Title

MOLECULAR BEAM STUDIES OF INFRARED MULTI-PHOTON DISSOCIATION OF POLYATOMIC MOLECULES

Author

Lee, Y.

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Abstract

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Yuan T. Lee

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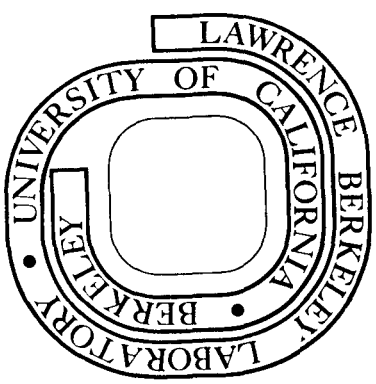

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Molecular Beam Studies of Infrared Multiphoton Dissociation of Polyatomic Molecules

Yuan T. Lee

Dept. of Chemistry & Lawrence Berkeley Laboratory
University of California, Berkeley, California
U.S.A 94720

Dynamics of infrared multiphoton dissociation of polyatomic molecules has been investigated in a crossed laser-molecular beam arrangement for some 14 different molecules. The direct identification of dissociation channels, the measurements of angular and velocity distributions of dissociation products as a function of laser energy and the lifetime of an excited molecule estimated from the onset of secondary decomposition, all indicated that the energy distributions in the excited molecules were considerably randomized before decomposition and that mode selected bond breaking did not occur. Instead, excited molecules mainly dissociate through the lowest energy channels. Although the average level of excitation beyond the dissociation energy was found to vary from 1 (for N_2F_4) to 9 (for SF_6) excess CO_2 laser photons, the molecules were found to be excited to levels with a characteristic lifetime of approximately 10 nsec, in the cases where a statistical (RRKM) theory could be applied. Apparently, the absorption of infrared photons from a high power laser was eventually limited by the lifetime of excited molecules. In systems where 3-center and 4-center eliminations are observed, it is possible to derive some information on the nature of potential energy surfaces for elimination processes. A phenomenological model calculation that exhibits the realistic qualitative behavior of multiphoton excitation and dissociation of polyatomic molecules will also be discussed.

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TECHNICAL INFORMATION DEPARTMENT
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720