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MECHANISMS OF MULTIPHOTON DISSOCIATION OF MOLECULAR IONS.(U)
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The dynamics of multiphoton absorption and dissociation of polyatomic ions will be studied to characterize the details of the process. Ion fragments will be identified and the energy released into translation will be measured using a coaxial laser ion beam spectrometer.

Single photon absorption studies of positive and negative polyatomic ions failed to reveal any vibrational absorptions in the infrared region investigated. A single photon electronic transition absorption in $O_2^{(+)}$ was found which resulted in dissociation.

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SUMMARY QUESTIONNAIRE

ONR Contract N00014-76-C-1035
Mechanisms of Multiphoton Dissociation of Molecular Ions

SRI Project PYU 5808

1. Contract Description

The dynamical details of the dissociation of molecular ions occurring by multiphoton absorption will be studied. Fragment identities will be determined as well as the average energy released into translation.

2. Scientific Problem

The multiphoton dissociation (MPD) of neutral polyatomic molecules has been shown to be an isotopically selective process whose energetics are governed by statistical principles. These conclusions are based upon detailed studies of the dissociation dynamics in collision free molecular beams. Similar information for polyatomic ions is unavailable, so that the mechanisms of MPD in ionic species remain to be determined.

Multiphoton processes are finding applications in isotope separation schemes, frequency conversion techniques and laser assisted chemistry. Elucidation of the detailed mechanisms involved was essential to the understanding and application of MPD to these problems in neutral environments, and will be of similar importance for ionic environments. In particular, if MPD of ions were to be non-statistical, then it might be possible to control the outcome of mode-specific ion-molecule reactions. Such mode-selective reactions would find wide applications in chemical synthesis and analysis. At present, however, neither the products produced from MPD of ions are known, nor are the energetics leading to fragmentation.

3. Scientific and Technical Approach

Positive or negative ions are produced in an electron impact source, then accelerated to 3 keV and mass analyzed to form a highly collimated beam. An electrostatic (quadrupole) field bends the ions through 90° where they are merged along a 40-cm path with the output from a grating tuned CO_2 TEA laser. Laser fluences of 1-50 J/cm^2 are available using a variety of focusing optics. Ion fragments will be energy analyzed using a second quadrupole field. Simple conservation principles will establish the mass of these fragments from a knowledge of the monoenergetic parent ion identity. Additional higher resolution energy analysis of the fragment ions using a hemispherical sector will provide a measure of the energy released into translation during dissociation. The energy resolution of this arrangement ($\geq$ 50 meV) will allow an estimate to be made of the number of infrared photons absorbed beyond the dissociation limit. The coaxial beams spectrometer to be used has already demonstrated the necessary capabilities in studies of the visible photodissociation of molecular ions.

4. Progress

The previous contract period was devoted to a systematic search for infrared absorptions of simple molecular ions. These absorptions would provide information regarding the initial steps of multiphoton processes in ions. Using the same coaxial beams spectrometer, a CW CO_2 laser was employed as an excitation source. We attempted to identify the vibrational absorptions by their effect on either the visible photodissociation of positive ions or the visible photodetachment of negative ions. Despite a thorough search in a number of ions, no evidence was found for any such IR absorptions.

We did, however, observe IR absorption leading to the direct dissociation of highly vibrationally excited O_2^+ ions. This process was shown to be a single photon absorption involving an electronic transition.

5. Publications

No publications have yet resulted from this work.

6. Extenuating Circumstances

None

7. Personnel

The following SRI personnel have participated in this work during the past year: M. J. Coggiola, P. C. Cosby, J. T. Moseley, J.-B. Ozenne, and J. R. Peterson. Dr. Ozenne participated in this work while a visiting scientist at SRI, on leave from the University of Paris, Orsay, France.

8. Graduate Student Degrees

No graduate students participated in this work.