Triply Differential Studies of Atomic and Molecular Photoionization Using Synchrotron Radiation

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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 hν ~ 35 eV on...
20 (contd). several atomic and molecular species, e.g., Ar, Kr, Xe, H2, N2, O2, CO, SO2, C2H2, C2N2, HCN, CH3CN, BF3, and SF6. Photoelectron branching ratios 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 photol-ionization. This was also a novel experiment, allowing the direct measurement of the alignment of molecular ions produced by photol-ionization and, simultaneously, the branching ratios for degenerate photoelectron channels. The third experimental approach was, again, a novel experiment, and involved determining the photoelectron spectrum of an atomic cluster (Xe3) in a mixture of clusters formed in a supersonic expansion by the technique of photol-ion-photoelectron coincidence.
SUMMARY QUESTIONNAIRE

Triply Differential Studies of Atomic and Molecular Photoionization
Using Synchrotron Radiation (Contract No. N00014-81-F-0005)

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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 photoionization 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 utilizes 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 fluorescence 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

Triply differential measurements were conducted in the vacuum ultraviolet wavelength range up to $h\nu \sim 35$ eV on several atomic and molecular species, e.g., Ar, Kr, Xe, H$_2$, N$_2$, O$_2$, CO, SO$_2$, C$_2$H$_2$, C$_2$N$_2$, HCN, CH$_3$CN, BF$_3$, and SF$_6$. Photoelectron branching ratios 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. Prototype fluorescence polarization spectroscopy measurements were performed on the $B^2\Sigma_u^+$ state of N$_2^+$ formed by photoionization of N$_2$ in the range $450 \, \AA < \lambda < 660 \, \AA$. The feasibility and theoretical interpretation of this new class of experimental probe was established. Finally, in the third part of the project, the photoelectron spectrum of Xe$_3$ was determined by photoelectron-photoion coincidence technique using a supersonic jet source of xenon clusters.

6. Publications

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


ABSTRACTS OF CONTRIBUTED PAPERS


INVITED TALKS


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**

