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<td>Dr. Cheuk-Yiu Ng</td>
<td>University of California, Davis</td>
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<td>Cheuk-Yiu Ng</td>
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<td>19b. TELEPHONE NUMBER (Include area code)</td>
<td>530-754-9645</td>
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INSTRUCTIONS FOR COMPLETING SF 298

1. REPORT DATE. Full publication date, including day, month, if available. Must cite at least the year and be Year 2000 compliant, e.g., 30-06-1998; xx-06-1998; xx-xx-1998.

2. REPORT TYPE. State the type of report, such as final, technical, interim, memorandum, master’s thesis, progress, quarterly, research, special, group study, etc.

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4. TITLE. Enter title and subtitle with volume number and part number, if applicable. On classified documents, enter the title classification in parentheses.

5a. CONTRACT NUMBER. Enter all contract numbers as they appear in the report, e.g. F33615-86-C-5169.

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FINAL AFOSR REPORT (Dec. 1, 2008-Nov. 30, 2011)

I. Grant Title:

Vacuum Ultraviolet Laser Probe of Chemical Dynamics of Aerospace Relevance

II. Principal Investigator and Address

Cheuk-Yiu Ng

Address: Department of Chemistry
          University of California at Davis
          Davis, California 95616

III. Contract Number: FA9550-09-1-0054
IV. Objective:

A primary goal of this research program is to provide pertinent information about the energetics, photochemistry, and chemical dynamics of spacecraft effluents, including H₂O, NH₃, hydrazine (N₂H₄), monomethylhydrazine, and unsymmetrical dimethylhydrazine. By using the unique VUV laser facilities developed in our laboratory, we plan to perform high-resolution photodissociation, photoionization, and photoelectron studies of these molecules, and to examine their chemical interactions with major atmospheric ions, aiming to identify possible sources of optical emission signatures that can be used for reliable tracking and detection of spacecraft maneuvers and rocket launches. Rovibrationally excited O₂⁺(X^2Π_{3/2,1/2}; v⁺), O₂⁺(a^4Π_u; v⁺) and N₂⁺(X^2Σ_g^+; v⁺) ions are known to be among the major ionic species produced by solar VUV photoionization and electron impact ionization. These excited O₂⁺ and N₂⁺ species are long-lived and expected to play a significant role in atmospheric reactions. Nevertheless, reaction cross sections involving these rovibrational excited ions with atmospheric neutrals remain mostly unknown. We have successfully developed a novel, high-resolution VUV laser pulsed field ionization-photoion (PFI-PI) scheme for the preparation of rovibrationally selected O₂⁺(X^2Π_{3/2,1/2}; v⁺=0-38, N⁺), O₂⁺(a^4Π_u; v⁺=18, N⁺), and N₂⁺(X^2Σ_g^+; v⁺=9, N⁺) ions with high intensities and high laboratory kinetic energy resolutions (ΔE_{lab} = 50 meV), allowing the measurement of reactions cross sections down to thermal energies. We proposed to conduct detailed absolute total cross section measurements for reactions between these state-selected diatomic ions and atmospheric neutrals, including atomic neutrals, Ar, O(^3P), and O(^1D), diatomic neutrals, H₂, O₂, N₂, NO, and CO, and polyatomic neutrals, HCN, H₂O, CO₂, NH₃, CH₄, C₂H₂, N₂H₄ and C₂H₄. Particular emphasis will be placed on the reaction dynamics involving reactant ions in highly vibrationally excited states. These state-resolved absolute total cross sections obtained as a function of kinetic energy are valuable for modeling aerospace environments using the Direct Simulation Monte Carlo and Particles in Cell methods. Environments of interest to the Air Force include nonequilibrium, hypersonic air plasma, such as those associated with communications blackout of reentry space vehicles and the plasmas of electric propulsion thrusters. They are also important for accurate simulation of density profiles of electron, ions, and neutrals in planetary atmospheres.

V. Technical developments

1. Technique for the preparation of State-selected ions

By employing an electric field pulsing scheme for vacuum ultraviolet (VUV) laser pulsed field ionization-photoion (PFI-PI) measurements, we have been able to prepare a rovibrationally selected PFI-PI beam of N₂⁺(v⁺=1, N⁺) with not only high intensity and high quantum state purity, but also high kinetic energy resolution. This represents a technical breakthrough for the preparation of state-selected ions for ion-molecule reaction studies. This work has just been published as a communication in the Journal of Chemical Physics [see Chang et al., “Communication: Rovibrationally selected study of the N₂⁺(X; v⁺ = 1, N⁺ = 0-8) + Ar charge transfer reaction using the vacuum ultraviolet laser pulsed field ionization-photoion method”, J. Chem. Phys. 134, 201105 (2011)].
Using the $\text{N}_2^+(X^2\Sigma_g^+; v^+) + \text{Ar}$ charge transfer process as a demonstration reaction, we show that the low $\Delta E_{\text{lab}}$ spread for the rovibronically selected $\text{N}_2^+$ beam achieved here allows the cross section measurements to be extended down to thermal energies. We compare in Fig. 4 the charge transfer cross sections for $\text{N}_2^+(v^+=1, N^+=0-8)$ at $E_{\text{cm}}=0.04-10.00$ eV obtained in this study with previous theoretical and experimental results, which include cross sections converted from reported rate coefficients. The previous experimental and theoretical results are scattered and are mostly higher than the present measurements. However, the cross section curve observed here is in excellent agreement with the theoretical predictions based on the Landau-Zener-Stückelberg formulism and the $ab\ initial$ potential energy surface calculated for the $[\text{N}_2+\text{Ar}]^+$ reaction system.

In addition to achieving the high kinetic energy resolution, the internal energy selection by the VUV laser PFI-PI method has made possible the examination of the rotational effect on the charge transfer cross section.

**Figure 1.** Comparison of $\sigma(v^+=1, N^+=0-8)$ values measured at $E_{\text{cm}}=0.04-10.00$ eV, obtained here with previous theoretical predictions and experimental results, which include cross sections converted from rate constant measurements.

After this successful demonstration experiment, we are currently working on the ion-molecule reactions $\text{N}_2^+(v^+=0-2, N^+ = 0-8) + \text{CH}_4 (\text{H}_2\text{O}, \text{and} \text{C}_2\text{H}_4)$. Interesting preliminary results on the vibrational effect for the reaction cross sections of these processes have been obtained.

2. **VUV photodissociation by VUV photoionization probe**

We have demonstrated that the VUV photodissociation dynamics of $\text{N}_2$ and $\text{CO}_2$ can be studied using VUV photoionization with time-sliced velocity-
mapped ion imaging (VUV-PI-VMI) detection. This represents the first experimentao study employing two tunable VUV lasers. This work has just published as a communication in Journal of Chemical Physics [see Pan et al., “Communication: Vacuum ultraviolet laser photodissociation studies of small molecules by the vacuum ultraviolet laser photoionization time-sliced velocity-mapped ion imaging method”, J. Chem. Phys. 135, 071101 (2011)].

In the case of CO₂, we show for the first time that O(^1D) atom is produced with CO(^1Σ⁺) at 92.21 nm as depicted in Fig. 2.

Figure 2. Photodissociation of CO₂ at 108451.5 cm⁻¹. Oxygen atoms in the O(^1D₂) state are detected via the autoionization transition 2s²2p⁵(2D³/₂,3/₂)3d^1D₂ ← O(^1D₂), (a) The sliced VUV-PI-VMI-image and (b) the P(Eₚ) derived from the image.
Currently, the ion-imaging apparatus is used for the VUV-photodissociation and VUV-photoionization studies of H₂O, NH₃, and N₂H₄.

3. **High-resolution near VMI-TPE detection**

We have recently implemented the VUV laser VMI-photoelectron imaging technique with the ion-imaging apparatus by adding appropriate µ-metal shields to the velocity-mapped ion-imaging apparatus. We found that this method can provide an energy resolution close to that achieved in VUV-PFI-PE measurements if the VUV photoionization energy is set near the photoelectron band of interest.

Further to this development, we have successfully established the VUV laser VMI-threshold photoelectron imaging method for high-resolution TPE measurements, achieving an electron energy resolution of 2 cm⁻¹ (FWHM). The TPE measurements are made by gating the TPE signal imaged at the center of the imaging detector. Figure 3 compares the VUV-TPE imaging (VUV-VMI-TPE) spectrum (upper curve) thus obtained with the VUV-PFI-PE spectrum (lower curve) of C₆H₅Cl in the energy range of 9.06-9.48 eV. The energy resolution for the VUV-PFI-PE measurement has been measured to be 1.5-2.0 cm⁻¹ (FWHM). Since the FWHMs of all the VUV-VMI-TPE bands are nearly identical to those of the corresponding VUV-PFI-PE bands, we conclude that the energy resolution achieved in this VUV-TPEI measurement is 2 cm⁻¹ (FWHM).

**Figure 3.** Comparison of the VUV-TPEI spectrum (upper spectrum) with the VUV-PFI-PE spectrum (lower spectrum), showing that the resolution achieved for the VUV-TPEI measurement is close to 2 cm⁻¹ (FWHM) observed in VUV-PFI-PE measurements.

VI. **Selected scientific findings**


The ionization energy (IE) of NiC and the 0 K bond dissociation energies (D₀) and heats of formation at 0 K (ΔH°₀) and 298 K (ΔH°₂₉₈) for NiC and NiC⁺ are predicted by the wavefunction based CCSDTQ(Full)/CBS approach and the multi-reference configuration interaction (MRCI) method with Davidson correction (MRCI+Q). The CCSDTQ(Full)/CBS calculations presented here
involve the approximation to the complete basis set (CBS) limit at the coupled cluster level up to full quadruple excitations along with the zero-point vibrational energy (ZPVE), high-order correlation (HOC), core-valence electronic (CV), spin-orbit coupling (SO), and scalar relativistic effect (SR) corrections. The present calculations provide the correct symmetry predictions for the ground states of NiC and NiC\(^+\) to be \(^1\Sigma^+\) and \(^2\Sigma^+\), respectively. The CCSDTQ(Full)/CBS IE(NiC) = 8.355 eV is found to compare favorably with the experimental IE value of 8.37209 ± 0.00006 eV. The predicted IE(NiC\(^+\)) value at the MRCl+Q/cc-pwCV5Z level, including the ZPVE, SO, and SR effects is 8.00 eV, which is 0.37 eV lower than the experimental value. This work together with the previous experimental and theoretical investigations supports the conclusion that the CCSDTQ(Full)/CBS method is capable of providing reliable IE predictions for 3d-transition metal carbides, such as FeC and NiC. Furthermore, the CCSDTQ(Full)/CBS calculations give the prediction of \(D_0(\text{Ni}^-\text{C}) - D_0(\text{Ni}^+\text{C}) = 0.687\) eV, which is also consistent with the experimental determination of 0.73221 ± 0.00006 eV, whereas the MRCl+Q calculations predict a significantly lower value of 0.39 eV for \(D_0(\text{Ni}^-\text{C}) - D_0(\text{Ni}^+\text{C})\). The analysis of the correction terms shows that the CV and valence-valence electronic correlations beyond CCSD(T) wavefunction and the relativistic effect make significant contributions to the calculated thermochemical properties of NiC/NiC\(^+\). For the experimental \(D_0\) and \(\Delta H^\circ_{\text{f0}}\) values of NiC/NiC\(^+\), which are not known experimentally, we recommend the CCSDTQ(Full)/CBS predictions \[D_0(\text{Ni}^-\text{C}) = 4.047\) eV, \(D_0(\text{Ni}^+\text{C}) = 3.361\) eV, \(\Delta H^\circ_{\text{f0}}(\text{NiC}) = 749.0\) kJ/mol and \(\Delta H^\circ_{\text{f0}}(\text{NiC}^+) = 1555.1\) kJ/mol].

2. 

Vacuum ultraviolet (VUV) laser pulsed field ionization-photoelectron (PFI-PE) spectroscopy has been applied to the study of the sulfur monoxide radical (SO) prepared by using a supersonically cooled radical beam source based on the 193 nm excimer laser photodissociation of SO\(_2\). The vibronic VUV-PFI-PE bands for the photoionization transitions SO\(^+\)(\(X_2^2\Pi_{1/2}; v^+ = 0\)) ← SO(\(X^3\Sigma^−; v = 0\)) and SO\(^+\)(\(\tilde{X}_3^2\Sigma^−; v^+ = 0\)) ← SO(\(X^3\Sigma^−; v = 0\)) have been recorded. On the basis of the semi-empirical simulation of rotational branch contours observed in these PFI-PE bands, we have obtained highly precise ionization energies (IEs) of 83 034.2 ± 1.7 cm\(^{-1}\) (10.2949 ± 0.0002 eV) and 83 400.4 ± 1.7 cm\(^{-1}\) (10.3403 ± 0.0002 eV) for the formation of SO\(^+\)(\(X_2^2\Pi_{1/2}; v^+ = 0\)) and SO\(^+\)(\(\tilde{X}_3^2\Sigma^−; v^+ = 0\)), respectively. The present VUV-PFI-PE measurement has enabled the direct determination of the spin-orbit coupling constant (\(A_0\)) for SO\(^+\)(\(X_2^2\Pi_{1/2,3/2}\)) to be 365.36 ± 0.12 cm\(^{-1}\). We have also performed high-level ab initio quantum chemical calculations using the wavefunction based CCSDTQ(Full)/CBS approach, which involves the approximation to the complete basis set (CBS) limit at the coupled cluster level up to full quadruple excitations. The zero-point vibrational energy correction, the core-valence electronic correction, the spin-orbit coupling, and the high-level correction are included in the calculation. The IE[SO\(^+\)(\(X_2^2\Pi_{1/2,3/2}\))] and \(A_0\)
predictions thus obtained are found to be in remarkable agreement with the experimental determinations.

VII. Personnel Supported:

1. Professor Cheuk-Yiu Ng: Principal investigator
2. Dr. Yih-Chung Ian Chang: Postdoctoral associate
3. Mr. Yuntao Xu: Graduate student (4th year student)
4. Mr. Zhou Lu: Graduate student (2nd year student)

VIII. Publications (2009-present):

5. Kai-Chung Lau, Yih-Chung Chang, Chow-Sheng Lam, and C. Y. Ng, “High-level ab initio predictions of the ionization energy, bond dissociation energies and heats of formations for Iron carbide (FeC) and its cation (FeC⁺)”, J. Phys. Chem. A (invited), 113, 14321 (2009).


IX. Interactions/Transitions:

A. Invited talks at workshops, conferences, and seminars (2009-present)


3. C. Y. Ng, “Role of Chinese Scientists in Advocating Science Research in China”, *Public Lecture* sponsored by the 2008/2009 Siu Lien Wong Visiting Fellow


8. C. Y. Ng, “Spectroscopy, energetics, and reaction dynamics of ions and neutrals by high-resolution VUV photoion-photoionization methods”, Cluster Research Laboratory, Toyota Technological Institute in East Tokyo Laboratory, Japan, June 5, 2009.

9. C. Y. Ng, “Spectroscopy, energetics, and reaction dynamics of ions and neutrals by high-resolution VUV photoion-photoionization methods”, Kyoto University, June 8, 2009.


16. “Spectroscopy and Dynamics of Neutrals and Ions by High-Resolution IR-VUV Laser Photoion-Photoelectron Methods”, Symposium on 25 years of ZEKE


29. C. Y. Ng, “The Role of Basic (Chemical) Research in Modern Society”, Public Lecture, sponsored by Hong Kong Royal Society of Chemistry Lecture, the Chinese University of Hong Kong, Hong Kong, Dec. 2, 2010.


34. C. Y. Ng, “High resolution photoionization and photoelectron studies of radicals”, Airlie Conference Center, DOE Contractor’s Meeting, Warrenton, Virginia, May 31-June 3, 2011.
36. C. Y. Ng, “Spectroscopy and Dynamics of Ions and Neutrals by High-Resolution IR-VUV Two-Color Photoion-Photoelectron Methods”, Department of Chemistry, National Tsing-Hua University, Taiwan, Nov. 9, 2011.
38. C. Y. Ng, “Absolute state-selected total cross sections for ion-molecule reactions of relevance to the ion chemistry of Titan: N₂⁺ (X²Σ⁺; v⁺ = 0-2, N⁺=0-9) + CH₄ (D₂ H₂O, C₂H₂, and C₂H₄)”, “Titan Workshop”, Miami Beach, March 11-14, 2012.

B. Interactions
This project represents a collaborative project between our group at UC Davis and Dr. Y.-H. Chiu, Dr. Dale J. Levandier, James Dodd at the Hanscom Air Force Research laboratory. Dr. Albert Viggiano of the Hanscom Air Force Research Laboratory is also interest to collaborate with us to measure the vibrational energy distributions of product O₂⁺, NO⁺, and N₂⁺ formed in the reactions of O⁺ + O₂ (N₂), which are the most reactions occurring in planetary atmospheres.

C. Transitions
The thermochemical data obtained by our group based on VUV photoion and photoelectron measurements have been used by the “Active Thermochemical Table” project (project leader: Dr. Branko Ruscic) of the Argonne National Laboraotry. We have recently extended these measurements to include transition metal species. Currently, the energetic predictions for transition metal-containing molecules are poor. Our measurements are expected to be valuable in guiding the further development of ab initio quantum calculation procedures.

X. New Discoveries:
XI. Honors/Awards:

**Lifetime achievement honors**
- 2009  Siu-Lien Wong Visiting Fellow, Chung Chi College, The Chinese University of Hong Kong
- 2005  Elected Fellow, American Association for Advancement of Science
- 2003  Distinguished Professor, UC Davis
- 1998  Alexander von Humboldt Senior Scientist Award
- 1997  Senior Fellow, Japanese Society for the Promotion of Science
- 1996  Distinguished Professor of Liberal Arts and Sciences, Iowa State University
- 1994  Iowa Regents Award for Faculty Excellence
- 1993  Elected Fellow, American Physical Society
- 1985  Honorary Professor of Chemistry, Zhengzhou University, China
- 1982  Camille and Henry Dreyfus Teacher-Scholar
- 1981  Alfred P. Sloan Foundation Fellow