# Abstract

The absorption of energy from intense, ultrashort laser pulses by molecules leads to the removal of one or more electrons and may induce fragmentation in molecular gas-phase targets. The amount of energy absorbed is dependent on many factors, including laser parameters and electronic properties of the molecule. This thesis experimentally considers the interaction of $C_{60}$ with femtosecond laser pulses of moderate laser intensities ($10^{11}$ - $10^{13}$ W/cm$^2$) in order to understand the processes of absorption and redistribution of energy in complex molecules.

Photo-electron spectra of $C_{60}$ molecules reveal an electronic structure, which has been determined to primarily belong to three Rydberg series, corresponding to the angular momenta $\ell = 3$, 5, and 7 (f, h, and j orbitals, respectively). The excitation and time dynamics of these peaks have been investigated with photo-electron and time-resolved pump-probe spectroscopy. The excitation of the Rydberg series has been interpreted to be a manifestation of the excitation of multiply active electrons into an intermediate state, which is then broadened by non-adiabatic transitions. From this "doorway" state, the Rydberg states are populated. The experimental results indicate that this intermediate excited state has a lifetime greater than 1 ns.

The unique fragmentation pattern of $C_{60}$ observed with nanosecond laser excitation has been successfully explained through statistical models. However, the nature of fragmentation under femtosecond laser irradiation is not obvious since the deposition of energy occurs on time scales much shorter than the internal energy redistribution. Time-resolved photo-ion time of flight spectroscopy and measurement of the spatial distribution have been used to study the fragmentation of $C_{60}$ after femtosecond laser excitation. Three processes describe the fragmentation of $C_{60}$ under fs-laser irradiation: sequential $C_2$ loss, asymmetric fission, and multiple fragmentation. The importance of these mechanisms depends on the charge state and the internal energy. The experimental results further indicate that the majority of the fragmentation that occurs after femtosecond laser excitation can be explained through statistical models, however, a small portion may be from the existence of non-statistical fragmentation channels.

The fragmentation of $C_{60}$ has been enhanced by altering the temporal envelope of the interacting laser field with the femtosecond pulse shaping technique. Using a closed loop optimization algorithm, an optimal pulse form has been found indicating that the ionic state is excited first, from which additional energy is easily absorbed, leading to an enhancement in fragmentation.