TRIPPLY DIFFERENTIAL STUDIES OF ATOMIC AND MOLECULAR PHOTOIONIZATION (U)

JUL 82  J L DEMMER, A C PARR, R STOCKBAUER  N00014-82-F-0011

UNCLASSIFIED
**REPORT DOCUMENTATION PAGE**

**RECIPIENT'S CATALOG NUMBER**

**TITLE (and Subtitle)**
Triply Differential Studies of Atomic and Molecular Photoionization Using Synchrotron Radiation

**AUTHOR(s)**
J. L. Dehmer (Argonne National Laboratory)
A. C. Parr
R. Stockbauer

**PERFORMING ORGANIZATION NAME AND ADDRESS**
National Bureau of Standards
Washington, D.C. 20234

**REPORT DATE**
19 July 1982

**NUMBER OF PAGES**
11

**ABSTRACT**
Basic studies of the dynamics and spectroscopy of atomic and molecular photoionization 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 \( h\nu \approx 35 \text{ eV} \) on a large variety of atomic and molecular systems. Photoelectron branching ratios...

**Keywords**
Photoionization, atoms, molecules, branching ratio, photoelectron spectroscopy, photoelectron angular distribution, synchrotron radiation, autoionization, shape resonance, atomic clusters, photoelectron-photoion coincidence, fluorescence polarization.
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$_4$) in a mixture of clusters formed in a supersonic expansion by the technique of photoion-photoelectron coincidence.
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

Approved for public release; distribution unlimited.
Reproduction in whole or in part is permitted for any purpose
of the United States Government.
1. Principal Investigators:

J. L. Dehmer, Senior Physicist
Argonne National Laboratory
Argonne, IL 60439
Tel: 312-972-4194

Albert C. Parr
Synchrotron Ultraviolet Radiation Facility
National Bureau of Standards
Washington, D.C. 20234
Tel: 301-921-2031

Roger Stockbauer
Surface Science Division
National Bureau of Standards
Washington, D.C. 20234
Tel: 301-921-2096

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 photionization 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 fluora-
cence from excited ionic states produced by photolization 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 SF$_6$ 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-photolon 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


ABSTRACTS OF CONTRIBUTED PAPERS


ABSTRACTS OF CONTRIBUTED PAPERS (CONTINUED)


ABSTRACTS OF CONTRIBUTED PAPERS (CONTINUED)


INVITED TALKS, COLLOQUIA, AND SEMINARS


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.