In recent years, in part due to progress made in generating and controlling intense laser fields, the timescale of dynamical processes in atomic and molecular systems has been pushed into the attosecond regime (1 as=10^{-18} s). In parallel with experimental work, theoretical methods are being developed to treat explicitly time-dependent electronic motion after photoexcitation on ultrashort timescales. This talk describes correlated, explicitly time-dependent N-electron methods and their application to selected problems.

In the presentation, we will review wave-function based, correlated many-electron methods and their application to femtosecond-laser driven dynamics in molecules. Specifically, we will be concerned with multi-determinant expansions of the N-electron wave-function such as time-dependent configuration interaction (TD-CI), where only the coefficients of the Slater determinants are time-dependent, and, for comparison, also with time-dependent complete active space SCF (TD-CASSCF), for which both the coefficients and the determinants depend on time. Extensions to include ionization, dissipation, and optimal control strategies are presented.

These methods will then be applied (i) for laser-pulse excitation and switching of real molecules without and with a dissipative environment, (ii) for the calculation of response properties of small molecules when driven by intense laser pulses, (iii) for long-range intermolecular charge transfer between molecules, (iv) for controlled electron dynamics, and (v) for ionization. Some fundamental questions associated with time-dependent electron structure theory will also be addressed.