SPECTRA AND DYNAMICS OF MOLECULES IN LONG-LIVED, HIGHLY REACTIVE TRIPLET STATES

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Molecules in their excited triplet states are highly chemically reactive. They are metastable, with long radiative lifetimes ($> 100\mu s$), carry chemically significant energy ($> 50kcal/mol$), and can exist in several isomeric forms. These characteristics make molecules in triplet states act as chemical protagonists in reactions initiated by ultraviolet radiation, and in energetic environments where collisions are the reaction initiators. Despite their key role in chemical reactions, molecules in triplet states can easily elude detection, largely because they do not make their presence known via fluorescence. The excitation and de-excitation dynamics of triplet states is not well understood. As an alternative to direct optical pumping, excitation transfer from a metastable atom to a target organic molecule for selective, efficient population of triplet states is very promising. In this scheme, a donor atom is optically excited via a two-photon transition, and the excitation energy is transferred from the atom to the acceptor molecule in a pulsed jet expansion. Hg atoms are used in this indirect, collision-induced photosensitized excitation transfer experiment to populate the metastable triplet states of target molecules such as acetylene and ethylene. SEELEM (surface electron ejection by laser excited metastables) is the detection method for both the laser excited atomic species and the product metastable molecules. SEELEM is employed as a probe of triplet state excitation and de-excitation dynamics. A double-resonance 'SEELEM dip' experiment will be described in which one of the metastable species is selectively depopulated via laser. The goal is characterization of the molecular triplet states, leading to illumination of the dynamical processes which underlie their complex spectra.