CFEL SCIENCE

SEMINAR

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CFEL-bldg. 99, seminar room I

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The two-layer multiconfiguration time-dependent Hartree method with variational Gaussian wave packets

The prevalent methods to calculate the quantum dynamics (of the nuclei) of molecules are the Multiconfiguration Time-Dependent Hartree Method (MCTDH) and variants thereof. They rely on expanding the system's overall wave function into a product basis of *time-dependent* wave functions of lower dimensionality ("Single Particle Functions", SPFs) to alleviate the exponential scaling of the numerical effort with the number of the system's degrees of freedom. The SPF's equations of motion are determined by the Dirac-Frenkel variational principle and generically involve mean-fields.

Not restricting the form of admissible SPFs (as in standard MCTDH) allows for small basis sets but makes propagating each SPF numerically expensive. Allowing only SPFs of a fixed form (e.g. Gaussian wave packets in G-MCTDH) reduces the cost of their propagation but necessitates larger basis sets.

We describe a novel two-layer variant of G-MCTDH which improves on the performance and convergence properties of quantum propagation based on frozen Gaussians (FGs). While the standard scheme uses factorizable multi-dimensional FGs, the present approach combines these into flexible, MCTDH-like SPFs.

At the same time, the still expensive variational evolution of the Gaussian parameters is reduced to low-dimensional subspaces. As a result, the novel scheme significantly alleviates the current bottleneck to accurate propagation in G-MCTDH and its variational multiconfigurational Gaussian (vMCG) variant.

Since the first-layer single-particle functions are chosen to be orthogonal, the present approach can be straightforwardly combined with existing multi-layer MCTDH schemes.

Host: Robin Santra – Theory Division