

SRI International
Molecular Physics Laboratory
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This report covers the period from October 1999 through September 2001. Additional information is available at the web site <http://www.sri.com/psd/physics>.

1. INTRODUCTION

SRI International, founded as Stanford Research Institute in 1946, is an independent nonprofit organization conducting research for government and commercial clients worldwide. Of SRI's 1400 employees, 374 hold Ph.D. or equivalent degrees. Many are principal investigators on basic research grants from NASA, NSF, NIH, DOE, or DOD.

The Molecular Physics Laboratory conducts research in many areas, including telescope observations of terrestrial and planetary atmospheres, and laboratory experimental and theoretical investigations of atmospheric chemical processes, combustion mechanisms, non-linear optics, biomedical optical diagnostics, and new techniques for trace species detection. The astronomical and atmospheric research activities are briefly summarized below.

2. PERSONNEL

2.1 Senior Research Staff

Principal investigators during the period were Michael J. Coggiola, Richard A. Copeland, Philip C. Cosby, David R. Crosley, Gregory W. Faris, David M. Golden, David L. Huestis, Laura Iraci (now at NASA Ames), Konstantinos S. Kalogerakis, Jochen Marschall, Harald Oser, James R. Peterson (emeritus), Tom G. Slanger, Felix T. Smith (emeritus), and Gregory P. Smith.

2.2 Postdoctoral Fellows and Visiting Scientists

The Molecular Physics Laboratory has an active postdoctoral research program. Graduate students interested in a postdoctoral position should contact the Laboratory Director at richard.copeland@sri.com. Postdoctoral Fellows during the period were Gabriel Amaral, Samantha Ashbourn, James E. Boulter, Seonkyung Lee, and Julie-Ann Reppel. K. S. Kalogerakis was promoted from Postdoctoral Fellow to become a regular staff member. Visiting scientists were Murthy Gudipati, Jorge Luque-Sanchez, Chung Park, and Robert Robertson.

2.3 Undergraduate Students

Each year since 1993 the Molecular Physics Laboratory has hosted 8-10 summer students through a Research Experiences for Undergraduates (REU) program funded by the NSF Physics Division. Undergraduate students are encouraged to apply through the web site <http://www.sri.com/psd/reu>.

2.4 Collaborations and Other Professional Activities

International collaborations are an important part of our research activities. Collaborators during the period included Kurt S.J. Anderson (New Mexico State U.), Thomas A. Barlow (Caltech), Thomas A. Bida (Lowell Obs.), Gert D. Billing (U. Copenhagen), A. Lyle Broadfoot (U. Arizona), Nancy J. Chanover (New Mexico State U.), David Crisp (JPL), Stephen T. Gibson (Aus. Nat. U.), Marshall Ginter (U. Maryland), Hanspeter Helm (U. Freiburg), A. Peet Hickman (Lehigh U.), Jeffrey R. Kuhn (U. Hawaii), Mats Larsson (U. Stockholm), Brenton R. Lewis (Aus. Nat. U.), Heidi L.K. Manning (Concordia C.), Victoria Meadows (JPL), Jeff S. Morrill (NRL), Donald E. Osterbrock (Lick Obs.), C. Wesley Walter (Denison U.), J.-H. (Sam) Yee (APL/Johns Hopkins U.), and Wim J. van der Zande (FOM/AMOLF Amsterdam).

Huestis leads the AAS/DPS Community Panel on Planetary Atmospheres in support of the NAS/NRC Decadal Survey of NASA Solar System Exploration programs. Golden serves on the NASA Panel for Evaluation of Chemical Kinetics and Photochemical Data for Use in Stratospheric Modeling.

3. RESEARCH

3.1 Telescope Observations

Slanger, Huestis, and Cosby: *Observations of the Venus nightglow with the Keck and APO telescopes*. Optical emissions from the Venus night atmosphere were first observed in 1975 by the visible-wavelength spectrometers on the Venera 9 and 10 orbiters. The emission features were subsequently assigned to excited states of molecular oxygen, which was a surprise because the Venus atmosphere was known to consist predominantly of carbon dioxide. A similar surprise resulted from the 1978 observations by the Pioneer Venus ultraviolet spectrometer that were subsequently assigned to emissions from excited states of nitric oxide. Contemporaneous ground-based telescope observations in the infrared near 1.27 microns showed additional intense emissions from molecular oxygen. These findings have been confirmed by subsequent ground-based studies. In 1999 we performed the first high-resolution ground-based observations in the visible, which confirmed the molecular oxygen emissions observed by the Venera orbiters, but surprisingly found strong emission from atomic oxygen that should have been conspicuous in the Venera spectra. Follow-up visible-wavelength observations at the Apache Point Observatory in 2001 again found molecular oxygen emissions but no evidence of atomic oxygen emission, suggesting that additional observations will be needed to understand this variability.

Slanger, Cosby, and Huestis: *Nightglow studies with astronomical sky spectra*. In order for ground-based astronomers to obtain accurate spectra of cosmological objects, it is necessary for them to separately measure spectra of the blank sky, because atmospheric emissions represent an interference

for their measurements. These so-called sky spectra, obtained with broad spectral coverage and high resolution, are an outstanding source of nightglow information on the terrestrial atmosphere. Since 1997, sky spectra have been obtained from astronomers using the 10-meter Keck telescopes on Mauna Kea. There is now a substantial list of atmospheric emissions seen for the first time. Numerous improvements have also been made to literature data. The new observations include (1) a broad range of vibrational transitions in the O₂ Atmospheric band system, (2) identification of the potassium D1 line, (3) a large number of atomic oxygen Rydberg emissions throughout the visible and near-IR region, (4) identification of emission from high rotational levels of vibrationally excited OH and emission from OH($v = 10$), (5) ionospheric emission of the O₂ Atmospheric bands, and (6) the identification of O₂ emissions throughout the 400-600 nm region, the decay of ionospheric emissions throughout the night, apparent atmospheric emissions from rare gases, and twilight glows.

Huestis, Cosby, and Slanger: *National Virtual Aeronomical Observatory (NVAO)*. As described above, astronomers' echelle spectrographs are already recording the best available survey spectra of optical emissions from excited atoms and molecules in the Earth's night atmosphere. These spectra would have tremendous value to atmospheric scientists if collected, processed, and presented in a manner suitable for subsequent analysis. With appropriate access and inquiry tools, students and professional scientists could perform "observations" on the "real atmosphere" from their desktops, either as educational exercises, publishable research, or "dry run" experiments before taking the field. SRI is creating a National Virtual Aeronomical Observatory as a component of the overall National Virtual Observatory. Astronomers will benefit from better understanding of the origins and expected strengths of emissions from terrestrial and extraterrestrial sources. The availability of broad-coverage spectra for every hour of every night at numerous locations would represent an unimagined and revolutionary resource for atmospheric science that could be assembled with modest incremental effort and cost. One dataset is available at <http://www-mpl.sri.com/NVAO/download/Osterbrock.html>

3.2 Laboratory and Theoretical Studies

Copeland, Huestis, Kalogerakis, Marschall, and Slanger: *Oxygen atom recombination and O₂ collisional relaxation*. In the atmospheres of the Earth and Venus, oxygen atoms are generated by the photodissociation of O₂ and CO₂ respectively. The three-body recombination of these oxygen atoms results in the formation of a multitude of electronically and internally excited states of O₂. On Earth, emission from these electronic states exhibits a peak at approximately 100 km and is part of the naturally occurring chemiluminescence of the atmosphere called nightglow. Since all the low-lying states of molecular oxygen are metastable, their radiative lifetimes are long enough that even at very high altitudes (pressure about 0.001 mbar at 100 km), collisions play a dominant role in determining the fate of these excited states. An understanding of the energy transfer rate constants and pathways is critical for elucidating the details of these atmo-

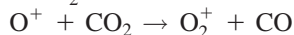
spherically important phenomena and for developing the ability to model them. Our laboratory has developed experimental methods to study collisional processes involving electronically excited O₂ molecules under temperature conditions relevant to the upper atmosphere (150-200 K). The recombination process produces oxygen molecules near the dissociation limit with significant electronic and vibrational excitation. In our experiments, we use direct laser excitation to a highly vibrationally excited level of the O₂(A³Σ_u⁺) state to access a similar energy regime. A second laser pulse with variable time delay probes the excited or neighboring levels to extract the relevant pathways, relative yields, and rates as the energy flows through numerous intermediate excited states. Our laboratory has developed REMPI detection schemes for many of the electronic states of molecular oxygen, including the A³Σ_u⁺, A³Δ_u, c¹Σ_u⁻, ⁵Π_g states, and high vibrational levels of the a¹Δ_g and b¹Σ_g⁺ states. Our studies have shown that excitation to high vibrational levels of the O₂(A³Σ_u⁺) state leads to population of several electronic states of O₂. Rates and branching ratios have been measured for vibrational energy transfer in the O₂(A³Σ_u⁺) and O₂(c¹Σ_u⁻) states. An interesting resonant electronic energy transfer path leads to population of highly vibrationally excited levels of the a¹Δ_g and b¹Σ_g⁺ states ($v' = 10-15$ and $v' = 16-19$ respectively). We have also generated O₂ in the previously unobserved but theoretically predicted ⁵Π_g state (important in recombination) and studied its collisional rate constants versus temperature.

Copeland, Kalogerakis, Marschall: *OH vibrational relaxation*. We are performing laboratory experiments needed to make quantitative inferences about atmospheric chemistry from the intensities of the atmospheric OH Meinel bands. Vibrationally excited hydroxyl radicals ($v = 6-9$) are generated in the 80 to 100 km altitude range of the Earth's atmosphere by reaction of atomic hydrogen with ozone. Lower vibrational levels ($v = 0-5$) are populated from higher vibrational levels via collisions with molecular oxygen. These lower vibrational levels are removed by collisions with atomic oxygen.

Copeland: *N₂ spectroscopy and collisional relaxation*. The excited electronic states a¹Π and A³Σ_u⁺ of N₂ play key roles in modeling aurora and airglows. Precise data on the formation, relaxation dynamics, and collisional energy transfer processes involving these states of N₂ are crucial to appropriate modeling. Current models still show some deviations from the observed data. It is the aim of our work in collaboration with Prof. Marshall Ginter and Dr. Murthy Gudipati of U. Maryland and with Dr. Jeff Morrill of NRL to measure accurate laboratory data on selectively prepared rovibrational states of the above-mentioned electronic states. These data will provide valuable input to atmospheric models. Typical experiments involve two-photon excitation of N₂ from the ground-state to the a-state, followed by a 1+1 resonance-enhanced ionization of N₂ from upper electronic states. Novel methods for vibrational state specific preparation of the A-state are being examined. Future experiments will study the excited molecules generated in collisions with N₂ in specific vibrational levels of the A-state.

Cosby, Huestis, and Peterson: *O₂⁺ dissociative recombina-*

tion. In the ionospheres of Venus and Mars, O_2^+ is the most abundant molecular ion, followed by CO_2^+ and NO^+ . As shown by previous laboratory work at SRI, more than 60% of the O_2^+ formed in the reaction



is vibrationally excited. This is expected to have a significant effect on the distribution of atomic oxygen excited states produced in dissociative recombination with electrons. We are measuring the specific product yields and absolute cross sections for dissociative recombination of O_2^+ in excited vibrational levels at the CRYRING heavy-ion storage ring facility in Stockholm, using a high-resolution detector system developed at AMOLF in Amsterdam and an ion source characterized and developed in our laboratory at SRI.

Crosley: *Laser fluorescence measurements of atmospheric species.* Measurements of atmospheric trace gases are an important key to understanding the photochemistry of the troposphere. Extremely low levels, on the order of a part per trillion and less, are often involved. Laser-induced fluorescence (LIF) instruments that directly detect OH and NO have flown or will soon fly on Global Tropospheric Experiment Missions. Combined with photolysis or chemical conversion of the parent species, LIF of these two molecules can also be used to measure HO_2 , NO_2 , and HONO. Proper, quantitative application of the LIF technique requires laboratory measurements of the radiative and collisional properties of the radicals involved. These processes govern the relationship, for the measurement conditions, between the observed signal and the concentration of the species involved. We are performing supporting studies pertinent to NO and NO_2 detection by making laboratory measurements on the NO molecule. We are measuring quenching of the $D^2\Sigma^+$ state at reduced temperature, rotational energy transfer in the $A^2\Sigma^+$ and $D^2\Sigma^+$ excited states, vibrational energy transfer in the ground $X^2\Pi$ state, and the final ground state rovibrational level distribution following collisional quenching of the $A^2\Sigma^+$ state.

Huestis: *Charge transfer collisions in ionospheres and exospheres.* The ionization potentials of $O(^3P_1)$ and H atoms are equal to within the current experimental uncertainty. This means that the charge transfer reactions



have very large cross sections in both directions, and thus play central roles in interactions with the solar wind and atomic escape in the exospheres of Venus, Earth, and Mars. In collaboration with Prof. A. Peet Hickman (Lehigh U.) we are performing quantum mechanical coupled channels calculations, including spin-orbit coupling, of the differential and momentum transfer cross sections needed for exospheric modeling.

Marschall: *Oxygen atom chemistry on mesospheric ice surfaces.* We are exploring the interactions of atomic oxygen with ice surfaces at temperatures relevant to the high-latitude summertime mesosphere. The specific experimental objectives are to quantify the loss rate of atomic oxygen on ice surfaces and to investigate atomic oxygen effects on the heterogeneous formation and growth rates of ice. Oxygen atom recombination is studied using a Knudsen cell reactor, together with laser-induced fluorescence and mass spectro-

scopic species detection. Atomic oxygen effects on ice formation and growth are investigated using a cryogenic test chamber, Fourier-transform infrared (FTIR) spectroscopy, and optical thin-film measurement techniques. The overall goal of this work is to provide experimental information to identify and clarify possible roles of mesospheric atomic oxygen in heterogeneous atmospheric chemistry and ice dynamics. The results of this project will aid in interpreting measurements of oxygen-atom and ice-particle distributions in the high-latitude summertime mesosphere and will provide insight for constructing numerical models for mesospheric phenomena such as noctilucent clouds (NLCs) and polar mesospheric summertime echoes (PMSEs).

G. P. Smith: *Chemical reactions of O, OH, and HO_2 in the terrestrial mesosphere.* The photochemistry of the Earth's mesosphere and upper stratosphere is controlled by the local reactions of odd oxygen and HO_x species. Models of this atmospheric region predict ozone, OH, and HO_2 concentrations that disagree with recent satellite and balloon measurements. To help resolve this discrepancy, we are performing laser photolysis experiments to measure the rates of the reactions $O + OH$, $O + HO_2$, and $OH + HO_2$ between 220 K and 300 K.

G. P. Smith and Huestis: *Giant planet hydrocarbon photochemistry— $CH_3 + CH_3$ recombination.* Planetary emissions of the methyl radical CH_3 were observed for the first time in 1998 on Saturn and Neptune by the ISO (Infrared Space Observatory) mission satellite. CH_3 is produced by VUV photolysis of CH_4 and is the key photochemical intermediate leading complex organic molecules on the giant planets and moons. The CH_3 emissions from Saturn were found to be about 10 times weaker than predicted by current models. The suggested remedy is to increase the rate of the recombination reaction



at 140 K to a value at least 10 times that measured experimentally at room temperature in rare gases. We are performing laboratory experiments at low temperature and very low pressure, below 0.01 mbar. The experiments are supported by RRKM theoretical modeling that is calibrated using the extensive combustion literature.

F. T. Smith: *Hyperbolic Poincare group and relativistic kinematics in hyperbolic space.* A new hyperbolic Poincare kinematic symmetry group has been developed that is applicable in an expanding, hyperbolic, homogeneous relativistic geometry, and reduces to the ten-parameter Poincare group of flat space-time. It reveals new aspects of particle kinematics in hyperbolic space and permits proving the covariance of the Dirac equation for n-body systems both in hyperbolic space and in its flat-space limit. This hyperbolic Poincare group has thirteen parameters, and is the direct product of two Lorentz subgroups and a translational subgroup in proper time. One Lorentz subgroup is generated by velocity boosts and operates in velocity space, the other is generated by hyperbolic translations and operates in position space. This structure expresses a fundamental kinematic symmetry between hyperbolic position space and relativistic velocity space, which persists in the flat-space limit and may be broken by gravitational inhomogeneities. The group unifies the

Hubble effect, describing the response of a velocity vector to spatial translation, with the effect of the Lorentz transformation, describing the response of a position vector to a velocity boost, as converse operations in a larger structure. These two operations have the characteristic alternation in sign with respect to each other that is also seen in Hamilton's equations of dynamics.

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