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EXPERIMENTAL STUDIES OF PHOTOIONIZATION PROCESSES IN AIR

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An experimental study is described NO in the nonequilibrium vacuum ultraviole The experiment utilizes a splitter-plate sh tion behind a reflected shock is used as a 1 by gases processed only by the incident sho radiation profiles behind strong incident sh the identification of the relevant excitation This report describes the apparatus needed to establish testing parameters.	whose object et (VUV) rad ock tube tech ight source t ock wave. In ock waves a processes. and present	tive is to iation fro inique wh o measur addition re also m s the res	o assess the role of om shock-heated air. erein the VUV radia- re the absorption a, nonequilibrium heasured to aid in sults of calculations		

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TABLE OF CONTENTS

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I.	INTRODUCTION AND DISCUSSION	1
	I.1 Radiative Properties of NO in the Vacuum Ultraviolet	3
	I.2 Multiple-Step Photoionization Processes	5
II.	RESEARCH PROGRAM	7
	II. 1 Vacuum Ultraviolet Radiation-Measurements	7
	II.1.1 Description of Experiment and Results in N ₂	7
	II.1.2 Definition of NO Splitter-Plate Experiment	9
	II.1.3 Nonequilibrium Radiation Overshoot Measurements	12
111.	CURRENT STATUS AND FUTURE PLANS	14
	REFERENCES	15
	FIGURES	17

i

TECHNICAL REPORT SUMMARY

Technical Problem:

An experimental program to investigate basic vacuum ultraviolet photoionization processes in air is currently underway at CAL. Particular emphasis is directed toward the interaction of shock-induced nonequilibrium radiation with gaseous species.

Methodology:

The experimental shock-tube program consists of three tasks;

- 1. Perform quantitative measurements to determine the contribution of the NO molecule to the nonequilibrium, vacuum ultraviolet emission spectrum from shock-heated air.
- 2. Conduct experiments to determine the relative importance of multiple-step vs. one-step photoionization processes in air.
- 3. Obtain quantitative, spectrally resolved measurements of the nonequilibrium, vacuum ultraviolet flux from advancing shock waves to verify previously developed excitation and radiative flux models.

Technical Results:

Emphasis thus far has been directed toward the first task, that of determining the role of NO in defining the resultant shock-induced vacuum ultraviolet radiative flux. This semi-annual report presents a brief discussion of the problem, a presentation of experiment definition, calculations, and the experimental design and fabrication of diagnostic instrumentation to date. Calculations have been completed to determine the important experimental parameters, such as shock speed, test gas mixture, initial pressure, etc. An equilibrium shock-wave computer program was utilized, and various test gas mixtures of N₂, O₂ and NO in Ne were considered. The requirements of the splitter-place absorption experiment discussed in detail in Section II. 1. 2 indicated that mixtures of Ne-NO and Ne-N₂-O₂ will be utilized for the initial

ii

series of tests.

In addition to the absorption measurements, additional measurements will be made of the nonequilibrium radiation overshoot behind the incident shock wave. The optical requirements for highly spatially resolved radiation overshoot measurements are very stringent, and are discussed in Section II. 1. 3. Special radiometers have been designed and fabricated for use in this phase of the program, and will be bench-tested prior to depolyment in the test series.

Comments:

The major emphasis will now be directed toward the NO absorption and nonequilibrium overshoot measurements, to determine the role of NO in VUV flux emitted from shock-heated gas mixtures.

I. INTRODUCTION AND DISCUSSION

The interactions of radiation with gases, producing excited as well as photoionized atoms and molecules, are of fundamental importance to a broad range of problems. Many of these problems may be treated by drawing upon a large body of knowledge derived through research in spectroscopy and related disciplines. Most of the available data, however, apply to equilibrium conditions, and in the case of gases, to those at room temperature. Thus many current problems, in which gases are found at high temperatures or under highly nonequilibrium conditions, require additional information for their solution. The radiative properties of shock heated gases are much different from those measured at equilibrium conditions, and extrapolations to nonequilibrium situations cannot readily be made.

Under strong-shock conditions, vibrational and rotational excited states of the molecular species are populated in nonequilibrium distributions, where different temperatures must be assigned to each of the internal energy modes. Further, the excitation of electronic states in atoms and molecules populates energy levels, especially metastables, which normally are unpopulated and optically inactive. Thus, for example, the absorptive processes which must be considered in a given problem are critically effected by the state of the gas.

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The calculation of the ionizing radiative flux emerging from a shockheated gas involves a knowledge of the gas dynamics and chemistry of the flow field, the population of the excited states responsible for the radiation, the spectral shape and distribution of the radiation in the wavelength region of interest, and a knowledge of the radiation transfer process including emission and absorption in the shock-heated gas. Examples of such situations are found in areas of nuclear effects as well as reentry physics. The rate of early fireball growth, for instance, involves the interaction of photons with shocked atmospheric gases. Hence, not only the normal absorption but also the extent of the photoionized region is affected by the prevailing mechanisms of photon interaction with shocked gases in excited states.

The tasks addressed in the present research program are based on results obtained in the course of a reentry-related research program in which the photoionization of the ambient atmosphere in front of a blunt reentry body was treated. For this work, the data were acquired for the specification of the vacuum ultraviolet (VUV) flux emerging from the shocked gas. The nonequilibrium radiative and absorptive properties of shock heated N₂, at wavelengths below 1000 Å were measured and used to predict the net VUV flux from the bow shock. ^(1, 2) Further calculations then permitted the determination of the properties of the resultant precursor photoelectron plasma ahead of this shock, which in turn affects the radar cross section of the system. ⁽³⁾

In the course of these experimental and analytical studies, two basic problems were identified which may affect these analyses. They are (1) the question of the contribution of the NO molecule in the determination of the nonequilibrium VUV emissive and absorptive properties of shock-heated air, and (2) the assessment of the role of multiple-step photoionization processes in air.

In general, the model of interest for production of photoelectrons in the atmosphere considers the strong ultraviolet radiation arising from shockheated air as the source of the ionizing radiation. This occurs, for example, in the production of a precursor plasma ahead of a reentering vehicle and also contributes to the intense ionization associated with an atmospheric nuclear explosion. Photons leave the shock-heated gas and interact with the ambient atmosphere. Of the principle constituents composing the ambient atmosphere, moleculer oxygen has the lowest ionization threshold of 12. 1 eV. Thus, considering a one-step photoionization process, only photons with an energy in excess of 12.1 eV are capable of producing photoionization. This energy corresponds to a wavelength of 1027 Å, and hence a one-step photoionization process has a sharp onset at this wavelength and extends to shorter wavelengths. For the precursor plasma formation, as an example, the primary source of the ionizing flux has been attributed to radiation from high-lying (> 12 eV) energy states of the nitrogen molecule. Since these states lie above

the dissociation energy of N_2 , the radiative transitions are functions of the rate of population of these states as compared with the rate of dissociation of the nitrogen molecule. The studies at CAL have shown this excitation process to be extremely rapid, giving rise to a highly nonequilibrium overshoot in the VUV radiation extremely close to the shock front. (1, 2, 3)

The importance of absorption close to the shock front in determining the net radiative flux emanating from a reentry gas cap was demonstrated in radiative flux calculations for typical reentry vehicles.⁽³⁾ A radiative transfer program developed at CAL⁽²⁾ was used to obtain the integrated flux from a reentry body to the bow shock wave, including nonequilibrium emission and absorption processes. The emission was determined from the instantaneous population of high-lying electronic states in N2, while the absorption term was obtained using the population of the ground state vibrational levels of nitrogen only. Very close to the shock front, it was found that absorption was the dominant process and the integrated flux was decreased from the calculated maximum value before leaving the shock surface. However, this calculation did not include possible absorption due to molecular NO, which also reaches its peak concentration close to the shock front. Based upon available data for cold NO absorption, it may be expected that NO will also affect the VUV flux intensity, which highlights the need for absorption coefficient data as a function of temperature.

I.1 Radiative Properties of NO in the Vacuum Ultraviolet

Behind a strong shock wave in air, NO is formed primarily from the fast reactions.

 $O + N_2 \longrightarrow NO + N,$ and $N + O_2 \longrightarrow NO + O$

where the initial O atoms arise from the rapid dissociation of O_2 . The NO concentration reaches a peak very close to the shock front, then decreases to its final equilibrium value. Thus, the possible role of NO in the emission and absorption of VUV flux in the shock-heated gas is dependent upon its formation rate as compared with the rate of excitation of the high-lying N₂

states and the vibrational relaxation rate of both N_2 and NO. Finite-rate chemistry shock wave computations $^{(4, 5)}$ have shown that the peak NO concentration occurs close to the peak in the N_2 excited state population, and the question arises as to the role of NO in the emission and absorption processes of VUV radiation in the shock layer.

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The studies at $CAL^{(1, 2, 3)}$ have shown that the low-lying vibrational levels of the ground state N₂ molecule contribute to absorption in the 800-1100 Å wavelength region. Thus, absorption due to N₂ is a function of the vibrational temperature, which determines the vibrational level population behind the shock wave. There have been numerous investigations reporting substantial absorption in cold NO in the same wavelength region. ^(6, 7, 8) These cold NO measurements can be compared with similar measurements in cold N₂, ^(9, 10) which indicate that the absorption coefficient of NO is comparable to, and at some wavelengths, substantially greater than that of cold N₂. For example at 897 Å, $k_{N_2} \approx 670$ cm⁻¹ while $k_{NO} \approx 9000$ cm⁻¹. ⁽⁶⁾ Thus, although the peak concentration of NO is calculated to be about 1/10 that of N₂, the much larger absorption coefficient at selected wavelengths may be expected to affect the spectral distribution as well as the net intensity of the VUV flux leaving the shock surface.

The measurement of absorption coefficients in cold gases, such as N_2 and NO, delineate overall wavelength regions where absorption is important, aid in identifying the high lying molecular states of the transitions, and provide, in some cases, the relevant spectroscopic constants. In general, however, these cold measurements are inappropriate for radiative flux calculations in shock-heated gases. First, in the heated gas, higher-lying vibrational levels are populated (rather than just $V''_z 0$) and lead to additional absorption in other wavelength intervals. Secondly, the extremely high rotational temperatures encountered in a shock layer alters the shape of the emission and absorption spectrum, toward a continuum-like distribution. Thus, absorption experiments in a heated gas are required to obtain accurate measurements applicable to radiative flux calculations in shock heated gases.

I.2 Multiple-Step Photoionization Processes

One of the dominant photoionization process in air is the direct, onestep process in which molecular O_2 is ionized. As mentioned earlier, the energy required for this process is 12.1 eV, corresponding to a wavelength of 1027 Å. Therefore, of all the radiation leaving the gas cap of a blunt reentry vehicle at velocities near 25 kft/sec, only that component of the radiation in the VUV below $\lambda = 1027$ Å can produce photoelectrons by this process. It is for this reason that the radiation from N₂ between 800 and 1100 Å was measured in the previous precursor ionization study. ⁽³⁾ At $\lambda < 800$ Å, the emergent radiation intensity decreases sharply because of the temperatures involved; this sets the lower lumit to the wavelengths considered in that problem.

The fundamental premise of one-step photoionization, however, may be of considerable moment to the problem. It is possible, for example, that the high photon flux at $\lambda > 1027$ Å, which cannot produce photoelectrons in the ambient O_2 , can nevertheless "pump" or process the gas, such that ionization can subsequently be induced by photons of lower energy (longer wavelength). Furthermore, the fraction of the flux at $\lambda < 1027$ Å which does not produce electrons is also available to "pump" the gas. Therefore, because of the high flux levels over the entire spectrum, even an inefficient multiplestep photoionization process may produce a significant amount of photoelectrons relative to the one-step process.

Approximate guidelines for the wavelength region of study can be deduced from the known properties of O_2 . For example, no absorption by O_2 takes place at wavelengths longer than 2000 Å. Therefore, the entire spectrum from 2000 Å through the near UV, visible and IR is ineffective in pumping the O_2 molecules. Below 2000 Å however, and extending to 1300 Å is a broad, strong continuum absorption in O_2 which leads to photodissociation. One therefore obtains, in the region ahead of the shock wave a zone of O-atom production. These atoms can recombine and react in a number of ways to produce O_2 in metastable states. Photo-absorption with subsequent cascading

to excited states can also result in the production of long-lived excited states. Photoionization cross sections for the various likely states are not known but there is no a priori reason to assume them small or negligible. Thus, because of the high radiative flux levels, photoelectron production via "pumped" excited states is a process whose significance in the overall ionization of air should be investigated.

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The experimental research program is designed to provide data pertinent to vacuum ultraviolet photoionization processes in air. To date, the effort has been directed toward the first task, that of determining the role of NO in the spectral distribution of shock-induced VUV radiation.

II. 1 Vacuum Ultraviolet Radiation-Measurements

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II.1.1 Description of Experiment and Results in N2

Prior to 1967, measurements in cold N₂ had been made, ^(9, 10) wherein absorption from the v''=0 level of the ground state N₂ molecule was observed. In 1967, absorption measurements (via shock-tube techniques) from shockheated, vibrationally-excited nitrogen were reported. ⁽¹¹⁾ Data were obtained in the window region (i. e. $\lambda > 1050$ Å), wherein absorption was attributed to transitions from the v''=5 thru v''=13 vibrational levels of the ground state. The research program at CAL extended these measurements into the windowless region of the VUV, and absorption measurements were obtained from 800 Å - 1100 Å in shock-heated nitrogen. ^(1, 2) Absorption from the v''=0through v''=5 vibrational levels are responsible for N₂ absorption in this wavelength interval. Since a similar approach is planned to obtain data in shock-heated NO, the experimental technique and previous absorption data will be briefly discussed.

Absorption measurements of shock-heated nitrogen were obtained in a high-purity shock tube designed for radiation studies. The pertinent feature of the experimental arrangement is a 3-channel vacuum ultraviolet spectrometer coupled to the shock tube by an explosively-driven plunger unit, which serves as a fast-acting valve-shutter combination. This windowless plunger was developed under a research program in which photoionization cross sections for N, O, and C were obtained (12, 13, 14) from emission measurements in the windowless region of the vacuum ultraviolet ($\lambda < 1050$ Å). For the N₂ investigation, a splitter plate was designed which fits into the shock tube at

the reflecting wall (Fig. 1a). The oncoming incident shock is divided and proceeds down two separate channels. One channel is obstructed and the shock is reflected. This pocket of gas serves as the light source (I_0). The light passes through a small aperture in the splitter plate, continuing through the gas in the second channel and into the spectrometer through the shutter-valve. The gas in this channel has been processed by the incident shock only, and hence is heated to a significantly lesser degree. The operation of the splitter plate is shown in Fig. 1a, with typical radiation data given in Fig. 1b. For the series of N₂ absorption experiments performed at CAL, (1, 2, 3) a test gas mixture of 10% N₂ + 90% Ne was used, giving a reflected-shock temperature of 11, 300° K which provided a continuum source of radiation (I_0) over the wavelength region of interest. (12) The absorbing gas behind the incident shock was at a temperature of 6000° K.

The lower trace in Fig. 1b is from a detector monitoring the total reflected-shock radiation history at a wavelength of about 1300 Å. The upper trace is the recorded signal from one channel of the VUV spectrometer viewing radiation at 760 Å which has been partially absorbed by the incident shockheated gas. A comparison of this signal to that obtained with the reflectedshock light source on the spectrometer side of the splitter plate gives the fraction of light transmitted through the heated gas.

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Figure 2 shows the absorption measurements from the experimental series. The splitter plate data are in excellent agreement with that predicted from the absorption measurements given in Ref 11. It can be seen that for wavelengths greater than about 1150 Å, the shock-heated N_2 is transparent, the higher vibrational levels responsible for absorption in this wavelength region not being sufficiently populated at 6000° K. However, as the wavelength decreases, the absorption increases markedly. This is to be expected, since the more highly populated lower vibrational levels (v'' = 0, 1, 2, 3) \sim are responsible for absorption coefficients. In the wavelength region data (I/I_0) have been converted to absorption coefficients. In the wavelength region of high absorption, the k value reaches a maximum near 800 cm⁻¹ atm⁻¹.

II. 1.2 Definition of NO Splitter-Plate Experiment

An absorption measurement program for NO, similar in scope to that described above for N_2 is currently underway. Prior to obtaining actual experimental measurements, calculations have been completed which serve to define the experimental test conditions. From the description of the previous N_2 absorption experiments, it can be noted that several criteria must be fulfilled in order to obtain suitable absorption data. The test gas mixture, pressure and incident shock speed must be compatible with: (1) attainment of reflected shock conditions (i. e., temperature) so as to obtain a suitable pocket of gas to serve as a continuum light source I_0 , and (2) incident shock conditions of temperature and species density, so as to obtain a measurable range of absorption in the test gas.

Equilibrium shock wave computations have been completed⁽¹⁵⁾ for various test gas mixtures. Based on the N₂ experiments, a test gas mixture consisting of 90% Neon as the carrier gas, at a total pressure of 2 torr was used in the computations. Neon is used to obtain high reflected shock temperatures and is optically inactive in the wavelength range of interest, $800 < \lambda < 1100$ Å. Both incident and reflected shock conditions, over a range of shock strengths, were calculated for the following aas mixtures:

> 90% Neon + 10% NO 90% Neon + 10% Air (8% N_2 + 2% O_2) 90% Neon + 5% N_2 + 5% O_2 90% Neon + 2% N_2 + 8% O_2 ,

and compared with the previous calculation for

90% Neon + 10% N₂.

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Figure 3 shows the computed incident shock temperature as a function of shock speed. For the previous N_2 experiments, a shock velocity of 13,700 ft/sec was used, yielding an incident shock temperature of 6000° K for the 10% N_2 test gas mixture. It can be seen that the temperature curves are similar for the various gas mixtures, with temperatures in the range 4500 - 6500° K

under consideration for the NO experiments. The corresponding reflected shock temperatures are shown in Fig. 4 for the various gas mixtures. For the incident shock temperatures of interest, the corresponding reflected shock temperatures range from about 8000 - 13,000° K.

Figure 5 presents the NO species concentration behind the incident shock wave (also shown is the N_2 calculation as a reference). It is the NO concentration, in combination with the wavelength-dependent absorption coefficient that determines the degree of absorption due to NO taking place in the test gas. The calculations presented in Fig. 6 show the ratio of N_2 molecule concentration to that of the NO concentration existing behind the incident shock wave. It can be seen that this ratio reaches a maximum at shock speeds near 13,000 ft/sec.

The combined results of the computations shown in Fig. 3-6 are employed to define the shock velocity and test gas mixture to be used in the initial absorption experiments. Since the N_2 molecule contributes substantially to the absorption process in the wavelength region of interest, it is desirable that the N_2/NO ratio be kept to a minimum. From Fig. 6, it can be seen that the 10% air-test gas mixture yields ratios greater than 100; thus this mixture will not be used in the experimental series. As the initial percentage of N_2 is decreased, the resultant N_2/NO ratio is also decreased, as can be seen from the 2% $N_2 - 8\% O_2$ computation. Here the ratio is over an order of-magnitude lower than the air results. From Fig. 6, then, it can be determined that mixtures other than air are required as the test gas. In addition, due to the shape of the N_2/NO ratio curves, it is recommended that shock velocities either larger or smaller than 13,000 ft/sec be utilized so as to obtain a minimum ratio for a given test gas mixture.

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At lower shock velocities, the reflected shock temperature is decreased (see Fig. 4); thus the resultant source radiation, I_0 , is reduced substantially. At higher shock velocities, the reflected shock temperature is increased; however, the absolute value of the NO concentration is substantially decreased (see Fig. 5), which in turn may yield very small values of absorption. The

curves in Fig. 5 indicate that the absolute value of NO concentration is relatively insensitive to the initial test gas composition, differing by about a factor of 2 or 3 for the various mixtures investigated. This can be compared to the N₂/NO ratio shown in Fig. 6, which was shown to be very sensitive to the initial test gas composition. Based upon the previous experimental determination of the N₂ absorption coefficient, ^(1, 2, 3) higher shock velocities near 16,500 ft/sec for the pure N₂ mixture would yield a value of I/I₀ \approx 0.80, or about 20% absorption. The curves in Fig. 5 indicate that at this velocity, the concentration of NO is approximately 2-3 orders of magnitude below the pure N₂ concentration, indicating that the test gas would be essentially transparent for comparable values of absorption coefficient.

Thus, the calculations indicate that the initial tests in the experimental series will be conducted at lower shock speeds (10,000 - 11,000 ft/sec), where much larger values of NO concentration can be obtained, see Fig. 5. Although the lower reflected shock temperatures yield a reduced source flux, I_0 , the experimental results from the previous N_2 tests indicate that suitable radiance levels will be obtained with the VUV spectrometer detectors.

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Approximate values of total absorption due to both N_2 and NO in the wavelength interval of interest (800 Å $\leq \lambda \leq 1100$ Å) have been estimated for several incident shock velocities and test gas mixtures. The measured^(1, 2, 3) values of vibrationally excited nitrogen absorption coefficients were used for the N_2 contribution, while absorption coefficients obtained in cold NO^(6, 7) were used to estimate the NO contribution. From the previous measurements, it is felt that the use of room temperature data yields a conservative estimate, since the higher vibrational and rotational temperatures existing in the shocked gas gives rise to larger values of absorption coefficient for a specified wavelength interval, $\Delta \lambda$. These estimates indicate that the absorption in the test gas is increased from 20% - 50% due to the presence of NO, over that which would occur with just N_2 present in the shocked gas. The initial experiments will be conducted with both N_2 and NO gas mixtures to obtain an assessment of the NO absorption contribution.

II.1.3 Nonequilibrium Radiation Overshoot Measurements

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In addition to the absorption measurements described above, additional measurements will be made of the nonequilibrium radiation overshoot behind the incident shock, such as shown in Fig. 7. These measurements, showing overshoot radiation at several wavelengths, have proven a powerful diagnostic tool, enabling excitation mechanisms for various N_2 band systems to be formulated. ^(1, 2, 3) Similar, highly-resolved measurements will be obtained for the NO test gas mixtures described above.

The optical requirements for spatially resolved radiation overshoot measurements are very stringent. As can be seen from Figure 7, the overshoot lasts about 3 μ sec, and the time to peak can be as short as 1 μ sec. Thus, its resolution requires sampling at about .1 to .2 μ sec. This is not an excessive electronic requirement. However, at shock speeds near 5 mm/ μ sec, the equivalent spatial sampling volume lies between 1/2 and 1 mm in extent. Thus, the radiometer field of view is restricted to no more than 1 mm wide in the direction normal to the shock wave. It can be higher in the other dimension, to increase signal intensities. The ideal beam geometry thus is a narrow fan-shaped beam.

The definition of this beam is complicated by scattered light effect. If slits are used near the shock tube window, considerable scattering from any practical slit occurs as the shock wave approaches the viewing station. This gives rise to a precursor "foot" ahead of the true signal rise that occurs when the shock wave enters the field of view. In the radiometers that have been designed for these studies, this problem has been circumvented by designing an optical system that makes use of slit images near the shock tube, see Figure 8. A spherical mirror is used to focus two fixed slits. One slit is focussed at the window of the shock tube, while the second focusses on the far wall of the tube. In this manner only radiations from the beam precisely defined by these images is permitted to enter the detector. Furthermore, no physical slits or window edges are illuminated by the advancing

shock wave, giving rise to crisp, sharply defined radiation profile measurements. The optical properties of such a radiometer has been discussed in Reference 16.

These radiation profile records will be used to assess the influence of NO on the nonequilibrium emission from the shock heated test gases. Differences will be sought between gases with varying NO, N_2 and O_2 concentrations that result from the formation and excitation kinetics of NO behind the shock waves.

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III. CURRENT STATUS AND FUTURE PLANS

The major emphasis for the next six weeks will be directed toward the NO absorption and nonequilibrium emission overshoot measurements, to determine the role of NO in VUV flux emitted from shock-heated gas mixtures. Based upon the calculations presented in Section II, neon-NO and neon-N₂-O₂ mixtures will be used in the experimental program, utilizing the splitter-plate technique.

High performance radiometers designed to measure the profiles of the nonequilibrium radiation behind strong shock waves in various test gases have been designed and assembled. They are presently being bench tested and will be ready for deployment in the aforementioned test program.

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FIG. 1 (a) WV EXPERIMENT (b) TYPICAL RADIATION DATA



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TRANSMISSION THROUGH SHOCK-HEATED NITTOGEN F/G. 2



FIG. 3 CALCULATED EQUILIBRIUM TEMPERATURE BEHIND INCIDENT SHOCK WAVE





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SHOCK - HEATED NITROGEN; P=0.5 TORE, VS = 5.05 Mm use

