2609

SRI International

6P

(NASA-CR-195727) PREDISSOCIATION IN N2(C'4, 1 SIGMA u +) AND OTHER N2 STATES AND ITS IMPORTANCE IN THE ATMOSPHERES OF TITAN AND TRITON Annual Report (SRI International Corp.) 6 p

N94-28816

Unclas

G3/91 0002609

Annual Report • April 1994

PREDISSOCIATION IN N_2 (C_4' $^1\Sigma_u^+$) AND OTHER N_2 STATES AND ITS IMPORTANCE IN THE ATMOSPHERES OF TITAN AND TRITON

Prepared by:

Tom G. Slanger and Richard A. Copeland
Molecular Physics Laboratory

SRI Project 4725
Contract No. NAGW-3581
MP 94-058

Prepared for:

National Aeronautics and Space Administration
Washington, DC 20546

Attn: Jay T. Bergstralh, SLC

Tom G. Slanger

Tom Slanger, Principal Investigator

333 Ravenswood Avenue • Menlo Park, CA 94025-3493 • (415) 326-6200 • FAX: (415) 326-5512 • Telex: 334486

Annual Report • April 1994

PREDISSOCIATION IN N_2 ($c_4' \, {}^1\Sigma_u^+$) AND OTHER N_2 STATES AND ITS IMPORTANCE IN THE ATMOSPHERES OF TITAN AND TRITON

Prepared by:

Tom G. Slanger and Richard A. Copeland
Molecular Physics Laboratory

SRI Project 4725
Contract No. NAGW-3581
MP 94-058

Prepared for:

National Aeronautics and Space Administration
Washington, DC 20546

Attn: Jay T. Bergstralh, SLC

Approved:

Donald J. Eckstrom, Director
Molecular Physics Laboratory

David M. Golden
Vice President
Physical Sciences Division

SRI International 333 Ravenswood Avenue • Menlo Park, CA 94025-3493 • (415) 326-6200 • FAX: (415) 326-5512 • Telex: 334486

STATEMENT OF WORK

SRI International will provide the appropriate personnel, materials, facilities, and support services and will exert its best efforts within time and funding constraints to: 1) Investigate predissociative effects in the $c_4'^1\Sigma_u^+$ state of N_2, and other nearby states and 2) Identify and quantify the atomic nitrogen states that are created from the predissociative process.

OBJECTIVES

The objectives of this program are to further our understanding of the upper atmospheres of Titan, Triton, and the earth in terms of the observed emissions of the 13-14 eV states of N_2. These states are generated at quite high rates, yet very little emission is observed from them. The reasons are complex, involving resonance trapping and predissociation, and it is our goal to quantify the effects of predissociation, particularly on the $c_4'{}^1\Sigma_u^+$ state of N_2.

Earlier experiments (Ajello et al., 1989) had indicated that predissociation of the c_4' state was of little importance, yet over the last two years a growing body of evidence has shown that for levels above v = 2, predissociation is in fact a major process (Walter et al., 1993). It is the v = 0 level for which production by electron bombardment and photoexcitation is highest, and so it has been most important to evaluate the effects of predissociation on this particular level.

Our goal has been to target $c_4'(v = 0)$ for a thorough analysis, in which we wish to determine both the extent of predissociation as a function of rotational level, and the atomic product branching ratio, where the only possible products are $N(^4S) + N(^4S)$ and $N(^2D) + N(^4S)$. For the first year of funding, our intention was to demonstrate two-photon excitation of the intermediate $N_2(a\,^1\Pi_g)$ state, so that we could bridge the gap to the 13 eV energy region, and then use a second laser to reach the c_4' state itself. In these tasks we have been successful.

ACCOMPLISHMENTS

In the first year of funding, our intention has been to arrive at a level of system development previously achieved by others whose goal was not the same. In subsequent years, the studies will differ from the earlier work.

Kompa and co-workers (Opitz et al., 1990) have demonstrated excitation of the $N_2(c_4')$ state by two-photon excitation of the $N_2(a)$ state, followed by excitation in the $a \rightarrow c_4'$ transition. However, their interest was in studying subsequent ionization processes. In our case, the important issue is to *avoid* ionization and observe the undisturbed fate of the c_4' state, particularly its dissociation to atoms.

In Figure 1 we show the 2 + 2 REMPI (resonance-enhanced multiphoton ionization) spectrum of the v = 1 level of the $N_2(a)$ state. This level lies approximately two-thirds of the way toward the target c_4' state, which is to be reached by the addition of a third photon of approximately the same color (~296 nm). We have generated a very similar spectrum by observing vacuum ultraviolet fluorescence rather than measuring ion currents.

Excitation of the c_4' state in this manner does not produce a spectrum, as in Figure 1 but generates isolated lines. This result is obtained because the initial level, a particular rotational level J of $a\,^1\Pi_g(v = 1)$, can only access levels in c_4' by $\Delta J = 0, \pm 1$. To obtain J-dependent information, it is necessary to excite different J in the multiphoton $a \leftarrow X$ step, which we have done.

In Figure 2 we show a spectrum in which the first laser is set at the bandhead of Figure 1, while the $a \rightarrow c_4'$ laser is scanned around 296 nm. As predicted, the spectrum is very sparse, and a few low-J lines are seen. In both figures, detection is carried out through ionization, either of the a state in a 2-photon nonresonant process, or of the c_4' state by one photon. When the output profiles overlap temporally, then the 283-nm laser, being substantially more intense, will act as the

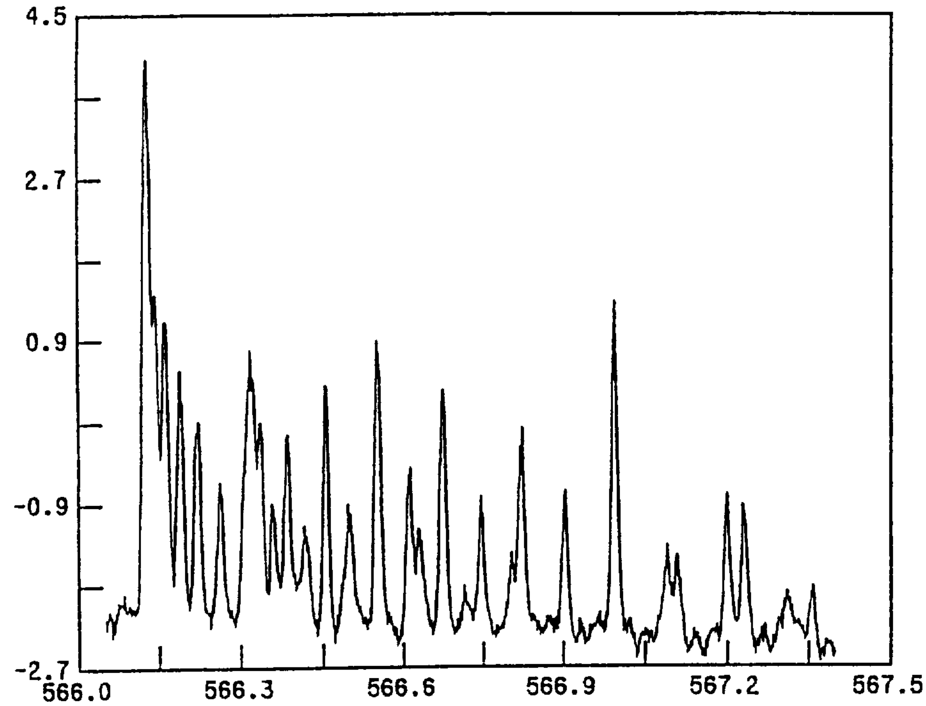

Figure 1. 2+2 REMPI spectrum of N_2(a-X) 1-0 band.

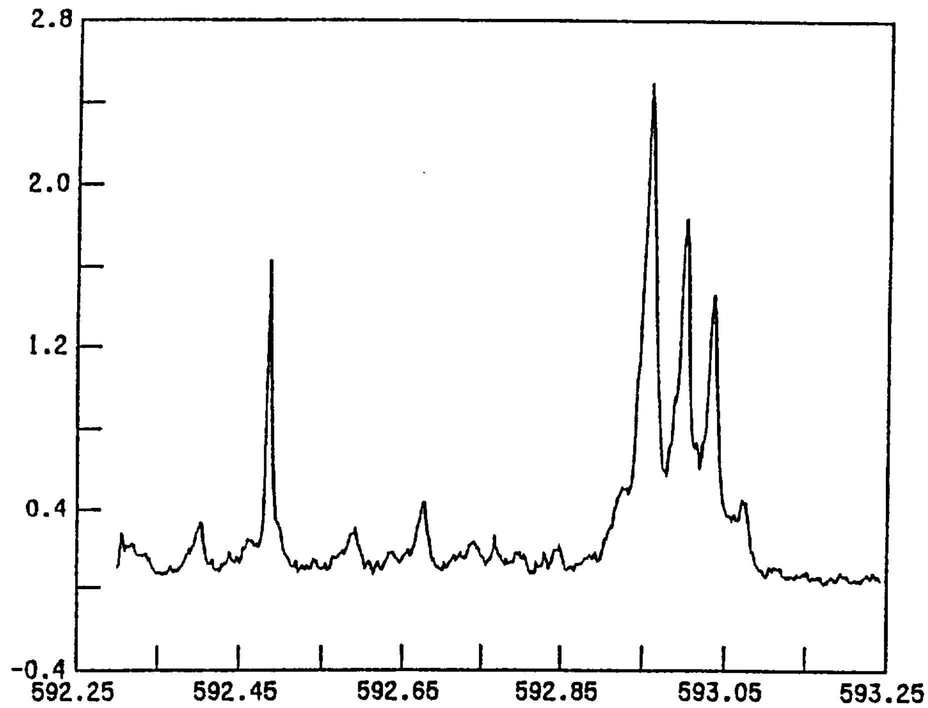

Figure 2. One-photon ionization spectrum of $N_2(c_4',v=0)$ generated from N_2(a) near a-X 1-0 bandhead.

ionizer for c_4'. If a temporal delay is introduced, ionization of c_4' by 296-nm radiation is substantially less efficient. For our purposes, these ionization pathways are just phenomenological effects, since ionization of c_4' is not our goal; it is only a marker for knowing that the c_4' state has been produced.

We have thus succeeded in generating $c_4'(v = 0)$, which was our goal for the first year. All of the other 13-14 eV states can be reached in this way, and higher levels of $N_2(a)$ may present advantages in terms of Franck-Condon factors; we will ascertain whether they do. Once we have collected other spectra to demonstrate the range of accessible states, then we will introduce the third laser and commence measurements of the atomic products of predissociation.

PLANS

Now that the $c_4'(v = 0)$ target level has been accessed, the remaining issue is to determine its predissociative products and yields. To do this, we will rely on two pieces of information. Walter et al. (1993) have investigated the higher vibrational levels and measured products and yields, and we expect it to be straightforward for us to access the same levels. Thus, we can normalize our v = 0 data against their higher level data. Second, we can excite 13-14 eV levels that are known to be fully predissociated, such as $b^1\Pi_u(v = 0)$, so that we have a 100% level calibration. From what we will have learned about the higher c_4' levels—which are reported by Walter et al. (1993) to dissociate to either $N(^4S) + N(^2D)$ or $N(^4S) + N(^2P)$—we will be in a position to determine the $c_4'(v = 0)$ predissociation products and yields.

As seen in Figures 1 and 2, the generation of substantial signals through the two-laser experiment has been accomplished. We now need to add the third laser to interrogate the N-atoms. We were concerned earlier that resonance trapping might be an issue, and that it would be necessary to work at low N_2 pressure, but this now seems unlikely to be a problem, since photons trapped within the cell (for a few passes) will not significantly add to the N-atoms generated within the laser focal volume. In other words, resonance trapping may affect VUV observations, but it should not affect localized N-atom yields.

Our specific plan for the second year's funding is to demonstrate that we can use the $N_2(a)$ state as a stepping stone to a range of 13-14 eV states and levels. The usual diagnostic will be ionization of these levels with the focused energy of the first laser. We will then incorporate the third laser, for atom detection, and start exploration of the atomic yields, initially by ascertaining the relative sensitivity of the system to the three atomic products, $N(^4S)$, $N(^2D)$, and $N(^2P)$. We note that the atoms will not be lost rapidly in a N_2 environment, and they can be sampled without any interference from the photon fluxes of the first two lasers.

REFERENCES

Ajello, J. M., G. K. James, B. O. Franklin, and D. E. Shemansky, Phys. Rev. A **40**, 3524 (1989).

Opitz, S., D. Proch, T. Trickl, and K. L. Kompa, Chem. Phys. **143**, 305 (1990).

Walter, C. W., P. C. Cosby, and H. Helm, J. Chem. Phys. **99**, 3553 (1993).

SRI International
Proposal for Research PYU 92-109R
11 April 1994

COST ESTIMATE SUMMARY
(05/15/94) - (05/14/95)

	Hours	Rates	Dollars		
Direct Labor		$	$		
Senior Professional	127	38.14	4,843.43		
Professional	658	21.34	14,039.39		
Technical	50	19.03	951.44		
Clerical	50	18.74	936.88		
Reports	9	21.93	197.40		
Total SRI Labor	894		$ 20,968.54		
Staff Benefits at 41.5%			8,701.94		
Total Labor and Staff Benefits				$	29,670.48
	Percent of Labor + Benefits		$		
Research Overhead	65.2%%		19,345.15		
Total Overhead					19,345.15
Other Direct Costs					
Report Production	C-1		8.01		
Materials and Supplies	C-2		5,237.00		
Communication	C-3		200.00		
Travel & Subsistence	C-4		1,887.00		
Equipment Purchases	C-5		8,140.00		
Computer	C-6		500.00		
Shipping and Receiving	C-7		200.00		
Subtotal Other Direct Costs			$ 16,172.01		
Burden at 0.70% of ODC			13.21		
Burden at 5.70% of ODC			814.25		
Subtotal Burden			827.46		
Total Other Direct Costs					16,999.47
G & A at 23.80%			13,774.28		
Total G & A					13,774.28
Total Estimated Cost				$	79,789.38

NOTE: The SRI cost estimating system calculates using exact amounts (i.e., to the penny). For purposes of negotiation and contract documents, the amounts shown above should be rounded to whole dollars.