

**Stony Brook University
The Graduate School**

Doctoral Defense Announcement

Abstract

DC Slice Ion Imaging Study of Atomic Orbital Orientation and Alignment in
Photodissociation

By

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A complete study of atomic photofragment polarization has been achieved by using DC slice imaging. In this dissertation, the theoretical treatment adapted for the sliced images has been derived to extract the angular momentum polarization anisotropy parameters for any recoil speeds, and the important photodissociation dynamics of small polyatomic molecules has been presented.

The first demonstration of DC slice imaging of orbital polarization was a study of the 193 nm photodissociation of ethylene, followed by detailed investigation of ozone and OCS. In ozone, the speed-dependent orientation was measured for O(1D_2) atom produced from the photodissociation in the 248-285 nm region. Strong recoil speed-dependent orientation following photolysis by linearly polarized light at all wavelengths was observed. The origin of this polarization is ascribed to nonadiabatic transitions at avoided crossings and at long range. The atomic orbital alignment and orientation including the higher order moments ($K=3,4$) has been carried out for the photodissociation of OCS at 193 nm. The observed speed-dependent β and polarization parameters of S(1D_2) atom support the interpretation that there are two main dissociation processes: a simultaneous two-surface excitation and the initial single-surface excitation followed by the nonadiabatic crossing to ground state. The density matrix can be constructed containing the higher order contributions for the circularly-polarized dissociation light. It was suggested in one case that the higher order contributions should not be overlooked for an accurate picture of the dissociation dynamics in this system.

In addition, photofragment angular anisotropy are reported for the photodissociation of acetaldehyde cations in the wavelength range 354-363 nm. In particular, the distinct angular distribution of the CH_3CO^+ fragments shows a large value of the higher order Legendre polynomial term, providing evidence that acetaldehyde cations are spatially aligned during the ionization process.

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