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TRIPLY DIFFERENTIAL STUDIES OF ATOMIC AND MOLECULAR PHOTOIONIZA--ETC(U)
JUL 82 J L DEHNER, A C PARR, R STOCKBAUER N00014-82-F-0011
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Basic studies of the dynamics and spectroscopy of atomic and molecular photo- ionization have been carried out using three experimental probes. The first and most extensively used experimental approach involves triply differential (differential in incident wavelength, electron energy, and ejection angle) photoelectron measurements using synchrotron radiation. Measurements were conducted in the vacuum ultraviolet wavelength range up to $h\nu \approx 35$ eV on a large variety of atomic and molecular systems. Photoelectron branching ratios		

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20. (contd). and angular distributions were obtained for all accessible states. A major emphasis of this work involved the initial exploration of novel effects of autoionization and shape resonances on alternative vibrational ionization channels. The second experimental approach entailed measuring the polarization of fluorescence following production of excited molecular ions by photoionization. This experiment allowed the direct measurement of the alignment of molecular ions produced by photoionization and, simultaneously, the branching ratios for degenerate photoelectron channels. The third experimental approach involved determining the photoelectron spectrum of an atomic cluster (Xe_3^+) in a mixture of clusters formed in a supersonic expansion by the technique of photoion-photoelectron coincidence.

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ANNUAL SUMMARY REPORT

Triply Differential Studies of Atomic and Molecular Photoionization
Using Synchrotron Radiation (Contract No. N00014-82-F-0011)

Submitted to

Office of Naval Research
Physical Sciences Division
Physics Program Office
Department of Navy
Arlington, VA 22217

Attn: Dr. Bobby R. Junker

Submitted by

National Bureau of Standards
19 July 1982

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2. Contract Description

The research covered by this contract involves basic studies of photoionization processes in atoms and molecules using three novel and/or advanced experimental approaches outlined in item 4. Topics of main interest include photoelectron branching ratios, photoelectron angular distributions, alignment of molecular ions by photoionization, and photoelectron spectra of atomic clusters.

3. Scientific Problem

This study is aimed at resolving roughly four unknown or incompletely known aspects of this problem area: First, this program seeks to characterize major aspects of photoionization dynamics, such as the effects of shape resonances and autoionizing resonances on alternative ionization channels, which can only now be studied in a definitive way with the advanced techniques employed in this work. Second, we seek to develop new probes of the phototization process, e.g., fluorescence polarization spectroscopy and photoelectron spectroscopy of atomic clusters, which will yield new types of information. Third, this project produces data crucial to testing theoretical predictions and, thus, contributes to the development of realistic theories of atomic and molecular photoionization. Fourth, the data produced by this project contributes to characterizing all the pathways by which radiation interacts with matter, and hence contributes to the macroscopic modeling of such interactions.

4. Scientific and Technical Approach

This program utilized three experimental approaches: First, the main effort involves measuring triply differential photoelectron cross sections using synchrotron radiation. Thus the intensity of photoelectrons ejected from atoms and molecules are measured as a function of three independent parameters -- the wavelength of the incident synchrotron radiation, the kinetic energy of the photoelectron, and the ejection angle relative to the polarization direction of the light. Second, the polarization of fluores-

cence from excited ionic states produced by photoionization is measured as a function of the wavelength of the incident light. Third, photoelectron spectra are measured in coincidence with the mass of the ion produced in order to obtain the photoelectron spectra of specific atomic clusters in the presence of a whole range of clusters formed in a supersonic expansion.

5. Progress

The scientific accomplishments of this program during the last contract period are reflected in the papers, abstracts of contributed talks, and invited lectures listed in Section 6. In particular, papers 5, 7, and 9 were previously submitted, but have appeared in this contract period. Papers 10-13 were submitted and appeared, and papers 14 and 15 were submitted. Two other manuscripts are in final draft stage. In addition, abstracts 16-21 and invited talks 8-19 were added during this contract period.

The highlights of the past year, not all of which are represented by the papers, abstracts, and talks, and be organized into six categories: First, paper 15 continues what has been a major theme of this program - effect of shape resonances on photoionization branching ratios and photoelectron angular distributions. This paper resolves a long standing controversy regarding the ordering of the valence levels in SF₆ and, using both experimental and theoretical evidence, shows the importance of channel interaction in the vicinity of a large shape-resonant feature. Second, papers 12-14 continue our unique series of triply differential studies as a function of position within autoionizing resonances. Third, we are presently conducting a second series of experiments on polarization of fluorescence from molecular photoions -- a technique we pioneered in the last period. Fourth, paper 11 is a major paper detailing our unique study of photoelectron-photoion studies of individual polyatomic clusters formed in a supersonic expansion. Fifth, we have completed the data analysis and are proceeding well toward writing up several more data sets involving triply differential photoionization studies of molecules. One significant study involves a new constant-photoelectron-energy approach for studying molecular photoionization. This involves setting the energy analyzer for a particular kinetic energy and scanning the incident wavelength. The scientific benefit of this technique will be explained in a paper to be published in the next period. Sixth, and most important for future work, we have devoted a major effort in last period to design, fabricate, and partially assemble a new generation double electron spectrometer system for triply differential photoionization studies. This system, when fully developed, should enhance our sensitivity/resolution by two to three orders of magnitude and qualitatively improve our scientific capability. This development has involved substantial investments by ONR, DOE via Argonne National Laboratory, and the National Bureau of Standards over the last two years and is expected to have a major impact on the field in future years.

6. Publications

The papers, abstracts of contributed talks, and invited talks are listed on the following pages.

PAPERS

1. B. E. Cole, D. L. Ederer, R. Stockbauer, K. Codling, A. C. Parr, J. B. West, E. D. Poliakoff, and J. L. Dehmer, "Wavelength and Vibrational-State Dependence of Photoelectron Angular Distributions. Resonance Effects in 5σ Photoionization of CO," J. Chem. Phys. 72, 6308 (1980).
2. A. C. Parr, D. L. Ederer, B. E. Cole, J. B. West, R. Stockbauer, K. Codling, and J. L. Dehmer, "Triply-Differential Photoelectron Studies of Molecular Autoionization Profiles," Phys. Rev. Letters 46, 22 (1981).
3. E. D. Poliakoff, J. L. Dehmer, D. Dill, A. C. Parr, K. H. Jackson, and R. N. Zare, "Polarization of Fluorescence Following Molecular Photoionization," Phys. Rev. Letters 46, 907 (1981).
4. K. Codling, A. C. Parr, D. L. Ederer, R. Stockbauer, J. B. West, B. E. Cole, and J. L. Dehmer, "The Effects of Autoionization on Vibrational Branching Ratios and Photoelectron Angular Distributions in Molecular Photoionization: The Formation of the Ground State of O_2^+ Between 574 Å and 600 Å." J. Phys. B 14, 657 (1981).
5. D. L. Ederer, A. C. Parr, B. E. Cole, R. Stockbauer, J. L. Dehmer, J. B. West, and K. Codling, "Vibrational-State Dependence of Partial Cross Sections and Photoelectron Angular Distributions through Autoionizing Resonances: The $n = 3$ Rydberg Level Converging to the $B^2\Sigma^+$ State of CO^+ ," Proc. Roy. Soc. Lond. A 378, 423 (1981).
6. A. C. Parr, G. Rakowsky, D. L. Ederer, R. L. Stockbauer, J. B. West, and J. L. Dehmer, "Current Research at NBS Using Synchrotron Radiation at SURF-II," IEEE Transactions on Nuclear Science, Volume NS28, 1210 (1981).
7. J. B. West, K. Codling, A. C. Parr, D. L. Ederer, B. E. Cole, R. Stockbauer, and J. L. Dehmer, "Partial Photoionization Cross Sections and Photoelectron Angular Distributions through the Hopfield Bands in N_2 Between 650 Å and 730 Å," J. Phys. B 14, 1791 (1981).
8. K. Codling, J. B. West, A. C. Parr, J. L. Dehmer, and R. L. Stockbauer, "Measurement of β Values and Branching Ratios in the Region of the $3s3p^64p^1P_1^0$ Resonance in Ar and the $5s5p^66p^1P_1^0$ Resonance in Xe," J. Phys. B 13, L693 (1980).
9. E. D. Poliakoff, P. M. Dehmer, J. L. Dehmer, and R. L. Stockbauer, "The Photoelectron Spectrum of Xe_3 by the Photoelectron-Photoion Coincidence Technique," J. Chem. Phys. 75, 1568 (1981).

PAPERS (CONTINUED)

10. D.M.P. Holland, A. C. Parr, D. L. Ederer, J. L. Dehmer, and J. B. West, "The Angular Distribution Parameters of Argon, Krypton, and Xenon for Use in Calibration of Electron Spectrometers," *Nucl. Instr. and Meth.* 195, 331 (1982).
11. E. D. Poliakoff, P. M. Dehmer, J. L. Dehmer, and R. Stockbauer, "Photoelectron-Photoion Coincidence Spectroscopy of Gas-Phase Clusters," *J. Chem. Phys.* 76, 5214 (1982).
12. A. C. Parr, D. L. Ederer, J. B. West, D.M.P. Holland, and J. L. Dehmer, "Triply Differential Photoelectron Studies of Non-Franck-Condon Behavior in the Photoionization of Acetylene," *J. Chem. Phys.* 76, 4349 (1982).
13. D. L. Ederer, A. C. Parr, J. B. West, D.M.P. Holland, and J. L. Dehmer, "Measurement of the Spin-Orbit Branching Ratios and the Angular Asymmetry Parameters in the Region of the $4s4p^65p$ Resonances in Krypton and the $5s5p^66p$ Resonances in Xenon," *Phys. Rev. A* 25, 2006 (1982).
14. A. C. Parr, D. L. Ederer, J. L. Dehmer, and D. M. P. Holland, "Characterization of Some Autoionizing Resonances in CO_2 Using Triply Differential Photoelectron Spectroscopy," *J. Chem. Phys.*, in press.
15. J. L. Dehmer, A. C. Parr, S. Wallace, and D. Dill, "Photoelectron Branching Ratios and Angular Distributions for the Valence Levels of SF_6 in the Range $16eV \leq h\nu \leq 30eV$," *Phys. Rev. A*, submitted for publication.

ABSTRACTS OF CONTRIBUTED PAPERS

1. A. C. Parr, J. L. Dehmer, B. E. Cole, D. L. Ederer, R. L. Stockbauer, and J. B. West, "An Angle-Resolved Photoelectron Spectrometer for Triply-Differential Photoionization Studies," Sixth International Conference on Vacuum Ultraviolet Radiation Physics, June 2-6, 1980, Charlottesville, VA. Book of Extended Abstracts, p. III-70.
2. K. Codling, J. B. West, A. C. Parr, J. L. Dehmer, B. E. Cole, D. L. Ederer, and R. L. Stockbauer, "Partial Cross Sections, Vibrational Branching Ratios, and Angular Distributions in the 570-600 Å Window Resonance in O₂," *ibid.*, p. II-14.
3. R. Stockbauer, A. C. Parr, J. L. Dehmer, B. E. Cole, D. L. Ederer, J. B. West, and K. Codling, "Perturbation of Vibrational Intensities and Angular Distributions by Autoionization of Molecular Photoionization," *ibid.*, p. II-15.
4. E. D. Poliakoff, J. L. Dehmer, A. C. Parr, D. Dill, K. H. Jackson, and R. N. Zare, "Polarized Fluorescence Excitation Spectroscopy of N₂," *ibid.*, p. II-25.
5. J. L. Dehmer, A. C. Parr, J. B. West, K. Codling, D. L. Ederer, B. E. Cole, E. D. Poliakoff, and R. Stockbauer, "Effects of Shape Resonances on Vibrational Branching Ratios and Photoelectron Angular Distributions in Molecular Photoionization," *ibid.*, p. II-86.
6. A. C. Parr, R. L. Stockbauer, K. Codling, J. B. West, and J. L. Dehmer, "Photoelectron Branching Ratios and Angular Distributions in the Region of the 3s3p⁶4p¹P₁^o Resonance in Ar and the 5s5p⁶6p¹P₁^o Resonance in Xe," Annual DEAP Meeting, 1-3 December 1980, Los Angeles, CA, *Bull. Am. Phys. Soc.* 25, 1131 (1980).
7. J. L. Dehmer, J. B. West, K. Codling, R. Stockbauer, A. C. Parr, D. L. Ederer, and B. E. Cole, "Triply-Differential Photoionization Studies of Molecular Autoionization Profiles," *ibid.*, p. 1136.
8. E. D. Poliakoff, J. L. Dehmer, D. Dill, A. C. Parr, K. H. Jackson, and R. N. Zare, "Polarization of Fluorescence Following Molecular Photoionization," *ibid.*, p. 1136.
9. K. Codling, J. B. West, A. C. Parr, J. L. Dehmer, and R. L. Stockbauer, "Photoelectron Angular Distribution Measurements through Autoionizing Resonances in Argon and Xenon," *Molecular Spectroscopy and Dynamics with Synchrotron Radiation-A European Workshop, Maria Laach, West Germany, 29 September-1 October 1980, Book of Abstracts, p. 52.*
10. R. Stockbauer, A. C. Parr, B. E. Cole, D. L. Ederer, J. Dehmer, J. West, and K. Codling, "Effects of Two-Electron Resonances on Photoelectron Energy and Angular Distributions," presented orally at the Gordon Research Conference on Electron Spectroscopy, Wolfsboro, NH, July 1980 (no abstract available).

ABSTRACTS OF CONTRIBUTED PAPERS (CONTINUED)

11. A. C. Parr, D. L. Ederer, R. Stockbauer, J. B. West, K. Codling, D.M.P. Holland, and J. L. Dehmer, "Triply-Differential Photoelectron Studies of Atomic and Molecular Photoionization," Twelfth International Conference on the Physics of Electronic and Atomic Collisions, 15-21 July 1981, Gatlinburg, Tennessee, Book of Abstracts.
12. E. D. Poliakoff, P. M. Dehmer, J. L. Dehmer, and R. Stockbauer, "The Photoelectron Spectrum of Xe₃ by the Photoelectron-Photoion Coincidence Technique," *ibid.*
13. E. D. Poliakoff, J. L. Dehmer, D. Dill, A. C. Parr, K. H. Jackson, and R. N. Zare, "Polarization of Fluorescence Following Molecular Photoionization," *ibid.*
14. A. C. Parr, D. L. Ederer, J. West, J. L. Dehmer, "Resonance Effects in the Angular Distribution and Branching Ratios of the Photoelectrons in C₂H₂ and C₂N₂," Annual Meeting of the American Society for Mass Spectrometry and Allied Topics, 25-29 May 1981, Minneapolis, Minn., Book of Abstracts.
15. D.M.P. Holland, A. C. Parr, D. Ederer, J. L. Dehmer, and J. B. West, "The Angular Distribution Parameters of Selected Rare Gases for Use in Calibration of Electron Spectrometers," National Synchrotron Instrumentation Conference, Cornell University, 15-17 July 1981, Book of Abstracts.
16. A. C. Parr, D. L. Ederer, R. Stockbauer, J. B. West, D.M.P. Holland, K. Codling, and J. L. Dehmer, "Triply Differential Photoelectron Studies of Autoionization and Shape-Resonance Effects in Molecular Photoionization," Annual DEAP Meeting, 3-5 December 1981, New York, New York, *Bull. Am. Phys. Soc.* 26, 1322 (1981).
17. E. D. Poliakoff, P. M. Dehmer, J. L. Dehmer, and R. Stockbauer, "Photoelectron-Photoion Coincidence Spectroscopy of Gas-Phase Clusters," *ibid.*, p. 1322.
18. A. C. Parr, D.M.P. Holland, D. L. Ederer, J. L. Dehmer, "Effects of Resonances in Molecular Photoionization Measured with Triply Differential Photoelectron Spectroscopy," Ninth International Conference on Mass Spectrometry, Vienna, Austria, July, 1982, Book of Abstracts.
19. A. C. Parr and H. M. Rosenstock, "Resonance, Autoionization, and Kinetic Effects in Photoionization," (invited talk) Annual Meeting of the American Society for Mass Spectrometry, 7-11 June 1982, Honolulu, Hawaii, Book of Abstracts.

ABSTRACTS OF CONTRIBUTED PAPERS (CONTINUED)

20. J. L. Dehmer, D. Dill, and A. C. Parr, "Molecular Photoionization Dynamics with Emphasis on Shape and Autoionizing Resonances," Abstract of Invited Lecture at the NATO Advanced Study Institute on Photophysics and Photochemistry in the Vacuum Ultraviolet, 15-28 August 1982, Lake Geneva, Wisconsin, Book of Abstracts.
21. A. C. Parr and J. L. Dehmer, "Photoelectron Spectrometer for High Resolution Angular Resolved Studies," International Conference on X-Ray and VUV Synchrotron Radiation Instrumentation, 9-13 August 1982, Hamburg, W. Germany, Book of Abstracts.

INVITED TALKS, COLLOQUIA, AND SEMINARS

1. J. L. Dehmer and Dan Dill, "Shape Resonances in Molecular Photoionization," Plenary talk presented at Molecular Spectroscopy and Dynamics with Synchrotron Radiation-A European Workshop, Maria Laach, West Germany, September 29-October 1, 1980, Book of Abstracts, p. 43.
2. A. C. Parr, "Current Research at NBS Using Synchrotron Radiation at SURF-II," Invited talk presented at the Sixth Conference on the Application of Accelerators in Research and Industry, Denton, TX, November 3-5, 1980. See paper 6.
3. J. L. Dehmer, "Potpourri of Current and Future Studies of Molecular Photoionization-Synchrotron Radiation, Supersonic Jets, and Multiphoton Ionization," Chemistry Department Colloquium, Boston University, Boston, MA, 13 April 1981.
4. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Photoelectron-Photoion Coincidence Spectroscopy of Atomic Clusters and Fluorescence Polarization Analysis," Atomic and Molecular Science Seminar, Argonne National Laboratory, Argonne, Illinois, 6 May 1981.
5. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," Molecular Spectroscopy Division Seminar, National Bureau of Standards, Gaithersburg, MD, 21 May 1981.
6. E. D. Poliakoff, "Alignment of Molecular Ions Produced by Photoionization," Seminar on Collision Experiments in Their Theoretical Frame (Fano Workshop), The University of Chicago, Chicago, IL, 23 May 1981.
7. A. C. Parr, "Status of Programs at NBS SURF-II," National Synchrotron Instrumentation Conference, Cornell University, 15-17 July 1981.
8. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," Physics Department Colloquium, Georgetown University, Washington, D.C., 6 November 1981.
9. A. C. Parr, "Triply Differential Photoelectron Spectrometry of Atoms and Simple Molecules," Center Colloquium, Center for Absolute Physical Quantities, National Bureau of Standards, January 6, 1982.
10. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Fluorescence Polarization and Photoelectron - Photoion Coincidence Studies of Clusters," Brookhaven National Laboratory, December 2, 1981.
11. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Fluorescence Polarization and Photoelectron - Photoion Coincidence Studies of Clusters," Exxon Research Laboratory, January, 1982.

INVITED TALKS, COLLOQUIA, AND SEMINARS (CONTINUED)

12. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Fluorescence Polarization and Photoelectron - Photoion Coincidence Studies of Clusters," Department of Chemistry, Dartmouth University, January, 1982.
13. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Fluorescence Polarization and Photoelectron - Photoion Coincidence Studies of Clusters," Department of Chemistry, Boston University, January 1982.
14. E. D. Poliakoff, "Two Novel Probes of Molecular Photoionization: Fluorescence Polarization and Photoelectron - Photoion Coincidence Studies of Clusters," Department of Chemistry, University of Pennsylvania, February, 1982.
15. A. C. Parr and H. M. Rosenstock, "Resonance, Autoionization, and Kinetic Effects in Photoionization," Invited talk presented at the 30th Annual Conf. on Mass Spectrometry and Allied Topics (American Society for Mass Spectrometry) Honolulu, June 6-11, 1982.
16. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," Department of Physics Colloquium, University of Alabama, March 31, 1982.
17. J. L. Dehmer, "Molecular Photoionization Dynamics - Progress and Prospects," Physics Colloquium, University of Chicago, May 27, 1982.
18. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," California Institute of Technology, June 14, 1982.
19. A. C. Parr, "Resonance Phenomena in Molecular Photoionization," University of California, Santa Barbara, June 15, 1982.

7. Extenuating Circumstances

None.

8. Unspent Funds

None will remain unspent at the end of the current contract period.

9. Graduate Students Receiving Degrees

None.

10. Other Federal Contract Support

During this period, J. L. Dehmer was a co-principal investigator for Office of Naval Research Contract N00014-81-F-0051, "Selectivity of Multiphoton Processes," 6/1/81 - 5/31/82. Renewal of this contract is currently pending.

During this period, R. Stockbauer was a co-principal investigator for Office of Naval Research Contract N00014-81-F-0021, "Characterization of Surface Bonding Using Photon and Electron Stimulated Desorption", 7/1/81-6/31/82. Renewal of this contract is currently pending.