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by

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EXPERIMENTAL AND THEORETICAL STUDIES IN PLANETARY AERONOMY

Quarterly Progress Report

Covering the Period 1 April 1966
through 30 June 1966

Prepared under Contract No. NASW-1283

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I. INTRODUCTION

This is the Fifth Quarterly Progress Report which describes the technical progress from 1 April 1966 through 30 June 1966 under NASA Contract No. NASW-1283. Scientific investigations accomplished during the current reporting period resulted in the generation of the following papers submitted and/or accepted for publication in accredited scientific journals, books and/or GCA Technical Reports or presented at scientific meetings.

Technical Papers Submitted and/or Accepted for Publication

	Publication
a. Submitted	
On the Quenching of 6300Å Airglow (P. Warneck and J.O. Sullivan)	Planetary and Space Sciences
Reactions of ^1D Oxygen Atoms III. Ozone Formation in the 1470Å Photolysis of O_2 (P. Warneck and J.O. Sullivan)	J. Chem. Phys.
Studies of Ion-Molecule Reactions by a Photoionization Mass Spectrometer Technique II. Charge Transfer Reactions of Argon Ions at Near Thermal Energies	J. Chem. Phys.
b. Accepted	
Ionization Potential of Molecular Xenon and Krypton (J.A.R. Samson and R.B. Cairns)	J. Opt. Soc. Am.

Technical Papers Presented at Scientific or Professional Meetings

On Ultraviolet and X-ray Spectroscopy of Laboratory and Astrophysical Plasmas (R.B. Cairns and J.A.R. Samson) - Presented at Culham Laboratory, Abingdon, Berks, England, on March 29 - April 1, 1966.

The Measured Photoionization Cross Section of Atomic Hydrogen (R.B. Cairns and J.A.R. Samson) - Presented at 47th Annual American Geophysical Union Meeting held in Washington, D.C., on April 19-22, 1966.

A Satellite Experiment on the Detection of Noctilucent Clouds in the VUV Region (F.F. Marmo and T.C. Degges) - Presented by F.F. Marmo and T.C. Degges at the 47th Annual American Geophysical Union Meeting held in Washington, D.C., on April 19-22, 1966.

In Section II, technical summaries are given on the work performed under the present contract. During the current reporting period, significant progress has been achieved in the following areas:

A. Photochemistry of planetary atmospheres

B. Laboratory investigations in the VUV and the EUV spectral regions

C. Theoretical studies

D. Other pertinent information.

II. TECHNICAL SUMMARIES OF WORK PERFORMED UNDER THE PRESENT CONTRACT

A. PHOTOCHEMISTRY OF PLANETARY ATMOSPHERES

During the current reporting period, work has been completed in three scientific areas: (1) reactions of ^1D oxygen atoms with ozone, (2) the quenching of 6300Å airglow, and (3) charge transfer reactions of argon ions at near thermal energies. In each case the subject matter has been submitted for publication in a scientific journal so that only brief discussion is given here since the details are available elsewhere.

1. Reactions of ^1D Oxygen Atoms with Ozone

In the previous quarterly report an analysis was presented concerning the role of ^1D oxygen atoms in reactions with ozone, wherein the ^1D species were produced by the photolysis of oxygen at 1470Å, which lies in the Schumann-Runge continuum. Prior to this experiment, it was generally assumed that the primary dissociation process would generate metastable O^1D atoms and O^3P atoms in equal amounts. It was decided to test this assumption since its validity had never been ascertained photochemically and the required capability was presently available in this laboratory. In the previous report, the following kinetic mechanisms were suggested for analytical consideration:

$$O_2 + h\nu \xrightarrow{1470Å} O + O^* \qquad (1)$$

$$O + 2\,O_2 \rightarrow O_3 + O_2 \quad \text{fast} \qquad (2)$$

$$O^* + 2\,O_2 \rightarrow O_3 + O_2 \quad k_3 \qquad (3)$$

$$O^* + O_2 \rightarrow O_2 + O \quad k_4 \qquad (4)$$

$$O^* + O_3 \rightarrow 2\,O_2 \quad k_5 \qquad (5)$$

where O^* signifies excited oxygen atoms in the ^1D metastable state in (1) above. For this mechanism it can be shown that the ozone quantum yield (ϕ) in pure oxygen is given by the following expression

$$\left[\frac{k_3}{k_5} [O_2] + \frac{k_4}{k_5} \right] [O_2] = \frac{I}{V} \frac{\phi^2 - 2(1-\alpha)\phi}{2-\phi} \tag{6}$$

where:

I = the light source intensity

V = the volume flow rate in the reactor

α = the quantum yield for O^1D generation

Although a value of $\alpha = 1$ (equal amounts of ^1D and ^3P atoms) was assumed in the previous report, it should be noted that Equation (6) provides a lower limit to this parameter since $\alpha \geq (1 - \phi/2)$. Since the observed ozone quantum yield,[1] $\phi \leq 0.5$, it follows that $\alpha \geq 0.75$. Since α can theoretically be less than unity, the initial assumption involving equivalent generation of O^1D and O^3P atoms is not necessarily valid. Although an α-value of unity would be consistent with the present investigation, the minimum value of 0.75 would indicate that the Schumann transition leading to ^1D oxygen atoms is at least the dominant factor in the photolysis of oxygen at 1470Å. An α-value of unity appears to be a reasonable choice.

With this criterion, additional analyses have been performed on new experimental data on the 1470Å photolyses of the following gas mixtures: 1:1 oxygen with nitrogen, 1:1 helium with carbon dioxide and 4:1 oxygen with N_2O to provide relative quenching rates for O^1D by nitrogen and helium, i.e.:

$$O^* + N_2 \rightarrow O + N_2 \tag{7}$$

$$O^* + He \rightarrow O + He \tag{8}$$

The results are presented in Table 1 along with new updated rate constant ratios involving reactions (3), (4), (5), (7) and (8). Absolute rate constants in Table 2 were obtained using the value for $k_4 = 10^{-15}$ cm^3 sec^{-1} provided by Hunten and McElroy[2] and $k_5 = 3 \times 10^{-12}$ cm^3 sec^{-1} presented by Fitzsimmons and Bair.[3]

TABLE 1

Ratios of Reaction Rate Constants
for Selected Temperatures

$T(^oK)$	$(k_4/k_5) \times 10^3$	$(k_3/k_5) \times 10^{23}$	k_6/k_4	k_7/k_4
273	0.91 ± 0.23	3.7 ± 2.5	-	-
297	0.61 ± 0.12	2.8 ± 2.0	0.40 ± 0.2	0.40 ± 0.20
323	0.47 ± 0.15	1.9 ± 1.5	-	-

TABLE 2

Estimates of O^1D Rate Constants

Reaction	Rate Constant
$O^* + 2\ O_2 \rightarrow O_3 + O_2$	$k_3 = 2 \times 10^{-34}$ $cc^2/molecule^2$ sec
$O^* + O_2 \rightarrow O + O_2$	$k_4 = 4 \times 10^{-15}$ cc/molecule sec
$O^* + O_3 \rightarrow O_2 + O_2$	$k_5 = 7 \times 10^{-12}$ cc/molecule sec
$O^* + N_2 \rightarrow O + N_2$	$k_7 = 1.5 \times 10^{-15}$ cc/molecule sec
$O^* + He \rightarrow O + He$	$k_8 = 1.5 \times 10^{-15}$ cc/molecule sec

Analysis of the O_2 - N_2O mixture experimental results indicates the requirement for a drastic reduction of the ozone quantum yield. Since N_2O

is known[4] to react rapidly with O^1D, these results are further evidence that ^1D oxygen atoms are indeed produced in the photolysis of oxygen at 1470Å according to the assigned mechanism.

The detailed work is described in a paper (which is the last in a series on laboratory investigations of O^1D reactions) entitled "Reactions of ^1D Oxygen Atoms III. Ozone Formation in the 1470Å Photolysis of O_2," which has been submitted for publication to the Journal of Chemical Physics.

2. <u>The Quenching of 6300Å Airglow</u>

Interpretation of the 6300Å dayglow observations[5-8] has created the argument[8-10] that the responsible emitter, O^1D, is rapidly quenched by either oxygen or nitrogen with a required quenching coefficient of about 10^{-10} cc/molecule sec.[2,8,9] Alternatively, Hunt[11] has demonstrated recently that in a dry atmosphere such large quenching rates are irreconcilable with the known distribution and abundance of ozone at lower altitudes.[12] His results indicate a more appropriate quenching coefficient of about 5×10^{-15} cc/molecule sec.

The results of the preceding section are directly applicable to the current problem. In brief, application of the mechanism indicated by reactions (3), (4), (5) and (7) and the results of Tables 1 and 2 indicate that oxygen and/or nitrogen quenching is not pertinent to the 6300Å dayglow problem. As such, it appears that a reexamination of the 6300Å dayglow mechanism in the Earth's atmosphere is clearly required.

The detailed work is described in a paper entitled "On the Quenching of 6300Å Airglow," which has been submitted for publication to Planetary and Space Sciences.

3. Charge Transfer Reactions of Argon Ions at Near Thermal Energies

In the Third Quarterly Report it was shown that the apparatus which was designed, developed, and employed to perform the measurements on the mass analysis of photoionization products from N_2 and O_2 could be modified for ion-molecule reaction studies. A detailed description of the instrumentation is available elsewhere.[13] For the present purpose, the main equipment modification involved the employment of relatively high pressures (above 50 microns) in the ion-molecule reaction cell. In brief, a new experimental technique has evolved which eliminates the difficulties and disadvantages of electron impact. An extensive program was undertaken to perform specific ion-molecule measurements which would be relevant to the interpretation of the Mariner-IV ionospheric observations. For a variety of reasons it was most propitious to investigate initially those chemical reactions involving A^+. Specifically, charge transfer processes were studied which involved $^2P_{3/2}$ argon ions at ion source pressures in the range 15-60 microns at a source acceleration potential of 0.5 eV. The average energy of the reacting ions was shown to be near thermal (i.e., less than 0.17 eV). The investigated charge transfer reactions and their respective rate constants are:

$$A^+ + NO \rightarrow A + NO^+ \qquad k = 3.9 \times 10^{-10} \text{ cc/molecule sec} \qquad (9)$$

$$A^+ + O_2 \rightarrow A + O_2^+ \qquad k = 1.1 \times 10^{-10} \text{ cc/molecule sec} \qquad (10)$$

$$A^+ + CO_2 \rightarrow A + CO_2^+ \qquad k = 7.0 \times 10^{-10} \text{ cc/molecule sec} \qquad (11)$$

$$A^+ + CO \rightarrow A + CO^+ \qquad k = 1.25 \times 10^{-10} \text{ cc/molecule sec} \qquad (12)$$

$$A^+ + N_2 \rightarrow A + N_2^+ \qquad k = 0.66 \times 10^{-10} \text{ cc/molecule sec} \qquad (13)$$

It is now evident that the above experimental technique can be employed to investigate a variety of ion-molecule reactions of potential importance to an understanding of the kinetics of planetary ionospheres.

No additional details are given here since they can be found in a paper entitled "Studies of Ion-Molecule Reactions by a Photoionization Mass Spectrometer Technique II. Charge Transfer Reactions of Argon Ions at Near Thermal Energies," which has been submitted for publication to the Journal of Chemical Physics.

B. LABORATORY INVESTIGATIONS IN THE VUV (1000-2000Å) AND THE EUV (BELOW 1000Å) SPECTRAL REGIONS

Under this phase of the program, two important scientific areas of investigation have not presently been completed: (1) investigation of the electron energy spectrum due to EUV and VUV photoionization, and (2) atomic hydrogen photoionization cross sections in the EUV and VUV regions.

1. Investigations of the Electron Energy Spectrum Due to EUV and VUV Photoionization

As discussed in the previous report, the electron energy analyzer is operating satisfactorily with a limited energy resolution of approximately 3 percent. Valuable experimental data have been obtained for the cases of molecular oxygen and nitrogen. However, a limiting experimental factor is the presence of the ambient magnetic field as it affects very slow electrons as discussed previously. During the current reporting period, emphasis has been directed to improving the energy resolution and minimizing the ambient magnetic field effects on slow electrons. The magnetic effect is due to the fact that when the energy of the electrons is investigated at wavelengths near the threshold for an electronic transition, ambiguous results may be obtained owing to magnetic trapping. To minimize the magnetic field effect, a Helmholtz coil has been constructed and employed to produce a uniform field which can be oriented in a direction opposite to the ambient field and adjusted in magnitude to produce a field free region. The field is produced by two coils of radius a cm separated by

a distance a cm and having each n turns of wire with a current of I amps flowing through each coil. The field at the center of the coil is given by

$$H = 0.285 \frac{\pi n I}{a} \text{ gauss} \tag{14}$$

The design parameters employed herein were a = 51 cm and n = 100 turns so that to produce a field of 0.5 gauss, a current of 280 mA was required. Using a gaussmeter, direction and magnitude of the ambient field were determined at the location of the experiment and an applied current of 270 mA was employed for complete neutralization. Work is now in progress to reassemble the electron energy analyzer and operate the Helmholtz coil in situ.

2. Atomic Hydrogen Photoionization Cross Sections for EUV-VUV Regions

Preliminary measurements of the photoionization cross sections of atomic hydrogen were reported in the previous quarterly report for the spectral region 912 to 850Å. At shorter wavelengths, there was a disturbing scatter of the experimental points and consequent disagreement with theory. It was pointed out that this scatter might be attributable to the presence of certain metastable impurities in the ionization chamber. In order to investigate this possibility, a number of experiments were designed and performed to identify the metastable states and to evaluate their contribution and role in the quantitative determination of the photoionization cross sections of atomic hydrogen.

Concerning electronic metastable states, recent reports[14,15] indicated that the role of the $C^3\Pi_u$ required evaluation. The appearance

potential of this state is 11.87 eV which is about 3.54 eV less than the ionization potential of ground state H_2 so that photoionization from the metastable state could be achieved by photons with energy $\geq$ 3.54 eV (i.e., any wavelength $\leq$ 3505Å). Accordingly, although an experiment was performed to search for the ionization onset at wavelengths below 3503Å, none was observed. Although this negative result does not preclude the existence of the metastable $C^3\Pi_u$ state, it does yield an estimate for the upper limit abundance which is too low to affect the present measurements. Thus, it has been concluded that the role of this specific metastable in the subject experiment was insignificant. To estimate the integrated effects of all metastable states, an additional experiment was performed in which the metastables were allowed to collide with a number of variable size ion collectors, so that from the resultant photoelectron ejection, an estimate of their total abundance could be obtained. The largest observed current (from the largest collector) was about 1×10^{-11} A corresponding to an upper limit estimate of 2×10^8 molecules sec^{-1} for the flow of metastables through the ionization chamber. Assigning a representative value to the photoionization cross section of these metastable species, it becomes evident that the resultant number density of metastables is still orders of magnitude too low to yield any measurable photoionization current. On the basis of these experiments, it has been concluded that metastable electronic states do not play a significant role in the present experimental measurement of atomic hydrogen cross sections.

Another source of spurious ionization was discovered during the performance of these experiments. It was noted that when a hydrogen discharge light source was employed on the monochromator, irradiation of the ionization

chamber resulted in ion currents only at those discrete wavelengths corresponding to the Lyman resonance series lines: 1215.7, 1025.7, 972.5, 949.7, and 937.8Å. It was important to ascertain the source of these currents since they could affect the measurements. Two mechanisms were considered:

(a) Possible Formation of H_3^+ in the Gas Phase

$$H + h\nu \text{ (Lyman-}\alpha \sim 10.3 \text{ eV)} \rightarrow H^*$$

$$H^* \text{ (10.3 eV)} + H_2 \rightarrow H_3^+ \text{ (10.07 eV)} + 0.23 \text{ eV} + e^-$$

where the other resonance lines behave similarly except that the photoelectron has a higher kinetic energy;

(b) Resonant Scattering by Atomic Hydrogen

$$H^* \rightarrow h\nu \text{ (Lyman resonance series)} \rightarrow H^*$$

$$H^* \rightarrow H + h\nu \text{ (Lyman resonance series)}$$

where the scattered photons eject electrons from the ion collectors only at the Lyman resonance series wavelengths.

The performance of an appropriate experiment demonstrated conclusively that the dominant cause of the spurious ionization was due to the resonant scattering mechanism described above. Since extremely low ion currents of the order of 10^{-13} A were obtained, no corrections were required in the present experiment with respect to the current measurements.

This interesting effect suggests that an appropriate cell can be employed as a narrow band optical detector since it would be totally nonresponsive to radiation at wavelengths other than those of the resonance series.

The possibility of exploiting this technique in the design of pertinent rocket and/or satellite experiments is being investigated.

The above results indicate that no significant effects are due to H_2 metastables or resonance scatter radiation in the electrodes. In order to evaluate the effects of vibrationally excited hydrogen molecules, the experimental photoionization experiments were extended to the EUV region. At wavelengths below 912Å, the following processes pertain:

$$H + h\nu \ (\lambda \leq 912\text{Å}) \rightarrow H^+ + e \tag{15}$$

$$H_2 + h\nu \ (\lambda \leq 804\text{Å}) \rightarrow H_2^+ + e \tag{16}$$

so that between 912 and 804Å the observed ionization current should be due to process (15). However, a large increase has been observed in the vicinity of 830Å and a smaller increase at about 860Å. If H_2 is excited in the discharge into vibrational levels of its ground state, then ionization from the lowest and presumably most densely populated level, should occur at 831.5Å and from the second vibrational level at 859.5Å. The existence of an appreciable number of H_2 molecules in the first vibrational level as observed in the performance of the above experiment for $\lambda < 912$Å results in semi-quantitative measurements of the photoionization cross section of atomic hydrogen using the present discharge technique. As a consequence, it was decided to generate atomic hydrogen by thermal dissociation for measurements in this spectral region wherein it is well known that the fraction of vibrational metastables is too low to affect the measurements. An appropriate oven has been designed, constructed and tested during the current reporting period.

C. THE DAILY VARIATIONS OF ELECTRON AND ION TEMPERATURES

In previous work under this contract,[16] a study was carried out of the heating and cooling of the ionospheric electrons which confirmed NASA rocket observations that the electron temperature is substantially in excess of the neutral particle temperature. Further work[17] showed that the positive ion temperature would also exceed the neutral particle temperature at altitudes above about 300 km. This prediction is consistent with the Explorer XVII satellite observations[18] and with back scatter data.[19]

These earlier calculations made use of the concept of a critical energy in computing the heat input arising from the collisions with the fast photoelectrons produced by the absorption of solar ultraviolet radiation. Because the neutral particle density decreases rapidly with increasing altitude in the F-region and the electron density does not and because the electron-electron collision cross section varies rapidly with electron energy and the electron-neutral particle collision cross sections do not, it is possible to define at any given altitude a critical energy E_c such that if the electron energy E is less than E_c, all its energy is converted into heat and if E is greater than E_c, E_c is converted into heat.

The approximation is not a necessary one and we have now replaced it by an accurate description of all the energy loss processes. This has required an extensive and detailed investigation of the electron-neutral particle collision processes and a new program has been written which incorporates this information.

Some results are shown in Figure 1. They are about a factor of two higher than our earlier predictions based on the critical energy concept.

The figure is interesting in its demonstration that the heat source is symmetrical about midday at altitudes below 300 km (in harmony with the analysis of Explorer XVII data), but it becomes asymmetrical at higher altitudes. The asymmetry reflects the well-known asymmetry in the neutral particle density.

Since our earlier work, it has become clear that thermal conduction must be included as an important influence on the electron and ion temperatures at high altitudes. We are now engaged in solving numerically the heat equilibrium equation including thermal conduction for various times during the day.

There is at present a serious discrepancy between electron temperatures derived from back scatter data and electron temperatures measured with rocket-borne probes in the region of 200 km. It can be anticipated that our calculations will aid in resolving the present discrepancy.

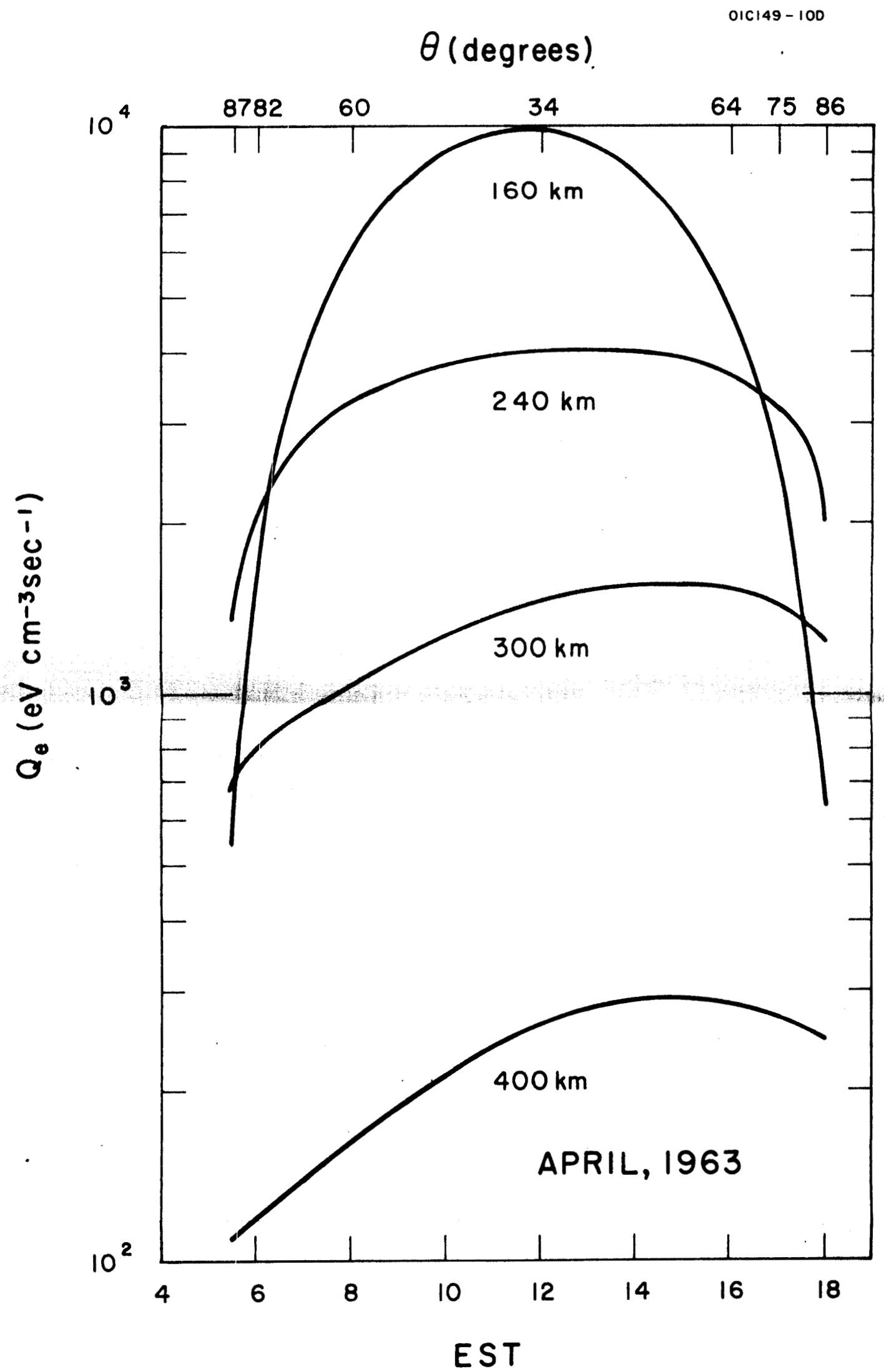
01C149 - 10D
θ (degrees)
87 82
60
34
64
75
86
10^4
10^3
10^2
Q_e (eV cm^{-3}sec^{-1})
160 km
240 km
300 km
400 km
APRIL, 1963
4
6
8
10
12
14
16
18
EST

Figure 1

III. OTHER PERTINENT INFORMATION

During this quarter, three scientific papers were presented at professional meetings.

Dr. J.A.R. Samson presented a paper entitled "Study of Kinetic Energy of Photo Electrons" at a Meeting on Ultraviolet and X-ray Spectroscopy of Laboratory and Astrophysical Plasmas at Culham Laboratory, Abingdon, Berks, England on March 29 - April 1, 1966. The talk provided considerable interest to the several laboratories represented at this international meeting. In particular, scientists at the University College, London, displayed a keen interest in our work since it is directly applicable to in situ determinations of electron temperature in the upper atmosphere. This group is active in this area. In addition, several papers were presented which were of particular interest to our current program. These included: (1) A paper presented by Huffman et al on the photographic absorption spectrum of atomic nitrogen. Unfortunately, no quantitative measurements were reported. (2) A paper by Prof. Weissler on fluorescent radiation induced by VUV photons, and (3) A paper by Dr. Fano on the photoionization of gases and in particular on autoionization. Note: Travel and trip expenses were supplied by GCA Corporation.

Dr. Cairns presented a paper entitled "The Measured Photoionization Cross Section of Atomic Hydrogen" at the 47th Annual American Geophysical Union Meeting held in Washington, D.C. on April 19-22, 1966. The paper was well received since it reported on quantitative absorption of atomic hydrogen which to date had not been reliably investigated. No other papers were given which were directly concerned with laboratory measurements in the VUV and EUV region.

Dr. F.F. Marmo presented a paper entitled "A Satellite Experiment on the Detection of Noctilucent Clouds in the VUV Region" at the American Geophysical Union Meeting held in Washington, D.C. on April 19-22, 1966. The paper was the subject of considerable discussion since there were others in the audience who had proposed noctilucent cloud observations in the visible and infrared regions. It turns out, however, that a major advantage of this VUV is that a satellite probe can operate on a world wide basis independent of the position of the sun. A number of other papers were presented at this meeting which were of direct interest to our program requirements. These included: (1) A paper entitled "An Analysis of Rocket Spectrometer Measurements in the Middle Ultraviolet" by R.T. Brinkman et al, in which the ultraviolet data obtained by Barth were analyzed. (2) "Vacuum UV and 3914Å Photometric Measurements of Auroras from a Satellite" by J.E. Evans et al, in which the daytime background of a solar illuminated atmosphere amounted to only 0.5 kR for λλ1240-1800Å. These data are directly applicable to the noctilucent cloud experiment, and finally (3) "Examination of Extra-Terrestrial Particles Collected by the Luster Sounding Rocket Experiment by N.H. Farlow et al, in which estimates were obtained on the size distribution and number density of particles associated with noctilucent clouds.

IV. QUARTERLY PROGRESS REPORT FOR HOURS WORKED IN THE PERIOD 1 APRIL 1966 THROUGH 30 JUNE 1966

In compliance with the requirements of the subject contract, the following tabulation of hours worked by GCA personnel on the subject contract is submitted as well as an integrated tabulation of total hours worked by labor category and grade.

Labor Category	Labor Grade	Total Hours
*Junior Technician	2	553
*Technician Experimental Machinist	3	212
*Senior Technician Senior Experimental Machinist	4	422
*Junior Scientist Junior Engineer	5	235
*Scientist Engineer	6	636
Senior Scientist Senior Engineer	7	221
Staff Scientist	8	603
Principal Scientist	9	215
Group Scientist	10	250
ODC (Overhead Direct Charges)	10	130

*and other equivalent categories

GCA TECHNOLOGY DIVISION

Project No. 1-218-01
Direct Labor Hours
1 April 1966 - 30 June 1966

Foley, C.	50	
Ridgeway, E.	65	
Abbott, P.	244	
Carvalho, J.	235	
Good, L.	248	
Hunter, R.	53	
Kachadorian, M.	86	
Kelley, F.	344	
Padur, J.	195	
ShardaNand	23	
Sharma, A.	58	
Shahood, D.	252	
Sullivan, J.	140	
Warneck, P.	246	
Nicolopoulos, H.	1	
Andonian, E.	12	
Cairns, R.	357	
Chandler, J.	12	
Fanning, J.	19	
Lombardo, C.	18	
O'Connor, A.	11	
Batchelder, P.	11	
Bordonaro, V.	14	
Castro, E.	5	
Christensen, A.	5	
Cotoni, J.	19	
Degges, T.	32	
Breau, M.	51	
Crivello, V.	3	
Murphy, J.	3	
Samson, J.	215	
Savoie, R.	11	
Rosendahl, R.	4	
Victor, G.	38	ODC only
Lopez, W.	4	
Suprenard, A.	1	
Bordini, L.	6	
Rawson, J.	4	
Nilsson, G.	35	
Marmo, F.	250	
Dalgarno, A.	92	ODC only
Dahglian, C.	5	
	3,477	

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