New Algorithms for Large Scale First-Principles Molecular Dynamics

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Since the pioneering work of Car and Parrinello, the history of First-Principles Molecular Dynamics has been closely associated with the development of high-performance computers. The efficiency of the algorithms proposed in the original Car-Parrinello method—Fast Fourier Transforms and dense linear algebra—have made it possible to successfully exploit multiple generations of supercomputers during the past three decades, including vector computers and distributed memory parallel computers. Today, a substantial fraction of available computing cycles worldwide is devoted to first-principles simulations.

As new computers become more powerful and their architecture becomes more complex, the design of first-principles simulation programs must strike the right balance between algorithmic simplicity—which is desirable for efficient scaling—and more elaborate algorithms that potentially reduce the computational complexity of the calculations but may compromise scalability or reduce accuracy.

We present a recently developed reduced-scaling algorithm for the computation of Hartree-Fock exchange in FPMD simulations. The approach is based on a recursive subspace bisection method that allows for a compact representation of Kohn-Sham orbitals, while preserving accuracy in a controlled manner. Applications to simulations of the solvation of a chloride ion in water using the PBE0 density functional will be discussed.

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