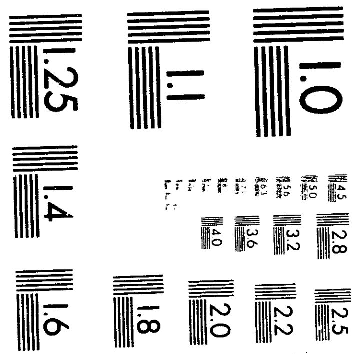

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Margaret M. Graff
Georgia Institute of Technology
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Performance report 1990

The object of this research is to study reactivity at superthermal collision energies using a fast neutral beam that is generated by photodetachment. Systems scheduled for initial study include basic oxygen-hydrogen reactions. Unfortunately, we can not yet report realization of this goal, but during this funding period we have made advances that are anticipated to lead to successful measurements during the next year. The parameters described below refer to the model system $O+H_2 \rightarrow OH+H$.

The basic design involves the collision of fast neutrals, created by photodetachment of the corresponding negative molecular ion, with a stable reactant gas in a collision cell. Products are detected by ionization and mass analysis. We are equipped to study rotational effects on reactivity by comparing results for rotational levels J=0 and 1 of H_2.

Highlights during the funding period are given below.

Development of a fast neutral beam

Neutral beam currents have been measured by phase-sensitive detection of the differential negative ion current with and without the photodetachment laser. Currents slightly in excess of 10 pA (6×10^7 s^{-1}) have been produced for ion beam currents of 3 nA. This ratio is lower than the projected neutral current (1.5×10^8 - 4.8×10^9) for several reasons.

Ion beam currents through the apparatus have, until recently, been ~1-3 nA (10^{10} s^{-1}) at 30 eV and ~0.1 nA (6×10^8 s^{-1}) at 10 eV. These are a factor of five lower than the projection ion currents. We have attempted to increase the ion current in several ways.

- The beam line has been kept scrupulously clean, with Aquadag (conductive coating) used wherever feasible.
- All apertures have been widened as much as possible.
- The electron-impact ion source has been optimized (electrode configuration, pressure, electron energy, extraction mechanism) for the dissociative attachment formation process $N_2O + e^- \rightarrow O^- + NO$.
- Ion trajectories through the apparatus have been simulated (SIMION) to optimize electrode configurations and potentials. (See figure)

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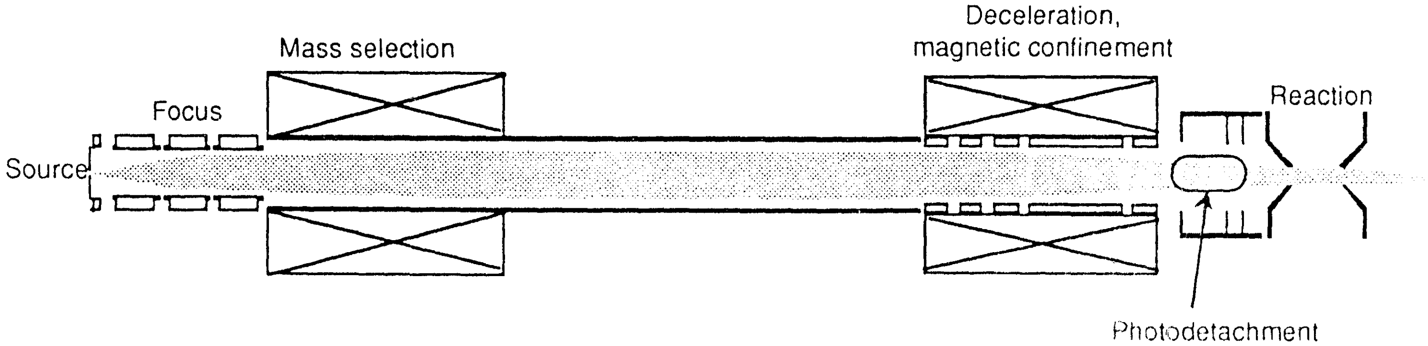
- The mass filter is operated with the highest transmission that is consistent with adequate mass resolution.
- The most recent modification is the installation of a coaxial magnetic field (~500 Gauss) in the deceleration region to limit the radial velocity acquired in deceleration. Other focusing potentials upstream are adjusted to take advantage of the magnetic force, resulting in a more tightly collimated beam. The use of a magnetic field has been observed to improve beam currents by 50% with insignificant degradation of mass resolution.



The dual goals of maximizing photodetachment efficiency and maximizing transmission through the reaction region are, to some extent, in conflict. When a reaction cell is used, the total beam current is maximized by effecting a tight focus within the cell. Photodetachment, however, is maximized by focusing the beam in the laser interaction region, ~3 cm before the reaction cell. We are optimizing the production of neutrals in several ways.

- We can now produce neutral beams of sufficient intensity to enable tuning of beam parameters to maximize production of neutrals. This becomes possible with phase-sensitive detection for neutral beams of ~10 pA but is unrealistic at lower neutral currents, where we are limited to tuning to maximize the *ion* current.
- The reaction cell has been replaced (at least temporarily) by a slit nozzle expansion. Several benefits result from this modification. The requirement of focusing the beam in the reaction region is removed, since the effective reaction region of the expansion is broader. The nozzle height (and, with it, the width of the reaction region) can be varied as one of the optimization parameters for production of neutrals. The effective path length with the present jet (slit length 2.5 cm, slit width 0.1-mm, backing pressure 1.0 T) is lower than for the reaction cell. This is limited primarily by the pumping

requirements of the experiment: the necessary cw operation precludes pulsing the jet (although a 50% duty cycle might be feasible), and geometric constraints preclude differential pumping. The slit nozzle geometry may be more consistent with the photodetachment geometry, however. Diagnostic tests are continuing.

Photodetachment powers of 1600 W can be obtained in a four-pass intracavity configuration with sufficient attention to the cleanliness and alignment of optics. The present configuration, with all mirrors outside the vacuum system, produces a laser cavity of length ~10 m (compare stock mode of 2 m). The resulting lasing modes are not fully stable, and frequently we are forced to operate in a two-pass intracavity configuration, with total photodetachment power limited to 800 W.

- We are in the process of installing the two multipass mirrors into the vacuum system, thereby shortening the cavity to ~4 meters. A further benefit is the fewer number of transmissions through the Brewster windows on the vacuum chamber (four per round-trip instead of the present sixteen). We anticipate greater laser stability in this configuration, with routine operation at the full 1600-W level.
- Additionally, the new in-vacuum optical setup will enable us to implement an extracavity multipass system as a backup method.

Detection stage

The detection system consists of a high-efficiency electron-impact ionizer, a quadrupole mass filter, and a channeltron electron multiplier. The maximum theoretical detection efficiency of the system 10^{-5}, limited by the short time that the fast neutrals spend in the ionization region. (The large momentum of the reaction products toward the detection system is used to advantage to discriminate against ionization of background molecules.) We have marginally detected the ionized fast neutral reactant O (30 eV) using phase-sensitive methods. Anticipated signal levels are much higher than those observed, indicating that the detection system is not operating at peak efficiency. Several improvements have been made.

- Dual tungsten filaments have been installed in the ionizer to effectively flood the region with electrons, ensuring that the space-charge limit is reached.
- Ion trajectory simulations have been used to improve the ion optics within the ionizer. The ramp potential that serves to discriminate against ionized background molecules also effectively functions as a focusing element for ionized fast neutrals entering the mass filter.

- The quadrupole mass filter is operated at the highest transmission that is consistent with adequate mass separation.

Diagnostic tests are continuing.

Equipment purchases

A uv photon detection system (Thorn EMI) has been purchased for detection of chemiluminescent products, an anticipated extension of the experiment following the mass spectrometric detection methods described above. (This purchase was initiated during the previous funding period.) The vacuum system for this experiment has been designed.

A closed-cycle helium refrigerator is scheduled for purchase during this funding period, replacing the present liquid-nitrogen cooling of the reaction cell. The refrigerator will allow us to operate the reaction cell at temperatures of ~30 K for improved velocity resolution. It will also be used in parahydrogen studies.

Personnel

The experiment is presently staffed by one graduate student. One student graduated this year with a terminal M.S. degree. Several undergraduates have made important contributions during the last year. Recruitment efforts for an additional graduate student continue.

A postdoctoral research associate, Peter Armstrong, has accepted a position and will begin working on this experiment in November, 1990, with an initial one-year appointment extendable to two years. He will be supported by remaining startup funds from Georgia Tech. His extensive experience in ion traps, laser operation, and small-signal detection will make him a valuable addition to the group. He is presently completing his thesis in the Department of Physics at the University of Virginia.

Theoretical studies of reaction dynamics

A study of fine-structure effects in the reaction $O+OH \rightarrow O_2+H$ (with A. F. Wagner) was completed and published. This study has important implications for reactions with long-range forces (dipole-quadrupole, quadrupole-quadrupole) in general, including many that are important to combustion. Application has been made to reactivity of neutral species at the low temperatures of interstellar clouds.

Publications and conference presentations

"Theoretical studies of fine-structure effects and long-range forces: Potential-energy surfaces and reactivity of O(^{3}P) + OH($^2\Pi$)", M. M. Graff and A. F. Wagner, J. Chem. Phys. 92, 2423 (1990).

"Theoretical studies of fine-structure effects and long-range forces: Approximating the reactive surface of O(^{3}P) + OH($^2\Pi$)", M. M. Graff and A. F. Wagner, Chem. Phys. Letters, in press (1990).

"Experimental study of reactivity using a fast neutral beam", T. L. Fountain, C. L. Parker, M. F. Steakley, and M. M. Graff, Annual Meeting of the Southeast Section of the American Physical Society, November 1990.

"Half-collision study of fine-structure effects in simple neutral reactions," M. T. Finch and M. M. Graff, Annual Meeting of the Southeast Section of the American Physical Society, November 1990.

Comparison of achievements with objectives:

Progress has been slower than we had hoped during the past year. We have encountered unanticipated problems, and the desired level of graduate student participation has not been achieved. Nevertheless, we believe that with the modifications described above we will succeed in fulfilling most of the objectives that were outlined in the original proposal (reaction cross sections for O+H_2, HD, and D_2, OH+H_2, O_2+H_2, rotational effects, and chemiluminescence detection).

We have concluded, however, that the development of the laser-induced fluorescence detection system is not realistic with the low signal levels inherent to this experiment. Georgia Tech funds that had originally been earmarked for the purchase of a ring dye laser will be used instead to support a postdoctoral research associate for two years. This change of direction is expected to be a major benefit to the research program.

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