

Time-resolved imaging of purely valence-electron dynamics during a chemical reaction

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Chemical reactions are manifestations of the dynamics of molecular valence electrons and their couplings to atomic motions. Emerging methods in attosecond science can probe purely electronic dynamics in atomic and molecular systems^{1–4}. By contrast, time-resolved structural-dynamics methods such as electron^{5–10} or X-ray diffraction¹¹ and X-ray absorption¹² yield complementary information about the atomic motions. Time-resolved methods that are directly sensitive to both valence-electron dynamics and atomic motions include photoelectron spectroscopy^{13–18} and high-harmonic generation^{19,20}; in both cases, this sensitivity derives from the ionization-matrix element^{16,21}. Here we demonstrate a time-resolved molecular-frame photoelectron-angular-distribution (TRMF PAD) method for imaging the purely valence-electron dynamics during a chemical reaction. Specifically, the TRMF PADs measured during the non-adiabatic photodissociation of carbon disulphide demonstrate how the purely electronic rearrangements of the valence electrons can be projected from inherently coupled electronic-vibrational dynamics. Combined with ongoing efforts in molecular frame alignment²² and orientation^{23,24}, TRMF PADs offer the promise of directly imaging valence-electron dynamics during molecular processes without involving the use of strong, highly perturbing laser fields²⁵.

Figure 1 provides a conceptual overview of our method. Carbon disulphide, CS₂, is a molecule that exhibits all the features generic to polyatomic molecular dynamics: vibrational mode coupling, conical intersections, spin conversion and photodissociation. As such, its non-adiabatic photodissociation reaction CS₂(X) + hν(201.2 nm) → CS₂⁺(C) → CS(X) + S(¹D, ³P), shown in Fig. 1, provides an excellent test. In this work we combine experimental measurements with theory to demonstrate how the TRMF PAD images the evolution of the valence-electronic structure of an excited-state wavepacket during the complex, coupled electron–nuclear processes inherent to chemical reactions. TRMF PADs probe both nuclear and electronic degrees of freedom through the photoionization matrix elements, $\mathbf{d}(t) = \langle \Psi_{+} | \hat{\mathbf{d}} | \Psi_{-}(t) \rangle$, and photoelectron (Ψ_{+}) states, through dipole coupling ($\hat{\mathbf{d}}$) with the laser field (\mathbf{E} ; refs 18,24). In the case of polyatomic molecules, $\Psi_{+}(t)$ and Ψ_{-} are composed of coupled electronic and vibrational components (see Supplementary Information). The significance of the TRMF PAD can be understood by considering the intimate relationship between Ψ_{+} and the electronic part of $\Psi_{-}(t)$. For example, under the well-known Born (plane-wave) approximation,

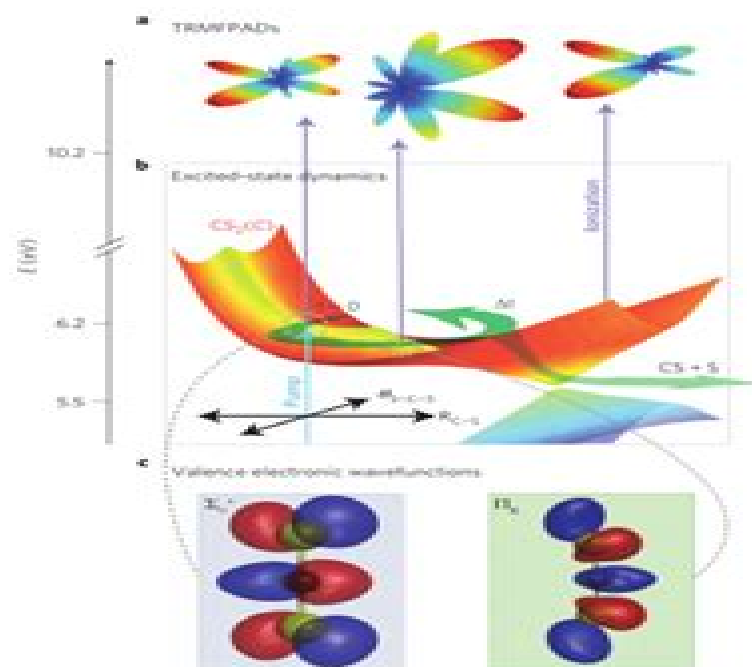


Figure 1 | Conceptual picture of TRMF PADs as a probe of valence-electronic dynamics during CS₂ photodissociation.

a, b, TRMF PADs (**a**) resulting from the wavepacket that evolves on the excited-state potential-energy surface (**b**). Dissociation to form free CS + S products occurs along the asymmetric stretch coordinate, through conical intersections to lower-lying singlet and triplet states. **c**, Left, Valence electronic wavefunctions in the initial linear geometry, of dominant Σ_u^+ character. Right, The appearance of Π_u character on wavepacket evolution. The time evolution of the MF PAD (**a**) directly reflects the evolution in valence-electronic character (**c**).

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Imaging In Chemical Dynamics

**Thomas Laurell, Johan Nilsson, Jed
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High Resolution Imaging Probes of Chemical Dynamics Wen Li, 2006 *Modern Trends in Chemical Reaction Dynamics* Xueming Yang, Kopin Liu, 2004 The field of chemical reaction dynamics has made tremendous progress during the last decade or so This is due largely to the development of many new state of the art experimental and theoretical techniques during that period It is beneficial to present these advances both theoretical and experimental in a review volume published in two parts Parts I and II The primary purpose of this review volume is to provide graduate students and experts in the field with a rather detailed picture of the current status of advanced experimental and theoretical research in chemical reaction dynamics All chapters in these two parts have been written by world renowned experts active in such research Modern Trends In Chemical Reaction Dynamics - Part I: Experiment And Theory Kopin Liu, Xueming Yang, 2004-03-09 The field of chemical reaction dynamics has made tremendous progress during the last decade or so This is due largely to the development of many new state of the art experimental and theoretical techniques during that period It is beneficial to present these advances both theoretical and experimental in a review volume published in two parts Parts I and II The primary purpose of this review volume is to provide graduate students and experts in the field with a rather detailed picture of the current status of advanced experimental and theoretical research in chemical reaction dynamics All chapters in these two parts have been

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medicine The Handbook of Modern Biophysics represents an aspect of that effort **Microtas 2004** Thomas Laurell,Johan Nilsson,Jed Harrison,Klavs Jensen,J. P. Kutter,2004 The Eighth International Conference on Miniaturized Systems in Chemistry and Life Science MicroTas 2004 is an annual meeting focusing on the research development and application of miniaturized technologies and methodologies in chemistry and life science The conference is celebrating its tenth anniversary after the first workshop at the University of Twente The Netherlands in 1994 This research field is rapidly developing and changing towards a domain where core competence areas such as microfluidics micro and nanotechnology materials science chemistry biology and medicine are melting together to a truly interdisciplinary meeting place This volume is the first in a two volume set a valuable reference collection to all working in this field Research in Chemical Kinetics Bozzano G Luisa,2012-12-02 Research in Chemical Kinetics Volume 1 focuses on authoritative review articles on a wide range of developing topics in the kinetics of gaseous and condensed phases The selection first elaborates on gas phase kinetics of free radicals studied by pulse radiolysis combined with time resolved infrared diode laser spectroscopy and solid liquid reactions of environmental significance Discussions focus on coprecipitation of phosphate with calcite reactions of silica and quartz infrared spectroscopy of free radicals and kinetics of methyl radicals The book then examines the collision energy dependence of reaction cross sections and photoelectrochemical dynamics including organometallic photoelectrochemistry and photofragmentation voltammetry contrasting reactivity of ion radical excited states photoelectrochemical reaction mechanisms and experimental methods The publication tackles collisional electronic energy transfer in CN free radicals photo ion imaging techniques and future directions in reactive scattering and photoelectrochemical dynamics Topics include photoelectrochemical reaction mechanisms photoelectrochemical measurement of quantum yields photofragment translational spectroscopy and velocity distributions The selection is a valuable reference for researchers interested in the kinetics of gaseous and condensed phases **Advanced MR Imaging in Clinical Practice, An Issue of Radiologic Clinics of North America** Hersh Chandarana,2015-06-14 Editor Hersh Chandarana MD and authors review Advanced MR Imaging in Clinical Practice Articles will include Current Status of Diffusion Weighted Imaging Current Status of Perfusion Weighted Imaging Non gadolinium Enhanced MR Angiography Pearls and Pitfalls of 3 T imaging Implementing MR Neurography in Clinical Practice Imaging around Hardware and Metal Recent Advances in T1 and T2 Weighted Imaging of the Abdomen and Pelvis Recent Advances in Neuro and Spine Imaging Advances in MR Hardware and Software and more **NMR Imaging in Chemical Engineering** Siegfried Stapf,Song-I Han,2006-02-10 How to use nuclear magnetic resonance imaging in chemical engineering Written by the internationally recognized top experts from academia and industry this first book dedicated to the topic provides an overview of existing methods and strategies to solve individual problems in chemical engineering Written in a simple and lively manner and backed by various industrial examples the book begins with a look at hardware and methods continuing on to cover porous

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The Zebrafish: Disease Models and Chemical Screens H. William Detrich, Monte Westerfield, Leonard I. Zon, 2011-09-23 This volume of *Methods in Cell Biology* is the 3e and provides comprehensive compendia of laboratory protocols and reviews covering all the new methods developed since 2004 This new volume on *Disease Models and Chemical Screens* covers two rapidly emerging and compelling applications of the zebrafish Details state of the art zebrafish protocols delineating critical steps in the procedures as well as potential pitfalls This volume concentrates on *Disease Models and Chemical Screens*

Synchrotron Radiation Techniques in Industrial, Chemical, and Materials Science Kevin L. D'Amico, Louis J. Terminello, David K. Shuh, 2012-12-06 The individual papers that comprise this monograph are derived from two American Chemical Society ACS Fall National Meetings that focused on the current uses of synchrotron radiation SR research techniques The first Symposium was held in Washington DC in August 1994 and the second convened in Chicago IL in August 1995 The intent of these symposia was to present a broad overview of several current topics in industrial chemical and materials based SR research to a chemically inclined audience The SR techniques covered were divided roughly into the

three general fields of industrial chemical and materials science for this purpose Included within these four categories are environmental geologic atomic molecular analytical solid state physics surface science and biological applications of SR There is little doubt that structural biology and environmental science are the largest growth areas in SR research as this monograph goes to press The spirit of these symposia was to bring together the expert synchrotron radiation user with new and potential users of SR techniques There are now a preponderance of particle storage rings located throughout the world devoted exclusively to the production of SR There have been great improvements in the particle accelerators and storage rings from which SR emanates These newest third generation SR sources are the result of the successful collaboration between SR users and accelerator physicists which has made a reality out of experiments never before possible

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