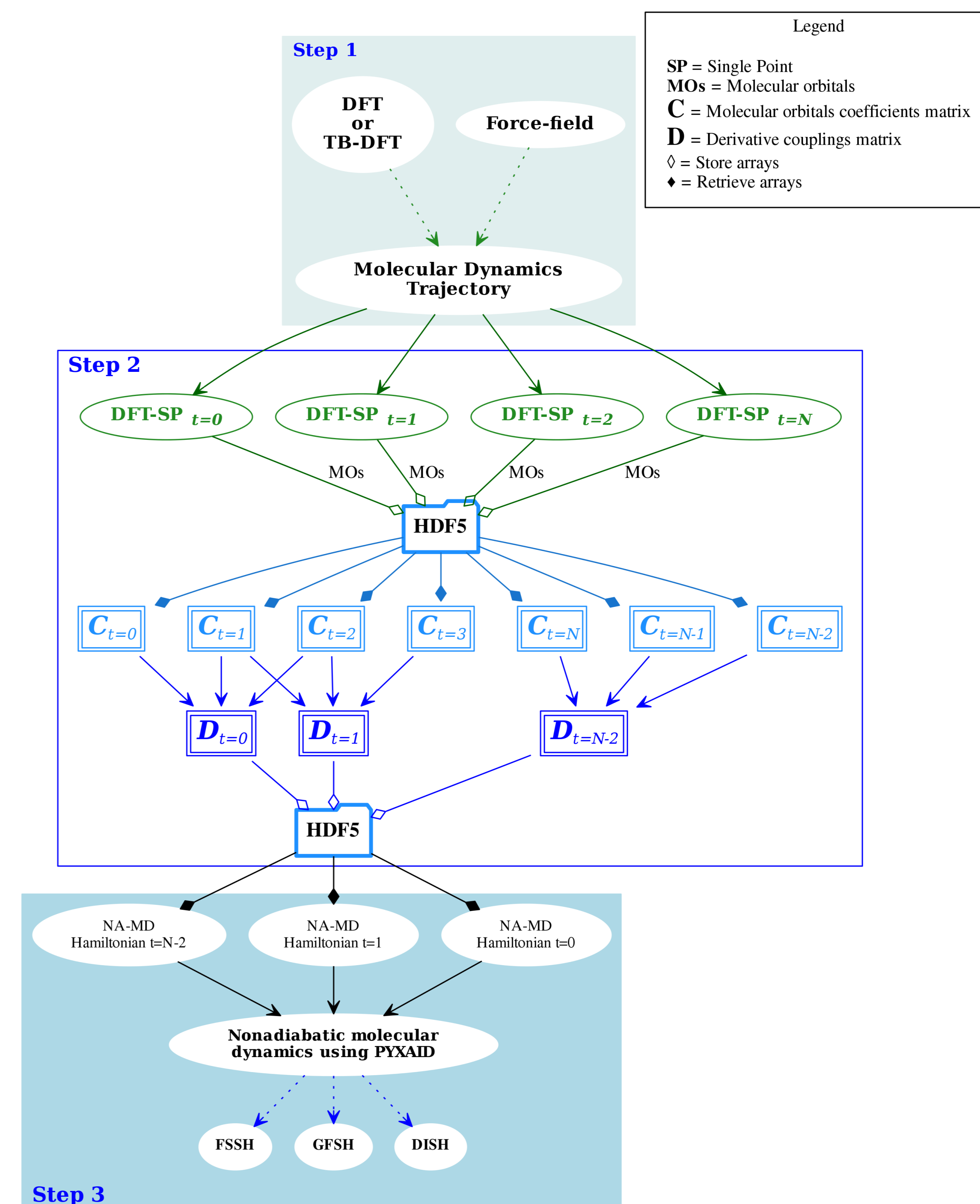
References

- [1] J. C., Tully. *J. Chem. Phys.* 1990, 93, 1061.
[2] Akimov, A. Prezdho. *J. Chem. Theory Comput.*, 2013, 9 (11), 4959.

Acknowledgements

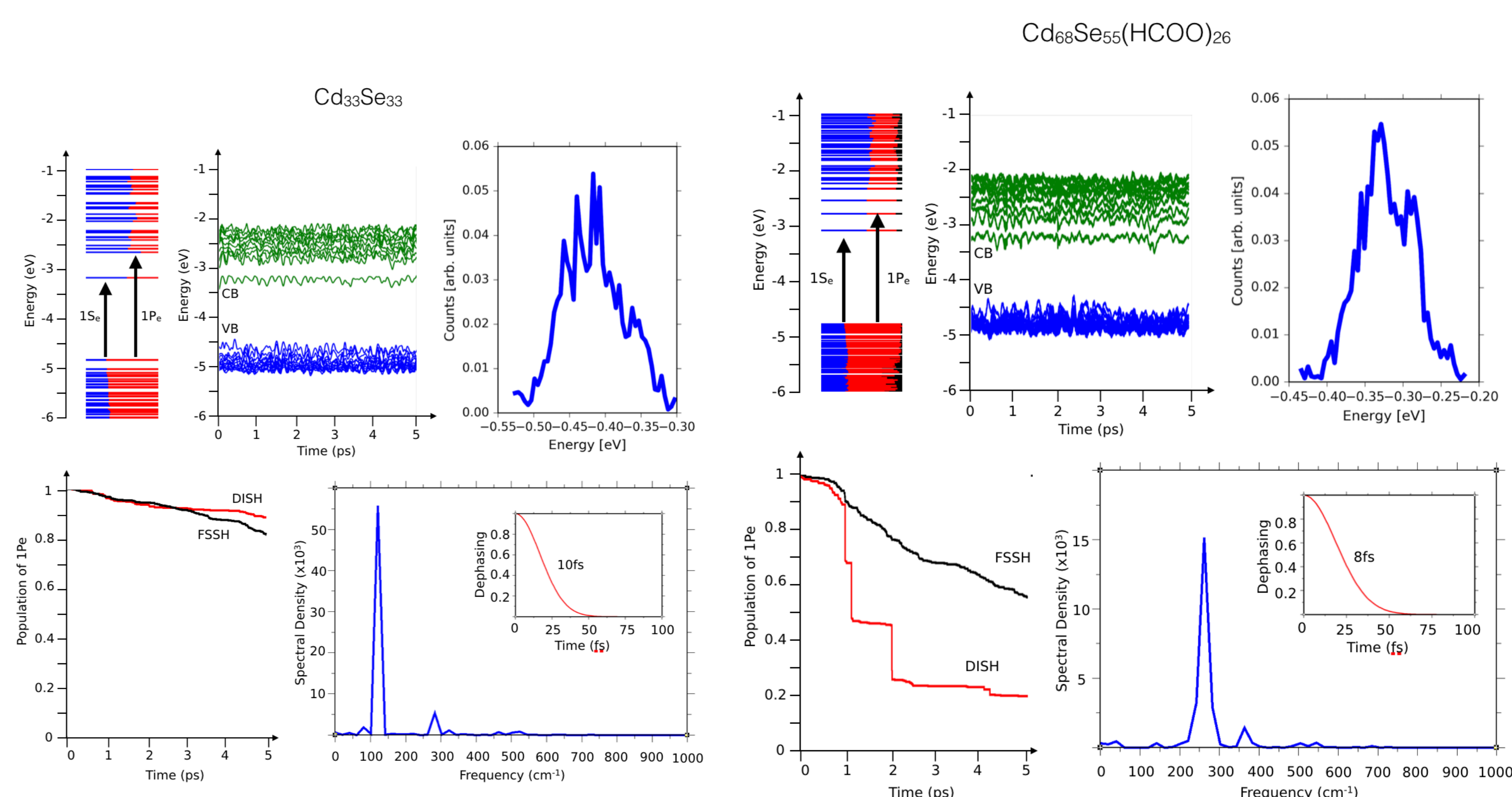
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Case Study Workflow



The excited states molecular dynamics simulations are performed in three steps:

1. Carrying out an *ab initio* or classical molecular dynamic simulation in the ground state.
2. Constructing the right-side of equation (2) at each time step t of the pre-computed molecular dynamics trajectory from step 1 by retrieving the energies E_j of the adiabatic states and the nonadiabatic coupling matrix σ_{ij} from an electronic structure calculations based on, for example, density functional theory (DFT). These terms can be calculated using a computational chemistry code like *CP2K*, *ADF*, etc.
3. Solving equation (2) at each time step using the *PYXAID*² code.



Conclusions

- A General software infrastructure have been developed to carry out complex simulations.
- The Software has been successfully tested on the simulation of nonadiabatic molecular dynamics of quantum