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ATOMIC AND MOLECULAR PROCESSES

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Pittsburgh University

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PROJECT SCIENTIST: M. A. Biondi, Director
Professor of Physics

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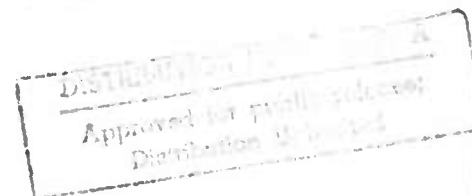
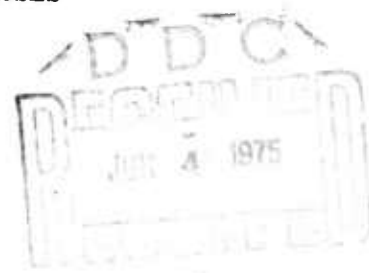
TITLE OF WORK: ATOMIC AND MOLECULAR PROCESSES

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April 30, 1975



Pittsburgh Atomic Sciences Institute

Faculty Members:

J. N. Bardsley
M. A. Biondi
W. L. Fite
F. Kaufman
E. C. Zipf

Post-Doctoral Personnel:

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E. Graham IV
H. H. Lo
R. L. Myers
T. L. Patterson

Graduate Research Assistants:

T. Barlak
J. S. Chang
Y. P. Chong
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Visiting Faculty:

None

TECHNICAL PROGRESS REPORT NO. 18

Pittsburgh Atomic Sciences Institute

Physics and Chemistry Departments

University of Pittsburgh

Pittsburgh, Pennsylvania

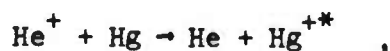
I. Summary of Research

The following research projects are identified by title, senior investigator(s) in charge of the work, and the general program to which they belong. This summary only covers advances during the period since our last progress report (dated October 30, 1974).

A. Laser Studies

1. Laser Ion-Molecule Reaction Rates (M. A. Biondi)

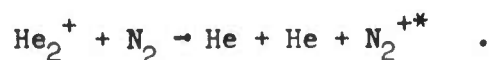
During the report period, the drift tube-mass spectrometer - optical spectrometer apparatus was used to study excited state production by non-resonant charge transfer. The fast atomic species transfer reaction,



was first investigated to determine the final state(s) in which the mercury ions are produced. The rate coefficient had been accurately determined earlier (Johnsen, Leu and Biondi, Phys. Rev. A8, 1808, 1973) and found to be very fast, $k > 10^{-9} \text{ cm}^3/\text{sec}$, at ion energies of interest for laser plasmas, 0.04 - 0.25 eV.

The current studies have indicated that the majority of the mercury ions produced by charge transfer from helium are formed in the $7p \text{ } ^2P_{3/2}$ excited state, which results in strong emission of $6149\text{\AA}$ radiation. Studies are currently in progress concerning the vibrational level populations in

excited molecular ions produced by the molecular species charge transfer reaction



Our initial findings are that the N_2^+ ions are produced principally in the $\text{B } ^2\Sigma_u^+$ state, with higher vibrational levels weakly populated. This charge transfer is evidently the principal inversion mechanism in a laser currently under investigation by Collins' group in Texas.

2. Metal Atom Chemi-excitation (F. Kaufman, M. A. Biondi)

The computer program which generates theoretical line shapes for the sodium resonance lines was improved further by incorporating all six hyperfine components in convolution with the instrument function consisting of the Airy, plate defect, and aperture functions. Calculated half-widths for Na, based on the finesse obtained by He-Ne laser calibration at 632.8 nm, range from 0.115 cm^{-1} at 600°K to 0.134 cm^{-1} at 1400°K . Experimental Na half-widths in fluorescence excited in the chemiluminescence cell by a greatly broadened Na vapor lamp gave a width of 0.122 cm^{-1} , corresponding to a temperature of about 900°K even though the actual temperature of the cell was 520°K . This large difference may be due to either of two causes: (1) A change in the reflectivity of the Fabry-Perot plates over the more than 40 nm wavelength range; or (2) the persistence of the broadened line shape of the excitation lamp due to geometry effects, i.e. the spreading fan of the poorly collimated incident beam, only roughly perpendicular to the optical axis of the interferometer, which favors absorption and reemission in the wings of the resonance line by fast Na atoms. The fluorescence line shape experiment is about to be repeated with excitation by a sodium microwave discharge lamp in 1 torr He carrier gas whose line shape is expected to be much narrower than that of the vapor lamp.

Several series of chemiluminescence experiments were run with Na excita-

tion by "active oxygen" coming from a microwave discharge in pure O_2 at 2 to 4 torr mixed with Na at concentrations of 10^{11} to 10^{12} cm^{-3} in argon carrier gas. Bright emission was observed and the Doppler temperature was $1200^\circ K$ based on the He-Ne laser finesse measurement at 632.8 nm or $800^\circ K$ based on the measured Na fluorescence line width. As the mixing region is about 30 to 50 msec downstream from the discharge, only O-atoms, $O_2(^1\Delta_g)$, and excited species arising from O-atom processes need to be considered. O_3 is expected to be present in very small concentration, because it is rapidly redissociated by $O_2(^1\Delta_g)$ which is known to be a major product of such discharges.

A new NO_2 addition and titration system was added to the apparatus which permits the wide variation of $[O]$ by partial or total titration with measured amounts of NO_2 . In a first series of experiments, $[O]$ was held constant and $[Na]$ was changed at constant total flow and pressure. The emitted intensity was seen to rise non-linearly with $[Na]$, and apparently dropped to zero at finite $[Na]$. When this threshold concentration, possibly an artefact of the experiment, is subtracted, a roughly quadratic dependence of I on $[Na]$ is obtained.

The NO_2 titration experiments showed (a) that atomic oxygen (or its products) is a necessary reactant, i.e. complete titration of O with NO_2 , which leaves the $O_2(^1\Delta_g)$ concentration virtually unchanged, quenches the emission totally; and (b) the chemiluminescent intensity is directly proportional to $[O]$.

These results suggest fairly strongly that, under our experimental conditions, the reaction $Na_2 + O \rightarrow NaO + Na^*$ is responsible for the emission. This would put a lower limit of about 67 kcal/mole (2.9 eV) on the Na-O bond strength whose value is listed as 65 ± 10 kcal/mole in the JANAF tables. The small exothermicity of the reaction would also be compatible with the

rather small translational energy release shown in our interferometer experiments. Attempts will now be made to examine the temperature dependence a constant $[Na]$ which should be a negative one, i.e. lower I for higher T if Na_2 is directly involved. Spectral information will also be obtained, as the present mechanism rules out the formation of more highly excited states of Na .

3. Production of Excited States by Dissociative Electron-Ion Recombination and Dissociative Excitation (M. A. Biondi and E. C. Zipf)

A microwave heating-microwave afterglow apparatus employing grating spectrometric and Fabry-Perot interferometric techniques to study the afterglow radiation has just been put into operation for studies of dissociative recombination production of atomic excited states. Initial investigations are of a well-documented dissociative recombination reaction, $Ne_2^+ + e \rightarrow Ne^* + Ne$, for which the total electron capture rate and its dependence on electron temperature (energy) have previously been determined. At present there is no information concerning relative production of various excited states as a function of electron energy. Our initial investigations are aimed at determining the change in excited state production rate with electron temperature and comparing it with theoretical models of the direct dissociative capture process for simple molecular ions.

We have completed a major modification to a second recombination apparatus that will permit a direct measurement of the vibrational and rotational temperatures of the molecular ions in our plasma through laser photoluminescent spectroscopy. A 1 megawatt tuneable dye laser is now in operation in our laboratory. We are currently testing and optimizing the composite laser-plasma system while we continue with our studies of rare-gas excimers using the laser to measure the concentration of the metastable atoms involved in the excimer formation.

4. and 5. Time-of Flight Experiments Involving the Production and Collisional Depopulation of Metastable Species (E. C. Zipf)

During December (1974) we conducted an extensive series of experiments at the Plum Brook vacuum chamber facility operated by NASA which exploited the remarkable size of this chamber (a cylinder 120' H x 100' D) and the enormous pumping speed provided by thirty-six 48" diffusion pumps. These time-of-flight experiments were designed to study the excitation of a number of metastable species with possible laser applicability. These included $N_2(A^3\Sigma_u^+)$, $O(^1S)$, $N(^2D)$, $N(^2P)$ as well as rare gas metastable atoms. Both chemiluminescent and surface detector TOF data were obtained. Analysis of these results will lead to cross section values as a function of velocity and a clearer understanding of molecular relaxation processes in an environment where the degree of collisional interactions could be carefully controlled and documented.

6. Theoretical Studies - Laser Physics (J. N. Bardsley)

Our technique for computing symmetric ion-atom interactions has been written up, and will be published shortly in the Physical Review. Ms. Sinha is completing the calculations of the resonant charge transfer cross sections for alkali atoms. Studies of the cross section for charge transfer of U^+ are underway.

The Monte-Carlo method has been applied in calculations of the velocity distributions of H^- and O^- in drift tubes with helium as the buffer gas. The distributions obtained for O^- ions was used in an analysis of the drift tube measurements of the rate of the charge transfer process $O^- + O_2 \rightarrow O + O_2^-$, and the cross section for this reaction was deduced. Similar calculations are in progress for O^+ ions with the aim of interpreting recent drift-tube studies of $O^+ - N_2$ and $O^+ - O_2$ reactions.

Studies of resonant rotational excitation have been initiated in order to assess the importance of rotational excitation as a source of heat in a molecular plasma. The theory is being applied to N_2 , O_2 and CO, the last one being of most current interest for laser applications.

B. Atmospheric Radiation Backgrounds

7. Particulate Technology (W. L. Fite)

I. Laboratory Experimentation

A. Detection of natural airborne particulates. Using a simple detector operating on the pyrolysis plus surface ionization principles, we have done extensive studies on particulate matter in ambient air. The instrument used operated at near atmospheric pressure and therefore was limited to (a) having a platinum filament and (b) having no electron multiplication. It has been established that such an instrument can detect particulate matter down to about 0.1 microns in diameter before being limited by circuit noise. To go any further seems to require electron multiplication which implies that high vacuum will have to be used. This simple device has been calibrated against other instruments, e.g., a Thermal Systems, Inc. Total Mass Monitor, and it appears that particulate concentrations in the range of 10 micograms/ m^3 can readily be detected, using a small sampling rate (30 cc/sec).

B. Water droplet effect. The possibility of using two filaments in a given particulate detector, one being made of platinum, which does not respond to water droplets, and the other being made of tungsten which has substantial alkali impurity in it and which does release alkali ions when water droplets strike it, is an attractive one in that both solid particulate matter and water droplets could perhaps be monitored simultaneously but separately in the same instrument. An experiment has been set up to study the water droplet effect on tungsten. It uses electrosprayed water droplets

introduced through a height expansion into high vacuum wherein the velocity and charge on "a droplet" are determined by a Faraday induction cage. Analysis in a transverse electric field determines the quantity q/mv^2 , where q and m are the charge and mass on the droplet. From these quantities m is directly determined without interference with the droplet, when it then strikes the hot surface and liberates ions. Although the apparatus is not quite fully operating in a quantitative sense, progress has been satisfactory and it is anticipated that good quantitative information will shortly be forthcoming.

II. Field Experiment Preparation

The device described in I-A, above has been scaled down in size, suitable circuitry has been procured and laboratory tests have been made on most of the hardware to be mounted on a balloon-parachute experiment, which we anticipate will be made piggy-back fashion in August with the cooperation of Dr. J. G. Anderson of the University of Michigan. We have no reason to believe that the instrument will perform any differently than the laboratory instrument described above. This means that with approximately 5 mm of 0.003" wire and a parachute velocity of the order of 2×10^4 cm/sec, the active region of the detector will sweep out a volume of about $80 \text{ cm}^3/\text{sec}$. Particles 0.1 micron in diameter at a concentration of 1 per cm^3 would give, after consideration of detection efficiency, in the range of 1-10 counts/sec. This corresponds to dust concentrations at the level of about 10^{-3} micrograms/cubic meter. This is 4 or 5 orders of magnitude less than concentrations near the surface of the earth. Since it is possible that the stratosphere has at least this dust concentration, we are optimistic that the detector now under construction will yield some interesting results in its use on the parachute experiment.

8. Radiative Properties of Simple Molecules and Atoms for Optical Discrimination Studies (E. C. Zipf)

We are nearing completion of a major modification to the electron scattering apparatus that we have been using to measure absolute excitation cross sections. The changes will improve the overall sensitivity of the apparatus so that studies of Rydberg atoms and molecules as well as other intermediate, pseudo-metastable states can be carried out expeditiously. The emphasis continues to be two-fold: (1) providing detailed absolute cross section data for the electron-impact excitation of potential lasing states and (2) garnering insights into the quantum mechanics of dissociative excitation and a measurement of specific product yields. The actual experiments will resume shortly upon the completion of our calibration work.

C. Infrared Early Warning

9. Infrared Emission Resulting from Fuel-Oxidizer Reactions (F. Kaufman)

Excellent progress can be reported in our infrared chemiluminescence studies of vibrational excitation and relaxation. In our greatly improved apparatus, a background-limited InSb detector is mounted in a liquid nitrogen cooled enclosure which also houses interchangeable circularly variable filters, tuning fork chopper and first stage preamplifier components. The reaction flow tube is viewed through a low emissivity window and the field of view ends in a liquid nitrogen dewar. Calibration with a black body has indicated a $D_{\lambda \text{ max}}^*$ of about $10^{13} \text{ cm Hz}^{1/2} \text{ watt}^{-1}$, about two orders of magnitude larger than that of commercial detectors.

In a further improvement the flow tube was connected to our large Roots blower through a 2 inch ball valve so that linear flow velocities up to 10^4 cm/sec are routinely attainable in the 3.8 cm i.d. flow tube. A new, movable mixing loop injector was fabricated and tested by viewing the uniformity of visible light emission due to the $\text{O} + \text{NO}$ chemiluminescence when NO in

He was added to discharged He-O₂ mixtures. It was found necessary to add a relatively large fraction of the flow (~ 5%) through the injector but this brought about uniform mixing within about 2-3 cm (0.2-0.3 msec). Another advantage of the much faster flows is the larger available range of modulation frequencies for the microwave discharge in order to avoid the difficult problem of large signal drifts at infrared wavelengths beyond about 3 μ m due to variable background emission (such as from slight warming of the windows). At flow velocities of 10⁴ cm/sec the discharge is easily modulated at frequencies up to 10 Hz without interference from axial diffusion effects in the flowing gas. Such double modulation using two lock-in amplifiers eliminates problems due to thermal drift.

Measurements of initial vibrational energy release in the reactions H + Cl₂, H + NOCl, and H + NO₂, i.e. production of HCl[‡] and OH[‡], were repeated in the new apparatus with the highly encouraging results that virtually unrelaxed distributions were observed in spite of the high total pressure (~ 1 torr) because of the high detectivity. Thus, comparing our HCl[‡] from H + NOCl with Nazar, Polanyi, and Skrlac's recent results at 10⁻⁵ to 10⁻⁴ torr pressure, the following relative concentrations of HCl^v with v = 3 to 8 are reported, normalized to v = 5, and giving our value first and Polanyi's second: v = 3 - 0.58, 0.82; v = 4 - 0.93, 0.96; v = 5 - 1.0, 1.0; v = 6 - 0.53, 0.48; v = 7 - 0.18, 0.10; and v = 8 - 0.03, 0.00. This is made possible by keeping the concentrations of the reacting species, H and NOCl, as low as 1 x 10¹² cm⁻³ and thereby eliminating homogeneous relaxation processes except those with He which are fast for rotation but very slow for vibration. Our apparatus thus makes possible, for the first time, the systematic study of vibrational relaxation processes by adding quencher molecules through movable injectors to a known manifold of vibrationally excited species.

Other successful experiments include measurement of $\text{OH}^\ddagger$ and $\text{NO}^\ddagger$ from the $\text{H} + \text{NO}_2$ reaction. Using recently published, reliable information on the A-values of the OH vibration-rotation transitions, the much discussed total extent of vibrational excitation of $\text{OH}^\ddagger$ appears to be in the range 10 to 15%, in good agreement with as yet unpublished dye laser experiments but not with beam and low pressure chemiluminescence results. A wide field of important and as yet inaccessible processes is thus being opened to experimental measurement and theoretical interpretation.

II. Publications and Technical Presentations

A. Publications

Complex Coordinate Studies of Resonant Electron-Atom Scattering, R. A. Bain, J. N. Bardsley, B. R. Junker and C. V. Sukumar, J. Phys. B7, 2189, (1974).

Calculations of the Positions and Residues of Regge Poles, C. V. Sukumar and J. N. Bardsley, J. Phys. B. 8, 568 (1975).

Regge Poles and the Watson-Sommerfeld Transformation in Low-Energy Atomic Collisions, J. N. Bardsley, J. Phys. B. 8, 577 (1975).

Associative Ionization of Ti, Zr, Gd, and Th in Collisions with O and O₂, H. H. Lo and W. L. Fite, Chem. Phys. Letters 29, 39 (1974).

Electrical Detection of Airborne Particulates Using Surface Ionization Techniques, R. L. Myers and W. L. Fite, Environmental Science and Technology 9, No. 4, 334 (1975).

Detection of Radicals and Excited Molecules in Dynamic Systems, W. L. Fite, International Journal of Mass Spectrometer and Ion Physics, 16, 109 (1975).

An Automatic Real-Time Detector and Sizer for Submicron Airborne Particulate Matter, R. L. Myers, W. L. Fite and T. M. Barlak, submitted to the Review of Scientific Instruments, 1975.

Kinetics of the Reaction of OH with HCl, M. S. Zahniser, F. Kaufman and J. G. Anderson, Chem. Phys. Letters 27, 507 (1974).

Kinetics of the Reactions of N₂H⁺, N₄⁺, and N₃⁺ with H₂O in the Gas Phase, V. M. Bierbaum and F. Kaufman, J. Chem. Phys. 61, 3804 (1974).

Vibrational Emission of NO₂ from the Reaction of NO with O₃, M. F. Golde and F. Kaufman, Chem. Phys. Letters 29, 480 (1974).

Kinetics of the Reaction OH + HNO₃ → H₂O + NO₃, J. J. Margitan, F. Kaufman and J. G. Anderson, to be published in International J. of Chem. Kinetics.

Hydrogen Chemistry: Perspectives on Experiment and Theory, F. Kaufman, in Physics and Chemistry of Atmospheres, B. M. McCormac, Ed.

Gas Phase Hydrogen Atom Recombination, L. P. Walkauskas and F. Kaufman, 15th International Combustion Symposium, The Combustion Institute, September 1974.

Reevaluation of the BEBO Method, R. M. Jordan and F. Kaufman, submitted to J. Chem. Phys., 1975.

Non-Thermal Rotational Distribution of CO(A¹Π) Fragments Produced by Dissociative Excitation of CO₂ by Electron Impact, M. J. Mumma, E. J. Stone and E. C. Zipf, J. Geophys. Res. 80, No. 1, 161, 1975.

The Dissociation of Molecular Nitrogen by Electron Impact, A. C. Zipf, and R. W. McLaughlin, J. Geophys. Res., 1975.

On the Excitation of the $N_2(b' \ ^1\Sigma_u^+ \rightarrow X^1\Sigma_g^+)$ System of N_2 by Electron Impact, R. W. McLaughlin and E. C. Zipf, J. Chem. Phys., 1975.

On the Dissociative Excitation of Ammonia by Electron Impact, B. L. Carnahan, T. G. Finn and E. C. Zipf, J. Chem. Phys., 1975.

Evidence for Vibrationally Excited CO_2^+ Ions in the Cytherean Ionosphere, G. L. Unger and E. C. Zipf, J. Geophys. Res., 1975.

The Dissociative Excitation of H_2 , HD, and D_2 by Electron Impact, B. L. Carnahan and E. C. Zipf, Phys. Rev. Letters, 1975.

Energy Spectra of Long-Lived Excited Nitrogen Atoms Produced by Electron-Impact Dissociation of N_2 , Ninth International Conference on the Physics of Electronic and Atomic Collisions, Seattle, Washington, 1975.

B. Technical Presentations

"Applications of Pseudopotentials in Atomic and Molecular Physics", J. N. Bardsley, Theoretical Seminar at Joint Institute for Laboratory Astrophysics, Boulder, January 1975.

"Vibrational and Rotational Excitation of Homonuclear Diatomic Molecules", J. N. Bardsley, Theoretical Seminar at Joint Institute for Laboratory Astrophysics, Boulder, February 1975.

"Dissociative Processes in Electron-Molecule Collisions", J. N. Bardsley, invited paper at the Gaseous Electronics Conference, Houston, October 1974.

"Monte Carlo Simulation of the Drift of H^- Ion in He", S. L. Lin and J. N. Bardsley, contributed paper at the Gaseous Electronics Conference, Houston, October 1974.

"Complex Analysis for Real Experiments", J. N. Bardsley, Colloquium at Kansas State University, October 1974.

"Resonant Charge Transfer", J. N. Bardsley, theoretical seminar at the Joint Institute for Laboratory Astrophysics, Boulder, November 1974.

"Interactions Between Ions and Their Parent Atoms", J. N. Bardsley, B. R. Junker and S. Sinha, contributed paper at the American Physics Society (DEAP) meeting, December 1974.

"Symmetric Charge Transfer in Low Energy Ion-Atom Collisions, J. N. Bardsley, Colloquium at University of Manchester, March 1975.

"Associative Ionization of Uranium and Other Metallic Atoms with Oxygen", colloquium talk by W. L. Fite, Illinois Institute of Technology, February 1975.

"Associative Ionization and Possible Applications to Uranium Isotope Separation", W. L. Fite, lecture, Lawrence Livermore Laboratories, March 18, 1975.

"Hydrogen Atom Recombination", F. Kaufman, seminar at University of Maryland, College Park, Maryland, November 7, 1974.

"Laboratory Studies of Stratospheric OH and Cl Reactions", J. G. Anderson, J. J. Margitan, M. S. Zahniser, and F. Kaufman, paper presented at the 4th Conference on the Climatic Impact Assessment Program, Cambridge, Massachusetts, February 6, 1975.

"NO₂ Fluorescence and O-NO Chemiluminescence", F. Kaufman, seminar at Florida State University, Tallahassee, Florida, February 13, 1975.

"Stratospheric Ozone, the SST, and Laboratory Studies of OH Reactions", F. Kaufman, colloquium at Florida State University, Tallahassee, Florida, February 4, 1975.

"Evidence for Vibrationally Excited CO₂⁺ Ions in the Ionosphere of Venus", G. L. Unger and E. C. Zipf, Trans. Am. Geophys. Union 55, XXXX, 1974.

"Energy Spectra of Long-Lived Excited Nitrogen Atoms Produced by Electron-Impact Dissociation of N₂", W. C. Wells, W. L. Borst and E. C. Zipf, Ninth International Conference on the Physics of Electronic and Atomic Collisions, Seattle, Washington, 1975.

C. Other Activities Relating to ARPA

M. A. Biondi and F. Kaufman served on the Review Committee of the ARPA Visible Laser Program Review conducted January 21-22, 1975 at the Naval Research Laboratories and at ARPA in Arlington, Virginia on January 23, 1975.

F. Kaufman attended meetings of the Climatic Impact Committee of the National Academy of Sciences in Washington, D. C. on November 1, 2 and 27, 1974.

E. C. Zipf launched three rockets into magnetically active aurora above Fort Churchill, Manitoba during January - February 1975.

E. C. Zipf attended the final planning meetings for the Project Antiquari rocket expedition to be conducted from a launch site south of Lima, Peru during May 1975.

E. C. Zipf attended the preliminary planning meetings for a series of midlatitude sounding rocket experiments aimed at measuring important minor constituents in the upper atmosphere (stratosphere and above).

III. Visiting Scientists

M. Ackerman, Institut d' Aeronomie Spatiale de Belgique

J. Bauerle, Westinghouse Research Laboratories

D. R. Herschbach, Harvard University

G. Herzberg, National Research Council, Ottawa, Canada

W. C. Lineburger, University of Colorado

E. Mueller, Pennsylvania State University

J. P. Toennies, Director, Max Planck Institut fur Stromungsforschung,
Gottingen, Germany

IV. Degrees Awarded

Ms. V. M. Bierbaum, Ph.D., December 1974.

Senior InvestigatorEst. Funds Expended and Committed (Thousands)

J. N. Bardsley	7.0
M. A. Biondi	42.4
T. M. Donahue	8.9
W. L. Fite	15.1
F. Kaufman	33.2
E. C. Zipf	<u>10.7</u>
Total Expended and Committed	117.3
Available Funds	<u>179.5</u>
Estimated Funds Remaining as of 3/31/74	62.2

University Accounting of Funds

Expended as of 3/31/74	111.8
Available Funds	<u>179.5</u>
Remaining Funds as of 3/31/74	67.7