"The fundamental barriers to calculating the quantum dynamics of polyatomic molecules driven in modern and next-generation nonlinear ultrafast laser experiments will be reviewed. The wave function representation of MCTDHF directly addresses the need for a quantum mechanically coherent description of molecules with many electrons interacting with many photons. It is an open question whether the method can be scaled or elaborated to successfully describe a range of nonlinear laser dynamics in polyatomic molecules with more than a few atoms and dozens of electrons.

The extension of MCDTHF to the description of nonlinear quantum dynamics in larger molecules will be discussed. These dynamics involve (1) dozens of electrons and (2) polyatomic nuclear motion. We argue that some of the fundamental barriers are being surmounted whereas others remain.

The barriers to a (1) scalable treatment of the electronic part of the problem are being attacked in several ways currently, restricted Slater determinant spaces (pruning the basis) and multilayer strategies in particular. Scalable implementations for the electronic problem will be discussed including (a) polyatomic orbital basis sets, (b) restricted Slater determinant spaces, (c) the multilayer framework, (d) Lindblad equation methods for multiple ionization, and (e) possible future directions including elaborations specific to the fermion problem.

Less progress has been made on the inclusion of nuclear motion (2). The fundamental barriers to including quantum vibration or rotation in the MCDTHF method will be identified, and different implementations of the fully nonadiabatic quantum electronic + nuclear problem that have already been implemented will be reviewed. The most promising strategy going forward is probably the combination of semiclassical nuclear dynamics with quantum electronic dynamics. Contemporary work on combined semiclassical and quantum methods may supply ingredients that are needed for a scalable all-purpose method for calculating the electronic and nuclear dynamics of molecules driven in modern ultrafast nonlinear laser experiments."