

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

Paul Hockett¹, Christer Z. Bisgaard^{1,2}, Owen J. Clarkin³ and Albert Stolow^{1,3} *

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, $\mathbf{d}(t) = \langle \Psi_+ | \hat{\mathbf{d}} | \Psi_-(t) \rangle$, where Ψ_+ 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,

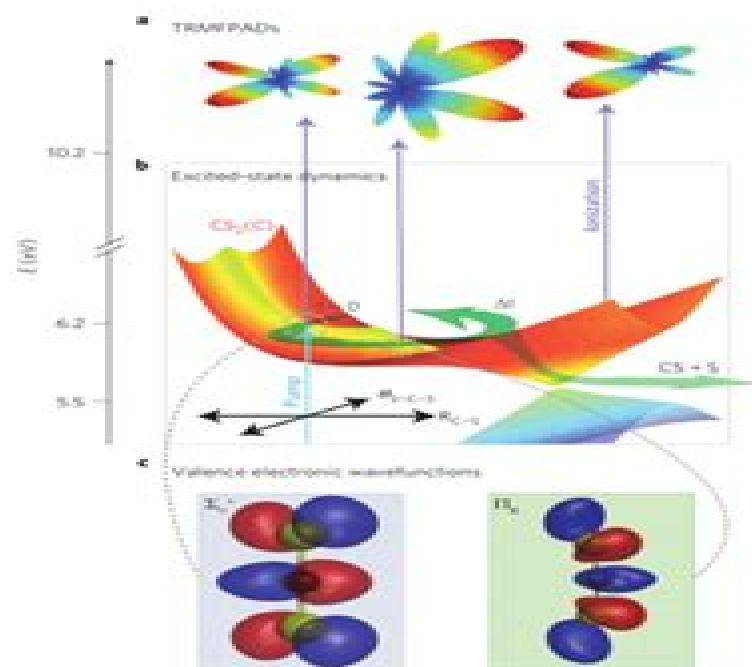


Figure 1 | Conceptual picture of TRMF PADs as a probe of valence-electronic dynamics during CS₂ photodissociation.

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_g⁺ character. Right, The appearance of Π_g character on wavepacket evolution. The time evolution of the MF PAD (**a**) directly reflects the evolution in valence-electronic character (**c**).

¹Steele Institute for Molecular Sciences, National Research Council, 100 Sussex Drive, Ottawa, Ontario K1A 0R6, Canada, ²Department of Chemistry, University of Copenhagen, Copenhagen DK-2100, Denmark, ³Department of Chemistry, Queen's University, Kingston, Ontario K7L 3N6, Canada. *e-mail: albert.stolow@nrc.ca.

Imaging In Chemical Dynamics

Hersh Chandarana



Imaging In Chemical Dynamics:

Imaging in Chemical Dynamics Arthur G. Suits, 2001 This book provides a comprehensive review of the rapidly growing field of imaging based probes of chemical dynamics It includes discussions of state resolved photodissociation dynamics orbital alignment and vector correlations radical photodissociation surface scattering imaging photoelectron spectroscopy ultrafast dynamics and coincidence techniques An Introduction to Nonlinear Chemical Dynamics Irving R. Epstein, John A. Pojman, 1998-10-22 Just a few decades ago chemical oscillations were thought to be exotic reactions of only theoretical interest Now known to govern an array of physical and biological processes including the regulation of the heart these oscillations are being studied by a diverse group across the sciences This book is the first introduction to nonlinear chemical dynamics written specifically for chemists It covers oscillating reactions chaos and chemical pattern formation and includes numerous practical suggestions on reactor design data analysis and computer simulations Assuming only an undergraduate knowledge of chemistry the book is an ideal starting point for research in the field The book begins with a brief history of nonlinear chemical dynamics and a review of the basic mathematics and chemistry The authors then provide an extensive overview of nonlinear dynamics starting with the flow reactor and moving on to a detailed discussion of chemical oscillators Throughout the authors emphasize the chemical mechanistic basis for self organization The overview is followed by a series of chapters on more advanced topics including complex oscillations biological systems polymers interactions between fields and waves and Turing patterns Underscoring the hands on nature of the material the book concludes with a series of classroom tested demonstrations and experiments appropriate for an undergraduate laboratory 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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Dynamic Hyperpolarized Nuclear Magnetic Resonance Thomas Jue, Dirk Mayer, 2021-05-21 This is the first book in the series to focus on dynamic hyperpolarized nuclear magnetic resonance a burgeoning topic in biophysics The volume follows the format and style of the Handbook of Modern Biophysics series and expands on topics already discussed in previous volumes It builds a theoretical and experimental framework for students and researchers who wish to investigate the biophysics and biomedical application of dynamic hyperpolarized NMR All contributors are internationally recognized experts lead the dynamic hyperpolarized NMR field and have first hand knowledge of the chapter material The book covers the following topics Hyperpolarization by dissolution Dynamic Nuclear Polarization Design considerations for implementing a hyperpolarizer Chemical Shift Imaging with Dynamic Hyperpolarized NMR Signal Sampling Strategies in Dynamic Hyperpolarized NMR Kinetic Modeling of Enzymatic Reactions in Analyzing Hyperpolarized NMR Data Using Hyperpolarized NMR to Understand Biochemistry from Cells to Humans Innovating Metabolic Biomarkers for Hyperpolarized NMR New Insights into Metabolic Regulation from Hyperpolarized ^{13}C MRS MRI Studies Novel Views on Heart Function from Dynamic Hyperpolarized NMR Insights on Lactate Metabolism in Skeletal Muscle based on ^{13}C Dynamic Nuclear Polarization Studies About the Editors Dirk Mayer is Professor of Diagnostic Radiology and Nuclear Medicine at the University of Maryland and is the Director of Metabolic Imaging He is a recognized expert on dynamic nuclear polarization DNP MRI based imaging techniques and has optimized acquisition and reconstruction techniques has constructed kinetic modeling for quantitative analysis and has developing new probes Thomas Jue is Professor of Biochemistry and Molecular Medicine at the University of California Davis He is an internationally recognized expert in developing and applying magnetic resonance techniques to study animal as well as human physiology in vivo He served as a Chair of the Biophysics Graduate Group Program at UC Davis where he started to redesign a graduate curriculum that balances physical science mathematics formalism and biomedical perspective in order to promote interest at the interface of physical science engineering mathematics biology and

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 materials fluids and flow of increasing

complexity from different fields of Chemical Engineering before finishing off with a review of reactors and reactions The result allows engineers industrial and academic researchers and decision makers to gain a detailed insight into the NMR toolbox such that they can estimate the benefit of NMR imaging with regard to cost efficiency and scientific results

Understanding Light Microscopy Jeremy Sanderson, 2019-03-04 Introduces readers to the enlightening world of the modern light microscope There have been rapid advances in science and technology over the last decade and the light microscope together with the information that it gives about the image has changed too Yet the fundamental principles of setting up and using a microscope rests upon unchanging physical principles that have been understood for years This informative practical full colour guide fills the gap between specialised edited texts on detailed research topics and introductory books which concentrate on an optical approach to the light microscope It also provides comprehensive coverage of confocal microscopy which has revolutionised light microscopy over the last few decades Written to help the reader understand set up and use the often very expensive and complex modern research light microscope properly Understanding Light Microscopy keeps mathematical formulae to a minimum containing and explaining them within boxes in the text Chapters provide in depth coverage of basic microscope optics and design ergonomics illumination diffraction and image formation reflected light polarised light and fluorescence microscopy deconvolution TIRF microscopy FRAP super resolution techniques biological and materials specimen preparation and more Gives a didactic introduction to the light microscope Encourages readers to use advanced fluorescence and confocal microscopes within a research institute or core microscopy facility Features full colour illustrations and workable practical protocols Understanding Light Microscopy is intended for any scientist who wishes to understand and use a modern light microscope It is also ideal as supporting material for a formal taught course or for individual students to learn the key aspects of light microscopy through their own study

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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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