

STATE SELECTIVE TIME OF FLIGHT SPECTROSCOPY*

J. D. Campbell[¶], M. H. Yu, M. Mangir and C. Wittig

University of Southern California, University Park, Los Angeles, Calif.

90007 (U.S.A.)

In this paper we discuss recent experiments in which we have applied the newly developed technique of optical time of flight (OTOF) spectroscopy to detailed studies of energy partitioning in photodissociation processes. In these experiments we combine the unique features of laser induced fluorescence (LIF) with TOF measurements to provide a state selective, time resolved, and highly sensitive probe of the recoil energy content of photofragments produced via either single or multiple quantum dissociation processes.

The principal components of the experimental arrangement consist of (1) a photolytic source; (2) a N₂ laser pumped dye laser diagnostic probe; (3) a fluorescence chamber containing a small concentration of the species to be dissociated and equipped with a sensitive photomultiplier tube detector; (4) a signal acquisition/data processing system. The photolytic source can be either a high power, pulsed ir laser (typically a CO₂ TEA laser) for multiple quantum dissociation experiments, or a pulsed rare gas excimer laser operating in the UV for studies of single quantum dissociation processes. The dye laser is constructed in a basic, grating-tuned oscillator/amplifier configuration which is easily adapted for operation at bandwidths of ~1 GHz using intracavity, pressure-tuned etalons.

*Supported by the National Science Foundation.

[¶]Present address: Math Sciences Northwest, Bellevue, WA 98009 (U.S.A.)

In this work, the photolysis and probe laser beams are spatially separated, at a fixed distance, and the intensity of LIF produced by spatially selected photofragments which encounter the probe laser beam at a given delay between the photolysis and probe lasers is measured. By varying the delay, the TOF spectrum for the particular fragment states probed by the dye laser is obtained from which the recoil energy spectrum in the center of mass reference frame can be deconvoluted using Fourier transform techniques.

In the present bulk gas type experiments only a small fraction of the isotropically scattered photofragments are selected for analysis, and the resultant, weak LIF intensities must be acquired and processed using photon counting techniques. A second generation fluorescence chamber, now under construction, will incorporate molecular beam techniques to improve signal intensities as well as provide information for the angular distribution of recoiling fragments.

Using the bulk gas arrangement, we have examined fragment energy recoil spectra for two distinct types of multiple quantum dissociation processes in intense infrared fields. In one of these processes, we have monitored the C_2 molecule produced in the $a^3\pi_u$ state via *sequential* multiple photon dissociations for various polyatomic parent molecules. In the other process we have monitored the NH_2 molecule produced in the $\tilde{X}(^2B_1)$ state as the primary photofragment of the lowest energy dissociation channel for the N_2H_4 parent molecule. The recoil spectra for these fragments are compared and discussed in terms of the essential features of energy partitioning which have been observed in representative single quantum dissociation processes.