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Broida Prize Talk: Imaging Photodissociation Dynamics

HANNA REISLER, Chemistry Department, University of Southern California, Los Angeles, CA 90089-0482

Advances in the technique of charged particle imaging (photofragments and photoelectrons) have enabled recent progress in understanding complex interactions involving electronically excited states of molecules, radicals, and transient species. In this talk, studies of the photodissociation dynamics of the NO dimer, a weakly covalently bound molecule, will be described in which imaging of photoions and photoelectrons is used to: (i) characterize the nature of the electronically excited states of the NO dimer; (ii) study and model the photofragmentation dynamics to NO fragments at the pair-correlation level; (iii) elucidate the role of nonadiabatic transitions, in particular interactions between Rydberg and valence states; (iv) determine the branching between channels in which one of the NO products is in an electronically excited state; and (v) clarify when the dimer behaves as a covalently bound ONNO molecule and when it exhibits properties similar to those of a van-der-Waals complex. With the aid of electronic structure calculations and time-resolved measurements, an intriguing picture of the dynamics emerges, in which interactions between valence and Rydberg states that vary during the dissociation play a crucial role.

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