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13. ABSTRACT (Maximum 200 words)

The goal of this project is to learn about the electronic and vibrational spectroscopy of size-selected clusters. This is done using high resolution negative ion photodetachment techniques developed in our laboratory during the last four years. In our experiments, a beam of cluster anions (typically C_n^- or Si_n^-) is generated using a laser vaporization source, mass-selected, and photodetached with a fixed-frequency or tunable laser. In the fixed-frequency experiments, we measure the electron kinetic energy distribution, thereby obtaining the anion photoelectron spectrum. This enables us to map out the electronic and vibrational states of the neutral cluster with about 10 meV resolution. For higher (0.4 meV) resolution studies, we use a technique called threshold photodetachment spectroscopy. Here, the cluster anions are detached with a tunable laser, and only those photoelectrons produced with nearly zero kinetic energy are collected. In principle, this yields the same information as the photoelectron spectrum, but the considerably higher resolution allows us to make much more definite assignments of the spectral features.

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OFFICE OF NAVAL RESEARCH

FINAL REPORT

for

CONTRACT NUMBER N0014-87-K-0495

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THRESHOLD PHOTODETACHMENT SPECTROSCOPY

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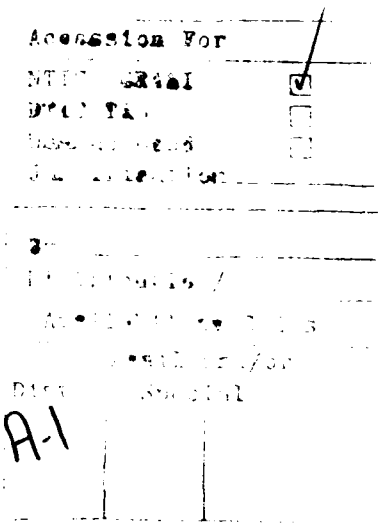
September 3, 1992



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Summary of Project:

The goal of this project was to learn about the electronic and vibrational spectroscopy of size-selected clusters. This is done using high resolution negative ion photodetachment techniques developed in our laboratory during the last four years. In our experiments, a beam of cluster anions (typically C_n^- or Si_n^-) is generated using a laser vaporization source, mass-selected, and photodetached with a fixed-frequency or tunable laser. In the fixed-frequency experiments, we measure the electron kinetic energy distribution, thereby obtaining the anion photoelectron spectrum. This enables us to map out the electronic and vibrational states of the neutral cluster with about 10 meV resolution. For higher (0.4 meV) resolution studies, we use a technique called threshold photodetachment spectroscopy. Here, the cluster anions are detached with a tunable laser, and only those photoelectrons produced with nearly zero kinetic energy are collected. In principle, this yields the same information as the photoelectron spectrum, but the considerably higher resolution allows us to make much more definite assignments of the spectral features.

We have made considerable progress in applying our combination of photodetachment techniques to silicon and carbon cluster anions. We have obtained vibrationally-resolved photoelectron spectra for Si_n^- (n = 2 - 4) and C_n^- (n = 2 - 11). These spectra represent the first observations of vibrationally resolved spectra for several of the polyatomic clusters. A comparison of the Si_4^- spectrum with *ab initio* calculations indicates that the ground states of Si_4 and Si_4^- have a planar rhombus structure. The C_n^- spectra are dominated by transitions between linear forms of the anion and neutral clusters for $n \leq 9$, but the C_{10}^- and C_{11}^- spectra are dominated by transitions between cyclic

clusters. We have also obtained threshold photodetachment spectra of Si_2^- and C_5^-. The Si_2^- spectrum allows us to definitively order the several low-lying electronic states of Si_2 and Si_2^-. The C_5^- spectrum yields frequencies for low-frequency vibrational modes in the anion and neutral as well as the spin-orbit splitting in the anion.

List of Publications/Technical Reports

1) T. N. Kitsopoulos, I. M. Waller, J. G. Loeser, and D. M. Neumark, "High Resolution Threshold Photodetachment Spectroscopy of Negative Ions," Chem. Phys. Lett. 159, 300 (1989); Technical Report #1.

2) D. M. Neumark, "Transition State Spectroscopy of Hydrogen Transfer Reactions," in Electronic and Atomic Collisions: Invited Papers of the XVI ICPEAC (AIP Conference Proceedings #205), edited by A. Dalgarno, R. S. Freund, P. Koch, M. S. Lubell, and T. B. Lucatorto (American Institute of Physics, New York: 1990), pp. 33-48; Technical Report #2.

3) I. M. Waller, T. N. Kitsopoulos, and D. M. Neumark, "Threshold Photodetachment Spectroscopy of the I + HI Transition State Region," J. Phys. Chem. 94, 2240 (1990); Technical Report #3.

4) T. N. Kitsopoulos, C. J. Chick, A. Weaver, and D. M. Neumark, "Vibrationally-Resolved Photoelectron Spectra of Si_3^- and Si_4^-," J. Chem. Phys. 93, 6108 (1990); Technical Report #4.

5) T. N. Kitsopoulos and D. M. Neumark, "Photoelectron Spectroscopy of Si_2^-," Materials Research Symposium Proceedings, Vol. 206 (1990 Fall Meeting), Materials Research Society (1991), p. 71; Technical Report #5.

6) T. N. Kitsopoulos, C. J. Chick, Y. Zhao, and D. M. Neumark, "Study of the Low-Lying Electronic States of Si_2 and Si_2^- Using Negative Ion Photodetachment Techniques," J. Chem. Phys. 95, 1441 (1991); Technical Report #6.

7) D. M. Neumark, "Negative Ion Photodetachment as a Probe of the Transition State Region: The I + HI Reaction," in Advances in Molecular Vibrations and Collision Dynamics, Vol. IA, ed. J. M. Bowman, (JAI Press, Greenwich, 1991) pp. 165-185; Technical Report #7.

8) D. W. Arnold, S. E. Bradforth, T. N. Kitsopoulos, and D. M. Neumark, "Vibrationally Resolved Spectra of C_2-C_{11} by Anion Photoelectron Spectroscopy," J. Chem. Phys. 95, 8753 (1991); Technical Report #8.

9) T. N. Kitsopoulos, C. J. Chick, Y. Zhao, and D. M. Neumark, "Threshold Photodetachment Spectroscopy of C_5^-," J. Chem. Phys. 95, 5479 (1991); Technical Report #9.

10) T. N. Kitsopoulos and D. M. Neumark, "Studying the Low Lying Electronic States of Small silison and Carbon Clusters Using Negative Ion Photodetachment Techniques," Proceedings of ONR Conference on Clusters, January 1990 (in press).

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